Correlation of the shape of light echo signals with the shape of the excitation pulses

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A correlation effect of the shape of the primary (PLE) and stimulated (SLE) light echo signals with the shape of the excitation laser pulses has been found in a ruby crystal. It is established that the shape of the PLE is inverted in time compared with the shape of the first low-intensity pulse, while the shape of the SLE duplicates the shape of the second low-intensity pulses.

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The unique property of light echo signals lies in their capability of being emitted at specified time intervals after the action on the resonance medium of laser pulses that are separated from each other.[1] In a number of crystals these time intervals can be appreciable (for example, at a temperature of 2.2 K in ruby they amount to tens of microseconds,[2] and in the $LaF_3:Pr^{3+}$ crystal they even amount to a few minutes[3]). Therefore the signals of the primary and stimulated light echo can be used to make optical delay lines if the shape of the echo signals duplicates the shape of the given excitation pulses.

In this paper we report the discovery of a correlation effect between the shape of the PLE and SLE signals and the shape of the excitation pulses, and we investigate the

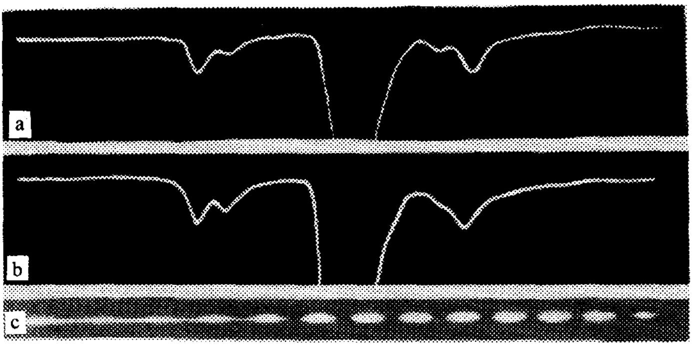

FIG. 1. Oscillograms, illustrating correlation effect of the shape of the primary light echo with the shape of the first laser pulse. The SLE signal is the first at the right; the rest of the signals are excitation pulses. The upper oscillogram (demonstrating the presence of correlation) corresponds to a power ratio of 10 for the first and second pulses, and oscillogram "b" (demonstrating partial breakdown of correlation) corresponds to a power ratio of 2. The power of the second pulse is 300 kW. The value (H_0) of the applied constant magnetic field is equal to 450 Oe. The time interval between pulses is equal to 24 nsec. The markers ("c") represent 10 nsec.

conditions for realizing it. A ruby crystal with a Cr^{3+} ion concentration of 0.05 wt.% was chosen as the resonance medium [working transition: ${}^4A_2$–${}^2E(\bar{E})$].

A set of oscillograms, demonstrating the correlation of the shape of the PLE and SLE with the shape of the excitation pulses, is shown in Fig. 1 and Fig. 2. Following the terminology of flash radiospectroscopy,[4] we shall call the pulse that is to be reproduced the code, and the next pulse the reading pulse. For three-pulse excitation; the first pulse is the triggering pulse. This experiment showed that the shape of the PLE and SLE signals correlates with the shape of the code pulse only if the electric field intensity of the latter is less than the inhomogeneity of the local electric field at the location of the paramagnetic centers, and if the field intensity of the reading and triggering pulses is considerably greater than it. It was established that such a correla-

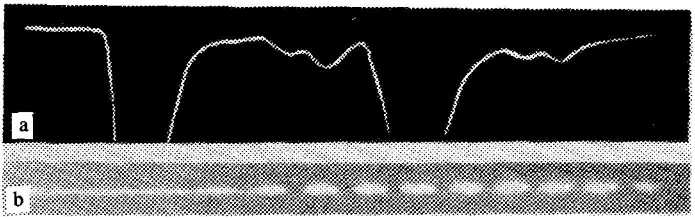

FIG. 2. Oscillogram, illustrating the identical shape of the stimulated light echo signal (first at the right) and the shape of the second excitation signal. the power ratio of the third and second pulses is equal to 10. The power of the third pulse is 200 kW. $H_0 = 450$ Oe. The time between the first and second pulses is equal to 24 nsec, and between the second and third pulses—20 nsec. The markers ("b") represent 10 nsec.

tion is realized when the following conditions are satisfied:

$$W_{(c)}^{1/2}\hbar^{-1}p\left(\frac{8\pi}{cS}\right)^{1/2}T_2^{*-1} << 1;\quad \Delta t_{(c)}^{-1}T_2^{*} << 1; \tag{1}$$

$$W_{(r;t)}^{1/2}\hbar^{-1}p\left(\frac{8}{cS\pi}\right)^{1/2}\Delta t^{-1}\approx 1, \tag{2}$$

where T_2^* is the transverse reversible relaxation time ($\approx 10^{-10}$ sec); p is the modulus of the electric dipole moment of the resonance transition ($\approx 4.8\times 10^{-21}$ CGS units); c is the velocity of light; S is the beam cross-sectional area ($\approx 10^{-2}$ cm^2); W and Δt are the power and duration of the laser pulse (the subscripts "c", "r" and "t" refer to the code pulse, reading and triggering pulses, respectively).

The experimental apparatus, on which these studies were performed, was similar to the apparatus described in Ref. 5. The duration of the excitation pulses was 10 nsec, and the time between pulses could be varied from 5 to 120 nsec. The apparatus had provision for the application of a constant magnetic field (directed along the optical axis of the crystal) with an intensity of up to 1.5 kOe although the PLE and SLE signals were also observed in a zero magnetic field. A unique feature of the excitation involved using a code pulse formed from two laser pulses delayed (by 5–8 nsec) with respect to one another, so that the shape of the code pulse could be varied.

In order to understand the reasons for the existence of shape correlation between the echo signals and the shape of the pulses let us turn to the three-pulse and four-pulse resonance excitation schemes for a two-level system, depicted in Fig. 3. In the three-pulse excitation case (Fig. 3,A), the system is capable of emitting two PLE signals at the times $2\tau_2+\tau_1$ and $2\tau_2+2\tau_1$ in the direction $2\mathbf{k}_{(r)}-\mathbf{k}_{(c)}$ and an SLE signal at the time $2\tau_1+\tau_2$ in the direction $\mathbf{k}_{(r)}$. The form factors of the PLE and SLE signals for the conditions of partial excitation of an inhomogeneously broadened line by a spectrum of rectangular pulses ($\Delta t \gg T_2^*$) have been determined in Ref. 6. If the code pulse is of such low intensity that $W_{(c)}^{1/2}\left[\frac{5\hbar^{-1}p}{(cS)^{1/2}}\right] \ll T_2^{*-1}$, then the form factor of the PLE and SLE turns out to be proportional to $W_{(c)}^{1/2}$. Thus, for these conditions, the ratio of the PLE signal intensities is equal to the ratio of the intensities of the first and second pulses; however, their sequential arrangement in time was the reverse of the order of these pulses. Convergence of the first two pulses leads to a convergence of both PLE signals, so that they form a single echo signal, the shape of which is reversed in time relative to the shape of the code pulse. A numerical analysis leads to a similar result in the case of a code pulse of arbitrary shape.[7] Obviously, if the code pulse had a symmetrical shape, then the PLE signal will also have a symmetrical shape under the stated conditions. Upon increase in τ_1, the intensity of the PLE decreases exponentially with the transverse irreversible relaxation time T_2. We note that if the code pulse and reading pulse change places, then the shape of the PLE signal will duplicate the shape of the code pulse. An analysis of the shape of the SLE signal for the three-pulse excitation conditions leads to a similar result if the code pulse is the second pulse. In the simplest case the problem reduces to the case of four-pulse excitation (Fig. 3,B),

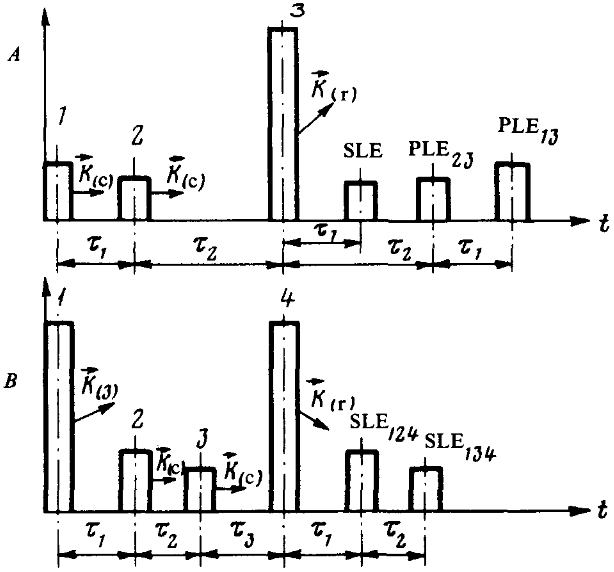

FIG. 3. Three-pulse (*A*) and four-pulse (*B*) excitation schemes of two-level system, explaining the correlation effects of the shape of the echo signals with the shape of the excitation pulses. $\mathbf{K}_{(c)}$, $\mathbf{K}_{(t)}$, $\mathbf{K}_{(r)}$ are the wave vectors of the code, triggering and reading pulses, respectively. PLE$_{\alpha\beta}$ are the PLE signals from the action of the αth and βth excitation pulses. SLE$_{\alpha\beta\gamma}$ are the SLE signals from the action of the αth, βth and γth excitation pulses.

from which it follows that with the convergence of the second and third pulses the total SLE signal will have a shape identical to the shape of the low-intensity code pulse. This result agrees with the conclusions of our numerical analysis of the SLE shape in the case of excitation pulses of arbitrary shape. With an increase in τ_3 the SLE signal decreases exponentially with the longitudinal relaxation time T_1, which can be extremely large.[2,3] From the viewpoint of possible technical applications of the discovered effect, it may be necessary for the polarization of the echo signals to coincide with the polarization of the code pulse. In ruby, for example, for pulses propagating in a direction close to the direction of the optical axis *C*, the following relations are valid: $\psi_{\rm PLE} = 2\psi_2 - \psi_1$ and $\psi_{\rm SLE} = -\psi_1 + \psi_2 + \psi_3$, where ψ_η is the angle of the polarization vector of the ηth linearly polarized pulse with respect to the chosen **X** axis in a plane perpendicular to the **C** axis; $\psi_{\rm PLE}$ and $\psi_{\rm SLE}$ are the angles of the polarization vectors of the PLE and SLE signals, respectively, relative to the **X** axis. For $\psi_\eta = 0$ (for any $\eta;\eta = 1,2,3$) the PLE and SLE signals will have a polarization coinciding with the polarization of the code pulse. In the SLE case the same alignment of polarizations is also realized in the case: $\psi_1 = \psi_3;\psi_{\rm SLE} = \psi_2$. The validity of these polarization rules was verified by us experimentally.

We note that we have also found a correlation of the shape of the echo signals with the shape of the excitation pulses in crystals of $CaWO_4{:}Nd^{3+}$ and $Y_3Al_5O_{12}{:}Nd^{3+}$, indicating the common character of the discovered effect.

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