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Optogalvanic effects in a hollow cathode glow discharge plasma

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The hollow cathode glow discharge is a very suitable source to measure the optogalvanic effect (OGE). A detailed study of this effect in neon has been made using a tunable, single-frequency dye laser. It was found that the lower level of the irradiated transition together with the plasma conditions determine the sign of the effect. Three situations occur: the lower level can either be metastable, resonant, or highly excited. With a five-level model taking into account electronic collisions and radiative decay a qualitative description of all the cases found in the experiment can be given. Further, it is shown that from a frequency scan of the OGE, line profiles can be obtained which, under certain conditions, contain the gas temperature in their Doppler part. Temperatures ranging from 700 to 1300 K determined this way are consistent with other results. This method makes it possible to measure the temperature as a function of the position, showing that inside the glow the gas temperature is constant. Finally, the time-dependent OGE is measured and analyzed in terms of ambipolar diffusion. This way an average transport temperature for the plasma electrons in the hollow cathode discharge in neon between 0.2 and 1.2 eV is found.

I. INTRODUCTION

When a discharge plasma absorbs radiation, the electrical circuit parameters may vary. This phenomenon is called the optogalvanic effect (OGE) and has been reported by Penning in 1928, who discovered that an increase in breakdown voltage occurs in a mixture of neon and argon when discharge tubes are irradiated by the emission of an identical discharge.¹ The development of tunable dye laser systems made it possible to study optogalvanic effects under much better defined conditions, and this has stimulated an increase in OGE studies of a fundamental as well as of a more applied nature.

The first applications were in the field of element analysis. It is possible to detect resonant transitions of spurious elements in flames or discharges by monitoring the discharge voltage as a function of the wavelength of the radiation.^{2,3} Also, wavelength calibration is possible.⁴ Hyperfine structures have been obtained by using Doppler-free two-photon⁵ or intermodulation techniques.^{6,7} Recently OGE profiles have been compared with line profiles obtained by absorption spectroscopy^{8,9} and the question arises whether OGE profiles can be used for diagnostic studies of discharge plasmas in the same way as profiles from optical spectroscopy. This is one of the subjects this paper deals with.

Therefore, an experimental study of the OGE has been carried out in a hollow cathode glow discharge in neon with a copper cathode. In Sec. II the experimental setup is described. To interpret our results we develop in Sec. III a description of the optogalvanic effect. Based upon this model it is possible to explain the positive as well as the negative effect, as we show in Sec. IV A. Here also some pathological cases are reported. In Sec. IV B we show that from the width of measured OGE profiles, discharge temperatures can be obtained. This is confirmed by comparison of OGE measurements with line profiles of spontaneous emission obtained through Fabry-Perot interferometry. Finally, in Sec. IV C we report measurements of the time dependency of the

OGE and give an interpretation of these measurements yielding a value of the average energy of the plasma electrons for various values of the discharge parameters.

II. EXPERIMENTAL SETUP

We studied the plasma of a hollow cathode glow discharge in neon with a cathode made of copper. The dimensions of the inner bore of the water-cooled cathode were a length of 50 mm and an inner diameter of 7 mm. The tube was prepared by the proper vacuum technique and was usually operated at pressures between 260 and 1040 Pa. Currents were between 1 mA and 1 A. Special care was taken that the discharge took place within the cathode only. Annular anodes were positioned at both sides of the cathode so that the direct discharge between cathode and anode was obstructed. The discharge vessel was made out of stainless steel (see Fig. 1).

The cylindrical hollow cathode discharge volume was observed in an end-on fashion. The optical axis of the detection system was moved parallel to the symmetry axis of the discharge. For passive spectroscopy a spatial resolution of 0.2 mm was reached. The resolution of active diagnostics was determined by the spatial quality of the laser beam. Here values of 0.3 mm were obtained.⁹

Using several spectroscopic techniques we determined gas temperatures, excited state densities, copper ground state densities, and electron densities. This data has been published elsewhere,¹⁰ but were used in this paper when needed.

For the measurements of the optogalvanic effect we used a single frequency ring dye laser (Spectra Physics 370 D) that was actively stabilized both in frequency and intensity. The wavelength range of the system was determined by the use of Rhodamine 6G. The frequency scan yielding the OGE profiles was calibrated with a bifocal étalon with a free spectral range of 144.86 MHz. The power of the laser system at the position of the discharge could be varied between

FIG. 1. Hollow cathode discharge tube. 1: cathode; 2: anodes; 3: quartz viewing ports; 4: quartz insulating tubes; 5: cooling water.

0.001 and 200 mW. For the time-dependent studies the laser beam was modulated by an optoelectric modulator with a rise time of 25 ns. The variations in the discharge voltage caused by the modulation of the incident radiation were coupled out at the anode through a capacitor of 50 nF. The cathode was at ground potential.

III. THEORY

A problem in interpreting the OGE is that in constructing a model a solid description of the discharge studied should be available. Some authors study the effect in a positive column,¹¹ but mainly because of its applicability most of them use a hollow cathode glow discharge. The parameters of this discharge have not been measured consistently, so in order to give our model a more solid base we decided to study the OGE in a hollow cathode glow discharge (HCG), determining the relevant parameters at the same time.¹⁰ In this way we were able to evaluate the contribution of the various ionization processes to the enhancement of the ionization caused by the absorption of radiation.

A striking feature of the OGE is that the sign of the effect can be positive or negative. This depends on the position of the irradiated discharge volume, the discharge current, the atomic transition, and when working with high resolution, even on the frequency of the laser radiation. The explanation suggested is that ionization through metastable-metastable collisions is dominating at low-discharge currents. By optical pumping from metastable levels an overall loss of these metastables occurs, resulting in an increase of the discharge voltage, i.e., a positive OGE. Some authors¹⁴ explain the negative OGE by saying that the absorption of laser radiation increases the electron temperature resulting in an enhancement of the conductivity of the plasma. It is clear that these two ideas cannot easily be brought into one model capable of predicting the sign of the OGE. Most authors, however, agree on the basic notion that small changes in the discharge voltage ΔV_b may be considered as proportional to the change in local population of excited states Δn_i as generated by the absorption of radiation through

$$\Delta V_D = \sum \alpha_i \Delta n_i, \quad (3.1)$$

where the parameters,

$$\alpha_i = \left(\frac{\partial V}{\partial n_i} \right), \quad (i = 1, 2, 3, \dots), \quad (3.2)$$

are proportionality constants. This concept is based on a variation in density of each particular excited state within the discharge volume. It should be clear that it requires a

major effort to evaluate the constants α_i for each particular case.

However, for the purpose of interpreting the experiments reported in this paper we formulate a model of the HCG in simplifying terms. The simplifications are partly based on the cylindrical symmetry of the HCG. Also we may assume that the discharge can be considered as a central large-sized plasma with high conductivity and a cathode fall region acting as an ion collecting sheath. Corresponding electrons are collected at the anode side of the plasma. Extra ionization brought about locally in the negative glow plasma will eventually be observed as extra ion current through the cathode sheath. This current change will cause a change in discharge potential,

$$\Delta V_D \sim D_a \left. \frac{d}{dr} \right|_{r=r_0} \Delta n(r, t), \quad (3.3)$$

where D_a is the ambipolar diffusion coefficient and the gradient is taken at the plasma boundary r_0 . The change in plasma density $\Delta n(r, t)$ is the solution of the equation

$$D_a \nabla^2 [\Delta n(r, t)] = S(r, t) + \frac{\partial}{\partial t} [\Delta n(r, t)]. \quad (3.4)$$

Here volume recombination of the plasma has been neglected¹⁰ and the source term solely represents the source of extra ions from the optogalvanic effect. The source S can be expressed in local disturbances $\Delta n_i(r, t)$ of excited states i caused by absorption of laser radiation through

$$S(r, t) = \sum_i k_i^c n(r, t) \Delta n_i(r, t), \quad (3.5)$$

where k_i^c is the cumulative ionization rate from the excited level i . Assuming that replacing $n(r, t)$ by the equilibrium distribution $n_0(r)$ with the laser off gives only second-order effects, the source term becomes

$$n_0(r) \sum_i k_i^c \Delta n_i(r, t). \quad (3.6)$$

The local disturbances $\Delta n_i(r, t)$ of the excited state densities may be evaluated independently from a collisional radiative model of the plasma considered.

A general expression for the OGE may now be written as

$$\Delta V_D = c_1 D_a \left. \frac{d}{dr} \right|_{r=r_0} \left[\sum_i k_i^c \int_{\rho_0}^{\rho} n_0(\rho) \Delta n_i(\rho, \tau) \times G(r, \rho/t, \tau) d\rho d\tau \right], \quad (3.7)$$

where c_1 is a constant and $G(r, \rho/t, \tau)$ is the Green's function satisfying Eq. (3.4) with the boundary conditions

$$G(r_0, \rho/t, \tau) = G(r_0, \rho/0, \tau) = 0. \quad (3.8)$$

Under our experimental conditions we can also show that $\Delta n_i(r, t)$ may be expressed as

$$[\Delta n_{i,f} \delta(t) + \Delta n_{i,0} U(t)] \delta(r), \quad (3.9)$$

that is, the increase in population of level i due to absorption of laser radiation is concentrated on the axis of the discharge. The time dependency can be considered to be the sum of a fast stimulus $\Delta n_{i,f}$ active at $t = 0$ on a much smaller time scale than the ambipolar diffusion time and a constant stim-

FIG. 2. OGE as a function of the current for three wavelengths. The transitions have different lower levels: 588.2 nm (metastable), 603.0 nm (resonant), and 576.4 nm (highly excited). The pressure was 650 Pa. The current is plotted on a logarithmic scale.

ulus $\Delta n_{i,0}$ acting from $t = 0$ and onwards, representing the steady state value of the increase in population of level i .

Now the OGE signal turns into

$$\Delta V_D = c_1 D_a \left. \frac{d}{dr} \right|_{r=r_0} \left[n_0(0) \sum_i k_i \Delta n_{i,f} G(r,0/t,0) + n_0(0) \sum_i k_i \Delta n_{i,0} \int_0^t G(r,0/t,\tau) d\tau \right]. \quad (3.10)$$

From this expression we see that the value of the OGE is linear in both $\Delta n_{i,f}$ and $\Delta n_{i,0}$. For t to infinity a steady-state value of the OGE will result; in that case the first term goes to zero.

FIG. 3. (a) Diagram of the five-level model to explain the qualitative aspects of the OGE. 1 is a metastable level, 2 can be either resonant or metastable or highly excited, and 3 is a highly excited level. (b) Simplified energy diagram of neon showing the transitions used in the experiments. M_1 and M_2 are the $3s(3/2)_2$ and the $3s'(1/2)_1$ metastable levels, R_1 and R_2 the $3s(3/2)_1$ and the $3s'(1/2)_0$ resonant level. H_1 and H_2 are the $3p$ and $4d$ highly excited levels. The wavelengths of the transitions are: 1: 588.2 nm; 2: 603.0 nm; 3: 616.3 nm; 4: 585.2 nm; 5: 576.4 nm.

For our calculations we have taken¹⁵

$$G(r,0/t,\tau) = \frac{1}{\pi r_0^2} \sum_{n=1}^{\infty} \frac{J_0(r\alpha_n)}{J_1(r_0\alpha_n)} \exp[-D_a \alpha_n^2(t-\tau)], \quad (3.11)$$

which represents the response to a unit instantaneous line source at the axis of a cylindrical configuration. Here ($n = 1, 2, 3, \dots$) are the roots of $J_0(r_0\alpha)$.

IV. EXPERIMENTAL RESULTS

A. Sign of the OGE

1. The OGE as a function of current

In Fig. 2 experimental results for the steady-state OGE as a function of discharge current for various absorbed NeI lines are given. The observed transitions are indicated in Fig. 3(b). It can be seen that for the 576.4-nm line the effect first increases with current and after reaching a maximum decreases. The increase can be explained by the increase in density of the higher lying states together with the enhancement of the cumulative ionization of the upper state. A further increase of the current and consequently the electron density leads to collisional electronic deexcitation compensating the resonant pumping and the OGE decreases to zero. Lines starting from either a metastable state (588.2 nm) or a resonant level (603.0 nm) show a different behavior. The effect from the 588.2-nm line is first positive, becomes negative at 200 mA, and will eventually become zero at higher currents. The 603.0-nm effect starts negative, successively becoming positive and negative before decreasing to zero.

From Fig. 3(b) it can be seen that the 588.2-nm line has the $3s(3/2)_2$ metastable state of neon as a lower level, whereas the 603.0-nm line has the $3s(3/2)_1$ resonant state as its lower level. Analysis of other NeI transitions shows that the positive OGE is an exception. Most current explanations in literature for the positive effect are that metastable-metastable ionization is dominating at these low discharge currents and that the optical pumping destroys metastables decreasing the total ionization rate. In our discharge we measured the metastable densities to be about 10^{18} m^{-3} , the electron densities to be from 1×10^{19} up to $9 \times 10^{19} \text{ m}^{-3}$, and the electron temperature to be about 1 eV.^{9,10} Taking cross sections for metastable-metastable and electron-metastable ionization of 100 and $6 \times 10^{-20} \text{ m}^2$, respectively,^{12,13} we find that at currents of about 100 mA and pressures of 260 Pa metastable-metastable ionization is only about 1% of the electron-metastable ionization. So to explain the positive OGE the cumulative electronic processes need to be taken into account. Here we should keep in mind that the excitation of ground state atoms is mainly by fast electrons entering the glow from the cathode fall.¹⁰ The observed features as shown in Fig. 2 can be explained in more detail by a five-level model.

Figure 3(a) shows the five levels used in the model, 0 is the ground state, + is the ion state, 1 is a metastable level, 2 is a resonant, a metastable or a highly excited level, and 3 is a highly excited level. In this model the laser coupling is always between 2 and 3. The transitions that have been studied in the experiment are indicated in the partial energy diagram of neon as shown in Fig. 3(b). These irradiated transitions are

either between a $3s$ and a $3p$ level or between a $3p$ and $4d$ level. The $3s(3/2)_2$ and the $3s'(1/2)_1$ are metastable, the $3s(3/2)_1$ and the $3s'(1/2)_0$ are resonant. For the five-level model of Fig. 3(a) the rate equations can be written down after defining the production rate of states in level i by excitation from the ground state as P_i and the total deexcitation rate of that level by k_i . The laser coupling rate is represented by L . The electronic coupling rate between level 1 and level 2, which are energetically close, is represented by k_{12} . The part of k_i leading to cumulative ionization is k_i^c . The rate equations for the levels are

$$\frac{dn_3}{dt} = L_{23}n_2 - L_{32}n_3 - k_3n_3 + P_3, \quad (4.1)$$

$$\frac{dn_2}{dt} = L_{32}n_3 - L_{23}n_1 - k_2n_2 + P_2 + k_{12}n_1, \quad (4.2)$$

$$\frac{dn_1}{dt} = -k_1n_1 + P_1 + k_{21}n_2. \quad (4.3)$$

Here $g_2L_{23} = g_3L_{32}$ and $g_1k_{12} = g_2k_{21}$, where g_i is the statistical weight of level and it is assumed that the energy difference between 1 and 2 is much smaller than the thermal electron energy. Equations (4.1)–(4.3) can be solved for the stationary state giving a change in the densities Δn_i^0 compared to the situation with the laser off. The OGE will, in first order, depend linearly on these changes, so Eq. (3.1) can here be rewritten as

$$\Delta V_D = -c_2 \sum_{i=1}^3 k_i^c \Delta n_i^0, \quad (4.4)$$

where c_2 is a constant. When substituting the solutions for Δn_i^0 in Eq. (4.4) a general expression for the OGE results, which can be used to discuss the measured effects of Fig. 2.

The first case to be considered is when the electronic coupling between 1 and 2 is negligible ($k_{12} = k_{21} = 0$). Then the system is reduced to the four levels 0, 2, 3, and 4. For the steady state, Eqs. (4.1) and (4.2) can now be solved with $k_{12} = k_{21} = 0$. Substituting this result in Eq. (4.4) we obtain

$$\Delta V_D = -c_2(k_3^c k_2 - k_2^c k_3) \frac{g_3 n_2^0 - g_2 n_3^0}{g_3 k_3 + g_2 k_2}, \quad (4.5)$$

where n_i^0 is the level density with the laser off. Under the conditions in our discharge it is evident that $g_3 n_2^0 > g_2 n_3^0$, so the sign of the OGE depends on the sign of the term $k_3^c k_2 - k_2^c k_3$. In this case, where an isolated lower level is considered, two possibilities occur: the state is radiatively coupled to the ground state or it is not. If there is radiative coupling we may assume from general arguments that the ionization ratio k_3^c/k_2^c is always larger than the deexcitation ratio k_3/k_2 . In those cases the factor $k_3^c k_2 - k_2^c k_3$ is positive and the OGE negative. In the experiment these conditions are met if the lower level of the irradiated transition is highly excited, i.e., the $3p$ level of Fig. 3(b). In Fig. 2 this is the case for the 576.4-nm line which shows a negative OGE for all currents. But also the 603.0-nm transition from the resonant $3s(3/2)_1$ level fulfills this condition when the electronic coupling with the metastables is low, i.e., at low currents. This is observed for the 603.0-nm line in Fig. 2 at currents below 5 mA.

In this four-level model it is also possible that the lower level is metastable. Then the radiative coupling at low currents is nonexistent. Therefore, the density becomes very high compared to other excited states and the cumulative ionization from the metastable state becomes relatively important. When the laser radiation depopulates this level the density is lowered dramatically and the number of ionizations decreases strongly. The $3p$ states which are created by absorption more readily decay to the ground state instead of being ionized, due to the low electron density, so the somewhat enhanced ionization from the $3p$ level does not compensate for the loss of ionizations from the metastable level. This loss of ionizations leads to the positive effect as measured for the 588.2-nm line in Fig. 2 at currents below 200 mA. This argument is in effect the same as saying that k_2 is very small for a metastable, so the term $k_3^c k_2 - k_2^c k_3$ becomes negative and the OGE positive.

At high currents the metastable level will be deexcited by electron collisions. This means that k_2 is raised with the electron density and above a certain value the term $k_3^c k_2 - k_2^c k_3$ will become positive, as was the case when the lower level of the irradiated transition was highly excited. Then the OGE turns to negative. This is observed in Fig. 2 for the 588.2-nm line at currents above 200 mA. One can say that above this current the metastable character of the $3s(3/2)_2$ state is lost.

To explain the effect of the 603.0-nm line at intermediate currents we must return to the five-level model. The only case where the five-level model gives different results compared to the four-level model is when the lower level of the pumped transition is resonant and there is a metastable level at a small energy difference. At very low currents this level will act as a highly excited level as described above. When the current is raised, coupling with other states occurs. Because the metastable is very near, the coupling with this level will be by far the strongest. This coupling then gives a deexcitation of the metastable state when the resonant state is pumped, and less ionization from the metastable state is the result. If this loss is larger than the extra ionizations from the $3p$ level, positive OGE will occur. This positive effect is seen for the 603.0-nm in Fig. 2 at currents between 5 and 50 mA. Under these circumstances the resonant $3s(3/2)_1$ level behaves like a metastable.

It is worthwhile to note that the OGE of the 585.2-nm line ending on the $3s'(1/2)_0$ resonant level always gives a negative OGE with almost the same shape as the 576.4-nm line of Fig. 2. Here apparently the coupling with the metastable states is much less, as is explained from the larger energy difference between the levels as well as from the difference in spin of the states involved.

At high currents the OGE for this line becomes negative also. Here the argument of the disappearing metastable character can again be used. At extremely high currents the OGE will always go to zero. There the laser radiation has a negligible influence with respect to the electronic coupling.

The conclusions of this section are that the OGE consists out of two major contributions: increase of ionization from the upper level which gives a negative effect and decrease of ionization from the lower level which leads to a

FIG. 4. OGE as a function of the radial position for three currents at a wavelength of 576.4 nm and a pressure of 650 Pa. The lower level of the transition is highly excited.

positive contribution. Which of these dominates depends on the discharge parameters before the laser pumping. All ionizations are performed by electronic collisions.

2. The OGE as a function of the position

The changing of the sign of the OGE is observed not only as a function of the current but also as a function of the radial position within the discharge. We reported this previously¹⁶ and it was also found by Zalewski¹⁷ and Bachor.⁸ In Fig. 4 the OGE of the 576.4-nm transition is given; in Fig. 5 the OGE of the 588.2-nm line is shown. In the glow the behavior is as expected from our description in Sec. IV A 1. In the cathode dark space, the effect is enhanced. This may be due to an extra electron originating in the dark space creating more than one electron in the glow. When the lower level is metastable, as in the case of the 588.2-nm transition shown in Fig. 5, it can be seen that the effect is positive everywhere at lower currents, but at higher currents both positive and negative effects occur. In the negative glow the strong electronic interactions cause a negative effect becoming positive at the edges where the electron density decreases. The reason of the negative effect at 0.4 A which occurs when irradiating near the cathode surface cannot be understood from our model, and has to be classified as a pathological effect.

FIG. 5. OGE as a function of the radial position for three currents at a wavelength of 588.2 nm and a pressure of 650 Pa. The lower level of the transition is metastable.

3. The OGE as a function of the frequency

The most striking change of the sign of the OGE is seen in Fig. 6.

This gives the OGE of the 588.2-nm transition measured at 0.3 mm from the cathode as a function of the frequency of the laser. This position corresponds with the place where the OGE becomes zero for a current of 200 mA in Fig. 5. The measurement shows that the OGE is positive in the line wings and negative in the center. The effect of the Ne isotope is also positive. This means that the sign of the term $k_3 k_2 - k_2 k_3$ changes as a function of the frequency. The factors k_3 , k_2 , and k_2 are governed by electron collisions so they will be independent of the frequency. But k_3 contains the radiative decay to the ground state. Because resonance radiation is less imprisoned in the line wings, a positive OGE can be expected there. Also radiation from the ²²Ne isotope is less imprisoned and will contribute to a positive effect.

It should however be emphasized that measurements of the OGE as a function of the frequency well within the negative glow plasma when using transitions between highly excited states give line profiles of a regular appearance. These will be analyzed in the next paragraph.

B. OGE line profiles and gas temperatures

In plasma diagnostics it is a standard procedure to obtain gas temperatures from the Doppler broadening of an emission profile. Johnston studied OGE profiles, but found line widths 20% to 50% larger than those to be expected from Doppler broadening.¹⁸ To see whether it is possible to determine line width and gas temperature from the OGE, we compared OGE profiles with emission profiles obtained with a Fabry-Perot interferometer. Starting from the notion that the OGE will be proportional to the change in level density of excited states involved in the ionization, we limit ourselves to a treatment with two excited states, a lower state 1 and a higher state 2. The laser frequency is within the absorption profile of the transition 1-2.

From the density matrix formalism¹⁹ it is known that for a certain frequency ω the difference in probabilities to find the state either in 1 or 2 is

$$\rho_{11} - \rho_{22} = (\rho_{11}^0 - \rho_{22}^0) \frac{1 + (\omega - \omega_0)^2}{1 + (\omega - \omega_0)^2 \Gamma^2 + I/I_s}, \quad (4.6)$$

where $\rho_{11}^0 - \rho_{22}^0$ is the difference when the laser intensity is zero, ω_0 the central frequency of the transition, I_s the saturation

FIG. 6. OGE as a function of the laser frequency. The radial position of the laser beam was 0.3 mm from the cathode. The pressure was 420 Pa and the laser wavelength 588.2 nm.

FIG. 7. OGE as a function of the laser power for three currents at 260 Pa. Line profiles have been obtained at a laser power of approximately 50 mW. The wavelength was 576.4 nm.

tion intensity, and Γ the decay rate of the off-diagonal elements. To calculate the OGE the actual densities $\Delta n_1 = \rho_{11} - \rho_{11}^0$ and $\Delta n_2 = \rho_{22} - \rho_{22}^0$ can be evaluated under steady-state conditions. It follows that

$$\Delta n_1 = \frac{Ak_2(\rho_{22}^0 - \rho_{11}^0)}{A(k_1 + k_2) + k_1k_2}, \quad (4.7)$$

and

$$\Delta n_2 = \frac{Ak_1(\rho_{22}^0 - \rho_{11}^0)}{A(k_1 + k_2) + k_1k_2}, \quad (4.8)$$

where

$$A = \frac{\Gamma I / I_s}{\Gamma^2 r(\omega - \omega_0)^2}, \quad (4.9)$$

and k_1 and k_2 are the decay rates of level 1 and 2, respectively. Consequently, the OGE will now be proportional to

$$\Delta V_D \sim \frac{(\alpha_1 k_2 + \alpha_2 k_1)(\rho_{22}^0 - \rho_{11}^0)}{k_1 + k_2 + k_1k_2/A}, \quad (4.10)$$

where α_1 and α_2 are proportionality constants relating the level density changes to the OGE. Combining Eqs. (4.10) and (4.9) we see that the OGE profile is a Lorentzian with a half-width

$$\Delta\omega = \Gamma \sqrt{1 + \Gamma k_1 k_2 / I_s (k_1 + k_2)}, \quad (4.11)$$

which is identical to the width of an absorption profile showing saturation broadening.¹⁹ From Eq. (4.11) it can also be seen that for $I < I_s$ the OGE will be linear with the laser power, whereas for $I > I_s$ the effect will saturate.

When dealing with absorbers having a certain velocity distribution the OGE will be the convolution of the Lorentzian derived above and the velocity distribution specified. In

FIG. 8. OGE as a function of the laser frequency in the center of the glow at 576.4 nm, pressure 260 Pa, current 200 mA. The lower trace is five times the difference between the measured profile and a least-squares Voigt fit.

FIG. 9. Temperature as a function of the current obtained with emission spectroscopy and OGE. $\triangle$: Fabry-Perot at 540.0 nm; $\circ$: OGE at 576.4 nm; $\bullet$: OGE at 585.2 nm. The experimental error for both methods is 50 K, the pressure was 640 Pa. The 585.2-nm line was strongly absorbed.

the case of a Maxwell distribution the OGE should clearly give a Voigt response as a resulting line shape if several conditions are fulfilled.

From these considerations we can derive conditions on how to measure temperatures of atomic absorbers with the OGE. The first thing to be considered is the dependency of the laser intensity. In Fig. 7 the results are shown of the OGE as a function of the laser beam intensity I . The line used was the 576.4-nm $3p$ - $4d$ transition. When the OGE is linear with I , no saturation occurs and a correct profile can be obtained. In practice some other things should be kept in mind. When measuring the effect with lock-in techniques, the chopping frequency should be low in order to obtain a real steady-state effect. Second, the lower level should preferably not be metastable or a resonant near a metastable because than effects as described in Sec. IV A will obscure the results. An important condition is also that the laser beam should be only weakly absorbed by the discharge because otherwise the laser intensity will not be homogeneous along the irradiated volume.

For the measurement of the OGE profile we used a $3p$ level of neon (576.4 nm). The laser power was about 50 mW, which was enough to get a good signal to noise ratio. The absorption was never more than 5% at the line center. A typical profile is shown in Fig. 8.

The lower trace is five times the residue after fitting the measurement to the expression for the Voigt profile. The effect of the Ne isotope was taken into account. In Fig. 9 the results for the temperature as a function of the discharge current are given at a filling pressure of 650 Pa. Also indicat-

FIG. 10. Temperature as a function of the radial position inside the glow obtained with the OGE at 576.4 nm for three currents at a pressure of 640 Pa.

ed are results obtained from Fabry-Perot measurements of spontaneous emission from the 540.0-nm line. The agreement is, within the experimental error of 100 K, good. In contrast to Fabry-Perot experiment the temperature measurement with the OGE offers the possibility to measure the temperature in the glow as a function of the radial position. For various discharge currents results of this experiment are shown in Fig. 10. Another advantage of the OGE method is that the line width of the single-frequency dye laser is negligibly small so there is no need for a correction of an apparatus profile. The temperature within the glow appears to be nearly constant. This underlines the fact that the electric field in the glow is very small and that elastic loss of energy by either electrons or ions within the glow is nonexistent. The beam electrons indeed lose their energy only by inelastic collisions. The source of heat is located in the cathode fall. Here the energy loss of accelerated ions entering the sheath is the main source of translational energy.

C. Time-dependent optogalvanic studies

The time dependence of the optogalvanic effect has been the subject of several studies. Experiments were carried out either by investigating the response to a light pulse of a dye laser pumped by a pulsed nitrogen laser²⁰ or by using a cw dye laser with a mechanical chopper.¹⁹ We used an electro-optical modulator with a risetime of 25 ns and measured the development of the OGE after the laser beam had been switched on. Repetition frequencies were between 1 and 10 kHz, so the effect could well be observed until a steady state had been reached. A typical response can be seen in Fig. 11. It was recorded with a boxcar integrator with a gate width of 1 μ sec. Here the response to the stepwise laser stimulus at 576.4 nm has been recorded. The pressure was 650 Pa and the current 100 mA. The result should be analyzed with help of the general expression (3.10) for the OGE in a cylindrical geometry.

In our interpretation we limit ourselves to the case where only cumulative ionization from the two levels involved takes place. Because the levels in the experiment are highly excited states complications arising from coupling with the metastable levels are avoided. The scheme in Fig. 12 gives the relevant levels. The production rates of levels 1 and 2 and the direct ionization rate are P_1 , P_2 , and P_+ , respectively. The total deexcitation rates of 1 and 2 are k_1 and k_2 , respectively. During the optical pumping, with pumping rate L the levels 1 and 2 are solidly coupled on a time scale of

FIG. 11. Measurement of the time dependence of the OGE of the 576.4-nm line with the laser frequency at the center of the transition. Pressure 650 Pa, current 100 mA.

FIG. 12. Diagram of the four-level model used in the analysis of the time-dependent OGE.

the inverse Rabi frequency. For the laser power we used, this amounts to about 10 ns. Because generally speaking the decay of 2 will be larger than that of 1 the levels both will go to a steady-state value $n_{1,s} = n_{2,s}$ related to the initial values $n_{1,0}, n_{2,0}$ through

$$n_{1,s} = n_{2,s} = \frac{k_1 n_{1,0} + k_2 n_{2,0}}{k_1 + k_2}. \quad (4.12)$$

This steady state will be reached on a time scale $(k_1 + k_2)^{-1}$. The decay rates k_1 and k_2 are governed by either radiative transitions or electronic deexcitations. Cumulative ionization is also included in the latter process. Radiative deexcitation of a 4d level may amount to 40 ns. Electronic deexcitation also will be fast; e.g., the cumulative ionization rate k_2^c of level 2 will be larger than 10^6 s^{-1} for an electron density of 10^{19} m^{-3} , an average electron temperature of 1 eV and an ionization cross section at that energy of 10^{-19} m^2 . So the time scales governing the extra ionization are at least one order of magnitude shorter than the time scale of the observed OGE. In practical cases both level 1 and 2 will be coupled to other excited states whose populations will also change under the influence of the laser radiation. But the time dependence of these effects will not be different from those of levels 1 and 2.

On the time scale where the maximum of the OGE occurs the source of extra ionization may be considered to be the sum of a delta stimulus and a step. This concept has been incorporated in expression (3.8) for the time dependence of an excited level contributing to the OGE. Using Eqs. (3.9) and (3.10) the OGE can now be described by

$$\Delta V_D = \sum_{n=1}^{\infty} \frac{z_n^0}{J_1(z_n^0)} \{ A_1 \exp(-z_n^0 t / t_D) + A_2 [1 - \exp(-z_n^0 t / t_D)] / z_n^0 \}, \quad (4.13)$$

FIG. 13. Comparison of the calculated and the measured time dependence of the OGE; conditions as in Fig. 11.

FIG. 14. Average electron transport temperatures derived from the time-dependent OGE of the 576.4-nm line as a function of current and pressure. O: 260 Pa; X: 650 Pa; ●: 1040 Pa.

where A_1 is proportional with the number of extra ionizations in the peak and A_2 is proportional to the number of extra ionizations in a time interval $t_D = r_0^2/D_a$. The factors z_n^0 are the zeros of the Bessel function J_0 , whereas J_1 denotes the Bessel function of the first order. The summation has been calculated as a function of the reduced time $\tau = t/t_D$ with the ratio A_2/A_1 as a parameter. This ratio defines the position of the maximum in the response on the α axis as well as the ratio M/S of the maximum response and the stationary value of the response.

Experimental values are now fitted to the calculated response curves by analyzing the experimental values in terms of M/S . From the position of the maximum both in the experimental and calculated curve a value of t_D and henceforth a value of the ambipolar diffusion coefficient D_a can be obtained. In Fig. 13 a fit of the experiment to the calculated curve is shown. The relevant ratio of A_2/A_1 is 4.1.

The ambipolar diffusion coefficient is related to the electron temperature through

$$D_a = D_i(1 + T_e/T_g), \quad (4.14)$$

where D_i is the ion diffusion coefficient. When D_i and T_g are known, a value of T_e may be obtained. A main difficulty in this analysis is that the electron energy distribution function in the case of a hollow cathode glow plasma is non-Maxwellian. A distinction should be made between beam electrons, carrying the full cathode fall energy into the plasma and losing energy in inelastic collisions, and the slow plasma electrons which are essentially secondaries as well as slowed down fast electrons. The interaction between fast and slow electrons is very small also because the fraction of fast electrons is small, but a strong interaction exists between slow electrons. This results in a quasi-Maxwellian distribution of slow electrons, governing the transport properties of the plasma. The measured value of T_e is the average kinetic energy of this distribution of slow electrons and may be called transport temperature. It should be noted that this temperature may be very useful in estimating, e.g., the recombination in the negative glow or the collisional rearrangement of excited levels. A final remark that should be made is that under our experimental conditions charge transfer of neon ions to copper atoms occurs. The diffusion coefficient of copper ions in neon is about twice that of neon ions. However, the value of the ion densities in the glow as obtained in another analysis¹⁰ of our experimental setup yield a ratio of copper to neon ions of at most 0.1 in the observed current range. Therefore,

the value of the diffusion coefficient of neon,²² with a correction for temperature effects²³ was used in our calculations of T_e . The results are shown in Fig. 14.

Here the temperature T_e is displayed as a function of discharge current with the pressure as parameter. The increase with pressure of T_e is not in accordance with results of, e.g., the Schottky theory for the positive column. However one should realize that the ionization mechanism in a HCG is quite different from that in a positive column. Taking into account that the discharge voltage decreases about 15% when the pressure changes from 260 to 1040 Pa, a drop in ionization by the beam electrons may have to be compensated by an increase of the average kinetic energy of the bulk of the electrons.

V. CONCLUSIONS

We have performed measurements and calculations on the optogalvanic effect in a hollow cathode glow discharge. In a simplified model it is possible to relate the variation in the population densities of the various excited states to a change in the discharge voltage. It is shown that under certain conditions the OGE is proportional to the population density variations within the irradiated volume. Then with a simple five-level model the qualitative aspects of the OGE can be explained in terms of discharge current, position, and frequency of the laser beam. In the model only electron-atom collisions and radiative decay have been taken into account. It shows that metastable-metastable ionization is not necessarily the process that causes the positive OGE. In a HCG under the conditions we used, this process is negligible. The positive OGE should be explained by the loss of ionization from the metastable level because the metastable state is depleted almost completely by the laser radiation. At higher currents where the electronic transitions start to dominate radiative transitions the depletion is less strong and the extra ionization from the upper level will turn the OGE sign into negative. At still higher currents the pumping effects will be obscured by the strong electronic coupling and the OGE will go to zero. In general it can be stated that the sign of the OGE depends on the character of the lower level and the plasma conditions before switching on the laser.

Pumping of the $3s(3/2)_1$ level of neon, which lies in-between the metastable levels, shows the influence of the coupling of these $3s$ states by electronic collisions. Here at low currents the OGE is similar to that occurring in transitions between highly excited states. Increasing the current first makes the OGE positive and then negative again. This is due to the electronic coupling between the $3s$ states. The $3s'(1/2)_0$ state is not strongly coupled to the metastable states, however, its OGE is always negative.

Both from the model and the experiment we have shown that the OGE profile, obtained by scanning an optical transition, matches the Voigt profile of the emission. So from the OGE profile a value of the gas temperature along the laser beam can be obtained by analysis of the Doppler part of the profile. Conditions for this procedure are: (1) the transition is between two highly excited states, (2) the laser beam is weakly absorbed, (3) the discharge is in a stationary state, and (4) the laser intensity used during the measurements has

to be in the domain where the OGE is linear with this intensity. Under these conditions we have shown that the OGE profiles of the 576.4-nm transition give the same temperatures as Fabry-Perot line profiles. The OGE method has the advantage that in a discharge like the HCG the temperature can be measured as a function of the position. Results show that in the glow the temperature is constant, indicating that the production of heat takes place in the cathode dark space and inelastic processes dominate the energy flow in the glow.

Finally we have studied the time dependency of the OGE. Transients in this effect occurring when switching the laser on or off can be explained in terms of ambipolar diffusion of the extra plasma created. By comparing the model with the experiment the ambipolar diffusion coefficient can be obtained. From this coefficient an average transport temperature of the plasma electrons has been derived.

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