Interference of fields with frequencies ω and 2ω in external photoelectric effect

B. Ya. Zel'dovich and A. N. Chudinov
Institute of Electrophysics, Ural Branch of the Academy of Sciences of the USSR

(Submitted 10 October 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 10, 405–407 (25 November 1989)

An interference of the amplitudes of the one-photon and two-photon photoelectric effects, predicted previously, has been observed during the exposure of a photomultiplier cathode to the beam from a pulsed neodymium laser and to the second harmonic of this beam simultaneously. A specific polar asymmetry of the interference term in the photocurrent has been found.

Baranova and Zel'dovich[1] have predicted a polar asymmetry in the probability for photoionization during the application of fields with a nonzero mean cube, $\langle \mathbf{E}^3 \rangle \neq 0$, to a medium. If a medium is exposed to a laser beam $E_L \exp(-i\omega_1 t)$ which causes two-photon ionization and also the second harmonic of this beam, $2\omega_1$, which causes one-photon ionization, the quantum-mechanical amplitude for thee process is a sum of the type $C_{ioniz} = a_2 E_1^2 e^{-2i\omega_1 t} + a_1 E_2 e^{-2i\omega_1 t}$, so the probability for the process contains three terms: $|a_2|^2 |E_1|^4$, $|a_1|^2 |E^2|^2$, and the interference term $Re(a_2 a_1^* E_1^2 E_2^*)$. It is this interference term in which we are interested here. Because of its tensor nature, this term changes sign under an inversion of the coordinate system, so an asymmetric component, e.g., of the type

$$\mathbf{j} \propto \mathrm{Re}[e^{i\gamma}(\mathbf{E}_2 \mathbf{E}_1^*)\mathbf{E}_1^*]. \qquad (1)$$

appears in the photocurrent.

In an effort to observe this predicted effect, we have used the experimental apparatus shown in Fig. 1, which is based on the 'EKSMA pulsed Nd:YAG laser. The length of an individual pulse in the output, which had single transverse mode, was $\tau_p \sim 27$ ps. Some of this light was converted into the second harmonic in a KDP crystal. After the beams passed through a Glan prism and a plane-parallel plate, they underwent Fresnel reflection from a glass wedge and were directed without focusing to the photocathode of an FÉU-127 photomultiplier. The red boundary of the photoelectric effect of the antimony-cesium cathode of the FÉU-127 is at the wavelength 0.6 μm, so laser light with $\lambda_1 = 1.06\ \mu$m should have caused only a two-photon photoelectric effect, while for the second harmonic, with $\lambda_2 = 0.53\ \mu$m, the one-photon effect would be possible. In some preliminary experiments in which we used only the λ_1 wave for illumination, we verified that the photoelectric effect was of a two-photon nature. The experimental I_1 dependence of the photocurrent turned out to be quadratic over a wide range of the intensity I_1. At values of I_1 above a certain level, there was an abrupt change in regime: The photocurrent reached saturation, and the photocathode began to be destroyed. In order to improve the accuracy of the measurements of I_1^2, we carried out measurements on the basis of the second-harmonic signal generated in a separate $LiNbO_3$ crystal in these control experiments.

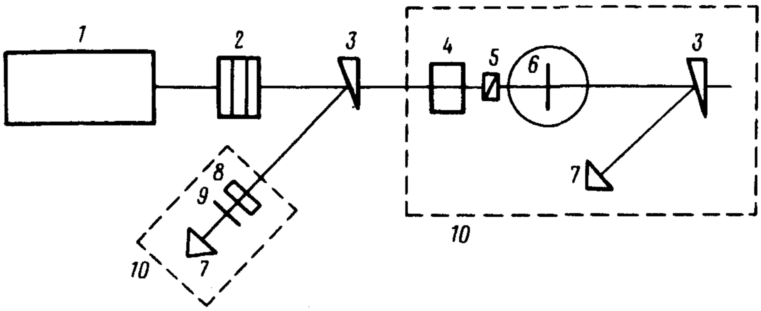

FIG. 1. Experimental layout. 1—ÉKSMA picosecond-pulse YAG laser; 2—cell holding an aqueous solution of $CuSo_4$; 3—glass wedge; 4—KDP crystal; 5—Glan prism; 6—plane-parallel glass plate, on rotating table, which introduces a phase shift; 7—FÉU-127 photomultiplier; 8—$LiNbO_3$ crystal; 9—SZS-21 filter; 10—darkened box.

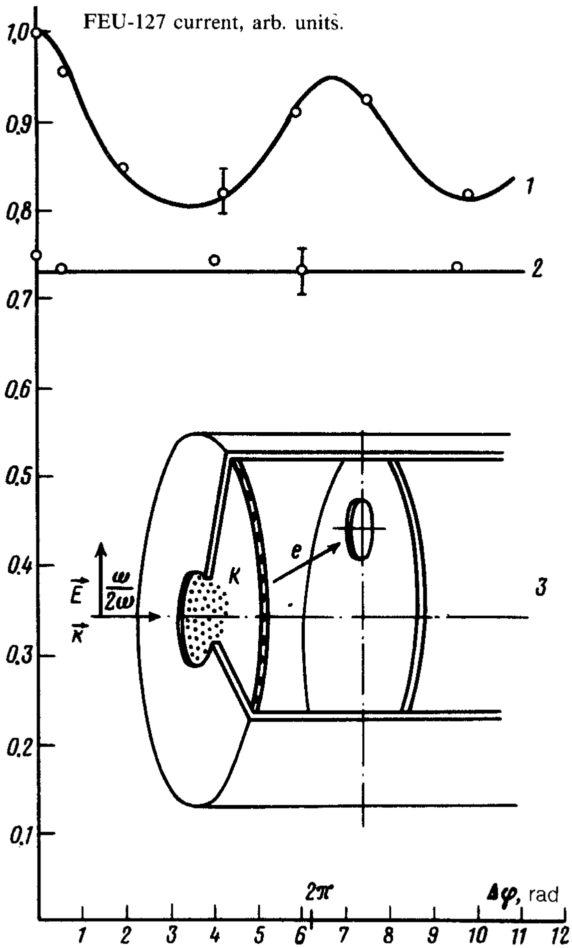

FIG. 2. Experimental results on the photomultiplier current versus $\Delta\varphi$, 1,3—The polarizations of the ω and 2ω beams are directed toward the hole; 2—the polarizations of the ω and 2ω beams are directed perpendicular to the hole.

The measurements were carried out at intensities I_1 corresponding to an unsaturated I_1^2 dependence of the photocurrent. The coefficient of the conversion into the second harmonic in the KDP crystal was chosen at a level such that the signal representing the one-photon photocurrent, proportional to I_2, would be approximately equal in magnitude to the signal $\propto I_1^2$.

To observe the interference, we varied the phase difference $\Delta\varphi = \varphi_2 - 2\Delta\varphi_1$ by rotating the plane-parallel glass plate, of thickness d = 1.4 mm, through an angle θ, so we had a phase difference $\Delta\varphi \approx [n(2\omega)_1) - n(\omega_1)]d\theta^2$, where n is the refractive index. The θ dependence of $\Delta\varphi$ was calibrated experimentally in an arrangement in which the plate was placed between two nonlinear crystals generating the second harmonic. To avoid disrupting the matching of the wavefronts of the interfering beams, we did not focus them or and pass them through dispersive wedges.

Figure 2 shows the photocurrent (in arbitrary units) versus thee phase difference $\Delta\varphi$. The polar asymmetry is embodied in the design of the FÉ-127. The light was incident normally on the surface of the photocathode. The points on the upper curve, where there is an obvious interference, correspond to a linear polarization of both waves in the direction toward the hole. The relative amplitude (a) of the interference term $I = I_1[1 + \cos(\Delta\varphi + \alpha)]$ was $a = 0.1 \pm 0.03$.

The points on the lower curve correspond to a linear polarization of both waves, transverse with respect to the direction toward the hole. In this case there was no interference, within the experimental error.

We were not able to determine the absolute phase α of the interference term (this phase is an intrinsic property of the photocathode, whose thickness was ~ 0.03 μm), since the glass window of the photomultiplier, ~ 1 mm thick, was in the path of the light and introduced a phase shift which we do not know.

In summary, we have observed a polar asymmetry of the photoionization caused by a field with $\langle \mathbf{E}^3 \rangle \neq 0$. This asymmetry results from an interference of the one-photon and two-photon transitions. An asymmetry of this type might have contributed to the writing of holograms of a quadratic optical polarizability which was recently observed in optical fibers (see Ref. 2; see also the discussion in Refs. 1 and 3).

We wish to thank N. B. Baranova, who collaborated in the prediction of the effect, for useful discussions.

[1]N. B. Baranova and B. Ya. Zel'dovich, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 562 (1987) [JETP Lett. **45**, 717 (1987)].
[2]U. Osterberg and W. Margulis, Opt. Lett. **11**, 516 (1986).
[3]R. H. Stolen, Opt. Lett. **12**, 585 (1987).

Translated by Dave Parsons