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The photoemission optogalvanic effect in a Ne–Nd hollow cathode

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Abstract. Laser-induced photoelectric and photoemission optogalvanic effects in a Ne–Nd hollow cathode discharge have been studied using a continuous wave laser source. The potential barrier for photoinduced electron emission from the cathode decreases as the applied voltage is increased. Owing to secondary electron emission in the plasma, the photocurrent is greater than that without discharge. The multiplication of secondary electrons and the quantum efficiency are also investigated.

1. Introduction

In recent years there has been much interest in studying the photoelectric (PE) effect for numerous applications of photoelectrons emitted from solid surfaces. It is well known that, when radiation is allowed to fall on the surface of a solid maintained at a negative potential, a PE current will be generated if the photon energy of the incident radiation is greater than the work function of the cathode material. In this process the electrons are first optically excited into states of higher energy and then move to the surface of the solid with or without scattering and escape into vacuum. As compared with other methods, laser-driven PE emission can produce intense, short electron beams of very high current density. Generation of such an intense electron beam from U and Mg elements using a pulsed XeCl laser has been reported (Ivri and Levin 1993). Multiphoton photoelectron emission also takes place with such intense laser beams (Yen *et al* 1980).

Experiments on the PE effect are usually carried out in a high-vacuum environment. However, studying the PE effect in the presence of an electric discharge is of importance for understanding various characteristics of the discharge plasma. The total current in the discharge under laser irradiation will be the resultant of the original plasma current without irradiation and that caused by interaction of photoelectrons emitted from the cathode surface with the plasma medium. Emitted photoelectrons on their way from the cathode to anode will normally collide with atoms and more secondary electrons will be generated, so that an enhancement in current is observed. This phenomenon, known as the photoelectric emission optogalvanic (POG) effect, caused by single-photon absorption (Downey *et al* 1988) as well as two-photon absorption (Sasi Kumar *et al* 1991, 1993) has been observed under various discharge

conditions. The essential difference between OG and POG is that the former is due to resonant absorption of radiation by the species present in the discharge medium whereas the latter is a non-resonant process, which will occur at any wavelength provided that the energy of the incident radiation is greater than that of the effective work function of the cathode. Applications of the POG effect for plasma surface characterization (Downey *et al* 1988), real-time monitoring of metal or semiconductor surfaces (Selwyn *et al* 1988) and the diagnostics of electrodes in a discharge lamp (Schulman and Woodward 1989) have been reported. In this paper we have studied both PE and POG effects in a Ne–Nd hollow cathode in a stable region of the discharge using an Ar⁺ laser as excitation source. The voltage-dependence of these processes and the quantum efficiency in both cases were also investigated.

2. Experimental

Figure 1 shows the experimental set-up, in which a commercial Ne–Nd hollow cathode (Cathodean, UK), commonly employed as a light source in atomic absorption spectroscopy, is used as the discharge cell. It consists of a hollow cylindrical cathode and a ring-shaped anode with an inter-electrode separation of about 2 mm, which is filled with neon gas at about 10 Torr. The discharge is produced by applying a highly stable DC voltage from a power supply (Thorn EMI) through a ballast resistance (66 k Ω) and is operated in the negative glow regime showing the characteristic colour of the neon glow discharge. Radiation from a CW argon ion laser (Spectra Physics 171) with multilines, consisting of a mixture of 514, 496, 488 and 478 nm radiation, is mechanically chopped and allowed to fall onto the cathode. The laser beam

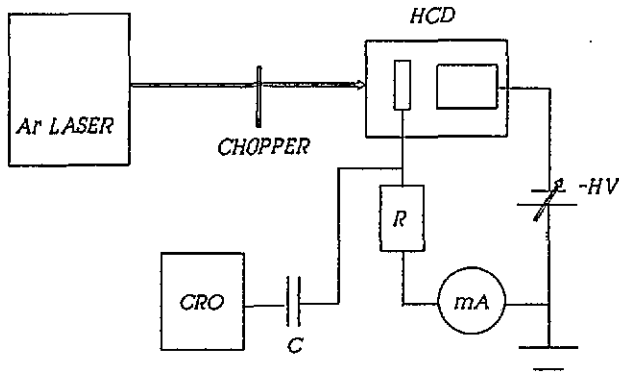


Figure 1. The experimental set-up ($R = 66 \text{ k}\Omega$, $C = 0.1 \mu\text{F}$).

has a TEM_{00} mode spatial distribution with a beam diameter of 1.58 mm. The capacitor ($0.1 \mu\text{F}$) blocks the DC voltage and allows detection of the AC signal arising from photoelectric emission. The strength of the photoelectric signal generated is measured as a function of discharge voltage and laser power using a digital storage oscilloscope (Iwatsu DS-8621). There is no resonant absorption by the discharge medium corresponding to these wavelengths and hence no OG signal is observed by direct absorption. Checking the response of the discharge, by passing the laser without striking the cathode it is confirmed that no OG effect is generated due to the plasma itself. All the measurements were carried out by taking the average of two signals. Since the power supply used was highly stable and ripple-free, it was possible to maintain a stable discharge so as to get a noise-free signal.

3. Results and discussion

3.1. Voltage-dependence of the photocurrent

The quantum yield of a photoelectric emitter depends upon three basic processes, namely photoexcitation, transport to the emitter surface and escape over the surface barrier, and is given by (Powell 1970)

$$Y(\hbar\omega - E) = C \int_0^{\hbar\omega - E} N_i(-E')(\hbar\omega - E - E') dE' \quad (1)$$

where $N(-E')$ represents the energy distribution of electrons and E is the barrier energy of the cathode. Y depends on both the frequency and the power of the radiation. According to (1), if the field is increased then the barrier height will decrease and hence the yield and strength of the photocurrent will increase. The photocurrent (I) and the quantum yield (Y) are related by

$$I = PY/(\hbar\omega) \quad (2)$$

where P is the absorbed light power and $\hbar\omega$ is the photon energy in electron-volts. Thus

$$I = A(\hbar\omega, P) \int_0^{\hbar\omega - E} N_i(-E')(\hbar\omega - E - E') dE' \quad (3)$$

where A depends on the photon energy $\hbar\omega$ and the laser power P . The mechanism responsible for the field-dependent barrier lowering depends on processes like the image-force lowering and the field penetration through the electrodes, given by Powell (1970) as

$$E = E(V) = E_0 - KV^{1/2} \quad (4)$$

where V is the applied voltage and K is the barrier-lowering constant in units of $\text{eV V}^{-1/2}$. This phenomenon of barrier-lowering is the same as the Schottky effect (Raizer 1991). Thus from equations (3) and (4) the voltage-dependence of the photocurrent can be written as

$$I = A \int_0^{\hbar\omega - E_0 + KV^{1/2}} N_i(-E')(\hbar\omega - E_0 + KV^{1/2} - E') dE' \quad (5)$$

The energy distribution of electrons can be expressed in the form

$$N_i(-E') \propto \exp(E'/k_B T) \quad (6)$$

where T is the temperature. Hence the functional form of the photocurrent is obtained from equations (5) and (6) as

$$I \propto A(k_B T)^2 \left[\exp\left(\frac{\hbar\omega - E_0 + KV^{1/2}}{k_B T}\right) + \left(\frac{\hbar\omega - E_0 + KV^{1/2}}{k_B T}\right) - 1 \right] \quad (7)$$

For large enough $(\hbar\omega - E_0 + KV^{1/2})/(k_B T)$ this expression reduces to

$$I = A(k_B T)^2 \exp\left(\frac{\hbar\omega - E_0 + KV^{1/2}}{k_B T}\right) \quad (8)$$

For emission from the Fermi tail in metals, as for degenerate semiconductors in which the electron energy distribution is an exponential function of the form of equation (6) and the photocurrent is approximately given by equation (8). According to this equation, the strength of the photocurrent depends on the photon energy, applied voltage, the work function of the cathode material and temperature. The increase in voltage will modify the surface work function according to equation (4) and will show an increase in photocurrent. The parameter K also depends on the dielectric constant of the medium and the inter-electrode separation (Powell 1970). On taking the logarithm of the above equation it can be seen that a plot of $\log I$ versus $V^{1/2}$ will be a straight line.

3.2. The photoelectric effect

The work function of the cathode (Nd) material is 2.95 eV (Gray 1972) and the energy of the laser radiation used corresponds to 2.6 eV per photon. The work function of a material depends on the state of the surface, namely its contamination, roughness, temperature and so on (Raizer 1991). Even at minimum voltage (100 V) a

PE signal is observed, even though there is a mismatch of 0.35 eV between the work function of the cathode material and the photon energy. The effective work function of the cathode material will be reduced in the presence of an electric field due to the combined effect of Schottky and field electron emission so that a lower photon energy threshold is sufficient to generate a signal. Surface-enhanced ionization, in which radiation interacts with positive ions of the metal surface, can also lower the work function sufficiently to allow efficient generation of photoelectrons by the photon flux (Naaman *et al* 1983). Before striking the discharge the PE signal increases (figure 2(a)) with voltage and it becomes maximum at a laser power of about 2 W, above which it begins to decrease (figure 2(b)). The signal generated is measured as a voltage change across a resistance ($R = 66 \text{ K}$). At higher laser powers, nonlinearities in laser–target interactions reduce the PE effect. The dependence on laser power (P) of the PE signal ($\Delta V_{PE} = IR$) can be fitted to the quadratic equation

$$\Delta V_{PE} = aP - bP^2. \quad (9)$$

The coefficients a and b depend on the discharge voltage and also the temperature near the irradiated region on the cathode. It is observed that a and b increase with voltage. From the slope of the $\log I$ versus $V^{1/2}$ plot (figure 2(c)), the value of the barrier-lowering constant K evaluated is $0.019 \text{ eV V}^{-1/2}$. In these calculations we have assumed an average temperature of 300 K. The value of K obtained from exponential fitting of the experimental values was 0.018, which is in close agreement with the former. This indicates that $E(V) < E_0$, so that the voltage applied between the electrodes reduces the barrier energy of the cathode surface according to equation (4).

3.3. The photoelectric emission optogalvanic effect

The magnitude of the POG signal strongly depends on the escape depth of the photoelectrons through the surface of the cathode and also on the plasma characteristics. The dependence of the signal on the discharge voltage and laser power are shown in figure 3. Variation of the POG signal with voltage strongly depends on the behaviour of the discharge. It is observed that, at higher voltages (about 149 V), the signal increases considerably. In this voltage region we have noticed, in the absence of laser irradiation, the existence of quasi-periodic oscillations in the plasma current, indicating a change in discharge characteristics. Another noticeable observation is that, after striking the discharge, if the tube current is reduced, then the POG signal is found to increase, reaching a maximum value at the point at which the discharge is turned off (figure 4). The electrons emitted from the cathode on their way to the anode are multiplied, producing a large number of secondary electrons. Thus, if the multiplication factor given by

$$\alpha(V) = \Delta V_{POG} / \Delta V_{PE} \quad (10)$$

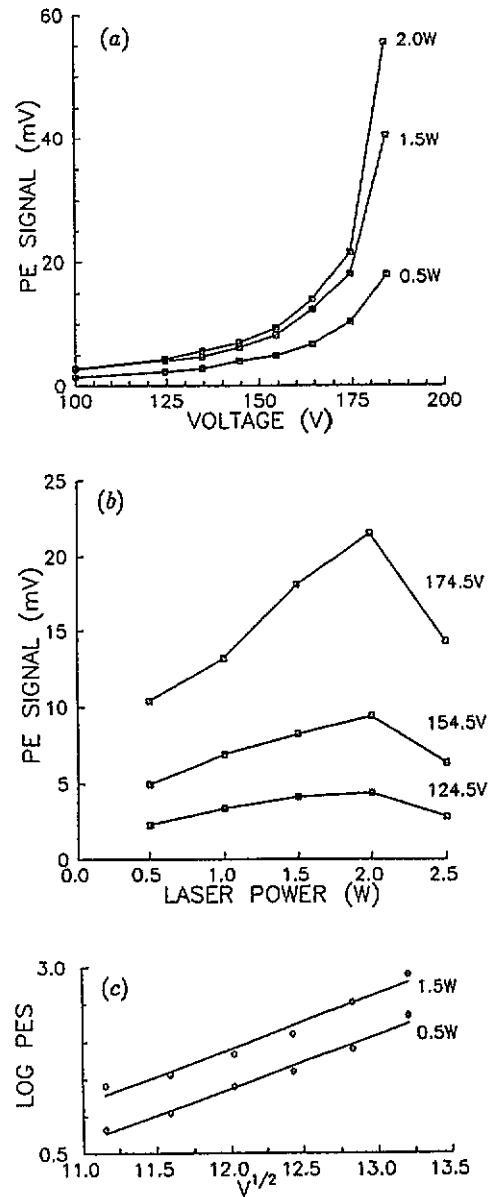


Figure 2. (a) The voltage-dependence of the photoelectric effect. (b) The laser-power-dependence of the photoelectric effect. (c) A $\log I$ versus $V^{1/2}$ plot.

is greater than unity, then the current increases and the POG signal (ΔV_{POG}) will be greater than the PE signal (ΔV_{PE}). The dependences of the multiplication factor on laser power and voltage are shown in figure 5. In the presence of a discharge under certain conditions, the $\log I$ versus $V^{1/2}$ plot (figure 3(c)) is a straight line. In the presence of a discharge, the barrier-lowering constant depends on both the applied field and the cathode sheath in the discharge. Electron multiplication in the discharge also affects the value of K . In this case, the photocurrent can be obtained from equation (8) by replacing the barrier lowering constant by K_d , where

$$K_d = \alpha K. \quad (11)$$

As the laser power is increased, the value of K_d increases, showing an intensity-dependence.

The effective quantum efficiency (Y_e), which is the ratio between the number of photoelectrons (N_e) and the

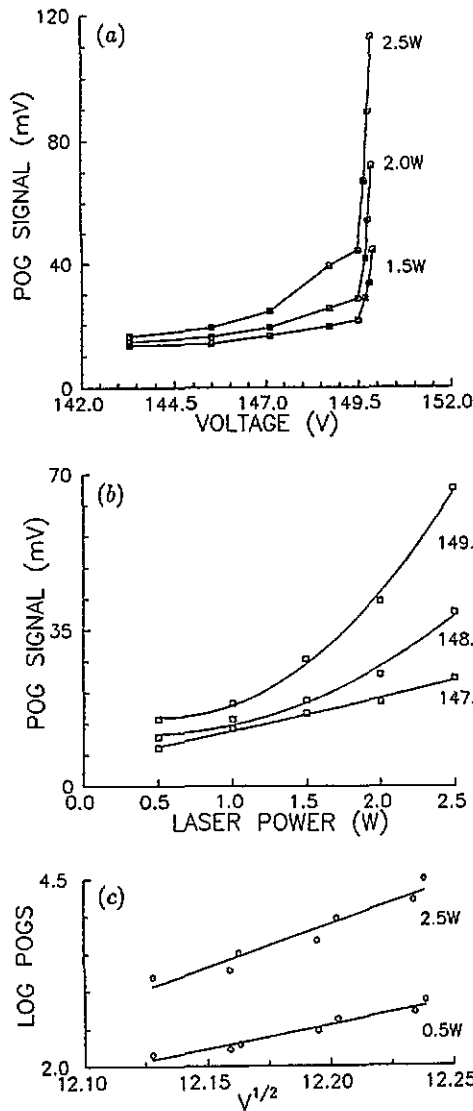


Figure 3. (a) The voltage-dependence of the photoelectric emission optogalvanic signal. (b) Its laser-power-dependence. (c) A plot of $\log I$ of the photoelectric emission optogalvanic signal versus $V^{1/2}$ in the presence of a discharge.

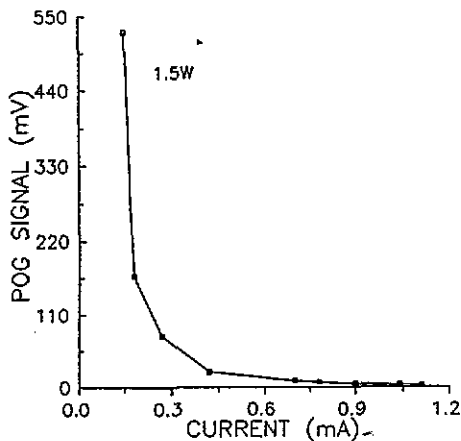


Figure 4. Variation of the photoelectric emission optogalvanic signal with current below the striking voltage in the presence of a discharge.

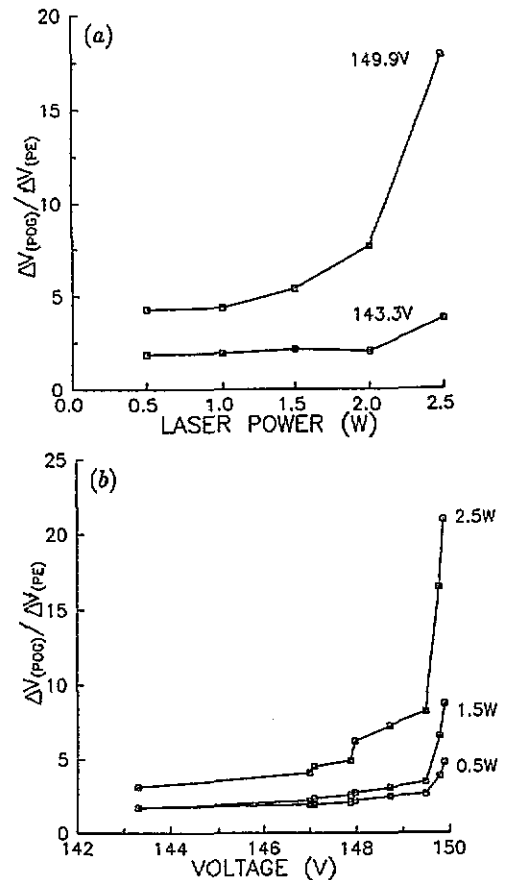


Figure 5. The dependence of the multiplication factor on (a) laser power and (b) voltage.

number of absorbed photons (N_{ph}), can be written as

$$Y_e = N_e/N_{ph} \propto I\hbar\omega/P. \quad (12)$$

In general the quantum efficiency (QE) in the plasma is larger than that in the vacuum (Downey *et al* 1988) due to the collisionally produced secondary electrons resulting from ionizing collisions with neon atoms on their way from cathode to anode. The calculated value of Y_e is in the range 10^{-6} – 10^{-7} electrons per photon, depending on the experimental conditions. These values are comparable to those obtained by Ivri and Levin (1993) using pulsed laser excitation. In the presence of a discharge, if the electron energy distribution is the same for both conditions, then it is given by the product of Y_e and the electron multiplication factor (α).

4. Conclusions

In this work we have studied the voltage-dependence of the photocurrent in a Ne-Nd hollow cathode under laser excitation. The potential barrier of the cathode material is found to decrease according to equation (4). Assuming an exponential function for the electron distribution, the functional form of the photocurrent is also given. In the presence of a discharge the photocurrent is greater than that without discharge, due to secondary electron generation in the plasma. Depending on the experimental conditions, the calculated quantum efficiency of the processes was in the range 10^{-6} – 10^{-7} photoelectrons per photon.

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