FIZIKA, 8 (1976) 61—71

TEMPERATURE DEPENDENCE OF THE RESISTIVITY DUE TO THE LOCALIZED SPIN FLUCTUATIONS IN TRANSITION METAL BASED ALLOYS

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Received 27. January 1976

Abstract: We show that the anomalous behaviour of the resistivity of nearly-magnetic transition-metal based alloys and of some actinide alloys and compounds can be understood in the frame of the LSF approximation of the one-band Wolff model. Theoretical results are compared with the experimental data and it is shown that the observed »universal« behaviour (different systems exibiting the same temperature dependence when plotted as a function of the reduced temperature T/T_K) follows as a consequence of the scattering of the conduction electrons on the fluctuations of the local magnetization.

1. Introduction

Since the work of Matthias et al.[1] and Clogston et al.[2] on the formation of local moments, transition-metal based alloys have often been subject of interest in metal physics and have stimulated many experimental and theoretical investigations. The resistivity data of these alloys showed a variety of behaviours including deep minima for *Mo* Fe, constant impurity resistivity for *Nb* Fe and anomalous low temperature decrease in *Rh* Fe. Detailed investigations of ternary alloys of $(Nb_x - Mb_{1-x})$ Fe revealed[3] that the resistivity minima are strongly correlated with the appearence of a permanent local moment on the iron sites, detected from the susceptibility measurements, and could be nicely explained in the s-d model.

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In this paper we want to discuss the transport properties of nearly magnetic transition-metal based alloys. We shall use a simple localised spin fluctuation model of Lederer and Mills[4] and Rivier and Zuckermann[5] and our attention will be mostly confined to the resistivity results since they provide clearest evidence and need for the LSF approach. Experimentally, these alloys exibited a new type of resistance anomaly, characterised by the extraordinary low temperature decreases in the resistivity, first seen by Coles in *Rh* Fe[6]. Similar resistance anomalies were subsequently found in the variety of systems: *Ir* Fe[7], *Pd* Ni[8], *Rh* Mn, *Rh* Co[9], *Pt* Fe[10], *Pt* Co[11], *Ru* Fe[12], *V* Ti[13], *V-Cr* Fe[14], etc. It emerged that in a large number of transition-metal alloys and several alloys and compounds of actinides (Brodsky[15]) the resistivity behaves in a more or less similar fashion. Contrary to Kondo alloys, it increases rapidly with increasing temperature up to a certain characteristic temperature T_K, above which it starts to flatten out, eventually reaching the high temperature plateau. At the same time the impurity susceptibility changes from temperature independent to Curie-Weiss behaviour with a negative Θ_{C-W} of order of T_K (Knapp[16], Loram et al.[10]).

In an early attempt to explain the resistivity data of these alloys they were considered as Kondo systems with an additional potential scattering channel (Fischer[17]; Kondo[18]; Nagaoka[19]). In calculations valid above T_K they find that the presence of non-magnetic potential scattering from the magnetic impurity gives rise to an extra factor in the coefficient of the logarithmic term which is proportional to sin $[2\eta_2(\varepsilon_F)]$. Then, if the potential scattering is strong enough, i. e. the phase shift $\eta_2(\varepsilon_F)$ is larger thant $\pi/4$, the sign of the Kondo resistivity effect will be reversed. However, a strong objection to the application of this mechanism to explain the behaviour of above mentioned alloys arises from the small magnitude of their residual resistivities. If the phase shift $\eta_2(\varepsilon_F)$ is large, the residual resistivity which is proportional to $\sin^2\eta_2(\varepsilon_F)$ should be large as well. In fact, just the opposite is found: residual resistivities of *Rh* Fe or *Ir* Fe, for example, are one order of magnitude smaller than in normal Kondo systems (Nagasawa[20]).

The reason for such a small amount of potential scattering is that in all these alloys the electronic structure of solute and solvent is rather similar. As a rule, in all the systems in which the *Rh* Fe — like anomaly is observed, both the impurity (3d-metal) and the host (4d or 5d-metal) belong to the same or neighbouring columns of the periodic table. This situation is exactly the opposite of what one has in normal-metal transition-metal alloys. There, the relevant part of the impurity electronic structure has a d-like character while the band electrons are s- or p-like (Friedel[21]). As shown by Rivier and Zitkova[22], in the former case the localised state can be expressed as a linear combination of the host electronic states, while in the latter case they are orthogonal to each other. If a dilute alloy is described by the nonorthogonal Anderson Hamiltonian, we could, in principle, deal with the intermediate situations. However, because of similarity between the impurity and the host, we shall assume that the one-band Wolff Hamiltonian with

small potential scattering is a sufficiently good representation of these particular transition-metal alloys (Coles alloys).

Finally, before explicating our model in detail, we want to discuss briefly the two-band LSF model of Lederer and Mills[4] besause the concepts they developed to explain the low temperature resistivity of *Pd* Ni are of great importance in our treatment as well. They start from the observation that although an isolated Ni ion is non-magnetic in Pd, if the Ni concentration exceeds 2at%, the Ni sites acquire a moment and the spins order ferromagnetically. Thus, one should expect to find the low-frequency fluctuations of the spin density to be enhanced in the vicinity of the impurity cell, compared to their amplitude far from the impurity.

To calculate the transport properties of such a system they assumed that the electrical current is carried by the s-electrons of Pd, while the principal low temperature contribution to the transport relaxation rate comes from the scattering of the s-electrons by the already described spin density fluctuations in the d-band. The electrical resistivity produced by inelastic collisions of s-electrons with d--electron spin fluctuations can be obtained by applying the standard variational procedure to the Boltzmann equation.

The low temperature resistivity is given by the expression

$$\varrho(T) = \varrho_0 \frac{\pi}{3}(T/T_K)^2,$$

which agrees with the experimental data. Parameter T_K is the characteristic temperature of the localised spin fluctuations. Extending these calculations and applying them to isoelectronic transition metal based alloys, Kaiser and Doniach[23] have shown that at higher temperatures the resistivity becomes linear in T. However for the enhancement factor T_K independent of T the resistivity never saturates, nor even decreases below the linear law. In order to fit the experimental data and obtain the saturation one has to assume that T_K is extremely large, on one hand, and that it is temperature dependent, on the other hand. Yet, it is not at all clear that the assumptions made to arrive at expression for $\varrho(T)$ will hold in this situation. The temperature dependence of T_K can be understood if one assumes the temperature dependent band susceptibility (which would involve another parameter, the paramagnon lifetime). The numerical results of Jullien, Beal-Monod and Coqblin[24]show that in the Kaiser and Doniach theory the saturation limit is practically never reached unless the local degeneracy temperature is of the order of the conduction electron degeneracy temperature (paramagnon temperature). Thus, it is clear that in the two-band LSF theory the high temperature behaviour of nearly magnetic alloys is not described in the satisfactory manner.

2. *Spin fluctuations in one band model*

We propose here a simple model to describe the transport properties of a system in which both solute and solvent are transition metals. As already mentioned we assume that in an alloy like *Rh* Fe or *Ir* Fe the hybridisation between s- and d-electrons is sufficiently strong to allow the transport properties to be discussed in terms of one electron band only*).

We suppose that the appropriate Hamiltonian to deal with the above mentioned systems is the Wolff Hamiltonian (Wolff[27], Moriya[28]) which reads

$$H = H_0 + H_i, \tag{1}$$

where H_0 describes the conduction band and H_i is the interaction part due to the presence of impurity. Explicitly, we write H_i as

$$H_i = W \Sigma_\sigma a_{0\sigma}^+ a_{0\sigma} + U n_{0\uparrow} n_{0\downarrow}, \tag{2}$$

where $n_{0\sigma} = a_{0\sigma}^+ a_{0\sigma}$, $a_{0\sigma}^+ = (1/N) \Sigma_K a_{k\sigma}^+$ is the creation operator in conduction band, N is the number of atoms in the unit volume, W is the one-body scattering potential and U is the extra Coulomb repulsion between electrons of the oposite spin which takes place in the impurity cell.

Here, we would like to point out that in contrast to Lederer and Mills[4] and Kaiser and Doniach[23], who calculated the resistivity by applying the variational procedure to the Boltzmann equation, we calculate the conductivity, i. e. we calculate the self-energy of a local electron and relate it to the transport relaxation time $\tau(\varepsilon)$[29]. As is well known, in the multiple scattering approximation we can write for $\tau(\varepsilon)$

$$\frac{1}{\tau} = - N_i \, Im \, T_1, \tag{3}$$

where T_1 is the scattering matrix for the single impurity and N_1 is the number of impurities. In the case of the Wolff Hamiltonian, T_1 can be written in terms of the localised electron self-energy Σ_0 as

$$T_1(\varepsilon) = \frac{W + \Sigma_0}{1 - G_0 (W + \Sigma_0)}. \tag{4}$$

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*) We are extending the results of Rivier and Zlatić[25] to include the effect of the impurity potential in addition to the LSF scattering. Similar extension is also attempted, on the slightly different basis, by K. Fischer[26] but the results he obtains for the resistivity of »Coles alloys« are different from ours since the self-energy he is using is incorrect.

In equation (4), $G_0(\varepsilon)$ is the bare ($U = 0$) local Green function defined by

$$G_0(\varepsilon) = \Sigma_k \frac{1}{\varepsilon - \varepsilon_k + i\delta}, \tag{5}$$

and in what follows we shall assume for G_0 to describe a parabolic band

$$G_0(\varepsilon) = \frac{1}{\varepsilon - E_0 + i\Gamma}, \tag{6}$$

where E_0 is the centre and Γ is the width of the electron band.

To evaluate $\Sigma_0(\varepsilon)$ we notice first that by introducing a new propagator G_1.

$$G_1 = \frac{G_0}{1 - W G_0},$$

or

$$(\tilde{E}_0 = E_0 + W) \tag{7}$$

$$G_1 = \frac{1}{\varepsilon - \tilde{E}_0 + i\Gamma},$$

Dyson equation can be written as

$$G(\varepsilon) = G_1(\varepsilon) + G_1(\varepsilon)\Sigma_0(\varepsilon)G(\varepsilon). \tag{8}$$

Next, recalling that the effect of adding the 3d-impurity to the 4d or 5d host results in an increase of the amplitude of the local spin fluctuation, we assume that the dominant contribution to $\Sigma_0(\varepsilon)$ comes from the scattering of electrons on the LSF which takes place at the impurity site. Thus, in the LSF approximation (Rivier and Zuckermann[5]), we have for $\Sigma_0(\varepsilon)$

$\Sigma_0 =$  (9)

where the wriggly line is the LSF propagator $\chi(\varepsilon)$ and the straight line is the electron propagator $G(\varepsilon)$. Since in the systems we are concerned with the electron--hole resonance (repeated scattering of an electron-hole pair) forms the most important Bose like excitation, the LSF propagator is well approximated by the local dynamic susceptibility

$$\chi_0(\varepsilon) = \frac{1}{T_K + i\varepsilon}. \tag{10}$$

Although Equs. (9) and (10) are reminiscent of the RPA results of Lederer and Mills[4] and Rivier and Zuckermann[5], here $\chi_0(\varepsilon)$ is a phenomenological expression and T_K is in principle determinable from the measurements of the local susceptibility. (For review of the NMR experiments in dilute alloys see Narath[30]).

Finally, since $\chi_0(\varepsilon)$ is a phenomenological expression, in calculating $\Sigma_0(\varepsilon)$ we replace $G(\varepsilon)$ by $G_1(\varepsilon)$ and obtain

$$\Sigma_0(\varepsilon) = k_B T \sum_n \chi_0(i\omega_n)\, G_1(i\omega_n + \varepsilon). \tag{11}$$

Making the analytical continuation to the real axes (Abrikosov et al.[29]) we arive at the expression for $\Sigma_0(\varepsilon)$

$$\Sigma_0(\varepsilon) = \Gamma^2 (I_1 + I_2 + I_3), \tag{12}$$

where

$$I_1 = \frac{1}{\{\varepsilon - \tilde{E}_0 \pm i(M+\Gamma)\}} \frac{\pi}{M\beta}$$

$$I_2 = \frac{\varepsilon - \tilde{E}_0 \pm \Gamma}{\{\varepsilon - \tilde{E}_0 \pm i(\Gamma + M)\}\{\varepsilon - \tilde{E}_0 \pm i(\Gamma - M)\}} \cdot$$

$$\cdot \left\{\psi\left(1 + \frac{\Gamma\beta}{2\pi} \mp i\frac{(\varepsilon - \tilde{E}_0)\beta}{2\pi}\right) - \psi\left(1 + \frac{M\beta}{2\pi}\right)\right\} \tag{13}$$

$$I_3 = \frac{\mp i\Gamma \cdot \psi\left(1/2 + \frac{M\beta}{2\pi} \mp i\frac{\varepsilon\beta}{2\pi}\right)}{\{(\varepsilon - \tilde{E}_0) + i(\Gamma \pm M)\}\{\varepsilon - \tilde{E}_0 - i(\Gamma \mp M)\}} \mp$$

$$\mp \frac{1}{2}\left\{\frac{\psi\left(\frac{1}{2} + \frac{\Gamma\beta}{2\pi} + i\frac{\tilde{E}_0\beta}{2\pi}\right)}{\varepsilon - \tilde{E}_0 \pm iM + i\Gamma} - \frac{\psi\left(\frac{1}{2} + \frac{\Gamma\beta}{2\pi} - i\frac{\tilde{E}_0\beta}{2\pi}\right)}{\varepsilon - \tilde{E}_0 \pm iM - \Gamma}\right\}.$$

In Equ. (13) parameter $M = k_B T_K$ defines the width of the LSF spectrum and $\psi(x)$ denotes digamma function (Abramovitz and Stegun[31]). From Equs. (13), (4) and (3) the relaxation time follows at once.

However before discussing our resistivity results in more details we want to make a comment on the specific heat of »Coles« alloys. We are concerned with the low temperature contribution to the electronic specific heat due to the localised spin fluctuations. If we calculate the entropy of the local interacting Fermi gas, then the impurity part of the specific heat is simply obtained as

$$C_V = T\left(\frac{\mathrm{d}S}{\mathrm{d}T}\right)$$

and we have (Abrikosov et al.[29], Chp. 4;)

$$C_V = \frac{\pi^2 k_B^2 T}{3} \Sigma_\sigma \frac{1}{\pi} Im \left\{ (G_0^R)^{-1} \frac{\partial G_0^R}{\partial \varepsilon} \right\} T = 0, \tag{14}$$

where $G_0^R(\varepsilon)$ is the Fourier transform of the retarded local Green function evaluated at zero temperature. Using for $G_0^R(\varepsilon)$ expression (8) with Σ_0 given by equation (11) we obtain (see also Lederer[32])

$$C_V = \frac{2\pi k_B T}{3 T_K}. \tag{15}$$

The dependence of C_V on the inverse of the spin fluctuation temperature is a characteristic feature of the local enhancement and it is in sharp contrast with the uniform enhancement theory (Berk and Schrieffer[33]; Doniach and Engelsberg[34]).

3. *Temperature dependence of the resistivity*

To calculate the temperature dependence of the resistivity we use the well known expression

$$\frac{1}{\varrho} = 2 \frac{e^2 v_F \varrho_F}{3} \int d\varepsilon \left(-\frac{\partial f}{\partial \varepsilon} \right) \tau(\varepsilon), \tag{16}$$

where v_F is the Fermi velocity, ϱ_F is the density of states at the Fermi level and $f(\varepsilon)$ is the distribution function.

It is important to notice that for $k_B T \ll \Gamma$ (which is always the case since $\Gamma \simeq 10^5$K) and for $\widetilde{E}_0 \ll M$, the structure of the self energy $\Sigma_0(\varepsilon)$ is such that it enables one to define the reduced temperature and energy: $\widetilde{T} = T/T_K$ and $\widetilde{\varepsilon} = \varepsilon/k_B T_K$. Thus, as long as the dominant contribution to the resistivity of a dilute alloy comes from the scattering by the localised spin fluctuations, the temperature dependence of the resistivity will have the same functional form, no matter how different the systems might appear. This universality is clearly seen in the resistivities of *Rh* Fe and *Ir* Fe which scale to the same curve if we plot them as a function of the reduced temperature $T = 2\pi(T/T_K)$ and chose $T_K = 15$K for *Rh* Fe and $T_K = 225$K for *Ir* Fe, (Rivier and Zlatić[25]*).

In Fig. 1 (1) we plotted numerically evaluated temperature dependence of $\varrho(T)$ (Equ. 16) normalized to one at $T \to \infty$, as a function of the reduced tempe-

*) These values for T_K are about eight times those extracted from the low temperature regimes by Kaiser and Doniach[24] but agree with those derived from the susceptibility measurements of Waszink[35] and of Knapp and Sarachik[36].

rature $\widetilde{T}$ and for various values of potential scattering. With regard to this figure we observe that even large changes in the bare energy shift $\widetilde{E}_0$ are not immediately reflected in the corresponding change of the resistivity curves. Although surprising, this result does not violate the Friedel's theorem since in the presence of the LSF scattering the bare $\widetilde{E}_0$ has little physical meaning. Rather, it is the renormalised shift, $\widetilde{E}_0 + Re\,\Sigma_0$, which is related to the charge difference between the impurity and the host, and which has to be in agreement with the Friedel's theorem. The renormalised shift, however, is only a slowly varying function of $\widetilde{E}_0$, with the consequence that even with large E_0 we can still have a small charge difference and hence a small residual resistivity (see Fig. 1).

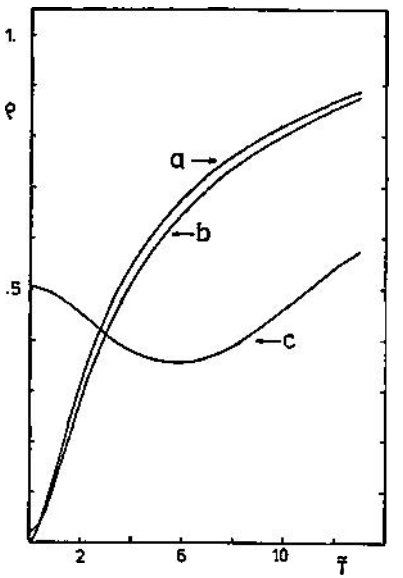

Fig. 1. The normalised resistivity, $\varrho\,(T)$ as a function of reduced temperature $\widetilde{T}$, $\widetilde{T} = 2\pi\,T/T_K$, is plotted for various values of $\widetilde{E}_0$ ($\widetilde{E}_0 = E_0 + W$). Curves a), b), c) correspond to $\widetilde{E}_0 = 0.0$; $\widetilde{E}_0 = 0.6$; $\widetilde{E}_0 = 2.0\ \Gamma$.

Next, we remark that the characteristic temperature T_K separates two different physical regimes. Below T_K, the impurity appears to be non-magnetic and the resistivity follows the »simple power low« behaviour. Above T_K, one cannot distinguish between the LSF and the permanent moment at the impurity site, the life time of the thermal fluctuations becomming shorter than the characteristic time of the magnetic fluctuations; in this region the resistivity exhibits first the logarithmic behaviour and then tends to a constant which we recognise as the Yosida[37] spin disorder limit.

The presence of a high temperature plateau in addition to the logaritmic temperature dependence of the resistivity is a further indication that, above T_K, the LSF describe a true spin in the sea of the conduction electrons.

4. *Comparison with experiment and conclusion*

In analysing experimental data, we assume that the impurity part of the resistivity is simply given by $\varrho_{\text{imp}}(T) = \varrho_{\text{alloy}} - \varrho_{\text{host}}$. Indeed, linear variations of $\varrho_{\text{imp}}(T)$ with concentration support our assumption that for dilute alloys the addition of impurities does not significantly change the electron-phonon resistivity. In what follows we restrict our theoretical considerations to the results which correspond to the alloys with small residual resistivities. We recall, see Fig. 1, that for $T \ll T_K$ the resistivity is a parabolic function of temperature, $\varrho(T) = \frac{\pi^2}{4} \cdot (T/T_K)^2$, while above $T_K/2\pi$ the resistivity changes linearly with T.

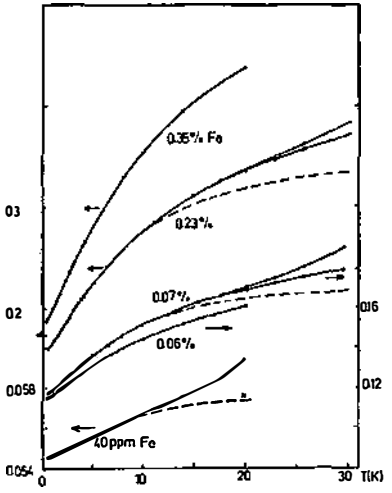

Fig. 2. Resistivity data on various *Rh* Fe alloys against temperature. The concentration effects tend to decrease the LSF temperature and push the onset of the logarithmic part of the resistivity towards lower temperatures. Dashed lines are theoreticaal curves with $\tilde{E}_0 = 0$.

Experimentally, parabolic and linear low temperature regimes are observed in a number of systems: *Rh* Fe ($T_K = 15\text{K}$[38]), *Rh* Co, *Rh* Mn[9], *Ir* Fe ($T_K = 225\text{K}$[7]), *Pt* Fe ($T_K = 2\text{K}$[10]), *Pt* Co ($T_K = 1\text{K}$[11]), *Ru* Fe ($T_K = 700\text{K}$[12]), but in most of the cases the resistivity was measured only over a limited temperature range so that it was difficult to determine the characteristic temperature accurately. Also, in analysing the experimental data one has to be sure that they correspond to single impurity limit since the interaction effects tend to decrease the LSF tem-

perature. As can be seen in Rusby's data on the resistivity of several *Rh* Fe alloys (Fig. 2), the coefficient of the T^2 term increases and the onset of the linear region is depressed towards lower temperatures as the concentration of Rh atoms becomes larger.

Aparat from *Rh* Fe and *Ir* Fe the logarithmic behaviour of the resistivity has been clearly seen in *Pt* Fe (Loram et al.[10]) and in *Pt* Co (Williams et al.[11]).

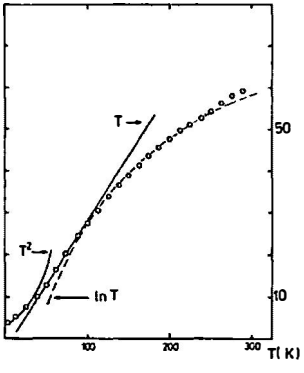

Fig. 3. Resistivity vs. temperature for Np Rh compound, with T^2, T and ln (T) temperature dependence schematicaly indicated. Ordinate unit is $1/\mu\Omega$ cm.

Finally, we mention that the detailed examination of the resistivity and susceptibility data of a great number of actinide systems (Brodsky[15]) has shown clearly that their properties can be understood in the framework of the LSF model. Fig. (3) shows an example of the resistivity vs. temperature curve of an actinide compound (Np Rh_3; $T_K = 150 \pm \pm 50$ K) in which various temperature regimes, as discussed in this paper, are clearly seen.

In conclusion, we have shown in this paper that the resistivity of »Coles« alloys can be described by a simple model involving a single band of host electrons scattered by localised spin fluctuations. The resistivity is given by the universal function of temperature which decreases as T^2 for $T \to 0$ and reaches the finite spin disorder value as $T \to \infty$.

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ZAVISNOST ELEKTRIČNOG OTPORA LEGURA PRIJELAZNIH METALA O TEMPERATURI

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Sadržaj

Ispituje se ponašanje onih legura, dobivenih otapanjem paramagnetskih primjesa u nemagnetskim prijelaznim metalima, koje obilježava a) sklonost lokalnom magnetizmu i b) sličnost elektronskih struktura primjese i matrice.

Pokazuje se da se anomalno ponašanje električnih otpora takovih legura može objasniti u LSF aproksimaciji Wolffovog hamiltonijana. Eksperimentalno opažena »univerzalnost« (isto temperaturno ponašanje različitih legura na reduciranoj temperaturnoj skali T/T_K) posljedica je raspršenja vodljivih elektrona na fluktuacijama lokalne magnetizacije.