Chapter IV

POG STUDIES WITH COPPER AS TARGET ELECTRODE

Introduction

Photoemission Optogalvanic (POG) studies with copper as target electrode is included in this chapter. The fundamental (1064 nm) and frequency doubled (532 nm) radiations from Nd:YAG laser have been used in the present studies to generate POG effect.

Only limited studies on photoelectric emission with copper had been carried out by earlier workers. Honig et al [1] observed laser induced emission of electron, ion and neutral atoms from copper. But the emission of electrons and ions were attributed only to thermal excitation. Berglund et al [2] studied photoemission from copper both theoretically and experimentally. They have shown that measurements of the spectral distribution of the photoelectric yield and of the energy distribution of photoemitted electrons at individual photon energies can be used to study both the optical excitation and the electron scattering processes in solids. At first photoelectrons are to be optically excited into states of higher energy; then they move to the surface of the solid with or without scattering and get escape to the
Fig. 4.1. Variation of discharge current with respect to applied voltage. (a) - 180 μbar, (b) - 190 μbar, (c) - 200 μbar, (d) - 210 μbar, (e) - 400 μbar.
outside of the metal. They also obtained theoretical expressions for the quantum yield and for the energy distribution of photoelectrons.

Tsnang et al [3] studied surface plasmon field enhanced multiphoton photoelectric emission from copper metal film. It is observed that electron yield increases by several orders of magnitude with fairly high quantum efficiency, when photons are coupled to the surface plasmon modes. Chen et al [4] also observed photoemission from copper films. Srinivasa Rao et al [5] analyzed the influence of electric field associated with the photons in enhancing photoemission on diamond turned copper with laser beam. Also they observed that at the optimum incident angle, the electron yield with p-polarized light was 75 times larger than that with s-polarized light.

In the present study we use copper target as a source for generating electron beams for POG effect.

4.1. Two-photon induced photoemission optogalvanic effect with copper as target electrode.

POG studies using copper as target electrode has been studied under two conditions (a) keeping the target electrode as cathode (forward bias condition) and (b)
Fig. 4.2. Variation of discharge current with respect to discharge gas pressure. (a) - 700 V, (b) - 800 V, (c) - 900 V
keeping the target electrode as anode (reverse bias condition).

Initially the dependence of discharge plasma current on applied voltage and discharge gas pressure has been characterized. For different pressures, the dependence of discharge plasma current on applied voltage is noted, (fig.4.1.). The discharge plasma current increases with the applied voltage. But, above a threshold value of the gas pressure discharge plasma current diminishes. Fig.4.2. shows the dependence of discharge plasma current on pressure. Plasma current increases to a threshold value of pressure, and then decreases. Around a pressure of 200 μbar to 210 μbar there is a sharp increase in the discharge current. At this pressure as the applied voltage is increased beyond a threshold an abrupt increase in the plasma current (plasma breakdown) is noted.

(a) keeping the target electrode as Cathode (forward biased condition)

A solid copper target of 2 mm thickness was sandwiched to one of the caps of the discharge cell. Nitrogen gas was continuously flown through a needle valve and an optimum pressure of 180 μbar was maintained in the discharge cell. The outlet of the discharge cell was connected to a rotary
Fig. 4.3. A typical POG signal in the forward biased condition
vacuum pump. Discharge was excited using a low noise high voltage power supply. A negative polarity was given to the target (forward biased condition) electrode. Frequency doubled pulsed laser radiations (532 nm) were normally irradiated on the target electrode. POG signal amplitude was directly measured from the storage oscilloscope. A typical POG signal shape (forward biased case) is given in fig. 4.3. The signal shows a FWHM of a few micro seconds which is much larger than the laser pulse duration ( ~ 10 ns). This difference occurs because, the POG signal arises due to the transportation of injected photoelectrons into the discharge. The propagation of electron avalanche in discharge result into POG signal, which sustains longer than the laser pulse duration. The discharge plasma will be left with strongly perturbed ion and electron densities, which afterwards relax into their respective steady states.

The work function of copper is 4.4 eV [6] which is slightly lower than the two photon energy (4.6 eV) at 532 nm so that we can expect a two photon induced photoemission from copper target.

According to generalized Fowler-Dubridge theory [7,8,9] the total electron density emitted from copper target under laser irradiation is
Fig. 4.4. Dependence of POG signal strength on laser power in the forward biased condition (a) - 600 V, (b) - 700 V, (c) - 800 V
\[ J(r,t) = \sum_{n=0}^{\infty} J_n(r,t) \quad (4.1) \]

where
\[ J_n(r,t) = a_n \left[ \frac{e}{\hbar \nu} \right] A(1-R)^n P(r,t)^n \quad (4.2) \]

In this experiment one can expect a two-photon induced photoemission so that the contribution to POG signal will be due to
\[ J_2(r,t) = a_2 \left[ \frac{e}{\hbar \nu} \right] A(1-R)^2 P(r,t)^2 \quad (4.3) \]

From (4.3) it is clear that the slope of the log - log plot of POG signal against laser intensity gives the number of photons taking place in the multiphoton process. It is two in the present case.

The variation of POG signal as a function of laser intensity for different discharge voltages (keeping the target as cathode) at 180 \( \mu \)bar of nitrogen gas pressure is shown in fig. 4.4. The signal strength increases with the
Fig. 4.5. Log-log plot of POG signal versus laser power in the forward biased condition. (a) - 600 V, (b) - 700 V, (c) - 800 V.
laser intensity as well as with the applied voltage. At lower laser intensity, the POG signal is almost the same for different values of applied voltage. But, at higher laser power, POG signal increases with the applied voltage. Electric field at metal surfaces reduces the image and ion fields and thereby cause reduction of the work function [10], which will result into an enhancement in the POG signal as observed in the present studies. But, at higher laser power much larger number of photoelectrons will be generated such as to surpass the recombination rate and hence an increase in the POG signal may be expected. In fig 4.5, log-log plot of signal strength against laser power for three different applied voltages are given. The three plots are parallel and have a slope of 2, which clearly confirms the two-photon process taking place in POG effect here, for different applied voltages.

As we go on increasing the laser intensity, a threshold intensity was observed above which, a complete breakdown (an abrupt enhancement in the discharge current) of the discharge plasma is observed. At this condition, the discharge plasma became so turbulent that the discharge current shoots up to five to ten times the normal discharge plasma current. Now, the POG signal shape becomes very different indicating the vigorous processes of transportation of the perturbed electrons and ions in the
Fig. 4.6. POG signal trace just before breakdown [700 V, 0.66 mA, 180 μbar, 0.85 W laser power]

Fig. 4.7. Trace of POG signal at the set off of plasma breakdown. [700 V, 6.45 mA, 180 μbar, 1.05 W laser power]
Fig. 4.8. POG signal after breakdown, [700 V, 6.44 mA, 1.05 W laser power]

Fig. 4.9. Discharge plasma oscillations after breakdown (6.42 mA)
Fig. 4.10. Spectra of discharge plasma before breakdown
Fig. 4.11. Spectra of discharge plasma after breakdown
Fig. 4.12. Spectra of plasma at the breakdown discharge current by increasing the applied voltage
plasma medium. Figures 4.6. and 4.7. give the trace of the POG signal shape at the onset of the breakdown. Fig. 4.8. shows a typical POG signal just at discharge breakdown. After the break down a typical discharge plasma nature is depicted in fig. 4.9.

At breakdown and before breakdown discharge plasma spectra were recorded and the difference in plasma spectral lines were analyzed. Figures 4.10. and 4.11. show the plasma spectra before and after breakdown. The spectra reveal well resolved bands corresponding to C-X transition of $N_2$ molecule present in the discharge. Before breakdown, the spectrum shows a few bands of nitrogen molecule, while at breakdown more number of lines with greater intensity are seen. The same discharge current at breakdown was also achieved by increasing the discharge voltage. However the corresponding spectrum of the plasma (fig. 4.12.) differs with the one recorded at low voltage breakdown concentration. The (0,1) band is predominant at low voltage breakdown condition. While (0,3) is predominant at high voltage (i.e. without breakdown) condition. The intensity distribution among the bands are almost same in the absence of breakdown at low and high voltage cases. However at breakdown the intensity distribution of the spectrum gets modified.
4.13. Dependence of threshold laser power for plasma breakdown on applied voltage
Fig. 4.14. POG signal trace at 290 µbar discharge gas pressure (700 V, 4.98 mA, 0.85 W).

Fig. 4.15. Trace of POG signal at a discharge gas pressure of 450 µbar (700 V, 4.55 mA, 0.85 W)
It is observed that, once the breakdown was initiated the discharge plasma turbulence sustains for a long time without resuming the original state of the plasma and the discharge current also remains enhanced.

As the applied voltage is increased the threshold laser intensity for which breakdown initiates decreases. Fig.4.13. shows the dependence of threshold breakdown laser intensity on the applied voltage.

The dependence of POG signal on the pressure in the discharge cell was also looked into. There exists an optimum pressure below which we get sharp POG signals (fig.4.3). As the pressure is increased above the optimum pressure a second peak is found to grow in strength and width with a delay of a few micro seconds with respect to the first peak (figs. 4.14, 4.15, and 4.16). As the pressure is increased, the population density of ions and electrons in the plasma medium will also increase. Hence the triggered interaction of the bunch of photoelectrons with the plasma medium will sustain for a longer duration through collisions and secondary electron emissions, leaving more perturbed ions and electrons. At higher laser intensity the second peak is found to increase in strength and width due to an enhancement in the above process. (fig.4.16). As the laser intensity is further increased ripple like structures are
Fig. 4.16. POG signal trace at 500 μbar of discharge gas pressure (700 V, 4.07 mA, 0.85 W).

Fig. 4.17. Ripple like structures in POG signal at large pressure (500 μbar) and high laser power (1.25 W).
Fig. 4.18 POG instability generated at larger pressures (780 bar, 1.25 W)

Fig. 4.19. A typical laser induced instability in discharge plasma.
observed in the second peak (fig. 4.17.). This can be attributed to the onset of discharge oscillations arising out of the abundance of the perturbed ions and electrons in the discharge plasma. At this condition an increase in pressure causes instability in the POG signal (fig. 4.18) and a strong nonlinear effect sets in. Similar instabilities (fig. 4.19) are observed for higher laser intensities also.

A ringing effect on POG signal (fig. 4.20) is observed in the present studies. A similar ringing effect was observed by Mitchell et al. [11] while studying the POG effect by using steel and Si electrodes. As a result of photoemission, the ion sheath near the cathode contracts slightly, because of the increase in the local charge density increase, as the avalanche developed. As the cathode sheath contracts, bulk electrons diffuse behind the moving sheath edge. But near the anode, the plasma potential first decreases below the anode potential, to allow excess negative charges to escape. This results in a rapid electron flux to the anode. However, too much charge is extracted and the plasma potential subsequently increases above the initial value to constrain further electron loss. This overshoot in the plasma may be partially responsible for the observed ringing.

We have observed that the amplitude of ringing gets
Fig. 4.20. Ringing of POG signal

Fig. 4.21. Ringing amplitude increases with discharge current
enhanced on increasing the discharge current (fig.21.). At higher current, the nonlinearity in the discharge plasma causes complex plasma oscillations to set off. Details of such discharge plasma instabilities are given in chapter VII.

(b) keeping the target electrode as anode ( reverse biased condition )

The polarity of biasing of the target electrodes are reversed from the earlier observations. Except this all other experimental conditions are kept the same as for the forward biased condition. Now, the copper target was given a positive polarity ( reverse biased condition ). In the absence of discharge plasma, signal was not obtained in the case of reverse biased condition, contrary to a sharp signal observed in the forward bias case (fig.22.). In forward biased condition as the pulse repetition rate is increased the signal becomes very prominent and for high laser intensity the width of the signal increases (fig.4.23.) showing the thermal contribution in electron emission.

In presence of discharge, electrons in the discharge plasma will be moved to the target electrode ( which is reverse biased ), while the positive ions will be clouded at
Fig. 4.22. Typical photoemission signal

Fig. 4.23. Thermal contribution to photoemission signal at high laser intensity.
Fig. 4.24. POG signal trace giving three peaks under reverse biased condition.

Fig. 4.25. POG signal trace shows two peaks at larger biasing voltages.
the opposite electrode, the cathode. The trace of POG signal in the reverse biased condition is shown in fig. 4.24. In the forward biased condition the signal trace has a prominent single peak only, while in the reverse biased condition the trace has three peaks: one almost same as that of the prominent peak observed in the forward biased condition and another peaks with higher amplitude with a delay of few micro seconds adjacent to the prominent peak. However, as the applied voltage is increased two peaks were observed (fig. 4.25). A satisfactory model was proposed by Mitchell et al [12] and Debontride et al [13] for forward biased case. However, in the reverse biased case this model is inadequate due to the presence of multiple peaks in the signal. Peaks with larger FWHM and delay indicate that in the reverse biased case the perturbed ions and electrons left in the discharge plasma get transported for a longer time and the perturbation prolongs. In the reverse biased condition, the presence of space charge effect due to electron clouds in the vicinity of the target electrode and positive ions present in the discharge plasma will generate ambipolar diffusion. The multiple peak structure can be attributed to the signal due to such ambipolar diffusion.

Fig. 4.26 shows the variation of POG signal as a function of laser intensity at various reverse bias voltages across the discharge cell. When we change the polarity of the
Fig. 4.26. Dependence of POG signal on laser power (reverse biased condition) (a) - 600 V, (b) - 700 V, (c) - 800 V
Fig. 4.27. Log-log plot of FOG signal versus laser power. (reverse biased case) (a) - 600 V, (b) - 700 V, (c) - 800 V
electrodes, the direction of the drift of electrons and positive ions in the discharge plasma will also change with respect to the laser beam. Log-log plot of signal strength versus laser intensity again gives a slope of ~2 indicating that the two-photon process taking place in reverse bias case also (fig. 4.27.). In fig. 4.28. dependence of forward and reverse biased signal strengths on laser intensity is given. From fig. 4.28. it can be observed that in the reverse biased condition, POG signal shows a saturation at higher laser intensity. But, within that range of laser intensity, no such saturation is observed in the forward biased case. In lower laser intensity level, initial values of POG signals are almost the same for both reverse bias and forward bias voltages. But, at still higher laser power, at a given discharge voltage, signal strength in the case of reverse bias is found to be larger than that in the case of forward bias. Above a certain laser power, signal strength saturates in the reverse bias, unlike in the forward biased condition. In this region, the signal strength in the forward biased case becomes larger than that in the case of reverse biased condition. But, the point of cross over at which signal strength in the case of forward bias over that of reverse bias, gets shifted to lower laser power region at higher voltages. The observation shows that under reverse bias case, signal gets saturated due to space charge effect, while in forward bias case such space charge effect may be
Fig. 4.28. Dependence of POG signal on laser power under both forward and reverse biased conditions

Forward biased case: (a) - 600 V, (b) - 700 V, (c) - 800 V,
Reverse biased case: (d) - 600 V, (e) - 700 V, (f) - 800 V
negligible.

4.2. Study of quantum efficiency of POG effect both under forward biased and reverse biased conditions.

The quantum efficiency (Q) of photoemission is a measure of the emission of photoelectrons per irradiance of photons. It is defined as [11]

\[ Q = \frac{N_e}{N_{ph}} \]  (4.4)

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

\[ N_e = \frac{It}{e} \]  (4.5)

where \( I \) is the electron current and \( t \) is the current pulse duration.

\[ N_{ph} = \frac{\text{Laser Pulse Energy}}{h\nu} \]  (4.6)

Fig.4.29. shows the variation of overall quantum
Fig. 4.29. Variation of overall quantum efficiency of POG effect with laser power under forward biased condition. (a) - 600 V, (b) - 700 V, (c) - 800 V.
efficiency $Q$ as a function of laser intensity and applied voltage in the forward biased condition. The quantum efficiency is 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 intensity is increased enhancement in the value of $Q$ is noticed. This apparently is an indication of the probable role played by thermionic electron released by rapid heating of the target surface by the intense laser pulses. An increase in the applied voltage also enhances the electron density (due to collisional ionization) and thus overall quantum efficiency in POG effect increases with field. Also enhancement in the value of $Q$ due to the applied voltage across the cell can be accounted for, as the dependence of photoelectric efficiency $\eta$ on electric field $E$ [11] given by

$$\eta = A \left[ 2h\nu - \phi - \frac{eE\beta}{4\pi\varepsilon_0} \right]$$  \hspace{1cm} (4.7)

where $A$ depends on the cathode material parameter, $\phi$ is the work function and $\beta$ is the enhancement factor, which is related to the roughness of the cathode surface.

Fig.4.30. shows the variation of the overall quantum efficiency $Q$ as a function of laser intensity and the applied field in the reverse biased condition. For
Fig. 4.30. Variation of overall quantum efficiency under reverse biased condition. (a) - 600 V, (b) - 700 V, (c) - 800 V.
forward biased condition (fig. 4.29.) one can see that $Q$ shows an enhancement with laser intensity and applied voltage and a tendency of saturation at higher laser intensity, whereas in the reverse biased condition $Q$ tends to decrease above an optimum laser power. This decrease in $Q$ may be attributed to the saturation of POG signal in the reverse biased case. Above the saturation level, $N_e$ does not increase even though $N_{ph}$ increases, resulting into a drop in the value of $Q$. The existence of this optimum laser intensity is more predominant at higher discharge voltage. Occurrence of saturation effect makes it possible to write an empirical relationship between signal strength ($S$) and laser intensity ($I$) in the reverse bias as

$$S = \frac{aI^2}{1 + I/I_s} \quad (4.8)$$

where $I_s$ is the saturation laser intensity.

At lower laser intensity $I < I_s$, one can approximate $S$ as

$$S = aI^2 - bI^3 \quad (4.9)$$

where $a/I_s = b$.

In general $a$ and $b$ coefficients depend on the discharge voltage. The second term in equation (4.9) causes saturation
effect in the POG signals in the reverse bias case as observed in fig.4.6.

The plot of Q versus laser intensity for various voltages show a quadratic behavior. From equation (4.8) and (4.9) one can write

\[ Q = K_1 I - K_2 I^2 \quad (4.10) \]

Above equation describes the dependence of Q on I empirically, indicating that the quantum efficiency in the reverse bias case is a nonlinear function of laser intensity. The presence of space charge effect which opposes the generation of secondary electrons lead to this nonlinearity. Equation (4.10) shows that Q will have an optimum value

\[ Q_{opt} = \frac{K_1^2}{4K_2^2} \quad (4.11) \]

with the corresponding laser intensity (critical)

\[ I_c = \frac{K_1}{2K_2} \quad (4.12) \]

Equation (4.11) and (4.12) show that the heights of the
Fig. 4.31. POG signal amplitude versus laser energy using 1064 nm laser pulses: (a) - 800 V, (b) - 900 V, (c) - 1000 V.
humps in fig. 4.8. depend on discharge voltages implicitly through $K_1$ and $K_2$.

4.3. POG effect with copper target electrode using 1064 nm pulsed laser radiation.

Irradiation of the target with 1064 nm radiation also gives POG signals. In 1064 nm radiation induced photoemission also we observe an enhancement in POG signal with laser intensity and discharge voltage as shown in fig. 4.31. Here, we expect a four photon process, similar to the two photon induced POG signal from copper using 532 nm radiation, since the energy corresponding to 1064 nm radiation is only 1.16 eV.

According to the generalized Fowler-Dubridge theory, the component

$$J_4 = a_4 \left[ \frac{e}{h \nu} \right] A(1-R)^4 P(r,t)^4$$

$$T(r,t)^2 \frac{4h \nu - \phi}{kT(r,t)}$$

(4.13)

has to contribute the photoemission current and obviously, the log-log plot of signal strength versus laser intensity should give a slope of 4. But, the log-log plot of signal strength versus laser intensity gives a slope of ~2 only
Fig. 4.32. Log signal amplitude versus log laser power (a) - 800 V, (b) - 900 V, (c) - 1000 V.
That is we observe a near quadratic dependence of signal strength as

\[ S = bI^2 \] (4.14)

That is the photoemission from copper using 1064 nm radiation is also mostly due to two photon process similar to the observation with 532 nm.

Yen et al [12] showed that the current density for an n-photon photoemission can be written as

\[ J_n = K[(1-R)I_0]^n \] (4.15)

where R is the reflectivity, \( I_0 \) is the incident laser intensity and K is a constant. When infrared radiations are used as pump beam, thermally assisted photoemission process is also possible.

The mismatch of workfunction and photon energy using 1064 nm laser radiation implies that the resulting process is a thermally assisted two-photon POG effect. Thermionic process due to heating of sample by laser pulse does indeed cause electron emission from the target complimenting the multiphoton excitation eventually constituting the POG signal [13]. Heating of the target by intense laser pulses
Fig. 4.33. Schematic representation of (a) two photon emission with 532 nm radiation and (b) thermally assisted multiphoton emission with 1064 nm laser radiation.
produce electrons with energies above the Fermi level, so that emission from the tail of Fermi distribution can take place (fig. 4.33.), with the absorption of fewer than four photons. This masks the fine details of the photoelectric effect. Thermionic current is entirely governed by the temperature of the cathode surface, which depends on the absorbed laser power. Heating of the copper cathode surface disturbs the equilibrium between electron and the lattice and this should effect strongly the thermionic emission because of the low specific heat of the degenerate electron gas. The laser power dependence of POG effect therefore deviates from a simple power law.
References


