Electric dipole echo in glasses

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A theory is developed of the electric dipole two-pulse echo produced in glasses by the interaction between an oscillating electric field and two-level systems (TLS) that determine the low-temperature properties of glasses. It is indicated that the echo signal differs substantially in glasses and in spin systems. The shape of the echo pulse is studied as a function of the pump-pulse amplitude over a wide range of amplitudes. The time dependence of the echo damping is determined. It is shown that this dependence in many cases follows a power law rather than an exponential law.

1. INTRODUCTION

Many low-temperature properties of glasses are known to be determined by two-level systems (TLS) that exist in them.[1-3] A two-level system is an atom or a group of atoms that move (relative to some generalized coordinate) in a field whose potential relief has the form of two wells separated by a barrier. At low temperature this barrier is overcome by quantum-mechanical tunneling. The energy E of a TLS (the distance between its first two levels) is a random quantity distributed over a rather wide range, with an approximately constant density of states. In such a TLS ensemble an echo appears, after some time, consisting of the response of the system to several short high-frequency pulses of an oscillatory field. Since the TLS interacts in dielectric glasses (which have an electric dipole moment) with both sound and an electric field, the echo can be either of the acoustic (sound) or electric-dipole type (mixed forms of echo are also possible).

We develop here a theory of the electric dipole echo that appears in glasses after the action of two pulses of a high-frequency electric field—known as the two-pulse or spontaneous echo.

Echo (acoustic and electric) in glasses has been the subject of many studies.[4-8] As a rule, echoes in glasses are described there in terms of results obtained for spin echo in paramagnetic salts. There is, however, a fundamental difference between these two physical systems.

In a paramagnetic system the Zeeman splitting is practically the same for all spins. As a rule there is only a small spread in the transition frequencies, due to the inhomogeneous broadening. The form of the spin-echo signal coincides in this case with the Fourier transform of the inhomogeneous line-broadening profile.[9]

The situation in glasses is different. The TLS energy (just like the tunnel parameters of glasses; see below) is uniformly distributed over a wide interval. This is the main reason why the results of spin-echo theory cannot be used directly to describe echo in glasses. This difference in the description is most strongly manifested in the theoretically predicted form of the echo signal. This form is quite complicated and varies with the pump-signal amplitude. This does not occur in paramagnets, where the form of the echo is independent of the amplitude.

The second aspect to which we call attention in the present paper is the character of the damping of the echo signal as the time interval between the pump pulses increases. The main cause of echo damping in glasses at low temperature is spectral diffusion[10-13] due to interaction of the resonant TLS with the surrounding thermal TLS.

It is customarily assumed that the decrease of the echo amplitude with the delay time is exponential.[4-6,12] In the work known to us, however, no attention is paid to the scatter of the tunnel transparencies of resonant TLS. Yet they are what governs the interaction with the surrounding thermal TLS. In particular, symmetric resonant TLS (produced in a symmetric two-well potential) do not interact at all with the environment. The contribution of the echo signal from such TLS is attenuated only because of their interaction with phonons.

Allowance for the spread of the tunnel transparencies, as shown in the present paper, leads to a power-law rather than exponential decrease of the echo signal. This raises the question of explaining the exponential decrease observed in a number of experiments.[4-6,14] This important problem is discussed in the next section.

2. FUNDAMENTAL RELATIONS

In our model for the TLS its Hamiltonian in the "node" representation is

$$H_{TLS}=\frac{1}{2}\begin{pmatrix} \Delta & -\Delta_0 \\ -\Delta_0 & -\Delta \end{pmatrix}, \tag{1}$$

where Δ_0 is the overlap integral due to the possible quantum-mechanical tunnelling between the walls, and Δ is the asymmetry of the two-well potential (the difference between the potential energies at the minimum of each well). The Hamiltonian of the TLS interaction with an external electric field $\mathscr{E} = \mathscr{E}_0 \cos \omega t$ in this representation takes the form

$$H_{int}=\vec{\mathscr{E}}\mathbf{m}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{2}$$

where $\mathbf{m} = (1/2)\partial\Delta/\partial\mathscr{E}$ is the electric dipole moment of the TLS. There is also a TLS–phonon interaction whose Hamiltonian is

$$H_{ph}=\Lambda_{ik}U_{ik}^{ph}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{3}$$

where Λ_{ik} is the tensor of the strain potential of the TLS and $U_{ik}^{\rm ph}$ is the strain tensor at the location of the TLS. After diagonalizing $H_{\rm TLS}$, i.e., changing to the proper representation, we get

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$$H=\frac{1}{2}\begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix}+\frac{1}{2}\begin{pmatrix} \mu & 2\mu' \\ 2\mu' & -\mu \end{pmatrix}\vec{\mathscr{E}} + \Lambda_{ik}U_{ik}^{ph}\begin{pmatrix} \Delta/E & \Delta_0/E \\ \Delta_0/E & -\Delta/E \end{pmatrix}, \tag{4}$$

where $E=(\Delta^2+\Delta_0^2)^{1/2}, \mu = 2\Delta \mathbf{m}/E, \mu' = \Delta_0\mathbf{m}/E$.

The contribution to the macroscopic dipole moment of the system from one TLS is

$$\mathbf{d} = -\operatorname{Sp}\left(\rho\frac{\delta H}{\delta\vec{\mathscr{E}}}\right), \tag{5}$$

where ρ is the density matrix averaged over the phonons (the TLS density matrix). It can be represented in the form

$$\rho=\begin{pmatrix} n & -ife^{i\omega t} \\ if^*e^{-i\omega t} & 1-n \end{pmatrix}, \tag{6}$$

where n is the average population of the TLS upper level and f is the off-diagonal part that differs substantially from zero only for the so-called resonant TLS, for which $E\approx\hbar\omega$. Using (6), we obtain from (5)

$$\mathbf{d}=\frac{1}{2}(1-2n)\mu-2\mu'\operatorname{Im}(fe^{i\omega t}). \tag{7}$$

To obtain the electric dipole moment per unit volume this expression must be summed over all the resonant TLS per unit volume of the glass.

The equation for the density matrix of the resonant TLS is (see, e.g. Ref. 11)

$$\frac{\partial n}{\partial t} = -\gamma(n-n_0)-F\operatorname{Re} f, \tag{8a}$$

$$\frac{\partial f}{\partial t}=i\left[\frac{E(t)}{\hbar}-\omega\right]f-\frac{\gamma}{2}f+\frac{F}{2}(2n-1), \tag{8b}$$

where $n_0=(1+e^{E/T})^{-1}$ is the equilibrium population of the TLS upper level; $F=\Delta_0\mathbf{m}\mathscr{E}_0/\hbar E$ is the Rabi frequency of the resonant TLS; γ is the TLS intrinsic damping due to its interaction with the phonons.

Equations (8) for the density matrix take into account spectral diffusion, which consists of the following. Consider a resonant TLS in which the distance between levels is close to $\hbar\omega$. The TLS interacts with neighboring thermal systems, i.e., with those in which the distance between levels is of the order of T. This interaction is due to elastic stresses and electric fields produced by the thermal TLS. Of importance for our problem is the dynamic part of the interaction, i.e., the part whose value depends on whether the thermal TLS is in the ground state or an excited state. But since the energy of a resonant TLS depends on the strain and on the electric field at its location, the quantum transitions (jumps) of thermal TLS cause the spacing of the resonant TLS levels to fluctuate with time. These fluctuations lead to loss of phase coherence of the wave function of the resonant-TLS wave function, and by the same token to damping of the echo signal. Thus,

$$E(t)=E+\hbar\Delta\omega(t), \tag{9}$$

where $\hbar\Delta\omega(t)$ is the contribution made to the energy of the resonant TLS by its interaction with surrounding thermal TLS.

3. SOLUTION OF EQUATIONS FOR THE DENSITY MATRIX

Let the TLS in glass be acted upon by two pulses of a high-frequency electric field of equal amplitude $\mathscr{E}_0$ and, generally speaking, of different durations τ_1 and τ_2. We denote the time interval between the pulses by τ_{21}.

The solution of the equations for the density matrix is:

a) in the absence of a pump field

$$n(t)=n_0+[n(0)-n_0]e^{-\gamma t}, \tag{10}$$

$$f(t)=f(0)\exp\left[-\frac{\gamma}{2}t+izt+i\int_0^t\Delta\omega(t')dt'\right], \tag{11}$$

where $z=E/\hbar-\omega$;[1)]

b) in the presence of a pump field but in the absence of damping and spectral diffusion

$$n(t)=n(0)-2\left[n(0)-\frac{1}{2}\right]\frac{F^2}{g^2}\sin^2\frac{gt}{2} - \frac{F}{2g}\left[f(0)\left(\sin gt+i\frac{2z}{g}\sin^2\frac{gt}{2}\right)+\text{c.c.}\right], \tag{12}$$

$$f(t)=\frac{F}{g}\left[n(0)-\frac{1}{2}\right]\left(\sin gt+i\frac{2z}{g}\sin^2\frac{gt}{2}\right) + f(0)\left(\cos gt+\frac{F^2}{g^2}\sin^2\frac{gt}{2}+i\frac{z}{g}\sin gt\right)-f^*(0)\frac{F^2}{g^2}\sin^2\frac{gt}{2}, \tag{13}$$

where

$$g=(F^2+z^2)^{1/2}. \tag{14}$$

Neglect of damping and of spectral diffusion during the action of a pump field means that the pulse is so short that the following inequalities hold:

$$\gamma\tau_1,\quad \gamma\tau_2\ll 1;\quad \left|\int_0^{\tau_1}\Delta\omega(t)dt\right|,\quad \left|\int_0^{\tau_2}\Delta\omega(t)dt\right|\ll 1.$$

Assuming that the values at the beginning of the first pulse were

$$n(0)=n_0,\ f(0)=0$$

and using the above equations, at $t=t_2^e$ when the second pulse ends we have

$$\begin{aligned} n(t_2^e)={}&n_0-2\frac{F^2}{g^2}\left(n_0-\frac{1}{2}\right)\sin^2\frac{g\tau_1}{2}\exp(-\gamma\tau_{21}) \\ &-2\frac{F^2}{g^2}\left(n_0-\frac{1}{2}\right)\sin^2\frac{g\tau_2}{2} \\ &+4\frac{F^4}{g^4}\left(n_0-\frac{1}{2}\right)\sin^2\frac{g\tau_1}{2}\sin^2\frac{g\tau_2}{2}\exp(-\gamma\tau_{21}) \\ &-\frac{F^2}{2g^2}\left(n_0-\frac{1}{2}\right)\left[\left(\sin g\tau_2+2i\frac{z}{g}\sin^2\frac{g\tau_2}{2}\right)\right. \\ &\times\left(\sin g\tau_1+2i\frac{z}{g}\sin^2\frac{g\tau_1}{2}\right)\exp\left[-\frac{\gamma}{2}\tau_{21}+iz\tau_{21}\right. \\ &\left.\left.+i\int_0^{\tau_{21}}\Delta\omega(t)dt\right]+\text{c.c.}\right], \end{aligned} \tag{15}$$

$$\begin{aligned} f(t_2^e)={}&\frac{F}{g}\left(n_0-\frac{1}{2}\right)\left(\sin g\tau_2+2i\frac{z}{g}\sin^2\frac{g\tau_2}{2}\right) \\ &-2\frac{F^3}{g^3}\left(n_0-\frac{1}{2}\right)\sin^2\frac{g\tau_1}{2}\left(\sin g\tau_2+2i\frac{z}{g}\sin^2\frac{g\tau_2}{2}\right). \end{aligned}$$

$$\times\exp(-\gamma\tau_{21})+\frac{F}{g}\left(n_0-\frac{1}{2}\right)\left(\sin g\tau_1+2i\frac{z}{g}\sin^2\frac{g\tau_1}{2}\right)$$
$$\times\left(\cos g\tau_2+\frac{F^2}{g^2}\sin^2\frac{g\tau_2}{2}+i\frac{z}{g}\sin g\tau_2\right)\cdot$$
$$\times\exp\left[-\frac{\gamma}{2}\tau_{21}+iz\tau_{21}+i\int_0^{\tau_{21}}\Delta\omega(t')\,dt'\right]$$
$$-\frac{F^3}{g^3}\left(n_0-\frac{1}{2}\right)\sin^2\frac{g\tau_2}{2}\left(\sin g\tau_1-2i\frac{z}{g}\sin^2\frac{g\tau_1}{2}\right)$$
$$\times\exp\left[-\frac{\gamma}{2}\tau_{21}-iz\tau_{21}-i\int_0^{\tau_{21}}\Delta\omega(t')\,dt'\right].\quad(16)$$

It can be seen from (15) and (10) that the population of the upper level of the resonant TLS is an even function of the cosine of the angle between $\mathscr{E}_0$ and **m**. Therefore the first term in expression (7) for **d** makes no contribution to the total dipole moment of the system after averaging over all the orientations of the TLS dipoles **m**, since there is no preferred direction in glass.

4. ECHO SIGNAL

A contribution to the echo is made only by an off-diagonal component of the density matrix, f, and furthermore only by the last term of Eq. (1) for $f(t_2^e)$. The first two terms in this expression, on the other hand, describe the damping of the polarization after the second pulse, the next (third) term describes the polarization damping after the first pulse, and only the last (fourth) determines the echo signal. The contribution to this signal from one resonant TLS is given by

$$\mathbf{d}=-\frac{\Delta_0}{E}\mathbf{m}\,\mathrm{th}\frac{\hbar\omega}{2T}$$
$$\times\ \mathrm{Im}\left\{e^{i\omega t}\frac{F^3}{g^3}\sin^2\frac{g\tau_2}{2}\left(\sin g\tau_1-2i\frac{z}{g}\sin^2\frac{g\tau_1}{2}\right)\right.$$
$$\left.\times\exp\left[-\gamma\tau_{21}+iz(t-2\tau_{21})-i\int_0^{2\tau_{21}}\Delta\omega(t')s(t')\,dt'\right]\right\},\quad(17a)$$

where

$$s(t)=\begin{cases}+1, & 0<t<\tau_{21},\\ -1, & t>\tau_{21}.\end{cases}\quad(17b)$$

We introduce new variables $p=(\Delta_0/E)^2$, E, and θ, the angle between $\mathscr{E}_0$ and **m**. The TLS distribution function with respect to these parameters is

$$\frac{N_0}{2p(1-p)^{1/2}}\frac{\sin\theta}{2},\quad(18)$$

where N_0 is the TLS density of states. The dipole moment per unit volume is

$$\mathbf{P}=\frac{\hbar N_0}{2}\int_{-\infty}^{+\infty}dz\int_0^1\frac{dp}{p(1-p)^{1/2}}\frac{1}{2}\int_{-1}^{+1}d\cos\theta\langle\mathbf{d}\rangle_{c,\xi},\quad(19)$$

where $\langle\cdots\rangle_c,\xi$ denotes respectively averaging over the configurations of the thermal TLS (the subscript c) and the random transitions (jumps) in thermal TLS (subscript ξ) surrounding the given resonant TLS.

Since there are no other preferred directions in glass, it is clear that the dipole moment **P** given by (19) is directed along the applied oscillatory electric field $\mathscr{E}_0$. We have therefore for its value

$$P=\frac{N_0\mathscr{E}_0^3m^4}{2\hbar^2}\mathrm{th}\frac{\hbar\omega}{2T}\mathrm{Im}\left\{e^{i\omega t}\int_0^1dy\,y^4\int_0^1\frac{p\,dp}{(1-p)^{1/2}}\exp(-\gamma\tau_{21})\right.$$
$$\left.\times\left\langle\exp\left[-i\int_0^{2\tau_{21}}\Delta\omega(t')s(t')\,dt'\right]\right\rangle_{c,\xi}I\right\},\quad(20)$$

where $y=\cos\theta, F=m\epsilon_0p^{1/2}y/\hbar$, and I is an integral over z:

$$I=\int_{-\infty}^{+\infty}\frac{dz}{g^3}\sin^2\frac{g\tau_2}{2}\left(\sin g\tau_1-2i\frac{z}{g}\sin^2\frac{g\tau_1}{2}\right)\exp[iz(t-2\tau_{21})].\quad(21)$$

The approximation usually employed in paramagnetic systems is that since the inhomogeneous-broadening line profile is narrow the spread in the spin-splitting frequencies (and correspondingly the detuning z) is assumed to be small compared with the Rabi frequency, $z\ll F$. In this case [see (14)] $g=F\gg z$ and the second term in the parentheses of (21) can be neglected. On the other hand, the distribution function in z is not constant, and it must be inserted as a factor in the integrand of (21). Next, taking outside the integral sign the combination

$$\left(\sin^2\frac{g\tau_2}{2}\sin g\tau_1\right)\Big/g^3,$$

which does not depend on $z(g=f)$, we obtain the known result that the form of the echo signal is determined by the Fourier transform of the inhomogeneous-broadening line profile, and its amplitude is proportional to the product

$$\sin^2\frac{F\tau_2}{2}\sin F\tau_1.$$

This result is used in fact in this form to describe echos in glasses.[4-8]

That this approach is invalid is quite obvious, since the distribution function in z in glasses is a constant and a contribution to the echo signal is made also by TLS with $z\gtrsim F$. Putting $t-2\tau_{21}\equiv t_2$, we have

$$I=I_1(\tau_1)-\frac{1}{2}I_1(\tau_1-\tau_2)-\frac{1}{2}I_1(\tau_1+\tau_2)+I_2(\tau_1)$$
$$-\frac{1}{2}I_2(\tau_1-\tau_2)+I_2(\tau_2)-\frac{1}{2}I_2(\tau_1+\tau_2),\quad(22)$$

where

$$I_1(\tau)=\int_0^\infty dz\frac{\sin[(F^2+z^2)^{1/2}\tau]}{(F^2+z^2)^{3/2}}\cos zt_2\quad(23)$$

is odd in τ and even in t_2, and

$$I_2(\tau)=\int_0^\infty\frac{z\sin zt_2}{(F^2+z^2)^2}\{1-\cos[(F^2+z^2)^{1/2}\tau)\}dz\quad(24)$$

is even in τ and odd in t_2.

As can be seen, a connection exists here between I_2 and I_1:

$$I_2(\tau)=-\int_0^\tau\frac{\partial I_1}{\partial t_2}d\tau.\quad(25)$$

The expression for $I_1(\tau)$ should lead to the form

$$I_1(\tau)=-\frac{\pi}{2}\Theta(\tau^2-t_2^2)\int\limits_{|t_2|}^{\tau}dx(\tau-x)J_0[F(x^2-t_2^2)^{1/2}]$$
$$+\frac{\pi\tau}{2F}\exp(-F|t_2|) \qquad (26)$$

and I_2 can be calculated using (25). It can be shown here that the term outside the integral sign in (26) makes no contribution to (22), since the corresponding terms cancel. Allowing for this (to within these terms) we obtain

$$I_2(\tau)=I_1(\tau)+\tilde{I}_2(\tau),$$

where

$$\tilde{I}_2(\tau)=-\frac{\pi F}{4}\Theta(\tau^2-t_2^2)$$
$$\times\int\limits_{|t_2|}^{\tau}dx(\tau-x)^2\left(\frac{x-|t_2|}{x+|t_2|}\right)^{1/2}J_1[F(x^2-t_2^2)^{1/2}], \qquad (27)$$

and $J_0(x)$ and $J_1(x)$ are Bessel functions of order zero and one, respectively. Here $\Theta(x)$ is the Heaviside unit step function [$\Theta(x)=0$ for $x<0$ and $\Theta(x)=1$ for $x>0$]. It follows from (26), (27), and (22) that an echo signal can exist only within a time $\pm(\tau_1+\tau_2)$ relative to the point $\tau_2=0$ (i.e., $t=2\tau_{21}$). There is no echo signal at any other time. This is, in final analysis, a consequence of the constancy of the TLS density of states in glasses, and differs from the situation in paramagnetic salts. In the latter the echo-signal duration is determined by the frequency spread of the paramagnetic impurities, but in glasses, as shown above, by the total duration of the pump pulses.

5. FORM OF ECHO SIGNAL

Let us investigate the form of the echo signal for times τ_{21} so short that the contribution from relaxation processes connected both with the intrinsic damping of the resonant TLS and with spectral diffusion can be neglected. The form of the echo signal is then given by

$$\Phi=\int\limits_0^1 dyy^4\int\limits_0^1 dp\frac{p}{(1-p)^{1/2}}I. \qquad (28)$$

We consider next the case in which both pump pulses have the same length $\tau_1=\tau_2=\tau$; then

$$I=I_1(\tau)-\frac{1}{2}I_1(2\tau)+2I_2(\tau)-\frac{1}{2}I_2(2\tau). \qquad (29)$$

Expanding the Bessel function in (26) in a series in the external-field amplitude and integrating term by term, we get

$$\langle I_1(\tau)\rangle_{p,y}=\int\limits_0^1 dyy^4\int\limits_0^1\frac{dp\,p}{(1-p)^{1/2}}I_1(\tau)$$
$$=-8\pi\tau^2\Theta(1-\vartheta)\sum_{k=0}^{\infty}(-1)^k\frac{a^{2k}(k+1)^2(k+2)}{(2k+5)!}C_k, \qquad (30)$$

where $a=m\mathscr{E}_0\tau/\hbar$ is the dimensionless amplitude of the pump pulses.

$$C_k=B_k-\frac{(1-\vartheta^2)^{k+1}}{2(k+1)},\qquad \vartheta=\frac{|t_2|}{\tau}. \qquad (31)$$

The coefficient B_k satisfies the recurrence relation

$$B_k=\frac{(1-\vartheta^2)^k}{2k+1}-\frac{2k}{2k+1}\vartheta^2B_{k-1},\qquad B_0=1-\vartheta. \qquad (32)$$

Similarly

$$\langle\tilde{I}_2(\tau)\rangle_{p,y}$$
$$=-8\pi\tau^2a^2\Theta(1-\vartheta)\sum_{k=0}^{\infty}(-1)^k\frac{a^{2k}(k+1)(k+2)^2(k+3)}{(2k+7)!}A_k, \qquad (33)$$

$$A_k=(1-\vartheta^2)^{k+1}\left[\frac{k+1+\vartheta^2}{2(k+1)(k+2)}+\frac{4\vartheta-1+2k(\vartheta-1)}{2(k+1)(2k+3)}\right]$$
$$-B_k\vartheta\left[\frac{\vartheta(\vartheta+2)}{2k+3}+1\right]. \qquad (34)$$

In weak fields with $a\ll 1$ we have

$$\Phi=\frac{\pi}{15}\begin{cases}0, & -2\tau<t_2<-\tau,\\ (\tau+t_2)^2, & -\tau<t_2<0,\\ (2\tau-t_2)^2-3(\tau-t_2)^2, & 0<t_2<\tau,\\ (2\tau-t_2)^2, & \tau<t_2<2\tau.\end{cases} \qquad (35)$$

The form of the echo in this approximation is symmetric about the point $t_2=\tau/2$ (see Fig. 1). The echo amplitude is proportional to a^3.

This symmetry is violated, however, even when second-order terms are taken into account: a nonzero contribution of negative polarity is produced on the interval $-2\tau<t_2<-\tau$, namely,

$$\Phi=-\frac{\pi}{1260}\omega_1^2(2\tau+t_2)^4,\qquad -2\tau<t_2<-\tau, \qquad (36)$$

where $\omega_1=m\mathscr{E}_0/\hbar$ is the Rabi frequency for symmetric TLS. In the intermediate field region the echo-signal form can be obtained only by numerically summing the series (30) and (33). Figure 1 shows how the form of the echo evolves as the pump amplitude is increased, with a complicated oscillatory structure appearing on the echo-signal envelope.

We consider now the case of strong fields, namely $a\gg 1$, i.e. $\omega_1\tau\gg 1$. Making in (28) the change of variables

$$y=\frac{\zeta}{(1-\eta^2)^{1/4}},\qquad p=1-\eta^2 \qquad (37)$$

and integrating with respect to η, we get

$$\Phi=2\int\limits_0^1\zeta^3(1-\zeta^2)^{1/4}I\,d\zeta, \qquad (38)$$

where now $F=\omega_1\zeta$. Substituting (23) in (38), making the change of integration variable $z\to F\sinh z$, and integrating with respect to ζ with the aid of the equation

$$\int\limits_0^1 d\zeta\,\zeta(1-\zeta^2)^{1/2}\sin x\zeta=\frac{\pi}{2}\frac{J_2(x)}{x}, \qquad (39)$$

where $J_2(x)$ is a Bessel function of second order, we get

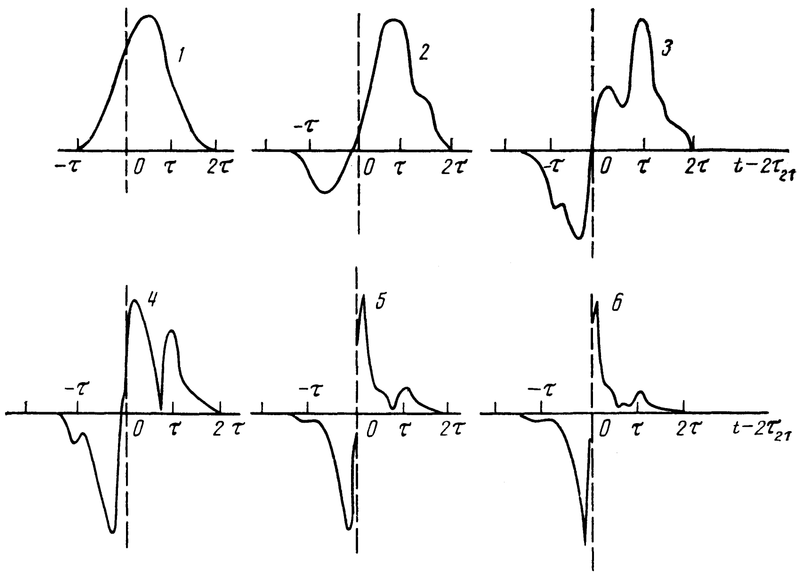

FIG. 1. Normalized envelope of echo pulses for various pump amplitudes a: 1—a = 1, 2—4, 3—7, 4—12, 5—20, 6—25.

$$\int_0^1 \zeta^3(1-\zeta^2)^{1/2} I_1(\tau)\,d\zeta = \frac{\pi}{4\omega_1^2}\int_0^\infty \frac{dz}{\mathrm{ch}^2 z}\,[F^+(z)+F^-(z)], \tag{40}$$

where

$$F^\pm = \frac{J_2(\omega_1\tau\,\mathrm{ch}\,z \pm \omega_1 t_2\,\mathrm{sh}\,z)}{\omega_1\tau\,\mathrm{ch}\,z \pm \omega_1 t_2\,\mathrm{sh}\,z}. \tag{41}$$

Similarly

$$\int_0^1 \zeta^3(1-\zeta^2)^{1/2} I_2(\tau)\,d\zeta = \frac{\pi t_2}{4\omega_1}\int_0^1 d\zeta\,\zeta^2(1-\zeta^2)^{1/2}\exp(-\omega_1\zeta|t_2|) - \frac{\pi}{4\omega_1^2}\int_0^\infty dz\,\frac{\mathrm{sh}\,z}{\mathrm{ch}^3 z}\,[F^+(z)-F^-(z)]. \tag{42}$$

We used Eqs. (40) and (42) to calculate the form of the echo numerically at a field value $a \geqslant 10$, when the alternating series (30) and (33) are difficult to sum numerically. The results of these calculations are also shown in Fig. 1.

An analytic expression for the echo form can be obtained for $a_1 \gg 1$ by evaluating the integrals of the Bessel function by the saddle-point method. This calculation is given in the Appendix. As a result we get

$$\Phi = -\frac{\pi}{\omega_1^4\tau^2}\left(1+\frac{2t_2}{\tau}\right)\cos[\omega_1(\tau^2-t_2^2)^{1/2}] + \frac{\pi}{8\omega_1^4\tau^2}\left(1+\frac{t_2}{2\tau}\right)\cdot$$
$$\times\cos[\omega_1(4\tau^2-t_2^2)^{1/2}] + \frac{3}{4}\frac{\pi t_2}{\omega_1}\int_0^1 dx\,x^2(1-x^2)^{1/2}\exp(-\omega_1|t_2|x)$$
$$\text{for } |t_2| < \tau \tag{43a}$$

and

$$\Phi = \frac{\pi}{8\omega_1^4\tau^2}\left(1+\frac{t_2}{2\tau}\right)\cos[\omega_1(4\tau^2-t_2^2)^{1/2}] + \frac{\pi}{\omega_1^4|t_2|^3}\left(-\frac{t_2}{2}+2\tau+6\frac{\tau^2}{t_2}\right)$$
$$\text{for } \tau \leqslant t_2 < 2\tau. \tag{43b}$$

Examination of these equations convinces readily that for large $\omega_1\tau$ the form of the echo is determined by the last term of (43a). The remaining terms are small and lead to oscillations on the tails of the echo-signal envelope.

At large pump amplitudes the form of the echo pulse thus becomes asymmetric. Its width decreases as a function of ω_1 in inverse proportion to ω_1, and the amplitude, conversely, increases in proportion to ω_1, since it is proportional to $\omega_1^3\Phi$. Figure 2 shows the dependence, plotted on the basis of our numerical calculations, of the echo amplitude on the pump amplitude a.[2)]

6. AREA OF ECHO PULSE

Interest attaches frequently to the area under the echo-signal envelope

$$S = \int_{-\infty}^{+\infty} P\,dt = \pi N_0 m\hbar\,\mathrm{th}\frac{\hbar\omega}{2T}\int_0^1 dy\,y^4\int_0^1 \frac{dp}{[p(1-p)]^{1/2}}\sin^2\frac{F\tau_2}{2}$$
$$\times\sin F\tau_1\exp(-\gamma\tau_{21})\left\langle\exp\left[-i\int_0^{2\tau_{21}}\Delta\omega(t')s(t')\,dt,\right]\right\rangle_{c,\xi}.$$

For $\tau_{21}\to 0$ (small interval between the pulses) and $\tau_1 = \tau_2 = \tau$, leaving out the factor $\pi N_0 m\hbar\tanh(\hbar\omega/2T)$, reduces the expression for the area to the form

$$S = \varphi(\tau) - \frac{1}{2}\varphi(2\tau), \tag{44}$$

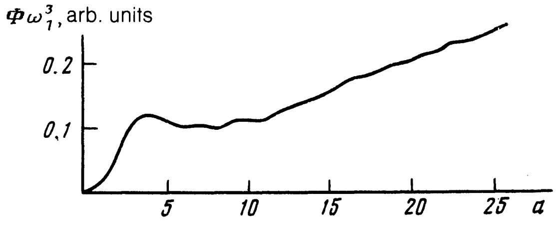

FIG. 2. Dependence of echo amplitude ($\Phi\omega_1^3$) on the pump-pulse amplitude a.

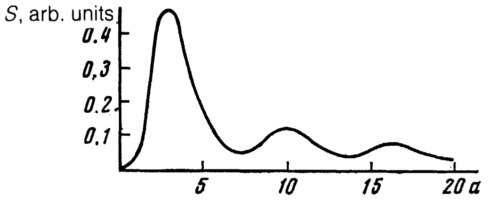

FIG. 3. Dependence of area under the echo signal on the pump amplitude.

where

$$\varphi(\tau)=\int_0^1(1-x^2)^{1/2}\sin(\omega_1\tau x)\,dx=\frac{\pi}{2\omega_1\tau}H_1(\omega_1\tau), \tag{45}$$

and $H_1(z)$ is the Struve function.

For weak fields ($\omega_1\tau\ll1$) we obtain

$$S=\frac{(\omega_1\tau)^3}{15}. \tag{46}$$

For large field amplitudes ($\omega_1\tau\gg1$) we have

$$S=\frac{3}{4\omega_1\tau}\left\{1+\frac{2}{3}\left(\frac{2\pi}{\omega_1\tau}\right)^{1/2}\left[\frac{1}{2^{5/2}}\sin\left(2\omega_1\tau+\frac{\pi}{4}\right)-\sin\left(\omega_1\tau+\frac{\pi}{4}\right)\right]\right\}. \tag{47}$$

Figure 3 shows plots of $S(a)$.

7. SPECTRAL DIFFUSION

It is known that in glasses at low temperatures the decrease of the echo signal as a function of the interval τ_{21} between the pulses is due to spectral diffusion.[10-13] Owing to the interaction of the resonant TLS with the thermal TLS surrounding them, the energy of the former contains time-dependent increment $\hbar\Delta\omega(t)$ defined by Eq. (9). It can be represented in the form[11]

$$\Delta\omega(t)=\sum_l J_l\xi_l(t), \tag{48}$$

where $\hbar J_l$ is the energy of the interaction between the given resonant TLS and the lth thermal one. It is proportional to the product of the factors (Δ/E) of the resonant and thermal TLS and inversely proportional to the cube of the distance between them (Ref. 10):

$$\hbar J_l=(1-p)^{1/2}\left(\frac{\Delta}{E}\right)_l\frac{G_l}{r_l^3}. \tag{49}$$

Here G_l depends on products of the components of the strain-potential tensor Λ_{ik} of the resonant TLS and $\Lambda_{ik}{}^{(l)}$ of the thermal TLS, and also on the direction of the vector $\mathbf{r}_l$ relative to the principal axes of the tensors Λ_{ik} (Ref. 10):

$$G_l=\frac{1}{4\pi\rho}\Big\{\left(\frac{1}{W_t^2}-\frac{1}{W_l^2}\right)[\Lambda_{ii}\Lambda_{kk}^{(l)}-3(\Lambda_{ii}\Lambda_{kn}^{(l)}+\Lambda_{kn}\Lambda_{ii}^{(l)})e_ke_n+15\Lambda_{ik}\Lambda_{mn}^{(l)}e_ie_ke_me_n]-6\left(\frac{1}{W_t^2}-\frac{2}{W_l^2}\right)\Lambda_{ik}\Lambda_{in}^{(l)}e_ke_n-\frac{2}{W_l^2}\Lambda_{ik}\Lambda_{ik}^{(l)}\Big\} \tag{50}$$

where we have set $\mathbf{e}=\ =\mathbf{r}_l/r_l$, φ is the density of the glass, and W_t and W_l are the transverse and longitudinal sound velocities in the glass.

It would also be possible to calculate similarly the contribution from the electric (dipole–dipole) interaction. It depends on the distance r_l in the same manner as (49). Recognizing, however, that for a number of glasses, such as fused quartz, the characteristic value of Λ is about 1 eV and the characteristic values of the TLS dipole moments are about 0.5 D, it turns out that the elastic contribution is somewhat larger for them than the dipole–dipole contribution. For simplicity we accordingly take into account only the elastic contribution.

The function $\xi_l(t)$ describes a random telegraph process.[11] It takes on values ±1 at random instants of time, with the average frequencies of the downward (from $+1$ to -1) and upward jumps equal to Γ_l^+ and Γ_l^-, respectively. They are equal respectively to the reciprocal of the lifetime of the thermal TLS in the upper and lower states. Since only the thermal TLS, i.e., those with energies $E\approx T$, execute transitions, we have $\Gamma_l^+\approx\Gamma_l^-$ for them. In the present paper we assume for simplicity that these two quantities are equal and denote them by Γ_l. This approximation leads only to a certain inaccuracy in the calculation of the numerical coefficient in the argument of the exponential in (55), which will appear in front of the result $\langle|G_l|\rangle$ of averaging $|G_l|$ over the directions.[3)] The present lack of the necessary information on the probability distribution of the strain potential in glasses justifies this simplification.

The dependence of the echo-signal amplitude on the time τ_{21} is determined by the values, averaged over the time (i.e., over the telegraph processes ξ_l) and over the configuration of the thermal TLS, of the quantity

$$K(\tau_{21})=\exp\left[-i\int_0^{2\tau_{21}}\Delta\omega(t')s(t')\,dt'\right], \tag{51}$$

where $s(t)$ is the step function (17b). Owing to the contributions to $\Delta\omega(t)$ [Eq. (48)] from the different thermal TLS and to the absence (which we assume) of a correlation between the different telegraph processes $\xi_l(t)$, the time averaging can be carried out independently for each of the thermal TLS. We denote this average by

$$k(\tau_{12})=\left\langle\exp\left[-iJ\int_0^{2\tau_{21}}\xi(t')s(t')\,dt'\right]\right\rangle_\xi. \tag{52}$$

For simplicity we have left out here the subscript l from $k(\tau_{21})$, J, and ξ.

The function $k(\tau)$ can be calculated by the method described in Klyatskin's book.[15] These calculations were actually carried out in Ref. 11, and $k(\tau)$ can be obtained from Eq. (3.18) of that paper, by putting $\tau'=0$ in it. As a result we have

$$k(\tau)=e^{-2\Gamma\tau}\Big\{\Big[\operatorname{ch}[(\Gamma^2-J^2)^{1/2}\tau]+\frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\operatorname{sh}[(\Gamma^2-J^2)^{1/2}\tau]\Big]^2+\frac{J^2}{\Gamma^2-J^2}\operatorname{sh}^2[(\Gamma^2-J^2)^{1/2}\tau]\Big\}. \tag{53}$$

The average over the telegraph processes (51) is expressed in terms of products of the mean values (52) of each of the thermal TLS separately. Our task is to average the resulting expression over all the configurations and parameters of the thermal TLS.

The influence of a thermal TLS on a resonant one depends on the distance r_l, on the angles that determine the orientation of r_l relative to the principal axes of the tensors Λ_{ik} and $\Lambda_{ik}^{(l)}$, and on the frequency of the jumps Γ_l. Therefore the configuration averaging reduces to averaging over these quantities. As to averaging over $\mathbf{r}$, we assume that all the positions of the thermal TLS in space are equally probable. The distribution in Γ (i.e., physically, in the tunnel transparency of the barrier) takes the form $1/\Gamma(1-\Gamma/\Gamma_0)^{1/2}$ (Ref. 3), where Γ_0 is the characteristic jump frequency of the TLS for which we have $E \approx T$. In order of magnitude we have

$$\Gamma_0 \simeq \frac{\Lambda^2 T^3}{\rho \hbar^4 W^5}, \qquad (54)$$

where Λ is the characteristic value of the strain-potential tensor components $\Lambda_{ik}^{(l)}$, and W is the average speed of sound.

Using the Holtsmark method,[16] we obtain

$$\langle k(\tau)\rangle_c = \exp\left[-(1-p)^{1/2}\frac{S(\tau,0)}{\tau_d}\right], \qquad (55)$$

where (we use now the notation of Ref. 11)

$$S(\tau,0)=\int_0^\infty \frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-k(\tau)], \qquad (56)$$

and

$$\frac{\hbar}{\tau_d}=\frac{4\pi}{3}N_0 T\langle|G|\rangle_\Omega \approx \frac{\Lambda^2 N_0}{\rho W^2}T. \qquad (57)$$

The quantity $|G_l|$ [see Eq. (50)] is averaged in (57) over the orientations of the vector $\mathbf{r}_l/r_l$. In contrast to Ref. 11, it is now important to take into account in (55) the asymmetry factor $(1-p)^{1/2}$ of the resonant-TLS two-well potential, since in a number of cases it determines the law governing the decrease of the echo amplitude with increase of τ_{12}.

As follows from Ref. 11, the behavior of the function $S(\tau,0)/\tau$ depends only on the product $\Gamma_0\tau$, with

$$S(\tau,0)=\tau\begin{cases}\pi\Gamma_0\tau, & \Gamma_0\tau\ll 1,\\ S(0), & \Gamma_0\tau\gg 1,\end{cases} \qquad (58)$$

where $S(0)$ is a constant of order unity introduced in Ref. 11.

8. DAMPING OF ECHO SIGNAL

The law governing the decrease of the echo amplitude with time is different at temperatures that are low and high relative to the temperature

$$T_d \approx (N_0\hbar^3 W^3)^{1/2} \approx 0.1-1\ \mathrm{K},$$

determined from the condition $\Gamma_0\tau_d = 1$.

In the low-temperature region $T \ll T_d$ ($\Gamma_0\tau_d \ll 1$), where most experiments on echo in glasses are performed as a rule,[4-6,14] the value of $S(\tau,0)/\tau_d$ in the argument of the exponential (55) becomes comparable with unity at

$$\tau \approx \tau_\varphi \equiv (\tau_d/\Gamma_0)^{1/2} \ll 1/\Gamma_0, \qquad (59)$$

where

$$\tau_\varphi \sim (\hbar^5\rho^3 W^7/N_0\Lambda^4)^{1/2} T^{-2}$$

is the time over which the wave function of the resonant TLS loses phase coherence by spectral diffusion.[11] Over times τ of this order (but not longer than Γ_0^{-1}), as follows from (58), we have

$$\frac{S(\tau,0)}{\tau_d}=\pi\frac{\tau^2}{\tau_\varphi^2}. \qquad (60)$$

It is easiest to determine the law of decrease of the echo amplitude as a function of the time τ_{21} in the region of low amplitudes $a \ll 1$. We confine ourselves here to this case. Since we have $a \ll 1$, the quantity I in (20) is independent of F, hence also of the tunnel transparency p. The dependence of the echo amplitude on the time interval τ_{21} is determined by the following integral:

$$A=\int_0^1 \frac{dp\,p}{(1-p)^{1/2}}\exp\left[-p\frac{\tau_{21}}{\tau_\omega}-(1-p)^{1/2}\frac{S(\tau_{21},0)}{\tau_d}\right]. \qquad (61)$$

In (61) we have taken into account $\gamma = p/\tau_\omega$, where τ_ω is the minimum relaxation time of a resonant TLS with distance $E=\hbar\omega$ between levels.

For $\tau_\omega \gg \Gamma_0$, the decrease of the echo signal begins at delays $\tau_{21} > \tau_\varphi$, and in the interval $\tau_\varphi < \tau_{21} < \Gamma_0^{-1}$ follows the power law

$$A \propto \frac{1}{\tau_{21}^2}. \qquad (62)$$

In this case $S(\tau,0)$ is determined by Eq. (60), and the main contribution to the integral (61) is made by p close to unity, i.e., almost symmetrical resonant TLS. When τ_{21} increases in the interval $\Gamma_0 < \tau_{21} < \tau_\omega$ we have $S(\tau_{21},0) = \tau_{21}S(0)$ and (62) gives way to a smoother dependence

$$A \propto \frac{1}{\tau_{21}}. \qquad (63)$$

Finally, at $\tau_{21} > \tau_\omega$, the power-law decrease changes to exponential:

$$A \propto \frac{1}{\tau_{21}}\exp\left(-\frac{\tau_{21}}{\tau_\omega}\right). \qquad (64)$$

For $\tau_\varphi \ll \tau_\omega \ll \Gamma_0^{-1}$, the decrease of the echo signal begins as before at delays $\tau_{21} > \tau_\varphi$, and is given by (62) in the interval $\tau_\omega < \tau_{21} < \tau_\omega$. As τ_{21} increases this variation becomes exponential:

$$A \propto \frac{1}{\tau_{21}^2}\exp\left(-\frac{\tau_{21}}{\tau_\omega}\right), \qquad (65)$$

which holds in the interval $\tau_\omega < \tau_{21} < \Gamma_0^{-1}$. Finally, for $\tau_{21} > \Gamma_0^{-1}$ the decrease is given by (4).

For $\tau_d \ll \tau_\omega \ll \tau_\varphi$ the decrease of the echo amplitude begins with times $\tau_{21} > \tau_\omega$ and in the interval $\tau_\omega < \tau_{21} < \tau_\varphi$ has the power-law variation (62). The main contribution to the integral (61) is made in this case by small p, i.e., by strongly asymmetric resonant TLS. In the interval $\tau_\varphi < \tau_{21} < \tau_\varphi^2/\tau_\omega$ this variation becomes exponential

$$A \propto \frac{1}{\tau_{21}^2}\exp\left(-\frac{\pi\tau_{21}^2}{\tau_\varphi^2}\right), \qquad (66)$$

and for $\tau_\varphi^2/\tau_\omega < \tau_2 < \Gamma_0^{-1}$ it takes the form (65). The main contribution is now made by values of p close to unity. Ultimately, for $\tau_{21} > \Gamma_0^{-1}$ we again obtain the decrease (64).

Finally, if the case $\tau_\omega \ll \tau_d \ll \tau_\varphi$ is realized, the decrease (62) holds for $\tau_\omega < \tau_{21} < \tau_\varphi$, Eq. (66) for $\tau_\omega < \tau_{21} < \Gamma_0^{-1}$, while for $\tau_{21} > \Gamma_0$ we get

$$A \propto \frac{1}{\tau_{21}^2} \exp\left(-\frac{\tau_{21}}{\tau_d}\right). \tag{67}$$

We consider now the case of high temperatures $T \gg T_d$ ($\Gamma_0 \tau_d \gg 1$). In this case the quantity $S(\tau,0)/\tau_d$ in the argument of the exponential (55) becomes comparable with unity at $\tau \approx \tau_d \gg 1/\Gamma_0$. Hence for $\tau_\omega \gg \tau_d$ the decrease of the echo signal begins at times $\tau_{21} > \tau_d$, following the power law (63) for $\tau_d < \tau_{21} < \tau_\omega$ (a contribution to (61) is made by values of p close to unity), and the exponential law (64) for $\tau_\omega \ll \tau_{21}$. If, however, $\tau_\omega \ll \tau_d$ holds, the decrease begins at $\tau_\omega < \tau_{21}$, while for $\tau_\omega < \tau_{21} < \tau_d$ we obtain the decrease (62), and the main contribution is made by asymmetric resonant TLS with $p \ll 1$, while for $\tau_{21} > \tau_d$ the following law is valid:

$$A \propto \frac{1}{\tau_{21}^2} \exp\left(-\frac{\tau_{21} S(0)}{\tau}\right). \tag{68}$$

It follows from the picture we have presented that owing to the distribution of the tunnel transparencies of the resonant TLS, the initial section of the echo-signal decrease is practically always not exponential but is a power-law variation, and only for longer times τ_{21} does the damping become exponential. This raises the question of the cause of the exponential dependence, observed in some experiments,[4-6,14] of the echo signal amplitude A on the delay time τ_{21}.

We wish to point out situations in which the echo-signal damping is exponential. First is the case when there is no spread in the tunnel transparencies, i.e., the values of the parameter p, which then has only one characteristic value. A probable example of such a situation may be TLS produced by OH impurity groups in amorphous SiO_2.

A second situation is one in which a combination of two damping mechanisms "operate," for example spectral diffusion and the intrinsic damping due to interaction with phonons.

Finally, one more explanation of the observed experiments may be the most realistic. Its essential idea is that besides the interaction of type $S_z - S_z$ between TLS pseudospins, which we have been considering here, a role can be played also by interactions of the form $S_z - S_x$ and $S_x - S_x$ (Ref. 17). The latter, insofar as can be judged from experimental data, play a lesser role in relaxation processes. Thus during the initial stage the damping of the echo pulses should obey the theory developed in the present paper. It then acquires an exponential character and is determined by these unaccounted-for interactions.

APPENDIX

The integral (4) is given by (the factor $\pi/8\omega_1^2$ is omitted)

$$Y = \int_{-\infty}^{+\infty} \frac{dz}{\mathrm{ch}^2 z} [F^+(z) + F^-(z)]. \tag{A1}$$

We introduce the variables $\alpha = \omega_1 \tau$ and $\beta = \omega_1 |t_2|$ and denote the integral of $F^\pm$ by $Y^\pm$. We have then $Y = Y^+ + Y^{-1}$, where

$$Y^\pm = \int_{-\infty}^{+\infty} \frac{dz}{\mathrm{ch}^2 z} \frac{J_2(\alpha\,\mathrm{ch}\,z \pm \beta\,\mathrm{sh}\,z)}{\alpha\,\mathrm{ch}\,z \pm \beta\,\mathrm{sh}\,z}. \tag{A2}$$

For the case $\beta > \alpha$, using (26), we can obtain

$$Y = 4\omega_1 \tau \int_0^1 \zeta^2 (1-\zeta^2)^{1/2} \exp(-\omega_1 |t_2| \zeta)\, d\zeta. \tag{A3}$$

For $|t_2| > \tau$ integration yields $8\tau/\omega_1^2 |t_2|^3$.

We have thus to calculate Y^+ and Y^- under the condition $\alpha > \beta$. Substituting the asymptotic expression for the Bessel function

$$J_2(x) = -\left(\frac{2}{\pi x}\right)^{1/2} \cos\left(x - \frac{\pi}{4}\right)$$

in the integral Y^+, we obtain

$$Y^+ = -\left(\frac{2}{\pi}\right)^{1/2} \mathrm{Re} \exp\left(-i\frac{\pi}{4}\right) \times \int_{-\infty}^{+\infty} \frac{dz}{\mathrm{ch}^2 z} \frac{1}{(\alpha\,\mathrm{ch}\,z + \beta\,\mathrm{sh}\,z)^{3/2}} \exp[if(z)], \tag{A4}$$

where $f(z) = \alpha \cosh z + \beta \sinh z$.

At the stationary point z_0 we have

$$f'(z_0) = 0, \quad \mathrm{th}\,z_0 = -\frac{\beta}{\alpha}, \quad f(z_0) = (\alpha^2 - \beta^2)^{1/2},$$

$$f''(z_0) = (\alpha^2 - \beta^2)^{1/2}, \quad \mathrm{ch}\,z_0 = \frac{\alpha}{(\alpha^2-\beta^2)^{1/2}}, \quad \mathrm{sh}\,z_0 = \frac{-\beta}{(\alpha^2-\beta^2)^{1/2}}.$$

Expanding $f(z)$ in the vicinity of the point z_0 and calculating the integral, we obtain

$$Y^+ = -\frac{2}{\omega_1^2 \tau^2} \cos[\omega_1 (\tau^2 - t_2^2)^{1/2}]. \tag{A5}$$

One can show analogously that

$$Y^- = Y^+, \quad Y = 2Y^+.$$

With allowance for the factor $\pi/8\omega_1^2$ in (40) we find that

$$2\int_0^1 \zeta^3 (1-\zeta^2)^{1/2} I_1(\tau)\, d\zeta = -\frac{\pi}{\omega_1^4 \tau^2} \cos[\omega_1 (\tau^2 - t_2^2)^{1/2}]. \tag{A6}$$

The second term of (42) yields

$$\frac{\pi}{\omega_1^4 \tau^2} \frac{|t_2|}{\tau} \cos[\omega_1 (\tau^2 - t_2^2)^{1/2}], \quad |t_2| < \tau. \tag{A7}$$

In the case $\tau < |t_2|$ we have for (42)

$$\int_0^1 \zeta^3 (1-\zeta^2)^{1/2} I_2(\tau)\, d\zeta = \frac{\pi}{4} \tau^2 \int_0^1 \zeta^3 (1-\zeta^2)^{1/2} \exp(-\omega_1 |t_2| \zeta)\, d\zeta \cdot \mathrm{sign}\, t_2. \tag{A8}$$

For $\omega_1 |t_2| > \omega_1 \tau$ the integral (A8) is equal to

$(\operatorname{sign} t_2)\cdot 3\pi\tau^2/\omega_1^4|t_2|^4$.

We have used here the fact that for $\tau > |t_2|$

$$I_2(\tau)=\frac{\pi}{4}\tau^2\exp(-F|t_2|)\cdot\operatorname{sign} t_2. \qquad \text{(A9)}$$

As a result, designating the integrals of $I_1(\tau)$ and $I_2(\tau)$ respectively by $\Phi_1(t)$ and $\Phi_2(t)$, we arrive at the expressions

$$\Phi_1(\tau)=\begin{cases} -\dfrac{\pi}{\omega_1^4\tau^2}\cos[\omega_1(\tau^2-t_2^2)^{1/2}], & |t_2|<\tau, \\ \dfrac{2\pi\tau}{\omega_1^4|t_2|^3} & |t_2|>\tau, \end{cases} \qquad \text{(A10)}$$

$$\Phi_2(\tau)=\begin{cases} \dfrac{\pi t_2}{2\omega_1}\displaystyle\int_0^1 dx\, x^2(1-x^2)^{1/2}\exp(-\omega_1|t_2|x) \\ \qquad -\dfrac{\pi t_2}{\omega_1^4\tau^3}\cos[\omega_1(\tau^2-t_2^2)^{1/2}], & |t_2|<\tau, \\ \dfrac{3\pi\tau^2}{\omega_1^4|t_2|^3 t_2}, & |t_2|>\tau. \end{cases} \qquad \text{(A11)}$$

The shape of the echo is determined in accordance with (29), by the expression

$$\Phi=\Phi_1(\tau)-\frac{1}{2}\Phi_1(2\tau)+2\Phi_2(\tau)-\frac{1}{2}\Phi_2(2\tau).$$

Therefore, using $(A>10)$ and (A11), we arrive at Eqs. (43a, b).

[1)] It must be borne in mind that the presence of the frequency ω in (11) is due only to representation of the nondiagonal density matrix in the form (8), i.e. separation of the factors $e^{\pm i\omega t}$. Actually, of course in the absence of the pump, ω does not enter in the expression for the density matrix.

[2)] When the echo signal contained several maxima the largest one was chosen to determine the amplitude.

[3)] This coefficient was calculated in Ref. 12.

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