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Electrical resistivity of $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ and $R\text{Fe}_{11.3}\text{W}_{0.7}$ alloys

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Abstract

We have measured the electrical resistivity of $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ and $R\text{Fe}_{11.3}\text{W}_{0.7}$ ($R = \text{Tb, Dy, Ho, Er, and Lu}$) polycrystalline systems over a temperature range of 4–700 K. The overall behaviour of the resistivity is determined mainly by the transition-metal ions. However, some interesting features are related to rare-earth atoms. At low temperatures, the resistivity, ρ , increases with temperature as AT^2 , where the coefficient A varies systematically with the total spin of rare-earth ions. We also find that ρ depends on the M element in our $R\text{Fe}_{12-x}\text{M}_x$ iron-rich compounds. Close to the Curie temperature, the resistivities of the alloys studied clearly show a change in the slope of ρ versus T . Saturation values of the high temperature magnetic resistivity follow the prediction by de Gennes and Friedel for spin disorder scattering.

1. Introduction

The pursuit of new permanent-magnet materials has become quite an active field recently. Iron-rich $R\text{Fe}_{12-x}\text{M}_x$ alloys, where R belongs to the lanthanide series and M is an *nd*-type element like Ti, Nb, or Ta, are attractive candidates for this purpose [1]. The addition of a third nonmagnetic element M stabilizes $R\text{Fe}_{12-x}\text{M}_x$ compounds in a ThMn_{12} type structure. However, it lowers their Curie temperature and spontaneous magnetization significantly [1, 2]. Therefore, the smallest possible contents of M are desirable in these alloys. A value of x as small as 0.5 has been achieved for $M = \text{Mo and Ta}$ [3, 4]. Single-phase alloys can also be obtained for Nb with $x = 0.65$ and for W with $x = 0.7$ [5, 6]. They form preferably for $M = \text{Ta and W}$ with heavy rare-earth elements.

The structural and magnetic properties of $R\text{Fe}_{12-x}\text{M}_x$ compounds with various M elements and different x contents have been extensively studied. As mentioned above, these alloys

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crystallize in a body-centred tetragonal ThMn_{12} type structure which has one R site and three nonequivalent transition-metal sites (8i, 8j, and 8f), each occupied by four ions. The M atoms preferentially occupy the 8i crystallographic sites. R and Fe sublattice moments couple antiferromagnetically for heavy rare-earth elements below the Curie temperature which lies in the 400–610 K range. It has been found that the iron sublattice has a large *c*-axis anisotropy. $\text{RFe}_{12-x}\text{M}_x$ alloys show usually uniaxial magnetic anisotropy at room temperature for all heavy rare earths except $R = \text{Tb}$ for which the easy axis lies in the basal plane. In addition, some compounds exhibit a change in magnetic structure (spin reorientation) upon cooling down from room temperature [7, 6, 5]. Magnetic-field-driven spin-reorientation transitions are also observed in the Dy and Ho compounds for various M (=W, Ta, Nb, Mo and Ti) elements. A systematic variation of the Curie temperature versus the unit cell volume, for a given R element but different M atom in $\text{RFe}_{12-x}\text{M}_x$, suggests that the R–Fe intersublattice exchange coupling depends on the M element type and on its concentration [6]. Theoretical studies of $\text{RFe}_{12-x}\text{M}_x$ compounds [8, 9] and recent magnetic structure calculations using novel techniques for $M = \text{Mo}$ [10] show the importance of the local environment of iron atoms in their electronic structure. In spite of the relative abundance of results on structural and magnetic properties of these alloys, we could not find any report on their transport properties.

The electronic transport properties are closely related to the magnetic state of the material. In particular, the resistivities of intermetallic compounds with transition metals exhibit unusually strong temperature variations in the regions where the magnetic order changes significantly. This may be related to the itinerant magnetism of the d bands, a point which is not usually emphasized in the interpretation of magnetic bulk properties, even though it can play a significant role in metallic compounds [11]. The magnetism of the inner 4f-shell electrons of the rare-earth ions is also important in transport properties [12, 13]. Even though transport measurements in polycrystalline materials yield averages over large numbers of randomly oriented crystallites, many interesting effects (such as anisotropic magnetoresistance or the extraordinary Hall effect) arise as a consequence of the symmetry properties of the magnetic state. Therefore, the electrical resistivity should give relevant information about the coupling of the conduction electron spins to magnetic sublattices. In addition, in $\text{RFe}_{12-x}\text{M}_x$ compounds we expect to see how *nd* electrons of the M element affect exchange couplings between R and Fe sublattices. Our aim is to infer what role the *nd* and localized 4f electrons play in electrical transport by finding correlations between the electrical resistivity and the magnetic moment of rare-earth atoms in the $\text{RFe}_{12-x}\text{M}_x$ series.

In this paper we report resistivity measurements in $\text{RFe}_{12-x}\text{M}_x$ ($R = \text{Tb, Dy, Ho, Er, and Lu, } M = \text{Ta, W}$) polycrystalline samples, with a very small content of a nonmagnetic element M, as a function of temperature in the range from 4 to 700 K. The results obtained enable us to determine what the dominant electron scattering processes are and how the rare-earth and the *nd* ions contribute to them.

The experimental procedure is described in section 2. Results of resistivity measurements are reported and discussed in section 3. Conclusions are drawn in section 4.

2. Experiment

The $\text{RFe}_{12-x}\text{M}_x$ alloys we studied were synthesized by melting pure elements (rare earths 3N, iron 3N 7, Ta 3N 5 and W 3N 5 purity) under an Ar atmosphere in a high frequency induction furnace. The ingots obtained were annealed at 800–900 K for one week and were checked by x-ray diffraction. A small amount of α -Fe ($\leq 3\%$) and TaFe_2 were found in $\text{RFe}_{11.5}\text{Ta}_{0.5}$ while in $\text{RFe}_{11.3}\text{W}_{0.7}$ traces of the 2:17 secondary phases were observed in addition to α -Fe. However, their concentration was always less than 10%. Magnetic properties of the very same samples

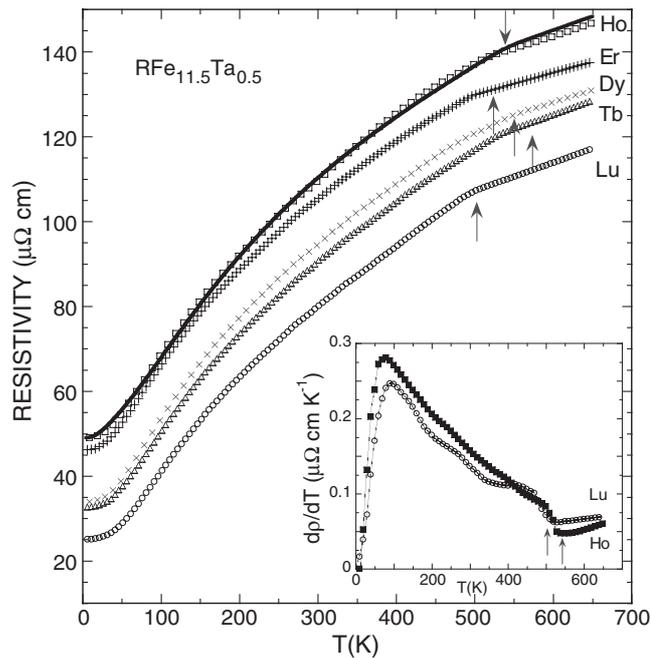


Figure 1. Resistivity data points versus temperature for $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ alloys ($R = \text{Tb}, \text{Dy}, \text{Ho}, \text{Er},$ and Lu). The arrows indicate the Curie temperature. The solid curve shows numerical fitting to the data points of $\text{HoFe}_{11.5}\text{Ta}_{0.5}$. The inset shows the temperature derivatives of ρ , obtained numerically from the experimental data, for $R = \text{Ho}$ and Lu .

as used in our study have been previously investigated in much detail [4, 6, 7]. The results obtained assure us that their properties are hardly affected by the secondary phase content. The electrical resistivity measurements were performed with a six-probe method on bar-shaped polycrystalline samples. The samples had typical dimensions of $1 \times 2 \times 10 \text{ mm}^3$. Before measurements were made, each sample was polished and checked for possible cracks. Platinum pressure contacts were applied to the samples in the whole temperature range (4–700 K). The mean relative error in our resistivity measurements is approximately 0.2%; absolute values were determined to within 5%.

3. Results and discussion

Figures 1 and 2 show how the resistivity ρ of the $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ and $R\text{Fe}_{11.3}\text{W}_{0.7}$ alloys varies with temperature, respectively. The overall behaviour of the resistivity is alike in these compounds. At low temperatures, the measured resistivity tends to some constant value. ρ is observed to increase with temperature in the whole range of temperatures studied. This increase becomes slower as temperature increases. All $\rho(T)$ curves show a small change of slope near the Curie point. Thereafter, the resistivity increases linearly with temperature. No anomalies are observed at the spin-reorientation transitions in the Dy and Er compounds.

We assume that the total resistivity is given by $\rho = \rho_{\text{res}} + \rho_{\text{ph}} + \rho_{\text{mag}}$, where ρ_{res} is the temperature-independent residual resistivity, ρ_{ph} is the resistivity brought about by electron–phonon scattering, and ρ_{mag} is the magnetic resistivity. The phonon contribution to resistivity, ρ_{ph} , can be calculated using the Bloch–Grüneisen formula [15]. However, we do not know

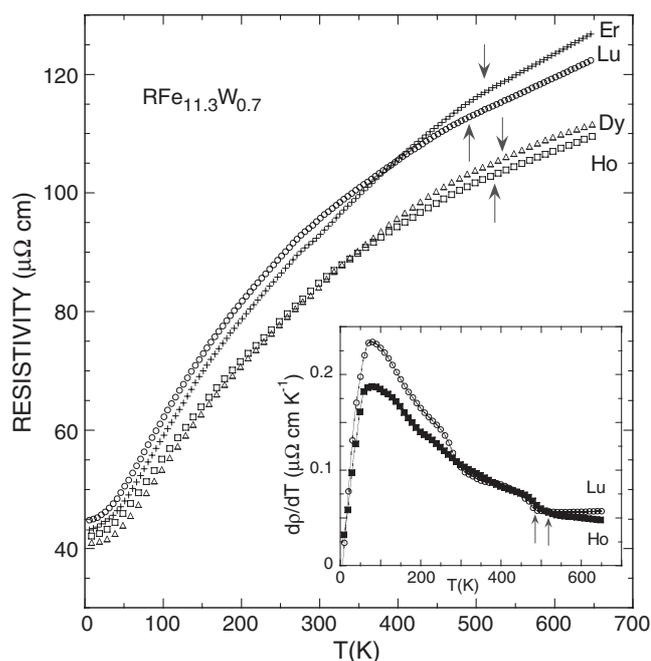


Figure 2. Resistivity data points versus temperature for $R\text{Fe}_{11.3}\text{W}_{0.7}$ alloys ($R = \text{Dy}, \text{Ho}, \text{Er},$ and Lu). The arrows indicate the Curie temperature. The inset shows the temperature derivatives of ρ , obtained numerically from the experimental data, for $R = \text{Ho}$ and Lu .

the Debye temperature θ_D of our alloys. Values between 350 and 450 K can be estimated for θ_D from Mössbauer studies [14], so we assume a value of 400 K for it. A comparison of the lattice resistivities at low and high temperatures T_1 and T_2 , respectively, using a relation $\rho_1/\rho_2 = 497.6(T_1/\theta_D)^4(T_1/T_2)$ [15], yields θ_D values in fair agreement with the one we have assumed. A plot of $\rho_{\text{mag}} = \rho - \rho_{\text{ph}} - \rho_{\text{res}}$ versus T ($T \leq 30$ K) is shown in figure 3 for the $R\text{Fe}_{11.3}\text{W}_{0.7}$ samples. We find that ρ_{mag} is proportional to T^2 in this temperature range. We have tried to fit the low temperature data without a T^2 term but this leads to quite a poor fitting and unreasonably low values for the Debye temperature. The values of the constant term ρ_{res} , obtained from a least squares fit to the relation $\rho - \rho_{\text{ph}} = \rho_{\text{res}} + AT^2$, vary between 25 and 50 $\mu\Omega$ cm for all $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ and $R\text{Fe}_{11.3}\text{W}_{0.7}$ alloys studied. We attribute scatter in these values to the impurity content of the starting elements used in the preparation. The disorder, inherent to our alloys, may be responsible in part for quite high values of ρ_{res} .

The T^2 variation of the low temperature resistivity ($\rho = AT^2$) may arise from electron–electron scattering. However, in this case typical A values found in ferromagnetic compounds are much smaller than the values obtained by us for A in the $R\text{Fe}_{11.5}\text{Ta}_{0.5}$ and $R\text{Fe}_{11.3}\text{W}_{0.7}$ alloys. In addition, we find that A varies systematically with the spin value of rare-earth ions in the materials studied as shown in figure 4. This can be likely attributed to the scattering of electrons by spin fluctuations which involve rare-earth ions. Indeed, one can expect to find in ferromagnetic materials spin waves with a quadratic dispersion relation in the long wavelength limit that gives rise to a T^2 resistivity dependence [17]. An acoustic mode involving spins of both rare-earth and iron ions with a dispersion given roughly by the R–Fe exchange interaction has been found to be important for electron scattering in $R_2\text{Fe}_{14}\text{B}$ compounds [16]. In this case, $A \propto 1/[(g_R - 1)J_R J_{R\text{Fe}}^2]$. Here, g_R and J_R are the rare-earth g -factor and total angular

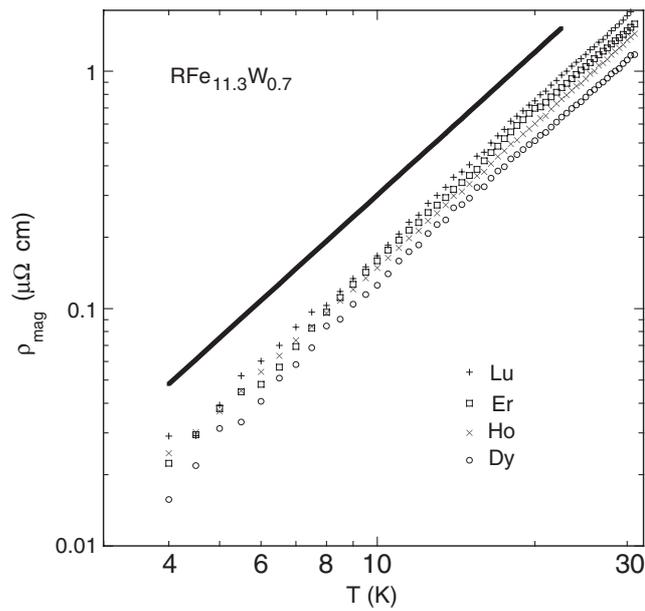


Figure 3. The total resistivity less the phonon and residual contribution versus T in the temperature range $4 \text{ K} \leq T \leq 30 \text{ K}$. The solid line shows a T^2 dependence.

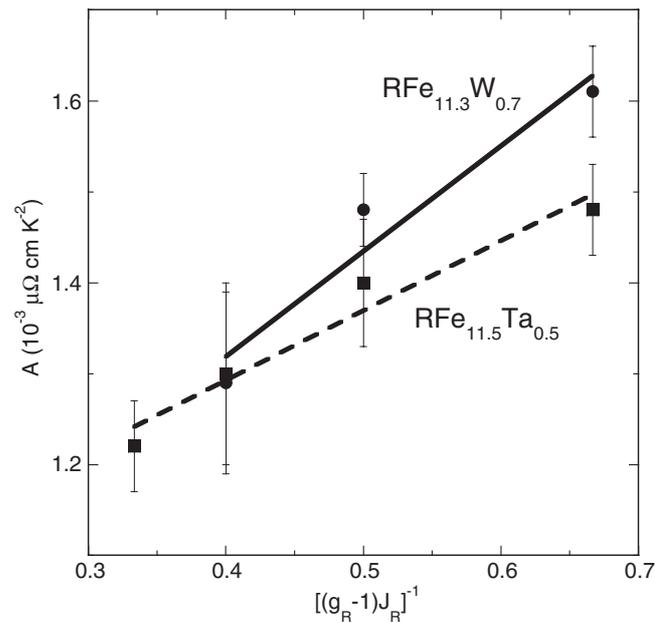


Figure 4. The values of coefficient A (from $\rho_{\text{mag}} = AT^2$) versus the inverse of the total rare-earth ion spin in $RFe_{11.5}Ta_{0.5}$ and $RFe_{11.7}W_{0.3}$ alloys.

momentum, respectively. Taking into account that the strength of R–Fe exchange interaction, J_{RFe} , in $RFe_{12-x}M_x$ [2], and specifically in $RFe_{11.5}Ta_{0.5}$ and $RFe_{11.3}W_{0.7}$ alloys [6], is hardly dependent on the R component for heavy rare-earth ions, a linear variation of A with the

inverse of $(g_R - 1)J_R$ should be obtained for such scattering. The values found for A follow this prediction quite well. We note that the A coefficients are larger in $RFe_{11.3}W_{0.7}$ alloys than in $RFe_{11.5}Ta_{0.5}$, for a given R element. Tentatively, we may attribute this to different perturbations in the 5d electron density induced by the the W and Ta ions. The R–Fe exchange interaction in intermetallic materials can be related to the coupling of 4f electrons to the 5d, 6s and 6p electrons at the rare-earth site, of which the 4f–5d interaction is most important [18]. Although an increase in the number of 5d electrons generally favours R–Fe exchange interactions, it also produces a decrease in the local 4f–5d and 5d–5d exchange [19]. The competition between these effects may lead, upon adding 5d electrons, to the decrease of J_{RFe} in $RFe_{12-x}M_x$ alloys [6]. This is in agreement with the observation that the values of A for $M = W$, which has one more 5d electron than $M = Ta$, are higher.

The derivatives of ρ with respect to T , obtained numerically from the experimental data of some samples studied, are exhibited in the insets of figures 1 and 2. As the temperature increases, the rate of variation of the resistivity goes through a maximum at $T \approx 80$ – 100 K; after that, it decreases and shows a step close to T_c . $d\rho/dT$ is almost constant for $T > T_c$. A self-consistent renormalization theory of spin fluctuations for weakly ferromagnetic metals predicts a maximum in $d\rho/dT$ as the low-temperature T^2 dependence of resistivity changes to a $T^{5/3}$ variation at higher temperatures [11]. However, the evidence for this in the systems studied is not conclusive.

In classical spin-disorder scattering models [12, 13], spin fluctuations are substantially reduced below T_c , and so is the magnetic resistivity. For localized magnetic moments, the thermal spin-disorder resistivity is given by $\rho_{\text{mag}}(T) \propto 1 - m^2$, where m is the normalized magnetization. The observed behaviour of ρ_{mag} in the alloys studied follows this prediction. We have tried to fit this simple relation to the magnetic resistivity data obtained for $HoFe_{11.5}Ta_{0.5}$. Since both localized 3d and 4f moments contribute to spin disorder scattering, $\rho_{\text{mag}} = \beta(1 - m_T^2) + \beta'(1 - m_R^2)$, where m_T and m_R are transition-metal and rare-earth sublattice magnetizations, respectively, and β and β' are the fitting parameters. As argued above, ρ_{mag} can be obtained by subtracting out the phonon and impurity contributions from the total resistivity. We note that ρ_{res} and $\rho_{\text{ph}}(T)$ are known as the former can be determined from the $T \rightarrow 0$ limiting value of ρ and the latter has no adjustable parameter once the slope of $\rho(T)$ at high temperatures and the Debye temperature are given. However, we need to know $m(T)$ in the two antiferromagnetically coupled sublattices. We have calculated m_T and m_R within a phenomenological molecular-field model for an anisotropic ferromagnet [20], assuming the free-ion moment for Ho and a value of unity for the Fe spin (values between 1.7 and 2.2 μ_B have been found for the average iron magnetic moment in various $RFe_{12-x}M_x$ compounds [2, 5, 6]). Values of exchange–interaction coefficients which yielded the best fit to the rare-earth and iron sublattice magnetizations, obtained from neutron diffraction studies in $HoFe_{11.5}Ta_{0.5}$ for $2 \text{ K} \leq T \leq 300 \text{ K}$ [7], have been used. The solid curve in figure 1 is the calculated resistivity for this compound. Our fit reproduces very well the shape of the experimental curve. However, the lack of data on $m_T(T)$ and $m_R(T)$ variations for a sufficiently wide temperature range in the alloys studied does not allow us to perform any further meaningful fittings.

In the paramagnetic region the magnetic resistivity becomes constant. Scattering from the completely disordered spin system gives a resistivity contribution proportional to GJ_{cf}^2 , where G is the de Gennes factor and J_{cf} is the interaction constant between conduction electrons and 4f localized spins. Figure 5 shows how the values of $\rho_{\text{mag}}^{\text{HT}}$, obtained by the extrapolation of the high temperature linear part of $(\rho - \rho_{\text{res}})$ to $T = 0$ K, vary with the de Gennes factor for the heavy rare-earth ions in $RFe_{11.5}Ta_{0.5}$ and $RFe_{11.3}W_{0.7}$ alloys. Good agreement with classical models is found in this case. The relative variation of the high temperature $\rho_{\text{mag}}^{\text{HT}}$ with R is

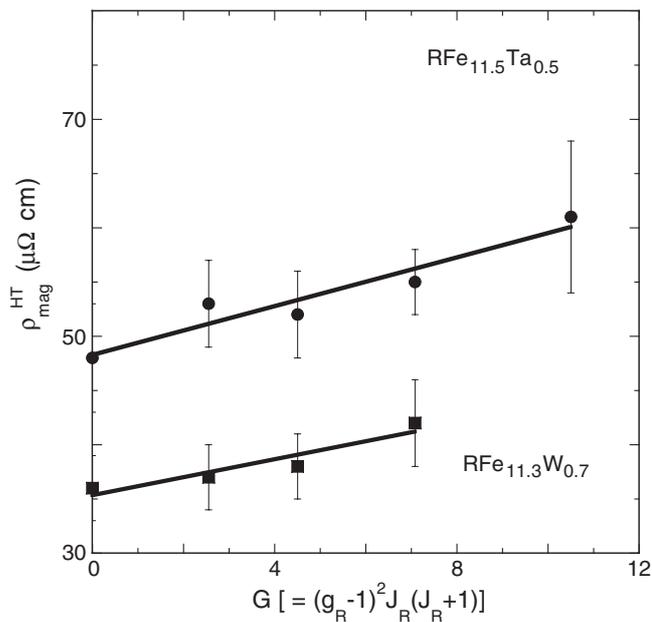


Figure 5. Saturation values of the magnetic resistivity versus the de Gennes factor for heavy rare-earth ions in RFe_{12-x}M_x compounds.

rather weak. However, its values for M = Ta are systematically higher than those for M = W. This can be explained by an increase in the conduction electron density going from Ta to W and likely decrease of J_{cf} on the base of the arguments given above.

4. Concluding remarks

In summary, we have measured the resistivity as a function of temperature in various RFe_{11.5}Ta_{0.5} and RFe_{11.3}W_{0.7} compounds. The low temperature variation of the magnetic resistivity ($\rho_{mag} \propto T^2$) can likely arise from the scattering of electrons by magnons that involve iron atoms and both rare-earth and iron atoms. The contribution of the rare-earth sublattice is inversely proportional to the total spin of R. It also depends on the R-Fe coupling which becomes weaker as the number of 5d electrons increases. In the intermediate temperature range, the electrons are scattered mainly by spin disorder. At high temperatures ($T > T_c$), the magnetic resistivity saturates. The total resistivity increases linearly with temperature due to phonon scattering. The saturation values of magnetic resistivity are smaller for higher 5d-electron densities. A simple model which includes spin disorder, phonon, and impurity scattering accounts well for the temperature variation of the resistivity in HoFe_{11.5}Ta_{0.5}.

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