

EXCITED ATOMS IN ARGON GAS DISCHARGE PLASMA

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General principles are discussed for a gas discharge plasma involving excited atoms where electron–atom collision processes dominate. It is shown that an optimal kinetic model of this plasma at not large electric field strengths may be based on the rate constants of quenching excited atom states by electron impact. The self-consistent character of atom excitation in gas discharge plasma is important and results in the tail of the energy distribution function of electrons being affected by the excitation process, which in turn influences the excitation rate. These principles are applied to an argon gas discharge plasma where excitation and ionization processes have a stepwise character and proceed via formation of argon atom states with the electron shell $3p^54s$.

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1. INTRODUCTION

A gas discharge plasma is in principle a nonequilibrium system [1–3] because an electric field energy is first injected into the gas through plasma electrons and then electrons transfer this energy to atoms. Hence, this system requires a kinetic description [4] based on the cross sections and rate constants of elementary processes. A general approach to this problem [5, 6] is based on the simultaneous analysis of the kinetic equation for the energy distribution function of electrons and the balance equations for excited atoms based on the parameters of elementary processes in the gas discharge plasma. Usually, elastic and inelastic collisions are important for the kinetics of a gas discharge plasma, and the peculiarity of this description is such that the theory does not allow evaluating the cross sections of electron–atom processes reliably, and therefore experimental data or certain scaling models based on experimental results are required.

Currently, there are numerous computer simulations based on this approach (see, e. g., [7–11]), but all these raise questions. First, processes of formation of fast electrons and excited atoms have a self-consistent character, that is, the process of atom excitation leads

to a sharp decrease in the electron distribution function with the increasing electron energy, and this in turn causes a decrease in the excitation rate. In this paper, the coupling of these processes is taken into account for an argon gas discharge plasma with a moderate electron number density $N_e < 10^{13} \text{ cm}^{-3}$. Second, the dependence of the atom excitation cross section on the electron energy is accounted for in this paper based on the quenching rate constants that are independent of the electron energy at low energies.

2. ENERGY DISTRIBUTION FUNCTION OF ELECTRONS

The gas discharge plasma under consideration is an ionized gas that is supported by an external stationary electric field. We consider the regime of high electron number densities where the electron equilibrium results from electron–electron collisions, which leads to the Maxwell distribution function

$$\varphi_0(v) = N_e \left(\frac{m_e}{2\pi T_e} \right)^{3/2} \exp \left(-\frac{m_e v^2}{2T_e} \right), \quad (2.1)$$

where v is the electron velocity, m_e is the electron mass, N_e is the electron number density, and T_e is the electron temperature. The normalization condition for the electron distribution function has the form

$$\int \varphi_0(v) \cdot 4\pi v^2 dv = N_e. \quad (2.2)$$

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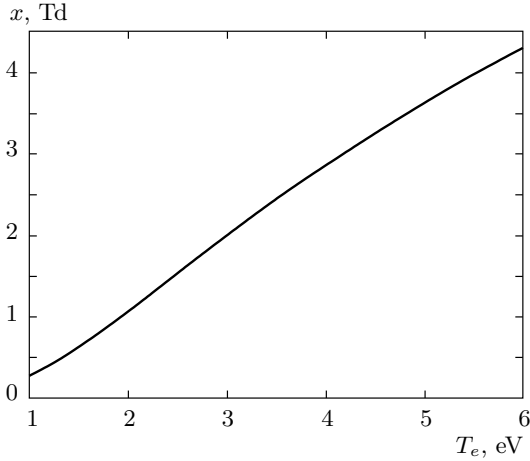


Fig. 1. The electron temperature T_e in an argon gas discharge plasma as a function of the reduced electric field strength $x = E/N_a$ given in townsend (1 Td = 10^{-17} V · cm²)

In the simplest method to account for the influence of inelastic processes, we take the distribution function to be zero at the atom excitation energy $\Delta\varepsilon$, which leads to the following form of the distribution function [12]:

$$\varphi_0(v) = N_e \left(\frac{m_e}{2\pi T_e} \right)^{3/2} \times \left[\exp\left(-\frac{\varepsilon}{2T_e}\right) - \exp\left(-\frac{\Delta\varepsilon}{T_e}\right) \right], \quad \Delta\varepsilon \gg T_e, \quad (2.3)$$

where $\varepsilon = m_e v^2/2$.

The difference of the electron T_e and atom T temperatures at not large electric field strengths E is established in elastic electron–atom collisions and is given by [13]

$$T_e - T = \frac{M a^2}{3} \frac{\langle v^2 / \nu_{ea} \rangle}{\langle v^2 \nu_{ea} \rangle}. \quad (2.4)$$

Here, M is the atom mass, $a = eE/m_e$, where e is the electron charge, E is an electric field strength, $\nu_{ea} = N_a v \sigma_{ea}^*$, and σ_{ea}^* is the diffusion cross section of electron–atom scattering. Being guided by the argon gas discharge plasma, we use formula (2.4) to determine the relation between the electron temperature T_e and the reduced electric field strength $x = E/N_a$, assuming the electron distribution function (2.3) and the cross sections for elastic electron collisions with an argon atom in [14]. The results are given in Fig. 1.

It follows that the energy distribution function of electrons in the regime of high electron number densities has the Maxwell form in the main part and becomes distorted at the tail of the distribution function.

This distortion is schematically taken into account in formula (2.3), and below we consider the tail of the electron distribution function under certain conditions. We use a general approach to this problem [5, 6] based on the kinetic equation for the distribution function of electrons and the balance equation for excited atoms. With the self-consistent character of processes of formation of fast electrons and excited atoms, we have the kinetic equation for the distribution function f_0 of electrons in the form

$$-\frac{a^2}{3v^2} \frac{d}{dv} \left(\frac{v^2}{\nu_{ea}} \frac{df_0}{dv} \right) = I_{ea}(f_0) + I_{ee}(f_0) - N_a \int_{\Delta\varepsilon}^{\infty} k_{ex}(\varepsilon) f_0(\varepsilon) d\varepsilon + N_m \int_0^{\infty} k_q(\varepsilon - \Delta\varepsilon) f_0(\varepsilon - \Delta\varepsilon) d\varepsilon - \frac{f_0(\varepsilon)}{\tau}, \quad (2.5)$$

where $I_{ea}(f_0)$ and $I_{ee}(f_0)$ are the electron–atom and electron–electron collisions, k_{ex} is the rate constant of excitation of atoms in the ground state by electron impact, k_q is the rate constant of quenching of an excited atom in collision with electrons, and we restrict ourselves to one excited state for simplicity. In the equation, we include the processes



and Eq. (2.5) is coupled to the balance equation for excited atoms

$$\frac{dN_m}{dt} = N_a \int_{\Delta\varepsilon}^{\infty} k_{ex}(\varepsilon) f_0(\varepsilon) d\varepsilon - N_m \int_0^{\infty} k_q(\varepsilon - \Delta\varepsilon) f_0(\varepsilon - \Delta\varepsilon) d\varepsilon, \quad (2.7)$$

where N_m is the number density of excited atoms. The set of equations (2.5) and (2.7) allows constructing the electron distribution function in the range that is responsible for atom excitation, and Fig. 2 gives this distribution function for the metastable state 3P_2 of the argon atom. In this figure, range 1 accounts for the character of formation of fast electrons in a gas discharge plasma; the distribution function drops sharply in range 2 due to excitation of this state, and the equilibrium between excitation and quenching processes is established in the region 3. We note that the energy electron distribution function in ranges 2 and 3 are determined by different processes of electron kinetics. Indeed, most part of the electrons penetrate in range 2

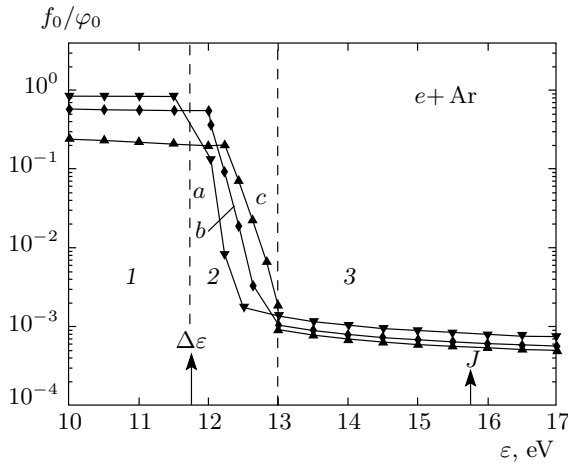
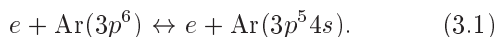


Fig. 2. Reduced energy distribution function of electrons in an argon gas plasma as a function of their energy ε , where f_0 is the electron distribution function and φ_0 is Maxwell distribution function (2.1). Arrows indicate the atom excitation threshold $\Delta\varepsilon$ and the atom ionization potential J . Curve a corresponds to the electron temperature $T_e = 2$ eV, curve b relates to $T_e = 3$ eV, and curve c corresponds to $T_e = 4$ eV

as a result of diffusion in the energy space from the range of lower energies and account for the loss of fast electrons as a result of atom excitation. On the contrary, range 3 results from quenching of excited atoms by slow electrons.

3. PROCESSES IN ARGON GAS DISCHARGE PLASMA

We start the analysis of processes in an argon gas discharge plasma involving electrons from inelastic processes:



The principle of detailed balance establishes a relation between the atom excitation cross section $\sigma_{ex}(\varepsilon)$ and the cross section $\sigma_q(\varepsilon - \Delta\varepsilon)$ of quenching an excited atom by electron impact as [15]

$$g_0\varepsilon\sigma_{ex}(\varepsilon) = g_*(\varepsilon - \Delta\varepsilon)\sigma_q(\varepsilon - \Delta\varepsilon), \quad (3.2)$$

where g_0 and g_* are the statistical weights for the ground and excited states. The excitation cross section near the excitation threshold $\Delta\varepsilon$ depends on the energy ε of the incident electron as $\sigma_{ex} \propto \sqrt{\varepsilon - \Delta\varepsilon}$ [16–18]. Therefore, the quenching rate constant k_q is independent of the electron energy at moderate energies. Hence, including the quenching rate constants in

the kinetic scheme, we can take the energy dependence of other cross sections and rate constants into account, and such a kinetic model would be optimal at not large electric field strengths.

In considering atom excitation by an electron impact, we divide this process into two parts such that the first corresponds to the formation of fast electrons that are able to excite the atom and the second self-consistent process is atom excitation due to electrons located in the tail of the energy distribution function of electrons. The rate constant $k_>$ of formation of fast electrons is determined by diffusion of electrons in the space of electron energies due to collisions between electrons and owing to the action of the electric field in accordance with Eq. (2.5). Using the Landau collision integral [19] at large electron energies, we obtain the rate constant in the form [20]

$$k_< = \frac{8\sqrt{2\pi}}{3} \frac{e^4 \Delta\varepsilon \ln \Lambda}{m_e^{1/2} T_e^{5/2}} \exp\left(-\frac{\Delta\varepsilon}{T_e}\right) \times \left(c_e + \frac{x^2}{4\pi e^2 \sigma_{ea}^* \ln \Lambda}\right), \quad (3.3)$$

where $c_e = N_e/N_a$ is the concentration of electrons, $\ln \Lambda$ is the Coulomb logarithm, and we set $\ln \Lambda = 7$ here and hereafter.

The rate constant $k_>$ of atom excitation by fast electrons at energies above the atom excitation energy follows from the kinetic equation for fast electrons that in the stationary case has the form

$$\frac{a^2}{3v^2} \frac{d}{dv} \left(\frac{v^2}{\nu_{ea}} \frac{df_0}{dv} \right) + \frac{4\pi e^4 N_e \ln \Lambda}{3m_e^2 v^2} \frac{d}{dv} \times \left(\frac{df_0}{dv} + \frac{f_0}{T_e} \right) - \nu_{ex} f_0 = 0, \quad (3.4)$$

where $\nu_{ex} = N_a k_{ex}$ is the excitation rate. We use the semiclassical solution of this equation in the form [21, 22]

$$f_0(\varepsilon) = f(\Delta\varepsilon)e^{-S}, \quad S = \kappa \left(\frac{\varepsilon - \Delta\varepsilon}{\Delta\varepsilon} \right)^{5/4}, \quad (3.5)$$

with the parameters

$$\kappa = \frac{2v_0\nu_{eff}}{5a}, \quad \nu_{eff} = \sqrt{3\frac{g_*}{g_0}\nu_0\nu_q}. \quad (3.6)$$

Here, $\nu_0 = N_a v \sigma_{ea}^*(v_0)$ is the rate constant of elastic electron–atom collisions at the excitation threshold, v_0 is the electron velocity at the excitation threshold, $\nu_q = N_a k_q$ is the quenching rate, and k_q is the quenching rate constant. We also use the principle of

detailed balance, Eq. (3.2). The excitation rate constant is given by

$$k_{ex} = \frac{g^*}{g_0} k_q \int \sqrt{\frac{\varepsilon - \Delta\varepsilon}{\varepsilon}} \varphi_0(\varepsilon) d\varepsilon. \quad (3.7)$$

In particular, if Maxwell distribution function (2.1) applies to all electron energies, we obtain the excitation rate constant

$$k_{th}(T_e) = k_q \frac{g^*}{g_0} \exp\left(-\frac{\Delta\varepsilon}{T_e}\right), \quad (3.8)$$

in accordance with the principle of detailed balance if thermodynamic equilibrium holds for electrons. In the case under consideration, where the electron distribution function decreases sharply with an increase in the electron energy, we use formulas (3.5) and (3.8) to obtain [21, 22]

$$k_{ex} = \frac{f_0}{\varphi_0} k_{>}, \quad k_{>} = 4.6 \frac{g^*}{g_0} k_q v_0^3 \frac{f_0}{N_e \kappa^{1.2}}, \quad (3.9)$$

where $f_0 = f_0(\Delta\varepsilon)$. Matching the electron fluxes in the electron energy space, we obtain [20]

$$\frac{f_0}{\varphi_0} = \frac{k_{<}}{k_{<} + k_{>}}, \quad k_{ex} = \frac{k_{<} k_{>}}{k_{<} + k_{>}}. \quad (3.10)$$

The criterion of validity of semiclassical solution (3.5) is [21]

$$\kappa \gg 1. \quad (3.11)$$

In particular, for the excitation of an argon atom on the lowest state 3P_2 ($1s_5$ in the Paschen notation), formula (3.6) gives

$$\kappa = \frac{330}{x}, \quad (3.12)$$

where the reduced electric field strength $x = E/N_a$ is given in townsend. The range $T_e = (2-6)$ eV is of importance for a plasma of gas discharge, and criterion (3.11) is fulfilled in this case according to the data in Fig. 1. In Fig. 3, we give the partial rate constants of excitation of an argon atom in the state 3P_2 , where $k_q = 4 \cdot 10^{-10}$ cm³/s [23]. We can see that neglecting the self-consistent character of atom excitation by electron impact in a gas discharge plasma leads to an error by one to two orders of magnitude.

In this analysis, we take into account that the numerical evaluation of the cross sections of electron-atom collisions is not reliable at not large electron energies, and experimental data are required in this case. This follows from the difficulties in accurate description

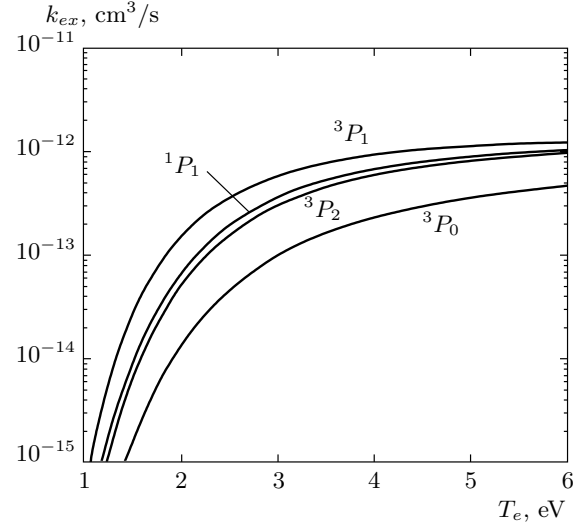


Fig. 3. The partial rate constants of atom excitation in the state ${}^3P_2(3p^54s)$ by electron impact in a gas discharge plasma at the electron concentration $c_e = 10^{-6}$ for the range 2 (see Fig. 2)

of the exchange interaction between an incident electron and valence atom electrons. In the case of collision transitions between resonant states, when the radiative dipole transition is possible between these states, the cross section of electron-atom collisions can be expressed in terms of the squared matrix element of the atom dipole moment [24, 25], as in the Born approximation, but the Born cross section can be continued to small collision energies on the basis of experimental data. In particular, at small electron energies, the quenching rate constant is given by [22, 26]

$$k_q = \frac{k_0}{(\Delta\varepsilon)^{7/2} \tau_r}, \quad (3.13)$$

where the atom excitation energy $\Delta\varepsilon$ is expressed in electronvolts and the radiative time τ_r for this transition is expressed in nanoseconds. The reduced rate constant is [22, 26] $k_0 = (4.3 \pm 0.7) \cdot 10^{-5}$ cm³/s for the s - p -electron transition in accordance with the experimental data in [27-29] for the excitation of K(4^2P), Rb(5^2P), and Cs(6^2P). We use it below for electron collisions with an argon atom for transitions from the ground state to resonantly excited states 3P_1 ($1s_4$) and 1P_1 ($1s_2$), where the respective quenching rate constant are $k_q = 8.2 \cdot 10^{-10}$ cm³/s and $k_q = 3.9 \cdot 10^{-9}$ cm³/s [30]. Figure 4 shows the rate constants of the excitation of an argon atom in its lower states with the electron shell $3p^54s$, obtained using formulas (3.3), (3.9), and (3.10).

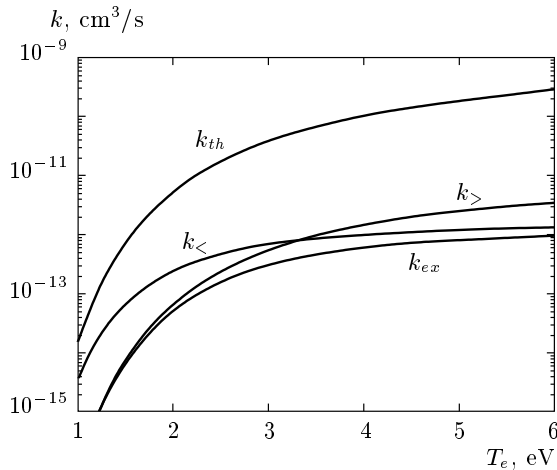


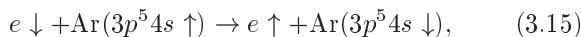
Fig. 4. Rate constants of the excitation of an argon atom by electron impact in a gas discharge plasma for lowest excited states with the electron shell $3p^54s$

Inelastic collision processes according to the scheme



may be responsible for the decay of lower excited states of an argon atom with the electron shell $\text{Ar}(3p^54s)$, and resonance transitions are more effective than other transitions. Although there are some measurements [31–34] for transitions between excited states of an argon atom, their accuracy is worse than in formula (3.13) which is based on measurements [27–29] for alkali metal atoms. Figure 5 contains the quenching rates for transitions between some states of this group, and Fig. 6 represents a simplified scheme where in accordance with the block model [6], the states related to the electron shell $\text{Ar}(3p^54p)$ are joined in one term. We use the rates of radiative transitions between the states under consideration according to measurements of the NIST group [35].

Another process of detachment of excited argon atoms with the electron shell $3p^54s$ can be related with the mixing processes that result from spin exchange between an incident and valence electron as a result of collision according to the scheme



where an arrow indicates the spin direction for electrons. Exchanging the spin direction of a valence electron changes its coupling to the rest of the atom, which leads to transitions between states of the electron shell $3p^54s$. Unfortunately, there is only an estimate $k_{mix} \sim 10^{-7} \text{ cm}^3/\text{s}$ [36, 37] for the rate constant of process (3.15), but this value is smaller than the

rate of process (3.14). In addition, Fig. 7 gives the rate constants k_Q of collision transitions from the states of the electron shell $3p^54s$ to all states with the electron shell $3p^54p$:

$$k_Q = \sum_i k_{qi} \exp\left(-\frac{\Delta\varepsilon_i}{T_e}\right).$$

4. TRANSPORT OF RESONANCE RADIATION IN ARGON GAS DISCHARGE PLASMA

Propagation of resonance radiation due to transitions involving excited states can be of importance for the kinetics of a gas discharge plasma because the lifetime of resonantly excited atoms increases due to the reabsorption process. The reabsorption process leads to the broadening of spectral lines, and the width ν of a spectral line as a result of atom collisions is given by [38]

$$\nu = \frac{1}{2} \langle N_a v \sigma_t \rangle, \quad (4.1)$$

where σ_t is the total cross section of atom collisions. The resonant lines impact broadening (or the Lorenz broadening of spectral lines) results from interaction between atoms in states between which the radiative transition proceeds. The interaction potential of such atoms is proportional to the square of the matrix element of the dipole moment operator between the transition states. The same proportionality also holds for the rate of radiative transitions between these states, and therefore the absorption coefficient k_0 at the line center does not depend on this value. For the radiative transition from a p to an s electron state, the absorption coefficient at the line center is

$$k_0 = \frac{3.5}{\lambda}, \quad (4.2)$$

where λ is the wavelength for resonant photons. This formula accounts for a complicated process of collision of atoms with p valence electrons, including elastic scattering of atoms, excitation transfer, and depolarization of p electron in the course of atom collisions. The partial and total cross section of this process were determined in [39, 40], and the results are used in formula (4.2). We note that in [11], the numerical coefficient in formula (4.2) is seven times lower.

At a moderate gas pressure, the broadening at the center of spectral lines is determined by the Doppler mechanism, but the Lorenz mechanism operates at the line wing. Using the Veklenko theory [41, 42] for the reabsorption of radiation for the Lorenz shape of a spectral line, we can find the effective lifetime τ_{eff} of a

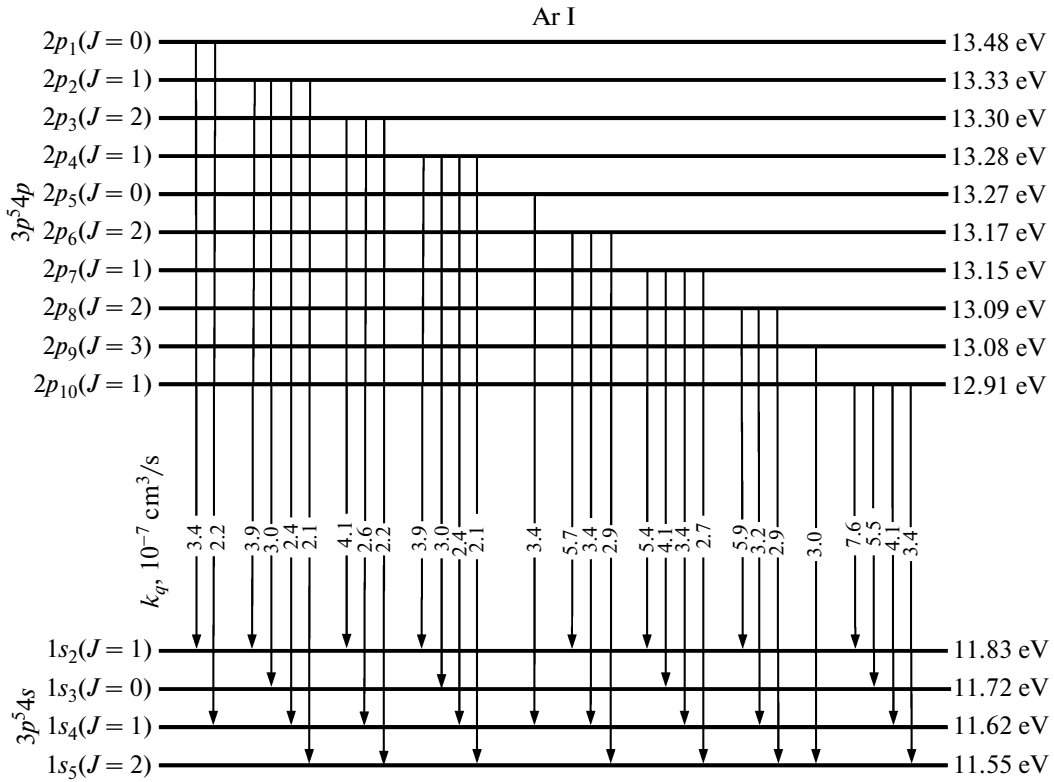


Fig. 5. Quenching rate constants for electron–atom collisions in a gas discharge plasma for transitions between argon atom states related to electron shells $3p^5 4s$ and $3p^5 4p$

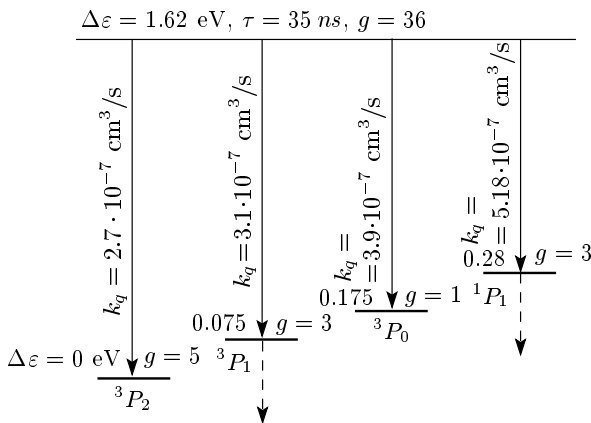


Fig. 6. Quenching rate constants for electron–atom collisions in a gas discharge plasma for transitions from the electron shell $3p^5 4p$, where all the electron terms of an argon atom are joined in one term, to atom states related to electron shells $3p^5 4s$

$$\tau_{eff} = 4.9\tau_r \sqrt{\frac{R}{\lambda}}, \quad (4.3)$$

where τ_r is the radiative lifetime of an isolated atom for this transition.

We also find the ratio of the atom number densities in states i and f if a radiative transition between these states is possible,

$$A(f) \rightarrow A(i) + \hbar\omega. \quad (4.4)$$

We can find the ratio of the number densities N_f and N_i in these states from the balance equations for the atom number densities in these states, and this ratio is

$$\frac{N_f}{N_i} = \frac{k_{ex}}{k_q} \left(1 + \frac{N_e}{N_0}\right)^{-1}, \quad N_0 = \frac{1}{k_q \tau_{eff}}, \quad (4.5)$$

where k_{ex} and k_q are the rate constants for the transition between i and k states and the reciprocal transition by electron impact in a gas discharge plasma, and τ_{eff} is the lifetime of the excited state f as a result of radiation with the reabsorption process taken into account. In particular, if a uniform argon gas discharge plasma is located inside a cylinder tube of the radius $R = 1 \text{ cm}$,

resonantly excited atom inside a uniform gas discharge plasma located in a cylinder tube of a radius R as:

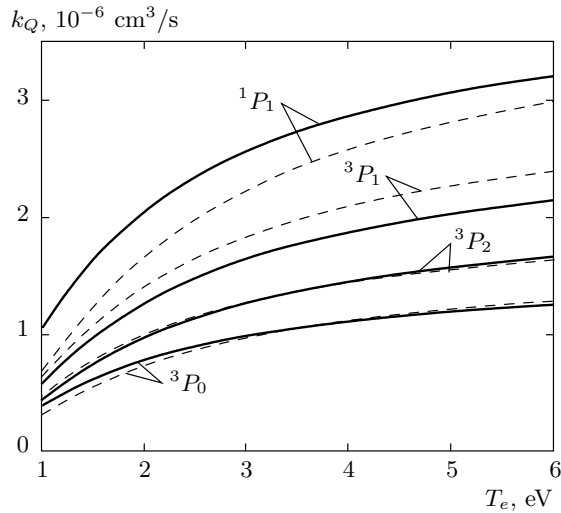


Fig. 7. Excitation rate constants by electron impact in an argon gas discharge plasma with transitions from states 3P_2 , 3P_1 , 3P_0 , and 1P_1 of the electron shell $3p^54s$ to all states with the electron shell $3p^54p$. Dotted curves correspond to joining terms with the electron shell $3p^54p$, and solid curves are the sums of partial cross sections

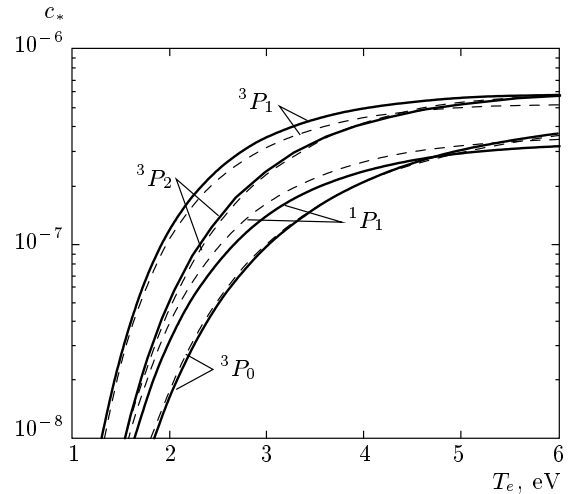


Fig. 8. Dependence on the electron temperature for the concentration of excited argon atoms 3P_2 , 3P_1 , and 1P_1 of the electron shell $3p^54s$ in a gas discharge plasma in the limit of high electron number density. Solid curves represent the transition to all states of the electron shell $3p^54p$, and the dashed curves correspond to a unified electron term of this electron shell

the parameter N_0 in (4.5) is $N_0 \approx 3 \cdot 10^{13} \text{ cm}^{-3}$ for the transition from the argon atom states $^3P_1(3p^54s)$ and $^1P_1(3p^54s)$ to the ground state. It follows that transfer of resonant radiation is of importance for gas discharge plasma.

5. POPULATION OF EXCITED ARGON ATOMS IN GAS DISCHARGE PLASMA

The concentration of excited atoms follows from the balance of processes of formation and detachment of these atoms. In a widespread case, where formation and decay of excited atoms result from electron-atom collisions, the concentration of excited atoms in a given state is

$$c_* = \frac{k_{ex}}{k_Q}, \quad (5.1)$$

where k_{ex} is the rate constant of formation of excited atoms in a given state and k_Q is the total quenching rate constant of this state. There are various channels of detachment of excited atoms in the case electron-atom collisions. Figure 8 shows the concentration of excited atoms related to the electron shell $3p^54s$ if radiation of excited atoms with the electron shells $3p^54s$ and $3p^54p$ is locked, and the rate constant k_Q of detachment of the states of the electron shell $3p^54s$ re-

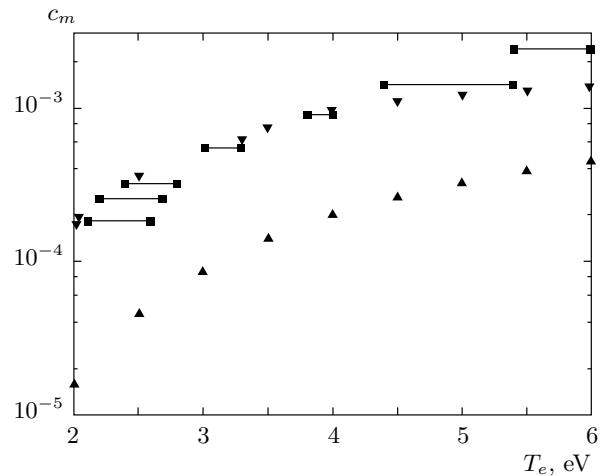


Fig. 9. Concentration c_m of argon metastable atoms in the lowest excited state 3P_2 as a function of the electron temperature for the range 3 (see Fig. 2). Triangles correspond to formula (5.1), where quenched leads to transitions of the electron shell $3p^54p$ and upturned triangles relate to the quenching due to atom ionization; experimental data are located between filled squares

sults mostly from transition to states with the electron shell $3p^54p$.

At low electron number densities, radiation for the transition between states of the electron shell $3p^54p$

and $3p^54s$ becomes locked, and the quenching of excited states results from atom ionization as an irreversible process because transitions to highly excited states are weaker. Another channel for quenching of the metastable state $^3P_2(3p^54s)$ consists in transition to resonantly excited states of this electron shell if radiation with transitions from these states to the ground state is locked. Figure 9 shows the electron temperature dependence for the concentrations of the metastable atoms $^3P_2(3p^54s)$ in an argon gas discharge plasma for different channels of quenching these states. It follows that conditions of the experiment in [43] corresponds to the channel of the decay of this state due to the ionization process. We note that the experimental data in [43] relate to an inductively coupled argon plasma of a low number density of argon atoms, and the method of optical absorption spectroscopy is used to determine the number density of metastable atoms.

We also note that the presence of metastable atoms in a gas discharge plasma influences the energy distribution function of electrons. In particular, from Eqs. (2.5) and (2.7), the ratio of the energy distribution function of electrons f_0 to the Maxwell distribution function φ_0 in (2.1) at energies above the atom excitation energy

$$\frac{f_0}{\varphi_0} = \frac{g_*k_q}{g_0k_Q}. \quad (5.2)$$

This relates to curve 3 in Fig. 2 and is used there.

6. CONCLUSION

We considered the argon gas discharge plasma where electron–atom collisions dominate and excited atoms influence the plasma kinetics. This occurs if a typical collision time of electron–atom collisions is small compared with other times of this plasma, in particular, a typical time of transport to plasma boundaries. The self-consistent character of inelastic electron–atom processes is of importance for this plasma. As a result, the energy distribution function of electrons decreases sharply above the atom excitation threshold with an increasing electron energy. For this reason, only the lowest excited states of atoms take part in the kinetics of a gas discharge plasma whereas formation of highly excited atoms does not proceed from the ground atom state and results from lowest excited states in a stepwise way. Correspondingly, ionization of such a gas discharge plasma has a stepwise character. We note that in spite of a simple kinetic scheme, many regimes of this plasma may be realized.

This analysis shows that the existing schemes of computer simulation of gas discharge plasma (see, e.g., [7–11]) are not reliable because they do not account for the self-consistent character of the gas discharge plasma kinetics the energy dependence of electron–atom processes. Based on the above analysis, a computer model describing the kinetics of gas discharge plasma can be constructed. In our analysis, we include a restricted number of excited states since excitation of other ones has the stepwise character.

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