Photon Echo in Gaseous Media

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Optical echos in gaseous media are investigated in order to determine the conditions under which an echo would be expected. The results are similar to those for a solid, except that the time between exciting pulses is limited when the two pulses do not travel in the same direction. With increasing pulse separation collisions reduce the echo intensity in a way which is characteristic of the type of collision process.

ALTHOUGH the existence of a photon echo in an inhomogeneously broadened solid is well established experimentally,[1] no one has reported a photon echo in a Doppler-broadened gas. In this article we treat the problem theoretically[2] in order to determine under what conditions a photon echo would be expected to occur in a gaseous medium.

An ideal photon echo in a solid would result from a 90° pulse followed by a 180° pulse. The 90° pulse produces a macroscopic polarization which decays rapidly if the atomic line is inhomogeneously broadened. The 180° pulse effectively reverses the time so that the atoms will rephase at a later time equal to the pulse separation.

In the case of a gas, after the first pulse the induced macroscopic polarization decays because of the rapid atomic motion. After the second pulse the atomic velocities are not reversed and the atoms do not return to their initial position to reform a macroscopic polarization. When the effects of atomic motion are taken into account and the correct retarded times are used it is found that a macroscopic polarization does reform. The formation of an echo only depends on the atoms retaining a memory of their condition immediately following the first pulse. Collisions of the atoms lead to a reduction in the size and a change in the shape of the echo. Observation of these effects could provide a convenient method for studying collision processes in gases.

In order to produce a photon echo we pass two pulses through the medium, separated by a time τ and in the directions represented by the unit vectors $\mathbf{n}_1$ and $\mathbf{n}_2$, respectively. We look for an additional pulse, the echo, to emerge from the medium in a direction $\mathbf{n}$ at a time approximately 2τ after the first pulse. We take our medium to consist of two level atoms located at positions $\mathbf{r}_i(t)$ (measured from an origin inside the medium) with resonant frequencies $\omega_i = E_{a,i} - E_{b,i}$, where $E_{a,i}$ is the energy of the upper level a and $E_{b,i}$ is the energy of the ground state b of the ith atom. The electric field radiated by such a set of atoms is

$$\mathbf{E}(t,\mathbf{R}) = (c^2R)^{-1} \sum_i \omega_i^2 \mathbf{n} \times[\mathbf{n}\times\mathbf{P}_i(t-|\mathbf{R}-\mathbf{r}_i(t_i)|/c)], \quad (1)$$

where R is a distant observation point, $\mathbf{n}=\mathbf{R}/|\mathbf{R}|$ is the unit vector from the origin to the field point, c is the velocity of light, and $\mathbf{P}_i(t)$ is the atomic polarization which has been evaluated at the retarded time $t_i = t - |\mathbf{R}-\mathbf{r}_i(t_i)|/c$. The polarization is taken to be that due to independent atoms.

In order to evaluate the effect of the incident pulses on the ith atom we define an interaction picture

$$|\psi(t)\rangle_I = e^{iH_0(t-t_{1i})}|\psi(t)\rangle_S, \quad (2)$$

where the states on the left and right of Eq. (2) correspond to the interaction and Schrödinger pictures, respectively, H_0 is the atomic Hamiltonian, and t_{1i} is the arrival time of the first pulse. The effect of the pulse is to multiply the state (in the interaction picture) by the time transformation operator U_{1i}:

$$|\psi(t)\rangle_I = U_{1i}|\psi(t_{1i})\rangle_I = U_{1i}|\psi(t_{1i})\rangle_S. \quad (3)$$

The resulting Schrödinger state is

$$|\psi(t)\rangle_S = e^{-iH_0(t-t_{1i})}U_{1i}|\psi(t_1)\rangle_S. \quad (4)$$

The effect of the second pulse on the atom is similar. After both pulses the state of the ith atom is

$$|\psi_i(t)\rangle = \exp[-iH_0(t-t_{2i})]U_{2i} \exp[-iH_0(t_{2i}-t_{1i})]U_{1i}|\psi_i(t_{1i})\rangle. \quad (5)$$

Calculating the polarization from (5) and retaining only terms leading to an echo, it is found that

$$P_i(t_i) = P_i \cos[\omega_i(t_i+t_{1i}-2t_{2i})], \quad (6)$$

where

$$p_i = 2\langle b|ex_i|a\rangle|\langle a|U_{2i}|b\rangle|^2|\langle a|U_{1i}|b\rangle|\langle b|U_{1i}|b\rangle.$$

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[1] I. D. Abella, N. A. Kurnit, and S. R. Hartman, Phys. Rev. **141**, 391 (1966).

[2] See also H. Pendleton, in *Proceedings of the International Conference on the Physics of Quantum Electronics*, edited by P. L. Kelly, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Co., New York, 1965), p. 822.

The times of arrival of the pulses at the ith atom are determined by the equations

$$ct_{1i}=\mathbf{n}_1\cdot\mathbf{r}_i(t_{1i}), \quad (7)$$

$$ct_{2i}=c\tau+\mathbf{n}_2\cdot\mathbf{r}_i(t_{2i}). \quad (8)$$

Equation (6) must also be evaluated at the retarded time

$$ct_i=ct-R+\mathbf{n}\cdot\mathbf{r}_i(t_i), \quad (9)$$

where we have assumed that $R\gg r_i$. We will now discuss briefly the situations when (a) the atoms are fixed as in a solid, (b) the atoms move uniformly with random velocities, and (c) the atoms suffer collisions while in motion.

(a) For fixed atoms, as in a solid, $\mathbf{r}_i$ is independent of time and we find from (1) and (6)–(8) that the radiated field at R at time t is proportional to

$$E(R,t)\propto\sum_i p_i\cos\omega_i \times\{(t-2\tau-R/c)+\mathbf{r}_i\cdot(\mathbf{n}+\mathbf{n}_1-2\mathbf{n}_2)/c\}. \quad (10)$$

When there is a spread in frequencies ω_i (and p_i is a slowly varying function of ω_i) Eq. (10) gives a peak in $E(R,t)$ (the echo) at a time $2\tau+R/c$. Since the medium is much larger than a wavelength, $r_i\gg c/\omega_i$, E is largest if $\mathbf{n}=2\mathbf{n}_2-\mathbf{n}_1$. If the angle α between $\mathbf{n}_1$ and $\mathbf{n}_2$ is small, this condition is satisfied to order α^2 when the angle between the first pulse $\mathbf{n}_1$ and the echo $\mathbf{n}$ is 2α. Under these conditions there will be a reduction in the size of the echo if the length L of the sample and the angle are such that $(\alpha^2\omega L/c)\sim1$.[1]

(b) In the case of Doppler broadening we have no spread in the frequencies ω_i but the motion of the atom must be considered in finding t_{2i} and t_i. For uniformly moving atoms

$$\mathbf{r}_i(t)=\mathbf{r}_i(t_{1i})+(t-t_{1i})\mathbf{v}_i.$$

Substituting t_{2i} and t_i as determined from (7) and (8) into (6), we find that the radiated field in this case is proportional to

$$E(R,t)\propto\sum_i p_i\cos\{\omega(1+\mathbf{v}_i\cdot\mathbf{n}/c) \times[(t-2\tau-R/c)+\mathbf{r}_i\cdot(\mathbf{n}-2\mathbf{n}_2+\mathbf{n}_1)/c]+\omega\theta_i\}, \quad (11)$$

where

$$\theta_i=2\tau\mathbf{v}_i\cdot(\mathbf{n}-\mathbf{n}_1)/c+2\mathbf{r}_i\cdot(\mathbf{n}_2-\mathbf{n})\mathbf{v}_i\cdot(\mathbf{n}-\mathbf{n}_2)/c^2. \quad (12)$$

Except for the term in θ_i Eq. (11) is the same as that obtained for the inhomogeneously broadened system (7) but with $\omega_i=\omega(1+\mathbf{n}\cdot\mathbf{v}_i/c)$. Hence we see that an echo is expected in a gaseous medium. The first term in θ_i [Eq. (12)] causes a dephasing of the echo which is proportional to $\alpha\tau$ for small α. Experimentally[1] it is convenient to have $\alpha\neq0$ for the detection of the echo, so that for large τ this effect could pose a serious problem for the study of collision effects by the photon echo technique. The second term in θ is of order $r_iv_i\alpha^2/c^2$ and is negligible compared to the dephasing effect proportional to $r_i\alpha^2$ already discussed for the solid.

In the forward direction ($\mathbf{n}_1=\mathbf{n}_2=\mathbf{n}$), assuming a Maxwellian distribution of velocities in (11), the field at R is proportional to

$$E(R,t)\propto(N/R) \times\exp[-(\omega^2\langle v^2\rangle/2c^2)(t-2\tau-R/c)^2], \quad (13)$$

where N is the density of atoms and $\langle v^2\rangle$ is the mean-square atomic velocity. The time duration of the echo is then the inverse Doppler width, i.e., of order 10^{-9} sec for visible transitions in room-temperature gases.

(c) Restricting our attention to the foreward direction, the effects of collisions of the atoms are determined by a correlation function of the form

$$\left\langle\exp\left\{i\left[\int_\tau^{t-R/c}\Delta\omega(t)dt-\int_0^\tau\Delta\omega(t)dt\right] \pm i\frac{\omega}{c}\left[\int_\tau^{t-R/c}v(t)dt-\int_0^\tau v(t)dt\right]\right\}\right\rangle,$$

where $\Delta\omega(t)=\omega(t)-\omega$ and v is the velocity in the $\mathbf{n}$ direction. The first term represents the integrated frequency shifts induced by the collisions and the second term the effects of collision on the velocity of the atoms. Such a correlation function has been considered by Gyorffy, Borenstein, and Lamb.[3]

If, following Ref. 3, we divide the collisions into "hard" and "soft" varieties the echo (9) is attenuated by a factor of the form

$$\exp-\{\xi_1(2\tau)+\xi_2(2\tau)^3+\xi_3(2\tau)^{1/2}\},$$

where ξ_1 is the reciprocal of the mean hard collision time, ξ_2 is associated with the fact that many soft collisions lead to a Brownian motion of the atomic velocity, and the third factor ξ_3 comes from phase interruption of the radiating atoms due to soft collisions.

Our treatment has avoided calculating the excitation operators U_{ni} because they are not relevant to the kinematics which produces the photon echo. We required only that U_{ni} be independent of ω_i for a band of frequencies. The width of this band determines the temporal length of the echo. In addition we have assumed that the two incident pulses are not attenuated (i.e., a short sample).

Under certain circumstances coherent reradiation effects[4] can prevent the formation of a full photon echo; the atomic excitation produced by a pulse can radiate away before the atoms dephase, thereby destroying the memory of the pulse. The assumption of small attenuation of the exciting pulses is sufficient to prevent such effects unless the spectral content of the pulses is broader than the Doppler linewidth. An even shorter sample may be required for very short exciting pulses in order to observe the full echo.

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[3] B. L. Gyorffy, M. Borenstein, and W. E. Lamb, Jr., Phys. Rev. (to be published).
[4] D. C. Burnham and R. Y. Chiao (to be published).