# Strong exchange interaction effects in a homogeneous electric field

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The exchange interaction between states localized at different centers in the presence of an external electric field is considered. Exact (in the field) expressions for the wave function, the Green function, and the particle-scattering operator are found in the long-wave approximation for an arbitrary number of interaction centers. A characteristic equation is obtained which is exact in all the energy parameters and which determines the energy levels of the system with allowance for the exchange interaction, the Stark effect, and the field ionization. Different limiting cases are considered, and the auto-ionization widths of the multicenter states are found. It is shown that near the field points of pseudointersection of the terms the exchange interaction leads to the exponential growth of the rate of auto-ionization of the state with the higher binding energy. The problem of the distinctive features of the interaction between the field and quasi-stationary states is discussed. The possibility of a marked field stabilization of resonances is established. The effects considered are of interest in connection with the physics of atomic collisons in a field, the field ionization of negative molecular ions, and charge transfer between impurities in a solid.

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## 1. INTRODUCTION

The exchange interaction between states localized at different centers, i. e., that part of the total interaction which is due to the overlap of the coordinate wave functions, plays an important role in many physical processes and systems. The pertinent examples are well known, and may pertain to different quantum objects. In the present paper we consider the effects of the exchange interaction in the presence of a homogeneous electric field. Physical examples—the interaction of negative ions with neutral atoms in the presence of a field, charge transfer between impurities in a solid, the photodetachment of an electron from a negative ion—are discussed in the Conclusion.

It is well known that an exchange interaction cannot be properly taken into account in the framework of perturbation theory based on a power-series expansion in a small energy parameter. The presence of a constant external force gives rise to additional characteristics of the systems in question. In an external electric field the energy spectrum of an atomic system becomes strictly speaking, continuous.[1] The escape to infinity of a particle is described by analytical expressions having essential singularities with respect to the field. The field-ionization effects cannot, in principle, be described in the framework of the conventional iteration schemes for solving the Schrödinger equation (or the equivalent Lippmann–Schwinger equations).

The self-consistent solution to the problem of the exchange interaction of localized and quasi-localized states is possible to obtain in the presence of a small coordinate parameter in the system. The exchange interaction is most effective for states with small binding energies $E$, for which states the particle wavelength $\lambda$ is long: $\lambda \gg a$ ($\lambda = (2|E|)^{-1/2}$, $m = \hbar = e = 1$), where $a$ is the range of the potential in whose isolated field the unperturbed state is realized. The short-range interaction can be taken into account either through a boundary condition imposed on the wave function,[2–4] or by the use of a separable representation of the pair scattering operators (see, for example,[5]) in the equations of multiple interactions.[6–11] Under these conditions the problem reduces to one of solving algebraic equations written in terms of the Green function for long-range forces and the physical amplitudes of scattering of a particle by the individual short-range centers. The generalization of the corresponding equations to interactions of the general type (including the nonpotential interactions connected, for example, with the exchange-particle creation-annihilation events) is contained in[11].

The long-wave approximation (the short-range interaction model) has been widely used in recent years to describe the exchange interaction between the stationary states of atoms,[7–12] negative molecular ions,[4,13,14] and impurities in a solid.[15,16] It is in this same approximation that the exact (in the field) solution to the problem of the interaction between an external field and a weakly-bound $S$ electron of a negative atomic ion has been obtained.[17–19]

Two single-center systems (the negative ion[17] and the hydrogen-like atom[1,19]) exhaust the known examples of systems for which the three-dimensional problem of the interaction of a bound state with an electric field has been solved. In the present paper we consider multicenter systems. Exact (in the field) analytical expressions are found in the long-wave approximation for the one-particle Green function, the scattering operator, and the wave functions of quasi-bound states. A characteristic equation is obtained which is exact in all the energy parameters of the system, and which determines the electron-energy levels with allowance for the exchange interaction, the Stark effect, and the field ionization. Using the two-center system as an example, we carry out a detailed investigation of the problem of the distinctive features of the exchange interaction in the presence of an electric field. It is shown that the electron-field interaction leads to the appearance in the term system of "field" points of pseudointersection[1] in the vicinity of each of which the autoionization width of the state with the higher binding energy exponentially increases. The distinctive features of the decay of quasi-stationary states in an electric field are considered. It is shown that a marked stabilization of resonances in a strong electric field is possible. As the intensity of the field is decreased, the width of the resonance level oscillates with a decreasing amplitude and an increasing frequency, assuming its minimum values under the conditions of maximal influence of the Stark effect.

## 2. THE GENERAL SOLUTION. COMPUTATION OF THE FIELD GREEN FUNCTION

The Hamiltonian of the system under consideration is given by

$$H=-\Delta/2+\sum_{s} U_s(\mathbf{r}-\mathbf{R}_s)-\mathbf{f}\cdot\mathbf{r}\ (e=m=\hbar=1). \qquad (1)$$

Here $\mathbf{r}$ is the radius vector of the electron, $U_s$ is its interaction with the $s$-th center located at the point $\mathbf{R}_s$, and $\mathbf{f}$ is the electric-field intensity.

The equations determining the Green function $\hat{G}=(E-H)^{-1}$ of the system, the scattering operator $\hat{T}$, and the wave function of the quasi-bound state have the form[7–11, 15]

$$\Psi(\mathbf{r})=\int G_f(\mathbf{r},\mathbf{r}_1)\sum_{s}\tau_s(\mathbf{r}_1)\,d\mathbf{r}_1, \qquad (2)$$

$$\tau_s(\mathbf{r})=\int t_s^f(\mathbf{r},\mathbf{r}_1)\sum_{s'\neq s} G_f(\mathbf{r}_1,\mathbf{r}_2)\,\tau_{s'}(\mathbf{r}_2)\,d\mathbf{r}_1\,d\mathbf{r}_2, \qquad (3)$$

$$T(\mathbf{r},\mathbf{r}')=\sum_{s} T_s(\mathbf{r},\mathbf{r}'), \qquad (4)$$

$$T_s(\mathbf{r},\mathbf{r}')=t_s^f(\mathbf{r},\mathbf{r}')+\int t_s^f(\mathbf{r},\mathbf{r}_2)\sum_{s'\neq s} G_f(\mathbf{r}_2,\mathbf{r}_3)\,T_{s'}(\mathbf{r}_3,\mathbf{r}')\,d\mathbf{r}_2\,d\mathbf{r}_3, \qquad (5)$$

$$G(\mathbf{r},\mathbf{r}')=G_f(\mathbf{r},\mathbf{r}')+\int G_f(\mathbf{r},\mathbf{r}_1)\sum_{s}\Omega_s(\mathbf{r}_1,\mathbf{r}')\,d\mathbf{r}_1, \qquad (6)$$

$$\Omega_s(\mathbf{r},\mathbf{r}_1)=\int t_s^f(\mathbf{r},\mathbf{r}_2)\,G_f(\mathbf{r}_2,\mathbf{r}_1)\,d\mathbf{r}_2 + \int t_s^f(\mathbf{r},\mathbf{r}_2)\sum_{s'\neq s} G_f(\mathbf{r}_2,\mathbf{r}_3)\,\Omega_{s'}(\mathbf{r}_3,\mathbf{r}_1)\,d\mathbf{r}_2\,d\mathbf{r}_3. \qquad (7)$$

Here $\hat{G}_f=(E+\Delta/2+\mathbf{f}\cdot\mathbf{r})^{-1}$ is the electron Green function in a homogeneous electric field. In the coordinate representation[20, 21]

$$G_f(\mathbf{r},\mathbf{r}',E)=-\frac{i}{(2\pi i)^{3/2}}\int_0^\infty\frac{dt}{t^{3/2}}\exp\left\{i\left[\frac{(\mathbf{r}-\mathbf{r}')^2}{2t}+E_f(\mathbf{r},\mathbf{r}')t-\frac{f^2t^3}{24}\right]\right\}, \qquad (8)$$

$$E_f(\mathbf{r},\mathbf{r}')=E+{}^1/_2\mathbf{f}(\mathbf{r}+\mathbf{r}'). \qquad (9)$$

In Eqs. (3), (5), and (7) $t_s^f(\mathbf{r},\mathbf{r}')$ is the operator of scattering by the $s$-th center of an electron interacting with the field:

$$t_s^f=U_s+U_s(E+\Delta/2+\mathbf{f}\cdot\mathbf{r}-U_s)^{-1}U_s\ ; \qquad (10)$$

$E$ is the total electron energy measured from the electric-field potential at the point $\mathbf{r}=0$. The electron energy enters into the Eqs. (1)–(7) only in the combination (9); therefore, they are invariant with respect to the choice of the coordinate origin.

The functions $\tau_s$ and the operators $\hat{T}_s$ and $\hat{\Omega}_s$ are different from zero only in finite regions of space where the potentials $U_s$ act. It is precisely this circumstance that allows us in the most general case to formulate algebraic equations for quantities that are the projections of $\tau_s$, $T_s$, and $\Omega_s$ on states with definite values of the orbital momentum $l_s$ relative to the centers $R_s$.[10, 11] In the long-wave approximation the corresponding equations get substantially simplified. For $\lambda\gg a_s$, allowance for higher angular momenta ($l_s\neq 0$) offers no difficulties and does not introduce any fundamental changes into the nature of the general solution. Therefore, we shall restrict ourselves to the solution of the problem with allowance for only $S$-scattering. The changes that must be made in the final expressions when describing the exchange interaction of $p$ and $d$ states will be indicated below.

In the leading approximation in the small parameter $a_s/\lambda$ the form of the operators $t_s^f(\mathbf{r},\mathbf{r}')$ is known (see[9], formula (10)):

$$t_s^f(\mathbf{r},\mathbf{r}')=\frac{2\pi}{\varkappa_s+i\sqrt{2E}-2\pi G_f^r(\mathbf{R}_s,\mathbf{R}_s)}\,\delta(\mathbf{r}-\mathbf{R}_s)\,\delta(\mathbf{r}'-\mathbf{R}_s). \qquad (11)$$

Here $\varkappa_s^{-1}$ is the scattering length for scattering of a free electron by the $s$-th center and $G_f^r(\mathrm{R}_s, \mathrm{R}_s)$ is the regularized, equal-argument field Green function:

$$G_f^r(\mathbf{R}_s,\mathbf{R}_s)=\lim\,[G_f(\mathbf{r},\mathbf{R}_s)-G_0(\mathbf{r},\mathbf{R}_s)],\ \mathbf{r}\to\mathbf{R}_s; \qquad (12)$$

$G_0$ is the Green function for free motion:

$$G_0(\mathbf{r},\mathbf{R}_s)=-\frac{1}{2\pi}\frac{\exp(i\sqrt{2E}\,|\mathbf{r}-\mathbf{R}_s|)}{|\mathbf{r}-\mathbf{R}_s|}. \qquad (13)$$

According to (6), (7), and (11), the algebraic equations determining in the long-wave approximation the Green function of the system have the form[2]

$$\Omega_s(\mathbf{R}_s,\mathbf{r})=2\pi\left[G_f(\mathbf{R}_s,\mathbf{r})-\sum_{s'\neq s}G_f(\mathbf{R}_s,\mathbf{R}_{s'})\,\Omega_{s'}(\mathbf{R}_{s'},\mathbf{r})\right]\cdot\left[\varkappa_s+i\sqrt{2E}-2\pi G_f^r(\mathbf{R}_s,\mathbf{R}_s)\right]^{-1}. \qquad (14)$$

The poles of the Green function, i.e., the complex roots of the equation

$$\det\left|\delta_{ss'}-\frac{2\pi(1-\delta_{ss'})\,G_f(\mathbf{R}_s,\mathbf{R}_{s'})}{\varkappa_s+i\sqrt{2E}-2\pi G_f^r(\mathbf{R}_s,\mathbf{R}_s)}\right|=0. \qquad (15)$$

characterize the energy spectrum of the system.

The problem thus reduces to the problem of the computation of the field Green function $G_f(\mathbf{r}, \mathbf{r}', E)$ at the points $\mathbf{R}_{s'}$, with the subsequent solution of the transcendental equation (15). In the coordinate representation it is possible to obtain a closed expression for $G_f(\mathbf{r}, \mathbf{r}', E)$, i.e., to solve the problem exactly with respect to the energy parameters. Indeed, let us align the $z$ axis along the field $\mathbf{f}$ and introduce the new variables defined by the relation

$$2\eta=z+z'+|\mathbf{r}-\mathbf{r}'|,\quad 2\zeta=z+z'-|\mathbf{r}-\mathbf{r}'|. \qquad (16)$$

Notice that the integral

$$I=\frac{-i}{(2\pi i)^{1/2}}\int_0^\infty\frac{dt}{\sqrt{t}}\exp\left\{i\left[\frac{(\zeta-\eta)^2}{2t}+E_f(\zeta,\eta)t-\frac{f^2t^3}{24}\right]\right\} \qquad (17)$$

is equal to the one-dimensional Green function for an electron in a homogeneous field, the regular method of construction of which is well known (see, for example,[22] p. 136). Taking into account the fact that $\eta \geqslant \zeta$ for all values of **r** and **r**′, we obtain

$$G_f(\mathbf{r},\mathbf{r}',E)=\frac{1}{2\pi(2f)^{1/3}|\mathbf{r}-\mathbf{r}'|}\left(\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\zeta}\right)V(x_1)[U(x_2)+iV(x_2)];$$
$$x_1=-\frac{2E_f(\zeta,\zeta)}{(2f)^{2/3}},\quad x_2=-\frac{2E_f(\eta,\eta)}{(2f)^{2/3}}. \tag{18}$$

Here $V(z)$ and $U(z)$ are the Airy functions of the first and second kinds respectively.[23]

It is easy to notice the analogy between the obtained expression and the well-known Hostler representation for the Coulomb Green function.[24] The similarity between the analytic expressions for the operators $(E+\Delta/2+1/r)^{-1}$ and $(E+\Delta/2+\mathbf{f}\cdot\mathbf{r})^{-1}$ in the coordinate representation is natural and is connected with the fact that the field of a remote Coulomb center can always be approximated in a finite region of space by a homogeneous field.

For $\mathbf{r}\to\mathbf{r}'$, the function $G_f(\mathbf{r},\mathbf{r}',E)$ diverges like $(2\pi|\mathbf{r}-\mathbf{r}'|)^{-1}$, and the regularized Green function (12) remains finite:

$$G_f'(\mathbf{R}_0,\mathbf{R}_0)=\frac{(2f)^{1/3}}{2\pi}\{x_0V(x_0)[U(x_0)+iV(x_0)]$$
$$-V'(x_0)[U'(x_0)+iV'(x_0)]\}+\frac{i\sqrt{2E}}{2\pi};$$
$$x_0=-2E_f(\mathbf{R}_0,\mathbf{R}_0)/(2f)^{2/3}. \tag{19}$$

Notice that the formula (19) coincides up to the substitutions

$$R_0\to(r-2n^2),\quad f\to-1/2n^4,\quad i\to\operatorname{ctg}\pi n$$

with the regularized Green function of the attractive Coulomb center for a negative energy $E=-1/2n^2$ close to the classical reversal point (see the formula (20) in[7]).

The problem of the computation of the one-particle Green function in a homogeneous electric field has been the subject of a large number of papers (see, for example,[21,25]), but the closed representation (18) was apparently unknown. The approach used above also allows us to find in its explicit form the multidimensional Green function

$$G_f(\{\mathbf{r}_i\},\{\mathbf{r}_i'\},E)=\left(E+\frac{1}{2}\sum_{i=1}^{N}\Delta_i+\mathbf{f}\sum_{i=1}^{N}\mathbf{r}_i\right)^{-1},$$

which describes the motion of an odd number $(N=2k+1)$ of non-interacting (among themselves) electrons located in a homogeneous electric field. We give the final result without derivation:

$$G_f=\frac{N^{N/2-1/3}}{(2f)^{1/3}}\left\{\frac{1}{2\pi N^{1/6}}\left[\sum_i(\mathbf{r}_i-\mathbf{r}_i')^2\right]^{-1/2}\left(\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\zeta}\right)\right\}^{(3N-1)/2}$$
$$\cdot V(x_1)[U(x_2)+iV(x_2)];$$
$$2\eta=N^{-1/2}\sum_i(z_i+z_i')+N^{-1/6}\left[\sum_i(\mathbf{r}_i-\mathbf{r}_i')^2\right]^{1/2};$$
$$2\zeta=N^{-1/2}\sum_i(z_i+z_i')-N^{-1/6}\left[\sum_i(\mathbf{r}_i-\mathbf{r}_i')^2\right]^{1/2};$$
$$x_1=-\frac{2E_f(N,\zeta,\zeta)}{N^{1/3}(2f)^{2/3}},\quad x_2=-\frac{2E_f(N,\eta,\eta)}{N^{1/3}(2f)^{2/3}},\quad E_f(N,\zeta,\zeta)\equiv E+N^{1/2}f\zeta. \tag{20}$$

For an even number of particles interacting with an electric field, we can find only an integral representation of the Green function, the situation here being entirely analogous to the situation that obtains for multidimensional oscillators.[26]

The formulas (14), (15), and (18) solve the formulated problem in its general form. The asymptotic representations of the Airy functions are well known,[23] and this allows us to write out without difficulty the solutions to all the equations in the various particular and limiting cases. The corresponding examples are considered below in Secs. 3 and 4. It should, however, be noted that for a weak electric field $(f/|E|^{3/2}\ll 1)$ the asymptotic expressions for $G_f(\mathbf{r},\mathbf{r}',E)$ can be found directly from the representation (8). Such an approach is physically very graphic, and can be used in the case of quasiclassical motion in the field of a potential of the general type.

With that end in view, let us consider the integral (8) for $E_f>0$. It is not difficult to verify that in the lowest approximation with respect to the small parameter $f/E_f^{3/2}$ the dominant contribution to (8) is made by the two slightly overlapping integration domains:

$$t\sim t_1=|\mathbf{r}-\mathbf{r}'|/\sqrt{2E_f},\quad t\sim t_2=2\sqrt{2E_f}/f.$$

The time $t_1$ is equal to the classical time of direct transit of a particle from the point **r** to the point **r**′ and $t_2$ is equal to the classical time of travel of a particle to the point **r**′ after its reflection from the field potential barrier. For $t\sim t_1$, we have $f^2t^3\ll 1$, i.e., that the role of the external field is insignificant. In the second integration domain the instant $t_2$ is the stationary-phase point, the vicinity of which makes a contribution having an essential singularity with respect to the field. As a result, we obtain

$$G_f(\mathbf{r},\mathbf{r}',E)=-\frac{\exp[ik_f\rho]}{2\pi\rho}+\frac{if}{4\pi k_f^2}\exp\left[i\left(\frac{2k_f^3}{3f}+\frac{f\rho^2}{4k_f}\right)\right]$$
$$(k_f=(2|E_f(\mathbf{r},\mathbf{r}')|)^{1/2},\quad \rho=|\mathbf{r}-\mathbf{r}'|). \tag{21}$$

For negative energy values, i.e., for $E_f<0$, the saddle point of the integrand is located on the lower imaginary semiaxis of the complex $t$ plane and corresponds to a purely imaginary "time" $t=-i2k_f/f$ of escape of a particle from under the field potential barrier. Therefore, in computing the integral it is convenient to deform the contour, integrating first along the imaginary semiaxis up to the saddle point, and then along the right branch of the saddle contour parallel to the real axis and going to infinity. The first integration determines the real part of the Green function, the dominant contribution being again given by the values of $t$ close in absolute value to $t_1=\rho/k_f$, for which $f^2t^3\ll 1$. This allows us to expand $\exp(f^2t^3/24)$ in a series and, with exponential accuracy, replace the upper limit of integration by infinity. As a result, we have:

$$\operatorname{Re}G_f=-2\sum_{m=0}^{\infty}\left(\frac{\rho}{k_f}\right)^{3m-1/2}K_{3m-1/2}(k_f\rho)\frac{f^{2m}}{(2\pi)^{3/2}m!2^{3m}3^m}, \tag{22}$$

where $K_\nu(z)$ is the Macdonald function. The imaginary part of the Green function, which corresponds to the escape of a particle to infinity, is determined by the remaining part of the contour:

$$\operatorname{Im} G_f = -\frac{f}{8\pi k_f^2}\exp\left[-\frac{2}{3}\frac{k_f^3}{f}-\frac{f\rho^2}{4k_f}\right]. \tag{23}$$

(Because of the Stokes phenomenon, the asymptotic formulas (21), (22), and (23) do not coincide when analytically continued.)

The asymptotic representations of the Green function in a strong electric field ($f/k_f^3 \geq 1$) can easily be found from the exact formula (18). The plots of the functions $\operatorname{Re}G_f^r(x)$ and $\operatorname{Im}G_f^r(x)$ for $x = -2E/(2f)^{2/3} \sim 1$, which will be needed below, are shown in the figure.

## 3. THE POLARIZABILITY OF AND DISINTEGRATION PROBABILITY FOR NEGATIVE MOLECULAR IONS LOCATED IN A HOMOGENEOUS ELECTRIC FIELD

Equation (15), together with the above-found representations of the field Green function, determines the energy levels of quasistationary, multicenter states for arbitrary relations between the energy parameters of the system. Of the greatest interest physically is the case of weak ($f/k_f^3 \ll 1$) electric fields, to the analysis of which case we here restrict ourselves.

Let us consider a negative diatomic ion. Retaining in the expansion (22) the two leading terms, we have3)

$$\prod_{s=1}^{2}\left[\varkappa_s - k_f(\mathbf{R}_s,\mathbf{R}_s)+\frac{f^2}{8k_f^5(\mathbf{R}_s,\mathbf{R}_s)}+\frac{if}{4k_f^2(\mathbf{R}_s,\mathbf{R}_s)}\exp\left\{-\frac{2}{3}\frac{k_f^3(\mathbf{R}_s,\mathbf{R}_s)}{f}\right\}\right]=\left[\frac{\exp\{-k_f(\mathbf{R}_1,\mathbf{R}_2)R\}}{R}+\frac{f^2}{8k_f^3(\mathbf{R}_1,\mathbf{R}_2)}\cdot\left(\frac{1}{k_f^2(\mathbf{R}_1,\mathbf{R}_2)}+\frac{R}{k_f(\mathbf{R}_1,\mathbf{R}_2)}+\frac{R^2}{3}\right)\exp\{-k_f(\mathbf{R}_1,\mathbf{R}_2)R\}+\frac{if}{4k_f^2(\mathbf{R}_1,\mathbf{R}_2)}\exp\left\{-\frac{2}{3}\frac{k_f^3(\mathbf{R}_1,\mathbf{R}_2)}{f}-\frac{fR^2}{4k_f(\mathbf{R}_1,\mathbf{R}_2)}\right\}\right]^2. \tag{24}$$

Here $R = |\mathbf{R}_1 - \mathbf{R}_2|$ is the interatomic distance, $\varkappa_{1(2)}^{-1}$ are the scattering lengths for scattering of an electron by the atom $A(B)$, and $k_f(\mathbf{R}_s, \mathbf{R}_{s'}) = [-2|E_f(\mathbf{R}_s, \mathbf{R}_{s'})|]^{1/2}$. For $f \to 0$, Eq. (24) goes over into the Firsov–Smirnov equation,[4] which describes the exchange interaction in the absence of a field. The zeros of the left-hand side determine the Stark effect and the auto-ionization of negative atomic ions.[17] For $f \neq 0$ the obtained equation with allowance for the right-hand side describes the exchange interaction between resonances of the "field" type. The procedure for solving Eq. (24) and the explicit form of its roots are determined by the energy parameters

$$\varkappa_1^2,\quad \Delta,\quad u(\mathbf{f}\cdot\mathbf{R}),\ \Delta_2,\quad \omega(k,R),\quad \Gamma_s(\varkappa_s,f). \tag{25}$$

Here $\varkappa_1^2/2$ is the electron affinity for the atom $A$, $\Delta = (\varkappa_1^2 - \varkappa_2^2)/2$ is the resonance defect of the noninteracting states, $u(fR) = \mathbf{f}\cdot\mathbf{R}$ is the voltage potential of the field between the points $\mathbf{R}_1$ and $\mathbf{R}_2$, $\omega(k, R)$ is the electron-transition frequency in the term system of the ion $AB^-$, $\Delta_2 = \beta f^2$ is the quadratic (in the field) Stark shift of the ionic level ($\beta(R)$ is the polarizability of the molecular ion $AB^-$), and $\Gamma_s$ is the auto-ionization width of the $s$-th atomic ion. The possible relations between the above-indicated parameters determine the physically different situations in the system under consideration.

For example, in the case of weak interaction between the field and the homoatomic ion $A_2^-$ ($\Delta = 0$, $u \ll \omega \sim \alpha R^{-1} \times e^{-\alpha R}$, $\beta f^2 \ll \omega$), expanding $k_f(\mathbf{R}_s, \mathbf{R}_{s'})$ in powers of the small parameter $u/E$, we can transform Eq. (24) into the form

$$\alpha-\varkappa\mp\frac{e^{-\alpha R}}{R}=\frac{f^2}{8}\left[\frac{1}{\alpha^5}+\frac{R^2\cos^2\theta}{\alpha^3}\pm\frac{R^3\cos^2\theta e^{\alpha R}}{\alpha^2}\pm e^{-\alpha R}\left(\frac{1}{\alpha^5}+\frac{R}{\alpha^4}+\frac{R^2}{3\alpha^3}\right)\right]+\frac{if}{8\alpha^2}\exp\left(-\frac{2}{3}\frac{\alpha^3}{f}\right)\left[e^{-\alpha R\cos\theta}+e^{\alpha R\cos\theta}\pm 2\exp\left(-\frac{fR^2}{4\alpha}\right)\right]$$

$$(\alpha=\sqrt{-2E},\ E<0). \tag{26}$$

Here $\cos\theta = \mathbf{f}\cdot\mathbf{R}_2/fR_2$ and the signs ± pertain respectively to the symmetric (+) and antisymmetric (-) states of the system. Solving Eq. (26) by iterating with respect to the terms containing the field, we obtain

$$E^{(\pm)}(f,R)=\varepsilon_0^{(\pm)}(R)-{}^1/_2\beta^{(\pm)}(R)f^2-{}^1/_2 i\Gamma^{(\pm)}(R). \tag{27}$$

Here $\varepsilon_0^{(\pm)} = -[\alpha_0^{(\pm)}(R)]^2/2$ denote the terms of the ion $A_2^-$, $\alpha_0^{(\pm)}(R)$ are the zeros of the left-hand side of Eq. (26), and $\beta^{(\pm)}(R)$ is the polarizability of the molecular ion for arbitrary $\mathbf{R}$. For $\theta = 0$ and $\theta = \pi/2$ the general expression for the polarizability of the two-center system $A_2^-$,

$$\beta^{(\pm)}(R)=\frac{1}{4[1\pm e^{-\alpha_0 R}]}\left[\frac{1}{\alpha_0^4}+\frac{R^2\cos^2\theta}{\alpha_0^2}\pm\frac{R^3\cos^2\theta e^{\alpha_0 R}}{\alpha_0}\pm e^{-\alpha_0 R}\left(\frac{1}{\alpha_0^4}+\frac{R}{\alpha_0^3}+\frac{R^2}{3\alpha_0^2}\right)\right]. \tag{28}$$

coincides with the components of the polarizability tensor for the homoatomic ion $A_2^-$, $\beta_{zz}^{(\pm)}$, $\beta_{xx}^{(\pm)} = \beta_{yy}^{(\pm)}$, computed earlier (by a direct method) in[27]. The auto-ionization widths of the negative molecular ion in the corresponding states are equal to

$$\Gamma^{(\pm)}(R)=\frac{f}{4\alpha_0}\exp\left(-\frac{2}{3}\frac{\alpha_0^3}{f}\right)\frac{1}{1\pm e^{-\alpha_0 R}}\left[e^{-\alpha_0 R\cos\theta}+e^{\alpha_0 R\cos\theta}\pm 2\exp\left(-\frac{fR^2}{4\alpha_0}\right)\right]. \tag{29}$$

The dependence of the polarizability tensor on the interatomic distance has already been analyzed.[27] Let us consider the asymptotic expressions for the auto-ionization width.

For small $R$, when we go over to the one-center situation $\alpha R \ll 1$, $\Gamma^{(+)}(R)$ coincides with the well-known expression for the auto-ionization width of a negative atomic ion with a weakly-bound $S$-electron,[17] while $\Gamma^{(-)}(R)$ determines the rate of disintegration of an atomic ion with a weakly-bound $p$-electron.[19] We have:

$$\Gamma_{l=1,\,m=0}(f,\alpha)=\frac{fR}{8}\exp\left(-\frac{2}{3}\frac{\alpha^3}{f}\right),\quad \theta=0, \tag{30}$$

$$\Gamma_{l=1,\,m=\pm1}(f,\alpha)=\frac{f^2R}{32\alpha^3}\exp\left(-\frac{2}{3}\frac{\alpha^3}{f}\right),\quad \theta=\frac{\pi}{2}. \tag{31}$$

Thus, the auto-ionization probability for a $p$-electron turns out to be less than that for an $S$-electron by a factor ($\alpha R$) equal to the centrifugal-barrier penetration factor, while the orientation of the electron cloud along a direction perpendicular to the field reduces the decay probability by a factor equal to the quasi-classical field parameter $f/\alpha^3$. These conclusions illustrate the nature of the changes that should be made in the final expressions when we go over to the $l \neq 0$ and the general cases. cases.4)

Let us now consider the case of large values of the interatomic distance. For $\alpha R \gg 1$ the polarizability (28) and the decay probability (29) for a two-center system increase exponentially. This effect is directly connected with the strong exchange interaction of states localized at different centers, states between which an electron makes resonance transitions. The exponential growth of $\beta^{(\pm)}(R)$ and $\Gamma^{(\pm)}(R)$ occurs as long as the conditions for a weak interaction of the system with the field are fulfilled, i.e., as long as $R \leqslant \alpha^{-1}\ln(\alpha^3/f)$. For larger $R$ the interaction of the system with the field is not weak ($u \geqslant \omega$). This case requires a different method for solving Eq. (24).

Let us consider, for example, the heteroatomic ion $AB^-(\Delta \neq 0)$. It is not difficult to see that there occurs in the system of "field" terms $E_s^0(fR_s) = -\varkappa_s^2/2 - \mathbf{f}\cdot\mathbf{R}_s$ ($s = 1, 2$) pseudointersecting in the plane defined by the equation

$$\mathbf{f}(\mathbf{R}_2 - \mathbf{R}_1) = \Delta. \tag{32}$$

Solving Eq. (24) in the vicinity of the plane of pseudointersection, we find the terms of the system with allowance for all the interactions:

$$E_{1,2}(f, R) = {}^1\!/_2\{V_1(R_1) - V_2(R_2) \pm [(V_1(R_1) - V_2(R_2))^2 - 4G_f^2\varkappa_1\varkappa_2]^{1/2}\}. \tag{33}$$

Here the $V_s(R_s)$ are the terms of the zeroth (in the exchange interaction) approximation:

$$V_s(R_s) = E_s^0(f, R_s) - {}^1\!/_2\beta_s^0 f^2 - {}^1\!/_2 i\Gamma_s^0, \tag{34}$$

$$\beta_s^0 = \frac{1}{4\varkappa_s^4}, \quad \Gamma_s^0 = \frac{f}{4\varkappa_s}\exp\left(-\frac{2}{3}\frac{\varkappa_s^3}{f}\right). \tag{35}$$

The exchange interaction between disintegrating states (the term $4G_f^2(R)\varkappa_1\varkappa_2$ under the radical sign in (33)) not only separates the terms $V_s(R_s)$ in the region of pseudointersection, it also substantially changes the autoionization rate. For example, in the case of strong exchange interaction of slowly decaying states ($|\omega| \gg |V_1 - V_2|$)

$$E_{1,2} = \frac{E_1^0 - E_2^0}{2} - \frac{\bar{\beta} f^2}{2} - \frac{i\bar{\Gamma}}{2} \pm (\varkappa_1\varkappa_2)^{1/2} G_f(R). \tag{36}$$

Here $\bar{\beta} = (\beta_1^0 + \beta_2^0)/2$ is the mean polarizability and $\bar{\Gamma} = (\Gamma_1^0 + \Gamma_2^0)/2$ is the mean decay width of the noninteracting states. Separating the real and imaginary parts in the formula (36), we find the polarizability and the autoionization width with allowance for the exchange interaction:

$$\beta_{1,2}(R) = \frac{1}{8}\left[\frac{1}{\varkappa_1^4} + \frac{1}{\varkappa_2^4} \pm e^{-kR}(\varkappa_1\varkappa_2)^{1/2}\left(\frac{1}{k^5} + \frac{R}{k^4} + \frac{R^2}{3k^3}\right)\right]. \tag{37}$$

$$\Gamma_{1,2}(R) = \frac{\Gamma_1^0 + \Gamma_2^0}{2} \pm \frac{f(\varkappa_1\varkappa_2)^{1/2}}{4k^2}\exp\left(-\frac{2}{3}\frac{k^3}{f} - \frac{fR^2}{4k}\right) \tag{38}$$

($k^2 = \frac{1}{2}(\varkappa_1^2 + \varkappa_2^2)$). Thus, the exchange interaction in the case under consideration leads to the exponential growth of the rate of disintegration in a field of the state with the higher binding energy. We discuss some physical consequences of this result in the Conclusion.

## 4. DISTINCTIVE FEATURES OF THE INTERACTION OF QUASI-STATIONARY STATES WITH AN ELECTRIC FIELD

The question of the distinctive features of the interaction between an electric field and quasi-stationary states is of fundamental interest in connection with the problem of the dipole moment of unstable particles (see, for example, [22], Chap. XI). This question has been discussed in the framework of the nonrelativistic Lee model with a specially chosen hypothetical spectrum of the charges and masses by Zel'dovich and Perelomov, [28,29] who take the interaction of the system with the field into account with the aid of perturbation theory. Such an approach is, however, inapplicable in the general case, which admits of disintegrations into charged particles whose interaction with the field should be allowed for exactly.

The decay of a potential (centrifugal) $p$-resonance in an external electric field is exactly described by the two-center equation (15) of the antisymmetric state for $R < 1/\varkappa_1 (\varkappa_1 = \varkappa_2)$, in which the asymptotic representations of the Green function for positive energies (see the formula (21)) should be used. The case of the multichannel (Feshbach) resonance can, on the whole, be described in similar fashion, but it allows us to simultaneously consider the question of the competition between the elastic and inelastic auto-ionizations of an excited negative ion in an electric field.

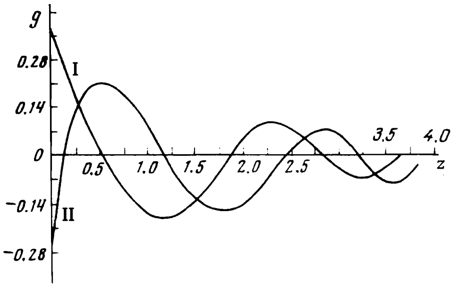
In the long-wave approximation the characteristic equation determining the energy levels of a two-channel system in a field has the form[7,10,30]

$$[\varkappa_1 - \alpha_1 - 2\pi G_f^r(\alpha_1)][\varkappa_2 - \alpha_2 - 2\pi G_f^r(\alpha_2)] = 4\pi^2\gamma^2 G_f^r(\alpha_1) G_f^r(\alpha_2)$$
$$(\alpha_1 = \sqrt{-2\varepsilon_1}, \quad \alpha_2 = \sqrt{-2\varepsilon_2}, \quad \varepsilon_1 = \nu + \varepsilon_2). \tag{39}$$

Here $\nu = \varepsilon_1 - \varepsilon_2$ is the excitation energy of the atomic core, the $\varkappa_{1(2)}^{-1}$ are the scattering lengths for electron scattering by the excited ($\varkappa_2$) and unexcited ($\varkappa_1$) core respectively, and $\gamma$ is the channel-coupling parameter, which is proportional to the amplitude of inelastic electron scattering by the atomic core. [30,31] It should be noted that the limitations of the long-wave approximation are of little significance in the problems under consideration. For $f \ll 10^9$ V/cm the external field only slightly distorts the electron wave function in the region of action of the potential of the core, i.e., the interactions of the electron with the core and the field virtually do not overlap. A system of exact algebraic equations describing the motion of an electron in the resultant field of nonoverlapping interactions (some of which may be long-range interactions) is derived in[10,11]. The use of the multichannel variant of these equations in the case under consideration (momentum-conserving $S$-electron transitions) leads to results differing from those obtained below only by the overdetermination of the phenomenological constants.

For $\varepsilon_1 > 0$ and $\varepsilon_2 < 0$, the solution to Eq. (39) determines the complex shift $\delta E^f$ of the resonance level in an external electric field:

$$\delta E^f = 2\pi\varkappa_2 G_f^r(\alpha_2) + \frac{\gamma^2\varkappa_2}{\varkappa_1 + ik - 2\pi G_f^r(k)} \tag{40}$$



Plots of the functions: I) $g = 2\pi(2f)^{-1/3}\,\mathrm{Re}\,G_f^r(x)$, II) $g(x) = 2\pi(2f)^{-1/3}\,\mathrm{Im}\,G_f^r(x)$; $x = -2E/(2f)^{2/3}$.

($k=\sqrt{2|\varepsilon_1|}$). Here the first term describes the Stark effect and the "elastic" field ionization of the resonance state of the negative ion and the second term determines the resonance-level shift induced by the interaction in the final state. With allowance for the previously computed values of the functions Re $G_f^r(E)$ and Im $G_f^r(E)$ (see the figure), it is easy to establish the occurrence in a strong field with $f \sim \varepsilon_1^{3/2}$ (the condition $f \ll \varepsilon_2^{3/2}$ may also be fulfilled in this case) of a substantial (up to 20–30% when $f \sim 4\varepsilon_1^{3/2}$) decrease in the resonance-decay rate. The effect of the stabilization of the low-energy resonances by the electric field could, apparently, be observed in a different sort of ionization experiments: in the resonance photodisintegration of negative ions, in Penning processes, etc.

In a field that is weak with respect to the parameters of both channels ($f \ll \alpha_1^3$, $f \ll \alpha_2^3$) the complex resonance-level shift is equal to

$$\begin{gathered}\delta E^f = -\frac{f^2}{4\varkappa_2^4} - \frac{if}{4\varkappa_2}\exp\left(-\frac{2}{3}\frac{\varkappa_2^3}{f}\right) \\ -\gamma^2\varkappa_2\left\{\left[\varkappa_1 + \frac{f}{2k^2}\sin\left(\frac{2}{3}\frac{k^3}{f}\right)\right] - i\left[k - \frac{f}{2k^2}\cos\left(\frac{2}{3}\frac{k^3}{f}\right)\right]\right\} \\ \cdot\left\{\left[\varkappa_1 + \frac{f}{2k^2}\sin\left(\frac{2}{3}\frac{k^3}{f}\right)\right]^2 + \left[k - \frac{f}{2k^2}\cos\left(\frac{2}{3}\frac{k^3}{f}\right)\right]^2\right\}^{-1},\end{gathered} \tag{41}$$

i.e., as the field is decreased the resonance-energy shift and the decay rate undergo damped oscillations, the level shift assuming its minimum value under conditions of maximum field stabilization. This effect is the quantum-mechanical analog of the well-known phenomenon of "lightening" of optical devices, and can qualitatively very graphically be interpreted in terms of the interference of waves diverging from the region of localization of a quasi-stationary state and reflected from the field potential barrier. It should, however, be emphasized that the absolute magnitude of the effect depends on the result of the interference of three-dimensional spherically nonsymmetric waves, and can, therefore, be found only from the exact solution to the problem. Also of interest is the nontrivial nature of the transition to conditions of free resonance decay: As the field intensity is decreased the effective dipole moment does not decrease, but oscillates with increasing frequency and constant amplitude whose magnitude is, generally speaking, of the same order as the dipole moment due to the asymmetry of the electron cloud in the initial state.[22]

For $\varepsilon_1<0$ and $\varepsilon_2<0$, the solution to Eq. (39) determines the total width of the excited negative ion $(A^-)^*$ in the electric field:

$$\Gamma = \Gamma_e + \Gamma_i:$$

$$\Gamma_e = \frac{f}{4\alpha_2}\exp\left(-\frac{2}{3}\frac{\alpha_2^3}{f}\right), \tag{42}$$

$$\Gamma_i = \frac{\gamma^2\varkappa_2 f}{4\alpha_1}\exp\left(-\frac{2}{3}\frac{\alpha_1^3}{f}\right)\left\{\left[\varkappa_1 - \alpha_1 + \frac{f^2}{4\alpha_1^5}\right]^2 + \frac{f^2}{16\alpha_1^4}\exp\left(-\frac{4}{3}\frac{\alpha_1^3}{f}\right)\right\}^{-1}.$$

Here $\Gamma_e$ and $\Gamma_i$ are the "elastic" and "inelastic" auto-ionization widths corresponding to the formation of the atom $A$ either in the excited ($\Gamma_e$) or in the ground ($\Gamma_i$) state. The obtained formulas show that the commonly observed "elastic" auto-ionization of excited negative ions[32] predominates over the "inelastic" process only at sufficiently large values of the field intensity

$$f > f_k = |\alpha_2^3 - \alpha_1^3| / \ln(\alpha_1^4/\gamma^2\alpha_2^2).$$

In a weak electric field $f<f_k$ the decay accompanied by the formation of the unexcited atom is more probable.

## 5. CONCLUSION. PHYSICAL EXAMPLES

The possibility of the rearrangement by the field of the terms of multicenter systems is of considerable interest in many respects. With the presence of "field" points of pseudointersection can be related diverse physical effects, examples of which are given below.

### A. Field charge transfer

In the term system for negative molecular ions of the type $AB^-$ there is, as a rule, no pseudocrossing; therefore, at low collision rates the probability of the nonresonance charge transfer

$$A^- + B \rightarrow A + B^-$$

is exponentially small. However, in a field with $f \sim \alpha\Delta \ln^{-1}(\Delta^2 v/\alpha^5)$ ($v$ is the speed of the atoms) the probability of a nonadiabatic transition in the vicinity of the plane of pseudointersection rises sharply. An entirely analogous situation obtains in a system of neutral atoms. At low collision rates the nonresonance charge transfer process

$$A + B \rightarrow A^- + B^- \tag{43}$$

proceeds, owing, as is well known, to nonadiabatic transitions at the points of pseudointersection (PI) of the ionic term with the covalent terms.[12] In typical cases significant changes in the PI parameters can be expected even for $f \sim 10^5$–$10^6$ V/cm. As is easy to verify, in these cases the charge-transfer probability on the whole increases. The process (43) is the initial stage of the chemical transformations that occur during the interaction of halogen molecules with alkali-metal atoms.[38] The change in the PI parameters that occurs in an external field indicates an interesting possibility of a catalytic influence of an electric field on an elementary chemical-reaction event.

### B. Field ionization

Atomic interactions in an external electric field are also of considerable interest in connection with the theory of autoionic spectroscopy.[34] The interaction of an atom $A$ of the representative gas with a neutral atom $B$ adsorbed in the surface of an emitter results in a sharp increase in the field-ionization probability near the PI plane. This should lead to the appearance in the energy spectra of the $A^+$ ions of resonance peaks whose intensity is directly determined by the magnitude of the exchange interaction.

### C. The effects of resonance tunneling

The above-considered mechanism of resonance tunneling of an electron can also change appreciably the rate of field disintegration of the negative ion $A^-$ (or of the neutral atom $A$) in a medium of atoms $B$ of an alien gas ($\varepsilon_B < \varepsilon_A$, where $\varepsilon_{A(B)}$ is the electron affinity of the atom $A(B)$). Averaging the two-center auto-ionization width (38) over the positions of the $B$ atoms, we obtain

$$\langle\Gamma_A\rangle=\Gamma_A^0+\langle\Gamma_{AB}\rangle,$$

$$\Gamma_A^0=\frac{f}{4\varkappa_A}\exp\left(-\frac{2}{3}\frac{\varkappa_A^3}{f}\right),\quad \langle\Gamma_{AB}\rangle\approx\frac{2\pi n_B}{f}\exp\left(\frac{\varkappa_A\Delta}{f}\right)\Gamma_A^0. \qquad (44)$$

Here $n_B$ is the density of the alien gas that constitutes the medium in which the field disintegration of the $A^-$ ion (or $A$ atom) occurs. It is not difficult to see that for $n_B \sim f\exp(-\varkappa_A\Delta/f)$ the resonance-tunneling mechanism will predominate.

Let us note, finally, the possible effects of electron tunneling through a cooperative (two-center) impurity level in experiments with dielectric films.[35] If the opposite surfaces of a film are heavily doped with different impurities ($\Delta \neq 0$), then a resonance-tunneling current will be observed only when $f=\Delta/a$ ($a$ is the thickness of the film and $\Delta$ is the energy defect of the one-center impurity levels). It is important that the volt-ampere characteristic of a junction, doped in the way indicated above, have a pronounced resonance character only for a totally definite polarity of the applied voltage.

The detailed analysis of the above-noted processes and systems is of independent interest, and the corresponding results will be published separately.

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[1)] Such points move to infinity as the field is switched off.

[2)] The equations for the wave function are given in[7] (formulas (9) and (10)).

[3)] The next terms in the expansion determine the static hyperpolarizabilities of the system, which are not calculated here.

[4)] The polarizability of the multicenter states with allowance for the orbital momenta $l_s \neq 0$ has already been considered in[27].

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