Spin Disorder Scattering in Magnetic Metallic Alloys

Jolanta Stankiewicz* and Juan Bartolomé

Instituto de Ciencia de Materiales de Aragón, Consejo Superior de Investigaciones Científicas and Universidad de Zaragoza, 50009-Zaragoza, Spain

Daniel Fruchart

Laboratoire de Cristallographie, CNRS, F-38042 Grenoble 09, France

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Anomalous behavior of the resistivity at or just below the Néel temperature in antiferromagnetic metals is usually attributed to the formation of superzone gaps. However, we find that $R\mathrm{Mn}_{12-x}\mathrm{Fe}_x$ alloys which have no such gaps exhibit a similarly anomalous resistivity. We show that electron scattering by substitutional *spin* disorder can account for such behavior of itinerant magnets. This mechanism, which has not been studied before, leads to a relaxation rate that is proportional to $x(12-x)m^2$, where m is the staggered magnetization. Together with spin fluctuations, phonon, and impurity scattering, it accounts well for the resistivity data we obtain for $\mathrm{HoMn}_{12-x}\mathrm{Fe}_x$, for $0 \leq x \leq 9$, in the temperature range of 4 to 400 K.

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Substitutional disorder in magnetic $A_{1-x}B_x$ alloys gives rise to a magnetic resistivity which has sometimes been found to follow Nordheim's rule $\rho_{\mathrm{mag}} \propto x(1-x)$ [1]. There are various explanations for this behavior. The scattering potential associated with alloy disorder can affect the electron scattering rates [2]. When only *s-s* scattering is important, Nordheim's formula obtains. Calculation of electron-magnon scattering in dilute ferromagnetic alloys [3] as well as an extension of the theory of de Gennes and Friedel [4] for binary alloys [5] yield the same expression. Magnetic order can also modify electronic states and, consequently, affect nonmagnetic relaxation processes [6], again leading to a Nordheim's like rule for impurity-induced magnetic scattering.

In addition, the resistivity of antiferromagnetic metallic alloys often shows, upon cooling, an upturn at or just below the Néel temperature T_N. Carrier scattering by critical spin fluctuations near T_N can lead to such anomalies [7]. However, this behavior is usually attributed to the formation of magnetic superzone gaps in the ordered phase [8,9]. These gaps reduce the effective number of conduction electrons below T_N giving rise to the observed resistivity upturn [10]. On this basis we have explained the anomalous temperature variation of the resistivity of some $R\mathrm{Mn}_{12-x}\mathrm{Fe}_x$ alloys studied earlier [11]. However, it has recently been found that the antiferromagnetic order in $R\mathrm{Mn}_{12-x}\mathrm{Fe}_x$ alloys has the same symmetry as the crystalline lattice [12]. Consequently, there are no superzone gaps and some different mechanism must be responsible for the anomalous resistivity in these alloys.

In this Letter, we report experimental results of the electrical resistivity of the complete $\mathrm{HoMn}_{12-x}\mathrm{Fe}_x$ series ($0 \leq x \leq 9$). We find that the resistivity anomaly at T_N varies with the composition. At low temperature the resistivity follows the $x(12-x)$ dependence, and, owing to disorder, its value can be quite high. We also find that, in contradiction with existing theory, the *magnetic* resistivity does not vanish as $T \rightarrow 0$. We show below that electrons at low temperatures can be effectively scattered by magnetic disorder arising from the random substitution of Mn by Fe atoms. Electron scattering by substitutional *spin* disorder itself has not, as far as we know, been studied. This mechanism, which leads to Nordheim's like relation, can be important in disordered magnetic alloys. Our simple theoretical model seems to provide a satisfactory account of the temperature and composition dependence of the measured resistivity in $\mathrm{HoMn}_{12-x}\mathrm{Fe}_x$. The itinerant character of the magnetic *3d* ions is also important in our resistivity calculations.

$R\mathrm{Mn}_{12-x}\mathrm{Fe}_x$ (R = lanthanide series) alloys crystallize in a body-centered tetragonal ThMn_{12} type structure which has one R site and three nonequivalent Mn sites, each occupied by four transition-metal ions. They can be obtained for x up to nine with R = Er and Ho. A strong antiferromagnetic coupling between *3d* ions leads to a noncollinear antiparallel arrangement of magnetic moments in the basal plane, with a [001] propagation vector [12,13]. In addition, *3d* ions in Fe-rich alloys contribute to the *c*-axis ferromagnetic moment. Thus, the magnetic structure of $R\mathrm{Mn}_{12-x}\mathrm{Fe}_x$ varies strongly with the nature of the *3d*-ions sublattice, from antiferromagnetic ordering in the basal plane for manganese-rich compounds to an almost ferromagnetic ordering along the *c* axis for iron-rich systems.

The cold crucible method was used to synthesize the $\mathrm{HoMn}_{12-x}\mathrm{Fe}_x$ alloys we studied. The ingots obtained were checked by x-ray diffraction. In samples with $x = 0$, small amounts (less than 5%) of $\mathrm{Ho}_6\mathrm{Mn}_{23}$ were detected. The electrical resistivity measurements were performed with a six probe method on bar-shaped polycrystalline samples in the temperature range from 4 to 400 K. We have applied a magnetic field of 6 and 12 T in some of these

measurements. The mean relative error obtained for resistivity measurements is approximately 0.1%; absolute values were determined to within 5%.

How the resistivity, ρ, of some of the $HoMn_{12-x}Fe_x$ samples we studied varies with temperature is shown in Fig. 1. At low temperatures, the measured resistivity is rather constant. It increases with increasing temperature up to a maximum, located within the 100–200 K range, for alloys which order antiferromagnetically ($x < 9$). ρ decreases thereafter up to the Néel temperature T_N, and increases nearly linearly with temperature beyond T_N. On the other hand, the resistivity of $x = 0$ alloys which order at $T_N = 90$ K shows no anomaly. For $x = 9$, the system is ferromagnetic in the range of temperature we have studied, and ρ increases monotically with T. We did not observe any change in $\rho(T)$ for $x = 5$ and 6 alloys in magnetic fields of 6 and 12 T. Such behavior confirms that the resistivity anomalies in $HoMn_{12-x}Fe_x$ are unrelated to magnetic superzones.

Before we fit our calculations to the resistivity data, we note that: (i) $\rho(4\text{ K})$ is large; (ii) the variation of ρ with T is within 10% of $\rho(4\text{ K})$ for most disordered ($4 \leq x \leq 8$) alloys. Therefore, appreciable localization effects are expected in these compounds. Accordingly, we include quantum corrections to the calculated Boltzmann conductivity

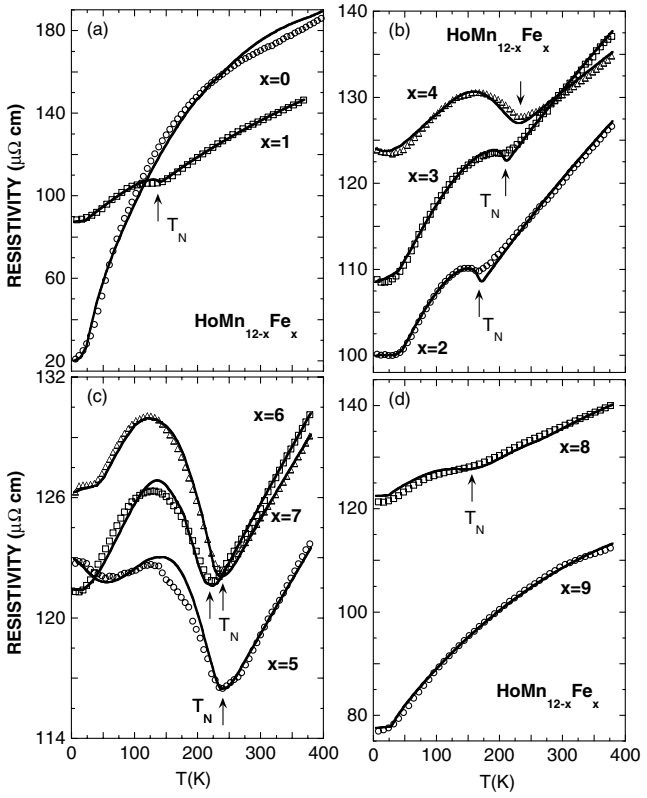

FIG. 1. Resistivity data points versus temperature for $HoMn_{12-x}Fe_x$ alloys. Only a small number of the data points are shown for the sake of clarity. The solid lines show numerical fittings to the data points.

$\sigma_B(T)$, which, following Kaveh and Mott [14], for the three-dimensional case, is

$$\sigma(T) = \sigma_B(0)\left[1 - \frac{3}{(k_F l_e)^2}\right] + \frac{e^2}{\pi^2\hbar}\left[\frac{L_{\rm in}(T) + l_{\rm in}(T)}{L_{\rm in}(T)[l_e + l_{\rm in}(T)]} - \frac{(k_F l_e)^2}{3[l_e + l_{\rm in}(T)]}\right], \quad (1)$$

where l_e ($l_{\rm in}$) is the elastic (inelastic) mean free path and $L_{\rm in}$ the inelastic diffusion length, defined by $L_{\rm in} = (\frac{1}{2} l_e l_{\rm in})^{1/2}$, $\sigma_B(0) = e^2 k_F^2 l_e / 3\pi^2\hbar$, and k_F is the Fermi wave vector. The first term in Eq. (1) is the zero-temperature, localization-corrected conductivity $\sigma(0)$. The second term represents competition between the quantum interference effects (positive contribution) and their degradation by inelastic scattering processes, which are thermally excited. These corrections account for no more than 3% of $\sigma_B(T)$. However, we use Eq. (1) in our calculations for $\rho(T)$ since the variation of the resistivity in some ($x \simeq 6$) alloys studied is of this order.

We assume that the total resistivity is a sum of the temperature-independent residual resistivity $\rho_{\rm res}$ ($\propto l_e^{-1}$), the phonon resistivity $\rho_{\rm ph}$, and the magnetic resistivity $\rho_{\rm mag}$. The phonon contribution is given by $1/l_{\rm in}^{\rm ph} = \beta k_F^2 T G(\theta_D/T)$, where $G(\theta_D/T)$ is the Bloch-Grüneisen function and θ_D is the Debye temperature. We use a $\theta_D = 300$ K for all compositions [15]. $\rho_{\rm ph}$ has no adjustable parameter since β can be obtained from the slope of ρ at high temperatures. Assuming that the effective number of carriers is constant, we can estimate the magnetic part of ρ. The upper panel of Fig. 2 shows the magnetic resistivity $\rho'_{\rm mag}$ versus temperature, obtained by subtracting out the phonon and an impurity contribution from the total resistivity. At low temperatures, $\rho'_{\rm mag}$ vanishes as $T \to 0$ for small and large (ferromagnetic limit) x values, respectively, as expected for scattering by thermal fluctuations in spin systems. However, this does not occur for the $x = 6$ and $x = 7$ alloy, shown in Fig. 2, nor for some other compositions. In addition, $\rho'_{\rm mag}$ decreases quite rapidly as the critical temperature is approached from below; beyond T_N, it increases almost linearly with temperature. Such behavior is quite different from the predictions of classical spin-disorder scattering models [4,16].

We first address the observation that the magnetic resistivity does not vanish as $T \to 0$. We consider spin-disorder scattering in a binary alloy, with two magnetic species, A and B. The conduction s electrons are exchange coupled to $3d$ moments, which are antiferromagnetically aligned and, for simplicity, we assume to be localized. Accordingly, we make use of the exchange Hamiltonian $H_{sd} = \sum_i J_i(\mathbf{R_i} - \mathbf{r})\mathbf{S_i}\cdot\mathbf{s}$, where J_i and S_i vary with site. Here, J_i and S_i are the exchange constant and spin of the $3d$ moment in the ith unit cell, respectively. Only terms with S^α contribute to elastic scattering of electrons at low temperatures if the system is magnetically ordered in the α

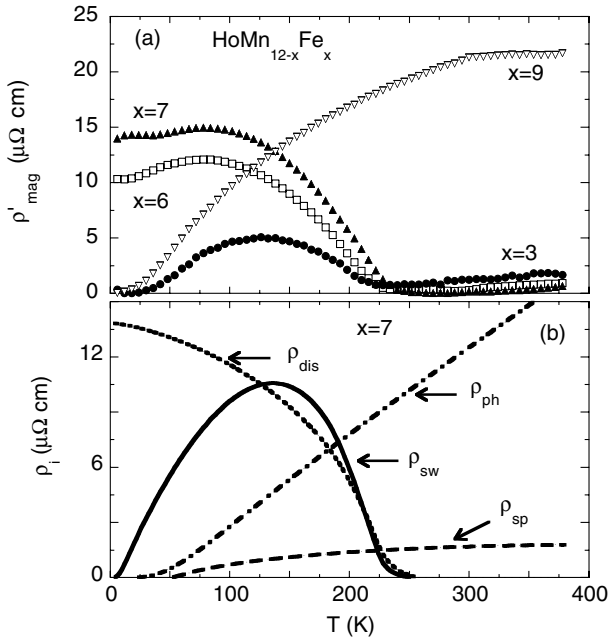

FIG. 2. (a) Magnetic contribution to the resistivity of $HoMn_{12-x}Fe_x$ alloys obtained by subtracting out the phonon and some constant residual term from the total resistivity. (b) Temperature variations of phonon ($\rho_{\rm ph}$), spin wave ($\rho_{\rm sw}$), single-particle ($\rho_{\rm sp}$), and magnetic disorder ($\rho_{\rm dis}$) contributions to the total resistivity of $HoMn_5Fe_7$ alloy.

direction. To find the transition rate from substitutional spin-disorder (SSD) scattering, we replace S_i^α in H_{sd} by $m_i + \delta S_i^\alpha$, where $m_i = \pm m$ and $m = (1-x)m_A + xm_B$ is the mean value of the staggered magnetization. Here, $m_A = |\langle S_A^\alpha \rangle|$ and $m_B = |\langle S_B^\alpha \rangle|$ are magnetic moments at site A and B, respectively. We assume $J_A \approx J_B$. From the Boltzmann equation, one then finds for the transition rate $W(\mathbf{k}\uparrow \rightarrow \mathbf{k}'\uparrow)$:

$$W(\Delta\mathbf{k}) \propto N^2 m^2 \delta_{\Delta\mathbf{k},\mathbf{0}} + N\sum_{ij} \delta S_i^\alpha (\delta S_j^\alpha)^* e^{i\Delta\mathbf{k}\cdot(\mathbf{R_i}-\mathbf{R_j})} + 2Re\left[m\sum_{ij} (\delta S_i^\alpha)^* e^{i\Delta\mathbf{k}\cdot(\mathbf{R_i}-\mathbf{R_j})} \right], \quad (2)$$

where $\Delta\mathbf{k} = \mathbf{k} - \mathbf{k}'$. The first term gives no contribution to the scattering rate. The third term vanishes upon summing over i. In the second term, only terms with $i = j$ give nonzero contributions to W, while all other terms average to zero for a random alloy. In this contribution to W, the wave vector is not conserved. It then follows, after some straightforward algebra, that

$$1/l_e^{\rm dis} \propto W(\Delta\mathbf{k}) \propto x(1-x)(m_A - m_B)^2 = x(1-x)m^2(S_A - S_B)^2/[(1-x)S_A + xS_B]^2, \quad (3)$$

where $l_e^{\rm dis}$ is the elastic mean free path limited by SSD and m is the staggered $3d$-ions magnetization. Thus, $1/l_e = 1/l_e^{\rm dis} + 1/l_e^{\rm res}$ where the $l_e^{\rm res}$ arises from impurity scattering. For S_A not too different from S_B, $\rho_{\rm dis} \propto x(1-x)m^2$, where $\rho_{\rm dis}$ is the resistivity arising from substitutional spin disorder. The same result, where m is equal to the total magnetization, holds for ferromagnetic alloys .

We treated the amplitude γ of the SSD scattering ($1/l_e^{\rm dis} = \gamma m^2$) as an adjustable parameter in our initial fitting procedure to the resistivity data. We used in our calculations $3d$-electron magnetic-moment data, obtained from neutron-scattering experiments [12,13]. When these were not available, an approximate relation $m(T) \propto (T_N^{3/2} - T^{3/2})^{1/2}$ has been assumed [17]. We found that γ follows the Nordheim's rule as predicted by Eq. (3). This is shown in the upper panel of Fig. 3. Consequently, in our definitive fittings we have $\gamma = 3.25 \times 10^{-4} x(12-x)$ Å^{-1}.

Next, we consider thermal fluctuations in a spin system and their effect on resistivity. Usually, when magnetic moments are localized, the thermal spin-disorder contribution is approximated by $1/l_{\rm in}^{\rm mag} \propto (1 - m^2)$ [4,16]. However, the magnetic resistivity in the system we studied depends on temperature quite differently, as shown in Fig. 2. Substitutional spin disorder does lead to the observed behavior. Nevertheless, to improve our fittings, we also consider additional inelastic scattering processes in itinerant d-electron system. We use here the results of self-consistent renormalization theory of spin fluctuations in a two-band model in which only s electrons contribute to the conductivity and d electrons are scattering centers

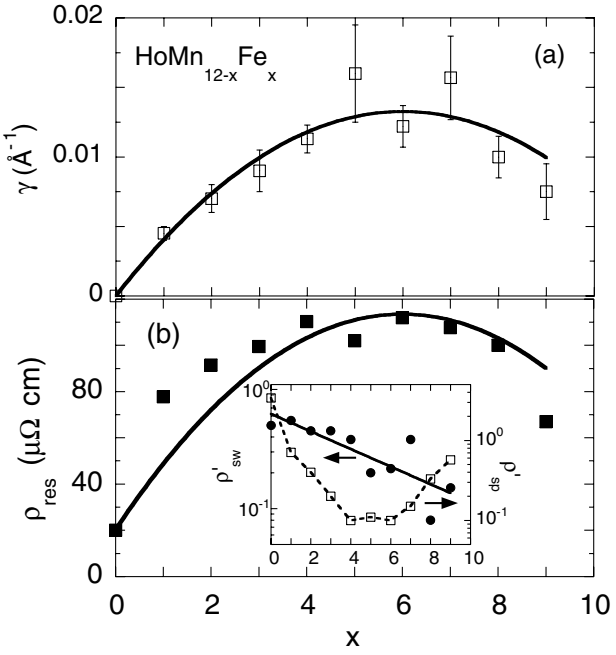

FIG. 3. (a) The values of γ, the amplitude of substitutional magnetic disorder scattering, and (b) of $\rho_{\rm res}$ as a function of composition. Solid lines show $x(12-x)$ dependence. The inset shows values of $\rho'_{\rm sw} = m^{-3}\rho_{\rm sw}/T$ and of $\rho'_{\rm sp} = \rho_{sp}/T$ in units of $\mu\Omega$ cm K^{-1} as a function of x. The solid and dashed lines are a guide to the eyes.

[17,18]. There are two different elementary excitations associated with transverse magnetization fluctuations, namely, Stoner single-particle excitations and spin waves. The spin wave contribution to the resistivity increases with decreasing magnetization at low temperatures. However, when the spin wave energy becomes comparable to the energy of Stoner excitations at some cutoff wave vector, spin waves are damped. Their contribution to the resistivity becomes then less important since it decreases as m^3. On the other hand, the scattering from single-particle excitations, which is negligible at low temperatures, becomes more important as the temperature increases. Above T_N, the resistivity arising from this mechanism is almost linear in temperature and shows tendency to saturate. In our calculations we use the simplified expressions [19] for the magnetic resistivity given in Ref. [18]. The amplitudes of spin wave and single-particle scattering as well as the cutoff wave vector are adjustable parameters. We found that our fits are not very sensitive to the magnitude of the cutoff wave vector. A value of 20–50 K is used for the effective temperature at which spin waves start to be damped. Therefore, three adjustable parameters are left in our fittings: two amplitudes for magnetic scatterings (spin wave and single-particle scattering) and the value of k_F. The latter turns out to be quite close to 1 Å^{-1} in all alloys studied.

Assuming that the parameters we look for are isotropic for polycrystalline $HoMn_{12-x}Fe_x$ samples, the total resistivity is then given by $(2\rho_{xx} + \rho_{zz})/3$, where ρ_{xx} and ρ_{zz} are different owing to magnetic scattering. SSD contributes to the resistivity only when there is a nonvanishing magnetic moment along the corresponding axis. For instance, ρ_{dis} is important in all resistivity components of $HoMn_6Fe_6$ since it orders antiferromagnetically in the basal plane below 220 K and ferromagnetically along c axis below 80 K. The results of our least square fittings are shown in Fig. 1 as solid lines. The overall agreement between the calculated and measured resistivities is very satisfactory. Our fits reproduce all important features of the experimental curves. We find that the magnetic scattering contributions to the total resistivity are much smaller than the temperature-independent residual resistivity ρ_{res} in all but the $x = 0$ alloy. [The latter is given by $\rho_{\text{res}} = \rho(T \rightarrow 0) - \rho_{\text{dis}}(T = 0)$.] How they vary with temperature is shown in the lower panel of Fig. 2 for a $x = 7$ alloy. The x variation of the spin wave and Stoner scattering amplitudes is exhibited in the inset of Fig. 3. The change of ρ_{res} with composition is also shown in Fig. 3. It follows quite closely a $x(12 - x)$ dependence as expected for binary alloys with substitutional atomic disorder [20].

Finally, we note that the low-temperature resistivity of $HoMn_{12-x}Fe_x$ alloys exhibits some features related to the ferromagnetic ordering of rare earth ions. These are, however, much weaker than the ones arising from antiferromagnetic ordering of transition-metal moments.

In conclusion, we have found anomalies in the magnetic resistivity of $HoMn_{12-x}Fe_x$ antiferromagnetic compounds. In particular, ρ_{mag} does not vanish as $T \rightarrow 0$, and exhibits, upon cooling, an upturn at or just below T_N. We have also shown that electron scattering by spin disorder, arising from random substitution of Mn by Fe atoms, leads to the observed anomalies. This mechanism, in addition to phonon and impurity scattering, accounts for main resistivity features in the $x \neq 0$ alloys. In order to improve the fits to our experimental data we have also included scattering by thermal spin disorder. Equally satisfactory fits are obtained for other $RMn_{12-x}Fe_x$ (R = Y, Er) alloys [21]. SSD scattering can be as well important in colossal magnetoresistance manganites which are often substitutional compounds or in ferromagnetic metallic alloys.

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*Electronic address: jolanta@posta.unizar.es

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