

Quantum oscillations caused by laser pulses in layered ϵ -GaSe

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The oscillations with a period of 248 fs which have been observed (for the first time) in the optical transmission spectra of ϵ -GaSe subjected to laser pulses 60 fs long with an energy of 1.68 eV are explained under the assumption of a coherent excitation of A_1 nonpolar optical phonons (134.6 cm^{-1}).

The crystals of ϵ -GaSe were grown at the Institute of Physics of the Academy of Sciences of the Azerbaijanian SSR by the Bridgman method. Test samples were cleaved just before the measurements from cylindrical single-crystal blocks 18×10 mm in size. The experiments were carried out at the Institute of Semiconductor Electronics of the Technical University of Aachen (FRG) by a excitation-probing technique.¹ All the measurements were carried out at room temperature, for crystals with thicknesses of 5, 10, 80, 110, and 861 μm and 1.925 and 2.32 mm. We will not discuss the results for the thin crystals (with thicknesses less than 110 μm) in the present letter. Figure 1 shows differential curves of the transmission of a crystal 2.32 mm thick at various excitation densities as a function of the time delay between the excitation pulse and the probe pulse.

The most characteristic features of the curves of the transmission of the crystals more than 100 μm thick are as follows: 1) with $\Delta t = 0$ we observe a significant absorption which increases linearly with the excitation density and reaches saturation at a density $\sim 4 \text{ mJ/cm}^2$. 2) At excitation densities above 0.02 mJ/cm^2 and at positive time delays $\Delta t > 0$ (the probe pulse comes after the excitation pulse) we observe damped oscillations with a period of 250 fs. Their amplitude decreases to zero in $\sim 5\text{--}6$ ps. 3) The initial phase of the oscillations, their frequency, and the time over which they are observed do not depend on the density of the exciting or probing beam or on the angle between the optic axis \bar{C} and the wave vectors of the incident beams. 4) As the pump density is increased as $\Delta t > 0$, we observe an induced absorption, which increases with increasing pump density in an approximately linear fashion.

The nature of the absorption at $\Delta t = 0$ and of the induced absorption as a function of the excitation density might qualify as a topic for a separate discussion and will not be discussed in this letter.

According to the theory of quantum beats in optical spectra,² the excitation of GaSe by pulses with an energy $E = 1.98 \text{ eV}$, a length $\tau = 60 \text{ fs}$, and a spectral width $\Delta E = 70 \text{ meV}$ may result in oscillations whose frequency corresponds to two excited states of direct free excitons. These states would differ in energy and would be coupled

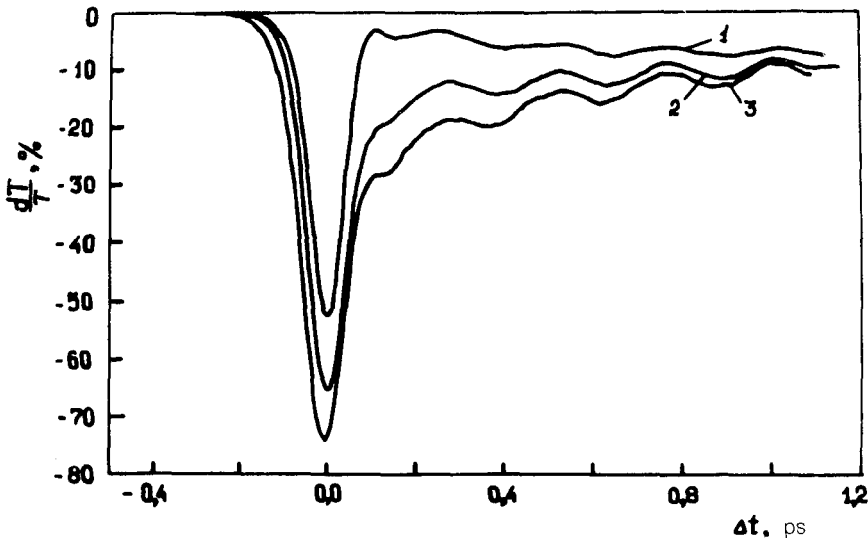


FIG. 1. Time evolution of the relative transmission $(T-T_0)/T_0$ of an ϵ -GaSe crystal 2.32 mm thick at room temperature at various excitation densities: 1—0.56; 2—2.8; 3—5.6 mJ/cm^2 . The energy of the excitation pulse and also of the probe pulse is $E = 1.98$ eV; their length is $\tau = 60$ fs.

by dipole transitions to a common ground state (this is an analog of the beats which arise in the course of quantum transitions between two excited states of an atom³). The oscillation period of 248 fs (which corresponds to an energy of 16.5 meV), however, does not match the periods of the oscillations which would be expected to accompany optical transitions between different exciton states,⁴ $2 \rightarrow 1$ (44 fs), $3 \rightarrow 1$ (37 fs). Furthermore, oscillations are also observed at fairly high excitation densities, at which the renormalized band gap is smaller than the energy of the ground state of the direct free excitons, and the concentration of photoexcited pairs exceeds the concentration corresponding to a Mott transition in GaSe ($n_M \approx 5 \times 10^{17} \text{ cm}^{-3}$; Ref. 5). The renormalized band gap at $F = 9.1 \text{ mJ}/\text{cm}^2$ and the concentration of excitons are estimated to be 1.96 eV and $1.5 \times 10^{18} \text{ cm}^{-3}$, respectively. The observed oscillations are thus not a consequence of quantum beats between different exciton states of gallium selenide. Furthermore, in the case of an exciton mechanism we would expect a strong angular dependence of the oscillation amplitude, since direct transitions are only poorly allowed in the geometry $\vec{E} \perp \text{LC}$ in the case of gallium selenide, and the ratio of the absorption coefficients for the two principal directions is⁴ $\alpha_{\parallel}/\alpha_{\perp} = 27$. Oscillations might be caused by a coherent excitation of nonpolar optical phonons of the A'_1 type (134.6 cm^{-1}), which are active in the Raman spectra of gallium selenide.⁶ A coherent excitation of Raman-active phonons is possible when a light pulse with a length smaller than one vibrational period of a phonon, $\tau_{\text{ph}} = 2\pi/\omega_{\text{ph}}$, is applied to the crystal, and the spectral width is greater than the energy of the corresponding half-width of the spectral band of the phonon.^{7,8} This condition is met, in particular, for the Raman-active A'_1 mode.⁶ The time corresponding to the vibrational period of a phonon of symmetry

A_1' (the most intense Raman-active band) with a frequency of 134.6 cm^{-1} is $\tau_{\text{ph}} = 247.7 \text{ fs}$. This value agrees fairly well with the period of the oscillations which we observed. Further evidence for a phonon nature of the oscillations comes from the circumstance that the lifetime of the A_1' phonons (134.6 cm^{-1}) which we estimate from the half-width of the spontaneous-Raman-scattering band is 5.6 ps and agrees with the time at which the oscillation amplitude drops to zero. These arguments are consistent with the conclusion that the frequency of the oscillations observed here for the first time is the fundamental frequency of a vibrational mode of symmetry A_1' . We have observed oscillations with a period of 176 fs for a GaS crystal. The period of those oscillations agrees well with the vibrational period of 177.5 fs of the strongest nonpolar Raman-active mode of a crystal with a symmetry A_1' and a frequency of 188 cm^{-1} .

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