

THE DISTRIBUTION FUNCTION OF ATOMIC LEVEL POPULATIONS IN A PLASMA

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An analytic form is obtained for the population distribution function in an atomic plasma as a series in successive time derivatives of the population of the first level. The first approximation includes the well-known method of a stationary sink. The quasistationary distribution obtained for hydrogen agrees well with numerical calculations of recombination and ionization, and for lithium, helium, and argon the quasistationary distribution gives qualitative agreement with numerical calculations.

The determination of the population distribution function of the discrete levels in an atomic plasma is of great importance in problems of ionization, recombination, and emission of plasmas and also the design and optimization of atomic-level lasers. Its role has been especially emphasized by the results of molecular kinetics, in which quasistationary distributions over the vibrational levels of molecules have recently been obtained. These are the Treanor distribution [1] and various generalizations thereof. The simple analytic form of such distributions had made it possible to establish a number of new laws in vibrational kinetics and has been widely used in laser theory [2-4].

In atomic kinetics the situation is more complicated. Here there are two approaches. One of them is based on a numerical solution of the balance equations for the populations. Very frequently, one has used a numerical solution of the simplified equations in the approximation of "stationary sink" (see, for example, [5-8]).

In the other approach [9-12], the motion of an electron between levels is treated as a stochastic process like Brownian motion in the energy space, the motion being described by the Fokker-Planck equation. In [11, 12], the authors used the modified diffusion approximation of the Fokker-Planck equation in finite differences and were able to preserve the real discrete structure of the energy spectrum. While the first approach is inconvenient because it is so cumbersome, the second, although entirely satisfactory for upper levels, is not accurate for the lower levels.

In the present paper we propose an analytic solution of the kinetic relaxation equations of a hydrogen plasma and, under certain simplifying assumptions, any atomic plasma. This solution is based on an expansion of the distribution in a series in the time derivatives of the population of the first level. In principle, any approximation can be calculated analytically, but in the majority of the cases of practical interest the first approximation is sufficient. The corresponding quasistationary distribution has, in some fundamental features, similarities with the Treanor distribution for molecules.

1. Method of Analytic Solution of the System of Kinetic Equations

Let us consider the bases on which we solve the equations of atomic kinetics, taking as an example the relaxation of a hydrogen plasma. We introduce simplifying assumptions. For the free electrons we assume a Maxwellian distribution function with constant electron temperature T_e . Of the various elementary collision processes, we take into account collisions of the first and the second kind of atoms with

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electrons and we ignore the infrequent processes of atom-atom and atom-ion collisions and also ionization, radiation, and triple recombination. In addition, the plasma is assumed to be optically thin and spatially homogeneous.

These assumptions enable us to write down the system of equations for the populations of the discrete levels of a hydrogen plasma without allowance for radiation decay of the levels as follows:

$$\frac{dN_n}{dt} = -V(n, n+1)N_nN_e + V(n+1, n)N_{n+1}N_e - V(n, n-1)N_nN_e + V(n-1, n)N_{n-1}N_e \quad (1.1)$$

Here N_n is the population of the n -th level of the hydrogen atom, N_e is the electron density, and V are the probabilities of collisions of the first and the second kind of electrons and atoms, averaged over the electron Maxwellian distribution. After times comparable with the collision time between free electrons, equilibrium with the continuous spectrum described by the Saha formula is established for the highest levels:

$$N_n = n^2 N_e^2 \left(\frac{2\pi\hbar^2}{mT_e} \right)^{3/2} \exp\left(-\frac{R}{n^2 T_e} \right) \quad (n > n_0) \quad (1.2)$$

where R is the hydrogen ionization potential. The probabilities $V(m, m+1)$ of the inverse processes are related to the probabilities of the direct processes by the principle of detailed balance:

$$\frac{V(m, m+1)}{V(m+1, m)} = \frac{(m+1)^2}{m^2} \exp\left(-\frac{E_{m+1, m}}{T_e} \right), \quad E_{m+1, m} = E_{m+1} - E_m \quad (1.3)$$

To solve the system (1.1) we make the change of variables

$$\tau = \int_0^t N_e(t') dt', \quad d\tau = N_e dt \quad (1.4)$$

Summing the resulting equations from 1 to m , we obtain

$$N_{m+1} = \frac{V(m, m+1)}{V(m+1, m)} N_m + \frac{1}{V(m+1, m)} \sum_{n=1}^m \frac{dN_n}{d\tau} \quad (1.5)$$

The solution of the system (1.5) can be represented in the form

$$N_m = \sum_{i=0}^{m-1} \alpha_m^i N_1^{(i)} \quad (1.6)$$

where the populations N_m are determined by the derivatives of the populations of the first level $N_1^{(i)} = d^{(i)} N_1 / d\tau^{(i)}$ and the quantities α_m^i , which do not depend on τ . To determine α_m^i , we substitute (1.6) into (1.5):

$$\sum_{i=0}^m \alpha_{m+1}^i N_1^{(i)} = \frac{V(m, m+1)}{V(m+1, m)} \sum_{i=0}^m \alpha_m^i N_1^{(i)} + \frac{1}{V(m+1, m)} \sum_{n=1}^m \sum_{i=1}^n \alpha_n^{i-1} N_1^{(i)} \quad (1.7)$$

Reversing the order of summation in the second term in accordance with the formula

$$\sum_{n=1}^m \sum_{i=1}^n \rightarrow \sum_{i=1}^m \sum_{n=i}^m$$

and equating coefficients of corresponding derivatives, we obtain the recursion relations

$$\alpha_{m+1}^i = \frac{V(m, m+1)}{V(m+1, m)} \alpha_m^i + \frac{1}{V(m+1, m)} \sum_{n=i}^m \alpha_n^{i-1} \quad (1.8)$$

which can also be represented as

$$\alpha_n^i = \sum_{m=i+1}^n \frac{n^2}{m^2} \frac{\exp(-En, m/T_e)}{V(m, m-1)} \sum_{k=i}^{m-1} \alpha_k^{i-1} \quad (1.9)$$

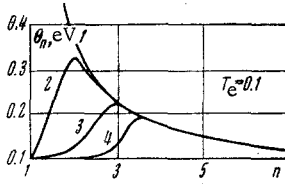


Fig. 1

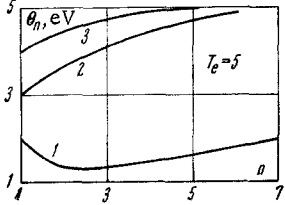


Fig. 2

Hence,

$$\alpha_m^0 = m^2 \exp(-Em, 1 / T_e) \quad (1.10)$$

$$\alpha_{m+1}^m = \left[\prod_{i=1}^m V(i+1, i) \right]^{-1} \quad (1.11)$$

$$\alpha_n^1 = \sum_{m=2}^n \frac{n^2}{m^2} \frac{\exp(-En, m / T_e)}{V(m, m-1)} \sum_{k=1}^{m-1} k^2 \exp\left(-\frac{Ek, 1}{T_e}\right) \quad (1.12)$$

The solution (1.6) describes the process of relaxation to a Boltzmann distribution. The departure from the Boltzmann distribution with temperature T_e that holds for $N_1^{(i)} = 0, i \neq 0$, is given by the terms with the first, second, etc., derivatives of the population of the ground state of the atoms. The process of recombination and ionization occurs in such a way that with the course of time the atoms begins to "forget" their initial conditions. In other words, the terms with the highest derivatives may be important only in the initial stage of the process and unimportant at the end. For the time

tail of the relaxation it is sufficient to retain the term with the first derivative; then for the populations we have

$$N_m = \alpha_m^0 N_1 + \alpha_m^1 N_m^{(1)} \quad (1.13)$$

where α_m^0 is given by (1.10) and α_m^1 by (1.12).

2. Recombination and Ionization of a Hydrogen Plasma

Let us consider the recombination and ionization of a hydrogen plasma at relatively high densities of the free electrons, when collisional transitions play the main role. The level population distribution can be conveniently characterized by temperatures θ_n between neighboring levels, these being determined by

$$\frac{N_{n+1}}{N_n} = \frac{(n+1)^2}{n^2} \exp\left(-\frac{E_{n+1, n}}{\theta_n}\right) \quad (2.1)$$

In the case of recombination of a plasma whose degree of ionization is greater than the equilibrium value corresponding to the free-electron temperature $T_e = 0.05-0.5$ eV, we obtain, using the expression (1.13) for the populations,

$$\theta_n = T_e \left[1 + \frac{T_e}{E_{n+1, n}} \ln \frac{1 + \kappa \beta_n^1}{1 + \kappa \beta_{n+1}^1} \right]^{-1} \quad (2.2)$$

where

$$\beta_n^1 = \frac{1}{n^2} \exp(E_{n, 1} / T_e) \alpha_n^1 = \sum_{m=2}^n \frac{\exp(E_{m+1, 1} / T_e)}{m^2 V(m, m-1)}, \quad \kappa = \frac{N_1^{(1)}}{N_1}$$

The parameter $\kappa(\tau)$ can be represented in the form

$$\kappa(\tau) = 4V(2,1) \left[\exp\left(-\frac{E_{2,1}}{\theta_1}\right) - \exp\left(-\frac{E_{2,1}}{T_e}\right) \right] \quad (2.3)$$

Since the population of the lower level increases with the time as a result of recombination, the parameter κ is positive (this follows directly from its definition), and therefore all the temperatures are always greater than T_e . In addition, $\kappa(\tau) \rightarrow 0$ and $\theta_1(\tau) \rightarrow T_e$ as $\tau \rightarrow \infty$. Thus, the temperature between the first and the second levels during recombination approaches the electron temperature asymptotically.

We calculated the temperatures θ_n for fixed T_e and different values of the parameter θ_1 . The probabilities of collisions of the first and the second kind between atoms and electrons were taken from expressions based on Bethe's formula (see, for example [6]). For $T_e = 0.1$ eV, the results of the calculation are represented in Fig. 1 in the form of the dependence of θ_n on n for different θ_1 . It is interesting that when θ_1 decreases to the value determined by the condition

$$\exp\left(-\frac{E_{2,1}}{\theta_1}\right) \gg \exp\left(-\frac{E_{2,1}}{T_e}\right) \quad (\theta_1 < 0.102 \text{ eV})$$

the values of the other temperatures remain virtually constant. In the well-known approximation of a "stationary sink" [6], the nature of the distribution obtained as a result of numerical calculations is also independent of the temperature between the first and the second level and agrees with the analytic solution.

It can be seen from Fig. 1 that, as a result of recombination, equilibrium is established fairly rapidly between the electrons on the upper levels and the free electrons. There is then an accumulation of the first level, the distribution in the upper levels remaining essentially unchanged (stationary sink, curve 1). Further, once the temperature θ_1 approaches equilibrium, there is a decrease in the temperature between the second and the third level and an accumulation of the second level, while the distribution at the upper levels remains essentially the same (curves 2 and 3), etc.

Thus, the recombination process can be regarded as a certain successive process of population of levels, beginning with the first.

Let us consider the problem of ionization of a hydrogen plasma whose degree of ionization at the initial time is less than the equilibrium value corresponding to the free-electron temperature $T_e = 5-10$ eV. In contrast to recombination, the parameter κ is negative, since the population of the ground state decreases to a certain equilibrium value. The temperature between the first and the second level increases to the electron temperature. The populations are described by the previous formula (2.2). The results of the calculation for $T_e = 5$ eV and different values of the parameter θ_1 are shown in Fig. 2. In accordance with the distributions obtained, the ionization proceeds fairly rapidly in the initial phase, when the electron temperature is very different from the temperature between the first and the second level. During this stage of ionization, many-quantum transitions to the upper levels play an important role. Therefore, curve 1 in Fig. 2 (in which only one-quantum transitions are taken into account) does not turn sufficiently toward the electron temperature. Once the difference between the electron temperature and the temperature between the first and the second level becomes insignificant, the populations of the excited states are determined primarily by the population of the neighboring lower level (stepwise ionization). At the same time, the temperatures between upper neighboring levels relax successively to the electron temperature. The nature of the relaxation of the distribution to the equilibrium state during the stage of stepwise ionization is shown in Fig. 2 by curves 2 and 3.

3. Condition of Applicability of the Quasistationary

Distributions. External Influences on a Relaxing System

The quasistationary distributions were obtained in the one-quantum approximation (we allowed for only transitions between neighboring levels). This approximation is not satisfied for the upper levels, for which the Saha distribution (1.2) holds. To obtain a single distribution, we fit the two distributions, using the normalization relation

$$\sum_{n=1}^{n_0} N_n = N - N_e$$

As a result, we obtain a system of two equations:

$$ZN_1 + Z^1 N_1^{(1)} = N - N_e, \quad N_1 + \beta_{n_0}^1 N_1^{(1)} = CN_e^2 \quad (3.1)$$

where

$$C = \left(\frac{2\pi\hbar^2}{mT_e}\right)^{3/2} \exp\left(\frac{R}{T_e}\right), \quad Z = \sum_{n=1}^{n_0} n^2 \exp\left(-\frac{E_{n,1}}{T_e}\right)$$

$$Z^1 = \sum_{n=1}^{n_0} \alpha_n^1$$

Solving the system for N_1 and $N_1^{(1)}$, we can readily relate the parameter κ to the electron density:

$$\kappa = \frac{N - N_e - ZCN_e^2}{Z^1 CN_e^2 - \beta_{n_0}^1 (N - N_e)} = \frac{1}{\beta_{n_0}^1} \left(\frac{CN_e^2}{N_1} - 1 \right)$$

Hence we obtain the admissible values of the κ for which relaxation processes can be considered. For recombination

$$\kappa < \frac{1}{\beta_{n_0}^{-1}} \left(\frac{CN_e^2(0)}{N_1(0)} - 1 \right) \approx \frac{1}{\beta_{n_0}^{-1}} \frac{CN_e^2(0)}{N_1(0)}$$

For ionization

$$|\kappa| < \frac{1}{\beta_{n_0}^{-1}} \left(1 - \frac{CN_e^2(0)}{N_1(0)} \right) \approx \frac{1}{\beta_{n_0}^{-1}}$$

Since the temperature θ_1 relaxes to T_e , it follows that $\kappa \rightarrow 0$, and the electron density tends to a certain limit determined by the equation

$$N - N_e - ZCN_e^2 = 0, \quad N_e = \frac{1}{2ZC} [V\sqrt{1+4ZCN} - 1]$$

In the case of recombination

$$N_e \rightarrow \sqrt{N/C} \quad (3.2)$$

In the case of ionization

$$N_e \rightarrow N(1 - ZCN) \quad (3.3)$$

Note that the same limit can be obtained from the exact solution of the system (3.1) as $\tau \rightarrow \infty$.

Differentiating the system (3.1) with respect to τ and solving it for $N_1^{(1)}$ and $N_1^{(2)}$, we obtain the condition under which the next term in the expansion in the derivatives can be ignored:

$$\frac{N_1^{(2)}}{N_1^{(1)}} = \frac{2ZCN_e + 1}{2ZCN_e + \beta_{n_0}^{-1}} \ll \frac{\alpha_{n_0}^{-1}}{\alpha_{n_0}^{-2}}$$

In the case of recombination with $T_e = 0.05-0.5$ eV, this inequality can be simplified, since

$$Z \approx 1, \quad Z^1 \approx \frac{1}{V(2,1)}, \quad \frac{\alpha_{n_0}^{-1}}{\alpha_{n_0}^{-2}} \approx V(2,1)$$

As a result

$$N_e \ll V(2,1)\beta_{n_0}^{-1}/2C \quad (3.4)$$

For the case of ionization, the condition under which the second derivatives can be ignored is transformed to

$$1/\beta_{n_0}^{-1} \ll \alpha_{n_0}^{-1}/\alpha_{n_0}^{-2} \quad (3.5)$$

While the inequality (3.4) enables one to find the times after which the time tail of the recombination is well described by the first derivative, the inequality (3.5) gives a lower bound for the electron temperature. When $T_e = 5$ eV, this inequality is satisfied with one or two orders of magnitude to spare. Note that in the calculations we have throughout assumed $n_0 = 9$. However, the higher values of n_0 had practically no influence on either the nature of the distribution nor the fulfillment of the conditions (3.4) and (3.5).

Thus, our distributions describe a fairly broad class of phenomena. This also applies to the distribution that takes into account only the first derivative, which, like Treanor's distribution for the vibrational levels of molecules, depends on the two parameters T_e and θ_1 . The electron temperature T_e characterizes both the external condition and the level distribution functions. The temperature θ_1 for given T_e characterizes the internal properties of the distribution. Without dwelling on questions of an external influence on a relaxing system, we point out only the obvious simplicity of the treatment of relaxation in varying external conditions for characteristic times of variation greater than the time required to establish a quasiequilibrium distribution.

4. Allowance for Radiative Decay of Levels

Hitherto we have considered a relatively dense plasma in which collisional transitions predominate. In a plasma with a low electron density, radiative decay of the levels also influences the population kinetics of the excited states in addition to collisions of the first and the second kind between atoms and electrons. Let $A(n, m)$ be the probability of a spontaneous radiative transition from level n to level m . Then the radiative flux from level n to all the lower levels is

$$-\sum_{m=1}^{n-1} A(n, m) N_n = -A(n) N_n, \quad A(1) = 0 \quad (4.1)$$

In the flux from the upper levels to level n we restrict ourselves to spontaneous transitions between levels $n+1$ and n . As a result, the kinetic equations acquire two further terms, which greatly complicate the problem, since the electron density depends on the time. However, in the case of a "stationary sink," when N_e remains virtually constant, or in the quasistationary case when N_e varies slowly compared with the change of the populations of the discrete levels, radiative decay can be included in the general scheme of solution of system (1.1), and one can obtain the corrections to the coefficients for arbitrary α_m^i , which are calculated from the recursion relation

$$\begin{aligned} \left[1 + \frac{A(m+1, m)}{N_e V(m+1, m)}\right] \alpha_{m+1}^i &= \frac{(m+1)^2}{m^2} \exp\left(-\frac{E_{m+1, m}}{T_e}\right) \alpha_m^i + \frac{1}{V(m+1, m)} \sum_{n=i}^m \alpha_n^{i-1} + \\ &+ \frac{1}{N_e V(m+1, m)} \sum_{n=i+1}^m [A(n) - A(n, n-1)] \alpha_n^i \end{aligned} \quad (4.2)$$

The recursion relation comes out particularly simply for the coefficients α_{m+1}^m :

$$\alpha_{m+1}^m = \left[\prod_{i=1}^m W(i+1, i) \right]^{-1}, \quad W(m+1, m) = V(m+1, m) + \frac{A(m+1, m)}{N_e} \quad (4.3)$$

Then the formula for the temperatures becomes

$$\theta_n^{-1} = E_{n+1, n}^{-1} \ln \left[\frac{(n+1)^2}{n^2} \frac{\alpha_n^0 + \alpha_n^1 \kappa}{\alpha_{n+1}^0 + \alpha_{n+1}^1 \kappa} \right] \quad (4.4)$$

where

$$\kappa = 4W(2, 1) \left[\exp\left(-\frac{E_{2,1}}{\theta_1}\right) - \frac{V(2, 1)}{W(2, 1)} \exp\left(-\frac{E_{2,1}}{T_e}\right) \right]$$

If the radiative correction is small,

$$A(m+1, m) / V(m+1, m) \ll N_e$$

Eq. (4.4) can be readily transformed to the old expression by the substitution

$$V(m+1, m) \rightarrow W(m+1, m)$$

If the electron density varies rapidly, radiative decay can be taken into account by the method of successive approximation, in which in the first step one must assume that N_e depends on the time in these formulas.

5. Relaxation of the Populations in an Atomic Plasma

In contrast to hydrogen, the structure of the discrete levels of arbitrary atoms is fairly complicated. For example, in the case of the lithium atom the level with the principal quantum number n is split into sublevels with different values of the orbital number l .

The balance equations that take into account inelastic collisions between atoms and electrons are

$$\frac{dN_{nl}}{dt} = -N_e N_{nl} \sum_{n'l' \neq nl} V(nl, n'l') + \sum_{n'l' \neq nl} V(n'l', nl) N_{n'l'} N_e \quad (5.1)$$

Since the probabilities of transitions between neighboring levels increase with decreasing distance between them, equilibrium is established most rapidly at the lower levels. Introducing the assumption of a Boltzmann distribution over $l = s, p, d, \dots$:

$$N_{nl} = N_{ns} (2l + 1) \exp \left[- \frac{\Delta E_l(n)}{T_e} \right], \quad \Delta E_l(n) = E_{nl} - E_{ns} \quad (5.2)$$

and taking into account the transitions $\Delta n = 0, \pm 1, \Delta l = \pm 1$, we go over in (5.1) to the variable τ :

$$\begin{aligned} \frac{dN_{ns}}{d\tau} = & -V(ns, n+1p) N_{ns} + V(n+1p, ns) 3 \exp \left(- \frac{\Delta E_p(n+1)}{T_e} \right) N_{n+1s} + \\ & + V(n-1p, ns) 3 \exp \left(- \frac{\Delta E_p(n-1)}{T_e} \right) N_{n-1p} - V(ns, n-1p) N_{ns} \end{aligned} \quad (5.3)$$

These equations are similar to (1.1) for the hydrogen atom; however, here, because of the branching of the flux, the coefficients of the populations (regarded as effective probabilities) are no longer related by an equation of the type (1.3) for the direct and inverse processes. As in the case of hydrogen, a solution is sought in the form of the series

$$N_{ns} = \sum_{i=0}^{n-2} \alpha_n^i N_{2s}^{(i)} \quad (5.4)$$

As a result, we obtain recursion relations for α_n^i , from which we obtain

$$\begin{aligned} \alpha_m^0 &= \exp \left(- \frac{E_{m,2}(s)}{T_e} \right), \quad E_{m,n}(s) = E_{ms} - E_{ns} \quad (m=3, 4, \dots) \\ \alpha_m^1 &= \alpha_m^0 \sum_{n=2}^{m-1} \frac{1}{V(ns, n+1p)} \left[1 + \sum_{k=3}^n \prod_{q=k}^n \frac{V(qs, q-1p)}{V(q-1s, qp)} \right] = \alpha_m^0 \beta_m^1 \end{aligned} \quad (5.5)$$

The parameter that determines the recombination and ionization kinetics is in this case the temperature $\theta_2(s)$ between the levels 2s and 3p:

$$\kappa = \frac{N_{2s}^{(1)}}{N_{2s}} = V(2s, 3p) \left[\exp \left(E_{3,2}(s) \left(\frac{1}{T_e} - \frac{1}{\theta_2(s)} \right) \right) - 1 \right] \quad (5.6)$$

Finally, we write down expressions for the populations of the discrete levels and the temperature between the levels $(n+1)s$ and np :

$$N_{nl} = N_{2s} (2l + 1) \exp \left[- \frac{E_{n,2}(s) + \Delta E_l(n)}{T_e} \right] (1 + \kappa \beta_n^1) \quad (5.7)$$

$$\theta_n^{-1}(sp) = [E_{n+1,n}(s) - \Delta E_p(n)]^{-1} \left[\frac{E_{n+1,n}(s) - \Delta E_p(n)}{T_e} - \ln \frac{1 + \beta_{n+1}^1 \kappa}{1 + \beta_n^1 \kappa} \right] \quad (5.8)$$

Because of the branching of the relaxation flux it is possible for population inversion to arise between levels $(n+1)s$ and np during the recombination of the plasma if

$$\frac{1 + \beta_{n+1}^1 \kappa}{1 + \beta_n^1 \kappa} > \exp \left[\frac{E_{n+1,n}(s) - \Delta E_p(n)}{T_e} \right] \quad (5.9)$$

This condition is especially simple for the transition 3s-2p:

$$\theta_2(s) > \frac{E_{3,2}(s)}{\Delta E_p(2)} T_e \quad (5.10)$$

The results of calculations of the populations in Li in accordance with these formulas is in good agreement with the numerical calculations [13] for relaxation in a dense lithium plasma and also in qualitative agreement with the results of [14] for the helium atom and [15] for the argon atom. Evidently, this approach can be used to calculate the populations of discrete levels of all atoms, and also to produce population inversion under different nonequilibrium regimes.

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