Effect of antiferromagnetic RKKY interaction and magnetic field in a two-impurity Kondo system

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We explore the magnetic field dependence of the single-particle excitation spectra (SPES) of a magnetic dimer on a conducting surface using the numerical renormalization-group technique. When the antiferromagnetic Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction is dominant, a dip structure is observed near the Fermi level in the SPES. However, upon application of a certain external magnetic field, a peak structure appears near the Fermi level. Our results show that this peak structure originates in two reasons; (1) combination of the parity splitting and the Zeeman splitting, (2) enhancement of the formation of the Yosida-Kondo singlet in the ground state caused by the cancellation of the antiferromagnetic RKKY interaction.

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The Kondo effect has been investigated intensively since it was identified as a resistance anomaly in diluted magnetic alloys.[1] In particular, recent successes in observations of the Kondo effect through the characteristic peak structure (Kondo peak) in differential conductance (dI/dV) of semiconducting quantum dots (QDs) and magnetic atoms on nonmagnetic metal surface has accelerated the investigations of the Kondo effect in various environments.[2–5] What is more, in these systems, we can observe the localized spin-spin interaction mediated by the conduction electrons, Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction,[6] through the Kondo effect. Though the RKKY theory has long been accepted, there are still very few methods for direct observation and measurement.[7] Because the RKKY interaction decays much slower in two-dimensional systems than in three-dimensional systems, the two magnetic atoms placed on a conducting surface should provide great opportunities to study the RKKY interaction. From this point of view, we present in this paper the calculation results of the single-particle excitation spectra (SPES), which corresponds to the dI/dV in such system, under an external magnetic field. Finally, we propose that we can measure the antiferromagnetic (AF) RKKY interaction through the dI/dV observation by scanning tunneling microscopy (STM).

As the model Hamiltonian, we use the two-impurity Anderson Hamiltonian,

$$H = \sum_{i\sigma} E_d d_{i\sigma}^{\dagger} d_{i\sigma} + \sum_i g\mu_B \mathbf{B}\cdot S_i + \sum_{k,\sigma} E_k c_{k\sigma}^{\dagger} c_{k\sigma} + \sum_{ik\sigma} (V_{kdi} c_{k\sigma}^{\dagger} d_{i\sigma} + \text{H.c.}) + \sum_i U n_{i\uparrow} n_{i\downarrow}. \quad (1)$$

The definition of the operators such as c_k and coefficients such as U are the same in Refs. 8 and 9. $\mathbf{B}=(0,0,B_z)$ is the magnetic field. We set $U=300$ meV and $\pi\rho V^2=22$ meV so as to make the Kondo temperature in the case of a single magnetic atom (T_{K0}) be around 3 K. ρ is the density of states of conduction electrons. Here, we assume the direction of the magnetic field is perpendicular to the surface. g is the Lande g-factor and μ_B is the Bohr magnetron.

We assume that the dispersion of the surface conduction electrons is $E_k=\frac{\hbar^2 k^2}{2m^{*2}}-D$. Here D is the half bandwidth and m^* is the effective mass of the conduction electrons. As typical values, we set $D=1.0$ eV and $m^*/m_e=0.5$. Here, m_e is the mass of electron. [For example, on Cu(111) surface, the bottom of the surface state is at 0.46 eV below the Fermi level and $m^*/m_e=0.46$.[10]]

When we set the origin point on the center of the two localized spins, the corresponding model Hamiltonian should be invariant under parity transformation. Thus, it is convenient to rewrite the model Hamiltonian using the creation and annihilation operator for each orbital which is either invariant to parity (+) or is changed in sign by parity (−).[8,9,11–14] We rewrite Eq. (1) as

$$H = \sum_k E_k c_k^{\dagger} c_k + \sum_{p=\pm} E_p d_p^{\dagger} d_p + \sum_i g\mu_B \mathbf{B}\cdot S_i + \sum_i U n_{i\uparrow} n_{i\downarrow} + V\left(\sum_k \sqrt{2}\cos\frac{k\cdot R_{12}}{2} c_k^{\dagger} d_+ + i\sqrt{2}\sin\frac{k\cdot R_{12}}{2} c_k^{\dagger} d_- + \text{H.c.} \right). \quad (2)$$

The RKKY interaction is introduced by the k- and R_{12}-dependent hybridization, $V\sqrt{2}\cos\frac{k\cdot R_{12}}{2}$ and $Vi\sqrt{2}\sin\frac{k\cdot R_{12}}{2}$. With these settings, the AF RKKY interaction becomes maximum at $R_{12}=7.0$ Å.[8,9] Thus, in the rest of this Brief Report, we set $R_{12}=7.0$ Å. On the bases of this model Hamiltonian, we calculate SPES using the numerical renormalization-group (NRG) technique.[15–17] For the discretization of conduction band with this kind of dispersion, we use the method by Campo and Oliveila.[14] Though the NRG technique is very powerful tool for accurate calculation of equilibrium properties, there are fundamental problems associated with calculation of dynamical quantities. In particular, conventional methods fail for finite magnetic field. A recent improvement is to use the density matrix.[18–21] Among them, we adopt the complete-Fock space NRG method[20,21] in the SPES calculation. In Fig. 1 we show the total SPES calculation results under a finite magnetic field.

When $B_z=0$ T, a dip structure is observed near the Fermi level in total SPES. This dip structure indicates that the AF spin-ordering effect caused by the RKKY interaction is much more dominant than the Kondo effect.[22] As the magnetic field increases, the width of the dip structure becomes narrow

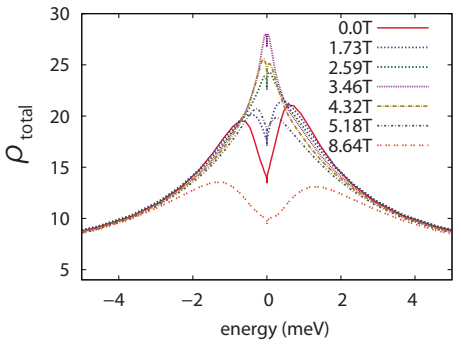

FIG. 1. (Color online) Calculation results of total single-electron excitation spectra (ρ_{total}) for several values of magnetic field.

and a peak structure appears near the Fermi level when B_z =3.46 T. Further increase in the magnetic field makes the peak split again. The magnetic dependency of total SPES is similar to that of a QD connected to ferromagnetic leads.[23–26] These studies reveal that the spin polarization in the leads results in the dip structure at B_z=0 T. And, compensation of the spin polarization by an external magnetic field or the gate voltage restores the Kondo peak. In our model, there is no spin polarization when B_z=0 T and we have to consider the other explanation. We focus on the effect of the parity symmetry in our model. In Fig. 2, we show the calculation results of spin- and parity-resolved SPES (ρ_{ps}).

p represents the parity and p=+ or −. s represents the spin direction and s=↑ or ↓. When B_z=0 T, the resolved SPES is the same for spin up and spin down $(\rho_{+\uparrow}+\rho_{-\uparrow})=(\rho_{+\downarrow}+\rho_{-\downarrow})$. However, the peak position differs by the parity. This results in the dip structure. Under a small magnetic field, this parity splitting is kept, but asymmetry with spin develops. The peak height of $\rho_{+\downarrow}$ and $\rho_{-\uparrow}$ ($\rho_{+\uparrow}$ and $\rho_{-\downarrow}$) becomes higher (lower). When the magnetic field becomes close to 3.46 T, the peaks in $\rho_{+\downarrow}$ and $\rho_{-\uparrow}$ are positioned near the Fermi level, and side peaks of $\rho_{+\uparrow}$ and $\rho_{-\downarrow}$ are merged. Further increase in the magnetic field, all of the peaks shifted away from the Fermi level. From these results, we conclude that the change in SPES under the external magnetic field originates in the following two causes. The first is the shift of the renormalized energy level of the localized state of the adatom by the combination of the parity splitting and Zeeman splitting. The low-temperature electron state of our model Hamiltonian exhibits local Fermi-liquid behavior. It means that the SPES near the Fermi level is reproduced by the quasiparticle spectra centered at the renormalized energy levels. As mentioned above, the renormalized energy levels of the adatoms are parity dependent. In our case, the state with +(−) parity lies above (below) the Fermi level. As B_z increase, the Zeeman energy increases the energy of up-spin state and decreases that of down-spin state. This makes the p=+, s=↓ level and p=−, s=↑ level come close to the Fermi level under a magnetic field of a proper magnitude. Further increase in the magnetic field makes all of the renormalized levels shifted far away from the Fermi level. We show a schematic illustration of this shift of the renormalized energy levels in Fig. 3.

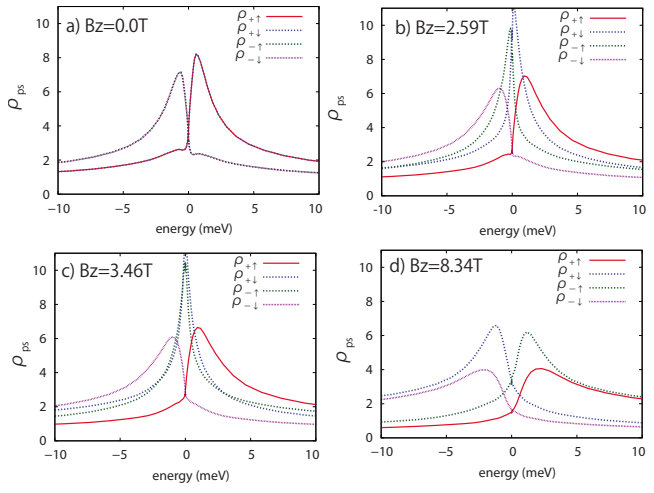

FIG. 2. (Color online) Calculation results of spin- and parity-resolved single excitation spectra (ρ_{ps}, p=+ or −, and s=↑ or ↓).

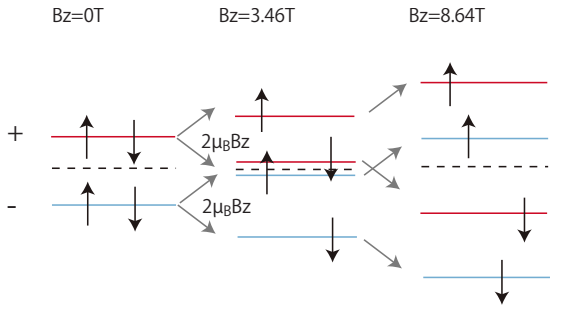

FIG. 3. (Color online) Schematic of the Zeeman splitting and parity splitting of the renormalized energy levels of adatoms. ± indicates the parity of the levels. ↑ and ↓ indicate the spin.

The second is the suppression of the formation of localized spin-spin singlet. In the case of our model, the ground state consists of combination of the localized spin-spin singlet and Yosida-Kondo singlet when the RKKY interaction is antiferromagnetic because of the e-h asymmetry. The external magnetic field of proper magnitude cancels the energy gain from formation of the localized spin-spin singlet. This increases the ratio of the Yosida-Kondo singlet state in the ground state, which results in the large value of the SPES at the Fermi level. Meanwhile, the excessive magnetic field aligns the localized spins, which suppresses the formation of the Yosida-Kondo singlet. We can verify the above discussions by calculating the phase shift. The phase shift can be estimated from energy flows of the low-lying many-particle energy (EFLE) shown in Fig. 4.

In the EFLE, eigenstates are labeled by the total charge Q, z component of the total spin S_z, and the total parity P. We estimate the parameters of the low-temperature fixed-point effective Hamiltonian from the EFLEs and calculate the phase shift. In Fig. 5, we show the results of the phase shift δ_{ps} calculation as a function of magnetic field. The definitions of p and s are the same for Fig. 2. The change in phase shift strongly depends on the parity and spin. $\delta_{+\downarrow}$ and $\delta_{-\uparrow}$ rapidly change as a function of the applied external magnetic field. On the other hand, $\delta_{+\uparrow}$ and $\delta_{-\downarrow}$ changes more slowly. If $\delta_{ps}=\pi/2$, it means that the ground state consist of the Yosida-Kondo singlet. On the contrary, δ_{ps}=0 indicates that

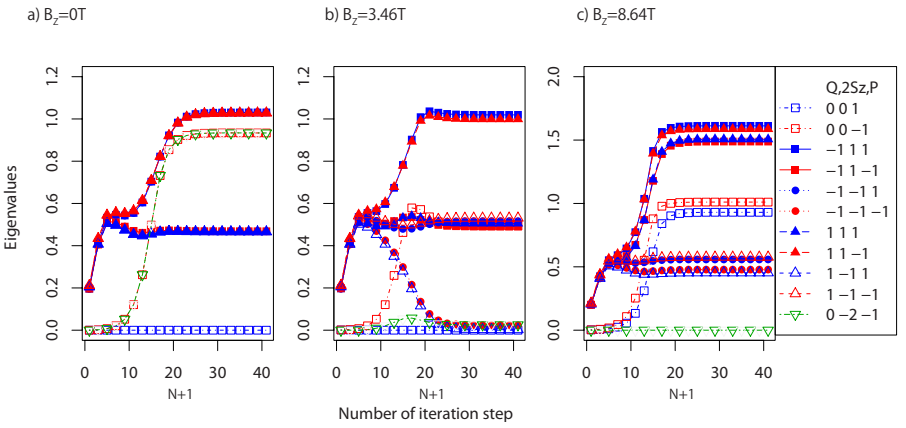

FIG. 4. (Color online) Energy flows of the low-lying many-particle energy levels with several values of B_z in odd iterations. $N+1$ corresponds to the iteration step number. In this calculation we calculate for magnetic field B_z, (a) B_z=0 T, (b)B_z=3.46 T, and (c) B_z=8.64 T. The states are labeled by the total charge Q, z component of the total spin S_z and the total parity P.

the ground state consist of the localized spin-spin singlet. The result that $\delta_{+\downarrow}$ and $\delta_{-\uparrow}$ become $\pi/2$ around B_z=3.5 T also supports the enhancement of the formation of the Yosida-Kondo singlet. Further increase in the B_z makes the phase shift far away from $\pi/2$. Considering the fact that the ground state changes from $(Q,S_z,P)=(0,0,1)$ to $(0,-2,1)$ state, the change in the phase shift means that the formation of the Yosida-Kondo singlet is suppressed by the spin aligning effect of the external magnetic field. We will now comment on how our results can be experimentally confirmed through currently available methods. One potential experiment which can be compared by our results is the dI/dV observation by STM for a magnetic dimer consisting of two same atoms placed on a metal surface covered by an insulating layer. Because the insulating layer suppresses the Fano effect, the dI/dV are almost proportional to the total SPES in such system. For example, Otte have investigated the dI/dV of a Co-Co dimer on $Cu_2N/Cu(100)$ surface under a magnetic field.[27,28] Figure 6 is the experimental results by Otte.[28] Experimentally estimated T_{K0} of a Co adatom on $Cu_2N/Cu(100)$ is also around 3 K.[29] And, Otte's results of Co-Co dimer and our calculation results have same tendency regarding the response to magnetic field, i.e., a dip the Fermi level at a 0 T magnetic field change to a peak at a 3 T magnetic field. The triple peak structure at the 3.0 T magnetic field in Fig. 6 would originate in the side peak caused by the parity splitting (Fig. 2).

As discussed in Refs. 27–31, $S_z=|1/2|$ state becomes most stable in a Co atom on $Cu_2N/Cu(100)$ surface because of a large positive longitudinal magnetic anisotropy caused by the Cu_2N layer, even though an isolate Co atom has $S=3/2$. In such a case, the mapping to spin-1/2 Kondo model is a good approximation and anisotropy is interpreted as the difference of g factor in direction of the magnetic field.[30,31] Thus, we believe our results can provide a qualitative explanation for the dI/dV results of the Co-Co dimer in low-temperature and low-energy region, in which the $S_z=|3/2|$ states are freeze out. Further investigations including the effect of anisotropy and using more realistic model are required. Another candidate is a Ti-Ti dimer on $Cu_2N/Cu(100)$ surface. The dI/dV results of a Ti atom on $Cu_2N/Cu(100)$ surface show a behavior typical of a magnetic atom with isotropic $S=1/2$.[29] Though there is no experimental results now, if dI/dV spectra of a Ti-Ti dimer on $Cu_2N/Cu(100)$ surface is measured under magnetic field in future, the results would be comparable to our results.

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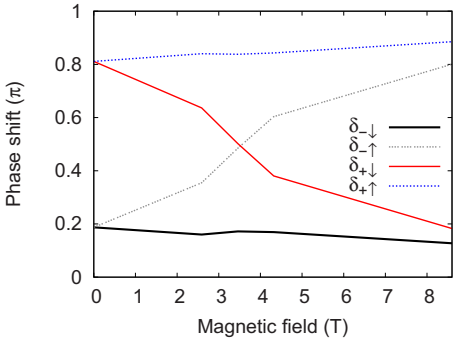

FIG. 5. (Color online) Phase shift as a function of adatom separation (in units of π radians).

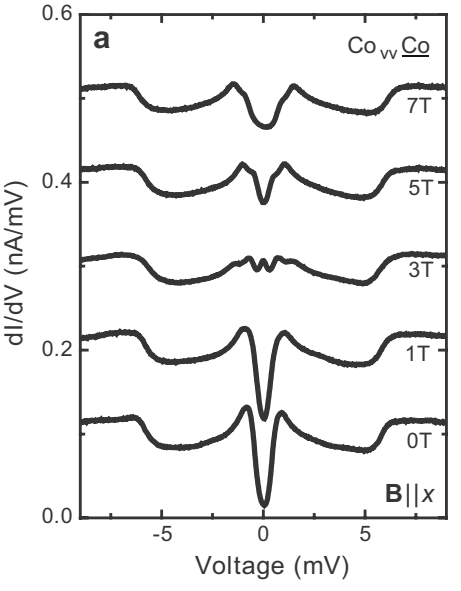

FIG. 6. Experimental dI/dV results of Co-Co dimer on $Cu_2N/Cu(100)$ surface by Otte. This figure is a reproduction of Fig. 4.14 in Ref. 28 with permission from Otte.

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