Reversible picosecond change in the transparency of gallium arsenide during interband absorption of intense light pulses

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Experiments reveal an increment in the transmission of gallium arsenide during illumination in its transparency spectrum by an intense picosecond light pulse. The experimental results agree qualitatively with the theory derived by Kumekov and Perel' {Fiz. Tekh. Poluprovodn. **18**, 835 (1984) [Sov. Phys. Semicond. **18**, 520 (1984)]}, which is based on the concept of Galitskiĭ-Goreslavskiĭ-Elesin quasiparticles {Zh. Eksp. Teor. Fiz. **57**, 207 (1969) [Sov. Phys. JETP **30**, 117 (1969)]}.

Several experimental studies have shown that the illumination of thin samples of direct-gap semiconductors by intense light pulses with a length τ in the picosecond range and with a photon energy slightly greater than the gap width E_g causes the transparency for light at the same frequency to increase with increasing energy of the pulses. This increase eventually reaches saturation, which persists for a time approximately equal to the recombination time τ_R (Refs. 3–5). A study of this phenomenon in the theory of Kumekov and Perel',[1] which is based on the Galitskiĭ-Goreslavskiĭ-Elesin quasiparticle concept,[2] predicts an additional optical absorption of a reversible nature which has not previously been confirmed experimentally. According to Ref. 1, the interband absorption occurs in an irreversible manner with respect to the intensity (I) of the incident light up to the time at which the populations of the upper and lower optical states coupled by the resonant optical transition become comparable; here the frequency of the energy relaxation of the photoexcited carriers, ν_r, is taken into account. The absorption is thereafter caused by a change, reversible with I, in the spectrum of quasiparticle states which are formed from the wave functions of the single-particle states of the valence band and of the conduction band that are coupled by the optical transitions. Conditions for the applicability of the theory of Ref. 1 are $\tau < \tau_R$ and that the scale time of the field change must be greater than ν_r^{-1}. The experiments

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described below were stimulated by the prediction in Ref. 1 of a reversible optical absorption.

In the present experiments, carried out at room temperature, we studied the change in the transparency spectrum of epitaxial GaAs films during and immediately after the transmission of intense light pulses through the samples. The samples are $Al_{0.5}Ga_{0.5}As$–GaAs–$Al_{0.5}Ga_{0.5}As$ heterostructures with layer thicknesses of 1.1, 2.1, and 0.6 μm, respectively, grown by MOS-hydride epitaxy on a (100) n^+-GaAs substrate. The donor density in the GaAs layer is $\sim 3\times 10^{15}$ cm^{-3} at a degree of compensation $\sim 60\%$. The substrate is etched away from an area of 4×4 mm^2. The $Al_{0.5}Ga_{0.5}As$ layers, intended for stabilizing surface recombination and mechanical strength, are transparent to light with $\hbar\omega < 2.2$ eV (Ref. 6).

The laser differential picosecond spectrometer of Ref. 7 is used. Two light pulses, the exciting and probing pulses, are focused on the sample nearly along the normal to its surface (the angle between the beams is $\sim 7°$). The two beams have an identical polarization. The intense exciting pulse, with a photon energy $\hbar\omega = 1.437$ eV ($\lambda = 863$ nm) (this energy is 11 meV greater than the value of E_g found from the photoluminescence spectrum) has a maximum energy transmitted through the probing region of $E_e^M \approx 100$ μJ at a pulse length ~ 30 ps (at half-maximum). The probe pulse, tuned to the wavelength λ_p, has roughly the same length and an energy two or three orders of magnitude lower. The diameter of the probe beam is ~ 0.8 mm. The energy (E_e) of the exciting pulse is adjusted by the thickness of a neutral absorber with a calibrated attenuation coefficient. An adjustable optical delay τ_d is used in the probe beam. We assume that the value $\tau_d = 0$ (there is an error of a few picoseconds because of readjustments) corresponds to the position of the maximum of the cross-correlation function $G(\tau_d)$ of the exciting and probe pulses.

The change in the transparency of the sample, T, is determined from the integrated signal (E_p) from a photodetector at the exit from the probe channel for various values of the energy E_e and of the delay τ_d. Measurements are carried out alternately with and without the exciting pulse. Because of the fluctuations in the laser output, the quantity $\log(T^1/T^0) = \log[(E_p^1/E_r^1)/(E_p^0/E_r^0)]$, where E_r is the reference signal in the probe channel, and the indices 1 and 0 specify the presence and absence of the excitation pulse, respectively, is calculated from ~ 100 measurements by an on-line computer. After a series of such measurements, the sample retains its original optical properties.

Figure 1 illustrates the behavior $\log(T^1/T^0)(E_e) = f(E_e)$ with the results of measurements at $\lambda_p = 794$ nm and $\tau_d = 5$ and 60 ps. During probing at the end of the excitation pulse ($\tau_d = 60$ ps), the transparency reaches a saturation level θ, as was observed in Ref. 5 during probing at the frequency of the exciting light. During probing synchronized approximately with the excitation ($\tau_d = 5$ ps), however, we observe an increment in the transparency, $\Delta = \log(T^1/T^0) - \theta > 0$, which increases monotonically with increasing E_e and which disappears at the end of the excitation, as can be seen from Fig. 1. This increment in the transparency agrees qualitatively with the prediction in Ref. 1 of a reversible absorption of light. Also in agreement with that interpretation is the saturation on the curve of the transparency versus the pulse energy after the end of the pulse. Similar results were found at $\lambda_p = 750$ nm.

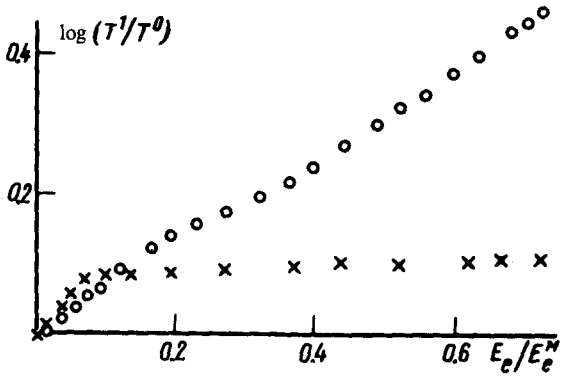

FIG. 1. Change in the transparency of GaAs with the energy of the exciting pulse at $\lambda_p = 794$ nm. ●—$\tau_d = 5$ ps; x—$\tau_d = 60$ ps.

The spectrum of the saturation level, $\theta(\lambda_p)$, is shown by the curves of $\log(T^1/T^0) = f(\lambda_p)$ in Fig. 2, obtained at $\tau_d = 60$ ps. These curves agree well for the two values E_e. The results of similar measurements at $\tau_d = 5$ ps, shown in the same figure, give an idea of the spectrum of the additional contribution $\Delta(\lambda_p)$. It should be noted that for photons of the probe beam with an energy in the interval $E_g < \hbar\omega_p < E_g + 0.341$ eV the transparency is determined primarily by the probability for electron transitions between the bands of heavy and light holes and the conduction band, while at high energies it is also determined by transitions from the completely filled valence band which is split off by the spin-orbit interaction, but these transitions go to correspondingly lower levels of the conduction band.[8] The presence of a clearly defined maximum at $\lambda_p < 700$ nm in Fig. 2 is a consequence of the transparency contribution from transitions from the split-off valence band to the conduction band. The changes in the transparency as a function of E_e at $\lambda_p = 794$ nm and $\lambda_p = 750$ nm described above are thus determined by the probabilities for transitions between energy levels lying substantially above the resonant levels. Measurements near the fundamental absorption edge and (especially) an unambiguous interpretation of these measurements are complicated by interference effects, which intensify as the transmission of the sample increases.

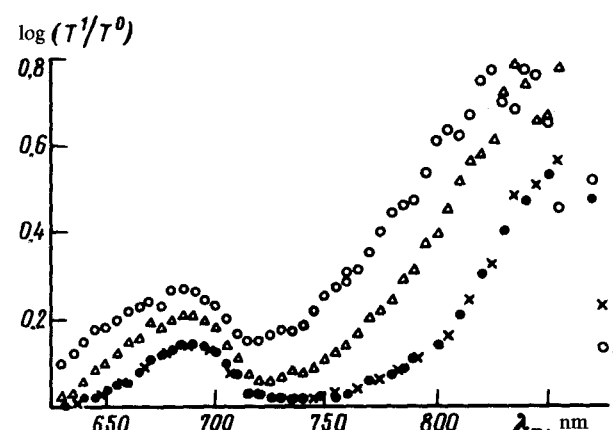

FIG. 2. Spectrum of the change in the transparency at $E_e = 0.85E_e^M$. ○—$\tau_d = 5$ ps; ●—$\tau_d = 65$ ps. $E_e = 0.53E_e^M$. Δ—$\tau_d = 5$ ps; x—$\tau_d = 65$ ps.

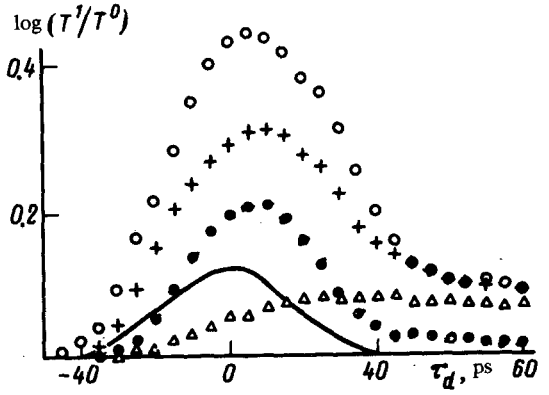

FIG. 3. Variation of the transparency with the time of the delay at $\lambda_p = 794$ nm. Δ— $E_e = 0.074E_e^M$; +—$E_e = 0.52E_e^M$; ○— $E_e = 0.72E_e^M$. $\lambda_p = 750$ nm: ●— $E_e = 0.72E_e^M$. The solid line is the cross-correlation function $G(\tau_d)$.

Figure 3 shows the results of measurements of the function $\log(t^1/T^0) = f(\tau_d)$. Because of the fluctuational nature of the laser radiation and the roughly equal lengths of the exciting and probe pulses, the measured results are statistical in nature (correlational), as is easily understood. It is logical to compare them with the cross-correlation function $G(\tau_d)$ measured under the same conditions. A statistical analysis shows that as the curves of the change in the transparency and $G(\tau_d)$ become closer in shape, the contribution of reversible processes to the transparency increases. This similarity of the curves is seen most clearly for values of λ_p in the part of the spectrum with $\Delta > \theta$ (the curve for $\lambda_p = 750$ nm in Fig. 3; see also Fig. 2). As τ_d increases to the value at which $G(\tau_d) = 0$, the value of $\log(T^1/T^0)$ falls off to the level of θ (Ref. 9), and beyond this point it falls off exponentially with a time constant ~ 500 ps $\sim \tau_R$.

In summary, these studies qualitatively confirm a conclusion that can be drawn from the theory of Ref. 1: that reversible changes occur in the transparency of a direct-gap semiconductor upon changes in the intensity of intense illumination. However, the comparison of experiment with theory is incomplete at this point, since quantitative comparisons cannot be made with the theory in its present state. Work in this direction is presently being carried out.

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