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Anomalous optogalvanic signal. Spectrometric applications

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Along with the conventional optogalvanic (OG) signal an anomalous light-induced response has been detected in hollow cathode discharge. This response manifests itself both in the amplitude and time-resolved OG reactions. The anomalous OG signal is found to be less dependent on the absorbing optical transition and yet, more informative on the parameters of the plasma medium, that is the OG detector. This circumstance is proposed for usage in four applications based on the behaviour of the anomalous OG signal. The latter is taken as a spectral marker, as an amplified quasi-amplitude signal as well as a sensitive tool in monitoring procedure.

Keywords: hollow cathode discharge, optogalvanic effect, dynamic optogalvanic signal, inflection operating point, spectral marker.

1. Introduction

The optogalvanic effect (OGE) is known as a light-induced change of gas-discharge conductivity. It manifests itself in the discharge current or voltage variation. Two kinds of OG signals (OGS) can be detected depending on the experimental arrangement. When the OG detector plasma is irradiated by a short enough light pulse, a dynamic (time-resolved) DOG response $\Delta U(\lambda,t)$ may be detected. Generally, it informs on the relaxation processes concerning the levels irradiated. When a chopped light beam is irradiating, the detecting lock-in system gives an averaged amplitude OG response $\Delta U(\lambda)$. The two types of OG responses are the base of OG spectroscopy [1,2].

Dynamic and amplitude OG reactions of specific kinds were observed in Refs. 3–8. Here instead of the normal DOG signals (1–3 peaks, duration 40–80 µs, amplitude in mV scale) in a hollow cathode discharge (HCD) a slow dumped oscillation [duration (0.8–1.0) ms, amplitude in Volts] was detected. Lately, frequency changes in spontaneously oscillating d.c. discharge (pseudo-sonic waves) have been used for detection of ^{21}Ne in pure Ne [9]. Close to the last application is that of Rusak and co-authors [10]. In the same context the stratification of d.c. discharge plasma should be taken in mind [11] as a plasma instability manifestation.

As for the amplitude OG signal a local peak-like response takes place at the same discharge current corresponding to the oscillating DOG response [6]. We called these kinds of unconventional OGS anomalous ones [12] and considered them as an instrumental manifestation at lower degree of stability to disturbance (SD) of the DOG circuit.

In this communication a concise statement of the anomalous OGS is developed by using a macroapproach and four applications are discussed. Two spectral applications are based on the relation between the optical transition and oscillating reaction and on the transformation of the oscillating reaction into a quasi-amplitude OG signal. The other applications are of monitoring character and are based on the sensitivity of the DOG oscillation to the gas discharge operating point.

2. Experiments. Dumped oscillating OG response in HCD

In performing various DOG measurements in HCD-OG detector in a standard OG measuring scheme (R_m = 1.5 KΩ, connected in series with a 10^4 pF decoupling capacitor C) (Fig. 1), i.e., in both trademark HCD lamps ("Pye

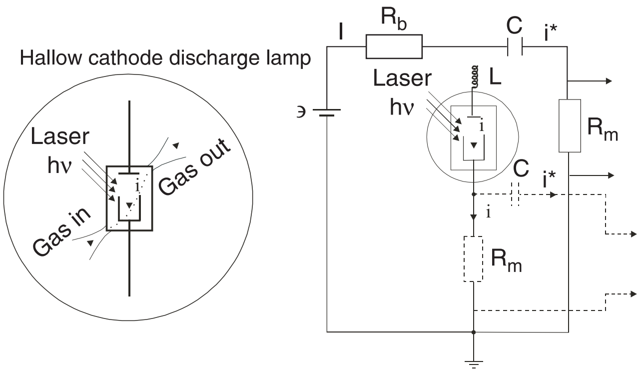

Fig. 1. Experimental set up, э is the power supply, R_b is the ballast resistor, HCD-OG detector, *L* is the equivalent inductance, *C* is the decoupling capacitor, R_m is the measuring resistor, *i** is current in the measuring circuit, and *I* is the current through R_b.

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Unicam", "Narva", "Hilger") and home-made ones, anomalous OG responses were detected (Figs. 2 and 3). They arise in a narrow Δi region on *i-V* curve where the dynamic resistance is negative: $dV/di < 0$ (Fig. 3, curve *d*). Here, the normal relaxation of the pulse (Rhodamine 6G, 10 ns, 100 Hz, 100 mW in the yellow region) light-populated/depopulated levels manifests itself as a dumped oscillation (Fig. 2). It starts after the initial peak as a phase change, i.e., in the first 15–20 µs and dumps in about 1 ms. If the discharge current interval is scanned by a short enough step, a smooth transition between the normal and anomalous DOG responses may be observed. As for the amplitude OG response, it manifests itself as a peak near the region $dV/di < 0$ (Fig. 3, curve *a*). The noise background near the inflection $\{i\text{-}V\}$ points was measured to be of higher level [6].

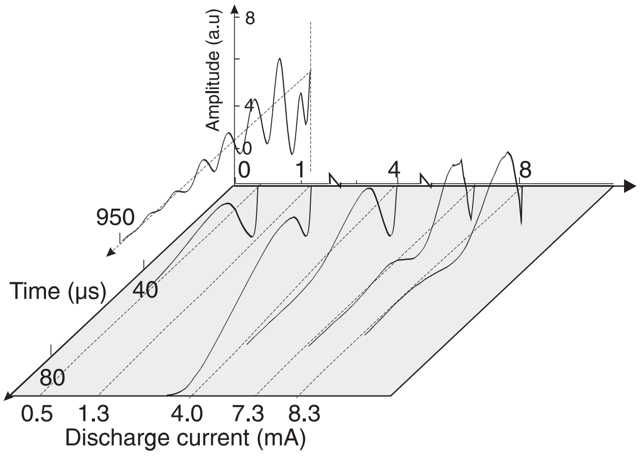

Fig. 2. Transition normal-to-anomalous DOG signal in Ne/Cu ("Narva") HCD lamp induced by DYE laser pulse (594.5 nm, 10 ns) at discharge current changing with small enough step. The time scale (0–80 µs) is not large enough for the oscillation by (0–980 µs) at i = 1.3 mA.

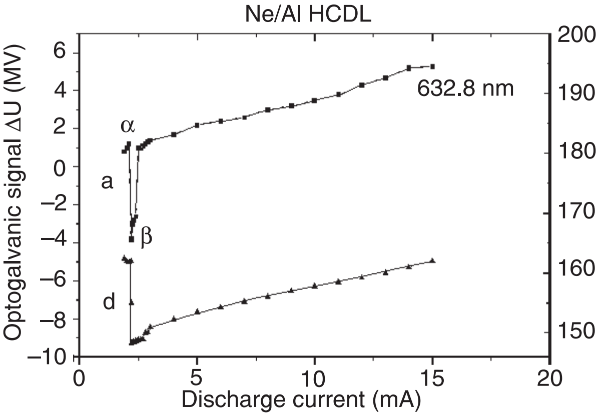

Fig. 3. Amplitude OG signal ΔU(i) (curve *a*) at 632.8 nm irradiation and *i-V* (curve *d*) in Ne/Al HCDL ("Narva").

The above-mentioned peculiarities of the anomalous response draw the attention to the reaction of the whole DOG circuit. A macroapproach was developed in Ref. 12 by using an equation describing the DOG circuit reaction to a Heaviside's type small galvanic perturbation. Figure 4 illustrates graphically the local solutions. They depend on the relation q/ω, where the coefficients q and characterize the reaction dumping and frequency

$$q = \{L + C[R_b R_m + R^*(R_b + R_m)]\}[2CL(R_m + R_b)]^{-1},$$

$$\omega = (R_b + R^*)[CL(R_b + R_m)]^{-1}$$

where R^* is a macroparameter depending on the HCD plasma resistance.

The solution in Fig. 4.1 is the closest in form to the initial perturbation. The solution in Fig. 4.4 takes place at $q << \omega$ and it is the closest in form to the observed oscillation. The oscillating solution requires negative dynamic resistance $R^* = dV/di < 0$. The corresponding *i-V* curves turned out to contain such negative R^*-branch.

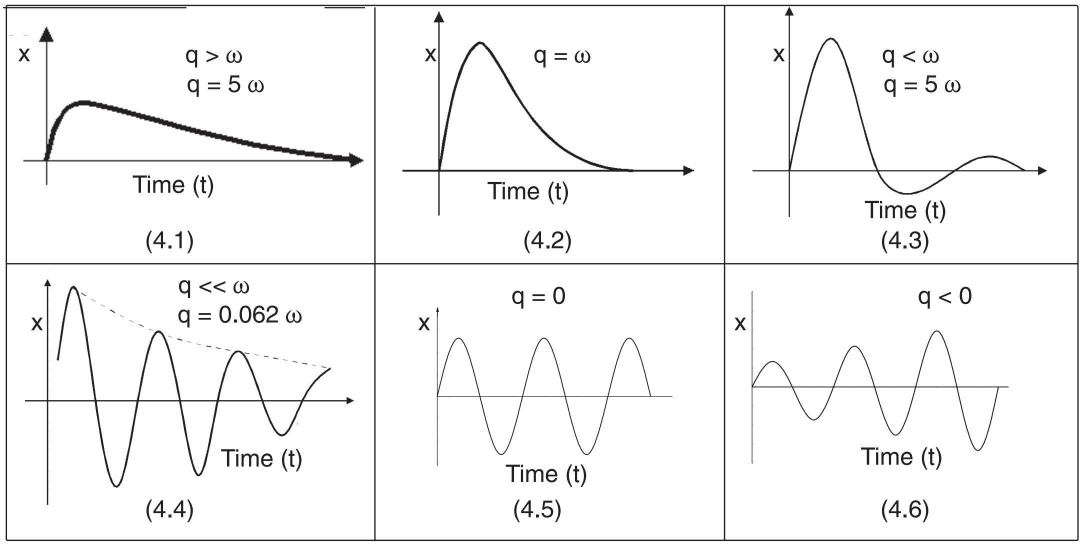

Fig. 4. Solutions of the time dependent equation, describing the OG circuit.

The solutions are stable excluding the vicinity of the inflection point $R^* < 0$. Figure 2 illustrates the transition normal-to-oscillating DOG response. This behaviour and the correlation with *i-V* curves are confirmed in simulating experiments. They are realised by adding an alternating component to the power supply (2 V, 0.4 ms). Without any illumination the oscillations and normal reactions are found to take place also due to this galvanic perturbation. This result imparts instrumental accent to the local solutions in Fig. 4, i.e., they represent transfer function manifestation at low stability to disturbance of the OG circuit (SDOGC). Obviously SDOGC depends on the current *L-*, *C-*, R_b-, R_m-, R^*-values combinations. The lower SDOGC means that the current OG circuit properties dominate the adequate OG reaction.

Thus the anomalous OG signal gives more information about the plasma-detector than about the absorbing optical transition.

The inflection point has been observed to drift in about 2 mA during about 250 hours of HCD tube operation. The spectral emission analysis shows a constant spectrum. It suggests that the inflection point drifting can be ascribed to the changing of the cathode surface due to sputtering process. The sputtered atoms were found responsible for the inflection point appearance after Penning's ionisation $A^{(M)} + B \rightarrow A + B^+ + e$ in HCD, where $A^{(M)}$ is the metastable atom of the buffer gas, B the sputtered atom [6,12]. Earlier the generated additionally charged particles were taken causing plasma instability [11].

3. Spectrometric applications of the oscillating DOG responses

3.1. Oscillating response as spectral OG marker

The oscillating response was found to arise at a population perturbation of levels, which essentially contribute to the ionisation, such as NeI ($1s_i$–$2p_j$) metastable levels, for example. On the other hand, when the upper level is closer than 1 eV to the ionisation limit this transition gives a single DOG peak. Its form repeats that of the pulse illuminating. No oscillation gives the illuminated sputtered atom in DOG measurement. Thus close enough to the inflection point $R^* < 0$ the DOG spectrum should contain also a certain number of dumped oscillations. Therefore the oscillations may be used as spectral markers.

This possibility is applied under identification of Nd I 581.39 nm and Na I 588.99 nm spectral lines in a real experiment with HCD in Ne buffer gas. A DAY laser (Rhodamin 6G) has to be tuned at these wavelengths for monitoring appearance of NdI and NaI isotopes. The spectral region of interest might be localised by using three consecutive oscillating DOG signals, belonging to three of ($1s_i$–$2p_j$) optical transitions of NeI, i.e., of NeI 597.55 nm, NeI 594.50 nm and 588.19 nm spectral lines. A homemade HCD tube (2 mm radius, 13 mm length, 3.5 Torr Ne gas pressure) is used. The value 7.2 mA discharge current turned out an inflection operating point. At this *i*-value oscillating OG reactions take place for metastable NeI ($1s_i$–$2p_j$) transitions being irradiated. Figure 5 illustrates the observed DOG spectrum within (597.55–576.40 nm). The single DOG signals of the searched lines are easy localisable among the multitude of oscillating and single NeI DOG ones.

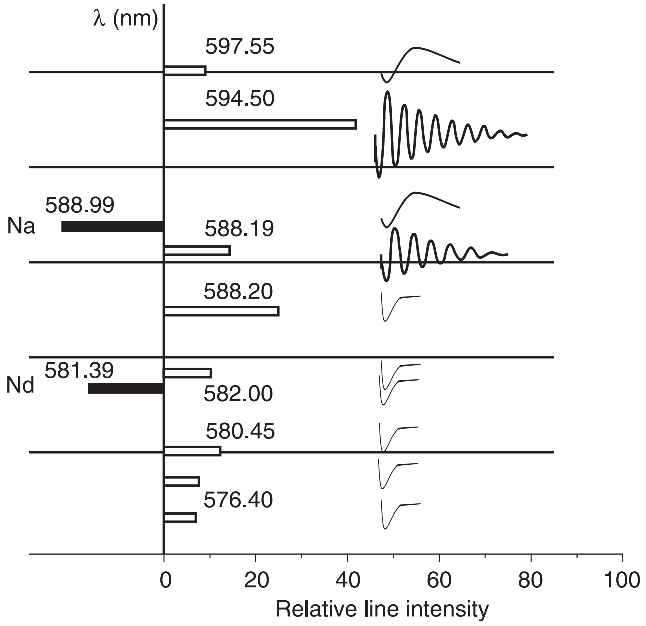

Fig. 5. The searched Nd and Na lines, a part of NeI optical spectrum (open bars) and their OG signals as markers in the DYE laser spectrum.

3.2. Quasi-amplitude OG spectrum. Enhancement of the OG signal

Just by adding a simple integrating scheme, the above mentioned DOG signals *ΔU(t,λ)* received another application, based on their peak transformation to a stationary *ΔU(λ)* spectrum. In this arrangement some $\Delta U(\lambda_i)$ peaks may be increased by integrating their oscillating DOG response.

A filter-integrating *RC* circuit (Fig. 6) averages either of *ΔU(t,λ)* polarities, usually the dominating one. Hence a scanning DYE laser induces an OG spectrum where for every λ

$$\Delta U(\lambda) = \int_{\tau} \Delta U(\lambda, t) dt,$$

where *t* is the average constant adjusted by the resistor R_2. The latter together with the capacitor C_a determine the smoothness of every peak *ΔU(λ)*. Figure 6 shows also a part of OG spectrum obtained by integrating DOG signals, where $R_1 = 0.5$ kΩ, $R_2 = 4$ kΩ, $C_a = 0.33$ mF. Obviously, in this case the amplitude *ΔU(λ)* depends also on the integrated polarity.

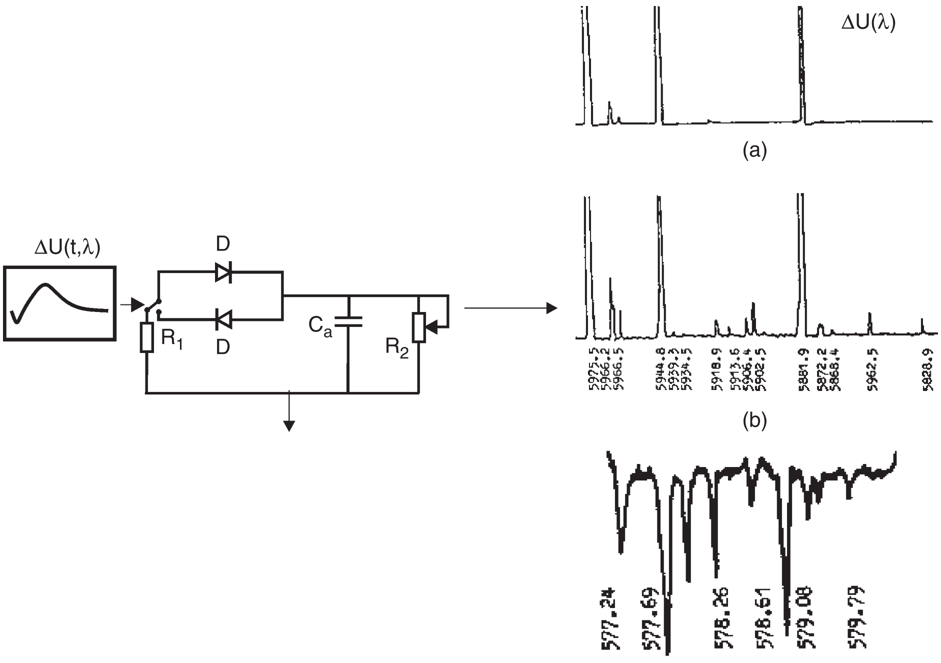

Fig. 6. Quasi-amplitude OG spectrum ΔU(λ). In Ne/Cu HCDL (“Narva”) within 583–589 nm at 3 mA (a) and 12 mA (b) discharge current. In Ne/V HCDL (“Narva”) at 5 mA discharge current within 577–580 nm (c).

The scheme allows the most optimal discharge current or buffer gas pressure in OG detector to be taken in relation to each signal $\Delta U(\lambda)$ of interest. This scheme gives one more specific advantage in the case of a weak DOG signal. Its quasi-amplitude analogue may be enhanced in amplitude by using a DOG circuit of lower degree of stability. Then, the transformed signal is of higher amplitude due to the larger integrated area.

4. Conclusions

An anomalous OG response in HCD- detector is applied as useful spectroscopic tool. This anomalous OG signal appears near the operating *i-V* point of negative dynamic resistance. Here, the weak perturbation initiates nonlinear amplitude and oscillating galvanic reaction. The galvanic simulation confirms the instrumental essence of the observed anomalous OG signal.

The specific oscillating form of the light pulse induced OG signal is proposed as a spectral marker in an OG unidentified spectrum. A transformation of the normal and oscillating DOG responses in quasi-amplitude one is realised.

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