Theory and observation of polar asymmetry of photoionization in a field with $\langle E^3\rangle \neq 0$

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The polar asymmetry in the emission of photoelectrons (photoemissive effect) with interference of two-photon absorption of radiation of frequency ω and one-photon absorption at frequency 2ω is calculated and recorded experimentally. The presence of polar asymmetry of the photocurrent is established on the basis of the dependence of the photocurrent on the phase difference of the interfering waves $\Delta\varphi = \varphi_2 - 2\varphi_1$. The dependence of the phase shift between the interference pattern of the field $\propto \langle \mathbf{E}^3\rangle$ and the interference pattern of the photocurrent on the direction of polarization of the interfering waves is demonstrated experimentally.

1. INTRODUCTION

Efficient generation of the second harmonic in an optical fiber has been detected experimentally.[1] To achieve generation the fiber must be "specially prepared" beforehand. The most effective way of doing this consists of simultaneously irradiating the fiber with radiation at the fundamental frequency ω and at the second harmonic 2ω, obtained in a nonlinear crystal.[2] A longer preparation time and special selection of the material of the fiber are required if the fiber is irradiated only by radiation at the frequency ω.

The explanation of the second harmonic generation (SHG) reduces to the following.[2,3] The real part of the amplitude of the field that is introduced into the fiber during the preparation process has the form

$$E_{\text{real}}(r,\ t) = \mathbf{E}_1 \cos(\omega t - \varphi_1) + \mathbf{E}_2 \cos(2\omega t - \varphi_2). \qquad (1)$$

Such a field has a zero mean value $\langle \mathbf{E}\rangle = 0$, but possesses polar asymmetry, which is characterized by the value

$$\langle \mathbf{E}^3\rangle = \frac{3}{4} E_1^2 E_2 \cos(\varphi_2 - 2\varphi_1),$$

(see Fig. 1, where $E_1 = E_2 = 1$ and $\varphi_2 - 2\varphi_1 = 0$). In the presence of the field

$$E_1(r)\exp(ik_1 z - i\omega t) + E_2(r)\exp(ik_2 z - i2\omega t)$$

a steady nonequilibrium charge distribution is formed in the fiber, proportional to the cube of the field:

$$(E_1^2)^* E_2 \exp(i\Delta k z) + E_1^2 E_2^* \exp(-i\Delta k z), \qquad (2)$$

where $\Delta k = k_2 - 2k_1$ is the mismatch of the wave vectors.

This interference pattern is transformed by some microscopic mechanism into a spatial modulation grating of the quadratic polarizability tensor $\delta\chi^{(2)}_{ijk}$:

$$\delta\chi^{(2)}(z) = \beta (E_1^2)^* E_2 \exp(i\Delta k z) + \text{c.c.}, \qquad (3)$$

where $\beta = \beta_1 + i\beta_2$ is a coefficient that depends on the exposure time. The nature of the writing mechanism (formation of the grating) is still unclear, in spite of numerous efforts.[4–10]

During the reading of the $\delta\chi^{(2)}$-hologram (3) the wave at the fundamental frequency $E_1(r)\exp(ik_1 z - i\omega t)$ induces a polarization at the doubled frequency in the fiber:

$$P_2 = \chi^{(2)} E_1^2 \exp(2ik_1 z - 2i\omega t) = \beta |E_1|^4 E_2 \exp(ik_2 z - 2i\omega t). \qquad (4)$$

Here, as in a usual volume hologram, the condition of phase synchronism is automatically satisfied. In the present case the polar asymmetry of the field is manifested in the medium in the form of an induced polarization proportional to $\chi^{(2)}$, whose presence, as is well known, indicates the absence of a symmetry center.

In the present article we consider another effect due to the polar asymmetry of the light field, namely the polar asymmetry of electron emission during photoionization. Besides that, the effect is interesting in itself; it can be used to explain the mechanism of writing of $\delta\chi^{(2)}$-holograms in optical fibers.

The assertion that the writing of $\chi^{(2)}$-holograms in optical fibers may be due to changes in the medium arising during the polar-asymmetric interference of single-photon absorption of the wave $E_{2\omega}$ and two-photon absorption of the wave E_ω was made in Ref. 3. The idea of the appearance of a photocurrent as a result of photoionization under the influence of pumping radiation and second harmonic radiation was expressed in Refs. 9 and 11. In Ref. 9 it was also suggested that this effect may explain SHG in optical fibers. In Ref. 11 it was shown that such a photocurrent consists of two components, one of which is determined by the interference of processes of single-photon absorption of a quantum of frequency 2ω and two-photon absorption of a quantum of frequency ω. A relation between the photoinduced SHG effect and the interference of single-photon ionization by radiation of the second harmonic and two-photon ionization by the pumping radiation was indicated in Refs. 12 and 13.

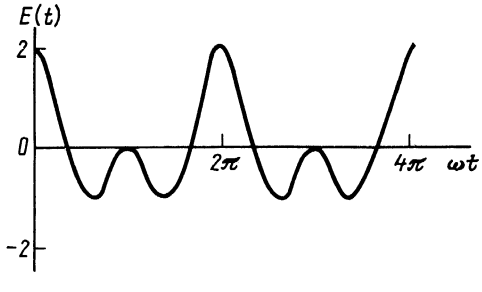

FIG. 1. Time dependence of the real part of the field strength in the case $E_1 = E_2$, $\varphi_2 - 2\varphi_1 = 0$, $\langle E\rangle = 0$. The quantity $\langle E^3\rangle = 4/3$ serves as a quantitative characteristic of the polar asymmetry of the field.

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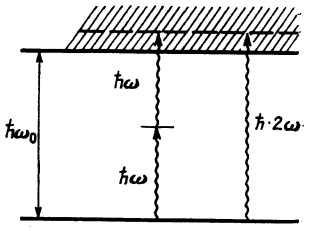

FIG. 2. Interference of two-photon $2\cdot\hbar\omega$ and single-photon $\hbar\cdot 2\omega$ ionization leads to polar asymmetry of the photoeffect.

2. POLAR ASYMMETRY OF PHOTOIONIZATION BY A FIELD WITH $\langle E^3\rangle \neq 0$

Consider the photoionization of an atom by a doubled-frequency field of the form (1). We assume that the ionization potential of the atom $\hbar\omega_0$ satisfies the condition $\hbar\omega < \hbar\omega_0 < 2\hbar\omega$, so that the laser field E_1 can cause only two-photon ionization, and its second harmonic can cause only single-photon ionization (Fig. 2). The transition probability $dW(\mathbf{n})/do$ depends on the direction of emission of the electron ($|\mathbf{n}| = 1$).

As is well known, during monochromatic irradiation $dW(\mathbf{n})/do$ possesses the symmetry property $dW(\mathbf{n})/do = dW(-\mathbf{n})/do$. The presence of polar asymmetry of the field $E(t)$, see Fig. 1, gives a reason to expect polar asymmetry in the probability of photoionization, $dW(\mathbf{n})/do - dW(-\mathbf{n})/do \neq 0$.

In our calculations we use the model of a short-range spherically symmetric δ-function potential in which there is only one bound s-state. The Hamiltonian of the system atom + field we take to be in the form

$$\hat{H} = -\frac{\hbar^2}{2m}\left(\frac{1}{r_0}+\varkappa\right)\delta^{(1)}(|\mathbf{r}|-r_0)+\left[p-\frac{e}{c}\mathbf{A}(t)\right]^2/2m; \tag{5}$$

where

$$\mathbf{A}(t) = -c\{2i\mathbf{E}_1 e^{-i\omega t}+i\mathbf{E}_2 e^{-2i\omega t}+\text{c.c.}\}/4\omega$$

is the vector potential of the light field. In the general case of elliptically polarized waves the vector amplitudes $\mathbf{E}_1$ and $\mathbf{E}_2$ are essentially complex. In the expression for the potential we have introduced the auxiliary parameter $r_0 \to +0$, which must tend to zero. In terms of the radial wave function of the s-wave $\psi(r) = \chi(r)/r$, this corresponds to the imposition of the boundary condition $\chi'/\chi = -\varkappa$ as $r \to +0$, see Ref. 14. In the absence of the field the Hamiltonian (5) has one bound s-state with energy $-\hbar\omega_0$:

$$\psi(\mathbf{r},t) = \exp(i\omega_0 t)\left(\frac{\varkappa}{2\pi}\right)^{1/2}\frac{e^{-\varkappa r}}{r}, \quad \varkappa = (2m\omega_0/\hbar)^{1/2}, \tag{6}$$

and states of the continuous spectrum with energies $\hbar^2\mathbf{k}^2/2m$, where $\mathbf{k}$ is the electron momentum far from the center. Since these states for a given energy are infinitely degenerate with respect to angle, additional considerations are necessary to choose appropriate linear combinations. If we are interested in the probability of emission in a given direction $\mathbf{n} = \mathbf{k}/k$, then, as is well known (see, e.g., Ref. 14), wave functions of the form

$$\psi_{\mathbf{k}}(\mathbf{r}) = (2\pi)^{-3/2}\left(e^{i\mathbf{k}\mathbf{r}}-\frac{1}{\varkappa-ik}\frac{e^{-ikr}}{r}\right)\exp\left(-i\frac{\hbar k^2}{2m}t\right), \tag{7}$$

in the asymptotic limit $r \to \infty$, containing a plane wave and a spherical wave converging on the center, are adequate to the problem.

The amplitude of the transition from the ground state (6) to the state with kinetic energy $\hbar^2 k^2/2m = \hbar(2\omega - \omega_0)$ consists of two terms: the first one describes the contribution of the single-photon transition under the influence of the field $E_2 \exp(-2i\omega t)$, and the second describes the two-photon transition under the influence of the field $E_1 \exp(-i\omega t)$. The first term corresponds to first-order perturbation theory with respect to E_2, and the second term to second-order perturbation theory with respect to E_1. Calculation of the second order requires the calculation of the matrix elements of the perturbations between ψ_0 and $\psi_{\mathbf{k}'}$ and between $\psi_{\mathbf{k}'}$ and $\psi_{\mathbf{k}}$ and integrating with the corresponding energy denominator over all values of the momentum of the virtual state $d^3\mathbf{k}'$. Without going into the technical details of these calculations, we mention only that the integration can be conveniently carried out by first integrating over $d^3\mathbf{k}'$ and then over the coordinates $d^3\mathbf{r}$. As a result we obtain the following expression for the photoionization probability:

$$\frac{dW(\mathbf{n})}{do} = B|f_1(\mathbf{E}_2\mathbf{n})+f_0(\mathbf{E}_1\mathbf{E}_1)+f_2[(\mathbf{E}_1\mathbf{n})^2-(\mathbf{E}_1\mathbf{E}_1)/3]|^2. \tag{8}$$

Here

$$B = \frac{2e^2}{\pi m\hbar}\frac{[\omega_0(2\omega-\omega_0)]^{1/2}}{(2\omega)^4}$$

$$f_1 = i(2\omega-\omega_0)^{1/2},$$

$$f_0 = -\left(\frac{e^2}{2m\hbar}\right)^{1/2}\frac{4(\omega_0-\omega)}{3\omega^3}[\omega_0^{1/2}-(\omega_0-\omega)^{1/2}][\omega_0^{1/2}-i(2\omega-\omega_0)^{1/2}],$$

$$f_2 = \left(\frac{e^2}{2m\hbar}\right)^{1/2}\frac{4}{\omega^2}(2\omega-\omega_0).$$

Let us clarify the meaning of the individual terms in Eq. (8). The term $f_1(\mathbf{E}_2\mathbf{n})$ corresponds to the single-photon transition. According to the selection rules of the electric dipole moment, from a state with orbital momentum $l = 0$ (an s-state) it is possible to transition only to a p-state, $l = 1$; this is due to the dependence of the amplitude on $\mathbf{n}$ of the form $\propto (\mathbf{E}_2\mathbf{n})$. In second order in the field E_1 two-photon transitions are possible: $s \to s$ with amplitude $\propto f_0(\mathbf{E}_1\mathbf{E}_2)$, and also $s \to d$, with amplitude $\propto f_2[(\mathbf{E}_1\mathbf{n})^2 - (\mathbf{E}_1\mathbf{E}_2)/3]$.

In the dipole approximation the excitation of an electron to the continuous spectrum occurs locally, near the center. The subsequent departure of the electron to infinity is accompanied by a phase shift of its wave function by the amount δ_l—the quantum-mechanical scattering phase in the state with orbital momentum l [S_l is the scattering matrix, which is diagonal in l and equal to $\exp(i\delta_l)$] and phase factor $\exp(i\pi l/2)$ associated with the expansion of the plane wave into spherical waves. It can be easily seen that in the short-acting potential there is only s-scattering, and it follows from expression (7) that $\delta_1 = \delta_2 = \ldots = 0$, and

$$\delta_0 = -\text{arctg}(k/\varkappa) = -\text{arctg}[(2\omega-\omega_0)/\omega_0]^{1/2}.$$

In complete accordance with this the quantity f_2 is real and the quantity f_1 is purely imaginary, and the quantity f_0 has the phase factor $\exp[i\delta_0(\omega)]$.

The ionization probability contains three terms, which

correspond to the single-photon process ($\propto |f_1|^2$), the two-photon process ($\propto |f_0 + f_2|^2$), and their interference. It is precisely this interference term that characterizes the polar asymmetry of ionization:

$$\frac{dW(\mathbf{n})}{do} - \frac{dW(-\mathbf{n})}{do} = 2B|f_1 f_2|\{i(\mathbf{E}_2\mathbf{n})[(\mathbf{E}_1^*\mathbf{n})^2 - (\mathbf{E}_1^*\mathbf{E}_1^*)/3] + \text{c.c.}\}$$
$$-2B|f_1 f_0|[i(\mathbf{E}_2\mathbf{n})(\mathbf{E}_1^*\mathbf{E}_1^*)\exp(-i\delta_0) + \text{c.c.}]. \quad (9)$$

The additional phase shift $i\delta_0$ in Eq. (9) can be explained in the following way. The expression for the ordinary single-photon ionization probability contains only even powers of the unit vector $\mathbf{n} = \mathbf{k}/k$. If we neglect the interaction of the emitted electron with the atom, $\delta_l = 0$, then its motion must be time-reversible, and the expression

$$\frac{dW(\mathbf{n})}{do} = C_1(\mathbf{E}_2\mathbf{E}_2) + C_2(\mathbf{E}_2\mathbf{n})(\mathbf{E}_2\mathbf{n})$$

will be symmetric with respect to time reversal since here $\mathbf{n} \to -\mathbf{n}$. Averaging over a few optical periods gives $\langle E_i E_k \rangle \to (E_i^* E_k + E_i E_k^*)/2$. To obtain the odd powers of $\mathbf{n}$ it is necessary to use dE/dt in order to retain the symmetry with respect to time reversal, for example, by introducing the conditions

$$\frac{dW(\mathbf{n})}{do} - \frac{dW(-\mathbf{n})}{do} = \text{const}\left(\mathbf{n}\frac{d\mathbf{E}_2}{dt}\right)(\mathbf{E}_1\mathbf{E}_1)$$

and for a monochromatic wave $d\mathbf{E}_2/dt = -2i\omega\mathbf{E}_2$. Quantum-mechanical scattering of the electron by an atom gives a time-irreversible factor, in our case $\exp(i\delta_0)$.

Let the polarization of the waves E_1 and E_2 be linear, i.e., $\mathbf{E}_1 = E_1\mathbf{e}_1$ and $\mathbf{E}_2 = E_2\mathbf{e}_2$ with real unit vectors $\mathbf{e}_1$ and $\mathbf{e}_2$. Then according to Eq. (9) the polar asymmetry is equal to

$$W(\mathbf{n}) - W(-\mathbf{n}) \propto \text{Re}(E_1^{*2}E_2 e^{i\gamma}),$$

where

$$\gamma = \arg\{i(\mathbf{e}_2\mathbf{n})[((\mathbf{e}_1\mathbf{n})^2 - 1/3)|f_2| + |f_0|\exp(i\delta_0)]\}.$$

As a result, the phase γ of the term with polar asymmetry, first of all, is no longer equal to zero and, secondly, it depends on the polarization of the interfering waves.

The polar asymmetry of the photoionization process inside the optical fiber under the influence of the waves E_ω and $E_{2\omega}$ can provide a mechanism for the writing of $\delta\chi^{(2)}$-holograms in the fiber.[2,3,9,12,13] Indeed, for $\langle E_x^3 \rangle > 0$, the electrons are emitted mainly along the x axis, while for $\langle E_x^3 \rangle < 0$ they are emitted in the opposite direction. Free electrons can then be caught in traps. The arising spatial charge distribution gives rise to the appearance of a static electric field

$$E_x^{\text{st}}(z) \propto \langle E_x^3(z) \rangle \propto \cos[(2k_1 - k_2)z].$$

The action of the static electric field then gives rise to the grating

$$\delta\chi^{(2)}(z) \propto \chi^{(3)}(0, \omega, \omega, -2\omega)E_x^{\text{ст}}(z),$$

as was assumed by Stolen and Tom.[2]

However, in contrast with the model of Stolen and Tom,[2] we assume that the static electric field arises as a result of active absorption of quanta of frequency ω and 2ω (see also Refs. 9–11) instead, in order to take into account the appearance of static polarization through the reactive cubic polarization

$$P_x \propto \chi^{(3)}(0, \omega, \omega, -2\omega)E_1^2 E_2^*.$$

As the example with the dynamic holograms in photorefractive crystals shows, active absorption gives a stronger effect than reactive processes.

Important results which follow from a consideration of this simple model are 1) calculation of the polar asymmetry as such and 2) the conclusion that in general there is a phase shift between the $\langle \mathbf{E}_{\text{real}}^3 \rangle$ pattern and the pattern of the polar asymmetry of the polarization. As is well known, in photorefractive crystals the phase shift between $\langle \mathbf{E}_{\text{real}}^2 \rangle$ and the variation of the refractive index plays an important role in the realization of energy exchange between waves. Since such a photoionization process can provide a mechanism for writing a grating $\propto \langle \mathbf{E}^3 \rangle$ in optical fibers, the phase of the $\delta\chi^{(2)}$-grating also has a large value (see Ref. 8).

After the completion of this paper Professor R. Glauber kindly informed us that the idea of interference of single-photon and two-photon absorption for fields with $\langle \text{EEE} \rangle \neq 0$ was discussed by him as early as 1967,[15] but without any special mention of polar photoionization.

3. EXPERIMENT

We experimentally investigated the polar asymmetry of electrons being emitted from the photocathode during normal incidence of two waves, the fundamental ω and the second harmonic 2ω.

The detection of interference of the fields E_ω and $E_{2\omega}$ during the external photoeffect was first reported in Ref. 16. In the present paper we report the experimental recording of polar asymmetry of the photoionization for two different polarizations of the light. This allows us to conclude that in agreement with theoretical predictions there exists a polarization-dependent phase shift between the interference pattern of the field and the polar asymmetry pattern of the emitted electrons. To observe the interference we chose an FÉU-127 photomultiplier. It possesses two characteristics that are necessary for this study. First, the red limit of the photoeffect of the antimony-cesium photocathode of the photomultiplier is 0.6 μm. Therefore laser radiation at $\lambda_1 = 1.06$ μm should cause only the two-photon effect, and its second harmonic $\lambda_2 = 0.53$ μm, only the single-photon effect. The two-photon character of the photoeffect was first checked by illuminating only with the wave λ_1. The experimental dependence of the photocurrent on I_1 was found to be quadratic over a large range of intensities I_1 (see Fig. 3b). As I_1 increases beyond some value the regime changes abruptly: the photocurrent saturates, and the photocathode begins to break down. Note that to increase the accuracy of the measurements the quantity I_1^2 was recorded in all the experiments from the signal of the second harmonic, obtained in a separate crystal of KTP.

The second necessary characteristic of the FÉU-127 photomultiplier is the presence of discrimination in the recording of the photoelectrons emitted in different directions. This anisotropy of the sensitivity was built into the FÉU-127 (see Fig. 3a). As it turned out, the sensitivity of the photomultiplier to radiation of frequencies ω and 2ω depends on the angle α between the direction to the "hole" in the photo-

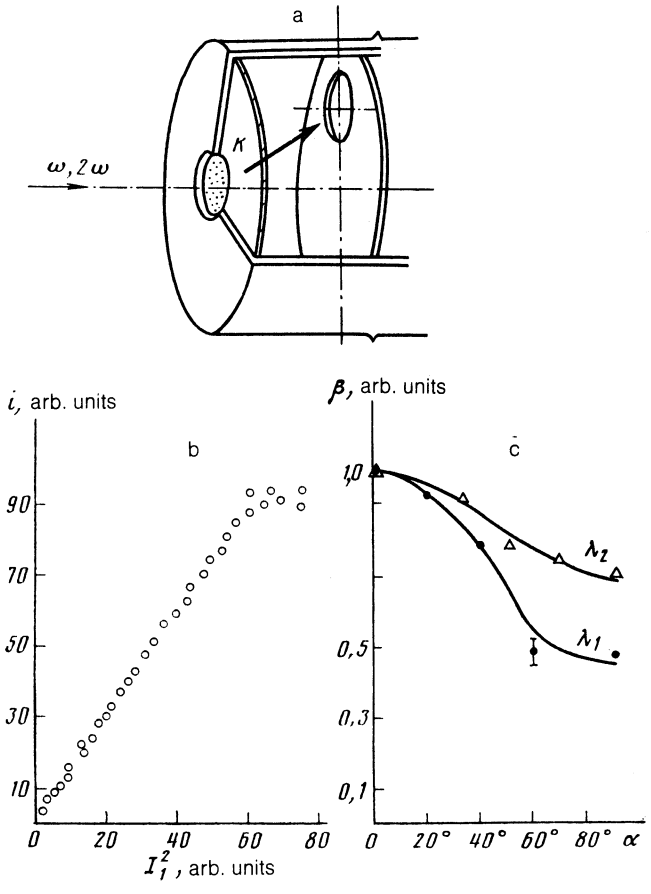

FIG. 3. Characteristics of the FÉU-127: a) construction; b) dependence of the photocurrent i on the square of the intensity I_1^2 of the radiation with $\lambda_1 = 1.064$ μm; c) angular dependence of the β-sensitivity of the photomultiplier for linearly polarized radiation with $\lambda_2 = 0.532$ μm (upper curve) and $\lambda_1 = 1.064$ μm (lower curve); α is the angle between the direction to the "hole" in the photomultiplier and the direction of the polarization vector; K is the cathode, the thick arrow indicates the direction of motion of the photoelectrons to the first dynode.

multiplier and the direction of the vector of the linear polarization of the incident radiation. Figure 3c presents the experimentally obtained dependence of the sensitivity of the FÉU-127 on the angle α for $\lambda_1 = 1.064$ μm and $\lambda_2 = 0.532$ μm in reduced units. We also measured the sensitivity of the FÉU-127 to circular polarization at $\lambda_1 = 1.064$ μm, and, as it turned out, within the limits of experimental error it is equal to the sensitivity to radiation with linear polarization at $\alpha = 0$ at the same wavelength.

An optical diagram of the experiment is shown in Fig. 4. As in Ref. 16, important use was made in the experiment of the interference nature of the polar asymmetry of the photoeffect. The presence of polar asymmetry was established not by direct recording of the angular distribution of the emitted electrons, but from the dependence of the photocurrent on the phase difference of the interfering waves $\Delta\varphi = \varphi_2 - 2\varphi_1$. In contrast with Ref. 16, in our experimental setup we added two dichroic polarizers immediately after the Glan prism. Such polarizers are commonly used in photography. The point here is that, conducting measurements with unknown polarizations of the waves E_1 and E_2, it is quite difficult to determine the phase shift between the interference pattern of the field $\propto \{E_1^2 E_2^* \exp[i(2k_1$

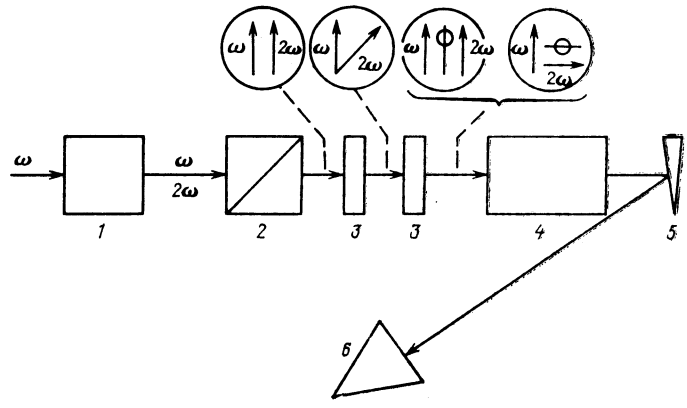

FIG. 4. Schematic of the experiment: 1) KDP crystal, 2) Glan prism, 3) dichroic polarizers, 4) hermetically sealed cuvette, air-pumped, 5) glass wedge, and 6) FÉU-127 photomultiplier.

$- k_2)z] + \text{c.c.}\}$ and the interference term in the photocurrent since the path to the photocathode passes through the glass window of the photomultiplier, which has a thickness of ~ 50 rad.[1)] We therefore posed the experimental problem of measuring the relative phase shift of the interference term for two different polarizations of the waves. The greatest experimental difficulty here lay in the necessity of varying the polarization state without introducing an additional phase shift $\Delta\varphi = \varphi_2 - 2\varphi_1$. As it turned out, the dichroic polarizer for radiation with $\lambda_1 = 1.064$ μm is a weak phase plate with maximum depolarization $\lesssim 5 \cdot 10^{-3}$. Therefore, arranging the two dichroic polarizers as shown in Fig. 4, we were able to vary the polarization of the wave $E_{2\omega}$ from the vertical to the horizontal by rotating the second polarizer by 90°, practically without changing the polarization of the wave E_ω or introducing an additional phase shift.

Radiation with frequencies ω and 2ω was normally incident simultaneously on the photocathode of the photomultiplier. The measurements were carried out at values of the intensity I_1 corresponding to unsaturated I_1^2-dependence of the photocurrent. The second-harmonic conversion coefficient in KDP was chosen so that the signal of the single-photon photocurrent, proportional to I_2, roughly coincided in magnitude with the signal proportional to I_1. In this situation in the first case the linear polarization of the waves E_ω and $E_{2\omega}$ coincided with the direction to the "hole" of the photomultiplier, as in Ref. 16. In the second case the polarization of the wave $E_{2\omega}$ coincided with the direction to the hole, but the polarization of the wave E_ω was perpendicular to this direction. We achieved such an arrangement by simultaneously rotating the polarization vector of the 2ω wave and the photomultiplier by 90° around the horizontal axis. Control experiments with a He–Ne laser on the interference of the reflected waves from the front and back surfaces of the glass window of the photomultiplier showed that such a rotation does not introduce an additional phase difference.

To observe the interference we measured the phase difference $\Delta\varphi = \varphi_2 - 2\varphi_1$, in contrast with Ref. 16, by varying the air pressure in the hermetically sealed cuvette with the glass windows. The calibration method is described in the Appendix. So as not to spoil the agreement of the wave fronts of the interfering beams, rather than focusing them we

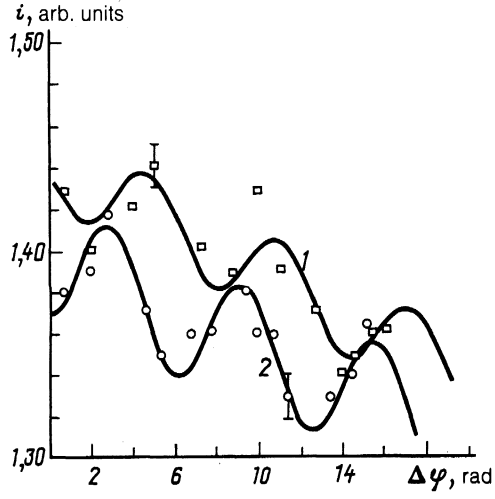

FIG. 5. Dependence of the photocurrent of the FÉU-127 on the phase difference $\Delta\varphi$ for two cases: 1) the polarization of the waves with frequencies ω and 2ω coincide with the direction to the "hole" in the photomultiplier, 2) the polarization of the wave with frequency 2ω coincides with this direction, while the polarization with frequency ω is perpendiculuar to it.

passed them through a dispersive wedge.

Figure 5 shows the dependence of the photocurrent on the phase difference $\Delta\varphi$ for the two cases described above. The calculated curves (solid lines) were obtained by an approximate fit of the form

$$I_{\text{FÉU}}=C_1+C_2(\Delta\varphi)+C_3\cos(a\Delta\varphi+\Delta\varphi_0)$$

to the experimental points. Decrease of the mean energy of the recorded field with increase in the pressure in the cuvette is possible due to induced birefringence in the glass windows. The phase shift between the two interference patterns of the photocurrent is equal to $\sim 98°$.

This also indirectly demonstrates the existence of a phase shift between the interference pattern of the field and that of the photocurrent.

4. CONCLUSION

In the present work we have calculated and experimentally recorded polar asymmetry of electron emission during interference of two-photon absorption of light at the frequency ω and single-photon absorption at the frequency of the second harmonic 2ω. We have experimentally demonstrated that the phase shift between the interference pattern of the field $\propto \langle \mathbf{E}^3 \rangle$ and the interference pattern of the photocurrent depend on the polarization of the interfering waves.

APPENDIX. TWO-CRYSTAL METHOD OF MEASUREMENT OF THE PHASE DIFFERENCE $\Delta\varphi=\varphi_2-\varphi_1$

In order to accurately measure the phase difference $\Delta\varphi = \varphi_2 - 2\varphi_1$ introduced by the various optical elements, we used a two-crystal scheme of second-harmonic generation. Optimization of such a scheme to increase the efficiency of conversion was studied in Refs. 17 and 18. We consider the case of weak pumping. The intensity of the second harmonic upon exit from the second crystal $I_{2\omega}$ is a result of the interference of the second-harmonic fields generated in the two crystals, and is governed by the phase difference

$$\Delta\varphi=\Delta\varphi_0+(k_{2\omega}-2k_\omega)z,$$

where z is the distance between the crystals, and k_ω and $k_{2\omega}$ are the wave numbers of the radiation in air at the frequencies ω and 2ω, respectively. $\Delta\varphi_0$ is the initial phase difference due to the differences between the two crystals. If the crystals are set in exact synchronism, then at $z = 0$ there should obtain either a maximum signal $I_{2\omega}$ (if the crystals are oriented identically) or a minimum signal (if one of the crystals is rotated 180° about the horizontal axis).

By way of an example, we will consider the method of measurement of the quantity $\Delta\varphi$ for the case of measurement of the phase difference $\Delta\varphi_0$ at the exit from the nonlinear optical crystal (KTP) and for the passage of a wave with frequency ω and its second harmonic 2ω through the hermetically sealed cuvette filled with various gases. Such a cuvette was used in experiments on the interference of waves with frequencies ω and 2ω in the external photoeffect.

A periodically pulsed laser with active Q-switching, based on an industrial laser of type LTI-403, was operated in the TEM_{00} mode with pulse repetition frequency of 3 Hz. As our nonlinear optical crystals we used KTP-cut crystals in such a way that the direction of synchronism ($oe\rightarrow o$) was perpendicular to the face of the crystal. Initially, the crystals were placed so that in the limit $z\rightarrow 0$ the dependence $I_{2\omega}(0)$ had a maximum, i.e., so that $\Delta\varphi_0 = 0$. The distance between the crystals was chosen so that the signal $I_{2\omega}$ was minimized. As it turned out, the experimental error in the determination of the separation z at which the signal $I_{2\omega}$ was minimum was much less than when it was maximum, and was $\lesssim 4\%$ of the period of the interference pattern $z_{2\pi} = 2\lambda_1/(n_{2\omega} - n_\omega)$.

When the first crystal is rotated by an angle θ in the plane perpendicular to the z axis, the value of the quantity $\Delta\varphi_0 = \Delta\varphi_0(\theta)$ varies, which leads to a variation in the maximum signal $I_{2\omega}$ and to a new position along the z axis of the minimum of $I_{2\omega}$. The variation of the phase difference at the exit of the first crystal was determined from the displacement of the position of the minimum of $I_{2\omega}$. A plot of the experimentally obtained dependence of the quantity $\Delta\varphi_0(\theta)$ on the angle θ is shown in Fig. 6. This figure shows how $I_{2\omega}$ depends on the angle of rotation in the absence of the second crystal. When the crystals depart from synchronism to one side, $\Delta\varphi_0(\theta)$ grows as a linear function of θ and reaches its maximum value of $+\pi$ radians in the region of the first intensity minimum. Unfortunately, here the contrast of the interference pattern falls to zero, so that the phase can no longer be measured, but after passing through this angle the phase jumps to zero and again grows to $+\pi$ radians over the extent of the subsequent Maker oscillation.

Let us consider how the phase of the second harmonic generated in the crystal varies as the crystal is rotated by an angle θ away from the direction of synchronism. We will assume that exact synchronism corresponds to normal incidence $\mathbf{e}_z = \mathbf{m}$, where $\mathbf{e}_z$ is the direction of the wave vector of the beam incident on the crystal, and $\mathbf{m}$ is the direction of the normal to the crystal (see Fig. 6).

The direction of synchronism in the KTP crystal is perpendicular to the z axis. From the form of the wave-vector surface it follows that when θ is parallel to the z axis[19] the additional phase change contains only terms $\propto \theta^2$. Terms linear in θ appear when θ lies in the xy plane. We will assume

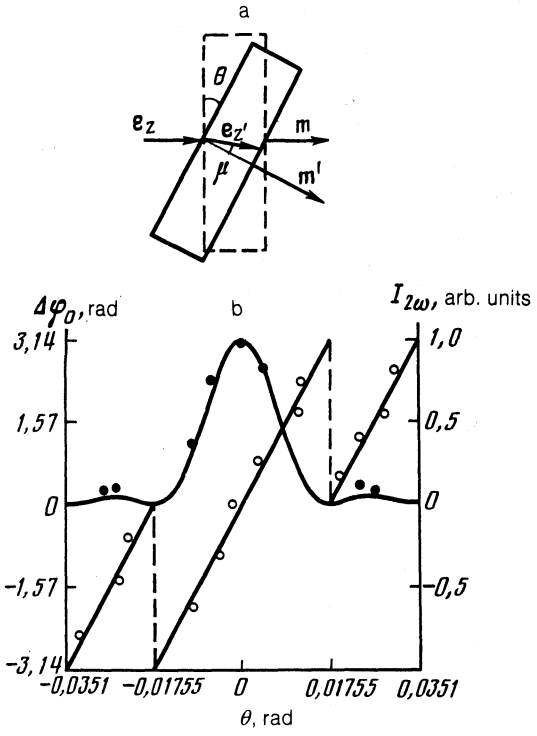

FIG. 6. a) Schematic diagram of the rotation of the KTP crystal; b) angular dependence of the intensity of the second harmonic (●) and of the phase difference $\Delta\varphi_0$ at the exit (○) of the first KTP crystal; points: experiment; solid curves: calculation.

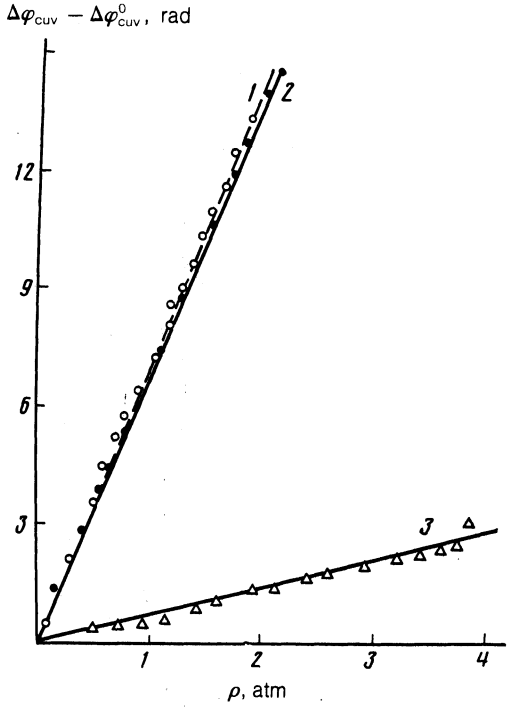

FIG. 7. Experimental dependence of the quantity $\Delta\varphi_{cuv} - \Delta\varphi^0_{cuv}$ on the pressure in the cuvette filled with air (1), nitrogen (2), and helium (3).

that θ is small and take into account only those terms linear in θ.

When the crystal is rotated by an angle of θ, to within a linear factor we have

$$\mathbf{m}'=\mathbf{m}+\theta,\ \mathbf{e}_z'=\mathbf{e}_z+(\theta-\mu),$$

where $\mathbf{e}_z'$ is the unit vector of the direction of the wave vector in the crystal. Snell's law is written in the form

$$n_a\theta = n_c\mu\ ,$$

where n_a and n_c are the refractive index in air and in the crystal. The wave vector in the crystal rotated by the angle θ is equal to

$$\mathbf{k}' = \frac{\omega}{c} n_c[\mathbf{m}'(1-\mu\eta)+\mu],$$

where η is the unit vector of the normal to the surface of the wave vectors. The intensity of the second harmonic is a result of the interference of the waves generated in the two nonlinear crystals, one of which is rotated by the angle θ:

$$I_{2\omega} \propto \left|1+\exp\left[i\left(\frac{\Delta kd}{2}\right)\right]\frac{\sin(\Delta kd/2)}{\Delta k\, d/2}\right|^2,$$

where d is the thickness of the crystal.

Taking into account Snell's law and the fact that $\mathbf{m}'$ is the synchronism direction, we obtain

$$\Delta\mathbf{k}=\mathbf{k}_1^0+\mathbf{k}_1^e-\mathbf{k}_2^0 = -\frac{\omega}{c} n_a\mathbf{m}'[(\eta_1^{(0)}+\eta_2^{(e)}-2\eta_2^{(0)})\theta].$$

Knowing the principal values of the dielectric tensor of the crystal, see Ref. 19, we find

$$(\eta_1^{(0)}\theta)=23{,}8\cdot10^{-3},\ (\eta_1^{(e)}\theta)=0,\ (\eta_2^{(0)}\theta)=-7{,}4\cdot10^{-3},$$

if as the positive θ direction we take rotation toward the x axis.

Hence we find $\Delta kd/2 \approx -3.3\cdot10^2\theta$ (rad) and the position of the first minimum of $I_{2\omega}$, which corresponds to $\Delta kd/2\approx\pi$, $\theta\approx9.56\cdot10^{-3}$ rad = 0.55 grad, which agrees with the experimental results.

An analogous method can be used to measure the additional phase difference introduced by the different optical elements. To do this, the element under investigation (a plane-parallel transparent plate, a hermetically sealed cuvette with transparent windows, etc.) is placed between the two nonlinear optical crystals. Complete synchronism of the crystals is not necessary. On the contrary, maximum contrast of the interference pattern is achieved when one of the crystals departs from synchronism.

Figure 7 presents the experimentally obtained dependence of the quantity $\Delta\varphi_{cuv} - \Delta\varphi^0_{cuv}$ on the pressure of various gases in the hermetically sealed cuvette ($\Delta\varphi^0_{cuv}$ is the phase difference introduced by the cuvette at standard pressure). From this behavior it is possible to estimate the magnitude of the quantity

$$\frac{d}{dp}\Delta n = \frac{d}{dp}(n_{2\omega}-n_\omega)$$

for air, nitrogen, and helium. Variation of the difference $n_{2\omega} - n_\omega$ with variation of the pressure at 1 atm in our case stands at $4.01\cdot10^{-6}$ for air, $3.89\cdot10^{-6}$ for nitrogen, and $4.07\cdot10^{-7}$ for helium.

[1)] All of the other optical elements and the phase difference at the exit of the nonlinear crystal can be calibrated with the help of the two-crystal technique presented in the Appendix.

[1] V. Osterberg and W. Margulis, Opt. Lett. **11**, 516 (1986); *ibid.* **2**, 57 (1987).

[2] R. H. Stolen and H. W. K. Tom, Opt. Lett. **12**, 585 (1987).
[3] N. B. Baranova and B. Ya. Zel'dovich, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 562 (1987) [JETP Lett. **45**, 717 (1987)].
[4] A. Krotkus and W. Margulis, Appl. Phys. Lett. **52**, 1942 (1988).
[5] H. W. K. Tom, R. H. Stolen, G. D. Anmiller, and W. Pleibel, Opt. Lett. **13**, 512 (1988).
[6] F. Ouellette, K. O. Hill, and D. C. Johnson, Opt. Lett. **13**, 515 (1988).
[7] M. V. Bergot, M. C. Farries, and M. E. Fermann, Opt. Lett. **13**, 592 (1988).
[8] W. Margulis, J. C. S. Carvalho, and J. P. Weid, Opt. Lett. **14**, 700 (1989).
[9] E. M. Dianov, P. G. Kazanskiĭ, and D. Yu. Stepanov, Kvantovaya Elektron. **16**, 887 (1989) [Sov. J. Quantum Electron. **19**, 575 (1989)].
[10] B. Lesche, J. Opt. Soc. Am. B **7**, 53 (1990).
[11] M. V. Éntin, Fiz. Tekh. Poluprovodn. **23**, 1066 (1989) [Sov. Phys. Semicond. **23**, 664 (1989)].
[12] E. M. Dianov *et al.*, *Proc. of the Conference on Integrated Photonics Research*, Hilton Head, S. C., 1990, p. MJ1.
[13] E. V. Anoikin, E. M. Dianov, P. G. Kazanskiĭ *et al.*, Pis'ma Zh. Tekh. Fiz. **15**, 78 (1989) [Sov. Tech. Phys. Lett. **15**, 33 (1989)].
[14] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory, 3rd ed.*, Pergamon Press, Oxford (1977).
[15] R. J. Glauber, in *Quantum Optics, Proc. of the Enrico Fermi Int. School in Physics*, ed. by R. J. Glauber, Academic Press, New York (1967), p. 15; R. J. Glauber, in *Proc. of the Tenth Session of the Scottish Universities Summer School in Physics*, ed. by S. M. Kay and A. Maitland, Academic Press, New York (1970), p. 53.
[16] B. Ya. Zel'dovich and A. N. Chudinov, Pis'ma Zh. Eksp. Teor. Fiz. **50**, 405 (1989) [JETP Lett. **50**, 439 (1989)].
[17] V. D. Volosov, A. G. Kalintsev, and V. N. Krylov, Pis'ma Zh. Tekh. Fiz. **2**, 85 (1976) [Sov. Tech. Phys. Lett. **2**, 32 (1976)].
[18] V. D. Volosov, A. G. Kalintsev, and V. N. Krylov, Kvantovaya Elektron. **3**, 2139 (1976) [Sov. J. Quantum Electron. **6**, 1163 (1976)].
[19] D. N. Nikogosyan and G. G. Gurzadyan, Kvantovaya Elektron. **14**, 1529 (1987) [Sov. J. Quantum Electron. **17**, 970 (1987)].

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