

Nonresonant charge transfer in the field of an intense light wave

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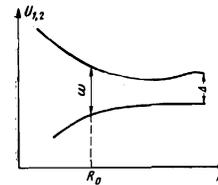
Nonresonant charge transfer in the field of an intense electromagnetic wave of frequency close to the resonance defect is investigated. It is shown that charge transfer is determined by electric dipole transitions between states of a quasimolecule. Expressions are obtained for the charge exchange cross section. It is shown that the cross section may significantly exceed the geometrical cross section.

In their recent papers Gudzenko and Yakovlenko^[1,2] have noted that the presence of an intense electromagnetic field may significantly increase the cross section for inelastic nonresonant collisions. They have investigated the exchange of excitation between two atoms in the presence of a field of frequency equal to the difference between the levels of the colliding atoms. In^[1] the cross section for the transfer of excitation was obtained only at the center of the absorption line. But the cross section for an inelastic collision will also be significantly increased if the frequency of the field does not coincide with the resonance defect. Indeed, the difference between the terms of the quasimolecule formed by the colliding atoms may at a certain distance between them coincide with the frequency of the external field. In the neighborhood of this point intense electronic transitions will occur. As a result the atoms on separation can turn out to be in different electronic states.

For the occurrence of such a process it is necessary that the matrix element of the electromagnetic perturbation $\mathbf{P} \cdot \mathbf{A}$ between the states of the quasimolecule should be different from zero. Of the greatest interest is the case when the electric dipole transition is allowed since the internuclear distances significant for the problem are considerably smaller than the wavelength of the resonance field. Just such a situation is realized in the case of nonresonant charge transfer. We consider this problem in greater detail.

In the production of positive or negative ions by charge transfer between neutral atoms the behavior of quasimolecular terms between which the transition occurs qualitatively corresponds to the diagram (the states are assumed to be nondegenerate). At a distance R much larger than the atomic radius a the molecular terms U_1 and U_2 contain two contributions. The first contribution depends exponentially on the distance and is of exchange origin. The second corresponds to the polarization interaction and is proportional to R^{-4} . The factor in front of the exponential in the exchange term contains different powers of R depending on the states of the atoms between which the electron is transferred. If the frequency of the field ω is close to the difference between the levels of the atoms Δ , so that $\Omega \equiv \omega - \Delta \ll \Delta$, then the resonance distance R_0 is much larger than the atomic dimensions (R_0 is a root of the equation $U_1 = U_2 + \omega$). In this case one should expect that the cross section for the process will appreciably exceed the geometric cross section, and the matrix element of the dipole moment can be calculated utilizing the asymptotic form of the molecular wave functions.

The simplest case is the case of charge transfer between negative ions when the approximation of short range potentials is applicable. The terms of the quasi-



molecule are determined by the relations (in atomic units) $U_{1,2}(R) = -\kappa_{1,2}^2(R)/2$, where $\kappa_{1,2}$ are roots of the equation

$$(\kappa - \alpha)(\kappa - \beta) = R^{-2}e^{-2\kappa R}. \quad (1)$$

Here $\alpha = (2|\epsilon_1|)^{1/2}$; $\beta = (2|\epsilon_2|)^{1/2}$; ϵ_1, ϵ_2 are the binding energies of the negative ions participating in the collision.

The molecular functions have the form

$$\begin{aligned} \psi_1 &= C_1 \left[\frac{e^{-\kappa_1 r}}{r} + (\kappa_1 - \alpha) R e^{\kappa_1 R} \frac{e^{-\kappa_1 |r-R|}}{|r-R|} \right], \\ \psi_2 &= C_2 \left[\frac{e^{-\kappa_2 |r-R|}}{|r-R|} + \frac{e^{-\kappa_2 R}}{R(\kappa_2 - \alpha)} \frac{e^{-\kappa_2 r}}{r} \right]; \\ C_1 &= \left(\frac{\kappa_1}{2\pi} \right)^{1/2} [1 + (\kappa_1 - \alpha)^2 R^2 e^{2\kappa_1 R}]^{-1/2}, \\ C_2 &= \left(\frac{\kappa_2}{2\pi} \right)^{1/2} \left[1 + \frac{e^{-2\kappa_2 R}}{R^2 (\kappa_2 - \alpha)^2} \right]^{-1/2}. \end{aligned} \quad (2)$$

The expression for the matrix element of the dipole moment in the general case is quite complicated. Therefore we reproduce its form in the limiting case $R\alpha, R\beta \gg 1$ (it is just this region that is significant when the condition $\Omega \ll \Delta = |\alpha^2 - \beta^2|/2$ is satisfied):

$$d_{12} = 2n \frac{(\alpha\beta)^{1/2}}{\alpha^2 - \beta^2} e^{-\alpha R}, \quad (3)$$

where n is the unit vector along the axis of the quasimolecule¹⁾. For the sake of definiteness we have assumed $\beta > \alpha$, so that $\exp[(\alpha - \beta)R] \ll 1$. In the same approximation we have for the terms $U_{1,2}$:

$$\begin{aligned} U_1 &\approx -\frac{\alpha^2}{2} + \frac{\alpha}{\beta - \alpha} \frac{e^{-2\alpha R}}{R^2}, \\ U_2 &\approx -\frac{\beta^2}{2} - \frac{\beta}{\beta - \alpha} \frac{e^{-2\beta R}}{R^2}. \end{aligned} \quad (4)$$

In the case of charge transfer between positive ions the coefficient in front of the exponential in the formula for d_{12} will depend on R . However, the principal part of the dependence—the exponential with the smaller of the two damping decrements—will be retained.

We now write down the system of equations for the amplitudes for the transition:

$$i\dot{a} = H_{11}a + V_{12}b, \quad i\dot{b} = H_{22}b + V_{21}a; \quad (5)$$

$$H_{11} = U_1 + V_{11}, \quad H_{22} = U_2 + V_{22}, \quad V = dE \cos \omega t,$$

where E and ω are the amplitude and the frequency of the light wave.

We assume the intensity of the field \mathbf{E} to be sufficiently small so that the condition $d \cdot \mathbf{E} \ll \omega$ is satisfied. Then in the diagonal elements of \hat{H} one can neglect according to the parameter dE/ω the rapidly oscillating additional terms V_{11} and V_{22} . In the neighborhood of the point R_0 we expand $U_{1,2}$ in series in terms of $R - R_0$ and we restrict ourselves to a linear approximation. We take the quantity d_{12} to be constant in the region of transition and equal to $d_0 = d_{12}(R_0)$. We arrive at the system of equations

$$\begin{aligned} i\dot{a} &= [U_1(R_0) + v(\partial U_1/\partial R)_0 t] a + d_0 E b \cos \omega t, \\ i\dot{b} &= [U_2(R_0) + v(\partial U_2/\partial R)_0 t] b + d_0 E a \cos \omega t, \end{aligned} \quad (6)$$

where v is the radial relative velocity at the point R_0 , and the time is measured from the moment of passage through this point.

By definition we have $U_1(R_0) - U_2(R_0) = \omega$; introducing

$$a = A \exp[-iU_1(R_0)t], \quad b = B \exp[-iU_2(R_0)t],$$

we obtain a system of equations in which the nondiagonal element has the form $(d_0 E/2)(1 + e^{2i\omega t})$. Neglecting the term with the factor $\exp(2i\omega t)$ with respect to the parameter $d_0 E/\omega$, we bring the equations to a form analogous to the equations of the Landau-Zener problem^[3].

$$i\dot{A} = v \left(\frac{\partial U_1}{\partial R} \right)_0 t A + \frac{1}{2} d_0 E B, \quad i\dot{B} = v \left(\frac{\partial U_2}{\partial R} \right)_0 t B + \frac{1}{2} d_0 E A. \quad (7)$$

The solution of these equations is well known. The probability of charge transfer in the case of two passages through the point R_0 (i.e., for one collision) is equal to

$$W = e^{-\delta_1} (1 - e^{-\delta_2}) + e^{-\delta_2} (1 - e^{-\delta_1}), \quad (8)$$

$$\delta_{1,2} = \frac{\pi}{2} d_0^2 (n_{1,2} E)^2 \left(\left| \frac{\partial U_1}{\partial R} - \frac{\partial U_2}{\partial R} \right|_0 v \right)^{-1}.$$

The vectors n_1 and n_2 determine the direction of the axis of the quasimolecule respectively when the atoms are approaching each other and separating. Using formulas (3), (4) we obtain

$$\delta_{1,2} = \frac{\pi}{4} \frac{\beta (E n_{1,2})^2}{\alpha^3 (\alpha + \beta)^2 (\beta - \alpha) v} \ln^2 \frac{4\alpha^3}{(\beta - \alpha)\Omega}. \quad (9)$$

We note that the term in d_{12} which is exponentially small cancels against a similar factor in $(\partial U_1/\partial R)_0$. The quantity $\delta \sim E^2/v \ln^2(\alpha^2/\Omega)$, so that under the assumptions made above δ can be both much greater, and much smaller than unity.

From the foregoing it is clear that the general case of an inelastic collision in the field of an intense light wave must be described by a formula of the Landau-Zener type. In various specific situations only the parameter δ will be altered. In particular, d_{12} is involved in all cases of charge transfer, but the probabilities of certain collisions accompanied by spin exchange or by an exchange of excitation can be determined by other multipole moments. In the dependence of the terms on R one can include polarization and Van der Waals interaction, etc.

The general limitation on the applicability of formula (8) in all these cases is the requirement of a sufficiently strong lack of parallelism of the terms at the point R_0 . In the opposite case a restriction to terms linear with respect to t in H_{11} , H_{22} becomes unjustified. In the problem under consideration this means a limitation on Ω from below. Estimating the quadratic terms of the expansion in terms of $R - R_0$ with the aid of formulas

(3), (4) we obtain the desired estimate in the form $\Omega \gg (d_0 E R_0)^2$ (in this case α , β and $\beta - \alpha$ are regarded to be quantities of the same order of magnitude). Another limitation arises from the requirement that the principal contribution to the probability of the transition should be made by the point R_0 , and not by the domain of large R , where the terms are practically parallel. It coincides with the well known criterion due to Massey and in the present case has the form $\Omega \gg \alpha v$.

In order to investigate the region of the detuning of Ω which is not encompassed by the Landau-Zener formula we utilize an approach analogous to the theory of charge transfer with a small resonance defect^[4]. Separating out in the transition amplitudes the factor

$$\exp \left[\frac{1}{2i} \int (H_{11} + H_{22}) dt \right],$$

we bring the system (5) to the form

$$i\dot{a} = -\frac{1}{2}(H_{11} - H_{22})a + V_{12}b, \quad i\dot{b} = -\frac{1}{2}(H_{11} - H_{22})b + V_{12}a. \quad (10)$$

Of all the values of R the only region of importance is the one in which $|H_{11} - H_{22}| \lesssim v_{12}$, since in the opposite case the equations become decoupled, i.e., there are no transitions between the terms of a quasimolecule. Utilizing once again the approximation $\alpha R \gg 1$, $\beta R \gg 1$ and omitting terms which contain rapidly oscillating factors we obtain

$$H_{11} - H_{22} = \Omega - \epsilon_0 e^{-2\alpha R}, \quad V_{12} = V_0 e^{-\alpha R}.$$

The meaning of the parameters ϵ_0 and V_0 is clear from formulas (3), (4); in ϵ_0 we have also included the factor $1/R^2$ which varies slowly compared with the exponential. The limits on the domain of transition are determined by the inequality

$$(\Omega - \epsilon_0 z)^2 \leq V_0^2 z, \quad z = e^{-\alpha R}$$

and are equal to

$$z_1 \approx \frac{\Omega}{\epsilon_0} \left[1 - \left(\frac{4V_0^2}{\epsilon_0 \Omega} \right)^{1/2} \right], \quad z_2 \approx \frac{\Omega}{\epsilon_0} \left[1 + \left(\frac{4V_0^2}{\epsilon_0 \Omega} \right)^{1/2} \right],$$

if $\Omega \gg V_0^2/\epsilon_0$;

$$z_1 \approx \Omega^2/4V_0^2, \quad z_2 \approx 4V_0^2/\epsilon_0^2, \quad \text{if } \Omega \ll V_0^2/\epsilon_0.$$

It can be easily seen that the parameter V_0^2/ϵ_0 is of the order of $(d_0 E)^2 R_0^2$, i.e., the case $\Omega \gg V_0^2/\epsilon_0$ corresponds to the Landau-Zener approximation. The region of transition in this case is small; its width is:

$$R_1 - R_2 = \frac{1}{2\alpha} \ln \frac{z_2}{z_1} = \frac{1}{\alpha} \left(\frac{4V_0^2}{\Omega \epsilon_0} \right)^{1/2} \ll \frac{1}{\alpha}; \quad R_{1,2} = -\frac{1}{2\alpha} \ln z_{1,2}.$$

In the general case we must solve the system (10) in the region $z_1 < z < z_2$ and join the solution obtained with the amplitudes a , b for $R > R_1$ and $R < R_2$. We expand $R(t)$ near the point $R_0 = (1/2\alpha) \ln(\epsilon_0/\Omega)$:

$$R = R_0 + v(R_0)(t - t_0)$$

and reduce the system (10) to the form

$$\begin{aligned} i\dot{a} + \frac{1}{2}(\Omega - \epsilon_0' e^{-2\alpha v t})a &= V_0' e^{-\alpha v t} b, \\ i\dot{b} - \frac{1}{2}(\Omega - \epsilon_0' e^{-2\alpha v t})b &= V_0' e^{-\alpha v t} a; \end{aligned} \quad (11)$$

$$\epsilon_0' = \epsilon_0 \exp[-2\alpha(R_0 - vt_0)], \quad V_0' = V_0 \exp[-\alpha(R_0 - vt_0)].$$

In future we omit the prime on ϵ_0 and V_0 .

The requirement of the validity of the expansion carried out above over the whole region between R_1 and R_2 again leads to a limitation on Ω from below, but a weaker one than in the Landau-Zener approximation.

Estimating the terms in the exponentials quadratic in terms of $t - t_0$ we obtain the criterion of applicability: $(R_1 - R_2)^2 \ll R_0/\alpha$. From this it follows that (11) is valid for $\Omega \gtrsim v_0^2/E_0$, and in the region $\Omega \ll V_0^2/\epsilon_0$ when the Landau-Zener approximation is not applicable the validity of (11) is limited by the condition

$$\ln^2(4V_0^2/\Omega\epsilon_0) \ll \ln(\epsilon_0/\Omega)$$

(it is understood that $\epsilon_0 \gg \Omega$). For comparison we recall that in the usual nonresonant charge transfer the diagonal elements of the system of equations for the amplitudes expressed in the base of atomic functions are constant (cf. [4]) and there exists a large region $0 < R < \alpha^{-1} \ln \Delta^{-1}$, where intense transitions take place. In the given problem H_{11} and H_{22} correspond to variable molecular terms which diverge as R is decreased. Therefore an adiabatic development of the system corresponds to the regions $R < R_2$ and $R > R_1$ when there are no electronic transitions.

The solution of the system (10) is expressed in terms of the degenerate hypergeometric function:

$$b = \exp\left(-\frac{i\epsilon_0 z}{2v\alpha}\right) F\left(\frac{iV_0^2}{2\alpha v\epsilon_0}, \frac{1}{2} + \frac{i\Omega}{2\alpha v}, \frac{i\epsilon_0 z}{2\alpha v}\right),$$

where $z = \exp(2\alpha vt)$. The initial condition is: $a = 0$, $b = 1$ as $t \rightarrow -\infty$. Making a connection with the adiabatic solutions for $R < R_2$ we obtain the expression for the probability of transition in the case when the critical region is traversed twice:

$$W = \frac{1 - \cos \varphi}{(1 - e^{-\pi\Omega/\alpha v})^2} [2(e^{-\pi\Omega/\alpha v} - e^{-(\theta_1 + \theta_2)}) + (1 - e^{-\pi\Omega/\alpha v})(e^{-\theta_1} + e^{-\theta_2})]; \quad (12)$$

$$\delta_{1,2} = \delta_0 \frac{(\mathbf{n}_{1,2} \cdot \mathbf{E})^2}{E^2(1 - \rho^2/R_0^2)^{1/2}}, \quad \delta_0 = \frac{\pi d_0^2 E^2}{4\epsilon_0 \alpha v \infty}, \quad \varphi = \int (U_1 - U_2) dt.$$

The integral which defines φ is taken over a time interval between two passages through the point R_2 . The phase φ is large if the relative velocity of the atoms is much smaller than the orbital velocity of the electrons.

Formula (12) goes over into the Landau-Zener formula (8) when the conditions $\Omega \gg \alpha v$, $\Omega \gg V_0^2/\epsilon_0$ are satisfied; we must also average over φ and use the relations

$$\left| \frac{\partial}{\partial R} (U_1 - U_2) \right|_0 = 2\alpha\epsilon_0, \quad d_0 E = 2V_0.$$

In the case $V_0^2 \gg \epsilon_0 \alpha v$, $\Omega \ll V_0^2/\epsilon_0$ ($\Omega/\alpha v$ is arbitrary) formula (12) coincides with the result of Demkov [4]:

$$W = \frac{\sin^2(\varphi/2)}{\text{ch}^2(\pi\Omega/2\alpha v)} \quad (13)$$

(in Demkov's formula we must replace Δ by $\omega - \Delta = \Omega$). Finally, for $\Omega < 0$, when the frequency of the field is smaller than the resonance defect the probability of transition falls off rapidly with increasing $|\Omega|$:

$$W \approx 2 \left[1 - \exp\left(-\frac{\pi V_0^2}{\alpha v \epsilon_0}\right) \right] \exp\left(-\frac{\pi |\Omega|}{\alpha v}\right). \quad (14)$$

Expression (12), as has been stated already is not applicable for very small $|\Omega|$, when the condition $\ln^2(4V_0^2/\epsilon_0\Omega) \ll \ln(\epsilon_0/\Omega)$ is violated. In this case the coefficients of the system (11) cannot be assumed to be the same at the points R_1 and R_2 as was assumed in deriving (12). Further, the quantities $U_{1,2}(R)$ contain contributions of the polarization interaction, while in the previous discussion only the exchange interaction was taken into account. Usually the exchange interaction dominates over the polarization interaction up to quite large R (for example, $R \sim 10$ atomic units in the H_2^+ ion). If R_0 falls in this region, then all the formulas

obtained above remain valid. In the opposite case the term $\Delta\kappa/R^4$ where $\Delta\kappa$ is the difference between the polarizabilities of the colliding atoms will be added to the difference $H_{11} - H_{22}$. Assuming that this quantity is slowly varying for $R \gg 1$ compared with the exchange exponential we can include it in Ω retaining the foregoing formulas with a renormalized value of the detuning.

The cross sections for the collisions are determined by the probabilities (8), (12) in terms of the usual formula

$$\sigma = 2\pi \int_0^{R_0} W(\rho) \rho d\rho,$$

where ρ is the impact parameter for the collision. In the case of collisions in a gas σ must be averaged over all the orientations of the vectors $\mathbf{n}_{1,2}$ with respect to the electric field of the wave. We shall treat the trajectory of the atoms as being rectilinear $R = \rho + vt$ and we shall average over ρ and \mathbf{v} taking into account the fact that $\rho \perp \mathbf{v}$. In the Landau-Zener case the averaged cross section is equal to

$$\begin{aligned} \langle \sigma \rangle &= 4\pi \int_0^{R_0} [\langle e^{-\theta_1} \rangle - \langle e^{-(\theta_1 + \theta_2)} \rangle] \rho d\rho, \\ \langle e^{-\theta_1} \rangle &= \int_0^1 \exp\left(-\frac{\delta_0 x^2}{(1 - \rho^2/R_0^2)^{1/2}}\right) dx, \\ \langle e^{-\theta_1 - \theta_2} \rangle &= \int_0^1 \exp\left(-\frac{2\rho^2 \delta_0 x^2}{R_0^2(1 - \rho^2/R_0^2)^{1/2}}\right) dx \\ &\times \int_0^\pi \exp\left(-2\delta_0 \left(1 - \frac{\rho^2}{R_0^2}\right)^{1/2} (1 - x^2) \cos^2 \varphi\right) \frac{d\varphi}{\pi}. \end{aligned} \quad (15)$$

In the case of exponential dependence of the terms, δ_0 in (15) has the same value as in (12); in the more general case

$$\delta_0 = \frac{\pi}{2} (\mathbf{d}_0 \mathbf{E})^2 \left(\left| \frac{\partial U_1}{\partial R} - \frac{\partial U_2}{\partial R} \right|_0 v_\infty \right)^{-1}.$$

We quote the results for two limiting cases:

$$\begin{aligned} \langle \sigma \rangle &\approx \frac{4\pi}{3} R_0^2 \delta_0 \infty \frac{E^2}{v_\infty}, \quad \delta_0 \ll 1, \\ \langle \sigma \rangle &\approx \frac{4\pi^3}{5} R_0^2 \delta_0^{-1/2} \infty \frac{v_\infty^{1/2}}{E}, \quad \delta_0 \gg 1. \end{aligned} \quad (16)$$

The maximum in the cross section is attained at $\delta_0 \sim 1$, the corresponding value of the velocity of the atoms at infinity $(v_\infty)_{\text{max}}$ is proportional to the intensity of the light wave $(v_\infty)_{\text{max}} \sim E^2$. At its maximum $\langle \sigma \rangle$ is of the order of magnitude $R_0^2 \sim \alpha^{-2} \ln^2(\alpha^2/\Omega)$, i.e., it can appreciably exceed the geometric cross section of the atom.

We quote some numerical estimates. For $\epsilon_1 = 0.7$ eV, $\epsilon_2 = 3.5$ eV, $\Omega = 0.1 \Delta$, $v \sim 10^4$ the maximum of $\langle \sigma \rangle$ is attained at flux densities of $cE^2/4\pi \sim 10^9$ W/cm². We proceed to average the probability (12). It can be easily seen that in the averaged probability $\langle W \rangle$ the case $V_0^2 \gg \epsilon_0 \Omega$ (formula (13)) opposite to the Landau-Zener case is not realized, since the operation of averaging automatically picks out the range of angles for which $V_0^2/\alpha v \epsilon_0 \lesssim 1$. Therefore we shall reproduce results for two limiting cases of small and large δ_0 , assuming the parameter $\Omega/\alpha v$ to be arbitrary:

$$\langle W \rangle = 2\delta_0/3 [1 + e^{-\pi\Omega/\alpha v}], \quad \delta_0 \ll 1, \quad (17)$$

$$\langle W \rangle = \frac{2e^{-\pi\Omega/\alpha v} + (\pi/\delta_0)^{1/2} (1 - e^{-\pi\Omega/\alpha v})}{(1 + e^{-\pi\Omega/\alpha v})^2}, \quad \delta_0 \gg 1.$$

The average cross section obtained from formulas (17) depends on two parameters: δ_0 and $\Omega/\alpha v_\infty$. In the case $\pi\Omega/\alpha v_\infty \gg 1$ we again arrive at formulas (16). In the opposite limiting case we have:

$$\begin{aligned} \langle\sigma\rangle &\approx \frac{2\pi}{3} \delta_0 R_0^2; & \pi\Omega/\alpha v_\infty \ll 1, & \delta_0 \ll 1, \\ \langle\sigma\rangle &\approx \frac{1}{2} \pi R_0^2; & \pi\Omega/\alpha v_\infty \ll 1, & \delta_0 \gg 1. \end{aligned} \quad (18)$$

In the formulas exhibited above for the cross sections and the averaged probabilities (15)–(18) it is assumed that we can neglect the term with $\cos \varphi$ in expression (12), i.e., we exclude from consideration the narrow region of impact parameters close to R_0 . Within this region the phase φ can be small, but for velocity $v \ll 1$ in atomic units the region indicated above is not significant for the calculation of the total cross section.

In conclusion we note an interesting characteristic feature of the processes considered above. The cross section for the transition is essentially determined by the value of R_0 which is a root of the equation $U_1 - U_2 = \omega$. Therefore, by varying the frequency of the electromagnetic wave and measuring the cross sections for inelastic collisions or the intensity of light absorbed in such collisions one can in principle reconstruct the shape of the curve for the interatomic interaction in definite states of the quasimolecule. It is essential that the corresponding band of frequencies, generally speaking, should not coincide with the resonance frequencies

of atomic transitions, so that self-absorption would not interfere with measurements.

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¹The average dipole moment of the quasimolecule is noninvariant with respect to the choice of the origin of coordinates since the system under consideration has a nonzero total charge. However d_{12} , d_{11} - d_{22} , which are essential for further discussion, possess such invariance in view of the orthogonality of ψ_1 and ψ_2 .

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180