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OBSERVATION OF TWO-PHOTON-INDUCED PHOTOEMISSION OPTOGALVANIC EFFECT USING COPPER TARGET ELECTRODE

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Two-photon-induced photoemission optogalvanic effect using copper as the target electrode has been observed in a nitrogen discharge cell using 532 nm radiation from a frequency-doubled Nd:YAG laser as the pump source.

1. Introduction

The interaction between coherent radiation and a gaseous discharge has been a subject of intensive investigations by many workers. One such study involves the measurement of the change in impedence of the discharge in the presence of absorbing radiation. This phenomenon known as optogalvanic (OG) effect has been perfected as an effective tool in the study of discharge plasma dynamics[1] and in high resolution spectroscopy.[2] A change in impedence of a discharge plasma can also be brought about by photoelectric emission or photoemission from a solid target kept as an electrode in the plasma medium under laser irradiation.[3] Such a perturbation due to photoelectric emission in a discharge medium is known as photoemission optogalvanic (POG) effect. Unlike in the case of OG effect, POG effect is a nonresonant phenomenon, since we can observe POG effect for all wavelength of radiation below a threshold value. POG effect has now been recognized as a technique to study the surface characteristics of solid targets (electrodes).[4,5] Photoelectric emission can also take place under multiphoton excitation mechanism. H. Chen *et al.*[6,7] observed plasmon-mediated enhancement in two-photon-induced photoelectric emission from aluminium and silver under laser excitation. The present paper deals with the observation of two-photon-induced POG effect from a copper target using a frequency-doubled pulsed Nd:YAG laser.

2. Experimental Setup

The discharge cell used in our experiment consists of a glass tube of 1 cm in diameter socketed into two metal end caps made of stainless steel. The separation between the

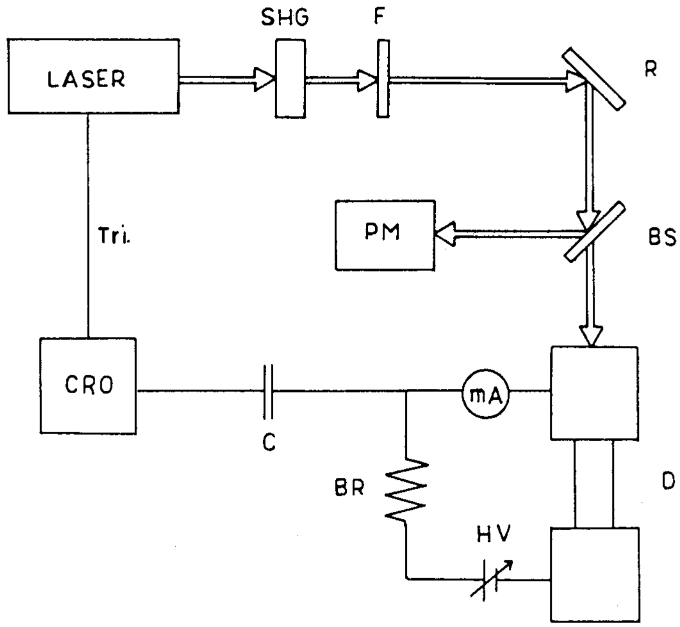

Fig. 1. Schematic of the experimental setup (SHG — second harmonic generator, F — harmonic separator, R — reflector, BS — beam splitter, PM — power meter, D — discharge cell, BR — ballast resistance).

ends of the metal caps is 3 cm and they act as electrodes. One end of the cylindrical cap is provided with a glass window while a copper target is fixed inside the metal cap at the other end of the discharge tube. The separation between the copper target and the end of the other electrode is about 4.5 cm. Suitable side tubes for continuous flow of gas into the cell are also provided. The schematic of the experimental setup is given in Fig. 1. The discharge is excited using a low noise high voltage dc power supply (Thorn EMI). A ballast resistance of 66 kΩ is also included in the circuit so as to limit the current in the circuit. The discharge in the cell was maintained steady at various pressures of nitrogen gas. Pulsed radiation at 532 nm (pulse width 10 ns, repetition rate 10 Hz) from a frequency-doubled Nd:YAG laser (Quantay Ray DCR-11) was used to excite POG signals from the copper target. The signal developed across the load resistor was fed to a digital storage oscilloscope (Iwatsu DS-8621, 200 MHz) through a capacitor (0.1 μF). The signal strength is measured directly from the oscilloscope trace.

3. Results and Discussion

According to the generalized Fowler–Dubridge theory, the total electron density emitted from a target under laser irradiation is

$$J(r, t) = J_0(r, t) + J'(r, t), \tag{1}$$

where $J'(r, t)$ is the photoemission photocurrent, while $J_0(r, t)$ is the contribution from thermionic emission. The photoemission photocurrent J' is given by

$$J'(r, t) = a\left(\frac{e}{h\nu}\right) A(1-R)PTF\left(\frac{h\nu - \phi}{k_B T}\right), \tag{2}$$

where R, A, P, ϕ, T, and a are the sample surface reflectivity, Richardson coefficient, laser power, work function of the sample, sample temperature, and sample-dependent constant, respectively. One can also observe n-photon-induced photoelectric effect if the work function of the target is equal to $nh\nu$. Then, we can write the n-photon-induced photoemission photocurrent as

$$J'_n(r, t) = a_n\left(\frac{e}{h\nu}\right)^n A(1-R)^n P^n T^n F\left(\frac{nh\nu - \phi}{k_B T}\right). \tag{3}$$

The function $F(x)$ known as Fowler function[8,9] in Eqs. (2) and (3) is given by

$$F(x) = \begin{cases} \displaystyle\sum_{m=1}^{\infty}(-1)^{m+1}\frac{e^{mx}}{m^2}, & \text{for } x \le 0, \\ \displaystyle\frac{\pi^2}{6} + \frac{x^2}{2} - \sum_{m=1}^{\infty}(-1)^{m+1}\frac{e^{-mx}}{m^2}, & \text{for } x \ge 0. \end{cases} \tag{4}$$

In the present case, $x > 0$.

From Eq. (3), it is clear that the slope of the log–log plot of POG signal strength against laser power gives the number of photons taking part in the multiphoton process. Studies were carried out by varying the gas pressure, laser power, and voltage across the cell. Irradiation of the copper target with 532 nm laser radiation does not give rise to any measurable signal in the absence of an applied voltage across the cell.

The variation of POG signal as a function of laser power for different discharge voltages (forward biased with copper as cathode) at 170×10^{-6} bar of nitrogen gas pressure is shown in Fig. 2. The signal strength increases with the laser power as well as with the applied voltage. At lower laser power, the POG signal is almost the same for different values of applied voltage. The work function of copper is 4.4 eV,[10] which is slightly lower than the two-photon energy (4.6 eV) at 532 nm so that we can expect two-photon-induced photoemission from copper target. In Fig. 3, log–log plots of laser power against signal strength for three different applied voltages are given. The three plots are parallel and have a slope of nearly 2 which clearly confirms the two-photon process in POG effect observed here.

The quantum efficiency (Q_f) of photoemission is defined by

$$Q_f = \frac{N_e}{N_{ph}}, \tag{5}$$

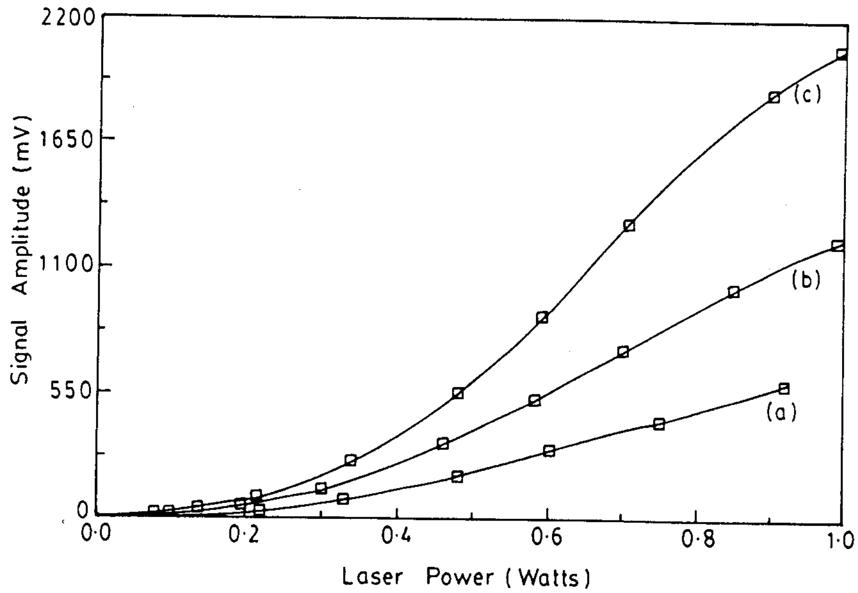

Fig. 2. Variation of the signal amplitude with laser power: (a) – 600 V, (b) – 700 V, (c) – 800 V.

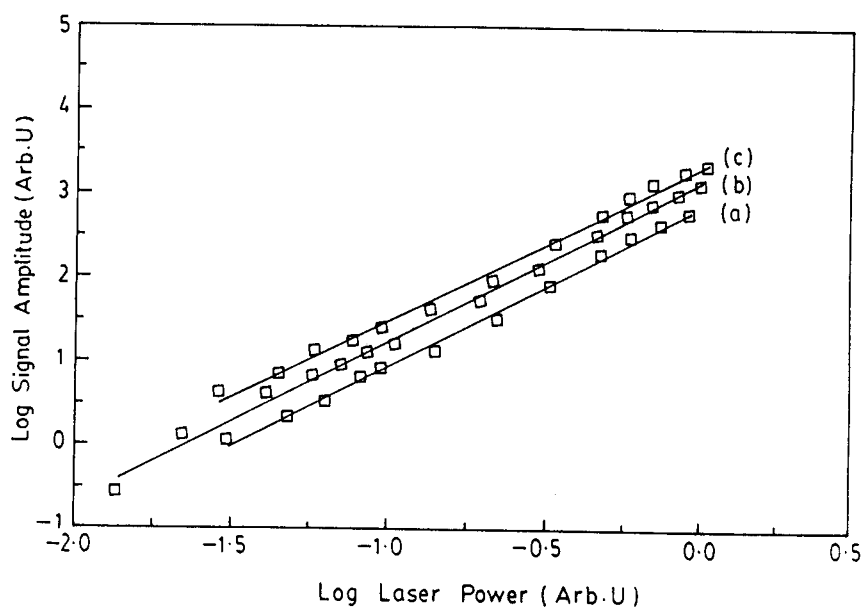

Fig. 3. Log–log plot of laser power against POG signal amplitude: (a) – 600 V, (b) – 700 V, (c) – 800 V.

where N_e and N_{ph} are, respectively, the number of photoelectrons ejected out and the number of photons incident on the target.

$$N_e = \frac{It}{e}, \tag{6}$$

where I is the electron current and t is the current pulse duration, and

$$N_{ph} = \frac{\text{laser pulse energy}}{h\nu}. \tag{7}$$

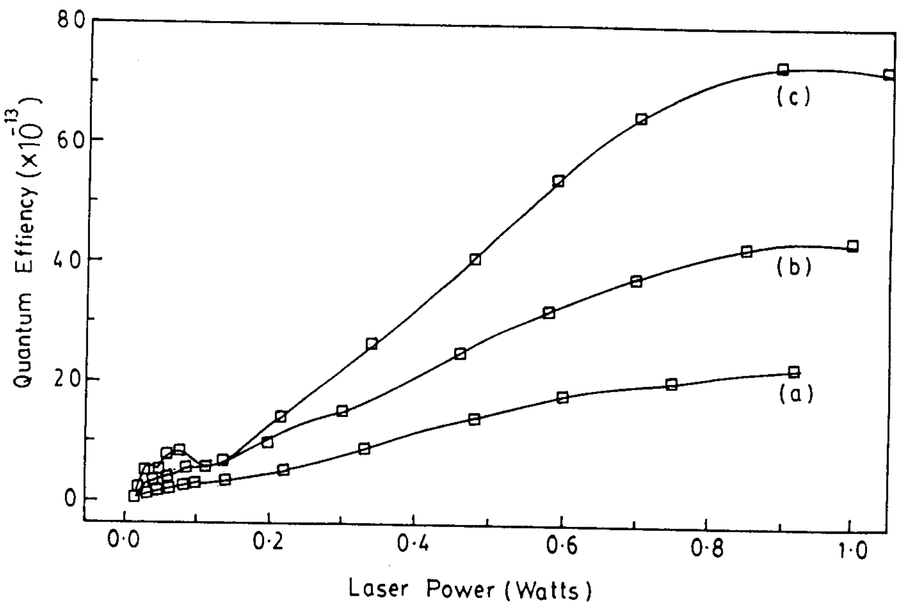

Fig. 4. Variation of quantum efficiency with respect to laser power: (a) – 600 V, (b) – 700 V, (c) – 800 V.

Figure 4 shows the variation of overall quantum efficiency Q_f as a function of laser power and the applied field E. The Q_f values are comparatively low because of the involvement of the two-photon process which has a much lower probability in the present case. However, as the laser power is increased, enhancement in the value of Q_f is noticed. This apparently is an indication of the probable role played by thermionic electrons released by rapid heating of the target surface by the intense laser pulse. An increase in the applied voltage also enhances the electron density (due to collisional ionisation) and thus the overall quantum efficiency in POG effect increases with the field. Also, enhancement in the value of Q_f due to the applied field across the cell can be accounted for as the dependence of photoelectric efficiency η on E is given by

$$\eta = A\left[2h\nu - \phi - \left(\frac{eE\beta}{4\pi\varepsilon_0}\right)^{1/2}\right], \tag{8}$$

where A depends on the cathode material parameter, ϕ is the work function, and β is the enhancement factor, which is related to the roughness of the cathode surface.

In conclusion we have observed two-photon-induced photoemission optogalvanic effect from a copper target which acts as one of the electrodes in the discharge cell using 532 nm pulsed radiation as the pump beam. The overall quantum efficiency in the POG has been found to increase with respect to laser power and electric field.

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