Photovoltaic effect in spin resonance in a quantizing magnetic field

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The photovoltaic effect in n-InSb in optical transitions between spin Landau-level subbands is investigated theoretically and experimentally in the ultraquantum limit. The current along the magnetic field is considered. The effect is due to Hamiltonian cubic terms that are caused by the absence of an inversion center. The current has a resonance dependence on the magnetic field and contains contributions both even and odd in the field. This character of the effect is due to resonance in the intermediate state and to interference between transition amplitudes of second order in the relativistic contributions to the Hamiltonian. The dependences of the photovoltaic effect on the crystal orientation and on the radiation polarization direction are investigated experimentally. The band parameters of InSb are determined by comparison with the theory.

1. INTRODUCTION

Starting with the work of Rashba and Sheka,[1] combined resonance (light absorption on account of the electric component of an electromagnetic wave and due to electronic transition with spin flip) continues to be of interest in solid-state physics. Thus, interference of magnetodipole and electrodipole resonances in the Voigt configuration in crystals without inversion center has been recently observed and investigated.[2-4]

The absence of an inversion center is known also to lead to an onset of a stationary current induced by homogeneous illumination—the photovoltaic effect (FVE).[5] The dependence on the light polarization and on the crystal orientation help distinguish it from among other photoelectric effects.

The FVE in a magnetic field was investigated by many,[6-8] but the case of a quantizing field has not been considered. We report here a theoretical and experimental study of the FVE in a quantizing magnetic field for spin transitions in the conduction band of a semiconductor of InSb type.

Study of the FVE under spin resonance can supplement light-absorption experiments as a method of measuring the band parameters, since the same Hamiltonian terms can lead both to electrodipole transitions and to an FVE current.

2. THEORY

We consider current flowing along the direction of the magnetic field **H** with the light propagating in the same direction (Faraday geometry). The light polarization and the orientation of **H** relative to the crystallographic axes are assumed arbitrary. Let the conditions corresponding to the quantum limit $\omega_s \gg T$, $\omega_s > \mu$, be satisfied, where $\omega_s = |g|\mu_B H$ is the spin-transition energy, μ is the Fermi level measured from the lower spin subband, g is the g factor, μ_B is the Bohr magneton, and $h = 1$.

Let $\mathbf{A}_0$ and $\mathscr{A}(t) = \mathrm{Re}(\mathbf{A}\exp[-i\omega t])$ be respectively the vector potential of the constant homogeneous magnetic field and of the electromagnetic wave,

$$U(\mathbf{r}) = \sum_i u(\mathbf{r}-\mathbf{r}_i)$$

the potential energy of the electron interaction with the randomly distributed impurities ($\mathbf{r}_i$ is the coordinate of the ith impurity center). The Hamiltonian of the system considered is

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1 + \mathscr{H}_2 + \mathscr{H}_U + U + \mathscr{F},$$

where $\mathscr{H}_0$ is the Hamiltonian of the free electron in the parabolic approximation:

$$\mathscr{H}_0 = \frac{k^2}{2m} + \frac{1}{2} g\mu_B(\mathbf{H}\sigma), \qquad \mathbf{k} = \mathbf{p} + \frac{e}{c}\mathbf{A}_0.$$

The terms $\mathscr{H}_1$, $\mathscr{H}_2$, and $\mathscr{H}_U$ correspond to three possible mechanical transitions with spin flip: $\mathscr{H}_1 = \delta_0(\sigma\chi)$, where in the principal crystal axes $\chi_i = k_j k_i k_j - k_l k_i k_l$ it is connected with the absence of an inversion center; the term

$$\mathscr{H}_2 = \tilde{g}\mu_B\{(\mathbf{Hk})(\sigma\mathbf{k}) + (\sigma\mathbf{k})(\mathbf{Hk})\}$$

is connected with the dependence of the g factor on the momentum, and the term

$$\mathscr{H}_U = \alpha([\nabla U, \mathbf{k}]\sigma)$$

is the spin-orbit interaction of the electron with the impurities.

The Hamiltonian terms designated by the letter $\mathscr{F}$ determine the interaction of the electrons with the electromagnetic wave, with

$$\mathscr{F} = \mathscr{F}_0 + \mathscr{F}_1 + \mathscr{F}_2 + \mathscr{F}_U,$$

where

$$\mathscr{F}_0 = \frac{e}{mc}(\mathbf{k}\vec{\mathscr{A}}), \qquad \mathscr{F}_1 = \frac{ie\delta_0}{c}((\sigma\chi)(\mathbf{r}\vec{\mathscr{A}})),$$

$$\mathscr{F}_2 = 2\tilde{g}\mu_B(\sigma\vec{\mathscr{A}})(\mathbf{Hk})\frac{e}{c},$$

$$\mathscr{F}_U = \alpha\frac{e}{c}(\sigma[\nabla U, \vec{\mathscr{A}}]).$$

For current to flow along the field **H**, the transition probability must be odd as a function of the longitudinal momentum p_z (the z axis is directed along the magnetic field). Obviously, this occurs if the probability is calculated in the nonzeroth order in the constant δ_0 that determines the absence of inversion center. We begin by solving a quantum-kinetic equation of the form

$$(I\delta f)_\alpha + G_\alpha = 0, \tag{1}$$

where δf_α is the increment to the equilibrium distribution function

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$$(I\delta f)_\alpha \equiv \sum_\beta (W_{\beta\alpha}\delta f_\beta - W_{\alpha\beta}\delta f_\alpha)$$

is the integral of electron collisions with the impurities, G is the generalization probability, $\alpha = (n,\mathbf{p},\sigma)$ is the set of quantum numbers indicative of the eigenstates of the Hamiltonian $\mathscr{H}_0$ in the gauge $A_0 = (0,H\chi,0)$, n is the number of the level, $\mathbf{p} = (p_y, p_z)$ is the electron momentum, $\sigma = \pm 1$ (we shall hereafter designate the spin projections by up and down arrows). Since we are interested in the Landau-level limits $n = 0$, we shall omit this subscript from all the quantities.

The generation functions $G_{0p\sigma} \equiv G_{p\sigma}$ are defined as

$$G_{\mathbf{p}\uparrow} = -\sum_{\mathbf{p}'} w_{\mathbf{p}\uparrow,\,\mathbf{p}'\downarrow}\delta\left(\frac{p_z^2 - p_z'^2}{2m} - \omega_s + \omega\right) f^{(0)}_{p_z'\uparrow}, \tag{2}$$

$$G_{\mathbf{p}\downarrow} = \sum_{\mathbf{p}'} w_{\mathbf{p}'\uparrow,\,\mathbf{p}\downarrow}\delta\left(\frac{p_z'^2 - p_z^2}{2m} - \omega_s + \omega\right) f^{(0)}_{p_z'\uparrow}. \tag{3}$$

Here $f\alpha^{(0)}$ is the equilibrium distribution function of the electrons, $w_{\alpha\beta}$ is the square of the matrix element of the electron transition with spin flip. Bearing in mind InSb,in which $g<0$, we assume that the lower spin subband corresponds to $\sigma = \pm 1$.

Since the Hamiltonian terms responsible for spin-flip processes are small, and the generation process itself requires that they be taken into account, we can neglect the contribution of such processes in the collisional integral.

We subdivide I, G, and δf into parts that are even and odd in p:

$$\begin{aligned} &I=I^+ + I^-, \quad G=G^+ + G^-,\\ &I^+\delta f^- + I^-\delta f^+ = G^-,\\ &I^+\delta f^+ + I^-\delta f^- = G^+. \end{aligned} \tag{4}$$

The current is determined by the odd part of δf_a:

$$j_z = -\frac{e}{V}\sum_\alpha v_\alpha^z\,\delta f_\alpha^-$$

(V is the volume of the system, v^z is the operator of the velocity z-component). For interaction with impurities, the term $I^-\,\delta f^+$ vanishes. Actually the probability of scattering by impurities can be written in the form $W_{\alpha\beta} = 2\pi N_i \langle |t_{\alpha\beta}|^2\rangle \delta(\varepsilon_{\alpha\beta})$, where t is the operator for scattering by a single impurity center (the angle brackets denote averaging over the impurity configurations) $\varepsilon(\alpha\beta) = \varepsilon(\alpha) - \varepsilon(\beta)$, and ε_i are the eigenvalues of $\mathscr{H}_0$. The scattering operator, however, satisfies the optical theorem and the condition that follows from the time reversal requirement:

$$\operatorname{Im} t_{\mathbf{pp}} = \pi \sum_{\mathbf{p}'} |t_{\mathbf{pp}'}|^2 \delta\left(\frac{p_z^2 - p_z'^2}{2m}\right), \tag{5}$$

$$t_{\mathbf{pp}'}(\mathbf{H}) = t_{-\mathbf{p}',\,-\mathbf{p}}(-\mathbf{H}). \tag{6}$$

(we do not write out here the inessential index σ.) Since the momentum operator and the magnetic field enter the Hamiltonian $\mathscr{H}_0$ in the form of the combination $p_y + Hxe/c$, simultaneous reversal of the sings of p_y and H does not change the Hamiltonian. It follows hence that for

$$\bar{t}_{p_z\,z} = \sum_{p_y} t_{\mathbf{pp}}$$

the condition $\bar{t}p_z p = \bar{t} - zp_z, -p_z$ is met. But this equation and the condition leads to

$$\sum_{p_y,\mathbf{p}'} [\,|t_{\mathbf{pp}'}|^2 - |t_{-\mathbf{p},-\mathbf{p}'}|^2]\,\delta\left(\frac{p_z^2 - p_z'^2}{2m}\right) = 0,$$

and this means that $\int_{p'} W_{pp'}$ is even in p_z. Using this fact and the energy conservation law, according to which $p_z' = \pm p_z$, we find that

$$(I^-\delta f^+)_{\mathbf{p}} = \sum_{\mathbf{p}'} [(W_{\mathbf{p}'\mathbf{p}} - W_{-\mathbf{p}'\,-\mathbf{p}})\,\delta f_{\mathbf{p}'}^+ - (W_{\mathbf{pp}'} - W_{-\mathbf{p},-\mathbf{p}'})\,\delta f_{\mathbf{p}}^+] \equiv 0.$$

This means that the odd probability of scattering by impurities in the superquantum limit does not lead, in contrast to the case of small H, to a photovoltaic effect.

Thus, the quantity δf_α^- we need to calculate the longitudinal current is determined by an equation that follows from the first equation of (4):

$$\delta f_{p\sigma}^- = G_{p\sigma}^-\tau_{pz\sigma}, \tag{7}$$

where the relaxation time $\tau_{p_z\sigma}$ is given by the expression

$$\frac{1}{\tau_{p_z\sigma}} = \sum_{\mathbf{p}'} W^+_{\mathbf{p}'\sigma,\mathbf{p}\sigma}\frac{p_z - p_z'}{p_z}.$$

Using Eqs. (2), (3), and (7), we obtain the relation for the current:

$$j_z = \frac{e}{V}\sum_{\mathbf{p},\,\mathbf{p}'} (l - l')\, f^{(0)}_{p_z\uparrow} w^-_{\mathbf{p}\uparrow,\,\mathbf{p}'\downarrow}\,\delta\left(\frac{p_z^2 - p_z'^2}{2m} + \omega - \omega_s\right), \tag{8}$$

where $l = v^z_{p_z}\uparrow\tau_{p_z}\downarrow$, $l' = v^z_{p_z'}\uparrow\tau_{p_z'}\downarrow$.

The asymmetric part of the photoexcitation probability can result also from neglect of interaction with impurities in first-order perturbation-theory series, on account of interference between F_1 and F_2:

$$w^{(1)}_{\alpha\beta} = {}^1\!/_2\pi\,\mathrm{Re}[(F_1)_{\beta\alpha}(F_2)_{\beta\alpha}^*], \tag{9}$$

where $\alpha = p\uparrow$, $\beta = p'\downarrow$. The expression for the matrix elements $(F_1)_{\beta\alpha}$ depends on the orientation of the magnetic field relative to the crystallographic axes.It follows from Eq. (27) of Ref. 1:

$$(F_1)_{\beta\alpha} = \frac{2^{1/2}eE_0\delta_0}{i\omega a^2}\, e_B\left(a^2p_z^2 - \frac{1}{2}\right)\delta_{\mathbf{pp}'}.$$

Here E_0 is the amplitude of the electric field of the electromagnetic wave, $e_B = e_-B_{(133)} + e_+B_{(233)}$, $e_\pm = 2^{-1/2}(e_x \pm ie_y)$, $\mathbf{e}$ is the polarization vector, and a is the magnetic length. The angular dependence is contained in the coefficients $B_{(j33)}$ (Ref. 1);

$$B_{(133)} = \cos 2\Phi\cos 2\Theta - {}^1\!/_2\, i\sin 2\Phi\cos\Theta(3\cos^2\Theta - 1), \tag{10}$$

$$B_{(233)}=-{}^3/_2\, i \sin 2\Phi \sin\Theta \sin 2\Theta, \qquad (11)$$

where Θ and Φ are the polar and azimuthal angles of the vector $\mathbf{H}$ in the system of the crystallographic axes.

For $(F_2)_{\beta\alpha}$ we have

$$(F_2)_{\beta\alpha}=2^{1/2}p_z\frac{eE_0\tilde{g}\omega_s e_+}{i\omega|g|}\delta_{pp'}.$$

from (9) we obtain from $W^{(1)}$:

$$w_{\alpha\beta}^{(1)}=\delta_{pp'}\frac{4\pi\delta_0\tilde{g}\omega_s e^2E_0^2}{|g|\omega^2a^2}p_z\left(a^2p_z^2-\frac{1}{2}\right)\mathrm{Re}(e_Be_+^*). \qquad (12)$$

It is seen from (8) that if one disregards scattering by impurities with spin flip and the nonparabolicity of the spectrum then the difference $l-l'$ vanishes because the spin subbands are identical and the momentum is conserved in the transition probability (12). For the effect not to become zero, account must be taken of the nonparabolicity of the spectrum in the calculation of $\tau_{pz\sigma}$ and $v^z_{p_z\sigma}$. Assuming the nonparabolicity to be small, we can represent the expression in the parentheses in (8) in the form $(\partial/\partial m)(\tau_{p_z\uparrow}v^z_{p_z\uparrow})|_{p_z}\Delta m$, where $\Delta mr\equiv m_\uparrow-m_\downarrow=-m\omega_s/\varepsilon_g$, and ε_g is the band gap. As a result we have

$$j_z^{(1)}=-\frac{e^3\delta_0\tilde{g}\omega_s^2E_0^2m}{\pi a^4|g|\varepsilon_g\omega^2}\int dp_z f^{(0)}_{p_z\uparrow}\frac{\partial}{\partial m}(\tau_{p_z\uparrow}v^z_{p_z\uparrow})|_{p_z}\times p_z\left(a^2p_z^2-\frac{1}{2}\right)P\delta_\eta(\Delta). \qquad (13)$$

Here $\delta_\eta(\Delta)=\eta/\pi(\Delta^2+\eta^2)$ is a "smeared" δ function, $\Delta=\omega-\omega_s$ is the detuning from resonance, and $P=\mathrm{Re}(e_Be_+^*)=\mathrm{Re}(e_-e_+^*B_{(133)})$. The damping $\eta\to+0$, however, as is customarily done, can be replaced in the final results, for comparison with experiment, by a finite quantity η indicative of the width of the transition.

For scattering by charged impurities

$$\tau_{p_i}=\varkappa^2|p_z|^3/2\pi e^4mn_i$$

($\varkappa$ is the dielectric constant and n_i is the impurity density). For this case we obtain from (13) where ε_B is the effective

$$j_z^{(1)}=-\frac{e^3\delta_0\tilde{g}\omega_s^2E_0^2(2m)^{3/2}n_e}{a^2|g|\varepsilon_gm\omega^2n_i\varepsilon_B}\langle\!\langle\varepsilon^{5/2}\rangle\!\rangle P\delta_\eta(\Delta), \qquad (14)$$

Bohr energy, and $\varepsilon=p_z^2/2m$ is the longitudinal energy of the electron. The double angle brackets denote averaging:

$$\langle\!\langle\ldots\rangle\!\rangle=\int dp_z f^{(0)}_{p_z\uparrow}(\ldots)\Big/\int dp_z f^{(0)}_{p_z\uparrow},$$

$$n_e=\frac{1}{(2\pi a)^2}\int dp_z f^{(0)}_{p_z\uparrow}$$

is the electron density. For nondegenerate case $\langle\!\langle\varepsilon^{5/2}\rangle\!\rangle=2\pi^{-1/2}T^{5/2}$.

Besides the considered contribution made to the current by interference of F_1 and F_2, there are also terms connected with allowance for the contribution made to w by the interaction of the electrons with the impurities. It may turn out that transitions with spin flip, with participation of impurities are not resonant, for in this case p_z is not conserved. It will be shown later, however, the FVE is determined in this case by a resonance in an intermediate state. The reason is similar to that of the onset of resonant FVE in a quantum film.[9,10]

Analysis shows that the resonant terms we need in the p_z-odd transition probability appear when account is taken of the interference of the transition amplitudes of the first and second order. It must also be borne in mind that the answer should contain the constant δ_0 and the second power of the impurity potential. For the corresponding contributions to w we obtain

$$w_{\alpha\beta}^{(2)}=\pi\Big\langle\,\mathrm{Im}\Big\{\Big[\frac{(F_1)_{\beta\gamma}U_{\gamma\alpha}}{i(\varepsilon_{\gamma\beta}+\omega)+\eta}+\frac{U_{\beta\delta}(F_1)_{\delta\alpha}}{i(\varepsilon_{\delta\alpha}-\omega)+\eta}\Big](F_U)_{\beta\alpha}^*\Big\}\Big\rangle, \qquad (15)$$

$$w_{\alpha\beta}^{(3)}=\pi\sum_{1,2}\Big\langle\,\mathrm{Re}\Big\{\frac{(F_{1,2})_{\beta\gamma}U_{\gamma\alpha}}{i(\varepsilon_{\gamma\beta}+\omega)+\eta}\Big(\frac{U_{\beta\delta}(F_{2,1})_{\delta\alpha}}{i(\varepsilon_{\delta\alpha}-\omega)+\eta}\Big)^*\Big\}\Big\rangle. \qquad (16)$$

Here $\varepsilon_{\gamma\beta}=\varepsilon_\gamma-\varepsilon_\beta$, ε_γ is the spectrum of the Hamiltonian $\mathscr{H}_0$; the subscripts $\gamma=\mathbf{p}'\uparrow$ and $\delta=\mathbf{p}\downarrow$ correspond to intermediate states. The transitions contributing to $w^{(2,3)}$ are shown schematically in Fig. 1. Equations (15) and (16) show that the resonance is due in this case to an intermediate state. The matrix elements of F_U and U contained in (15) and (16) are given by the relations

$$(F_U)_{\beta\alpha}=-\frac{2^{1/2}\alpha eE_0N_i}{i\omega}\sum_{\mathbf{q},j}u_{\mathbf{q}}\exp(-i\mathbf{q}\mathbf{r}_j)q_se_+J_{p',p}(q),$$

$$U_{p',p}=\sum_{\mathbf{q},j}u_{\mathbf{q}}\exp(-i\mathbf{q}\mathbf{r}_j)J_{p',p}(q),$$

where u_q is the Fourier component of the potential of an individual impurity, N_i is the total number of impurities, $J(\mathbf{q})=\exp(i\mathbf{q}\cdot\mathbf{r})$.

After substituting and averaging we get

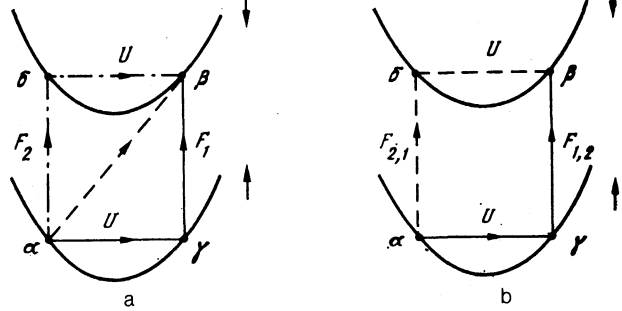

FIG. 1. Two types of transition that determine the contributions $j_z^{(2)}$ and $j_z^{(3)}$ to FVE in spin resonance; a—interference of transition matrix elements determined by F_U (dashed line), and of the composition matrix element of type $U\times F_1$ (solid line), or $F_1\times U$ (dash-dot line); b—interference of composite matrix elements $U\times F_{1,2}$ and $F_{2,1}\times U$. Momentum conservation in the transition dictates the resonant character of the process.

$$w_{\alpha\beta}^{(2)} = -\frac{2\pi e^2 E_0^2 \alpha \delta_0 N_i}{a^2\omega^2} \sum_{\mathbf{q}} q_z |u_{\mathbf{q}}|^2 |J_{p',p}(\mathbf{q})|^2$$

$$\times \operatorname{Im}\left\{ e_+^* e_B \left(\frac{a^2 p_z'^2 - 1/2}{\eta + i\Delta} + \frac{a^2 p_z^2 - 1/2}{\eta - i\Delta} \right) \right\},$$

$$w_{\alpha\beta}^{(3)} = \frac{4\pi e^2 E_0^2 \delta_0 \tilde{g}\omega_s N_i}{a^2\omega^2 |g|} \sum_{\mathbf{q}} |u_{\mathbf{q}}|^2 |J_{p',p}(\mathbf{q})|^2$$

$$\times \operatorname{Re}\left\{ e_+^* e_B \left[\frac{p_z (a^2 p_z'^2 - 1/2)}{(\eta + i\Delta)^2} + \frac{p_z' (a^2 p_z^2 - 1/2)}{(\eta - i\Delta)^2} \right] \right\}.$$

We introduce the notation

$$\mathscr{R}_l(\Delta) = \frac{n_i e V^2}{(2\pi)^5 m a^2} \int dp_z \int dp_z' \int d\mathbf{q}_\perp \exp\left(-\frac{q_\perp^2 a^2}{2} \right)$$

$$\times (\tau_{p_z p_z} - \tau_{p_z' p_z'}) f_{p_z\uparrow}^{(0)} |u_{\mathbf{q}_\perp, p_z - p_z'}|^2 \delta\left(\frac{p_z^2 - p_z'^2}{2m} + \Delta \right) \mathscr{B}_l,$$

where

$$\mathscr{B}_1 = (p_z' - p_z)(a^2 p_z^2 - 1/2),$$

$$\mathscr{B}_2 = (p_z' - p_z)(a^2 p_z'^2 - 1/2),$$

$$\mathscr{B}_3 = p_z (a^2 p_z'^2 - 1/2),$$

$$\mathscr{B}_4 = p_z' (a^2 p_z^2 - 1/2).$$

We have then for the current

$$j_z^{(2)} + j_z^{(3)} = \frac{e^2 E_0^2 \delta_0 2\pi}{a^2\omega^2} \left\{ -\alpha \operatorname{Im}\left(\left[\frac{\mathscr{R}_1(\Delta)}{\eta + i\Delta} + \frac{\mathscr{R}_2(\Delta)}{\eta - i\Delta} \right] e_+^* e_B \right) \right.$$

$$\left. + \frac{2\tilde{g}\omega_s}{|g|} \operatorname{Re}\left(\left[\frac{\mathscr{R}_3(\Delta)}{(\eta + i\Delta)^2} + \frac{\mathscr{R}_4(\Delta)}{(\eta - i\Delta)^2} \right] e_+^* e_B \right) \right\}.$$

In the case of short-range impurities ($u_q = u_0/V$, $u_0 = \text{const}$), the values of $\mathscr{R}_i$ are easily obtained:

$$\mathscr{R}_1 = \mathscr{R}_2 \left(1 - \frac{4\Delta}{\omega_c} \right),$$

$$\mathscr{R}_2 = \frac{n_e e}{2\pi} \left\langle\!\!\left\langle \theta(\varepsilon + \Delta) \frac{1}{(\varepsilon + \Delta)^{1/2}} [(\varepsilon + \Delta)^{1/2} + \varepsilon^{1/2}] \right\rangle\!\!\right\rangle,$$

$$\mathscr{R}_3 = -\frac{n_e e}{2\pi} \left(1 - \frac{4\Delta}{\omega_c} \right) \left\langle\!\!\left\langle \theta(\varepsilon + \Delta) \frac{\varepsilon^{1/2}}{(\varepsilon + \Delta)^{1/2}} \right\rangle\!\!\right\rangle$$

$$\mathscr{R}_4 = \frac{n_e e}{2\pi} \langle\!\langle \theta(\varepsilon + \Delta)(\varepsilon + \Delta) \rangle\!\rangle.$$

Here ω_c is the cyclotron frequency and $\theta(x)$ is the unit step function.

Let $\Delta \ll \langle\langle \varepsilon \rangle\rangle$, where $\langle\langle \varepsilon \rangle\rangle$ is the average energy of the longitudinal motion of the electron. In this case $p_z' \approx -p_z$ and $\mathscr{R}_1 = \mathscr{R}_2 = -2\mathscr{R}_3 = 2\mathscr{R}_4 = \mathscr{R} = \text{const}$. Assuming that the relaxation time is determined by scattering from the same potential U as in the Born approximation of $\mathscr{R}$, we get

$$\mathscr{R} = n_e e \langle\!\langle \varepsilon \rangle\!\rangle / \pi.$$

As a result we get for j_z

$$j_z^{(2)} + j_z^{(3)} = -\frac{4\pi\alpha\delta_0 e^3 n_e \langle\!\langle \varepsilon \rangle\!\rangle}{a^2\omega^2} E_0^2 S_\eta(\Delta) P', \qquad (17)$$

where

$$S_\eta(\Delta) = \delta_\eta(\Delta) - \frac{\tilde{g}\omega_s}{\alpha |g|} \delta_\eta'(\Delta)$$

is a function indicative of the frequency dependence of the current [$\delta_\eta'(\Delta)$ is the derivative of $\delta_\eta(\Delta)$], and the function

$$P' \equiv \operatorname{Im}(e_+^* e_B) = |e_+|^2 \operatorname{Im} B_{(233)} + \operatorname{Im}(e_+^* e_- B_{(133)})$$

determines its polarization dependence.

For the nondegenerate case we have $\langle\langle \varepsilon \rangle\rangle = T/2$.

Other contributions in the same order of theory (and proportional to $\delta_0 \tilde{g}$) could result from the contribution, odd in p_z, to the spectrum $\varepsilon_{p_z\sigma}$, but it is easy to verify that they vanish, at least in this order.

Note that expression (17) is valid for an arbitrary impurity potential.

In Eqs. (14) and (17) the line width was taken into account with the aid of a numerical parameter η. Such a model corresponds to line broadening by spin relaxation. A much greater role is played in InSb by broadening due to nonparabolicity of the electron spectrum. It can be shown that allowance for it reduces to the following replacement of the function $\delta_\eta(\Delta)$:

$$\delta_\eta(\Delta) \to F(\Delta)$$

$$= \int_0^\infty dx x^\nu f^{(0)}(Tx) \frac{\eta_s}{\eta_s^2 + (\Delta + \gamma x)^2} \Big/ \pi \int_0^\infty dx x^\nu f^{(0)}(Tx), \qquad (18)$$

where $\nu = 2$ for Eq. (14) and $\nu = 3/2$ for Eq. (17), $\gamma = 2\omega_s T/\varepsilon_g$ and η_s are parameters indicating respectively the nonparabolic broadening and the broadening to the spin relaxation. If the condition $\gamma \gg \eta_s$ is met, we have in the case of Boltzmann statistics

$$F(\Delta) = \frac{1}{\gamma} \left(-\frac{\Delta}{\gamma} \right)^\nu \frac{\exp(\Delta/\gamma)}{\Gamma(\nu + 1)} \theta(-\Delta), \qquad (19)$$

where $\Gamma(3) = 2$, $\gamma(\frac{5}{2}) = \frac{3}{4}\pi^{1/2}$.

Thus, allowance for nonparabolicity leads to an asymmetric line broadening. The contributions $j_z^{(1)}$ and $j_z^{(2)}$ remain constant in sign, while the sign of $j_z^{(3)}$ alternates.

Expression (17) shows that the polarization dependence of the current is the same for all parts near resonance. In the case of purely circular polarization it is determined by the quantity $\operatorname{Im} B_{(233)}$. In typical experimental geometries the field **H** is directed along [001], [110], or [111]. It can be seen from (10) that an effect on circular polarization is possible only in the [111] orientation, with $P' = -2/\sqrt{3}$ for cyclotron-inactive polarization (right-handed), and there is no effect for left-handed polarization. In the case of linear polarization the current differs from zero for the orientations [111] and [001]. In the first case there is no polarization dependence. In the second the sum $j_z^{(2)} + j_z^{(3)}$ has a polarization dependence of the form $\sin 2\varphi$, where φ is the angle between the vector **e** and the [100] direction. As for the term $j_z^{(1)}$, it is equal to zero for circular polarization, and differs from zero only at $\mathbf{H} \| [100]$, and depends on φ like $-\cos 2\varphi$. The values of P and P' for the principal directions

are listed in the table.

When comparing the sign of the current with experiment it must be remembered that the z axis is directed along the magnetic field. It is more convenient to use the current in the laboratory frame. In the [111] orientation $\mathbf{H} \to -\mathbf{H}$ and simultaneous change of the circular polarization or preservation of the linear one does not change the sign of the current. In the [001] orientation the polarization $j_z^{(1)}$ changes while $j_z^{(2)} + j_z^{(3)}$ does not change the sign when $\mathbf{H} \to -\mathbf{H}$.

In the orientation [100] and for linear polarization of the radiation it must be taken into account that the polarization is not preserved inside the sample, but is rotated by the Faraday effect. The measured signal is proportional in this case of $\int_0^d dz j_z$, where d is the sample thickness.

3. SAMPLE PREPARATION AND MEASUREMENT PROCEDURE

We used in the experiments n-type indium antimonide samples with concentration $n_{77\,K} = 1.3 \cdot 10^{14}$ cm^{-3}. The ingot was oriented by a standard x-ray procedure accurate to 30′. Samples measuring $3 \times 3 \times 10$ mm were cut from the ingot in such a way that the maximum dimension corresponded to one of the three crystallographic directions: ⟨100⟩, ⟨110⟩, and ⟨111⟩. Etching in a selective etchant made it possible to establish the crystallographic indices in absolute units.

Indium contacts were placed on the end faces of the samples, as shown in Fig. 2. The radiation source was an optically pumped submillimeter CH_3OH vapor laser. The experiments were performed at a fixed laser emission wavelength $\lambda = 118.8\,\mu$m. The intensity of the radiation incident on the sample was 5 mW/cm^2. The measurements were made with the radiation polarized both linearly and circularly by passing the beam through a crystalline quartz quarter-wave plate.

The samples were placed in an optical cryostat with a superconducting solenoid having a maximum induction 8 T. The inhomogeneity of the magnetic field in the volume occupied by the sample was $1 \cdot 10^{-4}$ of the maximum. The photoemf signal was measured by an amplifier with a lock-in detector. A germanium bolometer placed behind the sample measured the transmission coefficient. The Faraday-rotation angle of the polarization plane in the samples in a magnetic field was fixed by the maximum transmission through a polarizer located between the sample and the bolometer. The densities of the free carriers and that of the ionized impurities in the samples in a magnetic field were determined by measuring and analyzing the Hall effect and the mobility.

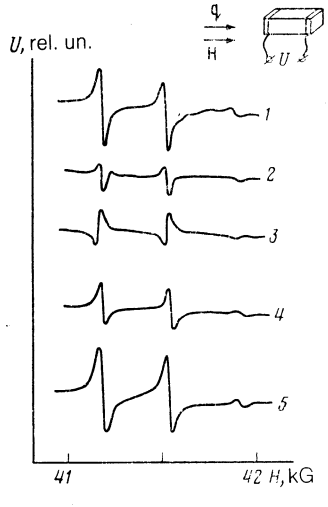

FIG. 2. Dependence of photoemf on the magnetic field at $\mathbf{H} \| [001]$: 1—$\varphi = 0°$, 2—30°, 3—60°, 4—90°, 5—120° (φ is the angle between the direction [100] and the polarization vector $\mathbf{e}$). Inset—geometry of experiment, $\mathbf{q}$—radiation wave vector.

4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

The experiments were performed in the Faraday geometry, $\mathbf{E} \perp \mathbf{H} \| \mathbf{q}$, where $\mathbf{q}$ is the radiation wave vector. The photoemf was measured along the magnetic field. For samples with principal crystallographic orientations we measured the magnetic-field dependences of the emf at various directions of the incident radiation, of the radiation polarization, and of the magnetic field. Typical plots are shown in Fig. 2. According to the results of an absorption experiment[11] the central peak corresponds to spin resonance by free carriers, and the peaks on the left and right of the peak correspond to impurity transitions.

The measured signals contain resonance peaks and nonresonant "base." For comparison with theory we shall analyze only the resonance peak corresponding to spin resonance by free carriers. The shape of the plot of the photoemf against the magnetic field contains a constant-sign component and an alternating-sign one. By analyzing the line shapes of the observed emf, photoconductivity, and absorption signals, we have verified that the line is better approximated by a Lorentz curve and its derivatives than by the dependences calculated from Eq. (19). It is possible that we failed to observe the line asymmetry due to nonparabolic broadening, owing to insufficient homogeneity of the magnetic field in the samples. We shall therefore describe the constant-sign peak by the function $\delta(\Delta) = \eta/\pi(\Delta^2 + \eta^2)$, and the alternating-sign peak by $-\delta'(\Delta) = -2\Delta\eta/\pi(\Delta^2 + \eta^2)^2$, where η is the half-width of the absorption line. Since the constant-sign component has a maximum at $\Delta = 0$, when the alternating-sign component vanishes, we shall compare hereafter the signal amplitudes at the extrema of the alternating-sign component, i.e., at $\Delta = \pm \eta/3^{1/2}$. The experimental curve was divided into two components by fitting the coefficient D in the expression. $\delta(\Delta) + D\delta'(\Delta)$ to obtain best agreement with the initial curve.

The photoemf signal contained a component that depends on the sign of the radiation vector, as well as an independent component. The FVE for a given direction of the polarization vector does not depend on the direction of $\mathbf{q}$. An FVE contribution odd in $\mathbf{q}$ can occur only in the orientation $\mathbf{H} \| [001]$, for in this orientation the current depends on the polarization, and the latter is varied in the sample by the Faraday effect which is odd in $\mathbf{q}$. In addition, polarization-independent effects odd in $\mathbf{q}$ exist, viz., the Dember emf[12] and the dragging effect,[13] as well as an emf even in $\mathbf{q}$ and due to inhomogeneity of the sample (these contributions are proportional to the absorption coefficient, which does not depend on the linear polarization). With account taken of this fact, the FVE effect was separated from the common signal in a different manner for each orientation.

Let us examine the behavior of the FVE for the princi-

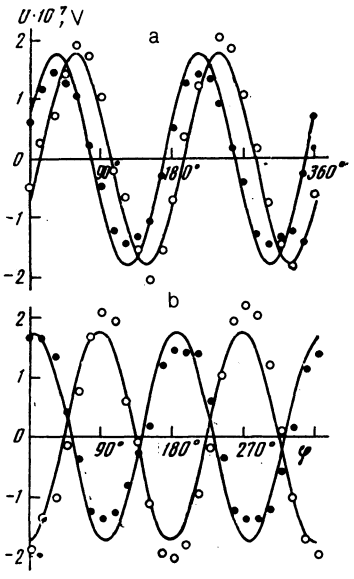

FIG. 3. Polarization dependence of FVE contribution even in Δ for two polarities of the magnetic field and directions of the incident radiation (a)—$\mathbf{q}\|[001]$, (b)—$\mathbf{q}\|[001]$. Here ○ corresponds to $\mathbf{H}\|[001]$ and ● to $\mathbf{H}\|[001]$. Curves—calculated from Eq. 20.

pal orientations of the crystal.

A. $\mathbf{H}\|[001]$. In this case the FVE was separated in accordance with its characteristic polarization dependence separately for the contributions even and odd in Δ. Figure 3 shows the experimental angular dependences of the even contribution to the FVE for two polarities of the magnetic field and directions of the incident radiation. To compare the theoretical relations with the experimental ones account must be taken of the Faraday effect. In a magnetic field corresponding to spin resonance, the density of the free carriers decreases on account of the magnetic freezeout, and the rotation of the polarization plane is due mainly to the Faraday effect on the "wing" of the impurity cyclotron resonance.[14] The Faraday angle remains practically unchanged within the limits of the spin-resonance line width.

Theoretically, contributions to the FVE are made in these orientations by $j_z^{(1)}$ and $j_z^{(2)}$—see the table and Eqs. (14) and (17). In this case $j_z^{(1)}$ depends on the magnetic-field direction, while $j_z^{(2)}$ does not. With allowance for the Faraday effect the expression for the FVE emf takes the form

$$U=\frac{(1-R^2)E_{\text{inc}}^2}{2\sigma_H n_\omega r}\delta(\Delta)(A^2+B^2)^{1/2}\sin(rd)\sin(rd+\varphi_0+2\varphi), \tag{20}$$

where

$$\operatorname{tg}\varphi_0=-\frac{A}{B}\operatorname{sgn}H,$$

$$\operatorname{sgn}H=\begin{cases}+1, & \mathbf{H}\|[001]\\ -1, & \mathbf{H}\|[00\bar{1}]\end{cases}$$

$$A=-\frac{2\delta_0\tilde{g}(2m)^{5/2}(KT)^{5/2}e^3n_e\omega_s^2}{\hbar^5\pi^{1/2}a^2|g|\varepsilon_g\varepsilon_B n_i\omega_0^2 m}$$

$$B=-\frac{2\pi\alpha\delta_0 e^3 n_e KT}{\hbar^4 a^2\omega^2}$$

φ is the angle between the polarization vector of the incident radiation and the [100] direction, R is the coefficient of reflection from the front and rear faces of the sample, σ_H is the longitudinal conductivity in the magnetic field corresponding to the spin resonance, E_{inc} is the intensity of the electric field of the wave incident on the sample, and n_ω is the refractive index. In this equation account is taken of the rays transmitted and reflected by the rear face of the sample, but their interference is neglected, since under the experimental conditions these faces were not parallel with accuracy sufficient for interference. We have also neglected here the damping of the radiation intensity in the sample, since the experimentally measured absorption coefficient was 0.01 cm^{-1}.

The Faraday angle rd was determined by measuring the rotation of the polarization plane of the wave passing through the sample, and turned out to equal 0.7 rad.

The angular dependence in (20) contains coefficients A and B determined by the band parameters δ_0, $\tilde{g}$, and α. The parameter δ_0 is determined from experiments on absorption under spin-resonance conditions,[3] where the interference between the electrodipole and magnetodipole transitions is clearly pronounced and turns out to equal $3.6\cdot10^{-34}$ $\text{erg}\cdot\text{cm}^3$. By varying the ratio A/B it is possible to approximate well the experimental polarization dependences for $A/B=1$ and 2 (see Fig. 3). Substituting in the theoretical expression

$$\frac{A}{B}=\frac{2^{5/2}\tilde{g}\omega_s^2 m^{3/2}(KT)^{3/2}}{\pi^{3/2}|g|\hbar\varepsilon_g\varepsilon_B n_i\alpha}$$

$\varepsilon_B=0.6$ meV, $m=0.0135m_0$ $\varepsilon_g=0.236$ eV, $|g|=51$, $n_i=1.1\cdot10^{14}$ cm^{-3}, we obtain $\tilde{g}/\alpha=26$. Substituting the experimental value of the signal in (20) and recognizing that $a=1.26\cdot10^{-6}$ cm, $n_\omega=4.2$, $\eta=5\cdot10^9$ s^{-1}, $n_e=1\cdot10^{13}$ cm^{-3}, $\sigma_H=2.5\cdot10^{-2}$ $\Omega^{-1}\cdot\text{cm}^{-1}$, we obtain $\tilde{g}=-1.9\cdot10^{-12}$ cm^2, and then $\alpha=-7\cdot10^{-14}$ cm^2. Such values of the parameters $\tilde{g}$ and α are in good agreement with the theoretical values calculated in Kane's model: $\tilde{g}=1.2\cdot10^{-12}$ cm^2 (Ref. 15), $\alpha=-\hbar^2/4m\varepsilon_g$

TABLE I. Values of P and P' for various orientations and polarizations.

Polarization	[001]		[110]		[111]	
	P	P'	P	P'	P	P'
Right-hand circular	0	0	0	0	0	$-2\cdot3^{-1/2}$
Left-hand circular	0	0	0	0	0	0
Linear	$-1/2\cos2\varphi$	$1/2\sin2\varphi$	0	0	0	$-3^{-1/2}$

$= -5\cdot10^{14}$ cm^2 (Ref. 16), but differ greatly from $g = -1\cdot10^{-11}$ cm^2 obtained in Ref. 17.

Figure 4 shows the angular dependences of the odd contribution to the FVE for two polarities of the magnetic field in the case of linear polarization of the radiation. In the theoretical part of the paper it is described by the expression for $j_z^{(3)}$ and is independent of the magnetic-field direction. A dependence on the direction of **H** appears when account is taken of the Faraday effect. The expression for the emf takes then the form

$$U = -\frac{(1-R)(1+R)E_{inc}^2 BC\delta'(\Delta)}{2\sigma_H n_\omega r}\sin(rd)\sin(rd+2\varphi), \tag{21}$$

where $C = -\tilde{g}\omega_s/|g|\alpha$. The angular dependence plotted in accordance with this equation describes the experimental one well, but the amplitude of the effect turned out to be $3.9\cdot10^{-4}$, much higher than the experimental $5\cdot10^{-7}$ V.

The polarization-independent resonant contribution to the total signal has components dependent on and independent of **q**. The more significant is the **q**-dependent component, which duplicates the line shape of the peak that is odd in Δ. We attribute this component to a photon dragging effect of the type of Ref. (13). The component independent of **q**, which has the line shape of the peak that is even in Δ, we attribute to the emf on the sample inhomogeneities. The dragging effect is 1.2 times larger than the FVE at the extrema of the polarization dependence of the latter, while the emf on the sample inhomogeneities is ~15% of the odd contribution to the FVE.

B. **H**||[110]. At this orientation there is not FVE theoretically and has not been observed in experiment.

C. **H**||[111]. In this case analysis of the experimental results has shown that the measured signals are independent of the angle between the linear-polarization vector and the crystallographic directions in the (111) plane. To separate from the experimental curves obtained for opposite directions of **q** the signal components that are dependent on or independent of the sign of the radiation vector, we have constructed the following combinations:

$$U^+ = [U(+\mathbf{q}) + U(-\mathbf{q})]/2,$$
$$U^- = [U(+\mathbf{q}) - U(-\mathbf{q})]/2.$$

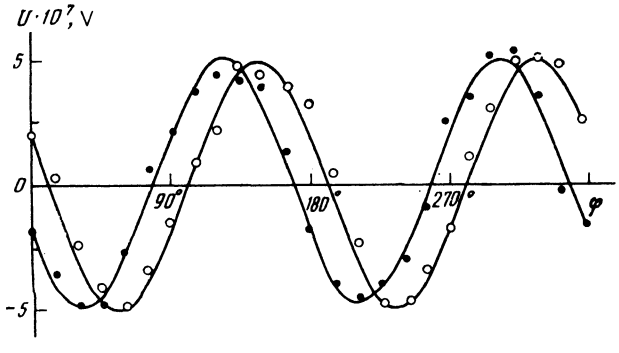

FIG. 4. Polarization dependence of the emf on FGE odd in Δ at two polarities of the magnetic field: ○—**H**||[001], ●—**H**||[001]. Curves—calculation by Eq. (21).

The values of U^+ and U^- were determined for two polarizations of the magnetic field relative to the [111] direction. Here U^+ is independent of the sign of q and is determined by the FVE and the resonance photoemf connected with the inhomogeneity of the samples. It is impossible to separate experimentally one from the other by our measurement procedure, but judging from analysis of the results in the **H**||[001] geometry for the contribution even in Δ, where the FVE can be separated by means of the polarization dependence, the value of the emf on the inhomogeneities is not more than 15% of the FVE. We shall assume with the same accuracy that U^+ is determined by the FVE.

Figure 5 shows the dependences of the FVE signals on the magnetic field for linear as well as right- and left-circular polarizations relative to the magnetic-field direction. It is seen from the figure that the effect exists only for linear and right-circular polarizations, with the signal amplitude for circular double that for linear, in agreement with the theory (see the table). Change of the sign of the magnetic field does not influence the magnitude of the effect for linear polarization of the radiation. The signal contains contributions even and odd in Δ. In this orientation, as seen from the table, only $j_z^{(2)}$ and $j_z^{(3)}$ are manifested. Since there is no angular polarization dependence at such an orientation, the Faraday effect does not influence the magnitude of the signal. The FVE emf for the even contribution is described, for linear polarization, by the expression

$$U = \frac{(1-R^2)E_{inc}^2 d}{3^{1/2}\sigma_H n_\omega} B\delta(\Delta).$$

From a comparison of the theoretical value of the signal and the experimental one we obtain $\alpha = 5.7\cdot10^{-14}$ cm^2, in good agreement with the value obtained for this parameter from the even contribution to the orientation **H**||[100]. The odd contribution for the FVE emf for linear polarization is given by

$$U = -\frac{(1-R^2)E_{inc}^2 d}{3^{1/2}\delta_H n_\omega} BC\delta'(\Delta).$$

Substituting here the parameter α, determined in the geometry **H**||[001] from the contribution even in Δ, we obtain a

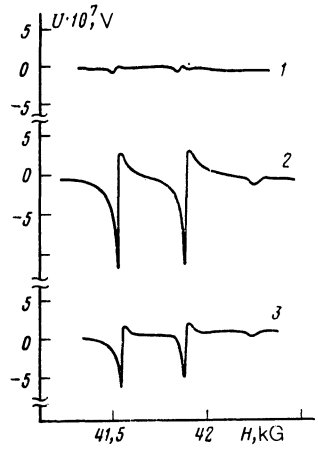

FIG. 5. Dependence of FVE emf on the magnetic field for various polarizations of the incident radiation: 1—left-hand circulation, 2—right circulation, 3—linear.

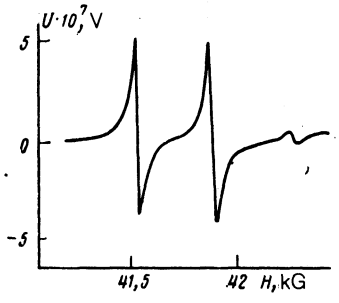

FIG. 6. Dependence of the photon dragging effect on the magnetic field at $\mathbf{H}\|[111]$.

signal emf equal to $4.7\cdot10^{-1}$ V, just as in the case of $\mathbf{H}$ [001].

The value of U^{-1} depends on the sign of $\mathbf{q}$ and is determined by the photon dragging effect and by the Dember photoemf. For comparison with the FVE, the dependence of U^{-} on the magnetic field is shown in Fig. 6, from which it is seen that the spin-resonance curve is antisymmetric, attesting to a small gradient effect, which has a constant-sign magnetic-field dependence.

Note that in our experiments the FVE emf was enhanced and the dragging-effect emf was weakened by reflection of the radiation from the rear face of the sample. In experiments with a tilted rear face of the sample the ratio of the dragging effect to the FVE was approximately double, so that the accuracy with which the FVE was separated was lower.

CONCLUSION

Thus, the proposed theory of the effect describes well the observed polarization dependences in the considered orientations of the magnetic field relative to the crystallographic directions. Comparison of the theoretical and experimental values of the signals for the FVE contribution even in Δ makes it possible to determine the parameters $\tilde{g}$ and α. These parameters are in good agreement with their values calculated in the Kane model. The theoretical value of the contribution odd in Δ is larger by approximately three orders than the experimentally observed one. This can be due in part to the fact that the inhomogeneity of the magnetic field in the volume occupied by the sample suppresses an alternating-sign signal, but has little effect on the value of the constant-sign contribution. Other contributions, not accounted for by the theory and describing the peak odd in Δ, are also possible.

The impurity peaks shown in Fig. 2 behave in experiment almost in the same way as the peak of the spin resonance on the free carriers. According to the selection rules, only intracenter transitions between bound states are possible. Free electrons are therefore created in such transitions via autoionization processes. The contribution of these states to the photomagnetic effects for impurity cyclotron transitions and impurity spin transitions have been discussed earlier. Excited impurity states act in the FVE as intermediate states, the final state being free. The frequency of the transition between impurity levels acts as a resonance frequency.

A detailed investigation of FVE on impurity transitions is beyond the scope of this article.

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