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Radiation of high-Z atoms sputtered by plasma

E. Marenkov[a,*], K. Gutorov[a], I. Sorokin[a,b]

[a] National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe shosse, 31, Moscow 115409, Russian Federation

[b] Kotel'nikov Institute of Radio Engineering and Electronics, Fryazino Branch, Russian Academy of Sciences, pl. Vvedenskogo 1, Fryazino, Moscow Oblast, 141190, Russian Federation

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ABSTRACT

In this paper we apply a time-dependent radiative-collision model for analysis of radiation of impurity atoms sputtered in uniform plasma. As an example we consider spectroscopy data of neutral molybdenum atoms sputtered in argon plasma at PR-2 linear plasma device. We use effective collision strengths for electron impact excitation/de-excitation and spontaneous emission obtained earlier by relativistic R-matrix calculations. The ionization rates resolved by excited levels are estimated by the approximate ECIP method. The main feature of the experimental spectra is the maximum of radiation intensity situated at a noticeable distance from the target surface, different for different wavelength. This feature is reproduced by our calculations proving that appearance of the maximum is due to slow excitation of some levels. In contrast to the previous works on the problem, we demonstrate that neither electron-impact ionization nor escape of atoms from the plasma column are necessary for appearance of the maximum. We also show that if the initial population of atoms in excited state exceeds a certain limit, 5% in our particular case, the maximum disappears completely, giving the upper estimate of the initial population.

1. Introduction

Radiation emission from sputtered atoms can be used to obtain parameters such as sputtering rate, velocity distribution and populations of excited states [1,2]. It also provides information on transport of sputtered particles in plasmas. High-Z materials are presently the leading candidate for divertor material in fusion devices. In addition to diagnostic possibilities, radiation of high-Z impurities also presumably plays a role in shielding effects mitigating the heat loads on the plasma-facing components [3]. However, atomic data for high-Z materials necessary for diagnostics are generally of poor quality. Recent efforts have been made to improve necessary atomic data [4].

During physical sputtering of metals in plasma by ions the most of the particles emitted from the surface are neutral atoms [5]. Due to ionization the number of neutral sputtered atoms decreases away from the target. Therefore, the intensity of radiation for neutral atoms lines also decreases. One can anticipate that, in idealistic case, when the radiating atoms have the same velocity, fly in the same direction, and the plasma is uniform, this decrease is exponential and the decay parameter depends on the electron impact ionization rate. However, observations of radiation from sputtered metal atoms conducted on different linear plasma devices, PSI-2 [6], Pilot-PSI [3] demonstrate that for some lines the dependence of intensity I on the distance x from the target has a maximum located on the order of 1 cm from the target. The experiments are routinely done for various metal targets (Cr, Mo, W) with different plasma species (He, Ne, Ar).

The most plausible explanation of this peak in emission intensity is the dominating role of excitation of metastable states. These long lived metastable states can take a significant time to reach an equilibrium state with the ground state. As far as we know, the only quantitative assessment of the effect is based on a “two-level” model, considering populations of only one excited and the ground level [7,8], transition between which corresponds to the line under consideration. The model is also incorporated in Monte-Carlo code ERO [9] simulating impurity transport in plasma of tokamaks or linear devices. Effective values are taken as the rates of electron impact excitation, deexcitation, spontaneous emission and ionization for transition between the two levels. Estimation of all these quantities is based on ADAS models considering one arbitrary chosen level as a metastable [10]. Of course, the two-levels model is only a zero-order approximation clearly not capturing much more complicated physics of non steady-state radiation from high-Z atoms in plasma where the number of levels contributing to the dynamics of populations can be very large.

In the present work we apply a collisional-radiative model with excessive number of excited levels to analysis of radiation of sputtered Mo atoms in plasma of PR-2 device. The calculations are based on the

* Corresponding author.
E-mail address: edmarenkov@gmail.com (E. Marenkov).

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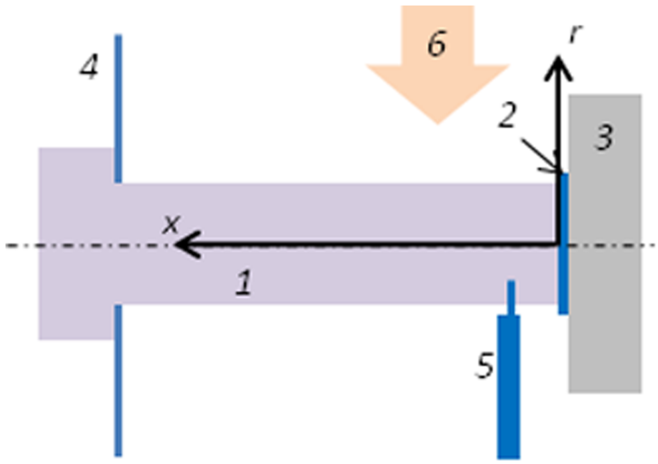

Fig. 1. Schematic view of the experimental setup. 1 – plasma column; 2 – Mo target; 3 – water cooled target holder; 4 – diaphragm, 5 – Langmuir probe movable along radial (r) direction, 6 – spectrometer view.

atomic data for neutral Mo obtained by fully relativistic R-matrix calculations [4]. We show that appearance of the maximum can indeed be associated with long-living excited states. Deficiencies of the two-level approximation are discussed. Effect of initial population of excited levels on the results is considered.

2. Experiments

The experiments were conducted at PR-2 facility [11]. PR-2 is a magnetic mirror linear plasma trap using the beam-plasma discharge. It allows for plasma conditions close to those in the edge plasma of tokamaks, with ions of energy 50–300 eV and the flux 10^{16}–10^{18} cm^{-2} s^{-1}. A polycrystalline molybdenum sample with diameter of 4 cm was situated on the watercooled target holder in the face of the plasma column, Fig. 1. Argon plasma was used for sputtering. The diameter of the plasma column was limited by a diaphragm of 3 cm diameter situated at a distance of 8 cm from the sample. A negative biasing was applied to the target resulting in Ar ions impinging on the surface with 250 eV energy.

Measurements of the plasma density and temperature were performed by a Langmuir probe situated 1 cm away from the target surface. The probe is movable along radial direction r which allows obtaining radial density and temperature plasma profiles shown in Fig. 2.

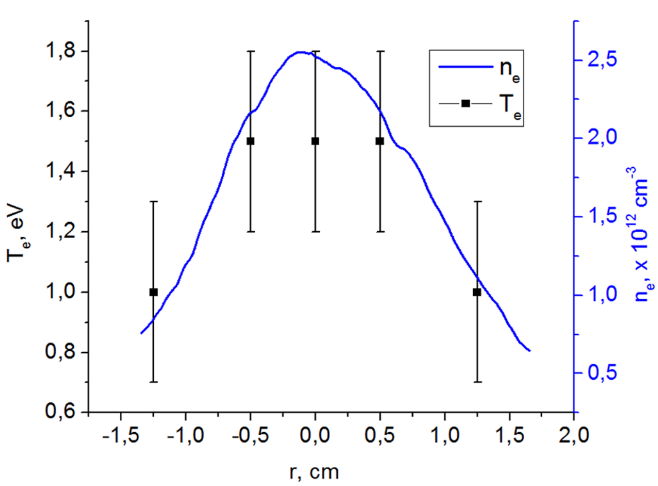

Fig. 2. Radial dependencies of the plasma temperature T_e and density n_e measured by the Langmuir probe. The relative error in determination of the density is estimated to be around 20%.

An automatic system for probe moving gives a possibility to quickly scan through radial direction [12]. However, determination of the electron density n_e and temperature T_e in each and every point requires full analysis of current-voltage characteristics which is impractical in our case for so many probe positions. Instead, the electron temperature was determined in 5 different radial positions shown in the figure. The density in between of these positions was then found from the values of ion saturation current supposing the constant $T_e = 1.5$ eV along r. This simplification does not contribute much to calculated n_e as the ion saturation current depends on the temperature as $\sqrt{T_e}$ which is virtually constant in our case.

We notice that the electron distribution function in a beam plasma discharge is often not Maxwellian. Usually groups of electrons centred around different temperatures are present [13]. We can directly observe two groups of electrons with temperatures 15 and 60 eV by the Langmuir probe, however the density of these electrons is about 10^8 cm^{-3} which is several orders lower than the main group $n_e \sim 10^{12}$ cm^{-3}.

The radiation of molybdenum atoms was detected by a system based on a monochromator equipped with a digital camera. The matrix of the camera was actively cooled to suppress thermal noise. Using the system line intensities integrated over radial direction were obtained at different x positions. To avoid possible signal distortions associated with a decrease in the sensitivity of the optical system at the matrix border, the viewing area included a portion of the sample holder. Also, when processing data, values near the borders of the matrix were also discarded, so effective registration region is 6 cm.

Two sets of lines are considered here. The first one consists of three transitions $4p^64d^55p^1\ {}^7P_{2,3,4}$ of the outer-shell electron to the ground level $4p^64d^55s^1\ {}^7S_3$. Their wavelengths are correspondingly 390 nm, 386 nm and 379 nm. The second one consists of three transitions from $4p^64d^55p^1\ {}^5P_{1,2,3}$ to an excited level $4p^64d^55p^1\ {}^5S_2$ with the wavelengths 557 nm, 553 nm, and 550 nm. For brevity, we will refer to these sets as "300 nm" and "500 nm" lines. The experimental results are shown in Figs. 5 and 6 by markers. Unfortunately, difficulties with accurate calibration of the spectrometer do not allow us to compare absolute values of the intensities. The lines intensities within a set are measured using the same calibration; however, the spectrometer has to be calibrated separately to compare the line intensities between the sets. This calibration could not be performed accurately enough. Therefore, the intensities in both sets are normalized to maximum intensity of the strongest line in the set.

The maxima are seen for both sets, but the maximum for 500 nm lines is located at larger distance, approximately 1 cm, than the maximum for 300 nm ones. As velocity of sputtered atoms is distributed over its magnitude and the emission angle, the decrease of the line intensity is due to both atomic processes and geometrical effects caused by escape of the Mo atoms through the sides of the plasma column. However, the geometry would effect the intensities of 300 nm and 500 nm lines in the same way and, therefore, the difference in the maxima positions should be attributed to atomic processes.

3. Main equations

We consider a collisional-radiative model for densities of excited levels n_i, $1 \leqslant i \leqslant m$. Evolution of the densities is due to electron impact excitation/deexcitation, spontaneous emission, and electron impact ionization:

$$\frac{dn_i}{dt} = -n_i\left\{\sum_{j=1}^{i-1}(A_{ij}+S_{ij}) + \sum_{j=i+1}^{m} S_{ij} + P_i\right\} + \sum_{j=1}^{i-1} n_j S_{ji} + \sum_{j=i+1}^{m} n_j(A_{ji}+S_{ji}) \quad (1)$$

Here A_{ij} is the Einstein coefficient for transition from level i to level j, $i > j$;$S_{ij} = n_e\langle\sigma v\rangle_{ij}$ is the excitation ($i < j$) or deexcitation ($i > j$) rate