Spin-orbit-induced Kondo size effect in thin films with 5/2-spin impurities

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(Received 3 March 1999)

Recently, for spin $S=5/2$ impurities, quite different size dependence of the Kondo contribution to the resistivity was found experimentally than for $S=2$. Therefore, previous calculation about the effect of the spin-orbit-induced magnetic anisotropy on the Kondo amplitude of the resistivity is extended to the case of $S=5/2$ impurity spin, which differs from the integer spin case as the ground state is degenerated. In this case the Kondo contribution remains finite when the sample size goes to zero and the thickness dependence in the Kondo resistivity is much weaker for Cu(Mn). The behavior of the Kondo coefficient as a function of the thickness depends on the Kondo temperature, which is somewhat stronger for larger T_K. Comparing our results with a recent experiment in thin Cu(Mn) films, we find a good agreement. [S0163-1829(99)04239-3]

I. INTRODUCTION

The Kondo effect[1] in samples with reduced dimensions (thin films, narrow wires) is one of the most challenging problems in the field. Most of the experiments[2] have shown that the Kondo contribution to the resistivity is suppressed when the sample size is reduced or the disorder in the sample is increased. In addition, the different thermopowers of samples with different thickness gave further evidences for the existence of size dependence.[3] The previously examined samples were Au(Fe), Cu(Fe), and Cu(Cr) alloys, i.e., alloys with integer spin impurities. Surprisingly, however, very weak size dependence has been found recently in Cu(Mn) alloys.[4] The first possible explanation related to the size of the Kondo screening cloud[5] was ruled out both theoretically[6] and experimentally.[7] In the limit of strong disorder the experiments might be well explained with the theory of Phillips and co-workers based on weak localization.[8]

In the dilute limit the theory of the spin-orbit-induced magnetic anisotropy proposed by the authors[9] was able to explain every experiment in samples with reduced dimensions, small disorder, and integer spin impurities for thin layers.[10,11] Recently an elegant method was developed by Fomin and co-workers[12] which can be applied for a general geometry. According to this theory[9,12] the spin-orbit interaction of the conduction electrons on the non-magnetic host atoms can result in a magnetic anisotropy for the magnetic impurity. This anisotropy can be described by the Hamiltonian $H_a=K_d(\mathbf{nS})^2$ where $\mathbf{n}$ is the normal direction of the experienced surface element, $\mathbf{S}$ is the spin of the impurity, and K_a is the anisotropy constant, which is always positive and inversely proportional to the distance of the impurity from the surface. Due to this anisotropy the spin multiplet splits according to the value of S_z. In the case of integer spin (e.g., $S=2$ for Fe), the lowest level is a singlet with $S_z=0$; thus at a given temperature the impurities close enough to the surface, where the splitting is greater than k_BT, cannot contribute to the Kondo resistivity. When the sample size becomes smaller, more and more impurity spins freeze out, reducing the amplitude of the Kondo resistivity.[10]

The theory predicts, however, different behavior for samples with half-integer impurity spin (e.g., $S=5/2$ for Mn) which is recently in the center of interest. In the one-half case when the anisotropy loses its meaning,[10] in fact no anomalous size dependence has been found for Ce impurities by Roth and co-workers.[13] In half-integer $S>1/2$ case the lowest level is a doublet ($S_z=\pm 1/2$) thus even an impurity close to the surface (large anisotropy) has a contribution to the Kondo resistivity. Even accepting that the surface anisotropy reduces the free spin of the manganese to a doublet, it is far from trivial that in this case the size dependence is drastically suppressed; therefore an elaborate theory is required.

In this paper, we calculate the Kondo resistivity in thin films of magnetic alloys with $S=5/2$ impurity with the help of the simple model and multiplicative renormalization group (MRG) calculation of Ref. 10. Fitting to the Kondo resistivity the $\Delta\rho=-B\ln T$ function, we calculate the coefficient B in terms of the film thickness which is quite different from the $S=2$ case. These results will be compared to the recent experiment by Jacobs and Giordano[4] presented in thin Cu(Mn) films and we compare also the case of alloys with different Kondo temperatures.

II. KONDO SIZE EFFECT IN THIN FILMS WITH IMPURITIES $S=5/2$

In the presence of the spin-orbit-induced surface anisotropy the Kondo Hamiltonian is

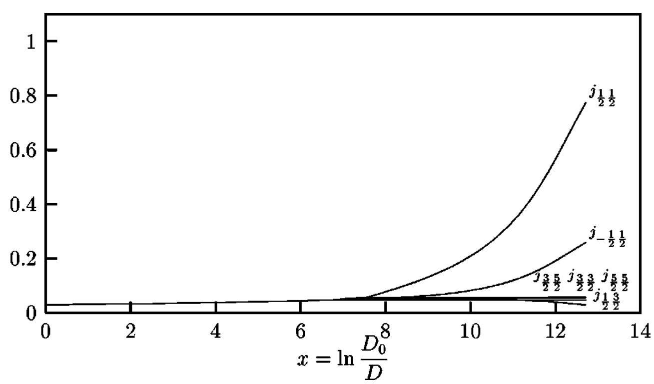

FIG. 1. The running couplings for $S=5/2$ as a function of $x=\ln D_0/D$ at $K=30$ K, $T=0.3$ K with parameters $D_0=10^5$ K and $j_0=0.0294(T_K=10^{-3}$ K).

$$H=\sum_{k,\sigma} \varepsilon_k a^\dagger_{k\sigma} a_{k\sigma} + H_a + \sum_{\substack{k,k',\sigma,\sigma' \\ M,M'}} J_{MM'} \boldsymbol{S}_{MM'} (a^\dagger_{k\sigma} \boldsymbol{\sigma}_{\sigma\sigma'} a_{k'\sigma'}), \qquad (1)$$

where $a^\dagger_{k\sigma}(a_{k\sigma})$ creates (annihilates) a conduction electron with momentum k, spin σ, and energy ε_k measured from the Fermi level. The conduction electron band is taken with constant energy density ρ_0 for one spin direction, with a sharp and symmetric bandwidth cutoff D. $\boldsymbol{\sigma}$ stands for the Pauli matrices, $J_{MM'}$'s are the effective Kondo couplings, and $H_a=KM^2$ is the anisotropy Hamiltonian when the quantization axis is parallel to **n**. Applying the Callan-Symantzik MRG method to the problem, the next to leading logarithmic scaling equations for the dimensionless couplings $j_{MM'}=\rho_0 J_{MM'}$ were calculated for any impurity spin in Ref. 10. There was a multiple step scaling performed, corresponding to the freezing out of different intermediate states due to the surface anisotropy. After exploiting the $j_{M,M'}=j_{M',M}=j_{-M,-M'}$ symmetries of the problem, the scaling equations [see Eqs. (20) and (21) in Ref. 10] were solved numerically in terms of the scaling parameter $x=\ln(D_0/D)$.

The results for $S=5/2$, $j_0=0.0294$, and $D_0=10^5$ K, i.e., $T_K=10^{-3}$ K for Cu(Mn), can be seen in Fig. 1. We can see from the plot that, when K/T is large enough, at $D=T$ every coupling can be neglected, except the $j_{\frac{1}{2}\frac{1}{2}}$ and $j_{-\frac{1}{2}\frac{1}{2}}$. This corresponds to the freezing out of the given higher S_z states, but it can be seen well that the two lowest energy states are still significant even for large anisotropy.

The Kondo resistivity calculated from the running couplings at $D=T$ by solving the Boltzmann equations (see Ref. 10) is

$$\rho_{\rm Kondo}(K,T)=\frac{3}{4}\frac{m}{e^2}\frac{2\pi}{\varepsilon_F \rho_0^2}\frac{c}{\int d\varepsilon\left(-\frac{\partial f_0}{\partial \varepsilon}\right)F^{-1}(\varepsilon)}, \qquad (2)$$

where c is the impurity concentration, ε_F is the Fermi energy, f_0 is the electron distribution function in the absence of an electric field, and F is a function of the running couplings at $D=T$, spin factors, the strength of the anisotropy K, and the temperature, defined in Ref. 10. For thin films the Kondo

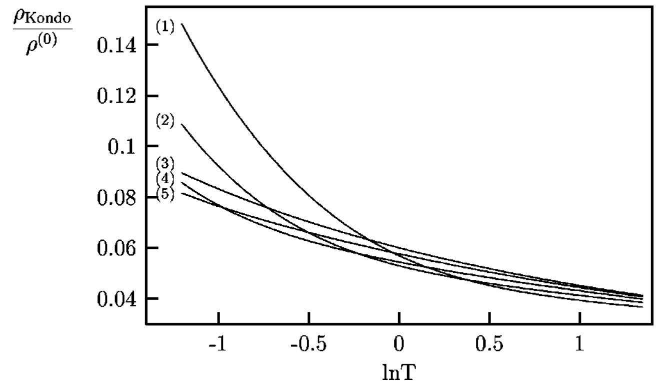

FIG. 2. The resistivity for $S=5/2$ for different values of t/α. (1) $t/\alpha=3/K$, (2) $t/\alpha=6/K$, (3) $t/\alpha=\infty(K=0)$, (4) $t/\alpha=13/K$, (5) $t/\alpha=40/K$. The initial parameters were chosen as $j_0=0.0294$ and $D_0=10^5$ K, $T_K=10^{-3}$ K, and $\rho^{(0)}=6\pi mc/4e^2\varepsilon_F\rho_0^2$.

resistivity calculated in the frame of a simple model where the two surfaces contribute to the anisotropy constant in an additive way as

$$K(d,t)=K_d+K_{t-d}=\frac{\alpha}{d}+\frac{\alpha}{t-d}, \qquad (3)$$

is

$$\overline{\rho}_{\rm Kondo}(t,T)\approx\frac{1}{t}\int_0^t \rho_{\rm Kondo}[K(x,t),T]dx, \qquad (4)$$

where α is the proportionality factor of the spin-orbit-induced surface anisotropy, t is the thickness of the film, and we have used the fact that the Kondo contribution to the resistivity is smaller by a factor of 10^{-3} than the residual normal impurity contribution (see for the details Ref. 10).

In Fig. 2 the resistivity as a function of $\ln T$ can be seen for different t/α for $S=5/2$, $j_0=0.0294$, and $D_0=10^5$ K, i.e., $T_K=10^{-3}$ K as for Cu(Mn). Thus reducing the thickness of the film for given α, the Kondo contribution to the resistivity is reduced comparing to the bulk value, but for given thickness there is a temperature below that the reduction turns into an increase. Fitting the logarithmic function $-B\ln T$ to the Kondo resistivity, the plots of the coefficient $B/B_{\rm bulk}$ as a function of the thickness can be seen in Fig. 3.

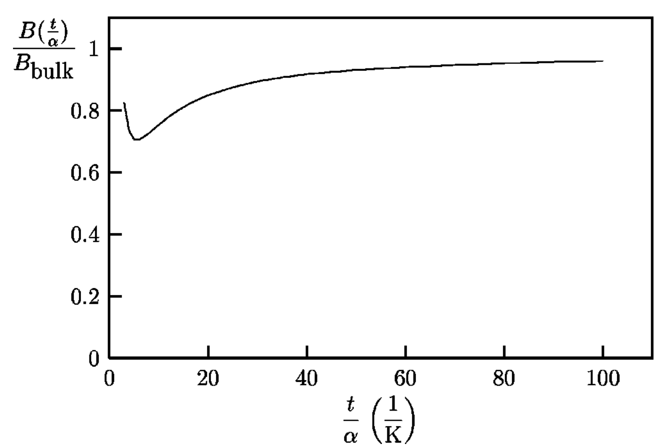

FIG. 3. The coefficient $B/B_{\rm bulk}$ as a function of t/α for $S=5/2$ and $T_K=10^{-3}$ K.

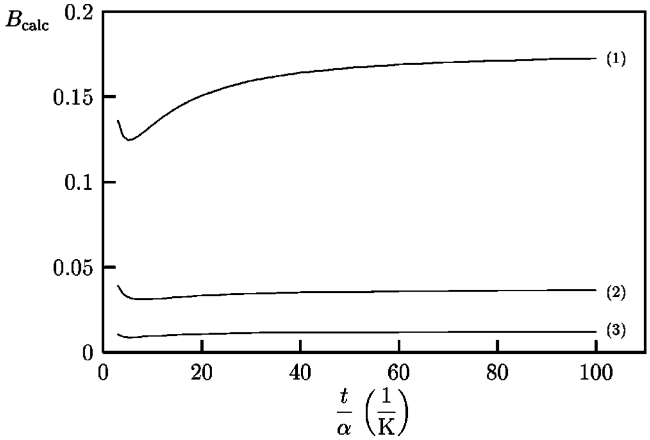

FIG. 4. The calculated coefficient $B_{calc}=(B/c)6\pi m/4e^2\varepsilon_F\rho_0^2$ as a function of t/α for $S=5/2$ for different Kondo temperatures. (1) $T_K=10^{-1}$ K, $B_{calc}^{bulk}=0.1793$; (2) $T_K=10^{-2}$ K, $B_{calc}^{bulk}=0.0377$; (3) $T_K=10^{-3}$ K, $B_{calc}^{bulk}=0.0127$.

Here we can better see this very different behavior from the $S=2$ case (cf. Ref. 10). First of all, the Kondo amplitude is reduced comparing to the bulk value, but the dependence on the thickness is much weaker than for $S=2$. Secondly, for small t/α's the coefficient does not go to zero as for $S=2$, but has a minima at $t/\alpha\sim 6/K$ and then changes sign and begins to increase. This corresponds to that for an $S=5/2$ impurity in the presence of the anisotropy, the lowest energy states are the $S_z=\pm\frac{1}{2}$ doublet which give a contribution to the resistivity even for large anisotropy (small distance from the surface) and which can also be larger than the bulk value as a consequence of the spin factors in the scaling equations. Because of the large domain of the microscopic estimation of the anisotropy constant ($\alpha\sim 100$ Å K-10^4 Å K), we cannot give a precise microscopic prediction for the place of the minima. According to the fits on the experimental results on Au(Fe)[10,11] and Cu(Fe),[14] $\alpha\sim 40-250$ Å K from which we can obtain a rough estimation for the place of the minima as $t_{min}\sim 240-1500$ Å. However, the theoretical calculation is not reliable in that region where $j_{\frac{1}{2}\frac{1}{2}}$ is already in the strong coupling limit. Thus, the minima may be a sign of the breakdown of the weak coupling calculation.

We have fitted in the $T=1.4-3.9$ K temperature regime, thus below 4 K where the electron-phonon interaction can still be neglected[4] and well above the Kondo temperature where the weak-coupling approximation is justified, thus our perturbative calculation and the logarithmic function for the Kondo resistivity is valid. These results are in good agreement with the recent experiments of Jacobs and Giordano[4] on thin films of Cu(Mn).

In Fig. 4 we have examined the function B/c for different Kondo temperatures corresponding to different 5/2-spin alloys. It can be seen from the figure that a minima in the $(B/c)(t/\alpha)$ function for small t/α's is present at pretty much the same place (i.e., $t/\alpha\sim 5-8/K$). The character of the B coefficient comparing to B_{bulk} would be roughly the same, but the absolute measure of the thickness dependence and the reduction from the bulk value become larger for larger T_K when we fit in the same temperature regime corresponding to the larger bulk value (see the figure caption of Fig. 4).

III. CONCLUSIONS

In this paper, we have reexamined our previous calculation[10] about the Kondo resistivity in thin films of alloys with $S=5/2$ impurities in the presence of spin-orbit-induced surface anisotropy. First, we presented our results on Kondo resistivity for $S=5/2$ and $T_K=10^{-3}$ K, i.e., for Cu(Mn), and fitted the $-B\ln T$ function on it. The B function in terms of the film thickness is plotted in Fig. 3 from which we can see that there is a reduction in resistivity comparing to the bulk value, but the B coefficient depends much more weakly on the thickness than in case of $S=2$, and for small t/α's B does not go to zero, but it has a minima at ca. $t/\alpha=6/K$ below that it increases which may be only a sign of the breakdown of the weak coupling calculation. These results are in good agreement with the recent experiment on Cu(Mn) films.[4] We do not get, however, their factor of 30 difference from the bulk value.

Then we have examined the B/c coefficient for different Kondo temperatures, i.e., for different 5/2-spin alloys. We have found that the character of the B/c function is the same, but the reduction from the bulk value and the thickness dependence is somewhat stronger for larger T_K.

Summarizing, the level structure of the impurity is very different for integer and half-integer spin and at the surface the spin has a degenerate ground state in the latter case, but we obtained an essentially suppressed size dependence, which is far from being trivial. The actually dependence is an interplay between different effects as the strength of the anisotropy and the different amplitudes and temperature dependence of coupling strength of the electron-induced transitions between levels. The drastically different behavior for the integer and half-integer cases found experimentally and determined theoretically provides a further support of the surface anisotropy as the origin of the size dependence.

ACKNOWLEDGMENTS

We would like to thank N. Giordano for helpful discussions. This work was supported by Grants Nos. OTKA T024005 and T029813. A.Z. benefited from the hospitality of the Meissner Institute and the LMU in Munich, and was supported by the Humboldt Foundation.

[1] J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).
[2] See, for a review, M.A. Blachly and N. Giordano, Phys. Rev. B **51**, 12 537 (1995); J.F. DiTusa, K. Lin, M. Park, M.S. Isaacson, and J.M. Parpia, Phys. Rev. Lett. **68**, 678 (1992); G. Neuttiens, J. Eom, C. Strunk, V. Chandrasekhar, C. Van Haesendonck, and Y. Bruynserade, Europhys. Lett. **34**, 623 (1996).
[3] C. Strunk, M. Henny, C. Schönenberger, G. Neuttiens, and C. Van Haesendonck, Phys. Rev. Lett. **81**, 2982 (1998).
[4] T.M. Jacobs and N. Giordano, Europhys. Lett. **44**, 74 (1998).
[5] G. Bergmann, Phys. Rev. Lett. **67**, 2545 (1991).

[6] V. Barzykin and I. Affleck, Phys. Rev. Lett. **76**, 4959 (1996).
[7] M.A. Blachly and N. Giordano, Phys. Rev. B **49**, 6788 (1994).
[8] I. Martin, Yi Wan, and Philip Phillips, Phys. Rev. Lett. **78**, 114 (1997).
[9] O. Újsághy, A. Zawadowski, and B.L. Gyorffy, Phys. Rev. Lett. **76**, 2378 (1996); O. Újsághy and A. Zawadowski, Phys. Rev. B **57**, 11 598 (1998).
[10] O. Újsághy and A. Zawadowski, Phys. Rev. B **57**, 11 609 (1998).
[11] L. Borda and A. Zawadowski, cond-mat/9903054 (unpublished).
[12] V.M. Fomin, V.N. Gladilin, J.T. Devreese, C. Van Haesendonck, and G. Neuttiens, Solid State Commun. **106**, 293 (1998).
[13] C. Roth, C. Sürgersand, and H.v. Löhneysen, Phys. Rev. B **54**, 3454 (1996).
[14] O. Újsághy and A. Zawadowski (unpublished).