Phase-matched emission from an optically thin medium following one-photon pulse excitation: 
Energy considerations

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Scully and coworkers [M. O. Scully, E. S. Fry, C. H. R. Ooi, and K. Wódkiewicz, Phys. Rev. Lett. 96, 010501 (2006)] demonstrated that there is directional, phase-matched emission following the excitation of an ensemble of atoms by a single-photon pulse. While the phase-matched emission intensity is proportional to the number of atoms, for optically thin samples the total energy emitted in the phase-matched direction is much less than that radiated in other directions. Moreover, even for optically thin samples, it is necessary to take into account effects related to cooperative decay if energy is to be conserved in the overall emission process. An analytic calculation is presented to show explicitly how cooperative decay reduces the incoherent emission and restores energy conservation in this low-density limit.

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In an interesting paper, Scully and coworkers [1] showed that there would be directional, phase-matched emission following the collective absorption of a one-photon pulse by an ensemble of two-level atoms, despite the fact that the atoms never acquire a dipole moment. Such emission can be viewed as a form of quantum super-radiance since there is no classical analog of a one-photon pulse. Directional emission of a similar nature from a single collective state created using a dipole blockade was discussed earlier by Saffman and Walker [2]. The paper of Scully et al. prompted a number of additional papers in which the directionality properties and nature of the emission for both product and entangled states of atoms were examined [3]. Most of these papers were concerned with optically thick samples, where the cooperativity factor associated with the emission process is greater than unity. In this high-density limit, the cooperative effects generally require a numerical solution of the equations of motion. The purpose of this Brief Report is to present an analytic calculation that is applicable to optically thin samples, in which the energy absorbed by the sample, on average, is considerably less than the energy in the incident pulse. In this limit, the energy radiated in the phase-matched direction, although proportional to the number of atoms in the sample, is always much less than the (single photon) energy radiated in other directions [1–3]. In fact this is a general result for any phased-matched emission in the low-density limit, such as that encountered in optical coherent transient experiments using classical fields [4].

It is assumed in Ref. [1] that a post-selection procedure can be used to project the medium into a fully symmetric, single collective excitation (FSSCE) of the atoms. Such a fully symmetric state is consistent with the assumption of an optically thin medium. In an optically thick sample, the field pulse creates an entangled state with absorptive weighting factors for different atoms [5], unless some “trick,” such as a dipole blockade [2] or an auxiliary ac Stark field [6] is used. The dipole blockade prevents multiple excitations in the sample owing to Rydberg atom interactions [2], while the ac Stark field shifts the transition frequency to render the medium optically thin during the excitation process [6]. Our discussion is limited to the low-density limit.

Even in the low-density limit, there is an interesting phenomenon that occurs related to energy conservation. In optical coherent transients, for example, it is possible to show that the energy radiated in all but the phase-matched direction is equal to the energy originally stored in the sample following an excitation pulse. The same is true for the one-photon pulse problem considered in this Brief Report. As a consequence, energy is not conserved since the total radiated energy, including the phase-matched emission, is greater than the original energy. This apparent paradox is resolved only when cooperative decay (i.e., the decay rate of each atom is modified by the other atoms in the sample) is taken into account. We include the effects of cooperative decay to first order in the density to show explicitly how it reduces the energy in all but the phase-matched direction by precisely the correct amount to restore energy conservation.

The underlying features of the emission process can be understood if we consider the sample to be composed of \( N \) stationary atoms placed at random positions in a volume \( V \). We assume that the average atom spacing is much greater than a wavelength, \( \bar{N} \lambda^3 \ll 1 \), where \( \bar{N} \) is the atomic density and \( \lambda \) is the transition wavelength. The atoms are excited by a \( \sigma_+ \) polarized, single-photon pulse directed along the z axis. This pulsed optical field imparts a spatial phase to each atom during the excitation process. Each atom is modeled as a two-level quantum system having lower state \( |g \rangle \), excited state \( |e \rangle \), and transition frequency \( \omega_0 \). The state \( |g \rangle \) corresponds to a ground state having angular momentum \( J = 0 \) and the state \( |e \rangle \) to an excited state having angular momentum \( J = 1 \) and magnetic quantum number \( m_J = 1 \) that is excited by \( \sigma_+ \)-polarized radiation. The incident pulse is centered at the transition frequency, implying that the average energy in the pulse is equal to \( \hbar \omega_0 \).
The initial state vector is taken to be the FSSCE

$$|\psi(0)| = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |j\rangle e^{i\epsilon_k r_j},$$  \(1\)

where \(r_j\) is the position of atom \(j\) and \(|j\rangle\) is the ket that corresponds to atom \(j\) in its \(J = 1, m_j = 1,\) excited state and all the other atoms in their ground states. At any later time, the state vector is given by

$$|\psi(t)| = \sum_{k,\epsilon_k} b_{k,\epsilon_k}(t)e^{-i\omega t}|k,\epsilon_k\rangle + \sum_{j=1}^{N} b_j(t)e^{-i\omega t}|j\rangle,$$  \(2\)

where the ket \(|k,\epsilon_k\rangle\) corresponds to the state in which all atoms are in their ground states and a photon having propagation vector \(k\), polarization \(\epsilon_k\), and frequency \(\omega_k = kc\) is emitted.

Neglecting any cooperative decay, each state amplitude \(b_j(t)\) decays simply as

$$b_j(t) = e^{-\gamma_e t/2}b_j(0) = \frac{1}{\sqrt{N}} e^{-\gamma_e t/2}e^{i\epsilon_k r_j},$$  \(3\)

where \(\gamma_e\) is the excited decay rate of an isolated atom. With this assumption, it is a simple matter to calculate the angular distribution of the emitted radiation using standard theories of spontaneous emission. By calculating \(|b_{k,\epsilon_k}(t)|^2\), summing over the polarizations \(\epsilon_k\), converting the sum to an integral over a continuum of emission modes, and integrating over frequency using the Weisskopf-Wigner approximation, one arrives at the angular distribution of the emitted radiation given by \([1, 7]\)

$$\frac{dI}{d\Omega} = \hbar\omega_0 \left[ \frac{3}{16\pi} (1 + \cos^2 \theta_k) \right] H(\theta_k, \phi_k),$$  \(4\)

where the structure factor \(H(\theta_k, \phi_k)\) is given by

$$H(\theta_k, \phi_k) = \frac{1}{N} \left| \sum_{j=1}^{N} \exp[-i(\mathbf{k} - \mathbf{k}_0) \cdot \mathbf{r}_j] \right|^2$$  \(5\)

$$= 1 + \frac{1}{N} \sum_{j, j' \neq j}^{N} \left| \exp[-i(\mathbf{k} - \mathbf{k}_0) \cdot \mathbf{r}_{jj'}] \right|^2,$$  \(6\)

\(\mathbf{k} = \mathbf{k}_0 \hat{\mathbf{k}}, \ \hat{\mathbf{k}}\) is a unit vector in the \(\mathbf{k}\) direction, \(\mathbf{r}_{jj'} = \mathbf{r}_j - \mathbf{r}_{j'}\), and \(\mathbf{k}_0 = \omega_0/c\). Implicit in these equations is the assumption that \(\gamma_e L/c \ll 1\), allowing us to replace \(\exp(-i \mathbf{k} \cdot \mathbf{r}_j)\) by \(\exp(-i\mathbf{k}_0 \hat{\mathbf{k}} \cdot \mathbf{r}_j)\) in carrying out the integration over frequency. The factor \((1 + \cos^2 \theta_k)/2\) in Eq. (4) corresponds to the radiation pattern emitted by an atom on a \(J = 1, m_j = 1\) to \(J = 0\) transition. The first term in Eq. (6) corresponds to incoherent radiation emitted in all directions, while the second term is associated with the directional, phase-matched emission.

It is interesting to note that we would arrive at similar equations had we considered free polarization decay from a medium prepared by a \textit{classical} pulse. The only difference is that the factor \(1/N\) would be replaced by the probability that each atom is excited by the pulse. Thus, the following discussion is valid equally for coherent transient emission as for emission from a collective atomic state excited by as single-photon pulse. In both cases it is the spatial phase imparted to the atoms by the field that leads to phase-matched emission, not the presence or absence of a nonvanishing dipole moment of the atoms.

As long as \(N\lambda^2/\zeta_0 \gg 1\), spatial fluctuations can be neglected and the double sum in Eq. (6) can be replaced by the double integral

$$\sum_{j, j' \neq j}^{N} e^{-i(\mathbf{k} - \mathbf{k}_0) \cdot \mathbf{r}_{jj'}} \rightarrow N^2 \int d\mathbf{r} \int d\mathbf{r}' e^{-i(\mathbf{k} - \mathbf{k}_0) \cdot (\mathbf{r} - \mathbf{r}')}.$$  \(7\)

\textbf{Emission pattern and radiated energy.} The emission pattern depends on the geometry of the volume. For a cylinder having radius \(a\) and length \(L\), it is a straightforward matter to use Eqs. (6) and (7) to calculate \([7]\)

$$H(\theta_k, \phi_k) = 1 + 16\pi a^2 L \left\{ \sin[k_0 L (1 - \cos \theta_k)/2]/k_0 L (1 - \cos \theta_k) \right\}^2$$

$$\times \left[ J_1(k_0 a \sin \theta_k)/k_0 a \sin \theta_k \right]^2,$$  \(8\)

where \(J_1\) is a Bessel function of order one. Consistent with the assumption that \(N\lambda^3 \ll 1\), we assume that \(k_0 a\) and \(k_0 L\) are both much greater than unity. Moreover, in the large Fresnel limit,

$$a^2 \gg \frac{\lambda L}{\pi},$$  \(9\)

it follows from Eqs. (4), (8), and (9) that, for \(k_0 a \gg 1\), the radiation pattern is given approximately by

$$\frac{dI}{d\Omega_k} = \hbar\omega_0 \left[ \frac{3}{16\pi} (1 + \cos^2 \theta_k) \right]$$

$$\times \left[ 1 + 4N a^2 L \pi \left( J_1(k_0 a \theta_k)/k_0 a \theta_k \right)^2 \right].$$  \(10\)

The angular divergence (full-width at half-maximum) of the phase-matched signal is of order \(\Delta \theta \approx 3.23/k_0 a\). The phase-matched emission is proportional to the atomic density for this one-photon excitation pulse. In coherent transients using classical fields, the incoherent term is linear in the density and the phase-matched term is quadratic in the density.

The total radiated energy is \([7]\)

$$I = \int d\Omega_k \frac{dI}{d\Omega_k} \sim \hbar\omega_0 \left[ \frac{3}{16\pi} (1 + \cos^2 \theta_k) \right]$$

$$\times \left[ 1 + 4N a^2 L \pi \left( J_1(k_0 a \theta_k)/k_0 a \theta_k \right)^2 \right].$$  \(11\)

assuming that \(k_0 a \gg 1\). The first term in Eq. (11) corresponds to the incoherent emission into \(4\pi\) and the second to the phase-matched emission. Thus the radiated energy is equal to the \textit{original} energy, \(\hbar\omega_0\), stored in the atoms, \textit{plus} the term from the phase-matched emission. Clearly if the theory is to have any validity at all, the second term in Eq. (11) must be much less than the first. Since the requirement that \(3N\lambda^2 L/8\pi \ll 1\) coincides with that to have an optically thin sample, it follows that, for optically thin samples, the \textit{energy radiated in the phase matched direction is always much less than the energy associated with the incoherent radiation.} Of course energy must be conserved \textit{exactly}, implying that there must be some additional contribution to Eq. (11), not yet considered, that exactly cancels the second term in Eq. (11). We return to this directly.

\textbf{Inclusion of cooperative decay.} Equation (11) implies that energy is not conserved in this calculation to order \(N\lambda^2 L\) for our one-photon pulse. To restore energy conservation, it is necessary to include cooperative decay effects. In the low-density limit, the cooperative decay effects \textit{increase}
the decay rate of each atom. The net effect is a reduction of the energy radiated in all but the phase-matched direction that compensates for the increased energy that is radiated in the phase-matched direction. It is possible to carry out a simple calculation to show this explicitly for an arbitrary volume of atoms, provided that the separation of each pair of atoms is much larger than a wavelength, \( \xi_{jl} = k_0 |r_{jl}| \gg 1 \).

The inclusion of cooperative decay leads to significant complications. The \( J = 1, m_J = 1 \) atomic state of atom \( j \) that is excited by the incident field is coupled to all the excited state magnetic sublevels of the other atoms. The radiated signal then results from contributions from all excited state magnetic sublevels. However in optically thin media, we will be able to neglect the contributions from all but the \( J = 1, m_J = 1 \) excited states, as you shall see.

To include the effects of cooperative decay to lowest order, the modification of the decay of \( b_j(t) \) must be taken into account. The appropriate equations are \[ b_{jm}(t) = -\gamma_e b_{jm}(t) - \sum_{\ell=1,\ell\neq j}^{N} \sum_{m=-1}^{1} \gamma_e G_{mm}^{1}(k_0 r_{jl})b_{\ell m'}(t), \]

where the \( G_{mm}^{1}(k_0 r_{jl}) \) are given in Ref. [8] and \( b_{jm} \) is the state amplitude for atom \( j \) to be in magnetic sublevel \( m \) of its excited state, and all other atoms to be in their ground states.

In optically thin media, Eq. (13) can be solved using perturbation theory. With initial condition \( b_{jm}(0) = e^{i k_0 r_j (\delta_{m,1} - C_{jm})} \), the solution is

\[ b_{jm}(t) \approx \frac{1}{\sqrt{N}} e^{-\gamma_e t} e^{i k_0 r_j (\delta_{m,1} - C_{jm})}, \]

where

\[ C_{jm} = \sum_{\ell=1,\ell\neq j}^{N} G_{mm}^{1}(k_0 r_{jl})e^{-i k_0 r_{jl}}, \]

is referred to as the cooperativity parameter. For the perturbation theory approach to be valid, we must require that \( |C_{jm}| \ll 1 \), which is satisfied if condition (12) holds and if the medium is optically thin. Since the radiated signal intensity depends on the square of the excited state amplitude, only the state amplitude \( b_{j,m=1} \) contributes, if terms of order \( |C_{jm}|^2 \) are neglected. In other words, we need only be concerned with

\[ b_j(t) \approx \frac{1}{\sqrt{N}} e^{-\gamma_e t} e^{i k_0 r_j [1 - C_{j}]} \approx \frac{1}{\sqrt{N}} e^{-\gamma_e t (1+C)} e^{i k_0 r_j}, \]

where \( b_j \equiv b_{j,m=1}, C \equiv C_{j,m=1} \), and [8]

\[ G_{11}(\xi) = \sqrt{4\pi} h_1(\xi) Y_{00}(\theta_e, \phi_e) - \frac{1}{2} \sqrt{4\pi \frac{5}{3}} h_2(\xi) Y_{20}(\theta_e, \phi_e), \]

where \( h_\ell \) is a spherical Hankel function of the first type and \( Y_{lm} \) is a spherical harmonic. Cooperative decay results in a change in the decay rate and a shift in the transition frequency of each decay mode.

To obtain the radiation pattern, we follow the same procedure used to arrive at Eq. (4). In integrating over time to obtain the spectrum, we encounter integrals of the form

\[ \int_0^\infty dt [A + Bt] e^{i(\omega_0 - \omega_0) - \gamma_e t} = \frac{A}{\gamma_e - i(\omega_0 - \omega_0)} + \frac{B}{[\gamma_e - i(\omega_0 - \omega_0)]^2}, \]

where

\[ A = \sum_{j=1}^{N} e^{-i(k-k_0)r_j}, \]

\[ B = -\gamma_e \sum_{j,j=1,j\neq j}^{N} e^{-i(k-k_0)r_j} G_{11}(k_0 r_{jl}) e^{-i k_0 r_{jl}}. \]

In turn, the absolute square of Eq. (18) must be integrated over frequency to arrive at the radiated intensity; that is, we need

\[ \int_{-\infty}^{\infty} d\omega_k \left| \frac{A}{\gamma_e - i(\omega_0 - \omega_0)} + \frac{B}{[\gamma_e - i(\omega_0 - \omega_0)]^2} \right|^2 = \frac{\pi}{\gamma_e} [|A|^2 + \text{Re}(AB^*/\gamma_e) + |B|^2/2\gamma_e]. \]

As a consequence, Eq. (4) must be replaced by

\[ \frac{dI}{d\Omega_k} = \hbar \omega_0 \frac{3}{16\pi} (1 + \cos^2 \theta) H_j(\theta_k, \phi_k), \]

where, to order \( C \),

\[ H_j(\theta_k, \phi_k) = \frac{1}{N} \left[ \sum_{j,j'=1}^{N} e^{-i(k-k_0)r_{jl}} - \text{Re} \sum_{j,j'=1}^{N} e^{-i(k-k_0)r_{jl}} G_{11}(k_0 r_{jl}) e^{-i k_0 r_{jl}} \right]. \]

To get the radiated power, we must integrate Eq. (21) over angles. Integration of the second summation in Eq. (22) over angles leads to an additional factor of \( \xi_{jl}^{-1} \ll 1 \) if \( j \neq j' \), implying that we need retain only the terms with \( j = j' \) in the last term in Eq. (22). As a consequence, we find

\[ I = \int \frac{dI}{d\Omega_k} d\Omega_k \approx \hbar \omega_0 (1 + F_1 + F_2), \]

where

\[ F_1 = \frac{3}{16N\pi} \sum_{j,j'=1}^{N} e^{i k_0 r_{jl}} \int d\Omega_k e^{-ik r_{jl}} (1 + \cos^2 \theta_k) \]

and

\[ F_2 = -\frac{1}{N} \sum_{j,j'=1}^{N} e^{i k_0 r_{jl}} \text{Re} G_{11}(k_0 r_{jl}). \]

The \( F_1 \) terms arise from the coherent, phase matched emission in the forward direction, but there is now an additional term, the \( F_2 \) term, which arises from cooperative
As a consequence, the second term in Eq. (23) exactly cancels the third term and
\[ I = \hbar \omega_0. \] (29)

Inclusion of cooperative decay has restored energy conservation. For a fixed arrangement of atoms satisfying condition (12), it is possible that \( F_1 \) can be negative since the atoms can be so placed as to have destructive interference even for the (spatially integrated) phase-matched emission. However, on averaging over a uniform density distribution by converting the sum to a double integral as in Eq. (7), the phase-matched emission energy \( F_1 \) is always positive, as we have seen for the cylindrical geometry. Thus, even though Eq. (25) was derived using condition (12), its range of validity appears to be wider, valid for atomic separations less than a wavelength. Thus the sum can be converted to a double integral and yields a negative value for \( F_2 \). The entire treatment, however, is valid only for optically thin media. An analogous calculation for emission from a phased array line of classical dipole oscillators has appeared recently [9].

In summary, while there is phase-matched emission following the excitation of an atomic ensemble by a single photon radiation pulse, most of the energy is radiated in directions other than the phase-matched direction in optically thin samples. As a consequence, the phase-matched emission does not constitute a replica of the one-photon excitation pulse. It appears that it is necessary to use an optically thick medium to achieve this result [1–3,10]. On average, cooperative decay results in an increased decay rate for the atoms and a corresponding decrease in the energy emitted in all but the phase-matched direction that exactly compensates the phase-matched super-radiant emission. The results of this Brief Report also characterize the signals radiated in optical coherent transient experiments.

[10] See, for example, S. A. Moiseev and S. Kröll, Phys. Rev. Lett. 87, 173601 (2001); J.-L. Le Gouët and P. R. Berman, Phys. Rev. A 80, 012320 (2009), and references therein. In the protocol discussed in these papers, an inhomogeneously broadened medium is used and the single-photon pulse is not stored in a symmetric, phased state. Nevertheless, by using an echo-like sequence, a faithful replica of the input pulse can be generated. The pulse emerges from the sample in a direction opposite to that of the input pulse.