Photoelectron emission and its instrumental effect on optogalvanic measurements in a hollow cathode discharge

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The photoelectron emission (PE) from the cathode surface of a hollow cathode discharge (HCD) with a sub-breakdown bias applied, and hence no discharge present, was measured within the framework of an optogalvanic (OG) experimental arrangement. The work function dependence on the applied sub-breakdown voltage was investigated. The PE component in a real OG measurement was found to manifest itself as an instrumental effect together with nonresonant ionization which we call here space ionization (SP). The convolution of these components was determined experimentally as an instrumental function. A deconvolution procedure to determine the actual OG signal was developed.

Keywords: hollow cathode discharge, photoelectron emission, work function, optogalvanic effect, instrumental function.

1. Introduction

Penning was the first to observe a light-induced change of gas discharge conductivity [1]. Any change of gas discharge conductivity due to resonant light absorption at an optical transition is known as an optogalvanic (OG) modulation or simply as an optogalvanic effect. The OG effect turns out to be more easily measured and of better accuracy than the measurement of the absorbed light fraction alone. The development and application of OG spectroscopy is based on this effect and benefits from these inherent advantages [2]. The hollow cathode discharge (HCD) is the preferred medium in OG spectroscopy for a number of reasons. Firstly, it exhibits quite a low level of conductivity fluctuations or so-called galvanic noise. In addition, due to the commensurability of the large contact surfaces: negative glow (NG) – cathode dark space (CDS) – cathode surface, a HCD is known to be a reservoir of sputtered atoms mostly residing in the ground state. Finally, the characteristic electron energy distribution function (EEDF), contains three groups of electrons, including one group with an energy of a few hundred eV. Thus, the HCD has contains energetic electrons which can participate in collisional processes with sputtered atoms/ions and buffer gas atoms. As a result, the HCD extends application of OG spectroscopy to the states of sputtered atoms/ions [3] and high lying states of buffer gases [4].

The above irrevocable HCD properties, however, also introduce some specific complications in the OG signal from HCDs [5] including the important contribution of photon or \( \gamma \)-processes to the OG signal [4,6]. In general, instrumental effects are a basic aspect of any measuring technique and must be quantified and removed from a measurement where they represent a significant interference with the true value of the measured signal. The transfer function between input light stimulus and the output galvanic signal has been only partially discussed within the context of OG spectroscopy. Some instrumental effects in time-resolved [7] and amplitude [5] OG spectroscopy, based on HCD, have been already reported.

In this study, two other light-induced instrumental effects in HCDs are discussed, i.e., photoelectron emission (PE) and nonresonant space ionization (SP). The PE component arises from the laser light-HC surface interaction.

Apart from this PE contribution, the laser light induces one further nonresonant galvanic process, i.e., space ionization (SP). The latter refers to ionization from atoms residing in excited states close enough to the ionization threshold to be easily ionized. Both these effects contribute to the measured OG signal.

2. Experiments

Time-resolved measurements of the modulation in the voltage across both the HC and the HCD, i.e., HCD with no discharge and discharge present, respectively, were performed. Figure 1 shows the experimental set-up used to observe time-resolved PE and OG signals. The trade marked HCD lamp Ne/Li (“Narva”), was used. The discharge was produced by applying a highly stable DC voltage and was operated in the negative glow (NG) regime. A standard experimental scheme for OG detection was employed.
Time-resolved PE and measured OG (MOG) signals were induced by irradiating the HC (HCD) with a commercial Surelite™ OPO system pumped by Nd-YAG laser operated at the third harmonic (355 nm). The system delivered pulses of duration 4 ns FWHM at a repetition rate of 10 Hz, tuneable in the 420–600 nm wavelength range. The OPO output had a bandwidth of 0.03 nm and an average output power 25 mW. Optogalvanic signals were detected across the resistor $R_m = 20 \, \text{k}\Omega$ and recorded with a digital oscilloscope (Tektronix™ TDS3032B). The RC constant was chosen low enough to allow one to resolve the time-dependent signal.

3. Results and discussion

Three types of light-induced time-resolved galvanic responses were measured, i.e., the photoelectron emission from the cathode surface in the absence of a discharge but with an applied bias, the conductivity change in the presence of a discharge (HCD) due to resonant irradiation and the conductivity change due to nonresonant irradiation of the HCD.

3.1. Photoemission signals

Figure 2 shows the light-induced PE time-resolved response $\Delta U_{PE}$ of the cathode surface to the absorbed photons at various subthreshold (pre-breakdown) voltages $0 < U < U_{thr}$ across the HCD lamp (i.e., no discharge present). The PE maximum $\Delta U_{PE,max}$ occurs at $t_{th}$ up to 1 µs depending on the voltage applied. The curves in Fig. 3 illustrate the relation $\Delta U_{PE,max} < U < U_{thr}$ for irradiation of the HC at three different laser wavelengths. There is no resonant absorption by the discharge medium corresponding to these wavelengths ($\lambda = 477$, 546, and 562 nm) and hence no OG signal exists, i.e., the observed signal is due to PE only. Increasing the applied bias and hence the electric field simply increases the photoemission yield.

The signals in Figs. 2 and 3 are effectively manifestations of the characteristic work functions (WF) of the cathode material in Ne/Li HCD lamps (in the presence of the laser light pulse). Spectral measurements of the spontaneous light emission showed that the cathode contains technological alloys and hence comments on the actual WF of a pure Li surface are not appropriate here. Ultimately, the data $\Delta U_{PE}$ measured at values $U < U_{thr}$ characterize the PE properties of the cathode surface material only but not the PE of the cathode surface in the presence of a discharge (HCD mode). Therefore the data of Figs. 2 and 3 serve to illustrate the fact only that laser light used in the experiment generates PE as a possible background superimposed on the actual OG signal.

3.2. Time-resolved OG signals. Instrumental aspects

The voltage values $U > U_{thr}$ change the initial conditions for the PE process discontinuously, i.e., the hollow cathode discharge strikes and the voltage applied becomes largely confined to a thin layer of 1–2 mm near to the cathode surface thereby forming the CDS region. Earlier, we estimated the electric field intensity in the CDS by measuring the peak shift of some He I spectral lines [8] and values in the...
range of 3–5 kV/cm were obtained. The latter E-field values exceed those used above (where $U < U_{thr}$) and hence there is in fact no reason for the PE contribution to be neglected a priori in the measured galvanic signal.

Together with the PE signal, the laser light induces another nonresonant galvanic process, i.e., space ionization. The latter refers to direct ionization of (in our case) Ne atoms residing in excited levels close enough to the ionization potential to be photoionized. Atoms, residing in highly excited levels are characteristic of HCD since, as the EEDF confirms, very energetic electrons are available for high excitation in collisions. Consequently the corresponding galvanic contribution $D_{USP}$ should also be borne in mind as they have an instrumental impact.

Figure 4 illustrates the measured light-induced OG time resolved responses of the discharge under conditions of resonant irradiation at two Ne I line wavelengths: $\lambda = 475.27$ nm and $\lambda = 478.89$ nm. It is clear from this figure that the amplitude of the OG peak depends on the discharge current. As for the actual OG signal $D_{UOG}(t)$ in Fig. 4, this problem requires the $D_{UPG}(t)(I > I_{thr}, h\nu)$ and $D_{USP}(t)$ contributions to be accounted for as an instrumental effect in the measured $D_{UMOG}(t)$ signals.

### 3.3. Determination of real OG signal in HCD

The signals $D_{UP}(t)$, $D_{USP}$ and $D_{UOG}(t)$ are generated by three different light-induced processes. Therefore, the measured signal $D_{UMOG}(t)$ represents a convolution of $D_{UOG}(t)$ and both instrumental contributions, $D_{UPG}(t)$ and $D_{USP}(t)$.

Fig. 4. Measured OG (MOG) signals in Ne/Li HCD lamp “Narva”. Laser lines irradiating 475.27 nm and 478.89 nm, $P = 25$ mW.

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Fig. 5. Instrumental function to the measured signals in Fig. 4 at different discharge currents for Ne/Li HCD. Curve “a” is the PE signal $\Delta U_{PG}(t)$.

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Really, the signal $U_{PG}(t)$ and $D_{USP}(t)$ manifest themselves as another convolution $\Delta U_{PE+SP}(t)$. Then,

$$\Delta U_{UMOG}(t) = \int_{-\infty}^{t} \Delta U_{OG}(t_1) \frac{d}{dt} \Delta U_{PE+SP}(t - t_1) dt_1. \quad (1)$$

Generally, the inverse problem, i.e., deconvolution of Eq. (1) is not an accurate one since the process requires very clean and noise free measured traces along with a very good knowledge of the instrument function which in our case is effectively the contribution $D_{PE+SP}$. $\Delta U_{PE+SP}$ may be obtained from the signal $\Delta U_{\Delta\lambda}(t)$, which is the nonresonant galvanic signal, measured in vicinity of the selected OG transition $\lambda_{scr}$, i.e., at a slightly detuned laser frequency. To obtain the nonresonant galvanic signal $\Delta U_{\Delta\lambda}(t)$ we set the OPO wavelength to 477 nm, which lies adjacent to the wavelength region of interest but does not correspond to any spectral lines in the Ne spectrum.

Figure 5 illustrates the instrumental function $\Delta U_{\Delta\lambda}(t)$ for the signals in Fig. 4. This function is measured at the same discharge current 0.6 mA and laser power 25 mW as

![Fig. 5](image)

Fig. 5. Instrumental function to the measured signals in Fig. 4 at different discharge currents for Ne/Li HCD. Curve “a” is the PE signal $\Delta U_{PG}(t)$.

![Fig. 6](image)

Fig. 6 Deconvolution of the measured signal $\Delta U_{UMOG}(t)$ by using the instrumental function $\Delta U_{\Delta\lambda}(t)$. $\Delta U_{OG}$—the proper OG signal.
the signal $\Delta U_{MOG}$. However, it is obtained at the wavelength of $\lambda = 477$ nm, close enough to the spectral lines $\lambda = 475.27$ nm and $\lambda = 478.89$ nm, but not resonant with them. The deconvolution of $\Delta U_{MOG}(t)$ by using the instrumental function $\Delta U_{\lambda_{ij}}(t)$ gives the actual signal $\Delta U_{OG}(t)$ in Fig. 6. The algorithm of Petrov [9], which is based on the Tikhonov regularization method [10], was used to compute the deconvolution.

4. Conclusions

In a typical optogalvanic experiment, the hollow cathode discharge-optogalvanic (HCD-OG) detector generates photoelectrons during irradiation by laser light. This PE component contributes to the real OG signal and manifests itself as an instrumental effect. Another instrumental impact, i.e., nonresonant space ionization (SP) is also present and appears in a convolution with PE. This instrumental convolution (function) is measured as a nonresonant galvanic signal $\Delta U_{\lambda_{ij}}(t)$ in the vicinity of the irradiated optical transition $\lambda_{ij}$. The actual OG signal $\Delta U_{OG}(t)$ is found by deconvolving the instrumental contribution from the measured signal $\Delta U_{MOG}$. The signal $\Delta U_{\lambda_{ij}}(t)$ represents the instrumental function, describing the galvanic contributions of both photoemission and nonselective space ionization.

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References