Ab initio calculation of the anisotropic magnetoresistance in $Ni_{1-c}Fe_c$ bulk alloys

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(Received 21 January 2003; published 2 July 2003)

By using the Kubo-Greenwood formula in combination with the fully relativistic spin-polarized Screened Korringa-Kohn-Rostoker method and the Coherent Potential Approximation we calculated the residual resistivity and the anisotropic magnetoresistance of bulk $Ni_{1-c}Fe_c$ alloys in the Ni-rich regime. While the calculated residual resistivities are typically 30–40% smaller than the measured values, for the anisotropic magnetoresistance ratios we obtained an excellent agreement between theory and experiment. Varying the angle between the directions of the magnetization and of the current we found a functional dependence of the resistivity consistent with the formula proposed originally by Döring.

DOI: 10.1103/PhysRevB.68.012402 PACS number(s): 75.30.Gw, 75.70.Rf, 75.70.Ak, 75.70.Cn

INTRODUCTION

Ab initio investigations of electric transport in solids attracted considerable interest in the last decades (see, e.g., Ref. 1). This interest was stimulated, on the one hand, by a spectacular progress in the field of *ab initio* band structure calculations, and, on the other hand, by a growing demand of experimental physics and technology, in particular, by intensive studies and applications of various complex devices which utilize the anisotropy of the resistivity in magnetically ordered alloys and heterostructures. Since in complex artificial structures (like spin valves) there are several possible contributions to the resistivity and its dependence on the direction of the current or the external magnetic field (e.g., random impurities, interfaces and their roughness, phonon scattering, etc.), it is often difficult to estimate theoretically their relative contributions. An accurate account of the residual resistivities and anisotropic (or spontaneous) magnetoresistance (AMR) ratios of the random magnetic alloys is, therefore, a rather promising, though still challenging task for *ab initio* theories.

In this paper we perform an *ab initio* study of the residual resistivity and the AMR of Ni-rich $Ni_{1-c}Fe_c$ bulk alloys in terms of the Kubo-Greenwood formula[2] of quantum linear response theory. Among the compounds showing high AMR, because of their low coercivity and high magnetic moment, $Ni_{1-c}Fe_c$ alloys are perhaps most commonly used in technological applications. Due to this fact, for these systems a large amount of resistivity data from high quality measurements is available in the literature.[3–7] After describing the computational method we used, we present and discuss our results focusing, in particular, on the concentration dependence of the residual resistivities and AMR ratios. In addition to experiments, we compare with the earlier theoretical work of Banhart and Ebert on the same system.[8] These authors also pointed out[9,10] the importance of spin-orbit coupling for the residual resistivities in magnetic binary substitutional alloys. In addition to AMR investigations, we furthermore study the dependence of the resistivity with respect to the angle between the magnetization and the current and successfully connect the obtained results to the general formulation of Döring.[11]

METHOD OF THE CALCULATIONS

In the present calculations of the electric conductivity we used the self-consistent potentials and effective fields from our previous work[12] in terms of the fully relativistic, spin-polarized screened Korringa-Kohn-Rostoker (KKR) method for layered systems[13,14] as combined with the single-site coherent potential approximation (CPA) to account for substitutional disorder.[15] Note that the experimental lattice constants of the fcc $Ni_{1-c}Fe_c$ alloys ($0<c<0.5$) were used. The conductivity for a disordered layered system can be written as[15]

$$\sigma_{\mu\mu}(n;c;\hat{\mathbf{M}}) = \sum_{p,q=1}^{n} \sigma_{\mu\mu}^{pq}(c;\hat{\mathbf{M}}), \tag{1}$$

where n is the number of layers considered, $\mu \in \{x,y,z\}$, c denotes the concentration of one of the constituents of a given binary alloy, and $\hat{\mathbf{M}}$ the direction of the magnetization, both of which are assumed to be uniform in all the layers of the bulk alloy. According to the Kubo-Greenwood formula,[2,15] the non-local conductivity between layers p and q, $\sigma_{\mu\mu}^{pq}$, can be calculated as

$$\sigma_{\mu\mu}^{pq} = \frac{\hbar}{\pi N_0 V_{at}} \mathrm{Tr}\langle J_\mu^p \, G^+(E_F) \, J_\mu^q \, G^+(E_F)\rangle. \tag{2}$$

Here N_0 is the total number of atoms per plane, V_{at} is the atomic volume, the brackets label an average over possible configurations of constituents A and B, J_μ^p stands for the μth component of the current operator with reference to the pth plane, and $G^+(E_F)$ is the (retarded) one-particle propagator at the Fermi energy, E_F. The corresponding resistivity is then defined by

$$\rho_{\mu\mu}(n;c;\hat{\mathbf{M}}) = 1/\sigma_{\mu\mu}(n;c;\hat{\mathbf{M}}). \tag{3}$$

Note, that the above formulation is, in general, valid only for the current-in-plane geometry (i.e., for $\mu \in \{x,y\}$). Since,

however, in the present study bulk systems are represented by a sequence of identical layers,

$$\mathrm{Ni}_{1-c}\mathrm{Fe}_c(001)/(\mathrm{Ni}_{1-c}\mathrm{Fe}_c)_n/\mathrm{Ni}_{1-c}\mathrm{Fe}_c(001), \qquad (4)$$

namely, n monolayers of permalloy capped from both sides by semi-infinite leads of the same material, translational symmetry of the electric fields and currents is retained in the direction normal to the planes and, therefore, Eqs. (1) and (3) also apply in the case of $\mu = z$.

Clearly, the calculated conductivity (and/or resistivity) of such layered systems converges to the bulk value in the limit of $n \to \infty$. The numerical procedure of performing this limit for the resistivity and the overall stability of the method was discussed in length in our previous work[16] and does not need be repeated here. As compared to the value of $n = 45$ taken in Ref. 16, in the present calculations we used a larger number of layers, namely, $n = 60$ that allowed us to perform a more stable fit for the resistivity of the bulk system.

All scattering channels up to and including a maximal angular moment quantum of two were taken into account. When performing the configurational average within the CPA [see Eq. (2)], no vertex corrections were taken into account.[17,15] The electrical conductivity was calculated using 3160 $k_\parallel$ points in the irreducible wedge of the surface Brillouin zone.[15] For some concentrations the stability of the obtained results was checked by increasing the number of $k_\parallel$ points up to 4950. In fact, we found that the two-dimensional Brillouin zone summations converge faster for $\rho_{zz}(n;c;\hat{\mathbf{M}})$ than for $\rho_{xx}(n;c;\hat{\mathbf{M}})$, therefore, all the results presented in this work refer to a current flowing normal to the planes (z), while we varied the orientation of the magnetization, $\hat{\mathbf{M}}$, with respect to this direction.[18]

Because of computational reasons a finite imaginary part, δ, of the Fermi energy has to be used in the calculation of conductivity.[16] The actual ''bulk'' resistivity is defined, therefore, as the following double limit:

$$\rho_{\mu\mu}(c;\hat{\mathbf{M}}) = \lim_{\delta \to 0} \lim_{n \to \infty} \rho^{cal.}_{\mu\mu}(n;c;\hat{\mathbf{M}};\delta). \qquad (5)$$

In Ref. 16 it was argued that for large enough n the slope of $n\rho^{cal.}_{\mu\mu}(n;c;\hat{\mathbf{M}};\delta)$ behaves linear in δ. This observation greatly simplifies taking the $\delta \to 0$ limit in Eq. (5). In Ref. 16 the lowest value of $\delta = 2$ mRy produced a resistivity still 3–4 times larger than expected in the limit of $\delta \to 0$, leading, therefore, to some uncertaintity in determining the bulk resistivity. In the present work we used much smaller values of δ ($\delta_{\min} = 0.1$ mRy), providing thus a more careful justification of the proposed numerical procedure. Figure 1 shows the calculated resistivities, $\rho^{cal.}_{zz}(c;\hat{\mathbf{M}};\delta)$, of $\mathrm{Ni}_{80}\mathrm{Fe}_{20}$ for $\delta = 0.1$, 0.25, 0.5, and 1 mRy and for two different directions of the magnetization ($\hat{\mathbf{M}} = \hat{z}$ and $\hat{\mathbf{M}} = \hat{x}$), together with a linear least square fit to the data. The estimated relative error of the residual resistivity turned to be about 1%. This accuracy of the fitting procedure applied in the entire concentration range, $0 < c < 0.5$.

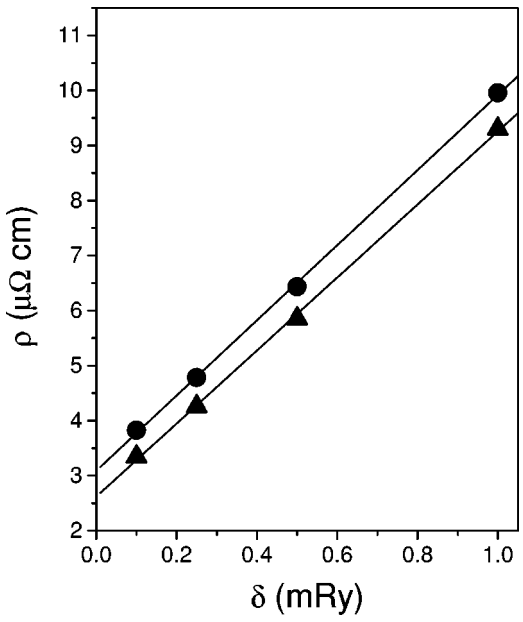

FIG. 1. Calculated resistivities, $\rho_{zz}(\hat{\mathbf{M}};\delta)$, of the $\mathrm{Ni}_{80}\mathrm{Fe}_{20}$ alloy with various choices of the imaginary part of the Fermi energy, δ. Circles and triangles refer to the cases when the current is perpendicular or parallel to the direction of the magnetization, $\hat{\mathbf{M}} = \hat{x}$ and $\hat{\mathbf{M}} = \hat{z}$, respectively. The solid lines stand for a least square fit to the data. The residual resistivity is provided by the interception of the lines with the ordinate axis.

RESULTS AND DISCUSSION

We adopted the commonly used definition[8,19] for the AMR ratio of bulk alloys,

$$\frac{\Delta\rho(c)}{\rho_{av}(c)} = \frac{\rho_\parallel(c) - \rho_\perp(c)}{\rho_{av}(c)}, \qquad (6)$$

with

$$\rho_{av}(c) = 1/3[\rho_\parallel(c) + 2\rho_\perp(c)], \quad \rho_\parallel(c) = \rho_{zz}(c;\hat{z}),$$
$$\rho_\perp(c) = \rho_{zz}(c;\hat{x}). \qquad (7)$$

Experimentally the above quantities are defined as an extrapolation of the measured results to zero applied magnetic field.

In Fig. 2 the calculated bulk resistivities, $\rho_\parallel$, $\rho_\perp$, and ρ_{av} of the $\mathrm{Ni}_{1-c}\mathrm{Fe}_c$ alloys are displayed in the concentration range, $0 < c < 0.5$. In full agreement with experiments, for all concentrations the resistivity for the current parallel to the field is found to be larger than the perpendicular one, indicating that the AMR ratio defined by Eq. (6) is always positive. The shape of the curve $\rho_{av}(c)$ compares well to the experimental observations: for small concentrations it rapidly increases and reaches a flat minimum at about $c = 0.25$. The calculated magnitudes of the averaged residual resistivity ρ_{av} are significantly larger for $c < 0.1$ and by about 30–40 % lower for $c > 0.1$ than the measured data. Similar observations were also made in the *ab initio* calculations by Banhart and Ebert,[8] with the exception that in the concentra-

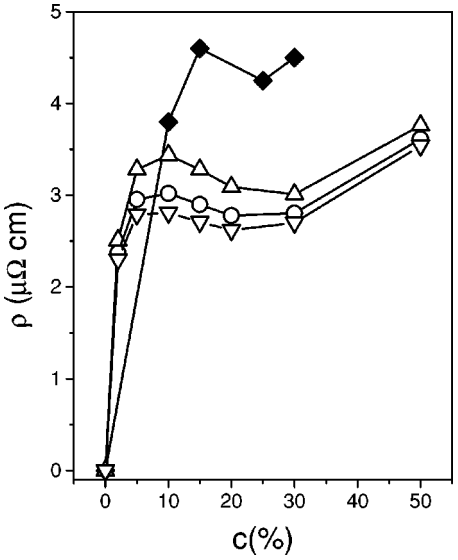

FIG. 2. Calculated (open symbols) and experimental (Refs. 3 and 4) (diamonds) residual resistivities of $Ni_{1-c}Fe_c$ alloys with respect to the concentration, c. For the definitions of $\rho_{\parallel}$ (up triangles), $\rho_{\perp}$ (down triangles), and ρ_{av} (circles); see expressions (7) in the text.

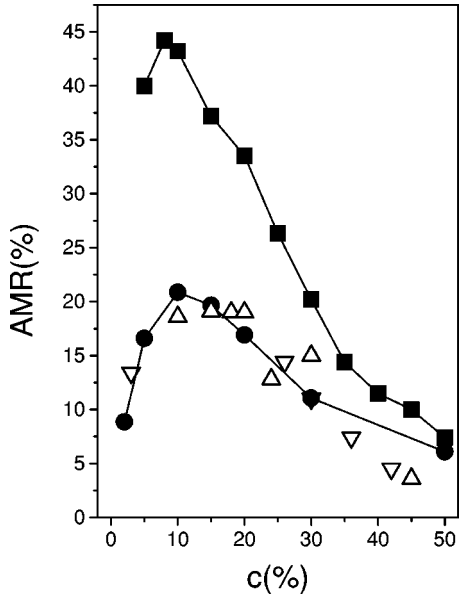

FIG. 3. Calculated and experimental AMR ratios of $Ni_{1-c}Fe_c$ alloys. Full circles: present work, full squares: calculations of Ref. 8, up-triangles: experiment (Ref. 3), down triangles: experiment (Refs. 5 and 6). The solid lines serve as a guide for eyes.

tion range $0<c<0.1$ they found a rather moderate increase of the resistivity. This difference between the two theoretical results can be fairly well understood, as in Ref. 8 vertex corrections were taken into account, which, in particular, for small concentrations (weak disorder) should considerably lower the resistivity. The systematic error of about -30–-40 % of the calculated resistivities with respect to the experimental data can be partially attributed to additional scattering mechanisms, such as grain boundaries, short-range order, etc., not taken into account in the *ab initio* calculations, giving rise, however, to an additional resistivity contribution. Clearly enough, missing correlations in the local density functional approximation in particular for the Ni constituent may add to the discrepancy between the measured and the calculated averaged residual resistivity. As presently no *ab initio* method is available that takes into account these correlations in the case of transport properties, it is very questionable to estimate their importance with respect to the above mentioned imperfections.

As can be seen from Fig. 3, both the functional shape and the magnitude of our calculated concentration dependent AMR ratios are in excellent agreement with the experimental data. In satisfactory agreement with experiments and the present calculations, the AMR ratio communicated in Ref. 8 shows a maximum at about $c=0.1$ and a steady decrease for larger concentrations; however, in particular, for small concentrations its magnitude is largely overestimated. Supposing that excess scattering effects give rise to an isotropic resistivity contribution, in that work the AMR ratios were corrected by taking the measured ρ_{av}, keeping, however, the calculated $\Delta\rho$ in Eq. (6). Although, the overall agreement of the AMR curve improved as compared to experiments, for $c<0.1$, the corrected AMR ratios were still too high by a factor of about two.[8]

The good quantitative description of the AMR of the $Ni_{1-c}Fe_c$ alloys provided by our theoretical approach indicates that the effects not considered in the calculations do contribute to the average resistivity as well as to $\rho_{\parallel}$ and $\rho_{\perp}$ in equal terms and, therefore, the AMR ratio can safely be calculated by neglecting them. In fact, random structural imperfections (grain boundaries or clusters) are not expected to give an anisotropic contribution to the resistivity.[8] Chemical fluctuations in the system (short-range order), however, and correlation effects do change the electronic structure without destroying the ''global'' cubic symmetry, which, in combination with spin-orbit coupling, is responsible for the observed anisotropic magnetoresistance. As the AMR is only one particular transport property, one cannot rule out the importance of the latter effects as in the case of structural imperfections.

By varying the direction of the magnetization, the dependence of the resistivity on the angle between the directions of the current and the magnetization can be studied. During the third decade of the last century Döring[11] put forward a general expression which describes the anisotropy of the resistivity in cubic crystals with respect to the direction of the magnetization and of the current relative to the crystallographic axes. In the special case, when the direction of the current is fixed along a certain crystallographic axis and the direction of the magnetization is varied between this and another crystallographic axis, the Döring expression reduces to

$$\rho(\vartheta)=\rho_0+B\cos^2\vartheta+C\cos^4\vartheta, \tag{8}$$

where ϑ is the angle between the magnetization and the current.

In Fig. 4 we present the results obtained for $Ni_{80}Fe_{20}$ and $Ni_{85}Fe_{15}$ alloys. In these calculations we fixed the current along the (001) direction of the fcc crystal and rotated the

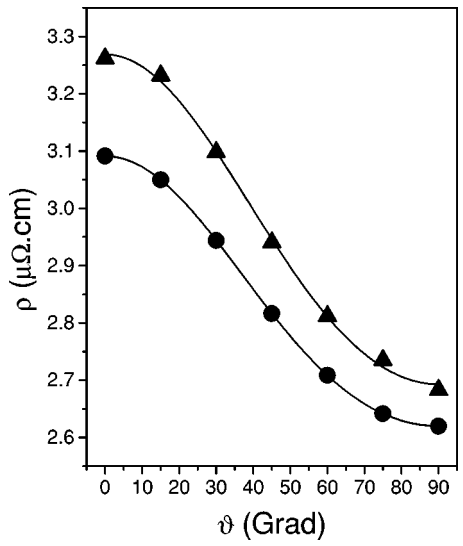

FIG. 4. Calculated resistivities with respect to the the angle, ϑ, between the current and the magnetization for $Ni_{85}Fe_{15}$ (triangles) and $Ni_{80}Fe_{20}$ (circles) alloys. Solid lines visualize the results of least square fits according to Eq. (7); see the text.

magnetization from the (001) to the (110) direction within the $(1\bar{1}0)$ plane. Note that the cases $\vartheta=0$ and $\vartheta=\pi/2$ correspond to $\rho_{\parallel}$ and $\rho_{\perp}$, respectively. As inferred from Fig. 4, the calculated results almost perfectly fit the functional dependence given in Eq. (8). Looking at the fitting parameters listed in Table I, it should be noted that even the $\cos^4\vartheta$ term has a non-negligible weight which cannot be omitted in the fitting procedure without a drastic loss in the overall quality of the fit.

CONCLUSIONS

In summary, by using the Kubo-Greenwood formula within the fully relativistic spin-polarized Screened KKR-CPA method for disordered layered system we performed *ab initio* calculations of the residual resistivities and anisotropic magnetoresistance ratios of bulk fcc $Ni_{1-c}Fe_c$ alloys in the Ni-rich regime. We obtained resistivities in satisfactory agreement with experiments. The differences relative to the measured data most likely have to be attributed to the missing vertex corrections within the single-site CPA and/or to additional scattering effects due to imperfections present in the experimental samples. Quite surprisingly, practically in the entire concentration range under consideration, the calculated AMR ratios were found in excellent quantitative agreement with the measurements, indicating that an accurate computational scheme, which includes spin polarization and relativity on the same level, can indeed account for magnetoresistive effects of alloys with high precision. In addition to the AMR ratios, for two permalloy systems, namely, for $Ni_{80}Fe_{20}$ and $Ni_{85}Fe_{15}$, we calculated the dependence of the resistivity on the angle between the current and the magnetization. The results fit well the general phenomenological expression given by Döring for the resistivity of saturated ferromagnetic cubic crystals.

TABLE I. Parameters (in units of $\mu\Omega$ cm) of the fit of the data presented in Fig. 4 to the function Eq. (7).

	ρ_0	B	C
$Ni_{85}Fe_{15}$	2.693	0.437	0.138
$Ni_{80}Fe_{20}$	2.620	0.315	0.156

ACKNOWLEDGMENTS

The authors gratefully acknowledge very encouraging discussions with Professor P. M. Levy. This paper resulted from a collaboration partially funded by the RTN network ''Computational Magnetoelectronics'' (Contract No. HPRN-CT-2000-00143) and by the Research and Technological Cooperation Project between Austria and Hungary (Contract No. A-23/01). Financial support was also provided by the Center for Computational Materials Science (Contract No. GZ 45.451), the Austrian Science Foundation (Contract No. W004), and the Hungarian National Scientific Research Foundation (OTKA T038162 and OTKA T037856).

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