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Photoemission optogalvanic effect studies in N_2, NO_2 and Ar discharges under pulsed laser excitation

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Abstract. A two-photon induced photoemission optogalvanic effect which brings about a change in the discharge voltage when a pulsed dye laser beam is focused on a tungsten electrode has been described. The experiment is performed with N_2, NO_2 and Ar discharges. The magnitude of the signal voltage is studied as a function of laser energy and discharge current. The effective quantum efficiency in the discharge is found to be larger than that in the vacuum condition.

1. Introduction

The optogalvanic (OG) effect is the change in the electrical properties of a gas discharge caused by illuminating the discharge with radiation (Green *et al* 1976). The OG effect is a sensitive technique for studying the spectra of atoms and molecules in a discharge plasma. The energy absorbed by the species perturbs the plasma characteristics and produces a measurable impedance change in the discharge. OG effects can also be produced by injecting electrons into the discharge via photoelectric emission from the electrode surface by laser irradiation (Downey *et al* 1988). Photoelectrons emitted may excite or ionize the atoms or molecules on their way to the anode and produce more secondary electrons by collision. This non-resonant phenomenon is called the photoemission optogalvanic (POG) effect. The magnitude of the POG signal depends on the work function of the electrode material, nature of the discharge medium, discharge conditions and the intensity of the laser beam. The effect of the discharge current and the laser energy on the magnitude of the POG signal is studied in this paper.

2. Experimental set-up

The experimental set-up used to observe the POG signal (Sasi Kumar *et al* 1991) is shown in figure 1. Radiation from a pulsed dye laser (Quanta Ray PDL-2), pumped by the second harmonic of a Q-switched Nd:YAG laser (Quanta Ray DCR-11), was used as the excitation source. The discharge cell was made of glass, had an inner diameter of 1 cm and tungsten electrodes at an inter-electrode separation of ~1 cm. The discharge was produced by DC excitation in the cell with a ballast resistance in series with the tungsten electrodes. Continuous flow of the sample gases N_2, NO_2 and Ar was maintained in the cell at low pressure. The pulsed dye laser beam was focused on the cathode using a lens. The POG signal taken through a capacitor can be displayed on a CRO. The amplitude of the signal was measured for different laser energies at 564 nm and discharge currents.

3. Results and discussion

The variation of the POG signal amplitudes at 564 nm wavelength for N_2, NO_2 and Ar are studied as a function of laser energy for different discharge currents. The work function of the electrode material (tungsten) is 4.3 eV (Weast 1974–75) which approximately coincides with twice the photon energy at 564 nm. It has been found that the dependence of the observed signal amplitude S on the laser pulse energy I can be fitted into a quadratic expression

$$S = AI + BI^2 \qquad (1)$$

where A and B are constants which depend on the discharge conditions. Figure 2 shows the above curve fitted to experimental observations. The first term can be interpreted as a one-photon process which is due to the direct absorption of radiation by the discharge

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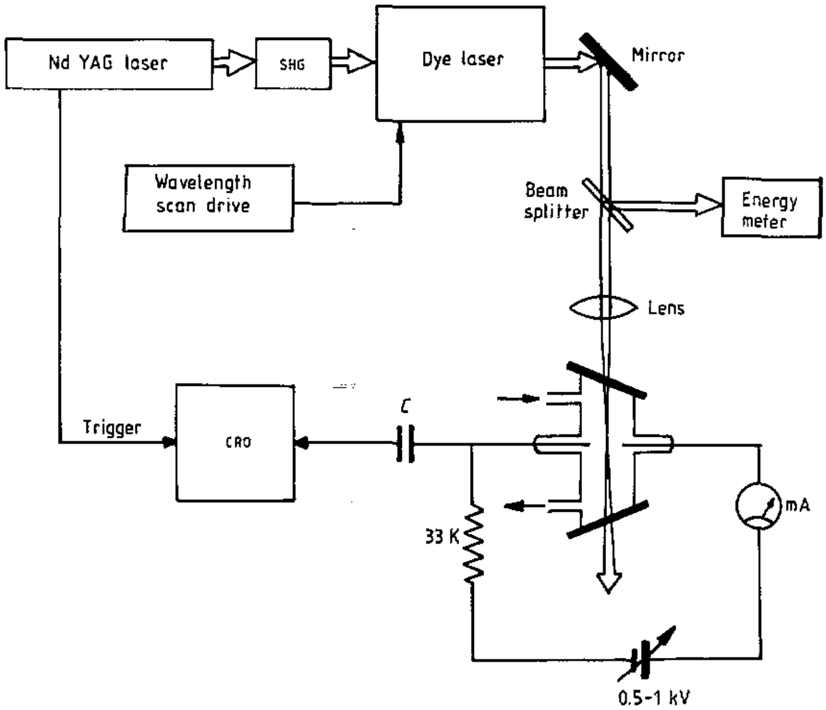

Figure 1. Block diagram of experimental set-up.

plasma (the OG effect) while the second term corresponds to the two-photon process due to photoemission from the electrode surface.

The magnitude of the POG signal in the case of the N_2 discharge is very much larger than that in NO_2 and Ar discharges. The first term in equation (1) results from the resonant absorption of laser radiation at 564 nm. It is noted that the N_2 molecule has absorption bands (Gaydon and Pearse 1965) in the spectral regions 700–500 nm (5,0 band of the first positive system) and at 640–500 nm (3,4 band of Gaydon's green system). One-photon-like signals can also arise due to thermionic emission from the target surface due to large temperature increases resulting from the laser irradiation. The second term arises as a result of the two-photon induced photoemission optogalvanic effect.

Figure 3 shows the variation of coefficients A and B as a function of discharge current. In the case of the N_2 discharge, coefficient A is initially negative and increases with current while for the discharge in Ar gas it decreases. For the NO_2 discharge the variation of A with the discharge current is nonlinear. The coefficient B for N_2 and NO_2 discharges decreases while for Ar it

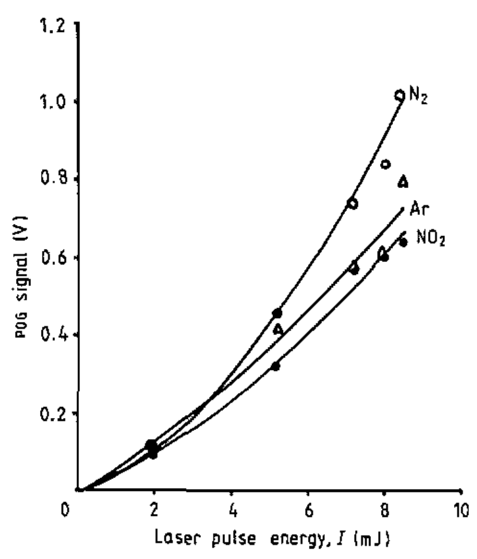

Figure 2. Curve for $S = AI + BI^2$ for N_2 at 15 mA ($A = 31.2$, $B = 10688$), Ar at 15.5 mA ($A = 57.8$, $B = 3423$) and NO_2 at 15.4 mA ($A = 43.43$, $B = 4144.5$). A is measured in VJ^{-1} and B in VJ^{-2}. The points correspond to experimental observations.

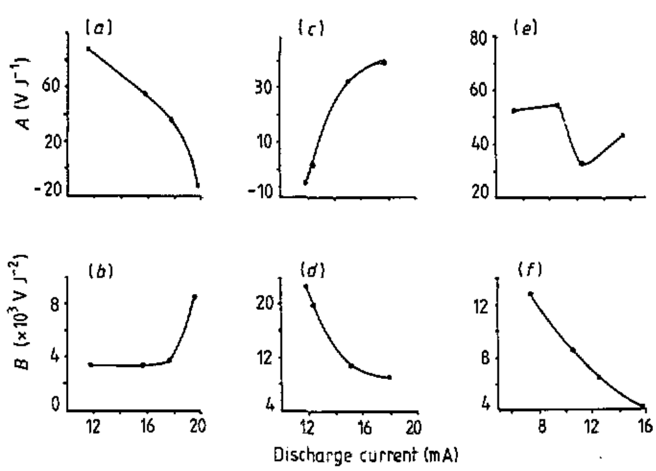

Figure 3. Dependence of A and B on discharge current for Ar ((*a*) and (*b*)), N_2 ((*c*) and (*d*)) and NO_2 ((*e*) and (*f*)).

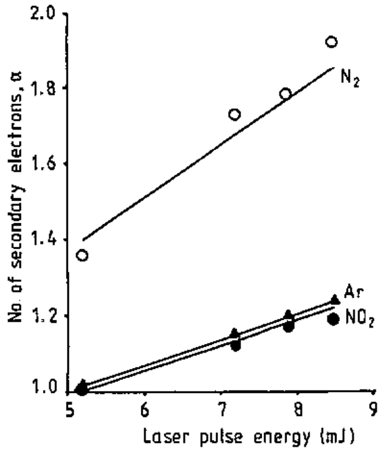

Figure 4. Number of secondary electrons produced per collision $\alpha = i(A + BI)$ for N_2 ($i = 17.8$ mA, $A = 39.6$, $B = 7450.27$), Ar ($i = 17.5$ mA, $A = 38.78$, $B = 3683.83$) and NO_2 ($i = 15.4$ mA, $A = 43.43$, $B = 4144.55$) and observed values.

increases with discharge current. This shows that at low currents the two-photon process in N_2 and NO_2 is more predominant than that in the Ar discharge.

4. Calculation of effective quantum efficiency

The quantum efficiency (photoelectrons emitted per photon) in the plasma is two to three times larger than that in the vacuum as a result of secondary electrons produced via collisions in the plasma. The effective quantum efficiency of the POG effect Φ_p in the gas discharge is

$$\Phi_p = \alpha\Phi \tag{2}$$

where Φ is the quantum efficiency in vacuum ($\sim 10^{-4}$) and α is the number of secondary electrons produced per collision and can be written as

$$\alpha = iS/I \tag{3}$$

where i is the discharge current, I the laser energy per pulse and S is given by equation (1). Then,

$$\alpha = i(A + BI). \tag{4}$$

Figure 4 shows the variation of α as a function of the laser pulse energy. α is found to be greater than one which means that effective quantum efficiency in the discharge is at least α times that in vacuum. However, for low laser pulse energy α is less than one—this is due to the predominance of recombination processes between electrons and positive ions in the discharge.

5. Conclusions

The change in the discharge characteristics as a result of the photoemission OG effect using tungsten electrodes in N_2, NO_2 and Ar discharge was studied. The effective quantum efficiency of the process in the discharge is found to be at least α (number of secondary electrons produced per collision) times that in vacuum.

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