Exchange interaction radically changes the behavior of a quantum particle in a classically forbidden region: A simple model

V. V. Flambaum
School of Physics, The University of New South Wales, Sydney, New South Wales 2052, Australia
(Received 10 September 2009; published 10 November 2009)

Exchange interaction strongly influences the long-range behavior of localized electron orbitals and quantum tunneling amplitudes. In the Hartree-Fock approximation the exchange produces a power-law decay instead of the usual exponential decrease at large distances. To show that this effect is real (i.e., not a result of the approximation) we consider a simple model where different effects may be accurately analyzed. Applications include huge enhancement of inner electron ionization by a static electric field or laser field considered by Amusia (e-print arxiv:0904.4395).

DOI: 10.1103/PhysRevA.80.055401 PACS number(s): 32.80.Rm, 03.65.Xp, 31.15.xr, 71.70.Gm

One of the first famous results of quantum mechanics was that a particle may tunnel through a potential barrier. The tunneling amplitude is exponentially small in the classical limit. This result may be incorrect if we take into account the exchange interaction and correlation effects. In the Hartree-Fock equations the exchange interaction is described by the nonlocal (integration) operator, and the well-known theorems proven for the Schrödinger equation with a local potential $U(\mathbf{r})$ are violated if we add the exchange term (or any other nonlocal operator). According to Flambaum [1] (see also [2–4]) the exchange can produce a power-law decay instead of the usual exponential decrease at large distances. For inner orbitals inside molecules decay is r^{-2} for macroscopic systems $\cos(k_F r)r^{-\nu}$, where k_F is the Fermi momentum and $\nu=3$ for one-dimensional, $\nu=3.5$ for two-dimensional, and $\nu=4$ for three-dimensional crystals. Slow decay increases the spin-spin interaction between localized spins in solids and the underbarrier tunneling amplitudes.

A very interesting manifestation of this phenomenon has been suggested by Amusia in Ref. [5]. He showed that the exchange interaction may increase probability of ionization of inner atomic electrons by an external electric field by many orders of magnitude (in one of the examples the enhancement factor was 10^{39}). Amusia claimed that this enhancement may explain experimentally observed enhancement of multielectron ionization by a strong laser field (see, e.g., Ref. [6]) (a different explanation, an "atomic antenna" mechanism, was suggested by Kuchiev [7] and rediscovered by Corkum [8]).

All theoretical results [1–5] mentioned above have been obtained in the Hartree-Fock approximation. Naturally, one may ask a question: is this enhancement real or is it just an artifact of an approximate solution? Indeed, the Hartree-Fock method ignores correlation effects which sometimes play a very important role. An estimate of the correlation effects has been done in Ref. [1]. The conclusion is that if the correlation corrections may be treated using perturbation theory, their long-range effect is less significant than that of the exchange. However, it is important to consider a simple model where different effects can be accurately analyzed and check if the enhancement of the quantum tunneling by the exchange interaction really takes place.

Let us consider a model of resonance tunneling from one potential well to another potential well. The case of symmetric double-well potential has been solved, e.g., in the textbook [9]. There are two levels corresponding to the symmetric (ground state) and antisymmetric wave functions. The tunneling produces the splitting of these levels, $E_{\pm}=E_1\mp t_1$, where $t_1\sim\exp(-\int|p|dr/\hbar)$ is the tunneling amplitude, $|p|=\sqrt{2m[U(r)-E_1]}$ is the semiclassical underbarrier momentum, and the integral is taken between the classical turning points.

If the first potential well (a) is slightly deeper than the second potential well (b), the ground state wave function may be presented as $\psi_g=\psi_{1a}+B_{t1}\psi_{1b}$, where $B_{t1}\sim t_1/(E_{1a}-E_{1b})$. Here we assume that the distance to other levels is large, $t_1\ll(E_{1a}-E_{1b})$, and the probability of the particle in the ground state to be in the well b is exponentially small [proportional to the squared tunneling amplitude, $B_{t1}^2\sim t_1^2/(E_{1a}-E_{1b})^2$].

Now we add a second particle (identical fermion or boson) to a higher state 2 which has energy close to the top of the barrier. We can present its wave function as $\psi_2=A_2\psi_{2a}+B_2\psi_{2b}$, where the coefficient B_2 is not necessarily small. In this case the probability of the particle in the ground orbital to be in the potential well b is no longer proportional to the exponentially small parameter t_1^2. Indeed, the following two-step process takes place.

Step 1. The second particle tunnels from the potential well a (orbital ψ_{2a}) to the potential well b (orbital ψ_{2b}).

Step 2. Two-body process $2b$, $1a\to 1b$, $2a$ due to a nondiagonal Coulomb exchange interaction which transfers the first particle from orbital $1a$ to the orbital $2a$ and the second particle from $2b$ to $1b$.

As a result of these two steps, we have no change in the occupation of state 2 and transfer of a particle from the ground state $1a$ to $1b$. This gives the amplitude for the ground state particle to be in the well b,

$$B_{G1}\sim\frac{G(2,1a;1b,2)}{E_{1a}-E_{1b}}, \tag{1}$$

where

$$G(2,1a;1b,2)=\int \psi_2(\mathbf{r})^\dagger \psi_{1a}(\mathbf{r})\frac{e^2}{|\mathbf{r}-\mathbf{r}'|}\psi_{1b}(\mathbf{r}')^\dagger \psi_2(\mathbf{r}')d\mathbf{r}'d\mathbf{r} \tag{2}$$

is the Coulomb exchange integral. Note that the potential wells here may have one, two, or three dimensions.

This result may also be derived from the Hartree-Fock equation for the orbital $\psi_1=\psi_{1a}+\delta\psi_1$,

$$-\frac{\hbar^2}{2m}\frac{d^2}{d\mathbf{r}^2}\psi_1(\mathbf{r})+[U(\mathbf{r})-E_1]\psi_1(\mathbf{r})=K(\mathbf{r}), \tag{3}$$

by projecting it to the orbital ψ_{1b}. Here

$$K(\mathbf{r})=\psi_2(\mathbf{r})\int \psi_2(\mathbf{r}')^\dagger \frac{e^2}{|\mathbf{r}-\mathbf{r}'|}\psi_1(\mathbf{r}')d\mathbf{r}' \tag{4}$$

is the exchange term. Note that the contribution of the direct term in the Coulomb interaction between particles 1 and 2 is included into the mean field potential $U(\mathbf{r})$.

Equation (2) gives us dependence of the amplitude B_{G1} on the distance $|a-b|$ between the wells a and b. If the distance $|a-b|\gg r_1$ where r_1 is the size of the orbital $1a$, we can expand $1/|\mathbf{r}-\mathbf{r}'|$ near $|\mathbf{r}-\mathbf{r}'|=|a-b|$. Integral with the first term $1/|a-b|$ of this expansion vanishes due to the orthogonality of the wave functions $\psi_{1a}(\mathbf{r})$ and $\psi_2(\mathbf{r})$. Therefore, the expansion starts from $1/|a-b|^2$.

Now we may discuss a contribution of the correlation effects. They correspond to higher orders in the perturbation theory in the Coulomb interaction integrals G, so they decay with distance faster than $1/|a-b|^2$.

Similarly, the enhancement of the tunneling takes place for the ionization of an inner atomic electron by an external electric field. We just need to make the length of the potential well b infinitely large, so the orbitals $1b$ and $2b$ will be in the continuum.

It is instructive to compare the exchange enhancement mechanism with the atomic antenna mechanism [7,8]. In the antenna mechanism an external electron is ionized by a strong laser field, oscillates in this field, and accumulates energy. Then this electron collides with the parent ion and ionizes it. In the exchange mechanism the external electron plays a passive role; it does not change its initial state. This may give an additional coherent enhancement if the number of electrons in an external subshell is large. Indeed, in many-electron atoms the exchange term in the Hartree-Fock equation [Eq. (3)] contains sum over all electron orbitals,

$$K(\mathbf{r})=\sum_q \psi_q(\mathbf{r})\int \psi_q(\mathbf{r}')^\dagger \frac{e^2}{|\mathbf{r}-\mathbf{r}'|}\psi_{1a}(\mathbf{r}')d\mathbf{r}'. \tag{5}$$

Therefore, all external electrons contribute coherently into the effective tunneling amplitude for an inner electron. Amusia [5] claimed that this may give an additional enhancement factor N_{ext}^2 in the probability of the ionization where N_{ext} is the number of external electrons (this dependence N_{ext}^2 is probably observed in the ionization of noble gas clusters in Ref. [10]; see discussion in Ref. [5]). Note, however, that different subshells may contribute to this sum in Eq. (5) with different signs, so the interference is not completely constructive. Consider, for example, the ionization of $1s$ electron, $\psi_{1a}=\psi_{1s}$. The sign of the integrals in Eq. (5) is determined by the sign of $\psi_q(\mathbf{r})$ near the origin where ψ_{1s} is located. The large distance behavior of the corresponding term in $K(\mathbf{r})$ is determined by the $\psi_q(\mathbf{r})$ which stays outside the integral. Therefore, the sign of the contribution of a subshell depends on the number of radial oscillations of the wave function $\psi_q(\mathbf{r})$ which determines the sign of the product ψ_q (near zero) ψ_q (outside the atom). One should take into account this fact when estimating the coherence enhancement factor N^2.

In the discussion above we assumed that the residual Coulomb interaction (beyond the mean field) between the electrons is sufficiently small to be treated perturbatively. We should check if the exchange enhancement survives in the case of a stronger Coulomb interaction. Consider the two-well problem with a very large Coulomb repulsion between two particles. A minimum of the Coulomb energy is achieved when the particles are in different wells in state $\psi_{1a}\psi_{2b}$ or $\psi_{1b}\psi_{2a}$. Mixing between these states may be produced by the nondiagonal exchange interaction $G(2b,1a;1b,2a)$, i.e., it does not require any tunneling at all. Two other states $\psi_{1a}\psi_{2a}$ and $\psi_{1b}\psi_{2b}$ are separated from the lower states by the large Coulomb energy $Q=Q_{aa}-Q_{ab}$, where Q_{aa} and Q_{ab} are the Coulomb energies for the particles in the same well and different wells, respectively. Mixing between the states $\psi_{1a}\psi_{2a}$ and $\psi_{1b}\psi_{2b}$ may be achieved in three steps.

Step 1. Tunneling of particle from $2a$ to $2b$ with creation of an intermediate state $\psi_{1a}\psi_{2b}$ separated by the energy interval Q.

Step 2. The nondiagonal exchange interaction $G(2b,1a;1b,2a)$ transfers $\psi_{1a}\psi_{2b}$ to $\psi_{1b}\psi_{2a}$. At this step we have mixing of the single-particle states $1a$ and $1b$, $B_{G1}\sim t_2 G(2b,1a;1b,2a)/Q^2$.

Step 3. The tunneling from $2a$ to $2b$.

We see again that we do not need the exponentially small tunneling amplitude t_1, i.e., the exchange enhancement works. The only suppression we have here is due to the large Coulomb matrix element Q in the denominator of the mixing amplitude, $t_{eff}\sim t_2^2 G(2b,1a;1b,2a)/Q^2$. (Note that a similar suppression due to a large value of Q transforms a half-filled conducting band in solids into the Mott insulator. This transition influences the exchange power tail for a localized electron in solids; see discussion in [1].)

Similar results may be obtained for an attraction between the particles. This may be a model for a tunneling of an inner electron through a Josephson junction.

Thus, the exponential enhancement of the tunneling due to the exchange interaction really exists.

This work was supported by the Australian Research Council. I am grateful to J. Berengut for useful comments.

[1] V. V. Flambaum, Phys. Rev. A **79**, 042505 (2009).
[2] V. A. Dzuba, V. V. Flambaum, and P. G. Silvestrov, J. Phys. B **15**, L575 (1982).
[3] C. Froese Fischer, *The Hartree-Fock Method for Atoms* (Wiley, New York, 1977).
[4] N. C. Handy, M. T. Marron, and H. J. Silverstone, Phys. Rev. **180**, 45 (1969); G. S. Handler and D. W. Smith, J. Chem. Phys. **73**, 3936 (1980).
[5] M. Amusia, e-print arXiv:0904.4395.
[6] L. A. Lompre, A. L'Huillier, G. Mainfray, and L. Y. Fan, J. Phys. B **17**, L817 (1984).
[7] M. Yu. Kuchiev, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 319 (1987); [JETP Lett. **45**, 404 (1987)].
[8] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[9] L. D. Landau and E. M. Lifshits, *Quantum Mechanics* (Nauka, Moscow, 1974), \$50, problem 3.
[10] A. B. Borisov, J. Davis, X. Song, Y. Koshman, Y. Dai, K. Boyer, and C. K. Rhodes, J. Phys. B **36**, L285 (2003).