Direct observation of a circular photocurrent in lithium niobate

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A new method is proposed for observing a spatially oscillatory photocurrent. A circular photocurrent has been observed by direct measurements for the first time in lithium niobate. The experimental results show that in lithium niobate the tensor component corresponding to the circular photovoltaic effect is larger than the off-diagonal component of the tensor of the linear photovoltaic effect, β^s_{15}.

An interesting property of crystals which lack a center of symmetry is that uniform illumination of the crystals with short-circuited electrodes gives rise to a steady-state current in the crystals.[1,2] This "bulk photovoltaic effect" is a consequence of the asymmetry of the elementary electronic processes in a crystal lacking a center of symmetry. It plays an important role in the recording of holograms in the familiar ferroelectric lithium niobate. The photovoltaic current density is described by the phenomenological expression[3–5]

$$j_i = \beta^s_{inl} E_n E^*_l + i\beta^{as}_{il} [\mathbf{E}\,\mathbf{E}^*]_l ,$$

where E_n and E_l are the components of the electric field of the light wave, and β^s_{inl} and β^{as}_{il} are the components of the third-rank symmetric tensor and the secondrank pseudotensor, respectively. The first term in this expression reaches a maximum in the case of linearly polarized light, and this component is accordingly called the "linear photocurrent." The second term is nonzero for circularly polarized light, and this current component is accordingly called the "circular photocurrent." The microscopic mechanisms which give rise to the linear and circular currents are different,[6] and they have received little study in ferroelectrics. The components of the tensor of the linear photovoltaic effect have been measured in many crystals lacking a center of symmetry, in particular, lithium niobate.[7] The circular current, on the other hand, has been observed in only a few crystals,[8,9] in which it has a spatially uniform component. In most crystals the circular polarization undergoes spatial oscillations with a period $\Lambda = \lambda/(n_o - n_e)$ (λ is the wavelength of the light, and n_o and n_e are the ordinary and extraordinary refractive indices for the light). Correspondingly, the circular current is spatially oscillatory and therefore difficult to see in direct measurements.[6] It has been suggested that the spatially oscillatory photocurrent might be observed by measurements in thin films, under conditions of strong absorption or by means of a periodic array of electrodes (with a period Λ), to provide a nonzero average value of the current over the thickness of the sample.[4,10] The spatially oscillatory current can be observed in photorefractive crystals indirectly, from the recording of gratings by waves with orthogonal polarizations,[11] and the circular current can be observed on the basis of the energy exchange between these gratings.[12,13] However, methods of this type require complicated experiments.

In the present letter we propose a new method for directly observing the spatially oscillatory photocurrent, and we report the first observation, by means of this method, of the circular photocurrent in lithium niobate.

The components of the photocurrent in lithium niobate are

$$\begin{aligned} j_x &= \beta^s_{22}(E_x E_y^* + E_x^* E_y) + \beta^s_{15}(E_x E_z^* + E_x^* E_z) + i\beta^{as}(E_z E_x^* - E_z^* E_x), \\ j_y &= \beta^s_{22}(E_x E_x^* - E_y E_y^*) + \beta^s_{15}(E_y E_z^* + E_y^* E_z) - i\beta^{as}(E_y E_z^* - E_y^* E_z), \\ j_z &= \beta^s_{31} E_x E_x^* + \beta^s_{31} E_y E_y^* + \beta^s_{33} E_z E_z^* . \end{aligned}$$

We see that the circular photocurrent, which flows perpendicular to the optic axis (the Z axis), is determined by a single independent component.[6] The linear photocurrent, determined by the tensor components β^s_{22} and β^s_{15}, flows in the same direction. Both the circular current and the linear photocurrent, which is related to the tensor component β^s_{15}, are spatially oscillatory. Let us consider the current that flows along the X axis of the crystal when light propagates along the Y axis:

$$j_x = \beta^s_{15} E_x^0 E_z^0 \cos(Ky + \Delta\varphi) + \beta^{as} E_x^0 E_z^0 \sin(Ky + \Delta\varphi),$$

where $E_x^0 = E_x \exp(-ikn_0 y)$, $E_z^0 = E_z \exp(-ikn_e y)$, $K = 2\pi/\Lambda$, $k = 2\pi/\lambda$, $\Delta\varphi$ is the initial phase shift between the field components E_x and E_y, and $\Lambda = 5\ \mu$m at the wavelength $\lambda \simeq 0.5\ \mu$m.

To observe the spatially oscillatory photocurrents, we propose using electrodes on a surface of the crystal which is perpendicular to the light propagation direction (Fig. 1). When the entire volume between the short-circuited electrodes is illuminated uni-

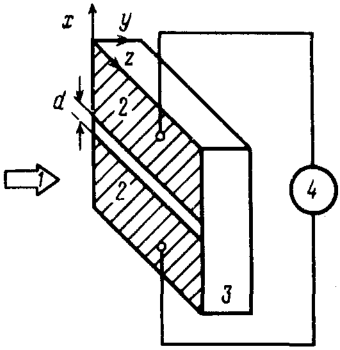

FIG. 1. Arrangement for measuring the spatially oscillatory photocurrent. 1—Light; 2—electrodes; 3—crystal substrate; 4—electrometer.

formly, a photocurrent determined by a thin surface layer of the crystal of thickness $h \ll \Lambda$ will flow. The spatial oscillations of the photocurrent will thus not influence the sign or magnitude of the measured current, which will instead be determined by the polarization state of the light incident on the crystal. Since the total current I for a given light power P is inversely proportional to the width (d) of the gap between the electrodes,

$$I \sim \beta P h / d$$

the width of this gap must be reduced in order to increase the current (i.e., to raise the sensitivity of the method).

In the present experiments, we use lithium niobate crystals annealed in air at 960 °C for 6 h. The crystals exhibit no significant absorption in the visible region. A pair of aluminum electrodes separated by a 10-μm gap is applied to polished Y-cut surfaces of the lithium niobate by a photolithographic method. These electrodes are used to measure the photovoltaic current j_x which flows in the direction parallel to the X axis. The beam from an argon laser with a wavelength $\lambda = 0.4765$ μm and a power ~ 13 mW is focused on the gap by an 8× objective. The degree of circular polarization of the light is varied by a volume electrooptic modulator with a half-wave voltage of 440 V. When no voltage is applied to the modulator, the light has a linear polarization making an angle of 45° with the optic axes of the modulator and the crystal. We use an

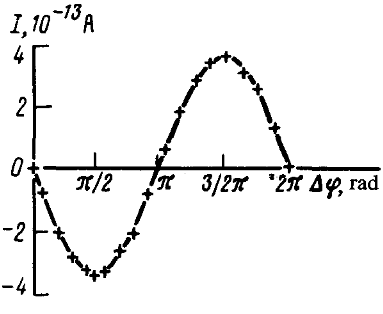

FIG. 2. The current I versus the phase shift $\Delta\varphi$.

ÉD-05M electrometer to measure the photovoltaic current with short-circuited electrodes. Figure 2 shows the resulting dependence of the current on the phase shift between the field components of the light along the X and Z axes (E_x and E_z). The dependence is sinusoidal, and we can clearly see extreme values of the photovoltaic current at phase shifts of $\pi/2$ and $3\pi/2$ in the circularly polarized light. These extrema show that we are directly observing a circular current in the lithium niobate. It also follows from this dependence that the maximum linear photocurrent, which is determined by the tensor component β^s_{15} and which flows in the crystal at phase shifts of 0, π, and 2π, is at least an order of magnitude lower than the maximum circular current. We may thus conclude $\beta^{as} \gg \beta^s_{15}$.

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