Galvanic manifestation of coherent degenerate Zeeman sublevels in a gas discharge

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Abstract. The degenerate Zeeman sublevels were found to contribute to the ionization in a gas discharge in accordance with their coherent state. The voltage across the discharge was monitored while the same ensemble of excited atoms was alternately self-aligned ↔ nonaligned or oriented ↔ aligned. In the latter case, the coherence was optically induced; the corresponding opto-galvanic signals, i.e. amplitude- and time-resolved ones, were compared. Each state of the ensemble of degenerate Zeeman sublevels, i.e. (self-) aligned, oriented or disordered was characterized by its own rate of ionization. Various hollow cathode discharge (HCD) media were studied, namely, Ne/As, Ne/Cu, Ne/Ni, Ne/Cd, Ne/Li and Ne/Si in the corresponding commercial HCD spectral lamps.

1. Introduction

Hanle was the first to align an excited atomic state, i.e. to prepare a coherent superposition of degenerate magnetic states \( m \) such that \( \Delta m = 0, \pm 2 \) [1] using linearly polarized resonant illumination for alignment. Later, the same coherence was found to be an attribute of gas discharge plasma [2]. It arises without any external reason (self-alignment), i.e. due to the space anisotropy of some internal process of excitation. In fact, all real gas discharge sources generate self-alignment. Another \( \Delta m \)- combination, i.e. \( \Delta m = \pm 1 \) is known as orientation [3]. It arises due to absorption of circularly polarized light. Both (self-) alignment and orientation manifest themselves optically, i.e., as linearly- and circularly-polarized spontaneous emission from interfering \( m \)- states.

In a real gas discharge medium, either (self-) alignment or orientation mean that the corresponding \( \Delta m = 0, \pm 2 \) or \( \Delta m = \pm 1 \) state dominate in number over the rest of the \( m \)- states permitted. Therefore, a certain definitive \( m \)-states disposition dominates along any arbitrary space axes. On the other hand, in low-temperature plasma the process of ionization is anisotropic. Thus, the yield of charged particles should also depend, generally, on the magnetic states \( m \). This nonenergetic contribution to the ionization (and conductivity, respectively) is present implicitly in some earlier investigations [4, 5].

In this study, the excited ensemble of atoms is investigated galvanically at the two \( \Delta m \)- transitions, i.e. i) self-aligned ↔ non-aligned and ii) aligned ↔ oriented.

A hollow cathode discharge (HCD) was used. Besides its spectroscopic properties, HCD is also known as a medium where the excited atoms are self-aligned by a characteristic beam, such as fast electrons [6, 7]. Since the self-alignment is an attributive property of HCD, the comparison in the

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ionization between self-aligned and nonaligned atoms is based on the magnetic destruction of this coherence (case i). As for the comparison aligned↔oriented atoms, these coherences are light induced (case ii).

2. Experimental set-up

The ionization was compared by measuring the voltage change $\Delta U$ across the discharge. Figure 1 illustrates the experimental set-up used. Commercial HCD spectral lamps Ne/As (“Pye Unicam”), Ne/Cu, Ne/Ni, Ne/Cd, Ne/Li and Ne/Si (“Narva”) were studied in a dc mode of operation The measurement of $\Delta U$ in a self-aligned → nonaligned ensemble (case i above) was based on an external scanning (by means of a stepper motor) magnetic field $B$ (Helmholtz coils) destroying the self-alignment. The values of $\Delta U(B)$ were measured by using a modulating (179 Hz) magnetic field $B_m$. In order to improve the S/N ratio, a phase-sensitive detection was applied. A lock-in amplifier type 232B (Unipan) provided the $\Delta U(B)$ dependence whose shape is close to that of the first derivative $\partial U(B)/\partial B$.

The experimental set-up is presented schematically in figure 1.

The measurement of an ensemble of Ne atoms being aligned ↔ oriented (case ii above) was performed by introducing light-induced alignment/orientation. Two lasers, He-Ne ($\lambda = 632.8$ nm) and He-Cd ($\lambda = 325.0$ nm), and a polarizer (linear or achromatic waveplate $\lambda/4$, 320-800 nm) were used. In essence, the set-up used was an opto-galvanic (OG) (in this case, a polarization OG) spectroscopy set-up [8]. The intensity of the illuminating light in the different polarizations was adjusted and controlled.

Figure 1. Experimental set-up: $\lambda/4$ – polarizer (linear or achromatic waveplate $\lambda/4$, 320-800 nm), $Ch$ – mechanical chopper, HCD – hollow cathode discharge, CDS – cathode dark space, $B$ – magnetic field, $B_m$ – modulating magnetic field, $C$ – decoupling capacitor, $R_m$ – measuring resistor, $NV$ – nanovoltmeter (selective type 237 or lock-in type 232B), $R_b$ – ballast resistor, $\varnothing$ – power supply.

3. Experimental results and discussion

3.1. Magnetic-field-induced transitions self-aligned–nonaligned ensemble of atoms

The weak scanning magnetic field $B$ applied was found to generate a peak-like galvanic change in the discharge. Figure 2a illustrates the galvanic behavior of Ne/As HCL as a function of $B$. The signals are close in shape to the first derivative $\partial U(B)/\partial B$. The maximum of the primitive function $\Delta U(B)$ (at $B = 0$) corresponds to the maximal self-alignment. The experimental circuit $HCD$ in $(B_0 \pm B_m)$ - $CR_m$ - $NV$ lock–in (figure 1) reduces the degree of self-alignment at any value of $B \neq 0$. Thus, the field $B$ reveals directly the difference in the conductivity at the transition self-aligned →nonaligned ensemble.

The magnetic field was directed along the geometric axis $OO$ of the cathode (figure 1). Thus, the field $B$ destroys the self-alignment along the radius $R \perp B$. This self-alignment arises due to the characteristic beam-like electrons along any radius $R$. In this geometry, resonances of magnetic depolarization (Hanle-signals) $\partial I_\lambda (B)/\partial B$ ($I_\lambda$ being the spectral line intensity in a given polarization) were observed earlier in the spontaneous emission from self-aligned levels in a HCD [7].

We should emphasize the fact that the behavior of the signals $\partial U(B)/\partial B$ as a function of the direction of $B$ is identical to that of the Hanle-signals $\partial I_\lambda (B)/\partial B$ observed earlier [7], i.e. a reduction of the signal when $B$ deviates from either $OO$ or $R$. The two kinds of signals are also of identical
behavior with respect to the discharge current $i$, i.e., their width rises with $i$. However, the manifestation of a Hanle-signal as a light emission polarization assigns it to a concrete quantum state. On the contrary, many excited (self-aligned) levels contribute to the plasma conductivity and the signal measured is an integral characteristic.

The resonances $\partial U(B)/\partial B$ were measured to be of a higher amplitude near the operating inflection $I$-$V$ points, where $\partial U/\partial I < 0$. Simultaneously, a lower signal-to-noise ratio was observed for $\partial U/\partial I < 0$. Earlier, this was found to be typical for HCD due to Penning ionization of the sputtered atoms [9]. Here, the external galvanic perturbation manifests itself as both a higher amplitude and higher noise.

3.2. Aligned/oriented ensemble of atoms

The set-up laser- Ch – HCD - CR$_m$ - selective NV (figure 1) generates and detects two types of amplitude OG (AOG) signals $\Delta U^{OG}$, i.e., due to illumination by either of linearly- or circularly-polarized light (figure 2(b, c)). The values of $\Delta U^{OG}$ were detected as a function of the laser power $P$; they characterize the conductivity of the same ensemble of atoms that are successively aligned $\leftrightarrow$ oriented. The comparison of the values $\Delta U^{OG}(P)$ reveals a general tendency, i.e. $\Delta U^{OG} \sim P$. However, within this trend, a different steepness of the dependence $\Delta U^{OG}(P)$ characterizes the aligned $\leftrightarrow$ oriented atoms. These results correlate with earlier measurements of time-resolved OG signals.

The signals $\Delta U^{OG}$ measured may be summarized as $\Delta U^{OG} \propto |\Delta U(\lambda_{21}) + \Delta U'(\Delta m = \pm 1, \pm 2)|$, where the dominating energy term $\Delta U(\lambda_{21})$ describes the conventional OG efficiency of the light-induced population transfer. The second term $\Delta U'(\Delta m)$ describes the galvanic contribution of the coherent magnetic sub-states.

The different values of $\Delta U^{OG}$ for linearly- and circularly-polarized irradiating light reveal a dependence of the cross-sections of excitation and ionization on the type of coherence $\Delta m = \pm 1, \pm 2$ of the degenerate state.

Figure 2. Magnetic-field-induced galvanic resonance $\partial U(B)/\partial B$ in a Ne/As HCD lamp (a), amplitude of the OG signals $\Delta U^{OG}$ in Ne/Li (b) and Ne/Cd (c) HCD lamps vs. the irradiating laser power in linear/circularly polarized light.
4. Conclusions

The galvanic signals $\partial U(B)/\partial B$ and $\Delta U^{OG}(P)$ measured are manifestations of both different type and degree of ordering of the atomic magnetic state $m$. The signals reveal implicitly different rates of ionization in the ensemble of atoms self-aligned ($\Delta m = 0, \pm 2$) → nonaligned and aligned ↔ oriented ($\Delta m = \pm 1$).

The magnetic disordering of the self-alignment induces additional conductivity due to the contribution of all self-aligned levels. This effect may be of opposite signs.

Two axes of self-alignment are found in the Ne/Cu HCD lamp.

Since the self-alignment is an a priori coherence, any real gas discharge medium generates the corresponding additional conductivity. The latter is sensitive to a weak magnetic field that destroys the coherence. Maximal $\partial U(B)/\partial B$ signals are observed in vicinity of the inflection $I/V$ point.

The difference in the amplitude distinguishes the signals $\Delta U^{OG}(P)$ as arising from aligned ↔ oriented ensemble of atoms having different contribution to the conductivity.

The different galvanic signals measured in both self-aligned → nonaligned and aligned ↔ oriented ensembles of atoms are manifestation of the galvanic contribution of the coherent degenerate Zeeman sublevels in a gas discharge.

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