Circular photovoltaic effect in the ferroelectric $Pb_5Ge_3O_{11}$

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A circular photovoltaic effect has been observed in the gyrotropic crystal $Pb_5Ge_3O_{11}$. The photovoltaic current changes sign because of the action of an electric field; this is a consequence of the enantiomorphous transformation of the right-hand crystal into left-hand crystal. The circular photovoltaic coefficient has been determined.

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The photovoltaic effect, as is known, involves the appearance of a steady-state electric current when a crystal is illuminated uniformly. A linearly polarized light produces a linear photovoltaic effect, which has been observed in several ferroelectrics,[1,2] insulators,[3] and semiconductors.[4] The appearance of a photovoltaic current due to the action of circularly polarized light[5,6] is possible in optically active crystals. This circular effect has been observed experimentally only in semiconductor Te crystals[7] and in insulators of the type $Bi_{12}SiO_{20}$.[3,8] Observation of this effect in ferroelectric crystals, in which the action of an external electric field can reverse the spontaneous polarization and, thereby, change the sign of the photovoltaic current (as well as the sign of the optical activity), is especially interesting in our opinion. For this reason, we chose crystals of the uniaxial ferroelectric material $Pb_5Ge_3O_{11}$ as the object of our investigation (the Curie temperature of this material is $T_C = 177\,^\circ C$). In the ferroelectric phase these crystals belong to the trigonal system (point group C_3); above the transition temperature they are hexagonal (C_{3h}).

The experiments were performed at room temperature. An LPM-11 helium-cadmium laser with a wavelength of 0.44 μm and a power of 5×10^{-3} W was used to illuminate the samples. The modulation method, which has been described in detail elsewhere,[3,8] was used to measure the circular photovoltaic effect. When this method is used, the rotation direction of the circularly polarized light incident on the crystal is varied periodically by means of an electro-optic modulator. In our experiments the modulation frequency was 20 Hz; the photovoltaic current was measured at the same frequency by means of a synchronous detector with a 10-sec time constant. Since the input resistance of the recording system (10^8 ohm) was much less than the sample resistance, the conditions of the experiment corresponded to the short-circuit regime.

Samples in the form of 0.5-mm-thick wafers cut perpendicularly to the Z (C_3) axis, with the lateral dimensions of about 10 mm were used for the measurements. Semitransparent platinum electrodes, which were used to measure the photovoltaic current, were attached to the wafer surfaces. Circularly polarized laser light, a 1- to

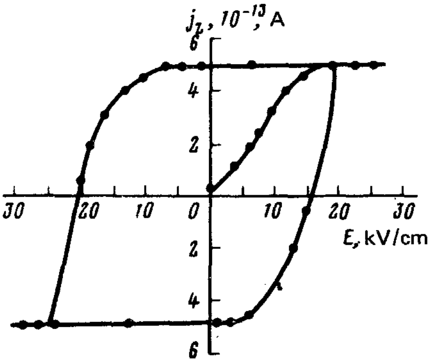

FIG. 1. Dependence of the circular photovoltaic current on the constant electric field applied along the Z axis.

2-mm-diam beam, was focused on the wafer. Before the measurements, the sample was heated to a temperature above the Curie point and cooled slowly to room temperature. Such a sample had essentially no photovoltaic current (see Fig. 1), consistent with a polydomain sample, since the signs of the currents are different for domains of different sign. A constant electric field was then applied to the sample along the Z axis, and the sample was held in this field for 10 min, after which the field was removed and the photovoltaic current was measured. The measurement results are shown in Fig. 1. As seen from the figure, the current dependence on the constant electric field exhibits a hysteresis, and at fields of +16 kV/cm and -20 kV/cm the polarization is reversed and the photovoltaic current changes sign (the reversal of polarization at the indicated fields was verified from the change in the sign of the optical activity). The difference in the coercive fields, as well as the small current in the polydomain sample, is attributable to unipolarity of the sample.

We then ran a series of experiments in order to confirm that the observed current is actually due to the circular photovoltaic effect, rather than due to the accompanying effects such as the pyroelectric effect or contact effects. The pyroelectric effect, in principle, can give an appreciable signal because of circular dichroism or possible parasitic amplitude modulation of the light; this signal changes sign as a result of reversal of the polarization. Direct measurements of the indicated parameters, as well as a very abrupt decrease of the measured current as a result of small deviation of the light beam from the Z axis make it possible to assume that the pyroelectric effect is insignificant. This conclusion is confirmed by our experiment. After rotating the crystal 180° about the axis perpendicular to the wave vector of the incident light, the measured current changes sign but not its magnitude, without a reversal of the electrodes. If the current were caused by the pyroelectric effect, then its sign would not change. An analysis of all the experimental data shows that the measured current is not produced by the contact effects.

In summary, the results shown in Fig. 1 are the first observation of a circular photovoltaic current, whose sign changes due to the action of an external electric field as a result of an enantiomorphous transformation of a right-hand crystal into a left-hand crystal. We note that the observation of a similar effect in Te or $Bi_{12}SiO_{20}$ crystals, in which the circular effect was observed previously, would require specially grown, right- and left-hand crystals. We see from the experimental results that the tensor component, which describes the circular photovoltaic effect, is equal to 2.5×10^{-10} V^{-1}.

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