Nonselective and polarization effects in time-resolved optogalvanic spectroscopy

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Abstract. Three interfering effects in optogalvanic (OG) spectroscopy are identified in a hollow cathode discharge (HCD) – OG detector. The laser beam is found to generate two nonselective processes, namely photoelectron emission (PE) from the cathode surface with a sub-breakdown bias applied, and nonresonant space ionization. The convolution of these galvanic contributions was determined experimentally as an instrumental function and a deconvolution procedure to determine the actual OG signal was developed. Specific plasma conductance is detected dependent on the polarization of the laser beam irradiating. Linearly/circularly polarized light beam is found to induce OG signals differ in amplitude (and their shape parameters in the time-resolved OG signals (TROGS)). The phenomena coherence and specific conductance are found to be in causal relationship. The additional conductance due to coherent states of atoms manifests itself as an intrinsic instrumental property of OG detector.

1. Introduction

The light portion absorbed on a selected optical transition $i \rightarrow k$ transforms itself into an additional conductance (OG effect) [1, 2] in the OG detector. The measured OG signal represents the galvanic effect due to both light induced small initial departure $\Delta n_{ik}$ from/to the steady state population density $n_i/n_k$ and its relaxation. When this effect is time-averaged an amplitude OG signal is measured; when a short enough laser pulse is absorbed the time-resolved OG signal (TROGS) is observable and the relaxing processes (spontaneous decay of $k$- state, ionization $\Delta n_e$ and electron gas heating $\Delta n_e$) may be identified.

The hollow cathode discharge (HCD) is the preferred medium – OG detector due to its inherent low level of conductivity fluctuations or galvanic noise. On the other hand, due to the commensurability of the large contact surfaces: negative glow (NG) – cathode dark space (CDS) – cathode surface, the HCD is known to be a reservoir of sputtered atoms mostly residing in the ground state. Another feature of the HCD is the characteristic electron energy distribution function (EEDF), which contains three groups of electrons, including one group with energy of up to a few hundred eV. Thus, the HCD extends the application of OG spectroscopy to states of sputtered atoms/ions [3] and also to high lying states of buffer gases [4]. Apart from the usual OG technique, Alternating Current Electro-Luminescent (ACEL) structure light emission has also been studied by using another OG

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scheme containing a HCD [5]. However, the above intrinsic HCD properties also introduce some specific complications to the measured OG signal in HCDs [6] including the important contribution of photon or $\gamma$- processes to the OG signal [4, 7].

Instrumental effects are a basic aspect of any measuring technique and a full understanding of their origin and magnitude is essential for the successful extraction of the real physical parameters from the measured parameter set. As for HCD the above spectral advantages are the reason for some interfering effects in HCD-OG detector. The laser light beam generates additional conductance due to its interaction with both cathode surface and highly excited states of buffer gas. To date the transfer function between input light stimulus and the output galvanic signal has been only partially discussed within the context of OG spectroscopy, based on the HCD, have already been reported [7].

In this study three light-induced instrumental effects in HCDs are discussed, namely: i/ the photoelectron emission (PE) from the cathode surface in the absence of a discharge but with an applied bias; ii/ nonresonant space ionization (SP) from the highly excited states of buffers; iii/ the state of polarization of the irradiating light beam. These instrumental issues, and the need to explore, quantify and account for them, came to light during a systematic study of the coherent conductivity of aligned and oriented ensembles of atoms [8] in HCDs irradiated by linearly and circularly polarized laser light. We present the results of this study here.

2. Experimental set-up

Time-resolved measurements of the modulation in the voltage across the hollow cathode lamp in the absence (HC) and presence (HCD) of a discharge respectively were performed. Figure 1 show the experimental set-up used to observe time-resolved PE and OG signals. The trade marked HCD lamps Ne/Ca, Ne/Li and Ne/Cd (“Narva”), were used. The discharge was produced by applying a highly stable DC voltage and was operated in 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 an OPO system which delivered a pulse of duration 4 ns FWHM at a repetition rate of 10 Hz and an average output power 25 mW and were detected across $R_m = 20 \, k\Omega$ with a digital oscilloscope. The RC constant was chosen low enough to allow one to resolve the time-dependent signal.

3. Experimental results and discussion

Three types of light-induced time-resolved galvanic responses were measured, i.e. the photoelectron emission $\Delta U_{PE}(t)$ from the cathode surface in the absence of a discharge but with an applied bias (figure 2a), the conductivity change $\Delta U_{PE+SP}(t)$ (figure 2b) due to the nonresonant irradiation of the HCD and the conductivity change of HCD due to resonant light pulse irradiation, i.e. the measured TROGS signal $\Delta U_{MOG}(t)$. TROGSs due to both circularly and plane polarized light irradiation are also compared in figure 4.
Figure 2. Light-induced time-resolved galvanic responses: (a) photoelectron emission (PE) from the cathode in absence of a HCD; (b) additive nonresonant TROG response $\Delta U_{PE+SP}(t)$ due to both PE- and space ionization effects; (c) measured resonant TROG signal $\Delta U_{MOG}(t)$.

3.1. Photoelectron emission (PE) from the cathode surface and nonresonant space ionization of buffers. Excluding of the background OG component

Figure 2a displays the light-induced PE time-resolved response $\Delta U_{PE}(t)$ of the cathode surface to the absorbed photons at various subthreshold voltage $U$, i.e. $0 < U < U_{thr}$ ($U_{thr}$, the threshold voltage of HCD on). The PE maximum $\Delta U_{PE,max}$ occurs at a time ($t_{max}$) up to $1 \mu$s after the onset of the signal depending on the voltage applied. The observed time decay of the responses is $(15 \pm 80) \mu$s which is typical for TROGS too. Ultimately, the data $\Delta U_{PE}(t)$ measured at values $U < U_{thr}$ characterize the PE properties of the cathode surface material but not the PE of the cathode surface in the presence of a discharge (HCD mode on). In HCD mode the voltage applied is confined to a thin layer of 1-2 mm thickness near to the cathode surface and forms the cathode dark space (CDS) region. Earlier we estimated the electric field in the CDS by measuring the peak shift of some He I spectral lines [9] where values in the range 3-5 kV/cm were obtained. This voltage (300 V ÷ 1000 V) exceeds the values used above of $U < U_{thr}$ and there is in fact no reason for the PE contribution to be neglected a priori in the measured galvanic signal.

Apart from this PE contribution, the laser light induces one more nonresonant galvanic process, i.e. space ionization. The latter takes place in HCD mode and refers to ionization from atoms residing in excited states close enough to the ionization threshold to be easily ionized. These ensembles of atoms, which reside in levels at high excitation energies, are characteristic of HCDs because they are intimately connected to its EEDF and the corresponding galvanic contribution $\Delta U_{SP}(t)$ should also be accounted for as they have an instrumental impact. The function $\Delta U_{SP}(t)$ manifests itself together with $\Delta U_{PE}(t)$ only in the cup-shaped HCD lamps studied. Figure 2b illustrates the additive effect of the two separate contributions as the function $\Delta U_{PE+SP}(t)$ measured at the nonresonant light irradiation by $\lambda = 655$ nm.

The measured resonant TROGSs $\Delta U_{MOG}(t)$ (figure 2c) are induced on the transition NeI (2s$^2$-1p$^2$) irradiated by its own line NeI 659.9 nm. The values of $\Delta U_{MOG}(t)$ are affected by the above PE- and SP processes. The function $\Delta U_{PE+SP}(t)$ is measured at the same discharge current 1 mA and laser power 2mW of the nonresonant line $\lambda = 655$ nm. The closest spectral lines 659.9 nm and 653.3 nm are far
enough away from the laser wavelength not to contribute to the resonant measurement. Thus $\Delta U_{PE+SP}(t)$ can be taken as an instrumental function.

Then the actual TROGS $\Delta U_{OG}(t)$ signal can be found by the deconvolution of $\Delta U_{MOG}(t)$:

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

The deconvolution of $\Delta U_{MOG}(t)$ by using the instrumental function $\Delta U_{PE+SP}(t)$ gives the actual signal $\Delta U_{OG}(t)$ in figure 3. The algorithm of Petrov [10], which is based on the Tikhonov regularization method [11], was used to compute the deconvolution.

Figure 3. Deconvolution of the measured signal $\Delta U_{MOG}(t)$ by using the instrumental function $\Delta U_{PE+SP}(t)$. $\Delta U_{OG}(t)$ - the actual OG signal in Ne/Ca HCD at discharge current $I = 1 \text{ mA}$, $P = 2 \text{ mW}$.

3.2. TROG signal and polarization of light irradiating

Linearly/circularly polarized light is known to induce coherence, namely alignment/orientation of the atoms [8]. Within the framework of a comparative study of aligned/oriented ensemble of atoms, the TROG signal $\Delta U_{OG}(t)$ due to linearly/circularly polarized light at one and the same optical transition and at identical discharge current and laser power using an OPO system (figure 1) were detected. The measured TROG signal $\Delta U_{OG}(t)$ signals (figure 4 (a, b)) give evidence two general regularities of their structure (the evolutions $\Delta U_{OG}(t)$ are composed by a fast rising peak followed by an exponential decay to either base line or signal with an opposite sign, which returns to the base line) and timeline duration/damping (different durations dependent on the optical transition) but allows in the $\mu$s time scale).

Figure 4. Time-resolved optogalvanic signals induced by linearly and circularly polarized laser light pulse.
In general, the corresponding evolutions $\Delta U^{\text{OG}}(t)^{\text{linear}}$ and $\Delta U^{\text{OG}}(t)^{\text{circal}}$ are not identical. They differ in waveform $\Delta U^{\text{OG}}(t)$ parameters:

i/ The peak $|\Delta U^{\text{OG}}(t)^{\text{linear}}^{\text{max}}|$ of the aligned atoms dominates in amplitude that of oriented ones. It is more evident on the transition $1s\rightarrow2p$ (figure 4(a, b));

ii/ The peaks $\Delta U^{\text{OG}}(t)^{\text{circal}}_{\text{max}}$ lie at $t$- coordinates different for aligned and oriented ensembles $\Delta U^{\text{OG}}(t)^{\text{linear}}$ and $\Delta U^{\text{OG}}(t)^{\text{circal}}$. A clear shift of nearby $4 \mu s$ is observable (figure 4a);

iii/ Different decay slopes characterize the evolutions $\Delta U^{\text{OG}}(t)^{\text{linear}}$ and $\Delta U^{\text{OG}}(t)^{\text{circal}}$. The best fit of the exponential part of the evolution gives different time of relaxation $T$ in the single-exponential approximation $y = A*\exp(-t/T)$ applied.

Figure 4 (a, b) displays that the waveform parameters of TROGS depend on the polarization if inducing light.

The TROGS in figure 4c has to be analyzed separately from the rest. Earlier TROGSs of similar oscillating shape were observed near $L-V$ inflection points [7] and taken as an extremal instrumental manifestation of the transient properties of the measuring circle. The oscillating TROGS characterizes the OG detector to stability for disturbance only and gives no information on the optical transition, manifestation of the transient properties of the measuring circle. The oscillating TROGS is different for aligned and oriented ensembles.

Generally, one may write down for the OG signal:

$$\Delta U_{\text{MOG}} = \pm |\Delta U(\lambda_{ik})^{\text{aligned}}\pm \Delta U(\Delta m = \pm 1) 0, \pm 2)|,$$  

(2)

where the dominating energy term $\Delta U(\lambda_{ik})$ describes the OG efficiency of population transfer $i \rightarrow k$. The second term $\Delta U = \Delta U(\Delta m)_{\text{aligned}}$ describes the OG contribution of the magnetic states $m$ of the aligned ($\Delta m = 0$, $\pm 2$) or oriented ($\Delta m = \pm 1$) atoms [8]. This term includes also the spontaneous coherence, namely self-alignment arising in any gas discharge due to space anisotropy of the excitation processes [8]. So far, the term $\pm \Delta U(\Delta m)$ has not been taken into account. On the other hand both (self)-aligned and oriented states can be influenced by a magnetic field [8, 12], including the lab. Therefore, the term $\pm \Delta U(\Delta m)$ manifests itself as an instrumental factor vs any measured OG signal.

4. Conclusions

Two nonresonant light induced processes, i.e. photoelectron emission (PE) from the cathode surface and space ionization SP are specific to the cup-shaped HCD lamps – OG detector. The measurements in HCD off illustrate the significance of PE process. The observed time decay of the responses $\Delta U_{\text{PE}}(t)$ is (15$\pm80$) $\mu s$, which is typical for TROGS too. In HCD mode for operation the two effects manifest themselves simultaneously as a galvanic background and may be excluded from the measured TROG signal by measuring their convolution $\Delta U_{\text{PE+SP}}(t)$ in vicinity of the selected optical transition.

The opto-galvanic signal depends also on the polarization of the illuminating light via the coherent state (alignment/orientation) of atoms. Therefore, an additional instrumental influence due to any change in either initial polarization or magnetic field around the OG detector may have to be accounted for in OG experiments.

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