Directed Spontaneous Emission from an Extended Ensemble of N Atoms: Timing Is Everything

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A collection of N static atoms is fixed in a crystal at a low temperature and prepared by a pulse of incident radiation of wave vector $\vec{k}_0$. The N atoms are well described by an entangled Dicke-like state, in which each atom carries a characteristic phase factor $\exp(i\vec{k}_0 \cdot \vec{r}_j)$, where $\vec{r}_j$ is the atomic position in the crystal. It is shown that a single photon absorbed by the N atoms will be followed by spontaneous emission in the same direction. Furthermore, phase matched emission is found when one photon is absorbed by N atoms followed by two-photon down-conversion.

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The central issue of this Letter, summarized in Fig. 1, is to suppose we prepare an ensemble of (two-level) atoms by absorbing one photon of wave vector $\vec{k}_0$; see Fig. 2. There is no dipole moment (i.e., no coherent superposition of levels) induced in the atoms. That is, we prepare the state in which one atom is totally excited, but we do not know which one. The absorbed photon will then be spontaneously emitted. We ask: Will the emitted photon go in 4π sr, or will it be directionally correlated with $\vec{k}_0$? The perhaps counterintuitive answer is the latter.

Directional radiation from a collection of coherently excited atoms is a well-studied problem [1]. Experiments such as photon echo [2], self-induced transparency [3], optical solitons [4], lasing without inversion [5], ultraslow light [6], and production of entangled light from phase coherent atoms [7] are examples of such physics. In his pioneering paper on superradiance, Dicke [8] pointed out that it is possible to have a radiating gas "such that spontaneous radiation occurs coherently in one direction."

Dicke states that a semiclassical treatment is generally adequate for the spontaneous radiation from superradiant states. He explains: "To calculate the radiation rate, the expectation value of the electric dipole moment is treated as a classical dipole. When the gas contains a large number of molecules, the dipole moment of the gas as a whole should be given by the sum of the expectation values of the individual dipole moments." That is, when a short pulse excites the atoms and passes on, the individual atoms radiate essentially as if they were a coherent superposition of classical dipole oscillators.

Let us begin by considering how to prepare an N-atom state in the crystal by the absorption of one photon. The Hamiltonian in the interaction picture is

$$V(t) = \sum_j \hbar g_0 \hat{\sigma}_j^\dagger \hat{a}_{\vec{k}_0} e^{i\vec{k}_0 \cdot \vec{r}_j} e^{-i(\nu_0 - \omega)t} + \text{adj.}, \qquad (1)$$

where $\hat{\sigma}_j^\dagger = |a_j\rangle\langle b_j|$ is the atomic transition operator, $\hat{a}_{\vec{k}_0}$ is the photon annihilation operator, ν_0 is the angular frequency of the incident radiation, ω is the angular frequency separation of the two atomic levels a and b, and g_0 is the atom-field coupling frequency. Then the unitary time evolution of the system is given by

$$\begin{aligned} U(\tau) &= \mathcal{T} \exp\left(-\frac{i}{\hbar} \int_0^\tau dt' V(t') \right) \\ &\simeq 1 - i g_0 \left(\int_0^\tau dt' e^{i(\nu_0 - \omega)t} \sum_j \hat{\sigma}_j^\dagger \hat{a}_{\vec{k}_0} e^{i\vec{k}_0 \cdot \vec{r}_j} + \text{adj.} \right), \end{aligned} \qquad (2)$$

where $\mathcal{T}$ is the time ordering operator, and we have assumed *weak* coupling between the atom and the field. Beginning with the state $|b_1, b_2, b_3 \ldots b_N\rangle \otimes |1_{\vec{k}_0}\rangle$ and assuming that the incident radiation has been chosen to be resonant with the atom, i.e., $\nu_0 = \omega$, we then have

$$\begin{aligned} &U(\tau)|b_1, b_2, b_3, \ldots, b_N\rangle \otimes |1_{\vec{k}_0}\rangle \\ &\quad \simeq |b_1, b_2, b_3, \ldots, b_N\rangle \otimes |1_{\vec{k}_0}\rangle \\ &\quad - i g_0 \tau \sum_j e^{i\vec{k}_0 \cdot \vec{r}_j} |b_1, b_2, \ldots, a_j, \ldots, b_N\rangle \otimes |0\rangle. \end{aligned} \qquad (3)$$

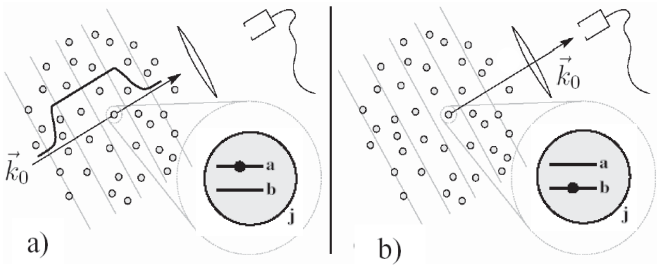

FIG. 1. N atoms are driven by a pulse having wave vector $\vec{k}_0$, and the transmitted radiation is monitored. (a) The incident pulse excites the atom. (b) The atom spontaneously emits predominantly in the direction of $\vec{k}_0$.

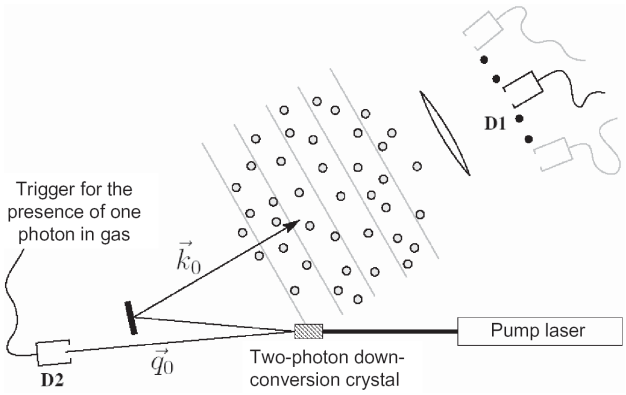

FIG. 2. Proposed setup for preparation of a conditional state and detection of a directed spontaneous emission photon from an ensemble of atoms prepared in the excited state by a pulse of wave vector $\vec{k}_0$.

We choose $g_0\tau$ to be small so that, most of the time, a $\vec{k}_0$ photon will pass through the atoms and register a count in the (perfect) detector as in Fig. 1(a). Thus, if we fail to detect a photocount [Fig. 1(b)] (knowing a photon is incident), we know that we have prepared the atom in the *entangled* state given by the second term in Eq. (3). Thus, we define the N-atom normalized conditional state vector times the vacuum state as

$$|\Psi(N|1_{\vec{k}_0})\rangle = \frac{1}{\sqrt{N}}\sum_j e^{i\vec{k}_0\cdot\vec{r}_j}|b_1, b_2, \ldots, a_j, \ldots, b_N\rangle|0\rangle, \quad (4)$$

where one atom is excited by a photon of wave vector $\vec{k}_0$.

In order to prepare the conditional state $|\Psi(N|1_{\vec{k}_0})\rangle$, we propose to use a correlated photon pair as in Fig. 2. A click in detector D2 tells us that a $\vec{k}_0$ photon is sent into the atomic array. It will usually pass through the atoms and be detected in D1. However, when D2 clicks and D1 does not, we know the state $|\Psi(N|1_{\vec{k}_0})\rangle$ has been prepared (assuming perfect detectors, etc.).

Next, we consider the spontaneous decay of the state, Eq. (4). We write the interaction Hamiltonian $W(t)$ and the matrix $U_W(t)$ for spontaneous emission,

$$W(t) = \sum_j\sum_{\vec{k}} \hbar g_{\vec{k}}\hat{a}^\dagger_{\vec{k}}\hat{\sigma}_j e^{-i\vec{k}\cdot\vec{r}_j}e^{i(\nu_{\vec{k}}-\omega)t} + \text{adj.} \quad (5)$$

and

$$U_W(t) = \mathcal{T}\exp\left(-\frac{i}{\hbar}\int_0^t dt' W(t')\right) = \prod_j U_W^{(j)}, \quad (6)$$

where $U_W^{(j)} = \hat{\gamma}_j^\dagger\hat{\sigma}_j$ is the matrix for the jth atom (in the limit $t\to\infty$) with the radiation operator

$$\hat{\gamma}_j^\dagger = \sum_{\vec{k}} \frac{g_{\vec{k}}e^{-i\vec{k}\cdot\vec{r}_j}}{(\nu_{\vec{k}}-\omega)+\frac{i}{2}\gamma}\hat{a}^\dagger_{\vec{k}}. \quad (7)$$

In Eq. (7) the Weisskopf-Wigner spontaneous emission rate is $\gamma = 2\pi\sum_{\vec{k}}|g_{\vec{k}}|^2\delta(\nu_{\vec{k}}-\omega) = 2\pi|g(\omega)|^2\mathcal{D}(\omega)$, where we introduce the density of states in free space $\mathcal{D}(\omega) = \frac{8\pi}{3}[\mathcal{V}\omega^2/(2\pi c)^3]$, with $\mathcal{V}$ as the quantization volume of the radiation field, and we have used $\sum_{\vec{k}}\to\int_0^\infty \mathcal{D}(\nu_{\vec{k}})d\nu_{\vec{k}}$. Then $U_W^{(j)}|a_j\rangle = \hat{\gamma}_j^\dagger|b_j\rangle$ and

$$\begin{aligned}\langle b_1,\ldots,b_N|U_W|\Psi(N|1_{\vec{k}_0})\rangle &= \frac{1}{\sqrt{N}}\sum_j e^{i\vec{k}_0\cdot\vec{r}_j}\hat{\gamma}_j^\dagger|0\rangle = \frac{1}{\sqrt{N}}\sum_{\vec{k}}\frac{g_{\vec{k}}}{(\nu_{\vec{k}}-\omega)+\frac{i}{2}\gamma}|1_{\vec{k}}\rangle\sum_j e^{i(\vec{k}_0-\vec{k})\cdot\vec{r}_j}\\ &\simeq \frac{\sqrt{N}}{V}\sum_{\vec{k}}\frac{(2\pi)^3 g_{\vec{k}}}{(\nu_{\vec{k}}-\omega)+\frac{i}{2}\gamma}|1_{\vec{k}}\rangle\delta^3(\vec{k}_0-\vec{k}),\end{aligned} \quad (8)$$

where we have used the atomic summation (for large number density as in a crystal)

$$\sum_j e^{i(\vec{k}_0-\vec{k})\cdot\vec{r}_j} = \frac{N}{V}\int_V d^3r e^{i(\vec{k}_0-\vec{k})\cdot\vec{r}} = \frac{N}{V}(2\pi)^3\delta^3(\vec{k}_0-\vec{k}), \quad (9)$$

with V as the volume of the gas. Thus, the emitted photon is directed along the exciting photon. This is another interesting consequence of many particle entanglement in a dense medium. Next, we consider the case of two-photon down-conversion in which the atoms are undergoing two successive spontaneous emissions: as in the case of a cascade scheme [Fig. 3(a)]. Here the two-photon operator for the jth atom is (see Ref. [9], p. 212)

$$\hat{\Gamma}_j^\dagger = \sum_{\vec{k},\vec{q}}\frac{g_{a,\vec{k}}g_{b\vec{q}}e^{-i(\vec{k}+\vec{q})\cdot\vec{r}_j}\hat{a}^\dagger_{\vec{k}}\hat{a}^\dagger_{\vec{q}}}{(\nu_{\vec{k}}+\nu_{\vec{q}}-\omega_{ac}+\frac{i}{2}\gamma_a)(\nu_{\vec{q}}-\omega_{bc}+\frac{i}{2}\gamma_b)}, \quad (10)$$

where γ_a and γ_b are the decay rates of level a and b, respectively. The two-photon state for the transition from state $|\Psi(N|1_{\vec{k}_0})\rangle$ [Eq. (4)] to the ground state also shows directionality

$$\begin{aligned}&\langle c_1,\ldots,c_N|\left(\sum_j\hat{\Gamma}_j^\dagger|c_j\rangle\langle a_j|\right)|\Psi(N|1_{\vec{k}_0})\rangle\\ &= \frac{1}{\sqrt{N}}\sum_j\sum_{\vec{k},\vec{q}}\frac{g_{a,\vec{k}}g_{b\vec{q}}e^{-i(\vec{k}+\vec{q}-\vec{k}_0)\cdot\vec{r}_j}|1_{\vec{k}},1_{\vec{q}}\rangle}{(\nu_{\vec{k}}+\nu_{\vec{q}}-\omega_{ac}+\frac{i}{2}\gamma_a)(\nu_{\vec{q}}-\omega_{bc}+\frac{i}{2}\gamma_b)}\\ &\simeq \frac{\sqrt{N}}{V}\sum_{\vec{k},\vec{q}}\frac{g_{a,\vec{k}}g_{b\vec{q}}(2\pi)^3\delta^3(\vec{k}+\vec{q}-\vec{k}_0)|1_{\vec{k}},1_{\vec{q}}\rangle}{(\nu_{\vec{k}}+\nu_{\vec{q}}-\omega_{ac}+\frac{i}{2}\gamma_a)(\nu_{\vec{q}}-\omega_{bc}+\frac{i}{2}\gamma_b)}.\end{aligned} \quad (11)$$

Thus, the directions of the emitted photons $\vec{k}$ and $\vec{q}$ are automatically phase matched relative to $\vec{k}_0$.

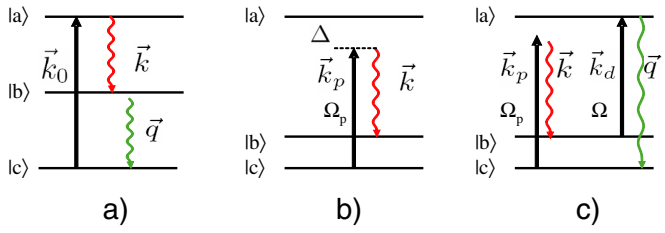

FIG. 3 (color online). Level schemes for: (a) cascade, (b) spontaneous Raman, and (c) double-Raman-quantum eraser schemes. In the cascade scheme (a), the levels a and c need to be coupled by a laser, so the transition should be dipole allowed; for example, level a is a superposition of "s" and "p" states.

In the following, we provide further discussions on the directionality of the spontaneously emitted photons by making connections with some important physical processes.

Connection with gain swept superradiance.—Gain sweeping behind this single photon collective emission can be gleaned by considering an ensemble of atoms that are excited by a short pulse at various times $t_j = \vec{k}_0 \cdot \vec{r}_j / c|\vec{k}_0|$. This has much in common with the notion of gain swept superradiance [10]. Although the mechanism of excitation is identical to our present problem, the directionality refers to the many photon emissions of the gain medium instead of one photon emission. The one photon state from all atoms is

$$|\tilde{\Psi}_N\rangle = \frac{1}{\sqrt{N}} \sum_j \sum_{\vec{k}} \frac{g_{\vec{k}}}{(\nu_{\vec{k}} - \omega) + \frac{i}{2}\gamma} e^{-i\vec{k}\cdot\vec{r}_j} e^{i\nu_{\vec{k}} t_j} |1_{\vec{k}}\rangle. \quad (12)$$

The single photon Glauber's correlation function is

$$G(r, t) = \langle \tilde{\Psi}_N | \hat{E}^\dagger(\vec{r}, t) \hat{E}(\vec{r}, t) | \tilde{\Psi}_N \rangle = |\langle 0 | \hat{E}(\vec{r}, t) | \tilde{\Psi}_N \rangle|^2, \quad (13)$$

where $\vec{r}$ and t are the detector position and detection time, respectively. Substitution of Eq. (12) and $\hat{E}(\vec{r}, t) = \sum_{\vec{k}} \mathcal{E}_{\vec{k}} \hat{a}_{\vec{k}} e^{i(\vec{k}\cdot\vec{r} - \nu_{\vec{k}} t)}$ with $\mathcal{E}_{\vec{k}} = \sqrt{\hbar \nu_{\vec{k}} / 2\varepsilon_o \mathcal{V}}$ into Eq. (13), we find

$$\begin{aligned} \langle 0 | \hat{E}(\vec{r}, t) | \tilde{\Psi}_N \rangle &= \frac{1}{\sqrt{N}} \sum_{\vec{k}} \sum_j \frac{\mathcal{E}_{\vec{k}} g_{\vec{k}} e^{i\vec{k}\cdot(\vec{r} - \vec{r}_j)} e^{-i\nu_{\vec{k}}(t - t_j)}}{(\nu_{\vec{k}} - \omega) + \frac{i}{2}\gamma} \\ &= \frac{\sqrt{N}}{V} \sum_{\vec{k}} \frac{\mathcal{E}_{\vec{k}} g_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} e^{-i\nu_{\vec{k}} t}}{(\nu_{\vec{k}} - \omega) + \frac{i}{2}\gamma} (2\pi)^3 \delta^3(\vec{k}_0 - \vec{k}), \end{aligned} \quad (14)$$

where we have used

$$\exp(i\nu_{\vec{k}} t_j) = \exp(i\nu_{\vec{k}} \vec{k}_0 \cdot \vec{r}_j / c|\vec{k}_0|) \simeq \exp(i\vec{k}_0 \cdot \vec{r}_j), \quad (15)$$

and the last step of Eq. (15) follows since $\nu_{\vec{k}} \simeq c|\vec{k}_0|$. This shows that the timing of the atomic excitation is the key physical process behind the present directional spontaneous emission, i.e., "timing is everything."

Young's interference pattern between spontaneously emitted photons.—Another window into the problem is provided by the resonant scattering of one photon off a pair of two-level atoms. After the photon is absorbed by the two-level atoms, we have (the EPR entangled state)

$$|\Psi\rangle' = \frac{1}{\sqrt{2}} (e^{i\vec{k}_0\cdot\vec{r}_1} |a_1, b_2\rangle + e^{i\vec{k}_0\cdot\vec{r}_2} |b_1, a_2\rangle) |0\rangle, \quad (16)$$

which is the conditional state of Eq. (4) for two atoms with equal phase. After spontaneous emissions, this state evolves into

$$|\Psi\rangle = \frac{1}{\sqrt{2}} (e^{i\vec{k}_0\cdot\vec{r}_1} \hat{\gamma}_1^\dagger + e^{i\vec{k}_0\cdot\vec{r}_2} \hat{\gamma}_2^\dagger) |0\rangle |b_1, b_2\rangle, \quad (17)$$

which implies

$$\begin{aligned} G(r, t) &= |\langle 0 | \hat{E}(r, t) | \Psi \rangle|^2 \\ &= \frac{1}{2} |\langle 0 | \hat{E}(r, t) | \gamma_1 \rangle + \langle 0 | \hat{E}(r, t) | \gamma_2 \rangle|^2 \\ &= \frac{\mathcal{E}_o^2}{r^2} \{1 + \cos[(\vec{k}_0 - \vec{k}) \cdot (\vec{r}_1 - \vec{r}_2)]\}. \end{aligned} \quad (18)$$

Here we see the contrasting conditions for high directionality (when $|\vec{k}_0 - \vec{k}|$ is small) and for high Rayleigh resolution (when $|\vec{k}_0 - \vec{k}|$ is large).

The spontaneous Raman from N atoms of Fig. 3(b) has no preferred direction. This can be shown using the interaction Hamiltonian

$$W_{\mathrm{R}} = \sum_{j,\vec{k}} \hbar G_{\vec{k}} \hat{a}_{\vec{k}}^\dagger |b_j\rangle\langle c_j| e^{i(\vec{k}_p - \vec{k})\cdot\vec{r}_j} e^{-i(\Delta - \Delta_{\vec{k}})t} + \text{adj.}, \quad (19)$$

where $G_{\vec{k}} = \Omega_p g_{\vec{k}} / \Delta$, $\Delta = \nu_p - \omega_{ac}$, and $\Delta_{\vec{k}} = \nu_{\vec{k}} - \omega_{ab}$. The spontaneous Raman state for N atoms follows as

$$\begin{aligned} U_{\mathrm{R}} |\{c_j\}\rangle |0\rangle &\simeq \left\{1 - \frac{i}{\hbar} \int_0^\tau dt' W_{\mathrm{R}}(t')\right\} |\{c_j\}\rangle |0\rangle \\ &= \sum_{\vec{k}} \frac{G_{\vec{k}}}{\Delta_{\vec{k}} - \Delta + \frac{i}{2}\Gamma_R} |1_{\vec{k}}\rangle \sum_j e^{i(\vec{k}_p - \vec{k})\cdot\vec{r}_j} |b_j, \{c_{i\neq j}\}\rangle, \end{aligned} \quad (20)$$

where all atoms are initially in c and Γ_R is the Raman decay rate. If we take the final state as $|\Psi(1)\rangle = \frac{1}{\sqrt{N}} \sum_j |c_1, \ldots, b_j, \ldots, c_N\rangle$, the photon state becomes

$$\begin{aligned} \langle \Psi(1) | U_W | c_1, \ldots, c_N \rangle |0\rangle &= \frac{1}{\sqrt{N}} \sum_{\vec{k}} \frac{G_{\vec{k}}}{\nu_{\vec{k}} - \nu_p + \omega_{bc} + \frac{i}{2}\Gamma_R} \\ &\quad \times |1_{\vec{k}}\rangle \sum_j e^{i(\vec{k}_p - \vec{k})\cdot\vec{r}_j}. \end{aligned} \quad (21)$$

The Lorentzian in Eq. (21) fixes the magnitude of $|\vec{k}|$ to be $|\vec{k}_p| - \omega_{bc}/c$. Therefore, the $\delta(\vec{k}_p - \vec{k})$ implied from Eq. (21) is always zero. Hence, the intensity goes as

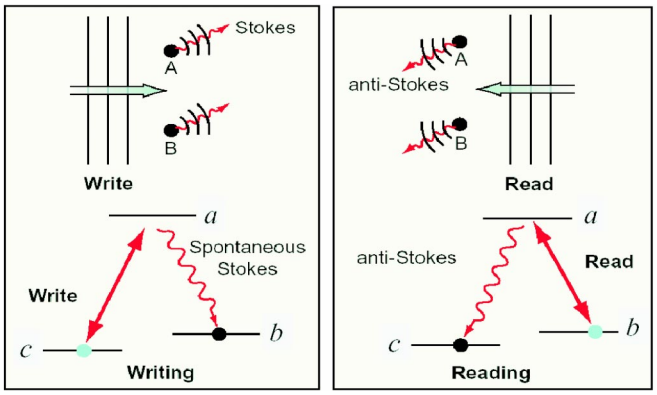

FIG. 4 (color online). The writing and reading processes of two atoms in a double-Raman scheme is actually a quantum erasure.

$|\sum_j e^{i(\vec{k}_p-\vec{k})\cdot\vec{r}_j}|^2 \simeq |\sum_j e^{i(\omega_{bc}/c)\hat{n}\cdot\vec{r}_j}|^2 = N$ since the sum of all the cross terms (of different atoms) is zero.

The Raman emission doublet (RED) from N atoms of Fig. 3(c) has the state vector obtained from a single atom case in Ref. [11] as

$$|\Psi_N\rangle_{\mathrm{RED}} = \frac{1}{\sqrt{N}}\sum_{j,\vec{k},\vec{q}} \frac{\Omega G_{\vec{k}} g_{\vec{q}} e^{i(\vec{k}_p+\vec{k}_d-\vec{k}-\vec{q})\cdot\vec{r}_j} L_{\vec{k},\vec{q}} |1_{\vec{k}}, 1_{\vec{q}}\rangle}{2\tilde{\Omega}(\nu_{\vec{q}} + \nu_{\vec{k}} - \nu_p - \nu_d + \frac{i}{2}\Gamma_R)}$$
$$= \frac{\sqrt{N}}{V}\sum_{\vec{k},\vec{q}} \frac{\Omega G_{\vec{k}} g_{\vec{q}} (2\pi)^3 \delta^3(\vec{k}+\vec{q}-\vec{k}_p-\vec{k}_d) L_{\vec{k},\vec{q}}}{2\tilde{\Omega}(\nu_{\vec{q}} + \nu_{\vec{k}} - \nu_p - \nu_d + \frac{i}{2}\Gamma_R)} \times |1_{\vec{k}}, 1_{\vec{q}}\rangle, \quad (22)$$

where $L_{\vec{k},\vec{q}} = (\nu_{\vec{q}} - \omega_{ac} + \frac{1}{4}i\gamma - \tilde{\Omega})^{-1} - (\nu_{\vec{q}} - \omega_{ac} + \frac{1}{4}i\gamma + \tilde{\Omega})^{-1}$ and $\tilde{\Omega} = \sqrt{\Omega^2 - (\gamma/4)^2}$. Here the directions of the Stokes ($\vec{k}$) and anti-Stokes ($\vec{q}$) photons are correlated, by the phase matching condition $\vec{k} + \vec{q} = \vec{k}_p + \vec{k}_d$.

Quantum eraser [12] is based on the absorption of one photon by two atoms. The atoms play the role of the two slits in Young's experiment. A photon is emitted as the atom makes the transition from c to b. There is no interference pattern, because there is now which path information for the atoms. Then an erasure pulse drives the atom to an excited state a and a second photon is emitted. The second order correlation function will now show interference. In other words, there is directionality in the correlation between the two-photon events as in Fig. 4. Thus, the two atom quantum eraser gives a simple physical insight into the directionality of the double-Raman process.

The experiments of the groups of Harris [13], *Lukin* [14], *and Kimble* [15] are an excellent example of the mapping of the quantum state of a photon onto an ensemble of gas atoms. The "writing" photon and the "reading" photon (Fig. 4) are similar in spirit to the Raman-quantum eraser scheme [16]. The directionality is a macroscopic property connected to the "quantum memory" stored in the atoms.

Correlated spontaneous emission is a well-studied process in laser physics and quantum optics [9]. There we are mostly concerned about temporally correlated spontaneous emissions (of two photons) from single atom.

In the present work, we are interested in the problem of spontaneous emission from many atoms. The directionality of the spontaneously emitted photon can be envisioned as "correlated spontaneous emission" process. Furthermore, a correlated ensemble of atoms acting as a two-photon parametric down-converter automatically leads to phase matching; see Eq. (11).

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