Photoinduced gyrotropy due to spin-density fluctuations of the free electrons in semiconductors

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An ultralinear dependence of the induced optical gyration in n-InAs on the electron density n in the c conduction band is observed. A 200 deg · cm/MW angle of rotation θ is attained. The proposed theory of the phenomenon relates the region of rapid increase in $\theta(n)$ to the spin splitting of the c band and describes the observed effects of spatial dispersion.

1. The propagation of an intense, circularly polarized pump light with a frequency ω in a crystal causes the linear-polarization vector of the probing beam of frequency Ω, which is directed nearly parallel to the pump light, to rotate through an angle

$$\theta = \alpha^{-1}(\omega)\{1-\exp[-\alpha(\omega)d]\}j_\omega\bar{\theta},$$
$$\bar{\theta} = \frac{4\pi^2\Omega}{c^2\kappa_0^2(\Omega)}\chi_a, \qquad \chi_a = \mathrm{Re}\,[\chi^{(3)}_{1212}(\Omega,\omega,-\omega)-\chi^{(3)}_{1221}(\Omega,\omega,-\omega)], \tag{1}$$

where $\alpha(\omega)$ is the absorption coefficient, $\kappa_0(\Omega)$ is the refractive index, d is the thickness of the sample, j_ω is the intensity of the pump light at the front face of the crystal, and $\chi^{(3)}_{ijkl}$ are the components of the cubic-susceptibility tensor.

The n-dependent photoinduced gyrotropy mechanism is related to the virtual interband transitions of the type $c_s \rightarrow v_{l,m} \rightarrow c_{s'} \rightarrow v_{l'm'} \rightarrow c_s$, where l is the valence subband index (v), and s and m are the spin indices of the c and v bands.[1)] To calculate χ_a, we will use the effective intraband Hamiltonian for the interaction of electrons with the wave field, in which the allowed virtual transitions $c_s \rightarrow v_{l,m} \rightarrow c_{s'}$ are taken into account,

$$H_c = \frac{e^2}{m}\sum_{i,j}\{a(\omega_i)(\mathbf{A}_i\mathbf{A}_j)\rho_{\mathbf{q}}^{(+)} + ib(\omega_i)[\mathbf{A}_i\mathbf{A}_j]\rho_{\mathbf{q}}^{(-)}\}, \tag{2}$$

where $\mathbf{A}_i$ is the vector potential of the wave with a frequency ω_i which takes on the values $\pm\omega$ and $\pm\Omega$, $\mathbf{q}=\mathbf{q}_{\omega_i}+\mathbf{q}_{\omega_j}$ is the sum of the wave vectors, $\rho_{\mathbf{q}}^{(\pm)}=\rho_{\mathbf{q}\uparrow}\pm\rho_{\mathbf{q}\downarrow}$, $\rho_{\mathbf{q}\uparrow}$ and $\rho_{\mathbf{q}\downarrow}$ are the electron-density operators with the spin $\pm 1/2$ projections along the direction $[\mathbf{A}_i\mathbf{A}_j]$, $a(\omega)$ and $b(\omega)$ are given in Ref. 2, and $b(-\omega)=-b(\omega)$, where $b=0$ at $\Delta_{so}=0$ (Δ_{so} is the spin-orbit splitting of the valence band). In InAs we have $b=5.1$ at $\hbar\omega=0.117$ eV.

Hamiltonian (2) was used in Ref. 2 to calculate Raman scattering by the electron density fluctuations and the spin-density fluctuations. We can express χ_a in terms of the real parts of those density correlation functions whose imaginary parts give the density fluctuations, the spin-density fluctuations and the scattering caused by these fluctuations. The effect of the term with $\rho_{\mathbf{q}}^{(+)}$ on χ_a is suppressed due to the screening

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of the relevant density fluctuations at $n > 10^{16}$ cm^{-3}. The effect we are discussing is attributable to the term with the spin density $\rho_{\mathbf{q}}^{(-)}$ in (2). The corresponding spin-density fluctuations do not carry a charge and are not screened.

The spin splitting of the c band has an appreciable effect on χ_a: $\Omega_0 \propto |\boldsymbol{\kappa}|$, where $\kappa_\alpha = p_\alpha \times (p_{\alpha+1}^2 - p_{\alpha+2}^2)$ (p_α are the Cartesian components of the momentum). In the case of InAs, at $n = 10^{17}$ cm^{-3} we have $\langle \Omega_{0F} \rangle \sim 10^{11}$ s^{-1} (the angle brackets denote averaging over the angles, and the subscript F refers to quantities at the Fermi level, $\mathscr{E}_F$).

At $T \ll \mathscr{E}_F$ a calculation yields the following expression for $\bar{\theta}$:

$$\bar{\theta} = \frac{\pi}{16\Omega}\left[\frac{eq_{FT}}{mc\,\omega\kappa_0(\Omega)}\right] \mathrm{Re}\,[\tilde{\theta}(\Omega,\omega) + \tilde{\theta}(-\Omega,-\omega) - \tilde{\theta}(\Omega,-\omega) - \tilde{\theta}(-\Omega,\omega)], \quad (3)$$

where q_{FT}^{-1} is the Thomas-Fermi screening length,

$$\tilde{\theta}(\Omega,\omega)' = 8b(\omega)b(\Omega)K(\Delta\omega, \Delta\mathbf{q}_\omega) + [b(\Omega)+b(\omega)]^2 K(\Omega-\omega, \mathbf{q}_\Omega - \mathbf{q}_\omega). \quad (4)$$

It is important to note that in our experiments both beams were generated by a single pulsed CO_2 laser, whose spectrum contained nine narrow ($\delta \ll qv_F$ wide) uniformly spaced lines (harmonics). $\Delta\omega$ and $\Delta\mathbf{q}_\omega$ in (4) represent the difference in the frequencies and in the wave vectors of the pump-beam harmonics. For the adjacent harmonics we have $\Delta\omega_1 = 3.66 \times 10^{11}$ s^{-1}. We can write the expression for $K(\omega, \mathbf{q})$ in (4) as

$$K(\omega,\mathbf{q}) = \langle |\vec{\kappa}|^{-2}[\kappa_z^2(\tilde{K}_{11} + \tilde{K}_{22}) + (\kappa_x^2 + \kappa_y^2)(\tilde{K}_{12} + \tilde{K}_{21})]\rangle, \quad (5)$$

$$\tilde{K}_{mn} = \omega_{mn}(\omega - \omega_{mn} + i\tau_{mn}^{-1})^{-1}[1 - i\tau_p^{-1}\langle(\omega - \omega_{mn} + i\tau_{mn}^{-1})^{-1}\rangle]^{-1}, \quad (6)$$

where τ_p and τ_s are the relaxation times of the momentum (in the case of elastic scattering) and of the electron spin, the exponents 1 and 2 refer to the lower and upper spin branches, $\tau_{mn}^{-1} = \tau_p^{-1} + \tau_s^{-1}(1 - \delta_{mn})$, $\omega_{mn} = \Omega_{mn} + \mathbf{v}_F\mathbf{q}$, $\Omega_{11} = \Omega_{22} = 0$, and $\Omega_{12} = -\Omega_{21} = \Omega_0$.

All possible four-wave processes such as $\Omega_r = \omega_{r'} - \omega_{s'} + \Omega_s$ contribute to the observable effect. If the phases of the individual harmonics are taken into account, we can replace the expression for $\tilde{K}_{mn}$ in (6) by

$$\tilde{K}'_{mn} = \sum_{r,s,t} \beta_r\beta_{r+t}\beta_s\beta_{s+t}\tilde{K}_{mn}(t\Delta\omega_1)\exp\left[i(\varphi_r^\Omega - \varphi_{r+t}^\Omega - \varphi_s^\omega + \varphi_{s+t}^\omega)\right], \quad (7)$$

where β_r is the relative intensity of the r-th harmonic $\left(\sum_r \beta_r^2 = 1\right)$.

2. The method used in this experiment is similar to that described in Ref. 3. However, to determine the function of the spatial dispersion, we aimed the beams at the crystal from one side and from the opposite sides (in both cases the angle between

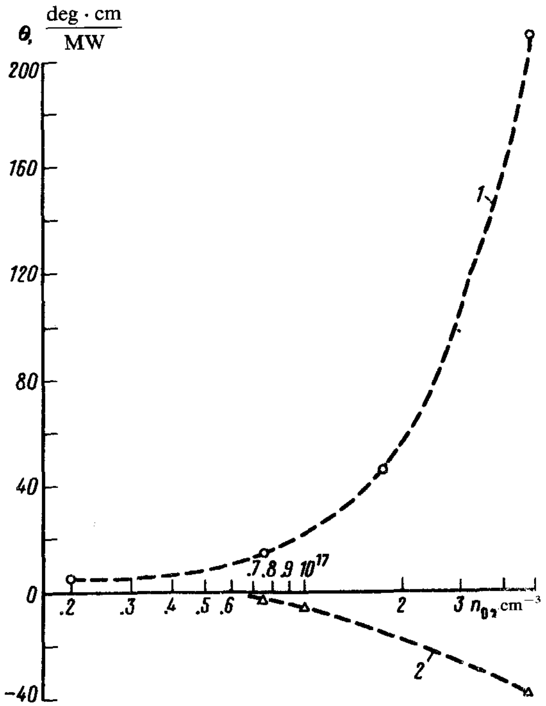

FIG. 1. Normalized angle of rotation $\overline{\theta}$ in n-InAs versus the free-carrier density n in the case of parallel and oppositely directed probing and pump beams. Each beam (here and for the curve in Fig. 2) contains nine spectral lines near $\hbar\omega = \hbar\Omega = 0$ and 117 eV at $T = 90$ K.

the beams in the sample was $\sim 1.5°$). Figure 1 shows the concentration dependences of $\overline{\theta}$ at $T = 90$ K, obtained when "parallel" and "bucking" geometries of the experiment were used. We see that the signs of the effect and the shape of the curves are different in these situations.

3. In the case of "parallel" beams, the principal contribution to $K(\omega, \mathbf{q})$ comes from the interaction of the adjacent harmonics. Here we have $\pm \Delta\omega_1$ in the argument of K, and $\widetilde{K}_{12}$ and $\widetilde{K}_{21}$ are the important components. If the conditions $\Delta\omega_1$, $v_F q$, $\langle \Omega_{0F} \rangle \ll \tau_p^{-1}$ and $\Delta\omega_1 - \langle \Omega_{0F} \rangle \gg \tau_s^{-1}$ hold, we can write

$$\mathrm{Re}\, K \simeq \frac{2}{3} \langle \Omega_{0F} \rangle [\Delta\omega_1 - \langle \Omega_{0F} \rangle]^{-1} . \tag{8}$$

Since $\langle \Omega_{0F} \rangle \propto n$, Eq. (8) can describe the sharp increase in $\overline{\theta}(n)$ (curve 1 in Fig. 1) at $n > (2\text{–}3) \times 10^{17}$ cm^{-3} when $\langle \Omega_{0F} \rangle$ approaches $\Delta\omega_1$. The order of magnitude of the effect is correctly predicted by the theory.

The collective interaction[2)] also plays a dominant role in the spectral $\overline{\theta}$ dependence (Fig. 2), where the sign changes four times over a 2-MeV interval, an occurrence that cannot be explained in terms of the given microscopic mechanism of the effect in the case of monochromatic beams. Since p-InAs and p-Ge, in which the photoinduced gyrotropy is different, have similar spectra,[4] we conclude that the spectrum is related to the distribution of the harmonic phases of the pump and the probing beams.

In the case of oppositely directed beams, we see that the first term in (4) does not change, while the argument of K in the second term acquires $q' = q_\Omega + q_\omega \simeq 2q_\omega$

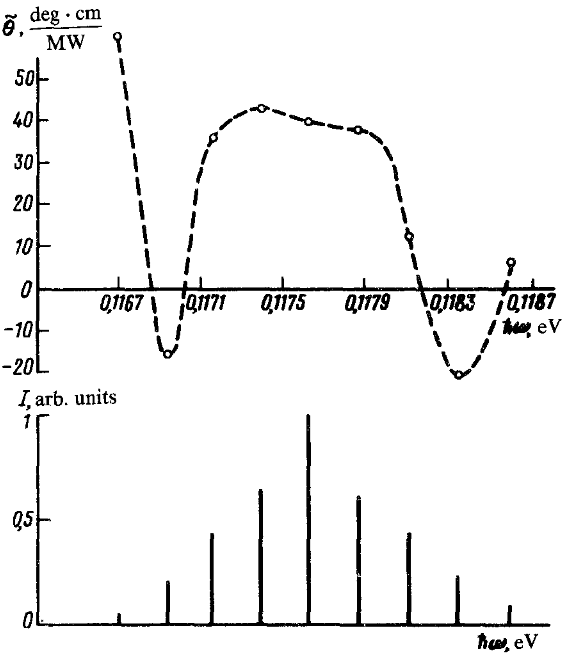

FIG. 2. Dependence of $\overline{\theta}$ on $\hbar\omega_r$ for the n-InAs sample ($n = 4.2\times10^{-17}$ cm^{-3}, $T = 300$ K). Both beams propagate in the same direction.

instead of $q = q_\Omega - q_\omega$. We now have $q'v_F \gtrsim \tau_p^{-1}$. Here $K\sim -1$. Since the collective interaction of all harmonics, rather than that of only the adjacent harmonics, contributes to K, the second negative term in (4) is larger in absolute value than the first positive term, which accounts for the observed change in the sign of the effect.

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1)Analysis of other possible photoinduced-gyrotropy mechanisms, including the contribution from the virtual allowed-forbidden transitions of the electrons of the filled valence band[1] and from the magnetization of the sample caused by the splitting of the c band by interaction (2) shows that these mechanisms play an insignificant role in these experiments.

2)The experiments involving a single probing beam line and a single pump beam line ($\omega = \Omega$) showed no evidence of a sharp increase in $\overline{\theta}$ as n was varied over the range $(2\text{–}5)\times10^{17}$ cm^{-3}, consistent with the theory.

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