Coherence and elastic scattering in resonance fluorescence

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Some coherence properties of the light emitted from a two-level atom which is excited by a monochromatic coherent field are investigated. It is shown that the elastically scattered light is coherent, and that this compoment of the emitted field can be separated from the total field by narrow-band filtering at the driving frequency. These results are then applied to the problem of interference between the fluorescent and the elastically scattered field. An interference experiment is analyzed in which the detection of one emitted photon signifies the beginning of a Rabi cycle and the detection of a subsequent photon fixes the instant in the Rabi cycle when the interference is detected. It is shown that the visibility of the interference is near zero at the time when the fluorescence is maximum.

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I. INTRODUCTION

The subject of resonance fluorescence from a single, coherently excited atom has received a great deal of attention, both theoretical and experimental, over many years. Resonance fluorescence provided the first experimental evidence for nonclassical states of light [1–3] and for the Mollow spectrum [4] showing the effects of Rabi oscillations [5,6]. Numerous variations on these themes, such as time correlations between the fluorescent sidebands [7], and the generation of the squeezed states of light from resonance fluorescence [8–10] have also been investigated.

However, a rather fundamental quantum effect, concerned with interference of light from single atoms, seems to have escaped observation so far. Whereas any two classical electromagnetic waves of similar frequencies can, in principle, give rise to observable second-order interference effects, this is not true for quantum fields, because the phase of a quantum field does not always have a value. For example, the fluorescent light produced by two excited single atoms cannot produce second-order interference for just this reason. Another way to understand this is to notice that in such cases the source of each detected photon can, in principle, be identified by an auxiliary measurement that does not significantly disturb the atoms [11,12]. For the same reason it has been suggested that, when the fluorescent light from a coherently driven atom is allowed to interfere with a strong classical light beam of similar frequency, the visibility of the resultant interference should vanish at just those moments when the driven atom is fully excited and the rate of fluorescence is maximum [13].

Such an experiment appears to be most practicable with a trapped atom confined within a region much smaller than a wavelength, so that the optical path differences in the experiment can be held constant to a fraction of the wavelength. With moving atoms in an atomic beam, such as those used in previous experiments [1–3,5–7] the proposed interference experiment appears to encounter formidable difficulties. The reason is that distances relative to the coherent pump field and relative to the coherent reference field have to be held constant to a fraction of a wavelength, and therefore the atomic beam cross section has to be held to this size also. This limits the atomic flux and consequently the photon rate so severely that the interference experiment is not feasible.

In the following we examine another approach to the same experimental problem, in which the position of the atom is not severely restricted and a larger atomic flux can therefore be used. We show below that the light which is elastically scattered from the coherently excited atom is itself coherent, and therefore can serve as the coherent reference in the investigation of interference between a fluorescent photon and a coherent or classical field [13]. Because both the elastically scattered light and the fluorescent light follow the same optical path, the path lengths are balanced automatically and it is only necessary to separate the two with a narrow-band filter and then to recombine them in order to study the interference.

Let $\hat{E}_1^{(+)}(\mathbf{r},t)$ and $\hat{E}_2^{(+)}(\mathbf{r},t)$ be the positive frequency parts of the elastically scattered and the fluorescent contributions to the optical field at $(\mathbf{r},t)$ from a coherently driven atom located at the origin. We identify Hilbert space operators by a caret. If both fields at $(\mathbf{r},t)$ are polarized, quasimonochromatic and can be approximated by plane waves, we may write

$$\begin{aligned}\hat{\mathbf{E}}_1^{(+)}(\mathbf{r},t)&=\boldsymbol{\epsilon}\hat{E}_1(t)e^{i(\mathbf{k}_1\cdot\mathbf{r})}\,,\\ \hat{\mathbf{E}}_2^{(+)}(\mathbf{r},t)&=\boldsymbol{\epsilon}\hat{E}_2(t)e^{i(\mathbf{k}_1\cdot\mathbf{r})}\,,\end{aligned}\tag{1}$$

where $\boldsymbol{\epsilon}$ is a unit polarization vector. If these fields are combined at a photodetector with a relative phase difference ϕ that can be varied, then the total field at the detector is

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)=\boldsymbol{\epsilon}[\hat{E}_1(t)e^{i(\mathbf{k}_1\cdot\mathbf{r}+\phi)}+\hat{E}_2(t)e^{i(\mathbf{k}_2\cdot\mathbf{r})}]\,,\tag{2}$$

and the photodetection probability at $(\mathbf{r},t)$ is proportional to [14]

$$\langle \hat{\mathbf{E}}^{(-)}(\mathbf{r},t)\cdot\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)\rangle = \langle \hat{E}_1^{(-)}(t)\hat{E}_1^{(+)}(t)\rangle + \langle \hat{E}_2^{(-)}(t)\hat{E}_2^{(+)}(t)\rangle + 2|\langle \hat{E}_1^{(-)}(t)\hat{E}_2^{(+)}(t)\rangle|\cos[(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{r}-\phi] \ . \quad (3)$$

In the special case in which field $\hat{E}_1^{(+)}(t)$ is in a coherent state and has eigenvalue $E_1(t)$, we have

$$\langle \hat{E}_1^{(-)}(t)\hat{E}_2^{(+)}(t)\rangle = E_1^*(t)\langle \hat{E}_2^{(+)}(t)\rangle \ , \quad (4)$$

so that the visibility of the interference is proportional to $\langle \hat{E}_2^{(+)}(t)\rangle$ and vanishes whenever $\langle \hat{E}_2^{(+)}(t)\rangle = 0$. We now proceed to show that if the total field at $\mathbf{r}$ is passed through a sufficiently narrow-band optical filter, then the light emerging is indeed coherent.

II. SCATTERED FIELD IN RESONANCE FLUORESCENCE

We suppose that a two-level atom of complex ($\Delta m = \pm 1$) transition dipole moment $\boldsymbol{\mu}_{12}$ and atomic level spacing $\hbar\omega_0$ is located at the origin and is subjected to a coherent, monochromatic exciting or pump field of electric-field amplitude $\boldsymbol{\epsilon}Ee^{-i\omega_1 t}$. We take $\boldsymbol{\epsilon}Ee^{-i\omega_1 t}$ to be the eigenvalue of $\hat{\mathbf{E}}_{\text{free}}^{(+)}(\mathbf{r},t)$ belonging to the multimode coherent state $|\{v\}\rangle$ that corresponds to the exciting field, and we shall suppose that $\boldsymbol{\epsilon}\cdot\boldsymbol{\mu}_{12} = |\boldsymbol{\mu}_{12}|$ and that this field vanishes at the detector. In the following we shall make frequent use of results for this problem derived previously by Kimble and Mandel [15], where it is shown that the total electric field at $(\mathbf{r},t)$ in the far field of the atom is expressible as

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r},t) = \frac{\omega_0^2|\boldsymbol{\mu}_{12}|}{4\pi\epsilon_0 c^2 r}\left[\boldsymbol{\mu}_{12} - \frac{(\boldsymbol{\mu}_{12}\cdot\mathbf{r})\mathbf{r}}{r^2}\right]\hat{b}(t-r/c) + \hat{\mathbf{E}}_{\text{free}}^{(+)}(\mathbf{r},t) \quad (t \ge r/c) \ . \quad (5)$$

$\hat{b}(t)$ is the atomic lowering operator at time t. The initial quantum state for the atom is the lower state $|1\rangle$, and since $\hat{\mathbf{E}}_{\text{free}}^{(+)}$ represents the driving field we have

$$\hat{\mathbf{E}}_{\text{free}}^{(+)}(\mathbf{r},t)|\psi_1\rangle = 0 \ , \quad \hat{b}(0)|\psi_1\rangle = 0 \quad (6)$$

for the combined state $|\psi_1\rangle \equiv |1\rangle_A \otimes |\{v\}\rangle_F$ of atom and field, provided $\mathbf{r}$ is outside the exciting field. For simplicity, we henceforth ignore the propagation delay r/c in Eq. (5), which can be made small. As in Ref. [15] we find it useful to introduce slowly varying dynamical variables which have had the highly oscillatory factor $\exp(\pm i\omega_0 t)$ removed. We define

$$\hat{b}_s(t) \equiv \hat{b}(t)e^{i\omega_0 t} \ . \quad (7)$$

Then from Eqs. (5)–(7) we obtain

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)|\psi_1\rangle = \mathbf{K}e^{-i\omega_0 t}\hat{b}_s(t)|\psi_1\rangle \ , \quad (8)$$

where $\mathbf{K}$ stands for the factor to the left of $\hat{b}(t-r/c)$ in Eq. (5).

III. NARROW-BAND FILTERING

As the coherent driving field is monochromatic and of frequency ω_1, so is the elastic scattering. We now consider more explicitly the action of a narrow-band optical filter centered on frequency ω_1 on the light emitted from the atom. Let $\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)$ represent the total field radiated by the atom falling on the filter at $\mathbf{r}$ at time t, and let $\hat{\mathbf{E}}_0^{(+)}(t)$ denote the field at the filter output. Then from linearity and the time translation property, if the exciting field is turned on at time $t=0$ we must have

$$\hat{\mathbf{E}}_0^{(+)}(t) = \int_0^t dt' f(t-t')\hat{\mathbf{E}}^{(+)}(\mathbf{r},t') \quad (t \ge 0) \ , \quad (9)$$

where $f(t)$ is the real impulse response of the optical filter, which always vanishes for $t<0$. Let the passband of the filter be centered at the coherent driving frequency ω_1 and be of bandwidth σ, with $\sigma \ll \beta$, where β is half the Einstein A coefficient for the atomic transition. For example, we could take $f(t)$ to be of the form

$$f(t) = \sigma e^{-\sigma t}\cos\omega_1 t \ , \quad (10)$$

in which case the frequency response $F(\omega)$ of the filter would be of the Lorentzian form

$$F(\omega) = \int_0^\infty f(t)e^{i\omega t}dt = \frac{1}{2}\left[\frac{1}{1-i(\omega+\omega_1)/\sigma} + \frac{1}{1-i(\omega-\omega_1)/\sigma}\right] \quad (11a)$$

$$\approx \frac{1/2}{1-i(\omega-\omega_1)/\sigma} \quad \text{when } \omega > 0 \ . \quad (11b)$$

Alternatively, if $f(t)$ is of the Gaussian form

$$f(t) = \frac{2\sigma}{\sqrt{2\pi}}e^{-\sigma^2 t^2/2}\cos\omega_1 t \quad (t>0) \ , \quad (12)$$

then the spectral response function of the filter is Gaussian and given by

$$F(\omega) = \int_0^\infty f(t)e^{i\omega t}dt = e^{-(\omega+\omega_1)^2/2\sigma^2} + e^{-(\omega-\omega_1)^2/2\sigma^2} \quad (13a)$$

$$\approx e^{-(\omega-\omega_1)^2/2\sigma^2} \quad \text{when } \omega > 0 \ . \quad (13b)$$

More generally, we shall take

$$f(t) = k(t)\cos\omega_1 t \ , \quad (14)$$

where $k(t)$ is any real function of t of width $1/\sigma$ that vanishes for $t<0$, whose peak value occurs close to $t=0$, which satisfies the condition

$$\int_0^t k(t')dt' = 1 \quad \text{when } \sigma t \gg 1 \ . \quad (15)$$

After combining Eqs. (5), (6), (9), and (14), we obtain

$$\hat{\mathbf{E}}_0^{(+)}(t)|\psi_1\rangle = \mathbf{K}\int_0^t dt' k(t-t')\cos\omega_1(t-t') \times e^{-i\omega_0 t'}\hat{b}_s(t')|\psi_1\rangle \ . \quad (16)$$

If the filter output is to be of bandwidth σ, then we must focus on sufficiently long times t such that $t \gg 1/\sigma$.

Then, since $\sigma \ll \beta$, it follows that $t \gg 1/\beta$ necessarily also. After expanding the cosine we obtain

$$\hat{\mathbf{E}}_0^{(+)}(t)|\psi_1\rangle = \tfrac{1}{2}\mathbf{K}\int_0^t dt' k(t-t')[e^{i\omega_1 t}e^{-i(\omega_1+\omega_0)t'} + e^{-i\omega_1 t}e^{i(\omega_1-\omega_0)t'}] \times \hat{b}_s(t')|\psi_1\rangle \ .$$

The term oscillating at the double optical frequency $\omega_1+\omega_0$ obviously will make a negligible contribution to the integral compared with the other one, so that we have, to a good approximation,

$$\hat{\mathbf{E}}_0^{(+)}(t)|\psi_1\rangle = \tfrac{1}{2}\mathbf{K}e^{i\omega_1 t}\int_0^t dt' k(t-t')e^{i(\omega_1-\omega_0)t'} \times \hat{b}_s(t')|\psi_1\rangle \ . \quad (17)$$

Because of the factor $k(t-t')$, which is nonzero only for t' in the range $t-1/\sigma$ to t, roughly speaking, we are concerned only with the long-time behavior of $\hat{b}_s(t')$. After a long time, when all transients have died out, $\hat{b}_s(t')|\psi_1\rangle$ either becomes constant in time or it exhibits oscillatory behavior with frequencies determined by the parameters β, γ, μ_{12}, $\omega_0-\omega_1$, $\{v\}$, etc. γ is the Lamb shift for the given transition of the two-level atom. Accordingly we write quite generally, for any initial state of the form $\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle$ with $|\psi_j\rangle \equiv |j\rangle_A|\{v\}\rangle_F$, $j=1,2$,

$$e^{i(\omega_1-\omega_0)t}\hat{b}_s(t)(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle) = \sum_{\{n\}}\{[A_{\{n\}}+B_{\{n\}}(t)]|1\rangle_A|\{n\}\rangle_F + [C_{\{n\}}+D_{\{n\}}(t)]|2\rangle_A|\{n\}\rangle_F\} \ . \quad (18)$$

Here $|\{n\}\rangle_F$ is the multimode Fock state of the electromagnetic field and $\{n\}$ stands for the set of all photon occupation numbers. We have chosen to separate the coefficients of $|1\rangle_A|\{n\}\rangle_F$ and $|2\rangle_A|\{n\}\rangle_F$ into constant parts and time-dependent parts, with the understanding that $B_{\{n\}}(t)$ and $D_{\{n\}}(t)$ are oscillatory in the long-time limit, but that $1/\sigma$ is much longer than any of the periods of oscillation.

We now use Eq. (18) to calculate the expectation of $\exp[i(\omega_1-\omega_0)t]\hat{b}_s(t)$ in the state $(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle)/(|\eta_1|^2+|\eta_2|^2)^{1/2}$. Recalling that the scalar product

$$\langle\{v\}|\{n\}\rangle = \prod_\lambda e^{-|v_\lambda|^2/2}\frac{v_\lambda^{*n_\lambda}}{\sqrt{n_\lambda!}} \ , \quad (19)$$

where λ is a mode label and the product is taken over all modes, we obtain

$$\frac{e^{i(\omega_1-\omega_0)t}}{(|\eta_1|^2+|\eta_2|^2)}(\eta_1^*\langle\psi_1|+\eta_2^*\langle\psi_2|)\hat{b}_s(t)(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle) = \frac{1}{(|\eta_1|^2+|\eta_2|^2)}\sum_{\{n\}}\{\eta_1^*[A_{\{n\}}+B_{\{n\}}(t)]+\eta_2^*[C_{\{n\}}+D_{\{n\}}(t)]\}\prod_\lambda e^{-|v_\lambda|^2/2}\frac{v_\lambda^{*n_\lambda}}{\sqrt{n_\lambda!}} \ . \quad (20)$$

Now it has been shown [15] that the expectation on the left-hand side is independent of t and independent of the initial state in the long-time limit for any $|\{v\}\rangle$. Hence we must have

$$\eta_1^*B_{\{n\}}(t)+\eta_2^*D_{\{n\}}(t)=0 \ ,$$

from which it follows that

$$B_{\{n\}}(t)=\eta_2^*Z_{\{n\}}(t) \ , \quad D_{\{n\}}(t)=-\eta_1^*Z_{\{n\}}(t) \ . \quad (21)$$

The $Z_{\{n\}}(t)$, like $B_{\{n\}}(t)$ and $C_{\{n\}}(t)$, are oscillatory functions with periods much less than $1/\sigma$. Then Eq. (20) becomes

$$\frac{\exp[i(\omega_1-\omega_0)t]}{|\eta_1|^2+|\eta_2|^2}(\eta_1^*\langle\psi_1|+\eta_2^*\langle\psi_2|)\hat{b}_s(t)(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle) = \frac{1}{|\eta_1|^2+|\eta_2|^2}\sum_{\{n\}}(\eta_1^*A_{\{n\}}+\eta_2^*C_{\{n\}})\prod_\lambda e^{-|v_\lambda|^2/2}\frac{v_\lambda^{*n_\lambda}}{\sqrt{n_\lambda!}} \ , \quad (22)$$

and if this is to be independent of η_1,η_2, we require

$$A_{\{n\}}=\eta_1 H_{\{n\}} \ , \quad C_{\{n\}}=\eta_2 H_{\{n\}} \ . \quad (23)$$

The expectation on the left-hand side of Eq. (22) has been evaluated explicitly in the long-time limit. It is given by [15]

$$\frac{e^{i(\omega_1-\omega_0)t}}{|\eta_1|^2+|\eta_2|^2}(\eta_1^*\langle\psi_1|+\eta_2^*\langle\psi_2|)\hat{b}_s(t)(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle) \Longrightarrow \frac{(-\frac{1}{2}\Omega/\beta)(1+iD/\beta)}{\frac{1}{2}\Omega^2/\beta^2+1+D^2/\beta^2} \ , \quad (24)$$

where Ω is the atomic Rabi frequency and $D\equiv(\omega_1-\omega_0+\gamma)$ is the detuning. Comparison of Eqs. (23) and (24) then shows that we must have

$$H_{\{n\}}=\left[\frac{-\frac{1}{2}(\Omega/\beta)(1+iD/\beta)}{\frac{1}{2}\Omega^2/\beta^2+1+D^2/\beta^2}\right]\prod_\lambda \frac{v_\lambda^{n_\lambda}}{\sqrt{n_\lambda!}}e^{-|v_\lambda|^2/2} . \tag{25}$$

We now use Eqs. (21), (23), and (25) in Eq. (18) and obtain

$$\begin{aligned}e^{i(\omega_1-\omega_0)t}\hat{b}_s(t)(\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle)\\ &=\frac{(-\frac{1}{2}\Omega/\beta)(1+iD/\beta)}{\frac{1}{2}\Omega^2/\beta^2+1+D^2/\beta^2}(\eta_1|1\rangle+\eta_2|2\rangle)\sum_{\{n\}}\prod_\lambda\left[\frac{v_\lambda^{n_\lambda}}{\sqrt{n_\lambda!}}e^{-|v_\lambda|^2/2}|n_\lambda\rangle\right]+\sum_{\{n\}}Z_{\{n\}}(t)|\{n\}\rangle(\eta_2^*|1\rangle-\eta_1^*|2\rangle)\\ &=\frac{(-\frac{1}{2}\Omega/\beta)(1+iD/\beta)}{\frac{1}{2}\Omega^2/\beta^2+1+D^2/\beta^2}[\eta_1|\psi_1\rangle+\eta_2|\psi_2\rangle]+\sum_{\{n\}}Z_{\{n\}}(t)|\{n\}\rangle[\eta_2^*|1\rangle-\eta_1^*|2\rangle] .\end{aligned} \tag{26}$$

Finally, we substitute Eq. (26) into Eq. (17) with $\eta_2=0,\eta_1=1$. With the help of Eq. (5) together with

$$\int_0^t k(t-t')Z_{\{n\}}(t')dt'\cong 0 , \tag{27}$$

because of the oscillatory behavior of $Z_{\{n\}}(t)$, we then obtain

$$\hat{\mathbf{E}}_0^{(+)}(t)|\psi_1\rangle=\tfrac{1}{2}\mathbf{K}e^{-i\omega_1 t}\frac{(-\frac{1}{2}\Omega/\beta)(1+iD/\beta)}{\frac{1}{2}\Omega^2/\beta^2+1+D^2/\beta^2}|\psi_1\rangle . \tag{28}$$

Hence $|\psi_1\rangle$ is the right eigenstate of $\hat{\mathbf{E}}_0^{(+)}(t)$ in the long-time limit, and the corresponding eigenvalue is the expectation $\langle\hat{\mathbf{E}}_0^{(+)}(t)\rangle$. After a long time t the field emerging from the narrow-band filter is therefore in the coherent state $|\{v\}\rangle$.

IV. APPLICATION TO AN INTERFERENCE EXPERIMENT

Having shown that the output of the narrow-band filter which is illuminated with the fluorescence from the atom is in a coherent state, like the pump field, we now consider the interference experiment illustrated in Fig. 1. An atom of an atomic beam is illuminated by a coherent pump field near resonance. A microscope objective collects some of the emitted light, which is then split into two parts with the help of beam splitter BS_1. One part enters the narrow band filter, whose output is combined with the other part at BS_3, and the mixed beams fall on detector D3. A phase shifter PS is inserted into one interferometer arm, as shown, and a portion of the fluorescent light in the other arm passes through beam splitter BS_2 to detector D2. The outputs of D2 and D3 go to the start and the stop inputs, respectively, of a time-to-digital converter (TDC), which measures the time difference between photoelectric pulses at its two inputs.

The appearance of a start pulse at time t signals the presence of an atom in the field of view of the microscope objective, which is in the ground state at the moment t. At a subsequent time $t+\tau$ the field $\hat{E}_3^{(+)}(t+\tau)$ at detector D3 is a linear superposition of the fluorescent field $\hat{E}_F^{(+)}(t)$ and the phase-shifted filter output $\hat{E}_0^{(+)}(t)$,

$$\hat{E}_3^{(+)}(t+\tau)=\eta_1\hat{E}_F^{(+)}(t+\tau)+\eta_2\hat{E}_0^{(+)}(t+\tau) . \tag{29}$$

Hence the probability of photodetection by D3 at time $t+\tau$ is proportional to

$$\begin{aligned}\langle\hat{E}_3^{(-)}(t+\tau)\hat{E}_3^{(+)}(t+\tau)\rangle\\ &=|\eta_1|^2\langle\hat{I}_F(t+\tau)\rangle+|\eta_2|^2I_0(t+\tau)\\ &\quad+\eta_1^*\eta_2\langle\hat{E}_F^{(+)}(t+\tau)\rangle E_0(t+\tau)\\ &\quad+\eta_1\eta_2^*E_0^*(t+\tau)\langle\hat{E}_F^{(+)}(t+\tau)\rangle .\end{aligned} \tag{30}$$

In this equation $E_0(t)$ is the eigenvalue of $\hat{E}_0^{(+)}(t)$ in the initial state. It follows that the interference terms involve $\langle\hat{E}_F^{(+)}(t+\tau)\rangle$ and its conjugate, and the interference vanishes or becomes small at those times when $\langle\hat{E}_F^{(+)}(t+\tau)\rangle$ vanishes or becomes small [13]. The time delay τ, following the return of the atom to the lower state at time t, determines whether the interference is strong or weak. Loosely speaking, the visibility of the interference is expected to be small at times when $\tau\sim\pi/\Omega$, when the atomic excitation is greatest in the Rabi cycle.

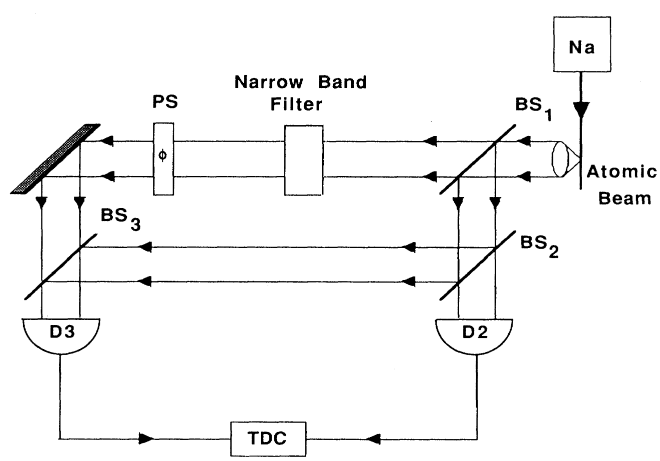

FIG. 1. Outline of the proposed experiment under discussion.

V. COINCIDENCE COUNTING RATES

Although the foregoing argument shows that there is no observable interference at times $t+\tau$ when $\langle \hat{E}_F^{(+)}(t+\tau)\rangle=0$, an experiment to study this effect would have to be based on delayed coincidence counting with the two detectors D2 and D3. Such a measurement cannot be adequately described by a second-order correlation function, as in Eq. (30). Let us therefore examine the joint probability density $P_2(t,t+\tau)$ for detections of a photon by D2 at time t and another photon by D3 at a later time $t+\tau$. If $\hat{E}^{(+)}(t)$ is the field incident on BS_1, then from Fig. 1 this is given by

$$P_2(t,t+\tau)=\alpha_3\alpha_2\langle \mathfrak{R}_1^*\mathfrak{T}_2^*\hat{E}^{(-)}(t)[\mathfrak{T}_3^*\hat{E}_0^{(-)}(t+\tau)e^{i\phi}+\mathfrak{R}_1^*\mathfrak{R}_2^*\mathfrak{R}_3^*\hat{E}^{(-)}(t+\tau)] \times[\mathfrak{T}_3\hat{E}_0^{(+)}(t+\tau)e^{-i\phi}+\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3\hat{E}^{(+)}(t+\tau)]\mathfrak{R}_1\mathfrak{T}_2\hat{E}^{(+)}(t)\rangle \; . \tag{31}$$

α_3 and α_2 are the quantum efficiencies of the detectors D3 and D2, respectively, and $\mathfrak{R}_j$ and $\mathfrak{T}_j$ are the complex reflectivity and transmissivity of beam splitter j ($j=1,2,3$). The visibility ϑ of the interference pattern is then given by

$$\vartheta=\frac{2|\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3\mathfrak{T}_3||\langle \hat{E}^{(-)}(t)\hat{E}^{(-)}(t+\tau)\hat{E}_0^{(+)}(t+\tau)\hat{E}^{(+)}(t)\rangle|}{|\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3|^2\langle \hat{E}^{(-)}(t)\hat{E}^{(-)}(t+\tau)\hat{E}^{(+)}(t+\tau)\hat{E}^{(+)}(t)\rangle+|\mathfrak{T}_3|^2\langle \hat{E}^{(-)}(t)\hat{E}_0^{(-)}(t+\tau)\hat{E}_0^{(+)}(t+\tau)\hat{E}^{(+)}(t)\rangle}\;, \tag{32}$$

provided $t \gg 1/\sigma \gg 1/\beta$. If we now use Eq. (9) to express $\hat{E}_0^{(+)}(t)$ in terms of $\hat{E}^{(+)}(t)$ via the convolution integral, then it is apparent from Eq. (31) that we need to evaluate certain three-time autocorrelation functions of the general form

$$\Gamma^{(2,2)}(t,t',t'',t)\equiv\langle \hat{E}^{(-)}(t)\hat{E}^{(-)}(t')\hat{E}^{(+)}(t'')\hat{E}^{(+)}(t)\rangle \quad (0\le t'\le t'') \; . \tag{33}$$

The procedure for the evaluation is outlined in Appendix A where it is shown that, for the special case of on-resonance excitation,

$$\Gamma^{(2,2)}(t,t',t'',t)=\begin{cases} K^4[\langle \hat{R}_3(t)\rangle+\frac{1}{2}]g(t'-t,t''-t)e^{-i\omega_1(t''-t)} & \text{when } t\le t'\le t'' \text{ or } t'\le t''\le t \\ K^4[\langle \hat{R}_3(t)\rangle+\frac{1}{2}]\left\{\langle \hat{b}_s^\dagger(t'-t)\rangle\langle \hat{b}_s(t''-t)\rangle e^{i\gamma(t'-t'')} + \frac{1}{4}\prod_{t_j=t',t''}\left[e^{-3\beta/2(t_j-t)}\left[\cos\Omega'(t_j-t)+\frac{\beta}{2\Omega}\sin\Omega'(t_j-t)\right] - e^{-\beta(t_j-t)}\right]\right\} & \text{when } t'\le t\le t'' \; . \end{cases} \tag{34}$$

The two-time correlation function

$$g(t,\tau)\equiv\langle \hat{b}_s^\dagger(t)\hat{b}_s(t+\tau)\rangle \tag{35}$$

is identical to the one defined in Ref. [15]. If ω_f is the frequency at which the filter passband is centered, the numerator $\mathcal{N}$ and denominator $\mathcal{D}$ in Eq. (32) become

$$\mathcal{N}\equiv\left[(\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3\mathfrak{T}_3^*e^{i\Phi})\left[\mathfrak{T}_1^*\int_0^{t+\tau}dt'k(t+\tau-t')\cos\omega_f(t+\tau-t')\Gamma^{(2,2)}(t,t',t+\tau,t)\right]+\text{c.c.}\right] \tag{36}$$

and

$$\begin{aligned}\mathcal{D}&\equiv|\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3|^2\Gamma^{(2,2)}(t,t+\tau,t+\tau,t) \\ &\quad+|\mathfrak{T}_3|^2|\mathfrak{T}_1|^2\int_0^{t+\tau}dt''\int_0^{t+\tau}dt'k(t+\tau-t'')k(t+\tau-t')\cos\omega_f(t+\tau-t'') \\ &\qquad\times\cos\omega_f(t+\tau-t')\Gamma^{(2,2)}(t,t',t'',t) \\ &=|\mathfrak{R}_1\mathfrak{R}_2\mathfrak{R}_3|^2\Gamma^{2,2)}(t,t+\tau,t+\tau,t) \\ &\quad+|\mathfrak{T}_3|^2|\mathfrak{T}_1|^2\left[\int_0^{t+\tau}dt''\int_0^{t+\tau}dt'k(t+\tau-t')k(t+\tau-t'') \right. \\ &\qquad\times\cos\omega_f(t+\tau-t'')\{\Gamma^{(2,2)}(t,t',t'',t)e^{i\omega_0(t'-t'')}+\text{c.c.}\} \\ &\qquad\left.-(t+\tau)\int_0^{t+\tau}dt'k^2(t+\tau-t')\cos^2\omega_f(t+\tau-t')\Gamma^{(2,2)}(t,t',t',t)\right] \; .\end{aligned} \tag{37}$$

On equating the filter center frequency ω_f with the laser frequency ω_1 (which is equal to $\omega_0-\gamma$ on resonance) and going to the long-time limit ($t \gg 1/\sigma$) and the narrow-bandpass condition $\sigma \ll \beta$, we see that

$$\mathcal{N}=\Re_1\Re_2\Re_3\mathfrak{T}_3^*\mathfrak{T}_1^*|\frac{k(\tau)}{\sigma}K^4(\mathbf{r})\left[-\frac{\Omega\beta}{\Omega^2+2\beta^2}\right][\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\langle\hat{b}_s(\tau)\rangle e^{-i\gamma\tau}\cos(\phi+\eta) \tag{38}$$

and

$$\mathcal{D}=|\Re_1\Re_2\Re_3|^2K^4(\mathbf{r})[\langle R_3(t)\rangle+\tfrac{1}{2}][\langle\hat{R}_3(\tau)\rangle+\tfrac{1}{2}]+|\mathfrak{T}_3|^2|\mathfrak{T}_1|^2K^4(\mathbf{r})[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\left[-\frac{\Omega\beta}{\Omega^2+2\beta^2}\right]^2\frac{k(\tau)}{\sigma}\ . \tag{39}$$

η is an arbitrary but constant phase associated with the complex transmissivities and reflectivities of all the beam splitters. On substituting Eqs. (38) and (39) into Eq. (32) we obtain for the visibility $\vartheta(\tau)$ in the long-time limit and for a narrow filter bandwidth

$$\vartheta(\tau)=\frac{2|\Re_1\Re_2\Re_3^*\mathfrak{T}_3^*\mathfrak{T}_1^*|\left[\frac{\Omega\beta}{\Omega^2+2\beta^2}\right]|\langle\hat{b}_s(\tau)\rangle|}{|\Re_1\Re_2\Re_3|^2[\langle\hat{R}_3(\tau)\rangle+\tfrac{1}{2}]+|\mathfrak{T}_3|^2|\mathfrak{T}_1|^2\left[\frac{\Omega\beta}{\Omega^2+2\beta^2}\right]^2}\ . \tag{40}$$

Figure 2 shows plots of visibility $\vartheta(\tau)$ as a function of τ for two values of Ω/β when all beam splitters are 50%:50%. The visibility falls to zero at those times when the atom is either fully excited or fully de-excited (cf. the graphs in Ref. [13]). The peak visibility of near 100% that is found for $\beta\tau \ll 1$ occurs at the time when the amplitude of the growing fluorescent field of the atoms equals the amplitude of the elastically scattered field that emerges from the narrow-band filter. At that time the two possible photon paths through the interferometer are completely indistinguishable, which makes the degree of coherence unity [16] as expected.

VI. CONCLUSION

Equation (40) confirms the conclusion that was already reached by the more general argument given in Sec. III above. It is possible to study the interference between fluorescent photons from a single atom and a coherent field by making use of the elastically scattered light from the atom. As the state of the atom goes through its Rabi cycle of oscillation, the visibility is expected to vanish at those times when the atom is close to being in the fully excited state, when the source of the detected photons can in principle be identified. This is yet another example of the relation between coherence and indistinguishability [16].

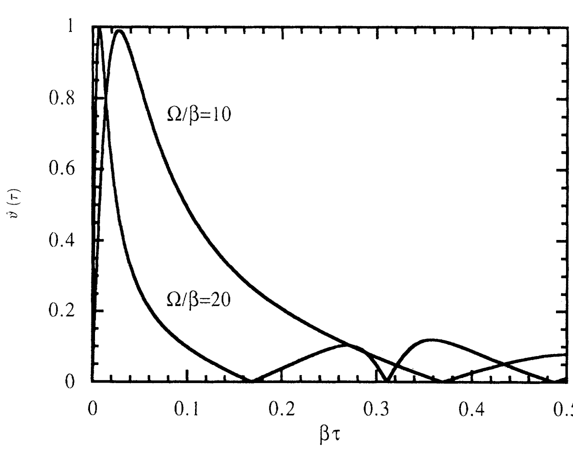

FIG. 2. Visibility $\vartheta(\tau)$ as function of delay τ given by Eq. (40), for $\Omega/\beta=10$, 20. All beam splitters are assumed to have $\mathfrak{T}_j=1/\sqrt{2}=\Re_j$ ($j=1,2,3$).

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APPENDIX A

We wish to evaluate

$$\Gamma^{(2,2)}(t,t',t'',t) \equiv \langle\hat{E}^{(-)}(t)\hat{E}^{(-)}(t')\hat{E}^{(+)}(t'')\hat{E}^{(+)}(t)\rangle \quad (t'\le t'')\ . \tag{A1}$$

After separating out the rapidly oscillating factor and writing

$$\hat{E}^{(+)}(t)=\hat{E}_s^{(+)}(t)e^{-i\omega_0 t}\ , \tag{A2}$$

where $\hat{E}_s^{(+)}(t)$ is given by the usual dipole formula

$$\hat{E}_s^{(+)}(t)=K(\mathbf{r})\hat{b}_s(t)+\hat{E}_{s,\text{free}}^{(+)}(\mathbf{r},t)\ , \tag{A3}$$

we have

$$\Gamma^{(2,2)}(t,t'',t',t) =K^4(\mathbf{r})e^{-i\omega_0(t''-t')}\langle\hat{b}_s^\dagger(t)\hat{b}_s^\dagger(t')\hat{b}_s(t'')\hat{b}_s(t)\rangle\ . \tag{A4}$$

We have made use of the fact that

$$\hat{E}_{s,\text{free}}(\mathbf{r},t)|\psi\rangle=0 \tag{A5}$$

for the initial state $|\psi\rangle$ and

$$[\hat{b}_s(t),\hat{E}^{(+)}_{s,\text{free}}(t')]=0 \quad \text{for all } t,t' , \tag{A6}$$

as shown in Appendix B. We have approached the calculation of the correlation function in (A4) in the Heisenberg picture via the same techniques that were used in Ref. [15].

We make use of the coupled integral Eqs. (75) and (76) of Ref. [15] to construct three different multitime correlations:

$$F(t,t',t'',t)\equiv\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}^\dagger_s(t'')\hat{b}_s(t)\rangle \times e^{i(\omega_0-\omega_1)(t_j+t'')} ,$$

$$G(t,t',t'',t)\equiv\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}^\dagger_s(t'')\hat{b}_s(t)\rangle e^{i(\omega_1-\omega_0)(t''-t_j)} , \tag{A7}$$

$$H(t,t',t'',t)\equiv\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{R}_3(t'')\hat{b}_s(t)\rangle e^{i(\omega_0-\omega_1)t} ,$$

where

$$t_j=\begin{cases} t & \text{when } t'\le t\le t'' \\ t' & \text{when } t\le t'\le t'' \text{ or } t'\le t''\le t \ . \end{cases} \tag{A8}$$

These inequalities are dictated by Markovian considerations. Using Eqs. (83) of Ref. [15] and the relation $\{\hat{b}^\dagger_s(t),\hat{b}_s(t)\}=1$ and proceeding along the same lines as in Eqs. (80)–(86) of Ref. [15], we arrive at the following Volterra integral equation for the combination $G(t,t',t'',t)+F(t,t',t'',t)$:

$$G(t,t',t'',t)+F(t,t',t'',t)=m_j(t,t',t'')+\int_0^{t''-t_j}dt_1\tilde{K}(t''-t_j-t_1)\{G(t,t',t_1,t)+F(t,t',t_1,t)\} \tag{A9}$$

with integral kernel $\tilde{K}(\tau)=(\Omega^2/\beta)(e^{-2\beta\tau}-e^{-\beta\tau})$ and with inhomogeneous term

$$m_j(t,t',t'')=\begin{cases} \langle b^\dagger_s(t)\hat{b}^\dagger_s(t')b^\dagger_s(t)\hat{b}_s(t)\rangle e^{-2i(\omega_1-\omega_0)t}e^{-\beta(t''-t)}-\dfrac{\Omega}{\beta}\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t)\rangle \\ \qquad\times e^{-i(\omega_1-\omega_0)t}[1-e^{-\beta(t''-t)}] \quad \text{when } t'\le t\le t'' \\ \langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t')\hat{b}_s(t)\rangle e^{-\beta(t''-t')}-\dfrac{\Omega}{\beta}\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t)\rangle \\ \qquad\times e^{-i(\omega_1-\omega_0)(t''-t')}[1-e^{-\beta(t''-t')}] \quad \text{when } t\le t'\le t'' \text{or } t'\le t''\le t \ . \end{cases} \tag{A10}$$

After eliminating $F(t,t',t'',t)$ from Eqs. (A7) and (A9) we find from the definition of $G(t,t',t'',t)$ in Eq. (A7)

$$\begin{aligned}\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t'')\hat{b}_s(t)\rangle &=\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t)\rangle\langle\hat{b}_s(t''-t)\rangle e^{-i(\omega_1-\omega_0)t} \\ &\quad+\tfrac{1}{2}\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}^\dagger_s(t)\hat{b}_s(t)\rangle e^{-2i(\omega_1-\omega_0)t}e^{-i(\omega_1-\omega_0)(t''-t)} \\ &\quad\times\left[e^{-3(\beta/2)(t''-t)}\left[\cos\Omega'(t''-t)+\frac{\beta}{2\Omega'}\sin\Omega'(t''-t)\right]-e^{-\beta(t''-t)}\right] \\ &\qquad\qquad \text{when } t'\le t\le t'' \qquad \text{(A11a)} \\ &=[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]g(t'-t,t''-t')e^{-i(\omega_1-\omega_0)(t''-t')} \quad \text{when } t\le t'\le t'' \text{ or } t'\le t''\le t , \qquad \text{(A11b)}\end{aligned}$$

where $\Omega'\equiv(\Omega^2-\beta^2/4)^{1/2}$. In Eq. (A11b) the two-time correlation function $g(t,\tau)$ is identical to the one defined in Eq. (77) of Ref. [15]. The two-time, third-order correlation function $\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t)\rangle$ can be calculated with the help of Eqs. (100)–(103) of Ref. [15] and is found to be

$$\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}_s(t)\rangle=[\langle R_3(t)\rangle+\tfrac{1}{2}]\langle\hat{b}^\dagger_s(t'-t)\rangle e^{i(\omega_1-\omega_0)t} . \tag{A12}$$

The two-time, fourth-order correlation function in Eq. (A11a) has not previously been evaluated. It can be obtained by proceeding along the same lines as in the calculation of $g(t,\tau)$ in Ref. [15]. This gives

$$\begin{aligned}\langle\hat{b}^\dagger_s(t)\hat{b}^\dagger_s(t')\hat{b}^\dagger_s(t)\hat{b}_s(t)\rangle &=\tfrac{1}{2}[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]e^{2i(\omega_1-\omega_0)t}e^{i\gamma(t-t')} \\ &\quad\times\left[e^{-3(\beta/2)(t'-t)}\left[\cos\Omega'(t'-t)+\frac{\beta}{2\Omega'}\sin\Omega'(t'-t)\right]-e^{-\beta(t'-t)}\right] .\end{aligned} \tag{A13}$$

On substituting Eqs. (A13) and (A12) into Eqs. (A11a) and (A4) we obtain finally

$$\Gamma^{(2,2)}(t,t',t'',t)=K^4[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]g(t'-t,t''-t)e^{-i\omega_1(t''-t)} \quad \text{for } t\le t'\le t'' \text{ or } t'\le t''\le t$$

$$=K^4[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\Bigg[\langle\hat{b}_s^\dagger(t'-t)\rangle\langle\hat{b}_s(t''-t)\rangle e^{i\gamma(t'-t'')} + \tfrac{1}{4}\prod_{t_j=t',t''}\Bigg\{e^{-3(\beta/2)(t_j-t)}\left[\cos\Omega'(t_j-t)+\frac{\beta}{2\Omega'}\sin\Omega'(t_j-t)\right] - e^{-\beta(t_j-t)}\Bigg\}\Bigg] \quad \text{for } t'\le t\le t'' . \tag{A14}$$

APPENDIX B

Proof that $[\hat{b}(t-r/c),\hat{\mathbf{E}}^{(+)}_{\text{free}}(\mathbf{r},t')]=0$ for all t,t'

Let

$$\hat{\mathbf{C}}\equiv[\hat{b}(t),\hat{\mathbf{E}}^{(+)}_{\text{free}}(\mathbf{r},t')]$$

$$=\frac{1}{L^{3/2}}\sum_{\mathbf{k},s} l(\omega)\boldsymbol{\epsilon}_{\mathbf{k},s}e^{i[\mathbf{k}\cdot\mathbf{r}-\omega(t'-t)]}[\hat{b}(t),\hat{a}_{\text{free }\mathbf{k},s}(t)] , \quad l(\omega)\equiv i(\hbar\omega/2\epsilon_0)^{1/2} . \tag{B1}$$

But from Eq. (5) the total field is

$$\frac{1}{L^{3/2}}\sum_{\mathbf{k},s} l(\omega)\boldsymbol{\epsilon}_{\mathbf{k},s}\hat{a}_{\text{tot }\mathbf{k},s}e^{i(\mathbf{k}\cdot\mathbf{r})}=\mathbf{K}(\mathbf{r})\hat{b}(t)+\frac{1}{L^{3/2}}\sum_{\mathbf{k},s} l(\omega)\boldsymbol{\epsilon}_{\mathbf{k}s}\hat{a}_{\text{free }\mathbf{k},s}(t)e^{i(\mathbf{k}\cdot\mathbf{r})} . \tag{B2}$$

We now scalar multiply each term in this equation by $\boldsymbol{\epsilon}^*_{\mathbf{k}'s}e^{-i\mathbf{k}'\cdot\mathbf{r}}$ and integrate over the volume L^3. Then we obtain

$$L^{3/2}l(\omega)\hat{a}_{\text{tot }\mathbf{k},s}(t)=\boldsymbol{\epsilon}^*_{\mathbf{k}s}\cdot\int_{L^3}\mathbf{K}(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}\,\hat{b}(t)+L^{3/2}l(\omega)\hat{a}_{\text{free }\mathbf{k},s}(t)$$

or

$$\hat{a}_{\text{free }\mathbf{k},s}(t)=\hat{a}_{\text{tot }\mathbf{k},s}(t)-\boldsymbol{\epsilon}^*_{\mathbf{k}s}\cdot\int\mathbf{K}(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}\frac{1}{L^{3/2}l(\omega)}\hat{b}(t) . \tag{B3}$$

After substituting from Eq. (B3) in Eq. (B1) we arrive at

$$\hat{C}=\frac{1}{L^{3/2}}\sum_{\mathbf{k},s} l(\omega)\boldsymbol{\epsilon}_{\mathbf{k}s}e^{i[\mathbf{k}\cdot\mathbf{r}-\omega(t'-t)]}[\hat{b}(t),\hat{a}_{\text{free }\mathbf{k},s}(t)]=0 . \tag{B4}$$

As this holds for all t,t', we may replace $\hat{b}(t)$ by $\hat{b}(t-r/c)$ in the definition of $\hat{C}$.

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