Photoinduced current in a low-density gas

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(Submitted 10 September 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **40**, No. 9, 374–377 (10 November 1984)

A new photovoltaic effect is predicted: the appearance of an electric current in a low-density gas as this gas interacts resonantly with electromagnetic radiation. Corresponding experiments reveal a current when sodium vapor is illuminated with the beam from a dye laser. This photoinduced current might be developed into a new method of intra-Doppler spectroscopy.

In this letter we discuss a new photovoltaic effect: a photoinduced current in a low-density gas. During the resonant application of light to a low-density neutral gas (in which the mean free path is much longer than the scale dimension of the absorbing cell), an electric current collinear with the wave vector can arise in the gas. The direction of the current reverses upon a change in the sign of Ω, the deviation of the frequency of the light from the center of the absorption line.

What is the physics involved in this effect? Let us assume that light in the form of a traveling quasimonochromatic wave is absorbed at a transition of a particle from its ground state to its excited state. Because of the Doppler effect, the only particles that will interact with the light are those whose velocity projections v_x onto the wave vector $\mathbf{k}$ are approximately equal to Ω/k (it is assumed that Doppler broadening is predominant). As a result of optical transitions, excited particles appear with a directed velocity $v_x = \Omega/k$. We next assume that the particle is ionized from its excited state by some mechanism, e.g., an auxiliary light beam. Since the particle goes into the ionization reaction with a definite velocity v_x, the resulting ion and the electron will have the same average velocity. The mass of the electron, however, is much smaller than that of the ion, so that the modulus of the electron's velocity will be substantially greater than the velocity of its directed motion, $v_x = \Omega/k$. In other words, the electrons will move in an essentially isotropic manner and (in a collisionless situation) will rapidly escape to the walls. Only the ions will be left in the volume, and their directed motion constitutes an electric current. Depending on the sign of Ω, this current will be directed either parallel or antiparallel to the wave vector. Exactly at resonance ($\Omega = 0$), there will be no current.

This effect is a true "galvanic" effect, distinguished from the familiar photovoltaic effect (see Refs. 1 and 2, for example), which results in a change in the conductivity of a gas (or plasma), and which requires an external potential difference. On the other hand, similar photovoltaic effects have been discussed in Refs. 3–5; the basic physics of the effects of Refs. 3–5 and that of the present effect is closely related to the physics of the photoinduced drift effect.[6] A difference between the effect discussed here and those of Refs. 3–5 is that we disregard slowing collisions in the volume. It turns out that in a Knudsen situation, in which the momentum relaxation of the ions and electrons occurs at the wall, the photoinduced current can reach a much higher level, so that it can easily be detected experimentally.

A quantitative description of the effect is simplest in a plane geometry: The absorbing cell is a plane capacitor, and the light is propagating parallel to its plates. If space charge is ignored, the current through the capacitor is described by the obvious expression

$$J = e \int d\mathbf{r} \int_0^\infty [q(v_x, \mathbf{r}) - q(-v_x, \mathbf{r})] \, d v_x , \tag{1}$$

where $q(v_x, \mathbf{r})$ is the number of ion-production events per unit time, per unit volume, per unit interval of the velocity v_x. If the ionization is caused by an auxiliary light beam, then we have $q(v_x, \mathbf{r}) = \xi p(v_x, \mathbf{r})$, where $p(v_x, \mathbf{r})$ is the "rate" of production of excited particles, and ξ is the probability for ionization after the excitation event. For a moderate intensity of the exciting light, we find

$$J = e w \xi \frac{2}{\pi} \operatorname{arc\,tg} \left(\frac{\Omega}{\Gamma} \right) e^{-(\Omega/k\bar{v})^2} . \tag{2}$$

Here Γ is the homogeneous half-width of the absorption line, $k\bar{v}$ is the Doppler parameter, and w is the number of photons of the exciting light which are absorbed in the cell per unit time.

Figure 1a is a sketch of the function $J(\Omega)$. The current switches direction in a narrow transition region $\Delta\Omega \sim \Gamma \ll k\bar{v}$.

To estimate the magnitude of the effect, we assume that $\sim 10^{-2}$ W of the exciting light is absorbed in the working volume. In the optical part of the spectrum, this power corresponds to $w \sim 10^{17}$ s^{-1}, so we can write

$$J \sim 10^{-2} \xi \ \text{A}. \tag{3}$$

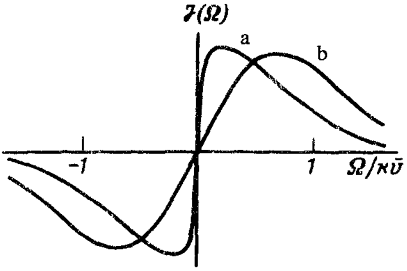

FIG. 1. The function $J(\Omega)$. a—For conditions corresponding to free motion of the ions, with $\Gamma/k\bar{v}=10^{-2}$; b—for plasma conditions.

Advanced instruments can easily detect currents $\sim 10^{-14}$ A and below. The photoinduced current can thus be observed at probabilities as low as $\xi \sim 10^{-12}$. If $\xi \sim 1$, then we find $J \sim 10^{-2}$ A from (3). Even as a much lower current, plasma conditions may prevail in the volume (the Debye length is much shorter than the spacing between the walls), so that (2) would be replaced by

$$J \cong eq_0\sqrt{T/T_e}\ (\Omega/k\bar{v})\, e^{-(\Omega/k\bar{v})^2}, \tag{4}$$

where q_0 is the number of ions produced in the working volume per unit time, and T and T_e are the temperatures of the gas and the electrons. In this case, the frequency dependence $J(\Omega)$ is smoother (Fig. 1b), with a single scale value $\Omega/k\bar{v} \sim 1$.

In several cases, it is not necessary to take special measures regarding the ionization of the excited particles, since this ionization may occur anyway, by virtue of certain types of collision processes (associative ionization, for example). Although collisions are extremely rare, the number of ions produced is quite sufficient for a detectable effect. Let us examine the particular case of the associative ionizaiton of sodium:

$$\mathrm{Na}(3p)+\mathrm{Na}(3p) \rightarrow \mathrm{Na}_2^+ + e, \tag{5}$$

for which we find the cross sections $\sigma = 1.5\times 10^{-16}$ cm^2 (Ref. 7) and $\sigma = 0.48\times 10^{-16}$ cm^2 (Ref. 8) in the literature. For q_0 we have the following estimate from (4):

$$q_0 \sim (w\tau)^2\, \sigma\bar{v}/V, \tag{6}$$

where $\tau \sim 10^{-8}$ s is the lifetime of the excited state of sodium, and V is the volume of the region in which the light is absorbed. If a power ~ 3 mW is absorbed in the sodium D line, and if we have $V \sim 5\times 10^{-5}$ cm^3, $\bar{v} \sim 5\times 10^4$ cm/s, and $\sqrt{T/T_e} \sim 1$, we estimate the resulting current to be $J \sim 10^{-9}$ A, which could easily be detected with standard instruments.

We carried out an experiment to detect this photoinduced current in sodium vapor excited by the quasimonochromatic beam from a dye laser. The light beam (10^{-2} cm in diameter) enters through 1-mm apertures in the plates of a plane capacitor on two side of the sodium vapor. The distance between the plates is 3 mm. At a laser power $\cong 100$ mW, $\cong 3$ mW is absorbed in the cell. To distinguish the effect of interest from the background of masking effects (effects that are symmetric in Ω and that

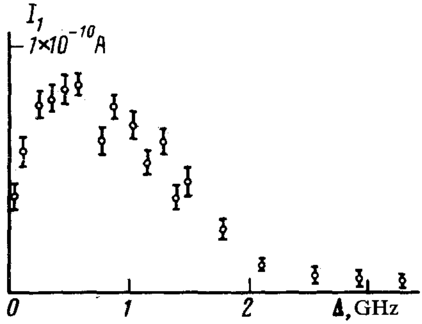

FIG. 2. Experimental current signal J_I at the deviation frequency versus the deviation amplitude Δ.

result from the ordinary photoelectric effect caused by the scattered light), we arranged a symmetric deviation of the light frequency from the center of one of the sodium D lines. The output signal at the deviation frequency from the current meter was measured as a function of the amplitude of the deviation. Figure 2 shows the results. They are evidence of a photoinduced current with its distinctive (antisymmetric) dependence on Ω. Assurance that the photoelectric effect has been suppressed comes from the absence of a signal at the deviation frequency from a photodetector that detects the scattered light. The magnitude of the photoinduced current reaches $\sim 10^{-10}$ A, in agreement with the estimates above for the associative-ionization mechanism.

Further study of this photoinduced current seems promising, particularly since this effect might be developed into a new method of intra-Doppler spectroscopy.

We wish to thank S. P. Pod'yachev and D. A. Shapiro for useful discussions throughout this study.

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Translated by Dave Parsons
Edited by S. J. Amoretty