Temporal and correlation properties of echo signals in two- and three-level systems under conditions of inhomogeneous broadening of resonant energy levels

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The temporal properties due to the Fourier-transforming properties of multilevel resonant systems are considered for coherent spontaneous emission of the echo type are discussed. The influences exerted on the photo-echo properties by qualitative differences between the mechanisms of inhomogeneous broadening of energy levels are considered in gases and also in crystals with paramagnetic impurities. It is shown that various time regimes can be realized in the coherent responses, viz., duplication of the temporal form of one of the exciting light pulses in either the forward direction or in a direction specularly reversed in time, as well as correlation analysis of excitation pulses with simultaneous scale transformations.

INTRODUCTION

Coherent interaction of optical pulses with resonant media is accompanied by many transition phenomena. Those investigated in greatest detail are optical nutations (ON), self-induced transparency (SIT), and photon (optical) echo (PE). The PE effect yields, in particular, otherwise unobtainable information on the energy-level fine structure masked under ordinary conditions by the inhomogeneous broadening of the spectral lines. Photon echo is therefore widely used in high- and ultrahigh-resolution spectroscopy. In addition to this already traditional application of the PE effect, great interest attaches to its use in systems for optical reduction of information, in view of the variety of the formed temporal and spatial structures of the optical echo responses.[2–5] Some similarities in the time dependences of the spin-echo signals and the external rf coding pulses were observed even earlier by Fernhach and Proctor[6] and also by Anderson *et al.*[7] It was also shown quite recently that under certain conditions the time envelope of a two-pulse PE signal can duplicate exactly, but with the time reversed, the temporal form of the coding optical pulse.[8] This similarity was later initially confirmed qualitatively in experiments on ruby crystals,[9] and subsequently also quantitatively for gaseous media.[10–13]

Of particular interest are investigations of the time features of three-pulse stimulated PE signals. It is known by now that a stimulated PE signal was observed in a Pr^{3+}-activated LaF_3 crystal when a third exciting pulse is applied even 30 minutes after the action of the second optical pulse[14]! In contrast to the primary or two-pulse PE, three-pulse excitation of a resonant medium permits both direct and mirror-reversed reproduction of the coding pulse in the stimulated and restored PE signals.[15] Interest attaches in this connection, from the standpoint of both the physical mechanism and the possible applications, to the recently observed time contraction of a PE signal compared with the external exciting light pulses.[16,17] For this to occur it is necessary that the Fourier-spectrum segment due to phase modulation be larger than the spectral region occupied by the time-dependent envelope of the amplitude.

Temporal correlations in coherent radiation are closely related to echo-signal spatial singularities that are produced in transformation of the wave fronts.[18–22] The resonant medium plays essentially here the part of a Fourier-analyzer of sorts. The Fourier spectrum of the exciting pulses is realized in the macroscopic polarization and population induced by the external field.[23–25] This last circumstance is particularly important for the understanding the mechanism that produces, by photochemical hole burning, a long-time holographic memory based on the accumulated stimulated PE.[26] The Fourier-transforming properties of a system two-level atoms are possessed also by spin systems in the region of nuclear magnetic resonance, and this is at present of interest for the development of radio-signal processing apparatus.[27,28] In our opinion, however, the scope for practical utilization of coherent responses in the optical band is much wider, since they possess also the dynamic properties of echo-holograms.

The temporal properties of the echo responses manifest themselves distinctly in excitation of multilevel systems, particularly three-level systems in gases.[29] Here, too, one can have both direct and mirror-reversed images of the temporal form of the coding pulse in signals of modified stimulated PE, as well as three-level PE.[33–35] A distinguishing feature of the regimes considered is the onset of a scale factor, due to the specific mechanism of inhomogeneous broadening of the energy levels in gases. It remains unclear, however, how the Fourier-transforming properties of multilevel atoms will manifest themselves under similar conditions in solids, where the character of the inhomogeneous broadening is determined by other than a Doppler mechanism.

The present paper is devoted to a theoretical analysis of the temporal properties of coherent echo responses connected with the Fourier-transforming properties of resonant multilevel systems. The primary two-pulse PE, stimulated

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PE in two-level systems, as well as combined PE, modified stimulated echo, and three-level PE in three-level systems with nonequidistant spectrum are considered. The influence exerted on the PE properties by qualitative differences in the inhomogenous energy-level broadening in gases, as well as in solids, viz., crystals with paramagnetic impurities, are discussed. It is shown that, depending on the relations between the amplitudes, on the durations of the exciting light pulses, and on the times of reversible and irreversible relaxation of the polarization of the medium, one can have either direct or mirror-reversed duplication of the form of one of the exciting light pulses (the coding one). Alternately, the temporal profile of the echo-signal envelope is a correlation function or a function in the form of a convolution of the excitation-pulse envelopes. Also discussed are the properties of the scale transformations connected with the character of the inhomogeneous broadening of the resonant energy levels.

§1. FUNDAMENTAL EQUATIONS. SOLUTION METHODS

We consider the formation of various reponses of PE in the interaction between coherent optical pulses and a system of three-level resonant atoms. The quantum-mechanical properties of the resonant system will be described by components of a density matrix, and the interaction of the light pulses with the medium will be considered in the dipole approximation. The fundamental equations take then the form

$$i\hbar\dot{\hat{\rho}}=[\hat{H},\ \hat{\rho}],\quad \hat{H}=\hat{H}_0-\hat{\mathbf{d}}\mathbf{E}(\mathbf{r},\ t), \tag{1.1}$$

where $\hat{\mathbf{d}}$ is the dipole-moment operator, and the intensity of the electric field of the light pulses is described by the expression

$$\mathbf{E}(\mathbf{r},\ t)=\mathrm{Re}\{\mathbf{E}_\Omega(\mathbf{r},\ t)e^{-i\Omega t}+\mathbf{E}_\omega(\mathbf{r},\ t)e^{-i\omega t}+\mathbf{E}_{\omega-\Omega}(\mathbf{r},\ t)e^{-i(\omega-\Omega)t}\},$$

with the frequencies Ω, ω, and ω–Ω close to the transition frequencies ω_{ba}, ω_{ca}, ω_{cb}.

We shall be mainly interested in cases when the inequality $\delta_i < T_2$ is satisfied, where δ_i is the duration of the exciting light pulses and T_2 is the characteristic time that determines the homogeneous width of the spectral line. Under these conditions, the relaxation processes that occur during the action of the light pulses can be neglected. Allowance for the relaxation processes in the intervals between the light pulses leads only to the onset of characteristic exponential factors, which we will also omit to simplify the ensuing expressions.

After changing to the component representation of the density matrix $\hat{\rho}$, the fundamental equations take the form

$$\begin{aligned}\dot{\rho}_{\alpha\alpha}&=\frac{i}{\hbar}\sum_\beta\{(\mathbf{Ed})_{\alpha\beta}\rho_{\beta\alpha}-\rho_{\alpha\beta}(\mathbf{Ed})_{\beta\alpha}\},\\ \dot{\rho}_{\beta\alpha}&=-i\omega_{\beta\alpha}\rho_{\beta\alpha}-\frac{i}{\hbar}(\mathbf{Ed})_{\beta\alpha}(\rho_{\beta\beta}-\rho_{\alpha\alpha})\\ &+\frac{i}{\hbar}\{(\mathbf{Ed})_{\beta\gamma}\rho_{\gamma\alpha}-\rho_{\beta\gamma}(\mathbf{Ed})_{\gamma\alpha}\},\quad \alpha,\beta=a,b,c.\end{aligned} \tag{1.2}$$

We separate in the elements of the density matrix ρ the slowly varying factors $\hat{\sigma}$, so that

$$\rho_{\beta\alpha}(\mathbf{r},\ t)=\sigma_{\beta\alpha}(\mathbf{r},\ t)\exp(-i\tilde{\omega}_{\beta\alpha}t), \tag{1.3}$$

where $\tilde{\omega}_{ba}=\Omega$, $\tilde{\omega}_{ca}=\omega$, $\tilde{\omega}_{cb}=\omega-\Omega$. After averaging over a time interval $T\gg\omega^{-1}$, Ω^{-1}, $(\omega-\Omega)^{-1}$ we have then

$$\begin{aligned}\dot{\sigma}_{\alpha\alpha}&=\frac{i}{2\hbar}\sum_\beta\{(\mathbf{Ed})_{\alpha\beta}\sigma_{\beta\alpha}-\sigma_{\alpha\beta}(\mathbf{Ed})_{\beta\alpha}\},\\ \dot{\sigma}_{\beta\alpha}&=-i\varepsilon_{\beta\alpha}\sigma_{\beta\alpha}-\frac{i}{2\hbar}(\mathbf{Ed})_{\beta\alpha}(\sigma_{\beta\beta}-\sigma_{\alpha\alpha})\\ &\quad+\frac{i}{2\hbar}\{(\mathbf{Ed})_{\beta\gamma}\sigma_{\gamma\alpha}-\sigma_{\beta\gamma}(\mathbf{Ed})_{\gamma\alpha}\},\end{aligned} \tag{1.4}$$

where

$$(\mathbf{Ed})_{ba}=\mathbf{E}_\Omega\mathbf{d}_{ba},\quad (\mathbf{Ed})_{ca}=\mathbf{E}_\omega\mathbf{d}_{ca},\quad (\mathbf{Ed})_{cb}=\mathbf{E}_{\omega-\Omega}\mathbf{d}_{cb},$$

and $\varepsilon_{\beta\alpha}$ stand for the detunings

$$\varepsilon_{ba}=\varepsilon_\Omega=\omega_{ba}-\Omega,\quad \varepsilon_{ca}=\varepsilon_\omega=\omega_{ca}-\omega,\quad \varepsilon_{cb}=\varepsilon_{\omega-\Omega}=\omega_{cb}-\omega+\Omega.$$

The polarization in the medium is determined by the mean value of the dipole-moment operator

$$\begin{aligned}\langle\mathbf{d}(\mathbf{r},\ t)\rangle&=\mathrm{Sp}\,(\hat{\rho}\hat{\mathbf{d}})=2\,\mathrm{Re}\,\{\langle\sigma_{ca}(\mathbf{r},\ t)\rangle\mathbf{d}_{ac}e^{-i\omega t}\\ &+\langle\sigma_{ba}(\mathbf{r},\ t)\rangle\mathbf{d}_{ab}e^{-i\Omega t}+\langle\sigma_{cb}(\mathbf{r},\ t)\rangle\mathbf{d}_{bc}e^{-i(\omega-\Omega)t}\},\end{aligned} \tag{1.5}$$

where the angle brackets in the right-hand side denote averaging over the frequency spreads of the corresponding transitions.

We shall solve the system (1.4) by successive approximations in the case of "small areas" of the exciting pulses, when the inequality

$$\left|\frac{\mathbf{d}_{\alpha\beta}}{\hbar}\int_{t_i}^{t_i+\delta_i}\mathbf{E}(\mathbf{r},t'-t_i)\,dt'\right|<1, \tag{1.6}$$

holds, where t_i are the times corresponding to the start of the pulse. In addition, we assume that the coherent light pulses have different frequencies Ω,ω and ω–Ω at different instants of time. Consider, for example, a detailed solution of the system (1.4) when a pulse of frequency Ω acts on a resonant medium. When the inequality (1.6) is satisfied, we can neglect in the zeroth approximation the change of the diagonal density-matrix elements. By the end of the pulse action we have then

$$\begin{aligned}\sigma_{ba}(t_i+\delta_i)&=\sigma_{ba}(t_i)\exp(-i\varepsilon_\Omega\delta_i)\\ &+\frac{i}{2\hbar}[\sigma_{aa}(t_i)-\sigma_{bb}(t_i)]\exp(-i\varepsilon_\Omega\delta_i)\,(\mathbf{d}\mathscr{E}_\Omega(\varepsilon_\Omega)),\end{aligned} \tag{1.7}$$

where $\mathscr{E}_\Omega(\varepsilon_\Omega)$ is the Fourier transform of the light-wave electric-field amplitude:

$$\mathscr{E}_\Omega(\varepsilon_\Omega)=\int_0^{\delta_i}\mathbf{E}_\Omega(\tau)\exp(i\varepsilon_\Omega\tau)\,d\tau. \tag{1.8}$$

To calculate the usual (primary) PE signal we must know the results of the action of the pulse on the "coherence" (off-diagonal density-matrix elements) produced by the preceding pulse. In next order of perturbation theory we obtain

$$\begin{aligned}\sigma_{ba}(t_i+\delta_i)&=\frac{1}{4\hbar^2}\{\sigma_{ab}(t_i)e^{-i\varepsilon\delta_i}(\mathbf{d}_{ba}\mathscr{E}_\Omega(\varepsilon))^2\\ &-\sigma_{ba}(t_i)e^{-i\varepsilon\delta_i}(\mathbf{d}_{ba}\mathscr{E}_\Omega(\varepsilon))\,(\mathbf{d}_{ba}\mathscr{E}_\Omega(\varepsilon))^*\}.\end{aligned} \tag{1.9}$$

Finally, for many problems it is necessary to calculate the responses of the media to fields of other frequencies, for example, combined PE or three-level echo (§3). Using again as an example the coherence produced in the ba transition, we easily obtain for σ_{ba}, using the right-hand sides of (1.4), the expressions

$$\sigma_{ba}(t_i+\delta_i)=\frac{i(\mathbf{d}_{bc}\mathscr{E}^*_{\omega-\Omega}(\varepsilon_\omega-\varepsilon_\Omega))}{2\hbar}\exp(-i\varepsilon_\Omega\delta_i)\sigma_{ca}(t_i) \tag{1.10a}$$

or

$$\sigma_{ba}(t_i+\delta_i)=-\frac{i(\mathbf{d}_{ca}\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega))}{2\hbar}\exp(-i\varepsilon_\Omega\delta_i)\sigma_{bc}(t_i), \tag{1.10b}$$

where $\mathscr{E}^*_{\omega-\Omega}(\varepsilon_\omega-\varepsilon_\Omega)$ and $\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)$ are the Fourier transforms of the fields at the corresponding frequencies. We obtain analogously the elements of the density matrix for other transitions. Note that the fundamental system (1.4) can also have a solution either in the "strong-field" approximation or in the "narrow-spectral-line" approximation (the latter was considered in detail earlier in Refs. 36 and 37). In the limit $\theta<1$ in the corresponding solutions, and in the limit $\varepsilon\tau<1$ in expressions (1.7)–(1.10), the two sets coincide.

We examine now the evolution of different PE signals. We begin for simplicity with the usual two-level system with nondegenerate energy levels.

§2. TEMPORAL PROPERTIES OF PE-SIGNAL FORMATION IN TWO-LEVEL SYSTEMS

1. Primary PE

Let the first pulse (frequency Ω, duration δ_1) be incident on the boundary $z=0$ of a resonant medium at the instant $t=0$. After a time δ_1, polarization is excited in the medium, owing to the nonzero dipole moment of the atoms in the $a\to b$ transition. Neglecting polarization effects, we can obtain for the average dipole moment of the atom, following the action of two light pulses, the expression

$$\langle d(t)\rangle=\mathrm{Re}\left\{-\frac{i|d_{ab}|^4\pi}{2\hbar^3}\times e^{-i\Omega t}\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\mathscr{E}_2^{\,2}(\varepsilon)\mathscr{E}_1^{\,*}(\varepsilon)e^{-i\varepsilon(t-t')}\right\}, \tag{2.1}$$

where

$$t'=2\tau+2\delta_1+z/v.$$

Expression (2.1) is basic for the explanation of many temporal properties of spin-echo and PE signals. It can be seen from (2.1), in particular, that two-level atoms transform the time dependence of the envelope of the amplitude of the light-signal electric field into a Fourier transform, and the Fourier transform of a sequence of two coherent light pulses in a PE signal are the product of the Fourier transforms of the individual pulses. Taking the inverse Fourier transform in (2.1), we obtain for the time dependence of the PE signal

$$F(t)=\int d\xi\, d\xi_1\, d\xi_2\, G(\xi)E_2(\xi_1)E_2(\xi_2)E_1^{\,*}(\xi_1+\xi_2+\xi-t+t'), \tag{2.2}$$

where the function $G(\xi)$ is the Fourier transform of the function $g(\varepsilon)$ that characterizes the distribution of the elementary emitters in the detunings within the limit of an inhomogenously broadening line. For a line of Gaussian form, the functions $g(\varepsilon)$ and $G(\xi)$ are respectively.

$$g(\varepsilon)=\frac{T_2^{\,*}}{\sqrt{\pi}}\exp[-(\varepsilon-\Delta\omega_0)^2T_2^{\,*2}],$$

$$G(\xi)=\frac{1}{2\pi}\exp\left[-i\Delta\omega_0\xi-\frac{\xi^2}{4T_2^{\,*2}}\right], \tag{2.3}$$

where T_2^* is the characteristic time that determines the inhomogeneous line width, while $\Delta\omega_0=\omega_{ba}^0-\Omega$ defines the deviation of the field frequency from the line center.

In the particular case when the exciting light pulses satisfy the strong-field conditions (in which case the inequality $\theta_i>\delta_i/T_2^*$ holds) or are spectrall broad (and then $\delta_i<T_2^*$), the dependence on the detuning ε can be neglected in $\mathscr{E}_2(\varepsilon)e^{-i\varepsilon\delta_2}$ and $\mathscr{E}_1^*(\varepsilon)e^{i\varepsilon\delta_1}$. Then

$$F(t)=cG(t-t_e), \tag{2.4}$$

where c is a constant independent of the time, and $t_e=z/v+2\tau+\delta_1+\delta_2$. This result means that the medium polarization, which is responsible for the echo signal, is a maximum at the instant t_e and occurs at a frequency that coincides with the central frequency of the spectral line. Note that in the small-areas approximation the two conditions—that the field be strong and that the spectral lines be narrow—are equivalent.

If only the second pulse is strong, the temporal form of the echo signal is given by an expression of the type

$$F(t)=c'\int d\xi\, G(\xi)E_1^{\,*}(\xi-t+t_e+\delta_1). \tag{2.5}$$

If, however, the characteristic duration of the first coding pulse exceeds the reversible polarization-relaxation time T_2^*, the function E_1^* can be taken outside the integral of (2.5). Then

$$F(t)=c'g(0)E_1^{\,*}(t_e+\delta_1-t), \tag{2.6}$$

where $g(0)=\pi^{-1/2}T_2^*\exp(-\Delta\omega_0^2T_2^{*2})$, and we obtain the mirror image of the temporal form of the first coding pulse in the PE signal. If the coding light pulse is the second one, and $\mathscr{E}_1^*(\varepsilon)e^{i\varepsilon\delta_1}=\text{const}$ for the first light pulse, we obtain

$$F(t)=c''\int d\xi\, d\xi'\, G(\xi)E_2(\xi')E_2(t-t_e+\delta_2-\xi-\xi'). \tag{2.7}$$

Again, if the inequality $\delta_2>T_2^*$ obtains,

$$F(t)=c''g(0)\int E_2(t-t_э+\delta_2-\xi)E_2(\xi)\,d\xi. \tag{2.8}$$

The waveform of the second exciting pulse is thus not reproduced, strictly speaking, in the primary PE signal, but is determined by an expression having the form of a convolution integral. We consider now the formation of stimulated PE signals in a two-level system.

2. Stimulated PE

It must be borne in mind, in the calculation of the atom's resonant dipole moment responsible for the formation of the stimulated PE signal in the medium, that the second exciting light pulse transforms the "coherence" produced by the first pulse into diagonal density-matrix elements, i.e., into population. We can then obtain for the mean value of the resonant dipole moment following the action of three light pulses the expression

$$\langle d(t)\rangle=\mathrm{Re}\left\{-\frac{i|d_{ab}|^4\pi}{\hbar^3}e^{-i\Omega t}\times\int\frac{d\varepsilon}{2\pi}\mathscr{E}_3(\varepsilon)\mathscr{E}_2(\varepsilon)\mathscr{E}_1^*(\varepsilon)g(\varepsilon)e^{-i\varepsilon(t-t'')}\right\}, \tag{2.9}$$

where $t''=z/v+2\tau_1+\tau_2+2\delta_1+\delta_2$. For the function $F_c(t)$ that determines the time dependence of the stimulated PE signal we obtain from (2.9)

$$F_c(t)=\int d\xi\, d\xi'\, d\xi''\, G(\xi)E_3(\xi')E_2(\xi'')E_1^*(\xi+\xi'+\xi''-t+t''). \tag{2.10}$$

We consider now particular cases of the general expression (2.10). If the amplitudes (durations) of all the exciting pulses satisfy the strong-field (narrow-spectral line) condition, we have

$$F_c(t)=cG(t-t_c), \tag{2.11}$$

where $t_c=z/v+2\tau_1+\tau_2+\delta_1+\delta_2+\delta_3$. In the upshot, that part of the medium polarization which is responsible for the stimulated PE signal is a maximum at the instant t_c and occurs at a frequency $\Omega+\Delta\omega_0$ corresponding to the center of the spectral line. If only the first pulse satisfies the strong-field (narrow spectral line) condition, we have

$$F_c(t)=c'\int d\xi\, d\xi'\, G(\xi)E_2(\xi')E_3(t-t_c+\delta_3-\xi-\xi'). \tag{2.12}$$

If the durations of the second and third pulses exceed the time T_2^*, then

$$F_c(t)=c'g(0)\int d\xi\, E_2(\xi)E_3(t-t_c+\delta_3-\xi), \tag{2.13}$$

i.e., F_c is a convolution-type integral, just as Eq. (2.12). If the second pulse in (2.10) is the strong one $\mathscr{E}_2(\varepsilon)=\text{const}$ or else $E_2(\xi)\sim\delta(\xi)$, we obtain the correlation integral

$$F_c(t)=c''\int d\xi\, d\xi'\, G(\xi)E_3(\xi')E_1^*(\xi'+\xi-t+t''). \tag{2.14}$$

Finally, if the third pulse in (2.10) satisfies the strong-field condition, we obtain ultimately a correlation integral between the envelopes of the first and second pulses:

$$F_c(t)=c\int d\xi\, d\xi'\, G(\xi)E_2(\xi')E_1^*(\xi'+\xi-t+t_c-\delta_1). \tag{2.15}$$

On the other hand, if two out of the sequence of three light pulses exciting a resonant medium are pairwise strong, and the inequality $\delta_{\rm cod}>T_2^*$ holds, the stimulated PE duplicates the waveform of the coding pulse either in the forward time direction (when the coding pulse is either the second or the third) or in the backward direction (the coding pulse is first):

$$F_c(t)\sim g(0)\begin{cases}E_3(t-t_c+\delta_3),\\ E_2(t-t_c),\\ E_1^*(t_c-\delta_1-t).\end{cases} \tag{2.16}$$

Note that the last result was obtained in Ref. 15.

We consider now the temporal features of the PE signals of coherent responseses in three-level systems.

§3. PE SIGNALS EXCITED IN THREE-LEVEL SYSTEMS

1. Combined PE

Let a three-level system with nonequidistant spectrum be excited by a sequence of two coherent light pulses at different frequencies Ω and ω with a time interval τ between them. It is known that a PE signal having the combination frequency $\omega-\Omega$ can be produced under these conditions, and the time of appearance of this signal is determined by a characteric nonequidistance parameter.[29,30]

The first light pulse incident on the boundary $z=0$ of the resonant medium at the instant of time $t=0$ produces in the atom a nonzero dipole moment that oscillates at the frequency Ω of the exciting pulse. The second light pulse, incident on the boundary between the resonant medium at the instant $t=\tau_1+\delta$, will act on the coherence created by the first pulse, so that the quantity σ_{bc} which determines the coherent response at the combination frequency $\omega-\Omega$, will satisfy Eq. (1.4)

$$\dot\sigma_{cb}+i\varepsilon_{cb}\sigma_{cb}=\frac{i}{2\hbar}(\mathbf{E}_\omega\mathbf{d})_{ca}\sigma_{ab}. \tag{3.1}$$

The solution of the last equation is similar to Eq. (1.10):

$$\sigma_{cb}(t_2+\delta_2)=\frac{i}{2\hbar}\sigma_{ab}(t_2)\exp(-i\varepsilon_{\omega-\Omega}\delta_2)\,(\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)\mathbf{d}_{ca}), \tag{3.2}$$

where $\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)$ is the Fourier transform of the time envelope of the second-pulse amplitude

$$\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)=\int_0^{\delta_2}d\tau\,\mathbf{E}_\omega(\tau)\exp\{i(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)\tau\}. \tag{3.3}$$

The resultant expression for σ_{cb} is

$$\sigma_{cb}(t)=\frac{d_{ca}d_{ab}}{4\hbar^2}\mathscr{E}_\Omega^*(\varepsilon_\Omega)\mathscr{E}_\omega(\varepsilon_{\omega-\Omega}+\varepsilon_\Omega)\times\exp\{-i\varepsilon_{\omega-\Omega}(t-t_2-2\delta_2)+i\varepsilon_\Omega(\delta_1+\tau)\}. \tag{3.4}$$

This expression must be averaged over all possible detunings ε_Ω and $\varepsilon_{\omega-\Omega}$. It must be borne in mind here that the mechanisms that broaden the resonant energy levels may be either different or the same (just as in the case of the Stark shift in the electric field of a crystal or else the Doppler shift in a gas). In the former case the procedure of averaging over the detunings $\varepsilon_{\omega-\Omega}$ and ε_Ω is statistically independent. This means physically that no echo signal is produced, and the second exciting pulse is followed only by a free-induction signal. In the second case, the detunings $\varepsilon_{\omega-\Omega}$ and ε_Ω should be functionally related. Assume that this relation is of the form

$$\omega_{ca}-\omega_{ca}{}^0=\varkappa(\omega_{ba}-\omega_{ba}{}^0),$$

where ω_{ca}^0 and ω_{ba}^0 are the frequencies corresponding to the "centers" of the spectral line, and $\varkappa$ is a coefficient that characterizes the line widths. Then,

$$\varepsilon_{\omega-\Omega}=\Delta\omega+(\varkappa-1)\varepsilon_\Omega, \tag{3.5}$$

where

$$\Delta\omega=(\omega_{ca}{}^0-\omega)-\varkappa(\omega_{ba}{}^0-\Omega). \tag{3.6}$$

From (3.4) we obtain for the atom's mean dipole moment responsible for the formation of the PE signal

$$\langle d_{\omega-\Omega}(t)\rangle = \mathrm{Re}\left\{\frac{d_{ca}d_{ab}d_{bc}}{\hbar^2}\pi\exp\left[-i(\omega-\Omega)t -i\Delta\omega\left(t-\frac{z}{v}-\delta_1-\tau\right)\right]F_k(t)\right\}, \tag{3.7}$$

where

$$F_k(t)=\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\mathscr{E}_1{}^*(\varepsilon)\mathscr{E}_2(\Delta\omega+\varkappa\varepsilon) \times\exp\left\{-i\varepsilon\left[\left(t-\frac{z}{v}\right)(\varkappa-1)-\varkappa(\tau+\delta_1)\right]\right\}. \tag{3.8}$$

Taking the inverse Fourier transform of (3.8), we readily obtain

$$F_k(t)=\int d\xi\, d\xi'\, G(\xi)E_2(\xi')E_1{}^* \times[\varkappa\xi'+\xi-(\varkappa-1)(t-z/v)+\varkappa(\tau+\delta_1)]e^{i\Delta\omega\xi'}. \tag{3.9}$$

We consider now different cases of (3.9). In the limit of strong first and second pulses it follows from (3.9) that

$$F_k(t)=cG[(\varkappa-1)(t-t_k)], \tag{3.10}$$

where

$$t_k=z/v+\tau+\delta_1+\delta_2+\tau/(\varkappa-1).$$

It can be seen from (3.10) that the combined-PE signal is a maximum at the instant $t=t_k$, the temporal width of the coherent response is $T_2^*/(\varkappa-1)$, and the polarization in the medium takes place at the frequency $\omega_{ca}^0-\omega_{ba}^0=\omega_{cb}^0$. If only the second pulse is strong, we get from (3.9)

$$F_k(t)=c'\int d\xi\, G(\xi)E_1{}^*[\xi+\delta_1-(\varkappa-1)(t-t_k)]. \tag{3.11}$$

If the inequality $(\varkappa-1)\delta_1>T_2^*$ holds, we have

$$F_k(t)=c'g(0)E_1{}^*[\delta_1-(\varkappa-1)(t-t_k)]. \tag{3.12}$$

The combined PE signal duplicates thus the time-reversed waveform of the first exciting light pulse and changes its scale by a factor $(\varkappa-1)$. If only the first light pulse satisfies the strong-field condition, we have

$$F_k(t)=\int d\xi\, G(\xi)E_2\left[\frac{(\varkappa-1)}{\varkappa}\left(t-\frac{z}{v}-\delta_1-\tau\right)-\frac{\tau}{\varkappa}-\frac{\xi}{\varkappa}\right]e^{i\Delta\omega T(\xi)}, \tag{3.13}$$

where

$$T(\xi)=\frac{(\varkappa-1)}{\varkappa}\left(t-\frac{z}{v}-\delta_1-\tau\right)-\frac{\tau}{\varkappa}-\frac{\xi}{\varkappa}.$$

If the inequality $(\varkappa-1)\delta_1>T_2^*$ holds, we get from (3.9)

$$F_k(t)=c''g(0)E_2\left[\frac{(\varkappa-1)}{\varkappa}\left(t-\frac{z}{v}-\delta_1-\tau\right)-\frac{\tau}{\varkappa}\right]. \tag{3.14}$$

The PE signal likewise duplicates in this case the waveform of the coding second light pulse in the forward direction, but with another scale coefficient.

Note that in contrast to gaseous media, where the numerical value of $\varkappa-1$ is determined by a specific Doppler effect or by the frequency ratio ($\varkappa-1=\omega_{cb}^0/\omega_{ba}^0$), in a solid the coefficient $\varkappa-1$ can take on different values (larger or smaller than zero, and even $\varkappa-1=0$). In the particular case $\varkappa=1$ no PE effect is produced at all, since there will be no reversible dephasing processes and the coherent-emission signal from the medium will be the induction signal.

2. Modified stimulated PE signals

In a three-level resonant medium it is possible to obtain modified stimulated-echo signals[29,31,32] which are analogs of the usual stimulated PE. However, the coherence is stored in the medium after the action of the second exciting light pulse of frequency in the population difference of levels a and b (and at the same time in the population of level c) is realized the the action of a third exciting pulse at the frequency ω–Ω. A coherent reponse is produced also at this frequency, in the form of a modified stimulated PE signal. We can then obtain for the mean value of the atom's dipole moment

$$\langle d_{\omega-\Omega}(t)\rangle=\mathrm{Re}\left\{i\frac{|d_{ab}|^2|d_{cb}|^2}{2\hbar^3}\pi\exp\left[-i(\omega-\Omega)t -i\Delta\omega\left(t-\frac{z}{v}-\delta_1-\delta_2-\tau_1-\tau_2\right)\right]F_m(t)\right\}, \tag{3.15}$$

where

$$F_m(t)=\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\mathscr{E}_3(\Delta\omega+(\varkappa-1)\varepsilon)\mathscr{E}_2(\varepsilon)\mathscr{E}_1{}^*(\varepsilon) \times\exp\left[-i\varepsilon(\varkappa-1)\left(t-\frac{z}{v}-\delta_1-\delta_2-\tau_1-\tau_2\right)-i\varepsilon(\delta_1+\tau_1)\right]. \tag{3.16}$$

If all three exciting pulses satisfy the strong-field condition, it follows from (3.16) that

$$F_m(t)=cG[(\varkappa-1)(t-t_m)], \tag{3.17}$$

where $t_m=z/v+\delta_1+\delta_2+\delta_3+\tau_1+\tau_2+\tau_1/(\varkappa-1)$. Just as in the case of combined PE, the onset of the modified stimulated PE and its duration are determined by the parameter $(\varkappa-1)$.

To investigate various limiting cases of the general relation (3.16) for the modified stimulated PE, it is convenient to transform to the temporal representation

$$F_m(t)=\int d\xi\, d\xi'\, d\xi''\, G(\xi)E_1^*(\xi')E_3(\xi'')$$
$$\times E_2[\xi'-\xi-(\varkappa-1)\xi''$$
$$+(\varkappa-1)(t-z/v-\delta_1-\delta_2-\tau_1-\tau_2)-\delta_1-\tau_1]e^{i\Delta\omega\xi''}. \qquad (3.18)$$

If only the third pulse satisfies the strong-field condition, we obtain for the temporal envelope of the modified stimulated-PE signal, accurate to an inessential exponential factor meaning a correction to the frequency $(\exp[-i\Delta\omega(t-t_m+\tau_1/(\varkappa-1))])$,

$$F_m(t)=c'\int d\xi\, d\xi'\, G(\xi)E_1^*(\xi)E_2[\xi'-\xi+(\varkappa-1)(t-t_m)-\delta_1]. \qquad (3.19)$$

If the conditions $\delta_1 > T_2^*$ and $(\varkappa-1)\delta_2 > T_2^*$ are met, this expression simplifies to

$$F_M(t)=c'g(0)\int E_1^*(\xi)E_2[-\delta_1+\xi+(\varkappa-1)(t-t_M)]\delta\xi. \qquad (3.20)$$

If the second pulse is also strong, the modified stimulated PE signal duplicates the waveform of the first pulse[34]:

$$F_M(t)\sim E_1^*[\delta_1-(\varkappa-1)(t-t_M)]. \qquad (3.21)$$

We can similarly obtain also other paired correlations for both the envelopes of the external pulses and the function $G(t)$. Thus, if the second pulse is strong, then

$$F_m(t)=c''\int d\xi\, d\xi'\, G(\xi)E_3(\xi')E_1^*[(\varkappa-1)\xi'$$
$$+\xi-(\varkappa-1)(t-t_m)-(\varkappa-1)\delta_3+\delta_1]e^{i\Delta\omega\xi'}. \qquad (3.22)$$

If, however, δ_3, $(\varkappa-1)\delta_2 > T_2^*$, we have

$$F_m(t)=c''g(0)\int d\xi\, E_1^*[(\varkappa-1)\xi-(\varkappa-1)(t-t_m)$$
$$-(\varkappa-1)\delta_3+\delta_1]E_3(\xi). \qquad (3.23)$$

3. Three-level PE

Three-level PE signals, just as those of modified stimulated PE, are formed in a three-level system by three exciting light pulses. In contrast to the modified stimulated PE, however, the third pulse does not act on the population produced by the second pulse. Let us examine the physical picture of formation of a three-level PE in a resonant medium.[30] The first light pulse of frequency Ω produces coherence in the transition $b\to a$. The second exciting light pulse, of frequency Ω, leads to the onset of coherence at the difference frequency $\omega-\Omega$ (combined-echo signals). The third exciting pulse acts again at the frequency ω, and as a result the medium polarization at the frequency of the $c\to b$ transition is again transferred to the $b\to a$ transition. To obtain three-level PE it suffices therefore to consider the action of the third pulse on the coherence responsible for the onset of the combined PE [expression (3.4)]. The total contribution to the average dipole moment is then

$$\langle d_\Omega(t)\rangle=\mathrm{Re}\left\{-i\frac{|d_{ca}|^2|d_{ba}|^2}{2\hbar^3}\pi\right.$$
$$\left.\times\exp[-i\Omega t+i\Delta\omega(\delta_2+\tau_2)]F_T(t)\right\}, \qquad (3.24)$$

where

$$F_T(t)=\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\mathscr{E}_1(\varepsilon)\mathscr{E}_2^*(\Delta\omega+\varkappa\varepsilon)\mathscr{E}_3(\Delta\omega+\varkappa\varepsilon)$$
$$\times\exp[-i\varepsilon(t-z/v-\varkappa\tau_2-\varkappa\delta_2)]. \qquad (3.25)$$

In the temporal representation, the envelope of the polarization of the three-level PE takes the form

$$F_T(t)=\int d\xi\, d\xi'\, d\xi''\, G(\xi)E_1(t-z/v-\varkappa\delta_2-\varkappa\tau_2+\varkappa\xi'-\varkappa\xi''-\xi)$$
$$\times E_2^*(\xi')E_3(\xi'')\exp[-i\Delta\omega(\xi'-\xi'')]. \qquad (3.26)$$

If the amplitudes of all three exciting pulses satisfy the strong-field conditions, we obtain from (3.26)

$$F_T(t)=cG(t-t_T), \qquad (3.27)$$

where $t_T=z/v+\delta_1+\delta_2+\delta_3+\varkappa\tau_2$.

We consider now some particular cases of the general relation (3.26). If the strong pulse is the third, then

$$F_T(t)=c'\int d\xi\, d\xi'\, G(\xi)E_1(\varkappa\xi'-\xi+t-z/v-\varkappa\delta_2-\delta_3-\varkappa\tau_2)$$
$$\times E_2^*(\xi')e^{-i\Delta\omega\xi}. \qquad (3.28)$$

If $\delta_1 > T_2^*$, expression (3.28) takes the simpler form

$$F_T(t)=c'g(0)\int d\xi\, E_1(\varkappa\xi+t-z/v-\varkappa\delta_2-\delta_3-\varkappa\tau_2)E_2^*(\xi')e^{-i\Delta\omega\xi}. \qquad (3.29)$$

If the second and third pulses are simultaneously the strong ones, then

$$F_T(t)=\int d\xi\, G(\xi)E_1(t-z/v-\delta_2-\delta_3-\varkappa\tau_2-\xi) \qquad (3.30)$$

or else at $\delta_1 > T_2^*$

$$F_T(t)=g(0)E_1(t-z/v-\delta_2-\delta_3-\varkappa\tau_2). \qquad (3.31)$$

If, however, the strong pulse is the first, it is easy to deduce from (3.26) that

$$F_T(t)=c''\int d\xi\, d\xi' G(\xi)$$
$$\times E_2^*(\xi')E_3\left(\xi'+\frac{t-z/v-\delta_1-\varkappa\delta_2-\varkappa\tau_2-\xi}{\varkappa}\right). \qquad (3.32)$$

If $\delta_3 > T_2^*$ expression (3.32) takes the form

$$F_T(t)=c''g(0)\int d\xi\, E_2^*(\xi)E_3\left(\xi+\frac{t-z/v-\delta_1}{\varkappa}-\delta_2-\tau_2\right). \qquad (3.33)$$

Note that the considered features of three-level PE signal formation are observed in both solids and gases. On the other hand, if the frequency of the second and third pulses is $\omega-\Omega$, the expression for the three-level PE signal amplitude is changed:

$$F_T(t)=\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\mathscr{E}_1(\varepsilon)\mathscr{E}_2(\Delta\omega+(\varkappa-1)\varepsilon)$$
$$\times\mathscr{E}_3^*(\Delta\omega+(\varkappa-1)\varepsilon)$$
$$\times\exp[-i\varepsilon(t-z/v-\delta_2(1-\varkappa)-\tau_2(1-\varkappa))]. \qquad (3.34)$$

When all three exciting light pulses are strong, the polarization time envelope of the three-level PE signal is given by

$$F_\tau(t)=cG(t-t_m'), \quad (3.35)$$

where $t_m' = z/v + \delta_1 + \delta_2 + \delta_3 + (1-\varkappa)\tau_2$.

In gases, by virtue of the specific energy-level inhomogeneous-broadening mechanism (the Doppler effect), $(1-\varkappa)$ is negative and is equal to $-\omega_{cb}^0/\omega_{ba}^0$. This is why the three-level PE effect cannot be observed in a gas when all vectors are collinear. To observe three-level PE under these conditions, the second and third exciting pulses have wave vectors $\mathbf{k}_2$ and $\mathbf{k}_3$ that are antiparallel to the wave vector $\mathbf{k}_1$ of the first exciting light pulse.[30] Note that in the latter case (when the vectors $\mathbf{k}_2$ and $\mathbf{k}_3$ are anticollinear to $\mathbf{k}_1$), the results (3.34) and (3.35) remain unchanged, and in the case of gases it is necessary to replace τ_2 by $-\tau_2$ in expressions (3.31) and (3.33) for gases. In solids, however, the three-level PE considered above [Eqs. (3.34) and (3.35)] can be realized if the coefficient $\varkappa$ is negative. In particular, the inequality $(1-\varkappa)\tau_2 > \tau_1 + \tau_2$ must be satisfied, hence $\varkappa < -\tau_1/\tau_2$.

CONCLUSION

The considered variety of temporal properties of PE signals is thus due to manifestations of the distinctive Fourier-transforming properties of multilevel resonant systems. From this standpoint, the use of an approximation linear in the field, or else of the small-area approximation, is promising in practice. In this case each atom, which is characterized by a perfectly definite difference between the frequencies of the transition and of the external field, becomes sensitive to the Fourier spectrum of the temporal envelopes of the amplitudes of the acting pulses. Thus, the Fourier spectrum of the coherent response in a resonant medium is the product of the corresponding Fourier components of the pulses by the function that determines the frequency spread of the elementary emitters. As a result, various temporal regimes can be effected in the coherent spontaneous emission signals, depending on the relations between the amplitudes of the applied pulses, their durations, as well as the time that characterizes the processes of reversible relaxation of the polarization. In particular, the temporal form of the coherent response can be described by a function that is the Fourier transform of the distribution function of the frequencies of the elementary emitters. It is possible also to duplicate the temporal form of one of the exciting light pulses (besides the form of the second pulse in the signal of the primary PE signal) either in the forward or in the mirror-reversed time duration. In some cases, the time envelope of the PE signal is described either by a correlation function or by a function that is a convolution either of paired combinations of the amplitudes of the exciting pulses, or of the function that determines the inhomogeneous broadening of the energy levels.

In multilevel systems, the temporal properties of the coherent responses in a resonant medium are similar, in many respects, to the two-level ones. However, both the duplication of the form of the exciting pulses, and the characteristic correlation transformations of the time envelopes are accompanied by scale changes that depend on the specific inhomogeneous-broadening mechanisms.

We note in conclusion that the main relations obtained for different responses of the PE type include not only time dependences but also spatial-synchronism conditions. This can be easily verified by separating in the slowly time-varying amplitudes $E(\mathbf{r},t)$ the phase factors proportional to $\exp[i\varphi(\mathbf{r})]$. Thus, in the particular case when a resonant medium is excited by pulses with planar wave fronts, when $\varphi_i(\mathbf{r}) = \mathbf{k}_i\cdot\mathbf{r}$, the primary PE is characterized by a wave vector $2\mathbf{k}_2-\mathbf{k}_1$, the combined PE by $\mathbf{k} = \mathbf{k}_2 - \mathbf{k}_1$, the stimulated PE (including the verified one) by $\mathbf{k}_c = \mathbf{k}_3 - \mathbf{k}_2 - \mathbf{k}_1$, and finally the considered three-level PE are characterized either $\mathbf{k} = \mathbf{k}_3 - \mathbf{k}_2 + \mathbf{k}_1$ or $\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$.

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