SPACE-OSCILLATING PHOTOCURRENT IN CRYSTALS WITHOUT SYMMETRY CENTER

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A theory of a new-type of photocurrent in crystals without symmetry center is suggested. The photocurrent is different from zero in the case when the light polarization does not coincide with the crystal optical axes. The current direction is constant for the whole crystal, however, the magnitude of the current oscillates along the direction of light propagation.

A constant photocurrent caused by homogeneous illumination without an electric field has been observed in some experiments [1–3] and is intensively studied at the present time. A consistent theory of the effect was suggested in the papers [4–6]. The microscopic origin of the photocurrent is the asymmetry of the processes of ionization and recombination of the electrons and holes that leads to the flow of carriers in the conduction band. However, the theory of the effect [4–6] was constructed for linearly polarized light. Let us take into account ellipticity of the light with frequency ω, that is the complexity of the light wave field $\boldsymbol{E}(\omega)$. We obtain a fundamentally new contribution in the photocurrent:

$$j_l^{\mathrm{P}}(\omega) = \mathrm{i}\beta_{ln}(\omega)[\boldsymbol{E} \times \boldsymbol{E}^*]_n, \quad c|\boldsymbol{E}(\omega)|^2 = 4\pi I, \tag{1}$$

where c is the light velocity, I is its intensity. The photogalvanic tensor β_{ln} is different from zero in crystals without symmetry center, because the current changes sign due to space reflections. The photocurrent (1) is not space-homogeneous. Indeed, crystals without symmetry center have one or two optical axes. Consequently, the eigenpolarizations of the light in those crystals are linear polarizations and the light wave field is space modulated:

$$\boldsymbol{E}(\boldsymbol{x}) = E_\mathrm{e}\boldsymbol{e}_\mathrm{e} \exp(\mathrm{i}n_\mathrm{e}\boldsymbol{q}\cdot\boldsymbol{x}) + E_0\boldsymbol{e}_0 \exp(\mathrm{i}n_0\boldsymbol{q}\cdot\boldsymbol{x}), \tag{2}$$

where E_e, E_0 are the projections of the electric field on the optical axes at the point $\boldsymbol{x} = 0$, $\boldsymbol{q}$ is the wave vector of the light, $|\boldsymbol{q}| = \omega/c$. Substituting eq. (2) in eq. (1) we obtain a space-oscillating current:

$$j_l^{\mathrm{P}}(\boldsymbol{x}) = \mathrm{i}\beta_{ln}[\boldsymbol{e}_\mathrm{e} \times \boldsymbol{e}_0]_n \times (E_\mathrm{e}E_0^* \exp(\mathrm{i}(n_\mathrm{e} - n_0)\boldsymbol{q}\cdot\boldsymbol{x}) - \mathrm{h.c.}). \tag{3}$$

From eq. (3) it follows that the photocurrent oscillates when the coordinate $\boldsymbol{x}$ changes along the wave vector $\boldsymbol{q}$ with period $l_0 = (q|n_\mathrm{e} - n_0|)^{-1}$. This type of photocurrent can be measured in thin films of a few microns thickness, $l \lesssim l_0$, or in crystals with a large absorption coefficient κ. Another method of registration of the photocurrent can be found on the use of electrodes in the form of a lattice with period l_0. The space structure of the photocurrent for a transparent ferroelectric crystal is represented in fig. 1. The current is directed along the vector $[\boldsymbol{q} \times \boldsymbol{c}]$ ($\boldsymbol{c}$ is the crystal axis) and it should lead [1] to the optical recording of the holographic lattice with period l_0.

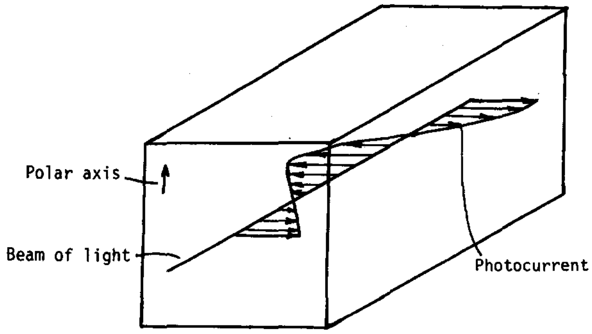

Fig. 1. Space structure of the photocurrent.

The microscopic mechanism of the effect is associated with the asymmetry of the process of electron and hole ionization. The processes of recombination do not contribute to current (1) because photocurrent (1) tends to zero after averaging with respect to the light polarization. For, without phonon optical transition of electrons (holes) in the conduction band, the photocurrent, associated with the process of ionization, can be represented in the form [5]:

$$j = e\int \nabla_k \epsilon_k f_k \mathrm{d}k$$
$$= (ec/4\pi\hbar\Gamma_i)(\kappa/\omega)(\overline{|D_k E|^2\,\nabla_k\epsilon_k})/\overline{|D_k E|^2}\,, \qquad (4)$$

where f_k is the electron distribution function with respect to momentum, ϵ_k is the dispersion law, Γ_i is the frequency of isotropization of the electron with respect to momentum, $\boldsymbol{D}_k$ is a dipole matrix element for electron transition in the band with momentum $\boldsymbol{k}$, the bar means averaging over the constant energy surface. Formula (4) is valid both for impurity-band transitions and for interband transitions. Only the particular expression for the absorption coefficient κ changes. The quantity $|\boldsymbol{D}_k \boldsymbol{E}|^2$ can be transformed:

$$2|\boldsymbol{D}_k\boldsymbol{E}|^2 = (|\boldsymbol{D}_k\boldsymbol{E}|^2 + |\boldsymbol{D}_k^*\boldsymbol{E}|^2) + [\boldsymbol{D}_k \times \boldsymbol{D}_k^*]\,[\boldsymbol{E}\times\boldsymbol{E}^*]\,. \qquad (5)$$

The symmetry property $\boldsymbol{D}_k^* = \boldsymbol{D}_{-k}$ results from the invariance under time inversion. Consequently the first term in eq. (5) is an even function of $\boldsymbol{k}$ and does not contribute to the current. The second term in eq. (5) is an odd function of $\boldsymbol{k}$ and leads to current (1) [‡1]. There is no necessity to take into account the Bloch function distortion of the electron in the neighbourhood of the impurity for the impurity-band transition and the electron–phonon interaction for the interband transition. Taking into account such effects is necessary for the calculation of the constant photocurrent [5].

The photocurrent can be easily calculated with a small momentum of excitation k_0, $k_0 a \ll 1$, where a is the lattice constant. If we neglect the crystal anisotropy then one has at small $\boldsymbol{k}$:

$$\epsilon_k = \epsilon_{-k} = k^2/2m, \qquad D_k^i = fc_i + \mathrm{i}gk_i + h_{iln}k_l k_n\,, \qquad (6)$$

where m is the electron mass, the constants f, h_{iln} characterize the crystal asymmetry. If the crystal polar axis is absent then $f = 0$. Let us substitute eqs. (5) and (6) in eq. (4) and obtain an expression for the photogalvanic tensor β_{ln}:

$$\beta_{ln} = \left(\frac{ec}{60\pi\hbar\Gamma_i}\right)\left(\frac{\kappa k_0^2\hbar}{m\omega}\right)\frac{g\epsilon_{nab}(5\delta_{al}c_b f \pm 2k_0^2 h_{abl})}{(3|\boldsymbol{c}\cdot\boldsymbol{e}|^2 f^2 + k_0^2 g^2)}\,, \qquad (7)$$

where $\boldsymbol{e}$ is the vector of light polarization, ϵ_{nab} is a unique antisymmetrical tensor. Let us estimate the magnitude of the photocurrent. We estimate f and h_{abl} in the following manner: $f \approx \xi g/a$, $h_{abl} \approx \xi g a$, where ξ is the characteristic parameter of the crystal asymmetry, $\xi \approx 10^{-1}$–10^{-2}. Let us choose $k_0 a \approx 10^{-1}$, $a \approx 10^{-8}$ cm, $\hbar/m\omega a^2 \approx 1$, $\Gamma_i \approx 10^{13}\mathrm{s}^{-1}$, the photocurrent associated with the polar axis has the order of magnitude $j^{\mathrm{p}} \approx \xi(eI/\hbar\Gamma_i)\kappa a \approx 10^{-7} \times \kappa I$(a/cm wt). The photocurrent associated with the third rank tensor h_{abl} is less by two orders according to our estimation. The space-oscillating photocurrent exceeds the constant photocurrent, which was calculated in ref. [5], a parameter $(\hbar^2/e^2 ma)(k_0 a)^{-1} \approx 10$.

The photocurrents observed in experiments also have their magnitude smaller by this order. However, special conditions for observation of the space-oscillating photocurrent are necessary.

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‡1 The asymmetry of a matrix element of dipole momentum was noted earlier in paper [7], however, the asymmetry of renormalization of the electron dispersion law due to the light which was considered in paper [7] does not lead to a photocurrent.