Resonance fluorescence in intense radiation felds

1978 J. Phys. A: Math. Gen. 11 1151

(http://iopscience.iop.org/0305-4470/11/6/016)

J. Phys. A: Math. Gen., Vol. 11, No. 6, 1978. Printed in Great Britain

Resonance fluorescence in intense radiation fields

V Sethu Raman

Iowa State University, Department of Chemistry and Ames Laboratory-USDOE, Ames, Iowa 50011, USA

Received 7 November 1974, in final form 28 November 1977

Abstract. The spectral density of resonance fluorescence from a two-level atom in a monochromatic radiation field of arbitrary strength and detuning is calculated by solution of the wave equation. A basis set in which the strong coupling of the atom and the quantised incident field is diagonal is used, and the resulting infinite set of coupled equations is solved by truncating them to a set of N equations and finally proceeding to the $N \to \infty$ limit. The resulting spectral densities of the inelastically scattered field for various levels of intensity of the incident field and degree of detuning are found to be identical with those of Kimble and Mandel obtained by solution of the coupled equations of motion of the atom and the radiation field in the Heisenberg picture.

1. Introduction

The elementary problem of interaction of a two-level atom with a monochromatic near-resonant field and the spectral density of the resultant fluorescence has received much attention in recent years theoretically and experimentally. Availability of single-mode, intensity-stabilised CW lasers, and atomic systems in collision-free conditions that closely approximate two-level systems, have enabled comparison of theoretical predictions with experiments (Hartig and Walther 1973, Schuda *et al* 1974, Wu *et al* 1975, Gibbs and Venkatesan 1976). In these experiments the atoms are exposed to the single-mode, near-resonant fields of intensity that correspond to a Rabi frequency of the order of 5–10 times the radiative linewidth for the transition of interest, for durations large compared with the radiative lifetime of the excited state.

On the theoretical side, Mollow (1969), treating the incident field classically, obtained a three-peaked power spectrum by solution of the equations of motion of the atomic system factorised from the complete system of atoms and the scattered field by a Markoffian approximation. Swain (1975) by employing a continued fraction procedure solved the wave equation and has obtained the spectral densities for arbitrary intensities and detuning of the incident field. Procedures involving solution of relevant master equations (Carmichael and Walls 1976) require the Markoffian assumption of δ-correlation of the free field and the quantum regression theorem (Lax 1967) for evaluation of the two-time correlation functions of interest. The validity of these assumptions has been proved subsequently for this problem (Mollow 1975, Kimble and Mandel 1976). Kimble and Mandel (1976, 1977) have obtained the spectral density of the scattered light by solving the coupled Heisenberg equations of motion of the atom–field system and have presented the spectra for various intensities, detuning and bandwidths. They demonstrate lucidly in their 1976 paper that the quantal features of the radiation field are tested only by examining the steady-state two-time correlation functions of the field (via the spectral density of the fluorescence) and the intensity. Thus their analysis illustrates the significance of the solution and experimental verification of this basic problem. The reader should also refer to the works of Agarwal (1974) and Hassan and Bullough (1975). A complete list of references may be found in the paper by Kimble and Mandel (1976).

In this paper we treat the incident field quantally, and by employing a basis in which the strong coupling of the atom and the incident field is diagonal, we solve the Schrödinger equation for long times and weak coupling of the atom with all the modes of the electromagnetic field from which the incident modes are excluded. Stroud (1971) using the above basis obtained short-term solutions that lead to incorrect spectral densities for the experiments of interest. Since submission of the first draft of this manuscript, Smithers and Freedhoff (1975) with this strong-coupling basis have solved the on-resonant excitation problem for times very much longer than the radiative lifetime. Since there is general agreement on the zero-detuning problem, it is of interest to solve the general case of arbitrary detuning for comparison with the results of Kimble and Mandel (1976). The paper is organised as follows. Section 2 presents the method of solution of the infinite set of coupled equations. In § 3 we calculate the spectrum of the resonantly scattered field. Section 4 analyses the decay spectrum for various limiting cases of intensity and detuning. Section 5 compares the method of this paper with those of Mollow (1969, 1975), Kimble and Mandel (1976) and the 'dressed atom' method of Cohen-Tannoudji and Reynaud (1977).

2. Method of solution

We consider a two-level atom interacting with a strong, near-resonant, monochromatic field of frequency ω. The ground level 0 is located at ω_0 below the excited level 1. The basis in which the interaction V_0 of the quantised radiation field with N quanta, and the atom is diagonal may be written as

$$|\pm\rangle = \binom{C}{S}|N;0\rangle \pm \binom{S}{C}|N-1;1\rangle. \qquad (1a)$$

Similarly the states in which N quanta of scattered photons of momenta $\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n$, and $N-n$ quanta of incident field are present yield the strong-coupling basis states

$$|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm\rangle = \left[\binom{C}{S}|N-n;0\rangle \pm \binom{S}{C}|N-n-1;1\rangle\right]|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n\rangle. \qquad (1b)$$

For intense incident fields $N \gg n$, the coupling coefficients C and S in equations (1) are taken to be independent of n. The energies of these eigenstates of equation (1b) of the Hamiltonian of the atom and the incident field are given by

$$\lambda^{\pm}_{1\ldots n} = \delta_{1\ldots n} + \tfrac{1}{2}(\Delta \pm \Omega) \equiv \lambda^{\pm} + \delta_{1\ldots n}$$

where

$$\Delta = \omega - \omega_0, \qquad \Omega = (\Delta^2 + 4\kappa^2)^{1/2}$$

$$\delta_{1\ldots n} = \omega_1 + \ldots + \omega_n - n\omega$$

$$\kappa = \langle N-n; 0|V_0|N-n-1; 1\rangle;$$

C and S are related to the Rabi frequency κ and the detuning frequency Δ as

$$C = 2\kappa/2\Omega(\Delta+\Omega), \qquad S \equiv (\Delta+\Omega)/2\Omega;$$

$\omega_1, \ldots, \omega_n$ are the frequencies of the scattered quanta. The state function at time t may be written in the basis in equation (1):

$$|t\rangle = b^+(t)|+\rangle + b^-(t)|-\rangle$$
$$+\sum_{n=1}^{\infty} \int \mathrm{d}\boldsymbol{k}_1 \int \ldots \int \mathrm{d}\boldsymbol{k}_n (b^+_{1\ldots n}(t)|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; +\rangle + b^-_{1\ldots n}(t)|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; -\rangle) \qquad (2)$$

where the amplitudes $b^{\pm}_{1\ldots n}(t)$, symmetric in their indices, are given by

$$b^{\pm}_{1\ldots n}(t) = \langle \boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm|t\rangle$$

and the basis vectors $|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n\rangle$ are normalised as

$$\langle \boldsymbol{k}'_1, \boldsymbol{k}'_2, \ldots, \boldsymbol{k}'_n|\boldsymbol{k}_1, \boldsymbol{k}_2, \ldots, \boldsymbol{k}_n\rangle = \frac{1}{n!}\sum_{\mathrm{P}} \delta(\boldsymbol{k}_1 - \boldsymbol{k}'_1) \ldots \delta(\boldsymbol{k}_n - \boldsymbol{k}'_n),$$

where the subscript P means the sum runs over all permutations of the indices $1, \ldots, n$ of the $\boldsymbol{k}'$ vectors only.

Considering the interaction V of the atom 'dressed' by the incident quanta, with the modes of the electromagnetic field (from which the mode of the incident field has been excluded) as a perturbation, the Schrödinger equation in the interaction picture may be solved by a Fourier transformation (Heitler 1954). The interaction may be written as

$$V(t) = -\boldsymbol{\mu} \cdot \boldsymbol{E} = \boldsymbol{\mu}(t) \cdot \int \mathrm{d}\boldsymbol{k} \boldsymbol{U}_k(\boldsymbol{r}) a_k(t) + \mathrm{adj} \qquad (3)$$

where the electric field $\boldsymbol{E}$ has been decomposed into its normal modes:

$$\boldsymbol{E}(\boldsymbol{r}, t) = \mathrm{i} \int (\hbar\omega_k/16\pi^3)^{1/2} \hat{\boldsymbol{\epsilon}}_k a_k \exp[\mathrm{i}(\boldsymbol{k} \cdot \boldsymbol{r} - \omega_k t)] \, \mathrm{d}\boldsymbol{k} + \mathrm{adj} = -\int \mathrm{d}\boldsymbol{k} \, \boldsymbol{U}_k(\boldsymbol{r}) a_k(t) + \mathrm{adj}.$$

Here $\boldsymbol{\mu}$ is the atomic dipole operator, $\omega_k = c|\boldsymbol{k}|$, $\hat{\boldsymbol{\epsilon}}_k$ is the polarisation vector and a_k is the annihilation operator for the mode $\boldsymbol{k}$. The Schrödinger equation may now be written as a system of coupled equations for the amplitudes $b^{\pm}_{1\ldots n}(t)$, for the initial condition $b^{\pm}_{1\ldots n}(t=0) = 0$ and $b^{\pm}(t=0) = b^{\pm}_0$,

$$\frac{\mathrm{i}\partial}{\partial t}\langle \pm|t\rangle = \mathrm{i}\dot{b}^{\pm}$$
$$= \int \mathrm{d}\boldsymbol{k}_1 \, \mathrm{e}^{-\mathrm{i}\omega_1 t} \boldsymbol{U}_1 \cdot (b^+_1\langle \pm|\boldsymbol{\mu}|+\rangle + b^-_1\langle \pm|\boldsymbol{\mu}|-\rangle) + \mathrm{i}\delta(t) b^{\pm}_0 \qquad (4a)$$

$$\frac{\mathrm{i}\partial}{\partial t}\langle \boldsymbol{k}_1; \pm|t\rangle = \mathrm{i}\dot{b}^{\pm}_1$$
$$= \boldsymbol{U}^*_1 \cdot (b^+\langle \pm|\boldsymbol{\mu}^+|+\rangle + b^-\langle \pm|\boldsymbol{\mu}^+|-\rangle) \, \mathrm{e}^{\mathrm{i}\omega_1 t}$$
$$-2^{1/2} \int \mathrm{d}\boldsymbol{k}_2 \boldsymbol{U}_2 \cdot (b^+_{12}\langle \pm|\boldsymbol{\mu}|+\rangle + b^-_{12}\langle \pm|\boldsymbol{\mu}|-\rangle) \, \mathrm{e}^{-\mathrm{i}\omega_2 t} \qquad (4b)$$

$$\frac{\mathrm{i}\partial}{\partial t}\langle \boldsymbol{k}_1, \boldsymbol{k}_2; \pm|t\rangle = \mathrm{i}\dot{b}^{\pm}_{12}$$

$$= 2^{-1/2}\boldsymbol{U}_2^* \cdot (b_1^+\langle\pm|\boldsymbol{\mu}^+|+\rangle + b_1^-\langle\pm|\boldsymbol{\mu}^+|-\rangle)\,\mathrm{e}^{\mathrm{i}\omega_2 t} + (1\leftrightarrow 2)$$

$$+ 3^{1/2}\int \mathrm{d}\boldsymbol{k}_3\, \boldsymbol{U}_3 \cdot (b_{123}^+\langle\pm|\boldsymbol{\mu}|+\rangle + b_{123}^-\langle\pm|\boldsymbol{\mu}|-\rangle)\,\mathrm{e}^{-\mathrm{i}\omega_3 t} \qquad (4c)$$

$$\vdots$$

$$\frac{\mathrm{i}\partial}{\partial t}\langle \boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm|t\rangle = \mathrm{i}\dot{b}^{\pm}_{12\ldots n}$$

$$= n^{-1/2}\sum_{j=1}^{n} \boldsymbol{U}_j^* \cdot (b_{1\ldots\not{j}\ldots n}^+\langle\pm|\boldsymbol{\mu}^+|+\rangle + b_{1\ldots\not{j}\ldots n}^-\langle\pm|\boldsymbol{\mu}^+|-\rangle)\,\mathrm{e}^{\mathrm{i}\omega_j t}$$

$$+ (n+1)^{1/2}\int \mathrm{d}\boldsymbol{k}_{n+1}\boldsymbol{U}_{n+1} \cdot (b_{1\ldots n+1}^+\langle\pm|\boldsymbol{\mu}|+\rangle + b_{1\ldots n+1}^-\langle\pm|\boldsymbol{\mu}|-\rangle)\,\mathrm{e}^{-\mathrm{i}\omega_{n+1} t}.$$

In the above equations $\boldsymbol{U}_j \equiv \boldsymbol{U}_{\boldsymbol{k}_j}(\boldsymbol{r}=0)$, and the matrix elements of the dipole operator, in the rotating-wave approximation, may be obtained from equation (1) as

$$\langle\pm|\boldsymbol{\mu}(t)|+\rangle = \pm\binom{S}{C} C\boldsymbol{\mu}_{10}\exp[\mathrm{i}(\lambda_\pm - \lambda_+)t]\,\mathrm{e}^{\mathrm{i}\omega_0 t}$$

$$\langle\pm|\boldsymbol{\mu}^+(t)|+\rangle = \binom{C}{S} S\boldsymbol{\mu}_{01}\exp[\mathrm{i}(\lambda_\pm - \lambda_+)t]\,\mathrm{e}^{-\mathrm{i}\omega_0 t}$$

$$\langle\pm|\boldsymbol{\mu}(t)|-\rangle = \pm\binom{S}{C} S\boldsymbol{\mu}_{10}\exp[\mathrm{i}(\lambda_\pm - \lambda_-)t]\,\mathrm{e}^{\mathrm{i}\omega_0 t}$$

$$\langle\pm|\boldsymbol{\mu}^+(t)|-\rangle = -\binom{C}{S} C\boldsymbol{\mu}_{01}\exp[\mathrm{i}(\lambda_\pm - \lambda_-)t]\,\mathrm{e}^{-\mathrm{i}\omega_0 t}.$$

In obtaining equations (4) we have used the results

$$\langle \boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm|a_{\boldsymbol{k}}|t\rangle = (n+1)^{1/2}\langle \boldsymbol{k}, \boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm|t\rangle$$

and

$$\langle \boldsymbol{k}_1 \ldots \boldsymbol{k}_n \pm|a_{\boldsymbol{k}}^\dagger|t\rangle = n^{-1/2}\sum_{i=1}^{n}\delta(\boldsymbol{k}-\boldsymbol{k}_i)\langle \boldsymbol{k}_1, \ldots, \not{\boldsymbol{k}}_i, \ldots, \boldsymbol{k}_n; \pm|t\rangle.$$

The notation $(1, \ldots, \not{j}, \ldots, n)$ stands for the set $(1, \ldots, n)$ from which j is excluded. With the transformation

$$b^{\pm}_{1\ldots n}(t) = \int -\frac{\mathrm{d}E}{2\pi\mathrm{i}}\exp[-\mathrm{i}(E-\lambda^{\pm}_{1\ldots n})t]\,b^{\pm}_{1\ldots n}(t),$$

equations (4) may be written as a set of coupled algebraic equations for $b^{\pm}_{1\ldots n}(E)$ as follows:

$$(E-\lambda^{\pm})b^{\pm} = b_0^{\pm} \pm \binom{S}{C}\int \mathrm{d}\boldsymbol{k}_1\, \boldsymbol{U}_1 \cdot \boldsymbol{\mu}(Cb^+ + Sb^-) \qquad (5a)$$

$$(E-\lambda_1^{\pm})b_1^{\pm}=\binom{C}{S}\,\boldsymbol{U}_1^*\cdot\boldsymbol{\mu}^*(Sb^+-Cb^-)\pm 2^{1/2}\binom{S}{C}\,\mathrm{d}\boldsymbol{k}_2\,\boldsymbol{U}_2\cdot\boldsymbol{\mu}(Cb_{12}^++Sb_{12}^-) \tag{5b}$$

$$(E-\lambda_{12}^{\pm})b_{12}^{\pm}=2^{-1/2}\binom{C}{S}[\boldsymbol{U}_1^*\cdot\boldsymbol{\mu}^*(Sb_2^+-Cb_2^-)+\boldsymbol{U}_2^*\cdot\boldsymbol{\mu}^*(Sb_1^+-Cb_1^-)]$$

$$\pm 3^{1/2}\binom{S}{C}\int \mathrm{d}\boldsymbol{k}_3\,\boldsymbol{U}_3\cdot\boldsymbol{\mu}(Cb_{123}^++Sb_{123}^-) \tag{5c}$$

$$\vdots$$

$$(E-\lambda_{1\ldots n}^{\pm})b_{1\ldots n}^{\pm}=n^{-1/2}\binom{C}{S}\sum_{j=1}^{n}\boldsymbol{U}_j^*\cdot\boldsymbol{\mu}^*(Sb_{1\ldots\not{j}\ldots n}^+-Cb_{1\ldots\not{j}\ldots n}^-)$$

$$\pm(n+1)^{1/2}\binom{S}{C}\int \mathrm{d}\boldsymbol{k}_{n+1}\,\boldsymbol{U}_{n+1}\cdot\boldsymbol{\mu}(Cb_{1\ldots n+1}^++Sb_{1\ldots n+1}^-).$$

This coupled hierarchy of equations is solved as follows. Ignoring, for the time being, the decay of the $|\boldsymbol{k}_1;\pm\rangle$ state (5*b*) may be solved approximately as

$$b_1^{\pm}(E)\approx\binom{C}{S}\boldsymbol{U}_1^*\cdot\boldsymbol{\mu}^*(Sb^+-Cb^-)/(E-\lambda_1^{\pm})$$

which on substitution in (5*a*) yields

$$(E-\lambda^{\pm})b^{\pm}=b_0^{\pm}\pm 2f\binom{S}{C}(Sb^+-Cb^-) \tag{6}$$

where we have defined

$$f=\tfrac{1}{2}\int \mathrm{d}\boldsymbol{k}_1|\boldsymbol{U}_1\cdot\boldsymbol{\mu}|^2/(E-\lambda_1^{\pm})=\tfrac{1}{2}(\sigma-\tfrac{1}{2}\mathrm{i}\gamma). \tag{7}$$

σ and γ are the radiative level shift and decay constant of the excited state. Specifically assuming that the atom is initially in its ground state, i.e. $b_0^{\pm}=\binom{C}{S}$, equation (6) yields the solution

$$b^+(E)=\frac{(E-\lambda^{\mp}-2f)\binom{C}{S}}{(E-R^+)(E-R^-)} \tag{8}$$

where $R^{\pm}$ are roots of the quadratic

$$(E-\lambda^+)(E-\lambda^-)-2fE+2f(\lambda^+C^2+\lambda^-S^2). \tag{9}$$

The neglect of the decay of the $|\boldsymbol{k}_1;\pm\rangle$ state in obtaining the solution for $b^{\pm}$ clearly amounts to neglect of corrections of the order α, the fine structure constant, to the energy of the $|\boldsymbol{k}_1;\pm\rangle$ state, and thus amounts only to the assumption of monotonic dependence on energy of σ and γ.

To solve for $b_1^{\pm}(E)$, similarly we start with the approximate solution

$$b_{12}^{\pm}(E)\simeq 2^{-1/2}\binom{C}{S}[\boldsymbol{U}_1^*\cdot\boldsymbol{\mu}^*(Sb_2^+-Cb_2^-)+\boldsymbol{U}_2^*\cdot\boldsymbol{\mu}^*(Sb_1^+-Cb_1^-)]/(E-\lambda_{12}^{\pm}) \tag{10}$$

from which we obtain

$$\int \mathrm{d}\boldsymbol{k}_2\, \boldsymbol{U}_2 \cdot \boldsymbol{\mu}(Cb_{12}^{+}+Sb_{12}^{-})$$

$$=(Sb_1^{+}-Cb_1^{-})\int \mathrm{d}\boldsymbol{k}_2 |\boldsymbol{U}_2\cdot\boldsymbol{\mu}|^2\Big(\frac{C^2}{E-\lambda_{12}^{+}}+\frac{S^2}{E-\lambda_{12}^{-}}\Big)$$

$$+\boldsymbol{U}_1^{*}\cdot\boldsymbol{\mu}\int \mathrm{d}\boldsymbol{k}_2\,\boldsymbol{U}_2\cdot\boldsymbol{\mu}(Sb_2^{+}-Cb_2^{-})\Big(\frac{C}{E-\lambda_{12}^{+}}+\frac{S}{E-\lambda_{12}^{-}}\Big). \tag{11}$$

The second integral on the right-hand side of (11), as will be clear below, leads to integrals of the type $\int \mathrm{d}\boldsymbol{k}_2|\boldsymbol{U}_2\cdot\boldsymbol{\mu}|^2/(E-\lambda_{12}^{\pm})(E-R_2^{\pm})$ which may be shown to vanish. Thus using equations (10), (11) and (7) we solve equation (5b) to obtain

$$b_1^{\pm}(E)=\frac{(E-\lambda_1^{\mp}-2f)\binom{C}{S}(\boldsymbol{U}_1^{*}\cdot\boldsymbol{\mu}^{*})\beta}{(E-R_1^{+})(E-R_1^{-})} \tag{12}$$

where $\beta=SC\Omega/(E-R^{+})(E-R^{-})$ and $R_1^{\pm}$ are roots of the quadratic of the form (9) with $\lambda^{\pm}$ replaced by $\lambda_1^{\pm}$. From an examination of the contribution of the two terms in equation (5c) involving $\boldsymbol{U}_1^{*}\cdot\boldsymbol{\mu}^{*}$ and $\boldsymbol{U}_2^{*}\cdot\boldsymbol{\mu}^{*}$ to $b_{12}^{\pm}$ in equation (5b), it is clear that the latter corresponds to corrections to the energy of the state $|\boldsymbol{k}_1;\pm\rangle$ due to virtual emission and re-absorption of quanta of momentum $\boldsymbol{k}_2$, while the former leads to diagrams corresponding to the sequence $|\boldsymbol{k}_1;\pm\rangle\to|\boldsymbol{k}_1,\boldsymbol{k}_2;\pm\rangle\to|\boldsymbol{k}_2;\pm\rangle\to|\boldsymbol{k}_1,\boldsymbol{k}_2;\pm\rangle\to|\boldsymbol{k}_1;\pm\rangle$ and thus their contribution vanishes in the limit of infinite normalisation volume.

Proceeding similarly we arrive at the general solution

$$b_{12\ldots n}^{\pm}(E)=\frac{(E-\lambda_{1\ldots n}^{\mp}-2f)\binom{C}{S}\beta_{1\ldots n}(E)(\boldsymbol{U}_1^{*}\cdot\boldsymbol{\mu}^{*}\boldsymbol{U}_2^{*}\cdot\boldsymbol{\mu}^{*}\ldots\boldsymbol{U}_n^{*}\cdot\boldsymbol{\mu}^{*})}{(E-R_{1\ldots n}^{+})(E-R_{1\ldots n}^{-})} \tag{13}$$

where

$$\beta_{1\ldots n}(E)=\frac{SC\Omega}{n^{1/2}}\sum_{j=1}^{n}\frac{\beta_{1\ldots\not{j}\ldots n}(E)}{(E-R_{1\ldots\not{j}\ldots n}^{+})(E-R_{1\ldots\not{j}\ldots n}^{-})}\quad\text{and}\quad \beta_1\equiv\beta. \tag{14}$$

$R_{1\ldots n}^{\pm}$ are roots of equation (9) with $\lambda^{\pm}$ replaced by $\lambda_{1\ldots n}^{\pm}$.

The solution in equation (13) may be verified to be exact in the limit of infinite normalisation volume, by direct substitution in equations (5).

3. Spectrum of the scattered field

The steady-state spectrum of the scattered field may be obtained from the amplitudes $b_{1\ldots n}^{\pm}(t\to\infty)$ for $n\gg 1$ as

$$b_{1\ldots n}^{\pm}(t\to\infty)=\mathrm{i}(2\pi)^{-1}\int_{-\infty}^{\infty}\mathrm{d}E\, b_{1\ldots n}^{\pm}(E)\exp[-\mathrm{i}(E-\lambda_{1\ldots n}^{\pm})t]$$

$$=\lim_{t\to\infty}\mathrm{i}(2\pi)^{-1}\binom{C}{S}v_1^{*}v_2^{*}\ldots v_n^{*}\int_{-\infty}^{\infty}\mathrm{d}E\frac{\beta_{1\ldots n}(E)\exp[-\mathrm{i}(E-\lambda_{1\ldots n}^{\pm})t]}{E-\lambda_{1\ldots n}^{\pm}+\mathrm{i}\alpha}$$

$$=\binom{C}{S}v_1^{*}\ldots v_n^{*}\beta_{1\ldots n}(E=\lambda_{1\ldots n}^{\pm}) \tag{15}$$

where we have defined $v_1^* = \boldsymbol{U}_1^* \cdot \boldsymbol{\mu}^*$ etc. In equation (15) we have replaced the solution in equation (13) by that obtained by turning off the decay of the $|\boldsymbol{k}_1 \ldots \boldsymbol{k}_n; \pm\rangle$ state. This is valid for $n \gg 1$ corresponding to the experimental situation of transit of the atom through the exciting field and detection of scattered quanta over intervals much larger than the lifetime for spontaneous decay of the excited state. In the appendix we show that the spectral densities obtained from the amplitudes in equation (15) are identical with those obtained by direct calculation of the photon emission rate spectrum. The probability that a photon of wavevector $\boldsymbol{k}_1$ is emitted is obtained from the probability of emission of n photons, $\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n$, by integrating the latter over the unobserved $n-1$ quanta as follows:

$$W_n(\boldsymbol{k}_1) = \int \mathrm{d}\boldsymbol{k}_2 \ldots \int \mathrm{d}\boldsymbol{k}_n (|b^+_{1\ldots n}(\infty)|^2 + |b^-_{1\ldots n}(\infty)|^2). \tag{16}$$

From equation (14) we can write

$$\beta_{1\ldots n}(E = \lambda^\pm_{1\ldots n}) = \frac{1}{(n!)^{1/2}}\left(\frac{SC\Omega}{2z}\right)^n \sum_{j=1}^{n} G_j^\pm \sum_{k\neq j} G_{jk}^\pm \sum_{l\neq j,k} G^\pm_{jkl} \ldots G^\pm_{1\ldots n} \tag{17}$$

where

$$G_j^\pm (E = \lambda^\pm_{1\ldots n}) = 1/(\lambda^\pm_{1\ldots n} - R^-_{1\ldots \not{j}\ldots n}) - 1/(\lambda^\pm_{1\ldots n} - R^+_{1\ldots \not{j}\ldots n})$$

and so on; z is complex for $\Delta \neq 0$ and is defined from equation (9) as follows:

$$R^\pm_{1\ldots n} = \delta_{1\ldots n} + \tfrac{1}{2}\Delta + f \pm z, \qquad z = \tfrac{1}{2}[(\Delta + 2f)^2 + 4\kappa^2]^{1/2} = R + I.$$

After some lengthy algebra, the sequence $W_2, \ldots, W_n$ is obtained in terms of integrals involving $G_j^\pm$ as follows:

$$W_2(\boldsymbol{k}_1) = \tfrac{1}{2}|v_1|^2(\gamma/2\pi)(\kappa^2/|z|^2)^2[A(|G_1^+|^2 + |G_1^-|^2) + 2F + 2\,\mathrm{Re}\,\tilde{C}(G_1^+ + G_1^-)]$$

$$W_3(\boldsymbol{k}_1) = \tfrac{1}{3}|v_1|^2(\gamma/2\pi)^2(\kappa^2/|z|^2)^4[A^2(|G_1^+|^2 + |G_1^-|^2) + 4AF + 4\,\mathrm{Re}\,\tilde{C}C + 2\,\mathrm{Re}(\tilde{C}H + \tilde{C}A)(G_1^+ - G_1^-)]$$

$$\vdots$$

$$\begin{aligned} W_n(\boldsymbol{k}_1) = \frac{1}{n}|v_1|^2(\gamma/2\pi)^{n-1}(\kappa^2/|z|^2)^{2n-2}[&A^{n-1}(|G_1^+|^2 + |G_1^-|^2) \\ &+ 2nA^{n-2}F + 4\,\mathrm{Re}\,C\tilde{C}(A^{n-3} + A^{n-4}H + \ldots + H^{n-3}) \\ &+ 4\,\mathrm{Re}\,C\tilde{C}A(H^{n-4} + 2AH^{n-5} + \ldots + (n-3)A^{n-4}) \\ &+ 2\,\mathrm{Re}\,\tilde{C}H(G_1^+ - G_1^-) + 2\,\mathrm{Re}\,A\tilde{C}(H^{n-3} + AH^{n-4} + \ldots + A^{n-3})]. \end{aligned} \tag{18}$$

The quantities A, C, $\tilde{C}$, F, H are defined as the integrals

$$A = \int \mathrm{d}\omega_2 |G_2^\pm|^2 = 2\pi\mathrm{i}(1/2\zeta - 1/2\zeta^* - 1/2f_+ - 1/2f_-)$$

$$\begin{aligned} F = \int \mathrm{d}\omega_2 |G_2^\pm|^2 |G_{12}^\pm|^2 &= 2\pi\mathrm{i}[|\xi_-|^2/(\delta - 2f_-) + |\xi_+|^2/(\delta - 2f_+) - \xi_+\xi_-/(\delta - 2\zeta) - \xi_+^*\xi_-^*/(\delta + 2\zeta^*) \\ &\quad + (\delta \to -\delta)] \end{aligned}$$

$$C = \int d\omega_2 G_{12}^{\pm} |G_2^{\pm}|^2$$

$$= 2\pi i[\xi_+/(\delta+2\zeta) - \xi_+/(\delta+2f_+) + \xi_-^*/(\delta-2\zeta^*) - \xi_-^*/(\delta+2f_-)]$$

$$\tilde{C} = C(\delta \to -\delta)$$

$$H = \int d\omega_2 (G_2^{\pm})^* G_{12}^{\pm}$$

$$= 2\pi i[1/(\delta+2\zeta) + 1/(\delta-2\zeta^*) - 1/(\delta+2f_-) - 1/(\delta+2f_+)]. \tag{19}$$

In equations (19) (and hereafter) we have dropped the subscript 1 on δ etc. We have also defined

$$\xi = R + f, \qquad f_\pm = f \pm I$$

$$\xi_+ = \tfrac{1}{2}(1/\zeta - 1/f_+), \qquad \xi_- = \tfrac{1}{2}(1/\zeta - 1/f_-).$$

Using the identities

$$H^{n-1} + H^{n-2}A + \ldots + A^{n-1} = (H^n - A^n)/(H-A)$$

and

$$H^{n-2} + 2AH^{n-3} + \ldots + (n-1)A^{n-2} = [H^n - nHA^{n-1} + (n-1)A^n]/(H-A)^2$$

we finally obtain from equation (18) the result, for the limit $n \to \infty$,

$$\lim_{n\to\infty} W_n(\mathbf{k}) = |v(\mathbf{k})|^2 \left(\frac{\gamma}{2\pi}\right)^n \left(\frac{\kappa^2}{|z|^2}\right)^{n+1} A^{n-1} \left[F + 2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right)\right]$$

$$= |v(\mathbf{k})|^2 \left(\frac{\gamma}{2\pi}\right) \left(\frac{\kappa^2}{|z|^2}\right)^2 \left[F + 2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right)\right]. \tag{20}$$

4. Analysis of the decay spectrum

For the resonant intense field limit equation (20) reduces to a simple form. For $\kappa/\gamma \gg 1$, $\kappa/\Delta_0 \gg 1$ we have:

$$z \simeq \kappa \qquad f_\pm \simeq f$$

$$A \simeq -2\pi i[f/(\kappa^2 - f^2) + 1/f]; \qquad H \simeq 2\pi i[1/(\delta+2\kappa+2f) + 1/(\delta-2\kappa+2f) - 2/(\delta+2f)]$$

$$C \simeq \tilde{C}^* \simeq -(2\pi i/2f)[1/(\delta+2\kappa+2f) - 1/(\delta-2\kappa+2f)]$$

$$C\tilde{C}^*/(A-H) \simeq -(2\pi i/4f^2)(\delta+2f/\delta)[1/(\delta-2\kappa+2f) - 1/(\delta-2\kappa+3f) + (\kappa \to -\kappa)].$$

The real part of the latter expression is regular at $\delta = 0$. For $|\delta| \ll \kappa$, the contribution from this term is $O(\gamma^2/\kappa^2)$ that arises from F. Thus we may set $(\delta + 2f/\delta) = 1$ and finally obtain

$$F + 2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right) = \frac{4\pi}{\gamma}\left(\frac{4}{\delta^2 - 4f^2} + \frac{3}{(\delta-2\kappa)^2 - 9f^2} + \frac{3}{(\delta+2\kappa)^2 - 9f^2}\right). \tag{21}$$

This spectral profile has been obtained by Mollow (1969), Smithers and Freedhoff (1975), Carmichael and Walls (1976), and Kimble and Mandel (1976). The ratios of the heights of the central and sideband maxima (3 : 1) and their width, at half maximum (3 : 2), differ from the prediction resulting from one-photon approximation of Stroud (1971). In the latter approximation we have

$$
\begin{aligned}
|b_1^+(\infty)|^2+|b_1^-(\infty)|^2 &= |v(\boldsymbol{k})|^2(|\beta(\lambda_1^+)|^2+|\beta(\lambda_1^-)|^2) \\
&= \frac{|v(\boldsymbol{k})|^2}{2\kappa}\left[\frac{2}{\delta^2-f^2}+\frac{1}{(\delta+2\kappa)^2-f^2}+\frac{1}{(\delta-2\kappa)^2-f^2}+\mathrm{O}\left(\frac{f^2}{\kappa^2}\right)\right].
\end{aligned}
\tag{22}
$$

In the limit of strong fields and large detuning, i.e. $\Delta_0 \gg \kappa \gg \gamma$, we can write

$$
\Delta \simeq \Delta_0, \qquad z \simeq \tfrac{1}{2}\Delta + f\Delta_0/\Delta, \qquad \xi_+ \simeq -\tfrac{1}{4}f, \qquad \xi_- \simeq -\Delta^2/4\kappa^2 f
$$

$$
f_+ \simeq 2f, \qquad f_- \simeq 2\kappa^2 f/\Delta^2
$$

$$
A \simeq -2\pi \mathrm{i}\Delta^2/4f\kappa^2
$$

$$
A-H \simeq -\frac{2\pi \mathrm{i}\Delta^2\delta}{4\kappa^2 f(\delta+2f_-)}
$$

$$
C \simeq \frac{2\pi \mathrm{i}}{4f[1/(\delta+2f_-)-1/(\delta+\Delta+2f)]}
$$

$$
\frac{C\tilde{C}^*}{A-H} \simeq -\frac{2\pi \mathrm{i}}{f}\left(\frac{\Delta^2}{2\kappa^2}\right)^2\frac{1}{(\delta+2f_-)^2[(\delta+2f)^2-\Delta^2]}
$$

$$
\begin{aligned}
2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right) &\simeq \frac{4\pi}{\gamma}\left(\frac{\Delta^2}{\kappa}\right)^2\frac{1}{(\delta^2-4f_-^2)[(\delta-\Delta)^2-4f^2][(\delta+\Delta)^2-4f^2]} \\
&\simeq \frac{4\pi^2}{\gamma^2}\left(\frac{\Delta}{\kappa}\right)^4\delta(\omega-\omega')
\end{aligned}
$$

$$
F \simeq -\frac{2\pi \mathrm{i}}{16f^2}\left[\frac{(\Delta/\kappa)^4}{\delta-2f_-}+\frac{1}{\delta-2f_+}+\left(\frac{\Delta}{\kappa}\right)^2\left(\frac{1}{\delta-\Delta-2f}+\frac{1}{\delta+\Delta-2f}\right)+(\delta\to-\delta)\right]
$$

$$
F+\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right) \simeq \frac{2\pi}{\gamma}\left[\frac{4\pi}{\gamma}\left(\frac{\Delta}{\kappa}\right)^4\delta(\omega-\omega')+\frac{2}{\delta^2-16f^2}+\frac{(\Delta/\kappa)^2}{(\delta-\Delta)^2-4f^2}+\frac{(\Delta/\kappa)^2}{(\delta+\Delta)^2-4f^2}\right].
\tag{23}
$$

The ratio of the heights of the peaks at $\delta=0$ and $\delta=\pm\Delta$ of the inelastically scattered part in equation (23) of $1:2(\Delta/\kappa)^2$ does not agree with the result $2:(\Delta/\kappa)^2$ of Kimble and Mandel (1976); though their relative widths are in agreement. The one-photon approximation cannot predict two outer peaks of equal intensity in this limit for the following reason. For large detuning the $|\pm\rangle$ and $|\boldsymbol{k};\pm\rangle$ states are approximately $|N-1,1\rangle$, $|N,0\rangle$ and $|N-2,\boldsymbol{k},1\rangle$, $|N-1,\boldsymbol{k},0\rangle$ states respectively. Thus only $|+\rangle$ and $|\boldsymbol{k};-\rangle$ states couple with a sizeable matrix element under V.

Another important limiting case where the spectrum in equation (20) reduces to a simple form is for fields of moderate strength, and large detuning, i.e. $|\Delta_0| \gg |\kappa|$, γ. Results for the cases where $\kappa^2+f^2>0$ and $\kappa^2+f^2<0$ differ.

In both cases we have the approximation

$$
I \simeq f\Delta_0/(\Delta_0^2+\kappa^2+f^2)^{1/2}, \qquad \xi_+ \simeq -\tfrac{1}{4}f, \qquad \xi_- \simeq -\tfrac{1}{2}f_-.
$$

Case (*i*) $\kappa^2+f^2>0$. Here f_- lies on the negative imaginary axis.

$$A\simeq -2\pi i/2f_-, \qquad H\simeq -2\pi i/(\delta+2f_-)$$

$$F\simeq -\frac{2\pi i}{f_-}\left(\frac{1}{\delta^2-4f_-^2}+\frac{f_-}{2f}\frac{1}{\delta^2-16f^2}+\frac{1}{2[(\delta-\Delta)^2-4f^2]}+\frac{1}{2[(\delta+\Delta)^2-4f^2]}\right)$$

$$\frac{C\tilde{C}^*}{A-H}\simeq -\frac{2\pi i}{2\delta f_-}\left(-\frac{1}{\delta+2f_-}+\frac{1}{\delta-\Delta+2f}+\frac{1}{\delta+\Delta+2f}+\frac{(f_-/f)}{\delta+4f}\right) \tag{24}$$

and

$$F+2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right)$$

$$\simeq -\frac{i\pi}{f_-}\left(\frac{5(f_-/f)}{\delta^2-16f^2}+\frac{(3\delta-2\Delta)/\delta}{(\delta-\Delta)^2-4f^2}+\frac{(3\delta+2\Delta)/\delta}{(\delta+\Delta)^2-4f^2}\right).$$

Case (*ii*) $\kappa^2+f^2<0$. Here f_- lies on the positive imaginary axis. Now

$$A\simeq 2\pi i/2f_-, \qquad H\simeq 2\pi i/(\delta+2f_-)$$

$$F\simeq \frac{2\pi i}{f_-}\left(\frac{1}{\delta^2-4f_-^2}-\frac{f_-}{2f}\frac{1}{\delta^2-16f^2}+\frac{1}{2[(\delta-\Delta)^2-4f^2]}+\frac{1}{2[(\delta+\Delta)^2-4f^2]}\right)$$

$$\frac{C\tilde{C}^*}{A-H}\simeq \frac{2\pi i}{2\delta f_-}\left(-\frac{1}{\delta+2f_-}+\frac{1}{\delta+\Delta+2f}-\frac{(f_-/f)}{\delta+4f}+\frac{1}{\delta-\Delta+2f}\right) \tag{25}$$

$$F+2\,\mathrm{Re}\left(\frac{C\tilde{C}^*}{A-H}\right)\simeq \frac{i\pi}{f_-}\left(-\frac{5(f_-/f)}{\delta^2-16f^2}+\frac{(3\delta-2\Delta)/\delta}{(\delta-\Delta)^2-4f^2}+\frac{(3\delta+2\Delta)/\delta}{(\delta+\Delta)^2+4f^2}\right).$$

The results in equations (24) and (25) lead to the same limit as $\kappa^2+f^2\to 0$. In the weak-field limit, i.e. κ/γ, $\Delta_0/\gamma\ll 1$, we have

$$z\simeq \tfrac{1}{2}\Delta_0+f\simeq\zeta, \qquad \xi_+\simeq\tfrac{1}{2}f, \qquad \xi_-\simeq 8f/\Delta^2\simeq -\tfrac{1}{2}f_-$$

$$A\simeq 8\pi i f/\Delta^2, \qquad H\simeq -2\pi i/(\delta+2f_-)$$

$$C\simeq -(\pi i/f_-)/(\delta+2f_-)\simeq -\tilde{C}^*$$

$$F\simeq 2\pi i|\xi_-|^2[1/(\delta-2f_-)-1/(\delta+2f_-)]\simeq 16\pi^2\gamma^2\delta(\omega-\omega')/\Delta^2$$

$$F+2\,\mathrm{Re}[C\tilde{C}^*/(E-H)]\simeq 14\pi^2\gamma^2\delta(\omega-\omega')/\Delta^2.$$

Thus in the weak-field limit, excitation by a monochromatic field leads to an elastic contribution only. This result naturally agrees with that obtained by the one-photon approximation (Heitler 1954).

In figures 1–3 we have computed the expression in equation (20) for weak ($\kappa=0{\cdot}025$), medium ($\kappa=1{\cdot}0$) and strong ($\kappa=2{\cdot}5$) incident fields, for different values of detuning frequency $\Delta=\omega-\omega_0$. All frequencies are in units of γ. Choices of parameters for these figures correspond to those of Kimble and Mandel (1976) in their figures 5, 6 and 7. The two sets of results agree closely with each other. Elastic contributions are not shown in either set of results. Their comparison of the theoretical results with the experiments of Wu *et al* (1975) and related comments need not be repeated here.

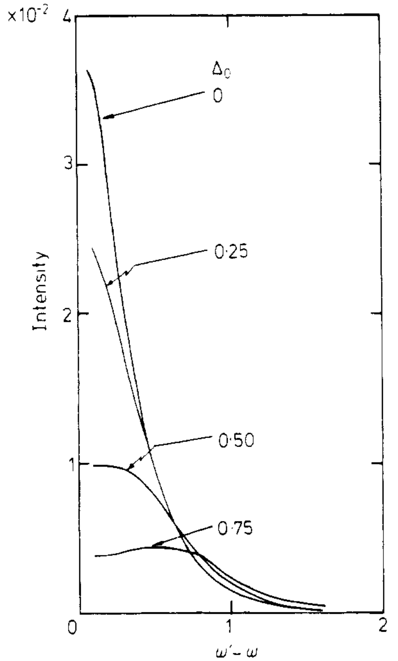

Figure 1. Frequency distribution of spectral density of inelastically scattered light for weak fields ($\kappa = 0{\cdot}025\gamma$). Intensity scales of figures 1, 2 and 3 are relative. Frequency scales are in units of γ. ω', ω are the frequencies of scattered and incident light. Detuning frequency $\Delta = \omega - \omega_0$.

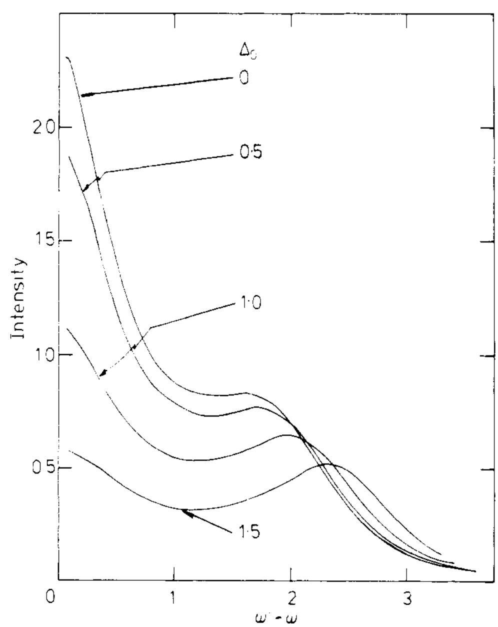

Figure 2. Frequency distribution of spectral density of inelastically scattered light for intermediate fields ($\kappa = \gamma$) for detuning frequencies $\Delta = 0$, $0{\cdot}5\gamma$, γ and $1{\cdot}5\gamma$.

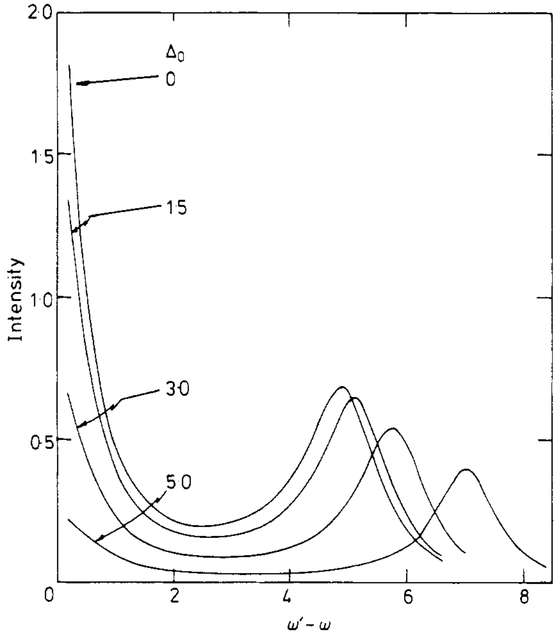

Figure 3. Frequency distribution of spectral density of inelastically scattered light for strong fields ($\kappa = 2{\cdot}5\gamma$) for detuning frequencies $\Delta = 0$, $1{\cdot}5\gamma$, 3γ and 5γ.

5. Discussion

Since the first draft of this manuscript was submitted, Mollow (1975) has rederived his earlier (1969) result by a procedure that does not involve the assumption of factorisation of the density matrix into that of the atom and the field. In that paper he concludes that the solution for the amplitudes $b^{\pm}_{1\ldots n}(t)$ obtained by turning off the decay of a state with a large number of scattered quanta leads to scattered electric fields that are non-vanishing only over a spherical shell of approximate thickness c/γ, whereas the exact solution obtained from equation (13) or equation (A.7) leads to fields uniformly distributed between $r = 0$ and $r = ct$. We shall take up a detailed comparison of our solution with that of Mollow in a forthcoming publication. Here in the appendix we simply demonstrate that the solution in equation (13) or equation (A.7) leads to a photon emission rate spectrum identical with that obtained in equation (20). Thus the Heitler-type procedure of turning off the decay of the state with a large number of scattered quanta leads to correct power spectra.

It is interesting to compare the methods of Mollow (1969, 1975), Cohen-Tannoudji and Reynaud (1977) and Kimble and Mandel (1976) with the method of this paper.

For dipole coupling in a two-level system one can show from the Heisenberg equation of motion, that the far electric field operator is proportional to the sum of the atomic raising and lowering operators. Thus to determine the spectral density of the scattered field one should calculate the two-time correlation function of the atomic dipole operator. Though this is usually done with the aid of the quantum regression theorem (Mollow 1969, Cohen-Tannoudji and Reynaud 1977), Kimble and Mandel

(1976) have shown that they can be obtained by a simultaneous solution of a set of related correlation functions.

Though Mollow's original derivation (1969) involved the Markoff assumption of factorisation of the density matrix of the atom and the radiated field, his later (1975) derivation avoids this assumption by solving the coupled equations of motion of bilinear products of amplitudes of vectors in the four subspaces in which the atom is in one of its two states and the scattered field contains 0 or 1 quantum of the detected mode.

Cohen-Tannoudji and Reynaud (1977), using the strong-coupling basis similar to ours, start with the master equation for the density matrix σ of the atom (+ incident field) obtained by the factorisation assumption above. The equations for $\langle\alpha, N|\sigma|\beta, N\pm 1\rangle$ ($\alpha, \beta = \pm$ of our equation (1a)) are uncoupled under the 'secular approximation' ($|\gamma/\kappa| \ll 1$). The coupling of amplitudes of $b^{\mp}_{1\ldots n}(t)$ to those of $b^{\pm}_{1\ldots\not{r}\ldots n}(t)$ in equations (4) reveals itself as coupling of density matrix elements of adjacent N-values. This procedure yields the line position and their relative heights and widths only for strong resonant incident fields where the secular approximation is valid.

All these methods involve the assumption of atomic transit times in the incident field of duration being very much greater than γ^{-1}. For transit time of the order of γ^{-1} they all appear to involve comparable labour.

Acknowledgments

It is a pleasure to gratefully acknowledge the continued support and encouragement of Professor Gerald J Small of this laboratory. This work was supported by the Quantum Chemistry Division of the National Science Foundation (Grant No. MPS74-02787 A01). The author also wishes to acknowledge support of this work in its initial phase by Professor R Wallace, Chemistry Department, University of Manitoba, Canada.

Appendix

In this Appendix we demonstrate that the power spectrum obtained from the rate of emission of photons in the steady state is identical to that in equation (20) obtained by turning off the decay of a state with large enough number of scattered quanta and calculating the long-term distribution of conditional probabilities in the frequency of one of the scattered modes.

Though we can start from the results in equations (13) and (14), it is of interest to utilise the non-Hermitian Hamiltonian derived by Mollow (1975) valid for the case of a two-level atom in a coherent monochromatic field. For this case Mollow has shown that the incident field can be replaced by a c-number field and that the term that leads to absorption of quanta that had been previously emitted can be rewritten to yield a modified Hamiltonian, in the dipole approximation:

$$H(t) = H_0(t) + H_1(t) \tag{A.1}$$

where

$$H_0(t) = \hbar(\omega_0 - \tfrac{1}{2}\mathrm{i}\gamma)b^+b + H_{0\mathrm{F}} - (\boldsymbol{\mu}^* \cdot \boldsymbol{E}_c^*(0, t)b + \mathrm{adj}) \tag{A.2}$$

$$H_1(t) = -\boldsymbol{\mu}^* \cdot \boldsymbol{E}_R^+(0) b. \tag{A.3}$$

Here $b = |0\rangle\langle 1|$ is the atomic de-excitation operator, $\boldsymbol{E}_R^+(0)$ is the negative frequency part of the vacuum electric field operator at the atom, $\boldsymbol{E}_c$ and $\boldsymbol{E}_c^*$ are the positive and negative frequency parts of c-number field that accurately represents the incident single-mode coherent field, and $\boldsymbol{\mu} = \langle 1|\hat{\boldsymbol{\mu}}|0\rangle$. H_{0F} is the Hamiltonian for the free field and the decay constant γ has been defined in the text.

For simplicity we shall restrict our discussion to the case of intense resonant incident field. The eigenfunctions of $H_0(t)$ of eigenvalues $\lambda^{\mp}_{1\ldots n}$ may be written as

$$|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm\rangle = 2^{-1/2}|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n\rangle(|0\rangle + C_\pm|1\rangle)$$

$$C_\pm = (\mathrm{i}\gamma/4\kappa \pm z/2\kappa) \simeq \pm 1; \qquad \kappa = -\boldsymbol{\mu} \cdot \boldsymbol{E}_c(0) \tag{A.4}$$

$$\lambda^{\pm}_{1\ldots n} = \tfrac{1}{2}[\tilde{\Delta} \pm (\tilde{\Delta}^2 + 4\kappa^2)^{1/2}] + \omega_1 + \ldots + \omega_n - n\omega \xrightarrow{\Delta=0, |\gamma/\kappa| \ll 1} -\mathrm{i}\gamma/4 \pm \kappa + \delta_{12\ldots n}$$

$$\tilde{\Delta} = \Delta - \tfrac{1}{2}\mathrm{i}\gamma = \omega - \omega_0 - \tfrac{1}{2}\mathrm{i}\gamma \simeq -\tfrac{1}{2}\mathrm{i}\gamma$$

where the results have been adopted for a near-resonant intense incident field. The functions $|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm\rangle$ in (A.4) are not orthogonal to each other. Instead the biorthogonal set defined by

$$H_0^+ |\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm'\rangle = \lambda^{\pm *}_{1\ldots n}|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm'\rangle \tag{A.5}$$

where

$$|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm'\rangle = 2^{-1/2}|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n\rangle(|0\rangle \pm C^*_{\mp}|1\rangle)$$

are orthogonal to the kets $|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm\rangle$. Expanding the state vector in the kets in (A.4) in the Schrödinger picture as

$$|t\rangle = b^+ \mathrm{e}^{-\mathrm{i}\lambda^+ t}|+\rangle + b^- \mathrm{e}^{-\mathrm{i}\lambda^- t}|-\rangle$$
$$+ \sum_{n=1}^{\infty} \sum_{\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n} (b^+_{1\ldots n} \mathrm{e}^{-\mathrm{i}\lambda^+_{1\ldots n} t}|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; +\rangle + b^-_{1\ldots n} \mathrm{e}^{-\mathrm{i}\lambda^-_{1\ldots n} t}|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; -\rangle) \tag{A.6}$$

we can solve for the Fourier transform of the amplitudes $b^\pm_{12\ldots n}(t)$ as

$$b^\pm_{12\ldots n}(E) = \frac{C^{2n} v_1 v_2 \ldots v_n}{(n!)^{1/2}(E - \lambda^\pm_{12\ldots n})} \sum_{j=1,\ldots,n} \beta_{1\ldots \not{j} \ldots n} \times \left(\frac{C_+}{E - \lambda^+_{1\ldots \not{j} \ldots n}} + \frac{C_-}{E - \lambda^-_{1\ldots \not{j} \ldots n}}\right) \tag{A.7}$$

where

$$\beta_{1\ldots n}(E) = \sum_{j=1,\ldots,n} \beta_{1\ldots \not{j} \ldots n}(E)\left(\frac{C_+}{E - \lambda^+_{1\ldots \not{j} \ldots n}} + \frac{C_-}{E - \lambda^-_{1\ldots \not{j} \ldots n}}\right). \tag{A.8}$$

As pointed out before, in the intense-field limit, $C = 2^{-1/2}$, $C_\pm = \pm 1$:

$$b^\pm_{1\ldots n}(t) = -\frac{1}{2\pi \mathrm{i}} \int \mathrm{d}E \exp[-\mathrm{i}(E - \lambda^\pm_{1\ldots n})t] b^\pm_{1\ldots n}(E).$$

The probability of finding the system in the state $|\boldsymbol{k}_1, \ldots, \boldsymbol{k}_n\rangle$ is obtained from the scalar product of $|t\rangle$ with $\langle \boldsymbol{k}_1, \ldots, \boldsymbol{k}_n; \pm| \exp(\mathrm{i}\lambda^{\pm *}_{1\ldots n} t)$. The rate of emission of quanta in the mode $\boldsymbol{k}_1$ (frequency $= \omega_1$) is obtained from the integrals

$$\frac{1}{T}\int_0^T \mathrm{d}t\, \mathrm{e}^{-\gamma t/2}|b^{\pm}_{1\ldots n}(t)|^2$$
$$= \frac{1}{T}\int_0^T \mathrm{d}t \int \frac{\mathrm{d}E}{2\pi\mathrm{i}} \exp[\mathrm{i}(E-\lambda^{\pm *}_{1\ldots n})t]$$
$$\times \int -\frac{\mathrm{d}E'}{2\pi\mathrm{i}} \exp[-\mathrm{i}(E'-\lambda^{\pm}_{1\ldots n})t] b^{\pm *}_{1\ldots n}(E) b^{\pm}_{1\ldots n}(E')\, \mathrm{e}^{-\gamma t/2}$$
$$= \frac{1}{4\pi^2}\int_{-\infty}^{\infty} \mathrm{d}E |b^{\pm}_{1\ldots n}(E)|^2$$

as

$$W(\boldsymbol{k}_1) = \int_{-\infty}^{\infty} \mathrm{d}E \sum_{n=1}^{\infty} \sum_{\boldsymbol{k}_2,\ldots,\boldsymbol{k}_n} (|b^{+}_{1\ldots n}(E)|^2 + |b^{-}_{1\ldots n}(E)|^2). \tag{A.9}$$

From (A.7) and (A.8) it can be shown after some tedious calculations that

$$\int_{-\infty}^{\infty} \mathrm{d}E \sum_{2,\ldots,n} (|b^{+}_{1\ldots n}(E)|^2 + |b^{-}_{1\ldots n}(E)|^2)$$
$$= \frac{|v|^2}{8n}\frac{2\pi}{\gamma} [\![8n \operatorname{Re} g + 2 \operatorname{Re} l^2 + 2 \operatorname{Re} l^2(2-m) + \operatorname{Re} l^2[4-m(2-m)] + \ldots$$
$$+ 2^{3-n} \operatorname{Re} l^2(2^{n-3} - m\{2^{n-4} - m[\ldots - m(2-m)]\ldots\})$$
$$+ 2^{4-n} \operatorname{Re} l^2(2^{n-2} - m\{2^{n-3} - m[\ldots(2-m)]\ldots\})]\!] \tag{A.10}$$

where

$$g = [1/\delta - 1/(\delta+z)][1/(\delta+2f) - 1/(\delta+z+2f)] + (z \to -z)$$
$$= 1/(\delta+z+2f) - 1/(\delta-z+2f)$$
$$m = 2f[2/(\delta+2f) - 1/(\delta+z+2f) - 1/(\delta-z+2f)]$$
$$f = -\tfrac{1}{4}\mathrm{i}\gamma; \qquad v = \langle \boldsymbol{k}_1, 0|H_1|1\rangle; \qquad \delta = \omega_1 - \omega.$$

(A.10) may be used to yield the n-sum in equation (A.9) as

$$W(\boldsymbol{k}_1) = \int_{-\infty}^{\infty} \mathrm{d}E\Big(|b^{+}_{1}(E)|^2 + |b^{-}_{1}(E)|^2 + \sum_{n=2}^{\infty}\sum_{\boldsymbol{k}_2,\ldots,\boldsymbol{k}_n} (|b^{+}_{1\ldots n}(E)|^2 + |b^{-}_{1\ldots n}(E)|^2)\Big)$$
$$= \lim_{n\to\infty} \frac{\pi|v|^2}{2\gamma}\Big(4 \operatorname{Re} g + \frac{n-1}{n}\operatorname{Re} l^2 + \frac{n-2}{n}\operatorname{Re} l^2 m \ldots + \ldots + \frac{1}{n}\operatorname{Re} l^2 m^{n-2}\Big)$$
$$= \lim_{n\to\infty} \frac{\pi|v|^2}{2\gamma}\Big(4 \operatorname{Re} g + \operatorname{Re}\frac{l^2}{n}\{n(1+m+\ldots+m^{n-2})$$
$$-[1+2m+3m^2+\ldots+(n-1)m^{n-2}]\}\Big)$$
$$\simeq \frac{\pi|v|^2}{2\gamma}[4 \operatorname{Re} g + \operatorname{Re} l^2/(1-m)]. \tag{A.11}$$

This result is easily verified to be identical with the intense-field result in equation (21) in the text.

References

Agarwal G S 1974 *Springer Tracts in Modern Physics* vol. 70 (Berlin: Springer)
Carmichael H J and Walls D F 1976 *J. Phys. B: Atom. Molec. Phys.* **9** 1199–219
Cohen-Tannoudji C and Reynaud S 1977 *J. Phys. B: Atom. Molec. Phys.* **10** 345–63
Gibbs H M and Venkatesan T N C 1976 *Opt. Commun.* **17** 87–90
Hartig W and Walther H 1973 *Appl. Phys.* **1** 171–7
Hassan S S and Bullough R K 1975 *J. Phys. B: Atom. Molec. Phys.* **8** L77–81
Heitler W 1954 *The Quantum Theory of Radiation* (Oxford: Clarendon)
Kimble H J and Mandel L 1976 *Phys. Rev.* A **13** 2133–44
—— 1977 *Phys. Rev.* A **15** 689–99
Lax M 1967 *Phys. Rev.* **157** 213–31
Mollow B R 1969 *Phys. Rev.* **188** 1969–75
—— 1975 *Phys. Rev.* A **12** 1919–45
Schuda F, Stroud C R Jr and Hercher M 1974 *J. Phys. B: Atom. Molec. Phys.* **7** L198–202
Smithers M E and Freedhoff H S 1975 *J. Phys. B: Atom. Molec. Phys.* **8** 2911–21
Stroud C R Jr 1971 *Phys. Rev.* A **3** 1044–52
Swain S 1975 *J. Phys. B: Atom. Molec. Phys.* **8** L437–41
Wu F Y, Grove R E and Ezekiel S 1975 *Phys. Rev. Lett.* **35** 1426–9