Photogalvanic effects in bismuth silicate ($Bi_{12}SiO_{20}$)

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The existence of a stationary current is observed in a gyrotropic $Bi_{12}SiO_{20}$ crystal excited by both linearly and circularly polarized light.

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Photogalvanic effects (PGE), whose magnitude and sign are determined by polarization of the incident light exhibit a relationship between current and field of the light wave with arbitrary polarization given by the general expression[1,2]

$$j_i^{PG} = I\{\alpha_{ijk}[(e_j e_k^* + e_j^* e_k)/2] + \gamma_{il}[i(\mathbf{e}\times\mathbf{e}^*)_l]\}, \quad (1)$$

where I is light intensity, e_i is the projection of polarization vector **e**, α_{ijk} is a third-rank tensor which is nontrivial in any crystal without an inversion center, γ_{il} is a tensor which is nontrivial only in gyrotropic crystals and is analogous to the gyration tensor g_{ik} which determines the natural optical activity.

The PGE occurring in the case of illumination by linearly-polarized light (first term in Eq. (1)) have been observed in a number of materials (e.g., Refs. 3 and 4); those dependent on the sign of circular polarization were theoretically predicted in Ref. 2, and experimentally identified only in Te.[5] In a $Bi_{12}SiO_{20}$ crystal which pertains to the 23 cubic point group, both the aforementioned tensors contain nontrivial components, a fact which enabled the authors to observe both effects (subsequently called "linear" and "circular" PGE).

Experiments were carried out in an undoped $Bi_{12}SiO_{20}$ specimen prepared in the shape of a plate cut perpendicularly to the [100] axis (plate thickness $L = 0.08$ cm). The polished plate surface was coated by means of an ion-plasma sputtering method with semi-transparent platinum electrodes (electrode surface ~ 1 cm^2). The light source was an argon-ion laser operating at $\lambda = 0.488$-μm spectral line. Absorption in this spectral region in undoped $Bi_{12}SiO_{20}$ is associated with deep acceptor levels ($N_A \sim 10^{19}$ cm^{-3}).[6,7]

To measure PGE, modulation methods were used which are based on the variation of incident light polarization in the mutually orthogonal directions for experiments with linear polarization, and variation in the direction of rotation for experiments with circular polarization. The need to use such methodology was governed by the appearance of relatively high (~ 10 mbarn) photo-emf in the case of stationary illumination evidently due to the Dember effect and the photovoltaic effects at the boundary of the metallic electrode and a crystal. Polarization modulation was accomplished by means of a ML-3 electrooptic modulator with a remote polarization prism at the output. A control voltage with an amplitude equal to the half-wave voltage $U_{\lambda/2} = 450$ V was applied to the modulator at a 40-Hz frequency. The initial condition of

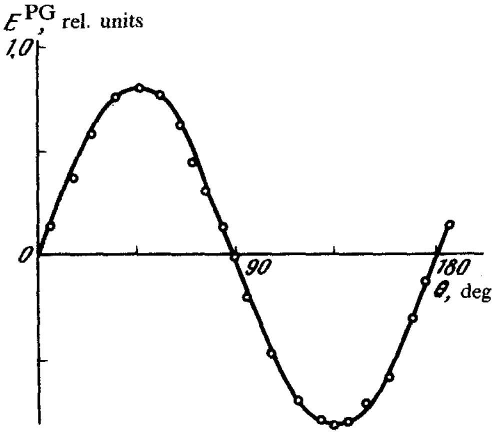

FIG. 1. Orientation dependence of "linear" photogalvanic effect (photo-emf) in $Bi_{12}SiO_{20}$.

light polarization at the modulator output was determined by the choice of a working point (value of a constant electrical bias).

The voltage drop across the input impedance of the selective amplifier, due to the photogalvanic current, was measured. Since the current changes its sign when the polarization changes orthogonally in the linear case and the direction of rotation changes in the circular case, the electrical signal is recorded at doubled frequency. All measurements were conducted at $T = 300$ K.

As can be seen from the form of the α_{ijk} tensor for a $Bi_{12}SiO_{20}$ crystal ($\alpha_{ijk} \neq 0$ only for $i \neq j \neq k$) under the experimental geometry (light propagates along the [100] axis), the magnitude and sign of "linear" PGE will depend on the orientation of the light polarization plane with respect to the crystal axes [010] and [001]

$$j_x^{PG} = I\,\alpha_{xyz}\,\sin 2\theta\,, \qquad (2)$$

where θ is the angle between the light polarization plane and the y- (or z) axis.

The above dependence on the orientation was actually observed in the experiment (Fig. 1). At the same time, the "circular" photogalvanic current remained unchanged as the specimen rotated around the x-axis, due to the fact that in cubic-symmetry crystals the γ_{il} tensor reduces to a pseudoscalar.

The dependence of "linear" and "circular" photogalvanic current on the intensity of incident light, measured in the range $I \sim (10^{-2}\text{–}1)$ W/cm^2, is linear with a coefficient of proportionality $K_1 = j^{PG}/KI$ (K is coefficient of absorption which equals 13 cm^{-1} for $\lambda = 0.488$ μm) being $(2.0 \pm 0.4) \times 10^{-9}$ A·cm/W and $(2.6 \pm 0.25) \times 10^{-9}$ A·cm/W, respectively.

An interesting feature of PGE in $Bi_{12}SiO_{20}$, at the aforementioned illumination levels, is the virtually-constant value of induced photo-emf E^{PG}. This is associated with a near-linear dependence of photoconductivity $\sigma_{photocond.}$ of the specimen on incident-light intensity (dark conductivity in $Bi_{12}SiO_{20}$ $\sigma_0 \ll \sigma_{photocond.}) \approx \text{const}(I)$. The value of E^{PG} obtained in the experiment is $(1.4 \pm 0.2) \times 10^{-2}$ V/cm and

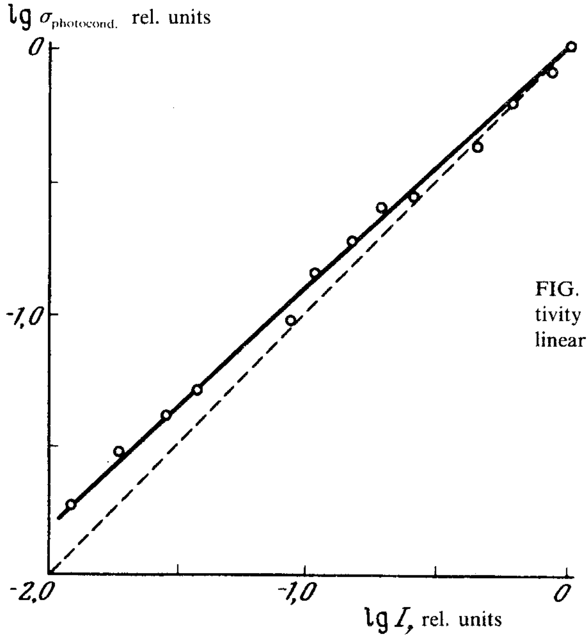

FIG. 2. Dependence of specimen photoconductivity on light intensity. Dashed line indicates a linear function $\sigma_{photocond.}(I)$.

$(1.8 \pm 0.1) \times 10^{-2}$ V/cm (for $I \sim 1$ W/cm^2) in the case of linear and circular polarization, respectively.

To describe the mechanism of "linear" PGE in $Bi_{12}SiO_{20}$ a model proposed for the case of impurity absorption in crystals without the inversion center,[8] may be used. Asymmetry in the electron distribution in this case is produced as a result of distortion of the electron wave function in the terminal state by the impurity field which is associated with its octupole moment $\langle Q_{ijk} \rangle = ea^{*3}$, where a^* is a characteristic dimension of charge density near an impurity. Comparison of the experimental values of the photogalvanic current with calculations involving Eq. (10) in Ref. 8 shows that a good agreement may be reached for $a^* \sim 10^{-18}$ cm. Clearly, the obtained value of a^* is fully realistic.

Explanation of a possible mechanism of "circular" PGE constitutes a more complex problem since accurate data for the zone structure of $Bi_{12}SiO_{22}$ crystals are currently unavailable. However, certain theoretical calculations may be made on the basis of a "circular" PGE model proposed in Ref. 2. Since, in contrast with the latter, in our case the electrons are excited from the impurity levels, calculations of their mean velocity must allow for terms that are linear with respect to the wave vector **k** only in the conduction band. Thus, the "circular" photogalvanic current is expressed as follows

$$j^{PG} = eKI\bar{v}^e \bar{\tau}_p^e , \tag{3}$$

where $\bar{v}^e = \beta_c/\hbar$ is the mean velocity of electrons excited by light, β_c is a coefficient of a term that is linear with respect to **k** in the conduction band, $\bar{\tau}_p^e$ is the relaxation time of pulse photo electrons. Equation (3) takes into account only the electron contribution, which is explained by the electronic nature of photoconductivity in undoped $Bi_{12}SiO_{20}$.[6,7] We shall calculate the values of $\bar{v}^e$ and β_c using Eq. (3) and the experimental value of j^{PG}. At $K = 13$ cm^{-1} and $\bar{\tau}_p^e \sim 10^{-13}$ sec, we get $\bar{v}^e \sim 7\times 10^4$ cm/sec and $\beta_c \sim 4\times 10^{-11}$ eV·cm. The relatively small value of $\bar{v}^e$ is compatible with a small mobility and large effective electron mass in $Bi_{12}SiO_{20}$.[6,9]

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