Metastable and resonance atoms in magnetron discharge.

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Abstract

The formation of metastable and resonance state densities of argon in cylindrical magnetron discharge is studied experimentally and theoretically at magnetic field strength B 20 mT, pressure p 2.8 Pa, current 72 mA. The excited particle densities were measured by laser absorption technique. Theoretical treatment is based on the solution of the system of balance equations for the densities of the 3p4s states with account of radiation transport, diffusion and numerous excitation and decay processes that take place in real magnetron discharge. The solution technique of the Biberman-Holstein equation of radiation transport is developed in conformity to magnetron discharge geometry. An influence of the radiation imprisonment and diffusion of metastable atoms is demonstrated, and results of the experiments and calculations are compared.

1. Introduction

Investigation of spatial distributions of the metastable and resonance atoms in discharge of magnetron configuration requires an accurate calculation of the excitation rates, and analysis of the transport processes connected with diffusion of the metastable atoms and resonance radiation imprisonment in magnetron discharges on the other hand. Recently developed non-local kinetic model of magnetron discharge plasmas [1] permits one to solve the spatially inhomogeneous Boltzmann kinetic equation in crossed electric and magnetic fields, calculate the electron distribution function and compute macroscopic properties, such as electron density and average energy, rates of excitation and ionization, etc. Solution of the problem on the metastable atom diffusion reveals no particular difficulties and can be obtained by standard methods. Formation of the resonance atom densities requires a special attention due to geometry properties of the discharge gap. Cylindrical magnetron discharge represents itself two extended coaxial electrodes in axially homogeneous magnetic field generated by coils. The photons of resonance radiation can escape from the discharge not only through the outer bounding surface as it happens in cylindrical discharge tubes, but also through the inner one.

The issues on population of resonance states in cylindrical discharge tubes are studied quite in detail for Doppler, Lorentz and Voigt profiles of spectral lines both under assumption of effective lifetime and with account of high modes. There are numerous analytical and numerical methods for solving the Biberman-Holstein equation in various geometry situations, however the discharge of magnetron configuration has not been considered in literature. At the same time there are experimental data on populations of metastable and resonance atoms in magnetron discharge, that require development of appropriate theoretical description.

In the present paper we study experimentally and theoretically the formation of the spatial distributions of the excited atoms in magnetron discharge in argon at magnetic field...
strength B – 20 mT, pressure p = 2.8 Pa, current 72 mA. The solution technique of the Biberman-Holstein equation is developed in conformity to magnetron discharge geometry. The approximation of effective lifetime is considered and radial dependence of the effective probability of radiation escape obtained for the discharge between two cylindrical electrodes. The balance equations for the particle densities of metastable and resonance atoms are considered with account of the excitation and decay processes which take place in the real discharge. The results of experiment and calculations are compared and discussed.

2. Transport equations for excited particles.

The spatial distribution of the resonance atoms in neglect for the intermixing processes is described by the Biberman-Holstein integral equation

\[ AN_r (r) - A \int_{r'} K(r, r') N_r (r') d' r' = W(r) \]  

(1)

where \( A \) is the probability of spontaneous radiation, \( W \) is the excitation rate, \( N_r \) is the density of the resonance atoms, \( K(r, r') \) is the probability that a photon emitted at the point \( r' \) will pass the distance \( |r - r'| \) without absorption and be absorbed at the point \( r \)

\[ K(r, r') = \frac{1}{4\pi|r - r'|} \int_0^{\epsilon_r, k_r} \exp \left(-k_r |r - r'| \right) dv \]  

(2)

and \( \epsilon_r, k_r \) are the contours of emission and absorption lines.

Under working pressures of magnetron discharge the contours of spectral lines are given by Voigt function with small value of Voigt profile parameter \( a \). Absorption coefficient in the center of spectral line \( k_0^{\nu} \) has large value at these pressures leading to complete absorption of the Doppler part of the contour at short distances, that permits us to use Lorentzian asymptotics of the Voigt function and assume \( k_{\nu} = a k_0^{\nu} / (\nu^2 + \omega_0^2) \), \( \epsilon_\nu = a / (\nu^2 + \omega_0^2) \)

where \( k_0^{\nu} = A \frac{c}{v_0} \frac{g_r}{g_0} \frac{2\sqrt{\ln(2)}}{8\pi^2 \Delta v_\nu} N_r \), \( \omega = 2\sqrt{\ln(2)} (v - v_\nu) / \Delta v_\nu \), \( \Delta v_\nu = 2\sqrt{\ln(2)} v_0 / c \) is half-width of the Doppler line, \( v_\nu \) is the frequency of the resonance transition, \( g_r \) and \( g_0 \) are the statistical weights of the resonance and ground states, \( c \) is the light velocity, \( N \) is the gas density and \( u \) is the thermal velocity. The presence of magnetic field of the order of hundreds Gauss and Seeman splitting of the order of natural width caused by this field, does not play any role in calculations since under large absorption coefficients the central part of the profile is completely absorbed.

The metastable atoms under the same approximations are described by the diffusion equation in the form

\[ W_m (r) = -D_m \frac{1}{r} \frac{d}{dr} r \frac{dN_m (r)}{dr} \]  

(3)

where \( W_m(r) \) and \( N_m(r) \) are the excitation rate and density of metastable atoms, \( D_m \) is the diffusion coefficient. A difference in the treatment of metastable and resonance atoms is connected with the presence of a finite free path for metastable atoms and with large paths of photons in the wings of spectral lines that does not permit to introduce diffusion coefficient for photons.

3. Approximation of the effective lifetime for magnetron discharge.
The approximation of effective lifetime $\tau_{\text{eff}}$ or effective probability of radiation escape $A_{\text{eff}}$ by Biberian's theory [2] can be obtained if eliminate the density $N_i(r')$ at the point $r$. This can be done if the kernel $K(|r-r'|)$ decreases more abrupt than the density $N_i(r')$. Then

$$A_{\text{eff}}(r) = A \left[ 1 - \frac{1}{4\pi} \int_{\omega_0}^{\omega} \int_{0}^{\infty} \int_{0}^{\infty} K(|r-r'|) dr' \right]$$

Integration over the volume in (4) includes integration over the magnetron length and the unscreened by the inner electrode region shown in Fig. 1. Defining by $z$ the axial distance between the points $r$ and $r'$, and by $\rho$ the distance between these points in the plane $z=\text{const}$, we may write $|r-r'|^2 = z^2 + \rho^2$. $r'^2 = r^2 + \rho^2 - 2\rho r \cos \psi$. The integral in (4) becomes

$$\int K(|r-r'|) d^3r' = \int_0^{\psi} \int_0^\rho \int_0^{\infty} \frac{d\psi}{\rho} d\rho I_0(\rho, \psi) + \int_0^{\psi} \int_0^{\infty} \frac{d\psi}{\rho} I_0(\rho, \psi),$$

where

$$I_0(\rho, \psi) = \frac{1}{2\pi} \int dz \frac{dz}{z^2 + \rho^2} k_0 \varepsilon_v \exp(-k_0 \sqrt{z^2 + \rho^2}) d\nu.$$

Integration limits over $\rho$ are $\rho_i = r \cos \psi - (R_i - r^2 \sin^2 \psi)^{1/2}$, $\rho_e = r \cos \psi + (R_i - r^2 \sin^2 \psi)^{1/2}$, and the angle $\psi_a = \arcsin(R_i/r)$. $R_i$ and $R_A$ are the radii of internal (cathode) and external (anode) cylinders. Integration can be carried out analytically and for the Lorentzian asymptotics of the Voigt line profile under large absorption coefficients we obtain

$$A_{\text{eff}} = A \frac{0.874}{\sqrt{\pi k_0 R_A}} \frac{1}{\pi} \left[ \int_0^\pi \frac{d\psi}{\sqrt{q_i(\psi,r)}} + \int_0^\pi \frac{d\psi}{\sqrt{q_A(\psi,r)}} \right]$$

where $q_i = \rho_i R_A$, absorption coefficient at the center of line is $k_0 = k_0^{1/2}(\alpha \pi)^{1/2} = k_0^{1/2}$. $k_0^{1/2} = AN(c/v_0)^2(g_i/\rho_0)/(4\pi^2\Delta v_f)$ is the absorption coefficient at the center of Lorentzian line contour. Thus, at pressures $p > 2.5$ Pa (or $k_0 > 10^4$, $x = |r-r'|$), i.e., in the range of applicability of the Lorentzian approximation of unabsorbed Voigt profile, the calculations with both profiles of spectral line give the same values of $A_{\text{eff}}$.

![Fig.1](image1.png)  ![Fig.2](image2.png)

Fig.1. The integration over the volume in Eq. (4) is equivalent to integration over the dashed region and along the discharge length. Fig. 2. Effective lifetime as a function of radial position and radius of cathode. The ratio $R_i/R_A$ equals: 0, that corresponds to cylindrical discharge tube; 0.1; 0.2
both electrodes. For comparison, the radial dependence of $A_{\text{eff}}$ for cylindrical tube ($R_c \to 0$) is shown at the same figure.

An accurate solution of the Biberman-Holstein equation (1) can be found with the help of eigenfunctions of the integral operator by transformation of the equation into the system of linear algebraic equation for the density of the resonance atoms. By splitting the inter-electrode distance into $M$ intervals and assuming the density of the resonance atoms within each interval to be constant, the integral term in Eq. (1) can be represented in the form

$$\int K(r, r') N_r(r') r' dr' = \sum_{m=1}^{M-1} N_r(r_m, \psi_m) \int_{r_m}^{r_{m+1}} K(r, r') r' dr dz d\psi$$

Integration of the kernel over the intervals $r_m < r' < r_{m+1}$ is analogous to above case and it gives the matrix of coefficients which determine the density $N_r(r_k)$.

In Fig. 3 the results of a model problem on resonance and metastable atom distributions are shown. The top figures show the densities of a resonance state obtained from the accurate solution of the Biberman-Holstein equation ($N_r$, solid line) and in approximation of effective lifetime ($N_r$, dashed line) for two profiles of excitation rate ($W$, dotted line). The solutions are normalized by the conditions that the maximal value of $W_0$ and $N_0$ is $0.87$ and $\sqrt{2\pi} R_c$, where $\lambda=1.47$ is the eigenvalue of the fundamental mode. It should be noted, that in the case of strong deviation of the excitation rate from the fundamental mode the densities calculated under approximation of effective lifetime are noticeably higher than those calculated accurately. In the bottom figure the normalized radial profiles of the excitation rate and densities of resonance and metastable states are represented to demonstrate the influence of diffusion of metastable atoms and transport of resonance radiation for magnetron discharge geometry.

![Fig. 3](image-url) Normalized densities of metastable and resonance atoms ($N_m, N_r$) for various profiles of excitation rate ($W$).
4. Results of experiment and modeling.

The excited particle number densities were measured by absorption spectroscopy using narrow bandwidth single mode diode laser. The laser set-up consisted of a grating stabilized diode laser in Littrow configuration. The bandwidth of 4.5 MHz and a tuning range of approximately 25 GHz were obtained. The wavelength scanning was achieved by a computer controlled voltage ramp to a highly stabilized current source. The whole laser set-up was actively temperature stabilized by a peltier element. Spectroscopic measurements were done in argon transitions $3p^54s-3p^54p$ in the range 810-826 nm. Electron density was determined from the measurements of the probe characteristics.

The recently developed kinetic theory of magnetron discharge permits us to solve the kinetic equation for a given spatially inhomogeneous electric field and obtaining the electron distribution function that can be used for further calculations of the macroscopic plasma parameters (electron density, excitation rates, etc.). In this study the radial profile of the electric field strength was chosen in a way that the electron distribution function calculated in this field gives the absolute values and radial profile of the electron density equal to the experimentally measured density of electrons. The radial dependence of the electric field strength is shown in Fig. 4. The electron density that corresponds to electron distribution function that is formed in this field has a radial dependence as shown in Fig. 5.

![Fig. 4. Model electric field strength as a function of radial position in magnetron discharge.](image1)

![Fig. 5. Electron density. Dots are the data of experiment and line is the result of calculation in a model electric field.](image2)

The electron distribution functions were used then to calculate the rates of excitation and decay of the resonance and metastable atoms in dependence on radial position. Under our condition the following processes are important in formation of the excited state densities:

A) Excitation processes (S)
1. Direct excitation of s-states from the ground state by electron impact.
2. Stepwise excitation due to intermixing in the system of lower s-states by electron impact.
3. Cascade excitation from the ground state by electron impact:
   Direct excitation to p-states and radiative transition to s-state.
   Direct excitation to higher states and radiative transition to s-state.
4. Cascade excitation from excited state by electron impact:
   Stepwise excitation from s-state to higher p-states and radiative transition to s-state.

B) De-Excitation processes (D)
1. Intermixing in the system of s-states by electron impact.
2. Cascade intermixing in the system of s-states by stepwise excitation to p-levels and subsequent radiation to s-level.
3. Stepwise ionization.

The rates of excitation (S) and de-excitation (D) were calculated in terms of the electron distribution function using corresponding cross-sections of the processes and probabilities of radiative transitions. The calculated rates were substituted into balance equations for the resonance and metastable atoms written in the form

$$AN_1(r) = A \int K(r, r')N_1(r')dr' = S - D;
-D_m \frac{d}{dr} \frac{dN_1(r)}{dr} = S - D,$$

which were transformed into a system of algebraic equations determining the densities of four lower excited levels.

In Fig. 6 the calculation results of the particle number densities of the $s_2$, $s_4$ and $s_6$ levels are compared with the data obtained in experiments for the following discharge conditions: argon, $B=20$ mT, $p=2.8$ Pa, $i=72$ mA, $R_0=0.5$ cm, $R_4=3$ cm, discharge length $L=12$ cm. The density profiles have maximum at the boundary of the cathode region and positive column where excitation rate attain maximal value, and then smoothly decrease towards anode.

![Fig. 6. Densities of the excited states. Symbols are the data of experiment, Solid lines are the results of calculations.](image)

The distribution of the metastable atoms is described quite well. In order to fit the experimental curve for the density of $s_2$ state, the probability of its spontaneous radiation was taken 2.5 times smaller than that given in literature. This difference can be explained by the finite length of magnetron discharge, an uncertainty in the constants of elementary processes and poor applicability of Lorentzian approximation of Voigt profile at short distances $x=|r-r'|$, where $k_0x<10^4$, $k_0=4 \times 10^4$ cm$^{-1}$ under our conditions.

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