Photon echo in nonlinear processes

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Optical induction and photon echo effects in nonlinear processes are shown to exist. They consist in instantaneous synchronization of the nonlinear polarization of the medium after two consecutive excitations of the medium by coherent fields. The echo phenomenon in combination (Raman) scattering is considered as an example. For the ordinary echo effect (resonant absorption processes), which is a particular case of the analysis performed here, the spatial distribution of the echo-signal intensity is determined by taking into account the real geometry of the active volume.

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High-power laser pulses of duration $\tau_p \sim 10^{-12}$–10^{-13} sec have by now been produced with the aid of lasers. These pulses allow us to observe, in all media, effects due to coherent interaction of light with matter (the condition for the coherence of the interaction is smallness of τ_p in comparison with the characteristic time T_2 of damping of the field-induced polarization of the medium). A central position among these effects is occupied by the photon echo (PE) effect,[1,2] which has been under intensive study recent years.[3] In the usual formulation of the experiment, the photon echo is produced spontaneously in the medium after it is excited by two successive coherent resonant pulses separated by a time interval $\tau \ll T_2, T_1$ (T_1 is the longitudinal-relaxation time). During the interval τ, the "phase memory" of the first pulse is erased by the reversible relaxation with characteristic time $T_2^* \ll T_1, T_2$ (T_2^{*-1} is the inhomogeneous broadening of the spectrum). The second pulse reconstructs the coherence of the radiators, and this leads to generation of a coherent echo signal at the instant 2τ. Interest in this phenomenon is due primarily to the fact that it serves as a convenient means of determining many important characteristics of resonant media (transition probabilities relaxation times, etc).

The present paper is devoted to establishment of the echo effect in nonlinear processes, and this should greatly extend the capabilities of the PE method. This effect arises when a medium interacts with nonresonant coherent fields.[1)] It will be shown below that the polarization of the medium, after two successive nonlinear excitation processes of rather general form at the instants 0 and τ ($\tau \ll T_2$), has the property of instantaneous synchronization ("collapse") at the instant 2τ and can be observed with the aid of a probing pulse that initiates a new nonlinear process that proceeds with high efficiency.

Assume that at the instants of time 0 and τ the system of molecules is acted upon by two successive excitations of duration τ_1 and τ_2, each consisting of a certain aggregate of coherent fields. The dynamic behavior of the system in various time intervals is clear from the following inequalities:

$$\tau_1, \tau_2 \ll T_2^* \ll \tau \ll T_2, T_1. \tag{1}$$

The interaction of the excitation pulses with the molecules is described by the Hamiltonian (i = 1 or 2 is the number of the excitation)

$$H_i = \hbar\omega_0\sigma_3 - \lambda_i(\sigma_+ Q_i^- + \sigma_- Q_i^+),$$

where ω_0 is the frequency of the working transition, while $\sigma_\pm$ and σ_3 are Pauli matrices (two-level description). The quantity λ_i is the matrix element of some nonlinear interaction leading to a transition between levels. The functions

$$Q_i^\pm = E_i \exp[\pm i(\omega t - \mathbf{k}_i\mathbf{r} - \varphi_i)], \quad E_i = \prod_{j=1}^{n} E_i^{(j)}$$

describe the aggregate of the excitation fields with the high-frequency factor separated; E_i is the product of the amplitudes of the fields making up the i-th excitation (n-th order process); $\omega = \omega_0 - \Delta\omega$, where $\Delta\omega$ is the inhomogeneous broadening; $\mathbf{k}_i$ and φ_i are the resultant wave vector and phase of the exciting field. For example, if the first excitation is realized by biharmonic pumping,[4] then

$$E_i = E_i^{(1)} E_i^{(2)}, \quad \omega = \omega_i^{(1)} - \omega_i^{(2)}, \quad \mathbf{k}_i = \mathbf{k}_i^{(1)} - \mathbf{k}_i^{(2)}, \quad \varphi_i = \varphi_i^{(1)} - \varphi_i^{(2)}, \tag{2}$$

where $E_1^{(1)}$, $E_1^{(2)}$; $\mathbf{k}_1^{(1)}$, $\mathbf{k}_1^{(2)}$; $\omega_1^{(1)}$, $\omega_1^{(2)}$ and $\varphi_1^{(1)}$, $\varphi_1^{(2)}$ are the amplitudes, wave vectors, frequencies, and phases of the fields making up the biharmonic excitation pulse. For two-photon absorption of pulses with unequal frequencies $E_1 = E_1^{(1)}E_1^{(2)}$, $\omega = \omega_1^{(1)} + \omega_1^{(2)}$, $\mathbf{k}_1 = \mathbf{k}_1^{(1)} + \mathbf{k}_1^{(2)}$, $\varphi_1 = \varphi_1^{(1)} + \varphi_1^{(2)}$ (the meaning of the notation is the same). For stimulated Raman scattering, relations (2) are satisfied, where the quantities with the superscript 1 pertain to the pump field and those with 2 to the Stokes wave.

We shall disregard the phase modulation of the excitation fields ($\partial\varphi_i(\mathbf{r}, t)/\partial t = 0$), assuming that the center of the symmetrical inhomogeneous-broadening line coincides with the carrier frequency ω of the exciting pulse, and that there is no phase modulation at the entrance into the medium ($\partial\varphi_i(0, t)/\partial t = 0$).

The equations of motion for the polarization of the medium with phenomenologically introduced relaxation terms take the following form (we omit here the index i, since the system of the obtained equations "works" in the entire time interval)

$$\frac{du}{dt} = -\frac{u}{T_2} - v\Delta\omega, \quad \frac{dv}{dt} = -\frac{v}{T_2} + u\Delta\omega + \frac{2\lambda}{\hbar}WE,$$
$$\frac{dW}{dt} = -\frac{W - W^{eq}}{T_1} - \frac{2\lambda}{\hbar}vE, \tag{3}$$

where W^{eq} is the equilibrium value of $\langle\sigma_3\rangle$ (we assume that at the instant t = 0 all the molecules are at the lower level, i.e., $W^{eq} = -1/2$),

$$u \pm iv = \langle\sigma_\pm\rangle \exp[\mp i(\omega t - \mathbf{k}\mathbf{r} - \varphi)]. \tag{3'}$$

The polarization of the medium at the frequency $\omega^{(j)}$ is given by

$$P_j^+ = P_j \frac{\partial Q^{(+)}}{\partial E^{(j)}} \exp[-i(\omega^{(j)}t - \mathbf{k}^{(j)}\mathbf{r} - \varphi^{(j)})], \tag{4}$$

where

$$P^+=\lambda N_v\int_{-\infty}^{+\infty}\langle\sigma_+\rangle f(\Delta\omega)\,d(\Delta\omega) \tag{5}$$

is the polarization at the frequency of the working transition; N_V is the molecule density; we average in (4) over the inhomogeneous-broadening spectrum,

$$\int_{-\infty}^{+\infty} f(\Delta\omega)\,d(\Delta\omega)=1.$$

We present the solutions of the system (3), (3′) for different time intervals. By virtue of condition (1), we put $T_1 = T_2 = \infty$.

After the passage of the first excitation pulse we have

$$u^{(1)}=0,\quad v^{(1)}=-{}^1/_2\sin\theta_1,\quad W=-{}^1/_2\cos\theta_1,$$

where

$$u^{(1)}+iv^{(1)}=\langle\sigma_+\rangle\exp\left[-i(\omega t-\mathbf{k}^{(1)}\mathbf{r}-\varphi^{(1)})\right],$$

$$\theta_1=\theta_1(\mathbf{r})=\frac{2\lambda_1}{\hbar}\int_0^{\tau_1}E_1(\mathbf{r},t')\,dt'$$

is the angle of rotation of the polarization vector under the influence of the exciting fields. Prior to the start of the action of the second excitation pulse, the inhomogeneous broadening leads to a dephasing of the polarizabilities:

$$\begin{aligned}u^{(1)}&={}^1/_2\sin\theta_1\sin(\Delta\omega\cdot\tau),\\ v^{(1)}&=-{}^1/_2\sin\theta_1\cos(\Delta\omega\cdot\tau),\quad W=-{}^1/_2\cos\theta_1.\end{aligned} \tag{6}$$

After the passage of the second excitation pulse we have

$$\begin{aligned}u^{(2)}&={}^1/_2\sin\theta_1\sin[\Delta\omega\cdot\tau-\chi],\\ v^{(2)}&=-{}^1/_2\cos\theta_2\sin\theta_1\cos[\Delta\omega\cdot\tau-\chi]-{}^1/_2\cos\theta_1\sin\theta_2,\\ W^{(2)}&=-{}^1/_2\cos\theta_1\cos\theta_2+{}^1/_2\sin\theta_1\sin\theta_2\cos[\Delta\omega\cdot\tau-\chi],\end{aligned} \tag{7}$$

where

$$u^{(2)}+iv^{(2)}=\langle\sigma_+\rangle\exp\left[-i(\omega t-\mathbf{k}^{(2)}\mathbf{r}-\varphi^{(2)})\right],$$

$$\chi=(\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}+(\varphi_2-\varphi_1),\quad \theta_2=\theta_2(\mathbf{r})=\frac{2\lambda_2}{\hbar}\int_{\tau}^{\tau+\tau_2}E_2(\mathbf{r},t')\,dt'$$

is the integral characteristic of the second exciting pulse. The condition $\tau_i \ll \tau$ was used in (6) and (7). Finally, at an arbitrary instant $t > \tau$ we have

$$\begin{aligned}u^{(2)}(t)&={}^1/_4\sin\theta_1(1+\cos\theta_2)\sin(\chi+\Delta\omega\cdot t)+{}^1/_2\cos\theta_1\\ &\times\sin\theta_2\sin[\Delta\omega(t-\tau)]+{}^1/_4\sin\theta_1(1-\cos\theta_2)\sin[\chi+\Delta\omega(2\tau-t)],\\ v^{(2)}(t)&=-{}^1/_4\sin\theta_1(1+\cos\theta_2)\cos(\chi+\Delta\omega\cdot t)-{}^1/_2\cos\theta_1\\ &\times\sin\theta_2\cos[\Delta\omega(t-\tau)]+{}^1/_4\sin\theta_1(1-\cos\theta_2)\\ &\times\cos[\chi+\Delta\omega(2\tau-t)],\\ W(t)&=-{}^1/_2\cos\theta_1\cos\theta_2+{}^1/_2\sin\theta_1\sin\theta_2\cos[\Delta\omega\cdot\tau-\chi].\end{aligned} \tag{8}$$

Substitution of formulas (6)–(8) in (4) and (5) shows that for an arbitrary distribution $f(\Delta\omega)$ the polarization of the medium P_j^+ differs from zero (more accurately, from the noise component) only in narrow time intervals at the instants 0, τ, and 2τ. We present the results for a Gaussian line $f(\Delta\omega)$. The nonlinear polarization P_j^+ is defined in accord with (4), where P^+ takes near $t = 0$ the value

$$P^+=-{}^1/_2 i\lambda N_V\sin\theta_1\exp(-t^2/T_2^{*2})\exp[i(\omega t-\mathbf{k}_1\mathbf{r}-\varphi_1)], \tag{9}$$

near $t = \tau$

$$\begin{aligned}P^+=-{}^1/_2 i\lambda N_V\cos\theta_1\sin\theta_2\exp[-(t-\tau)^2/T_2^{*2}]\cdot\\ \times\exp[i(\omega t-\mathbf{k}_2\mathbf{r}-\varphi_2)],\end{aligned} \tag{10}$$

and near $t = 2\tau$

$$\begin{aligned}P^+={}^1/_4 i\lambda N_V\sin\theta_1(1-\cos\theta_2)\exp[-(t-2\tau)^2/T_2^{*2}]\\ \times\exp[i(\omega t-(2\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}-(2\varphi_2-\varphi_1))].\end{aligned} \tag{11}$$

Expressions (9), (10), and (11) show that the polarization "collapses" at the instants 0, τ, and 2τ. This proves indeed the existence of a nonlinear PE. In analogy with the case of ordinary photon echo (linear PE), we can say that this behavior of the polarization during the course of its probing will initiate coherent induction signals at the instants 0 and τ, and echo signals at the instant[2)] 2τ.

To find the intensities of the induction and PE signals it is necessary to solve simultaneously Maxwell's equations and the equations for the polarization with the initial value of the latter as given by (4) and (11) at $t = 2\tau$.

Before we proceed to a concrete example of nonlinear PE, let us dwell the case of ordinary PE, which is a particular case of our analysis. The approach used here and based on the solution of three-dimensional Maxwell's equations makes it possible to obtain exact expressions for the intensity of the linear PE; these expressions contain a number of important singularities of which no notice has been taken in the literature so far. In this case $\mathbf{E}_1$, $\mathbf{E}_2$; $\mathbf{k}_1$, $\mathbf{k}_2$ and φ_1, φ_2 are the amplitudes, wave vectors, and phases of the resonant excitation fields. The spontaneous echo-signal amplitude $\mathbf{A}_e = \mathbf{E}_e\exp(i\omega t)$ satisfies the equation

$$\operatorname{grad}\operatorname{div}\mathbf{A}_3-\Delta\mathbf{A}_3+\frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}_3=-\mu_0\frac{\partial^2}{\partial t^2}\mathbf{P}_3, \tag{12}$$

where η is the refractive index, $\mu_0 = 4\pi/c^2$, and $\mathbf{P}_e$ is calculated from Eqs. (3), (3′), and (5) with an initial condition determined from (11) at $t = 2$. When solving (3) we can neglect the reaction of the echo on the medium, a procedure valid in a wide range of variation of the molecule density[10] and expressed by the condition

$$\alpha l\ll 1, \tag{13}$$

where $\alpha = 4\pi\lambda^2\omega^2(\hbar c\eta T_2^*)^{-1}$ is the absorption coefficient. Recognizing that the active volume V subtended in the medium by the excitation pulses is in fact an elongated cylinder (a is the radius and l the length, $a/l \ll 1$), we shall use in (12) a scalar approximation, and consider the radiation near the cylinder axis (the cylinder axis z coincides with the direction of $\mathbf{k}_1$). Using also the inequalities $\omega \gg |\partial/\partial t|$ and $\omega T_2^* \gg 1$, we obtain

$$\begin{aligned}-\Delta E_3-k^2E_3={}^1/_4 i\mu_0\omega^2\lambda N_V\sin\theta_1(1-\cos\theta_2)\cdot\exp[-(t-2\tau)^2/T_2^{*2}]\\ \times\exp\{-i[(2\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}+2\varphi_2-\varphi_1]\},\end{aligned}$$

where $k^2 = \omega^2\eta^2/c^2$. Hence

$$\begin{aligned}E_e=i\frac{\mu_0\omega^2\lambda N_V}{16\pi}\exp\left\{-\frac{(t-2\tau)^2}{T_2^{*2}}\right\}\int_{(V)}d\mathbf{r}'\frac{\exp\{-ik|\mathbf{r}-\mathbf{r}'|\}}{|\mathbf{r}-\mathbf{r}'|}\\ \times\sin\theta_1(1-\cos\theta_2)\exp\{-i[(2\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}'+2\varphi_2-\varphi_1]\}.\end{aligned}$$

Using the condition (13) and neglecting the contribution of φ_1 to the dispersion we find that in the observation zone ($|\mathbf{r}| \gg |\mathbf{r}'|$) the echo signal constitutes a spherical wave with unevenly distributed intensity

$$\begin{aligned}E_e=A\frac{\exp\{-i(k|\mathbf{r}|+2\varphi_2-\varphi_1)\}}{|\mathbf{r}|}\sin\bar\theta_1(1-\cos\bar\theta_2)\\ \times\exp\{-(t-2\tau)^2/T_2^{*2}\}\int_{(V)}\exp\{-i(2\mathbf{k}_2-\mathbf{k}_1-k\mathbf{n})\mathbf{r}'\}\,d\mathbf{r}',\end{aligned} \tag{14}$$

where

$$A=i\mu_0\omega^2\lambda N_V/16\pi,\quad \mathbf{n}=\mathbf{r}/|\mathbf{r}|,$$

and $\bar\theta_1$ and $\bar\theta_2$ are the areas of the exciting pulses at the entry into the medium. Integrating in (14), we have

$$I_e=|E_e|^2=16\pi^2a^2A^2\frac{1}{|\mathbf{r}|^2}\sin^2\bar\theta_1(1-\cos\bar\theta_2)^2. \tag{15}$$

$$\cdot\exp\left[-\frac{2(t-2\tau)^2}{T_2^{*2}}\right]\frac{\sin^2(1/2 l\Delta k_\parallel)}{(\Delta k_\parallel)^2}\frac{J_1^2(a\Delta k_\perp)}{(\Delta k_\perp)^2},$$

where

$$\Delta k_\parallel=|2\mathbf{k}_2-\mathbf{k}_1|\cos\psi_1-k\cos\psi_2,\quad \Delta k_\perp=|(2\mathbf{k}_2-\mathbf{k}_1)_\perp-k\mathbf{n}_\perp|; \qquad (16)$$

ψ_1 is the angle between the z axis and the vector $2\mathbf{k}_2-\mathbf{k}_1$; ψ_2 is the angle between the z axis and the vector $\mathbf{n}$: $J_1(x)$ is a Bessel function of first order.

In contrast to the customarily employed expressions derived with the aid of the plane-wave method,[2] formula (15) provides a complete picture of the spatial and temporal distribution of the PE signal intensity. We note in particular, the following important circumstance. The sharp maximum of the PE intensity at $\mathbf{k}_1 \parallel \mathbf{k}_2$ is realized in the direction of the z axis, which in this case coincides with the direction of $2\mathbf{k}_2-\mathbf{k}_1$. At the same time, if the exciting pulses are not parallel (it is customary to use, for the observation of the PE in condensed media, an experimental setup in which the excitation pulses can be spatially separated from the PE), the condition $k\mathbf{n} = 2\mathbf{k}_2-\mathbf{k}_1$ cannot be satisfied, since the vectors contained in this condition are of equal length. However, even in this case it is concluded on the basis of the plane-wave method (see[2] and all the subsequent papers) that the maximum of the intensity coincides with the direction of $2\mathbf{k}_2-\mathbf{k}_1$. An investigation of (15) with $\Delta\mathbf{k}_\parallel$ and $\Delta\mathbf{k}_\perp$ related by formulas (16) shows that this is not so. The intensity I_e can have two maxima. One is always directed along the z axis, i.e., along $\mathbf{k}_1$. The second maximum likewise does not coincide with the direction of $2\mathbf{k}_2-\mathbf{k}_1$, and its value depends very strongly on the real geometry of the volume V (on the parameters a and l). Moreover, as shown by a numerical analysis, this maximum appears far from always. Thus, the strong sensitivity of I_e to changes in the geometry of the volume is indeed the cause of the difficulty of observing the PE.

An analogous procedure is used to calculate the intensity of the induction signals observed at the instants 0 and τ and having sharp maxima in the directions $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively.

As a second example we consider the case when the polarization is probed by a scattering pulse of amplitude E_p, frequency ω_p, and wave vector $\mathbf{k}_p$. According to (3), (5), and (11), the polarization at the Stokes frequency $\omega_s = \omega_p - \omega_0$ is equal to

$$P_s^+=1/4 i\lambda_{cr}N_v\sin\theta_1(1-\cos\theta_2)E_p\exp\left(-(t-2\tau)^2/T_2^{*2}\right]$$
$$\times\exp\left[i(\omega_s t-(\mathbf{k}_p-2\mathbf{k}_2+\mathbf{k}_1)\mathbf{r})\right],$$

where λ_{cr} is the scattering matrix element, and θ_1 and θ_2 are the characteristics of the exciting pulses (the excitation method is not spelled out here concretely). Under the same assumptions as before, in the given-pump-field (E_p) approximation we have for the intensity of the Stokes wave

$$I_s=\frac{B}{|\mathbf{r}|^2}\sin^2\theta_1(1-\cos\theta_2)^2\exp\left[-2\left(\frac{t-2\tau}{T_2^*}\right)^2\right]$$
$$\times\frac{\sin^2(\Delta k_\parallel l/2)}{(\Delta k_\parallel)^2}\frac{J_1^2(a\Delta k_\perp)}{(\Delta k_\perp)^2}$$

where

$$B=(1/4 a\mu_0\omega_s^2\lambda_{cr}^2N_vE_p)^2,\quad k_s^2=\omega_s^2\eta_s^2/c^2,$$

$\Delta k_\parallel$ and $\Delta k_\perp$ are the lengths of the projections of the vector $\Delta\mathbf{k} = \mathbf{k}_p - 2\mathbf{k}_2 + \mathbf{k}_1 - n\mathbf{k}_s$ on the z axis ($\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ is the observation direction) and on the plane perpendicular to it. In this case the vectors $2\mathbf{k}_2-\mathbf{k}_1$, $\mathbf{k}_p$, and $k_s\mathbf{n}$ have different lengths. Therefore the coherent Stokes-scattering echo signal will have one sharp maximum in the direction defined by the relation $k_s\mathbf{n}_m = \mathbf{k}_p - (2\mathbf{k}_2-\mathbf{k}_1)$. The intensity of this signal

$$I_s=\frac{B}{16|r|^2}a^2l^2\sin^2\theta_1(1-\cos\theta_2)^2\exp\left[-2\left(\frac{t-2\tau}{T^*}\right)^2\right]$$

greatly exceeds the level of the spontaneous Raman noise scattering (comparative numerical data can be obtained by using the results of[11]). It is also easy to establish that an intense anti-Stokes scattering echo signal will be observed in the direction $k_{as}\mathbf{n}_m = \mathbf{k}_p + (2\mathbf{k}_2-\mathbf{k}_1)$, where $k_{as}^2 = \eta_{as}^2(\omega_0+\omega_p)^2/c^2$. We note also that the induction signals at the instants 0 and τ will be directed at angles satisfying the conditions $k_s\mathbf{n}_m = \mathbf{k}_p - \mathbf{k}_1$ and $k_s\mathbf{n}_m = \mathbf{k}_p - \mathbf{k}_2$ for the Stokes scattering and the conditions $k_{as}\mathbf{n}_m = \mathbf{k}_p + \mathbf{k}_1$ and $k_{as}\mathbf{n}_m = \mathbf{k}_p + \mathbf{k}_2$ for the anti-Stokes scattering. Thus, in contrast to the linear echo, a spatial separation of the induction signals from the excitation signals is possible here. The method of photon inductions and photon echo in Raman scattering can serve by the same token as a supplement to the method of active Raman scattering spectroscopy, which has been developed recently.[11,12]

It is easy to continue the list of examples of echoes in nonlinear processes. We shall indicate the main requirements that must be satisfied in the experiment. It follows from the inequalities (1) that the active medium must be chosen such that the homogeneous broadening be much smaller than the inhomogeneous broadening. At the present time there are no sufficiently well systematized data on the nature of the line broadening, particularly in condensed media. We can point out as examples, however, the media already used to obtain the ordinary echo effect, namely Cr^{3+} impurities in ruby and in Al_2O_3, Nd^{3+} impurities in $CaWO_4$, the gases SF_6, BCl_3, and CO_2, and Cs vapor. It is also possible to use media activated in Raman scattering and satisfying the condition $T_2^* \ll T_2$, i.e., fused quarts, Shpol'skiĭ systems, rare-earth impurities in glasses, gases of simple molecules (H_2, O_2, N_2, I_2) at sufficiently low pressures, etc. (see[13] and the literature cited there). We note that from the point of view of the experiment, the most convenient scheme seems to be one in which two short powerful excitation pulses are applied against the background of a stationary "pilot" probing pulse. In contrast to the linear induction and echo effects, it is possible to have here frequency and spatial separation of the induction and echo signals from the excitation pulses, the spatial separation being weakly dependent on the geometry of the active volume. We note in concluding that the use of echo phenomena in nonlinear processes makes it possible not only to measure the nonlinear polarizabilities of the medium and its relaxation characteristics, but also to extend greatly the possibilities of other applications of phonon echo,[2] which have been restricted so far only to resonant absorption processes.

[1)] In this sense, the usual PE effect, which corresponds to processes of resonant absorption and is described by perturbation theory of first order in the field, will be called by us the linear PE although this phenomenon itself is, of course, described by nonlinear electrodynamic equations.

[2)] It is appropriate to note that among echo phenomena that are apparently not connected with definite resonant transitions are the effects of

radio-frequency spontaneous echo in ferroelectrics, [5] in type-II superconductors, [6] and in ferromagnets. [7] In contrast to the nonlinear-optical processes considered by us, the nature of these effects is connected with the phasing of the electric or magnetic dipoles of the medium, and can be described within the framework of the theory of cyclotron echo. [8, 9]

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