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GaSe Faraday rotation near the absorption edge

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We measured the optical dispersion of the interband Faraday rotation of GaSe near the absorption edge at the temperature of 298°K. From our data it is possible to confirm that a direct allowed electronic transition takes place between the valence and the conduction bands in GaSe.

We measured the Faraday rotation of a thin sample of GaSe as a function of the wavelength, near the absorption edge, at a temperature of 298 °K. A pulsed magnetic field with a pulse duration of the order 5 msec was used. The system characteristics, as well as the calibration procedure of the magnetic field, are described elsewhere.[1] We used as a light source for the Faraday rotation measurements a argon pumped Rhodamine 6G dye laser. The laser wavelength, measured with a precision better than 0.1 nm, was varied from 649.4 nm up to 625.6 nm. This last value corresponds to an upper limit in the laser photon energy of 98.5% of the GaSe energy gap. The sample of a thickness of 215 μm was studied with the laser light polarization perpendicular to its c axis.

We carried out two sets of measurements corresponding to the values of the magnetic field peak of 60 and 30 kG, respectively. We found that the rotation angles were proportional to the strength of the applied magnetic field within experimental errors for the whole spectral range; so, we averaged the values obtained for the two series of measurements. The Faraday rotation of GaSe was found to be positive. Our results are summarized in Table I. Because the Verdet constant of the sample depends on its temperature (by shifting the energy gap), we kept the temperature stable within ±2 °K. By using the value given by Tatsuyama *et al.*[2] for the variation of the energy gap E_g with temperature

$$\frac{dE_g}{dT} = -5\times10^{-4}\ \text{eV/°K}, \tag{1}$$

we find that an uncertainty of ±2 °K in the temperature produces a shift in E_g of the order ±0.001 eV, which is unimportant in our experiment.

In Fig. 1 we plotted the measured Verdet constant of GaSe, multiplied it by the index of refraction, as a function of incident photon energy; the corresponding values of the index of refraction were taken from the Akhundov *et al.* measurements.[3] We estimated an experimental error for the Verdet constant measurements of the order ± 5%.

By measuring the frequency dependence of the interband Faraday rotation of a semiconductor, it is possible to obtain information about the value of its energy gap and the nature of the optical transition: direct or indirect, that takes place. According to Boswarva *et al.*[4] we have for a direct allowed optical transition the relation

$$nV = KF_2(x), \tag{2}$$

where n is the index of refraction of the sample, V is its Verdet constant (min G^{-1} cm^{-1}), K is a constant, and the variable x is defined by

$$x = E/E_g, \tag{3}$$

where E is the incident photon energy and E_g is the energy gap. The function $F_2(x)$ is given by

$$F_2(x) = (1/x)\left[(1-x)^{-1/2} - (1+x)^{-1/2}\right] - 1. \tag{4}$$

This last function applies when the contributions of the lower bands are taken into account. On the other hand, when it is assumed that only one valence band contributes to the magneto-optical rotation, we have the relation, as shown by Roth,[5]

$$nV = KF_1(x), \tag{5}$$

where the function $F_1(x)$ is given by

$$F_1(x) = (1/x)\left[(1-x)^{-1/2} - (1+x)^{-1/2}\right] - (4/x^2)\left[2 - (1-x)^{1/2} - (1+x)^{1/2}\right]. \tag{6}$$

The two functions are very similar and both approach $(1-x)^{-1/2}$ when $E \to E_g$. However, for any value of x, the function $F_1(x)$ is always smaller than $F_2(x)$.

For indirect allowed electronic transitions between valence and conduction bands, the phonon contribution must be included. Assuming one phonon emission and absorption process, Kolodziejczak *et al.*[6] found the relation

$$nV = KF_3(x') = \left(\frac{1}{x'^2}\ln\frac{1}{1-x'^2} + \frac{1}{x'}\ln\frac{1-x'}{1+x'} + 1\right)K, \tag{7}$$

while Boswarva *et al.*[4] obtained the result

$$nV = KF_4(x') = \left(\frac{1}{2}\ln\frac{1}{1-x'^2} + \frac{1}{2x'}\ln\frac{1-x'}{1+x'} + 1\right)K, \tag{8}$$

TABLE I. Optical dispersion of the Verdet constant of GaSe.

Wavelength (μm)	Photon energy (eV)	Verdet constant (min G^{-1} cm^{-1})
0.6494	1.909	0.59_5
0.6459	1.919_5	0.61_5
0.6420	1.931	0.65
0.6381	1.943	0.70
0.6356	1.950_5	0.74
0.6328	1.959	0.80_5
0.6306	1.966	0.91
0.6287	1.972	1.04_5
0.6275	1.976	1.15
0.6265	1.979	1.28_5

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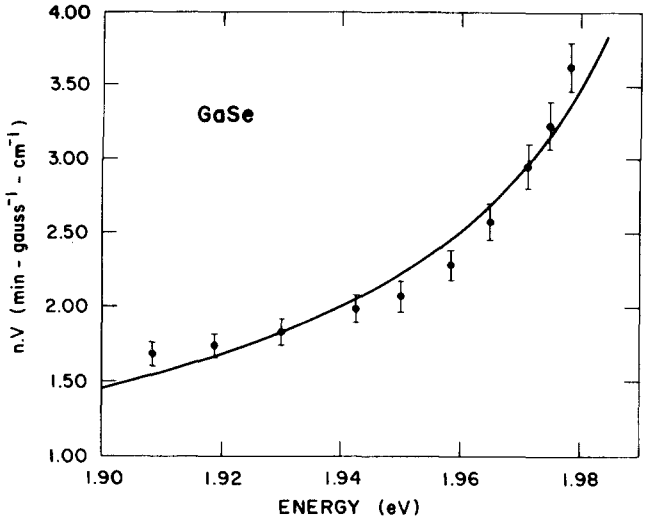

FIG. 1. GaSe Verdet constant, multiplied by its index of refraction, as a function of the incident photon energy.

where $x' = E/(E_g + E_{ph})$ and E_{ph} is the phonon energy. These two last functions are quite different from F_1 or F_2, in that they rise more slowly and do not go to infinity as E goes to E_g.

Our Faraday rotation data, as a function of the incident photon energy, were fitted to Eqs. (3), (5), (7), and (8). We found that when functions F_3 and F_4 are used the fitting is very poor; whereas, a much better agreement is obtained by using the functions F_1 or F_2. For function F_1 we get the parameter values

$$K = 0.720 \text{ min G}^{-1}\text{cm}^{-1} \text{ and } E_g = 2.02 \text{ eV}; \quad (9)$$

and by using function F_2,

$$K = 0.520 \text{ min G}^{-1}\text{cm}^{-1} \text{ and } E_g = 2.01 \text{ eV}. \quad (10)$$

Thus we can conclude from the observed frequency dependence of the GaSe Faraday rotation that an electronic direct allowed transition takes place between the valence and conduction bands. Nevertheless, from our experimental data it is not possible to decide if functions F_1 or F_2 give us a better description of the interband Faraday rotation effect in GaSe.

We also plotted in Fig. 1 (solid line) the theoretical curve represented by Eq. (3). A similar curve is obtained by using the function F_1 instead of F_2. It can be seen from the graph that the fitting is not quite satisfactory, the difference between the theoretical curve and the data points is slightly bigger than the assumed experimental error. Such a discrepancy can be explained in the following way: from optical absorption spectra data (Akhundov *et al.*[3]) and magneto-optical absorption (Brebner *et al.*[7]), it is known that there is an exciton absorption line in GaSe very near to the fundamental absorption edge (about 30 mV below of it) and the contribution of this excitonic transition to the Faraday rotation should be taken into account in order to get a better agreement between theory and experimental data.

The constant K in Eqs. (2) and (5) is given by the relation, according to Boswarva *et al.*,[3]

$$K = \frac{2^{1/2} e^2 |p_{cv}^x|^2}{c\hbar m_0^2} \left(\frac{\mu}{E_g}\right)^{3/2} \gamma \quad (11)$$

in cgs units, where e is the electron charge (in esu units), c is the light velocity in a vacuum, $\hbar$ is the Planck's constant divided by 2π, m_0 is the mass of the free electron, μ is the reduced effective mass for electrons and holes, E_g is the energy gap, γ is the Zeeman splitting parameter

$$\gamma = \mu_B (g_c + g_v)/2\hbar, \quad (12)$$

with μ_B the Bohr magneton and g_c and g_v the g factors of the conduction and valence bands, respectively; and $|p_{cv}^x|^2$ is the square of the matrix element of $-i\hbar(\partial/\partial x)$ between the valence and the conduction states at $k = 0$.

By using the values for GaSe given by Nishina *et al.*,[8] we get

$$g^* = g_c + g_v = 3.3 \text{ and } \mu = 0.14 m_0, \quad (13)$$

and the values of the parameters K and E_g given in Eqs. (9) and (10), we obtain the two possible values of $|p_{cv}^x|^2$,

$$|p_{cv}^x|^2 = 4.6 \times 10^{-39} \text{ cgs units} \quad (14)$$

or

$$|p_{cv}^x|^2 = 3.3 \times 10^{-39} \text{ cgs units}, \quad (15)$$

respectively. These are of the same order of magnitude as reported by Balkanski *et al.*[9] for the II–VI compounds, such as ZnS, ZnSe, CdS, and ZnTe,

$$|p_{cv}^{xx}|^2 = 1.53 \times 10^{-38} \text{ cgs units}. \quad (16)$$

Likewise, the value we obtained for the energy gap E_g is in complete agreement with the values reported from optical absorption experiments at room temperature. For instance, according to Tatsuyama *et al.*,[2] $E_g = 2.01$ eV at 282 °K.

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