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ELECTRON ENERGY RELAXATION LENGTH IN CONNECTION WITH THE PROBLEM OF ELECTRON ENERGY DISTRIBUTION LOCALITY IN GLOW DISCHARGE PLASMA IN A XENON-CHLORINE MIXTURE

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ДЛИНА РЕЛАКСАЦИИ ЭНЕРГИИ ЭЛЕКТРОНА И ПРОБЛЕМА ЛОКАЛЬНОСТИ РАСПРЕДЕЛЕНИЯ ЭЛЕКТРОНОВ ПО ЭНЕРГИЯМ В ПЛАЗМЕ ТЛЕЮЩЕГО РАЗРЯДА В СМЕСИ КСЕНОН-ХЛОР

The analytic calculations of relaxation length of electron energy based on a probabilistic approach and numerical calculations of nonlocality effect on the plasma-chemical processes rates have been carried out for plasma of an electronegative glow discharge in a mixture of xenon and chlorine. It is shown, that for total pressure higher than 6 Torr and with amount of chlorine more than 1/25 the effect of electron energy distribution nonlocality is negligibly small and the local approach can be used for modeling such discharges.

GAS DISCHARGE, ENERGY RELAXATION, ELECTRONEGATIVE GASES, ENERGY DISTRIBUTION LOCALITY.

На основе вероятностного подхода выполнены аналитические расчеты длины релаксации энергии электрона, а также численные расчеты влияния нелокальности на скорости плазмохимических процессов в электроотрицательном разряде в смеси ксенона и хлора. Показано, что при общем давлении более 6 Торр и при доле хлора более 1/25 влияние нелокальности энергетического распределения электронов пренебрежимо мало, и для моделирования таких разрядов можно пользоваться локальным приближением.

ГАЗОВЫЙ РАЗРЯД, РЕЛАКСАЦИЯ ЭНЕРГИИ, ЭЛЕКТРООТРИЦАТЕЛЬНЫЕ ГАЗЫ, ЛОКАЛЬНОСТЬ ЭНЕРГЕТИЧЕСКОГО РАСПРЕДЕЛЕНИЯ.

In recent years, there has been significant interest in investigating and modeling the physical processes in middle pressure (5 – 40 Torr) electronegative (EN) discharges in the mixtures of inert gases and halogens because of their practical applicability as effective and powerful sources of ultraviolet radiation [1 – 5].

The physical processes in strongly EN plasmas are extremely complex, and therefore one obtains physical information about these processes mainly from numerical modeling [2, 6 – 8]. The vast majority of EN plasma models use so-called local approach, which means that the electron energy distribution function (EEDF) and also all relevant plasma-chemical processes in the given place can be only expressed in terms of electric field E and neutrals density N being in the same place. We can accept that EEDF is local if

$$\lambda_W(W_e) < \lambda_{E/N} \quad \forall W_e, \qquad (1)$$

where $\lambda_W(W_e)$ – relaxation length of electron energy W_e, $\lambda_{E/N}$ – spatial scale of E/N ratio.

If (1) is not valid, then the kinetic energy of an electron cannot be expressed through the plasma conditions (at particular field E) measured in the place where this electron is located, i. e. EEDF is nonlocal [9, 10], and this fact

should be taken into account while modeling, especially if the field E is spatially nonuniform [10].

In the present paper we estimate the degree of EEDF nonlocality in EN plasmas of a gas mixture containing xenon and chlorine.

It is known that the nonisothermal plasma (at $T_e > T_i$ where T_e, T_i are electron and ion temperatures) of a gas discharge containing both positive and negative ions stratifies across the current direction into two regions with different ion compositions and properties [6 – 8, 11]. In the central region («core»), the densities of positive n_p and negative n_n ions are significantly more than the electron density n_e, i. e. we can speak about an ion-ion (*i-i*) plasma where $n_p \approx n_n >> n_e$. In the case of strong electronegativity the diffusion of electrons in the «core» occurs to be almost free diffusion, the radial profile of n_e is almost flat, and the radial space charge electric field E_r is weak, close to zero. In the outer plasma region («edge»), the density of negative ions is very low $n_p \approx n_e >> n_n$, and we can speak about an electropositive (EP) electron-ion (*e-i*) plasma. In the «edge» area, there is an ambipolar diffusion regime, and the finite radial space charge electric field E_r directed to the wall. The thickness of the «edge» can be estimated for cylindrical geometry as [12]:

$$\delta \approx \frac{\pi}{2} \frac{R_0}{\sqrt{2\sqrt{\alpha} + \alpha}}, \qquad (2)$$

where $\alpha = \nu_a e R_0^2 / (\mu_n T_e)$, ν_a – attachment frequency, μ_n – negative ion mobility, T_e – electron temperature, R_0 – discharge tube radius.

In the narrow transition region between *i-i*- and *e-i*-plasma, there are strong variations of both positive and negative ion concentrations.

The estimation of λ_W and EEDF nonlocality effects has got some difficulties because the value λ_W is not constant but it depends on the electron energy W_e. Therefore, at middle pressure it happens that inequation (1) can be valid for big electron energies (particulary, in pure inert gases where W_e is higher than the first excitation potential) while for lower energies (1) cannot be valid. In such a case, one should derive the kinetic Bolzmann equation taking into account the electric field inhomogeneity which is a difficult task. According to [9, 13], EEDF for EP inert gases can be accepted as local if $pR_0 \geq 10$ Torr·cm (p – gas pressure).

Three considerations prompted us to carry out the presented investigation.

Firstly, the value pR_0 for the part of our discharges is just ≤ 10 Torr·cm [1, 2], and an EEDF nonlocality can be suspected.

Secondly, for EP plasmas there is no stratification, radial field E_r is distributed over the whole plasma cross section, and $\lambda_{E/N} \cong R_0$. But in case of EN plasma, $\lambda_{E/N} \cong \delta$, and corresponding to (2), δ can be ~ $(0.1 - 0.2)R_0$ [8] i. e. $\lambda_{E/N}$ is smaller than R_0. Hence, a criterion (1) of EEDF locality for EP plasmas is weaker than for EN ones, and the problem of EEDF nonlocality in EN plasmas can become even more actual than for EP plasmas.

Thirdly, the gas mixtures considered here contain a noticeable amount of molecular EN gas where the electron energy losses can happen even for small initial energies – due to excitation of low lying vibrational and rotational molecular states and by dissociative attachment [14]. That can lead to a reduction of λ_W in comparison with λ_W in atomic EP gas. By numerical modeling an EN discharge in molecular oxygen [15], it has been shown that EEDF in the tube of 12 mm diameter is local at pressure $p \geq 1$ Torr, which corresponds to $pR_0 \geq 1.2$ Torr·cm. Hence, for discharges in molecular EN gases, the EEDF nonlocality can occur at smaller pressure than in inert gases. This is an encouraging result but, unfortunately, there are no literature data either about $\lambda_W(W_e)$ or about EEDF nonlocality in the discharge plasma containing molecular chlorine.

Before estimating electron energy relaxation length $\lambda_W(W_e)$ itself, let us obtain the expression for the electron free path length $\lambda_e(W_e)$. Definitely,

$$\lambda_e(W_e) = \frac{v_e}{\nu_{ea}(W_e)},$$

where v_e is electron velocity, $\nu_{ea}(W_e)$ is the frequency of any electron-atomic (EA) collisions. Since

$$\nu_{ea}(W_e) = \sigma^{full}(W_e) v_e N,$$

where $\sigma^{full}(W_e)$ – full EA collision cross-sec-

tion for electron energy W_e, we obtain

$$\lambda_e(W_e) = \frac{1}{N\sigma^{full}(W_e)}.$$

If we consider the discharge in a gas mixture, for example in a mixture of xenon and chlorine, then

$$\lambda_e(W_e) = \frac{1}{N_{\mathrm{Xe}}\sigma_{\mathrm{Xe}}^{full}(W_e) + N_{\mathrm{Cl}_2}\sigma_{\mathrm{Cl}_2}^{full}(W_e)}.$$

In one elastic EA collision, an electron losses almost no energy. Therefore, we can accept that the probability of preserving electron initial energy W_e after one EA collision is equal to the probability of elastic collision:

$$a_{el}(W_e) = \sigma^{el}(W_e) / \sigma^{full}(W_e),$$

where $\sigma^{el}(W_e)$ is the cross section for elastic EA collisions.

The same probability for a series of k successive collisions will be

$$a_{el}^{(k)}(W_e) = [a_{el}(W_e)]^k.$$

If the gas mixture contains halogen molecules, then the denominator of $a_{el}(W_e)$ is always bigger than the numerator due to attachment and excitation of low lying molecular states, and the value of $a_{el}(W_e)$ is always smaller than 1 for all W_e. Hence, we can define the fact of relaxation of initial electron energy W_e after k collisions as diminution of $a_{el}^{(k)}(W_e)$ value down to $1/e$, and from the equation

$$\frac{1}{e} = \left[\frac{\sigma^{el}(W_e)}{\sigma^{full}(W_e)}\right]^k$$

we can calculate the value of $k(W_e)$ – the number of collisions needed for the relaxation of electron energy W_e:

$$k(W_e) = \max\left(1, \left\{-\ln\left[\frac{\sigma^{el}(W_e)}{\sigma^{full}(W_e)}\right]\right\}^{-1}\right).$$

As electron motion in any gas at middle pressure is similar to the Brownian motion, the most probable electron displacement from an initial point after $k(W_e)$ collisions corresponds to the desired electron energy relaxation length $\lambda_W(W_e)$:

$$\lambda_W(W_e) = \sqrt{k(W_e)} \cdot \lambda_e(W_e) = \frac{\sqrt{k(W_e)}}{N_{\mathrm{Xe}}\sigma_{\mathrm{Xe}}^{full}(W_e) + N_{\mathrm{Cl}_2}\sigma_{\mathrm{Cl}_2}^{full}(W_e)}. \quad (3)$$

In Fig. 1 the results of the calculations of $\lambda_W(W_e)$, according to Eq. (3), with the use of the cross-section set taken from [14] are presented for a discharge in the mixture of Xe and Cl_2. One can see that if W_e exceeds the first excitation potential of Xe, the value of $\lambda_W(W_e)$ is small and we can state that the EEDF in this energy range is local. If N_{Cl_2} concentration is more than 10^{15} cm^{-3}, then the EEDF is local

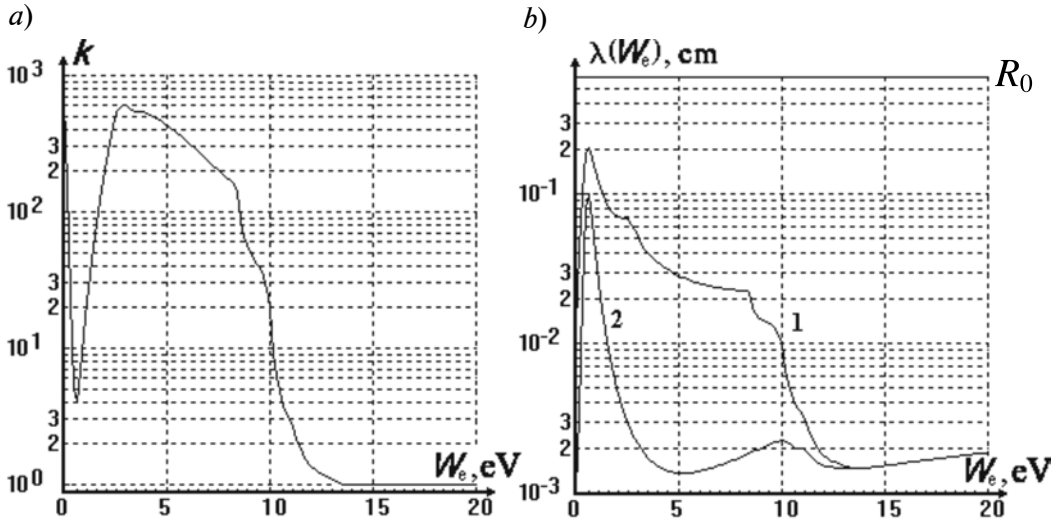

Fig. 1. Dependences on electron energy: *a* – number of collisions needed for energy relaxation; *b*: *1* – energy relaxation length, *2* – free path length; the horizontal line shows the tube radius $R_0 = 6$ mm. Gas mixture is: 6 Torr of Xe and 0.25 Torr of Cl_2, discharge current $I = 10$ mA, $\lambda_{E/N} \approx \delta \approx 0.1$ cm

if $W_e > 2.5$ eV (slightly higher than excitation threshold of the $B^1\Pi_u$ state of Cl_2 molecule). For the slowest electrons with $W_e < 0.5$ eV, the cross-section of dissociative attachment becomes large, and $\lambda_W(W_e)$ becomes small. However, we can see that in the electron energy range $0.5 < W_e < 2$ eV, $\lambda_W(W_e) \approx \lambda_{E/N} \cong \delta$, and expression (1) for these electrons is no more valid. The cross-sections of EA collision processes in this energy range (dissociative attachment, molecular vibrational level excitation) are small, hence the probability of electron energy losses is small as well.

So, the EEDF in discharge plasmas containing inert gases and chlorine can turn out to be nonlocal only in the narrow electron energy range of $0.5 < W_e < 2$ eV.

In discharges in EN gases and mixtures, the strong field inhomogeneity occurs in the «edge» (*e-i* plasma), where the radial field E_r can be large (up to 100 – 400 V/cm at $p \approx 6$ Torr) within the distances of ≤ 0.5 mm from the wall [6, 8]. Radial fields in the «edge» can even exceed the longitudinal field E_z (further in the «core» the radial field E_r quickly reduces to zero). But the outer region contains relatively few electrons [8] and, due to the large total field, the fraction of «hot» electrons with big probability of energy-consuming inelastic collisions is significant. This means that $\lambda_W(W_e)$ for such a fraction is small (see Fig. 1), and «hot» electrons do not penetrate into the region of the «core». Slow electrons can penetrate from the «edge» into the «core» but due to the small concentration of electrons in the «edge», they cannot seriously effect EEDF in the «core».

The radial field in the «core» is small [6, 8], and the total electrical field practically is equal to the longitudinal field E_z which is uniform, and hence there is no difference between local und nonlocal EEDF.

There is one more aspect which should be considered. Together with electron energy relaxation in isotropic Brownian motion, there is a radial drift of electrons to the discharge center in radial field E_r. If electrons during this drift have collisions with small energy losses only, they can receive an additional kinetic energy from the radial field.

But the slowest electrons with $0 < W_e < 0.5$ eV cannot gain energy: $\lambda_W(W_e)$ for such electrons is small (see Fig. 1, *b*, curve *1*) because the attachment cross-section is large in this energy range [16], and in the attachment acts electrons disappear as free particles.

The said receive of kinetic energy from radial field is mostly relevant to the group of electrons with $0.5 \leq W_e < 2$ eV where inelastic cross-sections are small and $\lambda_W(W_e)$ is comparable with δ. But let us notice that it is enough

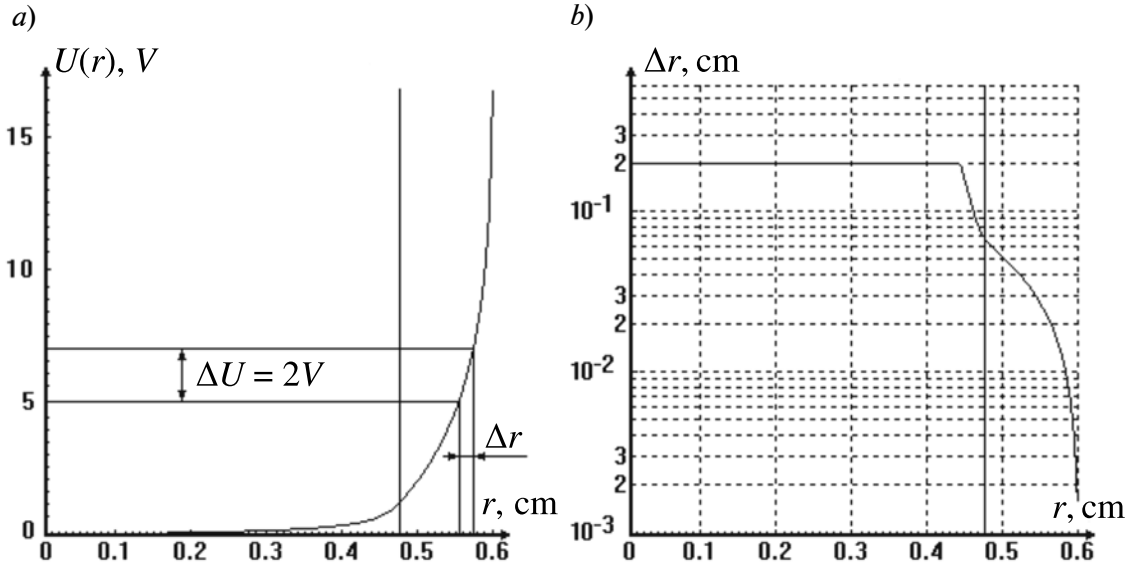

Fig. 2. Radial field potential (*a*) and the value of Δr (*b*), limited above by $\lambda_W \cong 0.2$ cm (see Fig. 1); discharge conditions are the same as in Fig. 1; vertical line shows the boundary between *i-i*- and *e-i*-plasmas

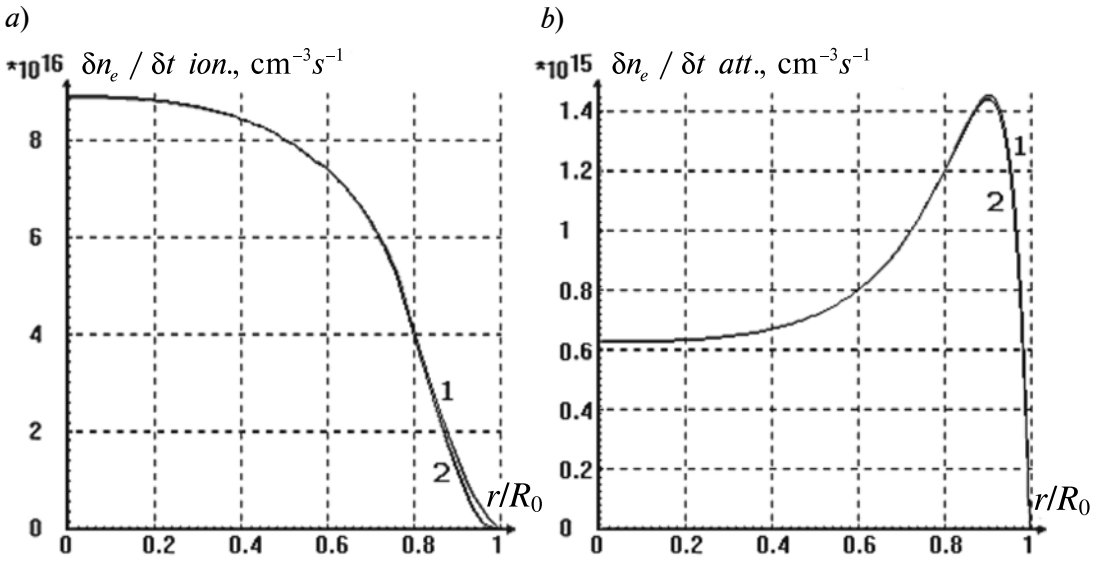

Fig. 3. Spatial distributions of ionization $N_{Xe} n_e \langle \sigma_i v_e \rangle$ (*a*) and attachment $N_{Cl_2} n_e \langle \sigma_a v_e \rangle$ (*b*) rates in local (*1*) and nonlocal (*2*) approach; the discharge conditions are the same as in Fig. 1

for these electrons to gain 2 eV, and their energy relaxation length becomes vanishingly small (see Fig. 1, *b*, curve *1*).

Hence, the real energy relaxation length of electrons with initial energy $0.5 \le W_e < 2$ eV can be defined as the piece of radial coordinate Δr along which the potential of radial field changes by $\Delta U \approx 2$ V (Fig. 2, *a*); more correctly, this length will be $\lambda_W^*(W_e) = \min[\Delta r, \lambda_W(W_e)]$ where $\lambda_W(W_e)$ can be estimated according to (3). In Fig. 2, there is an illustration to Δr computing and the result of such computing: $\Delta r(r) \approx 2 / E_r(r)$, where E_r is in V/cm.

It is seen that in the «edge» (*e*-*i* plasma), where nearly all the radial potential fall is concentrated, the values of Δr are vanishingly small, and we can hence accept the EEDF in the «edge» as local. Only at the boundary between *e*-*i*- and *i*-*i*-plasmas and inside the «core» (where E_r is close to zero), $\lambda_W^*(W_e) \approx \lambda_W(W_e)$, and EEDF in the energy range $0.5 < W_e < 2$ eV is nonlocal. But this group of electrons practically takes no part in plasma interaction processes due to smallness of all collision cross-sections for electrons of said energy range.

Assuming the afore-mentioned, we can suppose that the effect of EEDF nonlocality on properties of our discharges should be not significant. Model calculations [8] confirm this assumption. As an illustration, the results of computing of rates of some plama-chemical processes both in local and nonlocal approach are presented in Fig. 3.

For the mixture of 6 Torr Xe and 0.25 Torr Cl_2, the biggest difference among all collision rates was observed in the ionization rate $N_{Xe} n_e \langle \sigma_i v_e \rangle$, the residual for local and nonlocal approaches was of about 1.6 % with the main deviation taking place just near the tube wall (not more than 0.5 mm from the wall). The reason for such a weak reduction of ionization rate in the nonlocal approach lies in the small relaxation length (< 0.1 mm). As a result, the wall losses of «hot» electrons, which are not only able to produce ionization but also can penetrate through the wall potential barrier and die on the wall, are small.

For the attachment rate $N_{Cl_2} n_e \langle \sigma_a v_e \rangle$ – the most important process for EN discharges – where the process is maintained mainly by slow electrons [16], the residual turned out to be even smaller – about 1 %.

These differences have almost no effect on the spatial distributions of charged particles and excimer molecules densities (the concentration of residuals was less than 1%).

The residuals for the mixtures with higher gas pressure or with higher percentage of chlorine are smaller because $\lambda_W(W_e)$ decreases

proportionally both to total pressure and to amount of molecular chlorine. For the mixture of 18 Torr Xe and 0.7 Torr Cl_2, the residuals for rates of all the processes turned out to be no more than 0.4 %.

Hence, we can state that for gas discharges in a mixture of Xe and Cl_2 with total pressure not less than 6 Torr and with amount of chlorine not less than 1/25, the effect of EEDF nonlocality on properties of discharges is insufficient, and the local approach can be used in modeling such discharges.

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