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# Moderation of recombination in an ultracold laser-produced plasma

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Abstract. It is shown that the recent production [T C Killian et al. *Phys. Rev. Lett.* 83 4776 (1999)] of an ultracold laser plasma ( $N_e \sim 2 \times 10^9$  cm<sup>-3</sup>,  $T_e \sim 0.1$  K,  $T_i \sim 10 \mu$ K) can be considered as the first experimental demonstration of the metastable state of a supercooled plasma, which we predicted theoretically. Our theory provides an explanation for the moderation of three-body recombination observed in the above-cited paper. We present the calculations that simulate the conditions of the ultracold-plasma experiments.

# **1. Experimental results**

Killian et al. [1] reported the production of an ultracold laser plasma with unique parameters: the charge density  $N_{\rm e} \sim 2 \times 10^9$  cm<sup>-3</sup>, the electron temperature  $T_e \sim 0.1$  K, ion temperature  $T_{\rm i} \sim 10 \,\mu$ K, and the ionisation degree  $\alpha \sim 0.1$ . The plasma was produced by a two-stage ( $\lambda_1 \approx 882$  nm and  $\lambda_2 \approx 514$  nm) ionisation of the  $6s[3/2]_2$  metastable states of xenon.

The metastable Xe atoms produced in a discharge were slowed down employing the Zeeman deceleration technique, collected in a magneto-optical trap, and radiation-cooled on the  $6s[3/2]_2 - 6p[5/2]_3$  ( $\lambda_1 \approx 882$  nm) transition down to a temperature of ~ 10  $\mu$ K. The ionising photon energy ( $\lambda_2 \approx 514$  nm) was so selected that the electron detached in the photoionisation had a low kinetic energy  $E/k_{\rm B} = 0.1-1000$  K.

Such a plasma is strongly nonideal. The nonideality parameter  $\gamma$  of this plasma, equal to the ratio of the average potential energy of particles to their kinetic energy ( $\gamma = e^2/(aT)$ ), is rather large and equals  $\gamma_e = 34$  for the electrons and  $\gamma_i = 3.4 \times 10^5$  for the ions [e is the electron charge and  $a = (4\pi N_e/3)^{-1/3} \sim 5 \times 10^{-4}$  cm is the average distance between the charged particles]. According to the conventional theory of three-body recombination, a plasma with such parameters should decay instantly. The characteristic three-body recombination rate  $\tau_{\rm rec}^{(0)} \sim 0.3(m_e^{1/2}T_e^{9/2})/e^{10}N_e^2$  ( $m_e$  is the electron mass) is  $\sim 5 \times 10^{-16}$  s for  $N_e \approx 2 \times 10^9$  cm<sup>-3</sup>,  $T_e \approx 0.1$  K and  $\sim 2$  ns for  $N_e \approx 2 \times 10^8$  cm<sup>-3</sup>,  $T_e \approx 1$  K. However, the plasma lifetime observed in the experiments of Ref. [1] was many orders of magnitude longer, amounting to  $\sim 100$  µs.

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Received 12 October 2000 *Kvantovaya Elektronika* **30** (12) 1077–1079 (2000) Translated by E N Ragozin In our opinion, the experiments performed in Ref. [1] are of fundamental importance not only because of the unique plasma parameters achieved, but also for statistical physics and the theory of phase transitions.

# 2. Theoretical prediction of recombination moderation

The moderation of recombination in a supercooled nonideal plasma was first predicted in the late 1980s based on the analysis of the simulated behaviour of a great number of Coulomb particles (see reviews [2-5] and references therein). The analysis of numerical multiparticle dynamics (MPD) simulations revealed that a metastable state appears in the system, which is remote from the thermodynamic equilibrium in respect of the degree of ionisation and the subsequent relaxation is moderated. More precisely, the subsequent relaxation to the thermodynamically equilibrium state takes place to only the degree to which the time reversibility (time symmetry) of the numerical solution of dynamic equations is lost.

To verify these conclusions experimentally, it was proposed to produce a nonideal plasma bunch by ionising ions with laser radiation with the photon energy being close to the ionisation energy [6, 7]. The viewpoint that the recombination moderation can occur in a dynamic system received support in Ref. [8].

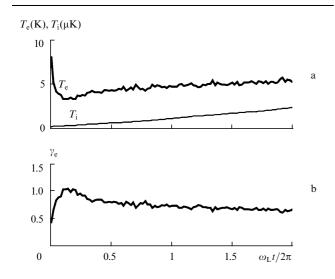
There are grounds to believe that it is the metastable state of a supercooled plasma predicted in our work that was realised in the experiments of Ref. [1]. Consider to what extent the results obtained agree with the theoretical predictions made earlier in Refs [2-5].

# 3. Initial stage of relaxation

Although in Ref. [1] the achievement of the electron temperature  $T_e \sim 0.1$  K was reported, experimental results were given only for  $T_e$  of the order of several Kelvins. This is unlikely to be accidental. The point is that electrons heat up, according to our results, due to collective interactions in a time  $t \approx 0.5\omega_L^{-1}$  [ $\omega_L = (4\pi e^2 N_e/m_e)^{1/2}$  is the Langmuir frequency]. During this period of time, the mixing of the phase trajectory of the multiparticle Coulomb system occurs, which is characterised by the Lyapunov index  $L \approx 2.4\omega_L$  [4, 5]. As a result of the mixing, the nonideality parameter  $\gamma$  is reduced to about 0.4 - 0.8.

We performed special MPD calculations by the method outlined in Refs [2, 3] to model the experimental conditions of Ref. [1]. Electrons and ions were treated, as before, as small charged spheres of diameter  $d \approx 0.05N_{\rm e}^{-1/3}$ . The charge density was  $2 \times 10^9$  cm<sup>-3</sup>. The initial conditions simulated the photoionisation of neutral atoms and were specified as follows. The initial coordinates of *n* ions (n = 512) were specified in the simulation range (a cube with the edge  $a = (n/N_{\rm e})^{1/3}$  and walls that reflect the particles specularly) with a probability density uniformly distributed over the cube volume. The initial ion velocities were specified in accordance with the Maxwell distribution for temperature  $T_{\rm i} = 10 \ \mu$ K. There was one electron per every ion in the simulation range (the initial coordinates of the ions and the electrons coincided), the electron velocities were uniformly distributed over directions, and their kinetic energy was set equal to the ionisation energy of a given pair of particles ('atom').

Then, the Newton equations for 2n particles were solved with the inclusion of all electrostatic interactions in the system and sufficient statistics was accumulated. The calculation showed that the electron thermalisation stage is preceded, for the given initial conditions, by the stage of escape from the potential wells (the descending portion of the temperature curve in Fig. 1a). Then, upon the onset of multiparticle interactions, the electrons thermalise, as we noted previously. Due to heating, in a time  $0.5\omega_L^{-1} \approx 0.2$  ns the electron temperature sets in at a level  $T_e \sim 3.3$  K, for which  $\gamma \approx 1$ . Then,  $T_e$  grows slowly, so that the average temperature  $T_e$  is 5 K.



**Figure 1.** Time evolution of  $T_e$  and  $T_i$  (a) and of the nonideality parameter  $\gamma_e = e^2/(aT_e)$  (b).

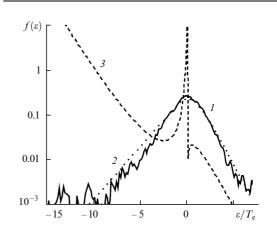
Note that the use of the classical approximation for the description of the motion of free particles is quite legitimate under the conditions of the experiments [1]. The parameter  $a(m_{\rm e}T_{\rm e})^{1/2}/\hbar$ , which characterises the ratio between the average interparticle distance and the de Broglie wavelength is high enough even for  $T_{\rm e} \sim 0.1$  K:  $a(m_{\rm e}T_{\rm e})^{1/2}/\hbar \sim 53$  [for  $T_{\rm e} \approx 5$  K we have  $a(m_{\rm e}T_{\rm e})^{1/2}/\hbar \sim 370$ ]. For ions and atoms,  $a(m_{\rm Xe}T_{\rm i})^{1/2}/\hbar \sim 260$  for  $T_{\rm i} \sim 10$  µK ( $m_{\rm Xe}$  is the mass of a xenon atom).

Thus, even if the laser-induced ionisation gives rise to electrons with a zero kinetic energy, the electron temperature of several kelvins should become settled in a time of less than a nanosecond. This is consistent with the data of Ref. [1].

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### 4. On the recombination mechanism

In MPD simulations [2-5], the electron velocity distribution function in the metastable state was shown to be Maxwellian. However, the distribution  $f(\varepsilon)$  of total electron energy is not the Boltzmann distribution. In the negative energy range  $\varepsilon < 0$ , the distribution  $f(\varepsilon)$  falls off exponentially as  $\sim \exp(-0.32|\varepsilon|/e^2N_e^{1/3})$ . This distinguishes radically the metastable state distribution from the Boltzmann distribution, which is characterised by an exponential growth. We obtained a similar result in this paper by simulating the experimental conditions of Ref. [1] (Fig. 2). The exponential decay of the total-energy electron distribution in the range of high negative energies is responsible for a significant recombination moderation.



**Figure 2.** Electron total-energy distribution function  $(2n = 1024, N_e = 2 \times 10^9 \text{ cm}^{-3})$ : the distribution in the metastable state obtained by averaging over a time  $\omega_L t = 3.1 - 12.4 (1)$ , microfield distribution calculated by the formulas of Refs [2-5] (2), and the Boltzmann distribution (3). The microfield and Boltzmann distributions were plotted for  $T_e = 3.6 \text{ K} - a$  value obtained by averaging the temperature over the time interval. The electrons were assumed to initially reside on the ions and possess the kinetic energy equal to the ionisation energy.

Earlier, we developed a recombination theory that is consistent with the results of simulations. The theory explains the absence of the effect of recombination moderation under usual conditions. The point is that for high-negative-energy electrons

$$\varepsilon < -\varepsilon_1 = \mathrm{Ry}\left(\frac{e^2 N_\mathrm{e}^{1/3}}{2\mathrm{Ry}}\right)^{2/3}, \ \mathrm{Ry} = \frac{m_\mathrm{e} e^4}{2\hbar^2} \approx 13.6 \ \mathrm{eV},$$
 (1)

the spectrum discreteness becomes important and the relaxation due to binary collisions, which are described by traditional kinetic models based on the principle of detailed balance, becomes dominant. The following expression for the recombination time was obtained in this case [3]:

$$\tau_{\rm rec} = \tau_{\rm rec}^{(0)} \xi, \ \xi = 1.82 \delta^{5/6} \xi_1 (\varepsilon_1 / T_{\rm e}) \xi_2 (N_{\rm e}) + 6.73 \delta^{7/6} (\xi_2 (N_{\rm e}) - 1).$$
(2)

Here,  $\tau_{rec}^{(0)}$  is the recombination time used in the conventional theory; and  $\xi$  is the correction factor;

$$\delta = \frac{2e^6 N_{\rm e}}{T_{\rm e}^2};$$

$$\begin{aligned} \xi_1(z) &= \frac{\exp z}{4z^{5/2}} \int_z^\infty dy y^{3/2} \left( 1 + 6y + 0.75y^2 + \frac{\pi y^3}{16} \right)^{1/2} \exp(-y); \\ \xi_2(N_{\rm e}) &= \exp\left[ \frac{-0.4(\varepsilon_1 - 1.5e^2N_{\rm e}^{1/3})}{2^{1/3}e^2N_{\rm e}^{1/3}} \right]. \end{aligned}$$

Expression (2) yields results close to those of the conventional three-body recombination theory in the range of nottoo-low temperatures ( $\xi \sim 1$  for  $T_e > 0.03 \text{ eV} \approx 350 \text{ K}$ ). For this reason, we paid earlier the main attention to the feasibility of producing a strongly supercooled ion-ion plasma wherein the role of quantum effects is insignificant. Attaining the parameters of an electron-ion plasma in the range where the recombination moderation is significant seemed hard to realise in experiments.

However, it was these parameters that were achieved in the experiments of Ref. [1]. Indeed, for  $T_e = 5$  K and  $N_e = 2 \times 10^9$  cm<sup>-3</sup>, we have  $\varepsilon_1 = 56$  K, and expression (2) yields a significant recombination moderation compared to the conventional theory:  $\xi = 2.4 \times 10^3$ . In this case, the characteristic  $\tau_{rec}$  following from our theory is 60 µs. For  $T_e = 5$  K and  $N_e = 10^9$  cm<sup>-3</sup>, we have  $\xi = 2.5 \times 10^3$ , and  $\tau_{rec} = 212$  µs, which is also consistent with the results of experiments [1]. Note that the radiative recombination under these conditions can be neglected because its time amounts to several seconds.

# 5. Conclusions

Thus, there are strong grounds to believe that Killian et al. [1] realised experimentally a supercooled plasma whose characteristic recombination time turned out to be several orders of magnitude larger than is predicted by the theories relying on the principle of detailed balance in its traditional form. The recombination moderation corresponds to the theory developed by us earlier. We believe that the recombination moderation will become far more significant as we pass to an ion-ion plasma.

Note especially the fundamental importance of the experimental realisation of the metastable state for the substantiation of statistical physics and the theory of phase transitions. The matter is that the difference between the traditional treatment and our approach is deep in nature and is not reduced to the difference in the kinetic models only (for more details, see Ref. [5]).

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