

Relaxation of Rydberg states in an ultracold plasma

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Abstract. The variations in the population densities of Rydberg states (with principal quantum number $n \sim 50 - 100$) in an expanding ultracold laser plasma ($N_e \sim 10^9 - 10^4 \text{ cm}^{-3}$, $T_e \sim 10 - 0.5 \text{ K}$) are calculated. The electron temperature measurement from the population densities of Rydberg states (T C Killian et al., Phys. Rev. Lett., vol. 86, p. 3759, 2001) is shown to be described by the conventional three-body recombination theory with the inclusion of recombination heating of free electrons.

Keywords: Rydberg states, laser plasma, three-body recombination.

1. Introduction

Our work was stimulated by the experiments performed in Refs [1–3]. The authors of paper [1] reported the production of an ultracold laser plasma with unique parameters: the electron density $N_e \sim 2 \times 10^9 \text{ cm}^{-3}$, the electron temperature $T_e \sim 0.1 \text{ K}$, the ion temperature $T_i \sim 10 \text{ K}$, and the degree of ionisation ~ 0.1 . The plasma was produced by the two-stage ionisation of metastable states of xenon precooled by laser radiation. It was also reported [1] that the plasma lifetime was anomalously long ($\sim 100 \mu\text{s}$) compared to the predictions of the three-body recombination theory.

In our papers [4–6], the experimental results [1] were analysed from the viewpoint of the relaxation theory of the isolated plasma summarised in reviews [7–10]. The observed lifetime was shown to be consistent with the unconventional concepts of these papers. An attempt to explain the anomalously long plasma lifetime observed in experiments [1] was also made in paper [11]. It was assumed that highly excited electronic states with large orbital angular momenta are not populated upon three-body recombination, resulting in a strong reduction in the recombination rate.

In a more recent experimental work [2], the expansion of an ultracold plasma bunch and the temporal variation of electron density during the expansion were studied. We analysed these data in papers [12, 13] using conventional concepts and considering the three-body recombination

moderation due to recombination heating. In this case, we proceeded from the following precedent. In the experiments [14, 15], the plasma bubble produced by laser evaporation of a metal target surface into gas was observed to glow for an anomalously long time. However, the recombination moderation under these conditions was attributed [15, 16] to the recombination heating of electrons. We showed [12, 13] that the measurements of the time dependence of the electron density performed in paper [2] do not permit us to unambiguously decide between one or other theory.

Measurements of the populations of Rydberg states (with principal quantum numbers $n \sim 50 - 100$) during the expansion of an ultracold plasma were reported in a recent paper [3], where the rate of recombination to the Rydberg states was found to be anomalously high compared to the predictions of the three-body recombination theory. In this paper, we analyse the experimental data [3] using a simple relaxation model of an expanding plasma. We showed that these data can be explained by the conventional three-body recombination theory taking the recombination heating of electrons into account.

2. Expansion of a plasma bunch

We analysed experiments [3] using the fact that the relaxation times of the relevant highly excited states were substantially shorter than the relaxation time of the density N_e and temperature T_e of free electrons (see, for example, Refs [17, 18]). This allows us to find the time dependences $N_e(t)$ and $T_e(t)$ taking into account the recombination and recombination heating using the plasma bunch expansion velocities measured experimentally [2, 3]. We determined the density of Rydberg atoms from the calculated $N_e(t)$ and $T_e(t)$ values and compared it with experimental data.

It was shown experimentally [2] that the time dependence of the average electron density is approximately described by the law of the particle density variation during the plasma expansion by neglecting recombination:

$$N(t) = \frac{n_0}{[4\pi(\sigma_0^2 + v_0^2 t^2)]^{3/2}}. \quad (1)$$

Here, n_0 is the total number of photoelectrons; σ_0 is the initial radius of a plasma bunch which has a Gaussian shape; and v_0 is the expansion velocity. Based on the data [8], the expansion velocity can be approximated by the following expression in a wide range of parameters investigated:

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Received 13 September 2001

Kvantovaya Elektronika 31 (12) 1084–1088 (2001)

Translated by E N Ragozin

$$v = \begin{cases} 30 - 50 \text{ m s}^{-1} & \text{for } E_e/k_B < 70 \text{ K} \\ (E_e/\alpha m_i)^{1/2} & \text{for } E_e/k_B > 70 \text{ K} \end{cases}, \quad (2)$$

where E_e is the initial energy of the electrons produced upon photoionisation; m_i is the xenon ion mass; $\alpha = 1.7$ is the fitting parameter; and k_B is the Boltzmann constant.

By using this fact, we can write the equations for the space-averaged values of the electron temperature and density as (cf. Refs [17, 18] and Ref. [8], p. 500, respectively)

$$\frac{dT_e}{dt} = \left(\frac{2}{3} \varepsilon^* + T_e \right) \frac{C}{T_e^{9/2}} N_e^2 - \frac{2}{3} \frac{f(t/t_0)}{t_0} T_e, \quad (3)$$

$$\frac{dN_e}{dt} = -\frac{C}{T_e^{9/2}} N_e^3 - \frac{f(t/t_0)}{t_0} N_e.$$

Here,

$$C = \frac{2^{9/2} \pi^{3/2} e^{10}}{45 m_e^{1/2} A(\gamma)} \approx 1.3 \times 10^{-7} \text{ cm}^6 \text{ K}^{9/2} \text{ s}^{-1} \\ \approx 1.7 \times 10^{-25} \text{ cm}^6 \text{ eV}^{9/2} \text{ s}^{-1}$$

is a constant that characterises the three-body recombination rate;

$$A(\gamma) = \begin{cases} (1/2) \ln[1 + 9/(4\pi\gamma^3)] & \text{for } \gamma < 0.5 \\ 1 & \text{for } \gamma > 0.5 \end{cases}$$

is the Coulomb logarithm; $\gamma \equiv e^2(2N_e)^{1/3}/T_e$; $f(\tau) = 3\tau \times (1 + \tau^2)^{-1}$ is the function that characterises the variation in the electron density and temperature during the plasma-bunch expansion; $\tau = t/t_0$; $t_0 = \sigma_0/v_0$ is the characteristic expansion time of the plasma bunch; and ε^* is the energy released in the electron gas in a single recombination event (see below).

Eqns (3) are valid for time intervals far shorter than the electron-ion energy exchange time

$$\tau_{ei} = \frac{m_i}{3m_e} \frac{3}{4(2\pi)^{1/2}} \frac{m_e^{1/2} T_e^{3/2}}{\Lambda e^4 N_e}.$$

As shown in papers [4–6], the initial electron temperature is not in fact very low. Even during the action of the laser pulse, the electrons manage to heat up to a temperature $T_{e0} \sim 4 - 5 \text{ K}$ due to collective interactions. For $N_e = 10^9 \text{ cm}^{-3}$ and $T_{e0} = 4 \text{ K}$, we have $\tau_{ei} \approx 200 \mu\text{s}$, and therefore the electron-ion collisions with energy exchange can be neglected at time scale of the order of the plasma bunch expansion time $t_0 \approx 5 \mu\text{s}$. Because the electron density decreases, this is also valid for later instants of time.

3. Relaxation of highly excited states

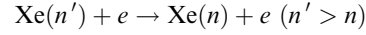
3.1 Transition rates

We will use the Kramers formula (see, for example, Ref. [19]) for the rates of spontaneous transitions between the hydrogen-like states characterised by principal quantum numbers n and n' :

$$A_{n,n'} \approx \frac{1.58 \times 10^{10} \text{ c}^{-1}}{n^3 n' (n^2 - n'^2)}, \quad (4)$$

$$A_n \approx \frac{\ln n}{n} 2.4 \times 10^{10} \text{ c}^{-1}.$$

For the rates $V_{m'}$ of collisional transitions



we use the interpolation Mewe formula [20]:

$$V_{m'} = \frac{1.58 \times 10^{-5} \text{ cm}^3/\text{c}}{T_e^{3/2}} \frac{F_{m'}}{\Delta(n', n, T_e)} \left\{ 0.15 + 0.28 \right. \\ \left. \times \left[\ln \left(1 + \frac{1}{\Delta(n', n, T_e)} \right) - \frac{0.4}{1 + 1/\Delta(n', n, T_e)} \right] \right\}. \quad (5)$$

Here, T_e is in electron-volts;

$$F_{m'} = \frac{32}{3\pi\sqrt{3}} \frac{1}{n'^2} \frac{n'^3 n^3}{(n^2 - n'^3)^3}; \quad \Delta(n', n, T_e) = \frac{\varepsilon_{n'} - \varepsilon_n}{T};$$

$$\varepsilon_n = \frac{\text{Ry}}{n^2}; \quad \text{Ry} = \frac{m_e e^2}{2\hbar^2} \approx 13.6 \text{ eV}.$$

The rates of direct and inverse transitions $n' \leftrightarrow n$ are related by the principle of detailed balancing:

$$V_{m'} = \frac{n^2}{n'^2} V_{n'n} \exp \left[\frac{\varepsilon_{n'} - \varepsilon_n}{T_e} \right]. \quad (6)$$

3.2 Rydberg state populations

According to experimental data [2, 3] and calculations [12, 13], the electron density and temperature vary within the following limits during the expansion time: $N_e \sim 10^9 - 10^4 \text{ cm}^{-3}$, $T_e \sim 10 - 0.5 \text{ K}$. The relaxation time of highly excited Rydberg states can be estimated by the expression $\tau_n \sim 1/(V_{m-1} N_e)$. For the above plasma parameters, $\tau_n \sim 10^{-4} - 0.3 \mu\text{s}$ for the level $n = 55$. However, τ_n is several orders of magnitude shorter than the characteristic time of the electron density and temperature variation at the corresponding instants of time. Therefore, the relaxation of the states can be studied in the quasi-steady-state approximation in which the level populations can be determined by solving linear algebraic equations, which depend on N_e and T_e parametrically (for more details, see, [17, 18]).

Furthermore, we show below that radiative transitions can be neglected for the states with $n \sim 50 - 100$ considered in paper [3]. Therefore, the so-called single-quantum approximation is valid for these states, which takes into account only the transitions between the nearest levels (for more details, see [8–10, 17, 18]). Within the framework of this approximation, the following expressions are valid for the excited-state populations:

$$N_n = N_n^{(B)} \xi_n(T_e), \quad (7)$$

where

$$N_n^{(B)} = 8\pi^{3/2} a_0^3 \left(\frac{\text{Ry}}{T_e} \right)^{3/2} n^2 N_e^2 \exp \left(-\frac{\varepsilon_n}{T_e} \right)$$

is the equilibrium population defined by the Saha–Boltzmann formula;

$$\xi_n(T_e) = \frac{\Sigma(n, T_e)}{\Sigma(\infty, T_e)}, \quad \Sigma(n, T_e) = \sum_{i=1}^{n-1} \frac{\exp(\varepsilon_i/T_e)}{2(i+1)V_{i,i+1}} \quad (8)$$

are the quantities characterising the deviation of the recombination distribution from the equilibrium one. This deviation becomes significant for the states with the binding energy of the order of the electron temperature. These states with $n \sim (\text{Ry}/T_e)^{1/2}$ are referred to as the bottleneck of three-body recombination. It determines the recombination flux.

The recombination flux, i.e. the number of recombination events per unit volume per unit time, in the single-quantum approximation is defined by the expression

$$\Gamma = \left(\frac{\text{Ry}}{T_e}\right)^{3/2} \frac{4\pi^{3/2}d_0^3N_e^3}{\Sigma(\infty, T_e)}. \quad (9)$$

In the range of the electron temperature and density under study, expression (9) gives virtually the same results as the expression $\Gamma = CN_e^3/T_e^{9/2}$ obtained on the basis of the diffusion approximation and employed in Eqn (3).

The total population of the Rydberg states with $n \sim 47$ –100, which were experimentally investigated, was determined by the summation:

$$N_{\text{Ry}} = \sum_{n=47}^{100} N_n. \quad (10)$$

Note that the population of the states lying below the bottleneck can be estimated by the expression

$$N_n \approx \frac{\Gamma}{V_{m+1}N_e} \propto N_e^2 T_e^{-4}. \quad (11)$$

Here, it was assumed that $V_{m+1} \propto T_e^{-1/2}$. Both the electron density and temperature decrease during the plasma expansion. It follows from expression (11) that the populations of Rydberg states depend on the electron temperature more strongly than on the electron density, resulting in the increase in the Rydberg-state population (see below).

3.3 Recombination energy release

The energy release per recombination event is commonly estimated by comparing the rates of radiative and collisional transitions. The approximate expression for the principal quantum number n^* defining the state for which the collisional and radiative relaxation rates become equal has the form [18]

$$n^* \approx 10^2 \left(\frac{T_e^{1/2}}{N_e \times 1 \text{ cm}^3} \right)^{1/9},$$

where T_e is expressed in electron-volts.

For $n > n^*$, the bound electron relaxes due to collisions with plasma electrons, and free electrons are heated. For $n < n^*$, the bound electron relaxes due to radiative transitions. Therefore, the energy released per one recombination event can be written as

$$\varepsilon^* = \text{Ry}/n^{*2} \approx 1.36 \times 10^{-3} \text{ eV} (T_e/1 \text{ eV})^{-2/9} \times$$

$$(N_e \times 1 \text{ cm}^3)^{1/9} \approx 44.7 \text{ K} \times (T_e/1 \text{ K})^{-2/9} (N_e \times 1 \text{ cm}^3)^{1/9}.$$

For the plasma obtained in the experiments [1–3], we have $n^* \sim 9$ –17 and $\varepsilon^* \approx 2000$ –500 K.

However, under the conditions considered, it is impossible to rigorously define the energy release per recombination event explicitly independent of the time. The matter is that the relaxation time of the quasi-steady-state population for $n = n^*$ is not so short as for $n \sim 50$ –100. For instance, for $N_e = 10^7 \text{ cm}^{-3}$ and $T_e = 5 \text{ K}$, we have $n^* = 11$ and $\tau_n \approx 10 \mu\text{s}$. For the states whose relaxation time is comparable with the plasma expansion time, the so-called excitation freezing occurs resulting in a decrease in the energy release per recombination event, because the excited atoms fly apart having no time to relax to stronger bound states [21, 22, 17].

Here, we will make a rough estimate of ε^* by neglecting the explicit time dependence ε^* . The recombination energy release determined by comparing the collision frequency of a highly excited electron with the inverse plasma expansion time t_0 (for more details, see Refs [12, 13]) has the form

$$\varepsilon^* \approx T_e \left(\frac{m_e^4 e^4}{2^8 \pi \hbar^6} \frac{1}{(N_e t_0)^2} \right)^{1/9}. \quad (12)$$

For instance, for $N_e \approx 10^9 \text{ cm}^{-3}$ and $t_0 \approx 3 \mu\text{s}$, we have $\varepsilon^* \approx 5.4 T_e$.

To adequately take into account the explicit time dependence of the energy release per recombination event in the plasma, is necessary to solve the transient equations for many excited states with $n \sim 10$ –30. This has no sense for the zero-dimensional expansion model. The consideration of a transient two-liquid one-dimensional model taking into account many excited levels involves great difficulties.

4. Results of calculations and experimental data

By using expressions (7) and (10), we reconstructed the dependence $T_e(t)$ from the experimental data for $N_{\text{Ry}}(t)$ [3] (Fig. 1). Fig. 2 shows the time dependences of the plasma parameters obtained by solving the zero-dimensional system of equations (3), the results of measurements of $N_e(t)$, and also $T_e(t)$ calculated from measured $N_{\text{Ry}}(t)$ values.

As before [12, 13], the time dependence of the electron density does not allow us to determine the intensity of recombination and, accordingly, the theory to which it corresponds. However, the time dependence of Rydberg state populations $N_{\text{Ry}}(t)$ allows a conclusion that the conventional recombination theory is in rather good agreement with the experimental data even within the framework of the coarse model employed here. The time dependence of the electron temperature is in qualitative agreement with the results of calculations using the zero-dimensional expansion model.

Note, however, that the model that we used cannot describe the nonmonotonic dependence of the electron temperature for $t \approx 20 \mu\text{s}$. This can be explained by the fact that we neglect the explicit time dependence of energy release per recombination event (see above). Note in this connection that the nonmonotone time dependence of the electron temperature related to the transient character of the

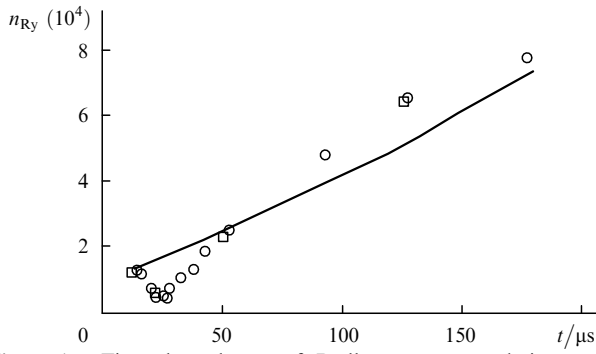


Figure 1. Time dependences of Rydberg state populations $n_{\text{Ry}} = N_{\text{Ry}}[4\pi(\sigma_0^2 + v_0^2 t^2)]^{3/2}$: experimental data of Ref. [3] (\circ); calculation for the electron temperatures obtained from the Rydberg-state population distributions over the principal quantum number n at different instants of time (\square , see Fig. 3); Rydberg-state populations corresponding to the time variation of the electron density and temperature obtained by solving zero-dimensional model equations (3) (the solid curve).

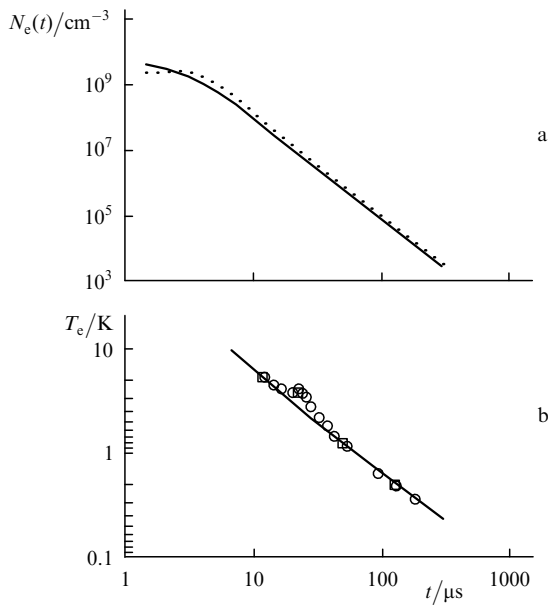


Figure 2. Time evolution of plasma parameters: the electron density in the expansion with [Eqn (3), the solid curve] and without [Eqn (1), the dotted curve] recombination (a); the electron temperature (the solid curve), the electron temperature reconstructed from the experimental data for the population distributions over the principal quantum number n at different instants of time (\square) and for the Rydberg state populations (\circ) for the initial parameters $\sigma_0 = 180 \mu\text{m}$, $v = 60 \text{ m s}^{-1}$, $N_{e0} = 2.7 \times 10^9 \text{ cm}^{-3}$, $T_{e0} = 6 \text{ K}$, $E_e = 9 \text{ K}$ (b).

energy release is known for conventional plasmas. It was predicted in paper [22] by simulating the helium plasma afterglow and later confirmed experimentally [23].

The Rydberg-state population distributions over the principal quantum number n at different instants of time were measured in paper [3]. The electron temperature was determined at instants of time by comparing these distributions with the recombination distribution (Fig. 3). The electron density for the recombination distribution was taken from the data of Fig. 2. The electron temperature was taken to provide the agreement between calculations and the experimental data for the states with $n \sim 55 - 75$.

The point is that the measuring technique used in paper [3] is most reliable for deep states. The electric-field pulse,

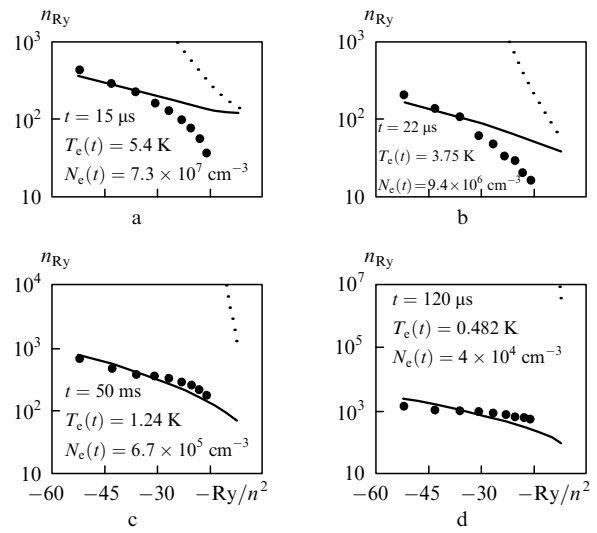


Figure 3. Population distributions n_{Ry} of Rydberg states over energy $-\text{Ry}/n^2$ at different instants of time calculated for given plasma parameters $N_e(t)$ and $T_e(t)$ (solid curves), Saha-Boltzmann distributions for these plasma parameters (dotted curves), and experimental points.

which ionised Rydberg states was supplied after prior application of a 5–10- μs pulse with the peak strength $F = 6 \text{ V cm}^{-1}$, which removed free electrons from the plasma bunch. This pulse should not only extract electrons, but also ionise the Rydberg states with $n = [(5.14 \times 10^9 \text{ V cm}^{-1}) \times (9F)^{-1}]^{1/4} = 99$. The collisional relaxation times for the states with $n \approx 100$ are short ($\tau_n \sim 10^{-6} - 10^{-2} \mu\text{s}$), and therefore the populations of not only the $n \approx 100$ state, but also of deeper states are lowered during the action of the prepulse.

The electron temperatures calculated from the Rydberg state population distributions over the principal quantum number n are in qualitative agreement with the results of calculations using the one-dimensional model [Eqn (3)] and are in excellent agreement with the electron temperatures calculated from the total Rydberg state populations (see Figs 2 and 3). The authors of paper [3] believe that there occurs an anomalously rapid three-body recombination to the Rydberg states with $n = 55$ under their experimental conditions. This conclusion is based on the approximate expression for the rate of variation of Rydberg state populations [3, 24]

$$\frac{dN_n}{dt} = 4.6 \times 10^{-38} \text{ m}^6 \text{ K s}^{-1} \left(\frac{1\text{K}}{T_e} \right) \times N_e^2 n^6 \left[1 - \exp\left(-\frac{\varepsilon_n}{T_e} \right) \right].$$

This expression takes into account only the events of three-body recombination directly to the state n and the events of ionisation of this state. The direct recombination to the $n = 55$ states is in fact very weak. However, these states are primarily populated not due to recombination events, but due to transitions from the adjacent states $n \leftrightarrow n \pm 1$. The rates of the $n \leftrightarrow n \pm 1$ transitions are several orders of magnitude higher than the rates of direct ionisation and direct recombination. This means that excited-state populations are established for the time $\tau_n \sim 1/(V_{n-1}N_e)$ far shorter than the recombination time $\tau_{\text{rec}} \sim N_e/F$. This

fact is well known from the theory of three-body recombination (see, for instance, [8–10, 17, 18]). The relaxation of highly excited states was studied in detail in paper [25].

The above discussion showed that the variation in the Rydberg-state population N_{Ry} follows the variation in the electron density and temperature. Because the dependence of N_{Ry} on T_e is stronger than on N_e (see above), N_{Ry} increases as the plasma expands. This fact is confirmed by comparing the results of calculations using the zero-dimensional model, which takes the recombination energy release into account.

5. Conclusions

Thus, the above calculations, which describe the variation in the Rydberg-state populations in an expanding ultracold laser plasma, show that the Rydberg-state populations measured in paper [3] are described by the conventional theory of three-body recombination taking into account the recombination heating of free electrons. Therefore, the assumptions that an ultracold plasma relaxes anomalously slow [1, 4–6, 11] or anomalously fast [3] should evidently be abandoned.

Acknowledgements. The authors thank T Killian for the discussion of the questions related to the experiments [1–3]. This work was supported by an ISTC Grant No. 1206.

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