Plasma resonance of high-density nonequilibrium carriers in GaSe single crystals

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A manifestation of a plasma resonance in the IR optical reflection has been studied at a picosecond time resolution in highly excited GaSe single crystals.

Analysis of the kinetics of the reflection of the beam from a CO_2 laser (10.6 μm) from pulse-excited semiconductors[1] has revealed fundamental opportunities for studying several properties of an electron-hole plasma. The use of picosecond light pulses, in order to generate a dense system of nonequilibrium charge carriers,[2] with densities up to values on the order of 10^{20} cm^{-3}, opens some new opportunities for studying the physics of collective phenomena. In the present study we have analyzed the picosecond kinetics of the reflection and transmission of IR light by highly excited GaSe single crystals as a result, we have observed a plasma resonance at the frequencies of the probing light. We have determined the values of the properties which characterize the recombination of the nonequilibrium carriers.

We studied single-crystal layers of ϵ-GaSe with a thickness of 5–10 μm. The laser oscillator generating the picosecond light pulses was a garnet laser ($h\nu = 1.17$ eV)

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with active-passive mode locking ($\tau = 25$ ps). Selected single pulses (at a repetition frequency of 10 Hz) were amplified by a four-pass amplifier. The samples were excited by light at the second harmonic of the fundamental frequency of the laser; the maximum energy in the pulse was 1 mJ. For probing we used an IR beam ($\lambda = 2.8$ μm) produced through stimulated Raman scattering in a cell filled with gaseous methane, into which part of the fundamental light from the picosecond laser was directed. Both the exciting beam and the probing beam were directed perpendicular to the plane of the sample ($\mathbf{k}\|\mathbf{c}$); the parameters of the Gaussian intensity distribution of the light were 1 and 0.3 mm, respectively. Photodiodes were used to detect the intensities of the pulses reflected from and transmitted through the crystal. A subsequent analysis of the experimental data was carried out with the help of a microcomputer; the experiment itself was controlled by the microcomputer.

Figure 1 shows the intensity of the reflected IR beam versus the time delay of this beam with respect to the exciting pulse for two values of the pump power density in a GaSe sample 10 μm thick. Initially, at a pump energy $\leqslant 385$ μJ, we observe a sharp decrease in the reflection during the exciting pulse; then comes a vastly slower relaxation to the original value (Fig. 1a). As the excitation intensity is raised, the kinetics becomes far more complicated (Fig. 1b). Immediately after a sharp decrease in the reflection comes a sharp increase; at the end of the exciting pulse the reflection decreases again, going through a minimum at a delay of 400 ps and at an excitation energy of 670 μJ. Later on, we again see a slow relaxation to the original value. Note that the interference effects which could in principle be manifested in a sample which constitutes a Fabry-Perot interferometer could not explain the observed behavior. Specifically, oscillations of this type were never observed in the kinetics of the intensity of the transmitted beam (see the inset in Fig. 1b); we can thus also rule out any manifestations of interference in the reflection. The same conclusion can be reached from an analysis of the reflection of thin GaSe layers as a function of the density of excited nonequilibrium carriers, whose presence at such high excitation levels is the primary reason for the modulation of the dielectric constant of the substance, $\tilde{\epsilon}$. In general, the real and imaginary parts of ϵ can be written as functions of the probing frequency ω as follows:

$$\epsilon_R = \epsilon_0 - 4\pi ne^2 \left[\frac{1}{m_e^*} \left\langle \frac{\tau_e^2}{1+\omega^2\tau_e^2} \right\rangle + \frac{1}{m_h^*} \left\langle \frac{\tau_h^2}{1+\omega^2\tau_h^2} \right\rangle \right] + 4\pi\chi,$$
$$\epsilon_I = \frac{4\pi ne^2}{\omega} \left[\frac{1}{m_e^*} \left\langle \frac{\tau_e}{1+\omega^2\tau_e^2} \right\rangle + \frac{1}{m_h^*} \left\langle \frac{\tau_h}{1+\omega^2\tau_h^2} \right\rangle \right] + \frac{4\pi\sigma}{\omega}. \quad (1)$$

Here $\epsilon_0 = 8.9$ is the low-frequency dielectric constant of the GaSe crystal; $\tau_{e,h}$ are the electron and hole momentum relaxation times; and σ and χ are the components of intraband transitions to the corresponding components of the dielectric constant. In the case of highly excited GaSe and probing light at 2.8 μm, these components should be substantial.[3] The procedure for calculating the reflection coefficient (R) of a dielectric medium of micron thickness with plane-parallel walls is described in Ref. 4; the results of these calculations are shown in Fig. 2. We see that the increase in the absorption at the probing frequency smooths out interference effects and gives rise to a

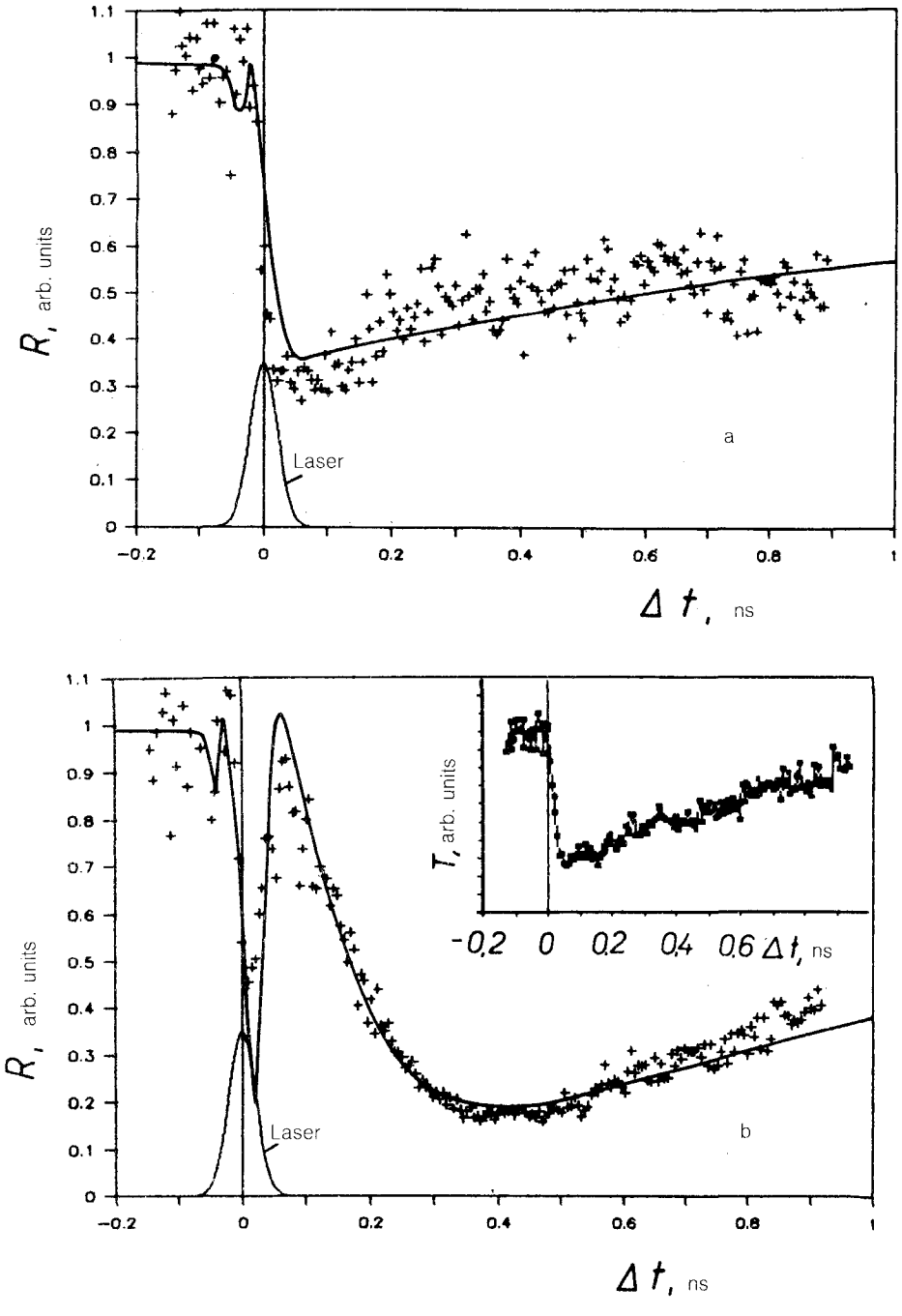

FIG. 1. Reflection coefficient of a single-crystal GaSe layer 10 μm thick at $\lambda = 2.8$ μm versus the time delay between the exciting and probing pulses. a—Pump energy of 670 μJ; b—385 μJ. Crosses) Experimental; solid lines) calculated from Eqs. (1) and (2). The shape of the laser pulse is shown at the bottom. The inset in part b shows the kinetics of the transmission of the probing beam.

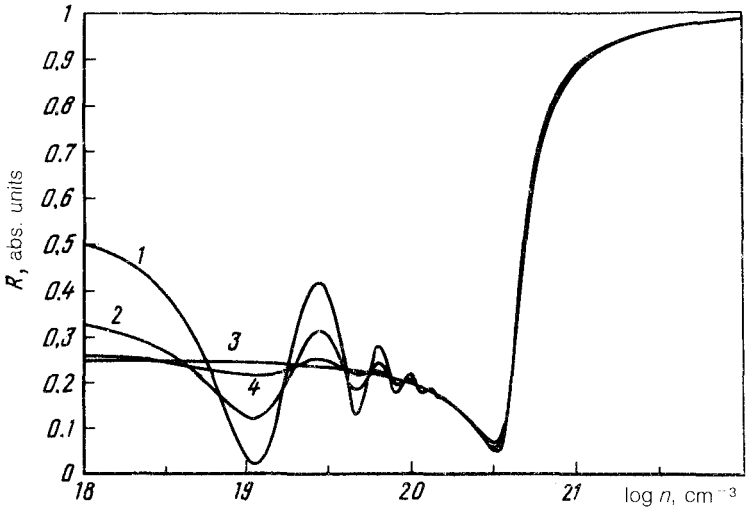

FIG. 2. Theoretical behavior of the reflection coefficient of GaSe films 10 μm thick (lines 1–3) and of a semi-infinite crystal (4) as a function of the density of nonequilibrium charge carriers for probing light at 2.8 μm. 1—$4\pi\sigma/\omega = 0$; 2—0.1; 3—0.2. The other parameter values used in the calculations are $\tau_{e,h} = 10^{-14}$s, $\mu = 0.31m_0$,$\epsilon_0 = 8.9$ and $\gamma = 3.4\times10^{-33}$ cm^6/s.

dependence characteristic of a semi-infinite sample (curve 4 in Fig. 2). Another characteristic feature here is that R reaches a minimum value at a certain density (4×10^{20} cm^{-3} in the case of GaSe). This behavior is characteristic of a plasma resonance. Two minima should be observed as this density point is passed in the course of the excitation and then again during recombination of the nonequilibrium charge carriers.

For a quantitative test we calculated the time evolution of the reflection coefficient R on the basis of a kinetic equation which gives the value of n, and which thus determines R, at any instant:

$$\frac{dn}{dt} = G_0 \exp\left(-\frac{t^2}{2\tau_0^2}\right) - \frac{n}{\tau} - \gamma n^3. \qquad (2)$$

The first term on the right side of this equation corresponds to generation, while the second and third describe linear recombination and Auger recombination, respectively (we are ignoring the quadratic term because of the indirect nature of the optical transitions[3] in GaSe).

The results of these calculations are illustrated by the solid lines in Fig. 1, a and b; the only difference between these two cases is in the rate at which the nonequilibrium carriers are generated. This rate corresponds to the particular power density of the laser excitation used in the experiment. The general agreement is extremely good; the Auger recombination coefficient is $\gamma = 3.5\times10^{-33}$ cm^6/s, and the maximum density of nonequilibrium carriers is 4.2×10^{20} cm^{-3}.

We note in conclusion that the presence of complicated "fast" regions on the time

dependence of R indicates that it would be worthwhile to use an even higher time resolution to study the kinetics of the IR reflection in highly excited semiconductors, with the goal of identifying the role played by the deviation from equilibrium in a high-density system of quasiparticles.

[1]G. N. Galkin, L. M. Blinov, V. S. Vavilov, and A. G. Solomatin, Pis'ma Zh. Eksp. Teor. Fiz. **7**, 93 (1968) [JETP Lett. **7**, 69 (1968)].
[2]M. I. Gallant and H. M. van Driel, Phys. Rev. **B26**, 2133 (1982).
[3]Y. Depeursinge, Nuovo Cimento **38B**, 153 (1977).
[4]M. Born and E. Wolf, Principles of Optics, Pergamon Press, New York, 1984.

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