Study of Excitation Rates in a Hollow Cathode Discharge

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In this work a dc discharge with a copper hollow cathode in argon used for deposition applications is investigated using atomic emission spectroscopy. Typical discharge parameters during our investigation are pressures between 80 and 200 Pa and current densities up to $7\,mA\,cm^{-2}$. The radial light intensity profiles of some selected copper and argon lines were measured. An analysis of these profiles gives insigths into the excitation rates of the argon filling gas and the sputtered metal atoms as well as into the radial dimension of the negative glow. The different excitation mechanisms lead to a sharp change of the radial light intensity profiles emitted by many copper spectral lines, which indicates the transition region between the cathode fall and the negative glow. The length of the cathode fall was observed to be very insensitive to all the macroscopic discharge parameters. A nearly constant value was found for the cathode fall length, which corresponds to about 22% of the cathode radius.

1 Introduction

The hollow cathode glow discharge is a good source of metal atoms and ions, which has been used in the processes of the thin film deposition [1] and as light source for spectroscopy and laser devices [2, 3]. The high energy of the secondary beam electrons, high current density and gas temperature make this discharge attractive for other applications in the field of plasma chemistry. Recently, discharges of microhollow cathodes operating at atmospheric pressure also enable their use in large-area plasma processing [4]. Because of the several technological applications, the hollow cathode configuration has been subject of intense research in the last years.

The spatial distribution of the sputtered metal atoms inside the cathode is particularly interesting for deposition applications. This can be obtained by laser-aided techniques [5, 6]. However, these techniques are usually difficult to be set up in many practical applications. Alternatively, atomic emission spectroscopy is a simple experimental technique that can provide useful information concerning the discharge. In this work a dc discharge with a copper hollow cathode in argon is investigated using atomic emission spectroscopy. The radial light intensity profiles of some selected copper and argon lines were measured and analyzed. A correlation between the emitted light intensities and the excitation rates of the argon filling gas and the sputtered copper atoms is proposed.

2 Experimental arrangement

The experimental arrangement is shown in Fig. 1. The discharge consists of a copper hollow cathode of 1 cm inner diameter and 7 cm length. The cathode was water cooled. Two cup-shaped anodes, also made of copper, were positioned on both extremities of the cathode. The effective area of the anodes is equal to the cathode area, each with $22\,cm^2$. The discharge was operated with 99.999% purity argon. The argon gas flow was measured with a flowmeter and regulated with a needle valve. The pressure was measured downstream with an absolute capacitance manometer and controlled by a throttle valve on the gas exhaust side. The HV-power supply with a load resistor of $1\,k\Omega$ was connected to both anodes. Current and voltage were measured, at the anode side, with a digital oscilloscope

For the measurements of the light intensity profiles, the cathode inner cross section was imaged into the entrance slit of the monochromator ($1180\,groove/mm$, $250\,mm$ focal length and $3.3\,nm/mm$ dispersion) with an object to image ratio of 0.5. To avoid stray light, due to reflections in the edge of the window, a fixed iris with $1.36\,cm$ diameter and $27\,cm$ away from the cathode was used, reducing, thus, the observation solid angle to $2 \times 10^{-3} sr$. An observation area of $0.1 \times 2.0\,mm$ was chosen, by adjusting the width and height of the monochromator slit to $500\,\mu m$ and $1\,mm$, respectively. The radial profiles of the light intensity were obtained by shifting the monochromator slit along the cathode image diameter in steps of $0.1\,mm$. In this way, the observation area was moved diametrally in steps of $0.2\,mm$, and the radial resolution, determined by the width of the monochromator slit, was $0.1\,mm$.

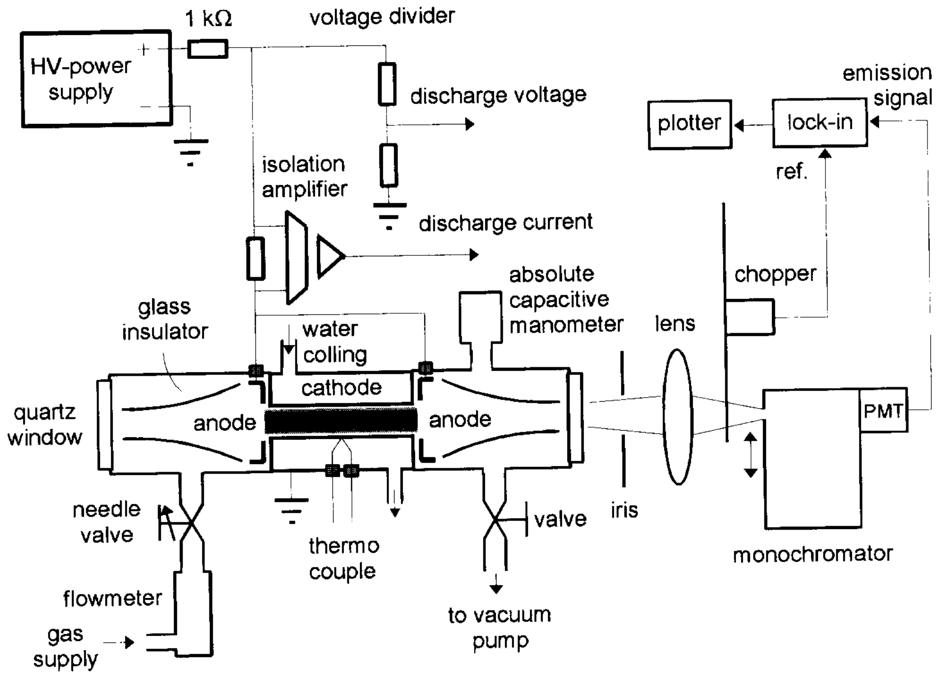

Figure 1. Schematic of the experimental set-up for emission spectroscopy measurements of light intensity profiles.

3 Results and discussion

3.1 The discharge characteristic

It has been observed that when a function of some discharge parameters is plotted against the applied voltage, the data points fall into the same curve. This curve is denominated the discharge characteristic and this function the invariant parameter [7]. The discharge characteristic curve is a simple way to characterize a gas discharge. The characteristic depends basically on the gas, the cathode material and geometry. For the hollow cathode discharge it has been observed that the function given by the product of the cathode length by the current density divided by the pressure, $l\,J/p$, is an invariant parameter, because it depends only on the voltage [8]. The discharge characteristic of our discharge is shown in Fig. 2. It was observed that for cathode wall temperatures $\leq 100\,^oC$ and gas flows $\leq 100\,sccm$ there is no alteration on the measured characteristic. There is a better overlapping of the experimental points in the low current range because the gas temperatures is close to the room temperature.

3.2 Light intensity emitted by noble gases

The light emission of a spectral line ($W\,m^{-3}\,sr^{-1}$) due to the spontaneous decay from a resonance state *n* to

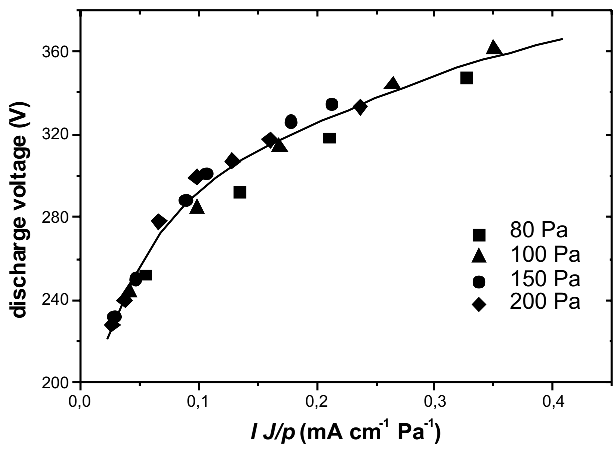

Figure 2. Characteristic curve of the Ar-Cu hollow cathode discharge.

lower state *m* is described by the emission coefficient [9],

$$\varepsilon(n \longrightarrow m) = \frac{h\nu}{4\pi} A\,(n \longrightarrow m)\,N_n\,, \qquad (1)$$

where N_n is the population density of the resonance upper state, $A(n \longrightarrow m)\,(s^{-1})$ is the transition probability, and $h\nu$ is the energy of the emitted photon. In the operation range of the discharge, the light emission can be described by the corona model [1, 10]. In this model, the atoms are excited from the ground state *g* to an upper state *n* by collisions with electrons and the upper state is depopulated by spontaneous radiative decay to lower states. In a steady state, the density of a resonance state N_n is related to the excitation rate $R_n(g \longrightarrow n)\,(m^{-3}s^{-1})$ by

$$R_n(g \longrightarrow n) = N_n\,A(n \longrightarrow) \qquad (2)$$

where $A(n \rightarrow) = \sum_{p<n} A(n \longrightarrow p)$. Then, the emission coefficient can be now written:

$$\varepsilon(n \longrightarrow m) = \frac{h\nu}{4\pi}\frac{A(n \longrightarrow m)}{A(n \longrightarrow)}\,R_n(g \longrightarrow n). \qquad (3)$$

For instance, in the hollow cathode discharge, the light emission profile follows the profile of the excitation rate.

The excitation and ionization of noble gases occurs due to collisions with the beam electrons, which are emitted from the cathode surface. The excitation rate is related to the energy distribution of the electron flux $J_{eb}(E)/e\,(m^{-2}\,s^{-1}\,eV^{-1})$ by

$$R_n = N_g \int_{I_n}^{E_{\max}} \sigma_n(E)\frac{J_{eb}(E)}{e}dE, \qquad (4)$$

where N_g is the population density of the ground state, $\sigma_n(E)\,(m^2)$ is the energy dependent excitation cross section, $E_{\max}\,(eV)$ is the highest energy of the beam electrons, and $I_n\,(eV)$ is the excitation potential [11]. For simplicity, R_n will be symbolic represented by $R_n = n_g\langle\sigma_n\,J_{eb}/e\rangle$. The excitation rate increases through the cathode fall as the electrons are accelerated by the strong electric field. In the negative glow the electric field falls to zero and the electrons are no more accelerated, but they are confined inside the potential well. As a result, the excitation rate increases through the cathode fall up to a maximal value, which remains constant in the negative glow. A typical light intensity profile measured for the $4p[1/2] \longrightarrow 5d[3/2]$ at $555.9\,nm$ decaying transition of the argon atoms is shown in Fig. 3. The excitation rate by collisions with the beam electrons, which is proportional to light emission, is also indicated in Fig. 3. The flat profile shows that the excitation rate is constant in the negative glow.

3.3 Light intensity profile emitted by the sputtered metal atoms

The situation is more complex for the sputtered metal atoms. The excitation and ionization in the cathode fall is again provided by collisions with the beam electrons, but excitation and Penning ionization by collisions with metastable argon atoms also play an important role. The excitation rate by collisions with the beam electrons should be similar to that of the argon atoms, i.e., it should increase through the cathode fall and stay constant in the negative glow. In the negative glow the excitation rate is enhanced by collisions with the fast electrons of the plasma. These electrons have energy of about $3\,eV$ [5], which is very close to the first resonance energy level of the copper atoms. Laser Induced fluorescence measurements of spatial profiles of $^3P_2$ metastable argon atoms in a glow discharge show that the maximum of the density profiles occurs in the middle of the

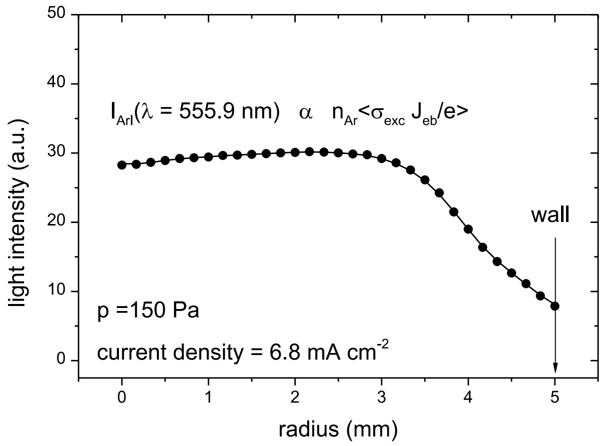

Figure 3. Radial profiles of the light intensity of the Ar I line at $555.9\,nm$ ($n_{Ar}\langle\sigma_{exc}\,J_{eb}/e\rangle$, excitation rate of the argon atoms by collisions with beam electrons).

cathode fall [5]. In the negative glow the metastable atoms density is reduced by one order of magnitude. The main loss mechanism is due to excitation collisions with thermal plasma electrons, with an energy of approximately $0.3\,eV$, to the $^3P_1$ resonant level, followed by a decaying transition to ground state. Therefore, the excitation and ionization rates of the copper atoms by collisions with metastable argon atoms and fast electrons have a strong variation in the interface between the cathode fall and the negative glow. Thus, these different excitation mechanisms lead to a sharp change of the radial light intensity profiles emitted by many copper spectral lines in the cathode fall - negative glow interface. This behavior can be observed from the radial profiles of light measured for the $^2P_{3/2} \rightarrow {}^2D_{5/2}$ (metastable) at $510.7\,nm$ decaying transitions of the copper atoms, shown in Fig. 4. The sudden increase in the light intensity profiles indicates the transition region between the cathode fall and the negative glow.

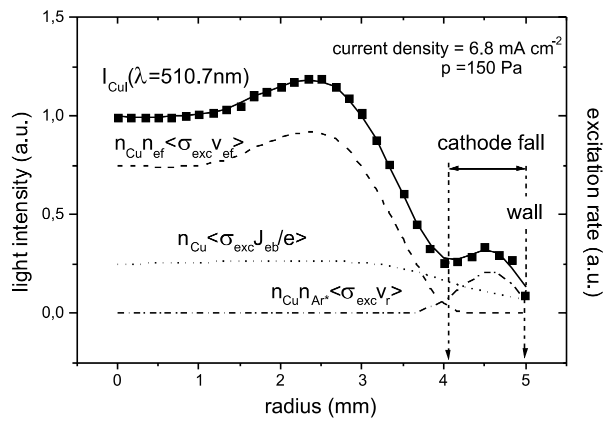

Figure 4. Radial profiles of light intensity of the Cu I line at $510.7\,nm$ showing qualitatively the contribution of the main excitation rates of the copper atoms ($n_{cu}n_{ef}\,\langle\sigma_{exc}\,\nu_{ef}\rangle$, excitation rate by collisions with fast electrons; $n_{cu}\langle\sigma_{exc}\,J_{eb}/e\rangle$ excitation rate by collisions with beam electrons; $n_{cu}n_{Ar^*}\langle\sigma_{exc}\,\nu_r\rangle$, excitation rate by collisions with metastable argon atoms).

The main excitation rates are also shown in figure 4. These curves were normalized with respect to the measured light intensity profile. A preliminary calculation with a simple model for the energy distribution of the electron flux, using the cross section for the optically allowed transition $^2S_{1/2} \rightarrow {}^2P_{3/2}$ reported by Msezane and Henry [12], gives for the excitation frequency by collisions with the beam electrons a value of $\langle \sigma_{exc}\, J_{eb}/e \rangle = 5.5 \times 10^1 s^{-1}$. This is approximately 3-fold lower than the excitation frequency by collisions with the fast electrons, which was estimated from the discharge current density, assuming a ratio of 10^3 between the density of the bulk plasma electrons and the fast electrons [5, 13] , to be $n_{ef} \langle \sigma_{exc}\, \nu_{ef} \rangle = 1.7 \times 10^2\, s^{-1}$. Therefore, we consider in the negative glow a ratio of 1/3 between the excitation rates of the beam electrons and the fast electrons. To represent the shape of the excitation rate of the copper atoms by collisions with the beam electrons, we use the experimental profile of light intensity measured for the argon atoms. As was pointed out previously, the dominant excitation in the cathode fall is by collisions with the metastable argon atoms. For simplicity, a parabolic density profile was assumed for the argon metastable to obtain the excitation rate profile shown in figure 4. The profiles for the different excitation rates of the copper atoms are, off course, only a crude estimation, which needs experimental evidence to be well established.

The cathode fall length can be more easily measured at higher current densities. Its length is about $1.2\, mm$ for pressures above $100\, Pa$, and is practically invariant over the investigated current range. A small decrease of the cathode fall thickness was observed with increasing pressures. However, the ratio between the cathode fall length d_c and the cathode radius r_w over all pressures was in the range $0.20 \leq d_c/r_w \leq 0.24$, which corresponds to a variation of only 20%.

4 Conclusions

By measuring the radial light intensity profiles of some selected copper and argon lines the length of the cathode fall was determined for a discharge with a copper hollow cathode in argon. The ratio between the cathode fall length and the cathode radius is about $d_c/r_w = 0.22$. This value was very insensitive to all the macroscopic discharge parameters showing only a slight increase, never higher than 20%, when the pressure was decreased from $200\, Pa$ down to $80\, Pa$. Similar results have been observed for discharges with hollow cathodes of different diameters made of uranium, but which were also running in argon. Thus, is seems that, so far the hollow cathode discharge is sustained by the ionization collisions of the beam electrons with the argon filling gas, the ratio between the cathode fall length and the cathode radius is, essentially, gas depend.

The analysis of the experimental radial emission profiles enables us to identify the main excitation rates of the argon filling gas and the sputtered copper atoms. The excitation of the argon occurs due to collisions with the beam electrons emitted from the cathode. In the case of the copper atoms, despite of the excitation by the beam electrons, the collisions with the plasmas fast electrons and metastable argon atoms play also an important role.

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