Photon localisation and spontaneous parametric fluorescence

A thesis submitted for the degree of Doctor of Philosophy
of the University of London by

David Mark Rosewarne
King's College London

September 1992

Abstract

During the past decade there has been much interest in non-classical states of light. This thesis concerns two such states: the single photon state and the two photon state produced in spontaneous parametric fluorescence.

Our interest in the single photon state centres around its localisation properties. We develop the theory of localisation of relativistic particles in the group-theoretical setting due to Wightman and Mackey and explicitly show the photon not to be localisable. We examine the notion of generalised localisability for massless relativistic particles which was introduced by Jauch and Piron and comment on the practical application of these ideas of relativistic localisation.

The theory of photon pair generation in spontaneous parametric fluorescence, as originated by Mollow and extended by Pike and Sarkar, is reviewed. This theory is applied to the analysis of fourth-order interference experiments, in which two beams emerging from a source of two-photon fluorescence are mixed on a beamsplitter, and to two-colour interference experiments, which mix four beams.

Two alternative analyses of fourth-order interference phenomena are presented. The former adapts Kleinman's analysis of spontaneous parametric fluorescence to an experimental arrangement of mirrors and beamsplitters. The latter regards the effect as produced by the quantum-mechanical interference of alternative possibilities, each with an associated amplitude calculable by simple rules.

Finally, we give an analysis of the quantum fluctuations in a model dielectric material which demonstrates the inadequacy of the quantised macroscopic Maxwell equations when dealing with fluctuations and squeezing in real dielectric materials.

For Mum and Dad

Contents

List of Figures

Acknowledgements

I would like to thank my supervisor, Professor E.R. Pike F.R.S., for help and encouragement during this period of research, and to Dr. S. Sarkar for arranging a summer visit to the then R.S.R.E., Malvern, which facilitated useful contact with the quantum optics group there. This work was supported by a S.E.R.C. quota studentship, and this thesis typeset using the LaTeX document preparation system.

Chapter 1

Introduction

In the past thirty or so years the field of quantum optics has grown to cover a diverse range of phenomena, but in the past decade there has been a special interest in the theoretical and experimental properties of non-classical states of light [26, 33]. These are states in which a classical wave picture of optical phenomena, or even a scheme in which optical effects are produced by particles called photons subject to classical probability laws, is inadequate.

The simplest examples of non-classical states arise when we may consider just one mode of the radiation field, and within this context the production, for example, of squeezed states [18] by such experimental techniques as non-linear optical interaction, has been much discussed. When we consider the electromagnetic field as a whole, the range of possible phenomena is much wider. This thesis is concerned with two such quantum states of the electromagnetic field, the single-photon state and the two-photon state produced in spontaneous parametric fluorescence.

Our interest in the single-photon state centres around its localisation properties. In classical physics the fundamental kinematic quantities describing the state of a particle at any instant in time are its position and momentum, and Newton's laws of motion (cast in Hamiltonian form) then allow us to predict the motion of the particle. In the new quantum mechanics of Schrödinger and Heisenberg, which describes the dynamics of non-relativistic particles, position and momentum are again the fundamental kinematic quantities, but this time represented by self-adjoint operators in a Hilbert space, the vectors of which represent possible states of the system.

After the tremendous success of this theory in predicting the properties of the hydrogen atom and then more complicated atoms and molecules, the next step was to create a quantum mechanics consistent with special relativity. The Klein–Gordon and Dirac relativistic wave equations are the two best-known attempts. For reasons explained in Chapter 2 the task of con-

structing a position operator for these equations is not quite trivial and we are led to a group-theoretical approach for the construction of position operators. This method also shows the impossibility of constructing a position operator for massless particles with spin, a fact which had been anticipated by Landau and Peierls [17]. However, from a practical point of view, we are used to associating a position with a photon in the sense that we may place a photodetector in a specific region of space and that if a photon is detected by this apparatus then it is natural to suppose that a photon was present in the region of space in which the detector rests. A model was developed by Glauber [10] of the radiation–matter interaction occurring in photodetection, which is of great practical use, but lacks a truly fundamental basis. Jauch and Piron [15] and Amrein [1] tried to generalise the group-theoretical approach to localisation to cover the case of massless particles with spin by the construction of a 'generalised imprimitivity'. In this way they attempted to define the localisation of the photon in a manner intrinsic to the photon, without reference to any specific model of photodetection. A feature of their formalism is that a photon localised in some region is associated with an energy density which ultimately decays as the inverse seventh power of distance from the region of localisation. We shall investigate the concept of generalised localisability to see how it squares with our intuition that a localised particle, which is not itself the source of a field, should only produce physical effects in the region in which it is localised.

The two-photon state in which we shall be interested is that produced by spontaneous parametric fluorescence where single photons are destroyed and a pair created in their place by certain materials with a second-order optical non-linearity. In Chapter 3 we are interested in the correlated times of arrival of these photon pairs at detectors placed external to the non-linear medium. As Mollow points out [21], such an effect cannot be treated by classical or semi-classical methods and he attempts to develop a fully quantum-mechanical analysis of this phenomenon, which we present in some detail. Unfortunately, it turns out that the details of this theory cannot yet be verified experimentally, for technological reasons. The fourth-order interference experiments described in Chapters 4 and 5 offer a possible experimental testing ground for Mollow's theory, in which instead of the fluorescence being directly incident on detectors, it is first incident on a beamsplitter and only subsequently on detectors. We also describe an extension of Mollow's theory, due to Pike and Sarkar [24, 25] which predicts power-law tails in the dual photocount amplitude, and which they suggested may be related to the power-law tails of Amrein's single-photon localised state. This possible link forms the original motivation for the study of these two rather different phenomena in this thesis.

The analysis of two-colour interference [23, 29], also in Chapter 4, shows that there is more to fourth-order interference effects than meets the eye, and that something definitively quantum-mechanical and counter-intuitive is happening in this experimental arrangement. For this reason, and because there is reason to question the fundamental basis of Mollow's analysis, we investigate in Chapter 5 two alternative approaches to the analysis of fourth-order interference, the first based on Kleinman's approach [16] to the description of spontaneous parametric fluorescence, and the second showing fourth-order interference to be explicable in terms of the interference of indistinguishable alternatives, each with an associated quantum-mechanical amplitude [30].

Appendix A describes enough of the theory of group representations and systems of imprimitivity associated with induced representations to understand the group-theoretical approach to localisability and to construct the irreducible unitary representations of the Poincaré group. These are required for a discussion of the quantum mechanics of elementary particles, the photon in particular.

Appendix D is a corrected version of the text of a paper by this author [31] which aims to highlight some of the problems involved in a quantum-mechanical description of dielectric materials, the non-classical aspects of which have recently provoked some interest [11].

Chapter 2

Localisability of quantum-mechanical particles

2.1 Localisability and the position operator in non-relativistic quantum theory

In this section, in the familiar setting of the non-relativistic Schrödinger equation for a spinless particle moving in a velocity-independent potential, we demonstrate the close link between the existence of a position operator for a particle and the Born interpretation of the wavefunction which tells us the probability of finding a quantum particle localised in a given region of space.

The state, ψ, of the particle at a given time is represented by an L^2 function $\psi : \mathbb{R}^3 \to \mathbb{C}$ and a scalar product between two states ψ and ϕ is defined by

$$(\psi, \phi) = \int \mathrm{d}^3\vec{x}\, \psi^*(\vec{x})\phi(\vec{x}), \tag{2.1}$$

so that we now have a Hilbert space, the vectors of which represent possible states of the system. The kinematical requirements of the theory are completed by adding self-adjoint position and momentum operators $\vec{q}$ and $\vec{p}$ whose action on the states is then

$$(\vec{q}\psi)(\vec{x}) = \vec{x}\psi(\vec{x}), \tag{2.2}$$
$$(\vec{p}\psi)(\vec{x}) = -i\nabla\psi(\vec{x}). \tag{2.3}$$

These operators are easily shown to satisfy the canonical commutation relations

$$\left((q_i p_j - p_j q_i)\psi\right)(\vec{x}) = i\delta_{ij}\psi(\vec{x}). \tag{2.4}$$

Having the position operator, we may then define the moment-generating function, $\mu(\vec{k})$, for the particle's position:

$$\mu(\vec{k}) = \langle\psi|e^{i\vec{k}\cdot\vec{q}}\psi\rangle = \int \mathrm{d}^3\vec{x}\,\psi^*(\vec{x})e^{i\vec{k}\cdot\vec{x}}\psi(\vec{x}), \tag{2.5}$$

and from this the probability distribution $p(\vec{x})$ for the particle position is available,

$$p(\vec{x}) = \int \frac{\mathrm{d}^3\vec{k}}{(2\pi)^3}\mu(\vec{k})e^{-i\vec{k}\cdot\vec{x}} = \psi^*(\vec{x})\psi(\vec{x}), \tag{2.6}$$

this being the standard Born interpretation of the wavefunction. We now have an entirely satisfactory picture of a particle's localisation properties, it being possible to calculate the probability of finding a particle in a given region, R, of space by calculating

$$\int_R \psi^*(\vec{x})\psi(\vec{x})\,\mathrm{d}^3\vec{x}. \tag{2.7}$$

2.2 The position operator in relativistic wave equations

In the case of the Schrödinger equation the matter of the position operator is simplified because Schrödinger determines the action of the position operator by fiat and there is plenty of experimental evidence to attest to the wisdom of his choice. Newton and Wigner [22] were the first to have much success in their attempt to construct a position operator for a relativistic elementary system (where by an elementary system we mean a system in which any state is a superposition of states attainable from a *single* state by the action of elements of the Poincaré group). Previous attempts had undesirable features, such as introducing negative-energy components to a state by their action, or not having three commuting components. As Newton and Wigner point out, the problem faced in the relativistic problem is that the theory only directly provides the components of the energy-momentum vector and the relativistic angular-momentum tensor. The challenge, then, is to construct a unique, well-defined and self-adjoint position operator satisfying the canonical commutation relations $[q_i, p_j] = i\delta_{ij}$ within the relativistic theory.

They start by attempting to construct the wavefunction of a state (or states) localised at the origin. From this the wavefunctions of states located at other points can be deduced by using the translation operator (which is constructable from the momentum operator) to generate all the other

position eigenstates. Knowing the set of eigenstates with their eigenvalues determines the position operator.

The postulates required for the states located at the origin are:

- They form a linear manifold S_0; the superposition of two states localised at the origin is also localised at the origin.
- S_0 is invariant under spatial rotations about the origin and reflections of both the space and time coordinates.
- A state in S_0 is made orthogonal to all states of S_0 by any spatial displacement.
- Certain regularity conditions, which amount to the requirement that all the generators of the Lorentz group may be applied to the localised states.

Although Newton and Wigner treat particles both with and without spin, only the spinless case will be presented here, as it demonstrates the essential features of the method.

The aim is to construct a position operator for the positive-energy solutions of the free Klein-Gordon equation. We represent the state ψ by the momentum space wavefunctions $\psi(\vec{p})$ with a relativistically invariant scalar product between states

$$(\psi, \phi) = \int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3 2p_0}\, \psi^*(\vec{p})\phi(\vec{p}), \tag{2.8}$$

where $p_0 = \sqrt{p^2 + m^2}$, and the wavefunction in coordinate space is

$$\phi(\vec{x}) = \int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3 2p_0}\, \phi(\vec{p}) e^{i\vec{p}\cdot\vec{x}}. \tag{2.9}$$

For a spin-zero particle the manifold of states localised at a point is one-dimensional, so, taking into account rotational invariance, the state localised at the origin, $\phi_{\bar{0}}(\vec{p})$, must be of the form

$$\phi_{\bar{0}}(\vec{p}) = f(p). \tag{2.10}$$

The action of the time reversal operator, Θ, is

$$(\Theta\psi)(\vec{p}) = \psi^*(-\vec{p}), \tag{2.11}$$

and according to the second postulate, the action of the time-reversal operator on a localised state also yields a localised state. Thus if $f(p)$ represents

a localised state then so does $f^*(p)$. By the first postulate the sum of two localised states is also localised, and so taking $f(p) + f^*(p)$ to represent the localised state we may have the state localised at the origin represented by a real function without loss of generality.

The operator $T(\vec{a})$, which implements a displacement $\vec{a}$ upon the origin of coordinates, acts on these momentum space wavefunctions by

$$(T(\vec{a})\psi)(\vec{p}) = e^{i\vec{p}\cdot\vec{a}}\psi(\vec{p}), \tag{2.12}$$

this form for the operator being a necessary and sufficient condition for the implementation of the canonical commutation relations. Using the crucial third postulate, that any displacement of the localised state creates a state orthogonal to the original we have

$$\int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3 2p_0}\, \phi_{\bar{0}}^*(\vec{p}) e^{i\vec{p}\cdot\vec{a}} \phi_{\bar{0}}(\vec{p}) = 0, \tag{2.13}$$

for any non-vanishing $\vec{a}$. This shows that $|\phi_{\bar{0}}(\vec{p})|^2/p_0$ is constant. With $\phi_{\bar{0}}(\vec{p})$ real we have

$$\phi_{\bar{0}}^2 = p_0. \tag{2.14}$$

As far as the first three postulates are concerned $\phi_{\bar{0}}$ could be discontinuous, being $+p_0^{1/2}$ for some $\vec{p}$ and $-p_0^{1/2}$ for others. However, the fourth localised state hypothesis rules out the discontinuous case since the generators of the Poincaré group are differential operators in momentum space, which are only defined on a domain of differentiable functions, and we may write for the localised state

$$\phi_{\bar{0}}(\vec{p}) \propto p_0^{1/2}. \tag{2.15}$$

The localised state in coordinate space is obtainable from equation 2.9. It is given by

$$\phi_r(r) \propto (\mu/r)^{5/4} H^1_{5/4}(i\mu r), \tag{2.16}$$

where $H^1_{5/4}$ is a Hankel function and μ is the particle's Compton wavelength. It goes to zero as $r \to \infty$ as $\exp(-\mu r)$, so, for massive particles, the coordinate space wavefunction has an approximate extent of a few Compton wavelengths.

Applying the translation operator to the state localised at the origin we obtain, for the wavefunction of the state localised at $\vec{z}$,

$$\phi_{\vec{z}}(\vec{p}) = T(-\vec{z})\phi_{\bar{0}}(\vec{p}) = p_0^{1/2} e^{-i\vec{p}\cdot\vec{z}}. \tag{2.17}$$

Using these eigenfunctions of position, which give us the matrix elements between position eigenstates and momentum eigenstates, the position operator can be constructed, its action in momentum space being

$$(q_k\psi)(\vec{p}) = -i\left(\frac{\partial}{\partial p_k} + \frac{p_k}{2p_0^2}\right)\psi(\vec{p}). \tag{2.18}$$

In coordinate space the non-local action of this position operator becomes clear:

$$(q_k\psi)(\vec{x}) = x_k\psi(\vec{x}) + \frac{1}{8\pi}\int \mathrm{d}^3\vec{y}\,\frac{e^{-\mu|\vec{x}-\vec{y}|}}{|\vec{x}-\vec{y}|}\frac{\partial\psi(\vec{y})}{\partial y_k}. \tag{2.19}$$

With this expression for the coordinate operators' action on the momentum space wavefunction, we can then construct a moment-generating function for the coordinates and a probability distribution for the position of the particle.

2.3 Group-theoretical approach to localisability

Although Newton and Wigner made significant progress towards the construction of mathematically acceptable position operators, their procedure is plausible rather than compelling. In particular, we are still left wondering whether we have constructed a unique position operator. For this reason we turn to a group-theoretical approach to localisation pursued by Wightman [34], which is in a mathematically rigorous framework and shows that the position operators they construct are, to within unitary equivalence, the only ones possible. An exposition of the necessary mathematical background for this approach is available in appendix A.

For motivation, we start with the one-dimensional motion of a free spinless particle along the entire real line. The group of symmetries of this system is the group of translations which is associated with a unitary representation of the additive group of the real numbers $U(s)$. By Stone's theorem this set of unitary operators may be written

$$U(s) = e^{isp}, \tag{2.20}$$

where p is a uniquely determined Hermitian operator, which in physics, being the generator of translations, is called the momentum operator. It is fundamental to Heisenberg's conception of the quantum theory of this system that

there also should exist a Hermitian position operator, q, which (as usual) obeys the canonical commutation relation

$$[q, p] = i. \tag{2.21}$$

A position operator would be associated, by Stone's theorem, with another representation of the additive group on the real line

$$V(t) = e^{-itq}. \tag{2.22}$$

As a consequence of the canonical commutation relations it is easy to prove, at least formally, that $U(s)$ and $V(t)$ are related by

$$U(s)V(t) = e^{-ist}V(t)U(s), \tag{2.23}$$

these being known as the commutation relations in global or Weyl form. We now make use of the Stone-von Neumann theorem, which assures us of the existence, to within equivalence, of unique representations $U(s)$ and $V(t)$ which satisfy the Weyl commutation relations. In turn this implies the existence of unique p and q, to within equivalence, and so we have position and momentum operators that are 'mathematically' unique, though we shall later ask whether this is adequate for the needs of physicists.

Our whole group-theoretical approach will be based on a generalisation of the above. We must first re-express the global commutation relations in a form which makes generalisation possible. We express $V(t)$ in terms of its associated resolution of unity

$$V(t) = \int_{-\infty}^{\infty} e^{-i\lambda t}\, \mathrm{d}P_\lambda. \tag{2.24}$$

This resolution can then be used to construct a projection-valued measure $E(S)$ on the Borel sets of the real line. The global commutation relations are then exactly equivalent to

$$E(S)U(s) = U(s)E(S - s), \tag{2.25}$$

where the Borel set $S - s$ is obtained from S by subtracting s from each of the elements of S.

We now turn to the physical interpretation of these equations. From the spectral decomposition of q

$$q = \int_{-\infty}^{\infty} \lambda\, \mathrm{d}P_\lambda, \tag{2.26}$$

$\langle\psi|P_\lambda|\psi\rangle$ is the probability that measurement of the coordinate q will give a result $\leq \lambda$ when the system is in the state $|\psi\rangle$ and so $\langle\psi|E(S)|\psi\rangle$ is the probability that measurement of the coordinate will give a result in the Borel set S. We may obtain a physical understanding of equation 2.25 by noting that if we have a system in a state ϕ which is localised in the Borel set $S-s$ so that it is in an eigenstate of the projection operator

$$E(S-s)|\phi\rangle = |\phi\rangle, \tag{2.27}$$

then the state $U(s)|\phi\rangle$ is by equation 2.25 an eigenstate of $E(S)$ and so is localised in the Borel set S which confirms our intuition as to the behaviour of a localised system when we shift it by s units.

Having the commutation relations in the form 2.25 we may generalise them both mathematically and in their physical application. Mackey found that for an arbitrary separable locally-compact group, G, a projection-valued measure on the Borel sets of G may be constructed which has the following commutation relations with the regular representation U

$$E(S)U(g) = U(g)E(g^{-1}S). \tag{2.28}$$

This pair may be shown to be unique up to equivalence and is an obvious generalisation of 2.25.

If G has a closed subgroup K Mackey's imprimitivity theorem [19], detailed in appendix A, provides a further generalisation of equation 2.28. It classifies the types of representation that may obey this equation but with S no longer a Borel set of G, but a Borel set of G/K, demonstrating that these representations are equivalent to representations induced from representations of the subgroup K.

It was Wightman [34] who first introduced these mathematical developments of group representation theory to the problem of the rigorous construction of position operators for quantum-mechanical particles. As in the one-dimensional example we proceed from the group of symmetries possessed by the system. We are trying to define the kinematical properties of the particle, as we did for the non-relativistic case, and so we are interested in the picture at one time only. The full symmetry group of a free relativistic particle is the Poincaré group which involves space-time translations, rotations in space and boosts, and we assume the existence of a unitary representation U in the particle's space of states which implements these transformations. However, if we are interested in the system at one time, we restrict the group of symmetries to the Euclidean group, $\mathcal{E}_3$, which is a semi-direct product of the group of space translations (a typical element of which we label by $\vec{a}$) with the group of spatial rotations (with typical element R).

An acceptable position operator $\vec{q}$ must be self-adjoint in the system's space of states and have the correct transformation properties under the action of the representation U of $\mathcal{E}_3$,

$$U^{-1}(\vec{a}, R)\vec{q}U(\vec{a}, R) = R^{-1}(\vec{q} - \vec{a}). \tag{2.29}$$

This is equivalent to the requirement that there exists a projection-valued measure on the Borel sets of $\mathbb{R}^3$ with the following property

$$E(S)U(\vec{a}, R) = U(\vec{a}, R)E(R^{-1}(\vec{x} - \vec{a})), \tag{2.30}$$

which expresses the same intuitive properties of localised states as discussed in the one-dimensional case.

We now see how such a position operator is provided by Mackey's imprimitivity theorem. We make the technical change of dealing not with the $\mathcal{E}_3$, but with its universal covering group, $\bar{\mathcal{E}}_3$, since, as described in appendix A, we need to encompass the double-valued representations of $\mathcal{E}_3$ in our scheme. $\bar{\mathcal{E}}_3$ is formed from a semi-direct product of the three-dimensional translation group T^3 with $SU(2)$ and possesses composition law

$$(\vec{a}, A)(\vec{b}, B) = (\vec{a} + R(A)\vec{b}, AB), \tag{2.31}$$

where $\vec{a}, \vec{b} \in T^3$, $A, B \in SU(2)$ and $R(A)$ is the rotation in three-dimensional Euclidean space associated with A. In constructing representations of this group induced from the $SU(2)$ subgroup the first requirement is a Mackey decomposition of the group elements, which may be trivially performed,

$$(\vec{a}, A) = (\vec{a}, 1)(0, A), \tag{2.32}$$

so that the cosets are labelled by a three vector, in future denoted by $\vec{x}$. The induced representations from the $SU(2)$ subgroup will have an associated projection-valued measure on the Borel sets of $\bar{\mathcal{E}}_3/SU(2) \sim \mathbb{R}^3$ which is exactly what is required for a position operator.

The group action on the cosets is

$$(a, A)^{-1}(\vec{x}, 1) = (R^{-1}(A)(\vec{x} - \vec{a}), 1)(0, A^{-1}). \tag{2.33}$$

From this Mackey decomposition, following the procedure of section A.2, taking advantage of the invariant measure defined on the cosets,

$$\mu_i = \mathrm{d}^3\vec{x}, \tag{2.34}$$

and using the well-known form $D(A)$ of the representations of $SU(2)$ we obtain

$$U(\vec{a}, A)\phi(\vec{x}) = D(A)\phi(R^{-1}(A)(\vec{x} - \vec{a})). \tag{2.35}$$

With this representation goes the canonically associated projection-valued measure which does indeed satisfy the required commutation relations of equation 2.30. This is exactly what is needed for the notion of a localisable system and so every representation which is induced from $SU(2)$ is a possible representation of a localisable system. But the imprimitivity theorem is yet stronger than this and we can in fact say that every localisable system is equivalent to a representation induced from the $SU(2)$ subgroup of the Euclidean group.

2.3.1 Localisability of elementary particles

We now turn to the question of localisability for elementary particles in this group-theoretical framework. First we need the irreducible representations of the Poincaré group for massive particles of positive energy and for massless particles of positive energy, which we derive in appendix A. From these we obtain representations of these particles under space translation and rotation by restricting the transformations $(a, \bar{\Lambda}) \in \bar{\mathcal{P}}_0$ to $(\vec{a}, A) \in \bar{\mathcal{E}}_3$ and then investigate localisability in the light of the previous section.

Massive particles The canonical form of the representation for massive particles we give in equation A.64. Restricting it to $\bar{\mathcal{E}}_3$ we obtain, for a spin-j particle,

$$U(\vec{a}, A)\phi(p) = e^{-i\vec{p}\cdot\vec{a}} D^j(A)\phi(R^{-1}(A)p), \tag{2.36}$$

where $R^{-1}(A)$ acts on the space components of the four vector p. We now construct a system of imprimitivity based on $\mathbb{R}^3$ following Amrein [1]. We map $\phi(p) \in \mathcal{L}^2(p, \mathcal{H}(D^j), \mu_m)$ onto $\varphi(\vec{p}) \in \mathcal{L}^2(\mathbb{R}^3, \mathcal{H}(D^j))$ by

$$\varphi(\vec{p}) = \frac{1}{\sqrt{p_0}}\phi(p), \tag{2.37}$$

and then Fourier transform to obtain $\bar{\varphi} \in \mathcal{L}^2(\mathbb{R}^3)$

$$\bar{\varphi}(\vec{x}) = \int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3}\, e^{i\vec{p}\cdot\vec{x}} \varphi(\vec{p}). \tag{2.38}$$

The action of $\bar{\mathcal{E}}_3$ on the $\bar{\varphi}$'s then becomes

$$U(a, A)\bar{\varphi}(\vec{x}) = D^j(A)\bar{\varphi}(R^{-1}(A)(\vec{x} - \vec{a})), \tag{2.39}$$

and we define the action of a projection-valued measure on the $\bar{\varphi}$'s

$$(E(S)\bar{\varphi})(\vec{x}) = \chi_S(\vec{x})\bar{\varphi}(\vec{x}), \tag{2.40}$$

which explicitly shows massive relativistic particles to be localisable.

Massless particles Looking at equation 2.35 for the form of a representation of $\bar{\mathcal{E}}_3$ induced by $SU(2)$ we may already anticipate that problems will arise in the attempt to localise massless particles. The appearance of $D(A)$ which has the form of a countable direct sum of irreducible representations of $SU(2)$ implies a spectrum of helicity states from $|s|, |s|-1 \cdots -|s|$ for some integer s, this being familiar from the theory of angular momentum for massive particles. However, in the case of the neutrino, we know that it possesses only one helicity state and not the two required by massive spin-half particles. The photon, the focus of our particular interest, is a spin-one massless particle which lacks the helicity-zero state possessed by a massive spin-one particle. We now discuss the different cases more carefully and show precisely why the photon and neutrino are not localisable.

The irreducible representation of the Poincaré group corresponding to spin-0 is

$$U(a,\Lambda)\phi(p) = e^{ipa}\phi(\Lambda^{-1}p), \tag{2.41}$$

the representation of the $\bar{\mathcal{E}}_2$ little group that appears being trivial. It is easy to show that the procedure we applied in the massive case to construct the imprimitivity may again be used (setting $m = 0$ and $D^j = 1$). Therefore massless spin-zero particles are localisable.

For the spin $s \neq 0$ irreducible representations, the formal proof that these representations are not localisable rests on the comparison of the helicity operator h for the representation of the Euclidean group formed by restricting the representation of the Poincaré group to the helicity operator h' constructed for the canonical form of a representation of $\bar{\mathcal{E}}_3$ induced from a representation of the $SU(2)$ subgroup. If these were equivalent then so also would be the helicity operators

$$h' = WhW^{-1}, \tag{2.42}$$

with W a bijective map between the two spaces. For a given representation of $\bar{\mathcal{E}}_3$ the helicity operator is in the commuting algebra and the helicity eigenspaces are therefore invariant subspaces of the representation. If two representations are equivalent then 2.42 implies the existence of a one to one correspondence between the helicity eigenspaces and eigenvalues in the Hilbert spaces of the two representations.

As we have seen, the representation of the Euclidean group for a system that is localisable is of the form

$$(U(\vec{a},A)\phi)(\vec{x}) = D(A)\phi(R^{-1}(A)(\vec{x}-\vec{a})), \tag{2.43}$$

which, on making the Fourier transformation

$$\bar{\phi}(\vec{p}) = \int \mathrm{d}^3\vec{x}\,\phi(\vec{x})e^{-i\vec{p}\cdot\vec{x}}, \tag{2.44}$$

has the following action on the momentum space functions that we have just defined

$$(U(\vec{a},A)\bar{\phi})(\vec{p}) = e^{-i\vec{p}\cdot\vec{a}}D(A)\phi(R^{-1}(A)\vec{p}), \tag{2.45}$$

and so any localisable system must have a representation equivalent to this also. To construct the helicity operator we first need the momentum and angular momentum operators which are available from the $U(\vec{a},A)$'s in the neighbourhood of $\vec{a}=0$, $A=1$. Near the origin $SU(2)$ and $O(3)$ are isomorphic and with each rotation, specified by a direction $\vec{n}$ and angle θ we may associate a member of $SU(2)$

$$A(\vec{\theta}) = \sigma_0 - \frac{i}{2}\vec{\theta}\cdot\vec{\sigma}, \tag{2.46}$$

where $\vec{\theta}=\theta\vec{n}$. In the neighbourhood of the origin the representation 2.45 becomes

$$(U(\vec{a},\vec{\theta})\phi)(\vec{p}) = e^{-i\vec{p}\cdot\vec{a}}(1-i\vec{S}\cdot\vec{\theta})\phi(\vec{p}-\vec{\theta}\times\vec{p}), \tag{2.47}$$

where $\vec{S}$ is a countable direct sum of the finite-dimensional generators of the irreducible representations of $SU(2)$. We obtain the momentum and angular momentum operators by

$$(\vec{p}\phi)(\vec{p}) = i\left(\frac{\partial}{\partial\vec{a}}U(\vec{a},\vec{\theta})\Big|_{\vec{a}=\vec{\theta}=0}\phi\right)(\vec{p}) \tag{2.48}$$

$$(\vec{J}\phi)(\vec{p}) = i\left(\frac{\partial}{\partial\vec{\theta}}U(\vec{a},\vec{\theta})\Big|_{\vec{a}=\vec{\theta}=0}\phi\right)(\vec{p}), \tag{2.49}$$

which gives

$$(p_i\bar{\phi})(\vec{p}) = p_i\bar{\phi}(\vec{p}) \tag{2.50}$$

$$(J_i\phi)(\vec{p}) = \left(S_i - i\varepsilon_{ijk}p_j\frac{\partial}{\partial p_k}\right)\phi(\vec{p}), \tag{2.51}$$

and so the helicity operator takes the form

$$h = \vec{J}\cdot\vec{p} = \vec{S}\cdot\vec{p}, \tag{2.52}$$

which has the spectrum $|s|, |s|-1\cdots-|s|$ for some integer s.

We now turn to the case of the representations of the Poincaré group corresponding to massless particles. The restriction of these to the Euclidean is given by Amrein [1]:

$$U(\vec{a},A) = \left[\frac{(p+p_3)v-(p_1-ip_2)w}{|(p+p_3)v-(p_1-ip_2)w|}\right]^{2s} e^{-i\vec{p}\cdot\vec{a}}\phi(R^{-1}(A)\vec{p}), \tag{2.53}$$

where v and w specify a member of $SU(2)$

$$A = \begin{pmatrix} v & w \\ -w^* & v^* \end{pmatrix}. \tag{2.54}$$

To calculate the generators of the group representation we put

$$v = 1 - i\theta_3/2 \tag{2.55}$$
$$w = -i\theta_1/2 - \theta_2/2, \tag{2.56}$$

and calculate the momentum and angular momentum

$$(p_i\bar{\phi})(\vec{p}) = p_i\bar{\phi}(\vec{p}) \tag{2.57}$$
$$(J_1\phi)(\vec{p}) = \left(\frac{sp_1}{p+p_3} - i\left(p_2\frac{\partial}{\partial p_3} - p_3\frac{\partial}{\partial p_2}\right)\bar{\phi}\right)(\vec{p}) \tag{2.58}$$
$$(J_2\phi)(\vec{p}) = \left(\frac{sp_2}{p+p_3} - i\left(p_3\frac{\partial}{\partial p_1} - p_1\frac{\partial}{\partial p_3}\right)\bar{\phi}\right)(\vec{p}) \tag{2.59}$$
$$(J_3\phi)(\vec{p}) = \left(s - i\left(p_1\frac{\partial}{\partial p_2} - p_2\frac{\partial}{\partial p_1}\right)\bar{\phi}\right)(\vec{p}). \tag{2.60}$$

The helicity is

$$(\vec{J}\cdot\hat{\vec{p}}\bar{\phi})(p) = s\bar{\phi}(\vec{p}), \tag{2.61}$$

and so there is only a single-helicity eigenspace for these representations which makes them unlocalisable except for $s = 0$.

We finally turn to the case of the photon which does not transform as an irreducible representation of the Poincaré group as it may exist in a superposition of helicity eigenstates. It transforms as the direct sum of the helicity-plus-one and helicity-minus-one representations, but, by the argument above cannot be localisable as it lacks the helicity-zero state.

2.3.2 The generalised imprimitivity for massless particles

Trying to develop some notion of localisation for the photon, and attempting to reconcile the lack of position operator for a photon with the experimental observation that photons may be detected at given points of space, Jauch and Piron [15] introduced the concept of a generalised imprimitivity, which we now outline. The photon is described by a four-component wave function $A^\mu(p^\mu)$ defined on the lightcone $p^\mu p_\mu = 0$. We demand that A^μ satisfies the Lorentz condition

$$p^\mu A_\mu = 0. \tag{2.62}$$

The functions thus defined form a linear space $\mathcal{G}$. In $\mathcal{G}$ we introduce the sesquilinear form

$$\langle A|B\rangle = \int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3 2p_0} A^*_\mu(p) B^\mu(p), \tag{2.63}$$

which would serve as a scalar product in a Hilbert space were $\langle A|A\rangle$ vanishing for $A \neq 0$. However there is a subspace $\mathcal{G}_0$ of functions of the form $\eta^\mu = p^\mu \Lambda(p)$ which satisfies $\langle \eta|\eta\rangle = 0$. In the space $\mathcal{G}/\mathcal{G}_0$ the form we defined does have all the required properties and we now work in this space by choosing from each equivalence class in $\mathcal{G}$ the representative element

$$A^\mu_c(p) = A^\mu(p^\mu) - \frac{p^\mu}{p^0} A^0(p^\mu), \tag{2.64}$$

which physically corresponds to choosing the Coulomb gauge. Since $A^0_c = 0$ we write $A^\mu_c(p^\mu) = \vec{A}(\vec{p})$, and the Lorentz condition and scalar product become

$$\vec{p}\cdot\vec{A}(\vec{p}) = 0 \tag{2.65}$$

$$\langle A|B\rangle = \int \frac{\mathrm{d}^3\vec{p}}{(2\pi)^3 2p_0} \vec{A}^*(\vec{p})\cdot\vec{B}(\vec{p}). \tag{2.66}$$

If we attempt to define a projection-valued measure on the space as we did earlier,

$$A(\vec{x}) = \left(\mathcal{F}\frac{1}{\sqrt{p_0}}\vec{A}\right)(\vec{p}) \tag{2.67}$$

$$(E(S)A)(\vec{x}) = \chi_S(\vec{x})A(\vec{x}), \tag{2.68}$$

where $\mathcal{F}$ represents the operation of Fourier transformation, we encounter the problem that the projection operators take transverse functions $\vec{p}\cdot\vec{A}(\vec{p}) = 0$ and map them to non-transverse ones. Therefore Jauch and Piron define a further projection operator, P,

$$(P\vec{A})(\vec{p}) = \vec{A}(\vec{p}) - \frac{\vec{p}}{p^2}\left(\vec{p}\cdot\vec{A}(\vec{p})\right), \tag{2.69}$$

which maps non-transverse functions onto transverse ones, and then define a so-called generalised system of imprimitivity on the Borel sets of $\mathbb{R}^3$ by

$$F(S) = \lim_{n\to\infty}(PE(S))^n. \tag{2.70}$$

This differs from an ordinary system of imprimitivity in that projections on different Borel sets do not commute unless the sets are disjoint or one is entirely contained within the other.

The terminology that they introduce is that a particle is weakly localised in S if its state vector is an eigenvector of $F(S)$. For this to be the case the state must be a simultaneous eigenstate of $E(S)$ and P. Heuristically this may be seen as follows,

$$\begin{aligned} F(S) &= PE(S)\cdots PE(S) = PF(S) \\ F(S) &= F^*(S) = E(S)P\cdots E(S)P = E(S)F(S) \\ F(S)\phi = \phi &\implies P\phi = \phi,\, E(S)\phi = \phi, \end{aligned} \tag{2.71}$$

so that functions localised in S are both transverse and disappearing outside S. Amrein investigated whether non-trivial such ϕ could be found for the massless representations of the Poincaré group. He concludes that it is not possible even to weakly localise single-helicity particles of non-zero spin. However the photon may be weakly localised, as a consequence of it existing in two possible helicity states. He also investigated the energy density associated with a weakly-localised photon and found it to vanish asymptotically as the seventh power of distance as we move further from the region of weak localisation.

2.4 Utility of relativistic position operators

In the previous sections we have constructed, for massive particles, a position operator on the space of positive-energy, free, single-particle states which has the correct transformation properties under the Euclidean group. Further, for a given spin of particle this operator is unique to within equivalence. For massless particles we have been less successful. The generalised imprimitivity of Jauch and Piron constructs projection operators on the Borel sets of $\mathbb{R}^3$, which do have satisfactory transformation properties under the Euclidean group, but they do not commute, so it is not possible to construct a position operator from them. Both these constructions are only likely to be of further interest if we can put them to further use, so we now investigate the problems that may be encountered in their application.

2.4.1 Massive case

The most obvious use to which we may put a position operator is in a problem where the original Euclidean invariance of the system is lost, making different

points of space inequivalent, and so the idea of a position operator of some interest. The simplest such case is a problem with an external potential. If we follow the model of non-relativistic quantum theory, we take the classical Hamiltonian with its kinetic and potential-energy terms, and replace the classical momentum by the quantum operator $\vec{p}$ and the classical position $\vec{x}$ by the quantum operator $\vec{q}$. Thus, in an external potential problem, the potential-energy term $V(\vec{x})$ is replaced by $V(\vec{q})$.

The first problem we encounter is one which would arise even if we had developed non-relativistic quantum mechanics from the group-theoretical scheme. If we abandon Schrödinger's explicit prescription for the position and momentum operators, equation 2.3, the best we can do is to obtain the p, q pair to within equivalence which is somewhat weaker. Having one position operator q we could define a new position operator q' by

$$(q'\psi)(\vec{x}) = (\vec{x} - \vec{a})\psi(\vec{x}). \tag{2.72}$$

q' is still an acceptable position operator in the sense that $[q'_i, p_j] = i\delta_{ij}$ and is unitarily related to q by

$$q' = e^{-i\vec{p}\cdot\vec{a}} q\, e^{i\vec{p}\cdot\vec{a}}. \tag{2.73}$$

However, if we were to use q' for the motion of an electron in the field of a proton located at $\vec{x} = 0$ then the predicted electron density, using the Born interpretation of the wavefunction would be centred at $\vec{x} = \vec{a}$. So we discover that, certainly if we want a measurement interpretation of our quantum theory, uniqueness to within equivalence is not good enough and we have to make an explicit choice out of all possible position operators. In the familiar non-relativistic regime we make the 'obvious' choice, but it is not clear what our approach should be in the relativistic case.

We next consider the acceptability of single-particle theories in relativistic quantum mechanics. Looking at the action of the position operator for the Klein-Gordon equation given in equation 2.18 we notice that this only differs significantly from the naïve multiplication by $\vec{x}$ when ϕ varies significantly on a scale equal to that of the particle's Compton wavelength. To talk about the localisation of a relativistic particle on this scale is dubious, as measurement of its position at this kind of resolution in a Heisenberg microscope experiment, or similar, would be expected to bring effects characteristic of quantum field theory into play, pair production in particular. Although Feynman [5] successfully develops a successful single-particle picture of such phenomena, negative-energy states are required to describe antiparticles so this is outside our framework. In trying to solve an external-potential problem with potential variations on this kind of length scale we would come up against

the usual problems of the Klein-Gordon equation its single-particle setting; the difficulties of negative-energy states and the Klein paradox [14].

A relativistic single-particle theory with a better pedigree, though still suffering from similar problems, is the Dirac equation. We can, for example, quite successfully use the Dirac equation with an added external Coulomb potential to solve for the relativistic corrections to the hydrogen atom. To do this we add the minimal coupling term to the Dirac equation to get

$$(i\partial\!\!\!/ - eA\!\!\!/(\vec{x},t))\psi(x,t) = m\psi(x,t), \tag{2.74}$$

with $A = (Ze/4\pi\varepsilon_0 x, \vec{0})$, and look for stationary solutions. When Dirac solved this equation he did so using $A(\vec{x},t)$ and correctly predicted the first relativistic corrections to Schrödinger's result for the hydrogen atom. If our relativistic position operator is to have any use we would insert $A(\vec{q},t)$ with its non-local action into 2.74 instead of $A(\vec{x},t)$. However, in so far as this would lead to results differing from Dirac's, this alternative procedure yields incorrect predictions. The higher the charge on the nucleus, the more the two approaches would be expected to differ, as the closer the scale of the atom is to the Compton wavelength, so we would expect that the more relativistic the electron's motion, the worse the relativistic position operator will perform.

A final comment concerns the measurement of a particle's position when it is in a localised state. Newton and Wigner give the wavefunction of a particle localised at the origin in equation 2.16, this being an eigenstate of the relativistic position operator with zero eigenvalue. What kind of measuring apparatus could yield this result? In a quantum field theoretical approach to the Klein-Gordon particle, $\phi(\vec{x},t)$ and $\dot{\phi}(\vec{x},t)$ are the local observables of the theory. So if we wanted to determine the particle as being localised at the origin, we would need to register ϕ at all points in space to pick up the field disturbance associated with the localised particle. The requirement of an entirely delocalised measuring apparatus to determine a particle as localised does not seem entirely satisfactory, at least from a physicist's point of view.

2.4.2 Massless case

The Jauch and Piron [15] and Amrein [1] approach to localisation attempts to construct the next best thing to a position operator for a single-photon for which the single-particle equation would be

$$\Box A^\mu = 0. \tag{2.75}$$

However, apart from the rather uninteresting case of free photons, one can question the advisedness of a single-particle theory of the photon, one

of this particle's most characteristic features being the readiness with which it is created and destroyed in interaction. A quantum field theory rather than a single-particle theory for light is important if we want to describe both quantum states of light and everyday optical phenomena in the same framework.

It is, therefore, instructive to examine the localised states of Amrein in the perspective of the full theory of quantum electrodynamics. Their single-photon, weakly-localised state has the form,

$$|1_{loc}\rangle = \sum_{\lambda=\pm 1} \int \frac{\mathrm{d}^3\vec{q}}{(2\pi)^3 2q_0} f_\lambda(\vec{q}) a^\dagger_\lambda(\vec{q})|0\rangle, \tag{2.76}$$

where $a^\dagger_\lambda(\vec{q})$ creates a photon of momentum $\vec{q}$ and helicity λ. However, the coherent state

$$|c_{loc}\rangle = \exp\left(\sum_{\lambda=\pm 1} \int \frac{\mathrm{d}^3\vec{q}}{(2\pi)^3 2q_0} f_\lambda(\vec{q}) a^\dagger_\lambda(\vec{q})\right)|0\rangle \tag{2.77}$$

is also said to have the same localisation properties. As we have already pointed out, although these states are said to be weakly localised in some Borel set, they have an associated energy density which falls off as r^{-7} from the region of localisation.

From an experimental point of view a natural way to attempt to create a localised disturbance of the electromagnetic field is to start from the vacuum and allow a current confined to a finite region of space to act for a finite amount of time. Causality then prohibits this current from influencing anything at space-like separation from this current. To be more specific, we imagine an electromagnetic vacuum at $t = 0$ in a space devoid of charges or currents. We then create a classical (c-number) charge and current distribution by separating a positive and a negative charge at the origin, taking them on separate journeys which end up in the same place at $t = t_0$. This activity is illustrated in figure 2.1. At $t = t_0$ we again have a free field so that we are considering the same physical system as that considered by Amrein, there being no difference at times $t > t_0$ between a field that was once in a vacuum state and has been acted on by a current which has now vanished, and a field that has always been free, but has been prepared in some suitable state. It is clear that no form of measuring apparatus outside the light cone with vertex at $(0, \vec{0})$ can be influenced by the photons emitted by the current, and in this practical sense it would be fair to say that the photons at any given time $t > t_0$ are localised within the (dashed) sphere of radius ct surrounding the origin. In contrast, a hypothetical measuring device sensitive to energy density would respond to the r^{-7} tail associated with Amrein's localised state.

Of course, mathematically, the concept of localisation may be defined in a variety of ways, but our aim is to point out that the weakly-localised states of Amrein are certainly not the most localised in terms of their physical effects.

The case of single-photon localisation is more complex. We were able to create the localised coherent state with a classical current which can be made to appear and disappear at will, so that we may use the theory of the free electromagnetic field and make a connection with the work of Amrein. This we could also attempt in the single-photon case if we were provided with a universe consisting of a single photon appropriately set up. However our considerations above show that the properties of the source are important in determining the localisation of the effects produced by the photons it emits.

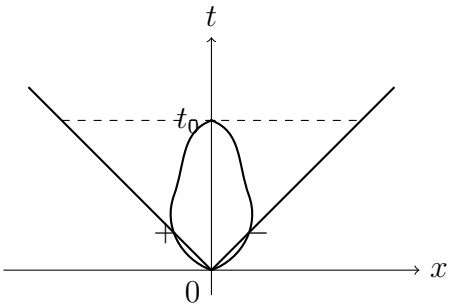

Figure 2.1: Generation of a localised disturbance of the free electromagnetic field

To create a single photon requires a quantum source of radiation which in turn will require the Hilbert space of the field to be extended to the Hilbert space of the source and field, so that we are no longer talking about the same physical system as Jauch, Piron and Amrein. However if, for example, we take our source of photons to be an atom at the origin with the whole system in its ground state at $t = 0$, and then stimulate the atom with a classical field to cause it to emit a quantum state of radiation, the same arguments as for the coherent state case show the impossibility of these photons influencing any system beyond a certain distance from the origin, and we would call these photons localised in a practical sense.

Chapter 3

Spontaneous parametric fluorescence

Spontaneous optical parametric fluorescence is a process in which single photons traversing non-linear optical materials with a second-order susceptibility are destroyed, and pairs of photons are produced in their place. The experimental arrangement in which we shall be interested in involves a crystal of such material in free space which is pumped by a source of plane-wave radiation of frequency ω_0, as in figure 3.1. We observe that, in any given direction, the intensity of the fluorescence is proportional to the intensity of the pumping radiation, and that the frequencies and wavevectors of the two outgoing photons satisfy energy and momentum conservation relations

$$\omega_0 = \omega_1 + \omega_2 \tag{3.1}$$

$$k_0^i = k^i(\omega_1) + k^i(\omega_2), \tag{3.2}$$

where the k^i's are wavevectors internal to the crystal which may be related to those externally observed by Snell's law. Arranging the system so that these equations may be satisfied involves some subtleties, which we will briefly deal with later, but this once achieved we find that these equations may be satisfied for a variety of k values so that there is a general broad-band emission of light. In any one direction, however, the frequency of the emitted photon is rather definite and its partner has a definite frequency and direction of travel.

There are also more subtle features of this fluorescence related to correlation properties of these pairs. Burnham and Weinberg [3] performed an experiment in which the signals from the photon detectors were fed into a correlator to measure the correlation in the photon arrival times. They found that, to within the time resolution of the experiment, the two members of

the pair arrive together, independently of the intensity of the pump beam (and so also of the arrival rate of the photons at the detectors). This may be understood qualitatively if we assume that the destruction of a pump photon and the creation of a pair in its place are simultaneous, and that each member of the pair propagates from their common point of origin to the detectors, part of the path in the crystal and part in free space. If the difference in propagation times were less than the time resolution of the experiment, the arrivals would be apparently simultaneous.

Although we can give the above prosaic explanation of the correlations observed in the Burnham and Weinberg experiment we might well wonder what an experiment with finer time resolution would be expected to yield. What, for example, would be the distribution of the differences in arrival times of the two photons? This reason alone is sufficient to motivate a theoretical investigation of this process and forms the principal subject of this chapter. Firstly however we discuss slightly more antique theories of second-order non-linear optical phenomena since they will form an important background to our later theoretical developments.

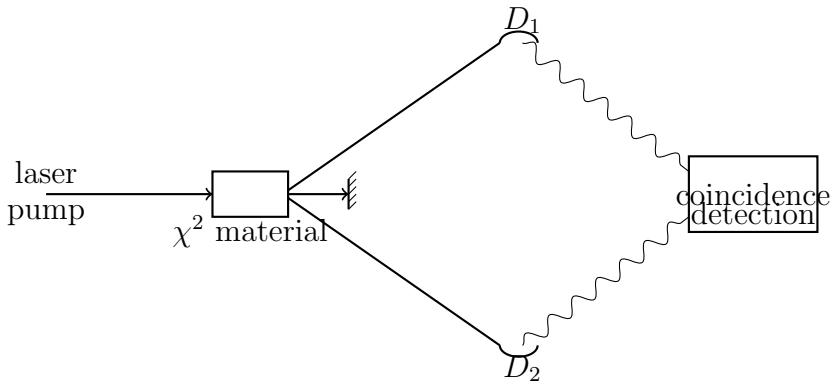

Figure 3.1: Experimental arrangement for generation of optical parametric fluorescence

3.1 Semiclassical and phenomenological theories of second-order non-linear optical phenomena

The field of non-linear optics was born in the experiment of Franken et al. [7] in which the red light of a ruby laser propagated through a quartz crystal

and was observed to generate ultra-violet radiation at twice the frequency of the red. Viewed in terms of photons this may be regarded as something like the reverse of parametric fluorescence where, instead of one photon being destroyed and two created, the fundamental process is the destruction of two red photons and the creation of one ultra-violet. In this experiment, however, it is not necessary to regard the electromagnetic field in quantum terms as the number of harmonic and pumping photons is large enough that we may treat the field as a classical quantity and just use quantum theory to analyse the properties of the material. This is the approach adopted by the early theorists and the one we now outline, following Shen [32].

The dynamics of the field are described by the classical Maxwell equations with the atomic currents in the non-linear material providing the source terms. In the dipole approximation, the interaction between atom and field is

$$\mathcal{H}_I = \vec{\mu} \cdot \vec{E}, \tag{3.3}$$

where $\vec{\mu}$ is the atomic dipole operator and $\vec{E}$ is the classical electric field. We solve for the motion of the atomic density matrix for which the equation of motion is

$$i\hbar \frac{\partial \rho}{\partial t} = [\mathcal{H}_{at} + \mathcal{H}_I, \rho] + \left(\frac{\partial \rho}{\partial t} \right)_{dis}. \tag{3.4}$$

The first term alone would gives the Hamiltonian evolution of the isolated atom, and the second term reflects the dissipation caused by environmental influence. For the diagonal elements of the density matrix this damping is often written

$$\frac{\partial}{\partial t}(\rho_{nn} - \rho^0_{nn}) = -\Gamma^1_n(\rho_{nn} - \rho^0_{nn}), \tag{3.5}$$

where ρ^0_{nn} is the equilibrium diagonal density matrix element at the nth atomic level and Γ^1_n is the reciprocal of the longitudinal decay time. For the off-diagonal elements we have

$$\frac{\partial}{\partial t}\rho_{nn'} = -\Gamma^2_{nn'}\rho_{nn'}, \tag{3.6}$$

where $\Gamma^2_{nn'}$ is the reciprocal of the transverse decay time for the coherence between levels n and n'. To obtain the non-linear polarisations and susceptibilities we proceed perturbatively in powers of the atom-field interaction and obtain

$$\rho = \rho^0 + \rho^1 + \rho^2 + \cdots \tag{3.7}$$

$$\langle \mu \rangle = \langle \mu^1 \rangle + \langle \mu^2 \rangle + \cdots, \tag{3.8}$$

where $\langle \mu^i \rangle = \mathrm{Tr}(\rho^i \mu)$ and the superscript indicates the order to which $\mathcal{H}_I$ appears. We assume that the medium consists of a concentration N of atoms and sum over the contribution that each atom makes. We then arrive at the following expressions for the first and second-order susceptibilities

$$\begin{aligned}\chi^1_{ij}(\omega) &= \frac{P^1_i(\omega)}{E_j(\omega)} \\ &= \frac{N}{\hbar}\sum_{gn}\left(\frac{\mu^i_{ng}\mu^j_{gn}}{\omega+\omega_{ng}+i\Gamma_{ng}} - \frac{\mu^j_{ng}\mu^i_{gn}}{\omega-\omega_{ng}+i\Gamma_{ng}}\right)\rho^0_{gg},\end{aligned} \tag{3.9}$$

$$\begin{aligned}\chi^2_{ijk}(\omega=\omega_1+\omega_2) &= \frac{P^2_i}{E_j(\omega_1)E_k(\omega_2)} \\ &= -\frac{N}{\hbar^2}\sum_{gnn'}\Bigg(\frac{\mu^i_{gn}\mu^j_{nn'}\mu^k_{n'g}}{(\omega-\omega_{ng}+i\Gamma_{ng})(\omega_2-\omega_{n'g}+i\Gamma_{n'g})} \\ &\quad + \frac{\mu^i_{gn}\mu^k_{nn'}\mu^j_{n'g}}{(\omega-\omega_{ng}+i\Gamma_{ng})(\omega_1-\omega_{n'g}+i\Gamma_{n'g})} \\ &\quad + \frac{\mu^k_{gn}\mu^j_{nn'}\mu^i_{n'g}}{(\omega+\omega_{ng}+i\Gamma_{ng})(\omega_2+\omega_{n'g}+i\Gamma_{n'g})} \\ &\quad + \frac{\mu^j_{gn}\mu^k_{nn'}\mu^i_{n'g}}{(\omega+\omega_{ng}+i\Gamma_{ng})(\omega_1+\omega_{n'g}+i\Gamma_{n'g})} \\ &\quad + \frac{\mu^j_{ng}\mu^i_{n'n}\mu^k_{gn'}}{\omega-\omega_{nn'}+i\Gamma_{nn'}}\left(\frac{1}{\omega_2+\omega_{n'g}+i\Gamma_{n'g}}+\frac{1}{\omega_1-\omega_{ng}+i\Gamma_{ng}}\right) \\ &\quad + \frac{\mu^k_{ng}\mu^i_{n'n}\mu^j_{gn'}}{\omega-\omega_{nn'}+i\Gamma_{nn'}}\left(\frac{1}{\omega_2-\omega_{n'g}+i\Gamma_{n'g}}+\frac{1}{\omega_1+\omega_{ng}+i\Gamma_{ng}}\right)\Bigg),\end{aligned} \tag{3.10}$$

These results apply to dilute non-linear media where we do not need to make a local field correction of the Clausius-Mosotti type. For our purposes, however, we note in particular that the nth order susceptibility arises in nth order perturbation theory. Having these non-linear susceptibilities immediately allows us to calculate the strength of such processes as harmonic generation and three-wave mixing but we do not have the means to describe parametric fluorescence as we have not introduced a quantum picture of the field which we certainly would need to describe the appearance of correlated photon pairs. Early attempts to describe the effect did try to use this framework, by introducing fictitious zero-point fluctuations into the classical field, the fluorescence being regarded as produced by parametric amplification or

by mixing of these fluctuations with the pump mode, but clearly such approaches are too contrived to be entirely satisfactory.

Kleinman [16] gave a completely quantum treatment of the fluorescence. To avoid the complexity of dealing with quantum matter and quantum radiation he adopts a quantised version of the classical phenomenological approach in which the polarisation of the medium is expanded as a power series in the electric field,

$$\vec{P} = \chi^1 \cdot \vec{E} + \chi^2 : \vec{E}\vec{E}. \tag{3.11}$$

Together with this polarisation there is an associated contribution to the energy of the system which is calculated as follows. In the unperturbed system $\vec{P} = \vec{0}$, so we start with $\vec{P} = \vec{0}$ and $\vec{E} = \vec{0}$, and build up to a given value of $\vec{E}$, changing $\vec{P}$ in accordance with equation 3.11 and finally reduce $\vec{P}$ to zero at constant $\vec{E}$. The work done by the field in this cycle is

$$H = -\int \mathrm{d}^3\vec{r} \int_0^{\vec{E}} \vec{P} \cdot \mathrm{d}\vec{E}, \tag{3.12}$$

and so the second-order susceptibility makes a contribution

$$H' = -\frac{1}{3} \int \mathrm{d}^3\vec{r}\, \vec{E} \cdot \chi : \vec{E}\vec{E} \tag{3.13}$$

to the energy of the system.

To quantise this theory we first quantise the free electromagnetic field in the usual way and then introduce an interaction Hamiltonian which is simply H' but with the classical electric field replaced with the quantum electric field. Of course this procedure cannot be justified on fundamental grounds (which would require a development of a full matter and radiation theory from a Lagrangian starting point) but this approach at least will give results consistent with the classical theory when a classical picture is valid, and is also capable of describing the quantised excitations of the field that occur in such effects as parametric fluorescence.

Kleinman uses this approach and Fermi's golden rule in first-order perturbation theory to calculate the transition rate of the system from states in which the pump mode is populated to states in which the pump mode is depopulated by one, with two photons in other modes. This occurs because the interaction Hamiltonian contains terms with one annihilation operator and two creation operators. In this way we may calculate the intensity and spectrum of the fluorescence in any direction and the correlation between the direction of propagation of the two photons. This theory will be further developed in chapter 5 as we can also use this approach in an analysis of fourth-order interference.

The golden-rule analysis is not, however, capable of describing the correlations existing in the Burnham and Weinberg experiment, since here we are interested in events (the clicks of photodetectors) at given points in space and at given times, where the golden-rule procedure merely gives scattering rates into delocalised modes. The theory of parametric fluorescence due to Mollow [21] is an attempt to describe these correlations in photon arrival times in a framework in which both matter and radiation are treated quantum-mechanically, and which we now describe in detail.

3.2 Photon correlations in spontaneous parametric fluorescence

Mollow's theory aims to provide an expression for the probability of obtaining photocounts at spacetime points x_1 and x_2 in the experimental arrangement of figure 3.1. This may be obtained from the four-point function

$$G^{(2,2)}(x_1', x_2'; x_2, x_1) \equiv \langle A^-(x_1')A^-(x_2')A^+(x_2)A^+(x_1)\rangle. \tag{3.14}$$

We shall be interested in the parametric process at low pump powers, corresponding to a small chance of a pair being present in the field. In this case we may write the state of the field as

$$|t\rangle \approx |0\rangle + c|2\rangle, \tag{3.15}$$

where $|c|^2 \ll 1$. This enables us to make a considerable simplification to the analysis because we can approximately factorise $G^{(2,2)}$:

$$G^{(2,2)}(x_1', x_2'; x_2, x_1) \approx G^{(2,0)}(x_1', x_2')G^{(0,2)}(x_2, x_1), \tag{3.16}$$

where

$$G^{(2,0)}(x_1', x_2') = \langle A^-(x_1')A^-(x_2')\rangle \tag{3.17}$$

$$G^{(0,2)}(x_2, x_1) = \langle A^+(x_2)A^+(x_1)\rangle \tag{3.18}$$

3.2.1 The role of atomic currents in photon pair emission

We work in the Heisenberg picture and assume that in the distant past the field is in the vacuum state. We also choose the Coulomb gauge so that

$\nabla \cdot \vec{A} = 0$ and the expressions for the retarded and advanced propagators are

$$D_C(x) = \left(1 - \frac{\nabla\nabla}{\nabla^2}\right) \frac{\delta(r - ct)}{4\pi r} \tag{3.19}$$

$$D_A(x) = \left(1 - \frac{\nabla\nabla}{\nabla^2}\right) \frac{\delta(r + ct)}{4\pi r} \tag{3.20}$$

and we shall also need the field commutator

$$\frac{i}{\hbar c^2}[A_I(x), A_I(x')] = D_C(x - x') - D_A(x - x') = D(x - x'). \tag{3.21}$$

The solution to the wave equation for the Heisenberg field operator $A(x)$ is

$$A(x) = A_I(x) + \int \mathrm{d}^4\bar{x}\, D_C(x - \bar{x}) J(\bar{x}), \tag{3.22}$$

where $J(\bar{x})$ is the Heisenberg electric current operator. From this $G^{(0,2)}(x_2, x_1)$ can immediately be written down

$$\begin{aligned} G^{(0,2)}(x_2, x_1) = &\int \mathrm{d}^4\bar{x}_2 \int \mathrm{d}^4\bar{x}_1\, D_C^+(x_2 - \bar{x}_2) D_C^+(x_1 - \bar{x}_1) \langle J(\bar{x}_2) J(\bar{x}_1)\rangle \\ &+ \int \mathrm{d}^4\bar{x}_1\, D_C^+(x_1 - \bar{x}_1) \langle [A_I^+(x_2), J(\bar{x}_1)]\rangle. \end{aligned} \tag{3.23}$$

In the second term we have made use of the fact that, as the expectation values are taken in the vacuum state,

$$\langle A_I^+(x_2) J(\bar{x}_1)\rangle = \langle [A_I^+(x_2), J(\bar{x}_1)]\rangle, \tag{3.24}$$

the A_I^+ annihilating the vacuum ket. We now wish to eliminate the field operators from the right hand side of equation 3.23. Starting from the observation that the full Heisenberg operators $A(x_2)$ and $J(\bar{x}_1)$ commute at equal times, by making use of equation 3.22 we get

$$[A_I(x_2), J(\bar{x}_1)] = -\int \mathrm{d}^4\bar{x}_2\, D_C(x_2 - \bar{x}_2) \langle [J(\bar{x}_2), J(\bar{x}_1)]\rangle, \tag{3.25}$$

and so at $t_2 = \bar{t}_1$ we may write

$$[A_I(x_2), J(\bar{x}_1)] = -\int \mathrm{d}^4\bar{x}_2\, \theta(\bar{t}_1 - \bar{t}_2) D(x_2 - \bar{x}_2) \langle [J(\bar{x}_2), J(\bar{x}_1)]\rangle. \tag{3.26}$$

However this equation is valid not only at $t_2 = \bar{t}_1$ but also at all other times, which is a consequence of both sides obeying the wave equation in the x_2 variable, equality at $t_2 = \bar{t}_1$ and also the equality of the time derivative

with respect to t_2 at $t_2 = \bar{t}_1$. By taking the positive-frequency part of 3.26 and substituting into equation 3.23 we obtain

$$G^{(0,2)}(x_2, x_1) = \int \mathrm{d}^4\bar{x}_2 \int \mathrm{d}^4\bar{x}_1 \left\{ D_C^+(x_2 - \bar{x}_2) D_C^+(x_1 - \bar{x}_1) \langle (J(\bar{x}_2) J(\bar{x}_1))_T \rangle \right. \\ \left. + \theta(\bar{t}_1 - \bar{t}_2) D_A^+(x_2 - \bar{x}_2) D_C^+(x_1 - \bar{x}_1) \right\} \langle [J(\bar{x}_2), J(\bar{x}_1)] \rangle. \tag{3.27}$$

Sufficiently far from the current distribution the term containing the advanced propagator makes a vanishing contribution, and so omitting the final term and transferring the positive-frequency signatures from the propagators onto the current, we obtain an expression which relates the dual photocount amplitude to the currents inside the material that the pumping laser excites.

$$G^{(0,2)}(x_2, x_1) = \int \mathrm{d}^4\bar{x}_2 \int \mathrm{d}^4\bar{x}_1 \left\{ D_C(x_2 - \bar{x}_2) D_C(x_1 - \bar{x}_1) \langle (J(\bar{x}_2) J(\bar{x}_1))_T^{++} \rangle \right\}. \tag{3.28}$$

3.2.2 The time-ordered current-current correlation function

The current correlation function 3.28 is first evaluated for the case of a single atom, with the intention of summing over atoms to obtain the result for an extended medium. For a single atom at the point $\vec{r}$ the correlation function, in the dipole approximation, is expressed as

$$\langle (J(\bar{x}_2) J(\bar{x}_1))_T^{++} \rangle = -\delta^3(\bar{r}_2 - \vec{r}) \delta^3(\bar{r}_1 - \vec{r}) \\ \times \frac{1}{2\pi} \int \mathrm{d}\omega_2 \int \mathrm{d}\omega_1 \, \omega_2 \omega_1 \, \tilde{g}_T^{(0,2)}(\omega_2, \omega_1) e^{-i\omega_2 \bar{t}_2 - i\omega_1 \bar{t}_1}, \tag{3.29}$$

where $\tilde{g}_T^{(0,2)}(\omega_2, \omega_1)$ is the Fourier transform of the time-ordered dipole-dipole correlation function

$$\tilde{g}_T^{(0,2)}(\omega_2, \omega_1) = \theta(\omega_2)\theta(\omega_1) \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}t_2 \int_{-\infty}^{t_2} \mathrm{d}t_1 \, e^{-i\omega_2 t_2 - i\omega_1 t_1} \langle \vec{\mu}(t_2) \vec{\mu}(t_1) \rangle \\ + (2 \leftrightarrow 1). \tag{3.30}$$

It is next assumed that the atom is driven by a prescribed classical field $E(t)$, with an electric dipole interaction

$$\mathcal{H}_I = \vec{\mu} \cdot \vec{E}. \tag{3.31}$$

The dipole operator may be expanded in a perturbation series

$$\vec{\mu}(t) = \vec{\mu}_I(t) + \frac{i}{\hbar}\int_{-\infty}^{t} \mathrm{d}t' \left[\mu_I(t), \mu_I(t')E(t')\right] + \cdots , \tag{3.32}$$

where $\mu_I(t)$ is the interaction picture dipole operator

$$\mu_I(t) = \sum_{jk} \mu_{jk} e^{i\omega_{jk}t} |j\rangle\langle k| \tag{3.33}$$

and

$$\hbar\omega_{jk} = E_j - E_k \tag{3.34}$$

We are particularly interested in the case where the atom is driven by a monochromatic field

$$E(t) = E_0 e^{i\omega_0 t} + \text{c.c.}. \tag{3.35}$$

If we retain only the first term in the perturbation series $\tilde{g}^{(0,2)}(\omega_2, \omega_1)$ is non-vanishing only for $\omega_1 + \omega_2 = \omega_0$ and so

$$\tilde{g}^{(0,2)}(\omega_2, \omega_1) = -i\hbar\phi(-\omega_2, -\omega_1, \omega_0)E_0 \tag{3.36}$$

where

$$\begin{aligned}\phi^{\lambda_2\lambda_1\lambda_0}(\omega_2, \omega_1, \omega_0) = &-\frac{1}{\hbar^2}\delta(\omega_2 + \omega_1 + \omega_0) \\ &\times \sum_p \sum_{j_2 j_1} \frac{\mu_{gj_2}^{\lambda_{p(2)}} \mu_{j_2 j_1}^{\lambda_{p(1)}} \mu_{j_1 g}^{\lambda_{p(0)}}}{(\omega_{p(2)} + \omega_{j_2 g})(\omega_{p(0)} - \omega_{j_1 g})}.\end{aligned} \tag{3.37}$$

The superscripts are vector indices, g is the ground state of the atom and p is a permutation of the indices $0, 1$ and 2. All permutations are summed over, giving a function symmetric in all three arguments.

3.2.3 Linear response of the fluorescent medium

Having evaluated the current-current correlation function which will give the non-linear optical effect of a single atom, we must sum over the atoms which compose the material. However we must also take into account the linear interaction of radiation and matter which gives rise to the refractive index, and because this interaction is strong we must deal with it non-perturbatively. In an infinite dielectric medium we would accomplish this by making the substitution

$$\tilde{G}(\vec{x} - \vec{x}', \omega) = -\frac{1}{4\pi|\vec{x} - \vec{x}'|} e^{ik(\omega)|\vec{x} - \vec{x}'|}, \tag{3.38}$$

where $\tilde{G}$ is the propagator for the Helmholtz wave equation and $k(\omega)$ is the wavenumber associated with frequency ω. However with a dielectric material filling a finite region of space we require a new propagator $\tilde{G}'(\vec{x}, \vec{x}', \omega)$, now a function of two spatial arguments, which reflects the lost translational invariance of the problem. The form of this propagator depends on the location of $\vec{x}$ and $\vec{x}'$ relative to each other and relative to the dielectric material. For example, with $\vec{x}$ and $\vec{x}'$ both well inside the medium $\tilde{G}'(\vec{x}, \vec{x}', \omega)$ should behave like the translation invariant $\tilde{G}(\vec{x} - \vec{x}', \omega)$, but with $\vec{x}$ and $\vec{x}'$ outside the medium and close together the behaviour should be that of the free-space propagator. A precise evaluation of $\tilde{G}'(\vec{x}, \vec{x}', \omega)$ is not practicable, but we suggest an *ansatz* that is appropriate when $\vec{x}$ is inside a plane-faced medium many wavelengths large and $\vec{x}'$ is in its Fraunhofer region:

$$\tilde{G}'(\vec{x}, \vec{x}', \omega) = -\frac{T}{4\pi x'} e^{i\phi(\vec{x}, \vec{x}', \omega)}, \tag{3.39}$$

where T is the amplitude transmission coefficient of the material's surface and $\phi(\vec{x}, \vec{x}', \omega)$ is the optical phase accumulated along the ray from $\vec{x}$ to $\vec{x}'$ which is refracted according to Snell's law at the surface. We illustrate this in figure 3.2. For the moment we choose our origin of coordinates, O, on the surface through which the ray emerges. The optical phase from $\vec{x}$ to $\vec{x}'$ is given by

$$\begin{aligned} \phi(\vec{x}, \vec{x}', \omega) &= k_e(OS - OR) + k_i PQ \\ &= k_e(x' - x_\perp \tan\theta_i \sin\theta_e) + k_i \frac{x_\parallel}{\cos\theta_i}. \end{aligned} \tag{3.40}$$

Using Snell's law

$$k_i \sin\theta_i = k_e \sin\theta_e, \tag{3.41}$$

we obtain

$$\begin{aligned} \phi(\vec{x}, \vec{x}', \omega) &= k_e x' + (x_\perp \cos\theta_i - x_\parallel \sin\theta_i) \\ &= k_e x' - \hat{k}_i \cdot \vec{x}. \end{aligned} \tag{3.42}$$

If we now shift the origin to a point O' such that $\vec{OO'} = \vec{a}$ we then obtain

$$\phi(\vec{x}, \vec{x}', \omega) = k_e(x' - \hat{a} \cdot \vec{x}') - \hat{k}_i \cdot (\vec{x} - \vec{a}), \tag{3.43}$$

where $\vec{x}$ and $\vec{x}'$ are now expressed relative to the new origin. Mollow's choice of propagator corresponds to $\vec{a} = 0$ which is satisfactory so long as all the rays exit the medium from the same face, so that a single $\vec{a}$ will suffice for all rays, leading to an unimportant overall phase factor.

To obtain the tensor propagator that is required for the vector wave equation we simply multiply the scalar propagator just obtained by the appropriate projection operator:

$$\tilde{D}(\vec{x},\vec{x}',\omega) = \left(\delta_{ij} - \frac{x_i' x_j'}{x'^2}\right)\tilde{G}(\vec{x},\vec{x}',\omega). \tag{3.44}$$

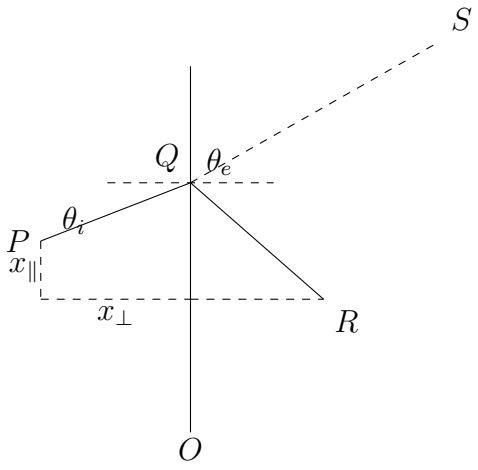

Figure 3.2: Geometry for calculation of optical phase between points inside and outside a refractive medium

3.2.4 Spacetime correlations in spontaneous photon pair production

With the modification to the photon propagator just proposed we are ready to evaluate the correlations in photon pairs emitted from a medium composed of many atoms.

Let us assume that the incident wave propagates in the z-direction, with amplitude in the transverse directions specified by the function $f(x,y)$

$$\vec{E}(\vec{r},t) = \vec{E}_0 f(x,y) e^{ik_0 z - i\omega_0 t}. \tag{3.45}$$

For a constant density, N, of atoms in the target, at points far away from the medium $G^{(0,2)}(x_2,x_1)$ may be approximated by

$$\begin{aligned} G^{(0,2)}(x_2,x_1) = i\hbar \frac{N}{(4\pi c)^2}\frac{P_1 P_2}{r_1 r_2}\frac{1}{2\pi}\int \mathrm{d}\omega_2 \int \mathrm{d}\omega_1\, e^{-i\omega_2 \bar{t}_2 - i\omega_1 \bar{t}_1} \\ \times T_2 T_1 \omega_2 \omega_1 E_0 \phi(-\omega_2,-\omega_1,\omega_0)\zeta(\vec{k}_2 + \vec{k}_1 - \vec{k}_0), \end{aligned} \tag{3.46}$$

where $P = 1 - \hat{r}\hat{r}/r^2$ for $\vec{r} = \vec{r}_{1,2}$, $\bar{t}_{1,2} = t_{1,2} - r_{1,2}/c$ and ϕ is the non-linear coefficient. $\vec{k}_1$ and $\vec{k}_2$ are the wavevectors in the target associated (through Snell's law) with the external observation directions $\hat{r}_1 = \vec{r}_1/r_1$ and $\hat{r}_2 = \vec{r}_2/r_2$. T_1 and T_2 are proportional to the corresponding amplitude transmission coefficients and $\zeta(\vec{k})$ is defined by the relation

$$\zeta(\vec{k}) = \int \mathrm{d}^3\vec{r}\, e^{-i\vec{k}\cdot\vec{r}} f(x,y)\eta(\vec{r}), \tag{3.47}$$

where $\eta(\vec{r})$ is unity inside the medium and zero outside.

We shall be particularly interested in the case where the target is a slab of material with parallel faces, placed perpendicular to the beam and larger than the beam cross section. In this case we may put

$$\eta(\vec{r}) = \theta(l/2 - |z|), \tag{3.48}$$

where l is the thickness of the target.

A cursory examination of equation 3.47 shows that, for targets in which the illuminated patch is many wavelengths large there is a strong preference for emission in which momentum is conserved. Also from the presence of the frequency delta function in equation 3.37 the sum of the energies of the two fluorescent photons is rigorously equal to that of a pump photon. Wave vectors internal to the target for which $\vec{k}_2 + \vec{k}_1 = \vec{k}_0$ and $\omega_2 + \omega_1 = \omega_0$ are said to lie on the phase-matching surface. The achievement of phase-matching is dependent on the details of the material's linear dispersion (in free space it could only be achieved for forward scattering of the pair) and we have to exploit the anisotropy of the medium to combat the normal increase of refractive index with frequency which would prevent phase-matching being attained, a simplified account of which we give in appendix B. When we observe the emission of pairs from the medium we assume that one observation direction $\hat{r}$ is such that the frequency ω_1^* is phase-matched. Then the complementary photon's frequency and wavevector are determined by

$$\omega_2^* + \omega_1^* = \omega_0 \tag{3.49}$$

$$\vec{k}_2^* + \vec{k}_1^* = \vec{k}_0. \tag{3.50}$$

When the illuminated area is infinite there is emission only in the directions of phase-matching: effectively in equation 3.47, $\zeta(\vec{k})$ becomes proportional to $\delta(\vec{k})$. This results in the emission at any angle being monochromatic. For finite sizes of illuminated region the form-factor $\zeta(\vec{k})$ is no longer vanishing for $\vec{k} \neq \vec{0}$ which allows a degree of uncertainty in the direction in which the complementary photon in emitted. This also implies that the radiation

emitted in any one direction is no longer monochromatic and, we shall see that, associated with this finite spectral width there is now correlation in the arrival times of the two photons.

If we are looking in a fixed direction then the internal wavevector possessed by one member of a pair is a function of the frequency of that photon. If the form-factor is such that the light emitted in any one direction is narrowband then we may make a linear expansion of the wavevector in terms of the frequency about the optimally phase-matched values:

$$\vec{k}_1 = \vec{k}_1^* + (\omega_1 - \omega_1^*)\frac{\mathrm{d}\vec{k}_1}{\mathrm{d}\omega} \tag{3.51}$$

$$\vec{k}_2 = \vec{k}_2^* + (\omega_2 - \omega_2^*)\frac{\mathrm{d}\vec{k}_2}{\mathrm{d}\omega} + \vec{B}. \tag{3.52}$$

$\vec{B}$ is the change in $\vec{k}_2$ that results when we shift our observation direction for photon 2 away from the optimal and is given by

$$\vec{B} = k_2 \left(1 - \frac{\hat{k}\cdot\vec{v}_2'}{v_2'}\right)\delta\hat{k}_2, \tag{3.53}$$

where $\vec{v}$ is the group velocity and $v' = \vec{v}\cdot\hat{k}$. For the case of an isotropic medium $\vec{B}$ is simply

$$\vec{B} = k_2\delta\hat{k}_2. \tag{3.54}$$

It is also useful to define

$$\vec{A} = \frac{\mathrm{d}\vec{k}_1}{\mathrm{d}\omega_1} - \frac{\mathrm{d}\vec{k}_2}{\mathrm{d}\omega_2}. \tag{3.55}$$

The expression for $G^{(0,2)}(x_2, x_1)$ now becomes

$$G^{(0,2)}(x_2, x_1) = \frac{i\hbar\omega_2^*\omega_1^*\chi(\omega_2^*, \omega_1^*)\cdot\bar{E}_0}{(4\pi c)^2 r_1 r_2} e^{-i\omega_2^*\bar{t}_2 - i\omega_1^*\bar{t}_1}\psi(\bar{t}_2 - \bar{t}_1; \hat{r}_2, \hat{r}_1), \tag{3.56}$$

where χ is defined by

$$\chi(\omega_2, \omega_1)\delta(\omega_2 + \omega_1 - \omega_0) \equiv NP_2P_1T_2T_1\phi(-\omega_2, -\omega_1, \omega_0)\cdot\bar{E}_0/E_0, \tag{3.57}$$

and ψ is defined by

$$\psi(\tau; \hat{r}_2, \hat{r}_1) = \int \mathrm{d}^3\vec{r}\, f(x, y)\eta(z)e^{-i\vec{B}\cdot\vec{r}}\delta(\tau - \vec{A}\cdot\vec{r}), \tag{3.58}$$

The coincidence counting rate at two points with effective time delay

$$\tau = t_2 - t_1 - (r_2 - r_1)/c \tag{3.59}$$

is thus proportional to $|\psi(\tau; \hat{r}_2, \hat{r}_1)|^2$. We assume that the pumping beam has a Gaussian profile:

$$f(x, y) = \exp\left[-\frac{x^2 + y^2}{2R^2}\right], \tag{3.60}$$

and we may now evaluate ψ in two particular cases of interest, $R\sqrt{A_x^2 + A_y^2} \ll A_z l$, for a thin beam propagating through a long stretch of the non-linear medium and $R\sqrt{A_x^2 + A_y^2} \gg A_z l$ for a thin slice of non-linear medium with broad transverse illumination. In the former case we obtain:

$$\begin{aligned}\psi(\tau; \hat{r}_2, \hat{r}_1) = &\frac{2\pi}{A_z}\eta(\tau/A_z) \\ &\times e^{i\tau\delta\theta_{2i}\mu} \exp\left[-\frac{1}{2}R^2k_2^2\left(\gamma^2(\delta\theta_{2i})^2 + \sin^2\theta_{2i}(\delta\phi_{2i})^2\right)\right],\end{aligned} \tag{3.61}$$

where $\mu = k_2 \sin\theta_{2i}/A_z$ and $\gamma = \vec{A}\cdot\hat{k}_2/A_z$. $\delta\theta_{2i}$ and $\delta\phi_{2i}$ are the internal polar and azimuthal angular shifts associated with the move away from optimum in the second observation direction in a coordinate system with polar axis defined by the pump and $\sin\phi_{2i} = 0$ defined by the plane of the pump and first observation direction. In the latter case:

$$\begin{aligned}\psi(\tau; \hat{r}_2, \hat{r}_1) = &\frac{\sqrt{2\pi}Rl}{A_z}e^{i\tau\delta\theta_{2i}\mu'} \\ &\times \exp\left[-\frac{1}{2}\frac{\tau^2}{R^2A_z^2} - \frac{1}{2}R^2k_2^2\sin^2\theta_{2i}(\delta\phi_{2i})^2\right]\operatorname{sinc}\left[\frac{1}{2}\gamma' k_2 l\delta\theta_{2i}\right].\end{aligned} \tag{3.62}$$

where $\mu' = k_2\cos\theta_{2i}/A_z$ and $\gamma' = \vec{A}\cdot\hat{k}_2/A_z$.

We look first of all at the behaviour of these as a function of τ. In the first case the dual photocount amplitude is uniform for

$$|\tau| < \frac{1}{2}l\left|\frac{\cos\theta_{1i}}{v_1'} - \frac{\cos\theta_{2i}}{v_2'}\right|, \tag{3.63}$$

and vanishing outside this range. This formula has a simple interpretation in terms of the differences of propagation times of the individual members of photon pairs from the point at which they simultaneously originate out to the detectors, and how this difference varies over the illuminated region of

the non-linear medium. Referring to figure 3.3, it is clear that a photon pair produced at O arrives with time difference τ_1 given by

$$c\tau_1 = r_1 - r_2. \tag{3.64}$$

A pair produced at O' arrives at the detectors with a time difference τ_2 given by

$$c\tau_2 = (r_1 + n_1 l \cos\theta_{1i}) - (r_2 + n_2 l \cos\theta'_{2i}). \tag{3.65}$$

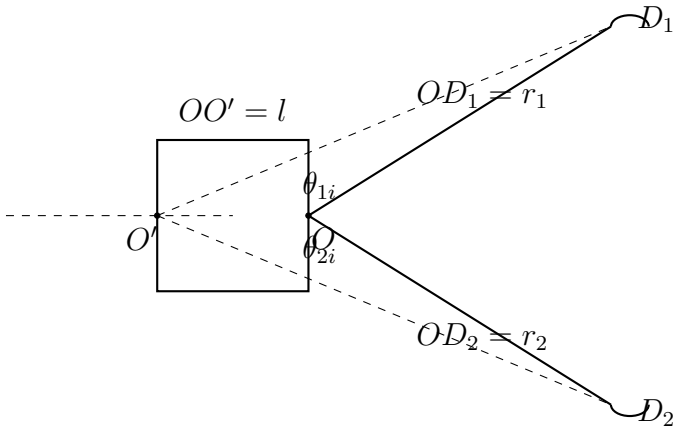

Figure 3.3: Propagation delays from different parts of the illuminated patch of non-linear medium

This range of time differences is the content of equation 3.63. Similarly the range of τ in equation 3.62 may be interpreted as a dispersion in arrival times due to the transverse extent of the beam, its Gaussian distribution a reflection of the Gaussian illumination profile. More generally, if a photon pair produced at the origin arrives at the detectors with $\tau = \tau_0$, then these simple propagation delays lead to a pair produced at a point $\vec{r}$ arriving at the detectors with $\tau = \tau_0 + \vec{A} \cdot \vec{r}$. In this light, equation 3.58 which gives the amplitude for two-photon detection with time delay τ, may be seen as a coherent sum of pair-production amplitudes, but, because of the delta function, only those parts of the medium from which propagation delays result in the specific time delay τ make a contribution.

We now turn to how the probability of joint photodetection depends upon the positioning of the detectors. In the thick-medium case the fall-off in the two-photon detection amplitude as the direction in which we look for the second photon moves away from the optimum is Gaussian with a width inversely proportional to the beam radius in both the polar and azimuthal directions. When the beam radius is large compared to the target thickness

the azimuthal fall-off is again Gaussian with width proportional to $1/R$ while the polar fall-off is as a sinc function with width proportional to $1/l$.

3.3 Power-law tails in two-photon correlations

The analysis of Mollow is appropriate when the form-factor effectively limits the radiation emitted in any one direction to a bandwidth much smaller than that over which the medium's linear and non-linear susceptibilities undergo appreciable variation. Pike and Sarkar [24] investigated the possibility of broad-band effects in the emission for the case of an ideal non-linear medium devoid of linear dispersion and dispersion of the non-linear susceptibility, such as might be obtained if the atomic resonances in the medium were at much higher frequencies than the pumping frequency. We outline a slight variant of their model. Instead of having a non-linear material and embedding it in space filled with a medium of matched refractive index, so that refraction at the medium's surface can be neglected, we consider simply a region of non-linearity in free space with phase-matching facilitated by the pump carrying anomalous momentum (ie. not consistent with the free-space dispersion relation $\omega = ck$). However the conclusions we draw differ only in trivial detail.

We look first at the case of a single atom at the origin in free space ($n = 1$) driven by an electric field of frequency $\omega = \omega_0$. Using Mollow's expression for the single-atom current-current correlation function, equation 3.29 we immediately obtain

$$G^{0,2}(x_2, x_1) \propto \frac{P_1 P_2}{r_1 r_2} \int \mathrm{d}\omega_2 \int \mathrm{d}\omega_1 \, \exp[-i\omega_2(t_2 - r_2/c) - i\omega_1(t_1 - r_1/c)] \times \omega_2\omega_1 \, \theta(\omega_2)\theta(\omega_1)\chi(\omega_2, \omega_1)E_0\delta(\omega_0 - \omega_2 - \omega_1). \tag{3.66}$$

Taking $\chi(\omega_2, \omega_1)$ to be constant over the frequency range zero to ω_0,

$$G^{0,2}(x_2, x_1) \propto \frac{2\omega_0}{(\bar{t}_2 - \bar{t}_1)^2} \exp\left[-i\frac{\omega_0}{2}(\bar{t}_1 + \bar{t}_2)\right] \times \left[\cos\frac{\omega_0}{2}(\bar{t}_2 - \bar{t}_1) - \mathrm{sinc}\frac{\omega_0}{2}(\bar{t}_2 - \bar{t}_1)\right], \tag{3.67}$$

where

$$\bar{t}_{1,2} = t_{1,2} - r_{1,2}/c. \tag{3.68}$$

Thus the asymptotic dependence of the two-photocount amplitude is as the inverse second power of the reduced time difference between the photodetections. Contrast this with our interpretation of Mollow's theory in terms of

propagation delays from different parts of the non-linear medium which with a single-atom source would lead to no dispersion in the photodetection time differences. In this new picture we can no longer imagine the creation of a pair at a given point at a given time, but we would have to imagine an uncertainty in the time difference with which the two photons leave the atom which leads to this inverse second power behaviour in the two-photocount amplitude.

Moving to the case of a medium pumped by a Gaussian beam we take the product of the transverse beam profile and the medium density to behave as

$$\vec{E}(\vec{r})\rho(\vec{r}) \propto \exp\left(-\frac{x^2+y^2}{\xi_1^2}\right)\exp\left(-\frac{z^2}{\xi_3^2}\right), \tag{3.69}$$

the z-dependence being the least realistic part since it would require the material density to have this Gaussian behaviour. This leads to

$$\begin{aligned} G^{0,2}(x_2,x_1) \propto \int_0^{\omega_0} \mathrm{d}\omega_2 \int_0^{\omega_0} \mathrm{d}\omega_1 \int \mathrm{d}^3\vec{r}\, \exp\left[-\frac{x^2+y^2}{\xi_1^2}-\frac{z^2}{\xi_3^2}\right] \\ \times \exp\left[-i\omega_2(t_2-r_2/c)-i\omega_1(t_1-r_1/c)\right] \\ \times \exp\left[iz(k_0-k_2z_2\cos\theta_2-k_1z_1\cos\theta_1)\right] \\ \times \exp\left[-ix(k_2x_2\sin\theta_2-k_1x_1\sin\theta_1)\right] F(\omega_2,\omega_1,\vec{r}_2,\vec{r}_1), \end{aligned} \tag{3.70}$$

where $k=\omega/c$, the observation points $\vec{r}_2$ and $\vec{r}_1$ are in the x–z plane making angles θ_2 and θ_1 with the pump direction, and where

$$F(\omega_2,\omega_1,\vec{r}_2,\vec{r}_1) = \frac{P_2P_1}{r_2r_1}\chi^{jmn}(\omega_0,\omega_1,\omega_2)E\omega_1\omega_2\delta(\omega_0-\omega_2-\omega_1). \tag{3.71}$$

For $\xi_\perp, \xi_3 \gg 1/k_0$ the values of ω which make the most significant contribution to the integral, for given observation points $\vec{r}_1$ and $\vec{r}_2$ are those close to the phase-matched frequencies. Mollow's approach is to take the ω_1, ω_2 and non-linear susceptibility factors out of the integral, giving them the phase-matched values and extending the limits of integration to plus and minus infinity, as the integrand is peaked at the phase-matched configuration. Pike and Sarkar consider the result with the original limits intact.

After performing the integration over the medium we obtain

$$\begin{aligned} G^{0,2}(x_2,x_1) \propto \int_0^{\omega_0} \mathrm{d}\omega_2 \int_0^{\omega_0} \mathrm{d}\omega_1\, \omega_2\omega_1 \exp\left[-i\omega_2\bar{t}_2-i\omega_1\bar{t}_1\right]\delta(\omega_0-\omega_1-\omega_2) \\ \times \exp\left[-\frac{\xi_3^2}{4}(k_0-k_2z_2\cos\theta_2-k_1z_1\cos\theta_1)^2\right] \\ \times \exp\left[-\frac{\xi_\perp^2}{4}(k_2x_2\sin\theta_2-k_1x_1\sin\theta_1)^2\right]. \end{aligned} \tag{3.72}$$

The ω_1 integration is immediate with the delta function, and using the convolution theorem the remaining integral may be written

$$G^{0,2}(x_2, x_1) \propto \exp(-i\omega_0 \bar{t}_1) \int_{-\infty}^{\infty} \mathrm{d}\tau \, I_1(\bar{t}_2 - \bar{t}_1 - \tau) I_2(\tau), \tag{3.73}$$

where

$$I_1(\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega \, \theta(\omega)\theta(\omega_0 - \omega)\omega(\omega_0 - \omega) \exp(-i\omega\tau), \tag{3.74}$$

$$I_2(\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega \, \exp\left[-\frac{1}{4}(U\omega^2 - 2V\omega) - i\omega\tau\right], \tag{3.75}$$

U and V being defined by

$$c^2 U = \xi_3^2(\cos\theta_2 - \cos\theta_1)^2 + \xi_\perp^2(\sin\theta_2 + \sin\theta_1)^2 \tag{3.76}$$

$$\begin{aligned} cV = {} & \xi_3^2 k_0(1 - \cos\theta_1)(\cos\theta_2 - \cos\theta_1) \\ & + \xi_\perp^2 k_0 \sin\theta_1(\sin\theta_2 + \sin\theta_1). \end{aligned} \tag{3.77}$$

Evaluating these integrals

$$I_1(\tau) = -\frac{2\omega_0}{\tau^2} \exp\left(-i\frac{\omega_0\tau}{2}\right)\left(\cos\frac{\omega_0\tau}{2} - \operatorname{sinc}\frac{\omega_0\tau}{2}\right) \tag{3.78}$$

$$I_2(\tau) = \sqrt{\frac{4\pi}{U}} \exp\left(\frac{V^2}{4U}\right) \exp\left(-\frac{\tau^2}{U} - i\frac{V\tau}{U}\right), \tag{3.79}$$

so the two-photodetection amplitude is the convolution of the single-atom result with a Gaussian in τ. The result of Mollow's theory would simply be the sharply localised Gaussian which could be regarded as the result of convolving $I_2(\tau)$ with $\delta(\tau)$. In the Pike and Sarkar extension the convolution causes the two-photodetection amplitude to acquire a power-law decay which they interpret heuristically in terms of single photons having a power-law photodetection amplitude and make a comparison with the power-law tails of the weakly-localised single-photon states of Amrein [1].

Having this prediction of power-law behaviour in the two-photon detection amplitude we now estimate the magnitude of the effect. We shall place bounds on its magnitude in the special case of detectors placed symmetrically in the phase-matched positions to pick up photons whose average frequency is half the pump frequency. To deal with the more general case would alter the results in detail but not character. In the symmetric case we are discussing we have

$$c^2 U = \frac{2V}{\omega_0} = (2\xi_\perp \sin\theta^*)^2, \tag{3.80}$$

where θ^* is the angle that both detectors make with the pump direction. In this case we may write the two-photon detection amplitude as

$$G^{0,2}(x_1,x_2) \propto \exp\left[-i\frac{\omega_0}{2}(\bar{t}_2+\bar{t}_1)\right] \times \int_{-\omega_0/2}^{\omega_0/2}\frac{\mathrm{d}\omega}{2\pi}\left(\frac{\omega_0^2}{4}-\omega^2\right)\exp\left[-\frac{U\omega^2}{4}-i\omega(\bar{t}_2-\bar{t}_1)\right], \quad (3.81)$$

which by elementary manipulation gives

$$G^{0,2}(x_1,x_2) \propto \exp\left[-i\frac{\omega_0}{2}(\bar{t}_2+\bar{t}_1)\right]\int_{-1}^{1}\frac{\mathrm{d}w}{2\pi}(1-w^2)\exp\left[-\frac{1}{2}\alpha w^2-iw\bar{t}\right], \quad (3.82)$$

where

$$\alpha = \frac{U\omega_0^2}{8} \quad (3.83)$$

$$\bar{t} = \frac{\omega_0}{2}(\bar{t}_1-\bar{t}_2). \quad (3.84)$$

The power-law tails in photodetection thus arise from an integral of the form

$$f(t) = \int_{-1}^{1}\frac{\mathrm{d}w}{2\pi}(1-w^2)\exp\left(-\frac{1}{2}\alpha w^2-iwt\right). \quad (3.85)$$

Splitting the range of integration,

$$\int_{-1}^{1} = \int_{-\infty}^{\infty} - \int_{1}^{\infty} - \int_{-\infty}^{-1}, \quad (3.86)$$

we may perform the first of these analytically

$$\begin{aligned} S(t) &= \int_{-\infty}^{\infty}\frac{\mathrm{d}w}{2\pi}(1-w^2)\exp\left(-\frac{1}{2}\alpha w^2-iwt\right) \\ &= \left(1+2\frac{\partial}{\partial\alpha}\right)I(\alpha,t), \end{aligned} \quad (3.87)$$

where

$$\begin{aligned} I(\alpha,t) &= \int_{-\infty}^{\infty}\frac{\mathrm{d}w}{2\pi}\exp\left(-\frac{1}{2}\alpha w^2-iwt\right) \\ &= \frac{1}{\sqrt{2\pi\alpha}}\exp\left(-\frac{1}{2}\frac{t^2}{\alpha}\right), \end{aligned} \quad (3.88)$$

giving

$$S(t) = \frac{1}{\sqrt{2\pi\alpha}} \exp\left(-\frac{1}{2}\frac{t^2}{\alpha}\right)\left(1 - \frac{1}{\alpha} + \frac{t^2}{\alpha^2}\right). \tag{3.89}$$

The remaining two integrals must contain the power-law behaviour and may be written

$$T(t) = 2\int_0^\infty \frac{\mathrm{d}w}{2\pi} w(w+2) \exp\left[-\frac{1}{2}\alpha(w+1)^2\right] \cos\omega t, \tag{3.90}$$

giving the following bound on T

$$\begin{aligned} |T(t)| &< 2\exp(-\alpha/2)\int_0^\infty \frac{\mathrm{d}w}{2\pi} w(w+2)\exp\left(-\frac{1}{2}\alpha w^2\right) \\ &= \frac{1}{\alpha}\exp(-\alpha/2)\left(\frac{1}{\sqrt{2\pi\alpha}} + 4\right). \end{aligned} \tag{3.91}$$

This bound is useful when $\alpha \gg 1$, the frequency spectrum of the core being much narrower than that of the tail, which is the case normally realised. In this case $|T(t)| \ll S(t)$, and the relative size of the power-law tail compared to the value of $f(t)$ near the centre of the core is approximately given by

$$\frac{|T(t)|}{S(0)} < \frac{4}{\sqrt{2\pi\alpha^3}} \exp(-\alpha/2). \tag{3.92}$$

If we wish to observe these tails we must endeavour to make α as small as possible, which requires $\xi_\perp$ to be small. In the experiment of Rarity and Tapster [28] we have a pumping wavelength of 413nm and $\xi_\perp$ equal to approximately 80μm, with the photon pairs emitted at approximately 15° to the pump direction before exiting the medium. This leads to a value of $\alpha \sim 10^3$ and so in this case the tails would be undetectably small. To achieve $\alpha \sim 1$ we must make $\xi_\perp$ similar to the wavelength of the pumping radiation which in a practical experiment requires us to focus the laser down to a beam-waist of a wavelength or so in a very thin slice of non-linear material. However, even if this arrangement were feasible there does remain the problem of whether real detectors would demonstrate these tails. It is clear from the analysis that the tails are caused by the finite range of integration on equation 3.85. This is a consequence of taking the double positive-frequency part of the current-current correlation function in equation 3.28 which in turn arises from choosing the positive-frequency part of the field to give the photodetection amplitude. However, in Glauber's theory of photodetection taking the positive-frequency part of the field arises from the rotating-wave approximation in the interaction between radiation and matter. For a given frequency

ω this approximation becomes valid on time scales $\gg 1/\omega$. However the tails we predict are caused by the positive frequencies that the detectors are exposed to, right down to zero frequency. Therefore there is no timescale in this problem for which the rotating-wave approximation is uniformly valid, and so it is possible that the prediction of tails is not even reliable in principle. These considerations apply even for a photodetector which behaves ideally, having a flat spectral response from zero to the pump frequency, but non-ideal behaviour will further modify this picture and thus it seems that any tails predicted in this experiment are not related to Amrein's and are consequent on particular models of photodetection.

Chapter 4

Fourth-order interference in spontaneous parametric fluorescence

We have discussed in some detail some of the expected properties of the photon pairs produced in parametric fluorescence, in particular the dependence of the two-photon correlations on the illumination profile of the non-linear medium and the possibility of observing power law tails in the two-photon detection probability. In making these predictions, however, we are far ahead of their experimental verification. The practical problem we experience is that we do not have photon-detecting devices with sufficient speed to measure the small dispersion in arrival time difference predicted by the theory. For example, with the fluorescence produced by a 1mm diameter laser traversing a thin crystal and with the detectors placed at $\pm 20^\circ$ equation 3.62 predicts that the spread of arrival time differences is on a scale of less than one picosecond. This is well beyond the resolution of the experiments of Burnham and Weinberg [3] (10ns) or even Friberg et. al. [8] (100ps). However, we may investigate the properties of the photon pairs emerging from the medium more thoroughly using so called fourth-order interference effects as follows.

The experimental set up is as in figure 4.1. For the moment we are interested in the pairs which result from a pump photon being split into two photons of approximately equal energy, both of which propagate at equal angles to the pump-beam direction. Such pairs are selected by apertures A_1 and A_2. Using two symmetrically placed mirror and prism arrangements (M_1, P_1 and M_2, P_2) we cause the two photons to be incident on beamsplitter BS, and detect photons in detectors D_1 and D_2 on the far side of the splitter. In one of the arms of the experiment we introduce an extra path difference, δ, and investigate how the coincidence rate between D_1 and D_2 depends on

this path length.

We motivate this experiment by imagining a classical analogue in which the medium is a source of classical pulses of light which are emitted simultaneously down the two arms. When the path lengths are equal there is an interference effect which causes the product of the intensities at the detectors to be depressed below what would be observed when the two paths have a difference much longer than the length of the pulses, since in this case the pulses reach the beamsplitter separately and do not interfere. A particular feature of this classical analysis is that the visibility of the fringes that occur, defined as the ratio of the interference term in the coincidence rate to the constant background coincidence rate is always less than one half [23]. In this respect, as we shall see, the quantum experiment with photon pairs differs in an unmistakable manner.

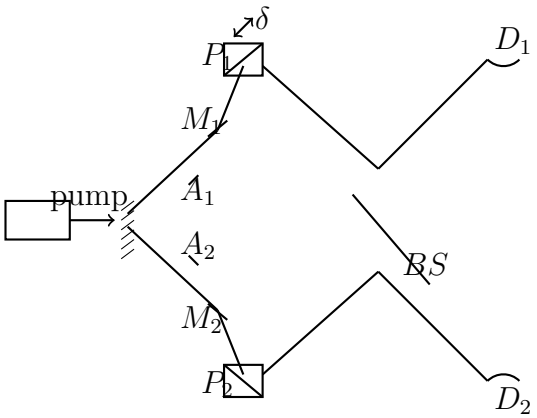

Figure 4.1: Experimental arrangement for the observation of fourth-order interference in spontaneous parametric fluorescence

4.1 Theory of fourth-order interference

In analysing the quantum experiment we follow the approach of Rarity and Tapster [28], the starting point for which is Mollow's expression for $G^{0,2}(x_1, x_2)$, the two-photon detection amplitude at spacetime points x_1 and x_2. This expression was derived for the emission of photons into free space, whereas in the experiment the photons may suffer extra delays, and are incident on mirrors and a beamsplitter. We now analyse the effect of these added optical components. Referring to figure 4.2, let $\alpha^+(\vec{r}, t)$ be the positive frequency part of the electromagnetic field after reflection at the beamsplitter. $B^+(\vec{r}, t)$ is the positive-frequency part of the field after a path delay is

introduced in one of the arms of the interferometer and $A^+(\vec{r},t)$ is the electromagnetic vector potential in the free space into which the medium emits the fluorescence. Firstly, we have

$$B^+(\vec{r},t) = A^+(\vec{r}+\vec{\delta},t), \tag{4.1}$$

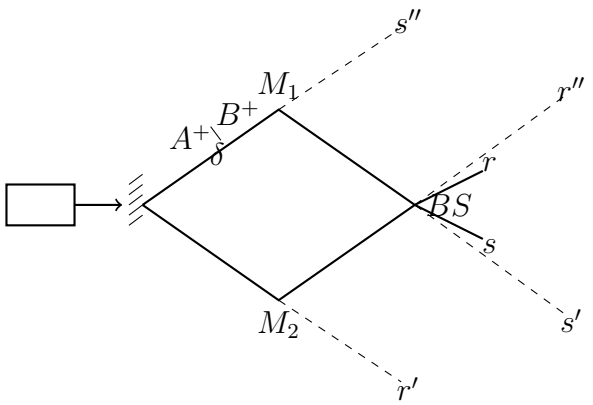

Figure 4.2: Definition of symbols used in the analysis of fourth-order interference

$\vec{\delta}$ being the extra path introduced in the upper arm of the interferometer, which we assume to be the only asymmetry between the upper and lower paths. We use the theory of mirrors and beamsplitters developed in appendix C to write

$$\alpha^+(\vec{r},t) = tA^+(\vec{r}',t) + rB^+(\vec{r}'',t) \tag{4.2}$$
$$\alpha^+(\vec{s},t) = -r^*A^+(\vec{s}',t) + t^*B^+(\vec{s}'',t), \tag{4.3}$$

where $\vec{r}'$ is the reflection of the detector position $\vec{r}$ in $M2$, $\vec{r}''$ the reflection of $\vec{r}$ in BS and then $M1$, $\vec{s}''$ that of $\vec{s}$ in $M1$ and $\vec{s}'$ that of $\vec{s}$ in BS and then $M2$. We have assumed the mirror to be non-dispersive over a frequency range at least as large as the bandwidth of the light that falls on it. The amplitude $C(t_1,t_2)$ for a dual photocount at points $\vec{r}$ and $\vec{s}$ at times t_1 and t_2 is given by

$$\begin{aligned} C(t_1,t_2) &= \langle \alpha^+(\vec{r},t_1)\alpha^+(\vec{s},t_2)\rangle \\ &= T\langle A^+(\vec{r}',t_1)A^+(\vec{s}'+\vec{\delta},t_2)\rangle - R\langle A^+(\vec{r}''+\vec{\delta},t_1)A^+(\vec{s}'',t_2)\rangle, \end{aligned} \tag{4.4}$$

where $T = tt^*$ and $R = rr^*$, the other two terms being negligible for a source with illuminated patch many wavelengths large, as they correspond

to emission very far from phase-matching. Taking Mollow's expression for the dual photocount amplitude

$$G^{0,2}(x_1, x_2) \propto e^{-i\omega_2^* \bar{t}_2 - i\omega_1^* \bar{t}_1} \psi(\bar{t}_2 - \bar{t}_1), \tag{4.5}$$

in the symmetric case we are considering

$$\omega_1^* = \omega_2^* = \omega_0/2, \tag{4.6}$$

we get

$$\begin{aligned} C(t_1, t_2) \propto\ & e^{i(\omega_0/2)[t_2 + t_1 - l/c]} \\ & \times \left[T\psi(t_2 - t_1 - (s'' - r' + \delta)/c) - R\psi(t_2 - t_1 - (s' - r'' - \delta)/c) \right], \end{aligned} \tag{4.7}$$

where

$$l = s'' + r' + \delta = s' + r'' + \delta, \tag{4.8}$$

δ being the extra path introduced, in addition to the Fraunhofer distances from the medium, to the emitted photons (and assumed not to introduce a frequency-dependent phase shift). We now write $\tau = t_1 - t_2$ and $d = s' - r'' = s'' - r'$.

The probability of joint photocounts at t_1 and t_2 is given by

$$\begin{aligned} |C(t_1, t_2)|^2 = {} & T^2 |\psi(\tau - (d + \delta)/c)|^2 + R^2 |\psi(\tau - (d - \delta)/c)|^2 \\ & - RT \left[\psi(\tau - (d + \delta)/c)^2 \psi^*(\tau - (d - \delta)/c)^2 + \text{c.c.} \right], \end{aligned} \tag{4.9}$$

where

$$\tau = t_1 - t_2, \tag{4.10}$$

$$d = s' - r'' = s'' - r'. \tag{4.11}$$

Due to the inadequate speed of current detectors what is measured is the integral over all τ of this quantity, which with suitable shifts in the time origin can be written

$$C \propto \int \mathrm{d}\tau \, |\psi(\tau)|^2 (T^2 + R^2) - RT \left[\psi(\tau - \delta/c) \psi^*(\tau + \delta/c) + \text{c.c.} \right], \tag{4.12}$$

or in terms of the Fourier transform of ψ,

$$C \propto \int \frac{\mathrm{d}\omega}{2\pi} \, |\tilde{\psi}(\omega)|^2 \left[T^2 + R^2 - 2RT \cos\left(\frac{2\omega\delta}{c} \right) \right]. \tag{4.13}$$

In the experiments of Rarity and Tapster a laser beam of Gaussian profile pumps a thin crystal, the appropriate ψ then comes from equation 3.62 which on substitution into equation 4.13 gives the coincidence rate

$$C \propto R^2 + T^2 - 2TR \exp\left(-\frac{\delta^2}{2\sigma^2 \sin^2\theta_*}\right), \tag{4.14}$$

Note that when the experimental set-up is symmetric ($\delta = 0$) the coincidence rate is minimum, and for the case of $T = R = 1/2$ is completely suppressed. In the case of complete suppression both members of the pair head for D_1 or both for D_2 as could be verified by evaluating the dual photocount probabilities integrated over all time in each arm separately, but also is clear from the observation that the beamsplitter does not destroy photons, nor does it affect the fluorescence from the medium, so the emitted photons must go somewhere. As we earlier intimated, this complete suppression has no classical explanation and is a manifestation of the quantum-mechanical nature of this phenomenon.

4.1.1 Tails in fourth-order interference

A proposal to experimentally observe the two-photodetection tails of section 3.3 was made by Pike and Walker and is currently the subject of SERC-funded experimental research. They hoped to find a reflection of the power-law tails in direct photodetection of the fluorescence in the results of a fourth-order interference at a beamsplitter in an experiment of the style of Rarity and Tapster.

The theoretical analysis of the proposal follows that of the fourth-order interference experiment that we have just described. However we use the expression for the two-photodetection amplitude of Pike and Sarkar [24, 25] which exhibits power-law tails to derive the appropriate $\psi(\tau)$,

$$\psi(\tau) \propto \int \frac{\mathrm{d}\omega}{2\pi}\, \theta(\omega_0/2 + \omega)\theta(\omega_0/2 - \omega) \left(\frac{\omega_0^2}{4} - \omega^2\right) \exp\left(-\frac{U\omega^2}{4} - i\omega\tau\right), \tag{4.15}$$

which leads to the following expression for the coincidence rate

$$C \propto \int_{-\omega_0/2}^{\omega_0/2} \frac{\mathrm{d}\omega}{2\pi} \left(\frac{\omega_0^2}{4} - \omega^2\right)^2 \exp\left(-\frac{U\omega^2}{2}\right) \left[T^2 + R^2 - 2TR\cos\left(\frac{2\omega\delta}{c}\right)\right]. \tag{4.16}$$

Rescaling the limits of integration gives

$$C \propto \int_{-1}^{1} \frac{\mathrm{d}w}{2\pi} (1 - w^2)^2 \exp\left(-\frac{1}{2}\alpha w^2\right) \left(T^2 + R^2 - 2TRe^{-iw\bar{\delta}}\right), \tag{4.17}$$

where

$$\alpha = \frac{U\omega_0^2}{4} \tag{4.18}$$

$$\bar{\delta} = \frac{2\omega_0\delta}{c}. \tag{4.19}$$

We may write

$$C(\delta) \propto (T^2 + R^2)g(0) - 2TRg(\bar{\delta}), \tag{4.20}$$

where

$$g(\bar{\delta}) = \int_{-1}^{1} \frac{\mathrm{d}w}{2\pi}(1-w^2)^2 \exp\left(-\frac{1}{2}\alpha w^2\right) e^{iw\bar{\delta}}. \tag{4.21}$$

Since $g(\bar{\delta})$ is the Fourier transform of a function defined on a compact support, by the Paley-Wiener theorem it must exhibit power-law decay as $\bar{\delta} \to \pm\infty$, and it is in this way that the power-law tails of Pike and Sarkar are reflected in fourth-order interference experiments. $g(\bar{\delta})$ has the form of a convolution.

$$g(\bar{\delta}) = J_1(\bar{\delta}) \otimes J_2(\bar{\delta}), \tag{4.22}$$

where

$$\begin{aligned} J_1(\bar{\delta}) &= \int \frac{\mathrm{d}w}{2\pi} \exp\left(-\frac{1}{2}\alpha w^2\right) e^{-iw\bar{\delta}} \\ &= \frac{1}{\sqrt{2\pi\alpha}} \exp\left(-\frac{1}{2}\frac{\bar{\delta}^2}{\alpha}\right), \end{aligned} \tag{4.23}$$

$$\begin{aligned} J_2(\bar{\delta}) &= \int_{-1}^{1} \frac{\mathrm{d}w}{2\pi}(1-w^2)^2 e^{-iw\bar{\delta}} \\ &= -\frac{16}{\bar{\delta}^3}\left(\sin\bar{\delta} + \frac{3\cos\bar{\delta}}{\bar{\delta}} - \frac{3\sin\bar{\delta}}{\bar{\delta}^2}\right). \end{aligned} \tag{4.24}$$

In contrast to the tails in the two-photon detection amplitude, which decayed as the inverse second power of the detection retarded time difference, these tails in the interference term fall-off as the inverse third power of the path difference introduced between the arms.

As before we may place an upper bound on the magnitude of the tails. We write

$$g(\bar{\delta}) = g_l(\bar{\delta}) + g_t(\bar{\delta}), \tag{4.25}$$

where g_l is the localised part of g and is given by

$$\begin{aligned} g_l &= \int_{-\infty}^{+\infty} \frac{\mathrm{d}w}{2\pi}(1-w^2)^2 \exp\left(-\frac{1}{2}\alpha w^2\right) e^{-iw\bar{\delta}} \\ &= \frac{1}{\sqrt{2\pi\alpha}} \exp\left(-\frac{1}{2}\frac{\bar{\delta}^2}{\alpha}\right)\left[\left(1-\frac{2}{\alpha}+\frac{3}{\alpha^2}\right)+\left(\frac{2}{\alpha^2}-\frac{6}{\alpha^3}\right)\bar{\delta}^2+\left(\frac{1}{\alpha}\right)\bar{\delta}^4\right], \end{aligned} \tag{4.26}$$

and g_t contains the power law decay

$$g_t = 2\int_0^{+\infty} \frac{\mathrm{d}w}{2\pi} w^2(2+w)^2 \exp\left[-\frac{1}{2}\alpha(w+1)^2\right]\cos w\bar{\delta}. \tag{4.27}$$

This leads to a bound on the size of the tail

$$\begin{aligned} |g_t| &< 2\exp(-\alpha/2)\int_0^{+\infty} \frac{\mathrm{d}w}{2\pi} w^2(2+w)^2 \exp\left(-\frac{1}{2}\alpha w^2\right) \\ &= \left[\frac{1}{\sqrt{2\pi\alpha}}\left(\frac{4}{\alpha}+\frac{3}{\alpha^2}\right)+\frac{16}{\alpha^2}\right]\exp(-\alpha/2). \end{aligned} \tag{4.28}$$

Not surprisingly, we again find the tails to be exponentially small in α.

4.2 Two-colour interference

We have so far discussed fourth-order interference effects observed by mixing the two beams emerging symmetrically with respect to the pump beam, each of which has average frequency equal to half the pump frequency. It is, however, possible to observe fourth-order interference in the photon pairs produced by an asymmetric split of the pump frequency. This has been investigated experimentally by Rarity and Tapster [29] and is an effect they call 'two-colour interference' as it involves the mixing of four different beams, of two different colours, on two beamsplitters, as shown in figure 4.3.

In contrast to the previous experiments it is now possible to introduce path length differences between the beams of each colour separately. The analysis of the two-colour interference experiments proceeds much as for the experiments we have already examined. Returning to figure 4.3, let us assume detectors at points $\vec{r}$ and $\vec{s}$. $\vec{r}'$ is the point obtained by reflecting $\vec{r}$ in $M2$, $\vec{r}''$ by reflecting $\vec{r}$ in $BS1$ and then $M1$, $\vec{s}''$ by reflection of $\vec{s}$ in $M1$ and $\vec{s}'$ by reflection in $BS2$ and then $M2$. Extra paths $\vec{\delta}_1$ and $\vec{\delta}_2$ may be introduced into the beams to break and otherwise completely symmetrical arrangement. We assume that $BS1$ and $BS2$ are non-dispersive for the range of frequencies that

fall on them, though not necessarily that they have the same transmission and reflection coefficients as each other. Instead of equation 4.3 we have

$$\alpha^+(\vec{r},t) = t_1 A^+(\vec{r}',t) + r_1 B^+(\vec{r}'',t) \tag{4.29}$$

$$\alpha^+(\vec{s},t) = -r_2^* A^+(\vec{s}',t) + t_2^* B^+(\vec{s}'',t). \tag{4.30}$$

The amplitude for dual photocounts at $\vec{r}$ and $\vec{s}$ at times t_1 and t_2 then becomes

$$\begin{aligned} C(t_1,t_2) &= \langle \alpha^+(\vec{r},t_1)\alpha^+(\vec{s},t_2)\rangle \\ &= t_1 t_2^* \langle A^+(\vec{r}',t_1)A^+(\vec{s}''+\vec{\delta}_2,t_2)\rangle - r_1 r_2^* \langle A^+(\vec{r}''+\vec{\delta}_1,t_1)A^+(\vec{s}',t_2)\rangle \end{aligned} \tag{4.31}$$

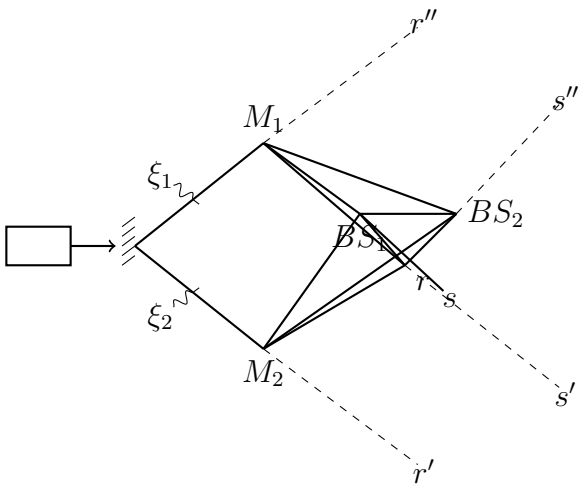

Figure 4.3: Schematic arrangement for observation of two-colour interference

Again using Mollow's expression for the two-photocount amplitude, equation 4.5, we obtain, to within an overall unimportant phase-factor,

$$C(t_1,t_2) = t_1 t_2^* e^{i\omega_1^* \delta_1/c}\psi(\tau - \delta_2/c) - r_1 r_2^* e^{i\omega_2^* \delta_2/c}\psi(\tau + \delta_1/c), \tag{4.32}$$

where the perfectly symmetrical experimental arrangement except for the path differences deliberately introduced implies

$$\vec{r}' - \vec{s}'' = \vec{r}'' - \vec{s}', \tag{4.33}$$

and we have set

$$\tau = t_2 - s''/c - t_1 + r'/c. \tag{4.34}$$

As before, if we take the modulus squared of this expression and integrate over all τ, we obtain the coincidence rate between detectors at $\vec{r}$ and $\vec{s}$. To be a little more specific we assume that a path difference δ is introduced for both colours and that the mirror is non-dispersive for all frequencies that fall on it, so that $t_1 = t_2 = t$, $r_1 = r_2 = r$, that the same path difference, δ, is introduced for each colour, and we take

$$\psi(\tau) = \exp\left(-\frac{1}{2}\frac{\tau^2}{R^2 A_z^2}\right), \tag{4.35}$$

where

$$A_z = \frac{c}{n_1}\sin\theta_1^* + \frac{c}{n_2}\sin\theta_2^*, \tag{4.36}$$

which is equation 3.62 for detector positions which lie at θ_1^* and θ_2^* when appropriately reflected in the mirrors and beamsplitter. Under these conditions the coincidence rate shows cosinusoidal oscillations damped by a Gaussian envelope as δ is altered

$$C \propto T^2 + R^2 - 2RT\cos\left[(\omega_1^* - \omega_2^*)\delta/c\right]\exp\left(\frac{\delta^2}{2R^2 A_z^2 c^2}\right), \tag{4.37}$$

4.3 Physical interpretation of fourth-order interference

For the two-beam fourth-order interference experiment, the range of path differences for which the depression in coincidence rate persists is proportional to the width of the function $\psi(\tau)$, requiring the overlap of $\psi(\tau + \delta/c)$ and $\psi(\tau - \delta/c)$ in equation 4.12. This has been interpreted as the necessity for single-photon wavepackets to overlap at the beamsplitter for the interference to occur, but it is in the two-colour interference experiment that we see how strange these fourth-order interference effects really are. Here the interference effect does not depend on the two members of the pair impinging on the same part of a single beamsplitter, and so two-colour interference cannot be interpreted in terms of single-photon wavepacket overlap, yet it is clear that four-beam interference and two-beam interference experiments differ only in detail, not in substance. The desire to understand more deeply what is at the root of this phenomenon was a principal motivation for the two alternative approaches to fourth-order interference which are described in the next chapter.

Chapter 5

Alternative approaches to the analysis of fourth-order interference

In this chapter we present two analyses of fourth-order interference phenomena. The first is based on Kleinman's [16] approach to the spontaneous parametric fluorescence problem in which a term cubic in the electric field is added to the Hamiltonian of the non-linear material to account for the non-linear susceptibility of the material which generates the pairs, and Fermi's golden rule provides the transition rate of the system into states in which photon pairs are present. All that is necessary to describe fourth-order interference is to take account of the way in which the field are altered by the presence of the beamsplitter.

The second approach is inspired by an approach to quantum electrodynamics and quantum optical phenomena due to Feynman [6]. We succeed in describing fourth-order interference in terms of the interference between quantum-mechanical alternatives each involving two photons, rather than in terms of interference between the separate members of each pair. These considerations are a simple extension of explanations of Young's experiment which involve the addition of quantum-mechanical amplitudes for a photon to pass through each slit to obtain the amplitude for a photon to arrive at a given point on the screen behind the slits. Unlike Young's experiment, however, which has a classical analogue in the sense that the interference pattern is unaltered if we turn up the intensity of the light illuminating the slits so that the light can be described by the classical Maxwell equations (instead of in terms of single photons), the fourth-order interference has no classical analogue, as we have already pointed out.

It may already appear that we have a perfectly satisfactory description of

fourth-order interference phenomena arising from Mollow's theory of spontaneous parametric fluorescence. However, apart from the desirability of multiple viewpoints to gain deeper insight, we believe there is serious reason to question the fundamentals of Mollow's theory for spontaneous parametric fluorescence. If we really had a complete description of the physics of this phenomenon, the spontaneous fluorescence could be described by the same theory as used to describe the mixing of three classical modes. In the theory of classical three-wave mixing we treat the field entirely classically, and the matter quantum-mechanically, so that this framework is inadequate for a description of spontaneous parametric fluorescence, which needs a quantum picture of the field. Mollow treats the atom as driven by a prescribed classical field and then uses the quantum equations 3.28 to apparently describe the fluorescence. However, he only takes the atom-field interaction to first order, whereas in section 3.1 we have to go to second-order perturbation theory to describe the non-linear susceptibility responsible for three-wave mixing. He derives a perturbation expansion for the Heisenberg dipole operator which conceals the fact that, even if he were to retain the full quantum field interacting with the atom, when the problem is viewed in the interaction picture the interaction evolution operator is linear in the electric field, and so the interaction ket only consists of vacuum and single-photon components. It is therefore difficult to see how his framework leads to a correct description of effects involving two photons. Nevertheless, we must address the question of why Mollow's theory seems to gives satisfactory predictions, at least for fourth-order interference experiments. Having the time-ordered current-current correlation function in equation 3.28 means that each atom makes a contribution to the two-photodetection amplitude which has a discontinuity at $t_1 = t_2$ which is then taken out to the observation points by the attached propagators. However, due to the phase-matching which happens when integration over an extended non-linear medium is performed, only a very narrow range of frequencies in this function make a contribution. Therefore, almost any quantity which has most of its change close to $t_1 = t_2$ (close in comparison to the final dispersion of arrival time differences) could replace the current-current correlation function without substantially affecting the predictions of the theory. In particular a quantity proportional to $\delta(t_1 - t_2)\delta(\vec{r}_1)\delta(\vec{r}_2)$ would perform satisfactorily as the current-current correlation function for a single atom, and this is at the root of our explanation of Mollow's theory in terms of the simultaneous generation of two photons at a point in the crystal, with subsequent propagation delays out to detectors.

A description of fourth-order interference which uses a phenomenological interaction Hamiltonian similar to that adopted by Kleinman, with two creation operators and one annihilation operator, and therefore capable of

describing the physical process of destruction of a pump photon and the creation of a pair in its place, has already been given by Ou et al. [9, 23]. They, in common with Mollow, evaluate a two-photodetection amplitude for the radiation emerging from the medium and then follow a similar approach to Rarity and Tapster in evaluating the fields after the beamsplitter and integrating over all time differences between photodetections to obtain the coincidence rate between detectors which only have coarse time resolution. The approach that we adopt calculates the emission rate of photons into final momentum directions using Fermi's golden rule which has the advantage of being technically simpler, not involving the calculation of information which is later integrated out, and also not resting on a particular model of photodetection, as is implied by calculating a Glauber-type photodetection amplitude.

5.1 Golden-rule analysis of spontaneous parametric fluorescence

Prior to an analysis of fourth-order interference experiments we will calculate the expected spectrum, distribution and intensity of spontaneous parametrically fluorescent light using the same theoretical machinery. As in the analysis of the power-law photodetection tails of Pike and Sarkar, the model we use is of a region of non-linear material embedded in free space with the pump carrying an anomalous momentum to facilitate phase-matching. We take the electromagnetic field to be scalar, which does not really represent a loss of generality, since the fluorescence is emitted only into the mode of ordinary polarisation, and provided we arrange for complete overlap of the polarisation vectors in the interference experiments we shall describe, the scalar theory is capable of describing the physics that would arise in the vector theory. Accordingly, the electromagnetic field in is modelled by a scalar field $\phi(\vec{x}, t)$ whose free Lagrangian density is

$$\mathcal{L} = \frac{\epsilon_0}{2}\left(\frac{\partial\phi}{\partial t}\right)^2 - \frac{1}{2\mu_0}(\nabla\phi)^2. \tag{5.1}$$

The momentum conjugate to ϕ is

$$\pi = \epsilon_0 \frac{\partial\phi}{\partial t}. \tag{5.2}$$

The Hamiltonian is therefore

$$H = \int \mathrm{d}^3\vec{x}\, \frac{\pi^2}{2\epsilon_0} + \frac{1}{2\mu_0}(\nabla\phi)^2. \tag{5.3}$$

Quantising this theory in the usual way leads to the familiar expressions for the fields in terms of the creation and annihilation operators

$$\phi(x) = \int \frac{\mathrm{d}^3\vec{k}}{(2\pi)^3}\, i \left(\frac{\hbar}{2\epsilon_0\omega_k}\right)^{1/2} \left[a^\dagger(\vec{k})e^{-i\vec{k}\cdot\vec{x}} - a(\vec{k})e^{i\vec{k}\cdot\vec{x}}\right] \tag{5.4}$$

$$\pi(x) = \int \frac{\mathrm{d}^3\vec{k}}{(2\pi)^3} \left(\frac{\hbar\epsilon_0\omega_k}{2}\right)^{1/3} \left[a^\dagger(\vec{k})e^{-i\vec{k}\cdot\vec{x}} + a(\vec{k})e^{i\vec{k}\cdot\vec{x}}\right], \tag{5.5}$$

where $a(\vec{q})$ and $a^\dagger(\vec{q}')$ satisfy the following normalisation conditions

$$\left[a(\vec{q}), a^\dagger(\vec{q}')\right] = (2\pi)^3\delta(\vec{q} - \vec{q}'). \tag{5.6}$$

Having the free field quantisation scheme the non-linear optical interaction is introduced as a perturbation

$$V = \frac{\chi}{3}\int_M \mathrm{d}^3\vec{x}\, E^3(\vec{x}), \tag{5.7}$$

where the integration proceeds over the volume of the non-linear medium and

$$E = \frac{\partial\phi}{\partial t}. \tag{5.8}$$

To calculate the rate of photon pair production we assume that the field starts in the state $|0, 0, \mathrm{coh}\rangle$, where the zeros refer to the occupation numbers of the fluorescent modes and coh labels the coherent state of the pumping radiation, and we look for the transition rate into states containing photons with wavevectors $\vec{q}$ and $\vec{q}'$ and the pump still in a coherent state, this state we label as $|\vec{q}, \vec{q}', \mathrm{coh}\rangle$. Fermi's golden rule gives the transition rate R as

$$R = \frac{2\pi}{\hbar}\,|V_{fi}|^2\,\delta(E_f - E_i), \tag{5.9}$$

with summation over a set of final states with energy close to that of the initial one. The transition matrix element is given by

$$V_{fi} = \frac{\chi}{3}\langle\vec{q}, \vec{q}', \mathrm{coh}|\int_M \mathrm{d}^3\vec{x}\, E^3(\vec{x})|0, 0, \mathrm{coh}\rangle. \tag{5.10}$$

Using

$$|\vec{q}, \vec{q}', \mathrm{coh}\rangle = a^\dagger(\vec{q})a^\dagger(\vec{q}')|0, 0, \mathrm{coh}\rangle, \tag{5.11}$$

and inserting the expression for the electric field from equations 5.5 and 5.8 into the expression for V_{fi} results in

$$V_{fi} = \frac{\hbar\chi}{2\epsilon_0}(\omega_q\omega_{q'})^{1/2}\int_M \mathrm{d}^3\vec{x}\, E_p^+(\vec{x})e^{-i(\vec{q}+\vec{q}')\cdot\vec{x}}. \tag{5.12}$$

As Mollow did, we consider the case of a parallel beam propagating in the z-direction through the medium,

$$E_p^+(\vec{x}) = E_0 f(x,y) e^{iq_0 z}, \tag{5.13}$$

where E_p^+ is the positive-frequency part of the pump electric field—the wavenumber of the pump being that appropriate to an extraordinary ray—and obtain

$$V_{fi} = \frac{\hbar\chi E_0}{2\epsilon_0}(\omega_q\omega_{q'})^{1/2}\tilde{f}(\vec{q}+\vec{q}'-\vec{q}_0), \tag{5.14}$$

where

$$\tilde{f}(\vec{k}) = \int_M \mathrm{d}^3\vec{x}\, f(x,y)e^{-i\vec{k}\cdot\vec{x}}. \tag{5.15}$$

If a Gaussian form is again assumed for the pumping laser beam's transverse profile and that the medium forms a slab of length l perpendicular to the beam

$$f(x,y) = \exp\left(-\frac{x^2+y^2}{2R^2}\right), \tag{5.16}$$

the form-factor then becomes

$$\tilde{f}(k_x,k_y,k_z) = 2V\,\mathrm{sinc}\left(\frac{k_z l}{2}\right)\exp\left[-\frac{R^2}{2}(k_x^2+k_y^2)\right], \tag{5.17}$$

where $V = \pi R^2 l$.

The most fundamental process we can measure is the emission of a photon pair into the solid angles $\mathrm{d}\Omega$ and $\mathrm{d}\Omega'$ with the unprimed photon having wavenumber in the range q to $q+\delta q$. Fermi's golden rule gives the rate for this as

$$\begin{aligned}\frac{\mathrm{d}^3R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'\,\mathrm{d}q} = \frac{2\pi}{\hbar}\left(\frac{\hbar\chi E_0 V}{\epsilon_0}\right)^2 \omega_q \frac{q^2}{(2\pi)^3}\int \frac{\mathrm{d}q'}{(2\pi)^3}\,\omega_{q'}\,\mathrm{sinc}^2\left[\tfrac{1}{2}(q_z+q_z'-q_0)\right] \\ \times\exp\left[-R^2\left((q_x+q_x')^2+(q_y+q_y')^2\right)\right]\delta(\hbar\omega_q+\hbar\omega_{q'}-\hbar\omega_0).\end{aligned} \tag{5.18}$$

Performing the integration over the delta function gives

$$\begin{aligned}\frac{\mathrm{d}^3R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'\,\mathrm{d}q} = I\omega_q\omega_{q'}\frac{q^2}{(2\pi)^3}\frac{q'^2(q)}{(2\pi)^3}\,\mathrm{sinc}^2\left[\tfrac{1}{2}(q_z+q_z'(q)-q_0)\right] \\ \times\exp\left[-R^2\left((q_x+q_x'(q))^2+(q_y+q_y'(q))^2\right)\right],\end{aligned} \tag{5.19}$$

where

$$I = \frac{2\pi}{\hbar}\left(\frac{\hbar\chi E_0 V}{\epsilon_0}\right)^2\frac{1}{\hbar c}. \tag{5.20}$$

$\vec{q}'(q)$ is determined by the direction specified by $\mathrm{d}\Omega'$ and $cq'/n = \omega_0 - cq/n$. We will now assume that the illuminated patch of the source is large enough so that knowledge of direction of the unprimed photon means that the primed photon is emitted into a small region of phase-space as a consequence of $\tilde{f}(\vec{k})$ being sharply peaked near $\vec{k} = \vec{0}$. In this case, as we have done before we may expand about the phase-matched $(*)$ configuration which is defined by

$$\vec{q}_* + \vec{q}'_* = \vec{q}_0 \tag{5.21}$$

$$\omega_{q_*} + \omega_{q'_*} = \omega_0. \tag{5.22}$$

We take polar coordinates with the polar axis along the pump direction and the direction of $\vec{q}$ defining the zero of azimuth and expand in small deviations from the phase-matched configuration allowing only the direction of the primed photon to deviate from the optimum.

$$q \to q_* + \delta q \tag{5.23}$$

$$q'(q) \to q'_* - \delta q \tag{5.24}$$

$$\theta' \to \theta'_* + \delta\theta' \tag{5.25}$$

$$\phi' \to \delta\phi' \tag{5.26}$$

5.1.1 Spectrum of fluorescent light

We first of all will examine the spectrum of the fluorescent light. For simplicity, we will restrict our attention solely to the phase-matched directions $\delta\theta' = 0$ and $\delta\phi' = 0$ since the spectrum only suffers slight shifts away from this configuration, but has the same width and functional form. Substituting equations 5.23 to 5.26 into the expression for the pair emission rate 5.19 we obtain

$$\begin{aligned}\frac{\mathrm{d}^3 R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'\,\mathrm{d}q} = I\omega_{q_*}\omega_{q'_*}\frac{q^2}{(2\pi)^3}\frac{q'^2(q)}{(2\pi)^3}\,\mathrm{sinc}^2\left[\tfrac{1}{2}(\cos\theta_* - \cos\theta'_*)\delta q\right] \\ \times \exp\left[-R^2(\sin\theta_* + \sin\theta'_*)^2(\delta q)^2\right].\end{aligned} \tag{5.27}$$

For a given θ_* and θ'_* there are two extreme cases:

- $R(\sin\theta_* + \sin\theta'_*) \ll l(\cos\theta_* - \cos\theta'_*)$. The spectra are of a sinc^2 form of width $2\pi/[l(\cos\theta_* - \cos\theta'_*)]$ to the first zero. It is the imbalance in longitudinal momentum carried by the photon pair as δq moves away from 0 which causes the loss of phase-matching.

- $R(\sin\theta_* + \sin\theta'_*) \gg l(\cos\theta_* - \cos\theta'_*)$. Each photon has a Gaussian spectrum centred about the phase-matched frequency of width (in

wavenumber) $1/[R(\sin\theta_* + \sin\theta'_*)]$ to the $1/e$ points in intensity. Here it is the imbalance in transverse momentum carried by the photon pair that causes the loss of phase-matching.

Of course what we typically have experimentally is R and l fixed with light emerging at a range of angles. Pairs whose individual members have half the pump photon energy emerge with $\theta_* = \theta'_*$ and inevitably have the Gaussian spectrum, whereas a pair emitted with one photon travelling forward and one backward will have the sinc^2 spectrum. Between these two extremes there will be a smooth transition whose exact form depends on the ratio R/l.

5.1.2 Emission into a pair of solid angles

To calculate the rate of emission into a pair of solid angles $\mathrm{d}\Omega$ and $\mathrm{d}\Omega'$ we simply integrate the previous expressions for the rate per unit wavenumber range over all δq. Corresponding to the former case above this results in

$$\frac{\mathrm{d}^2 R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = I\omega_{q_*}\omega_{q'_*}\frac{q^2}{(2\pi)^3}\frac{q'^2}{(2\pi)^3}\frac{2\pi}{l(\cos\theta_* - \cos\theta'_*)} \times \exp\left[-R^2 q'^2_* \left(\cos^2\theta'_*(\delta\theta')^2 + \sin^2\theta'_*(\delta\phi')^2\right)\right]. \tag{5.28}$$

As the primed direction moves away from the optimum there is a Gaussian fall-off in the two-photon emission rate. The polar and azimuthal angular widths $\Delta\theta'$ and $\Delta\phi'$ are given by

$$\Delta\theta' = \frac{1}{Rq'_*\cos\theta'_*} \tag{5.29}$$

$$\Delta\phi' = \frac{1}{Rq'_*\sin\theta'_*}. \tag{5.30}$$

Corresponding to the latter case we have

$$\frac{\mathrm{d}^2 R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = I\omega_{q_*}\omega_{q'_*}\frac{q^2}{(2\pi)^3}\frac{q'^2}{(2\pi)^3}\frac{\sqrt{\pi}}{R(\sin\theta_* + \sin\theta'_*)} \times \exp\left[-R^2 q'^2_* \sin^2\theta'_*(\delta\phi')^2\right] \mathrm{sinc}^2\left(\tfrac{1}{2}q'_* \sin\theta'_*\, l\,\delta\theta'\right), \tag{5.31}$$

so in this case the two-photon intensity falls off in a Gaussian fashion azimuthally and as sinc^2 in the polar direction with widths

$$\Delta\theta' = \frac{2\pi}{lq'_*\sin\theta'_*} \tag{5.32}$$

$$\Delta\phi' = \frac{1}{Rq'_*\sin\theta'_*}. \tag{5.33}$$

5.1.3 Intensity of fluorescent light

We may now calculate the single-photon emission rate in any direction by integrating over all solid angles $\mathrm{d}\Omega'$. It is convenient to express the results in terms of the pumping power rather than the electric field in the medium. For the transverse mode shape that we chose the relationship between these two quantities is

$$E_0^2 = \frac{P}{2\epsilon_0 c R^2}, \tag{5.34}$$

where P is the power with which the medium was pumped. We also find it convenient to express the results in terms of frequencies rather than wavenumbers. For the first case we obtain

$$\frac{\mathrm{d}R}{\mathrm{d}\Omega} = \frac{1}{32\pi}\frac{\chi^2}{\epsilon_0^3 c^4} P l \omega_*^3 \omega_*' \frac{1}{\cos\theta_*'(\cos\theta_* - \cos\theta_*')}, \tag{5.35}$$

and for the second

$$\frac{\mathrm{d}R}{\mathrm{d}\Omega} = \frac{1}{32\pi}\frac{\chi^2}{\epsilon_0^3 c^4} P l \omega_*^3 \omega_*' \frac{1}{\sin\theta_*'(\sin\theta_* + \sin\theta_*')}. \tag{5.36}$$

5.2 Analysis of fourth-order interference phenomena

Here we use the approach developed for the photon pair emission rate to analyse fourth-order interference effects. The principle is to express the annihilation operators for modes after the beamsplitter in terms of those for the free space modes into which the parametric fluorescence is emitted. We may then make use of the matrix elements of the previous section.

In appendix C it is shown that the action of a beamsplitter whose orientation is defined by the vector $\vec{l}$ and containing the point $\vec{r}$ is given by

$$a_{\mathrm{out}}(\vec{k}) = t a_{\mathrm{in}}(\vec{k}) + r e^{-2i(\vec{k}\cdot\vec{l})(\vec{k}\cdot\vec{l})} a_{\mathrm{in}}(L\vec{k}) \tag{5.37}$$

$$a_{\mathrm{out}}(\vec{k}') = -r^* e^{(\vec{k}'\cdot\vec{l})(\vec{k}'\cdot\vec{l})} a_{\mathrm{in}}(L\vec{k}') + t^* a_{\mathrm{in}}(k'), \tag{5.38}$$

where the operation L acting on a vector reflects it in the plane of the beamsplitter:

$$L\vec{x} = \vec{x} - 2(\vec{x}\cdot\vec{l})\vec{l}. \tag{5.39}$$

We assume a mirror to be equivalent to a beamsplitter with zero transmission coefficient.

The geometry for fourth-order interference is illustrated in figure 5.1. We assume that the two mirrors are defined by normals $\vec{l}$ and $\vec{m}$ and contain the points $\vec{r}$ and $\vec{s}$ The beamsplitter has normal $UV\vec{n}$ and we set the origin of coordinates in the plane of the beamsplitter. The modes we illustrate are related as follows:

$$d(\vec{q}) = tc'(\vec{q}) + rc(N\vec{q}) \tag{5.40}$$
$$d'(\vec{q}) = -r^*c'(N\vec{q}) + t^*c(\vec{q}) \tag{5.41}$$
$$c(\vec{q}) = b(L\vec{q})e^{-2i(\vec{r}\cdot\vec{l})(\vec{q}\cdot\vec{l})} \tag{5.42}$$
$$c'(\vec{q}) = a(M\vec{q})e^{-2i(\vec{x}\cdot\vec{m})(\vec{q}\cdot\vec{m})} \tag{5.43}$$
$$b(\vec{q}) = a(\vec{q})e^{i\vec{q}\cdot\vec{\delta}(\vec{q})}. \tag{5.44}$$

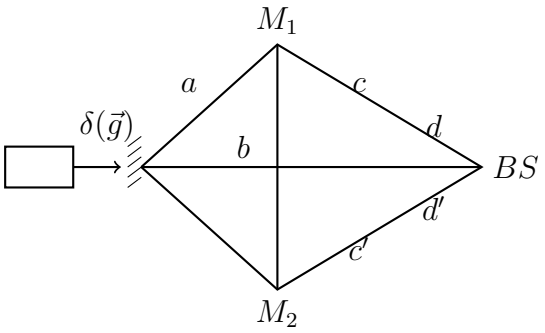

Figure 5.1: Mode transformations induced by optical components in fourth-order interference experiment

Solving these for d and d' in terms of the a's

$$d(\vec{q}) = \alpha_M(\vec{q})a(M\vec{q}) + \alpha_{LN}(\vec{q})a(LN\vec{q}) \tag{5.45}$$
$$d'(\vec{q}) = \alpha_{MN}(\vec{q})a(NM\vec{q}) + \alpha_L(\vec{q})a(L\vec{q}), \tag{5.46}$$

where

$$\alpha_M(\vec{q}) = te^{-2i(\vec{x}\cdot\vec{m})(\vec{q}\cdot\vec{m})} \tag{5.47}$$
$$\alpha_{LN}(\vec{q}) = re^{iLN\vec{q}\cdot\vec{\delta}(LN\vec{q})-2i(\vec{r}\cdot\vec{l})(N\vec{q}\cdot\vec{l})} \tag{5.48}$$
$$\alpha_{MN}(\vec{q}) = -r^*e^{-2i(\vec{x}\cdot\vec{m})(\vec{q}\cdot\vec{m})} \tag{5.49}$$
$$\alpha_L(\vec{q}) = t^*e^{iL\vec{q}\cdot\vec{\delta}(L\vec{q})-2i(\vec{r}\cdot\vec{l})(\vec{q}\cdot\vec{l})}. \tag{5.50}$$

Again we look for the emission of photons into solid angles $\mathrm{d}\Omega$ and $\mathrm{d}\Omega'$ the final states of the system being of the form

$$\langle f| = \langle \vec{q}, \vec{q}', \mathrm{coh}| = \langle 0, 0, \mathrm{coh}|\, d(\vec{q})d(\vec{q}'). \tag{5.51}$$

Using equations 5.46 the expression for the transition matrix element is

$$V_{fi} = \langle 0,0,\mathrm{coh}|\left[\alpha_M(\vec{q})a(M\vec{q}) + \alpha_{LN}(\vec{q})a(LN\vec{q})\right] \times \left[\alpha_{MN}(\vec{q}')a(MN\vec{q}') + \alpha_L(\vec{q}')a(L\vec{q}')\right]\frac{\chi}{3}\int \mathrm{d}^3\vec{x}\, E^3(\vec{x})|0,0,\mathrm{coh}\rangle \quad (5.52)$$

Using equation 5.14 we obtain V_{fi} in the form

$$V_{fi} = \frac{\chi\hbar}{2\epsilon_0}(\omega_q\omega_{q'})^{1/2}E_0\Big[\alpha_M(\vec{q})\alpha_L(\vec{q}')\tilde{f}(M\vec{q}+L\vec{q}'-\vec{q}_0) + \alpha_{LN}(\vec{q})\alpha_{MN}(\vec{q}')\tilde{f}(LN\vec{q}+MN\vec{q}'-\vec{q}_0)\Big], \quad (5.53)$$

the other terms disappearing for a source many wavelengths large. We can see in equation 5.53 that the general requirement for the attainment of fourth-order interference is the simultaneous non-vanishing of $\tilde{f}(M\vec{q}+L\vec{q}'-\vec{q}_0)$ and $\tilde{f}(LN\vec{q}+MN\vec{q}'-\vec{q}_0)$. The larger the region of illumination of the source, the more rapidly vanishing is $\tilde{f}(\vec{q})$ as q becomes larger. Thus, for a given misalignment of mirrors, the achieving of fourth-order interference is facilitated by making the illuminated patch of the source smaller so that the region of $\vec{q}$ for which $\tilde{f}(\vec{q})$ is non-vanishing encompasses both $M\vec{q}+L\vec{q}'-\vec{q}_0$ and $LN\vec{q}+MN\vec{q}'-\vec{q}_0$. However, we will specialise, as we have done before, to the case of a perfectly symmetrical experimental set-up, apart from the deliberately introduced path differences. In this case the mirrors and the beamsplitters all have the same normal so that $L = M = N$, $\vec{l} = \vec{m} = \vec{n}$ and the positions of mirrors L and M satisfy $\vec{r} = -\vec{s}$. The pump-beam wavevector $\vec{q}_0$ lies in the plane of the beamsplitter so that $\vec{q}_0 = N\vec{q}_0$. As a consequence of these assumptions we have

$$\tilde{f}(M\vec{q}+L\vec{q}'-\vec{q}_0) = \tilde{f}(LN\vec{q}+MN\vec{q}'-\vec{q}_0) = \tilde{f}(\vec{q}+\vec{q}'-\vec{q}_0), \quad (5.54)$$

and so obtain

$$|V_{fi}|^2 = \frac{\chi\hbar E_0}{2\epsilon_0}\omega_q\omega_{q'}|\tilde{f}(\vec{q}+\vec{q}'-\vec{q}_0)|^2 \times \left[T^2+R^2-2TR\cos\left(L\vec{q}\cdot\vec{\delta}(L\vec{q}) - \vec{q}\cdot\vec{\delta}\vec{q}\right)\right]. \quad (5.55)$$

Rather than re-analyse the two cases $R(\sin\theta_* + \sin\theta'_*) \gg l(\cos\theta_* - \cos\theta'_*)$ and $R(\sin\theta_* + \sin\theta'_*) \ll l(\cos\theta_* - \cos\theta'_*)$, we will assume the former to hold, in which case the light emerging from the medium in any one direction has a Gaussian spectrum. We will also integrate over all wavenumbers for the unprimed photons which enter $\mathrm{d}\Omega$ and $\mathrm{d}\Omega'$ which will be the appropriate experimentally measured quantity if our photon-counting devices have a spectral response which is unity over the frequencies emitted in their respective

directions. We look first at the case of symmetrically split pump photons for which $\vec{q}_* = \vec{q}'_*$, $\theta_* = \theta'_*$ and assume that we are looking in the optimum direction $\delta\theta' = 0$, $\delta\phi = 0$. The path difference $\vec{\delta}$ is introduced so that $\vec{\delta}(\vec{q}) = \delta\hat{q}_*$, perpendicular to the wavefront from the medium. Going through the by now familiar procedure we obtain

$$\frac{\mathrm{d}^2R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = I\omega_{q_*}^2\frac{q_*^4}{(2\pi)^6}\int \mathrm{d}(\delta q)\exp\left[-2R^2\sin^2\theta_*(\delta q)^2\right] \times\left[T^2+R^2-2TR\cos(2\delta(\delta q))\right], \tag{5.56}$$

giving

$$\frac{\mathrm{d}^2R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = R_0\left[T^2+R^2-2TR\exp\left(-\frac{\delta^2}{2R^2\sin\theta_*}\right)\right], \tag{5.57}$$

where

$$R_0 = I\omega_{q_*}^2\frac{q_*^4}{(2\pi)^6}. \tag{5.58}$$

This result agrees with 4.14.

In the case of two-colour interference we introduce path length differences of δ and δ' for photons travelling in directions defined by $\vec{q}$ and $N\vec{q}'$

$$\frac{\mathrm{d}^2R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = I\omega_{q_*}\omega_{q'_*}\frac{q^2}{(2\pi)^3}\frac{q'^2}{(2\pi)^3}\int \mathrm{d}(\delta q)\exp\left[-R^2(\sin\theta_*+\sin\theta'_*)^2(\delta q)^2\right] \times\left[T^2+R^2-2TR\cos((q'_*-\delta q)\delta'-(q_*+\delta q)\delta)\right], \tag{5.59}$$

giving

$$\frac{\mathrm{d}^2R}{\mathrm{d}\Omega\,\mathrm{d}\Omega'} = R_1\left[T^2+R^2-2TR\cos(q'_*\delta'-q_*\delta)\exp\left(-\frac{1}{2R^2}\left(\frac{\delta+\delta'}{\sin\theta_*+\sin\theta'_*}\right)^2\right)\right], \tag{5.60}$$

where

$$R_1 = I\omega_{q_*}\omega_{q'_*}\frac{q^2}{(2\pi)^3}\frac{q'^2}{(2\pi)^3}\frac{\sqrt{\pi}}{R(\sin\theta_*+\sin\theta'_*)}. \tag{5.61}$$

This agrees with equation 4.37.

5.3 Interfering-alternatives picture of fourth-order interference

We now give an analysis of fourth-order interference in which we regard the effect as due to the quantum-mechanical interference of indistinguishable alternatives. The physical processes which occur in fourth-order interference and their associated quantum-mechanical amplitudes are:

- Photon propagation. We shall consider only scalar photons for which we take the amplitude for propagation in free space between two points separated by a distance r to be proportional to $(1/r)\exp(ikr)$ for a photon of wavenumber k.

- Destruction of a photon of frequency ω_0 by a 'unit-cell' of the non-linear medium with the simultaneous creation of a pair of photons with frequencies ω_1 and ω_2 where $\omega_1 + \omega_2 = \omega_0$, the pump frequency. The amplitude for this we take to be $a(\omega)$ where $\omega = \omega_1 - \omega_2$.

- Transmission and reflection of photons at mirrors and beamsplitters. For a perfect mirror we take the amplitude for reflection to be unity. For a beamsplitter we take the amplitudes to be as indicated in figure 5.2.

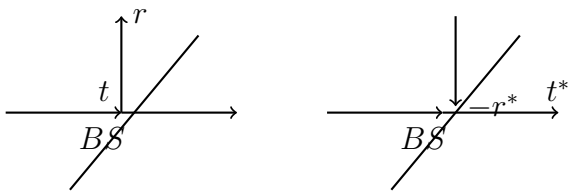

Figure 5.2: Amplitudes for photon transmission and reflection at a beamsplitter

We will firstly look at the case where a single 'unit-cell' of the non-linear medium rests in the pump beam. Calling the higher-frequency photon 'blue' and the lower-frequency one 'red' the probability of the photons scoring a coincidence count with the blue arriving at D_1 and the red at D_2 is determined by the two alternatives of figure 5.3. Taking the wavenumbers of the red and blue photons to be k_r and k_b, the total upper path length to be $l+\delta$, and that along the lower path to be l, the probability of obtaining the final state shown is given by

$$P_r^b \propto \left| a(\omega)\frac{e^{ik_r(l+\delta)}}{l+\delta}\frac{e^{ik_b l}}{l}(tt^*) + a(\omega)\frac{e^{ik_b(l+\delta)}}{l+\delta}\frac{e^{ik_r l}}{l}(-rr^*)\right|^2 \tag{5.62}$$

$$\approx \frac{|a(\omega)|^2}{l^4}\left[T^2 + R^2 - 2TR\cos(2\omega\delta/c)\right], \tag{5.63}$$

since $\delta \ll l$. Each term in equation 5.62 has a factor for the production of the pair, for the propagation of each member of the pair and for the behaviour of each member of the pair at the splitter, either transmission or reflection. Of course, if a blue photon ends by travelling up with a red one down this

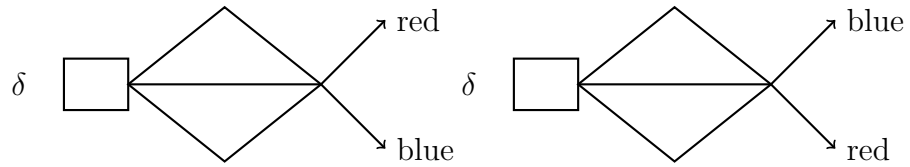

Figure 5.3: Interfering alternative paths which produce the fourth-order interference effect

will also produce a coincidence count if, as we assume, the detectors are of broad enough spectral response. The probability for this process, P_b^r, is easily shown to be $P_b^r = P_r^b$. As these two final states are in principle distinguishable there is no quantum-mechanical interference between them and to obtain the probability for either of these two processes we add the probabilities for each of them separately. Similarly when pairs with a range of ω's are emitted we add the probabilities for processes that contribute to coincidence counts.

$$P_c \propto \int \mathrm{d}\omega \, |a(\omega)|^2 \left(T^2 + R^2 - 2TR\cos(2\omega x/c)\right). \tag{5.64}$$

This starts to look very much like the expressions we have already seen for fourth-order interference patterns.

In fact we have cheated slightly in that a unit cell source can emit pairs with both photons travelling by the upper path or both by the lower, as we do not have the constraint of phase-matching that arises with an extended source. As a consequence there are in fact four indistinguishable alternatives to take into account. We chose to neglect the two in which the pairs travel by the same path so that a direct comparison of the formulae for the single-atom and extended-medium cases is possible.

5.3.1 Extended source of pairs: phase-matching

We will now see how an interfering alternatives picture can lead to the idea of phase-matching in the emission of pairs from an extended source. The model we work with is the usual one of a region of non-linearity in free space, with the pump photons carrying anomalous momentum. We will as usual have the pump beam coming in along the z-axis, the amplitude for finding a pump photon at a given point being proportional to the positive-frequency part of the pump electric field there. We look for a photon of frequency ω_1 at $\vec{r}_1$ and one of frequency ω_2 at $\vec{r}_2$. The indistinguishable alternatives over which we must sum are those for the production of pairs at different points inside the

medium. Let a unit cell be positioned at a point $\vec{x}$ in the medium, referred to some origin O. The amplitude for pump-photon propagation to the point $\vec{x}$, annihilation/pair production and propagation out to $\vec{r}_1$ and $\vec{r}_2$ is given by

$$A(\vec{r}_1, \vec{r}_2) = E_0 f(x, y) e^{ik_z z} a(\omega) \frac{e^{ik_1|\vec{r}_1 - \vec{x}|}}{|\vec{r}_1 - \vec{x}|} \frac{e^{ik_2|\vec{r}_2 - \vec{x}|}}{|\vec{r}_2 - \vec{x}|}. \tag{5.65}$$

Looking in the far field of the medium so that $r_{1,2} \gg x$ we approximate $|\vec{r}_{1,2} - \vec{x}|$ by $r_{1,2} - \hat{r}_{1,2} \cdot \vec{x}$ in the exponentials and by $r_{1,2}$ in the $1/r$ dependences of the propagators.

$$\begin{aligned} A(\vec{r}_1, \vec{r}_2) &= \frac{E_0}{r_1 r_2} a(\omega) e^{ik_1 r_1 + ik_2 r_2} \int \mathrm{d}^3\vec{x}\, e^{ik_z z - i\vec{r}_1 \cdot \vec{k}_1 - i\vec{r}_2 \cdot \vec{k}_2} \\ &= \frac{E_0}{r_1 r_2} a(\omega) e^{ik_1 r_1 + ik_2 r_2} \tilde{f}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0), \end{aligned} \tag{5.66}$$

$\tilde{f}$ being the form-factor that we have already met and $\vec{k}_1$ and $\vec{k}_2$ the wavevectors of the emitted photons. For an extended medium we thus find that the amplitudes for pair emission interfere destructively except for emission near the phase-matched condition.

5.3.2 Two-colour interference experiments

We may now combine the analyses of alternative paths and pair production in different parts of the medium to analyse the two-colour interference experiment of figure 5.4. The interference is between the dotted and solid alternatives. We will assume that the lower paths have lengths r_1 and r_2 for the 'red' and 'blue' paths respectively with the upper paths having lengths $r_1 + \delta_1$ and $r_2 + \delta_2$, the paths being measured from the origin to the beam-splitter via the mirrors. The amplitude A, for obtaining a coincidence count at detectors D_1 and D_2 is given by

$$\begin{aligned} A = \frac{E_0}{r_1 r_2} a(\omega) \Big[& e^{ik_1 r_1 + ik_2 (r_2 + \delta_2)} \tilde{f}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0)(tt^*) \\ & + e^{ik_1 (r_1 + \delta_1) + ik_2 r_2} \tilde{f}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0)(-rr^*) \Big]. \end{aligned} \tag{5.67}$$

Again in the perfectly symmetric case we have the two form-factors equal, and taking the modulus squared of this amplitude and integrating over the different ω values of pairs that may be produced we obtain

$$P_c \propto \frac{E_0^2}{r_1^2 r_2^2} \int \mathrm{d}\omega\, |a(\omega)|^2 |\tilde{f}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0)|^2 \left(T^2 + R^2 - 2TR \cos(k_2 \delta_2 - k_1 \delta_1) \right). \tag{5.68}$$

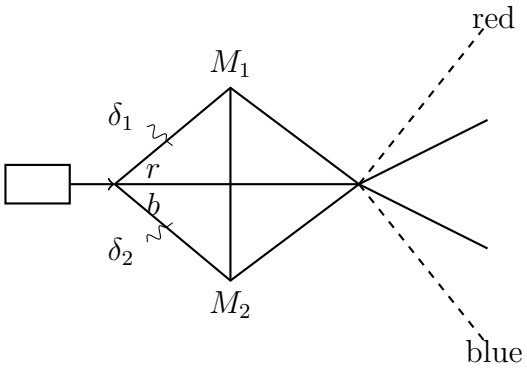

Figure 5.4: Interfering alternatives in the two-colour interference experiment

When the illuminated patch of the medium is large the form-factor is much more rapidly varying with ω than the pair production amplitude and we may take this term out as a constant factor in the integral. Substituting a familiar form-factor from chapter 5 yields the same predictions as the ones previously obtained for this experiment.

Chapter 6

Conclusions and outlook

In chapter 2 we started with a detailed investigation of the localisability of quantum-mechanical particles and gave an explicit demonstration of the impossibility of constructing a position operator for the photon. Thus if we use the word localisable as synonymous with the existence of a position operator and a wavefunction with a Born interpretation then the photon is not localisable. This does not mean, however, that it is impossible to construct excited states of the free electromagnetic field whose physical effects are localised within some definite region of space, as we showed in the thought experiment at the end of chapter 2.

In chapter 3 we investigated the theory of spontaneous parametric fluorescence in some detail and found Mollow's theory for the dispersion in photocount time differences to have a simple interpretation in terms of the travel times of individual members of the photon pair generated in different regions of the non-linear medium. The magnitude of the power-law tails in the two-photocount amplitude due to Pike and Sarkar was estimated and found to be unobservably small for the case of point detectors in the far field, and it is indeed possible that these tails are an artefact produced by making the rotating-wave approximation the theory of photodetection where this approximation does not hold good. The original interest in these tails was due to a possible link between the tails arising in spontaneous parametric fluorescence and those reported by Amrein in the energy density associated with weakly-localised single photons, but the arguments we have given concerning the production of localised disturbances of the electromagnetic field incline us to believe that the tails of Amrein have little to do with the experimental production of localised photons.

In chapter 4 we gave a recapitulation of a standard approach to the analysis of fourth-order interference experiments and applied this to an examination of how the Pike and Sarkar photodetection tails are reflected in fourth-

order interference experiments. However the curious nature of fourth-order interference effects, and doubts about the basis of Mollow's theory of two-photon detection in spontaneous parametric fluorescence (which formed the starting point for the analysis of fourth-order interference effects), motivated the alternative analyses of fourth-order interference of chapter 5. Although we deal with scalar photons and an idealised model of the non-linear medium these analyses capture the essential features of this phenomenon and could be extended to cover the more realistic case of a source for which we have to address the problem of refraction as the light leaves the medium.

The final strand of this thesis, in appendix D, concerned the quantum theory of dielectrics in which we evaluate the ground-state field fluctuations in a dielectric material modelled by a field of harmonic oscillators and find this to be incompatible with the results of Glauber and Lewenstein [11] who quantised the macroscopic Maxwell equations. The purpose of this was to demonstrate the inadequacy of the phenomenological model in describing truly quantum effects in these materials, rather than to propose a serious model of the matter that comprises a dielectric. However, this model does have some potential for future development. It could be extended to the vector electromagnetic field which would bring in the fluctuations associated with the longitudinal part of the vector and scalar potential and would allow a demonstration of how the spontaneous-emission rate of an atom placed inside a cavity in this model dielectric is modified by the surrounding material, which could be compared with the similar calculation of Glauber. The model also offers the possibility of extension to propagative problems in quantum optics, where the dielectric material only fills part of the space, and we might hope to calculate the propagation of fluctuations and squeezing through changing dielectric materials and make comparisons with the phenomenological approach.

Appendix A

Theory of induced group representations

In this appendix, following Barut and Raczka [2] and Mackey [20], we outline enough of the theory of group representations to follow the discussion of the group-theoretical approach to localisation of chapter 2. In doing so we concentrate on the algebraic aspects of representation theory, largely neglecting many topological and measure-theoretical subtleties. Nevertheless, we hope to communicate an overall structure of induced representation theory, as applied to locally compact topological groups of semi-direct product form, which may be filled in more detail by further reading.

A.1 Preliminaries

A.1.1 Group spaces

A set X with constituent elements x becomes a group space when we specify an action of the group G on X by providing a map $G \times X \to X$ satisfying

$$ex = x \tag{A.1}$$

$$(g_2 g_1)x = g_2(g_1 x) \tag{A.2}$$

for all $x \in X$, $g_1, g_2 \in G$ and where e is the identity element of G. For a given $x \in X$ the set gx for all $g \in G$ is called the *orbit* of x. Clearly, two orbits are either disjoint or equal and the set X may be partitioned into distinct orbits under the action of G. In the special case where the whole space X consists of one orbit, the action of G on X is said to be transitive.

A.1.2 Borel spaces and measures

We are going to require the notion of integration over groups G and group spaces X, and for this we need to define a measure on classes of subsets of G and X. We shall endow them with a Borel structure, and then define a Borel measure on this structure.

A Borel space $(X, \mathcal{B})$ is a set X and a family of subsets $\mathcal{B}$ of X called Borel sets satisfying:

- $\emptyset, X \in \mathcal{B}$
- $X_j \in \mathcal{B} \Rightarrow X - X_j,\ \bigcap_{j=1}^{\infty} X_j,\ \bigcup_{j=1}^{\infty} X_j \in \mathcal{B}$

Every set has at least one Borel structure. In fact, given a family F of subsets of X it is possible by countable intersections and unions to construct a unique smallest Borel structure which contains F. An important example is furnished by the set of real numbers in which, out of the many possible Borel structures, the usual one is generated by the family of all semi-closed intervals.

A Borel measure is a map $\mu : \mathcal{B} \to [0, \infty]$ satisfying:

- $\mu(X_i) \geq 0$ for $X_i \in \mathcal{B}$, $\mu(\emptyset) = 0$
- If X_i are disjoint, $\mu(\bigcup_{i=1}^{\infty} X_i) = \sum_{i=1}^{\infty} \mu(X_i)$
- There exist $X_k \in \mathcal{B}$ for $k = 1, 2, \ldots$ such that $\mu(X_k) < \infty$ and $X = \bigcup_{k=1}^{\infty} X_k$

Two measures μ and μ' are said to belong to the same *measure class* if they satisfy

$$\mu(X) = 0 \iff \mu'(X) = 0 \tag{A.3}$$

for all $X \in \mathcal{B}$.

Measures on group spaces

If a Borel space equipped with a Borel measure is also a group space then we may define the concept of invariant measure. A measure μ_i is right-invariant if

$$\mu_i(gX) = \mu_i(X), \tag{A.4}$$

where the Borel set gX is obtained by translating each element of X by the action of g. For a particular group space such a measure may or may not exist, but in the particular case in which the group space X is the group G itself then Haar's theorem assures us of the existence of a unique invariant

measure (to within a positive multiplicative constant) on the Borel sets of G. If an invariant measure on a group space is not obtainable the next best thing is a *quasi-invariant measure*. Such a measure μ_{qi} has the property that the translated measure μ'_{qi} defined by $\mu'_{qi}(X) = \mu_{qi}(gX)$ is in the same measure class as μ_{qi} for every $g \in G$.

The Radon-Nikodym theorem

A real non-negative function f on X is measurable if for $a > 0$ the set $\{x : f(x) > a\}$ is measurable. A complex function g is measurable if it may be written in the form $g = f_1 - f_2 + i(f_3 - f_4)$ with $f_1 \cdots f_4$ real measurable functions. The limit of a Lebesgue sum relative to the measure μ is called the integral $\int_X f(x)\, d\mu(x)$ of f. If $\int_X f(x)\, d\mu(x)$ is finite then f is said to be integrable. A measure ν is said to be absolutely continuous with respect to the measure μ if and only if $\mu(A) = 0 \Rightarrow \nu(A) = 0$ for all measurable sets A of X. The Radon-Nikodym theorem adds that ν is absolutely continuous with respect to μ if and only if there is a measurable function ρ such that

$$\nu(A) = \int \rho(x)\, \chi_A(x)\, d\mu(x) \tag{A.5}$$

for any measurable set A, where χ_A is the characteristic function of the set A defined by $\chi_A(x) = 1$ when $x \in A$ and zero otherwise. This theorem is often used in the differential form

$$\frac{d\nu}{d\mu}(x) = \rho(x), \tag{A.6}$$

where $\rho(x)$ is the Radon-Nikodym derivative of ν with respect to μ.

A.1.3 The spectral theorem for self-adjoint operators

Let $\lambda \in \mathbb{R}$. An operator function $E(\lambda)$ in some Hilbert space H is said to be a *resolution of the identity* if it satisfies

$$E(\lambda)^* = E(\lambda) \tag{A.7}$$
$$E(\lambda)E(\mu) = E(\min(\lambda, \mu)) \tag{A.8}$$
$$E(\lambda + 0) = E(\lambda) \tag{A.9}$$
$$E(-\infty) = 0, \quad E(\infty) = I. \tag{A.10}$$

Equations (A.7) and (A.8) (for $\lambda = \mu$) show that $E(\lambda)$ is a projection operator, and (A.8) for $\lambda < \mu$ shows that $E(\lambda)$ projects onto a subspace of that

projected onto by $E(\mu)$. Equation (A.9) demonstrates a right-continuity property of $E(\lambda)$.

With this spectral measure we now state Hilbert's spectral theorem for self-adjoint operators. Every self-adjoint operator A in H has the representation

$$A = \int_{-\infty}^{\infty} \lambda \, dE(\lambda), \tag{A.11}$$

where the spectral measure $E(\lambda)$ is uniquely determined by the operator A. The equality should be understood in the weak sense

$$(Au, v) = \int_{-\infty}^{\infty} \lambda \, d(E(\lambda)u, v) \tag{A.12}$$

for all $v \in H$ and all u in the domain of definition of A.

Projection-valued measures

From a spectral measure on the real numbers we will define a projection-valued measure defined on the Borel sets of the real line. Firstly we define projections on intervals $\Delta = [\lambda_1, \lambda_2]$ of the real line by

$$P(\Delta) = E(\lambda_2) - E(\lambda_1), \tag{A.13}$$

with the property from (A.8) that

$$P(\Delta_1)P(\Delta_2) = P(\Delta_1 \cap \Delta_2), \tag{A.14}$$

and also that

$$P(\Delta_1) + P(\Delta_2) = P(\Delta_1 \cup \Delta_2) \tag{A.15}$$

for Δ_1 and Δ_2 disjoint. From these two properties we may construct projection operators on the Borel sets of the real line by countable intersections and unions. In this way the existence of a resolution of the identity $E(\lambda)$ becomes equivalent to the existence of a projection-valued measure $P(X)$ on the Borel sets of the real line having the following properties:

- $P(\emptyset) = 0$, $P(\mathbb{R}) = I$
- $P(X_1 \cap X_2) = P(X_1)P(X_2)$
- $P(\bigcup X_i) = \sum P(X_i)$ for disjoint X_i.

The nomenclature 'projection-valued measure' arises because $(u, P(X)u)$ forms an ordinary measure on the Borel sets of the real line for all $u \in H$. The existence of a suitable projection-valued measure on the Borel sets of $\mathbb{R}^3$ is crucial to the mathematical definition of particle localisation of chapter 2.

A.1.4 Unitary group representations

Here we develop a minimal background to the theory of group representations required for the construction of induced representations of the Poincaré group.

A continuous unitary representation, U, of a group G consists of a unitary operator U_g for each $g \in G$ acting in some separable Hilbert space $H(U)$ and satisfies

$$U_{g_1} U_{g_2} = U_{g_1 g_2}, \tag{A.16}$$

with $u \to U_g u$ a continuous function of g for each fixed $u \in H(U)$.

Irreducibility and equivalence

A closed subspace M of $H(U)$ will be said to be invariant if

$$U_g \phi \in M \tag{A.17}$$

for all $g \in G$, $\phi \in M$. By restriction of the U_g's to M we obtain a subrepresentation U^M whose space is M. It is also easy to show that $M^\perp$ is also invariant and so U restricted to $M^\perp$ also forms a subrepresentation $U^{M^\perp}$. Because any vector in $H(U)$ may uniquely be expressed as a sum of one element in M and one in $M^\perp$ we say that U is the direct sum of U^M and $U^{M^\perp}$. A representation which has no invariant subspaces except for 0 and $H(U)$ is said to be *irreducible*.

Two representations U_g and V_g are said to be *equivalent*, and we regard them as mathematically the same, if there exists a bijective map $W : H(U) \to H(V)$ such that

$$V_g W = W U_g. \tag{A.18}$$

Similarly two representations are said to possess an *intertwining operator* if there exists a nonzero $T : H(U) \to H(V)$ satisfying

$$V_g T = T U_g. \tag{A.19}$$

Suppose that T has a null space $\mathcal{N}(T)$ and range $\mathcal{R}(T)$. $\mathcal{N}(T)$ is easily seen to be an invariant subspace of U and $\mathcal{R}(T)$ an invariant subspace of V. T sets up an equivalence between V restricted to $\mathcal{R}(T)$ and the representation $\tilde{U}$ in the space $H(U)/\mathcal{N}(T)$ defined by $\tilde{U}_g \tilde{u} = \widetilde{U_g u}$ where the tildes denote equivalence classes of vectors in $H(U)$ that differ by an element of $\mathcal{N}(T)$. The space $H(U)/\mathcal{N}(T)$ may be identified with the subspace $\mathcal{N}^\perp$ of $H(U)$, so this intertwining operator shows the equivalence of a subrepresentation of V to a subrepresentation of U. Conversely the existence of such an equivalence implies the existence of a non-zero intertwining operator.

We now obtain a useful criterion, Schur's second lemma, which enables us to determine whether a representation is irreducible. Consider the algebra $\mathcal{I}(U,U)$ of intertwining operators between U and U, also known as the *commuting algebra* of U. If there exists a nonzero member, S_0, of this algebra then $S_\lambda = S_0 - \lambda I$ is also a member where $\lambda \in \mathbb{C}$. Now, for some λ, S_λ must have a non-trivial null space N. If U is irreducible N must be the whole space since by definition its only non-trivial invariant subspace is $H(U)$. Thus for an irreducible representation the commuting algebra is the one-dimensional

$$S = \lambda I. \tag{A.20}$$

Representations of commutative groups

We demonstrate the important properties of the representation of commutative groups that we will use in the construction of representations of semi-direct products.

We first show that irreducible representations of commutative groups are all one-dimensional. Let U be an irreducible unitary representation of the commutative group N. For all $n_1 \in N$ and fixed $n_2 \in N$:

$$U_{n_1}U_{n_2} = U_{n_1 n_2} = U_{n_2 n_1} = U_{n_2}U_{n_1}. \tag{A.21}$$

By Schur's lemma

$$U_{n_2} = \alpha(n_2)I, \tag{A.22}$$

where $\alpha \in \mathbb{C}$, and so any one-dimensional subspace of $H(U)$ is invariant. But U is irreducible and so such a subspace must coincide with $H(U)$.

A *character* of commutative group N is a map $\hat{n} : N \to \mathbb{C}$ satisfying

$$|\hat{n}| = 1 \tag{A.23}$$

$$\hat{n}(n_1)\hat{n}(n_2) = \hat{n}(n_1 n_2). \tag{A.24}$$

It follows that $\hat{n}(e) = 1$ and $\hat{n}(n^{-1}) = \hat{n}(n)^*$, and the characters form a one-dimensional unitary representation of N. We denote the complete set of characters by $\hat{N}$. If $\hat{n}_1, \hat{n}_2 \in \hat{N}$ then $n \to (\hat{n}_1\hat{n}_2)(n) = \hat{n}_1(n)\hat{n}_2(n)$ satisfies

$$|(\hat{n}_1\hat{n}_2)(n)| = 1 \tag{A.25}$$

$$(\hat{n}_1\hat{n}_2)(n_1)\,(\hat{n}_1\hat{n}_2)(n_2) = (\hat{n}_1\hat{n}_2)(n_1 n_2), \tag{A.26}$$

and so the product of two characters is also a character. $\hat{e} = 1$ is the unit character and $\hat{n}^{-1}(n) = \hat{n}(n^{-1})$ provides the inverse character. Thus $\hat{N}$ forms an abelian group under the given composition laws.

We next state the SNAG theorem due to Stone, Naimark, Ambrose and Godement which is fundamental to the representation theory of abelian groups. Let U be a unitary continuous representation of an abelian locally compact group N in Hilbert space $H(U)$. Then there exists on the Borel sets of $\hat{N}$ a projection-valued measure E such that

$$U_n = \int_{\hat{N}} \hat{n}(n)\, dE(\hat{n}). \tag{A.27}$$

We may consider this to be a generalisation of the concept of a direct sum of irreducible group representations. Notice, in particular, that for an irreducible representation the measure is concentrated entirely on a single $\hat{n} \in \hat{N}$. A special case of the above is the Stone theorem, where $N = \mathbb{R}$, the additive group of the real numbers, for which the characters χ are also labelled by a real number s:

$$\chi_s(r) = e^{irs}. \tag{A.28}$$

Hence for any unitary representation of the additive group of the real numbers we may write

$$U_r = \int_{-\infty}^{\infty} e^{irs}\, dE(s), \tag{A.29}$$

where $E(s)$ is a spectral measure on the real numbers. This result is clearly related to the spectral theorem for self-adjoint operators. In fact the set U_r uniquely determines a self-adjoint operator H with spectral decomposition

$$H = \int_{-\infty}^{\infty} s\, dE(s), \tag{A.30}$$

and we write

$$U_r = e^{irH}. \tag{A.31}$$

The regular representation

How next are we to go about the construction of representations of a given group, which may not have the convenient property of commutativity? Suppose G acts on the group space X and we are provided with a right-invariant measure μ_i on X. Forming the Hilbert space $L^2(X, \mu_i)$ of all functions $f : X \to \mathbb{C}$ such that

$$\int |f(x)|^2\, d\mu(x) < \infty, \tag{A.32}$$

we obtain a unitary representation of G by setting

$$(U_g f)(x) = f(g^{-1}x), \tag{A.33}$$

as may instantly be verified, and with attention to its topological properties it can be shown to be continuous.

The existence of the representation hinges on the existence of the invariant measure μ. Haar's theorem assures the existence of a unique (to within a constant factor) invariant measure on G itself, and so we can construct a representation of G by setting $X = G$ and using the Haar measure; this yields what is called the *right-regular representation*.

A.2 Induced representations

The construction of induced representations is achieved by a generalisation of the method of the previous section which we elaborate in two stages. First, for motivation, we return to the representation constructed in $L^2(X, \mu_i)$ above. We suppose that G has a closed subgroup K and we obtain the right coset space G/K. The aim is to use G/K as the group space on which to build the representation. G acts on the cosets in a natural way

$$g_2(g_1K) = (g_2g_1)K, \tag{A.34}$$

but we are faced with the immediate problem that it is not in general possible to construct an invariant measure on G/K. It is possible, however, to obtain a quasi-invariant measure, and this proves sufficient to construct a unitary representation on $L^2(G/K, \mu)$ as below:

$$(U_{g_0}f)(\bar{g}) = \left(\frac{d\mu(g_0^{-1}\bar{g})}{d\mu(\bar{g})}\right)^{1/2} f(g_0^{-1}\bar{g}), \tag{A.35}$$

where $\bar{g} = gK$ and the Radon-Nikodym derivative compensates for the non-invariance of the measure.

The full construction of induced representations is more complicated. We start with a unitary representation L_k of K in a Hilbert space $H(L)$. The space in which the operators of the induced representation will act is $L^2(G/K, H(L), \mu)$, the space of vector-valued functions $u : G/K \to H(L)$ which satisfy

$$\int_{G/K} (u(\bar{g}), u(\bar{g}))\, d\mu(\bar{g}) < \infty, \tag{A.36}$$

which is a natural generalisation of the $L^2(G/K, \mu)$ that we used previously. Then we choose a representative member of each coset so that we may write each element of G as

$$g = s(\bar{g})\kappa(g), \tag{A.37}$$

where $\kappa(g) \in K$ and $s(\bar{g})$ is the standard member of each coset to which g belongs. The action of U^L, the representation induced by L, is then given by

$$U^L_{g_0} u(\bar{g}) = \left(\frac{d\mu(g_0^{-1}\bar{g})}{d\mu(\bar{g})} \right)^{1/2} L_{\kappa(g_0^{-1}g)}\, u(g_0^{-1}\bar{g}), \tag{A.38}$$

which is easily seen to have the required properties of a representation.

A.2.1 The imprimitivity theorem

Along with every induced representation, a (so-called canonically associated) projection-valued measure acting in $L^2(G/K, H(L), \mu)$ may be defined on the Borel sets of G/K:

$$(E(S)u)(\bar{g}) = \chi_S(\bar{g})\, u(\bar{g}), \tag{A.39}$$

where $\chi_S(\bar{g})$ is the characteristic function of the Borel set S, defined as unity when $\bar{g} \in S$ and zero otherwise. It is simple to verify that these projections obey the following commutation relations with the operators U_g of the induced representation:

$$E(S)U_{g_0} = U_{g_0} E(g_0^{-1}S), \tag{A.40}$$

where $g^{-1}S$ is the Borel set obtained by translating each element of S by g^{-1}. A projection-valued measure bearing this relationship with a group representation is known as a *system of imprimitivity* for the group representation.

Mackey's imprimitivity theorem [20] proves a converse result: given a representation U of a group G with closed subgroup K and a projection-valued measure E on G/K satisfying the commutation relations (A.40), then there exists a representation L of K such that the pair U, E is equivalent to the pair U^L, E^L consisting of the representation U^L induced by L and the canonically associated system of imprimitivity.

A.2.2 Induced representations of semi-direct products

Our aim here is to demonstrate a method by which a complete set of irreducible representations of a group of semi-direct product form may be constructed. We present it as a recipe, without justification, but at the heart of the proof are the SNAG and imprimitivity theorems. The interested reader is urged to consult Barut and Raczka [2] if the omission is not acceptable.

Suppose we are given two topological groups N and H and a homomorphism $h \to \alpha_h$ of H into the group of all automorphisms of N. We define a group called the *semi-direct product* of N and H with respect to α, denoted

by $N \times_\alpha H$, as the group whose elements are the ordered pairs (n, h) with the multiplication law

$$(n_1, h_1)(n_2, h_2) = (n_1\, \alpha_{h_1}(n_2),\, h_1 h_2). \tag{A.41}$$

We will only consider the case of N commutative. First we form the set of characters $\hat{N}$ and make $\hat{N}$ into a group space by defining the following action of H:

$$(h\hat{n})(n) = \hat{n}(\alpha_h n), \tag{A.42}$$

since $\hat{n}(\alpha_h n)$ is easily shown to be a character.

The whole set of characters is thus partitioned into separate orbits under the action of H. The procedure for the construction of irreducible representations of $N \times_\alpha H$ starts by taking, for each separate character, the set of all $h \in H$ such that

$$h\hat{n} = \hat{n}. \tag{A.43}$$

This set forms a subgroup of H and is called the *little group* of H at $\hat{n}$, labelled $H_{\hat{n}}$. If we are then given an irreducible representation of $H_{\hat{n}}$, say $V^{\hat{n}}$, we may then construct an irreducible representation W of $N \times_\alpha H_{\hat{n}}$ according to

$$W(n, h_x) = \hat{n}(n)\, V^{\hat{n}}(h_x), \tag{A.44}$$

and then we may construct an irreducible representation of $N \times_\alpha H$ as an induced representation from this representation of $N \times_\alpha H_{\hat{n}}$.

It may be shown that if $\hat{n}$ and $\hat{n}'$ lie on the same orbit then the two representations obtained by the above procedure are equivalent, so it suffices to choose one $\hat{n}$ from each orbit for the construction of inequivalent representations. Further, if we can choose so that the set of $\hat{n}$'s chosen form a Borel set in $\hat{N}$ then the set of induced irreducible representations encompasses all irreducible representations.

A.3 Irreducible representations of the Poincaré group

Mathematically, the Poincaré group, P, is the set of transformations on four-vectors x^μ, $\mu = 1 \cdots 4$:

$$x^\mu \to \Lambda^\mu{}_\nu x^\nu + a^\mu, \tag{A.45}$$

where a^μ specifies a space-time translation and $\Lambda^\mu{}_\nu$ is a Lorentz transformation which satisfies

$$\Lambda^\mu{}_\rho\, \Lambda^\nu{}_\sigma\, g^{\rho\sigma} = g^{\mu\nu}, \tag{A.46}$$

where $g^{\mu\nu}$ is the metric tensor with non-vanishing components

$$g^{00} = 1, \quad g^{11} = g^{22} = g^{33} = -1, \tag{A.47}$$

all other components vanishing. The set of transformations (a, Λ) does indeed form a group with composition law

$$(a, \Lambda)(a', \Lambda') = (a + \Lambda a', \, \Lambda\Lambda') \tag{A.48}$$

and is thus seen to be a semi-direct product of the four-dimensional translation group T^4 with the group of Lorentz transformations L. We will be particularly interested in the subgroup of the Poincaré group formed from the semi-direct product $T^4 \times_\alpha L_0$ where L_0 is the group of proper orthochronous Lorentz transformations, which satisfy

$$\Lambda^0{}_0 \geq 1, \quad \det \Lambda = 1, \tag{A.49}$$

and is labelled P_0. This subgroup is the one that is continuously connected to the identity and does not involve space reflections nor time reversal.

In quantum theory the symmetry of nature under change of coordinate system is manifested by the existence of a set of unitary operators implementing an automorphism on the system's Hilbert space of states

$$|\psi\rangle \to U(a, \Lambda)|\psi\rangle. \tag{A.50}$$

In order that the transformation (a, Λ) followed by (a', Λ') be physically equivalent to $(a + \Lambda a', \Lambda\Lambda')$ we must have

$$U(a, \Lambda)U(a', \Lambda') = \omega[(a, \Lambda), (a', \Lambda')] \, U(a + \Lambda a', \Lambda\Lambda'), \tag{A.51}$$

where ω is a complex number of modulus unity. In this way $U(a, \Lambda)$ is said to form a *projective unitary representation* of P_0. We cannot in general set $\omega = 1$ and obtain a faithful representation as the multiplication of all the vectors in the system's space of states by a complex number of modulus unity does not affect the physics. For P_0 we can however deal with double-valued representations $\omega = \pm 1$, the double-valuedness arising out of the necessity for double-valuedness in the representation of SO_3, the rotations subgroup of L_0.

We now consider $SL(2, \mathbb{C})$, the universal covering group for L_0. $SL(2, \mathbb{C})$ is the multiplicative group of 2×2 complex matrices with unit determinant. We may define a group action of $SL(2, \mathbb{C})$ on the set of 2×2 Hermitian matrices by

$$\bar{A} : X \mapsto AX\bar{A}^\dagger, \tag{A.52}$$

where $\bar{A} \in SL(2,\mathbb{C})$ and X is 2×2 Hermitian. The set of 2×2 Hermitian matrices may be put in 1:1 correspondence with the set of four-vectors x^μ by

$$X = x^\mu \sigma_\mu, \tag{A.53}$$
$$x^\mu = \tfrac{1}{2}\mathrm{Tr}(X\bar{\sigma}^\mu), \tag{A.54}$$

where $\sigma_\mu = (\sigma_0, \vec{\sigma})$, $\bar{\sigma}^\mu = \sigma^\mu$, so that

$$X = \begin{pmatrix} x^0 + x^3 & x^1 - ix^2 \\ x^1 + ix^2 & x^0 - x^3 \end{pmatrix}. \tag{A.55}$$

Now $x^\mu x_\mu = \det X = \det(AX\bar{A}^\dagger) = \det X' = x'^\mu x'_\mu$, so the magnitude of the four-vector associated with the Hermitian matrix is invariant under the action of $SL(2,\mathbb{C})$. Therefore each $\bar{A} \in SL(2,\mathbb{C})$ may be associated with a Lorentz transformation Λ, and more specifically because $SL(2,\mathbb{C})$ is simply connected and the maps $\bar{A} \to \Lambda^0{}_0$, $\bar{A} \to \det\Lambda$ are continuous, each element of $SL(2,\mathbb{C})$ is associated with a member of L_0. Explicitly the map $SL(2,\mathbb{C}) \to L_0$ is given by

$$\Lambda^\mu{}_\nu = \tfrac{1}{2}\mathrm{Tr}(\bar{A}\sigma^\mu\bar{A}^\dagger\bar{\sigma}_\nu), \tag{A.56}$$

a simple consequence of equations (A.52) and (A.55). The map is easily seen to be a 2:1 homomorphism, the elements $\pm\bar{A}$ of $SL(2,\mathbb{C})$ giving rise to the same Lorentz transformation.

We denote the semi-direct product of T^4 with $SL(2,\mathbb{C})$ by $\tilde{P}_0$. It is possible to show that every possibly double-valued representation of P_0 arises from a true representation of $\tilde{P}_0$ and for this reason we now restrict our attention to $\tilde{P}_0$.

A.3.1 Irreducible representations of the Poincaré group

We are now able to use the theory we developed for the irreducible representations of semi-direct products to construct the irreducible representations of $\tilde{P}_0$.

The characters associated with the T^4 subgroup may be labelled with a four-vector p (dropping Greek indices for convenience):

$$\chi_p(a) = e^{ip\cdot a}. \tag{A.57}$$

We then define the group action of $SL(2,\mathbb{C})$ on the characters:

$$(\bar{A}\chi_p)(a) = \chi_p(\Lambda a) = \chi_{\Lambda^{-1}p}(a), \tag{A.58}$$

where Λ is the Lorentz transformation associated with $\bar{A}$ by equation (A.56). Due to the Lorentz invariance of $p \cdot p$ for all four-vectors and the sign of p^0 for time-like and light-like four-vectors there are six kinds of orbits for these characters under the action of $SL(2, \mathbb{C})$:

- $\hat{O}_m^+$: p time-like, $p^0 > 0$.
  These orbits consist of four-vectors on the positive mass hyperboloid, i.e. four-vectors of the form $(E, \vec{p})$ with $E = \sqrt{m^2 + p^2}$.

- $\hat{O}_m^-$: p time-like, $p^0 < 0$.
  These orbits consist of four-vectors on the negative mass hyperboloid, i.e. four-vectors of the form $(E, \vec{p})$ with $E = -\sqrt{m^2 + p^2}$.

- $\hat{O}_0^+$: p light-like, $p^0 > 0$.
  These orbits consist of four-vectors on the forward light cone, i.e. four-vectors of the form $(p, \vec{p})$.

- $\hat{O}_0^-$: p light-like, $p^0 < 0$.
  These orbits consist of four-vectors on the backward light cone, i.e. four-vectors of the form $(-p, \vec{p})$.

- $\hat{O}_{im}$: p space-like.

- $\hat{O}_0^0$: p vanishing.

All these orbits can be used to generate irreducible representations of $\tilde{P}_0$, but only the representations generated by $\hat{O}_m^+$ and $\hat{O}_0^+$ have any physical meaning, corresponding to positive-energy massive and massless particles respectively. We accordingly concentrate exclusively on these two cases.

A.3.2 Massive case

We use our freedom to choose the stabilised character $\dot{p}$ from the $\hat{O}_m^+$ orbit to select $\dot{p} = (m, \vec{0})$. It is clear that the little group for this character is $SU(2)$, the covering group of the group of rotations in three-dimensional space.

The irreducible representations of $SU(2)$ are very familiar from the theory of angular momentum. They are conventionally labelled $D^j(A)$, where $A \in SU(2)$ and j is an integer or half integer, and exist in a $2j + 1$ dimensional Hilbert space $H(D^j)$.

The irreducible representation of $T^4 \times_\alpha SU(2)$ which we will use to induce a representation of $\tilde{P}_0$ is therefore

$$U(a, A) = e^{i\dot{p}\cdot a}\, D^j(A). \tag{A.59}$$

We next need a Mackey decomposition of $\tilde{P}_0$ in the form $g = s(\bar{g})\kappa(g)$ where $\kappa(g) \in T^4 \times_\alpha SU(2)$ and the $s(\bar{g})$ form a Borel set in $\tilde{P}_0$. We will demonstrate two ways in which this may be achieved, the canonical and helicity methods.

In the canonical method we use the polar decomposition of $\bar{A} \in SL(2,\mathbb{C})$ to write $\bar{A} = \lambda A$ where $A \in SU(2)$ and λ is the exponential of a traceless Hermitian matrix which physically represents a pure boost with no rotation. This leads immediately to a Mackey decomposition

$$(a, \bar{A}) = (0, \lambda(\bar{A}))\,(\lambda^{-1}(\bar{A})a, A(\bar{A})), \tag{A.60}$$

so that the coset that $(a, \bar{A})$ belongs to is labelled by the boost λ. However, we find it convenient to label the cosets by a four-momentum p on the mass hyperboloid, there being a 1:1 map between such momenta and boost matrices λ such that the boost λ_p boosts $(m, \vec{0})$ out to $(E, \vec{p})$. On these cosets we can define an invariant measure

$$d\mu_m = \frac{d^3\vec{p}}{E}. \tag{A.61}$$

Applying equation (A.38) to $\tilde{P}_0$ we choose $g = \lambda_p$ and $\kappa(g)$ is the identity element. Taking $g_0 = (a, \bar{A})$ we obtain the Mackey decomposition of $g_0^{-1}g$:

$$(a, \bar{A})^{-1}(0, \lambda_p) = (0, \lambda_{\Lambda^{-1}p})\,(-\lambda^{-1}_{\Lambda^{-1}p}\Lambda^{-1}a,\ \lambda^{-1}_{\Lambda^{-1}p}\bar{A}^{-1}\lambda_p), \tag{A.62}$$

where again Λ is the Lorentz transformation associated with $\bar{A}$. This decomposition leads to

$$\kappa^{-1}(g_0^{-1}g) = \lambda^{-1}_{\Lambda^{-1}p}\bar{A}^{-1}\lambda_p, \tag{A.63}$$

and then using equation (A.60) leads to the required irreducible representation of $\tilde{P}_0$:

$$U^{m,+,j}(a, \bar{A})\,\phi(p) = e^{ip\cdot a}\,D^j\left(\lambda^{-1}_{\Lambda^{-1}p}\bar{A}\lambda_{\Lambda^{-1}p}\right)\phi(\Lambda^{-1}p). \tag{A.64}$$

In the helicity method, useful when comparing the representations for massive and massless particles, the Mackey decomposition is performed differently. Instead of choosing $s(\bar{g})$ so that it performs a boost directly from $(m, \vec{0})$ to $(E, \vec{p})$, we choose it as the product of a boost λ_{p_s} which takes $(m, \vec{0})$ to $(E, 0, 0, p)$ and $A_{\vec{p}} \in SU(2)$ whose associated rotation takes $(E, 0, 0, p)$ onto $(E, \vec{p})$:

$$\lambda^h_p = A_{\vec{p}}\,\lambda_{p_s}, \tag{A.65}$$

giving an explicit representation the same as (A.64) but with λ replaced by λ^h.

A.3.3 Massless particles

The character $\dot{p}_0$ from $\hat{O}_0^+$ that we stabilise is $\dot{p}_0 = (1, 0, 0, 1)$. If we denote a general member of $SL(2, \mathbb{C})$ by

$$\bar{A} = \begin{pmatrix} \alpha & \beta \\ \gamma & \delta \end{pmatrix}, \tag{A.66}$$

where $\alpha, \beta, \gamma, \delta \in \mathbb{C}$ and $\alpha\delta - \beta\gamma = 1$, the stability group is given by members of $SL(2, \mathbb{C})$ such that

$$\begin{pmatrix} \alpha & \beta \\ \gamma & \delta \end{pmatrix} \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \alpha^* & \gamma^* \\ \beta^* & \delta^* \end{pmatrix} = \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix}, \tag{A.67}$$

giving elements of the form

$$(z, \theta) = \begin{pmatrix} e^{i\theta/2} & e^{-i\theta/2} z \\ 0 & e^{-i\theta/2} \end{pmatrix}, \tag{A.68}$$

where z is arbitrary complex and $\theta \in [0, 2\pi)$. This group may be seen to be isomorphic to a semi-direct product of the two-dimensional translation group $T^2 = (z, 0)$ with the group of rotations in the plane SO_2. However, since $(0, 2\pi) \neq (0, 0)$, this semi-direct product does not form the Euclidean group of the plane, E_2, but a two-fold covering of it. We shall refer to it as $\tilde{E}_2$.

We may again use the theory of induced representations of semi-direct products to construct the irreducible representations of the $\tilde{E}_2$ little group. We form the set of characters $\widehat{T^2} = \chi_{\tilde{z}}$ labelled by a complex number $\tilde{z}$:

$$\chi_{\tilde{z}}(z) = e^{i(\bar{\tilde{z}}z + \tilde{z}\bar{z})}, \tag{A.69}$$

and the group action of SO_2 on these is then

$$R(\theta)\chi_{\tilde{z}}(z) = \chi_{e^{-i\theta}\tilde{z}}(z). \tag{A.70}$$

The character orbits under the action of SO_2 are the sets $|\tilde{z}| = \text{const}$ and the little groups are of two types. For $\tilde{z} = 0$ the little group is SO_2 in full, and for $\tilde{z} \neq 0$ the characters are invariant only under the identity element of SO_2. The latter lead to the representations of *continuous spin*, discovered by Wigner, which have no known physical meaning. Therefore we concentrate on the representations of SO_2 which are single- or double-valued (since our final representations of $\tilde{P}_0$ must have this property). These are of the form

$$V(\theta) = e^{is\theta} \tag{A.71}$$

for $s = 0, 1/2, 1, \ldots$, leading to the representation of $\tilde{E}_2$:

$$V^0(z, \theta) = e^{is\theta}, \tag{A.72}$$

and then to the representation W of $T^2 \times_\alpha \tilde{E}_2$:

$$W(a, (z, \theta)) = e^{i\dot{p}_0 \cdot a}\, e^{is\theta}, \tag{A.73}$$

from which the final representation of $\tilde{P}_0$ may be induced.

Assume that under the action of $\bar{A} \in SL(2, \mathbb{C})$ the stabilised vector $\dot{p}_0 = (1, 0, 0, 1)$ is taken out to $(p, \vec{p})$. The Mackey decomposition of $T^4 \times_\alpha SL(2, \mathbb{C})$ may this time be performed using

$$\bar{A} = \nu_p\, \bar{\varepsilon}, \tag{A.74}$$

where ν_p is the product of a boost taking $(1, 0, 0, 1)$ out to $(p, 0, 0, p)$ with a rotation taking $(p, 0, 0, p)$ to $(p, \vec{p})$. We may again label the cosets by a four-vector $(p, \vec{p})$ on the light cone, on which the invariant measure

$$d\mu_0 = \frac{d^3\vec{p}}{p} \tag{A.75}$$

exists, and following the same procedure as for the massive case leads to the following irreducible representations of $\tilde{P}_0$:

$$U^{0,+,s}(a, \bar{A})\, f(p) = e^{ip\cdot a}\, \exp\bigl(is\theta(\nu_p^{-1} \bar{A} \nu_{\Lambda^{-1}p})\bigr)\, f(\Lambda^{-1}p). \tag{A.76}$$

Appendix B

Type-I phase-matching in uniaxial media

The emission from a non-linear medium emitting spontaneous parametric fluorescence is said to lie on the phase-matching surface when

$$\begin{aligned} \omega_1 + \omega_2 &= \omega_0 \\ \vec{k}_1 + \vec{k}_2 &= \vec{k}_0. \end{aligned} \tag{B.1}$$

ω_1 and ω_2 are the frequencies of the fluorescent photons, $\vec{k}_1$ and $\vec{k}_2$ their wavevectors and ω_0 and $\vec{k}_0$ those of the pump. Normally the refractive index of a dielectric substance increases with frequency which prevents the conditions (B.1), even for a photon pair travelling colinearly with the pump. It is however possible to exploit an anisotropic refractive index to make phase-matching possible, the principle being to put the pump in a mode with low refractive index and allow the fluorescent photons to enter modes of a high refractive index.

We shall consider the case in which the medium has a uniaxial non-linear susceptibility

$$\epsilon = \epsilon_0\left(\epsilon\hat{e}_1^2 + \epsilon\hat{e}_2^2 + \epsilon'\hat{e}_3^2\right), \tag{B.2}$$

where $\hat{e}_1$, $\hat{e}_2$ and $\hat{e}_3$ form a right-handed orthonormal set and $\hat{e}_3$ is the direction of the optic axis. In the absence of free charges and currents, the

Maxwell and constitutive equations in Fourier space are

$$\vec{k} \cdot \vec{D} = 0 \tag{B.3}$$

$$\vec{k} \cdot \vec{B} = 0 \tag{B.4}$$

$$\vec{k} \times \vec{H} = -\omega \vec{D} \tag{B.5}$$

$$\vec{k} \times \vec{E} = \omega \vec{B} \tag{B.6}$$

$$\vec{D} = \epsilon \vec{E} \tag{B.7}$$

$$\vec{B} = \mu_0 \vec{H}. \tag{B.8}$$

We evaluate the normal modes for a plane-wave with wavevector $\vec{k}$ which makes an angle θ with the optic axis:

$$\vec{k} = \sin\theta\, \hat{e}_1 + \cos\theta\, \hat{e}_3. \tag{B.9}$$

From B.3, B.4 and B.5 combined with B.8 we note that $\vec{k}$, $\vec{D}$ and $\vec{B}$ are mutually orthogonal whereas in free space we would have $\vec{k}$, $\vec{E}$ and $\vec{B}$ orthogonal. One normal mode has

$$\vec{D} = D\hat{e}_2 \tag{B.10}$$

$$\vec{B} = B(-\cos\theta\, \hat{e}_1 + \sin\theta\, \hat{e}_3), \tag{B.11}$$

for which equations B.5 to B.8 give the dispersion relation

$$\frac{ck}{\omega} = \frac{1}{n_o}, \tag{B.12}$$

where

$$n_o = \sqrt{\epsilon}. \tag{B.13}$$

This is a mode of ordinary polarisation with refractive index n_o. The other normal mode has

$$\vec{D} = D(\cos\theta\, \hat{e}_1 - \sin\theta\, \hat{e}_3) \tag{B.14}$$

$$\vec{B} = B\hat{e}_2, \tag{B.15}$$

for which the refractive index is given by

$$n_e = \left(\frac{\cos^2\theta}{\epsilon} + \frac{\sin^2\theta}{\epsilon'} \right)^{-1/2}. \tag{B.16}$$

This is a mode of extraordinary polarisation. We can achieve so-called type-I phase-matching by choosing a material with $\epsilon' < \epsilon$ and introducing the pump in a mode of extraordinary polarisation.

We will investigate the phase-matching conditions in the case where the refractive index experienced by a photon depends only on its polarisation and not its frequency. The momenta of the fluorescent photons are indicated in figure B.1. The pump momentum is $\overrightarrow{OP}$ and those of the fluorescent photons are $\overrightarrow{OQ}$ and $\overrightarrow{QP}$. It is immediately clear that $OQ + OP = \text{const}$, as a consequence of energy conservation, so the locus of Q is an ellipse. It is also simple to show that

$$\frac{\omega_0}{2c}\left(\frac{n_o^2 - n_e^2}{n_o}\right)(OP)^{-1} = 1 - \left(\frac{n_e}{n_o}\right)\cos\theta, \tag{B.17}$$

this being the polar equation of an ellipse with eccentricity n_e/n_o and semi-latus rectum l given by

$$l = \frac{\omega_0}{2c}\left(\frac{n_o^2 - n_e^2}{n_o}\right). \tag{B.18}$$

Of course this is a highly simplified picture, but it is the one relevant to most of the calculations we have done so far.

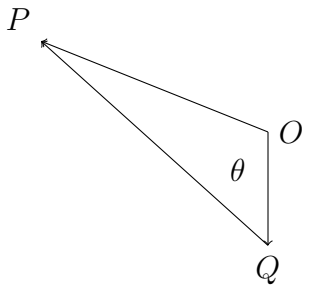

Figure B.1: Photon momenta in type I phase-matching.

Appendix C

Quantum theory of beamsplitters

The theory of beamsplitters has been given by several authors [4, 23, 27], and we here develop it in a form suitable for our analysis of fourth-order interference experiments. A fundamental approach would of necessity involve a microscopic model of the material composing the splitter, but we shall follow the approach generally accepted in quantum optics of regarding the splitter as a device which performs a linear transformation of the incoming field to produce the outgoing field so that, classically, the energy of the field is conserved, and, quantum-mechanically, the total number of photons in the field is conserved.

We assume the beamsplitter to be in a plane containing the point with position vector $\vec{r}_0$ and normal $\hat{I}$. Such a device mixes excitations of the field with wavevectors $\vec{k}$ and $\vec{k}'$ where $\vec{k}$ and $\vec{k}'$ are related by reflection in the plane of the mirror:

$$\vec{k}' = L\vec{k} = \vec{k} - 2(\vec{k} \cdot \hat{I})\hat{I}. \tag{C.1}$$

Suppose that two plane-waves are incident on the mirror, with positive-frequency parts given by

$$A^{+\,\mathrm{in}}_{\vec{k}}(\vec{r}, t) = A^{+}_{\vec{k}}\, e^{i(\vec{k}\cdot\vec{r} - \omega t)}. \tag{C.2}$$

Although we are not actually going to analyse the splitter in terms of its physical properties (its dielectric constant and thickness), such a procedure classically involves the application of boundary conditions. Therefore we apply similar conditions on the fields at the surface of the splitter to relate the outgoing fields:

$$A^{+\,\mathrm{out}}_{\vec{k}}(\vec{r}, t) = A^{+}_{\vec{k}}\, e^{i(\vec{k}\cdot\vec{r} - \omega t + i\phi)}. \tag{C.3}$$

These relationships ensuring the conservation of energy. We thus obtain the relationships between the Fourier components of the ingoing and outgoing fields:

$$A^{+\,\mathrm{out}}_{\vec{k}}(\vec{r}_0,t) = t\,A^{+\,\mathrm{in}}_{\vec{k}}(\vec{r}_0,t) + A^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r}_0,t) \tag{C.4}$$

$$A^{+\,\mathrm{out}}_{\vec{k}'}(\vec{r}_0,t) = -A^{+\,\mathrm{in}}_{\vec{k}}(\vec{r}_0,t) + t\,A^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r}_0,t) \tag{C.5}$$

to the ingoing fields at the splitter:

$$A^{+\,\mathrm{out}}_{\vec{k}}(\vec{r},t) = A^{+\,\mathrm{out}}_{\vec{k}}\,e^{-i\omega t+i\vec{k}\cdot\vec{r}} \tag{C.6}$$

$$A^{+\,\mathrm{out}}_{\vec{k}'}(\vec{r},t) = A^{+\,\mathrm{out}}_{\vec{k}'}\,e^{-i\omega t+i\vec{k}'\cdot\vec{r}} \tag{C.7}$$

Moving to the quantum theory of the splitter we simply replace expressions for the classical electric field by its quantum counterpart. By writing the positive frequency part of the electric field

$$\hat{A}^{+\,\mathrm{out}}_{\vec{k}}(\vec{r},t) = \int \frac{d^3k}{(2\pi)^3}\left(\frac{\hbar}{2\epsilon_0\omega}\right)^{1/2} a^{\mathrm{out}}_{\vec{k}}\,e^{i\vec{k}\cdot\vec{r}-i\omega t}, \tag{C.8}$$

we may regard the beamsplitter as causing the following mode transformations:

$$a^{\mathrm{out}}_{\vec{k}}(k) = t\,a^{\mathrm{in}}_{\vec{k}}(k) + r\,e^{i(\vec{k}'-\vec{k})\cdot\vec{r}_0}\,a^{\mathrm{in}}_{\vec{k}'}(k) \tag{C.9}$$

$$a^{\mathrm{out}}_{\vec{k}'}(k) = -r^*\,e^{-i(\vec{k}'-\vec{k})\cdot\vec{r}_0}\,a^{\mathrm{in}}_{\vec{k}}(k) + t\,a^{\mathrm{in}}_{\vec{k}'}(k), \tag{C.10}$$

or, alternatively,

$$a^{\mathrm{out}}_{\vec{k}}(k) = t\,a^{\mathrm{in}}_{\vec{k}}(k) + r\,e^{-2i(\vec{k}\cdot\hat{I})(\vec{r}_0\cdot\hat{I})}\,a^{\mathrm{in}}_{\vec{k}'}(k) \tag{C.11}$$

$$a^{\mathrm{out}}_{\vec{k}'}(k) = -r^*\,e^{2i(\vec{k}\cdot\hat{I})(\vec{r}_0\cdot\hat{I})}\,a^{\mathrm{in}}_{\vec{k}}(k) + t\,a^{\mathrm{in}}_{\vec{k}'}(k), \tag{C.12}$$

which are necessary for the analysis of fourth-order interference in chapter 5. If the mirror is non-dispersive so that t and r are independent of frequency, simple manipulations lead to

$$\hat{A}^{+\,\mathrm{out}}_{\vec{k}}(\vec{r},t) = t\,\hat{A}^{+\,\mathrm{in}}_{\vec{k}}(\vec{r},t) + \hat{A}^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r}',t) \tag{C.13}$$

$$\hat{A}^{+\,\mathrm{out}}_{\vec{k}'}(\vec{r},t) = -\hat{A}^{+\,\mathrm{in}}_{\vec{k}}(\vec{r}',t) + t\,\hat{A}^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r},t) \tag{C.14}$$

where $\vec{r}'$ is the image point of $\vec{r}$, for $\vec{r}$ on one side of the mirror, and

$$\hat{A}^{+\,\mathrm{out}}_{\vec{k}}(\vec{r},t) = t\,\hat{A}^{+\,\mathrm{in}}_{\vec{k}}(\vec{r}',t) + \hat{A}^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r},t)$$

$$\hat{A}^{+\,\mathrm{out}}_{\vec{k}'}(\vec{r},t) = -\hat{A}^{+\,\mathrm{in}}_{\vec{k}}(\vec{r},t) + t\,\hat{A}^{+\,\mathrm{in}}_{\vec{k}'}(\vec{r}',t)$$

for $\vec{r}$ on the other side. In this form the relationships are used in the analysis of fourth-order interference in chapter 4.

Appendix D

Ground-state quantum fluctuations in a scalar Hopfield dielectric

D.1 Introduction

To describe the effects of dielectric materials in quantum optics two different approaches may be adopted. The first is to start from a Lagrangian density including radiation, matter and interaction terms and quantise the system by the usual methods, as was done by Hopfield [13] in his investigation of the contribution of excitons to the complex dielectric constant. Excepting the usual pathologies of quantum field theory, no one would doubt that this procedure is capable of answering any question about the matter–radiation system if one is prepared to work hard enough to solve the resulting equations. The other, which is suitable if interest is principally in the type of radiation system, is to take care of it by a diagonalisation system which classically can be taken care of by a matter–radiation interaction which classically yields the macroscopic Maxwell equations [12].

In the linear case, these equations involve the dielectric displacement electric constant, and which is more usual in quantum optics is to start from a Lagrangian which yields the macroscopic Maxwell equations:

$$\mathcal{L} = \frac{1}{2} \int d^3r \, (\epsilon \vec{E}^2 - \vec{B}^2). \tag{D.1}$$

In a recent paper, Glauber and Lewenstein [11] investigate the consequences of quantising the dielectric displacement and the magnetic field fluctuations and squeezing of the dielectric displacement of dielectric constant for a uniform substance of dielectric constant.

D.2 Fluctuations predicted by the macroscopic Maxwell equations

$\vec{D} = \epsilon_0 \epsilon \vec{E}$, ϵ being the dielectric constant of the material of our system — so take into account from the start the linear interaction of radiation with matter. We then have a framework in which the only matter we have to take care of explicitly is that external to the dielectric, the so-called free charges and currents.

The question to which this paper is addressed is whether these two approaches can be expected to give compatible predictions. It would be convenient if, under conditions where light and matter are undergoing linear interaction, we could use the latter method, as it promises to deliver results with much less effort than working with matter and radiation as coupled dynamical systems.

Where, if anywhere, do we expect the difference between these two formalisms to be apparent? One possible area is in the predicted ground-state field fluctuations. It is to be expected that the zero-point current fluctuations of the matter to make a contribution to the field fluctuations that is not predicted by the quantised macroscopic Maxwell equations. To demonstrate this point explicitly, we will demonstrate a disagreement between the fluctuations predicted by the quantised macroscopic Maxwell equations with those expected in an exact solvable model of radiation–matter interaction.

The essential matters of principle can be demonstrated for a single mode of the radiation for which they take the coordinate to be

$$q = A/\omega \quad (= B/\omega) \tag{D.2}$$

where ω is the free-space frequency of a mode with the same wavenumber, dq/dt is proportional to the electric field strength,

$$\frac{dp}{dq} = \dot{q} = B = -\frac{c}{V}\frac{dq}{dt} \tag{D.3}$$

and the Lagrangian for the field mode becomes

$$\mathcal{L} = \frac{1}{2}\left(\frac{\partial q}{\partial t}\right)^2 \frac{1}{2}\omega^2 q^2 \tag{D.4}$$

The canonical momentum is

$$p = \frac{\partial \mathcal{L}}{\partial(\partial q/\partial t)} = \frac{\partial q}{\partial t} \tag{D.5}$$

They conclude that the frequency of the mode is changed from ω to $\epsilon' = \omega/\sqrt{\epsilon}$, this being the modification classically described by the refractive index. As for the manner in which the fluctuations are modified by the presence of matter, we have, in the ground state of the dielectric

The fluctuations in the magnetic field are

$$\langle (\Delta E)^2 \rangle = \frac{1}{2} \frac{\omega}{\epsilon_0 V} \tag{D.6}$$

$$\langle (\Delta B)^2 \rangle = \frac{1}{2} \frac{\omega}{\epsilon_0 V} \tag{D.7}$$

and we shall now go on to investigate the fluctuations predicted by a simple model for the linear interaction of radiation and matter.

D.3 Fluctuations predicted by the scalar Hopfield model

To investigate what the results of a fundamental approach might be, a model which is simply a scalar version of Hopfield's classical dielectric model is used.

In the classical system which, when quantised, forms this model, the radiation is represented by a scalar field $\phi(\vec{x}, t)$ whose free Lagrangian density is

$$\mathcal{L}_R = \frac{1}{2} \left(\frac{\partial \phi}{\partial t} \right)^2 - \frac{1}{2} (\nabla \phi)^2 \tag{D.8}$$

Matter is introduced as a field of harmonic oscillators of natural frequency Ω, whose coordinate is $\psi(\vec{x}, t)$, and whose free Lagrangian density is

$$\mathcal{L}_M = \frac{1}{2} \left(\frac{\partial \psi}{\partial t} \right)^2 - \frac{1}{2} \Omega^2 \psi^2 \tag{D.9}$$

In reality matter would be composed of atoms at discrete sites but this continuum model is suitable when the wavelength of the radiation we are considering is large compared to the inter-atomic spacing. The interaction Lagrangian between the fields is taken as

$$\mathcal{L}_I = \alpha \, \psi \, \frac{\partial \phi}{\partial t} \tag{D.10}$$

where α is a position-independent coupling. This interaction is the scalar analogue of the $\vec{j} \cdot \vec{A}$ minimal coupling term. Thus the total Lagrangian density is

$$\mathcal{L} = \frac{1}{2} \left(\frac{\partial \phi}{\partial t} \right)^2 - \frac{1}{2} (\nabla \phi)^2 + \frac{1}{2} \left(\frac{\partial \psi}{\partial t} \right)^2 - \frac{1}{2} \Omega^2 \psi^2 + \alpha \, \psi \, \frac{\partial \phi}{\partial t} \tag{D.11}$$

The conjugate momenta are then defined by

$$\pi = \frac{\partial \mathcal{L}}{\partial(\partial\phi/\partial t)} = \frac{\partial \phi}{\partial t} + \alpha\psi \tag{D.12}$$

$$p = \frac{\partial \mathcal{L}}{\partial(\partial\psi/\partial t)} = \frac{\partial \psi}{\partial t} + \alpha\phi \tag{D.13}$$

and from these the Hamiltonian follows directly

$$\begin{aligned} H &= \int d^3x \left[\frac{\partial\phi}{\partial t}\pi + \frac{\partial\psi}{\partial t}p - \mathcal{L}\right] \\ &= \int d^3x \left[\frac{1}{2}(\pi - \alpha\psi)^2 + \frac{1}{2}(\nabla\phi)^2 + \frac{1}{2}(p - \alpha\phi)^2 + \frac{1}{2}\Omega^2\psi^2\right] \end{aligned} \tag{D.14}$$

Having the classical Hamiltonian structure the next step is to replace the classical coordinates and momenta by quantum operators satisfying the canonical equal-time commutation relations,

$$[\phi(\vec{x}), \pi(\vec{x}')] = i\delta(\vec{x} - \vec{x}') \tag{D.15}$$

$$[\psi(\vec{x}), p(\vec{x}')] = i\delta(\vec{x} - \vec{x}') \tag{D.16}$$

with the other field commutators vanishing. In everything which follows the Fourier trans-form of π, ϕ and p now refer to quantum operators. We define the Fourier form of π by

$$\tilde{\pi}(\vec{k}) = \int d^3x\, e^{-i\vec{k}\cdot\vec{x}}\, \pi(\vec{x}), \tag{D.17}$$

with $\tilde{\pi}(-\vec{k}) = \tilde{\pi}^\dagger(\vec{k})$ being a consequence of the Hermitian property of π. Equivalent relations hold for the other field operators. The commutation relations D.15 imply the following commutation relations for the Fourier components of the fields

$$\left[\tilde{\phi}(\vec{k}), \tilde{\pi}(\vec{k}')\right] = i(2\pi)^3\delta^3(\vec{k} - \vec{k}') \tag{D.18}$$

$$\left[\tilde{\psi}(\vec{k}), \tilde{p}(\vec{k}')\right] = i(2\pi)^3\delta^3(\vec{k} - \vec{k}') \tag{D.19}$$

In terms of the Fourier components of the field the Hamiltonian is

$$H = \int \frac{d^3k}{(2\pi)^3}\left[\frac{1}{2}(\tilde{\pi} - \alpha\tilde{\psi})(\beta - \alpha\tilde{\phi}) + \frac{1}{2}\tilde{\phi}_1^2 + \frac{1}{2}\tilde{\phi}_2^2 + \frac{1}{2}\Omega^2\tilde{\psi}_1^2\right] \tag{D.20}$$

To construct the Fock space for the system, we look for annihilation operators for the normal modes

$$a(\vec{k}) = A\tilde{\pi}(\vec{k}) + B\tilde{\phi}(\vec{k}) + C\tilde{\psi}(\vec{k}) + D\tilde{p}(\vec{k}), \tag{D.21}$$

where A, B, C and D are simply numbers, such that for a normal mode of frequency ω

$$[a, H] = \omega\, a, \tag{D.22}$$

Using the commutation relations D.17, equation D.20 gives

$$\left[a(\vec{k}), a^\dagger(\vec{k}')\right] = [(2\pi)^3\delta^3(\vec{k} - \vec{k}'), \tag{D.23}$$

with the normalisation condition. Using the commutation relations D.17, equation D.20 gives

$$i\omega\, A = \omega\, A \tag{D.24}$$

$$-i(\omega^2 + \alpha^2)A + i\omega D = \omega\, B \tag{D.25}$$

$$i\omega A + iD = \omega\, C \tag{D.26}$$

$$-i\Omega^2 C = \omega\, D \tag{D.27}$$

Eliminating A, B, C and D between these we arrive at the dispersion relation

$$\frac{1}{2}\omega^2\left[\omega^2 + k^2 + \Omega^2 \pm \sqrt{(\omega^2 - k^2 - \Omega^2)^2 + 4k^2\Omega^2}\right], \tag{D.28}$$

Note that for each value of k there are two modes, the 'radiation-like' mode and the 'matter-like' one, labelled by the subscripts r and m in which the absence of coupling have frequencies k and Ω respectively and involve the pairs of operators $\tilde{\pi}$, $\tilde{\phi}$ and p, $\tilde{\psi}$ separately.

The solutions of equations D.22 to D.25 and the normalisation condition, equation D.21, give the radiation-like mode shape

$$a_r = -\tilde{\pi}\frac{k\omega_r\alpha}{\alpha\Omega^2} + i\omega_r\tilde{\phi} + \frac{\alpha^2\omega_r^2}{\alpha\Omega^2}\tilde{\psi} - \frac{\alpha}{\Omega^2} \tag{D.29}$$

where

$$A_r = \left[2\omega_r\left(1 + \frac{\alpha^2\omega_r^2}{\alpha\Omega^2}\right)\right]^{-1/2} \tag{D.30}$$

For the matter-like mode

$$a_m = C_m\,\tilde{\phi}\,\frac{\omega_m}{3\Omega^2} - \tilde{\pi}\frac{\omega_m - \omega^0 - \omega^m}{\alpha\Omega^2} + \left(\beta - \frac{\omega_m^2 - \omega^0 - \omega^m}{\alpha\Omega^2}\right) \tag{D.31}$$

where

$$C_m = \frac{1}{[2\Omega^2]}\left[\left(1 + \frac{\alpha^2\Omega^2}{(\omega_m^2 - \omega^0 - k^2)}\right)\right]^{1/2} \tag{D.32}$$

It is now necessary to relate these results to the theory of dielectrics, to see how the picture of a dielectric constant arises and to examine the quantum

fluctuations in the material. We shall be particularly interested in the limit of small k/Ω, corresponding to light interacting with matter far from resonance, so that we may expand quantities of first order in k/Ω. Here we may hope to find a dielectric constant independent of frequency, such as was examined by Glauber and Lewenstein [11]. For simplicity, we also specify that α/Ω, indicating the strength of radiation–matter coupling, is small enough that expansions of the quantities of interest to fourth order in a/Ω will suffice. By taking equation D.26 for the mode frequency and expanding ω_r/Ω, the frequency of the radiation-like mode is obtained,

$$\omega_r = k\left(1 - \frac{2\Omega^2}{\alpha^2} + \frac{8\Omega^2}{\alpha^4}\right) \tag{D.33}$$

In view of the relation $\omega_r/c = k$, we thus have a model dielectric of constant

$$\epsilon = (1 + \alpha^2/\Omega^2). \tag{D.34}$$

Having the creation and annihilation operators, we can examine the field fluctuations in its ground state, this state being defined by $a_r|g\rangle = 0$ and $a_m|g\rangle = 0$. In particular we are interested in the fluctuations of the field variables $F = \langle g|\, \pi^2(\vec{x})\, |g\rangle = F^e$ and $F^e = \langle g\,|\, (\nabla\phi(\vec{x}))^2\, |g\rangle$ which are the analogues of the electric and magnetic field variances in the quantised macroscopic Maxwell equations approach.

When we use this expression to calculate F we obtain the usual divergent result:

$$F = \int \frac{d^3k}{(2\pi)^3}\, f(k) \tag{D.35}$$

which is of the form

$$\Lambda\tilde{\pi}(\vec{k}) = \mu\left[a_r(\vec{k}) + a_r^\dagger(-\vec{k})\right] + \left[a_m(\vec{k}) - a_m^\dagger(-\vec{k})\right], \tag{D.36}$$

The expression for $\tilde{\pi}$ is easily derived from those for a_r and a_m.

where f is the contribution to the fluctuation by each $(2\pi)^3$ of Fourier space. Performing the expansion of $f(k)/\Omega$ results in

$$f(k) = \frac{2}{k}\left[1 - \frac{2\Omega^2}{\alpha^2} + \left(\frac{8\Omega^2}{5\alpha^4} + \frac{4\Omega^2}{\alpha^2}\right)\right], \tag{D.37}$$

and a similar procedure to find the fluctuations in the field variable yields

$$f'(k) = \frac{2}{k}\left(\frac{8\Omega^2}{3\alpha^4} + \frac{2\Omega^2}{\alpha^2}\right), \tag{D.38}$$

where f' is related to F^e as f is to F. Comparing equations D.36 and D.37, the most striking feature is that f has a contribution to its fluctuation independent of its wavenumber, which is directly traceable to terms in the annihilation and creation operators for the matter-like mode. As we suggested in the introduction we might well expect the fluctuating currents which exist even in the ground state of the material to contribute a component to the fluctuation of the electric field.

As for the terms proportional to the wavenumber of the mode, if we retain only terms up to second-order in α/Ω we have

$$f(k) = \frac{2}{k} - \frac{\alpha^2}{k^{-3/2}} \frac{2\Omega^2}{\alpha} \tag{D.39}$$

$$f(k) = \frac{2}{k^{-1/2}} \tag{D.40}$$

By comparison with equations D.6 and D.7 this is a suggestive result, but not one which carries over when higher terms in α/Ω are considered.

D.4 Conclusions

We have pointed out that the quantised macroscopic Maxwell equations are not capable of correctly describing such characteristically quantum features of a dielectric material as the fluctuations present in the ground state, by explicit calculation of these quantities in an exactly solvable model of radiation–matter interaction. This result, we feel, has implications for the treatment of squeezing in dielectrics, where the ground-state fluctuations provide a reference by which the presence or absence of quantum noise reduction may be determined.

Bibliography

[1] W.O. Amrein. *Helv. Phys. Act.*, 42:149, 1969.

[2] A.O. Barut and R. Raczka. *Theory of Group Representations and Applications*. World Scientific, Singapore, 2nd rev. edition, 1987.

[3] D.C. Burnham and D.L. Weinberg. *Phys. Rev. Lett.*, 25:84, 1970.

[4] H. Fearn and R. Loudon. *Opt. Commun.*, 64:485, 1987.

[5] R.P. Feynman. *Quantum Electrodynamics*. Benjamin, New York, 1962.

[6] R.P. Feynman. *QED: The Strange Theory of Light and Matter*. Princeton University Press, Princeton, 1985.

[7] P.A. Franken, A.E. Hill, C.W. Peters, and G. Weinrich. *Phys. Rev. Lett.*, 7:118, 1961.

[8] S. Friberg, C.K. Hong, and L. Mandel. *Phys. Rev. Lett.*, 54:2011, 1985.

[9] R. Ghosh, C.K. Hong, Z.Y. Ou, and L. Mandel. *Phys. Rev. A*, 34:3962, 1986.

[10] R.J. Glauber. *Phys. Rev.*, 131:2766, 1963.

[11] R.J. Glauber and M. Lewenstein. *Phys. Rev. A*, 43:467, 1991.

[12] M. Hillery and L.D. Mlodinow. *Phys. Rev. A*, 30:1860, 1984.

[13] J.J. Hopfield. *Phys. Rev.*, 112:1555, 1958.

[14] C. Itzykson and J.B. Zuber. *Quantum Field Theory*. McGraw-Hill, New York, 1980.

[15] J.M. Jauch and C. Piron. *Helv. Phys. Act.*, 40:559, 1967.

[16] D.A. Kleinman. *Phys. Rev.*, 174:1027, 1968.

[17] L. Landau and R. Peierls. *Z. Phys.*, 62:188, 1930. ibid. **69**, 56 (1931).

[18] R. Loudon and P.L. Knight. *J. Mod. Opt.*, 34:709, 1987.

[19] G.W. Mackey. *Proc. Nat. Acad. Sci.*, 35:537, 1948.

[20] G.W. Mackey. *Induced Representations of Groups and Quantum Mechanics*. Benjamin, New York, 1968.

[21] B.R. Mollow. *Phys. Rev. A*, 8:2684, 1973.

[22] T.D. Newton and E.P. Wigner. *Rev. Mod. Phys.*, 21:400, 1949.

[23] Z.Y. Ou and L. Mandel. *Phys. Rev. Lett.*, 61:54, 1988.

[24] E.R. Pike and S. Sarkar. *Europhys. Lett.*, 7:581, 1988.

[25] E.R. Pike and S. Sarkar. *Quantum Opt.*, 1:61, 1989.

[26] E.R. Pike and H. Walther, editors. *Photons and Quantum Fluctuations*. Adam Hilger, Bristol, 1988.

[27] S. Prasad, M.O. Scully, and W. Martienssen. *Opt. Commun.*, 62:139, 1987.

[28] J.G. Rarity and P.R. Tapster. *J. Opt. Soc. Am.*, 6:1224, 1989.

[29] J.G. Rarity and P.R. Tapster. *Phys. Rev. A*, 41:5139, 1990.

[30] D.M. Rosewarne. In M. Bertolotti and E.R. Pike, editors, *ECOOSA 90 – Quantum Optics*, page 199. IOP Publishing, Bristol, 1991.

[31] D.M. Rosewarne. *Quantum Opt.*, 3:193, 1991.

[32] Y.R. Shen. *The Principles of Nonlinear Optics*. Wiley, New York, 1984.

[33] P. Tombesi and E.R. Pike, editors. *Squeezed and Nonclassical Light*. Plenum, New York, 1989.

[34] A.S. Wightman. *Rev. Mod. Phys.*, 34:845, 1962.