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Low temperature electron spin resonance theory for systems with multichannel Kondo impurities

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Abstract

Recent experiments observed an electronic spin resonance (ESR) signal from localized ytterbium ions in a Yb-based compound, manifesting non-Fermiliquid properties. Motivated by these experiments we develop a theory for the low temperature ESR in metallic systems with multichannel magnetic (Kondo) impurities.

Some rare-earth and actinide compounds exhibiting properties of heavy fermions [1] and socalled non-Fermi-liquids [2] have been the topic of many studies because of their large variety of low temperature properties. The low energy physics of these systems is determined by a hybridization of rare-earth or actinide localized electrons of 4f or 5f states with conduction electron band(s). Such a hybridization in metals produces the Kondo effect [3], i.e. the screening of the spin of a localized electron by spins of conduction electrons. It is usually believed that in heavy fermion compounds this gives rise to spin fluctuations of localized spin moments, which are completely screened below some characteristic temperature, i.e. the ground state is a singlet with a finite magnetic susceptibility. Due to the screening effective masses of carriers are enhanced, compared to normal metals. This manifests itself in large values of the low temperature magnetic susceptibility, temperature linear Sommerfeld coefficient of the electron specific heat, and low temperature coefficient of the resistivity. Such a behaviour can be described in the framework of a standard Fermi liquid theory [4] with the enhanced effective electron mass. On the other hand, for non-Fermi-liquid compounds the magnetic susceptibility and the Sommerfeld coefficient of the specific heat are usually divergent at low temperatures, while the resistivity often reveals a power-law low temperature behaviour with exponents less than two (the latter is characteristic for Fermi liquids and it is observed in heavy fermion compounds [1]). It turns out that there is no magnetic ordering in heavy fermion or non-Fermi-liquid compounds (they are metals with zero order parameter). However, very often, by tuning some parameters, like an external pressure, or by a chemical substitution, such systems undergo phase transitions to ordered magnetic or superconducting states [1, 2].

Among many experimental methods of investigation, the ESR serves as a valuable tool to study the intrinsic spin dynamics of many systems. In ESR experiments a static magnetic field is applied to the investigated system, and one measures the absorption of an ac magnetic field, polarized perpendicular to the static field direction. Due to such a geometry, the ESR serves as a very good method to study the magnetic anisotropy, and it is very sensitive to local electronic properties of a magnetic ion. However, it is usually believed that Kondo ions in heavy fermion systems are not appropriate ESR probes. The reason is that the typical rate of spin fluctuations of Kondo ions is expected to cause a large ESR linewidth [5, 6]. The spin of a local magnetic moment has to be screened by the ones of conduction electrons for temperatures less than the Kondo temperature, $T_{\rm K}$. Hence, as it was believed, this makes the ESR signal undetectable. The Kondo temperature, according to known theories, also determines the characteristic linewidth of the ESR [7]. From this viewpoint, according to previously existing theories, the ESR linewidth for systems with their $T_{\rm K} \sim 10$ K has to be about 15 T. This means that such a signal is unobservable. This is why, up to the last year, a direct observation of the Kondo ion by the ESR has been reported only in dilute Kondo alloys with extremely small Kondo temperatures [8].

Very recently the observation of the ESR signal was reported for a localized (Kondo) Yb ion in a rare-earth compound YbRh₂Si₂ [9]. The authors showed that the observed ESR spectra could be ascribed to a bulk Yb^{3+} resonance. They observed narrow ESR lines, of which the linewidths were much smaller than the Kondo temperature known from different experiments. Previous measurements of the static magnetic susceptibility, electron specific heat, and resistivity in YbRh₂Si₂ [10] revealed the effective Kondo temperature $T_{\rm K} \sim 24$ K. Nuclear magnetic resonance (NMR) study observed the behaviour, which was consistent with that characteristic Kondo temperature, $T_{\rm K} \sim 15$ K [11], i.e. of the same order of magnitude as the one derived from experiments on static magnetic susceptibility and specific heat [10]. In the framework of known theories of the ESR for Kondo systems such a Kondo temperature should be related to the linewidth 37 T (for $T_{\rm K} \sim 24$ K) or 22 T (for $T_{\rm K} \sim 15$ K). On the other hand, the experiment [9] was performed down to 1.5 K, and the characteristic linewidths were about 30 mT (corresponding to the characteristic temperature 20 mK)⁴. Hence, for the first time, the ESR signal was observed at temperatures below the Kondo temperature of a magnetic ion. This fact can indicate the existence of unscreened magnetic moments of Yb^{3+} below the Kondo temperature in that compound.

Motivated by the above, in this work we develop the low temperature theory of the ESR for multichannel Kondo systems, which can reveal the non-Fermi-liquid behaviour⁵. Let us consider the Hamiltonian

$$\mathcal{H} = \sum_{\mathbf{k},\sigma,m} \epsilon_{\mathbf{k}} c^{\dagger}_{\mathbf{k},\sigma,m} c_{\mathbf{k},\sigma,m} - \mu_{\mathrm{B}} H \left[g_{i} S^{z} + \frac{g}{2} \sum_{\mathbf{k},m} (c^{\dagger}_{\mathbf{k},\uparrow,m} c_{\mathbf{k},\uparrow,m} - c^{\dagger}_{\mathbf{k},\downarrow,m} c_{\mathbf{k},\downarrow,m}) \right] - h(t) g_{i} \mu_{\mathrm{B}} S^{x,y} - h(t) g \mu_{\mathrm{B}} \sum_{\mathbf{k},\sigma,\sigma',m} c^{\dagger}_{\mathbf{k},\sigma,m} \sigma^{x,y}_{\sigma,\sigma'} c_{\mathbf{k},\sigma',m} + \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma',m} J_{m} \vec{S} c^{\dagger}_{\mathbf{k},\sigma,m} \vec{\sigma}_{\sigma,\sigma'} c_{\mathbf{k}',\sigma',m} + \mathcal{H}_{\mathrm{anis}},$$
(1)

 4 Notice that NMR experiments [11] also observed some crossover in the temperature behaviour of NMR characteristics at 200 mK.

⁵ We do not claim that our theory totally describes the ESR experiments on YbRh₂Si₂ (the dense system, while our theory is developed for Kondo impurities). However, we believe that some important conclusions from our theory are common for all non-Fermi-liquid Kondo-type systems, in which there is no coherence between localized magnetic moments and/or between conduction electrons.

where $c_{\mathbf{k},\sigma,n}^{\dagger}$ ($c_{\mathbf{k},\sigma,n}$) are creation (destruction) operators of conduction electrons ($\sigma = \uparrow, \downarrow, m = 1, \ldots, M$ denotes the number of a channel), \vec{S} , with $\vec{S}^2 = S(S+1)$, is the operator of the spin of an impurity, $J_m > 0$ are channel-dependent exchange constants of a local spin–spin interaction between the impurity and conduction electrons, g_i is the effective g-factor of the impurity, g is the effective g-factor of conduction electrons, μ_B is the Bohr magneton, H is the dc magnetic field, h(t) is the ac magnetic field, and \mathcal{H}_{anis} is the term which describes the magnetic anisotropy of the interaction (e.g., $\mathcal{H}_{anis} = \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma',m} J_m^{\alpha} S^{\alpha} c_{\mathbf{k},\sigma,m}^{\dagger} \sigma_{\sigma,\sigma'}^{-c} c_{\mathbf{k}',\sigma',m} + (1/2) A_m (S^{\alpha})^2 + O([S^{\alpha}]^4)$ for the impurity–host coupling with the uniaxial magnetic anisotropy along the axis α), here supposed to be small, compared to the values of the isotropic exchange interaction between the impurity and conduction electrons and the bandwidth of conduction electrons. Obviously, if g_i strongly differs from g, one should observe two different ESR signals in experiments. We shall not study such a situation in detail, supposing $g_i \sim g$ in what follows (and we shall use units in which $g_i \sim g = \hbar = \mu_B = 1$).

Consider the circular time dependence of the ac magnetic field, i.e. the h(t)-dependent part of equation (1) has the form $h[\exp(i\omega t)S_{tot}^+ + H.c.]$, where $h \ll H$ is the magnitude of the ac field, $\omega \sim H$ is its frequency, and $S_{tot}^{\pm} = S_{tot}^x \pm i S_{tot}^y$ are linear combinations of the projections of the total spin of the system. In such a case we can use a unitary transformation (turn all spins about the z-axis) to remove the explicit time dependence from the term of the Hamiltonian proportional to h. It is clear that if $\mathcal{H}_{anis} = 0$, the explicit time dependence is totally removed by such a unitary transformation from the Hamiltonian. This means that the ESR signal will have resonance at $\omega = H$, with zero linewidth (δ -function peak) and no shift of the resonance due to spin-spin interactions. Naturally, additional interactions, not included in equation (1), e.g., an electron-lattice coupling or electron-nuclear one, will produce the broadening of such a δ -function line and/or a shift of the position of the resonance. A similar situation will appear if the magnetic field H is directed along the axis of the uniaxial magnetic anisotropy: one can remove the explicit time dependence from the Hamiltonian with the help of the unitary transformation. Observe that if the polarization of the ac field is not circular one can drop other (nonresonance) terms proportional to h, present in the Hamiltonian, after the unitary transformation, because $h \ll H, \omega$. In the case of, e.g., the linear polarization of the ac field, i.e. $h \cos(\omega t) S_{tot}^x$, a resonance-antiresonance situation is possible with $\omega = \pm H$, usual for the ESR case. However, we shall consider only the resonance with $\omega = H$ in detail in what follows. In our work we study the general case $\mathcal{H}_{anis} \neq 0$, considering the weak magnetic anisotropy as a perturbation.

In the framework of the linear response theory the absorption of the ac magnetic field is given by

$$I(\omega) = \frac{h^2 \omega}{2} \chi_{aa}^{\prime\prime}(q=0,\omega), \tag{2}$$

where $a \perp z$ is the direction of the polarization of the ac field and $\chi''(q, \omega)$ is the imaginary part of the dynamical magnetic susceptibility, which we take at q = 0 because the ESR is a local characteristic (usually in the ESR experiments the wavelength of the ac field is larger than the size of a sample). We point out that if the magnetic anisotropy is along the *x* direction then $\omega^2 \chi''_{xx}(0, \omega) = H^2 \chi''_{yy}(0, \omega)$ (or, if there is an angle ϕ between the axis *x* and *a* one has $\omega^2 \chi_{aa}(0, \omega) = [H^2 \cos^2 \phi + \omega^2 \sin^2 \phi] \chi_{yy}(0, \omega)).$

The dynamical magnetic susceptibility can be calculated using spin-spin correlation functions of the problem. To find the latter we use a bosonization approximation. Then our approach becomes similar to the one of [12] for the ESR theory of quantum spin chains. Such a similarity is not unexpected, since the behaviour of a Kondo impurity and the behaviour of quantum spin chains is often described by similar mathematical apparatuses; see, e.g., [13].

It is well known that in the dilute limit one can linearize the dispersion law of conduction electrons about Fermi points, and re-formulate the Kondo Hamiltonian in a one-dimensional representation (only s waves scatter off the impurity) [13]. Then following, e.g., [14, 15] we use the boson representation of Fermi operators of chiral (left- and right-moving) electron fields and the boundary conformal field theory to calculate correlation functions (related to Green functions) of the problem. We point out here that creation and destruction operators of electrons are decomposed into charge, spin and channel parts in the bosonization description. In the considered multichannel case the magnetic susceptibility of the system is related, as usual, to the spin part of electron creation and destruction operators in the boson representation. However, each of those parts affects correlations of others in the Kondo problem.

It was found [14] that the low temperature two-point correlation function of two primary fields Φ with the scaling dimensions $\Delta^{(j)} = j(j+1)/(M+2)$, where j = 1/2, 1, ..., M/2, of the *M*-channel Kondo problem can be written as ($\tau = -it$)

$$\langle \Phi^{(j_1)}(r_1,\tau) \Phi^{(j_2)}(r_2,0) \rangle = \delta_{j_1,j_2} (r_1 - r_2)^{2j_1} \left[\frac{\pi T}{v_F \sin(\pi T \tau)} \right]^{2\Delta^{(j_1)}}.$$
 (3)

The dimension of spin operators is related to j = 1, while those for electron operators correspond to j = 1/2 (spin operators can be represented as a product of two fermion operators). We are interested in left–right correlation functions, due to the standard connection of left- and right-moving fields in an open chain $\psi_{\text{R},\sigma}(r,t) = -\psi_{\text{L},\sigma}(-r,t)$.

In [15], using the boundary conformal field theory, it was found that at low energies and long distances the two-particle correlation function (e.g., the spin–spin correlator, important for the calculation of the magnetic susceptibility) involves double s-wave terms and shows a nontrivial behaviour. Notice that to calculate the three-dimensional Green function of the system with the magnetic impurity at the origin one can decompose the wavefunctions into spherical harmonics, and then take into account that only the s-wave harmonic is modified (it is different from the noninteracting value for the system without impurities). The correction to the Green function of the spin part of electrons (in the bosonization representation) due to a single Kondo impurity can be written as

$$G(r_1, r_2, \omega) = G^0(r_1 - r_2, \omega) + G^0(r_1, \omega)T(\omega)G^0(-r_2, \omega),$$
(4)

where $T(\omega)$ is the scattering matrix. We emphasize again that we are interested in spinspin correlation functions, related to two-particle functions: in the bosonization language each boson operator is related to the product of *two* Fermi operators. It is important to emphasize that three-point functions (which are necessary to find the leading correction to the Green function) are completely specified by the conformal invariance [14, 15]. For a dilute set of impurities the second term of the left-hand side of equation (4) can be replaced by $c \int dr G^0(r_1 - r, \omega)T(\omega)G^0(r - r_2, \omega)$, where *c* is the concentration of impurities. The averaging over impurities' positions restores the translational invariance of the problem. Obviously, it can be performed for small values of *c*, ignoring impurity–impurity interactions. In this limit one can neglect the difference in the behaviour of three-dimensional and onedimensional ensembles of disordered impurities. The summation over multiple-impurity terms (ignoring impurity–impurity couplings) yields the standard Dyson's form of the retarded Green function in the boson representation

$$G(k,\omega) \approx [\omega^2 - k^2 - \Pi(\omega, k, T)]^{-1}.$$
(5)

Here $\Pi(\omega, k, T) = cT(\omega)$ is the self-energy for the dilute ensemble of noninteracting impurities in the first order in the concentration of impurities. Equation (4) is written for the Green function of the one-dimensional boson (spin) field, in the presence of an interaction

between bosons caused by the magnetic anisotropy in the impurity-conduction electron coupling (which is considered in our work as a perturbation; see below).

References [14, 15] derived the expression for the dynamical magnetic susceptibility as a function of (bosonic) Green functions for the multichannel Kondo problem. The channel anisotropy does not change the form of that expression. This is why, taking into account the above analysis, the dynamical magnetic susceptibility becomes proportional to $-\omega[k^2 - \omega^2]/[k^2 - \omega^2 - \Pi(\omega, k, T)]$. The static magnetic field in the bosonization language can be eliminated by the re-definition of boson field (equivalent to the shift of chiral fields). After the Fourier transformation of spin operators this means that quasi-momenta are shifted, i.e. $k \rightarrow H$. In the situation of a small magnetic anisotropy, we have the resonance at $\omega = H$, and, hence, the shift of the resonance position is determined by the real part of $\Pi(H, H)/2H \ll H, T$, while its imaginary part defines the linewidth of the ESR.

The correction to the Green function (i.e. the self-energy) can be found by following perturbation theory in a leading irrelevant operator at the fixed point(s) of the problem, see equation (4). Notice that for the case of the total screening of the impurity spin S = M/2, i.e. in the Fermi liquid situation, one has to perform calculations to the second order in the leading irrelevant operator at the Fermi liquid fixed point.

The behaviour of Kondo impurities with magnetically anisotropic interactions and different couplings between conduction electrons for M channels is characterized by M fixed points. This is why M low energy scales $T_1 < \cdots < T_M$ are generated due to the channel anisotropy. The system flows away from one fixed point to a new one. It is important to emphasize that at $T \rightarrow 0$ one has the Fermi liquid-like behaviour with the finite static magnetic susceptibility and the Sommerfeld coefficient of the specific heat. In the scaling limit for a small easy-plane $(|J_m^{\alpha} + J_m| \ll |J_m|)$ magnetic anisotropy one can parametrize $J_m = 2\gamma_m/\theta_m v, J_m^{\alpha}/J_m = \delta_m/3, \delta_m = (\theta_m^2/2) + (\gamma_m^2/8), A_m = -(J_m + J_m^{\alpha})\delta_m/3, \text{ cf } [13, 16].$ Hence, the magnetic anisotropy is characterized by the parameters $\gamma_m \approx \cos(J_m + J_m^{\alpha})/\cos J_m$ (the isotropic limit $A_m = J_m^{\alpha} = 0$ pertains to $\theta_m \to 0$, $\gamma_m \to 0$, $\gamma_m/\theta_m = -J_m/2$). For the most important case of two channels we can explicitly find (cf [17]) two low energy scales as $T_{\rm K} \approx v_{\rm F} \exp(-\pi |\theta_1|/\nu \gamma_1)$, and $T_a \approx v_{\rm F} \cos(\pi J_1/2J_2) \exp(-\pi |\theta_2|/\nu \gamma_2)$, where $v_{\rm F}$ is the Fermi velocity of conduction electrons, and we supposed $J_1 \leq J_2$. For $T, H > T_K$ (in resonance $H \sim \omega$) we are in a high-energy regime, where the spin of the impurity is not screened. For $T_a < T, H < T_K$, the intermediate fixed point of the multichannel Kondo problem provides the non-Fermi-liquid divergent behaviour of the static magnetic susceptibility and the Sommerfeld coefficient of the specific heat (it is logarithmic for M = 2and S = 1/2 [17, 18]). However, for $T, H < T_a$ that divergent behaviour is saturated [17, 18], characteristic to the Fermi liquid. If the channel anisotropy is absent, $J_1 = J_2$, then $T_a = 0$, and there is only the non-Fermi-liquid low energy regime. On the other hand, in the extremal channel-anisotropic situation $J_1 = 0$ only one channel of conduction electrons is connected with the impurity, then the only low energy scale T_a determines the standard Kondo crossover to the low temperature Fermi liquid behaviour, as it must be.

In fact, in our perturbation scheme we calculate the self-energy Π as usual, via the correction to the Green function, cf equation (4), considering the magnetic anisotropy as a perturbation. Then, the perturbation parameter (the coupling constant λ) is related to parameters of the magnetic anisotropy (we have shown above that in such an approach it results in Kondo scales T_1, \ldots, T_M). On the other hand, in our, in fact, conformal perturbation theory the number of channels actually defines the scaling dimension for the operator under consideration. If the leading irrelevant boundary operator has the dimension $1 + \Delta$, then the corresponding coupling constant of the perturbation λ has the dimension $-\Delta < 0$ (hence, it is irrelevant).

Let us concentrate on the most important case S = 1/2. This case is of interest for YbRh₂Si₂, because the first excited crystalline electric field (CEF) state is 115 K higher than the lowest one. It was shown that in the case of the *M*-channel uniaxial Kondo interaction the (only) leading irrelevant operator has the scaling dimension $1 + \Delta$, with $\Delta = 2/(2 + M)$ [19] for $M \ge 2$, i.e. in this case we have to take j = 1, and $\Delta = \Delta^{(1)}$, see above. On the other hand, for M = 1 (the single-channel case) the only leading irrelevant operator belongs to the conformal tower of the identity operator with the scaling dimension two [15, 19]. Notice that in the absence of the magnetic anisotropy, $J_m^{\alpha} = A_m = 0$, one has $\Pi = 0$ and the Green function has a pole at $\omega = H$. Hence, the ESR intensity is proportional to the δ -function, as it must be. We point out that our conformal perturbation theory describes all low temperature regions, comparing with the low energy Kondo scales T_1, \ldots, T_M . In each energy region, however, one has to take into account the necessary fixed point(s), which determine low energy physics.

Consider the most interesting case M = 2. At very low temperatures, $T \ll T_a$ (notice that we are interested in the singular part of the Green function), where the coupling constant λ is determined by T_a , we obtain after some algebra the Fermi liquid behaviour with the resonance shift due to Kondo impurities $\Delta \omega \sim HT_a \ln(T_a/T_K)/T_K$ and the linewidth of the ESR $\Delta H \sim$ $TT_a^2 \ln^2(T_a/T_K)/T_K^2$. In the intermediate range of temperatures $T_a < T < T_K$ the non-Fermiliquid regime persists. It is governed by the intermediate fixed point. In this regime the real part of the dynamical magnetic susceptibility is given by $\chi'(\omega, T) = c \ln[T_{\rm K}/\max(\omega, T)]/\pi^2 T_{\rm K}$ and the imaginary part can be written as $\chi''(\omega, T) = c \tanh(\omega/2T)/2\pi T_{\rm K}$. In the boundary limit, i.e. near an impurity, $v_{\rm F}|\tau| \gg r_{1,2}$, the temperature dependences of the shift of the resonance caused by Kondo impurities and the linewidth of the ESR are proportional to $\sqrt{T/T_{\rm K}}$. On the other hand, in the bulk limit, $v_{\rm F}|\tau| \ll r_{1,2}$, the temperature dependences are determined by the Fermi liquid exponent. Such a difference is the manifestation of the known 'bottleneck' effect, i.e. the difference in the behaviour of the ESR for impurities and conduction electrons even if their g-factors are equal. It is possible that in ESR experiments with multichannel Kondo systems one can observe (divided in such a way) contributions from conduction electrons and impurities separately, if the concentration of impurities is low. For $T > T_{\rm K}$, one has to use $\lambda \sim \nu J_{\rm eff}(T) \sim [\ln(T_{\rm K}/T)]^{-1}$ for $J_{\rm eff} \to \infty$, which yields the shift of the resonance due to Kondo impurities inversely proportional to $H \ln(T_{\rm K}/T)$, while the linewidth of the ESR is proportional to $T[\ln(T_{\rm K}/T)]^{-2}$.

Our treatment can only be applied for low enough temperatures. At higher temperatures lattice vibrations have to modulate the CEF of ligands, which, by means of spin–orbit interactions, cause the spin–lattice relaxation. That process determines the exponential temperature dependence of the linewidth of the ESR $\sim [e^{C/T} - 1]^{-1}$, where *C* is the value of the CEF, at high temperatures [20].

Let us apply our findings to the ESR signal observed in YbRh₂Si₂. YbRh₂Si₂ has the 'easy plane' magnetic anisotropy (in our notations the axis of the anisotropy α corresponds to the *c*-axis of a crystal). The experiment [9] really observed the strong dependence of the ESR signal on the direction of the field *H*. The effective *g*-factor was 0.17 ± 0.07 for *H* parallel *c* and it was much larger, 3.56, for *H* directed perpendicular to *c*. The experiment observed a negative shift of the effective *g*-factor, related by the authors to the antiferromagnetic Kondo coupling. On the other hand, in our theory the negative sign of $\Delta \omega$ is connected with the case $T_a < T_K$ (due to the logarithm, see above). For T > 12 K the ESR linewidth manifested an exponential temperature behaviour, related to the lowest CEF doublet. For lower temperatures an increase of the linewidth linear in *T* was observed, in accordance with our theory. Using the data of the experiment [9], $T \sim 5$ K and $T_K \sim 12$ K, we obtain $T_a \sim 0.9$ K, i.e. $T_a \ll T_K$. Hence, our theory manifests a qualitative agreement with the ESR experiments on YbRh₂Si₂. On the other hand, one cannot expect a quantitative agreement of our theory with the experimental data,

because our theory essentially uses a low concentration of magnetic ions, while in YbRh₂Si₂ the concentration of magnetic ions contributing to the ESR signal was, according to estimations of the ESR intensity, 0.6.

Summarizing, we have developed the low temperature theory of the ESR for metallic systems with multichannel magnetic (Kondo) impurities, coupled to conduction electrons with magnetically anisotropic and channel-anisotropic interactions. We have shown that in the absence of the magnetic anisotropy the ESR signal has a δ -function shape, if one does not take into account electron–lattice or electron–nuclear couplings. We predict that the anisotropy of couplings of conduction electrons of different channels to the Kondo impurity introduces additional low energy scales, which define the behaviour of the low temperature ESR characteristics. It is possible that two different low energy scales (two values of the Kondo temperature), observed in experiments with YbRh₂Si₂, are related to such a channel anisotropy.

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