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Structural transitions in GaAs during irradiation by a 100-fs laser pulse

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***Abstract*. It is shown experimentally for the first time that the pumping of a GaAs sample by a 100-fs laser pulse causes plasma-induced bandgap collapse and 'cold' melting of the material during the pulse.**

Keywords: *semiconductors, electron–hole plasma, femtosecond laser-induced structural transitions, bandgap collapse*

We experimentally observed in Ref. [1] the plasma-induced bandgap collapse and the 'cold' melting of a Si(100) sample during irradiation by a 100-fs pump laser pulse. A similar possibility of ultrafast 'cold' melting and structural transitions to new crystal phases was theoretically predicted for GaAs [2–4]; however, as far as we know, these phenomena were not yet observed in this semiconductor.

We studied laser-induced structural transitions in GaAs using a standard femtosecond laser system of the Institute of Laser and Plasma Physics, Essen University, Germany. The laser was similar to those described in Ref. [5]. It consisted of a master oscillator and amplifiers (regenerative and multipass), which used sapphire crystals. The laser produced 800-nm laser pulses (the fundamental emission with frequency ω) with 10-Hz repetition rate, which were approximately 100 fs long and had an energy of up to 1.5 mJ (TEM$_{00}$ mode). The relative amplitude of parasitic pulses was below 5 % – 7 %.

Polarised (s or p polarisation) focused pump radiation at the fundamental frequency in the form of single pulses, which were separated by a synchronised electromechanical shutter, was directed at an angle of 45° to a target made of undoped GaAs(100). The target was translated after each laser pulse. The energy of specularly reflected radiation was measured with a pyroelectric detector at different energies of incident radiation.

The experimental dependences of reflectivities R_s^{ω} and R_p^{ω} on the pump pulse energy were processed to eliminate their spatial averaging caused by a nonuniform distribution of pump energy density F in the light spot of the TEM$_{00}$ mode on the target. The resulting reflectivities R_{1s}^{ω} and R_{1p}^{ω} are shown in Fig. 1 as functions of the effective (integrated over a pulse) pump energy density $F_{\text{eff}} = (1 - R_{1s,p}^{\omega})F$. Using this procedure, we were able to compare the parts of these curves corresponding to identical sample excitation conditions.

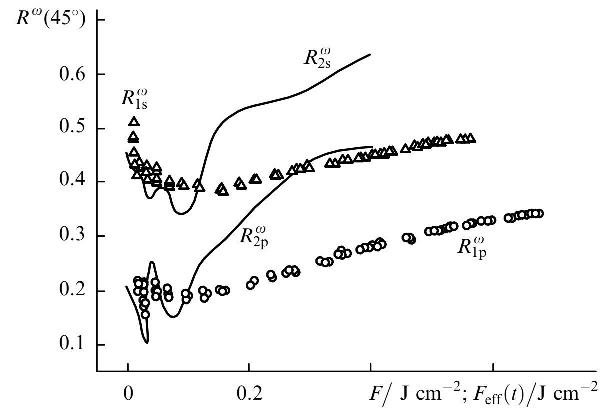

Figure 1. Plots of GaAs reflectivities $R_{1s}^{\omega}(F)$, $R_{1p}^{\omega}(F)$, $R_{2s}^{\omega}(F_{\text{eff}})$, and $R_{2p}^{\omega}(F_{\text{eff}})$.

Because of the 'self-action' of laser radiation [6, 7], which consists in a change in optical characteristics of a semiconductor during a pump laser pulse, the reflectivities $R_{1s}^{\omega}(F_{\text{eff}})$ and $R_{1p}^{\omega}(F_{\text{eff}})$ are averaged in time within the pump-pulse duration. To eliminate this averaging, we performed the graphical processing of the curves (the time T-transform) according to the expressions

$$R_{1s,p}^{\omega}(F_{\text{eff}}) = \int_{F_{\text{eff1}}}^{F_{\text{eff2}}} R_{2s,p}^{\omega}(F')\mathrm{d}F' \left[\int_{F_{\text{eff1}}}^{F_{\text{eff2}}} \mathrm{d}F' \right]^{-1}, \tag{1}$$

$$R_{2s,p}^{\omega}(F_{\text{eff}}) = R_{1s,p}^{\omega}(F_{\text{eff}}) + \frac{\mathrm{d}R_{1s,p}^{\omega}(F_{\text{eff}})}{\mathrm{d}F_{\text{eff}}} F_{\text{eff}}, \tag{2}$$

where R_{2s}^{ω} and R_{2p}^{ω} are the 'true' reflectivities of the sample for the instantaneous $F_{\text{eff}}(t)$, which represents the integral of radiation intensity over the time t within a laser pulse; and F_{eff1} and F_{eff2} are the boundaries of the integration region for which the transform is valid. Note that the transformation of the dependences of R_{1s}^{ω} and R_{1p}^{ω} on F_{eff} according to (1), (2) assumes a nonstationary optical excitation of GaAs, when one may neglect diffusion and recombination contributions in the kinetic equation for the density of electron–hole plasma. The conditions of applicability of this approximation were discussed in our paper [1].

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The dependences $R_{2s}^{\omega}(F_{\rm eff}(t))$ and $R_{2p}^{\omega}(F_{\rm eff}(t))$ have two well-pronounced minima in the region of low $F_{\rm eff}(t)$ (below 0.2 J $\rm cm^{-2}$) at 0.02 – 0.03 J $\rm cm^{-2}$ and 0.06 – 0.08 J $\rm cm^{-2}$. Note that the presence of one minimum and a subsequent increase in linear reflectivity with increasing F was many times experimentally observed in semiconductors, and this behaviour was attributed to the achievement of the edge of plasma reflection [6, 8]. However, two distinct minima observed here for the dependences $R_{2s}^{\omega}(F_{\rm eff}(t))$ and $R_{2p}^{\omega}(F_{\rm eff}(t))$ suggest a new interpretation of these features. For this purpose, we used these dependences and calculated by Fresnel formulas the optical constants n^{ω} and k^{ω} for excited GaAs (Fig. 2a). According to the dependences $n^{\omega}(F_{\rm eff}(t))$ and $k^{\omega}(F_{\rm eff}(t))$, two minima of the curves $R_{2s}^{\omega}(F_{\rm eff}(t))$ and $R_{2p}^{\omega}(F_{\rm eff}(t))$ correspond to two strong linear-absorption bands because one may neglect for $F_{\rm eff}(t) = 0.02 - 0.1$ J $\rm cm^{-2}$ the two-photon absorption and free-carrier absorption, which are lower than the residual absorption between the peaks of $k^{\omega}(F_{\rm eff}(t))$.

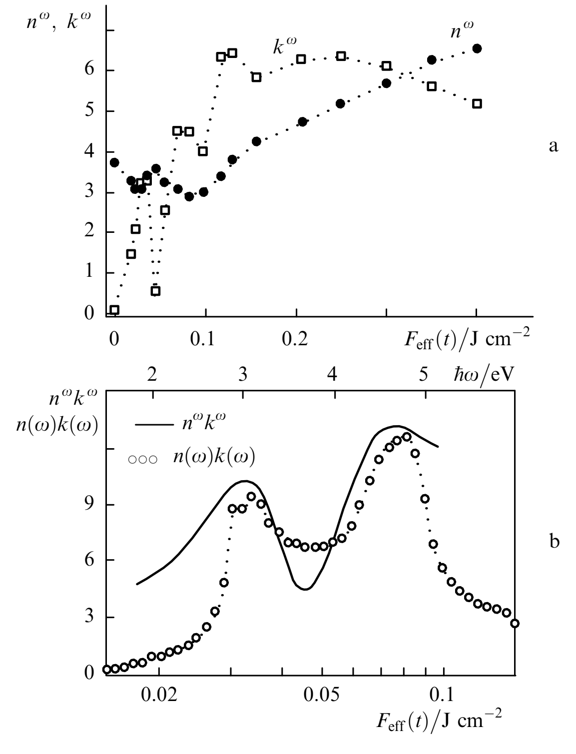

Figure 2. Dependences of the real (n^{ω}) and imaginary (k^{ω}) parts of the refractive index of GaAs on $F_{\rm eff}(t)$ (a) and the products $n^{\omega}k^{\omega}$ for excited GaAs on $F_{\rm eff}(t)$ and $n(\omega)k(\omega)$ for the unexcited sample on the photon energy $\hbar\omega$ (b).

Taking into account the plasma-induced red shift of the linear absorption spectrum of semiconductors at high electron – hole plasma densities [9, 10], it is reasonable to assume that we recorded during a 100-fs pump pulse both bands (E_1 and E_2) of interband transitions $L_{3'} \rightarrow L_1$ and $X_4 \rightarrow X_1$ in GaAs with maxima at 3.0 eV and 4.75 eV, respectively [11]. This shift of the spectrum, which corresponds to the bandgap collapse in GaAs, was observed earlier in Ref. [9] for a probe pulse delayed by more than 0.3 ps, the process duration decreasing with increasing F.

Although the values of ε_1^{ω} and ε_2^{ω} calculated by us for excited GaAs using the dependences $n^{\omega}(F_{\rm eff}(t))$ and $k^{\omega}(F_{\rm eff}(t))$ agree well with the corresponding data of Ref. [9, 10], we independently tested the assumptions concerning the bandgap collapse in GaAs and the red shift of its bands E_1 and E_2: the product $n^{\omega}k^{\omega}$ for the excited sample (Fig. 2b) was com-pared with the product $n(\omega)k(\omega)$ for the unexcited sample (the spectral dependences $n(\omega)$ and $k(\omega)$ were taken from Ref. [11]). A good agreement of the curves in amplitude and position of the peaks qualitatively supports the hypothesis of a red shift.

When $F_{\rm eff}(t) \geqslant 0.12$ J $\rm cm^{-2}$, GaAs changes to the state with $n^{\omega}(0.12\ \rm J\ cm^{-2}) \approx 3.4$ and $k^{\omega}(0.12\ \rm J\ cm^{-2}) \approx 6.3$ (Fig. 2a). These values are close to the optical constants of the equilibrium liquid phase l-Si: $n(1.5\ \rm eV) = 3.3$, $k(1.5\ \rm eV) = 5.7$ [12] (the corresponding data for the l-GaAs phase are unknown to us). This means that ultrafast nonthermal melting occurs in GaAs *during a pump laser pulse*. This statement is additionally supported by a good agreement of the threshold energy density for the formation of this phase $F_{\rm eff}(t) \approx 0.12$ J $\rm cm^{-2}$ and the threshold value presented in Ref. [13] for nonthermal melting of the material ($F \approx 0.15$ J $\rm cm^{-2}$ for 620-nm pump pulses and subpicosecond delays of a probe pulse).

In summary, our experimental data suggest the possibility of an ultrafast (during a 100-fs laser pulse) plasma-induced red shift of linear-absorption bands and the GaAs bandgap 'collapse' along the [111] and [100] crystallographic directions with subsequent formation of a 'cold' metal liquid phase.

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