# Interference between a fluorescent photon and a classical field: An example of nonclassical interference

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Fluorescent photons emitted from an atom will interfere with a classical field only if the atom is not in a pure excited state. This is a strictly quantum-mechanical condition, which can be tested, in principle, by allowing the light emitted from an atom undergoing Rabi oscillations to interfere with the coherent pumping field. The theory of this process is discussed.

## I. INTRODUCTION

Whereas any two classical light waves in principle can give rise to second-order interference effects, and indeed such effects have been observed even with two separate and independent sources,[1–5] the same is not true for quantum fields. The reason is that the phase of a classical field always exists, whereas that of a quantum field does not, even if the field is quasimonochromatic. In particular, because a single photon has no definite phase according to quantum electrodynamics, two single photons cannot exhibit second-order interference.[6,7] However, fourth-order interference effects involving the joint detection of two photons have been predicted[8–11] and observed.[12,13]

Somewhat similar conclusions apply to the interference of a photon with a classical field. This situation might be encountered when the fluorescence from a single atom is mixed with a coherent reference beam, and one looks for second-order interference effects. The interference effects are expected to vanish whenever the initial state of the atom is an eigenstate of the energy, because then the emitted photon has no phase. To put it another way, when one of the two sources used for the interference experiment source is a fully excited atom, then when a photon is detected in the interference plane, it is possible to determine from which source the photon must have come by examining the atom. This rules out any second-order interference, which is always a manifestation of the intrinsic indistinguishability of several possible paths of the detected photon. On the other hand, interference effects between the atomic and the classical source can occur when the source atom is in a superposition state and only partially excited. Needless to say, this phenomenon is nonclassical and does not exist in semiclassical radiation theory.

In the resonance fluorescence of a two-level atom in the presence of a coherent exciting field on or near resonance, the expectation of the atomic excitation changes continuously between lower and upper limits as a result of the Rabi oscillations that the atom undergoes. If the fluorescent light emitted by the atom were mixed with the pumping field, we would have just the situation discussed above; interference effects should come and go as the atomic excitation changes. We then have a prototype of the nonclassical kind of second-order interference experiment. This problem is treated quantitively below.

## II. THEORY OF THE INTERFERENCE PROCESS

Let us consider the interference of a polarized quantum field $\hat{E}_{\mathrm{qu}}(\mathbf{r},t)$ at position $\mathbf{r}$ at time $t$ (Hilbert space operators are labeled by the caret) with a classical field $E_{\mathrm{cl}}(\mathbf{r},t)$. When a photodetector is located at $\mathbf{r}$, the probability $P(\mathbf{r},t)$ of a detection at time $t$ is given by[14]

$$P(\mathbf{r},t)=K\langle(\hat{E}_{\mathrm{qu}}^{(-)}+E_{\mathrm{cl}}^{(-)})(\hat{E}_{\mathrm{qu}}^{(+)}+E_{\mathrm{cl}}^{(+)})\rangle , \tag{1}$$

where $E^{(+)},E^{(-)}$ are positive and negative frequency parts of the total field $E$, and $K$ is a constant characteristic of the detector. In particular, if both $\hat{E}_{\mathrm{qu}}^{(+)}(\mathbf{r},t)$ and $E_{\mathrm{cl}}^{(+)}(\mathbf{r},t)$ can be treated as single-mode fields with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively, and with equal frequencies $\omega$, we can write

$$\begin{aligned}\hat{E}_{\mathrm{qu}}^{(+)}(\mathbf{r},t)&=C\hat{a}_1e^{i(\mathbf{k}_1\cdot\mathbf{r}-\omega t)} ,\\ E_{\mathrm{cl}}^{(+)}(\mathbf{r},t)&=Cv_2e^{i(\mathbf{k}_2\cdot\mathbf{r}-\omega t)} ,\end{aligned} \tag{2}$$

where $\hat{a}_1$ is a photon annihilation operator and $v_2$ is a complex mode amplitude. Then from Eqs. (1) and (2) we have

$$\begin{aligned}P(\mathbf{r},t)&=K\,|\,C\,|^2(\langle\hat{n}_1\rangle+|\,v_2\,|^2+v_2^*\langle\hat{a}_1\rangle e^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}}+\mathrm{c.c.})\\ &=K\,|\,C\,|^2\{\langle\hat{n}_1\rangle+|\,v_2\,|^2+2\,|\,v_2\,|\,|\,\langle\hat{a}_1\rangle\,|\cos[(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}+\arg(\phi_1-\phi_2)]\} ,\end{aligned} \tag{3}$$

 

where $\hat{n}_1 \equiv \hat{a}_1^\dagger \hat{a}_1$, and we have written

$$\langle \hat{a}_1 \rangle = |\langle \hat{a}_1 \rangle| e^{i\phi_1} , \quad v_2 = |v_2| e^{i\phi_2} . \tag{4}$$

We note that $P(\mathbf{r},t)$ varies periodically with position, but only if $\langle \hat{a}_1 \rangle \neq 0$. The visibility $\mathscr{V}$, or the relative modulation amplitude of the interference pattern, is given by

$$\mathscr{V} = \frac{2|v_2||\langle \hat{a}_1 \rangle|}{\langle \hat{n}_1 \rangle + |v_2|^2} . \tag{5}$$

In the special case when the quantum field is derived from an initially fully excited atom, $\langle \hat{a}_1 \rangle = 0$ and there is no interference. On the other hand, for a partly excited atom in a superposition state we generally have $\langle \hat{a}_1 \rangle \neq 0$, and then interference effects are to be expected.

## III. APPLICATION TO RESONANCE FLUORESCENCE

We now apply these considerations explicitly to the fluorescent field produced by a two-level atom in a coherent driving field on resonance. If the atom of level spacing $\hbar\omega_0$ and transition dipole moment $\boldsymbol{\mu}$ is located at the origin, then the positive frequency part of the field at $\mathbf{r},t$ far from the atom is given by the usual dipole formula[15,16]

$$\hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t) = \frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r}\left[\boldsymbol{\mu} - \frac{(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}}{r^2}\right]\hat{b}(t-r/c) + \hat{\mathbf{E}}_{\rm free}^{(+)}(\mathbf{r},t) . \tag{6}$$

Here $\hat{b}(t)$ is the atomic lowering operator and $\hat{\mathbf{E}}_{\rm free}^{(+)}$ is the external or free field. At any point $(\mathbf{r},t)$ where the coherent excitation field vanishes, the expectation value of $\hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t)$ is then given by

$$\langle \hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t) \rangle = \frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r}\left[\boldsymbol{\mu} - \frac{(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}}{r^2}\right]\left\langle \hat{b}\left[t-\frac{r}{c}\right]\right\rangle , \tag{7}$$

so that $\langle \hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t) \rangle$ is completely determined by $\langle \hat{b}(t) \rangle$.

In order to calculate $\hat{b}(t)$ at an arbitrary time $t$ following the turn-on of the coherent driving field, we shall make use of the general integral relation between $\hat{b}(t)$ and the atomic inversion $\hat{R}_3(t)$ that was derived in Ref. 16 [Eq. (28)]. In the special case in which the exciting field is in a coherent state on resonance, and the electric field seen by the atom is given by $\boldsymbol{\varepsilon}\omega_0\mathcal{A}e^{-i\omega_0 t}e^{i\phi}$, with $\mathcal{A},\phi$ real, the integral relation leads to the following equation connecting the expectations of $\hat{b}(t)$ and $\hat{R}_3(t)$:

$$\langle \hat{b}(t) \rangle = e^{i(\phi-\omega_0 t)}\Omega\int_0^t dt' \langle \hat{R}_3(t') \rangle e^{-\beta(t-t')} . \tag{8}$$

Here $\Omega \equiv 2\omega_0\mathcal{A}\boldsymbol{\mu}\cdot\boldsymbol{\varepsilon}/\hbar$ is the atomic Rabi frequency in the external field, $\boldsymbol{\varepsilon}$ is a unit polarization vector, $\beta$ is half the Einstein $A$ coefficient, and it has been assumed that the atom is in the ground state at time $t=0$. The mean inversion $\langle \hat{R}_3(t) \rangle$ under these conditions has been calculated,[16,17] and it takes the form

$$\langle \hat{R}_3(t) \rangle = \left[\frac{-\frac{1}{2}}{\frac{1}{2}\Omega^2/\beta^2+1}\right] \times \left[1+\frac{\Omega^2}{2\beta^2}e^{-3\beta t/2}\left[\cos\Omega' t + \frac{3\beta}{2\Omega'}\sin\Omega' t\right]\right]$$

with

$$\Omega' \equiv (\Omega^2 - \tfrac{1}{4}\beta^2)^{1/2} . \tag{9}$$

When Eq. (9) is substituted under the integral in Eq. (8), we readily find that

$$\langle \hat{b}(t) \rangle = e^{i(\phi-\omega_0 t)}\left[\frac{-\Omega/2\beta}{\Omega^2/2\beta^2+1}\right] \times \left[1-e^{-3\beta t/2}\left[\cos\Omega' t - \frac{\Omega^2-\beta^2}{2\Omega'\beta}\sin\Omega' t\right]\right] , \tag{10}$$

and with the help of Eq. (7),

$$\langle \hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t+r/c) \rangle = \frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r}\left[\boldsymbol{\mu} - \frac{(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}}{r^2}\right]\left[\frac{-\Omega/2\beta}{\Omega^2/2\beta^2+1}\right]e^{i(\phi-\omega_0 t)}\left[1-e^{-3\beta t/2}\left[\cos\Omega' t - \frac{\Omega^2-\beta^2}{2\Omega'\beta}\sin\Omega' t\right]\right] . \tag{11}$$

It is interesting to examine the expectation of $\hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t+r/c)$ predicted by this equation at various times $t$. To make the situation as simple as possible we assume that the atom is subjected to a strong exciting field, so that $\Omega/\beta \gg 1$, and the natural atomic lifetime is very long compared with the period for Rabi oscillations.

(a) At time $t=0$, we always have from Eq. (11)

$$\langle \hat{E}_{\rm qu}^{(+)}(\mathbf{r},r/c) \rangle = 0 . \tag{12}$$

(b) At times $t=n(\pi/\Omega')$, $n=1,3,5,\ldots$, when the atomic excitation given by Eq. (9) is close to a maximum, we find

$$\langle \hat{\mathbf{E}}_{\rm qu}^{(+)}(\mathbf{r},t+r/c) \rangle = -\frac{\omega_0^2}{2\pi\varepsilon_0 c^2 r}[\boldsymbol{\mu}-(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}/r^2]\frac{\beta}{\Omega} \times e^{i(\phi-\omega_0 t)} . \tag{13}$$

(c) At times $t=n2\pi/\Omega'$, $n=0,1,2,\ldots$, when the atomic excitation given by Eq. (9) is at a minimum, we find that $\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle$ is close to zero, and numerically much smaller than the value given by Eq. (13).

(d) At times $t=n(\pi/2\Omega')$, $n=1,3,5,\ldots$, when the atomic excitation is close to 50%, we obtain

$$\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle = \pm\frac{\omega_0^2}{8\pi\varepsilon_0 c^2 r}[\boldsymbol{\mu}-(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}/r^2]e^{i(\phi-\omega_0 t)} . \tag{14}$$

Because we have taken $\Omega/\beta \gg 1$, we see that the expectation value of the field in case (d) is very much larger than that in cases (b) or (c) when the atom is either highly excited or unexcited. At times when $\langle \hat{R}_3(t)\rangle \approx -\frac{1}{2}$, corresponding to an unexcited atom, a zero field is to be expected. On the other hand, $\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}\rangle$ is very small in case (b) even though the emitted light intensity is high, because in this state the phase of the quantum field is indefinite. Therefore, any interference effects are expected to wash out under conditions (b). In the partly excited state corresponding to case (d), on the other hand, the quantum field has a phase and a nonvanishing expectation. Figure 1 shows graphs of $|\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle|$ as a function of time, for several values of the ratio $\Omega/\beta$. When $\Omega/\beta=20$, the kind of behavior expected from the foregoing discussion is indeed observed. For smaller values of $\Omega/\beta$ the competing effects of spontaneous emission modify the Rabi oscillations and cause increasing distortions of $\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}\rangle$.

In order to examine the visibility of the interference pattern we now substitute Eq. (11) in Eq. (1), and assume that the classical field has the same amplitude as the quantum field at time $t=\pi/2\Omega'$, with $\Omega/\beta \gg 1$, i.e.,

$$\mathbf{E}_{\text{cl}}^{(+)}(\mathbf{r},t) \approx \frac{\omega_0^2}{8\pi\varepsilon_0 c^2 r}[\boldsymbol{\mu}-(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}/r^2]e^{i(\mathbf{k}_2\cdot\mathbf{r}-\omega_0 t)} . \tag{15}$$

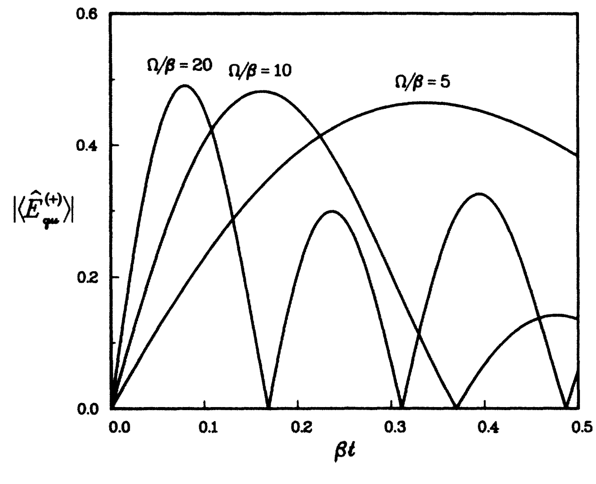


FIG. 1. The expected time variation of $|\langle \hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle|$ given by Eq. (11) in arbitrary units for several different values of $\Omega/\beta$.

For the expectation value

$$\langle \hat{\mathbf{E}}_{\text{qu}}^{(-)}(\mathbf{r},t+r/c)\cdot\hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle$$

of the fluorescent field we obtain from Eq. (6)

$$\langle \hat{\mathbf{E}}_{\text{qu}}^{(-)}(\mathbf{r},t+r/c)\cdot\hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle = \left[\frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r}\right]^2\left|\boldsymbol{\mu}-\frac{(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}}{r^2}\right|^2[\langle \hat{R}_3(t)\rangle+\tfrac{1}{2}] ,$$

when we make use of the relation $\hat{b}^\dagger(t)\hat{b}(t)=\hat{R}_3(t)+\frac{1}{2}$, and with the help of Eq. (9)

$$\langle \hat{\mathbf{E}}_{\text{qu}}^{(-)}(\mathbf{r},t+r/c)\cdot\hat{\mathbf{E}}_{\text{qu}}^{(+)}(\mathbf{r},t+r/c)\rangle = \left[\frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r}\right]^2\left|\boldsymbol{\mu}-\frac{(\boldsymbol{\mu}\cdot\mathbf{r})\mathbf{r}}{r^2}\right|^2\left[\frac{\Omega^2/4\beta^2}{\Omega^2/2\beta^2+1}\right]\left[1-e^{-3\beta t/2)}\left[\cos\Omega' t+\frac{3\beta}{2\Omega'}\sin\Omega' t\right]\right] . \tag{16}$$

When Eqs. (11), (15), and (16) are used in Eq. (1), we readily find for the visibility $\mathcal{V}$ of the resulting interference pattern

$$\mathcal{V}=\frac{\left[\dfrac{\Omega/2\beta}{\Omega^2/2\beta^2+1}\right]\left|1-e^{-3\beta t/2}\left[\cos\Omega' t-\dfrac{\Omega^2-\beta^2}{2\Omega'\beta}\sin\Omega' t\right]\right|}{\dfrac{1}{4}+\left[\dfrac{\Omega^2/4\beta^2}{\Omega^2/2\beta^2+1}\right]\left[1-e^{-3\beta t/2}\left[\cos\Omega' t+\dfrac{3\beta}{2\Omega'}\sin\Omega' t\right]\right]} . \tag{17}$$

Figure 2 shows a plot of visibility versus time $t$ given by this equation for several different values of the ratio $\Omega/\beta$.

## IV. DISCUSSION

Let us examine the curve for $\Omega/\beta=20$ more closely. It will be seen that the visibility $\mathcal{V}$ of the interference pattern vanishes close to certain times $t$ satisfying

$$\Omega t=n\pi \quad \text{or} \quad \beta t=n\pi/20 \quad (n=0,1,2,\ldots) . \tag{18}$$

There are just the times given by cases (b) and (c) above, when the atom is either most highly excited and the mean fluorescent light intensity is greatest, or it is deexcited

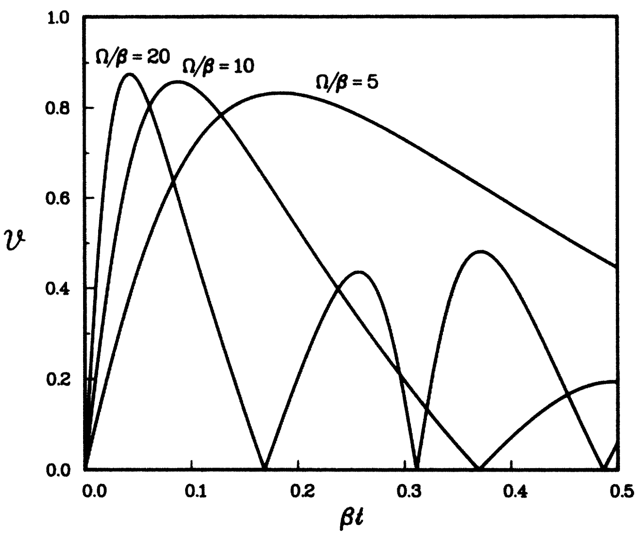


FIG. 2. Variation of the fringe visibility $\mathcal{V}$ with time for several different values of $\Omega/\beta$.

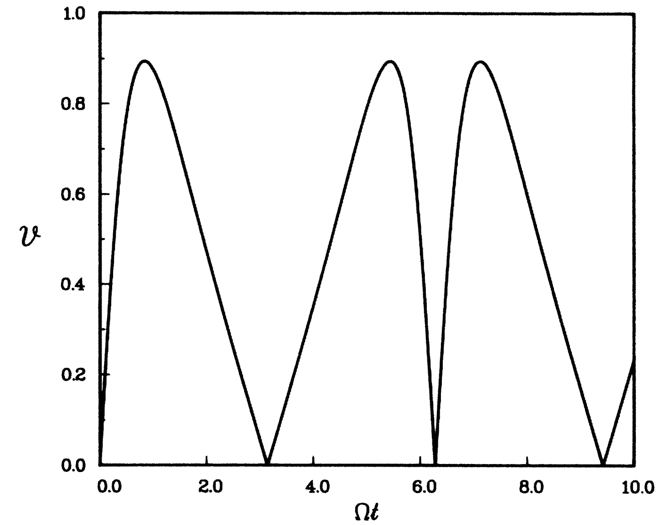


FIG. 3. The time variation of the visibility $\mathcal{V}$ in the limit $\Omega/\beta\rightarrow\infty$.

and the intensity is least. The visibility is greatest in between the zeros, but not half-way between. The reason is that the mean intensity in the denominator in Eq. (17) also exerts an influence on $\mathcal{V}$, and introduces an asymmetry, because it peaks at different times. Even in the limit $\Omega/\beta\rightarrow\infty$, when Eq. (17) reduces to

$$\mathcal{V}\rightarrow\frac{\sin\Omega t}{\frac{3}{2}-\cos\Omega t}\,, \tag{19}$$

this distortion is apparent in the plot given in Fig. 3. The visibility is greatest when $\Omega t=\arccos\frac{2}{3}\approx 0.84$ and reaches the maximum value $2/\sqrt{5}\approx 0.89$.

Although the behavior of this system is easiest to understand when $\Omega/\beta\gg 1$, it is evident from Fig. 2 that there are zeros in the visibility even when $\Omega/\beta$ is less than 10. Their positions are however displaced relative to those given by (b) and (c) above, for the reasons already mentioned. With this understanding the fundamental conclusion that second-order interference effects between photons and a classical field require an indefinite number of photons is borne out by the calculation.

There remains the question how this phenomenon might be exhibited experimentally. When the atoms in a weak atomic beam are exposed to a coherent laser beam close to an atomic resonance, they undergo Rabi oscillations and emit photons. Moreover, for atoms traveling with a certain velocity, position within the laser beam corresponds to the exposure time $t$. If a small portion of the coherent exciting field is diverted with the aid of a beam splitter and allowed to interfere with the fluorescent photons emitted by the atom, we have just the situation treated in Sec. III. The visibility of the interference pattern should vary with the position of the atom within the pumping beam according to Eq. (17). The nonclassical interference phenomenon we have been discussing is therefore not only of interest in principle, but it should be observable in practice.

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