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REVIEW ARTICLE

Operator reaction field theory in quantum optics: a study of one two-level atom in a broad-band squeezed vacuum without rotating wave approximation

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Abstract. We review the use of operator reaction field theory in quantum optics by considering the problem of one atom in an isotropic three-dimensional broad-band squeezed vacuum. In these terms we give a quantum electrodynamical analysis of this system exact up to all contributions of order one in the ratio $\gamma \omega_0^{-1}$, the ratio of the *A*-coefficient to the atomic resonance frequency. These terms include all of the counter-rotating terms at this order. At zero order, namely in the usual rotating wave approximation, we find the usual Einstein rate equation while we confirm that the fluorescence spectrum consists of two Lorentzian peaks. However, at order one we find an additional *oscillating* term in the rate equation and additional resonant dispersive terms correct for each of the two Lorentzian peaks in the fluorescence spectrum. Similar results are expected for broad-band correlated squeezed vacua of arbitrary, rather than isotropic, geometry.

Keywords: Operator self-fields, QED, screening of the *A*-coefficient, rotating-wave approximation, Einstein rate equation, squeezed vacuum, fluorescence spectrum

1. Introduction

The two-level atom is the simplest possible model of any real atom. It has a distinguished history in theoretical laser physics (see, e.g., [1]) and still more so generally in theoretical quantum optics (see, e.g., [2-4] as particular references). Even the strictly mathematical representations of the model as a quantum mechanical spin- $\frac{1}{2}$ model have significance. Thus in [5] one of us mentioned a number of different representations of this model (e.g. as the twodimensional representation of the su(2) Lie algebra (equals spin- $\frac{1}{2}$), as a restricted two-boson Schwinger representation, a one-fermion representation, as a restricted one-boson representation, etc) which could all be summarized as a one-fermion representation of the model. Coupled to the quantized electromagnetic field this means [5] a strictly nonlinear fermion-boson coupling between the atom and the field. This nonlinearity is one aspect of the importance of the model in theoretical quantum optics.

Indeed, particularly interesting physics emerges as soon as the single two-level atom is coupled to any quantized e.m.

radiation field, either vacuum or other field. In this paper we offer a brief review of the topical problem of a single two-level atom coupled to a broad-band isotropic squeezed vacuum field. The effects of squeezed vacua with other geometries can easily be extracted from the theory. Such a review may be particularly appropriate for the newly revised form of this journal, J. Opt. B: Quantum Semiclass. Opt. As our title suggests we also use this article to review very briefly some of the applications of operator reaction field theory (ORFT) in theoretical quantum optics. Early references to ORFT are [6-8] for example. A recent application of ORFT is given in [9], and this is particularly relevant to this review since it concerns two-level atoms in squeezed vacua. However, compared with the previous work using the ORFT for twolevel atoms in squeezed vacua, and especially that in [9] where $N_a \ge 1$ atoms are considered, a new feature in this review of one atom is that we shall work here consistently to order *one* in the ratio $\gamma \omega_o^{-1}$ and not order zero: γ is the A-coefficient of the two-level atom and ω_0 is its radiatively unshifted transition frequency. This order one in $\gamma \omega_0^$ makes the first correction to the usual analysis, made for

example in [9], in the so-called rotating wave approximation (RWA). But even at order zero, namely within the RWA itself, the effects of the quantized radiation field, which include radiative level shifts of each of the two levels of the atomic model induced by the ordinary e.m. vacuum of no photons, are further complicated by the photons of the squeezed vacuum. So the 'simple' single two-level atom, coupled into the squeezed vacuum, has already become quite complicated.

Still, by working at order one in $\gamma \omega_0^{-1}$ we shall find that we can extract two wholly new results, not reported before to our knowledge. One of these is a steady oscillation in the steady state atomic inversion induced by the broad-band squeezed vacuum; the other is additional *resonant* terms in the fluorescence spectrum induced by this squeezed vacuum. Both of these results derive from the counter-rotating terms correcting the RWA.

As noted, we shall deliberately work the whole analysis in terms of ORFT: this is particularly convenient for this (and many other) problem in theoretical quantum optics. By making this analysis in all its necessary detail for the single two-level atom in this paper we thus provide a review of at least *some* important general aspects of the application of ORFT in quantum optics.

Two very early papers on ORFT are [6, 7], already mentioned, while earlier reviews appeared in [8]: a recent book [8] also reviews the interplay between the radiation reaction and vacuum fluctuations and the interpretation of various operator ordering (normal, anti-normal, symmetric) in the case of the normal vacuum (see, in particular, [10]). ORFT is a powerful technique for introducing in a natural (and rather physical) way both the Einstein Acoefficient and the radiative level shifts for the low-energy quantum electrodynamics of one or more atoms interacting with the quantized e.m. field. Thus for the many-body problem of *linear dielectrics* consisting of N_a two-level (or N_a many-level) atoms, for example (N_a is here very large $\sim 10^{23}$), we have already constructed [11–13] selfconsistently, the refractive index theory of such a dielectric by adding to the interaction field between pairs of atoms the self-field. This self-field describes radiation reaction, an idea which goes back to Lorentz [14]. In the third edition of his book P A M Dirac [15] gives a relativistic theory of radiation reaction but discards this in the fourth edition [16] since he no longer considers point electrons analogous to classical electrons there. In [17], pp 581-94, Jackson explicitly introduces and analyses the classical Abraham-Lorentz model. This model has the additional radiative reaction *force* or self-force $\frac{2}{2}e^2v^{\prime\prime}/c^3$ acting on the electron due to its radiation, where v is the electron velocity. Since $v = x^{\dagger}$ and x is the position of the electron, the dipole moment for a bound electron held in an atom is ex, while the additional field on the electron is the self-field $\frac{2}{3}ex^{\cdots}(t)/c^3$. For $x(t) = x_0e^{-i\omega t}$ for the oscillating bound electron driven by the applied field E(t) = $E_0 e^{-i\omega t}$ this self-field is $\frac{2}{3}ex(-i\omega)^3/c^3$, which corresponds exactly to the $\frac{2}{2}i(\omega/c)^{3}p(r)$, the convergent part of the integral, equation (1) below. Moreover, in [11] it is shown that this expression for the convergent part of integral (1) changes the *polarizability* $\alpha(\omega)$ for a single atom at frequency ω rad s⁻¹ to $\gamma(\omega) = \alpha(\omega)/(1 - \frac{2}{3}ik_0^3\alpha(\omega))$. With the expression for $\alpha(\omega) = (e^2/m) \sum_{s} f_s (\omega_s^2 - \omega^2)^{-1}$ due to Kramers and Heisenberg referenced in [11], in which f_s is the 'oscillator strength' and m the electron mass, one finds near to a resonance, where $\omega \approx \omega_s$ for some atomic resonance frequency ω_s , that $\gamma(\omega) \approx (e^2/m) f_s / [\omega_s^2 - \omega^2 - \frac{2}{3}i\omega_s^3 c^{-3}(e^2/m) f_s]$. But $f_s = 2m\omega_s |x_{0s}|^2 \hbar^{-1}$ where $ex_{0s} = e\langle 0|x|s \rangle$ is the dipole matrix element between the ground atomic state $|0\rangle$ and the excited atomic state $|s\rangle$ so that $\gamma(\omega) \approx (e^2/m) f_s / [\omega_s^2 - \omega^2 - i\omega_s \gamma]$ and γ is the Einstein A-coefficient-see below. In this argument the A-coefficient, deriving from the classical expression (1), is a classical concept, associated with classical oscillators of strength f_s . However, \hbar is concealed in these f_s . These are pure numbers: $f_s > 0$ satisfy the Thomas–Reiche– Kuhn sum-rule $\sum_{s} f_s = 1$ for one 'optical' electron in the atom as is easily proved. Since $\gamma(\omega)$ replaces $\alpha(\omega)$ in the dielectric theory [11-13] it is easy to see that the Acoefficient in $\gamma(\omega)$ can become 'screened' (actually by a factor of refractive index and more generally by other manybody terms within that dielectric theory-see below). On the other hand, the classical radiative level shifts mentioned below equation (1) become transformed into nonclassical expressions by making use of the ORFT as is explained briefly in the second paragraph below equation (1) and then shown explicitly in section 2.2. Moreover, many-body terms further correct these radiative shifts, including in these the Lorentz field shifts, and their generalization [11-13]. In practice, the self-field in the dielectric theory [11-13] amounts to assigning a meaning to the mathematical expression

$$\int \boldsymbol{p}(\boldsymbol{r}) \cdot \boldsymbol{F}(\boldsymbol{r}, \boldsymbol{r}'; \omega) \delta(\boldsymbol{r} - \boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}'. \tag{1}$$

In this expression a dipole moment density p(r) induced in an atom at r (in three-dimensional space) radiates at radial frequency ω through the 'photon propagator' or Green function $F(r, r'; \omega)$ back onto itself at r = r' (we explain for the record that the tensor Green function F takes the explicit form $F(r, r'; \omega) = (\nabla \nabla + k_0^2 U)r^{-1} \exp(ik_0 r)$ where $r = |r - r'|, k_0 = \omega c^{-1}, U$ is the unit tensor in dyadic notation and ∇ is the usual gradient operator (see, e.g., [11, pp 254–255])). The meaning we naturally attach to expression (1), is $\frac{2}{3}ik_0^3 p(r)$, namely the strictly convergent part of this integral. However, there is also a linearly divergent part, and potentially still other divergences because of the dipole approximation [18]: the linear divergence becomes the 'classical' level shift and was known to Lorentz [14].

In ORFT the same two terms arise (see our equation (12) below), but now the radiative level shift is softened by normal ordering to a logarithmic divergence $\int^{\omega_c} \omega^{-1} d\omega$, and actual *convergence* is gained by formally choosing $\omega_c < \infty$ and equal to the Compton frequency, a prescription first adopted by Weisskopf [19]. By this prescription the level shift (of about 1000 MHz) is very much of the right order to match the subsequently observed Lamb–Retherford radiative level shift in the 2²s level of hydrogen shifted relative to the p-states (2¹p and 2²p) [20]. In this paper, we shall show by ORFT how the squeezed vacuum modifies these vacuum radiative shifts by adding appropriate 'light shifts' to them, for the

squeezed vacuum is an excited state of the ordinary vacuum (such light shifts induced by ordinary laser fields have come in to particular prominence recently in the context of laser cooling [21]).

Notice how in the many-body dielectric theory of [11-13] the δ -function in equation (1) survives as such throughout the whole connected many-body analysis. It emerges at the end [13, equation (7.5)] as the 'self-correlation' in the density fluctuations which determine the total optical scattering cross-section of the many-atom dielectric (for orientation, the argument for equation (7.5) in [13] uses the result of equation (6.9) in [13] which is $\int u_2(r) dr = \kappa_T k_B T$ where κ_T is the isothermal compressibility of the system, $k_{\rm B}$ is Boltzmann's constant and T is the absolute temperature: $u_2(r)$ is the first of the so-called Ursell functions [11, p 268, equation (4.12b)] and $u_2(r) = g_{12}(r) + n^{-1}\delta(r) - 1$ [11, pp 261, 263]. At last, in this expression [13, equation (7.5)] we find the δ -function $\delta(r)$ describing self-correlation or selfinteraction or radiation reaction! The number n is the mean number of atoms in unit volume.

This brief excursion into molecular correlation theory allows us to make a point concerning the ORFT applied to dielectrics. This is that the ORFT naturally provides a theory of the screening of the Einstein A-coefficient in a dielectric [22][†]. In the recent papers [23–29], for example, (as well as the recent papers on this topic referenced in these papers) it is shown that the effective A-coefficient γ in a dielectric gains at least a factor of the refractive index of that dielectric. It is worth commenting on these recent papers [24-29]. In [24], section 3 evaluates the transverse Green function which is the convergent part of an integral like that in equation (1) for the self-field. However, the refractive index factor $n(\omega)$ arises here by using the screened propagator \tilde{F} we used in [12] rather than the unscreened Fwhich appears in equation (1). The divergence associated with the longitudinal Green function of this paper [24] is already explained, and indeed properly handled, in our [12]. It is important to note that, except for neglect of certain subtle surface-dependent many-body terms, the use of F rather than F in [12] is shown to be wholly *equivalent* to the use of F: in fact, \tilde{F} derives from F otherwise as a gauge transformation (see particularly the reference to Mazur (1958) in [11]). Even so, the 'subtle surface-dependent terms' play a specific role in introducing the factor $n(\omega)$ into the scattering cross-section (and the A-coefficient)—see [30, 31]). The discussion [25] has also been handled previously by the self-field analysis (see, for example, chapter 2 of [11] and the later analysis).

The paper by de Vries and Lagendijkin [26] entitled 'Resonant scattering and spontaneous emission in dielectrics: microscopic derivation of local field effects' is also already *comprehensively* covered and extended in the papers [11–13] with [22] and the references to the authors' work listed there. In effect [11–13] together solve the whole many-body problem for the internal field (equally local-field [26]) in a disordered dielectric, but because the dielectric constant $\epsilon(\omega)$ (called $m^2(\omega)$ in [11–13]) is expressed *in terms* of $\epsilon(\omega)$, such expressions become formal 'integral equations' for $\epsilon(\omega)$. Various alternative integral equations of this type are given in [11–13] as well as a low-density expansion in a small parameter $n\alpha(\omega)$, *n* the number density of atoms and $\alpha(\omega)$ the atomic polarizability. The problem of the isotropic crystal lattice considered in the context of [26] was originally treated in the paper by Bullough and Thompson, referenced in [11].

Glauber and Lewenstein [27] makes a macroscopic quantization of a smooth dielectric which leads to a screening of the A-coefficient which is more compatible with the use of the screened propagator \tilde{F} as it was introduced in [12] (see [22,24]) rather than the unscreened propagator F quoted here below equation (1). Using equation (7.19*a*) in [12] a 'best result' for the dispersion relation is obtained (m^3 in this equation (7.19*a*) should be corrected to m^2). This best dispersion relation (also requoted as equation (2) in [22]) is not the Lorentz–Lorenz relation but takes the form

$$\left(\frac{m^2-1}{4\pi}\right)\left(\frac{2m^2+1}{3m^2}\right) = n_0\beta + \frac{m^2+2}{3m^2}(C+Q) + \frac{4\pi}{3m^2}(C+Q)^2 + \cdots$$

where n_0 is the number density of atoms, $m \equiv m(\omega)$ is the refractive index and *C* depends on *m* and β , and *Q* depends on the atomic polarizability α and the multi-body correlations, while β is an *effective* polarizability depending on radiation reaction (self-and other correlations). Evidently this dispersion relation is actually a complicated manybody integral equation for $m(\omega)$ or for $m^2(\omega) \equiv \epsilon(\omega)$ the dielectric constant. However, the 'screened internal field factor' $3m^2/(2m^2 + 1)$ in this dispersion equation, taken together with \tilde{F} instead of *F*, yields the exact result found in [27]. Evidently the actual situation is once again still more complicated than that found in [27].

Milonni [28] also considered a macroscopic quantization of a smooth linear dielectric at frequencies away from resonances. He investigates local field corrections to the *A*-coefficient of Lorentz field type (quoting Dexter and others, his [16], in this connection) as well as investigating a 'screened' local field of [27] above. Note, from [22], that the effective local field correction on the *A*-coefficient is nearer to $n[\frac{1}{3}(n^2+2)]^4$ rather than the Lorentz field correction $n[\frac{1}{3}(n^2+2)]^2$ obtained by Dexter and others (*n*= refractive index, called $m(\omega)$ in [27], and is as used in [11–13]).

Fleischhauer and Yelin [29] study radiative atom-atom interactions in optically dense media in a wholly quantum framework assuming Gaussian classical statistics for the interacting field and show, among other results, that the Lorentz-Lorenz dispersion relation holds for the mean amplitude of the local field seen by the probe atom (also see Bowden and Dowling [29]). Unfortunately for any comparisons the quantum basis to the many-body theoretical papers [11-13] still remains unpublished (cf [11-13] but also see [30]) but for two-level atoms at least the linearized quantum Bloch-Maxwell equations, formally extended to many atomic levels via the polarizabilities $\alpha(\omega)$, yield the classical many-body theory analysed in [11-13]. The 'internal field' theory thus becomes that presented there and its effects on the A-coefficient are those here summarized in [22, 24–28] above.

[†] Unfortunately the expression 13 lines below equation (1) in [22] should read $\bar{n}\kappa k_{\rm B}T = \int [\delta(r) + \bar{n}(g_2(n) - 1)] d(\text{vol})$ —the important $\delta(r)$ replacing the '1' appearing there. (Note that the authorship of [10] in [22] needs correction to that of our [30] below.)

In the present analysis the A-coefficient for a single twolevel atom at *r* effectively derives from the (convergent part) of the self-field $\frac{2}{3}ik_0^3 p(r)$ coming from equation (1) (as is already explained above equation (1)). This self-field for the single two-level atom appears at operator level as the $\frac{2}{3}i(\frac{\omega_o}{c})^3 p\sigma_-$ as it is given in expression (12) for the operator self-field exhibited below. Of course, the $k_0 \equiv \omega c^{-1}$ coming from equation (1) now goes to actual resonance at $\omega = \omega_0$ in the ORFT, expression (12) below. However, in the manybody problem of a real dielectric [11–13] such screening of the A-coefficient cannot be so simply expressed in terms of a single factor of the refractive index, nor indeed this factor with further 'internal field' factors as suggested in [23, 29]. The many-body theory shows [11-13] that the situation is much more complicated. Even so [22] the screening of the A-coefficient in a linear dielectric is in effect being observed by experiments measuring the total scattering cross section of the dielectric, the connecting link being through reaction field theory[†].

Recently [32,33] we have extended such linear refractive index theory to a nonlinear refractive index theory, and most recently to a theory of optical bistability in a squeezed vacuum [34, 35]. In effect the ORFT plays its role, consistently and naturally, in all of these theories, and very much as it does for the single atom in this paper.

With these remarks as an indication of the scale in which the ideas of ORFT can be applied in theoretical quantum optics we return to the simpler problem of the single twolevel atom in the squeezed vacuum to be addressed in this paper. The pure, zero-photon, vacuum field provides the appropriate vacuum level shifts [6-8], but of course there is no fluorescence spectrum from such a vacuum. For any observable fluorescence spectrum the natural correspondence to be drawn between unsqueezed and squeezed situations is that between an atom in a thermal (i.e. black-body isotropic and unsqueezed) field with a nonzero mean photon number \bar{n} at resonance, and an isotropic squeezed vacuum—which also has a nonzero mean photon number. We call this mean number in the isotropic squeezed vacuum the number N(>0) in this paper—in line with previous work on squeezed vacua [9].

We remark on this correspondence in this paper noting that counter-rotating terms correct the (unsqueezed) thermal field as well as the squeezed vacuum field. In contrast, the counter-rotating terms which induce the oscillation in $r_3(\infty)$ in the squeezed vacuum make no change at all in $r_3(\infty)$ for the thermal field and the oscillations in $r_3(\infty)$ are very much an effect of the squeezed vacuum.

For historical reference recall (see [6–8, 36–40] and especially [6, 7, 36]) that in the low-energy quantum electrodynamical description of vacuum (i.e. ordinary vacuum) field induced radiative level shifts in atoms one soon discovers that the RWA is a very bad approximation and that as much as one half of the total radiative shift in a two-level atom, and likewise in a real multi-level atom [39], is due to the counter-rotating terms. Depending on the actual photon number distributions a similar situation extends to the light shifts induced by any excited states of the quantized electromagnetic field with nonzero photon numbers [9, 38]. The *squeezed* vacuum is just such an excited state of the quantized e.m. field, and this already suggests that only by systematically investigating both the rotating *and* the counter-rotating terms in the theory will it become possible to give an adequate description of the properties of a single atom interacting with a squeezed vacuum.

The radiative level shifts present themselves in the theory at order $\gamma \omega_0^{-1}$ where γ is the *A*-coefficient ($\gamma = \frac{4\pi}{3} \frac{p^2}{\hbar} \frac{\omega_0^3}{c^3}$ for the two-level atom (e.g., [6,7]) and ω_0 is the atomic transition frequency in rad s^{-1} : *p* is the dipole matrix element for the transition between the two nondegenerate atomic levels). For ordinary atomic transitions with large A-coefficients, such as the *D*-line transitions of Na, $\gamma \omega_0^{-1} \approx 10^{-6}$ and is still small (but the radiative level shifts are still measurable-[20]); and this situation of smallness is little changed for high Rydberg transitions where p is relatively very large but ω_0 becomes much smaller [41]. In order to carry through a consistent investigation of the generalized A-coefficients and radiative level shifts for a single atom in a squeezed vacuum, as is intended in this paper, this situation suggests that it would indeed be helpful to make a systematic development of the theory, systematic to order $\gamma \omega_0^{-1}$, which includes all terms which arise at this order. ORFT is ideal for such an investigation [6-8, 36-39] although [9] in particular adopted the RWA for a comparable investigation of a system of one or more atoms in a squeezed vacuum. This paper therefore presents just such a systematic investigation for the *single* two-level atom in the broad-band squeezed vacuum. It extends [9] (for the single atom) by including the counterrotating terms, and in the course of the argument we derive the new results for $r_3(\infty)$ and for the fluorescence spectrum already mentioned.

From numerical work (see section 2) we find that the frequency of the oscillation induced in the steady inversion is close to $2\omega_0$ rad s⁻¹ as expected for counter-rotating term effects, while the *magnitude* of the oscillation is indeed $O(\gamma \omega_0^{-1})$ and small, of the order of the radiative shifts. Since we shall work with a single two-level atom throughout, there arises the question of the effects of the other levels in a real atom at this order especially if the oscillation in $r_3(\infty)$ is ever to be observed experimentally.

For the radiative level shifts from the ordinary vacuum these effects of many levels are well understood (e.g., [8, 39, 42]) and the radiative effects of each of the different level transitions available (each of those with nonvanishing dipole matrix elements p) must simply be added. For the effects of the squeezed vacuum and many levels (already $O(\gamma \omega_0^{-1})$ for two levels as noted) the situation is different: in the first place finite atomic inversion $r_3(\infty) > -1$ is a resonance phenomena; thus by calculating $r_3(\infty)$ for the steady inversion for a single two-level atom of resonance frequency ω_0 in the squeezed vacuum one legitimately assumes that the additional effects of the other levels in any real multi-level atom should be nonresonant and relatively small. Moreover, for the resonance fluorescence spectrum for a two-level atom in the presence of a squeezed vacuum we find *resonant* terms $O(\gamma \omega_0^{-1})$ from the counter-rotating terms

[†] To our knowledge the simple particular factor of refractive index on the scattering cross-section first emerged in the papers [30, 31] while it is also explained via the screened propagator \tilde{F} of [12] put into equation (1) instead of F (see references for details).

in addition to the *two* Lorentzian peaks (within the RWA) at O(1) [43, 44]. Thus by subtracting from any observed fluorescence spectrum the two Lorentzian peaks O(1) it may be possible to observe the additional terms $O(\gamma \omega_0^{-1})$ in an experiment. Evidently other levels in any real multi-level atom should again have relatively little effect since all of these other level contributions would be off-resonant and relatively small.

This viewpoint thus suggests that it makes physical sense to investigate in all detail and consistently up to $O(\gamma \omega_0^{-1})$ all terms contributing to the theory of a single two-level atom coupled to a squeezed vacuum. Additional multi-level effects in a real atom would only complicate but not significantly add to the understanding of an already complicated situation. This is a part of the background motivating the study of the two-level atom in this paper. The other part of that motivation (as explained) is to review the application of ORFT in quantum optics particularly by studying the single two-level atom in the squeezed vacuum this way.

2. Einstein rate equations

2.1. The case of the ordinary vacuum and the finite temperature effects

It is perhaps the 'Einstein rate equation' which has been most fundamental to our understanding of the interaction between atoms and radiation—ever since Einstein's original pioneering work [45] on his theory of the *A*- and *B*coefficients. For example, and well before the advent of the laser, Max Born [46] formulated this rate equation as (and see also, e.g., Louisell [47] and the review [48])

$$\dot{r_3}(t) = -\gamma \{ (1 + r_3(t)) + 2\bar{n}r_3(t) \}$$
(2)

in a modern notation. Max Born also adjoined

$$\frac{\mathrm{d}\bar{n}}{\mathrm{d}t} = \frac{\mathrm{d}|c_1|^2}{\mathrm{d}t} = -\frac{\mathrm{d}|c_2|^2}{\mathrm{d}t} = -\frac{1}{2}r^{\cdot}{}_3(t) \tag{3}$$

which conserves mean photon number \bar{n} seen as a function of time t ($r_3(t) \equiv dr_3(t)/dt$). In both equations (2) and (3), a two-level atom is implicitly understood. In this paper we consider a two-level atom with ground and excited states $|g\rangle$ and $|e\rangle$ respectively: these states have an energy spacing $\hbar\omega_0$ and γ is the A-coefficient defined in section 1. When the atom is in the arbitrary state $|\psi(t)\rangle = c_1(t)|g\rangle + c_2(t)|e\rangle$, $r_3(t)$ describes the inversion: $r_3(t) = |c_2|^2 - |c_1|^2$, while $|c_1|^2 + |c_2|^2 = 1$, so $\frac{1}{2}r_3(t) = d|c_2|^2/dt$ and this links $r_3(t)$, equation (2), to the radiation rate $d\bar{n}/dt$, equation (3), since conservation of mean photon number requires $\bar{n} + \frac{1}{2}r_3 + \frac{1}{2} =$ constant. We shall derive a generalized Einstein rate equation for the inversion $r_3(t)$ of a single two-level atom in the squeezed vacuum in this paper.

Under conditions in which \bar{n} is itself a constant equilibrium value, equation (2) as given by Born [46] becomes linear in $r_3(t)$ and its solution is

$$r_3(t) = (1+2\bar{n})^{-1} \{ (1+(1+2\bar{n})r_3(0))e^{-\gamma(1+2\bar{n})t} - 1 \}$$
(4)

and as $t \to \infty$, $r_3(t) \to -(1+2\bar{n})^{-1}$. If the atom is surrounded by ambient black-body radiation in equilbrium, \bar{n} is the Planck function, $\bar{n} = (e^{\beta\hbar\omega_0} - 1)^{-1}$. Then $|c_2|^2/|c_1|^2 =$ $(1 - r_3(t))/(1 + r_3(t)) = e^{-\beta\hbar\omega_0}$, so the state occupation in the atom is Boltzmann as was envisaged by Einstein [45–48].

The curly bracket in equation (2) shows that this rate equation embodies both stimulated absorption and the celebrated stimulated emission first envisaged by Einstein [45]: the spontaneous term is the $1 + r_3(t) = 2|c_2|^2$ in equation (2). Thus, as a working theory awaiting the advent of the laser and a deeper, quantum optical, analysis the theory of this 'Einstein rate equation' was already complete. But because of the obvious fundamental importance of the equation, two of the present authors subsequently gave the analysis of [49, 50] for it. Done at the level of low-energy q.e.d., particularly important conclusions from [49, 50] were (i) that, to reach equation (2) from a two-level atom Hamiltonian in which the interaction with the radiation field was taken in the RWA, it was necessary to replace expectation values taking the form $\langle e_0^{(-)}(t)R_3(t)e_0^{(+)}(t')\rangle$ by products of expectation values $\langle e_0^{(-)}(t)e_0^{(+)}(t')\rangle\langle R_3(t)\rangle$ in which $\langle R_3(t)\rangle$ is identified as $r_3(t)$ above: the $e_0^{(\pm)}(t)$ are (see below) free-field operators. If such steps, which replace averages of atom-field operator products by products of averages, are not made, and we describe such steps as an atom-field decorrelation below, the system is not closed in $r_3(t) = \langle R_3(t) \rangle$ simply, and equation (2) becomes a large system of coupled equations; (ii) in order to obtain the Planck radiation law (and the Planck function \bar{n} already introduced for black-body radiation in equilibrium by assuming the Boltzmann distribution $|c_2|^2/|c_1|^2 = e^{-\beta\hbar\omega_0}$ it was necessary to consider many atoms all coupled to the black-body radiation (which was to be expected). But then, in order to reach the rate equation (2) it was also necessary to make these atoms independent of each other by ignoring any cooperative effects. A calculation which reported measurable effects due to such cooperative action of the atoms was first given in [50] and appealed to the Hamiltonian of Dicke [51] for $N_a > 1$ atoms occupying the same single site. Experiments which confirmed the theoretical predictions of [50], namely that as $N_a \rightarrow \infty$ the system behaves in equilibrium as a giant quantum oscillator, were reported at the same time [41] (also see [52]) using Rydberg Na atoms interacting with black-body radiation in low-Q microwave cavities at a wavelength of 2.2 mm and temperatures of \sim 300 K. In [53] we successfully extended this giant quantum oscillator theory to the case of the squeezed vacuum.

These cooperative giant quantum oscillator states at equilibrium mean that the atoms now satisfy a Bose– Einstein statistics rather than a simple Boltzmann distribution individually [50]. The authors of [49] also investigated the theory if the atoms remained noncooperative and independent but the atom–field decorrelation referred to above was not made: various observable effects were described but, as far as we know, no experiments have been undertaken to observe these. Finally, concerning equation (2) itself, it is very relevant to the work now described in this paper to point out that exactly the same rate equation, (2), is obtained whether the RWA is made or not.

Thus, to sum up at this point, by studying single two-level atoms the purpose of the present paper is to derive by ORFT the novel results predicted for many two-level atoms under assumptions equivalent to atom-field decorrelation (i) and independent atoms (ii) for such two-level atoms in a broadband *squeezed vacuum* rather than black-body radiation: some of the correspondences between these two situations are already drawn in, e.g., [9] (and see, e.g., [54,55]).

Note that these results as described so far are all in the RWA; but as noted above the Einstein rate equation in black-body radiation is the same whether the RWA is made or not [49, 50]. However, one can see that this will not be the situation in the squeezed vacuum field. Typically this is an excited state of the radiation field comprised of even number photon states only, and within the RWA terms associated with two-photon processes oscillating at twice the atomic transition frequency, namely at $2\omega_0$, are discarded. This suggests that the rate equation (2) will show additional interesting effects in a squeezed vacuum but only outside the RWA. A point of this paper is thus to report these novel effects occurring outside the RWA for one, or many independent atoms in a broad-band squeezed vacuum. These effects are in particular the oscillatory terms in $r_3(\infty)$ already described; but they correspondingly modify the fluorescence spectrum in the squeezed vacuum as we show (we note that they also modify correlation functions such as the intensity-intensity correlation function like that investigated in [49]).

We must also note that we have already shown that in such a squeezed vacuum there actually are additional cooperative-atom effects from more than one atom in squeezed vacua. In particular, at microwave frequencies as we have remarked [53] that as $N_a \rightarrow \infty$ a giant quantum oscillator state is again formed. At $N_a < \infty$ there are however differences of detail between the effects of blackbody radiation and the effects of the squeezed vacuum on the atomic behaviour and on the fine structure of the cooperative fluorescent and absorption spectra [53, 56] and the situation needs an experimental investigation—one comparable with that already made [41, 52] for the black-body case, if this can be done.

As noted, some correspondences between black-body thermal field effects and the effects of the broad-band squeezed vacuum are already drawn in [9, 54, 55]. The investigations of [9, 53] use ORFT taken in the RWA. In this paper we use this ORFT as described originally in [36] (see also in [9]) to investigate the novel effects arising outside the RWA mainly in the rate equation generalizing equation (2) and in the fluorescence spectrum as already explained.

2.2. Analysis of the operator equations of motion

The Hamiltonian H for a single two-level atom with transition frequency ω_0 interacting with the quantized radiation field taken in dipole approximation and without the RWA can be put in the form

$$H = \frac{1}{2}\hbar\omega_0\sigma_z + \sum_{k,\lambda}\hbar\omega_k a^{\dagger}_{k,\lambda}a_{k,\lambda} - \underline{p}\cdot e.$$
 (5)

Operators $\sigma_{x,y,z}$ are Pauli spin operators while k, λ label modes of the quantized radiation field: $\omega_k = ck$. The dipole operator $p = p\sigma_x \hat{\underline{u}} = p(\sigma_+ + \sigma_-)\hat{\underline{u}}$: p is the matrix element and $\hat{\underline{u}}$ the direction of p. In the RWA, the operator $p\sigma_+$ couples only to the positive frequency part of the field while $p\sigma_-$ couples only to the negative frequency part. If we then pick-off one and only one mode k, λ from equation (5) (for example a resonant mode) there is also a total number operator commuting with H in the RWA. This is the source of Born's additional equation, (3), which he (Born) has over simplified. In this paper, outside the RWA we need to take into account the total reaction field and all of its modes. The action of this total reaction field outside the RWA is evident in equations (8) and (9) below.

As they are defined the raising and lowering Pauli operators σ_{\pm} satisfy, with σ_z the usual su(2) Lie algebra for angular momentum

$$[\sigma_+, \sigma_-] = \sigma_z, \qquad [\sigma_z, \sigma_\pm] = \pm 2\sigma_\pm. \tag{6a}$$

For the spin- $\frac{1}{2}$ two-level atoms there are also the anticommutators

$$[\sigma_+, \sigma_-]_+ = 1, \qquad [\sigma_\pm, \sigma_z]_+ = 0. \tag{6b}$$

The total field operator e, quantized in a large box of volume V, is

$$e(t) = i \sum_{k,\lambda} \underline{g}_{k,\lambda}(a_{k,\lambda}(t) - a_{k,\lambda}^{\dagger}(t))$$
(7)

and the vector coupling $\underline{g}_{k,\lambda} = (2\pi\hbar\omega_k V^{-1})^{1/2} \hat{\epsilon}_{k,\lambda}$: the $\hat{\epsilon}_{k,\lambda}$ are the unit polarization vectors.

From the Hamiltonian H, equation (5), Heisenberg's equations of motion for the atomic operators, taken in a normal ordering prescription [9, 36], are $(p = p\underline{u})$

$$\sigma_{z}^{\cdot}(t) = -2i\hbar^{-1}\underline{p}.[e^{-}(t)(\sigma_{-}(t) - \sigma_{+}(t)) + (\sigma_{-}(t) - \sigma_{+}(t))e^{+}(t)]$$
(8)

$$\sigma_{-}^{\cdot}(t) = -i\omega_{0}\sigma_{-}(t) -i\hbar^{-1}\underline{p}.[e^{-}(t)\sigma_{z}(t) + \sigma_{z}(t)e^{+}(t)] = (\sigma_{+}^{\cdot}(t))^{+}.$$
(9)

For normal ordering one splits e(t) into $e^{\pm}(t)$, positive and negative frequency part fields. In ORFT one distinguishes self-fields $e_{self}^{\pm}(t)$ from free fields $e_0^{\pm}(t)$ so that (for the one atom)

$$e^{\pm}(t) = e_0^{\pm}(t) + e_{\text{self}}^{\pm}(t).$$
 (10)

The free fields are

$$e_0^{\pm}(t) = \pm \mathrm{i} \sum_{k,\lambda} \underline{g}_{k,\lambda} \left\{ \begin{array}{c} a_{k,\lambda}(0) \\ a_{k,\lambda}^{\dagger}(0) \end{array} \right\} \mathrm{e}^{\mp \mathrm{i}\omega_k t} \tag{11}$$

while ([9, 36]; and references therein)

$$e_{\text{self}}^{\pm}(t) = \pm i \frac{2}{3} \left(\frac{\omega_0}{c}\right)^3 \underline{p} \sigma_{\mp}(t)$$

$$\pm \frac{2}{3\pi} \left(\frac{\omega_0}{c}\right)^3 \underline{p} \ln(\omega_c \omega_0^{-1}) [\sigma_+(t) - \sigma_-(t)]; \qquad (12)$$

 ω_c is the cut-off frequency for ω and can be taken as the Compton frequency.

Because $e_{\text{self}}^{\pm}(t)$, equation (12), depend on $\sigma_{\pm}(t)$, even though they depend only linearly, both of equations (8) and (9) are nonlinear operator equations. But from the definitions of the field operators and an appeal to the algebra of equations (6) one finds that the nonlinear equations (8), (9) reduce to the coupled set of *linear* operator equations

$$\sigma_{z}^{\cdot}(t) = -\gamma (1 + \sigma_{z}(t)) - 2\hbar^{-1}\underline{p} \cdot \\ \times \sum_{k,\lambda} \underline{g}_{k,\lambda} [a_{k,\lambda}^{\dagger}(0)\sigma_{-}(t)e^{i\omega_{k}t} \\ -\sigma_{-}(t)a_{k,\lambda}(0)e^{-i\omega_{k}t} + \text{h.c.}]$$
(13)

$$\sigma_{-}^{\cdot}(t) = (-\frac{1}{2}\gamma - i\omega_{0}^{\prime})\sigma_{-}(t) + (\frac{1}{2}\gamma - i\Delta_{0})\sigma_{+}(t)$$
$$-\hbar^{-1}\underline{p} \cdot \sum_{k,\lambda} \underline{g}_{k,\lambda} [a_{k,\lambda}^{\dagger}(0)\sigma_{z}(t)e^{i\omega_{k}t}$$
$$-\sigma_{z}(t)a_{k,\lambda}(0)e^{-i\omega_{k}t}] = (\sigma_{+}^{\cdot}(t))^{+}$$
(14)

and γ is the A-coefficient: ω'_0 is the atomic frequency ω_0 shifted by the *ordinary* vacuum ($\omega'_0 = \omega_0 - \Delta_0$; $\Delta_0 = \gamma \pi^{-1} \ln(\omega_c/\omega_0)$ [6,7,9] and $\Delta_0 \omega_o^{-1} = O(\gamma \omega_0^{-1})$).

From equations (13), (14) we first of all derive the equation for the mean atomic inversion $r_3(t) = \langle \sigma_z(t) \rangle$ and thus derive the generalization of equation (2) to the squeezed vacuum: $|\rangle$ is here the initial state of the field and atom together. A formal integration of the operator equation (14) yields

$$\sigma_{-}(t) = \sigma_{-}(0)e^{-(\frac{t}{2}\gamma + i\omega_{0})t}$$

$$+ \int_{0}^{t} e^{-(\frac{1}{2}\gamma + i\omega_{0}')(t-t')} \left\{ (\frac{1}{2}\gamma - i\Delta_{0})\sigma_{+}(t') -\hbar^{-1}\underline{p} \cdot \sum_{k,\lambda} \underline{g}_{k,\lambda} [a_{k,\lambda}^{\dagger}(0)\sigma_{z}(t')e^{i\omega_{k}t'} -\sigma_{z}(t')a_{k,\lambda}(0)e^{-i\omega_{k}t'}] \right\} dt' = (\sigma_{+}(t))^{+}$$
(15)

and if this result is substituted into equation (13) one finds that (compare [9, 49])

$$\sigma_z(t) = -\gamma (1 + \sigma_z(t)) + \mathcal{A} + \mathcal{B} + \mathcal{C}.$$
(16)

The *t*-dependent operators \mathcal{A}, \mathcal{B} and \mathcal{C} are

$$\mathcal{A} = -2\hbar^{-1}\underline{p} \cdot \sum_{\boldsymbol{k},\lambda} \underbrace{g_{\boldsymbol{k},\lambda}}_{\boldsymbol{k},\lambda}(0) [\sigma_{-}(0)e^{-\Gamma_{0}t} \\ -\sigma_{+}(0)e^{-\Gamma_{0}^{*}t}]e^{i\omega_{\boldsymbol{k}}t} + \text{h.c.} \}$$
(17*a*)
$$\mathcal{B} = -2\hbar^{-1}\underline{p} \cdot \sum_{\boldsymbol{k},\lambda} \underbrace{g_{\boldsymbol{k},\lambda}}_{\boldsymbol{k},\lambda}$$
(17*a*)
$$\times \int_{0}^{t} \{a_{\boldsymbol{k},\lambda}^{\dagger}(0)[(\frac{1}{2}\gamma - i\Delta_{0})\sigma_{+}(t')e^{-\Gamma_{0}(t-t')} \\ -(\frac{1}{2}\gamma + i\Delta_{0})\sigma_{-}(t')]e^{-\Gamma_{0}^{*}(t-t')}]e^{i\omega_{\boldsymbol{k}}t} + \text{h.c.} \}dt'$$
(17*b*)

and

$$\begin{aligned} \mathcal{C} &= -2\hbar^{-2}\underline{p}\,\underline{p}\sum_{k,\lambda}\sum_{k',\lambda'}\underline{g}_{k,\lambda}\underline{g}_{k',\lambda'} \\ &\times \int_{0}^{t} \{a_{k,\lambda}^{\dagger}(0)\sigma_{z}(t')a_{k',\lambda'}(0)e^{-\Gamma_{0}(t-t')+i(\omega_{k}t-\omega_{k'}t')} \\ &+ a_{k',\lambda'}^{\dagger}(0)\sigma_{z}(t')a_{k,\lambda}(0)e^{-\Gamma_{0}(t-t')-i(\omega_{k}t-\omega_{k'}t')} \\ &- a_{k,\lambda}^{\dagger}(0)a_{k',\lambda'}^{\dagger}(0)\sigma_{z}(t')e^{-\Gamma_{0}(t-t')+i(\omega_{k}t+\omega_{k'}t')} \\ &- \sigma_{z}(t')a_{k',\lambda'}(0)a_{k,\lambda}(0)e^{-\Gamma_{0}(t-t')-i(\omega_{k}t+\omega_{k'}t')} \\ &+ \text{h.c.}\}\,dt' \end{aligned}$$
(17c)

where $\Gamma_0 = \frac{1}{2}\gamma + i\omega'_0$. These somewhat monstrous operator expressions are complete so far; and their expectation values are to be taken with respect to the initial state of the atom and the initial state of the field which is a squeezed vacuum field. Within the RWA the same problem is negotiated in [9] and as there we shall need a model for a *three-dimensional* modeto-mode correlated squeezed vacuum such as might perhaps be created by an optical parametric amplifier (OPA). Our choice here is the same isotropic three-dimensional model of the *correlated* squeezed vacuum introduced in [9]. This is based on expectation values of pairs of free-field operators $\langle a_{k,\lambda}^{\dagger}a_{k',\lambda'}^{\dagger}\rangle$ and $\langle a_{k,\lambda}^{\dagger}a_{k',\lambda'}^{\dagger}\rangle$ for each mode pair $(k, \lambda), (k', \lambda')$.

As shown in [9] this isotropic model, together with similar specifications for three-dimensional squeezed vacua of different anisotropic geometries, coincide in results with those adopted more *ad hoc* in [57]. Following our work in [9] we shall therefore assume that the vacuum is characterized by

$$\sum_{\lambda,\lambda'} \hat{\epsilon}_{k,\lambda} \hat{\epsilon}_{k',\lambda'} \langle a_{k,\lambda}^{\dagger} a_{k,\lambda} \rangle = N_k (kk')^{-1} \delta(k-k') (U - \hat{k} \hat{k})$$
(18a)

$$\sum_{\lambda,\lambda'} \hat{\epsilon}_{k,\lambda} \hat{\epsilon}_{k',\lambda'} \langle a_{k,\lambda}^{\top} a_{k',\lambda'}^{\top} \rangle$$

= $M_{k,k'}^{*} (kk')^{-1} \delta(k+k'-2k_p) (U-\hat{k}\hat{k})$ (18b)

while, as befits the vacuum, squeezed or unsqueezed,

$$\langle a_{k,\lambda} \rangle = \langle a_{k,\lambda}^{\dagger} \rangle = 0.$$
 (18c)

The real numbers N_k and the complex numbers $M_{k,k'}^*$, depending only on wavenumbers for isotropy, now characterize this vacuum mode pair by mode pair. In practice, we shall work with N_k and the simpler $M_{k,k'}^* = M_k^*$ as characterizing it. For minimum uncertainty $|M_k|^2 = N_k(N_k+1)$ (e.g. [9,57] and references therein).

In these expressions (18a)-(18c) U is the unit tensor and \hat{k} is the direction of the wavevector $k = k\hat{k}$ with dyadic $\hat{k}\hat{k}$; then k is the mode wavenumber, $\omega = ck$ is the mode frequency and in particular $2\omega_p \equiv (2k_p)c$ is the frequency of the field pumping the squeezed vacuum (the OPA pump field). Evidently the δ -function in equation (18b) conserves energy; but momentum is *not* conserved (conservation of both energy and momentum together leads to a one-dimensional not a three-dimensional vacuum [9, 58]). Missing momenta are therefore supposed to be provided at the crystal downconverting the pump. Comments on conservation of energy, momenta and polarization (or otherwise) are given in [9,58]; and results for geometries other than isotropic are also given there. As was noted these results overlap with expressions adopted in [57].

Equation (16) is now to be traced over $|\rangle \equiv |\text{field}\rangle |\text{atom}\rangle$, and the resultant expectations $\langle \text{field} | \cdots | \text{field} \rangle$ are given by equations (18*a*)–(18*c*). However, the initial state $|\text{atom}\rangle$, for the atom, can remain arbitrary. The squeezed vacuum property equation (18*c*) immediately eliminates \mathcal{A} which therefore vanishes from the theory. For the terms in \mathcal{B} the substitution there of the integral form, equations (15) for $\sigma_{\mp}(t)$ makes these terms in \mathcal{B} of order $O(e^4)$: for present purposes we need to keep corrections $O(\gamma \omega_0^{-1})$ outside the RWA which are $O(e^2)$. So we drop these $O(e^4)$ terms (i.e. drop $O(\gamma^2 \omega_0^{-2})$) and eliminate \mathcal{B} .

The terms in C remain significant. We first use the unequal time commutation relation between the free-field operators $e_0^{\pm}(t)$ and arbitrary atomic operators (called $\rho(t')$ say) at different times t and t'. This is [49,59]

$$[e_0^{\pm}(t), \rho(t')] = 0 \tag{19}$$

under the condition t > t'. At this point, in order to isolate the free-field expectation values appearing in equations (18*a*), (18*b*) we must adopt the atom-field

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decorrelation approximation called (i) earlier: otherwise we gain a large system of coupled equations extending the equation (2). As explained this decorrelation is of the form $\langle \hat{A}\hat{F} \rangle \approx \langle \hat{A} \rangle \langle \hat{F} \rangle$ for atom and field operators \hat{A} and \hat{F} respectively. For black-body fields [49, 50] it is valid for weak fields and *short times* only. Observable corrections were noted in [50]. Investigation of comparable effects in the context of this analysis for the squeezed vacuum is not complete yet and will be reported in due course.

We thus make the atom-field decorrelation and shall assume the Markov approximation. It is known [60] that in the context of spontaneous emission into the normal vacuum there are additional terms in the decay at large times $t > \gamma^{-1}$ of the form $\gamma \omega_0^{-1} (\omega_0 t)^{-2} \gtrsim \gamma^3 \omega_0^{-3}$. Here we are concerned with outside the RWA at first order in $\gamma \omega_0^{-1}$, so we adopt the Markov approximation on this basis.

With the matter-field decorrelation and Markov approximation we find

$$\langle \sigma_{z}^{\prime}(t) \rangle = -\gamma (1 + (1 + 2N) \langle \sigma_{z}(t) \rangle) -\gamma (F(M) e^{-2i\omega_{p}t} + c.c.) \langle \sigma_{z}(t) \rangle$$
(20)

which is the generalization of the rate equation (2) we are looking for. The number F(M) is given by

$$F(M) = \{(2\omega_p - \omega'_0)/\omega'\}^3 M (2\omega_p - \omega'_0) + \{(2\omega_p + \omega'_0)/\omega'_0\}^3 M (2\omega_p + \omega'_0) - i\pi \mathcal{P} \int_0^\infty M(\omega_k) (\omega_k/\omega'_0)^3 \{(\omega'_0 - \omega_k + 2\omega_p)^{-1} - (\omega'_0 - \omega_k - 2\omega_p)^{-1}\} d\omega_k$$
(21)

and $M(\omega) = M_k$, $k = \omega c^{-1}$. Likewise, $N = N(\omega'_0)$ and $N(\omega)$ is N_k at $k = \omega c^{-1}$; the \mathcal{P} means principal value integral and provides (compare [9]) a form of renormalization.

With $\langle \sigma_z(t) \rangle$ identified as $r_3(t)$ one sees immediately that equation (20) is the rate equation, (2), to which the terms in F(M) are added once \bar{n} there is identified with N: in fact [9] $\bar{n} \equiv N(\omega'_0)$ for black-body radiation, for example. Thus the new feature is indeed the term in F(M) and its c.c. and these terms are entirely due to the inclusion of corrections outside the RWA (the anti-resonant terms) in the presence of the squeezed vacuum. Note that $M(\omega) = 0$ for black-body radiation so the RWA has no effect on the rate equation in this case as was recognized [49, 50]. Equation (20) is thus the Einstein rate equation modified by corrections outside the RWA induced entirely by the squeezed vacuum. We show that they include an oscillation even in the steady state $\lim_{t\to\infty} \langle \sigma_z(t) \rangle$.

For simplicity we choose exact resonance $\omega_p = \omega'_0$ for the pump of the squeezed vacuum and take $M(\omega) \equiv M_k = M$ (constant) in the range $\omega_1 \leq \omega'_0 \leq \omega_2$ and M = 0 elsewhere. Then the generalized rate equation, (20), is

$$r'_{3}(t) = -\gamma - \gamma [(1+2N) + 2|M|(1+\Delta_{M}^{2})^{1/2} \cos(2\omega_{0}'t-\delta)]r_{3}(t)$$
(22)

after the identification $r_3(t) \equiv \langle \sigma_z(t) \rangle$. The phase δ is

$$\delta = \phi + \tan^{-1} \Delta_M \tag{23}$$

$$\Delta_M = \pi^{-1} \left[\ln \left(\frac{\omega'_0 + \omega_k}{3\omega'_0 - \omega_k} \right) \right]_{\omega_2}^{\omega_1}$$
(24)

and $M = |M|e^{i\phi}$. When $M \equiv 0$, equation (22) is equation (2) with $\bar{n} \equiv N$, so it is the parameter M of the squeezed vacuum, outside the RWA, which provides the new features.

2.3. Analysis of the generalized rate equation

At first sight the initial radiation rate $-\frac{1}{2}r_3(0)$ is $\gamma[1 + N + |M|(1 + \Delta_M^2)^{1/2} \cos \delta]$ for an initially inverted atom $(r_3(0) = +1)$. For $|M| = \sqrt{N(N+1)}$ and $\delta = 0$ (for example), $\gamma[1 + N + |M|(1 + \Delta_M^2)^{1/2} \cos \delta] > \gamma(1 + 2N) > \gamma(1 + N)$ (case of black-body radiation) and the radiation rate at t = 0 is thus enhanced. However, this enhancement at t = 0 would in practice be scarcely observable since the rate is actually modulated at the very high frequencies $2\omega_0'$. The role of the phase δ and thus of ϕ is also significant. There is no particular reference phase for ϕ as there would be if the external driving field was a coherent driving field of frequency ω_p . Instead ϕ is determined by the processes of the OPA: we can expect to consider it a random phase (or a dynamical phase, as recently suggested in [61]), but in any realization it will take a particular value.

The solution of equation (22) when $|M| \neq 0$ is

$$r_{3}(t) = e^{-\gamma(1+2N)t} e^{-b\sin(2\omega'_{0}t-\delta)} \left\{ r_{3}(0)e^{-b\sin(\delta)} -\gamma \int_{0}^{t} dt' e^{\gamma(1+2N)t'+b\sin(2\omega'_{0}t'-\delta)} \right\}.$$
(25)

Comparison with equation (4) will show that because $|M| \neq 0$ in the squeezed vacuum, terms of order $b \equiv \gamma |M| \sqrt{1 + \Delta_M^2} / \omega'_0$ (in which |M| = O(1)) have emerged from the anti-resonant terms outside the RWA.

The integral in equation (25) is expressible in terms of a series in modified Bessel functions $I_n(b)$. We give the result for the steady state taking $\delta = 0$ for simplicity. This result is

$$r_{3}(\infty) = -\gamma (2\omega_{0}')^{-1} e^{-b \sin 2\omega_{0}' t} \left\{ \frac{1}{\alpha} I_{0}(b) + 2\operatorname{Re} \sum_{n=1}^{\infty} \frac{(\alpha - \operatorname{in}) I_{n}(b)}{(\alpha^{2} + n^{2})} e^{\operatorname{in}(2\omega_{0}' t - \frac{\pi}{2})} \right\}$$
(26)

in which $\alpha \equiv \gamma (1+2N)/2\omega'_0$. The factor $e^{-b \sin 2\omega'_0 t}$ also has an expression in terms of the $I_n(b)$ and the non-oscillatory component easily proves to be

$$(r_{3}(\infty))_{0} = -\frac{1}{1+2N}I_{0}^{2}(b)$$

= $-\frac{1}{1+2N}\left(1+\frac{1}{2}\left(\frac{\gamma|M|\sqrt{1+\Delta_{M}^{2}}}{\omega_{0}'}\right)^{2}+\cdots\right).$ (27)

In addition to this is the steady oscillatory component as is shown in figures 1 and 2: notice that the *onset* of the oscillatory behaviour is affected by the phase δ , figures 1(*a*), (*b*).

The expression (26) for $r_3(\infty)$ is plainly of period $T = 2\pi (2\omega'_0)^{-1} = \pi (\omega'_0)^{-1}$ where $\omega'_0 = \omega_0 - \Delta_0$ and Δ_0 is the ordinary vacuum radiative shift (i.e., $r_3(\infty)$ is of period 2π if plotted against normalized time $\tau \equiv 2\omega'_0 t$ (figure 1) and of period $2\pi/(\frac{2\omega_0}{\gamma}) \equiv 2\pi(\frac{\gamma}{2\omega_0})$ if plotted against normalized time $\tau_0 \equiv \gamma t$ (figure 2). The effect of this vacuum shift is clear for values of $\gamma/\omega_0 \sim 10^{-2}$ (figure 2(a)). It will be

where



Figure 1. The atomic inversion $r_3(t)$ against the normalized time $\tau = 2\omega'_0 t$ for $\gamma/(2\omega_0) = 10^{-2}$ and phase value $\delta = 0$ and for an initially inverted atom $(r_3(0) = +1)$ in the case of the squeezed vacuum with N = 1, $|M| = \sqrt{N(N+1)}$: the dotted curve represents the solution in the RWA (equation (4)), the full curve represents the solution without the RWA (equation (25)). Note $r_3(t)$ is only shown in the interval $-0.36 < r_3(t) < -0.31$ (*b*) as (*a*) but for the phase value $\delta = \pi$.

interesting to see whether it is *this* vacuum shifted frequency which is eventually observed, the shift being $O(\gamma \omega_0^{-1})$ as has been described already. Notice from the figures that the amplitude of the oscillation at the frequency $2\omega'_0$ is $O(\gamma \omega_0^{-1})$ as explained. Notice in figures 1, $2(a) \gamma \omega_0^{-1}$ is taken to be 10^{-2} (not the more realistic values 10^{-6} or 10^{-7} for optical frequencies)—but see our comment in section 5 where the ratio $\gamma \omega_0^{-1}$ could be larger for an electron spin- $\frac{1}{2}$ system.

3. The polarization

We have thus solved for the inversion $r_3(t) \equiv \langle \sigma_z(t) \rangle$ taken from equation (13). For the operator equation (14), we can follow the same procedure by formally integrating equation (13) for $\sigma_z(t)$ and inserting the result into (14); the use of the squeezed vacuum correlation functions (18) and of the relation (19) then leads finally to the following equations:

$$\langle \sigma_{-}^{\prime}(t) \rangle = (-\Gamma - i(\omega_{0} - \Delta_{N})) \langle \sigma_{-}(t) \rangle$$

$$+ (\Gamma - i\delta_{N}) \langle \sigma_{+}(t) \rangle - \gamma m_{1} \langle \sigma_{+}(t) \rangle e^{-2i\omega_{p}t}$$

$$- \gamma m_{3}^{*} \langle \sigma_{+}(t) \rangle e^{2i\omega_{p}t} + (\gamma m_{1}^{*} e^{2i\omega_{p}t}$$

$$- \gamma m_{3} e^{-2i\omega_{p}t}) \langle \sigma_{-}(t) \rangle$$

$$= \langle \sigma_{+}^{*}(t) \rangle^{*}.$$

$$(28)$$



Figure 2. The steady 'oscillatory' atomic inversion $r_3(\infty)$, equation (26), in the squeezed vacuum case N = 1, $|M| = \sqrt{2}$, $\delta = 0$ outside the RWA against the normalized time $\tau_0 = \gamma t$ and for values of $(\frac{\gamma}{2\omega_0}) = 10^{-2}$, 10^{-4} ; (*a*), (*b*) respectively. Full and broken curves represent the (ordinary) vacuum shift $\Delta_0 = 0$ and $\Delta_0 = \gamma$.

In these equations of motion $\omega'_0 \equiv \omega_0 - \Delta_0$ and Δ_0 is the radiative shift of the ordinary vacuum as before; $\Delta_N \equiv \gamma \pi^{-1} \mathcal{P} \int_0^\infty d\omega_k (\omega_k/\omega'_0)^3 N(\omega_k)$ $[(\omega'_0 - \omega_k)^{-1} + (\omega'_0 + \omega_k)^{-1}]$ is the corresponding shift due to the squeezed vacuum and $\delta_N = \Delta_0 + \Delta_N$ is the total radiative shift; $\Gamma = \frac{1}{2}\gamma(1+2N)$; $\gamma m_1 = \gamma((\omega_p + \Delta)/\omega'_0)^3 M(\omega_p + \Delta) - i\Delta_{M_1}$; $\gamma m_3 = \gamma((2\omega_p + \omega'_0)/\omega'_0)^3 M(2\omega_p + \omega'_0) + i\Delta_{M_3}$; $\Delta_{M_1} \equiv \gamma \pi^{-1} \mathcal{P} \int_0^\infty d\omega_k (\omega_k/\omega'_0)^3 M(\omega_k) (\omega_k - \omega'_0)^{-1}$; $\Delta_{M_3} \equiv \gamma \pi^{-1} \mathcal{P} \int_0^\infty d\omega_k (\omega_k/\omega'_0)^3 M(\omega_k) (3\omega'_0 - \omega_k)^{-1}$; and $\Delta = \omega_p - \omega'_0$ is the squeezed vacuum detuning parameter.

As compared with similar analysis for a thermal field assuming the RWA [49], there are two new features in equations (28): one is that $\langle \sigma_{-}^{-}(t) \rangle$ couples to $\langle \sigma_{+}(t) \rangle$ through the $(\Gamma - i\delta_{N})\langle \sigma_{+}(t) \rangle$ term, an anti-resonant term brought in by correction outside the RWA [36]: the second is the $-\gamma m_1 \langle \sigma_{+}(t) \rangle e^{-2i\omega_p t}$ induced by the squeezed vacuum $M \neq$ 0. Note that in m_1 , the parameter $M(\omega_p + \Delta)$ is, strictly speaking, renormalized by the $-i\Delta_{M_1}$, [9]. However, we simplify m_1 to M here and look for envelope solutions

$$\langle S_{\pm}(t) \rangle = \langle \sigma_{\pm}(t) \rangle e^{\pm i\omega_p t}.$$
 (29)

Then, since both terms in m_3 , m_3^* become highly oscillatory, or else by simply ignoring the squeezing produced at the higher frequency through $M(\omega)$ with $\omega \approx 3\omega_p$, we reach just

$$\begin{aligned} \langle S_{-}^{\cdot}(t) \rangle &= -(\Gamma - i\Delta) \langle S_{-}(t) \rangle - \gamma M \langle S_{+}(t) \rangle \\ &+ [(\Gamma - i\delta_{N}) \langle S_{+}(t) \rangle + \gamma M^{*} \langle S_{-}(t) \rangle] e^{2i\omega_{p}t} \\ &= \langle S_{+}^{\cdot}(t) \rangle^{*}. \end{aligned}$$
(30)

In the RWA anti-resonant terms in $e^{2i\omega_p t}$ are dropped so that the RWA solutions $\langle S_{\pm}(t) \rangle_0$ solve the system

$$\langle S_{-}(t) \rangle_{0} = -(\Gamma - i\Delta) \langle S_{-}(t) \rangle_{0} - \gamma M \langle S_{+}(t) \rangle_{0}$$

$$\langle S_{+}(t) \rangle_{0} = -(\Gamma + i\Delta) \langle S_{+}(t) \rangle_{0} - \gamma M^{*} \langle S_{-}(t) \rangle_{0}.$$

$$(31)$$

In the resonant case ($\Delta = 0$) the solution of (31) for the atomic dipoles $\langle S_x \rangle = \langle S_+ + S_- \rangle$ and $\langle S_y \rangle = i \langle S_+ - S_- \rangle$ shows there are two different (transverse) decay constants [43]. For $\Delta \neq 0$ the solutions of (31) were reported in [62] which show that for $\Delta > \gamma |M|$ the atomic dipoles oscillate with two different frequencies, namely ($\omega'_0 + \theta$) and ($\omega_p + \Delta - \theta$), where $\theta \equiv \gamma^2 |M|^2/2\Delta$ is a squeezinginduced Bloch–Siegert shift [62] (an analogue of the optical Bloch–Siegert shift ($\gamma^2/8\omega'_0$) in a normal vacuum outside RWA (cf [63]).

It is worth noting that equations similar to (31) can be reached within the context of the study of the spontaneous dynamics of a two-level atom in the presence of a phaseconjugating mirror (PCM), where the parameter M in this case is related to the vacuum field modes that are conjugated and reflected by the PCM to the atom [64,65]

4. The fluorescence spectrum

We now calculate the fluorescence spectrum outside the RWA according to equations (30) working up to the first correction in $O(\gamma/\omega'_0)$. To do this we substitute $\langle S_{\pm}(t) \rangle_0$ (the solutions of (31) in the RWA [62]) for $\langle S_{\pm}(t) \rangle$ in the terms multiplied by $e^{\pm 2i\omega_p t}$ in the more exact equations (30): so $\langle S_{\pm}(t) \rangle_0$ is a zeroth-order iteration $O(\gamma/\omega'_0)$. The first-order iterations $\langle S_{\pm}(t) \rangle_1$ then solve the system

$$\begin{aligned} \langle S_{-}^{\cdot}(t) \rangle_{1} &= -(\Gamma - i\Delta) \langle S_{-}(t) \rangle_{1} - \gamma M \langle S_{+}(t) \rangle_{1} \\ &+ [(\Gamma - i\delta_{N}) \langle S_{+}(t) \rangle_{0} + \gamma M^{*} \langle S_{-}(t) \rangle_{0}] e^{2i\omega_{p}t} \\ &= \langle S_{+}^{\cdot}(t) \rangle_{1}^{*}. \end{aligned}$$
(32)

Here we only report the solutions for the case $\Delta > \gamma |M|$, (other cases of $\Delta \leq \gamma |M|$ can be similarly considered). These solutions for $\Delta > \gamma |M|$ are

$$\langle \sigma_{+}(t) \rangle_{1} = (a_{0} + a_{1}) e^{-(\Gamma - i\omega_{p} - iw)t} + (b_{0} + b_{1}) e^{-(\Gamma - i\omega_{p} + iw)t} + (c_{1}e^{-(\Gamma + i\omega_{p} - iw)t} + c_{2}e^{-(\Gamma + i\omega_{p} + iw)t} + c_{3}e^{-(\Gamma - 3i\omega_{p} + iw)t} + c_{4}e^{-(\Gamma - 3i\omega_{p} - iw)t}) = \langle \sigma_{-}(t) \rangle_{1}^{*}$$
(33)

where $a_0, b_0 = [(w \mp \Delta)\langle \sigma_+(0)\rangle \pm i\gamma M^*\langle \sigma_-(0)\rangle]/2w;$ $w = \sqrt{\Delta^2 - \gamma^2}|M|^2 \approx \Delta - \theta$ and a_1, b_1, c_i (i = 1-4) are numbers calculated to $O(\gamma/\omega'_0)$ outside the RWA. Note that the terms in a_0, b_0 represent the zeroth-order solutions $\langle \sigma_{\pm}(t) \rangle_0$ in the RWA.

Based on (33) or its equivalent operator form the resultant fluorescence spectrum in the steady state, $G(\omega) \equiv$ Fourier transform of $\lim_{t\to\infty} \langle \sigma_+(t+\tau)\sigma_-(t) \rangle_1 + \text{c.c.}$, can be calculated either by appealing to the quantum regression theorem [66] or by direct methods [37]. For simplicity we take M = real and $G(\omega)$ is given, up to $O(\gamma \omega_0^{(-1)})$, by

$$G(\omega) = G_0(\omega) + G_1(\omega) \tag{34}$$

where

$$G_{0}(\omega) = \frac{\gamma N}{2(\Delta - \theta)} \left[\frac{(2\Delta - \theta)}{\Gamma^{2} + (\omega - (\omega_{0}' + \theta))^{2}} + \frac{\theta}{\Gamma^{2} + (\omega - (2\omega_{p} - \omega_{0}' - \theta))^{2}} \right]$$
(35)

is the two Lorentzian's structure calculated within the RWA [44] to O(1) in terms of $\gamma \omega_p^{-1}$ (this order replacing the order $\gamma \omega_0^{'-1}$, but $\omega_p \approx \omega_0^{'}$). The second quantity in (34), $G_1(\omega)$, represents four additional anti-resonant terms $O(\gamma \omega_p^{-1})$ —all of which vanish if M = 0. Two of these corrections are of 'resonant dispersive' type and induce asymmetric corrections $O(\gamma \omega_p^{-1})$ to each Lorentzian in (35). These two terms are

$$\frac{\gamma N M(\delta_N \omega_p^{-1})}{2(1+2N)(\Delta-\theta)} \left[\frac{-(\omega - (\omega'_0 + \theta))}{\Gamma^2 + (\omega - (\omega'_0 + \theta))^2} + \frac{(\omega - (2\omega_p - \omega'_0 - \theta))}{\Gamma^2 + (\omega - (2\omega_p - \omega'_0 - \theta))^2} \right].$$
(36)

The remaining two terms in $G_1(\omega)$ are not resonant and are

$$\frac{\gamma NM}{2(1+2N)(\Delta-\theta)} \left[\frac{-((\delta_N+\theta)\omega_p^{-1})(\omega+\omega_0'+\theta)}{\Gamma^2+(\omega+\omega_0'+\theta))^2} + \frac{((2\Delta-\theta+\delta_N)\omega_0^{-1})(\omega+2\omega_p-\omega_0'-\theta)}{\Gamma^2+(\omega+2\omega_p-\omega_0'-\theta)^2} \right].$$
 (37)

For actual detection, of the dispersive structure of (36), it thus remains to subtract the two Lorentzians in (35) from the observed spectrum and look for the corrections due to each of the resonant dispersive terms $O(\gamma \omega_p^{-1})$.

It is relevant that the comparable (exact) steady state spectrum outside the RWA for black-body radiation, rather than for the squeezed vacuum, is

$$G(\omega) = \frac{\gamma N (1 - \frac{2\delta_N}{\omega + \omega_0})}{[\Gamma^2 (\frac{2\omega}{\omega + \omega_0})^2 + (\omega - \omega_0 + \frac{2\omega_0 \delta_N}{\omega + \omega_0})^2]}$$
(38a)
$$\approx \frac{\gamma N}{2(\omega_o - \theta_N)} \left[\frac{(2\omega_0 - \theta_N)}{\Gamma^2 + (\omega - (\omega_0 - \theta_N))^2} - \frac{\theta_N}{\Gamma^2 + (\omega + (\omega_0 - \theta_N))^2} \right]$$
(38b)

where in (38*b*) we have approximated $\sqrt{\omega_0^2 - (\Gamma^2 + \delta_N^2)} \approx \omega_0 - \theta_N$ and $\theta_N = \frac{(\Gamma^2 + \delta_N^2)}{2\omega_0} = O(\frac{\gamma^2}{\omega_0}) \approx O(1)$ is a generalized Bloch–Siegert shift for the black-body field case. This shift θ_N reduces to the optical Bloch–Siegert shift ($\gamma^2/8\omega'_0$) in normal vacuum [63] if N = 0 and $\Delta_0 \simeq 0$. Note that the familiar $G(\omega)$ for the black-body field in the RWA [42,49] is regained if θ_N is dropped. Note too that the second term in (38*b*) is not of resonant type and is $O(\frac{\theta_N}{\omega_0 - \theta_N}) = O(\frac{\gamma^2}{\omega_0^2})$ which is still negligible at $O(\gamma \omega_0^{-1})$. Otherwise the first term in (38*b*) calculated outside the RWA, which is a Lorentzian centred at $\omega = \omega_0 - \theta_N$, is comparable with the first term in (35) for the squeezed vacuum calculated inside the RWA. Thus, in the black-body radiation case the fluorescence spectrum *is* affected outside the RWA and in this it is unlike the Einstein rate equation, (2), as noted before.

5. Summary

Thus by using ORFT we have been able to give for a broad-band isotropic squeezed vacuum a comprehensive analysis, exact within our present understanding of quantum mechanics, to $O(\gamma \omega_n^{-1})$ for both the generalized rate equation corresponding to the Einstein rate equation, (2), and for the fluorescence spectrum. The results are specific to the isotropic vacuum only in detail and comparable results should arise for all geometries of the vacuum. The particular effects $O(\gamma \omega_n^{-1})$ arising outside the RWA are small but evidently of considerable fundamental interest. Perhaps the oscillatory behaviour in the steady state arising in the rate equation at $O(\gamma \omega_p^{-1})$ is the most striking of these results; while concerning the *smallness* of these quantities $O(\gamma \omega_n^{-1})$ and their actual measurement it is worth noting again (and see section 1) that: (i) these terms are of the same order as that of the radiative (Lamb) shifts measured since 1947 [20]; (ii) the asymmetric dispersive structure correction $O(\gamma \omega_p^{-1})$ induced by equation (36) into the spectrum of two Lorentzians (35) is essentially a frequency shift of somewhat Bloch-Siegert type, and Bloch-Siegert shifts have been measured [67], and, (iii) both the behaviour of $r_3(\infty)$ and the corrections (36) to (35) are resonant phenomena, and the effects of the other off-resonant levels in a real multi-level atom should, broadly speaking, be of less physical significance.

One might also explicitly draw attention to the formal connection, even as actual physics, between an atomic twolevel system and a spin- $\frac{1}{2}$ electron. In the latter the splitting between two spin states can be adjusted by an external d.c. magnetic field. This suggests that a spin- $\frac{1}{2}$ electron in such a d.c. field immersed in the squeezed vacuum field will produce the effects decribed in this paper. The ratio $\gamma \omega_0^{-1}$ can be made larger simply by adjusting the strength of the d.c. field. As experimental techniques continue to be refined we thus can look forward to some actual observations in suitably chosen broad-band squeezed vacua of the novel behaviours predicted in this paper. For example, it may be now worth referring to the technique called 'spectrochronography' which combines spectral and temporal resolutions for an investigation of transient light-emitting excitation in matter [68].

Note added in proof. Concerning the *smallness* of the oscillations $O(\gamma/\omega_p)$ in the steady state predicted in this paper and particularly with reference to the points (i), (ii) and (iii) made in section 5, the ratio $(\gamma/\omega_p) \sim 10^{-6}$ for optical frequencies ω_p and proves to be of the same order for high Rydberg microwave transitions (section 1). However, for the microwave Lamb line frequency ~ 1040 MHz, Power [18, p 115] points out that (γ/ω_p) in our notation is about 1/10—some 5 times the values adopted for figures 1 and 2 and quite large enough (in itself at least) for a successful experimental observation.

References

- Sargent M, Scully M O and Lamb W E 1974 Laser Physics (New York: Addison-Wesley)
- [2] Scully M O and Zubairy M S 1997 *Quantum Optics* (Cambridge: Cambridge University Press) ch 5
- [3] Mandel L and Wolf E 1995 Optical Coherence and Quantum Optics (Cambridge: Cambridge University Press) ch 15

- [4] Allen L and Eberly J H 1975 Optical Resonance and Two-Level Atoms (New York: Wiley)
- [5] Bullough R K, Thompson B V, Nayak N and Bogoliubov N M 1995 Studies in Classical and Quantum Nonlinear Optics ed O Keller (New York: Nova) pp 87–132
- [6] Bullough R K 1973 Coherence and Quantum Optics ed L Mandel and E Wolf (New York: Plenum) pp 121–56
- [7] Ackerhalt J R, Eberly J H and Knight P L 1973 Coherence and Quantum Optics ed L Mandel and E Wolf (New York: Plenum) pp 635–44
- [8] Milonni P W 1976 Phys. Rep. C 25 1
 See also Milonni P W 1994 The Quantum Vacuum (New York: Academic)
- [9] Hassan S S, Wahiddin M R, Saunders R and Bullough R K 1995 Physica A 215 556
 - Hassan S S, Wahiddin M R, Saunders R and Bullough R K 1995 *Physica* A **219** 482
- [10] Senitzky I R 1973 Phys. Rev. Lett. 31 955
 Milonni P W, Acherhalt J R and Smith W A 1973 Phys. Rev. Lett. 31 958
 Agarwal G S 1974 Phys. Rev. A 10 717
 Dalibard J, Dupont-Roc J and Cohen-Tannoudji C 1982 J. Physique 43 1617
 Puri R R 1985 J. Opt. Soc. Am. B 2 447
- [11] Hyne F and Bullough R K 1984 Phil. Trans. R. Soc. A 312
 251
- [12] Hynne F and Bullough R K 1987 Phil. Trans. R. Soc. A 321 305
- [13] Hynne F and Bullough R K 1990 Phil. Trans. R. Soc. A 330 253
- [14] Lorentz H A 1909 The Theory of Electrons (Leipzig) (Reprinted in 1952) (New York: Dover)
- [15] Dirac P A M 1947 The Principles of Quantum Mechanics 3rd edn (Oxford: Oxford University press) p 290, equation (59)
- [16] Dirac P A M 1958 The Principles of Quantum Mechanics 4th edn (Oxford: Oxford University Press)
- [17] Jackson J D 1975 Classical Electrodynamics 2nd edn (New York: Wiley)
- [18] Power E A and Zienau S 1959 Phil. Trans. R. Soc. 251 427 Power E A 1964 Introductory Quantum Electrodynamics (London: Longmans Green)
- [19] Weisskopf V S 1939 Phys. Rev. 56 72
- [20] Lamb W E Jr and Retherford R C 1947 *Phys. Rev.* 72 241
 Lamb W E Jr and Retherford R C 1950 *Phys. Rev.* 79 549
 Lamb W E Jr and Retherford R C 1951 *Phys. Rev.* 81 222
 Lamb W E Jr and Retherford R C 1952 *Phys. Rev.* 85 259
 Lamb W E Jr and Retherford R C 1952 *Phys. Rev.* 86 1014
- [21] Cohen-Tannoudji C 1992 Fundamental Systems in Quantum Optics ed J Dalibard, J M Raimond and J Zinn-Justin (Amsterdam: Elsevier) pp 1–161
- [22] Bullough R K and Hynne F 1996 Local field corrections, frequency shifts and generalized A-coefficients in dielectrics *Coherence and Quantum Optics* vol 7, ed J H Eberly, L Mandel and E Wolf (New York: Plenum) pp 615–6
- [23] Barnett S M, Huttner B and Loudon R 1992 Phys. Rev. Lett. 68 3698
- [24] Barnett S M, Huttner B, Loudon R and Matloob R 1996 J. Phys. B: At. Mol. Opt. Phys. 29 3763
- [25] Juzeliunas G 1997 Phys. Rev. A 55 R4015
- [26] Pedro de Vries and Ad Lagendijk 1998 Phys. Rev. Lett. 81 1381
 - Bullough R K and Thompson B V 1970 J. Phys.: Condens. Matter **3** 1780
- [27] Glauber R and Lewenstein M 1991 Phys. Rev. A 43 467
- [28] Milonni P W 1995 J. Mod. Opt. 42 1991
- [29] Fleischhauer M and Yelin S F 1999 *Phys. Rev.* A **59** 2427
 Bowden C M and Dowling J 1993 *Phys. Rev.* A **47** 1247
 Bowden C M and Dowling J 1994 *Phys. Rev.* A **49** 1514
- [30] Bullough R K, Obada A-S F, Thompson B V and Hynne F

Review Article

1968 Microscopic theory of the Einstein optical scattering equations *Chem. Phys. Lett.* **2** 293

- [31] Bullough R K 1967 A unified approach to molecular scattering theory 2nd Interdisciplinary Conf. on Electro-Magnetic Scattering (Amherst, Ma., USA, June 1965.) Proc. 2nd ICES (New York: Gordon and Breach) pp 537–77
- [32] Hassan S S, Batarfi H A and Bullough R K 1995 Studies in Classical and Quantum Nonlinear Optics ed O Keller (New York: Nova) pp 609–23
- [33] Hassan S S, Bullough R K, Saunders R and Batarfi H A 1995 Quantum Communications and Measurements ed Belavkin, O Hirota and R L Hudson (New York: Plenum) pp 93–9
- [34] Bullough R K, Batarfi H A, Hassan S S, Ibrahim M N and Saunders R 1996 *Proc. SPIE* **2799** 320
- [35] Hassan S S, Jarad T and Bullough R K 1999 Dispersion relations and bistability in squeezed vacua within a complex refractive index theory in preparation
- [36] Saunders R, Bullough R K and Ahmad F 1975 J. Phys. A: Math. Gen. 8 759
- [37] Hassan S S and Bullough R K 1975 J. Phys. B: At. Mol. Phys. 8 L147
- [38] Hassan S S 1976 PhD Thesis University Of Manchester
- [39] Saunders R 1973 PhD Thesis University of Manchester
- [40] Ackerhalt J R, Knight P L and Eberly J H 1973 Phys. Rev. Lett. 30 456
- Ackerhalt J R and Eberly J H 1974 *Phys. Rev.* D 10 3350
 [41] Haroche S, Goy P, Raimond J M, Fabre C and Gross M 1982 *Phil. Trans. R. Soc.* A 307 659
 Raimond J M, Goy P, Fabre C and Haroche S 1982 *Phys. Rev. Lett.* 49 117
 - Raimond J M, Goy P, Gross M, Fabre C and Haroche S 1982 Phys. Rev. Lett. **49** 1924
- [42] Agarwal G S 1974 Springer Tracts in Modern Physics vol 70 (Berlin: Springer)
- [43] Gardiner C W 1986 Phys. Rev. Lett. 56 1917
- [44] Ficek Z and Sanders B C 1994 J. Phys. B: At. Mol. Opt. Phys. 27 809
- [45] Einstein A 1917 Z. Phys. 18 121
- [46] Born M 1948 Natural Philosophy of Cause and Chance (Oxford: Oxford University Press) p 82
- [47] Louisell W H 1973 Quantum Statistical Properties of Radiation (New York: Wiley)

- [48] Lewis H R 1973 Am. J. Phys. 41 38
- [49] Hassan S S and Bullough R K 1988 Physica A 151 397
- [50] Hassan S S, Hildred G P, Puri R R and Bullough R K 1982 J. Phys. B: At. Mol. Phys. 15 2635
- [51] Dicke R H 1952 Phys. Rev. 93 99
- [52] Hildred G P, Puri R R, Hassan S S and Bullough R K 1984 J. Phys. B: At. Mol. Phys. B 17 L535
- [53] Wahiddin M R, Hassan S S and Bullough R K 1995 J. Mod. Opt. 42 171
- [54] Palma G M and Knight P L 1989 Phys. Rev. A 39 1962
- [55] Wahiddin M R 1989 *PhD Thesis* University of Manchester [56] Wahiddin M R, Hassan S S and Bullough R K 1996
- Nonlinear Opt. **16** 333 [57] Ficek Z and Drummond P D 1991 Phys. Rev. A **43** 6247
- Ficek Z and Drummond P D 1991 *Phys. Rev.* A **43** 6285
- [58] Bullough R K, Hassan S S, Saunders R and Wahiddin M R in preparation
- [59] Mollow B R 1975 J. Phys. A: Math. Gen. 8 L130
- [60] Knight P L 1977 Phys. Lett. A 61 25
 Knight P L 1977 Coherence and Quantum Optics vol 4, ed L Mandel and E Wolf (New York: Plenum) pp 635–45
- [61] D'Ariano M, Fortunato M and Tombesi P 1995 Nuovo Cimento B **110** 1127
- [62] Ficek Z 1993 J. Mod. Opt. 40 2333
- [63] Stenholm S 1972 J. Phys. B: At. Mol. Phys. 5 878
 Cohen-Tannoudji C, Dupont-Roc C and Fabre C 1973 J. Phys. B: At. Mol. Phys. 6 L214; L218
 Hannaford P, Pegg D T and Series G W 1973 J. Phys. B: At. Mol. Phys. 6 L222
 Ahmad F and Bullough R K 1974 J. Phys. B: At. Mol. Phys. 7 L147
 - Ahmad F and Bullough R K 1974 J. Phys. B: At. Mol. Phys. 7 L275
 - Swain S 1974 J. Phys. B: At. Mol. Phys. 7 2363
- [64] Milonni P W, Bochove E J and Cook R J 1989 J. Opt. Soc. Am. B 6 1932
- [65] Hendriks B H W and Nienhuis G 1989 Phys. Rev. A 40 1892
- [66] Lax M 1968 Phys. Rev. 172 350
- [67] Morand M S and Theobald G 1969 C. R. Acad. Sci., Paris B 269 503
 Arimondo E and Moruzzi G 1973 J. Phys. B: At. Mol. Phys.

6 2382

[68] Freiberg A and Saari P 1983 IEEE J. Quantum Electron. 19 622