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Splitting of the Kondo resonance in anisotropic magnetic impurities on surfaces

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Abstract. Using the numerical renormalization group method, we study the splitting of the Kondo resonance by a magnetic field applied in different directions in the Kondo model for anisotropic magnetic impurities. Several types of magnetic anisotropy are considered: the XXZ exchange coupling anisotropy $J_\perp \neq J_z$, the longitudinal magnetic anisotropy DS_z^2 and the transverse magnetic anisotropy $E(S_x^2 - S_y^2)$. In the spin-1/2 model with the XXZ exchange coupling anisotropy, we find very small direction dependence in the magnitude of the splitting. In the spin-3/2 model with the easy-plane ($D > 0$) anisotropy, we observe very unequal magnitudes with further differences between the x- and y-directions in the presence of an additional transverse anisotropy. A simple and rather intuitive interpretation is that the splitting is larger in magnetically soft directions. The magnitude of the splitting is directly related to the energy differences between spin states and it is only weakly modified by some multiplicative factor due to Kondo screening. The results for the $S = 3/2$ model are in good agreement with recent scanning tunneling spectroscopy studies of Co impurities adsorbed on CuN islands on Cu(100) surfaces (Otte A F *et al* 2008 *Nat. Phys.* **4** 847).

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Contents

1. Introduction

Transition metal atoms with partially filled d shells tend to form local magnetic moments. When such magnetic atoms are embedded in simple metallic hosts or adsorbed on their surfaces, they induce various anomalies in the low-temperature thermodynamic and dynamic properties (Kondo effect). The most distinctive feature is the presence of a sharp resonance in the density of states near the Fermi level, which is known as the Abrikosov–Suhl resonance or the Kondo resonance. This resonance cannot be interpreted in the single-electron picture; it is of many-particle origin due to enhanced exchange scattering of the low-energy conduction-band electrons, which screen the impurity moment. By applying an external magnetic field, the impurity becomes spin polarized, the Kondo resonance splits and the Kondo effect is suppressed when the Zeeman energy $g\mu_B B$ is increased beyond the characteristic energy scale $k_B T_K$, where T_K is the Kondo temperature.

Experimental attempts to directly observe the Kondo resonance in dilutely doped metals using the photoemission spectroscopy are beset with difficulties in measuring spectral features near the Fermi level, weak signals and limited energy resolution. A more favourable approach is the transport spectroscopy of quantum dots with nonzero total spin of the confined electrons [1]–[4]; these nanostructures can be considered as artificial magnetic atoms with an additional benefit of being easily tunable by changing potentials on the external electrodes. In these systems, the Kondo effect manifests as a zero-bias anomaly in the transport properties and the Kondo resonance can be probed by measuring the differential conductance as a function of the bias voltage, although the interpretation of these results is not straightforward since the quantum dot is driven out of equilibrium [5]. The splitting of the Kondo resonance was observed by applying a magnetic field [1, 2, 5], but there remain open questions regarding the magnitude of the splitting [4].

The first successful probes of real magnetic impurities using scanning tunneling microscopy (STM) were performed for magnetic impurities adsorbed on noble metal surfaces almost a decade ago [6, 7]. The hybridization of the impurity with the substrate is relatively strong in these systems (chemisorption) and the tip of the STM acts essentially as a non-perturbing probe of the local density of states at the adsorption site; the impurity is thus in equilibrium with the substrate metal. Rather than a Lorentzian-like Kondo resonance, a Fano-resonance-like feature is seen in the dI/dV spectra, presumably because the electrons are mostly tunneling into the sp-like adatom levels, which hybridize strongly with the substrate and extend the furthest out into the vacuum region, while direct tunneling into the strongly localized inner d-shells is unlikely; this interpretation is confirmed by the very weak dependence of the tunneling spectra on the tip–surface separation. Strong hybridization results in relatively high Kondo temperatures (tens of K), thus laboratory magnetic fields are not strong enough to induce an observable Kondo resonance splitting (at most one could expect to observe a small broadening of the Kondo resonance which might be within the energy resolution of ultra-low-temperature STMs). Recently, the idea was put forward to adsorb magnetic atoms on ultra-thin 'isolating' CuN islands on the Cu(100) substrate, thereby reducing the Kondo temperature to the more appropriate 1 K range. The splitting of the Kondo resonance in the magnetic field can then be easily observed using fields in the T range [8, 9]. Furthermore, in these systems the zero-bias anomaly in the $\mathrm{d}I/\mathrm{d}V$ spectra takes the simple form of a Lorentzian-like resonance, which makes the comparison with theory more straightforward. In recent spin-excitation spectroscopy measurements using an STM, the magnetic field was applied in different directions in the Co/CuN/Cu(100) system, uncovering strongly anisotropic Kondo resonance splitting with magnitudes differing by a factor of two [9]. Prior studies have demonstrated that such magnetic adsorbates may be well described by simple Kondo-like impurity models with magnetic anisotropy terms [10]. In this paper, we thus study spectral functions of high-spin Kondo models with various sources of magnetic anisotropy in an external magnetic field. In the case of the $S = 3/2$ Kondo model with easy-plane magnetic anisotropy, as is relevant for the experiments in the Co/CuN/Cu(100) system, we find results which agree very well with the experiment.

2. Model and method

We consider the anisotropic Kondo impurity model [11]–[20]

$$H = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + J_z s_z S_z + J_\perp (s_x S_x + s_y S_y) \tag{1}$$

$$+ D S_z^2 + E(S_x^2 - S_y^2) + \sum_\alpha g_\alpha \mu_\mathrm{B} B_\alpha S_\alpha. \tag{2}$$

Operators $c_{k\sigma}$ describe conduction band electrons with momentum $\boldsymbol{k}$, spin $\sigma \in \{\uparrow, \downarrow\}$, and energy $\epsilon_{\boldsymbol{k}}$, while $\boldsymbol{s} = \{s_x, s_y, s_z\}$ is the spin density of the conduction-band electrons at the impurity site. Furthermore, $\boldsymbol{S} = \{S_x, S_y, S_z\}$ are the quantum mechanical impurity spin-S operators, J_z and $J_\perp$ are the longitudinal and transverse Kondo exchange coupling constants, D is the longitudinal and E the transverse magnetic anisotropy. Finally, the g_α are the g-factors, μ_B the Bohr magneton and B_α the magnetic field; index $\alpha \in \{x, y, z\}$ denotes a direction in space. Note that we have assumed that the magnetic anisotropy tensor and the g tensor are diagonal in the same frame; this is not necessarily always the case.

The Kondo resonance splitting in the $S = 1/2$ Kondo model and in the Anderson impurity model has been studied in the isotropic case by a number of techniques: Bethe ansatz [21, 22], slave-boson mean-field theory [23], local-moment approach [24, 25], spin-dependent interpolative perturbative approximation [26] and numerical renormalization group [19], [27]–[30]. Some analytical Fermi-liquid theory results are known in the low-field limit [24], while the high-field limit is accessible by perturbation theory. The nontrivial range is the cross-over regime, $g\mu_\mathrm{B} B \sim k_\mathrm{B} T_\mathrm{K}$.

We performed calculations using the NRG method [31]–[33]. The results for anisotropic impurities in the absence of the field were reported in [34]; here, we extend those studies to situations with a magnetic field oriented in an arbitrary direction. Spectral functions are computed using the density-matrix approach [35] with averaging over many interleaved discretization grids and using a narrow broadening kernel in order to reduce errors due to over-broadening that affect NRG results at high energies [36].

In the Kondo model, the Kondo resonance may be most conveniently observed in the T-matrix spectral function [27, 37, 38]. The T-matrix is defined through $G = G^{(0)} + G^{(0)} T G^{(0)}$, where $G^{(0)}$ and G are the conduction-band electron propagators in the clean system and in the system with impurity, respectively. The T-matrix completely characterizes the scattering on the impurity and, in particular, it contains information on both elastic and inelastic scattering cross sections [38, 39]:

$$\sigma_{\mathrm{total}}(\omega)/\sigma_0 = \rho\pi^2 \left[-\frac{1}{\pi} \operatorname{Im} T(\omega) \right], \tag{3}$$

$$\sigma_{\mathrm{el}}(\omega)/\sigma_0 = \rho^2\pi^2 |T(\omega)|^2, \tag{4}$$

$$\sigma_{\mathrm{inel}}(\omega) = \sigma_{\mathrm{total}}(\omega) - \sigma_{\mathrm{el}}(\omega), \tag{5}$$

where σ_0 is the cross section in the case of unitary scattering. We also note that the T-matrix spectral function, $-(1/\pi)\operatorname{Im} T(\omega)$, is the Kondo-model equivalent of the d-level spectral function in the closely related Anderson impurity model. As a first approximation, we may assume that the total scattering cross section is directly related (proportional) to the measured differential tunneling current for magnetic adsorbates on decoupling layers.

3. The splitting of the Kondo resonance

We introduce $\Delta_\alpha = g_\alpha \mu_{\mathrm{B}} B_\alpha$, i.e. the impurity energy level spacing due to the magnetic field in the direction α (Zeeman splitting). The g-factors g_α cannot be easily determined in experiments, but assuming that the orbital magnetism in the magnetic adsorbate is completely quenched, we may as a first approximation assume an isotropic g tensor, $g_\alpha \equiv g_{\mathrm{s}} \approx 2$. In theoretical calculations, the actual values of g_α do not need to be known as they enter the Hamiltonian only indirectly as energies Δ_α, however, they are important for the correct interpretation of experimental results [4]. For Co/CuN/Cu(100), isotropic $g \approx 2.2$ was established [9].

We designate by δ_α the position of the Kondo resonance, which we will determine as a function of the Zeeman splitting Δ_α for a field applied in the direction α. The δ versus Δ curve (in particular, in the cross-over regime at intermediate fields $\Delta \sim k_{\mathrm{B}} T_{\mathrm{K}}$) depends strongly on how the position δ is extracted from the spectral functions. First, we note that the Kondo resonance is not a simple Lorentzian peak even in the zero-field limit, but it rather has logarithmic tails [28, 36], [40]–[42]. Furthermore, the peak shape becomes increasingly asymmetric as the magnetic field is established [24, 28, 29]. As no unbiased peak fitting procedure can be introduced in general, it is only meaningful to define the peak position as the energy of its maximum. Further ambivalence arises here, since it is possible to extract the maximum either from the spin-averaged spectral function (procedure A) or from individual spin-dependent components (procedure B). The differences between the results extracted by the two procedures are the most pronounced at the onset of visible peak splitting in the spin-averaged spectral function, but the two approaches become equivalent in the high-field limit. Since the peak shapes themselves vary with Δ, there is no generic way to relate the results obtained using the two procedures. Experimentally one measures the spin-averaged spectral function, while theoretically one is more interested in the individual components, thus in this work we consider peak positions as obtained by both procedures. Where needed, we will distinguish them by a superscript, i.e. $\delta_\alpha^{\mathrm{A}}$ versus $\delta_\alpha^{\mathrm{B}}$.

3.1. Isotropic case

For reference, let us consider first the known case of the isotropic $S=1/2$ Kondo model. In the limit of high magnetic fields, the splitting is expected to be linear, i.e. $\delta=\Delta$ for $\Delta \gg k_{\mathrm{B}}T_{\mathrm{K}}$ (the distance between the two peaks in the spin-averaged spectral function is thus exactly twice the Zeeman energy); this is the usual Zeeman splitting for isotropic free spins. In the low-field limit, the splitting is reduced by strong correlations of the Kondo ground state and one expects to find linear splitting, but with a different slope, i.e. $\delta=2/3\Delta$ for $\Delta \ll k_{\mathrm{B}}T_{\mathrm{K}}$ [21, 24, 29]. The factor 2/3 can be derived using Fermi-liquid theory arguments [24]. The (slow) cross-over between the two limiting regimes occurs around $\Delta \sim k_{\mathrm{B}}T_{\mathrm{K}}$.

The Kondo-resonance splitting in the isotropic $S=1/2$ Kondo model with $\rho J=0.1$, i.e. $T_{\mathrm{K}}=1.16\times10^{-5}W$, is shown in figure 1. The extraction of δ is difficult and error-prone in the small-field limit (since the peak shift is much smaller than the peak width) and in the large-field limit (due to NRG discretization artefacts), but it is fortunately more reliable in the cross-over regime which is of main interest. Nevertheless, deviations by a few percent from the universal Kondo-scaling-limit results are expected for two reasons: (i) NRG artefacts make it difficult to pinpoint the exact position of the peak maximum; and (ii) for very large fields, B becomes comparable to the scale of J for the present choice of model parameters and non-universal behaviour must occur. We were thus not able to verify the approach to the expected limiting behaviours $\delta=2/3\Delta$ and $\delta=\Delta$. We note, in particular, that the ratio δ/Δ exceeds 1 in the large-field limit. For smaller $\rho J=0.05$ with much reduced Kondo temperature $T_{\mathrm{K}}=3\times10^{-10}$, we do not observe such behaviour for equivalent $\Delta/k_{\mathrm{B}}T_{\mathrm{K}}$, thus true universal properties are only observed in the extreme Kondo limit.

3.2. XXZ exchange anisotropy, $J_\perp \neq J_z$

We studied the XXZ exchange anisotropic $S=1/2$ Kondo model with $J_\perp=2J_z$ for $\rho J_z=0.1$. In the $S=1/2$ model, the exchange anisotropy is an irrelevant perturbation and the system flows to the same strong-coupling fixed point as in the isotropic Kondo model. Nevertheless, while the fixed point itself is isotropic, the expansion around the fixed point in terms of the irrelevant operators will be anisotropic with expansion parameters which are functions of both $J_\perp$ and J_z, thus dynamic response of the system is expected to exhibit some weak effects of the anisotropy. We indeed find that the dynamic magnetic susceptibility is somewhat larger in the transverse direction than in the longitudinal direction at all frequencies. Anisotropic polarizability results in unequal splitting of the Kondo resonance. We thus find that the splitting magnitude is slightly larger for a field applied in the transverse direction. The difference is largest for small fields (but still at most a few percent as extracted by procedure B) and it goes to zero in the large field limit where the impurity again behaves as a free isotropic spin.

The XXZ exchange anisotropy is a relevant perturbation in the $S\geqslant1$ Kondo models and it generates the DS_z^2 magnetic anisotropy term during the renormalization process [12, 13].

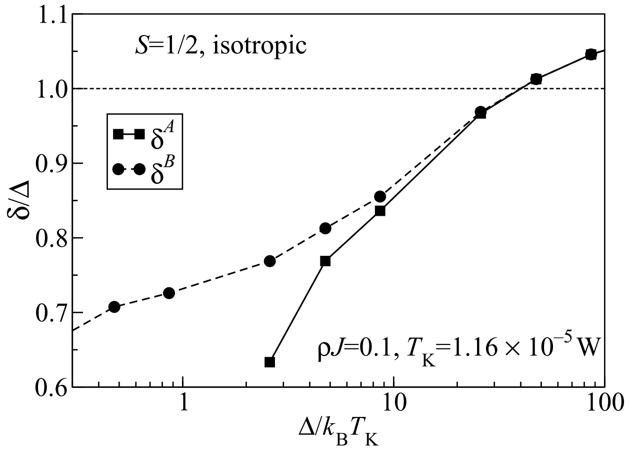

Figure 1. Kondo-resonance splitting in the isotropic $S = 1/2$ Kondo model as a function of the magnetic field. We plot the position of the Kondo peak, δ, rescaled by the Zeeman energy, $\Delta = g\mu_B B$, as a function of $\Delta/k_B T_K$. The peak position is extracted either from spin-averaged spectral function (δ^A) or from individual spin components (δ^B). Model parameters: $\rho J = 0.1$, flat band of width $2W$, so that $\rho = 1/2W$.

Therefore we will not consider the effects of the XXZ exchange anisotropy in the $S \geqslant 1$ models here, since the qualitative behaviour of these models is similar to that of models with bare DS_z^2 terms which are discussed in the following.

3.3. Longitudinal magnetic anisotropy, DS_z^2

We now study the effects of the longitudinal magnetic anisotropy term DS_z^2 by considering the prototype $S = 3/2$ Kondo model with easy-plane anisotropy ($D > 0$). This model undergoes effective $S = 1/2$ Kondo effect if $D > T_K^{(0)}$, where $T_K^{(0)}$ is the Kondo temperature for the isotropic $S = 3/2$ model [34]. In figure 2, we plot spectral functions for a range of magnetic fields. We find unequal splitting of the Kondo resonance for fields applied in the longitudinal and transverse directions with the ratio of the splitting magnitudes (defined as the slopes of δ versus Δ curves) being near two, see the inset in figure 2. The coefficient in the z-direction is approximately 1 (as in the isotropic $S = 1/2$ Kondo model in the high-field limit), while the coefficient in the directions x and y is twice as large. There is also some offset: the extrapolated δ versus Δ curves do not pass through the origin. This is a simple consequence of extracting δ by procedure A. We find that the splitting coefficients are to a good approximation independent of D as long as $D \gg T_K^{(0)} \sim 10^{-5}\ W$ (2.2 and 1.1 for $D = 0.001\ W$, 2.3 and 1.1 for $D = 0.01\ W$, 2.2 and 1.0 for $D = 0.1\ W$) and the ratio is thus quite generically $\sim$2. This is expected since the main role of the anisotropy term DS_z^2 is to enforce a projection to the low-energy $|S_z| = 1/2$ subspace.

In the absence of any anisotropy and in zero magnetic field, Kondo screening of the impurity spin by half a unit from $S = 3/2$ to 1 would occur on the temperature scale $T_K^{(0)}$ with a characteristically slow approach to the unitary scattering limit ('underscreening'); the Kondo resonance is cusp-like in this case [43, 44]. If $D < T_K^{(0)}$, the screening process stops abruptly

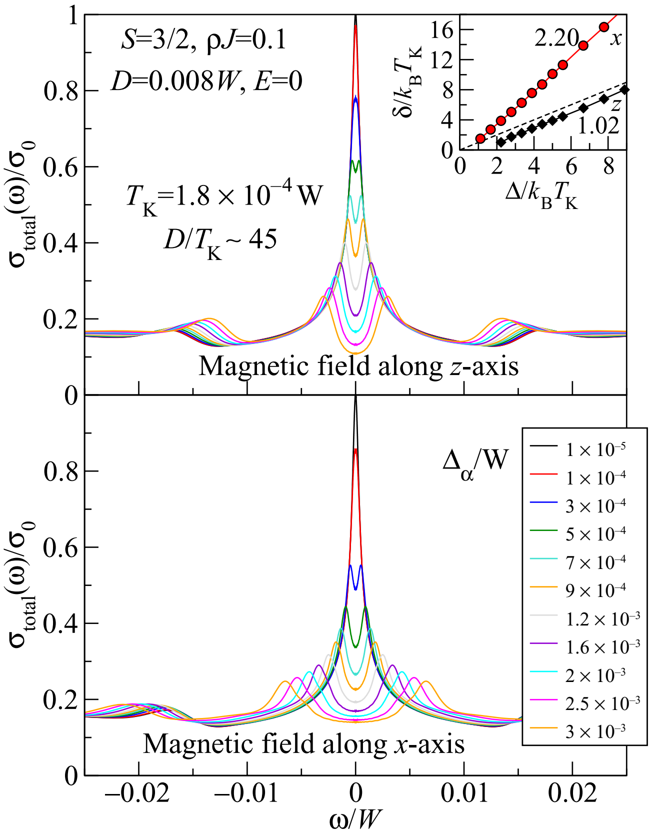

Figure 2. Spectral functions for the $S = 3/2$ Kondo model with easy-plane anisotropy, DS_z^2 with $D > 0$, for magnetic field applied in longitudinal (upper frame) and transverse directions (lower frame). Inset: magnitude of the Kondo peak splitting $\delta_\alpha^{\mathrm{A}}$ versus Zeeman energy Δ_α.

on the temperature scale of D and the impurity spin becomes completely compensated [34]. For $D > T_{\mathrm{K}}^{(0)}$, however, the $|S_z| = 3/2$ states freeze out on the scale of D, while the low lying $|S_z| = 1/2$ doublet behaves as an XXZ anisotropic spin-1/2 Kondo model [34]. For large D, the ratio of the effective exchange constants is $J_\perp/J_z = 2$; this stems from the fact that the $|S_z| = 1/2$ submatrix of the operator S_z is equal to $(1/2)\sigma_z$, while the submatrices of the operators S_x and S_y are equal to σ_x and σ_y, i.e. twice the spin-1/2 operators [34]. Furthermore, and more importantly, in the $|S_z| = 1/2$ subspace, the magnetic field couples to the x- and y-components of the effective spin with twice the usual strength Δ. In other words, the g-factor of the effective $S = 1/2$ model in the transverse directions is twice as large as the longitudinal g-factor. Having previously established that the XXZ exchange anisotropy in the $S = 1/2$ Kondo model leads to a difference of the magnitude of splitting by only a few percent, the ratio of the splitting magnitude around two in the $S = 3/2$ model can be explained essentially by the different g-factors.

There are additional features in the Kondo spectra at the excitation energy $2D$ (at zero field). These peaks shift anisotropically as the magnetic field is increased, similarly to the conductance steps in the experimental $\mathrm{d}I/\mathrm{d}V$ plots [9]. They correspond to magnetic excitations from the $|S_z| = 1/2$ states to the $|S_z| = 3/2$ states [8, 9]. The weight in these

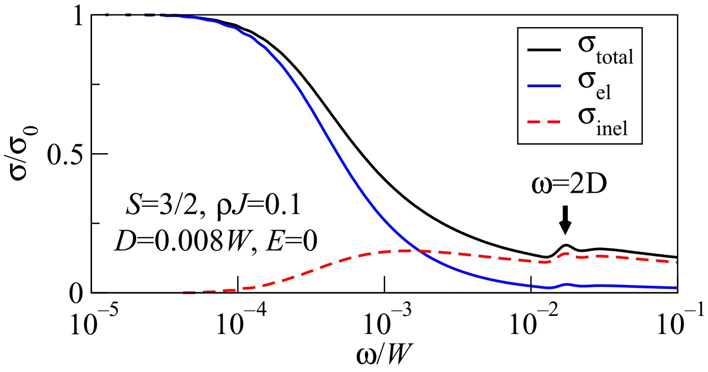

Figure 3. Decomposition into elastic and inelastic scattering rates for the anisotropic $S = 3/2$ Kondo model.

spectral features is due mostly to inelastic processes, see figure 3. These magnetic excitation peaks may be used to extract (experimentally) the g-factors if the magnetic field strength is calibrated [8, 9].

For Δ much smaller than T_K, the splitting can be extracted by procedure B. This regime is not accessible experimentally using probes which are not spin sensitive. In figure 4, we plot the splitting ratio δ/Δ as a function of the rescaled Zeeman energy $\Delta/k_B T_K$ extracted using both procedures (note that the splitting ratio δ/Δ is not the same as the slope of the 'experimental' δ versus Δ curve). We observe the transition from the low-field $\Delta \ll T_K$ Fermi liquid behaviour to the experimentally relevant intermediate-field behaviour, as well as deviations for very strong fields where nonlinear behaviour becomes manifest for the magnetic field applied in the transverse direction. Remarkably, for magnetic fields in the experimentally relevant range, the δ versus Δ curves are linear to an excellent approximation, even though the curves in the low-field and high-field limits have more complex behaviour. This is significant, since the results reported in [9] might find alternative interpretation as the peaks appear to simply follow the eigenenergies of the decoupled spin Hamiltonian, but we have shown that they are in agreement with the proposed model (i.e. the splitting of a Kondo resonance). It may be noted that in the calculations presented in figure 2, we had chosen a value of D such that the ratio D/T_K is comparable to the experimental one, as determined from the tunneling spectra shown in figure 2(b) in [9].

3.4. *Transverse anisotropy,* $E(S_x^2 - S_y^2)$

In the $S = 3/2$ Kondo model with easy-plane anisotropy we now add additional transverse anisotropy described by the term $E(S_x^2 - S_y^2)$. We observe that even a small transverse anisotropy E leads to an appreciable anisotropy in the directions x and y, figure 5. In the experimentally relevant range, the splitting curves are again linear to a good approximation. The splitting magnitude increases to 2.5 in the 'soft' direction y and it decreases to 1.8 in the 'hard' direction x. The absence of such effects in the Co/CuN/Cu(100) system suggests that the transverse anisotropy parameter E is indeed very small.

In the presence of transverse anisotropy, the effective spin-1/2 degree of freedom does not correspond to the $|S_z| = 1/2$ states, but rather to two degenerate linear combinations $\phi_{1,2}$

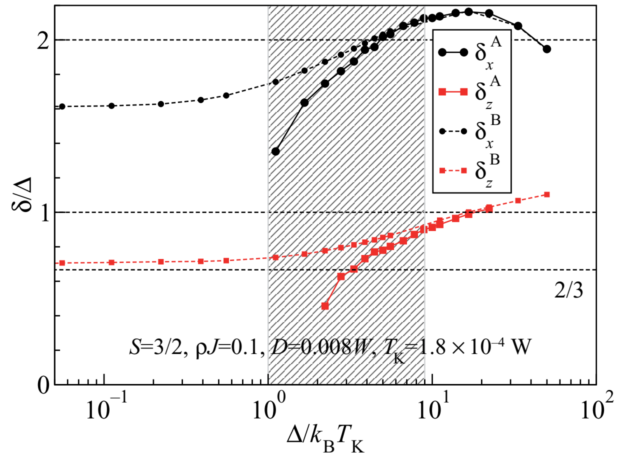

Figure 4. Anisotropic Kondo-resonance splitting in the $S = 3/2$ Kondo model as a function of the magnetic field. The dashed region corresponds to the 'experimentally accessible range'; see also the inset in figure 2.

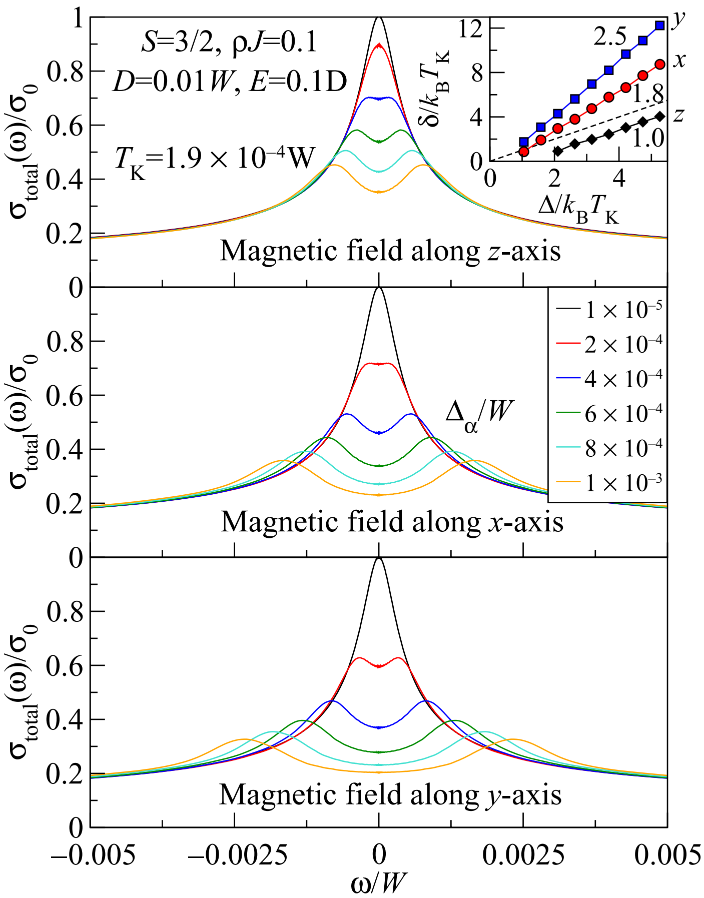

Figure 5. Spectral function for the $S = 3/2$ Kondo model with easy-plane anisotropy and weak transverse anisotropy.

of the four S_z states which depend on the ratio E/D. In order to determine the effective g-factors in this situation, one needs to project the spin-3/2 operators on the $\phi_{1,2}$ subspace, which gives

$$g_x^{\text{eff}} = \frac{1-3E/D}{\sqrt{1+3(E/D)^2}} + 1, \tag{6}$$

$$g_y^{\text{eff}} = \frac{1+3E/D}{\sqrt{1+3(E/D)^2}} + 1, \tag{7}$$

$$g_z^{\text{eff}} = \frac{2}{\sqrt{1+3(E/D)^2}} - 1. \tag{8}$$

For $E/D = 0.1$, as relevant for the case presented in figure 5, we find g-factors 1.69, 2.29, and 0.97, in fair agreement with the slopes of the δ/Δ curves shown in the inset.

4. Conclusion

For the example of the $S = 3/2$ Kondo model with easy-plane magnetic anisotropy which maps at low temperatures onto an anisotropic $S = 1/2$ Kondo model, we have shown that the effect of the external magnetic field may be interpreted in terms of effective anisotropic g-factors which result from different coupling of the magnetic field with the projected spin operators in the $|S_z| = 1/2$ subspace. The XXZ exchange coupling anisotropy, also present in the effective $S = 1/2$ model, plays a lesser role in this respect, although this anisotropy is very important in that it strongly increases the Kondo temperature of the spin-1/2 screening [34].

Generalizing these results to other high-spin models and to clusters of coupled impurities which behave as effective anisotropic spin-1/2 impurities, we expect that the magnitude of the splitting of the Kondo peak is given by the energy difference between the effective spin-1/2 states of the decoupled impurity, multiplied by some prefactor of order 1 which is a smooth function of B/T_{K} and which takes into account the effect of the Kondo screening. Since the prefactor varies very slowly (logarithmically) with B/T_{K}, it may be taken to be a constant in the experimentally relevant range of magnetic fields.

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