

ON THE THEORY OF IONIZATION AND RECOMBINATION IN A LOW-TEMPERATURE PLASMA

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Expressions for the recombination and ionization rates in a plasma of arbitrary composition are obtained on basis of the concept of the two processes being one of electron diffusion in discrete energy space. Electron-atom collisions and radiative processes are taken into account. The possible absence of Maxwellian energy distribution of free electrons is taken into account. Results of concrete calculations are presented and compared with the experiments.

THE kinetics of ionization and recombination in a partially ionized plasma has a number of distinguishing features, which are closely connected with the multi-level structure of the atoms and molecules. The recombining electron, before producing an atom in the ground state, passes through a set of excitation states. The recombination rate is determined by the time of passage through these states, and is therefore closely connected with the distribution of the atoms over the energy levels. In final analysis, the recombination rate is a complicated function of the concentration of the light and heavy particles, their energy distributions (the Maxwellian electron energy distribution may be violated), and by the condition for the yield of the radiation. All the foregoing applies equally well to ionization.

The rates of ionization and recombination are customarily represented as the product of the electron density, raised to a certain power, and an appropriate coefficient. However, owing to the dependence of these coefficients on a number of parameters, it is preferable to speak of ionization and recombination functions (IF and RF). The IF and RF can be determined by solving a system of balance equations¹⁾ written out for each of the excited states of the atom. This system should be solved simultaneously with the kinetic equation for the electrons.

Starting with the work of Bates and co-workers,^[4] a numerical method of solving this problem has gained wide acceptance, and has made it possible to compile tables of IF and RF for certain elements and for a limited range of parameters.^[4-6] It is important, however, that the electron energy distribution was assumed to be Maxwellian, and the emission yield was considered only in two limiting cases.

On the other hand, it was noted that although the levels in the atoms are not equidistantly located, the probabilities of the transitions between the energetically close levels are the largest. In this case, the recombination can be regarded as a certain slow probabilistic process of the Brownian-motion type in energy space,

and a suitable Fokker-Planck equation can be written, with the discrete spectrum replaced by a continuous one.^[7-9] The latter circumstance has narrowed down the limits of applicability of the indicated approach.

In this paper we use a modified diffusion approximation,^[10, 11] in which the real discrete structure of the energy spectrum is retained. This calls for the derivation and solution of a corresponding Fokker-Planck finite-difference equation. Account is taken of the collision and radiative processes. It is assumed that the Maxwellian distribution for the electrons is violated. Allowance for the discreteness makes it possible to consider not only recombination but also ionization, and the limits of the analysis of recombination in this case are greatly broadened. General expressions are obtained for the IF and RF, as well as approximation formulas. The connection between the IF and RF is discussed. Results of calculations are presented for a number of atoms and are compared with the experimental data.

FUNDAMENTAL EQUATIONS

We write the rate of change of the electron concentration in the form

$$\partial n_e / \partial t = n_1 n_e \beta - n_e^3 \alpha - \nabla \mathbf{J}, \quad (1)$$

where β and α are functions of the ionization and recombination,²⁾ $\nabla \mathbf{J}$ takes into account the transport phenomena. We are interested below only in volume processes (α and β).

The functions α and β should be determined from the solution of the system of nonstationary particle-balance equations for each of the levels of the atom. However, the problem can be greatly simplified by using the condition that the excited states are quasistationary. The point is, that in a wide range of conditions, the following conditions are satisfied in the plasma

$$\sum_{k>1} n_k \ll n_e, n_1$$

(n_k —concentration of the atoms in the state k). Then the concentration of the atoms in the excited states will

¹⁾The balance equation follows, under certain assumptions, from a more rigorous analysis of this problem, see, for example, [1,2]. Among the latest papers, mention should be made of [3].

²⁾The recombination rate is sometimes assumed equal to $\alpha^* n_e^2$. Then $\alpha^* = n_e \alpha$.

have time to adjust itself to relatively slow changes of n_e and n_i .

The problem can be solved quantitatively by comparing the ionization relaxation time τ_{1e} and the relaxation time τ_k of the individual excited states. Estimates performed in [5, 11] have shown that $\tau_{1e} \gg \tau_k$ ($k \geq 2$) in a wide range of conditions. It is clear that when $t \gg \tau_k$ it is possible to use the quasistationarity approximation. Quasistationarity means that at each given instant of time, in all cross sections of energy space, there is one and the same particle flux j , which determines the IF and RF. The assumption of quasistationary development of ionization or recombination makes it possible to calculate j in the stationary approximation.

In [10, 11] we investigated the distribution of the atoms over the levels and of the electrons over the energies in a stationary nonequilibrium plasma. The processes in the discrete energy spectrum were described with the aid of a Fokker-Planck finite-difference equation. The processes in the energy continuum were described with the aid of a differential Fokker-Planck equation. Account was taken of the influence of the inelastic electron-atom collisions on the free-electron distribution, as well as of the reaction of the non-Maxwellian behavior on the distribution over the excited states. It turned out that the final results can be represented in the form of formulas that are obtained when account is taken of only transitions between neighboring energy levels (the single-quantum approximation). However, the probabilities of these transitions differ from the single-quantum ones and are expressed in terms of moments that are characteristic of the diffusion approximation. Thus, for the transition $k \rightarrow k+1$, this effective probability is given by

$$\langle z_{k, k+1} \rangle = (E_{k-1} - E_{k+1})^{-1} (E_k - E_{k+1})^{-1} \sum_n \langle w_{kn} \rangle (E_n - E_k) (E_n - E_{k-1}), \quad (2)$$

where $\langle w_{kn} \rangle$ is the probability of the $k \rightarrow n$ transition, averaged over the free-electron distribution. It is easy to see that $\langle z_{k, k+1} \rangle \equiv \langle w_{k, k+1} \rangle$ on going over to the single-quantum approximation, and this quantity is proportional to the mean square of the energy transferred by the electrons to the atoms on going over to a quasi-continuous energy change.

Moments of the type (2) can be calculated with the aid of the sum rules, making it possible to represent $\langle z_{k, k+1} \rangle^0$ in the form

$$\langle z_{12} \rangle^0 = n_e \frac{4 \sqrt{2\pi} e^4 \Lambda_1}{\Delta E_1 \sqrt{m T_e}} \exp \left\{ -\frac{\Delta E_1}{T_e} \right\}, \quad \Delta E_k = E_k - E_{k+1},$$

$$\langle z_{k, k+1} \rangle^0 = n_e \frac{4 \sqrt{2\pi} e^4 \Lambda_k E_{k-1}}{\Delta E_k (E_{k-1} - E_{k+1}) \sqrt{m T_e}} \exp \left\{ -\frac{\Delta E_k}{T_e} \right\}, \quad k \geq 2, \quad (3)$$

where $\langle \rangle^0$ denotes averaging over the Maxwellian electron-energy distribution, and Λ_k is the so-called Coulomb logarithm for the bound states, a plot of which against $\Delta E_k/T_e$ is shown in Fig. 1.

In the absence of strong external fields, the main factor violating the Maxwellian distribution may be the inelastic collisions with the nonuniformly populated atoms. This process is most appreciable in the high-energy region, where the frequency of the Maxwellizing collisions may turn out to be insufficient. Formally this

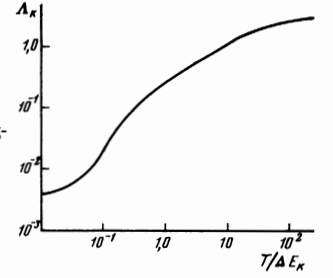


FIG. 1. Plot of the Coulomb logarithm for bound states.

circumstance can be taken into account by replacing $\langle z_{12} \rangle$ by $\langle z_{12} \rangle^0 F_1$, where

$$F_1 = \frac{1}{c} \frac{\sqrt{1+4c} - 1}{\sqrt{1+4c} + 1}, \quad c = \frac{n_i \langle z_{12} \rangle^0}{n_e \langle z_{ee} \rangle^0},$$

$$\langle z_{ee} \rangle^0 = \frac{2 \sqrt{2\pi} e^4 \lambda}{T_e \sqrt{m T_e}} \exp \left\{ -\frac{\Delta E_1}{T_e} \right\}, \quad (4)$$

λ is the Coulomb logarithm for the interelectron collisions; these collisions are usually the main Maxwellizing factor. [12] For $k \geq 2$ we assume $\langle z_{k, k+1} \rangle = \langle z_{k, k+1} \rangle^0$.

The motion of the bound electron over the energy spectrum, resulting from the radiation acts, has a directional character and does not constitute diffusion. In the section of energy space between the levels k and $k+1$, the flux due to the radiation processes can be approximately written in the form

$$n_{k+1} a_{k+1}^R - n_e^2 a_{k+1}' = n_{k+1} \sum_{i>k} A_{in} - n_e^2 \sum_{n<k} \alpha_{ek}, \quad (5)$$

where A_{in} is the probability of the spontaneous transition $i \rightarrow n$, and α_{ek} is the probability of radiative recombination at the level k . It can be shown from (5) that for each level k the transitions $k \rightarrow i$ ($i < k$), $k+1 \rightarrow k$, and the radiative recombination have been taken into account accurately. In the presence of reabsorption, A_{in} in (5) is replaced by $A_{in}^* = A_{in} \theta_{in}$, where θ_{in} is the probability that the given photon will leave the plasma without absorption. [13]

Writing now the expression for the total flux j in the section of energy space between each pair of levels, we obtain the following system of coupled equations:

$$j = n_k \langle z_{k, k+1} \rangle - n_{k+1} (\langle z_{k+1, k} \rangle + \alpha_k^R) - n_e^2 a_{k+1}'.$$

Solving the equations and connecting the populations of the last of the discrete levels realized in the plasma with the electron concentration, we obtain expressions for the IF and RF:³⁾

$$\beta = \left(n_e K_1^{-1} \prod_{h \geq 1} S_h \right)^{-1}, \quad \alpha = \left(1 + \sum_{h \geq 1} \alpha'_{h+1} S_h \right) \left(n_e n \sum_{h \geq 1} S_h \right)^{-1}; \quad (6)$$

$$\Pi_k = \prod_{n \geq k} \left(1 + \frac{\alpha_{n+1}^R K_n}{\langle z_{n, n+1} \rangle K_{n+1}} \right), \quad S_k^{-1} = \langle z_{k, k+1} \rangle K_k^{-1} \Pi_k,$$

$$K_k = 2 \sum_i (g_{kn} h^3)^{-1} (2\pi m T_e)^{3/2} e^{-E_k/T_e}, \quad (7)$$

Π_k is a factor that takes into account the influence of the radiation yield on the population of the $(k+1)$ -st

³⁾ At high temperatures, ionization by electrons from the ground state and electron-ion-electron recombination into this state may become noticeable. These processes are not steplike. Their contribution must be summed with (6).

level. Σ_i is the partition function of the ion, and g_k is the statistical weight of the level k . The energy E_k is reckoned from the continuum, $n = n_e + \sum_{k \geq 1} n_k$ is the total number of heavy particles.

This leads to a general relation between the IF and RF:

$$\beta = \alpha K_1 n \left[\left(1 + \sum_{k \geq 1} \alpha'_{k+1} S_k \right) \Pi_1 \right]^{-1}. \quad (8)$$

IONIZATION BY ELECTRON IMPACT AND THREE-PARTICLE RECOMBINATION

If radiative processes can be neglected, then

$$\alpha = \left(\sum_k \frac{n n_e K_k}{\langle z_{k, k+1} \rangle} \right)^{-1}, \quad \beta = \alpha K_1 n. \quad (9)$$

The detailed-balancing relation $\beta = \alpha K_1 n$ is valid also in the absence of a Maxwellian distribution, if due to inelastic electron-atom collisions ($F_1 < 1$).

We simplify the expression for α by using the fact that all terms of the sum in (9) make equal contributions. This can be verified directly. However, it is sufficient to recall the character of the distribution of the atoms over the excited states.^[10, 11] The highly-excited levels are in relative equilibrium with the continuum. The low-lying levels are close to equilibrium with the ground state. Obviously, upon ionization (recombination), the bound electron rapidly passes through energy intervals corresponding to these groups of levels. As to the group of intermediate essentially-nonequilibrium states, it constitutes the "bottleneck" for the flux in energy space. The passage through the bottleneck in fact determines the rate of the ionization (recombination).

The position of the bottleneck E^* can be estimated by using the approximate expression for the populations, obtained earlier in^[10], referred to the equilibrium values of T_e ($y_k = n_k/n_k^0$, $y_e = n_e/n_e^0$):

$$y(E_k) = y(E_1) \chi \left(\frac{E_k}{T_e} \right) + y_e^2 \left[\chi \left(\frac{E_1}{T_e} \right) - \chi \left(\frac{E_k}{T_e} \right) \right], \quad \chi(x) = \frac{4}{3\sqrt{\pi}} \int_0^x e^{-t^{3/2}} dt \quad (10)$$

From the condition $\partial^2 y / \partial E^2 = 0$ it follows that $E^* = 3T_e/2$. The lower limit of the bottleneck can be identified with the point where the derivative $\partial y / \partial E$ decreases by a factor e ; this yields a value $\sim 7T_e/2$.

A. Low temperatures. The bottleneck falls in the region of strongly excited states, in which the discreteness of the levels can be neglected. Going over in (9) from summation to integration, and assuming the excited states to be hydrogen-like, we obtain

$$\alpha^{-1} = \frac{3\pi \sqrt{\pi} m^2}{2h^3 e^4 \Lambda Ry^{3/2}} T_e^{3/2} \chi \left(\frac{E_1}{T_e} \right), \quad (11)$$

$\bar{\Lambda}$ is the value of Λ_k in the region of the bottleneck. We can assume

$$\bar{\Lambda} \approx 0.2; \quad Ry = e^4 m / 2h^2.$$

The formula of the type (11) was obtained earlier in^[9], where the discreteness of the levels was neglected from the very outset. By stipulating that the difference between the neighboring levels in the bottleneck zone be smaller than T_e , we can readily obtain a criterion for the validity of (11):

$$2^{(3/2)^{3/2}} \sqrt{T_e} / Ry \ll 1.$$

It is important that (11) does not depend on the type of the atom. This is due to the fact that the rate of the recombination (ionization) is determined by the passage through the highly-excited hydrogen-like states.

B. High temperatures. The bottleneck is located in the energy interval $(E_1 - E_2)$. Retaining in (9) only the first term, we obtain

$$\alpha = \frac{h^3 e^4 \Lambda_1 g_1}{\pi m^2 \Delta E_1 T_e^2 \Sigma_i} \exp \left\{ \frac{E_2}{T_e} \right\} F_1, \quad \beta = \langle z_{12} \rangle^0 F_1 / n_e. \quad (12)$$

Formula (12) for β corresponds to the approximation of "immediate ionization" of the excited atoms that appear during the course of the relaxation. The ionization rate is determined by the excitation rate. A criterion for the applicability of (12) is the inequality $S_1 \gg \sum_{k \geq 2} S_k$. Calculating $\sum_{k \geq 2} S_k$ in the quasicontinuous approximation, we can write this inequality in the form

$$\frac{2}{3} \frac{\Lambda}{\Lambda_1} \frac{\Delta E_1}{T_e} \left(\frac{Ry}{T_e} \right)^{3/2} \exp \left\{ -\frac{E_2}{T_e} \right\} \left[F_1 \chi \left(\frac{E_2}{T_e} \right) \right]^{-1} \ll 1.$$

It follows from (12) that when $F_1 \neq 1$ the IF and RF depend not only on the temperature but also on n_e . In the limit of a strong non-Maxwellian distribution ($F_1 \ll 1$), β is determined by the rate of the interelectron collisions for an electron with energy $\epsilon = E_1 - E_2$:

$$\beta = \frac{2\sqrt{2\pi} e^4 n_e \Lambda}{T_e \sqrt{m} T_e} \exp \left\{ -\frac{\Delta E_1}{T_e} \right\}. \quad (13)$$

C. General case. We obtain an approximate formula applicable to the entire region of T_e . To this end, we separate in the sum (9) the term with $k = 1$, and replace the remaining sum with an integral. We then obtain

$$\alpha^{-1} = \frac{\pi m^2 T_e^2 \Sigma_i}{h^3 e^4} \left[\frac{\Delta E_1 e^{-E_2/T_e}}{F_1 g_1 \Lambda_1} + \frac{3\sqrt{\pi} T_e^{3/2} \chi(E_2/T_e)}{2\Lambda \Sigma_i Ry^{3/2}} \right]. \quad (14)$$

This expression makes it possible to draw certain conclusions with respect to the dependence of α on the plasma composition. Thus, at low T_e , as already noted, α is the same for all the atoms. With increasing T_e , the specific structure of the atom comes into play. Under these conditions, α depends principally on the ratio Σ_i/g_1 and on the magnitude of the "gap" ΔE_1 .

D. Results of calculations, comparison with experiment. Figure 2 shows the results of the calculation of α for the atoms H, Ar, He, N, K, and Cs. For each of them, the calculations were made both by the approximate formula (14) and by formula (9). In some cases (depending on the specific structure of the atom) we used both $\langle z_{n, n+1} \rangle^0$ calculated with the aid of the sum rules, and $\langle z_{n, n+1} \rangle^0$ obtained by using directly presently known experimental data on the transition probabilities. In all cases, the simple formula (14) did not result in appreciable deviations from the more accurate values. It can be used to calculate IF and RF in plasmas of various composition under a wide range of conditions.

The same figure shows the calculated data of Bates and co-workers^[4] for H, obtained by numerically solving the system of balance equations of the excited atoms. Although our calculations were performed by a different method, on the whole the agreement is satisfactory. It is not so good in the region $T_e = (8-16) \times 10^3$ K. It is

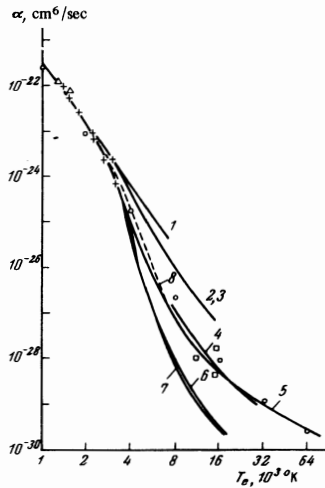


FIG. 2. Coefficient of three-particle recombination as a function of T_e for different atoms. 1—Calculation of α for all atoms by formula (11); 2, 3, 4, 5, 6, 7—calculation of α by formula (9) for K, Cs, N, H, He, and Ar, respectively; 8—calculation of α by formula (14) for H. \circ — calculated points of Bates et al. [4]. Experimental data: Δ — for H [15], + — for He [15], \square — for H [14].

important that it is precisely here that α is the most sensitive to the form of the cross section of the $1 \rightarrow 2$ transition at the threshold. This cross section was obtained by Bates from Gryzinski, i.e., not in the same way as we did. Figure 2 shows also the experimental data from [14], which are in fair agreement with the calculated ones.

We proceed to discuss the dependence of α on the singularities of the structure of the atom. At $T_e < 3000^\circ \text{K}$ we have $\alpha \sim T_e^{-9/2}$ for all elements, as is well confirmed by numerous measurements made on H, He, and Cs atoms, [15-17]. With increasing T_e , deviations from the $T_e^{-9/2}$ law set in. For atoms with a relatively uniform level density (K, Cs), the $T_e^{-9/2}$ law has, naturally, greater applicability. For He, which has the maximum gap ΔE_1 , the deviation from $T_e^{-9/2}$ begins much earlier (the case of Ar is discussed below). Hydrogen and nitrogen occupy an intermediate position.

The specific structure of the atom is manifest not only in the value of ΔE_1 . Formula (14) explains the practical coincidence of α for the pairs H, N and He, Ar. These atoms have close values of $\Sigma_i/g_i\Lambda_1$.⁴⁾

IONIZATION AND RECOMBINATION FUNCTIONS WITH ALLOWANCE FOR RADIATION

When the influence of the radiation yield is significant, the calculation of α differs from that of β , since these quantities depend differently on the electron concentration. In the presence of reabsorption, an additional difference is due to the influence of the linear dimensions of the plasma and other parameters that determine the effective lifetimes. Allowance for all these

⁴⁾In complicated atoms with splitting in l , it is convenient to combine into one level groups of nearly-equal-energy states that are in relative equilibrium. For example, for nitrogen such a level, with $k = 1$, is the state of the main configuration.

effects is based on formulas (6). Let us discuss the extreme cases and possible simplifications.

If we decrease n_e and by the same token increase the role of the radiation, then we obtain from (6) in the limit the so-called coefficient of "radiative recombination"

$$\alpha = (n_e n)^{-1} \sum_k \sigma_{pk}. \quad (15)$$

This approximation is valid if the following inequalities hold for the levels that contribute to $\sum_k \alpha_{ek}$:

$$\frac{a_{n+1}^R K_n}{\langle z_{n, n+1} \rangle K_{n+1}} \gg 1, \quad \frac{a_{h+1}^i}{n_e n} \left(n_e n \sum_k S_k \right) \gg 1. \quad (16)$$

The first inequality means that the excited atom is more likely to emit than to experience an inelastic collision. The second inequality indicates that the "radiative recombination" coefficient (15) must be larger under these conditions than the "triple recombination" function calculated in the preceding section, multiplied by n_e .

If the inequalities (16) are reversed, the radiation yield becomes negligible, and we return to the formulas of the preceding section. In the intermediate conditions there occurs the so-called "impact-radiation" recombination. We shall discuss its qualitative features.

The intensity of the radiative processes decreases rapidly with increasing k , and the intensity of the impact processes increases. Therefore, the energy interval can be broken up into two regions: $E_k > E_R$, where the recombination is determined by the radiation, and $E_k < E_R$, where collision processes predominate.

Let us consider the case of low temperatures and let us estimate the influence of the radiation on α . Obviously, this influence is significant only if the level E_R lies above the lower limit $7T_e/2$ of the bottleneck. By the same token, the length of the bottleneck decreases, as can be roughly estimated by modifying formula (11) in a natural manner:

$$\alpha^{-1} = \frac{3\pi \sqrt{\pi} m^2}{2h^3 e^4 \Lambda R y^{1/2}} T_e^{9/2} \chi \left(\frac{E_R}{T_e} \right). \quad (17)$$

The value of E_R can be determined from the condition $\Pi_{E_R} = 1$. If we take into account the fact that the strongly excited states are hydrogen-like, we can obtain the following approximate expression:

$$E_R \approx R y n_e^{1/2} \left(\frac{\sqrt{\pi} e^4 \Lambda}{R y \sqrt{m T_e} c_1} \right)^{1/2}, \quad c_1 = (3-4) \cdot 10^{10} \text{sec}^{-1}. \quad (18)$$

Inasmuch as the function χ is continuous, it follows from (18) that at low temperatures, when n_e is appreciably decreased, the radiation does not influence α as strongly. The situation is different at high T_e , when the radiation can exert a strong influence on α , and still not shift the bottleneck from the energy interval $E_1 - E_2$. Then

$$\alpha = \frac{h^2 e^4 \Lambda_1}{\pi m^2 \Delta E_1 T_e^2 \Sigma_i} g_1 F_1 \exp \left\{ \frac{E_2}{T_e} \right\} \Pi_1. \quad (19)$$

With further decrease of n_e , it is necessary to use the general expression, since S_1 becomes comparable with $\sum_{k \geq 2} S_k$.

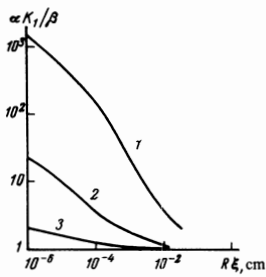


FIG. 3. Dependence of $\alpha K_1/\beta$ on the parameter $R\xi$ for a mixture Ar + K. $\xi = n_K/n_{Ar}$, R —linear dimension of the system, which determines the radiation yield. Curves: 1— $n_e = 10^{12}$, 2— 10^{13} , 3— 10^{14} cm^{-3} .

The radiation has entirely different effects on α and β . At high temperatures it has little effect on β . Indeed, the factor Π_1 in (19) drops out completely from the expression from β at high temperatures (when $S_1 \gg \sum_{k>1} S_k$). At low temperatures, to the contrary, the radiation has little effect on α , but it can radically alter β . In fact, under these conditions

$$\beta = K_1 \alpha \Pi_1^{-1}, \quad (20)$$

where α is given by formula (17). Figure 3 shows the dependence of $\alpha K_1/\beta$, for a mixture of argon with potassium on $R\xi$; R is the linear dimension of the plasma and ξ is the ratio of the number of potassium atoms and the number of argon atoms. It follows from Fig. 3 that Π_1 can reach quite large values, particularly for small n_e . Naturally, the use of the detailed-balancing relation $\beta = \alpha K_1 n$ is not valid under such conditions.

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