Observation of an interference of one- and two-photon ionization of the sodium 4*s* state

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A beam of Na atoms in the 4*s* state, with a binding energy $I = 1.97$ eV, was exposed to light pulses at $\lambda_1 = 1064$ nm from a Nd:YAG laser and also the corresponding second harmonic, at $\lambda_2 = 532$ nm. The light at these two wavelengths caused a two-photon ionization and a one-photon ionization, respectively; $\hbar\omega_1 < I < 2\hbar\omega_1$. The experiments reveal an oscillatory dependence of the flux of electrons emitted in a certain direction on the phase difference between $E^2(\omega_1)$ and $E(2\omega_1)$; i.e., the experiments reveal the interference in the title of this paper.

1. The detection of self-organized, optical, second-harmonic generation in fused-quartz optical fibers[1] has raised the idea that there might be a polar asymmetry in the emission of electrons during the ionization of atoms (and also during the ionization of molecules or defects in a solid). This asymmetry would result from an interference between two-photon absorption of the laser light, $E(\omega)\exp(-i\omega t)$, and one-photon absorption of the second harmonic of this light, $E(2\omega)\exp(-2i\omega t)$ (Refs. 2 and 3). This interference would occur because an emitted electron would be excited by two processes into the same state in the continuous spectrum, $\exp(i\vec{k}\vec{r})$ (Fig. 1). A plane wave in the continuous spectrum would be excited in a one-photon process through a p state and in a two-photon process through s and d states, if the process started with a bound s electron. The overall wave function would then lead to a polar asymmetry in the emission probability (Fig. 2). In some previous studies[4] with a photomultiplier cathode having a red sensitivity boundary $\lambda = 600$ nm, we experimentally observed an interference of this type when the cathode was illuminated by copropagating pulses with $\lambda = 1064$ nm and $\lambda = 532$ nm. Unfortunately, the theory derived in Refs. 2 and 3 for isolated atoms is not applicable to the photoelectric effect at a solid (i.e., at this cathode). It thus appeared worthwhile to attempt to detect a polar asymmetry caused in the emission by an $\omega/2\omega$ interference for atoms.

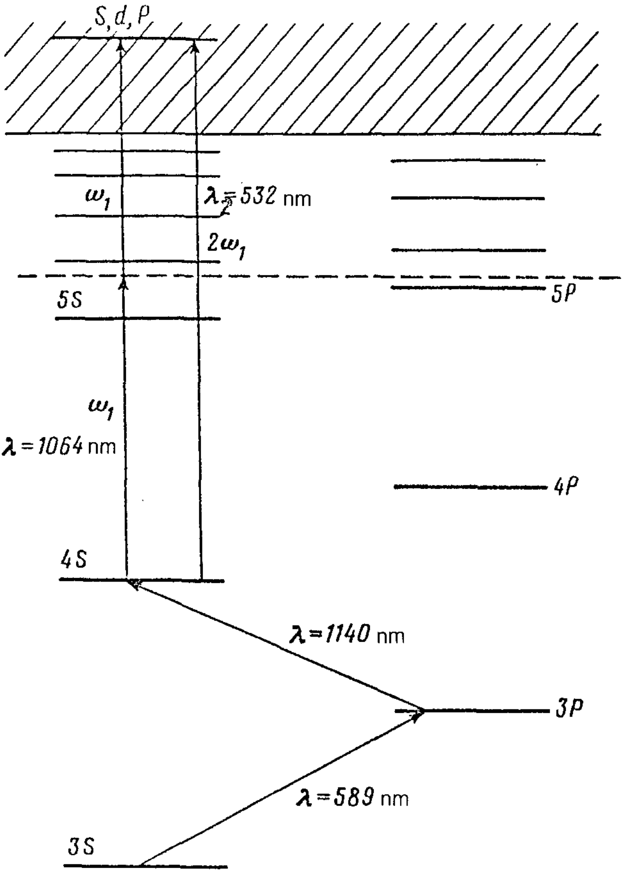

FIG. 1. Energy level diagram and processes describing the one-and two-photon ionization. The sodium atom, in the 4s excited state, has an ionization potential of 1.97 eV. It is exposed to a pump field $E(\omega_1)$ with $\hbar\omega_1 = 1.16$ eV and also to the second harmonic of this field, $E(2\omega_1)$, with $\hbar 2\omega_1 = 2.32$ eV.

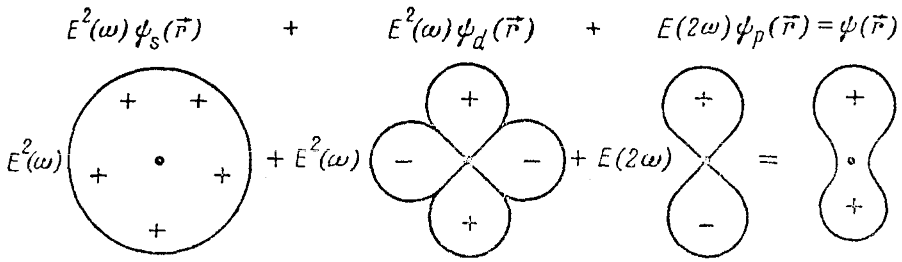

FIG. 2. Diagram illustrating how the *s*, *p*, and *d* waves in the continuous spectrum combine into a common function having a polar asymmetry in the emission probability distribution.

We selected sodium atoms in the 4*s* state for these experiments for several reasons. One is the presence of a 5*p* level with a transition energy $\hbar\omega(4s\text{–}5p) = 1.15$ eV, which is nearly resonant for the wavelength of a neodymium laser ($\hbar\omega_1 = 1.16$ eV). The probability for two-photon ionization is thus substantially increased. Phase-dependent interference effects were recently detected by Muller *et al.*[5] in the seven-photon and eight-photon ionization of krypton atoms.

2. The experimental apparatus is shown in Fig. 3. A source *S* is enclosed in a vacuum chamber *C*. Sodium vapor is produced in this source by evaporating sodium at $T = 513$ K. A vertically directed atomic beam is obtained as the Na vapor escapes through a hole $\simeq 1$mm in diameter. To excite the Na from the 3*s* state to the 3*p* state, we used a lens *A* to focus the light from laser *L* 1. This laser was a dye laser using the dye rhodamine 6*G*, with $\lambda = 589$ nm. This dye laser was pumped by the second harmonic produced inside the resonator of pulsed Nd:YAG laser *L* 2 [this was in-resonator second-harmonic generation (IRSHG)]. The further excitation from the 3*p* level to the 4*s* level was achieved by focusing the beam from laser *L* 3 in the same place. The latter laser was an F_2^-:LiF color-center laser, with $\lambda = 1140$ nm, pumped by another Nd:YAG laser, *L* 4. Both beams, that with $\lambda = 589$ nm and that with $\lambda = 1140$ nm, entered the chamber through window W_1. The beam from neodymium laser *L* 5 and its second harmonic ($\lambda = 1064$ and 532 nm, respectively) entered from the opposite direction, through window W_2. The second-harmonic generation occurred in a KTP crystal, at the exit from which we obtained a beam $E(2\omega_1)$ with a horizontal linear polarization and a beam $E(\omega_1)$ with a fixed (generally elliptical) polarization. The length of the $E(\omega_1)$ pulses was on the order of 10^{-7} s. These pulses were generated at a repetition frequency of 5 kHz and had an average power $\simeq 6$ W. The average power of the $E(2\omega_1)$ beam ranged from 0.06 to 0.2 W. The average power of the beam with $\lambda = 589$ nm was 0.025 W, and that of the beam with $\lambda = 1140$ nm was 0.01 W. The triggering of all three neodymium lasers was electrically synchronized. All these lasers generated pulses of about the same length.

A phase shift $\Delta\varphi$ between the square of the pump field, $E^2(\omega_1)$, and the second-harmonic field $E(2\omega_1)$ was introduced by rotating a plane-parallel glass plate (G) with a thickness of 7 mm. The dependence of $\Delta\varphi$ on the rotation angle θ was calibrated beforehand by the procedure of Ref. 6. For this particular plate, the corresponding

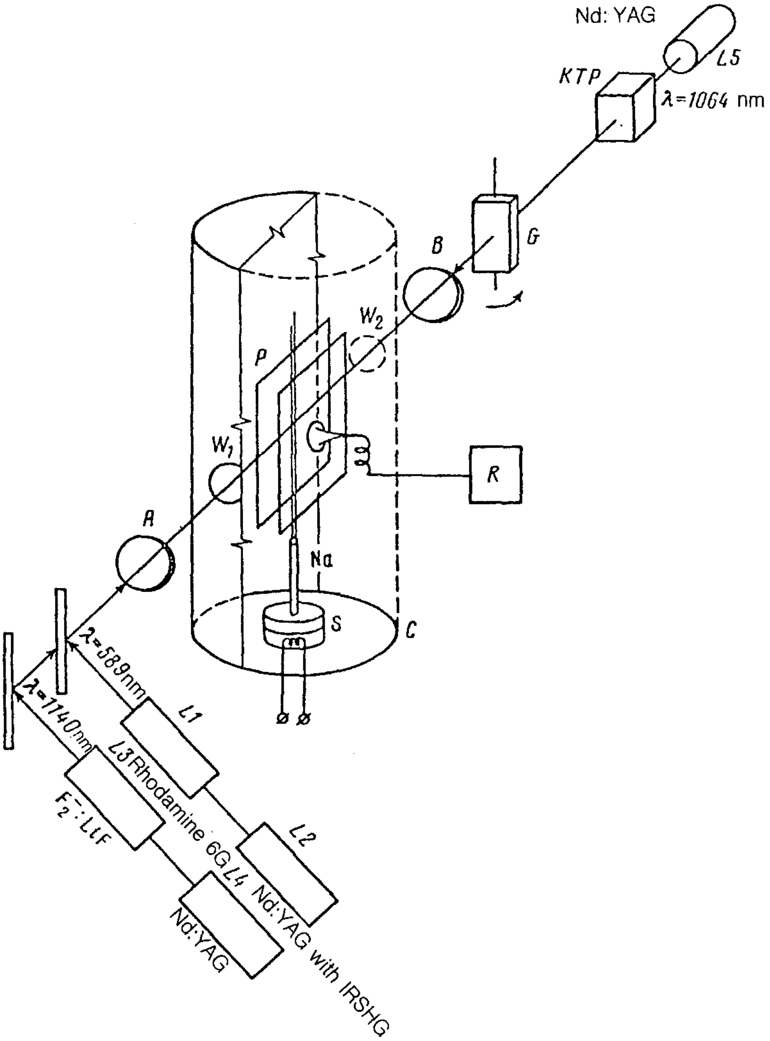

FIG. 3. Experimental layout.

relationship is $\Delta\varphi = 0.075 \cdot \theta^2$, where $\Delta\varphi$ is in radians, and θ in degrees. Both light beams, $\simeq 1$ mm in diameter, were focused on the atomic beam by lens *B*, with a focal length of 21 cm.

Although we realize that the chromatic aberrations induced by lens *B* and window W_2 pose a difficult problem, we took no measures to combat them. It proved to be exceedingly difficult to bring all four beams together. In particular, this difficulty has so far prevented us from carrying out experiments with all possible polarizations of the $E(\omega_1)$ and $E(2\omega_1)$ waves. The electrons produced in the ionization were detected by a VÉU-6 channel electron multiplier. The entrance aperture of the VÉU-6 was positioned opposite the illuminated part of the atomic beam in the direction of the polarization $\vec{E}(2\omega_1)$. It was mounted in a hole in one of two metal plates *P*. The distance between these plates was 8 mm. The output signal from the electron multiplier was sent to a chart recorder *R*.

When a positive voltage $U = 30$ V was applied to the plate attached to the electron multiplier, all the free electrons that formed apparently reached the entrance to this multiplier. When there was no voltage on the multiplier, in contrast, apparently the only electrons which were detected were those whose velocity vector was directed toward the aperture. This conclusion is supported, in particular, by the following results. At $U = 30$ V, the signal for both the two-photon ionization by the $\vec{E}(\omega_1)$ beam and the one-photon ionization by the $\vec{E}(2\omega_1)$ was independent of the polarization of the light. When the voltage was removed from the plates $(U = 0)$, the signals decreased by a factor of about 10. These signals reached a maximum in the case of a linear polarization in the direction of the aperture of the electron multiplier. They decreased by another factor of 3 or 4 when the polarization was rotated 90°, for both $\vec{E}(\omega_1)$ and $\vec{E}(2\omega_1)$. The signal from the electron multiplier had a small background $(\leqslant 10\%)$ due to one-photon ionization of the 4*s* state by the yellow light from laser *L* 1 $(\lambda = 589$ nm$)$. After this background was subtracted, the plots of the output signal of the electron multiplier versus $|E(\omega_1)|^4$ and $|E(2\omega_1)|^2$ were linear. By adjusting the conditions for the second-harmonic generation in the KTP crystal, we were able to make these two components of the signal comparable in magnitude. Unfortunately, it was difficult to equalize these components more precisely than within a factor of 2.

3. We turn now to the results of the interference measurements. Figure 4 shows

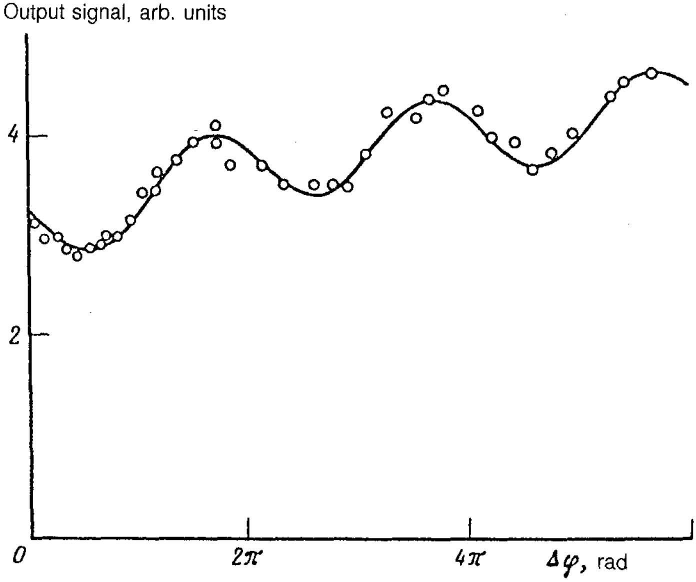

FIG. 4. Experimental output signal from the electron multiplier as a function of the phase shift $\Delta\varphi$ between the fields $E(2\omega_1)$ and $E(\omega_1)$.

the output signal from the electron multiplier versus the phase shift $\Delta\varphi$ between $E^2(\omega_1)$ and $E(2\omega_1)$. This phase shift was introduced by rotating the glass plate. The contrast $(S_{\max} - S_{\min})/(S_{\max} + S_{\min}) \simeq 15\%$ and the periodicity, with a period of 2π, can be seen absolutely reliably. We should point out that difficulties in bringing the beams into coincidence limited the number of cases with "good" results like these to about 6 out of 13. In another five or so cases the interference was obvious, but the constrast was slightly poorer. In two cases, the noise prevented reliable detection of an interference. The monotonic trend in the plot of the signal versus $\Delta\varphi$ or, more precisely, versus the plate rotation angle θ is apparently due to a transverse displacement of the beam.

4. Conclusion. In summary, a polar symmetry caused in the distribution of emitted electrons by an interference between one-photon and two-photon ionization of free atoms has been detected in these experiments (for the first time, to the best of our knowledge). The atoms were ionized by exposing them to a field with a nonzero average cubic value $\langle E^3\rangle = E_1^2(\omega)E_2^*(2\omega) + \text{c.c.}$ We think it would be interesting to test the predictions based on the quantum theory of scattering of electrons by an atomic core[2,6] regarding the behavior of the phase of the interference term as a function of the polarizations of the wave $\vec{E}(\omega_1)$ and $\vec{E}(2\omega_1)$.

[1] V. Osterberg and W. Margulis, Opt. Lett. **11**, 516 (1986).

[2] N. B. Baranov and B. Ya. Zel'dovich, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 562 (1987) [JETP Lett. **45**, 717 (1987)]; N. B. Baranov and V. Ya. Zel'dovich, J. Opt. Soc. Am. B **8**, 27 (1991).

[3] M. V. Éntin, Fiz. Tekh. Poluprovodn. **23**, 1066 (1989) [Sov. Phys. Semicond. **23**, 1066 (1989)].

[4] B. Ya. Zel'dovich and A. N. Chudinov, Pis'ma Zh. Eksp. Teor. Fiz. **50**, 405 (1989) [JETP Lett. **50**, 439 (1989)]; N. B. Baranova, A. N. Chudinov, and V. Ya. Zel'dovich, Opt. Commun. **79**, 116 (1990); N. B. Baranova, B. Ya. Zel'dovich, A. N. Chudinov, and A. A. Shul'ginov, Zh. Eksp. Teor. Fiz. **98**, 1857 (1990) [Sov. Phys. JETP **71**, 1043 (1990)]; A. N. Chudinov, A. A. Shulginov, and B. Ya. Zel'dovich, Opt. Lett. **16**, 1346 (1991).

[5] H. G. Muller, P. H. Bucksbaum, D. W. Schumacher, and A. J. Zavriyer, Phys. B **23**, 2761 (1990).

[6] A. N. Chudinov, Yu. E. Kapitsky, A. A. Shulginov, and B. Ya. Zel'dovich, Opt. Quantum Electron. **23**, 1055 (1991).

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