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GaSe, A NEW EFFECTIVE MATERIAL FOR NONLINEAR OPTICS

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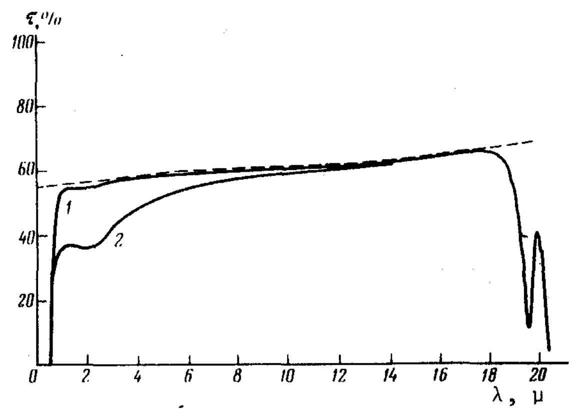
We have investigated the nonlinear optical properties of the semiconducting crystal GaSe, which belongs to the point group 6m2. We used p-type single crystals with carrier density 10^{15} cm^{-3} and mobility 25 cm^2/V-sec, grown by the Bridgman-Stockbarger method. The growth technique makes it possible to obtain single crystals up to 30 mm in diameter and more than 100 mm in length.

The GaSe crystal is transparent in the wavelength range from 0.65 to 18 μ (Fig. 1) and is of good optical quality. The absorption coefficient in the transparency region does not exceed 1 cm^{-1}.

We measured the ordinary refractive index and the birefringence index in the wavelength range from 0.63 to 10.6 μ. The dispersion of the refractive index in the transparency region is described with sufficient accuracy by the formulas

$$n_o^2 = A/\lambda^4 + B/\lambda^2 + C + D\lambda^2 + E\lambda^4,$$
$$n_e^2 = K + L/(\lambda^2 + M) + H\lambda^2, \qquad (1)$$

where A = -0.05466, B = 0.48605, C = 7.8902, D = -0.000824, E = -0.00000273, K = 6.0476, L = 0.3423, M = -0.16491, H = -0.001042, and λ is measured in microns.



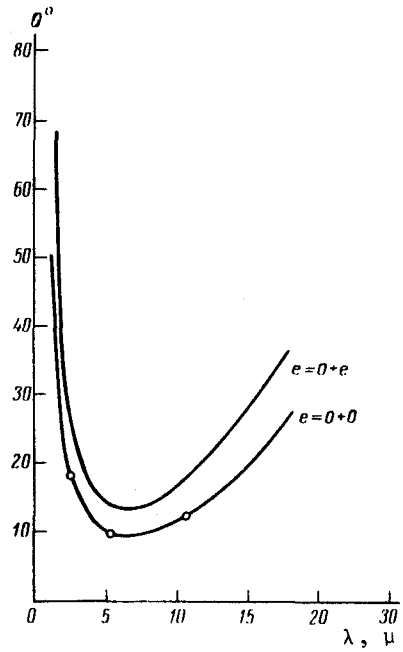


Fig. 1. Transmission spectrum of GaSe crystal: dashed curves - reflection loss, continuous curves - transmission spectrum: 1 - sample thickness 1.5 mm, 2 - 6.0 mm.

Fig. 2. Dependence of the phase synchronism angle for the second harmonic on the pump wavelength.

On the basis of the data obtained for the refractive indices, we calculated the synchronism angle Θ for second-harmonic generation as a function of the pump wavelength (Fig. 2) for the interaction types e = o + o and e = e + o. The effective nonlinear coefficients for these types of interactions are given by

$$d_{eff} = - d_{22} \cos \Theta \sin 3\phi \; (e = o + o)$$

$$d_{eff} = - d_{22} \cos^2 \Theta \cos 3\phi \; (e = e + o),$$

where Θ is the angle between the wave vector $\vec{k}$ of the pump radiation and the optical axis of the crystal (Z axis), ϕ is the angle between the crystallographic plane (XZ) and the (kZ) plane, d_{22} is the nonlinear optical coefficient (the only linearly-independent nonzero coefficient for the given type of symmetry). Second harmonic generation was obtained when the phase synchronism conditions were satisfied for the interaction of the type e = o + o with the pumping by CO (λ = 5.3 μ) and CO_2 (λ = 10.6 μ) molecular laser and by a $CaF_2:Dy^{2+}$ (λ = 2.36 μ) crystal laser. The values of the synchronism angles are: Θ (λ = 2.36 μ) = 18°40' ± 10', Θ (λ = 5.3 μ) = 10°10' ± 20', and Θ (λ = 10.6 μ) = 12°40' ± 20'.

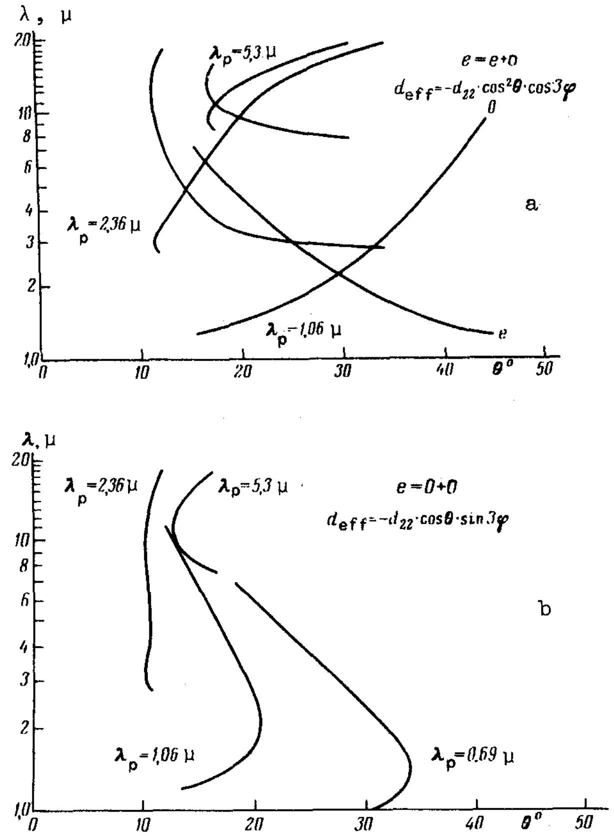


Fig. 3. Wavelength vs. angle curves of parametric radiation for different pump wavelengths.

In the experiments on the generation of the second harmonic of the CO_2 laser, we measured the value of the nonlinear optical coefficient d_{22} for GaSe relative to the known value of the coefficient d_{31} for CdSe [1]. The measurements were made in GaSe samples for thickness ℓ = 0.3 mm with the phase-synchronism conditions satisfied, and on CdSe samples cut in the form of a prism in the absence of phase synchronism, by the method described in [2]. The planes of the faces of the CdSe prism were parallel to the optical axis of the crystal, and the polarization vector of the laser radiation was perpendicular to the optical axis. As a result of the measurements we obtained a ratio d_{22}(GaSe)/d_{31}(CdSe) = 3 ± 0.6 (d_{31}(CdSe) = 0.68 × 10^{-7} cgs esu [1]).

We calculated the angle vs. wavelength curves for collinear parametric interaction with pump wavelength 0.69, 1.06, 2.36, and 5.3 μ (Fig. 3). We used in the calculation the refractive indices calculated from formulas (1).

From the measured value of d_{22}, and with account taken of the angular dependence of d_{eff}, we estimated the pump power density P_p needed to obtain 30% gain at the parametric frequencies for the degenerate case in a GaSe crystal 2 cm long. The results of the estimates are given in the table.

By illuminating the natural faces of the GaSe crystal with giant pulses from a $CaF_2:Dy^{2+}$ laser, we established that the surface of the GaSe sample becomes damaged at power densities >5 MW/cm².

λ_p, μ	P_p, kW/cm^2	
	e = o + o	e = e + o
0.69	15	100
1.06	28	32
2.36	120	125
5.3	600	610

The transparency in a wide spectral range, the high value of the nonlinear susceptibility, satisfaction of the phase-synchronism conditions for parametric generation in the wavelength band from 1 to 18 μ, and the possibility of effectively converting sum and difference frequencies, all make GaSe a very interesting material for nonlinear optics.

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SINGULARITIES OF THE SOUND ATTENUATION COEFFICIENT IN A PHASE TRANSITION OF ORDER 2.5

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The anomalies that the thermodynamic quantities of metals acquire as a result of a change in the topology of the Fermi surface $\varepsilon(p) = \varepsilon_F$ are customarily called a phase transition of order 2.5 [1, 2] (I. Lifshitz [1]).

In this paper we consider the singularities of the absorption coefficient of high-frequency sound ($k\ell >> 1$, $k = \omega/s$ is the wave vector of the sound, ω is its frequency, s is its velocity, and ℓ is the mean free path of the electrons) near the phase-transition point defined by the condition $\varepsilon_F = \varepsilon_c$, where ε_c is the critical value of the energy at which the topology of the equal-energy surfaces changes.

We investigated two variants: (a) appearance (or vanishing) of the cavity of the Fermi surface (for simplicity, a sphere) at $z = (\varepsilon_F - \varepsilon_c = 0)$

$$p^2/2m = z \qquad (z > 0), \tag{1}$$

(b) "breaking of the neck" (see [2]).

According to the results of [3], when $k\ell >> 1$ the electrons that take part in the absorption of sound are those located on the "strip"

$$(\mathbf{v}(\mathbf{p})\mathbf{k}) = 0, \qquad \epsilon(\mathbf{p}) = \epsilon_F, \tag{2}$$

where $\vec{v} = (\partial\varepsilon/\partial\vec{p})$ is the electron velocity.

Each "strip" makes the following contribution to the sound attenuation coefficient:

$$\Gamma \approx \frac{\pi\omega}{(2\pi\hbar)^3 \rho s} \int_0^{2\pi} \frac{|\Lambda|^2 d\phi}{v^2(\phi)\mathcal{K}(\phi)}, \tag{3}$$