

SOME APPLICATIONS OF RELAXATION THEORY OF A HIGHLY IONIZED HYDROGEN PLASMA

B. S. Gordiets, L. I. Gudzenko, and L. A. Shelepin

Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, Vol. 9, No. 6, pp. 115-120, 1968

Relaxation of a highly ionized uniform plasma is discussed and results are given concerning impulse recombination and ionization, in relation to their application to the amplification of radiation via the transitions of atomic hydrogen.

One of the current applications is to obtain a source of nonequilibrium radiation using a comparatively dense highly ionized plasma, and in particular to create an active medium capable of effective amplification of electromagnetic radiation. Papers [1, 2] use hydrogen as an example to carry out first estimates showing the possibility of creating an inverse population of certain discrete levels (of atoms or ions) on the impulse recombination of a plasma.

Paper [2] gave the solution of an algebraic system of equations for the populations of the lower discrete levels and free electron density, using the so-called "constant runoff" assumption [3], i. e. , that the populations  $N_n$  of the excited states and free electron density  $N_e$  do not change in time during the recombination process.

However, when the free electrons are cooled rapidly the solution of the constant-runoff equations may prove unsuitable for describing the true relaxation picture even in an optically thick plasma. Making allowance for the reabsorption of resonance radiation, which varies strongly in the course of recombination or ionization, involves the analysis of the non-steady-state problem, associated with the solution of the system of nonlinear differential relaxation equations.

In order to allow for radiation reabsorption, we make use of an effective probability of radiative transition

$$A^*(m, n) = F(m, n) A(m, n). \tag{1}$$

Here  $A(m, n)$  is the probability of spontaneous decay;  $m$  and  $n$  are the principal quantum numbers;  $F(m, n)$  is a coefficient which depends on the characteristic dimensions of the plasma, the form of the absorption line for the transition  $m \rightarrow n$  and the density of atoms on the level  $n$ .

Only the Lyman series can play an important part in the capture of radiation for the plasma parameters considered here. Assuming that these lines have a Doppler profile, we have the following expressions for an infinitely long cylinder of radius  $R$  [4]:

$$F(m, 1) = 1 \text{ for } k(m, 1)R < 2, \\ F(m, 1) = \frac{1.6}{k(m, 1)R\sqrt{\pi} \ln[k(m, 1)R]} \text{ for } k(m, 1)R \geq 2,$$

where  $k(m, 1)$  is the absorption coefficient at the center of the spectral line

The balance equation for the populations of the discrete levels of hydrogen has the following form:

$$\frac{dN_n}{dt} = -N_n \left\{ \sum_{m=n+1}^g V(n, m) N_e + \sum_{m=1}^{n-1} R(n, m) N_e + \right. \\ \left. + [B(n, e) + B(n, g)] N_e + A^*(n) \right\} + \sum_{m=n+1}^g [R(m, n) N_e + A^*(m, n)] N_m + \\ + \sum_{m=1}^{n-1} V(m, n) N_e N_m + N_e^3 [B(e, n) + B(g, n)] + N_e^2 [A(e, n) + A(g, n)]. \tag{2}$$

A system of nine equations was considered for the lower discrete levels  $n = 1-9$ . The upper levels ( $n > 9$ ) were

assumed to be in equilibrium with the free electrons, and their populations were determined from Saha's formula. The totality of these levels is denoted by the symbol  $g$ . It was assumed that they constitute (effectively for the given calculation) a quasi-continuous spectrum. The following symbols were introduced for the probabilities of the elementary processes:  $V(n, m)$  and  $R(n, m)$  corresponding to collisions of the first and second kind between atom and electrons,  $B(n, e)$  for ionization,  $B(e, n)$  for triple recombination,  $A(e, n)$  for radiation recombination. The coefficients  $B(n, g)$  and  $B(g, n)$  correspond to the transition probabilities of an electron passing from the lower discrete levels into the quasi-continuous spectrum and vice-versa, respectively, as a result of a collision with a free electron,  $A(g, n)$  is the result of spontaneous decay. The following expressions were used for the probabilities of radiative transitions\*:

$$A(n) = 1.66 \cdot 10^{10} n^{-2.5}, \quad A^*(n) = \sum_{m=1}^{n-1} A(n, m),$$

$$A(m, n) = 1.57 \cdot 10^{10} n^{-3} m^{-3} (n^{-2} - m^{-2})^{-1},$$

$$A(e, n) + A(g, n) = 5.2 \cdot 10^{-14} n^{-3} \left(\frac{Ry}{kT_e}\right)^{3/2} \exp\left(\frac{E_n}{kT_e}\right) i\left(-\frac{|E_{10, n}|}{kT_e}\right),$$

$$E_{n, n} = E_m - E_n, \quad E_n = Ry n^{-2}.$$

Here  $T_e$  is the free electron temperature;  $Ry$  is Rydberg's constant. The coefficients describing collision processes have the following form after we average the semiempirical cross sections obtained from Bethe's formula [6] and use a Maxwell velocity distribution for the free electrons,

$$V(n, m) = 1.73 \cdot 10^{-7} n^{-3} m^{-3} \left(\frac{Ry}{kT_e}\right)^{3/2} \left(\frac{E_{n, m}}{kT_e}\right)^{-4} \exp\left(-\frac{|E_{n, m}|}{kT_e}\right) U\left(\frac{|E_{n, m}|}{kT_e}\right),$$

$$R(m, n) = V(n, m) n^2 m^{-2} \exp\left(\frac{|E_{n, m}|}{kT_e}\right),$$

$$B(n, g) + B(n, e) = 8.65 \cdot 10^{-8} n^{-3} \left(\frac{Ry}{kT_e}\right)^{3/2} \int_{\zeta_n}^{\infty} z^{-4} U(z) \exp(-z) dz,$$

$$B(g, n) + B(e, n) = 5.5 \cdot 10^{-8} n^{-3} \left(\frac{Ry}{kT_e}\right)^{3/2} \exp\left(\frac{E_n}{kT_e}\right) \int_{\zeta_n}^{\infty} z^{-4} U(z) \exp(-z) dz,$$

$$U(z) = 1 + z \exp(z) E_i(-z) \quad (\zeta_n = E_n / kT_e).$$

The system of equations (2) must be supplemented by the equation of conservation of the total number of electrons per cubic centimeter of plasma, which coincides with the density of heavy particles

$$N = N_e + \sum_{n=1}^g N_n + \frac{N_e^2}{3} \left(\frac{2\pi\hbar^2}{m Ry}\right)^{3/2} \left(\frac{Ry}{kT_e}\right)^{3/2} \left(\frac{m^3 e^6}{8\pi\hbar^6 N}\right)^{3/4}.$$

The problem of the decay of a nonequilibrium plasma was solved numerically for initial densities  $N_e = 10^{14}, 10^{15} \text{ cm}^{-3}$  and temperatures  $kT_e = 0.05, 0.1, 0.2 \text{ eV}$  for an infinitely long cylinder of radius  $R = 3 \text{ mm}$ . The initial populations of the lower level ( $n \leq 0$ ) were taken to have a Boltzmann distribution with  $kT_e^0 = 2 \text{ eV}$ . It was assumed that the free electrons in the plasma were cooled instantaneously from  $2 \text{ eV}$  to  $kT_e$ . The calculations were carried out on an electronic computer using the Runge-Kutta method with a variable step length. The recombination coefficient  $\alpha = N_e^{-2} dN_e/dt \text{ (cm}^3 \text{ sec}^{-1}\text{)}$  is given in Fig. 1 as a function of the electron concentration  $N_e \text{ (cm}^{-3}\text{)}$  for  $kT_e = 0.1 \text{ eV}$  with  $N = 10^{14}$  and  $10^{15} \text{ cm}^{-3}$ . The part played by the reabsorption radiation, which slows down recombination, increases as the plasma density increases. Thus curve 2, corresponding to  $N = 10^{15} \text{ cm}^{-3}$ , lies below curve 1 ( $N = 10^{14} \text{ cm}^{-3}$ ). For the sake of comparison  $\alpha$  is also given as a function of  $N_e$  for  $N = 10^{15} \text{ cm}^{-3}$  for an optically thick plasma (curve 3). The reduced populations  $N_n^1 \text{ (cm}^{-3}\text{)}$  of the levels ( $N_n^1 = N_n/g_n$ , where  $g_n$  is the statistical weight of level  $n$ ) are given in Fig. 2 as a function of time  $t \text{ sec}$  for various  $N \text{ cm}^{-3}$  and  $kT_e \text{ eV}$ . It is clear from the graphs that in the recombination process for  $kT_e = 0.05-0.2 \text{ eV}$  and  $N = 10^{14}, 10^{15} \text{ cm}^{-3}$ , a series of levels have an inverse population. This is caused by the fact that the upper levels are populated more rapidly because of the flow of electrons from the continuous spectrum during impulse recombination, and the impoverishment of the lower excited states as the result of the greater probability that they will suffer radiative decay. The reabsorption of resonance radiation, which effectively diminishes the probability of radiative decay in a dense plasma, exerts a strong effect on the presence of inversion.

There is an inverse population of levels 3-2 in the plasma (for  $N = 10^{15} \text{ cm}^{-3}$ ) and of levels 3-2 and 4-2 (for  $N =$

\*For greater detail see [5].

$= 10^{14} \text{ cm}^{-3}$ ) immediately after the instantaneous cooling of the free electrons, when the population of the fundamental state is still small and reabsorption is practically absent.

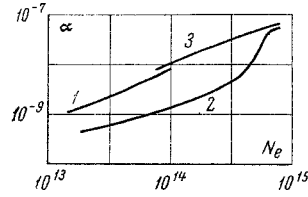


Fig. 1

As the number of atoms in the fundamental state increases, important reabsorption of the  $L_\alpha$  line commences; the population of the second level then increases somewhat and the inverse population relative to it disappears. We note that for a strongly ionized decaying plasma with parameters  $kT_e = 0.05, 0.1 \text{ eV}$  and  $N = 10^{14}, 10^{15} \text{ cm}^{-3}$ , an inverse population can exist as regards the second layer only in the first recombination period when the density of free electrons is still large ( $4 \cdot 10^{13} \text{ cm}^{-3}$ ).

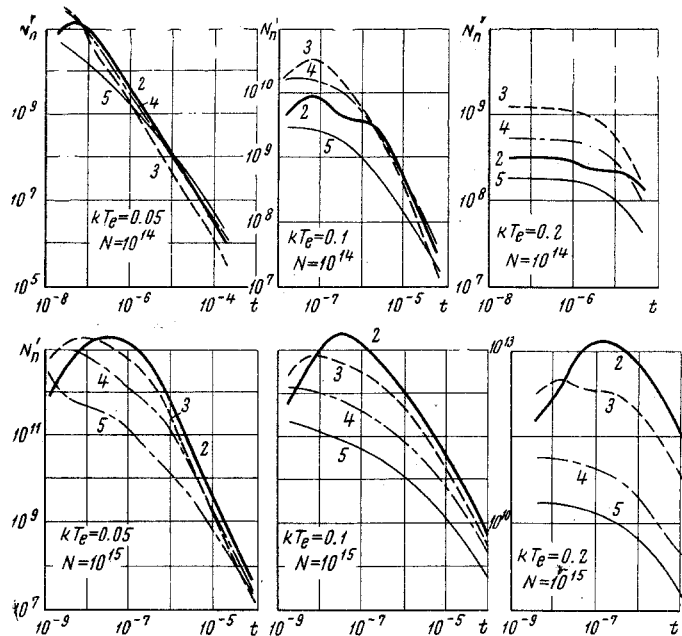


Fig. 2

When, as the result of recombination,  $N_e$  becomes smaller than  $4 \cdot 10^{13} \text{ cm}^{-3}$ , inversion of the fourth level appears relative to the third; for still smaller  $N_e$  inverse populations of the fifth and fourth layers can result. This is connected with the fact that for large densities the free electrons even out the populations of these states. When  $N_e$  falls off considerably the part played by radiation processes increases (particularly radiative decay of level 3, since the reabsorption of lines  $H_\alpha$  and  $L_\beta$  is small). Inversion between the third and fourth layers is interesting in view of the fact that it may exist even in the presence of a large number of atoms in the fundamental state, while inversion relative to the second level exists only until reabsorption of the  $L_\alpha$  line begins to play a part. The negative absorption coefficient per cm of photon path is calculated from the formula

$$\kappa_{m,n} = \frac{\lambda_{m,n}^2}{4\Gamma_{m,n}} A^*(m,n) (N_{m'} - N_n).$$

Here  $\lambda_{m,n}$  is the wavelength and  $\Gamma_{m,n}$  is the line width. The coefficients  $\kappa_{m,n}$  are given in Table 1 for various values of  $kT_e$  and times  $t$ ; they are quite sufficient for making a laser.

In order to obtain generation under laboratory conditions we must compensate for losses in the mirror over a path length of  $l \lesssim 1 \text{ m}$  in the medium; thus there is effective amplification in the visible range and in adjacent parts of

the spectrum if  $\chi \gtrsim 10^{-4} \text{ cm}^{-1}$ . This leads to restricting the density of the highly ionized hydrogen to values of  $N = 10^{13} - 3 \cdot 10^{14} \text{ cm}^{-3}$  (for  $N < 10^{13} \text{ cm}^{-3}$  there are few atoms which cause amplification, for  $N > 3 \cdot 10^{14} \text{ cm}^{-3}$  inversion decreases because of the increase in inelastic collisions with an increase of density  $N_e$ ); the mean energy of free electrons should not exceed 0.2 eV. Cooling of electrons from an energy  $kT_e^0 \approx 2 \text{ eV}$  corresponding, under equilibrium conditions, to practically fully ionized hydrogen, should occur in a time  $\Delta t \lesssim 10^{-7} \text{ sec}$ , according to the preliminary calculations of [1]. Up to the present there has been no analysis of the methods of sharp cooling of electrons, and the requirements formulated here seemed unrealistic to experimenters; thus it was important to show the feasibility of the undertaking.

Table 1. The Coefficients of Negative Absorption  $\kappa$  ( $\text{cm}^{-1}$ ) for Certain Transitions, for Various Values of Temperature  $kT_e$  (eV) and Density  $N$  ( $\text{cm}^{-3}$ ) at Various Times  $t$  (sec)

$kT_e$	$N$	$t$	Transition	$\kappa$
0.2	$10^{13}$	$1.5 \cdot 10^{-8}$	3 = 2	$3.4 \cdot 10^{-2}$
0.05	$10^{14}$	$6.4 \cdot 10^{-8}$	3 = 2	$5.3 \cdot 10^{-2}$
0.1	$10^{14}$	$1.7 \cdot 10^{-8}$	3 = 2	$4.9 \cdot 10^{-4}$
0.2	$10^{14}$	$10^{-8}$	3 = 2	$4.9 \cdot 10^{-4}$
0.2	$10^{15}$	$4.4 \cdot 10^{-5}$	3 = 2	$3.1 \cdot 10^{-5}$
0.05	$10^{15}$	$5.7 \cdot 10^{-6}$	4 = 3	$1.8 \cdot 10^{-3}$
0.05	$10^{15}$	$2.3 \cdot 10^{-5}$	4 = 3	$8 \cdot 10^{-4}$
0.05	$10^{14}$	$1.4 \cdot 10^{-8}$	4 = 3	$5.8 \cdot 10^{-3}$
0.1	$10^{14}$	$3.7 \cdot 10^{-6}$	4 = 3	$6 \cdot 10^{-4}$
0.1	$10^{14}$	$4.5 \cdot 10^{-5}$	4 = 3	$4.1 \cdot 10^{-4}$

Article [7] discussed the cooling of free electrons in the adiabatic expansion of a plasma into vacuum: gas-kinetic dispersion, the dispersion of a magnetized cluster, and the efflux of a magnetized jet. Estimates showed that under realistic engineering conditions the necessary cooling is attained in fairly short times on expansion. Paper [8] gives estimates of the cooling times of free electrons after a sharp cut off of the heating field, as the result of elastic collisions of electrons with cold heavy plasma particles, and also as the result of ambipolar diffusion to the walls of the gas discharge pipe. It turned out that the deep-freezing time could be  $\lesssim 10^{-7} \text{ sec}$  in the conditions of an ordinary gas discharge in pure hydrogen and in a mixture of hydrogen with helium.

Detailed calculations show that requirements formulated for the cooling times can be relaxed considerably. The lengths of time for which inversion can exist in hydrogen undergoing impulse recombination are given in Table 2 for some transitions. These lead to the general result that the cooling times should not exceed  $10^{-6} - 10^{-5} \text{ sec}$ . Reabsorption leads to a deterioration of the recombination conditions, and so it is convenient to work with gas discharge tubes of small diameter. On the other hand, the tube must not be made too narrow as this makes it difficult to trigger and maintain the discharge, and worsens the ratio between the useful (for the aims discussed here) volume recombination and the "harmful" recombination close to the wall which populates the lower levels without radiation. In order to decrease the recombination occurring in the vicinity of the wall without lowering the effective cooling, it is convenient to carry out the discharge in a mixture of hydrogen and a sufficient quantity of a gas-filler, having an ionization potential considerably higher than that of hydrogen. Such a gas could be helium (see [8, 9]) which creates a large capacity thermostat of cold heavy particles without exerting a direct effect on the kinetics of the relaxation. A rough estimate of the appropriate concentration of helium can be obtained from the condition that after cooling, volume recombination should predominate over recombination close to the wall

$$\alpha N_e^2 > \frac{DN_e}{(R/2.4)^2},$$

where  $D$  is the ambipolar diffusion coefficient. For  $T_e \approx 2000^\circ \text{ K}$ ,  $N_e \approx 10^{14} \text{ cm}^{-3}$ ,  $R \approx 0.3 \text{ cm}$  we obtain a helium pressure of  $p \approx 1.3 \text{ mm Hg}$ . To decrease the losses of free electrons due to diffusion to the wall, it is convenient to apply a longitudinal magnetic field (whose strength should not exceed the critical value), which facilitates triggering the discharge and stabilizing it.

In a plasma of more complicated chemical composition than pure hydrogen, the processes of ionization (and recombination) relaxation occur together through channels whose characteristic times are often very different (see also [10]). It is thus of interest to examine the possibility of creating an effective amplifying medium immediately on its rapid ionization. Such a situation cannot occur in the pure hydrogen plasma whose relaxation is discussed here.

It is convenient to dwell a while longer on the analysis of hydrogen ionization, since it is the initial stage in the impulsive change of the average energy of free electrons, and the assumption that before the rapid cooling both the free and bound electrons have an equilibrium distribution with a temperature  $kT_e \approx 2$  eV does not altogether correspond to judiciously framed experiments.

Table 2. Inversion Lifetimes for Various Transitions for Different Temperatures  $kT_e$  0.05; 0.1; 0.2 (eV) of the Free Electrons and Densities  $N$  ( $\text{cm}^{-3}$ ) of the Recombining Plasma

$N=10^{15}$	$N=10^{16}$	$N=10^{14}$	$N=10^{14}$
3=2	4=3	3=2	4=3
$10^{-8}$ $7 \cdot 10^{-9}$ $2 \cdot 10^{-8}$	$10^{-4}$ 0 0	$9 \cdot 10^{-8}$ $3 \cdot 10^{-7}$ $6 \cdot 10^{-8}$	$10^{-4}$ $10^{-4}$ 0

Practically the same equations as in system (2) with minor modifications, describing a wide range of relaxation processes in a hydrogen plasma, are convenient for carrying out this analysis.

Since an existing program was available for solving the non-steady-state problem on an electronic computer, a uniform analysis could be made of relaxation processes in a highly ionized atomic plasma, specifying various initial conditions. Thus in the simplest ionization regime the solution of the system of equations (2) was calculated for the initial conditions  $N_1(0) \neq 0$ ,  $N_2(0) = N_3(0) = \dots = N_g(0) = 0$ . The degree of ionization at the initial time was less than the equilibrium value corresponding to the temperature  $T_e$  which is assumed to be constant during the process of ionization. The results of solving system (2) for such a model are given in Fig. 3a, b, respectively, for  $N = 1.26 \cdot 10^{14} \text{ cm}^{-3}$ ,  $kT_e = 5$  eV and  $N = 1.26 \cdot 10^{15} \text{ cm}^{-3}$ ,  $kT_e = 10$  eV; the initial free electron density was taken to be equal to  $N_e(0) = 9 \cdot 10^{10} \text{ cm}^{-3}$ . It must be stressed that ionization, like recombination, is a complicated, many-stage process, and so to characterize it, as is often done, by an ionization cross section is usually impossible. At the same time, the results of systematic calculations concerning ionization are indisputably of interest not only in the analysis of the amplification properties of a highly ionized plasma, but also in a wide range of applied plasma problems, in particular in the problem of the passage of a plasma jet through matter.

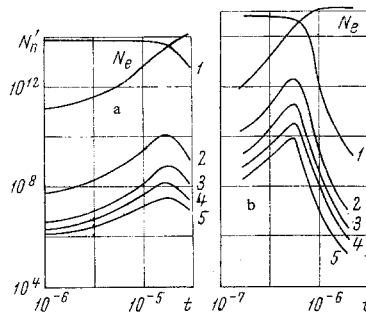


Fig. 3

The first experimental papers in which inversion in atomic hydrogen was obtained appeared not long ago. A note in [11] mentioned the population inversion of the fifth and fourth levels in the expansion regime of a jet of argon-hydrogen plasma. Despite the point of view expressed by the authors of [11] explaining the inversion by the collisions of hydrogen atoms with excited argon atoms, a recombination inversion mechanism seems more natural. Estimates show that a hydrogen plasma with the parameters mentioned in [11] undergoes population inversion when recombining, without any transfer of excitation from argon.

Paper [12] gave the first observations of generation for the  $4 \rightarrow 3$  transition for atomic hydrogen on an impulse discharge in a mixture of hydrogen and helium. The pipe diameter (7 mm), and the partial pressures of hydrogen and helium (0.03 and 3.5 mm Hg respectively) are in good agreement with the calculations and estimates given above.

However, even here we would be premature in concluding finally that the laser operates on the basis of recombination, since there are no data in [12] concerning development of the generation impulse in time.

The authors are grateful to A. T. Matachun for help in the numerical calculations.

#### REFERENCES

1. L. I. Gudzenko and L. A. Shelepin, "Negative damping in a non-equilibrium hydrogen plasma," *ZhETF*, vol. 45, no. 5, 1963.
2. L. I. Gudzenko and L. A. Shelepin, "Amplification in a decaying plasma," *Magnitnaya gidrodinamika* [Magnetohydrodynamics], vol. 1, no. 3, 1965.
3. D. R. Bates and A. E. Kingston, "Properties of a decaying plasma," *Planet. Space Sci.*, vol. 11, no. 1, 1963.
4. T. Holstein, "Imprisonment of resonance radiation in gases." *Phys. Rev.*, vol. 72, p. 1212, 1947.
5. B. F. Gordiets, L. I. Gudzenko, and L. A. Shelepin, "The relaxation of hydrogen level populations. Inversion in a decaying highly ionized plasma," Preprint FIAN, no. 29, 1967.
6. I. I. Sobel'man, *Introduction to the Theory of Atomic Spectra* [in Russian], Fizmatgiz, Moscow, 1963.
7. L. I. Gudzenko, S. S. Filippov, and L. A. Shelepin, "An accelerated recombining plasma jet," *ZhETF*, vol. 53, no. 4, 1966.
8. B. F. Gordiets, L. I. Gudzenko, and L. A. Shelepin, "Cooling of free plasma electrons," *Zh. tekhn. fiz.*, vol. 36, no. 9.
9. L. I. Gudzenko, V. N. Kolesnikov, N. N. Sobolev, and L. A. Shelepin, "Application of highly ionized plasma to laser construction," *Magnitnaya gidrodinamika* [Magnetohydrodynamics], vol. 1, no. 3, 1965.
10. L. I. Gudzenko and V. M. Finkel'berg, "Impulse discharge in a chemically active medium as a source of optical radiation," Preprint FIAN, no. 16, 1967.
11. V. M. Gol'dfarb, E. V. Il'ina, I. E. Kostychova, G. A. Luk'yanov, and V. A. Silant'ev, "The population of hydrogen levels in an argon-hydrogen plasma jet," *Optika i spektroskopiya*, vol. 20, no. 6, 1966.
12. K. Bockasten, T. Lundholm, and O. Andrade, "Laser lines in atomic and molecular hydrogen," *J. Opt. Soc. of America*, vol. 56, no. 9, 1966.

17 May 1967

Moscow