Measurement of the intraband relaxation time of electrons in a semiconductor

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We estimate the electron relaxation times T_1 and T_2 (energy and quasimomentum relaxation times) in the semiconducting single crystal $CdS_{0.75}Se_{0.25}$ ($T = 130$ °K) near the bottom of the conduction band ($\hbar\omega - E_g \approx 5$ meV), using the effect of self-induced transparency in single-photon excitation by ultrashort pulses of the second harmonic ($\hbar\omega = 2.34$ eV, $\tau \approx 5 \times 10^{-12}$ sec) of a mode-locked neodymium laser.

Several recent theoretical[1–3] and experimental[4–7] papers are devoted to coherent interaction of powerful laser radiation with semiconductors (the phenomenon of self-induced transparency) at a light-pulse duration τ shorter than the polarization relaxation time of the medium, $\tau \ll T_2$, and at $\theta_0 \equiv (\mu/\hbar) \int \mathcal{E}(t)\,dt > \pi$, where $\mathcal{E}(t)$ is the slow amplitude of the light pulse and μ is the dipole matrix element of the transition. In the present study, using a sounding pulse, we measured the relaxation times T_1 and T_2 in the semiconductor $CdS_{0.75}Se_{0.25}$ under coherent excitation of electrons in the conduction band by a high-power ultrashort pulse (USP) of light, which causes self-transparency. The self-induced transparency was the result of coherent excitation of the electron-hole pairs in the case of single-photon interband absorption of USP of the second harmonic

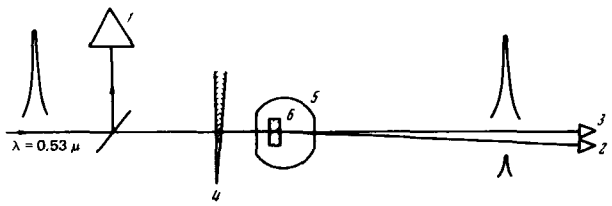

FIG. 1. Experimental setup: 1,2,3—calorimeters to measure the pulse energy entering the crystal, the sounding pulse energy, and the exciting-pulse energy past the crystal, 4—optical delay line, 5—cryostat, 6—$CdS_{0.75}Se_{0.25}$ ($T = 130$ °K).

($\lambda = 0.53\ \mu$, $\Delta\lambda \approx 5$ Å) of a mode-locked neodymium laser. The measurement method consists in the following: A weak sounding pulse is used to investigate the state of the medium (semiconductor) at individual instants of time after the passage of the powerful USP of light. The sounding pulse (a fraction of the sound-harmonic radiation) is produced with the aid of a wedge made up of two flat mirrors (Fig. 1). Such a device makes it possible to regulate smoothly the time delay between the two pulses in a range 1—10 by moving the wedge is moved perpendicular to the incident light, and 10—200 psec by longitudinal displacement of one plate of the wedge. The small angle between the mirrors (~30′) ensures overlap of the beams in the sample and their spatial separation at a sufficiently large distance (~1 m) from the sample. The sounding-pulse power was lower than the self-bleaching threshold of the semiconductor.

Using a calorimeter of high sensitivity (not worse than 10^{-6} J) we measured the energy of the sounding pulse passing through the crystal at different delays Δt relative to the high-power ultrashort light pulse (Fig. 2). The characteristic scatter of the energy values in the case of small delays can apparently be attributed to T_2 relaxation. Owing to the statistical character of the laser radiation, the "area" θ_{10} of the exciting pulse can vary in a wide range, and the medium can either amplify the sounding pulse, $\theta_{10} \approx \pi$, be transparent to it, $\theta_{10} \approx \pi/2$, or else strongly absorbing $\theta_{10} < \pi/2$. One can therefore expect the energy of the sounding light leaving the sample to vary in a wide range at $\Delta t \lesssim T_2$ (the corresponding calculations are given below). At $\Delta t > T_2$, the

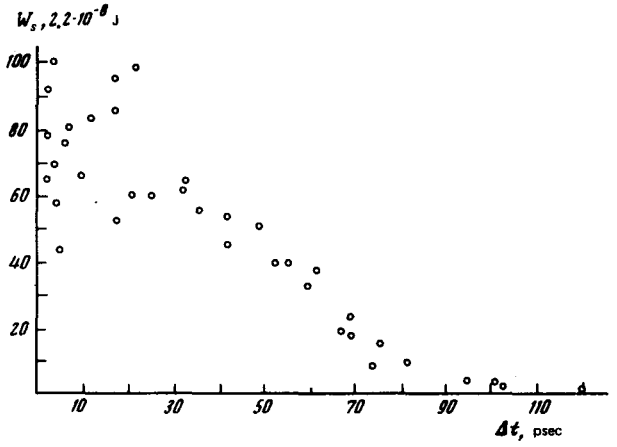

FIG. 2. Dependence of the sounding-pulse energy W_s passing through the crystal on its delay relative to high-power USP.

interaction is no longer coherent and the absorption of the sounding pulse is determined by the process of T_1 relaxation (energy relaxation). In this section of the curve, the scatter of the experimental energy values lies within the limits of the measurement accuracy. The smooth decrease of the curve (the absence of saturation in the semiconductor) can be attributed to the fast (with characteristic time 10^{-11}—10^{-10} sec) process of the binding of the electrons into excitons (see, e.g., [8], in which this time was measured for case crystals by using free-exciton luminescence kinetics). The delay ΔT with which the sharp decrease of the fluctuations of the energy W_s is observed determines the upper bound of the transverse relaxation time ($T_2 \lesssim 20$ psec). From the exponential decrease of W_s at delays $\Delta t > 20$ psec we determined the time $T_1 \approx 60$ psec.[1)]

Within the framework of the model of inhomogeneously broadened resonance line of a medium consisting of indentical two-level atoms, we have analyzed theoretically the dynamics of the propagation of two USP of light. It was assumed that the sounding and the exciting pulses propagate in the same direction. The coherent interaction of the USP was investigated by numerically solving a system of differential equations, some of which are Maxwell's equations and the remaining ones are analogous to the Bloch equations[9]:

$$\frac{\partial e}{\partial z} + Ke + \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} g(x) v_x \, dx = 0,$$

$$\frac{\partial v_x}{\partial \tau} + \gamma v_x + x u_x + n_x e = 0,$$

$$\frac{\partial u_x}{\partial \tau} + \gamma u_x - x v_x = 0,$$

$$\frac{\partial u_x}{\partial \tau} - v_x e = 0.$$

The quantities e, v, u, and n, which enter in the system, are dimensionless and stand for the slow field amplitude, the imaginary and real part of the polarization, and the inverted population. The connection between $e(z, \tau)$ and the true values of the electric field in the medium is determined by the expression $e(z, \tau) = (\mu/\hbar) T_2^* \mathcal{E}(z, t)$, where T_2^* is the time characterizing the inhomogeneous broadening of the energy levels. The coefficients K and γ describe the nonresonant losses in the medium and the polarization relaxation connected with the homogeneous broadening of the energy levels, $g(x)$ is the frequency distribution function of the sources, and $x = \Delta\omega T_2^*$. It is obvious that the solution of the problem within the framework of the model of a resonant medium consisting of identical atoms, with an inhomogeneously broadened transition line, is a very strong idealization. We were therefore interested not in a quantitative comparison of the theoretical calculation with the experimental results but in the feasibility in principle of measuring the time T_2 by the sounding-

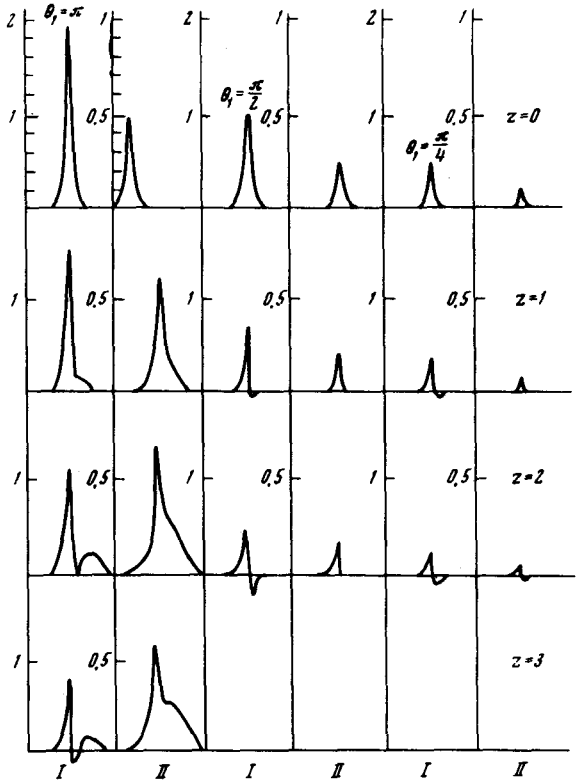

FIG. 3. Dynamics of propagation of two USP in a resonantly absorbing medium at $\theta_{10}=\pi$, $\pi/2$, $\pi/4$ and $z=0,1,2,3$ (I—exciting pulse, II—sounding pulse).

pulse method. We determined the qualitative features of the propagation of a delayed USP as a function of the parameters of the first pulse exciting the medium. Numerical calculations were performed with a computer for $\theta_{10}=\pi$, $\pi/2$, $\pi/4$ and $z=0,1,2,3$. The initial waveform of the pulses entering the medium was a hyperbolic secant. It turned out that the absorption of a weak USP at delays $\Delta t < T_2$ can vary strongly, depending on the value of the parameter θ_{10} for the first pulse (Fig. 3). The reversal of the sign of the amplitude in Fig. 3 corresponds to the case when the phase of the field produced in the medium lags the phase of the external field by π. Since the experiment yields the intensity or the energy of the radiation, the negative spikes manifest themselves as additional pulses. Thus, our calculation confirms qualitatively the feasibility of estimating the relaxation times by the method proposed above.

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[1)]Similar relations were obtained in[7] for the case of resonance excitation of an A-exciton in the same crystal ($T_2 \lesssim 0.15$ nsec, $T_1 \approx 1$ nsec).

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