Exchange interaction of an atom with a multiply charged ion

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An analytic expression for the exchange energy in the interaction of an atom with a multiply charged ion is derived within the limitations of the quasiclassical approach for large distances between the nuclei of the colliding systems. It is shown that in the case of interaction with a multiply charged ion one can distinguish two ranges of values for the distance R between the nuclei of the interacting systems in which the exchange potential is represented by two different functions of R.

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1. INTRODUCTION

Collision processes involving multiply charged ions (MCI) have recently become the object of intensive theoretical[1-13] and experimental[14-19] study since they permit the determination of parameters that are important for modern photonuclear machines and heavy-ion accelerators. In addition, collisions of charged particles with atoms are accompanied by capture of an electron from the target atom into highly excited states of the MCI with excitation energies E of the order of $z^2/2$, where z is the charge of the ion. The large capture cross sections ($\sigma \sim 50z$ a.u.) in slow collisions ($v \ll 1$) makes it possible to regard the process as one of the most efficient means of obtaining powerful x-ray and ultraviolet sources.

The probability for capturing an electron from the target atom into a bound excited state of the ion in slow collisions ($v < 1$; here and in what follows we use atomic units in which $\hbar = m_e = e = 1$) depends on the exchange-interaction energy (EIE) $\Delta(R)$, which determines the frequency $\Gamma = \Delta/\hbar$ at which the electron passes from one potential well to the other.

If the electron is captured at once into many excited levels of the MCI so that its spectrum in the field of the MCI can be regarded as continuous, then the frequency for the passage of the electron from one potential well to the other will be given in terms of the EIE Δ as follows[8]:

$$\Gamma = \pi\Delta^2/2\Delta E, \quad \Delta E \to 0,$$

where $\Delta E = z^2/n^3$ is the separation between neighboring energy levels of the electron in the field of the MCI.

A quasiclassical method developed in Ref. 20 makes it possible to calculate the trend of the terms in the two-center problem for the case of the collision of a proton with a hydrogen atom, provided the initial states of the colliding atomic particles are highly excited. This method[20] gives the level energies with an error of the order of $1/n$, where n is the principal quantum number of the state of the electron in the isolated atom. When this method is applied to a system consisting of an atom and an MCI, very cumbersome calculations are encountered, especially in the region $R < z$, where R is the internuclear distance and z is the ionic charge. Semiempirical expressions for the EIE that were obtained earlier[1] by approximation of numerical calculations[21] were apparently inadequate for the physical charge-exchange mechanism since they gave different forms for the dependence of the charge-exchange cross section on the ionic charge.[1-5] Chubisov[7] gave numerical estimates of the exchange energy for the interaction of an atom with a MCI in the quasiclassical approximation, but he did not take account of the contribution from electron states with nonvanishing orbital angular momentum l.

In this paper we develop a method that makes it possible, within the framework of the quasiclassical approach, to obtain an analytic expression for the exchange energy $\Delta(R)$ for the interaction of any atom with an MCI. The expression obtained by this method for the EIE is the first term in an expansion in powers of $z^{1/2}/R$, so the criterion for the validity of the results is that $R \gg z^{1/2}$, where R is the internuclear distance. This inequality justifies the use of the quasiclassical method to calculate the electron wave function in the subbarrier region.

Let us first consider the interaction of a hydrogen atom in its $1s$ ground state with a point (structureless) charge z, taking account of the transition of the electron from one charge to the other:

$$H + A^{+z} \to p + A^{+(z-1)}. \tag{1}$$

In the limit $R \to \infty$ (when the nuclei are far apart) there are terms of two types: $E_H(R)$ and $E_z(R)$, which correspond to the localization of the electron on only one of the two charges, i.e., they correspond to the initial and final states of process (1):

$$E_H(R) = -\frac{1}{2} - \frac{z}{R} - \frac{9}{4}\frac{z^2}{R^4}, \tag{2}$$

and

$$E_z(R) = -\frac{z^2}{2n^2} - \frac{1}{R} + \frac{3}{2}\frac{n(n_1 - n_2)}{zR}, \tag{3}$$

The last term in (3) corresponds to the Stark splitting of the ion levels in the field of the proton, n, n_1, and n_2 being the principal quantum number and the parabolic quantum numbers, respectively, of the Stark states of the electron in the ion.

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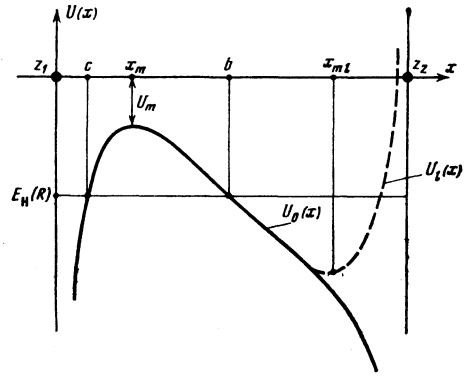

FIG. 1.

It is evident that the last term in each of (2) and (3) can be neglected when

$$R>n^2/z \tag{4}$$

In this case the positions R_c of the points where terms (2) and (3) cross depend only on the charge of the ion and the principal quantum number:

$$R_c \approx \frac{2n^2}{z(1-n^2/z^2)}, \quad n<z. \tag{5}$$

At the crossing points of terms (2) and (3) the states of the system in which the electron moves about only one center or the other have the same energy: $E_H(R_c) = E_z(R_c)$. However, the possibility that the electron may undergo a subbarrier transition from one potential well to the other leads to splitting of the terms into two terms E_H' and E_z' lying close together, which correspond to states in which the electron moves in both wells at once. Thus, the exchange splitting of the terms does not have a dynamical origin (is not associated with the motion of the nuclei), but is due to the penetrability of the potential barrier separating the two centers (see Fig. 1).

2. EXCHANGE SPLITTING AT LARGE NUCLEAR SEPARATIONS

Since electron transitions take place at large internuclear distances ($R>n^2/z$) one can use the previously developed asymptotic theory[22] to determine the EIE. The asymptotic method is based on the idea that when the electron is far from the nuclei the EIE

$$\Delta(R)=\int(\Psi_H\nabla\Psi_z-\Psi_z\nabla\Psi_H)\,dS \tag{6}$$

will be determined mainly by the electron distribution close to the line joining the two nuclei (the internuclear axis R). The problem therefore reduces to that of finding the values of the wave functions Ψ_H and Ψ_z centered on the hydrogen atom and the ion, respectively, in the vicinity of the internuclear axis.

Near the internuclear axis, the electron moves in a potential of the form

$$U=-\left(\frac{1}{x}+\frac{z}{R-x}\right), \tag{7}$$

where x is the distance of the electron from the proton along the axis R. The potential barrier (7) has its maximum at the point $x_m=R/(z^{1/2}+1)$, where we have

$$U(x_m)-E_H(R)=-(1+z^{1/2})^2/R+1/2+z/R\approx 1/2-2z^{1/2}/R. \tag{8}$$

When the internuclear distance R is smaller than $2(1+2z^{1/2})$, so that $|E_H(R)|<|U_m(R)|$, the electron moves in a generalized orbit that encompasses both nuclei; then the motion of the electron cannot be separated into motions in different potential wells and the concept of EIE is meaningless at these distances. Hence formula (6), which determines the frequency of subbarrier electron transitions from one nucleus to the other, is valid only when

$$R>2(1+2z^{1/2}). \tag{9}$$

By comparing (5) and (9) we can easily determine the values of the principal quantum number n for the states in the MCI that interact with the term $E_H(R)$ at distances $R>4z^{1/2}$:

$$z>n\geqslant 2^{1/2}z^{1/4}. \tag{10}$$

From inequality (10) we can find the values of the ionic charge z for which the number of ionic terms $E_z(R)$ that interact with the hydrogen term $E_H(R)$ will be greater than unity:

$$z>4.$$

To evaluate the wave functions Ψ_H and Ψ_z in (6) that specify the behavior of the electron below the potential barrier when it is centered on the proton or the MCI, respectively, we adopt the quasiclassical approach and express the wave functions in the form

$$\Psi_H=\frac{A}{r_1\sqrt{|p_H|}}\exp\left(-\int_c^{r_1}|p_H|dr_1\right), \tag{11}$$

and

$$\Psi_z=\frac{B}{r_2\sqrt{|p_z|}}\exp\left(-\int_b^{r_2}|p_z|dr_2\right), \tag{12}$$

with

$$p_H=\left(1+\frac{2z}{R}-\frac{2z}{|R-r_1|}-\frac{2}{r_1}\right)^{1/2}, \tag{13}$$

and

$$p_z=\left(\frac{z^2}{n^2}+\frac{2}{R}-\frac{2z}{r_2}-\frac{2}{|R-r_2|}\right)^{1/2}. \tag{14}$$

Here p_H and p_z are the quasimomenta of the electron centered on the proton and the ion, respectively, r_1 and r_2 are the distances of the electron from the proton and the ion, and A and B are constants. The constant A can be evaluated by matching wave function (11) to the well-known ground-state wave function of the hydrogen atom; the result is

$$A=e^{-1}/2\sqrt{\pi}. \tag{15}$$

The constant B can be evaluated from the condition that the oscillating wave function for the electron in the broad potential well of the MCI,[23]

$$\Psi_z(r_2<b)=\frac{C}{r_2\sqrt{p_z}}\cos\left(\int p_z\,dr_2-\frac{\pi}{4}\right)$$

be normalized; the result is

$$B=1/2C=z/2^{1/2}\pi n^{3/2}. \tag{16}$$

In calculating the integral (6) we make use of the fact that it converges rapidly near the internuclear axis. For the integration surface S we take a plane perpendicular to the internuclear axis at distances R_1 from the proton and R_2 from the MCI (so that $R_1+R_2=R$). Then the position of the electron on the plane S near

the axis R can be specified approximately as

$$r_1 \approx x+\rho^2/2x,\ r_2=R-x+\rho^2/2(R-x), \tag{17}$$

where x is the distance along the internuclear axis from the integration surface S to the proton and ρ is the radius vector in the plane S that specifies the distance of the electron from the internuclear axis, so that in formula (6) we have $d\mathbf{S}=2\pi\rho d\rho$ and $0\leq\rho\leq R_1$.

Let us evaluate the EIE near the term-crossing point where $p_H \approx p_g$. Substituting (11) and (12) into (6) and assuming that the integral in (6) is determined mainly by the exponential dependence of the wave functions (11) and (12) and converges rapidly in the region $\rho \leq R_1$, we obtain the following expression for the exchange interaction:

$$\Delta(R)=\frac{4\pi AB}{Rp_{max}}\exp\left(-\int_c^b |p|dx\right), \tag{18}$$

where

$$|p|=\left(1+\frac{2z}{R}-\frac{2}{x}-\frac{2z}{R-x}\right)^{1/2},\quad p_{max}=\left(1-\frac{4z^{1/2}}{R}\right)^{1/2},$$

and p_{max} is the maximum value of the electron quasimomentum at the point $x_{max}=R_1=R/(z^{1/2}+1)$, where $dU/dx|_{R_1}=0$.

Thus, when $p_H=p_g$ the EIE is independent of the position of the integration plane S with respect to the nuclei and, as should be expected, it is determined, except for a factor outside the exponential, by the penetrability of the potential barrier.

Let us divide the range of integration $x=\{c,b\}$ in (18) into the two intervals $c\leq x<R/(z+1)$, in which the interaction of the electron with the MCI can be treated as a perturbation, and $R/(z+1)<x\leq b$, in which the field of the proton can be treated as a perturbation. On expanding the electron quasimomentum (13) or (14) within each region in the small perturbation parameter we obtain a series of tabulated integrals whose sum gives the integral in formula (18) except for a term of order $z^{1/2}/R$:

$$\int_c^b p\,dx=R-1-\ln\frac{4R^2}{z}-\frac{2(z-1)}{(1+2z/R)^{1/2}}\ln\left[\left(\frac{R}{2z}\right)^{1/2}+\left(\frac{R}{2z}+1\right)^{1/2}\right]+O\left(\frac{z^{1/2}}{R}\right). \tag{19}$$

Substituting formulas (15), (16), and (19) into Eq. (18), we finally obtain the following expression for the exchange interaction:

$$\Delta_{n,l=0}(R)=2\left(\frac{2}{\pi}\right)^{1/2}\times\frac{R}{n^{3/2}}\exp\left\{-R+\frac{2(z-1)}{(1+2z/R)^{1/2}}\ln\left[\left(\frac{R}{2z}\right)^{1/2}+\left(\frac{R}{2z}+1\right)^{1/2}\right]\right\}\left(1+O\left(\frac{z^{1/2}}{R}\right)\right). \tag{20}$$

It is easy to obtain two limiting values for $\Delta_{n0}(R)$ from Eq. (20):

$$\Delta_{n0}(R)=2\left(\frac{2}{\pi}\right)^{1/2}\frac{R}{n^{3/2}}\exp\left(-\frac{R^2}{3z}\right),\quad 4z^{1/2}<R<2z, \tag{21}$$

and

$$\Delta_{n0}(R)=\frac{2^{z+1}e^{-z}R^ze^{-R}}{z\Gamma(z)},\quad R>2z. \tag{22}$$

Equation (21) agrees with the result of a recent calculation[9] in which the action of the MCI was replaced by an electric field of strength $\varepsilon=z/R^2$. The EIE (22) for $R>2z$ can be obtained from the asymptotic theory, in which[22]

$$\Delta=\pi\varphi_H(R_1)\varphi_g(R_2),$$

where $R_1=R/(z+1)$ and $R_2=zR/(z+1)$ are the distances from the integration plane S to the proton and the MCI, $R_1+R_2=R$, and φ_H and φ_g are electron wave functions centered on the proton and the MCI, respectively.

When $z=1$, the second term in the argument of the exponential in (20) vanishes, and the exchange interaction between a hydrogen atom and a proton is given by

$$\Delta(R)=2\left(\frac{2}{\pi}\right)^{1/2}Re^{-R},$$

which differs by only 8% from the well-known formula[24] $\Delta=(4/e)Re^{-R}$. This gives an idea of the magnitude of the errors in our calculation.

Expression (20) for the exchange potential for the interaction of a hydrogen atom with an MCI is valid for an s electron. Let us extend this result to the case in which the electron orbital angular momentum l becomes different from zero when the electron undergoes a transition to the MCI.

In this case the wave function for the electron in the MCI has the form

$$\Psi_z=(2l+1)^{1/2}P_l(\cos\theta_2)\varphi_{zl}(r_2), \tag{23}$$

at distances at which the field of the proton hardly distorts the central field of the MCI. Here $\varphi_{zl}(r_2)$ is the radial wave function for the electron and is given in the quasiclassical approximation by the known formula (12) in which the constant B is independent of l despite the fact that the quasimomentum of the electron in the MCI does depend on the orbital angular momentum l:

$$p_{zl}=\left[\frac{z^2}{n^2}-\frac{2z}{r_2}+\frac{l(l+1)}{r_2^2}\right]^{1/2}=\left[\left(\frac{z}{n}-\frac{n}{r_2}\right)^2-\frac{n^2}{r_2^2}\left(1-\frac{l(l+1)}{n^2}\right)\right]^{1/2}, \tag{24}$$

where r_2 and θ_2 are the spherical coordinates of the electron in the MCI (we recall that the projection of the electron angular momentum onto the axis vanishes, so that the electron wave functions are real and do not depend on the azimuthal numbers).

The following conclusions can be drawn from an analysis of expression (24) for the quasimomentum of the electron in the central field of the MCI:

1) When $l(l+1)\ll n^2$ the centrifugal energy $l(l+1)/r_2^2$ may be treated as a small addition to the kinetic energy $p_0^2=z^2/n^2-2z/r_2$ of the s electron centered on the MCI, and the quasimomentum (24) may be expanded in powers of the small ratio of the centrifugal energy to the kinetic energy:

$$p_{zl}\approx p_0+l(l+1)/2r_2^2p_0+\ldots;$$

2) For values $l\approx n$ of the orbital angular momentum, the potential barrier between the two Coulomb centers is virtually impenetrable and the contribution to the charge exchange from states with $l\sim n$ may be neglected.

For small values of l the quasiclassical expression for the radial wave function can be written in the form

$$\varphi_{zl}(r)=\varphi_{z0}(r)\exp\left(-\frac{1}{2}\int_b^r\frac{l(l+1)}{r^2p_0(r)}dr\right), \tag{25}$$

where φ_{z0} corresponds to zero orbital angular momentum and $p_0=(z^2/n^2+2/R-2z/r)^{1/2}$ is the quasimomentum of an s electron in the MCI.

We shall assume, as before, that the main contribution to the integral (6) comes from values of the angles such that $l\theta_2<1$; hence the angular dependence may be neglected and the Legendre polynominal in (23) replaced by unity when $l\ll n$. In that case the main dependence of the exchange interaction potential on the angular momentum l will be due to the radial wave function[9] (25):

$$\Delta_{nl}(R)=(2l+1)^{1/2}\exp[-l(l+1)/2z]\Delta_{n0}(R), \tag{26}$$

where $\Delta_{n0}(R)$ is the EIE (20) for zero orbital angular momentum.

Formula (26) admits a simple generalization to the case in which the electron moves in the field of two arbitrary charges z_2 and $z_1(z_2>z_1)$. It is sufficient to make the substitutions $R\to z_1R$ and $z\to z_2/z_1$ in Eq. (19) and take account of the fact that $p_{\max}=z_1$:

$$\Delta_{nl}(z_1,z_2,R)=2(2l+1)^{1/2}\exp[-l(l+1)z_1/2z_2]\left(\frac{2}{\pi}\right)^{1/2}\frac{Rz_1^{3}}{n^{1/2}}$$
$$\times\exp\left\{-z_1R+\frac{2(z_2/z_1-1)}{(1+2z_2/z_1^2R)^{1/2}}\ln\left[\left(\frac{z_1^2R}{2z_2}\right)^{1/2}+\left(\frac{z_1^2R}{2z_2}+1\right)^{1/2}\right]\right\}. \tag{27}$$

3. THE GENERAL CASE

If the atom and the MCI have complex electronic structures, the exchange energy for the interaction between them at large distances will be due to subbarrier transition of one of the outer atomic electrons to a level of the MCI that is available to it in accordance with the Pauli principle and the vector addition rules. Then we may assume that the electron moves in the Coulomb fields of the two atomic cores with charges z_a and z. In that case the wave function for system (1) in the initial state has the form

$$\Psi_{in}=\sum_i^{N_a}\frac{(-1)^{i-N_a}}{\sqrt{N_a}}\begin{bmatrix}s_i & S_a & I\\ m_s & M_S & M_I\end{bmatrix}\Psi^{z_a}_{LM_L S_a M_S}\Phi^{z}_{i m_{s_i} m_s}. \tag{28}$$

Here the summation is taken over the spin projections m_s and M_S for the ion and the atom, respectively, so that wave function (28) corresponds to a total spin of $\mathbf{I}=\mathbf{S}_a+\mathbf{s}_i$ with projection $M_I=m_s+M_S$ for the quasimolecule, and $\Psi^{z_a}_{LM_L SM_S}$ is the wave function of the atom, which, in accordance with the fractional parentage scheme,[25] can be written as

$$\Psi^{z_a}_{LM_LSM_S}=\sum G^{L_aS_a}_{ls}\begin{bmatrix}s_a & \frac{1}{2} & S_a\\ m_s & \sigma & M_S\end{bmatrix}\begin{bmatrix}l & l_e & L\\ m & \mu & M\end{bmatrix}\Phi^{z_a}_{lms m_s}\varphi^{z_a}_{l_e\mu\frac{1}{2}\sigma}, \tag{29}$$

where l_e is the orbital angular momentum of the outer electron, G^{LS}_{ls} is a fractional parentage coefficient, Φ^{z_a} and Φ^{z} are the wave functions of the atomic core and the MCI,

$$\begin{bmatrix}j_1 & j_2 & j_3\\ m_1 & m_2 & m_3\end{bmatrix}$$

is a Clebsch-Gordan coefficient, and lm, LM, and $l_e\mu$ are the orbital angular momenta of the atomic core, the atom, and the electron, respectively, together with their projections.

After transition of the atomic electron to the outer shell of the MCI, the wave function for system (1) becomes

$$\Psi_f=\sum\begin{bmatrix}s_a & S_i & I\\ m_s & M_S & M_I\end{bmatrix}\begin{bmatrix}s_i & \frac{1}{2} & S_i\\ m_s\sigma & & M_S\end{bmatrix}\begin{bmatrix}l_i & l_e' & L_i\\ m' & \mu' & M'\end{bmatrix}\Phi^{z_a}_{lm s_a m_s}$$
$$\times\Phi^{z}_{lm s_a m_s}\varphi^{z}_{l_e'\mu'\frac{1}{2}\sigma'}. \tag{30}$$

The functions φ^{z_a} and φ^{z} in (29) and (30) are the one-electron quasiclassical wave functions (11) and (12) centered on the atom and the MCI, respectively, with the coefficients A and B are given by

$$A=\frac{A_0e^{-z_a/\gamma}\gamma^{1/2}}{2\sqrt{\pi}}\left(\frac{z_a}{2\gamma^2}\right)^{z_a/\gamma},\qquad B=\frac{z}{2^{1/2}n^{3/2}\pi}, \tag{31}$$

where $\gamma^2/2$ is the binding energy of the outer electron in the atom.

After cumbersome calculations that we omit, one can obtain the following expression for the exchange energy in the interaction of any atom with a MCI in the quasiclassical approximation:

$$\Delta=\sqrt{N_a}\,[(2s_a+1)(2s_i+1)]^{1/2}\begin{pmatrix}s_a & \frac{1}{2} & S_a\\ s_i & I & S_i\end{pmatrix}G^{L_aS_a}_{l_as_a}\begin{bmatrix}l_a & l_e & L_a\\ M & 0 & M\end{bmatrix}$$
$$\times\begin{bmatrix}l_i & l_e' & L_i\\ m & 0 & m\end{bmatrix}\Delta_{l_el_e'}(R), \tag{32}$$

$$\Delta_{l_el_e'}(R)=A_0\frac{z(2\gamma^2/z)^{z_a/\gamma}}{(2\pi\gamma)^{1/2}n^{3/2}}[(2l_e+1)(2l_e'+1)]^{1/2}\exp\left[-\frac{l_e'(l_e'+1)\gamma}{2z}\right]R^{2z_a/\gamma-1}$$
$$\times\exp\left\{-\gamma R+\frac{2(z-z_a)}{\gamma(1+2z/\gamma^2R)^{1/2}}\ln\left[\left(\frac{\gamma^2R}{2z}\right)^{1/2}+\left(\frac{\gamma^2R}{2z}+1\right)^{1/2}\right]\right\}, \tag{33}$$

where

$$\begin{pmatrix}s_a & \frac{1}{2} & S_a\\ s_i & I & S_i\end{pmatrix}$$

is Wigner's 6-j symbol, A_0 is an asymptotic coefficient that specifies the magnitude of the electron at the periphery of the atom:

$$\varphi(r\to\infty)=A_0r^{z_a/\gamma-1}e^{-\gamma r},$$

N_a is the number of valence electrons in the atom, S_a, s_i, and I are the spins of the atom, the ion, and the whole system, $\mathbf{I}=\mathbf{S}_a+\mathbf{s}_i$, and s_a and s_i are the spins of the atomic core and the ion after change exchange.

4. CONCLUSION

In this paper we employed the quasiclassical approach to obtain an analytic expression for the exchange energy in the interaction of an atom with a multiply charged ion for the limiting case of large distances between the interacting systems ($R>z^{1/2}$). The exchange-interaction potential $\Delta(R)$ is expressed in terms of known parameters of the atom and the multiply charged ion: the charge z of the ion, the binding energy $\gamma^2/2$ of the electron in the atom, and the magnitudes of the orbital angular momentum and spin of the electron in the atom and in the ion (the projection of the orbital angular momentum onto the line joining the nuclei is zero—

the electron is in a σ state).

There are two ranges of values for the distance R between the nuclei of the interacting systems in which the exchange-interaction potential $\Delta(R)$ is represented by different functions of R. Thus, when $R \gg 2z$, Eq. (20) for the exchange interaction reduces to the limiting expression (22), which can be derived within the framework of the known asymptotic theory.[21] When $4z^{1/2} < R < 2z$, the asymptotic theory is inapplicable, owing to the fact that when $z > 4$ the asymptotic expression for the wave function for the electron centered on the ion cannot be used at these distances. At the same time, the calculations show that the quasiclassical method worked out here correctly gives the interaction both in the intermediate range $4z^{1/2} < R < 2z$ and in the asymptotic limit $R > 2z$. In addition, the quasiclassical approach allows the results to be easily generalized to the case in which the electron's orbital angular momentum l ceases to be zero when the electron makes a transition from the atom to the MCI.

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Stimulated photoassociation in the field of an intense electromagnetic wave

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The quantum-mechanical problem of the stimulated transition of a system in the field of an intense electromagnetic wave from the continuous spectrum to a bound state having a finite lifetime is considered. The formulas derived are used to calculate the stimulated production of mesic atoms and the mesic molecule $dd\mu$. It is shown that the probability for the production of such systems may be considerably enhanced in the presence of an external electromagnetic field.

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1. Transitions from the continuous spectrum to a bound state with spontaneous emission of a photon are possible in particle collisions. As a rule, however, the probability for such transitions is small. The probability for such transitions in atomic collisions may be considerably enhanced in the field of an intense electromagnetic wave, however, on account of stimulated photon emission.[1–3]

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