Manipulation of the RKKY exchange by voltages

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(Received 22 April 2019; revised manuscript received 20 June 2019; published 2 July 2019)

In the last years, electric fields have been used to control the magnetic exchange interactions and anisotropies in nanometric devices. In this paper, we study the spin-spin exchange interaction between two magnetic impurities embedded in a three-dimensional nonrelativistic electron gas, namely the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. The gas is confined in an insulating structure, and an applied voltage produces local changes in the electron density, which modulates the Fermi level of the system. Using a simple model, we demonstrate that this voltage modifies the strength and wavelength of the coupling between the impurities. Depending on the voltage, the effective RKKY exchange can change from a ferro- to an antiferromagnetic coupling, and vice versa. The spin-spin coupling can also be switched on and off by the voltage.

DOI: 10.1103/PhysRevB.100.014403

I. INTRODUCTION

The spin-spin exchange interaction is one of the most important couplings in condensed-matter physics. Exchange is responsible for the magnetic order; thus, its manipulation promises several applications in magnetic memory technologies. Conduction electrons mediate one of those interactions, namely the RKKY exchange, after Ruderman and Kittel [1], Kasuya [2], and Yosida [3]. RKKY interaction can be understood as follows. A local magnetic moment in a metal spin polarizes its surroundings and such polarization couples with nearby magnetic moments. The exchange coupling between the spins is characterized by changes in its sign as the distance between the spins is varied. Thus, depending on the separation between magnetic atoms, the RKKY exchange coupling may stabilize either a ferromagnetic or an antiferromagnetic order. The RKKY coupling is usually dominant at subnanometer distances because its magnitude decays with the separation distance r as $1/r^3$.

Besides the study of the exchange interaction in magnetic media, a topic of much interest is its manipulation. For example, the voltage-controlled coupling between magnets in heterostructures has been proposed and measured for several configurations and materials [4–8], including exchange-coupled layers separated by a nonmagnetic film [9–14]. Such systems exhibit a remarkable effect known as giant magnetoresistance (GMR), which is a relevant change of the electric resistance as a function of the relative orientation of the magnetization in each magnetic layer [15], the latter being controlled by the thickness of the spacer layer. The discovery of such a system in the late 1980s [16,17] opened the possibility of developing new devices, highlighting the relevance of the control of the exchange coupling, for example, by choosing the properties of the Fermi surface of the spacer [18–20]. Other realizations of voltage-controlled systems include nuclear spins [21], magnetic dimers between electrodes [22], two-dimensional materials [23–28], and ultrathin Co films [29–31]. Magnetization can be induced in platinum at metal|dielectric interfaces [32]. Other examples include exchange in antiferromagnetic Mott insulators [33], the interfacial Dzyaloshinskii-Moriya interaction [34], the *voltage-controlled magnetic anisotropy* effect [35–42], as well as phase transitions [43,44] and resonances [45] in magnetoelectric materials.

In this paper, we investigate the RKKY interaction between two spins in the presence of electric fields. Using a simple method, we observe that the field induces charge accumulation which shifts the Fermi wave number of the conduction electrons, and changes the RKKY sign and strength for a fixed separation distance between impurities. Our work provides a method to control the coupling between magnetic impurities in a three-dimensional electron gas.

II. ANALYTIC DESCRIPTION OF THE CONTROL OF THE RKKY EXCHANGE

Let us start reviewing the theory of the RKKY interaction in the strong screening limit [46]. The metal is modeled as a three-dimensional nonrelativistic ideal gas with spin density $\mathbf{s_c}(\mathbf{r}) = \psi^\dagger(\mathbf{r})(\hbar\boldsymbol{\sigma}/2)\psi(\mathbf{r})$, where $\hbar$ is Plank's constant divided by 2π, $\mathbf{r}$ is the position vector, $\boldsymbol{\sigma}$ is the vector of Pauli matrices, and $\psi(\mathbf{r})$ is the wave function. In the absence of magnetic fields, the ensemble average of the spin-density, $\langle\mathbf{s_c}\rangle$, is zero. This situation changes in the presence of a magnetic impurity at the origin $\mathbf{R} = 0$ with spin $\mathbf{S}$, due to the $s-d$ exchange interaction between $\mathbf{S}$ and the conduction electron spin density, $H_{s-d} = -2J_{\rm ex}\hbar^{-2}\int_{V_0}\mathbf{s_c}(\mathbf{r})\cdot\mathbf{S}\delta(\mathbf{r})$, where $\delta(\mathbf{r})$ is the Dirac delta, $J_{\rm ex}$ is the exchange coupling constant, and V_0 is the system volume. Within the strong and static screening approximation, $J_{\rm ex} = e^2 d_{\rm TF}^2\epsilon_0^{-1} = g_e^{-1}$, where $\epsilon_0 = 8.85\times10^{-12}$ F/m is the permittivity of the free space and $d_{\rm TF}$ is the Thomas-Fermi penetration length $d_{\rm TF} =$

$[\epsilon_0/(e^2 g_e)]^{1/2}$, which is of the order of a few Angstrom. The electron density of states for three-dimensional ideal gases is $g_e = m_e k_F/(\pi^2\hbar^2)$, the electron mass is m_e, and the wave number at the Fermi level is k_F. In this normalization, the spin densities have units of $\hbar/V_0$ and $J_{\rm ex} = g_e^{-1}$ has units of energy multiplied by volume. In the linear response regime, the ensemble-averaged spin density is

$$\langle \mathbf{s_c} \rangle(\mathbf{r}) = 2\chi(r)\mathbf{S}, \tag{1}$$

$$\chi(r) = \frac{1}{8\pi r^3}\left[\frac{\sin(2k_F r)}{2k_F r} - \cos(2k_F r)\right], \tag{2}$$

where $\chi(r)$ is the spin susceptibility [46] and $r = |\mathbf{r}|$. The divergence in the susceptibility for $r \to 0$ is due to the delta-function form of the localized spin density, $\mathbf{S}\delta(\mathbf{r})$.

Let us consider two spins, one $\mathbf{S}_1 = \sum S_{1,k}\mathbf{e_k}$ located at the origin, and the other $\mathbf{S}_2 = \sum S_{2,k}\mathbf{e_k}$ at $\mathbf{R}$. The Cartesian unit vectors for the $k = x, y, z$ axes are $\mathbf{e_k}$, $R = |\mathbf{R}|$, and the spins are independent, i.e., they commute $[S_{1,k}, S_{2,k'}] = 0$. Their interaction, as mediated by the conduction electron spin polarization, is described by the Hamiltonian $H_{\rm int} = -[4\chi(R)/(g_e\hbar^2)]\mathbf{S_1}\cdot\mathbf{S_2}$, where $\chi(R)$ exhibits changes in its sign as a function of the distance between the impurities. It is illustrative to use the basis $|\Psi\rangle = |S_1, S_2, S, S_z\rangle$ that satisfies $\mathbf{S_1}^2|\Psi\rangle = \hbar^2 S_1(S_1+1)|\Psi\rangle$, $\mathbf{S_2}^2|\Psi\rangle = \hbar^2 S_2(S_2+1)|\Psi\rangle$, $\mathbf{S}^2|\Psi\rangle = \hbar^2 S(S+1)|\Psi\rangle$, and $\mathbf{S_z}|\Psi\rangle = \hbar S_z|\Psi\rangle$ for $\mathbf{S} = \mathbf{S_1} + \mathbf{S_2}$, and $\hat{S}_z$ is the component of $\mathbf{S}$ along the quantization axis that we label z. Then, the Hamiltonian of the RKKY exchange is diagonal $H_{\rm int} = -2{g_e}^{-1}\chi(R)S(S+1) + H_0$ with the constant $H_0 = 2{g_e}^{-1}\chi(R)[S_1(S_1+1) + S_2(S_1+1)]$. Depending on the sign of $\chi(R)$, the energy is minimized by a symmetric (i.e., ferromagnetic-like) configuration [the total spin S is maximum for $\chi(R) > 0$] or antisymmetric [$S = 0$ for $\chi(R) < 0$]. The marginal case $\chi(R) = 0$ represents a system of uncoupled spins. In the next paragraphs, we study the control of the RKKY sign and strength via applied electric fields.

III. APPLICATION OF A VOLTAGE

Consider the insulating structure in Fig. 1(a). When a voltage is applied, there is no charge current but charge accumulation and deficit at the two interfaces. We model the metal as an electron gas subject to a voltage ϕ_0, such that the electric potential inside the gas $\phi(\mathbf{r})$ is smaller than the Fermi energy divided by e. In the zero-temperature limit, the Fermi-Dirac distribution becomes [47]

$$f(E_0 - e\phi) = \theta\left(-\left[\frac{\hbar^2\mathbf{k}^2}{2m_e} - e\phi - \frac{\hbar^2 k_F^2}{2m_e}\right]\right), \tag{3}$$

where θ is the step function with $\theta(x<0) = 0$ and $\theta(x>0) = 1$. When the potential ϕ is a slowly varying function, the last occupied level has the following kinetic energy [48]:

$$E_F(\phi) \equiv \frac{\hbar^2 \mathbf{q_F}^2}{2m_e} = \frac{\hbar^2 k_F^2}{2m_e} + e\phi. \tag{4}$$

Equations (3) and (4) show that without charge current, the kinetic energy must be smaller (larger) in the region where the electric potential is applied to compensate the increased

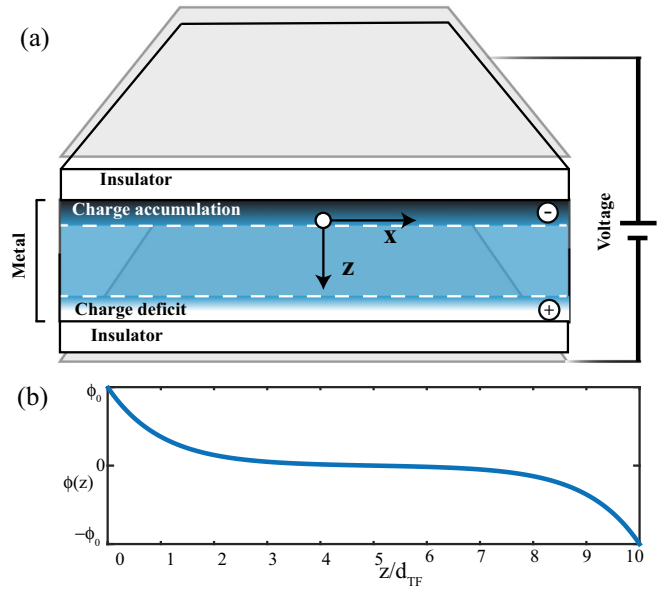

FIG. 1. Setup for the study of the voltage-induced control of the RKKY exchange. (a) Two insulators sandwich a metal and the stack is subject to a potential ϕ_0 along the z axis. As a result of the screening effect, there is an accumulation (deficit) of charge in the upper (lower) interface. The result is a space-dependent total potential inside the metal, as shown in (b). The accumulation/deficit of charge is approximately proportional [47] to the applied potential $\phi(z)$. Thus, a voltage shifts the Fermi level at the interfaces and, consequently, modifies the RKKY interaction.

(decreased) electrostatic energy $-e\phi$ and maintain the electrochemical potential constant [48]. If one writes the Fermi energy in terms of an electric potential-dependent wave vector $\mathbf{q_F}(\phi)$, one arrives at

$$q_F(\phi) \equiv |\mathbf{q_F}| = k_F\sqrt{1 + \frac{2m_e e\phi}{\hbar^2 k_F^2}} \approx k_F\left(1 + \frac{m_e e\phi}{\hbar^2 k_F^2}\right). \tag{5}$$

Then, the main effect of the voltage on the electronic system is a shift of the Fermi energy at the interface [49]. The problem of two magnetic impurities in a gas with a nonuniform Fermi energy is difficult in general. Indeed, two perturbations are acting on the electron gas, namely the voltage and the magnetic moments, and then the application of perturbation theory is not straightforward. A possible strategy in this regard is to consider that the Fermi wave number is smooth enough to be parameterized by the voltage. We follow this approach, which is based on the same assumptions of the Thomas-Fermi theory, and distinguish between the two following cases. The first one corresponds to two interacting particles that are at the same interface, i.e., at $z = 0$. The second case is of a particle at one interface ($z = 0$), and the other in the metal bulk ($0 < z < L$) or the opposite interface ($z = L$). The next subsections are devoted to each one of those situations.

A. Two particles at the same interface

We consider two spins, $\mathbf{S_1}$ and $\mathbf{S_2}$, at one of the insulator|metal interfaces. Both spins are subject to the same potential $\phi_0 = \phi(z = 0)$, and then the wave number q_F of

Eq. (5) can be used directly in the susceptibility χ, as well as in the density of states, to obtain the modified dynamic RKKY interaction. Note that since the $s-d$ exchange depends on the Coulomb interaction between localized and conduction spins, then the presence of the electrostatic potential ϕ modifies the number of states at the Fermi level as well as the screening length. This results in a modified exchange constant $J_{\rm ex} = g_e^{-1}$. The susceptibility χ becomes ϕ dependent, and at leading order reads

$$\tilde{\chi}(R;\phi) \approx \chi(R) - \phi_0 \frac{e m_e f_1(k_F R)}{16\pi\hbar^2 k_F^3 R^4}, \tag{6}$$

where $f_1(x) = \sin(2x) - 2x[\cos(2x) + 2x\sin(2x)]$. Note that $\tilde{\chi}$ is again an oscillatory function that decays with the radial distance. Also, the voltage-dependent part of the susceptibility scales as $1/R^2$ for large $k_F R$, while the voltage-independent one goes as $1/R^3$. The effect of this slowly varying voltage is to change the RKKY oscillation wavelength, while no phase shift appears at $R = 0$. This behavior is expected since the electric potential is assumed to be uniform inside the metallic region that contains the interacting impurities. The exchange coupling is also shifted, as given by

$$\frac{4\tilde{\chi}(R;\phi)}{g_e(q_F)\hbar^2} \approx \frac{4\chi(R)}{g_e(k_F)\hbar^2} - \phi_0 \frac{e\pi f_2(k_F R)}{2\hbar^2 k_F^4 R^4}, \tag{7}$$

where $f_2(x) = \sin(2x) - 2x[\cos(2x) + x\sin(2x)]$. Since the susceptibility diverges, it is convenient to plot $6\pi r\chi(r)$ as a function of $k_F x$. Note that $6\pi r\chi(r) \to 1$ when $r \to 0$ and $\phi \to 0$. Figure 2(a) shows the susceptibility for several voltages. We can see that a positive voltage, $\phi_0 > 0$, produces a stronger interaction and also faster spatial oscillations in the RKKY function. This behavior is because a positive electric potential diminishes the electrostatic energy [cf. Eq. (3)], and then the kinetic energy at the Fermi level, given by Eq. (4), is larger than in the $\phi_0 = 0$ case. On the other hand, a negative potential raises the electrostatic energy and decreases the Fermi wave number, which implies a slower spatial oscillation. Let us estimate the effect of the potential in two magnetic impurities separated a distance $R = 5$ Å, in a copper matrix, with Fermi energy [47] $E_F(0) = 7$ eV, $k_F = 1.24/$Å. We define the following phase shift: $\Delta\varphi \equiv 2R(q_F - k_F) = (k_F R)e\phi/E_F \sim \phi/(1V)$, which shows how much the potential shifts the oscillatory part of the susceptibility. Then, for an applied voltage of 0.1 V, the RKKY function is shifted in 0.1 radians. Figure 2(a) illustrates this phase shift, while Fig. 2(b) shows the type of coupling for several separation distances and voltages. In this figure, we observe that by using an external voltage, it is possible to tailor the type of order, ferromagnetic or antiferromagnetic, or switch the interaction off. The borders between the zones with a *ferro-* and an *antiferromagnetic* types of coupling are the zeros of the susceptibility function χ.

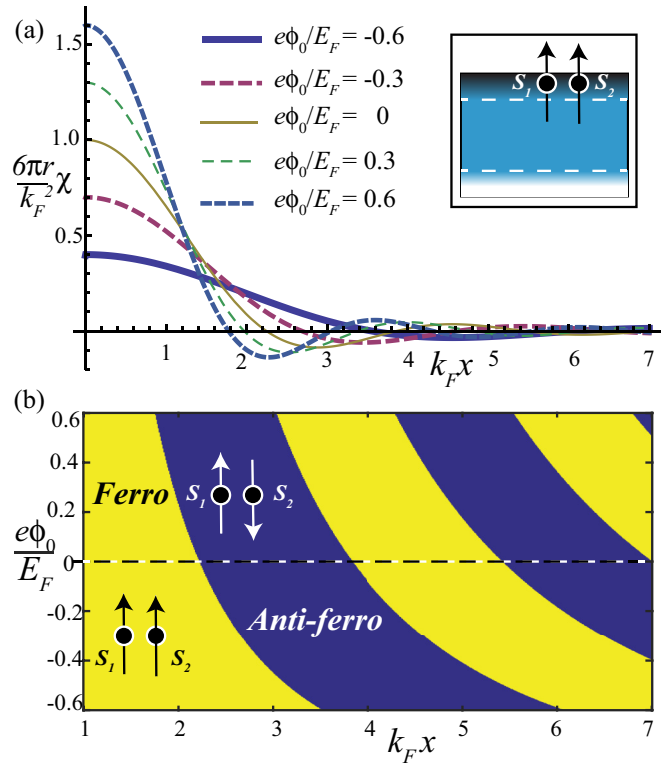

FIG. 2. Voltage-induced change in the electron susceptibility along the x axis. (a) The RKKY oscillations modify their wavelength as well as their magnitude. The inset shows the configuration of the interacting particles. Considering Cu with $E_F = 7$ eV, an applied voltage ϕ_0 produces the different curves shown in the upper panel. For example, the thick-dashed curve corresponds to $\phi_0 = -2.1$ V ($e\phi_0/E_F = -0.3$). (b) Type of coupling (*ferro-* or *antiferromagnetic*) as a function of the voltage and the distance between impurities. The zones with light (dark) color stand for a positive susceptibility $\chi > 0$ ($\chi < 0$).

B. Particles at different potential levels

In the Thomas-Fermi screening theory, the potential along the z axis is

$$\phi(z) = \phi_0[e^{-z/d_{\rm TF}} - e^{-(L-z)/d_{\rm TF}}], \tag{8}$$

where L is the metal thickness. This profile is shown in Fig. 1(b) for $\phi_0 > 0$. Consider one particle at the top interface, and the other along the z axis, as shown in the inset of Fig. 3(a). Since the potential is applied along the z axis, both impurities are at different potential levels. Let us introduce the following averaged wave number $Q_F(z)$:

$$Q_F(z) = \frac{1}{z}\int_0^z dz' q_F(\phi(z')), \tag{9}$$

which reduces to $Q_F \to k_F$ for $\phi \to 0$. It is worth noting that the above definition of Q_F is a generalization of the Wentzel-Kramers-Brillouin (WKB) approximation [50] for one-dimensional potentials as shown in the Appendix. Other choices of Q_F, such as the local approximation $Q_F = 1 + m_e e\phi_0/(\hbar^2 k_F^2)e^{-z/d_{\rm TF}}$ yield qualitatively the same results. Using the approximate formula Eq. (5) for q_F, and the potential of Eq. (8), one gets the following wave number:

$$\frac{Q_F(z)}{k_F} = 1 + \frac{e m_e \phi_0}{\hbar^2 k_F^2}\frac{d_{\rm TF}}{z}\left(e^{\frac{-z}{d_{\rm TF}}} - e^{\frac{-L}{d_{\rm TF}}}\right)\left(e^{\frac{z}{d_{\rm TF}}} - 1\right). \tag{10}$$

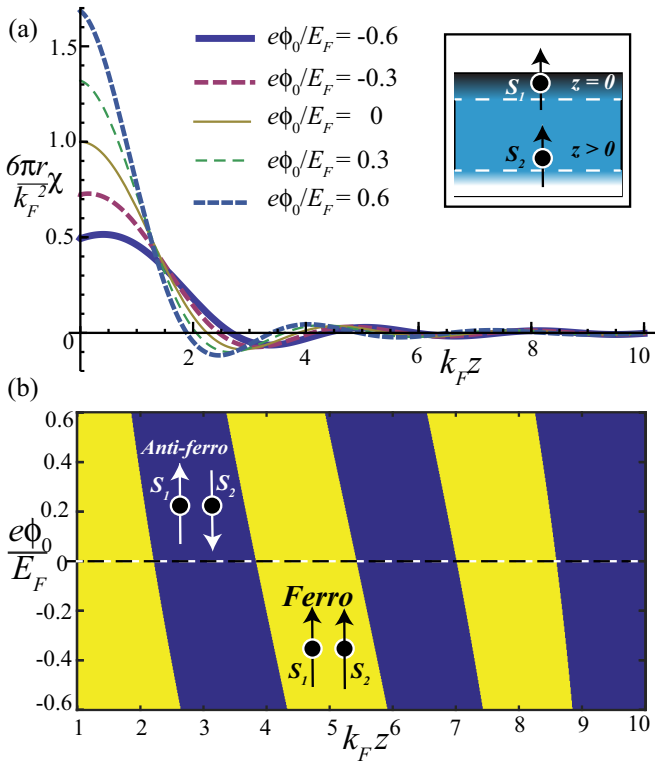

FIG. 3. Electric-potential-induced change in the electron susceptibility along the z axis with $x = y = 0$. (a) The RKKY oscillations modify their wavelength as well as their magnitude. The inset shows the configuration of the interacting particles. (b) Type of coupling (*ferro-* or *antiferromagnetic*) as a function of the voltage and the distance between impurities. The zones with light (dark) color stand for a positive susceptibility $\chi > 0$ ($\chi < 0$). We used $d_{\rm TF}k_F = 2$.

Figure 3 shows the susceptibility function along the z axis (with $x = y = 0$) for several applied potentials. Naturally, at $z = 0$,

$$Q_F \to k_F + \frac{em_e\phi}{\hbar^2 k_F}[1 - e^{-L/d_{\rm TF}}] \approx k_F + \frac{em_e\phi}{\hbar^2 k_F}, \qquad (11)$$

and we recover the analysis of the previous subsection.

The charge accumulation at one interface exactly cancels the charge deficit at the other interface. Consequently, the z-dependent phase shift of the RKKY oscillation sums zero when the interacting impurities are at opposing interfaces. As a result, $Q_F(L) = k_F$, and the susceptibility function at $z = L$ is the same regardless of the applied voltage. This scenario changes when we consider higher-order corrections in the small dimensionless quantity $\epsilon = e\phi_0/E_F$. In particular, the second-order Taylor expansion of q_F integrates the following $Q_F(L)$:

$$\frac{Q_F(L)}{k_F} = 1 + e^{-\frac{L}{d_{\rm TF}}}\left(\frac{\epsilon}{2}\right)^2\left(1 - \frac{d_{\rm TF}}{L}\sinh\left[\frac{L}{d_{\rm TF}}\right]\right) \neq 0,$$

where $\sinh(z)$ is the hyperbolic sine of z. Then, one could argue that the voltage-induced control of the RKKY coupling between magnetic impurities at different interfaces is a relatively small effect compared to one with impurities at the same interface [cf. Fig. 2(b)].

IV. CONCLUSIONS AND REMARKS

The control of microscopic interactions has attracted considerable attention during recent years. In particular, the modulation of the exchange coupling by electric fields is proposed as a candidate for the efficient manipulation of magnetic devices. While the interlayer coupling of magnets has been studied, the effect on magnetic impurities has not been fully clarified. Here we considered two impurities in an electron gas. The system under study consists of an electron gas perturbed by two magnetic impurities and a space-dependent potential. Thus, fully analytic treatment is beyond the linear response theory. Also, the numerical calculation of the susceptibility (correlation function) for the states that diagonalize the Hamiltonian with a Thomas-Fermi potential is not straightforward due to the large number of integration variables, divergences, and the separation of scale between rational and oscillatory functions. Thus, we have employed a simple approach valid for slowly varying potentials, in which the Fermi wave number is parametrized by the electric potential. In the presence of an electric potential, the Fermi level is shifted, which results in a voltage-dependent Fermi wave number. Since conduction electrons mediate the RKKY exchange, a shift in the Fermi level modifies the strength and wavelength of the interaction between magnetic impurities. This control can be used to change the interaction from ferromagnetic to antiferromagnetic or to decouple the spins.

Two cases were distinguished. First, for particles at the same insulator|metal interface, the application of the potential is equivalent to a uniform shift of the Fermi level due to the charge accumulation/deficit. Indeed, the application of the potential is analogous to changing the properties of the host metal. A potential that increases (decreases) the charge density at the interface produces faster (slower) RKKY oscillations because the conduction electrons have a larger (smaller) kinetic energy.

For impurities at different potential levels, we used a space-dependent wave number and found the sign of the susceptibility for several values of the applied voltage and positions. If one of the particles is at the interface, and the second is at the metal bulk, the control of the RKKY exchange interaction is stronger as compared to one of the impurities at opposing interfaces. In the latter case, the voltage-induced corrections on the RKKY function appear only at second order in the voltage.

ACKNOWLEDGMENTS

We acknowledge financial support in Chile from Postdoctorado FONDECYT 2019 Folio 3190030, FONDECY Grant No. 1160198, and Financiamiento Basal para Centros Científicos y Tecnológicos de Excelencia FB 0807. J.d'A.C. acknowledges financial support in Brazil from Project FAPERJ No. 150.038./2018. A.C. acknowledges financial support from Ministry of Research, Technology, and Higher Education of the Republic of Indonesia through PDUPT Research Grant No. 374/UN2.R3.1/HKP05.00/2018. This material is

based upon work supported by the Air Force Office of Scientific Research under Grant No. FA9550-18-1-0438.

APPENDIX: AVERAGED WAVE NUMBER

Let us consider a nonrelativistic three-dimensional electron gas described by the Schrödinger equation,

$$-\frac{\hbar^2\nabla^2}{2m_e}\psi + V(z)\psi = E\psi, \tag{A1}$$

where $V(z) = -e\phi(z)$ is the electrostatic potential that varies slowly along z. Let us use the following WKB-like ansatz:

$$\psi(\mathbf{r}) = A(z)e^{i\mathbf{Q}\cdot\mathbf{r}}, \tag{A2}$$

where the wave number $|\mathbf{Q}| = Q_F(z)$ and the amplitude $A(z)$ are slowly varying functions of z. Replacing the above Ansatz into Eq. (A1), one gets

$$(\nabla[\mathbf{Q}\cdot\mathbf{r}])^2 = \frac{2m_e}{\hbar^2}[E - V(z)]. \tag{A3}$$

In the *quasiunidimensional limit* ($\mathbf{Q} \to Q_F\mathbf{e_z}$, which is equivalent to classical particles moving mainly along the z axis), and at the Fermi level ($E = E_F$), the above equation reduces to

$$\frac{d(zQ_F)}{dz} = \sqrt{\frac{2m_e}{\hbar^2}[E_F - V(z)]}, \tag{A4}$$

and then

$$Q_F(z) = \frac{1}{z}\int_0^z dz'\sqrt{\frac{2m_e}{\hbar^2}[E_F - V(z')]}, \tag{A5}$$

which is the same Q_F of Eq. (9) for $V = -e\phi$.

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