agnetic Resonance in Solids

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[*] In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Scaling corrections to the static magnetic susceptibility of the heavy fermions compounds YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$

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We theoretically investigated the static magnetic susceptibility in the heavy fermion compounds YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$. The molecular field approximation together with the renormalization of the Kondo interaction by the high energy conduction electrons excitations result in the Curie-Weiss law and Van Vleck susceptibility with temperature dependent Curie and Weiss parameters.

1. Introduction

Heavy fermion compounds attracted the interest of many researches in the last decades, especially since the discovery of electron spin resonance (ESR) in YbRh$_2$Si$_2$ [1] at the temperatures well below the thermodynamically measured Kondo temperature $T_K \approx 25$ K [2, 3]. A series of theoretical approaches was proposed to understand unusual behavior of these systems following both a picture of itinerant heavy electrons [4, 5] and a picture of localized Yb^{3+} spins [6-16]. Peculiar properties of heavy fermion Kondo lattices are determined by the interplay of the strong repulsion of 4f-electrons on the rare earth ion sites, their hybridization with wide band conduction d-electrons and the crystal electric field (CEF) effects. Recent angle resolved photoemission measurements [17] revealed the dispersion of the CEF-split 4f states due to f-d hybridization which was interpreted within the Anderson model. At the same time the rather narrow 4f band near the Fermi energy points out the quasi-localized nature of the f-electron motion. The ESR experiments [1, 18, 19] also indicate the importance of local properties: the angular dependence of the ESR g-factor, linewidth and intensity reflects the tetragonal symmetry of the CEF at the Yb-ion position.

In earlier works [13-16] the spin relaxation in Kondo lattice systems was successfully studied basing on the local properties of an Yb-ion in the CEF. It was shown that the Kondo effect plays an important part in the spin kinetics of heavy fermion compounds leading to the mutual cancelation of the large relaxation rates in the collective spin mode. The strong coupling between the f- and d-electrons turned out to make a negligible contribution to the effective relaxation rate in the bottleneck regime. The ESR response is determined by the relaxation of the f- and d-electrons to the thermal bath rather than by their mutual relaxation. The peculiarities of the f-d hybridization and the 4f-states dispersion do not seem to be important for the study of the ESR phenomenon which also justifies the picture of localized f-electrons as far as it concerns the dynamical properties.

In another work [10] we investigated the static magnetic susceptibility of YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$ at temperatures below T_K. The simplest molecular field approximation resulting in the Curie-Weiss law and Van Vleck susceptibilities gave an excellent agreement with experimental data. However, the comparison with experiment showed that the Curie constant and Weiss temperature considerably differed for low and high temperature regions which may indicate the Kondo effect in the static susceptibility data. An importance of Kondo renormalization for spin kinetics suggests it to have an influence on the static properties as well. In this research we extend the earlier analysis of the static magnetic susceptibility taking into account its possible renormalization due to the Kondo effect.

2. Basic model

We start from the local properties of an Yb-ion in the tetragonal CEF. A free Yb^{3+}-ion has a $4f^{13}$ configuration with one term ^2F. The spin orbital interaction splits the ^2F term into two multiplets: $^2F_{7/2}$ with $J = 7/2$ and $^2F_{5/2}$ with $J = 5/2$, where J denotes the value of the total momentum $\mathbf{J} = \mathbf{L} + \mathbf{S}$ with $\mathbf{L}$ and $\mathbf{S}$ as the orbital and spin momentum of the ion. The excited multiplet $^2F_{5/2}$ is separated from the ground state $^2F_{7/2}$ by about 1 eV. Since this value is much larger than the CEF energy, we consider in the following the ground multiplet only. Within the lowest multiplet the spin and orbital momentums of the ion are expressed via its total electronic momentum and the Lande g-factor g_J: $\mathbf{S} = (g_J - 1)\mathbf{J}$, $\mathbf{L} = (2 - g_J)\mathbf{J}$. In this way the Zeeman energy of Yb-ions can be written as follows

$$H_{ZJ} = -\mathbf{H}\mathbf{M}_J , \qquad (1)$$

where $\mathbf{H}$ denotes the external magnetic field and $\mathbf{M}_J$ is the total magnetic moment of Yb-ions.

$$\mathbf{M}_J = -\mu_B g_J \sum_i \mathbf{J}_i . \qquad (2)$$

The Kondo exchange coupling of the Yb-ion with conduction electrons and the indirect interaction between Yb-ions via the conduction electrons (RKKY interaction) are also expressed in terms of the total momentum J:

$$H_{J\sigma} = A_0 (g_J - 1) \sum_i \boldsymbol{\sigma}_i \mathbf{J}_i , \qquad (3)$$

$$H_{RKKY} = (g_J - 1)^2 \sum_{ij} I_{RKKY}^{ij} \mathbf{J}_i \mathbf{J}_j . \qquad (4)$$

Here A_0 denotes the zero order term of the Kondo exchange integral expansion in multipoles (details can be found in [20-25]), $\boldsymbol{\sigma}_i$ is the spin density of the conduction electrons at the i-th ion site and I_{RKKY}^{ij} denotes the constant of the RKKY interaction between two Kondo ions.

The energy of the Yb-ion in the tetragonal CEF reads [26]

$$H_{CEF} = \alpha B_2^0 O_2^0 + \beta (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma (B_6^0 O_6^0 + B_6^4 O_6^4) , \qquad (5)$$

where $O_k^q(\mathbf{J})$ are the equivalent operators [26], B_k^q denote the CEF parameters and $\alpha = 2/63$, $\beta = -2/1155$, $\gamma = 4/27027$. The respective eigenstates and energy splitting were found as functions of the CEF parameters in the works [10-12]. They are four Kramers doublets related to the irreducible representations Γ_7^t and Γ_6^t of the double tetragonal group [26], which are labeled in papers [11, 12] by $^1\Gamma_7^t$, $^2\Gamma_7^t$, $^3\Gamma_6^t$, $^4\Gamma_6^t$. Each doublet is described by the wave functions of the type

$$|n\nu\rangle = \sum_{M=-J}^{J} C_{n\nu M} |JM\rangle \qquad (6)$$

with $n = 0,1,2,3$ and $\nu = \uparrow, \downarrow$. The index n numbers the energy levels of the Yb-ion in the CEF, ν marks the states within a Kramers doublet and M is the eigenvalue of operator J_z (z-axis coincides with tetragonal axis of CEF). In this paper we assume the Kramers doublets sequence $^2\Gamma_7^t$, $^4\Gamma_6^t$, $^1\Gamma_7^t$, $^3\Gamma_6^t$ as experimentally observed for YbRh₂Si₂ by the angle resolved photoemission method [17].

The kinetic energy of conduction electrons and their Zeeman energy can be written as

$$H_c = \sum_{ij\lambda} t_{ij} c_{i\lambda}^+ c_{j\lambda} - \mu \sum_{i\lambda} c_{i\lambda}^+ c_{i\lambda} , \qquad (7)$$

$$H_{Z\sigma} = -\mathbf{H}\mathbf{M}_\sigma . \qquad (8)$$

Here $\lambda = \pm 1$ labels the orientation of the conduction electron spin, μ is the chemical potential,

and $\mathbf{M}_\sigma$ is the magnetic moment of the conduction electrons with the g-factor g_σ:

$$\mathbf{M}_\sigma = -\mu_B g_\sigma \sum_i \boldsymbol{\sigma}_i \,. \tag{9}$$

Finally, we represent the total Hamiltonian as $H = H_0 + H_{int}$, where

$$H_0 = H_{CEF} + H_c + H_{ZJ} + H_{Z\sigma} \,, \tag{10}$$

$$H_{int} = H_{J\sigma} + H_{RKKY} \,. \tag{11}$$

3. Molecular field approximation

The total magnetic susceptibility includes both the Kondo ions and conduction electrons contributions:

$$\chi_{\gamma\gamma'}^J = \left.\frac{\partial \langle M_J^\gamma \rangle}{\partial H^{\gamma'}}\right|_{\mathbf{H}=0} \,, \quad \chi_{\gamma\gamma'}^\sigma = \left.\frac{\partial \langle M_\sigma^\gamma \rangle}{\partial H^{\gamma'}}\right|_{\mathbf{H}=0} \tag{12}$$

where $\langle \dots \rangle$ means the statistical average and $\gamma, \gamma' = x, y, z$.

At first we consider the CEF contribution to the static susceptibility neglecting the Kondo- and RKKY interactions. The CEF symmetry implies the relation $\chi_{\gamma\gamma'} = \delta_{\gamma\gamma'} \chi_{\gamma\gamma}$ with the non-zero diagonal components $\chi_{xx} = \chi_{yy} = \chi_\perp$ and $\chi_{zz} = \chi_\parallel$. At the temperatures low compared with the CEF excited levels $\Delta_1, \Delta_2, \Delta_3$ the Yb-ions susceptibility is divided into Curie and Van Vleck parts corresponding to the contributions of the ground and excited states, respectively:

$$\chi_{\perp,\parallel}^{0J} = \frac{C_{\perp,\parallel}^0}{T} + \chi_{\perp,\parallel}^{0,VV} \,, \tag{13}$$

$$C_{\perp,\parallel}^0 = N \frac{\mu_B^2 g_{\perp,\parallel}^2}{4k_B} \,, \qquad \chi_{\perp,\parallel}^{0,VV} = N \mu_B^2 g_J^2 \sum_{n=1,2,3} \frac{1}{\Delta_n} \sum_{\nu\nu'} \langle 0\nu | J_{x,z} | n\nu' \rangle \langle n\nu' | J_{x,z} | 0\nu \rangle \,. \tag{14}$$

Here index "0" indicates the statistical averaging in (12) with the Hamiltonian H_0 defined by the equation (10), N is the number of Yb-ions; the parameters $g_\perp$ and $g_\parallel$ in the Curie constants $C_{\perp,\parallel}^0$ are actually the g-factors of the Yb-ion which become anisotropic after projection on to the ground Kramers state:

$$g_\perp = 2g_J \langle 0\uparrow | J^x | 0\downarrow \rangle \,, \quad g_\parallel = 2g_J \langle 0\uparrow | J^z | 0\uparrow \rangle \,. \tag{15}$$

As regards the conduction electrons contribution it is, in this approximation, the usual anisotropic Pauli susceptibility $\chi_{\gamma\gamma'}^{0\sigma} = \delta_{\gamma\gamma'} \chi^{0\sigma}$ with

$$\chi^{0\sigma} = \frac{1}{2} N \mu_B^2 g_\sigma^2 \rho \,, \tag{16}$$

where ρ denotes the conduction electrons density of states per lattice site at the Fermi surface.

The next step is to analyze the static magnetic susceptibility as affected by the Kondo- and RKKY interactions in the simple molecular field approximation. In this case their effects are no other than renormalization of the Zeeman energy of the Yb-ions and conduction electrons, which reduces the total Hamiltonian to

$$\tilde{H} = H_{CEF} + H_c + \tilde{H}_Z \tag{17}$$

with

$$\tilde{H}_Z = -\mathbf{H}_J \mathbf{M}_J - \mathbf{H}_\sigma \mathbf{M}_\sigma \,. \tag{18}$$

Here

$$\mathbf{H}_J = \left(1 - \lambda_A \hat{\chi}^\sigma - \lambda_I \hat{\chi}^J\right)\mathbf{H}, \quad \mathbf{H}_\sigma = \left(1 - \lambda_A \hat{\chi}^J\right)\mathbf{H}, \quad (19)$$

where

$$\lambda_A = \frac{A_0(g_J - 1)}{N\mu_B^2 g_J g_\sigma}, \quad \lambda_I = \frac{(g_J - 1)^2}{N\mu_B^2 g_J^2} \sum_i I_{RKKY}^{ij}, \quad (20)$$

A_0 and I_{RKKY}^{ij} introduced in (3), (4) and $\hat{\chi}$ denotes susceptibility tensor.

The direct calculation with the Hamiltonian (17) leads to a system of coupled equations for the conduction electrons and Yb-ions susceptibilities:

$$\begin{cases} \left(1 + \lambda_I \hat{\chi}^{0J}\right)\hat{\chi}^J + \lambda_A \hat{\chi}^{0J}\hat{\chi}^\sigma = \hat{\chi}^{0J}, \\ \hat{\chi}^\sigma + \lambda_A \hat{\chi}^{0\sigma}\hat{\chi}^J = \hat{\chi}^{0\sigma}. \end{cases} \quad (21)$$

Keeping in mind that $\hat{\chi}^{0J}$ and $\hat{\chi}^{0\sigma}$ are diagonal we arrive to the relations $\chi_{\gamma\gamma'}^J = \chi_{\gamma\gamma}^J \delta_{\gamma\gamma'}$ and $\chi_{\gamma\gamma'}^\sigma = \chi_{\gamma\gamma}^\sigma \delta_{\gamma\gamma'}$. The non-zero components $\chi_{xx} = \chi_{yy} = \chi_\perp$ and $\chi_{zz} = \chi_\parallel$ are of the form

$$\chi_{\perp,\parallel}^J = \chi_{\perp,\parallel}^{0J} \frac{1 - \lambda_A \chi^{0\sigma}}{1 + (\lambda_I - \lambda_A^2 \chi^{0\sigma})\chi_{\perp,\parallel}^{0J}}, \quad (22)$$

$$\chi_{\perp,\parallel}^\sigma = \chi^{0\sigma} \frac{1 + (\lambda_I - \lambda_A)\chi_{\perp,\parallel}^{0J}}{1 + (\lambda_I - \lambda_A^2 \chi^{0\sigma})\chi_{\perp,\parallel}^{0J}}. \quad (23)$$

Both ionic and conduction electrons susceptibility are seen to be renormalized by the Kondo- and RKKY interactions, moreover, the latter becomes anisotropic due to the CEF effects.

The total susceptibility $\chi = \chi^J + \chi^\sigma$ can be represented as a sum of renormalized Curie-Weiss, Van Vleck and Pauli susceptibilities. Substituting (13) into (22), (23) we obtain

$$\chi_{\perp,\parallel} = \frac{C_{\perp,\parallel}}{T + \theta_{\perp,\parallel}} + \chi_{\perp,\parallel}^{VV} + \chi^{0\sigma} \quad (24)$$

with the temperature independent Curie constants, Weiss temperatures and Van Vleck susceptibilities:

$$C_{\perp,\parallel} = C_{\perp,\parallel}^0 \left(\frac{1 - \lambda_A \chi^{0\sigma}}{1 + (\lambda_I - \lambda_A^2 \chi^{0\sigma})\chi_{\perp,\parallel}^{0,VV}}\right)^2, \quad (25)$$

$$\theta_{\perp,\parallel} = C_{\perp,\parallel}^0 \frac{\lambda_I - \lambda_A^2 \chi^{0\sigma}}{1 + (\lambda_I - \lambda_A^2 \chi^{0\sigma})\chi_{\perp,\parallel}^{0,VV}}, \quad (26)$$

$$\chi_{\perp,\parallel}^{VV} = \chi_{\perp,\parallel}^{0,VV} \frac{(1 - \lambda_A \chi^{0\sigma})^2}{1 + (\lambda_I - \lambda_A^2 \chi^{0\sigma})\chi_{\perp,\parallel}^{0,VV}}. \quad (27)$$

The Pauli susceptibility in (24) is obviously negligible compared with the other terms.

The expression (24) coincides qualitatively with the previous results [10] but in this study we consider also the contribution of the CEF excited states to the Curie-Weiss susceptibility and the conduction electrons influence on the Van Vleck part. Although the Curie-Weiss law and Van Vleck susceptibilities (24) are well fitted to experimental data the fitting parameters C, θ and χ^{VV} proved to be different for low and high temperature regions. This temperature dependence may indicate the Kondo effect which leads to an additional renormalization of the static susceptibility.

4. Scaling corrections to the static susceptibility

At first glance, the standard perturbation expansion is the simplest way to develop a theory beyond the molecular field approximation. However, this is not sufficient at low temperatures: the calculations to the second order in the Kondo interaction show the logarithmic divergences of the type $\ln(T/W)$, where W is a conduction electron band width. The perturbation technique can be improved on the basis of the Anderson's "poor man's scaling" [27] method which allows one to take into account the influence of high energy conduction electrons excitations on the low energy physics by a renormalization of Kondo coupling constants.

In this research we consider the scaling procedure only within the ground Kramers doublet neglecting the renormalization effects on the CEF excited states. According to neutron scattering experiments the CEF excited levels are located at 17, 25, 43 meV (197, 290, 499 K) for YbRh$_2$Si$_2$ [28] and 18, 25, 36 meV (209, 290, 418 K) for YbIr$_2$Si$_2$ [29]. Therefore the low temperature physics ($T \ll 200$ K) is well described by the lowest Kramers doublet with the total electronic momentum of the Yb-ion represented in terms of the effective spin $S = 1/2$:

$$J^z = \frac{g_\parallel}{g_J} S^z, \qquad J^{x,y} = \frac{g_\perp}{g_J} S^{x,y}, \tag{28}$$

where $g_\parallel$ and $g_\perp$ are defined by (15). After projection on to the state the Kondo interaction (3) takes the form

$$H_{s\sigma} = \sum_i \left[J_\perp (S_i^x \sigma_i^x + S_i^y \sigma_i^y) + J_\parallel S_i^z \sigma_i^z \right], \tag{29}$$

with $J_{\perp,\parallel} = A_0 \dfrac{g_J - 1}{g_J} g_{\perp,\parallel}$.

The "poor man's scaling" procedure projects the original Kondo interaction (29) on to the low energy conduction electrons states yielding a Hamiltonian $\tilde{H}_{s\sigma}$ with new Kondo couplings $\tilde{J}_\perp$ and $\tilde{J}_\parallel$ (details see in [14, 15]). The renormalized parameters $U_\perp = \widetilde{\rho J_\perp}$ and $U_\parallel = \widetilde{\rho J_\parallel}$ become temperature dependent:

$$U_\perp = \bar{U}/\sin\varphi, \qquad U_\parallel = \bar{U}\cot\varphi. \tag{30}$$

Here $\bar{U} = \rho\sqrt{J_\perp^2 - J_\parallel^2}$, $\varphi = \bar{U}\ln(T/T_{\text{GK}})$, the abbreviation "GK" indicates the Kramers ground state and T_{GK} denotes a characteristic temperature given as follows

$$T_{\text{GK}} = W \exp\left[-\frac{1}{\bar{U}} \arccos\left(\frac{g_\parallel}{g_\perp} \right) \right]. \tag{31}$$

The quantities T_{GK} and $\bar{U}$ are scaling invariant which do not change with renormalizing the Hamiltonian $H_{s\sigma}$.

The renormalized static susceptibility is formally the same as that found in the molecular field approximation (24) with the Curie constant and Weiss temperature replaced by temperature dependent parameters:

$$\tilde{\chi}_{\perp,\parallel} = \frac{\tilde{C}_{\perp,\parallel}}{T + \tilde{\theta}_{\perp,\parallel}} + \chi_{\perp,\parallel}^{VV}, \tag{32}$$

where

$$\tilde{C}_{\perp,\parallel} = C_{\perp,\parallel}\left(a_{\perp,\parallel} - \bar{U}\cot\varphi \right), \qquad \tilde{\theta}_{\perp,\parallel} = \theta_{\perp,\parallel}\left(a_{\perp,\parallel} - \bar{U}\cot\varphi \right) \tag{33}$$

with

$$a_\perp = \bar{U}\left(\varphi_0 / 2 + \cot\varphi_0\right), \quad a_\parallel = \bar{U}\cot\varphi_0. \tag{34}$$

Here $\varphi_0 \equiv \varphi(T = W) = \arccos(g_\parallel / g_\perp)$; $\bar{U}$ and φ are introduced in (30). As we should expect the Curie constant and Weiss temperature are renormalized by the high energy conduction electrons excitations converting to functions of temperature, whereas the Van Vleck part related to the CEF excited states is not affected by the renormalization.

It is also interesting to compare our result with the one-impurity susceptibility found within the isotropic s-d model. If we drop the Weiss temperature arising from the molecular field contributions and neglect the Van Vleck contribution then, in the isotropic limit $g_\perp = g_\parallel = g$ ($J_\perp = J_\parallel = J$), we immediately obtain the well known result

$$\chi_\perp = \chi_\parallel = \frac{\mu_B^2 g^2}{4k_B T}\left[1 - \frac{1}{\ln(T/T_{GK})}\right], \tag{35}$$

where T_{GK} given by (31) takes the form of one-impurity Kondo temperature $T_{GK} = W\exp(-1/\rho J)$.

The expressions (32)-(34) describe the temperature dependence of the static magnetic susceptibility renormalized by the high energy conduction electrons excitations. The new Curie and Weiss parameters decrease upon lowering temperature and logarithmically diverge at $T \rightarrow T_{GK}$:

$$\tilde{C}_{\perp,\parallel}, \; \tilde{\theta}_{\perp,\parallel} \sim a_{\perp,\parallel} - \frac{1}{\ln(T/T_{GK})}. \tag{36}$$

However, one must remember that the "poor man's scaling" approach is only valid for the temperatures well above T_{GK} and the singularities indicate just a decrease of $\tilde{C}_{\perp,\parallel}$ and $\tilde{\theta}_{\perp,\parallel}$ with temperatures lowering to T_{GK}.

5. Summary

In conclusion, we carried out a theoretical analysis of the static magnetic susceptibility of YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$. An improved molecular field approximation approach shows that the CEF excited states give a contribution to the Curie-Weiss susceptibility and the conduction electrons influence on the Van Vleck part. Besides, an additional renormalization of the static magnetic susceptibility due to the Kondo effect converts the Curie and Weiss constants to temperature dependent functions, which agrees qualitatively with the experimental data [10]. The detailed quantitative comparison with experiment will be the subject of another paper.

Acknowledgments

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