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Collisional recombination in strongly coupled plasmas

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Abstract

The molecular dynamics model of collisional recombination in strongly coupled plasmas is developed in the frames of the fluctuation approach. A non-monotonic nonideality dependence of the collisional recombination rate is discovered for all multiplicities of ionization studied. The rate is drastically suppressed with respect to the extrapolation of the ideal plasma three-body recombination conventional expression. The mechanisms of the suppression are discussed. The value of the suppression agrees with the value which was measured for the ultracold plasma. Collisional recombination reduces to the three-body recombination at the decrease of the nonideality.

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1. Introduction

Three-body recombination resulting from the collision of two electrons and an ion is an important process in plasmas. It is well studied for ideal plasmas [1, 2]. It is also essential for strongly coupled plasmas (SCP) in a cluster [3–5], latent track [6–8] and ultracold [9–11] cases when multiple ionization is included (ions with charge $Z > 1$). Different approximations are used to analyze the influence of nonideality on the recombination [12–20]. In contrast to the ideal plasmas, the recombination in SCP can hardly be reduced to the three-body process. Therefore, we use the term *collisional recombination*.

The qualitative estimation of the suppression of the collisional recombination rate was first predicted in [12] on the basis of the hypothesis of a 'gap' adjoining to the ionization limit and replacing the highly excited atomic levels in SCP [21]. An attempt to estimate the three-body recombination rate in SCP was done subsequently in [13]. However, the authors [13] based their estimation on the Fokker–Planck approximation and ignored both the hypothesis [12, 21] and the arguments concerning the fluctuation nature of the 'gap' formation in SCP [22]. The influence of the highly excited atomic level cutoff on the three-body recombination

rate was estimated in [16]: a number of cutoff procedures used resulted in different degrees of suppression; however, the reason to exclude merged levels above the cutoff from the recombination process is not discussed; papers [12, 21, 22] are not mentioned. The influence of plasma microfields on the three-body recombination rate in SCP was taken into account in [17] resulting in a certain suppression of the rate; the many-body Green function approach was used in [14, 15]; the 'gap' ideas [21, 22] were not included.

The recombination in singly ionized SCP was studied by the method of molecular dynamics (MD) [18–20]. The authors [18] suggested to abandon the principle of detailed balance to explain their result on the lowering of the recombination rate; however, it was noted in [23] that uncommon mirror boundary conditions used in [18] could influence the MD results. Small plasma cluster with the Gaussian distribution of density and random distribution of particles was modeled in [19, 20]. The numerical procedure used was a very refined one; however, the processes of correlation establishment and fast electron exit the cluster could falsify the MD results.

As to ultracold plasmas it should be mentioned that there are recent experiments for weakly coupled ultracold plasmas that found good agreement with standard theory. Three-body recombination is measured in an ultracold neutral xenon plasma by detecting recombination-created Rydberg atoms using a microwave-ionization technique [24]. With the accepted theory and measured rates, the plasma temperatures are extracted, which are in reasonable agreement with previous measurements early in the plasma lifetime. The resulting electron temperatures indicate that the plasma continues to cool to temperatures below 1 K. The free expansion of ultracold neutral plasmas is used as a time-resolved probe of electron temperature evolution in combination with numerical simulations to characterize the crossover from an elastic-collision regime to the regime in which inelastic processes drastically heat the electrons [25]. Extensive Monte Carlo calculations of electron-impact-induced transitions between highly excited Rydberg states provide accurate rate coefficients [26]. For moderate energy changes, the calculations confirm the widely applied expressions [27] but reveal strong deviations at small energy transfer. Simulations of ultracold plasmas demonstrate that these corrections significantly impact the short-time dynamics of three-body Rydberg atom formation. The improved rate coefficients yield quantitative agreement with recent ultracold plasma experiments. The papers [24–26] address to the final ultracold plasma parameters and do not consider the initial stage when collisional recombination and disorder-induced electron heating could be competitive relaxation processes.

Disorder-induced heating is the relaxation process when potential energy associated with the initially disordered state is converted into kinetic energy of charged particles and therefore correspondingly higher particle temperatures [19, 20, 28].

All the approaches [12–20] are based on the essential approximations; their results do not agree with each other, and there are no reliable expressions to calculate the collisional recombination rate in SCP. We use the fluctuation approach and MD method [29, 30] to study the process of collisional recombination in SCP and to obtain the rate of the process for a certain range of nonidealities and a number of ion charges.

2. The model of the process

The plasma model [30] is used, in particular Coulomb potentials for charge–charge interactions with the cutoff $(-E_0)$ at $r < r_0 = e^2/E_0$ for the electron–ion interaction. An additional MD algorithm is developed to study relaxation [31]. It consists of two parts. The first part is preliminary: a certain procedure of excluding pairs from the equilibrium plasma is repeated several times to create an initial state without pairs. The second part is the main one: the

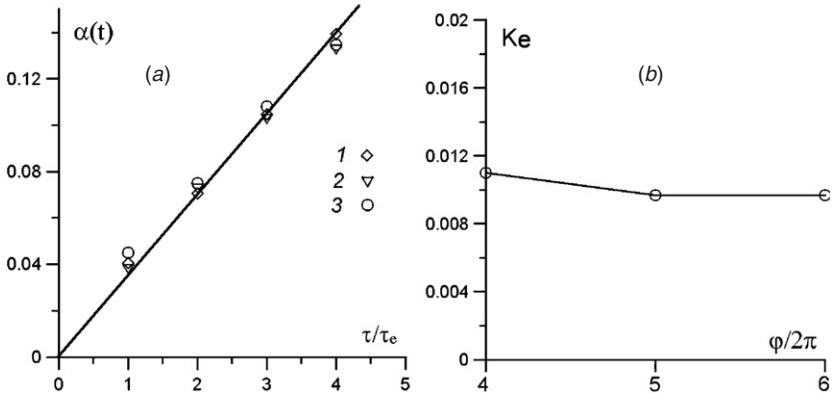

Figure 1. Tests of the correctness of the simulation procedure used. (*a*) Initial part of $\alpha(t)$ for different values of E_0: 1–2.5 eV, 2–5 eV , 3–7.5 eV for $T = 2$ eV, τ_e, the period of the plasma oscillation, (*b*) dependence of the K value on φ_0.

initial state created is used to start the relaxation MD run and to define the initial rate of the pair appearance according to the expression

$$K = \mathrm{d}\alpha/\mathrm{d}t|_{t=0}, \tag{1}$$

where α is the fraction of pairs in the system, and K is the frequency of pair production or the collisional recombination rate. To identify the electron–ion pairs, which can be associated with the atoms, the algorithm is used which is described in [29]. An example of the simulation is presented in figure 1(*a*) which points to the E_0 independence of $\alpha(t)$.

Another parameter used in the simulation is the minimum rotation phase φ_0 (figure 1(*b*)) which is the parameter that limits the minimum lifetime of a pair that can be considered as bound (see [29]). The rotation phase φ_0 is an integral angular velocity of the electron around the ion. The value $\varphi_0 = 8\pi$ is chosen in [29, 30] from the quantum consideration. The φ_0 and E_0 independences of K testify the correctness of our simulation procedure and physical significance of the K values obtained.

Independence of E_0 has a deep physical sense as well. First, it reveals the fact that the bottleneck for the collisional recombination is the energy region above $(-E_0)$ adjoining to the ionization limit. Second, as the energy region mentioned corresponds to the highly excited levels which are hydrogen-like for any atom, one is able to expect that our results for K can be applied to complex atoms.

3. Nonideality and charge dependences

The MD results are presented in figure 2 for the ion charges $Z = 1$–4 and a certain range of the electron nonideality parameter $\Gamma = (4\pi n_e)^{1/3}(e^2/k_BT)$, where n_e is electron number density. The more the Z value, the more time-consuming is the calculation since the number of electrons increases in order not to decrease the number of ions in the MD box. Therefore, the results become less representative with the increase of Z. Nevertheless, it is evident that $K\tau_e$ passes the maximum about 0.03 for all Z. τ_e is the period of plasma oscillations.

The less the Γ value the more time consuming is the calculation since it is necessary to increase the number of particles in the MD box. The MD results for small Γ available agree

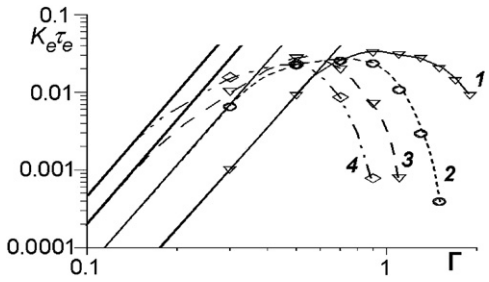

Figure 2. Dependence of the recombination rate on the nonideality parameter at the different values of the ion charge. 1: $Z = 1$, 2: $Z = 2$, 3: $Z = 3$, 4: $Z = 4$. Solid lines are the rate of three-body recombination (2).

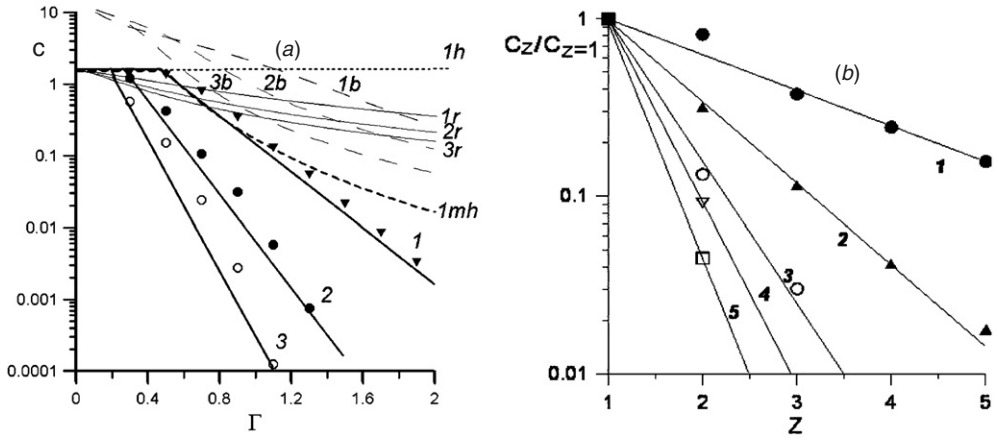

Figure 3. Different projections of the function $C(\Gamma, Z)$. (*a*) Dependences on Γ at the different values of the ion charge: 1: $Z = 1$, 2: $Z = 2$, 3: $Z = 3$; the line $C =$ const and exponential fit used in (5) are drawn through the MD points; the letter 'b' is related to the results of [13], the letter 'r' is related to the results [17], the letter 'h' is related to the original results [16], the letter 'mh' is related to the modified results [16]. (*b*) Dependences on the ion charge at the different values of nonideality: 1: $\Gamma = 0.3$, 2: $\Gamma = 0.5$, 3: $\Gamma = 0.7$, 4: $\Gamma = 0.9$, 5: $\Gamma = 1.1$.

with the rate of the three-body recombination for ideal plasma [1, 2, 32]:

$$K = C \cdot Z^3 e^{10} m^{-1/2} \cdot {n_e}^2 T^{-9/2}. \tag{2}$$

The value, $C = 1.4$, is chosen to fit the MD data. It agrees with the range $C = 1$–3 [1]. The agreement of the MD data with the limiting case (2) on both absolute value and functional dependence points to the reliability of the approach developed. If we allow the coefficient C to become a function $C(\Gamma, Z)$ and use that freedom to fit the calculated recombination rates for a SCP, we can reveal the influence of nonideality in a more apparent way. The results are presented in figure 3. The constant value $C(\Gamma, Z) = 1.4$ remains valid for $\Gamma < 0.5$ at $Z = 1$ (figure 3(*a*)). The Γ interval of the constant $C = 1.4$ reduces with increase of Z. The drop of C at the larger nonidealities can be approximated by

$$C = A(Z)\, \mathrm{e}^{-\lambda(Z)\Gamma}. \tag{3}$$

Introduction of functions $A(Z)$ and $\lambda(Z)$ permits to separate Z and Γ dependences. The Z dependences of $C(\Gamma, Z)$ for a number of Γ values (figure 3(*b*)) is approximated by

$$C = B(\Gamma)\,\mathrm{e}^{-\mu(\Gamma)Z}, \tag{4}$$

where Z and Γ dependences are separated again. Combination of (2)–(4) results in

$$K\tau_e = K_0 Z^3 \Gamma^{9/2}\,\mathrm{e}^{a_1 Z}\,\mathrm{e}^{-\lambda_0\Gamma}\,\mathrm{e}^{-\lambda_1 Z\Gamma}, \tag{5}$$

where $K_0 = 2.7$, $a_1 = 0$, $\lambda_0 = 1.5$, $\lambda_1 = 3$.

4. Mechanism of the recombination

The change of the recombination mechanism manifests itself through the change of the Γ dependence on C. The initial $C = \text{const}$ part corresponds to the conventional Fokker–Planck process via highly excited atomic levels [32]. The 'gap' between pair bound states and free electrons begins to exceed the temperature for the larger nonidealities, since the value of the 'gap' ΔE is about $2\Gamma kT$ for $Z = 1$ [29, 30]. The 'gap' is filled by the many-particle fluctuations [30] and they block the Fokker–Planck diffusion. Another argument is figure 1(*b*) which shows that the energy region adjoining to the ionization limit is the bottleneck for the collisional recombination as is mentioned above.

Since the one-step jump across the 'gap' becomes the limiting recombination mechanism for SCP, the Γ dependence of the recombination rate is supplemented by the additional exponential terms in (5) which can be interpreted as a probability for an electron to overcome the 'gap'. Two effects can be distinguished. The first effect is a Z-independent one and is related to the rapid varying interaction in the electron subsystem. The microfields produced smash up the excited atomic levels and create short-living multi-electron localized states reducing the recombination rate. The Z-dependent effect is related to the overlapping of the Coulomb wells and formation of the relatively long-living multi-electron localized states with the same influence on the recombination rate. The effect cuts the $C = \text{const}$ part at lower values of Γ and increases the slope of the decay of the recombination rate with the increase of Z in figure 3(*a*).

The change of the recombination mechanism from the Fokker–Planck to the one-step jump process can be elucidated by the additional arguments derived from the analysis of the distribution of pairs over their lifetime [29, 30] shown in figure 4. It has two peculiarities: exponential decay and strong energy dependence of the lifetime. The combination of these two features can be proven only if the pair energy is almost constant during its lifetime and its decay is a result of one strong collision with an electron. Let $\phi(t, E)$ be the probability that a pair with the initial energy E lives for the time t, then

$$\phi(2t, E) = \phi^2(t, E). \tag{6}$$

It follows from the exponential character of the decay. Let $p(E', t|E)$ be the probability of the transition from the state with energy E to the state with energy E' for the time t. If the process is a Markovian as is the case for the Fokker–Planck diffusion,

$$\phi(2t, E) = \int_\Delta p(E', t|E)\phi(t, E')\,\mathrm{d}E', \tag{7}$$

where Δ is the energy interval of the pair stability [29, 30]. The interval corresponds with the energies less than the restriction energy of pair states in plasmas. Besides,

$$\int_\Delta p(E', t|E)\,\mathrm{d}E' = \phi(t, E). \tag{8}$$

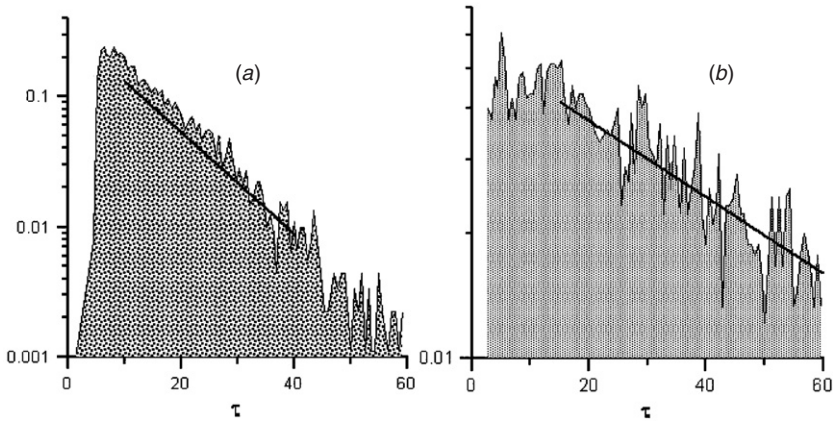

Figure 4. Distribution of pair fluctuations over lifetime at two binding energies. (*a*) −3 eV, (*b*) −6 eV for $T = 1$ eV.

Substituting (6) in (7) and subtracting (8) multiplied by $\phi(t, E)$, we obtain

$$\int_{\Delta} p(E', t|E)(\phi(t, E') - \phi(t, E))\,\mathrm{d}E' = 0, \tag{9}$$

which is valid for arbitrary values of E and t. Therefore, $p(E', t|E)(\phi(t, E') - \phi(t, E)) \equiv 0$. Because of the strong energy dependence of the lifetime $\phi(t, E) \neq \phi(t, E')$ at $E \neq E'$. Therefore, $p(E', t|E) = 0$ at $E \neq E'$. It means that the Fokker–Planck diffusion does not take place during the pair lifetime for the pair decay. It is the case for the recombination as well due to detailed balance principle.

The Fokker–Planck diffusion is no more the case in SCP and the gap becomes the governing factor for the recombination. The reasons are now evident for the drastic discrepancy between the estimations [13, 17] and MD results in figure 3(*a*). The authors [13] treat recombination as the Fokker–Planck process; both [13–15] and [17] do not take into account the 'gap' formation in SCP.

The influence of the highly excited atomic level cutoff on the three-body recombination rate was estimated in [16]. It is equivalent to the 'gap' in a certain sense. However, the direct application of Hahn's expression contradicts to the MD results: the K value remains constant within the Γ range in figure 3(*a*) (line 1h) and the decrease of K starts only for $\Gamma > 2$. However, if we substitute Hahn's recipe of the cutoff by $2\Gamma kT$ cutoff [29] in his expression for K the resulting values occur in agreement with the MD results in both absolute values and relative dependences for $\Gamma < 1$ (line 1mh in figure 3(*a*)). It is an additional argument in favor of our treatment of the recombination in SCP.

It is shown in [30] that there is no 'gap' in the electron density of states as was suggested in [12]. However, a 'gap' does exist between pair states and collective states of free electrons. The 'gap' is filled by the many-particle fluctuations [30] what is sufficient to block the Fokker–Planck diffusion when the 'gap' value exceeds the temperature as figure 3(*a*) shows. It points to the reason why and when one is able to exclude merged levels above the cutoff from the recombination process, what was postulated in [16]. The cutoff value, which was uncertain in [16], is also determined.

5. Comparison with experiments

The Killian–Rolston experiment [9] is the first where the collisional recombination rate is measured for SCP. It is discovered that only about 20% of ions recombined for 10^2 μs in the plasma with the electron number density 10^{10} cm^{-3} and temperature 1 K. It gives the estimation of the recombination rate as $2.3 \times 10^{-6}\tau_e^{-1}$ in agreement with the value $2.6 \times 10^{-6}\tau_e^{-1}$ which follows from (5). This comparison is only of qualitative value, since some subsequent theoretical and experimental papers on ultracold plasmas [24–26] have shown that the electron temperature varies during the evolution of the plasma and the electron Coulomb coupling parameter rapidly (within the first microsecond) goes to about $\Gamma = 0.2$. However, if the traditional expression for the recombination rate was valid for plasma obtained in [9], the value of the recombination rate of $2.3 \times 10^{-6}\tau_e^{-1}$ would give the temperature about 80 K. The authors [9] reports the temperature about several degrees K in agreement with [19, 20]. The computer simulation [19, 20] shows that disorder-induced heating is the main process at the initial relaxation stage. The subsequent recombination is rather slow and the authors [19, 20] relates their result to the suppression of the recombination rate predicted in [16]. The latter result is discussed in section 3.

Another indirect relevance of the MD data obtained can be derived from the experimental study of the initial stage of the latent track formation. The plasma model developed in [6–8] can give the self-consistent explanation of the transient x-ray emission observed only under assumption that the collisional recombination rate is much slower thanthat predicted by (2). The upper estimate derived agrees with (5).

6. Conclusions

Collisional recombination rate in SCP is calculated by the MD method in the frames of the fluctuation approach. Formula (5) is suggested which describes the numerical results for the whole range of Z and Γ studied and has a transparent physical meaning.

- A drastic suppression of the recombination rate is discovered with respect to the extrapolation of the conventional expression for the three-body recombination.
- The absolute value of the rate passes through a maximum with increase of nonideality and then decreases exponentially.
- The suppression is shifted to lower electron nonidealities with the increase of the multiplicity of ionization.
- The mechanism of the suppression is related to the formation of the 'gap' [30] adjoining to the ionization limit in the electron spectrum. The 'gap' substitutes the Fokker–Planck recombination process for the one-step jump across the 'gap' when the 'gap' begins to exceed the temperature.

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