

PROCESSES OF DECAY OF RYDBERG PLASMA

*E. A. Andreev^a, P. V. Kashtanov^{*b}, B. M. Smirnov^b*

^a*Semenov Institute of Chemical Physics, Russian Academy of Sciences
117977, Moscow, Russia*

^b*Institute for High Temperatures, Russian Academy of Sciences
127412, Moscow, Russia*

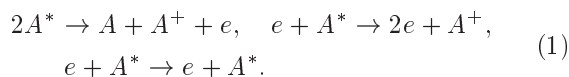
Received 24 May 2006

The lifetime of a molecule consisting of two Rydberg atoms with respect to electron release is determined from computer simulation of two classical electrons in the field of Coulomb centers. From this, the cross section of the Penning process of collision of two Rydberg atoms with an electron release is obtained. The rate constant for ionization of Rydberg atoms is evaluated for the Rydberg plasma within the Thomson model. On the basis of these processes, the kinetics is analyzed for the decay of a Rydberg plasma. Comparison with experimental data shows that these decay processes are responsible for the first stage of the decay of a magnetized and nonmagnetized Rydberg plasma located in a magnetic superconducting trap, whereas other processes become important at a subsequent stage of the plasma evolution.

PACS: 31.15.Gy, 34.60.+z, 52.20.-j, 52.65.Yy

1. INTRODUCTION

We consider a Rydberg plasma [1–3] that is initially an ensemble of highly excited atoms in almost identical states. In real experiments [4], these atoms are captured by a special trap and are located there until they decay. The decay of the Rydberg plasma proceeds according to the scheme



The goal of this paper is to determine the rate of the first process, the Penning process. We use computer simulation for two classical electrons located in the field of two motionless Coulomb centers.

Knowing the lifetime of this system or the width of its autoionization state allows analyzing the kinetics of the decay of a Rydberg plasma. In these evaluations, we are guided by parameters of the experiment in [4], where excited ^{85}Rb atoms in the state with the principal quantum number $n = 130$ are collected into a superconducting magnetic trap of the volume $V = 0.1 \text{ cm}^3$. The number density of excited atoms is $N_* = 10^6 \text{ cm}^{-3}$ and the atom temperature is 4 K. These parameters are

typical for such experiments. A strong magnetic field of the trap, $B = 2.9T$, strongly affects the kinetics of electrons, because the Larmor electron radius under these conditions is small, $r_H = 0.08\mu\text{m}$, and electrons are magnetized. This determines the special properties of the Rydberg plasma kinetics in a magnetic trap.

2. PENNING PROCESS INVOLVING TWO HIGHLY EXCITED ATOMS

We determine the parameters of the first process in (1) on the basis of computer simulation. We consider two electrons in the field of two Coulomb centers with a separation R between them (see Fig. 1). As the initial conditions, we take the distance r_1 of the first

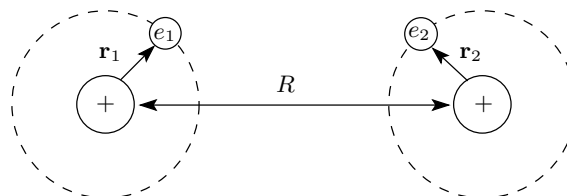


Fig. 1. Positions of electrons and cores for the interaction of two Rydberg atoms

*E-mail: kashtan@maryno.net

electron from the first Coulomb center to be equal to the distance r_2 of the second electron from the second Coulomb center,

$$r_1 = r_2.$$

We take the total energy of this classical system (which is conserved in the course of evolution) to be

$$U(\mathbf{r}_1, \mathbf{r}_2, R) = -\frac{e^2}{r_1} - \frac{e^2}{r_2} - \frac{e^2}{|\mathbf{r}_1 + \mathbf{R}|} - \frac{e^2}{|\mathbf{r}_2 - \mathbf{R}|} + \frac{e^2}{R} + \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} = -2J, \quad (2)$$

where J is the ionization potential of each atom. Throughout the paper, we use atomic units,

$$e^2 = \hbar = m_e = 1,$$

and let $J = \gamma^2/2$ denote the ionization potential in the atomic units; therefore, $n = 1/\gamma$ is the principal quantum number of the electron if it is an integer.

With the initial conditions in (2), we study the evolution of this system at fixed nuclei by solving the Newton equations for electrons,

$$\begin{aligned} \frac{d^2\mathbf{r}_1}{dt^2} &= -\frac{\partial U(\mathbf{r}_1, \mathbf{r}_2, R)}{\partial \mathbf{r}_1}, \\ \frac{d^2\mathbf{r}_2}{dt^2} &= -\frac{\partial U(\mathbf{r}_1, \mathbf{r}_2, R)}{\partial \mathbf{r}_2}. \end{aligned} \quad (3)$$

In the course of evolution, the electrons move in the fields of cores and interact with cores and with each other. As a result of energy exchange, one electron can be released, and then Penning process (1) develops. We assume that this process occurs if the distance of one electron from the center of this system exceeds $3R$, i.e.,

$$r_1 + \mathbf{R}/2 \geq 3R$$

or

$$r_2 - \mathbf{R}/2 \geq 3R.$$

The time τ when this distance is attained is said to be the lifetime of this system. We collect the statistics for these lifetimes. In this evaluation, we find the lifetimes for a given distance R between the nuclei for 875 initial conditions. Arranging the values of these lifetimes in decreasing order, we find the survival probability P at each time, i.e., the relative number of cases for which $\tau \leq t$, as is demonstrated in Fig. 2 for $R = 1.61a_0/\gamma^2$, where a_0 is the Bohr radius. The survival probability $P(t)$ is approximated by the dependence $\exp(-t/\tau_P)$. Table 1 contains the average lifetimes τ_P of the system of two excited classical atoms for different distances between them together with the error due to this method

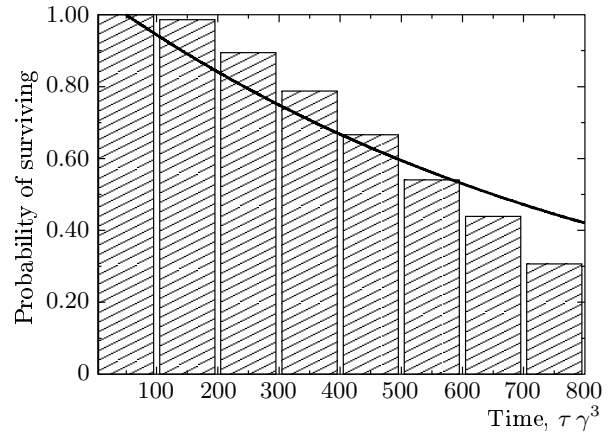


Fig. 2. Histogram for the time dependence of the survival probability for a system of two Rydberg atoms

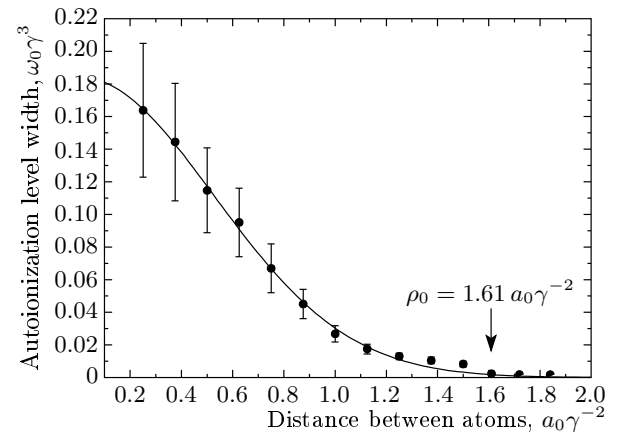


Fig. 3. The autoionization level width for a system of two Rydberg atoms as a function of the distance R between them. Dots are the results of computer simulation, the curve approximates these results by the dependence $\Gamma(R) = 0.18 \exp^{-1.81R^2}$

of finding the result. We note that the error increases as the distance between the nuclei decreases because of an increase in the relative time for an electron to reach the distance $3R$ when it is assumed to be free. Figure 3 shows that the dependence of the autoionization level width

$$\Gamma(R) = \frac{1}{\tau_P(R)}$$

is approximated by an exponential.

On the basis of these lifetimes τ_P for a system of two Rydberg atoms or the widths of the autoionization level $\Gamma = 1/\tau_P$ for this system, we find the cross section of a collision of two Rydberg atoms that leads to their decay. Assuming the trajectory of their collision

Table 1. The lifetime of an autoionization state of two interacting Rydberg atoms depending on the distance between them

$R\gamma^2$	0.25	0.50	0.75	1.00	1.25	1.50	1.61
$\tau\gamma^3$	6.1	8.7	15	37	77	120	430

to be a straight line, we express the probability of this process as

$$W(\rho) = 1 - \exp \left[- \int_{-\infty}^{\infty} \Gamma(R) \frac{RdR}{v\sqrt{R^2 - \rho^2}} \right],$$

where ρ is the impact parameter of the collision, v is the collision velocity, and R is the current distance between the nuclei. From this, we find the cross section of the Penning process involving two Rydberg atoms as

$$\sigma_P = \int_0^{\infty} W(\rho) 2\pi\rho d\rho.$$

We consider the case of a sharp dependence $\Gamma(R)$. The above cross section is then given by [5]

$$\sigma_P = \pi\rho_0^2, \quad \frac{\Gamma(\rho_0)}{v} \sqrt{\frac{\pi}{\alpha}} = e^{-C}, \quad (4)$$

where $C = 0.576$ is the Euler constant and

$$\alpha = d \ln \Gamma(R) / dR^2 \quad \text{at} \quad R = \rho_0.$$

Figure 4 gives the dependence of the cross section of this process on the collision velocity.

We also find the rate constant of the Penning process involving two Rydberg atoms as

$$k_P = \left\langle \sqrt{\frac{2E}{\mu}} \sigma_P \right\rangle,$$

where E is the relative energy and $\mu = m/2$ is the reduced mass of colliding atoms, with m being the mass of an individual atom, and brackets denote the average over the atom velocities. Assuming the Maxwell distribution of atoms over velocities and taking a weak dependence of the cross section σ_P on the collision velocity into account, we can represent this rate constant as

$$k_P = \sqrt{\frac{16T}{\pi m}} \sigma_P(v_P), \quad v_P = 2.24 \sqrt{\frac{2T}{m}}, \quad (5)$$

where T is the temperature of a Rydberg gas expressed in energy units. In particular, for the experimental conditions in [4], the rate constant of the Penning process is

$$k_P = 2.9 \cdot 10^{-4} \text{ cm}^3/\text{s}.$$

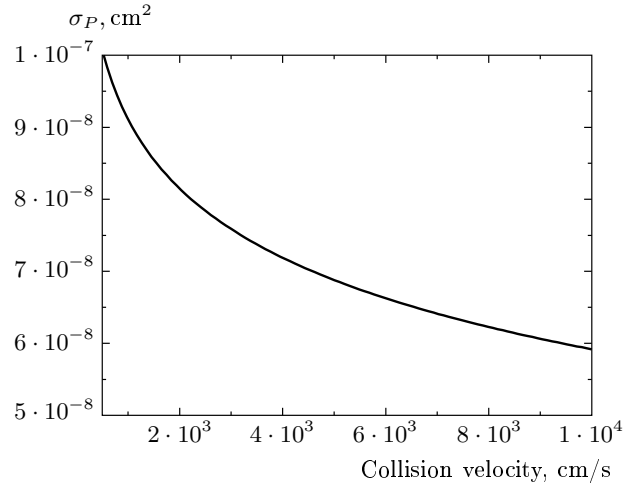


Fig. 4. The cross section of the Penning process involving two Rydberg atoms versus the collision velocity

3. SPECTRUM OF RELEASED ELECTRONS IN PENNING PROCESS

Free electrons resulting from Penning process (1) are important for the subsequent kinetics of the ensemble of Rydberg atoms in a Rydberg plasma. Hence, along with the width of the autoionization level for two interacting Rydberg atoms in the Penning process and the cross section of this process, the spectrum of released electrons influences the subsequent evolution of the Rydberg plasma. We therefore analyze the spectrum of released electrons for a fixed distance between two interacting Rydberg atoms.

Because of the nature of this process, the distribution function $f(\varepsilon)$ of released electrons can be expected to be a monotonic function of the electron energy ε . Figure 5 gives histograms for spectra of released electrons for different fixed distances between Rydberg atoms. Approximating these spectra by a simple dependence

$$f_0(\varepsilon) \sim \omega e^{-\beta\varepsilon},$$

where ω is a normalization constant. We give the values of the parameters β and ω at some distance between Rydberg atoms in Table 2. In addition, we determine the accuracy of the above approximation for the statistics used from the correlation function

$$\Delta = \frac{\int_0^{\infty} [f(\varepsilon) - f_0(\varepsilon)]^2 d\varepsilon}{\int_0^{\infty} f^2(\varepsilon) d\varepsilon}.$$

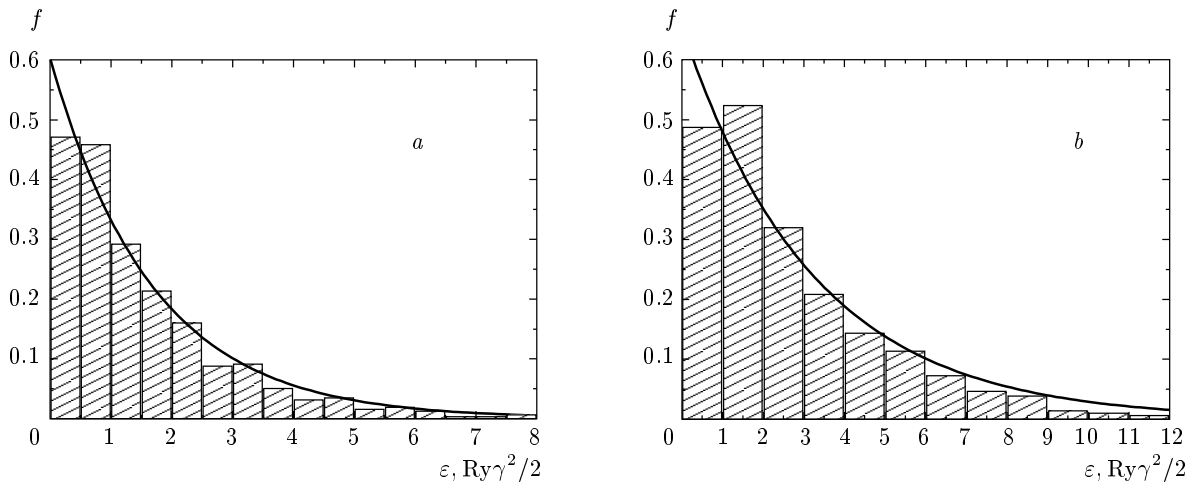


Fig. 5. Spectrum of released electrons at the distances $R = 1.00/\gamma^2$ (a) and $1.61/\gamma^2$ (b) between Rydberg atoms

Table 2. Parameters of the spectrum of released electrons as a result of the decay of an autoionization state of two interacting Rydberg atoms depending on the distance between them

$R\gamma^2$	0.25	0.50	0.75	1.00	1.25	1.50	1.61
$\beta\gamma^2/2$	0.57	0.73	0.70	0.60	0.50	0.71	0.62
$\Delta, \%$	1.70	0.12	4.50	1.30	4.00	3.90	3.20

4. COLLISIONS OF ELECTRONS AND RYDBERG ATOMS

When free electrons are formed in a Rydberg gas, they interact with Rydberg atoms effectively. Collisions between electrons and Rydberg atoms lead to ionization of Rydberg atoms or partial quenching of their excitation. Therefore, collisions involving electrons and Rydberg atoms may be important for kinetics of the decay of a Rydberg gas.

In analyzing these processes, we use the Thomson formula for the ionization cross section [6], which allows expressing the rate constant as

$$k_{ion} = \int_J^{+\infty} f(\varepsilon) d\varepsilon \sqrt{\frac{2\varepsilon}{m_e}} \frac{\pi e^4}{\varepsilon} \left(\frac{1}{J} - \frac{1}{\varepsilon} \right) = \mathcal{C} k_{max}, \tag{6}$$

$$k_{max} = v_{max} \sigma_{max} = \frac{2\sqrt{2} \pi e^4}{3\sqrt{3} m_e^{1/2} J^{3/2}},$$

where J is the ionization potential of the Rydberg atom, ε is the electron energy, m_e is the electron mass,

$$f(\varepsilon) = \omega e^{-\beta\varepsilon}$$

is the distribution function of the released electrons over energies, and the values v_{max} and σ_{max} correspond to the collision energy $\varepsilon = 3J$ at which k_{ion} reaches the maximum. In evaluating the factor \mathcal{C} in formula (6), we take the electron distribution function $f(\varepsilon)$ at the distance $R = 1.61/\gamma^2$ between Rydberg atoms, which makes the main contribution to cross section (4) of the Penning process. This gives $\mathcal{C} \approx 0.04$. We note that the accuracy of the Thomson formula is restricted, and we also assume that electrons do not thermalize when they ionize the Rydberg atom.

Returning to the conditions of the experiment in [4], we use formula (6) to find the ionization rate constant of a Rydberg atom by electron impact as

$$k_{ion} = 2.5 \cdot 10^{-3} \text{ cm}^3/\text{s}. \tag{7}$$

Evidently, this rate constant for ionization of Rydberg atoms by free electrons exceeds the rate constant of the Penning process,

$$k_P = 2.9 \cdot 10^{-4} \text{ cm}^3/\text{s}.$$

But if electrons are magnetized, we obtain an opposite relation between the rate constants of the electron impact and Penning process.

5. KINETICS OF THE RYDBERG PLASMA DECAY

We now analyze the evolution of a Rydberg plasma on the basis of the above rate constants for the processes in this system. Using processes (1), we have the following balance equations for the number densities of excited atoms N_* and electrons N_e :

$$\begin{aligned}\frac{dN_*}{dt} &= -k_P N_*^2 - k_{ion} N_e N_*, \\ \frac{dN_e}{dt} &= k_P N_*^2 + k_{ion} N_e N_*.\end{aligned}\quad (8)$$

If N_0 is the initial density of atoms, the number density of electrons formed at the end of the decay of the Rydberg gas is also N_0 , and

$$N_* + N_e = N_0$$

in the course of evolution. From this, for the number density of excited atoms, we have

$$\frac{dN_*}{dt} = -(k_P - k_{ion})N_*^2 - k_{ion}N_0N_*.\quad (9)$$

We note that in collisions with electrons, a change of an excited atom occurs. Assuming that a significant change of the atom excitation occurs at the last stage of the decay when ionization of excited atoms proceeds very rapidly, we ignore this change.

The energy of the electron subsystem is conserved in the course of evolution of a Rydberg gas. At the beginning, this energy is related to atom excitations. In the end of the evolution of the Rydberg gas, when the thermodynamic equilibrium is established, this energy is found in free electrons, and therefore the number density of electrons N_e and atoms N_a in the ground state at the end is determined by the Saha formula

$$\frac{N_e^2}{N_a} = K(T_e),\quad (10a)$$

$$\left(J_0 + \frac{3}{2}T_e\right) N_e = N_0(J_0 - J),\quad (10b)$$

where T_e is the electron temperature in thermodynamic equilibrium, J is the ionization potential of a Rydberg atom, J_0 is the ionization potential of an atom in the ground state, and $K(T_e)$ is the equilibrium constant for the Saha distribution. We note that in the thermodynamic equilibrium in an ideal plasma, which we consider here, the number density of excited atoms can be neglected in comparison with the total number density of electrons and atoms in the ground state [6, 7]. Because $J_0 \gg J$, we have $T_e \ll J_0$, and hence $K(T_e) \gg N_0$. From this, it follows that $N_e \approx N_0$, which allows us to transform expressions (10a) and (10b) to the form

$$N_a = \frac{K(T_e)}{N_0^2},\quad (11a)$$

$$N_a = N_0 \frac{\Delta\varepsilon + \frac{3}{2}T_e}{J}.\quad (11b)$$

This confirms the above assumption that

$$N_e \approx N_0, \quad N_a \ll N_0$$

at the end of the evolution process for a Rydberg gas. Thus, when a gas of Rydberg atoms reaches the equilibrium, almost all the initially excited atoms become ionized.

6. ANALYSIS OF THE EXPERIMENTAL RESULTS FOR THE DECAY OF AN ENSEMBLE OF RYDBERG ATOMS

We use the above expressions for the analysis of the experimental results in [4], where the parameters are $n = 130$ (the principal quantum number of a highly excited state), the initial number density of excited atoms is $N_0 = 10^6 \text{ cm}^{-3}$, the total number of excited atoms is equal to 10^5 , and the atom temperature is $T = 4 \text{ K}$. Under these conditions, the rate constants of processes (1) follow from formulas (5) and (6) as

$$k_P = 2.9 \cdot 10^{-4} \text{ cm}^3/\text{s}, \quad k_{ion} = 2.5 \cdot 10^{-3} \text{ cm}^3/\text{s}.$$

Below, we consider two limit regimes of the evolution of an ensemble of excited atoms. In the first regime, when a magnetic field is turned on, electrons are magnetized, and therefore the second and third processes in scheme (1) are absent. We can then take $k_{ion} = 0$ in balance equation (9), and its solution is given by

$$N_* = \frac{N_0}{1 + N_0 k_P t}.\quad (12)$$

In Fig. 6, this dependence is compared with the experimental one for a strong magnetic field.

We note that the dependence in (12) differs from the experimental one when the number density of Rydberg atoms decreases by several orders of magnitude. On the basis of the spectrum of final states of the Penning process, we can evaluate a change in the decay rate due to quenching processes because ionization of one atom in collisions of two Rydberg atoms is accompanied by quenching of the other colliding atom. But when this effect becomes essential, the decay of Rydberg atoms according to the experimental results is determined by another channel. We introduce this channel phenomenologically in balance equation (8), which now becomes

$$\frac{dN_*}{dt} = -k_P N_*^2 - \frac{N_*}{\tau},\quad (13)$$

where the parameter τ is responsible for the decay of Rydberg atoms in the second decay stage at large times,

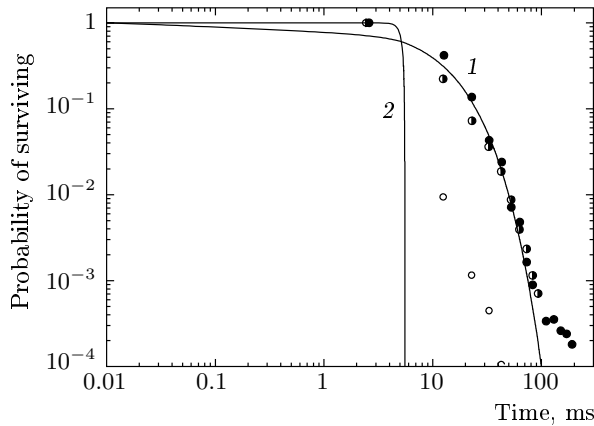


Fig. 6. Time dependences of the number density of Rydberg atoms at the experimental parameters in [4] according to experimental results and theoretical analysis. Black circles — experimental data, solid curves — theoretical data; the absence of a strong magnetic field — open circles and curve 2, the presence of a strong magnetic field — filled circles and curve 1

and experimental conditions are such that $\tau \approx 10$ ms. According to Eq. (13), this decay process may result from radiation of Rydberg atoms, from collisions with ions, and possibly with magnetized electrons if the decay products do not partake in a subsequent decay of Rydberg atoms.

We now consider the other limit case of decay of Rydberg atoms where the magnetic field is absent and the ionization of Rydberg atoms by electron impact dominates. Then the electron number density growth is determined by the second balance equation, which we analyze taking into account that $k_P \ll k_{ion}$. We find that in the first stage, when the electron number density is small, their decay is determined by the Penning process, and in accordance with formula (12), the electron number density is then given by

$$N_e = \frac{N_0^2 k_P t}{1 + N_0 k_P t}. \quad (14)$$

Therefore, in the case where $k_P \ll k_{ion}$, the Penning process gives seed electrons, and then their number density grows in accordance with the second equation in (8),

$$\frac{dN_e}{dt} = k_{ion} N_e N_*. \quad (15)$$

This equation describes the decay of Rydberg atoms starting from the electron number densities

$$N_e \sim N_0 \frac{k_P}{k_{ion}}.$$

This corresponds to a sharp variation in the electron number density in time, which is given in Fig. 6 for the experimental conditions. Although a typical time is in qualitative agreement with the experiment, the experimental decay curve is smoother, which testifies to a more complicated process occurring in reality.

7. CONCLUSIONS

The above analysis allows one to evaluate the rate constant of the Penning process involving two Rydberg atoms and to estimate the rate constant of the ionization of Rydberg atoms by electron impact if these electrons result from the Penning process. We obtain a qualitative agreement for typical times of decay of a Rydberg plasma due to the Penning process and ionization of highly excited atoms by electron impact. Comparison with experimental results shows that along with these two processes of the Rydberg plasma decay, additional processes are important at the late stage of the decay process.

This study is supported in part by the RFBR (grant № 04-03-32736).

REFERENCES

1. E. A. Manykin, M. I. Ozhovan, and P. P. Poluektov, *Sov. Phys. Doklady* **26**, 974 (1981).
2. E. A. Manykin, M. I. Ozhovan, and P. P. Poluektov, *Zh. Eksp. Teor. Fiz.* **57**, 256 (1983).
3. E. A. Manykin, M. I. Ozhovan, and P. P. Poluektov, *Chem. Phys. Rep.* **18**, 1353 (2000).
4. J. H. Choi, J. R. Guest, A. P. Povilus et al., *Phys. Rev. Lett.* **95**, 243001 (2005).
5. B. M. Smirnov, *Negative Ions*, Mc Graw Hill, New York (1981).
6. B. M. Smirnov, *Physics of Ionized Gases*, Wiley, New York (2001).
7. L. D. Landau and E. M. Lifshitz, *Statistical Physics*, vol. 1, Pergamon Press, Oxford (1980).