Formation kinetics and parameters of a photoresonant plasma

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The kinetics of a plasma produced by applying to a metal vapor pulsed resonant radiation whose frequency corresponds to the energy of a resonant transition in the atom is investigated. It is established that the principal ionization mechanism in such a plama, which ensures practically total ionization of the vapor at relatively low energy inputs, is stepwise ionization of the atoms by electron impact. Under conditions of above-equilibrium occupancy of the resonantly excited state, this process takes place at a constant electron temperature T_e. The calculated values of T_e agree with the measurement results. The possibility of producing a nonideal photoresonant plasma is analyzed; it is shown that the action of resonant radiation increases the nonideality parameter by 1.5 to 2 times compared with an equilibrium plasma. The parameters of the equilibrium plasma produced by relaxation of a photoresonant plasma are calculated. These parameters ($N_e \sim 10^{16}$–10^{17} cm^{-3}, $T_e \sim 0.4$–0.6 eV) are not reached if the plasma is produced by traditional methods. It is shown that an equilibrium plasma with arbitrary electron density can be produced by the photoresonance method.

1. A photoresonant plasma (PRP) is produced when a gas is acted upon by monochromatic radiation whose frequency corresponds to a resonance transition of the gas atom.[1] Even the first experiments[2] aimed at producing PRP and at investigating its properties have shown that this interesting physical object differs substantially from a plasma produced by traditional methods (gas discharge, charged-particle beam, etc.). Interest in the investigation of PRP has increased substantially in recent years in view of the advent of high-power tunable visible-light lasers, which are used to produce the PRP. Experiments performed with the aid of such sources[1] have shown that even relatively low-power laser emission suffices to obtain PRP of Na (Ref. 3), Li (Ref. 4), Cs (Ref. 5), Ba (Ref. 6), Mg (Ref. 7) and other vapors with quite high ionization. In contrast to a gas-discharge plasma, electrons in PRP are not subject to the accelerating action of an electric field, so that their temperature remains relatively low. By the same token, the resonant action of the radiation on the gas is a special method of producing a plasma having extremal physical properties, viz., high degree of ionization at a low electron temperature. Production of a supercooled plasma with close to one particle in the Debye sphere is reported in Ref. 8. Plasma with such parameters cannot be produced by other methods. The purpose of the present study was an investigation of the mechanisms and formation kinetics of a PRP and determination of the best attainable properties of this object.

2. We shall consider the PRP produced when metal vapor of density N is acted upon by monochromatic radiation of intensity J. For the sake of argument we consider the conditions of the experiment of Ref. 8, where Na vapor at a density 10^{16}–$4\cdot10^{17}$ cm^{-3} was exposed to 25-ns pulses of resonant radiation of intensity $6\cdot10^7$ W/cm^2, as a result of which PRP was produced with parameters $N_e \sim 10^{16}$ cm^{-3} and $T_e \sim 0.2$–0.3 eV. The cited paper contains a most complete description of the PRP parameters.

In the initial state of PRP production, the main source of the charged particles are the collisions between resonantly excited atoms, which either lead directly to formation of electrons and ions (associative ionization)[9]:

$$A^*+A^* \rightarrow A_2^+ + e, \tag{1}$$

or lead to formation of highly excited atoms[9]:

$$A^*+A^* \rightarrow A^{**}+A, \tag{2}$$

which undergo associative ionization

$$A^{**} + \left\{ \begin{matrix} A \\ A^* \end{matrix} \right\} \rightarrow A_2^+ + e \tag{3}$$

when colliding with normal or resonantly excited atoms.

At high substantially above-equilibrium density of the resonantly excited atoms, the processes (1)–(3) lead to a rapid growth of the electron density in the PRP. Thus, in the case of Na the rate constant of process (1) is $\approx 3\cdot10^{-11}$ cm^3/s (Ref. 9). Therefore the use of resonant radiation of saturating intensity ($J \gtrsim 10^2$–10^3 W/cm^2) at an Na vapor density $\sim 10^{16}$–10^{17} cm^{-3} ensures effective ionization at a level $\sim 10^{21}$ cm^{-3}/s. Thus, even in the first few tens of nanoseconds of the PRP lifetime the electron density in it exceeds $\sim 10^{13}$ cm^{-3}. This creates a more effective channel for the formation of charged particles, due to the stepwise ionization of the atoms by electron impact. In fact, the rate of formation of charged particle via associative ionization (1) does not depend on N_e, whereas the rate of stepwise ionization is proportional to N_e (we are considering the collisional-kinetics regime typical of the experimental conditions, when the excited atoms are destroyed mainly by electron-atom collisions, and the role of the radiative processes is negligible). The criteria for realization of such a regime have been discussed in detail, e.g., in Refs. 10 and 11. Rougly speaking, these criteria are of the form $T_e \ll \hbar\omega$, $N_e \gtrsim 10^{13}$–10^{14} cm^{-3} ($\hbar\omega$ is the atom-excitation potential). The electron density N_{el} above which the efficiency of stepwise ionization exceeds the efficiency of associative ionization is determined from the condition

$$N_{e1} \sim N_1 k_{ass}/k_{st},$$

where k_{st} is the rate constant of the stepwise ionization of the excited atoms. It will be shown below that $k_{st} \sim 10^{-7}$ cm^3/s. It follows hence that, for example in the case of Na, the main

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mechanism of charged-particle production, even at ionization degrees as low as $N_e/N \gtrsim 10^{-4}$, is stepwise ionization, and the features of its course determine in fact the character of the formation of PRP and its basic parameters.

3. Let us analyze the kinetics of PRP formation under conditions when the production and neutralization of the charged particles are connected with the stepwise ionization processes and with the detailed opposite process of triple recombination. Under the conditions of interest to us, when the main mechanism of destruction of the exciting atoms is via inelastic electron-atom collisions, the expressions for the rate constants of the indicated processes are of the form[11]

$$k_{st} = \frac{g_e g_i}{g_n} A \frac{e^{10}}{m^{1/2} T_e^{9/2}} \left(\frac{mT_e}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{I}{T_e}\right), \tag{4}$$

$$\alpha_{rec} = A \frac{e^{10}}{m^{1/2} T_e^{9/2}}. \tag{5}$$

The dimensionless parameter $A = 4 \pm 0.5$ is determined here by statistical reduction of a large number numerical model calculations, e and m are respectively the charge and mass of the electron, and I is the ionization potential of the atom. It should be noted that expression (4) is applicable not only to a description of the ionization of the atoms in the ground state, but also to a description of the ionization of the excited atoms (it is accordingly necessary to use in this case the ionization potential I_1 of the excited atom).

By virtue of the assumptions formulated above, the occupancies of the highly excited states of the atom are connected with the occupancy of the resonant level by the equilibrium Boltzmann relation, with an electron temperature T_e. To describe the kinetics of PRP formation is suffices therefore to consider the balance equations for the density of the atoms in the ground (N_0) and in the resonantly excited (N_1) states, and also for the temperature and density of the electrons. These equations are

$$\frac{dN_0}{dt} = -J\sigma\left(N_0 - \frac{g_0}{g_1}N_1\right) + \frac{N_1}{\tau} + N_e(N_1 k_{qu} - N_0 k_{exc}), \tag{6}$$

$$\frac{dN_1}{dt} = J\sigma\left(N_0 - \frac{g_0}{g_1}N_1\right) - \frac{N_1}{\tau} - N_e(N_1 k_{qu} - N_0 k_{exc}) - N_e(N_1 k_{ion}^{(1)} - N_e^2 \alpha_{rec}), \tag{7}$$

$$\frac{dN_e}{dt} = N_e(N_1 k_{ion}^{(1)} - \alpha_{rec} N_e^2), \tag{8}$$

$$\frac{3}{2}\frac{dT_e}{dt} = \hbar\omega(N_1 k_{qu} - N_0 k_{exc}) - \left(I_1 + \frac{3}{2}T_e\right) \times (N_1 k_{ion}^{(1)} - N_e^2 \alpha_{rec}). \tag{9}$$

Account is taken in Eqs. (6) and (7) of the transitions between the ground and resonantly excited states under the influence of the monochromatic radiation (σ is the cross section for such transitions), and also the transitions between the indicated states via electron impact:

$$A + e \leftrightarrow A^* + e. \tag{10}$$

The constants of processes (10) ar interrelated by the detailed balancing principle

$$k_{exc} = k_{qu} \exp(-\hbar\omega/T_e), \tag{11}$$

and the constant for quenching the resonant state by electron impact under the condition $T_e \ll \hbar\omega$ is independent of T_e and is given by[12]

$$k_{qu} = \frac{0.125 e^2 \lambda^{7/2}}{\hbar^{4} \tau 4\pi}\left(\frac{2m}{c}\right)^{1/2} \approx 1{,}1\frac{\lambda^{7/2}}{\tau}\left[\frac{\text{cm}^3}{\text{s}}\right], \tag{12}$$

where λ[cm] is the wavelength of the resonant transition, τ[s] is the radiative lifetime characterizing the considered resonant transition. Account is taken in the energy-balance equation (9) for the free electrons not only of the processes (10) but also of the stepwise ionization and recombination, which influence the change of the average electron energy.

Relations (4), (5), (11), and (12) are valid for a Maxwellian distribution of the electrons in energy, a distribution that obtains for the considered rather high degrees of plasma ionization. In addition, the approach developed is valid for the description of an ideal plasma in which the Debye sphere contains considerably more than one particle:

$$^4/_3\pi r_D^3 N_e \gg 1, \tag{13}$$

where $r_D = (T_e/8\pi e^2 N_e)^{1/2}$ is the Debye radius. It should be noted in connection with this requirment that the theory developed here cannot be used to determine the parameters of an ideal PRP if relation (13) is violated. It is possible, however, to determine on the basis of this theory the conditions under which the PRP is no longer ideal.

4. We have confined ourselves in the analysis of the solutions of the set of equations (6)–(9) to the most typical case of saturating laser-radiation intensity. In this case the relation between the occupancies of the ground and resonant states is:

$$N_0 = N_1 g_0/g_1, \tag{14}$$

and becomes valid before the collision processes come into play. Generalization of the results that follow to include the case of arbitrary intensity of the resonant radiation leads only to a certain complication of the calculations, but not to any qualitatively new physical conclusions.

We analyze first the character of the solution of Eq. (9). The initial condition for this equation should be chosen to be the value of the electron temperature T_{e0} determined from the energy of the electrons produced by associative ionization. It can be assumed that T_{e0} is close to zero. Since the ratio N_1/N_0 in PRP is substantially higher than the equilibrium value, the electrons in such a situation should be heated by quenching of resonantly excited atoms [the first term in the right-hand side of (9)]. A rise of T_e causes a rapid increase of the stepwise ionization intensity, characterized by a steeply increasing temperature dependence. Since the stepwise ionization process draws energy from the electrons, the temperature rise stops at a certain value T_{e1} and further increase of the electron density takes place at this quasistationary value of the temperature. This value is a solution of Eq. (9) in which it is assumed that $dT_e/dt = 0$ and only the principal electron heating and cooling mechanisms are considered:

$$\hbar\omega k_{qu} = (I_1 + {}^3/_2 T_{e1}) k_{ion}^{(1)}(T_{e1}). \quad (15)$$

We present below the quasistationary values of T_{e1} calculated on the basis of the solution of (15) with constants (4) and (12) for PRP of alkali metals:

Vapor	Li	Na	K	Rb	Cs
T_e, eV	0.45	0.37	0.34	0.32	0.30

The obtained electron temperature in PRP of Na (0.37 eV) is close to the measurement results (0.2–0.5, 0.35, and 0.35 eV in Refs. 7, 13, and 14, respectively) and exceeds somewhat the value of 0.2–0.3 eV of Ref. 8. The value $T_e = 0.08$ eV measured in Ref. 15 seems unjustifiably low. It may pertain to the initial stage of PRP formation, when quenching of resonantly excited atoms makes no noticeable contribution whatever to the electron heating. A strong overestimate $T_e \approx 1$ eV is obtained from the numerical calculations of Ref. 16, probably due to the neglect of the contribution of the stepwise ionization to the energy and electron-density balance, a contribution decisive under the considered conditions.

We shall use the normalization relation

$$N_0 + N_1 = N \quad (16)$$

(N is the total density of the atoms), where we neglect the contribution of all the excited states but the resonant one (this approximation is justified for an ideal plasma[11]), transforming Eqs. (8) and (9), with allowance for the detailed-balancing principle, into

$$\frac{dN_e}{dt} = N_e\left[\frac{\hbar\omega}{I_1 + {}^3/_2 T_e} k_{qu} N \frac{g_1}{g_1+g_0}\left[1 - \exp\left(-\frac{\hbar\omega}{T_e}\right)\right] - \frac{3}{2}\frac{dT_e}{dt}\frac{1}{I_1 + {}^3/_2 T_e}\right]. \quad (17)$$

The solution of this equation under the quasistationary conditions ($dT_e/dt = 0$) stipulated above is

$$N_e = N_{e0} \exp\left[\frac{\hbar\omega}{I_1 + {}^3/_2 T_e}\gamma t\right], \quad (18)$$

where $\gamma = k_{qu} N g_1/(g_1 + g_0)$. Thus, an exponential growth of the electron density in PRP takes place at a constant value of T_e. The characteristic time of this growth is $\gamma^{-1} \sim (k_{qu} N)^{-1}$ and for the typical experimental conditions of Ref. 8 ($N \sim 10^{17}$ cm^{-3}, $k_{qu} \sim 10^{-7}$ cm^3/s) it does not exceed several nanoseconds. It is just in this time that the energy of the resonant radiation is converted into the energy of the ionized gas. This explains also the repeatedly observed[1] experimental fact that the gas is rapidly and practically completely ionized by the resonant radiation if the transition is saturated.

The exponential growth of the electron density (18) at constant temperature continues until the ternary recombination, which keeps N_e from increasing further, becomes substantial. The characteristic time t_2 at which this occurs is estimated from the expression

$$N_e^2(t_2) = N \frac{g_1}{g_0+g_1}\frac{2g_i}{g_1}\left(\frac{mT_{e1}}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{I_1}{T_{e1}}\right), \quad (19)$$

where $N_e(t)$ is given by (18). The value

$$t_2 = \left(2\gamma\frac{\hbar\omega}{I_1 + {}^3/_2 T_{e1}}\right)^{-1} \ln\left[N\frac{g_1}{g_0+g_1}\frac{2g_i}{g_1}\left(\frac{mT_{e1}}{2\pi\hbar^2}\right)^{3/2} \times \exp\left(-\frac{I_1}{T_{e1}}\right) N_{e0}^{-2}\right] \quad (20)$$

obtained in this manner is substantially larger than $1/\gamma$. At $t > t_2$, determined by relation (20), a quasi-equilibrium state is established in the PRP. This state is described by the Saha relation between the electron density and the density of the resonantly excited atoms:

$$N_1 \frac{2g_i}{g_1}\left(\frac{mT_e}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{I_1}{T_e}\right) = N_e^2(t). \quad (21)$$

The $N_e(t)$ dependence is expressed here as before by the exponential (18), but the electron temperature no longer remains constant, but increases with time, since an additional electron-heating mechanism enters into the electron energy balance and its effectiveness is proportional to the square of the electron density. In this regime, the plasma becomes fully ionized almost instantaneously. This plasma state is of no interest to us, since a noticeable contribution to the absorption of the resonant radiation is made in this case by mechanism of collective interaction between this radiation and the plasma electrons, i.e., the PRP acquires features of a laser plasma.

5. Let us analyze the PRP properties caused by its deviation from ideal. These properties are characterized by a parameter[17] $\Gamma = e^2/r_D T_e$ that determines the ratio of the potential energy of the charged-particle interaction to their kinetic energy, and is inversely proportional to the number (13) of charged particles in the Debye sphere. In a PRP, where the electron density is connected by the Saha relation[21] with the above-equilibrium density of the resonantly excited atoms, the value of the parameter Γ is higher than in an equilibrium low-temperature plasma. Using relations (14) and (21) and the normalization relation

$$N_0 + N_1 + N_e = N, \quad (22)$$

we determine the maximum attainable values of the parameter Γ in a PRP. First of all, the expression for the electron density is of the form

$$N_e = \frac{nG}{2}\left[\left(\frac{4N}{nG} + 1\right)^{1/2} - 1\right], \quad (23)$$

where

$$n = \left(\frac{mT_e}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{I_1}{T_e}\right), \qquad G = \frac{2g_i}{g_0+g_1}.$$

At a fixed atom density N, the parameter Γ has a nonmonotonic dependence on the electron temperature and has a maximum at a certain value of T_e. This maximum Γ, calculated for PRP in Li and Cs vapor, is compared in Table I with the maximum value of Γ for an equilibrium plasma whose parameters are related by the equilibrium Saha equation. The data for other alkali metals lies in the interval between the corresponding values for Li and Cs. It can be seen that for a given vapor density the values Γ reached in a PRP are higher than in an equilibrium plasma. This is due to the lower value of T_e in a PRP having the same electron density as an equilibrium plasma.

6. We analyze now the properties of the plasma produced when a PRP relaxes to the equilibrium state when the

TABLE I. Comparison of the photoresonant- and equilibrium-plasma parameters for which the plasma parameter Γ is a maximum. The upper of the bracketed numbers corresponds to PRP, and the lower to an equilibrium plasma.

N, cm^{-3}	Li			Cs		
	Γ	T_e, eV	N_e, cm^{-3}	Γ	T_e, eV	N_e, cm^{-3}
10^{14}	0.0165	0.27	$7.4\cdot10^{13}$	0.027	0.19	$7.3\cdot10^{13}$
	0.011	0.37	$7.7\cdot10^{13}$	0.017	0.27	$7.7\cdot10^{13}$
10^{15}	0.041	0.31	$6.9\cdot10^{14}$	0.066	0.22	$6.8\cdot10^{14}$
	0.027	0.42	$7.4\cdot10^{14}$	0.042	0.31	$7.3\cdot10^{14}$
10^{16}	0.096	0.37	$6.3\cdot10^{15}$	0.16	0.26	$6.1\cdot10^{15}$
	0.066	0.49	$6.9\cdot10^{15}$	0.10	0.36	$6.8\cdot10^{15}$
10^{17}	0.22	0.44	$5.4\cdot10^{16}$	0.35	0.31	$5.2\cdot10^{16}$
	0.155	0.57	$6.2\cdot10^{16}$	0.24	0.43	$6.1\cdot10^{16}$
10^{18}	0.46	0.52	$4.3\cdot10^{17}$	0.73	0.38	$4.0\cdot10^{17}$
	0.35	0.68	$5.3\cdot10^{17}$	0.52	0.51	$5.1\cdot10^{17}$

action of the resonant radiation on the vapor is stopped. The characteristic time to establish equilibrium between the resonant and ground states of the atom in such a system is $\sim (N_e k_{qu})^{-1}$ and under the typical conditions of the experiment of Ref. 8 it turns out to be $\sim 10^{-9}$–10^{-8} s. As follows from Eq. (15), the time to establish ionization equilibrium is also of the same order. This time is shorter by 2–3 orders than the characteristic time of energy emission from the plasma volume, determined by the time of emission of the resonant radiation with allowance for strong dragging. Therefore the parameters of the PRP and of the equilibrium plasma that results from its relaxation are in a one-to-one relation, which we determine assuming a plasma-ionization degree $N_e/N \ll 1$ and neglecting, in accordance with the foregoing, the contribution of all but the resonant excited states of the partition function of the atom.

By virtue of the assumptions made, the main energy reserve in the PRP is contained in the resonantly excited atoms. This energy is equal to

$$N_1\hbar\omega \equiv N\hbar\omega g_1/(g_0+g_1).$$

As a result of relaxation of the PRP, this energy goes to raise the temperature of the electrons and to additional ionization of the plasma. The energy density in an equilibrium plasma is $N_e(I+(3/2)T_e)$. Equating the bulk reserve of energy in the PRP and in a the equilibrium plasma, and using the equilibrium Saha equation, we obtain the connection between the total vapor density and the electron temperature in an equilibrium plasma resulting from PRP relaxation:

$$N = \left(\frac{g_0+g_1}{g_1}\right)^2 \left(\frac{I+{}^3/_2T_e}{\hbar\omega}\right)^2 \frac{g_e g_i}{g_0}\left(\frac{mT_e}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{I}{T_e}\right). \quad (24)$$

The parameters calculated from (24) for an equilibrium plasma that results from relaxation of a weakly ionized PRP with saturated resonant transition are listed in Table II. The resonance level used in the calculation was the state $^2P_{1/2}$. As seen from a comparison of the data in Tables I and II, decay of the PRP results in an equilibrium plasma having a parameter Γ close to the maximum possible for an equilibrium plasma.

The equilibrium-plasma parameters listed in Table II were obtained under the assumption that the initial degree of ionization of the PRP is much less than unity. If this assumption does not hold, relaxation of a PRP results in an equilibrium plasma with a density higher than given in Table II, and hence also with a higher electron temperature. Specific values of these parameters, as functions of the initial degree of ionization of the PRP, can be easily calculated on the basis of the relation

$$N_1\hbar\omega + N_{e1}(I+{}^3/_2T_{e1}) = N_e(I+{}^3/_2T_e),$$

in which the initial electron density N_{e1} in the PRP is connected with the vapor density of the quasi-equilibrium relation (21). Thus, it becomes possible to produce a low-temperature plasma with an arbitrary electron density. This is achieved as a result of the relaxation of a PRP in which the variable parameters are the atom density and the initial electron density.

7. The analysis in this paper has shown that the PRP passes in the course of its development through a sequence of states, the parameters of each of which can be established with sufficient reliability in the limiting case of a saturated transition and of high electron density. The first of these states is characterized by an exponential growth of the electron density at a constant temperature whose values are given above. In this state, the energy of the resonant radiation is

TABLE II. Parameters of equilibrium plasma produced by relaxation of a PRP with saturated resonant transition.

N, cm^{-3}	Li			Cs		
	Γ	T_e, eV	N_e, cm^{-3}	Γ	T_e, eV	N_e, cm^{-3}
10^{14}	0.0082	0.30	$2.4\cdot10^{13}$	0.013	0.22	$2.5\cdot10^{13}$
10^{15}	0.021	0.34	$2.4\cdot10^{14}$	0.035	0.25	$2.5\cdot10^{14}$
10^{16}	0.054	0.39	$2.3\cdot10^{15}$	0.088	0.29	$2.5\cdot10^{15}$
10^{17}	0.13	0.46	$2.3\cdot10^{16}$	0.21	0.35	$2.4\cdot10^{16}$
10^{18}	0.32	0.55	$2.2\cdot10^{17}$	0.49	0.42	$2.4\cdot10^{17}$

effectively converted into plasma-electron energy. The PRP passes next into a quasi-equilibrium state in which all the excited levels are at equilibrium with the continuous spectrum, and the coupling of the resonant levels to the ground state is given by the saturation condition (14). The parameter of this state, which corresponds to the maximum value of the parameter Γ, are listed in Table I. Through the action of the resonant radiation the plasma in such a state is characterized by an above-equilibrium value of the electron density. Finally, when the action of the resonant radiation is stopped, the plasma relaxes to an equilibrium state whose parameters can be roughly estimated on the basis of (24) and are listed in Table II. It appears that the action of resonant radiation on vapor is the only means of producing an equilibrium plasma with so high an electron density at so low an electron temperature. The lifetime of such a plasma is determined either by the expansion time or by the time of emission of the resonant radiation. The latter, with account taken of the dragging of radiation, amounts under the conditions of the experiments of Ref. 8 to $\sim 10^{-6}$–10^{-5} s.

We were able to present here a consistent analysis of the PRP development kinetics because, in the considered range of plasma parameters ($N_e \sim 10^{15}$–10^{16} cm^{-3}, $T_e \approx 0.3$–0.5 eV) its behavior is determined by collisional processes whose characteristics are known with sufficient reliability. This pertains primarily to stepwise ionization and ternary recombination, as well as to the quenching of resonantly excited atoms by electron impact. At the lower electron density realized in PRP with low metal-vapor density, a noticeable role is assumed in the kinetics by radiative breakdown of the excited states, which leads to deviations of the atom distribution over the excited state from an equilibrium Boltzmann distribution. Description of the plasma behavior in such a situation requires a level-by-level kinetic theory whose results are highly sensitive to rate constants of the collisional transitions between the excited states of the atom. The large errors to which such constants are subject prevent us from describing a low-density PRP with as high an accuracy as obtained here for a dense plasma.

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