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Spin Correlations Effect on the Electrical Resistivity in Rare Earth Intermetallics

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The magnetic resistance anomaly in the paramagnetic phase of ordered cubic rare earths systems is considered. Within simple model of the free electrons scattering on the localized moments of magnetic ions system in the Born approximation it is shown that the negative temperature coefficient of the resistivity can be attributed to the spin correlations. The condition for the anomaly is formulated, which generalizes the ones known for the crystal field-free systems in the critical region. The conclusions are illustrated by the results of the resistivity calculations for DyAg and their comparison with the experimental data.

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1. Introduction

The magnetic part of resistivity of some rare-earth (RE) intermetallics, e.g. DyAg, GdAg [1], $TbIn_3$ [2], $GdIn_3$ [3], featuring strong localization of $4f$-electrons and stable rare-earth ion valency, exhibits the negative temperature coefficient within the whole paramagnetic region. This feature has not been explained to date. For the critical temperature vicinity $T \simeq T_c$, in the case of crystal field-free systems (e.g. Gd-ones), the anomaly has been interpreted as resulting from the conduction electrons elastic scattering on the spin fluctuations. The Ornstein–Zernike-type correlation functions applied to conceive the critical fluctuations resulted in showing that the crucial for the anomaly were the correlations at the wave vector close to the magnetic ordering one: $|\boldsymbol{q}| \simeq 0$ for ferromagnets, [4] and $|\boldsymbol{q}| \simeq |\boldsymbol{Q}_0| > 0$ for antiferromagnets [5].

In the present paper we consider the rare earths magnetic resistance anomaly in the paramagnetic region, within the same as in [4, 5] model for the resistivity and scattering. Describing the spin correlations within the molecular field — random field approximation (MFA–RPA) allows us to take into account the scattering on the fluctuactions as well as on the inelastic, crystal field induced, collective excitations. We show that the negative temperature coefficient of resistivity, above certain temperature $T > T_c$, can be attributed to the values of the wave-vector-dependent inter-ion exchange energy $\mathcal{J}(\boldsymbol{q})$ in the region $|\boldsymbol{q}| \leq 2k_F$. The energy depends on the range of magnetic interactions, as we show comparing the Ruderman–Kittel–Kasuya–Yosida (RKKY) with the nearest and the next nearest neighbors (N+NN) approximation. The anomaly condition that we have formulated supports and generalizes, as we argue, the ones presented in mentioned papers. As the illustration of the exchange range influence on the resistivity anomaly we present and discuss the results of the resistivity calculations for DyAg.

2. The model

We will use the standard for RE Hamiltonian $\mathcal{H}_{RE} = \mathcal{H}_0 + \mathcal{H}_{sf}$, where $\mathcal{H}_0 = \mathcal{H}_{el} + \mathcal{H}_{CF} + \mathcal{H}_{int}$, where in turn $\mathcal{H}_{el}$ describes the conduction electrons and the sum $\mathcal{H}_{CF} + \mathcal{H}_{int}$ — magnetic ions system. The first term in the latter relates to the crystal field and the second one reads $\sum_{n,m} \mathcal{J}(\boldsymbol{R}_n - \boldsymbol{R}_m)\boldsymbol{S}_n\boldsymbol{S}_m$ and describes magnetic interactions between spins $\boldsymbol{S}_n \equiv \boldsymbol{S}(\boldsymbol{R}_n)$, localized at lattice sites $\boldsymbol{R}_n$. $\mathcal{J}(\boldsymbol{R}_m - \boldsymbol{R}_n)$ denotes the energy of the inter-ion exchange interaction. The interaction between conduction electrons and magnetic ions, being the source of magnetic resistance, is given by $\mathcal{H}_{sf} = j_{ex} \sum_n \delta(\boldsymbol{r} - \boldsymbol{R}_n)\boldsymbol{s}\boldsymbol{S}_n$, where $\boldsymbol{s}$ denotes the spin of conduction electron. To calculate the magnetic resistivity we employ the formula derived from the variational solution of the Boltzmann equation with the Born approximation for the scattering, see e.g. [6]. For the cubic RE systems it reads

$$\rho(T) = \frac{\rho_2}{(2k_F)^4} \int_0^{2k_F} \mathrm{d}q q^3 \int \frac{\mathrm{d}\Omega_q}{4\pi} \int_0^\infty \mathrm{d}(\hbar\omega) \frac{\hbar\omega}{k_B T} \frac{\Im\chi_{ion}^{zz}(\boldsymbol{q},\omega,T)}{\sinh^2(\hbar\omega/2k_B T)},$$

$$\rho_2 = \frac{3V}{8N} \frac{j_{ex}^2}{e^2} \frac{m\pi}{\hbar\varepsilon_F}, \tag{1}$$

where m denotes electron mass, k_F and ε_F — Fermi momentum and Fermi energy respectively, and N/V is the density of magnetic ions. The formula expresses the spin correlation contribution to the resistivity by the imaginary part of the ionic magnetic susceptibility $\Im\chi_{ion}^{zz}(\boldsymbol{q},\omega)$ which, in virtue of the fluctuation-dissipation theorem, is expressed by the correlation function of ionic spin operators. Considering the spin correlations in the framework of MFA–RPA we get, for $T > T_c$, [7]:

$$\frac{\Im\chi_{RPA}^{zz}(\boldsymbol{q},\omega,T)}{\hbar\omega} = \pi\mathcal{W}_0(\boldsymbol{q},T)\delta(\hbar\omega) + \pi\sum_l \frac{\chi_{inel}(\mathcal{E}_l(\boldsymbol{q}),T)}{\mathcal{E}_l(\boldsymbol{q},T)[\mathrm{d}\chi_{inel}(\omega)/\mathrm{d}\hbar\omega]_{\hbar\omega=\mathcal{E}_l(\boldsymbol{q})}}$$
$$\times[\delta(\hbar\omega + \mathcal{E}_l(\boldsymbol{q},T)) + \delta(\hbar\omega - \mathcal{E}_l(\boldsymbol{q},T))], \tag{2}$$

$$\mathcal{W}_0(\boldsymbol{q},T) = \frac{\chi_{\rm el}(T)}{[1-\mathcal{J}(\boldsymbol{q})\chi_{\rm inel}(\omega=0,T)][1-\mathcal{J}(\boldsymbol{q})\chi_{\rm CF}(\omega=0,T)]},$$

with $\mathcal{J}(\boldsymbol{q})$ being the lattice Fourier transform of $\mathcal{J}(\boldsymbol{R}_m - \boldsymbol{R}_n)$ while $\chi_{\rm CF}(\omega,T) = \delta_{\omega,0}\,\chi_{\rm el}(T) + \chi_{\rm inel}(\omega,T)$ is the single ion susceptibility

$$\chi_{\rm el}(T) = (k_{\rm B}T)^{-1} \sum_{\substack{n=1\\ E_n=E_m}}^{2J+1} a_{nm}^2 p_n,$$

$$\chi_{\rm inel}(\omega,T) = \sum_{\substack{n,m=1\\ E_n\neq E_m}}^{2J+1} \frac{a_{nm}^2(p_n-p_m)}{E_m-E_n-\hbar\omega-\mathrm{i}\eta}, \tag{3}$$

where a_{nm} denotes the matrix element of the z-component of ionic magnetic moment between crystal field states $|n\rangle$, $|m\rangle$ of energies E_n, E_m; p_n is the thermal occupancy factor.

The sum at right hand side of (2) describes the contribution of the inelastic scattering on the collective excitations with the energies $\mathcal{E}_l(\boldsymbol{q})$ being, in MFA–RPA, the roots of the equation $1 - \mathcal{J}(\boldsymbol{q})\chi_{\rm inel}(\omega,T) = 0$. $\mathcal{W}_0(\boldsymbol{q},T)$ describes the elastic scattering on the spin fluctuations which appears to be a unique reason for magnetic resistance in the crystal field-free systems. In such a case the formula (1) is equivalent to the respective formulae in [4, 5], with the spin correlation function expressed by $\mathcal{W}_0(\boldsymbol{q},T)$ instead the Ornstein–Zernike-type function employed there.

3. Anomalous behavior of $\rho(T)$

Let us note that the sufficient condition for $\rho(T)$ to be a decreasing function of T to the constant $\rho(\infty)$, is the positive value of the coefficient which can be obtained by the high temperature asymptotic expansion of $\rho(T)$ with respect to $1/T$. Substituting (2)–(3) to (1) and making a number of tedious expansions of this formula we have obtained the following asymptotics:

$$\frac{\rho(T)}{\rho(\infty)} = 1 + \frac{1}{k_{\rm B}T} \sum_{n,m=1}^{2J+1} a_{nm}^2 \sum_{|\boldsymbol{q}|<2k_{\rm F}} |\boldsymbol{q}|\mathcal{J}(\boldsymbol{q}) + \mathcal{O}(T^{-2}). \tag{4}$$

Thus, the resistivity anomaly, i.e. the occurrence of an interval of temperatures in which the magnetic part of the resistivity decreases, is determined by the sign of the integral of the *exchange integral* taken over the range allowed for the conduction electrons scattering process, namely $|\boldsymbol{q}| < 2k_{\rm F}$.

Our calculations of this coefficient dependence on the Fermi energy for the exchange integral in the RKKY and the N+NN approximation, Fig. 1, show that the spin correlations may produce the anomaly regardless of the crystal field splitting. This conclusion refers to the systems mentioned in [4, 5] or in the Introduction, displaying the negative coefficient in the whole paramagnetic region, as well as to the case of $\mathrm{Pr_3Tl}$ [8] displaying a maximum of the magnetic resistivity at $T_{\max} > T_{\rm c}$. Therefore the anomaly condition appears to be more general than the ones pointed previously: $ak_{\mathrm{F}} < \pi$ for ferromagnets [4] and $|\boldsymbol{Q}_0| < 2k_{\mathrm{F}}$ for antiferromagnets [5]. It is easily seen, and it is illustrated in Fig. 1 for DyAg, that these conditions yield the coefficient being positive. The plots are characteristic of other RE systems. We conclude that in the systems not displaying any anomaly the exchange interaction is more properly described by the N+NN than the RKKY, as the latter gives a positive value of the coefficient in any case.

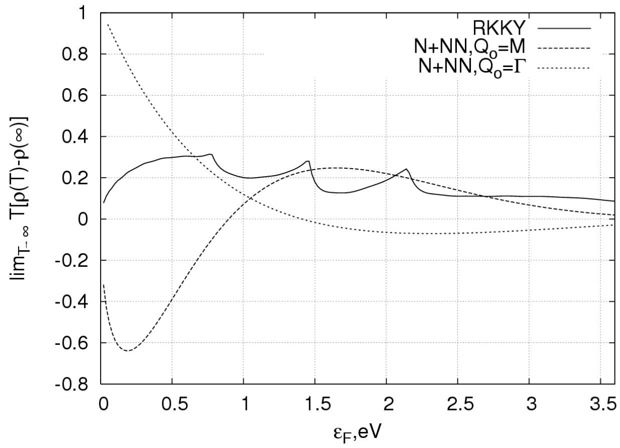

Fig. 1. Coefficient of $\rho(T)$ expansion (4) with respect to $1/T$. Cases of $\mathcal{J}_{\mathrm{RKKY}}$ and $\mathcal{J}_{\mathrm{N+NN}}$ for simple cubic lattice and the lattice parameter $a = 3.64$ Å for DyAg [9]. The plots for N+NN are at $\boldsymbol{Q}_0 \equiv \boldsymbol{M} = \pi/a(1,1,0)$ as proper for DyAg [9], and for some ferromagnet at $\boldsymbol{Q}_0 \equiv \Gamma = (0,0,0)$.

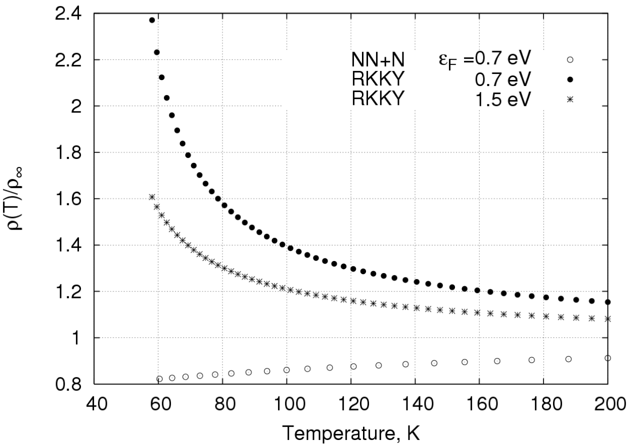

Fig. 2. Dependence $\rho(T)$ (1): comparison of exchange integrals RKKY and N+NN, data for DyAg [9].

We illustrate the dependence of the resistivity behavior on the range of the exchange interactions by the calculations for DyAg (Fig. 2). Let us note however that the best agreement with experiment [1], which we have obtained for the RKKY and $\varepsilon_{\mathrm{F}} = 0.7$ eV, is beyond the range $1.35 \div 2.00$ eV of ε_{F}, corresponding, according to our calculations, to the magnetic order vector $\boldsymbol{M} = \pi/a(1,1,0)$ for DyAg [9].

4. Conclusions

We have shown that the magnetic resistivity anomaly, recognized for the crystal field-free RE systems in the critical region, may occur in the whole paramagnetic region also for the systems with the crystal field and that the role of the spin correlations in these effects manifests itself by the exchange interactions. In the framework of MFA–RPA we have obtained a good agreement of calculations with experiment for DyAg as well as for the systems mentioned in the Introduction, which will be presented in a separate paper. For the interpretation of particular experimental results an essential significance has the approximation applied to the calculation of the inter-ion exchange interactions as well as the value of the Fermi energy, often not known from experiment. It must be admitted that the best agreement of our $\rho(T)$ calculations for various RE intermetallics with experiment we frequently obtained for ε_{F} beyond the interval determined by the RKKY for particular ordering vector $\boldsymbol{Q}_0$. The fact that in some cases a better qualitative agreement with experiment was obtained using close-range interactions rather than the RKKY also needs further discussion.

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