Observation of a photo-emf that depends on the sign of the circular polarization of the light

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A new photogalvanic effect, predicted by Ivchenko and Pikus [JETP Lett. **27**, 604 (1978)], has been observed. When a tellurium crystal oriented along the C_3 axis was exposed to CO_2-laser radiation a photo-emf was observed, which reversed sign with reversal of the circular polarization of the light. The magnitude of the effect agrees with the theoretical estimates.

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Ivchenko and Pikus[1] predicted a new photogalvanic effect in gyrotropic crystals, namely the occurrence of a photo-emf proportional to the degree of circular polarization P_{circ} of the exciting light, such that its sign changes with reversal of the sign of P_{circ}. The effect is due to singularities of the band structure of gyrotropic crystals and can occur in either interband or intraband absorption of light. In the present article we report observation of this effect in tellurium in which light was absorbed by free carriers.

The experiments were performed on undoped tellurium crystals with residual acceptor density $N_a \approx 2\times 10^{14}$ cm^{-3}. At $T=300$ °K these crystals have an intrinsic conductivity with $n_i \approx 5\times 10^{15}$ cm^{-3}. The samples were cylinders having axes coinciding with the C_3 axis of the crystal (cylinder length $L=0.8$ cm, area $S=3.1\times 10^{-2}$ cm^2). Annular contacts were deposited near the end faces of the cylinder. The radiation source was a pulsed CO_2 laser of 3 kW power and 100 nsec pulse duration ($\lambda=10.6$ μm). The linearly polarized laser light passed through a quarter-wave CdS plate, rotation of which could be used to vary $P_{\text{circ}}=\sin 2\phi$, where ϕ is the angle between the optical axis of the $\lambda/4$ plate and the laser-light polarization plane. We measured the photo-emf produced between the contacts when light was propagated along the C_3 axis, as a function of the rotation angle ϕ. Figure 1 shows the results of measurements made at $T=300$ K. It is seen that, in agreement with predictions of the theory,[1] the photo-emf is proportional to the degree of the circular polarization of the radiation $V_\phi = V_\phi^0 P_{\text{circ}}$. Besides the photo-emf proportional to R_{circ}, an emf V_{drag} due to the dragging of the carriers, and independent of P_{circ}, should appear at the employed geometry. As seen from Fig. 1, the dc component of the photo-emf is larger by at least one order of magnitude than V^0, in agreement with measurements of V_{drag} in[2].1) When the temperature is lowered to 150 K, the observed effect decreases sharply by more than one order of magnitude. As noted in[1], in the case of intraband light absorption with participation of phonons or impurity centers, the effect in question is determined only by the contribution of the virtual transitions through other bands, whereas transitions through states within one and the same band make no contribution to the effect.

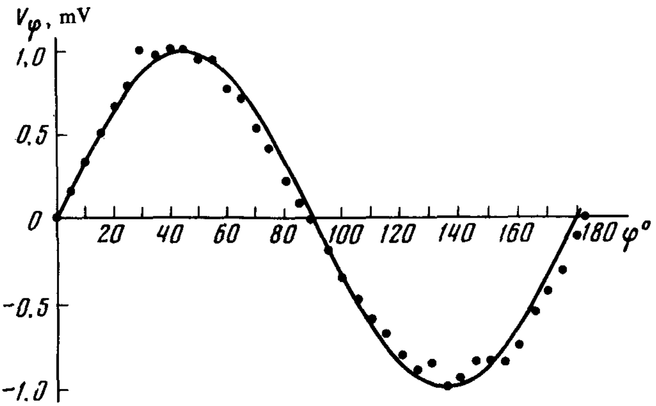

FIG. 1. Photo-emf produced in illuminated tellurium vs. the rotation angle ϕ of the $\lambda/4$ plate. The solid line is a plot of $V_\phi = V^0_\phi \sin 2\phi$ and $V^0_\phi = 1$ meV.

We shall consider the photo-emf connected with light absorption by free electrons in tellurium and produced as a result of virtual transitions through two nearest valence bands v_1 and v_2 (Fig. 2). The wave functions of the electrons in these bands are given by[3]

$$\Psi^e_{v_i} = C^i_{3/2}|3/2\rangle + C^i_{-3/2}| - 3/2\rangle, \tag{1}$$

where

$$C^1_{3/2} = -C^{(2)}_{-3/2} = \left(\frac{E+\beta k_z}{2E}\right)^{1/2}; \quad C^{(1)}_{-3/2} = C^{(2)}_{3/2} = \left(\frac{E-\beta k_z}{2E}\right)^{1/2}, \quad E = (\Delta^2+\beta^2 k_z^2)^{1/}$$

It is seen from (1) that in the band v_1 the predominant contribution is made at $k_z > 0$ by states with angular momentum $m_z = 3/2$, and at $k_z < 0$ by states with $m_z = -3/2$ (at $\beta > 0$); in the v_z band, on the other hand, the situation is reversed. It is precisely this connection between the angular momentum m_z and the momentum k_z which produces the effect in question. Upon excitation by light with right-hand circular polarization $\sigma_+ (e_- = (e_x - ie_y)/\sqrt{2} = 1)$ only optical transitions $|c, 1/2\rangle \to |v, 3/2\rangle$ and $|v, -3/2\rangle \to |c, -1/2\rangle$ are allowed. Therefore in the case of virtual interband transition through the v_1 band followed by absorption of a σ_+ photon and emission (absorption) of a phonon (Fig. 2a) the corresponding transition matrix element is proportional to $|C^{(1)}_{3/2}(k_{z1})^2|$ and does not depend on k_{z2}. In the subsequent emission (absorption) of a phonon and absorption of a photon (Fig. 2a), the transition matrix element is proportional to $|C^{(1)}_{-3/2}(k_{z2})|^2$ and does not depend on k_{z1}. Therefore, as seen from (1), the electrons with $k_{z1} > 0$ depart predominantly on going through the band v_1, and arriv-

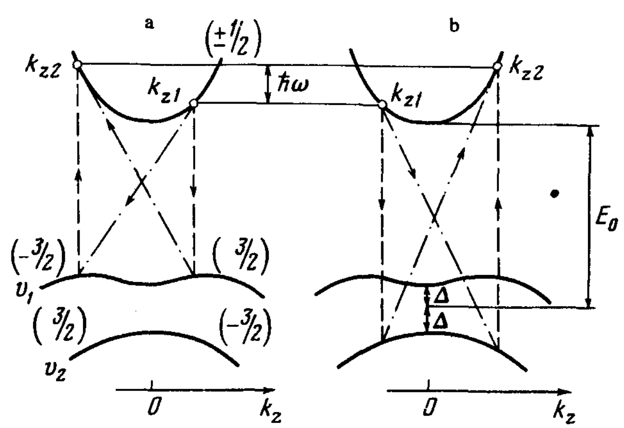

FIG. 2. Pattern of virtual transitions through the valence bands v_1 and v_2 upon absorption of light by free electrons. Dashed–transitions with absorption of σ_+ photon of energy $\hbar\omega$, dash-dot–transition with phonon participation.

ing electrons are predominantly those with $k_{z2} < 0$, and it is this which produces the current J^2_{z1}. In virtual transitions through the band v_2, on the contrary, the departing electrons are predominantly those with $k_{z1} < 0$ and the arriving ones with $k_{z2} > 0$ (Fig. 2b). Owing to the difference between the energy denominators in the composite matrix elements of the transitions through bands v_1 and v_2 there is no complete mutual cancellation of the currents, and the probability of transition of an electron from the state $\mathbf{k}_1$ to the state $\mathbf{k}_2$, with the contributions of both bands taken into account, is proportional to

$$1 + P_{\text{circ}}\, \beta (k_{z1} - k_{z2})/E_0 .$$

The general expression for the electron photocurrent can be written in the form

$$J^e_{\phi_z} = e I k' (\bar{V}_{z1} \tau_1 - \bar{V}_{z2} \tau_2), \tag{2}$$

where I is the total photon flux incident on the crystal, k' is the absorption coefficient connected with the virtual transtions through the bands v_1, v_2, and $\bar{V}_{1,2}$ is the average velocity of the optically excited electrons in the initial and final states, respectively, and $\tau_{1,2}$ is their momentum relaxation time. When account is taken of the short-range interaction Γ_{31} and Γ_{32} with the optical phonons,[4] under the conditions $k_B T \ll \hbar\omega \ll E_0$ and $\Delta \ll E_0$, the coefficient k' is given by

$$k' = \frac{e^2}{\hbar c n_\perp} \frac{m^c_\perp}{m^*} \left(\frac{2 m^c_\parallel}{\hbar\omega} \right)^{1/2} \frac{N}{\rho E_0} \sum_{\nu = 1,2} \frac{|D_\nu|^2}{\omega_\nu} \operatorname{cth} \frac{\hbar \omega_0}{2 k_B T} . \tag{3}$$

Here N is the electron concentration, $m^c_\parallel$ and $m^c_\perp$ are the effective masses of the electrons, ρ is the density, $1/m^* = 2|P_1|^2/m_0^2 E_0$, p_1 is the interband matrix element of the momentum operator, $n_\perp = 4.8$ is the refractive index, $D_{1,2}$ are the constants of the deformation potential for the interband transitions with participation of optical phonons,[5] and $\omega_{1,2}$ are the phonon frequencies ($\hbar\omega_1 = 11.4$ meV, $\hbar\omega_2 = 17.6$ meV). In the same approximation, the average electron velocities are[2]

$$V_{z1} = v_0 \frac{k_B T}{E_0} P_{\text{circ}} \quad , \quad V_{z2} = -\frac{2}{3} v_0 \frac{\hbar\omega}{E_0} P_{\text{circ}} \quad , \quad v_0 = \frac{\beta}{\hbar} = 4 \cdot 10^7 \text{ cm/sec} . \tag{4}$$

The expression for the photocurrent J^h_ϕ connected with light absorption by free holes differs from that given by $(2-4)$ by a factor of the order of $(\hbar\omega/2\sqrt{\Delta(\Delta+3\hbar\omega)}$. The experimentally measured emf is

$$V_{\phi_z} = L(J^e_{\phi_z} + J^h_{\phi_z})/S(\sigma_e + \sigma_h), \tag{5}$$

where σ_e and σ_h are the electron and hole conductivities.

Let us estimate the constants $D_{1,2}$ by comparing the theoretical formulas for V_ϕ with the experimental value of V^0_ϕ (Fig. 1). At room temperature the main contribution to the conductivity is made by the electrons, i.e., $\sigma_e > \sigma_h$, therefore the main contribution to J_ϕ is apparently also determined by the electrons. The predominant momentum-scattering mechanism is here the scattering by polar optical oscillations. If we confine ourselves to allowance for the electron contribution and assume that $\tau_1 = \tau_p$, where τ_p is the transport time $\tau_2 = \tau_p(\hbar\omega/k_B T)^{1/2}, m^c_\perp/m^* = 1$, $m^c_\parallel = 0.06 m_0$, and $|D_1| = |D_2|$, then the experimental values of I, S, L, and V^0_ϕ correspond to $D = 2 \times 10^8$ eV/cm. For the quantity $D\Omega^{1/3}$, where Ω is the volume of the unit cell, we obtain the value 9 eV. It is possible that this value of D is overestimated, since an appreciable contribution to k' can be made by transition with emission or absorption of acoustic phonons.

The experimentally observed decrease of V_ϕ with decreasing temperature can be due to the relative smallness of $J^h_{\phi_z}$ compared with $J^e_{\phi_z}$ as well to a decrease of the values of $\coth(\hbar\omega_\nu/2k_B T)$ and τ_2/τ_p.

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[1] We note that in the case of weak absorption of the light, its reflection causes V_{drag} to decrease by a factor $(1-R)/(1+R)$ (R is the reflection coefficient) whereas V_ϕ, which does not depend on the direction of the exciting beam, does not contain such a factor.

[2] We note that the electron currents have the same direction in intraband and interband absorptions.

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