

Opto-galvanic effect on degenerate magnetic states of sputtered atoms in a glow discharge

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Abstract. The opto-galvanic response of some degenerate states of sputtered atoms to linearly- and circularly polarized light is studied. On the same optical transition both time-resolved- and amplitude opto-galvanic signals are found depending on the polarizations of light absorbed. The latter induces galvanic responses differing in opto-galvanic efficiency, time-evolution and sensitivity to discharge current and laser power. The differences are ascribed to the rate constants of the decay processes, characterizing aligned and oriented atoms.

1. Introduction

The degenerate magnetic states m are known as the first ones excited in a coherent superposition, i.e. linearly polarized resonant light absorbed *aligns* them (coherence of the states of $\Delta m = 0, \pm 2$); circularly polarized resonant light *orients* m - states at either $m = 0 \rightarrow m = 1$ or $m = 0 \rightarrow m = -1$ [1]. Both *alignment* and *orientation* manifest themselves optically in linearly- or circularly polarized spontaneous emission. In a real gas discharge medium either *alignment* or *orientation* means that the resulting coherent m - states above appear along with other permitted m -states. In other words, the coherences add new spatial dispositions of valence electron. On the other hand both ionization and excitation of atoms are anisotropic in low-temperature plasma and its conductivity should depend, generally, on the magnetic states m too. So far, this dependence is not taken into account in modeling/calculation of the interaction atom-electron. For the first time, the relation *coherence* - *conductivity* j has been studied as a conductivity change Δj of the same ensemble of atoms *aligned* $\leftrightarrow$ *non-aligned* sequentially [2-4].

Recently, a preliminary study found some different correlations *coherence (alignment/orientation)* - *conductivity* of some states of neon atom in a hollow cathode discharge (HCD) [5]. It means that interaction electron $\leftrightarrow$ atom (*aligned/oriented*) depend on the coherence induced.

The main objective of this study is the conductivity of an ensemble of sputtered atoms in a HCD as a function of their *alignment/orientation*. Generally, one such dependence found in detailed comparisons would represent an important result, namely, that one or other light polarization strongly influences rate constants of electron-atom interaction.

HCD is known as a reservoir of sputtered atoms, first of all [6]. One and the same ensemble of sputtered atoms that is *aligned* $\leftrightarrow$ *oriented* in sequence is studied by using light-induced coherence. On the other hand, the resonant light absorption is the necessary condition for another effect in a gas

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discharge, i.e. opto-galvanic (OG) one [7]. The latter is the galvanic manifestation of the redistributing action of light absorbed vs. steady state population. Ultimately, a light-induced change of gas discharge conductivity, i.e. OG signal (OGS), may be detected. The time-resolved OGS (TROGS) describes the transient processes of relaxation related to the conductivity of the medium which is absorbing. Therefore the TROGS is the most appropriate technique for precisely comparing *aligned* ↔ *atoms* in their coherent conductivity. The amplitude (OGS) (AOGS) is an averaged galvanic replica of the chopped laser beam irradiating.

This investigation is realized as a comparison of opto-galvanic signals (OGSs) induced by linearly- and circularly polarized light. OGSs on the transitions Ca I ($3p^6 3d4s\ ^3D \rightarrow 3p^6 3d4p\ ^3D^0$) ($\lambda = 560.13$ nm), Li II transition ($1s2s\ ^3S \rightarrow 1s2p\ ^3P^o$) ($\lambda = 548.35$ nm), Li II ($1s2s\ ^3S \rightarrow 1s2p\ ^3P^o$) ($\lambda = 548.57$ nm) and Cd I transition ($4d^{10}5s5p\ ^3P^o \rightarrow 4d^{10}5s7s\ ^3S$) ($\lambda = 325.25$ nm) (designations under reference [8]) are compared.

Earlier the sputtering of Li atoms by Ne^+ ion in a HCD was modeled [9]. As for atoms of Ca they are difficult for atomization and a HCD represents the optimal medium for sputtering.

2. Experimental set-up

Two types of OGSs, i.e. time-resolved (TROGSs) and amplitude (AOGSs) are detected by using set-up in figure 1. The trademarked HCD lamps Ne/Ca-, Ne/Cd- and Ne/Li (all “Narva”) were used. The discharge was produced by applying a highly stable dc voltage and was operated in a NG regime. A standard experimental scheme for TROGS detection was employed. TROGSs were induced by irradiating the 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 of 25 mW and were detected across $R_m = 20\ \text{k}\Omega$ ($C = 10\ \text{nF}$) with a digital oscilloscope. The RC constant was chosen low enough to detect the time-dependent signal $\Delta U(t)$. The latter represents a galvanic time-resolved replica-evolution of the peak-light perturbation. Plane- and circularly polarized light pulses irradiate the discharge in sequence and their TROGSs are compared (see Figs. 2).

AOGSs are measured by using He-Cd laser (“KIMMON” electric co., ltd. $\lambda = 325$ nm, 25 mW), chopper and selective nanovoltmeter “Unipan” (type 237). The values of $R_m = 1\ \text{m}\Omega$ and $C = 10^2\ \text{mF}$ are used. The equality in intensity of the illuminating light polarizations is adjusted and controlled.

3. Experimental results and discussions

The analysis of the OGSs is based on the understanding that sputtered atoms which are not returned back to the cathode are thermalized by the buffers in the cathode dark space [10] and no overriding their velocity vector does not exist.

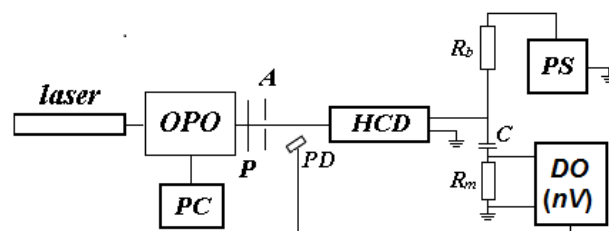


Figure 1. Experimental set-up: HCD – hollow cathode discharge lamp, OPO – optical parametric oscillator system, P – polarizer (linear or achromatic waveplate $\lambda/4$, 450 - 800 nm), A – aperture, PD – photo detector, C – decoupling capacitor 10 nF, R_b – ballast resistor, $R_m = 20\ \text{k}\Omega$ – measuring resistor, PC – personal computer, PS – power supply, DO – digital oscilloscope, nV – nanovoltmeter.

Figures 2 (a ÷ c) contain certain general TROGSs regularities: *i/* the signal is composed by a fast rising peak followed by an exponential decay to either base line or a part of opposite sign, which returns to the base line; *ii/* the discharge current (dc) I extends the waveforms $\Delta U^{OG}(t)$, namely both the first peak and the rest damping part; *iii/* dc stimulates the first peak amplitude, namely $\Delta U^{OG(max)} \propto I$ on Li II transitions; on Ca I transition the inverse relationship $\Delta U^{OG(max)}(t) \propto I^{-1}$ takes place. Within these regularities the waveforms $\Delta U^{OG}(t)^{linear/circular}$ induced by linearly- or circularly polarized light are compared.

On the whole, the shapes of the two types of signals are different from one another in their parameters.

As a rule linearly polarized light induces greater galvanic perturbation $S(t)^{linear}$, where

$$S(t)^{linear/circular} = \int_0^{t_1} \Delta U^{OG}(t)^{linear/circular} dt \quad (1)$$

and t_1 [μs] – the time of integration dictated by the concrete case. The program Origin Lab is used to calculate the value of the area $S(t)$, characterizing the magnitude of the perturbation. A detailed comparative analysis of TROGSs shows the following dependences on the light polarization:

- i. The inequality $|S(t)^{linear}| > |S(t)^{circular}|$ means a higher OG efficiency of linearly polarized light on irradiated transitions, namely the ratio $S(t)^{linear}/S(t)^{circular}$ varies within the limits [1.13÷1.87] in the signals in figure 2 and the relation $S(t)^{linear}/S(t)^{circular} \propto I$ takes place. Minimum values of this relation of 1.13 (at $I = 1$ mA) and 1.23 (1.6 mA) characterize TROGSs induced by Ca I 560.13 nm. The signals induced by Li II 548.35 nm are the closest in their OG efficiency (1.62 and 1.64). The maximum values of 1.58 (1 mA) and 1.87 (1.6 mA) characterize TROGSs induced by Li II 548.57 nm.
- ii. The TROGSs in figure 2 suggest characteristic decay time T of the evolution $\Delta U^{OG}(t)$. A single-exponential approximation $y = A \cdot \exp(-x/T)$ as in reference [11] is used here to compare T -values of each pair $\Delta U^{OG}(t)^{linear/circular}$ signals and one and the same starting t -point of the fitting is taken for each pair $a_{1,2}$, $b_{1,2}$, and $c_{1,2}$ TROGSs. The signal of lower amplitude dictates the starting point. The procedure of the best fit gives T -values (see figures 2) differing in the range of $\Delta T \in [0.96 \div 2.06]$ μs.
For each pair the relation $\Delta T \propto I$ take place. Despite the small values of ΔT the fact that $T^{linear} > T^{circular}$ systematically suggests that atoms perturbed by linearly polarized light relax slower than the atoms perturbed by circularly polarized light.
- iii. Control TROGSs measures on the transition Ne I ($1s_5 \rightarrow 2p_9$) (640.2 nm) in Ne/Li HCD lamp give other relationships, namely $S(t)^{linear}/S(t)^{circular} \propto I^{-1}$ and the values $S(t)^{linear}/S(t)^{circular} = 1.94$ (at $I = 0.22$ mA) and 1.11 (at $I = 1.6$ mA). Here discharge current equalizes the galvanic perturbations due to different polarizations. Another T -relation, namely $T^{circular} > T^{linear}$ characterizes TROGSs. For example, $T^{circular} = 26.88$ μs and $T^{linear} = 8.75$ μs at 1.6 mA. In OG context the line Ne I 640.2 nm is unique in that it corresponds to the only transition linking the $1s_5$ and $2p_9$ states. No optical redistribution of the population Ne I $p_9 \{n_{p9} + \Delta n(t)\}$ to any Ne I $s_{2,3,4}$ states takes place. The light induced departure $\Delta n(t)$ may relax only via both spontaneous transition $2p_9 \rightarrow 1s_5$ and interaction Ne I $p_9 \{n_{p9} + \Delta n(t)\} + e$. Therefore, the signal due to Ne I 640.2 nm line is a sensitive probe of collisional interactions. On the other hand, since the level Ne I $1s_5$ is the only in maintaining HCD at $I \leq 1$ mA [16] its disturbance is incomparable with that of the levels Ca I $3p^6 3d4s^3D$, Li II $1s2s^3S$ and Li II $1s2s^3S$. Therefore, the above comparison should be regarded only as an ambiguous manifestation of TROGSs induced by linearly and circularly polarized resonant light.

In general, the understanding of the TROGS [8, 11-15] is based on the galvanic effect due to relaxation of the light-induced population transfer $\Delta n_{i \rightarrow j}$ from/to the steady state population density n_i/n_j . It is a fast process (in the 10^{-9} s time region) which is non-resolved in our experiment. Further, this perturbation relaxes $\Delta n_j(t)$ typically in the μs range by various channels dictated by the plasma

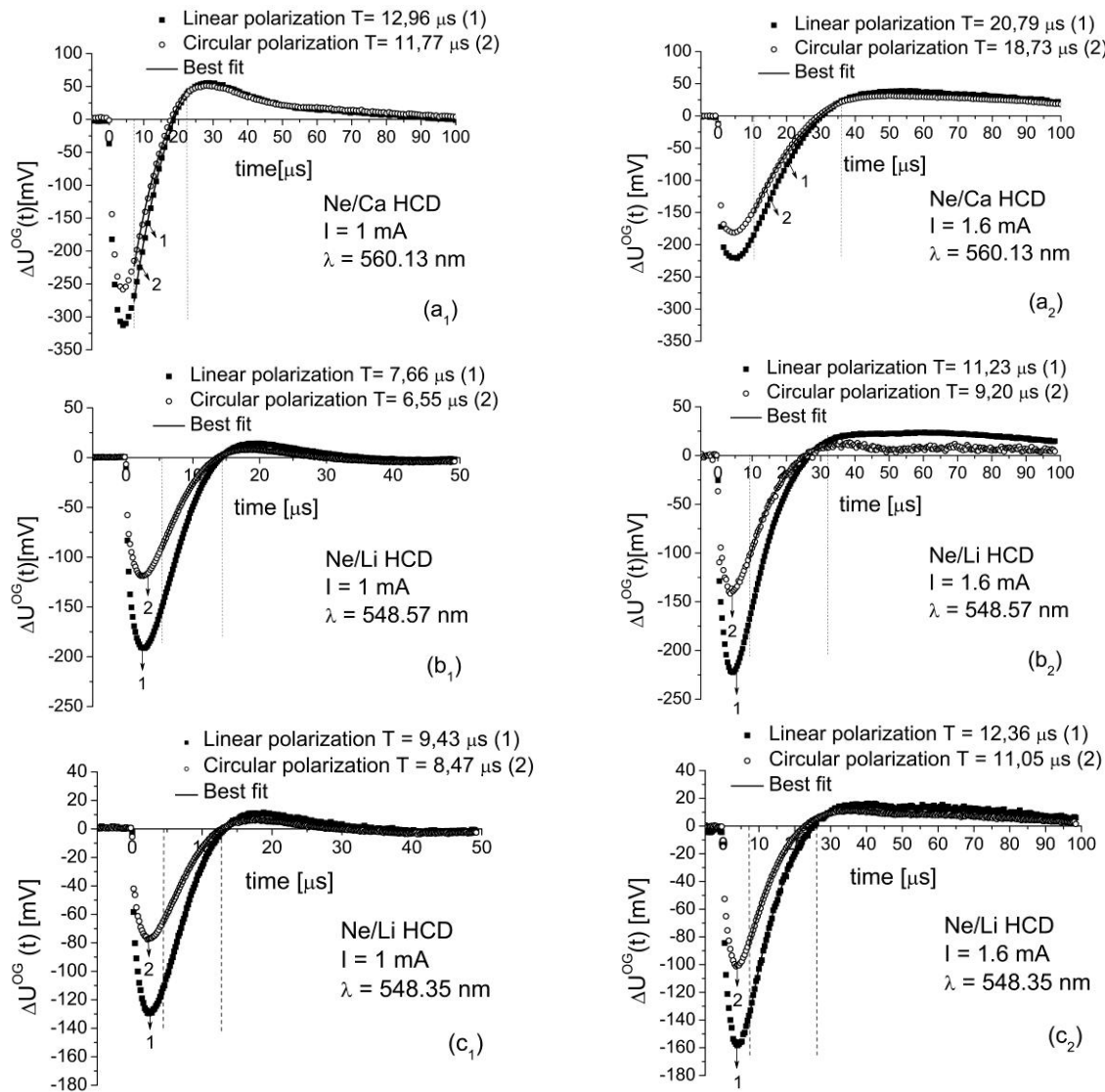


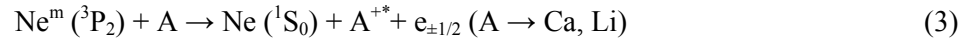
Figure 2. TROGSs induced by linear- and circular polarization of the lines Ca I ($3p^63d4s^3D \rightarrow 3p^63d4p^3D^0$) (560.13 nm), Li II ($1s2s^3S \rightarrow 1s2p^3P^o$) (548.57 nm) and Li II ($1s2s^3S \rightarrow 1s2p^3P^o$) (548.35 nm) in Ne/Ca- and Ne/Li HCD lamps. Broken lines point the time interval Δt [μs] of the fitting: a_1 -[7.2 ÷ 22.4] μs ; a_2 -[10.4 ÷ 35.2] μs ; b_1 -[5.4 ÷ 14.4] μs ; b_2 -[9.2 ÷ 32] μs ; c_1 -[4.4 ÷ 12.6] μs ; c_2 -[7.2 ÷ 26] μs .

as a whole, i.e. by the collective interaction of the corresponding level with the others via the electron collisions and radiative transitions (spontaneous decay of upper state); simultaneously, the ionization also changes due to the perturbed populations $n_i - \Delta n_{ij}(t)$ and $n_j + \Delta n_{ij}(t)$. Ultimately, the contributions of these processes to the conductivity j are reduced to changes of electron number density Δn_e and/or electron drift velocity Δv_e , i.e.

$$\Delta j = c_1 \Delta n_e + c_2 \Delta v_e \quad (2)$$

These terms define also the characteristic decay time T of the evolution $\Delta U^{OG}(t)$ at light pulse short enough. Concrete processes of the above transformation are not discussed in this comparative study.

However, a specific for HCD reaction should be taken into account vs measured signals. In a HCD the dc is known to play also another specific role, namely as a factor for the atomic sputtering yield. [7, 10] The latter is a prerequisite for Penning ionization in this discharge. In our case:



where $\text{Ne}^m(^3\text{P}_2)$ is the metastable. The ionization (3) competes with the stepwise one and was found to influence the current/voltage curve, particularly at low dc values. [2, 3] However, of greater importance is the circumstance that the metastable spin state (magnetic number) may affect the efficiency of the process (3). The ionization cross-section for Penning collisions has been studied theoretically [16] and the author has found that in a HCD the process (3) always contributes to the conductivity. Here the population transfer $\Delta n_{i \rightarrow j}$ between states of sputtered atoms the process (3) stimulates first of all the term $c_1 \Delta n_e$ in expression (2). As to the second term $c_2 \Delta v_e$ its contribution may be ignored due to lower in 10^{-3} density of sputtered Ca atoms in regard to the buffers [10]. This argument is stronger in regard to Li II ions.

Really, these measurements reveal the specific galvanic contributions of the atoms when their states $\Delta m = 0, \pm 2$ or $\Delta m = \pm 1$ dominate in number the rest m- states permitted, namely aligned and oriented. The TROGSs $\Delta \text{UOG}(t)$ align and $\Delta \text{UOG}(t)$ orient describe the appearance and evolution of two specific conductivities of one and the same ensemble of atoms that are aligned $\leftrightarrow$ oriented. Since, the processes of relaxation are the same on one and the same optical transition, the observed differences in the evolutions $\Delta \text{UOG}(t)$ linear/circular could be ascribed to different rate constants, characterizing the decay processes of the aligned/oriented ensembles of atoms.

The differences in TROGSs above are checked also on AOGSSs. He-Cd laser ($\lambda = 325.25$ nm) irradiates a Ne/Cd HCD lamp. Here the signal ΔUOG is the OG effect induced by the chopped laser beam and averaged by the τ - constant of the amplifier (figure 3). Differing from one another amplitudes ($\Delta \text{UOG}(P)$ linear and $\Delta \text{UOG}(P)$ circular) and steepness ($\partial\{\Delta \text{UOG}(P)\} \text{ linear} / \partial P$ and $\partial\{\Delta \text{UOG}(P)\} \text{ circular} / \partial P$) characterize the two types of AOGSSs. Here the logical trend $\Delta \text{UOG} \sim P$ is influenced by the dc (figure 3b) and the relation $\Delta \text{UOG}(P) \text{ circular} > \Delta \text{UOG}(P) \text{ linear}$ takes place at $I = 0.7$ mA and $P > 0.7$ mW. At least, these results confirm the differences in OG efficiency of linearly and circularly polarized light.

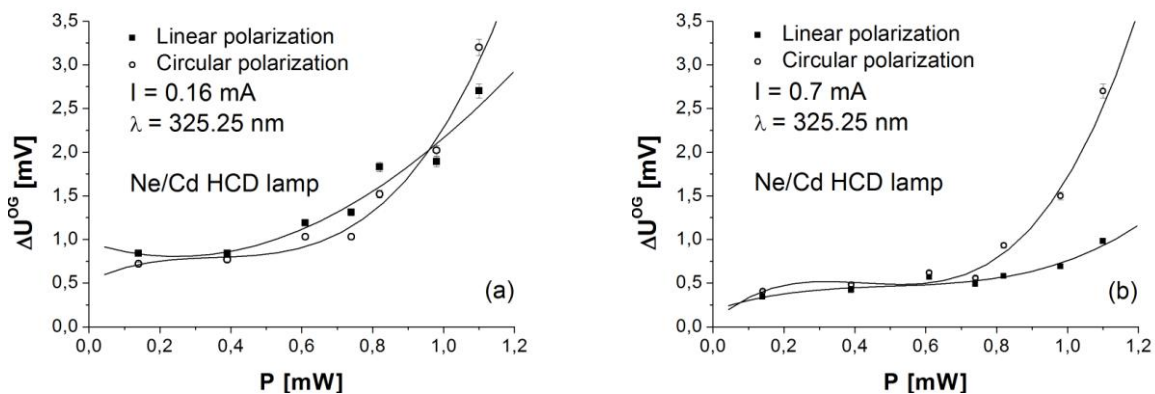


Figure 3. Amplitude OG signals induced by linear and circular polarizations of the spectral line CdI ($4d^{10}5s5p\ ^3P^0 \rightarrow 4d^{10}5s7s\ ^3S$) (325.25 nm): (a) $I = 0.16$ mA; (b) $I = 0.7$ mA.

Conclusions

Both time-resolved OGSs (TROGSs) on the transitions Ca I ($3p^6 3d4s^3D \rightarrow 3p^6 3d4p^3D^0$) (560.13 nm), Li II ($1s2s\ ^3S \rightarrow 1s2p\ ^3P^0$) (548.57 nm), Li II ($1s2s\ ^3S \rightarrow 1s2p\ ^3P^0$) (548.35 nm) and amplitude

(AOGSs) on the transition Cd I ($4d^{10}5s5p\ ^3P^0 \rightarrow 4d^{10}5s7s\ ^3S$) (325.25 nm) are observed to depend on the polarization of resonant light irradiating, i.e. either linear or circular.

Light polarizations induce TROGSs distinguishing in both OG efficiency and waveform $\Delta U^{OG}(t)$ parameters. As a rule linearly polarized light induces greater galvanic perturbation.

Characteristic decay times, namely $T^{linear} > T^{circular}$ of the evolution $\Delta U^{OG}(t)$ are found to depend on the dc. This T -relation is opposite to that on the transition Ne I ($1s_5 \rightarrow 2p_9$) (640.2 nm).

The TROGSs describe the appearance and evolution of two specific conductivities of one and the same ensemble of atoms that are aligned $\leftrightarrow$ oriented, namely $\Delta U^{OG}(t)^{align}$ and $\Delta U^{OG}(t)^{orient}$. The aligned ensemble relaxes slower than oriented one.

Since the processes of relaxation are the same on one and the same optical transition the observed differences in the evolutions $\Delta U^{OG}(t)^{linear/circular}$ mean different rate constants, characterizing the decay processes of the aligned/oriented ensembles of atoms. In gas discharge plasma (self-) illuminated the interaction atom $\leftrightarrow$ electron depends on the light polarization.

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