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Transport spin polarization of Ni_xFe_{1-x}: Electronic kinematics and band structure

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We present measurements of the transport spin polarization of Ni_xFe_{1-x} $(0 \leqslant x \leqslant 1)$ using the recently developed point-contact Andreev reflection technique, and compare them with our first-principles calculations of the spin polarization for this system. Surprisingly, the measured spin polarization is almost composition independent. The results clearly demonstrate that the sign of the transport spin polarization does not coincide with that of the difference of the densities of states at the Fermi level. Calculations indicate that the independence of the spin polarization of the composition is due to compensation of density of states and Fermi velocity in the *s* and *d* bands.

Spin-polarized transport in magnetic materials is beginning to play an increasingly important role in fundamental and applied research due to the rapid advance of magnetoelectronics.[1] The very definition of this new field, based on the ability of magnetic metals to carry spin-dependent current, implies that many physical phenomena and device applications are determined by the interplay of magnetic and transport properties of these materials. Although many materials are spin polarized, technical constraints limit the number actually used in practice to only a handful. In particular, Permalloy, a member of a family of binary alloys, Ni_xFe_{1-x} $(x=0.8)$, features an attractive combination of vanishingly small magnetostriction, low coercivity, and high permeability, which makes it the material of choice for magnetic recording media, sensors, and nonvolatile magnetic random access memory.

Impressive progress in understanding magnetic properties of $3d$-transition-metal ferromagnets has been made in the last decades, particularly due to the advances of the band-structure calculations, based on the local spin-density approximation (LSDA). However, many aspects of the deceptively simple model system of Ni_xFe_{1-x} alloys still elude quantitative explanations. One of the unresolved problems is the difficulty in reconciling the itinerant character of magnetic *d* electrons (which seems to be reliably established by de-Haas–van Alfen experiments[2]) and the value as well as the positive sign of the spin polarization measured by tunneling experiments. Since the electronic density of states (DOS) at the Fermi level is higher for spin-down *d* electrons than for *s* electrons, it is obvious that this effect cannot be explained within simple models based on the constant tunneling matrix element approximation. The qualitative explanation was suggested in a number of papers (e.g., Ref. 3), where it was pointed out that the tunneling matrix elements for *s* electrons are larger than for *d* electrons. Although this picture is instructive for a qualitative understanding of the transport spin polarization, it is not very useful for quantitative analysis, since in transition metals electrons can be only marginally divided into *s* and *d* types. Instead, it is more appropriate to speak in terms of different bands with different Fermi velocities. Within this approach we propose a natural quantitative interpretation of this effect based on band-structure calculations, consistent with our spin-polarization measurements and the most recent tunneling results.[4]

In order to make a meaningful comparison between spin-polarization measurements in various experiments and the theory, the spin polarization must be clearly defined.[5,6] One cannot generally expect that the tunneling spin polarization, P_T, which is determined by a fraction of the spin-polarized current, is the same as the spin polarization probed, for instance, by spin-resolved photoemission, P_N. While P_N is related to the electronic density of states (DOS) at the Fermi surface, $N(E_F)$, P_T is determined by a weighted average of the DOS and tunneling matrix elements, which are, in general, functions of the Fermi velocities. In the spin-polarized Andreev reflection experiments,[5,7] yet another spin polarization, P_A, is measured. In the clean (ballistic or Sharvin) limit, P_A is defined by the average projection of the Fermi velocity v_F on *z*, the direction normal to the contact plane,

and thus $P_A = P_{Nv} \propto \langle N(E_F) v_{Fz} \rangle$. In the opposite, dirty (diffusive or Maxwell) limit, P_A is determined by $P_{Nv^2} \propto \langle N(E_F) v_{Fz}^2 \rangle$.[5,6] The same $P = P_{Nv^2}$ characterizes the spin polarization of the bulk electric current, as well as the tunneling current in the case of specular, low transparency barrier.[6] In the Ni_xFe_{1-x} system, where the transport properties are determined by both heavy *d* electrons and light *s* electrons, the tunneling current as well as the current in the diffusive case of Andreev reflection are dominated by the majority spins, even though their density of states is smaller. Similarly, there is no reason to believe that P_T or P_A should be related to the magnetic moment, which is defined as a difference in the total number of spin-up and spin-down electrons. Since in Ni_xFe_{1-x} the Fermi surface is far from spherical, the effective mass is strongly dependent on the wave vector, and the bands are highly hybridized, it is unrealistic to expect that the spin-dependent transport of these compounds can be described by the simple model of the polarized homogeneous electron gas, as it was often assumed in earlier works. Consequently, the once popular idea that the spin polarization, as measured by the tunneling spectroscopy, is proportional to the bulk magnetization[3,8] is not applicable to this system.

In this article, we present direct detailed measurements of the transport spin-polarization of the Ni_xFe_{1-x} system by a newly developed point-contact Andreev reflection (PCAR) technique.[5] We also perform band-structure calculations of the spin polarization in this system, using a standard LSDA technique. The measured values of the transport spin polarization are almost independent of the composition,[9] in reasonably good agreement with the theory. Based on the band-structure calculations, we interpret this surprising result as a consequence of compensation of the numerous but heavy *d* electrons and scarce but light *s* electrons.

Many thin films and bulk samples were studied. They included a Ni single crystal, several Ni and Fe polycrystalline foils, a [100]-oriented single-crystal Fe film grown on a GaAs substrate by molecular beam epitaxy, and a number of variable composition Ni_xFe_{1-x} films grown on Si-[100] substrates deposited by thermal (*e*-beam) evaporation.[4] In order to make meaningful conclusions from the measurements and to compare the experimental results with the theory, we determined the structural phase of the Ni_xFe_{1-x} films for the entire composition range. X-ray-diffraction data (specular $\theta/2\theta$ scans) were recorded for each of the Ni_xFe_{1-x} compositions over two angular ranges, 35°–68° and 71°–86°. In all cases only a single phase was found:[10] the γ(fcc) phase is present for $x > 0.47$; the α(bcc) phase is present for $x < 0.30$. These results are consistent with the results for bulk samples[11] and for thin films.[12] The lattice parameters for the films were within 0.3% of the corresponding bulk values.

The details of the PCAR technique are described elsewhere.[5] The method measures the degree of suppression of Andreev reflection at a ferromagnet/superconductor interface due to the spin polarization of the ferromagnet.[13] The Andreev process allows propagation of a single electron with the energy below the superconducting gap Δ from the normal metal to the superconductor, by reflecting at the interface as a hole via a time-reversal process. In a nonmagnetic normal metal this is always allowed, because in such a metal each energy state has both spin-up and spin-down electrons.

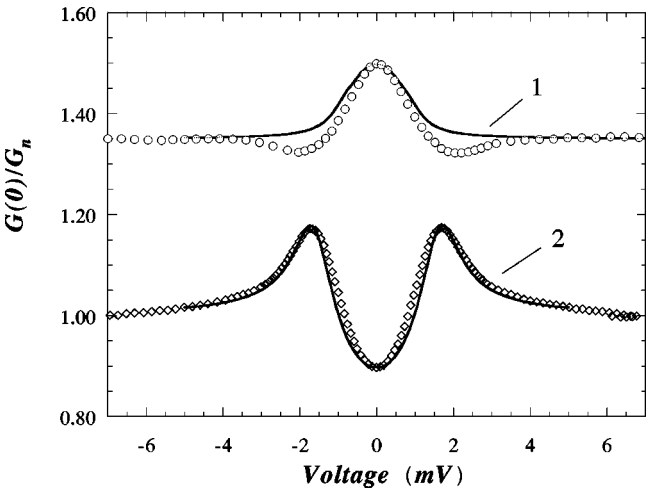

FIG. 1. $G(V)/G_n$ as a function of voltage *V* for two samples: 1 (○), Fe point/Ta foil base; 2 (◇), Nb point/$Ni_{80}Fe_{20}$ film base; solid curves—modified BTK fits ($T = 1.7$ K, $Z = 0$, $\Delta = 0.6$ meV and $T = 2.2$ K, $Z = 0.4$, $\Delta = 1.5$ meV, respectively). The values of *P* obtained from these fits are 42.5% for Fe and 49.5% for $Ni_{80}Fe_{20}$. Note that the bath temperature in the latter case was 1.8 K (see text). Curve 1 has an arbitrary vertical offset.

However, in a magnetic metal this is no longer true and Andreev reflection is limited by the spin direction with the smaller number of conductance channels, which drastically changes the subgap conductance. To account for finite temperatures and arbitrary barrier transparency *Z*, the normalized conductance data $G(V)/G_n$ (G_n is obtained at voltages $V \gg \Delta/e$, *e* is the electron charge) were compared to the modified[5] Blonder, Tinkham, and Klapwijk (BTK) (Ref. 14) model with only two adjustable parameters (*P* and *Z*).[15] The temperature was generally taken to be equal to the temperature of the helium bath and Δ was defined separately from the BCS dependence.[16] This procedure allowed us to determine the *magnitude* of the spin polarization.[5]

Our adjustment mechanism consisted of a sharpened rod (superconducting or ferromagnetic) which was driven by a micrometer until it touched the (ferromagnetic or superconducting) base. Superconducting Nb, V, and Ta were used for the measurements reported here. Typical normalized conductance data $G(V)/G_n$ obtained by the PCAR method are shown in Fig. 1 as a function of voltage *V*. For each sample a number of different contact junctions (with the contact resistance $1\ \Omega < R_c < 100\ \Omega$) were measured and then fitted with the modified BTK model. In Table I we present a summary of the data obtained for several samples for the end points (Ni and Fe) which were studied in more detail. Although we observed some variation in the values of *P* for the same material, the results are quite consistent and do not appear to depend strongly on whether the ferromagnet was a bulk single crystal, a foil, or a film. Furthermore, it does not seem to matter whether it was the point or base in the contact. Finally, the value of *P* does not depend strongly on the superconducting material. Accordingly, the values for *P* for individual samples of each material were averaged together. For Fe, $\langle P \rangle = (44 \pm 3)\%$ and for Ni, $\langle P \rangle = (46 \pm 3)\%$.

The PCAR results for the entire Ni_xFe_{1-x} are shown in Fig. 2. For the measurements of the thin-film series a Nb tip was used. The spin polarization is almost composition inde-

TABLE I. Spin-polarization results for pure Fe and Ni. *N* refers to the number of distinct point contacts made, *P* to the average polarization obtained for the *N* contacts, and *dP* to the standard deviation. C=crystal, Fl=foil, Fm=film.

Point/Base	*N*	*P*(%)	Point/Base	*N*	*P*(%)
Fe/V (C)	9	45±2	Nb/Ni (C-1)	8	45±2
Fe/Ta (Fl)	14	46±2	Nb/Ni (C-2)	8	41±4
Fe/Nb (Fl)	3	42±3	Nb/Ni (C-3)	11	48±4
Nb/Fe (Fm)	5	41±3	Nb/Ni (Fl)	10	45±2
Ta/Fe (Fm)	12	45±2	Nb/Ni (Fm)	14	45±3
			Ta/Ni (Fl-1)	8	44±4
			Ta/Ni (Fl-2)	10	50±1
Average, Fe	$\langle P \rangle$	44±3	Average, Ni	$\langle P \rangle$	46±3

pendent, whereas the measured magnetic moment (shown in the inset in Fig. 2) changes by a factor of 3. Evidently, our measurements do not show any correlation between the spin polarization and magnetic moment, which was observed in the early tunneling spectroscopy measurement.[17] Although our spin-polarization values differ substantially from these early results, they are quite close to the most recent tunneling measurements[4] obtained from the ''companion'' Ni_xFe_{1-x} samples.[9] This result is not necessarily to be expected as PCAR probes $N(E_F)v_{Fz}^2$ averaged over the entire Fermi surface, whereas tunneling through a thick barrier can be shown to probe $N(E_F)v_{Fz}^2$ only at those selected points of the Fermi surface where quasimomentum is perpendicular to the interface. Apparently, averaging over individual grains in the Ni_xFe_{1-x} films helps to bring the tunneling spin-polarization results close to the Fermi surface-averaged PCAR results.

To calculate the spin polarization, we performed LSDA band-structure calculations.[18] Our x-ray measurements allowed us to conclude that a single structural phase was present at any given Ni-Fe composition. Thus we were able

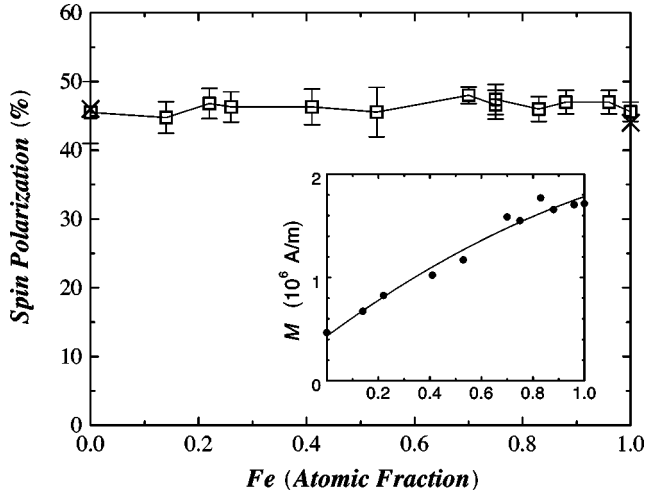

FIG. 2. PCAR results for the spin polarization *P* as a function of Fe content for Ni_xFe_{1-x} samples. (□), films; (×), pure Ni and Fe foils and single crystals. Inset: Magnetic moment, *M*, as a function of the Fe content for Ni_xFe_{1-x} samples. The lines are guides to the eye.

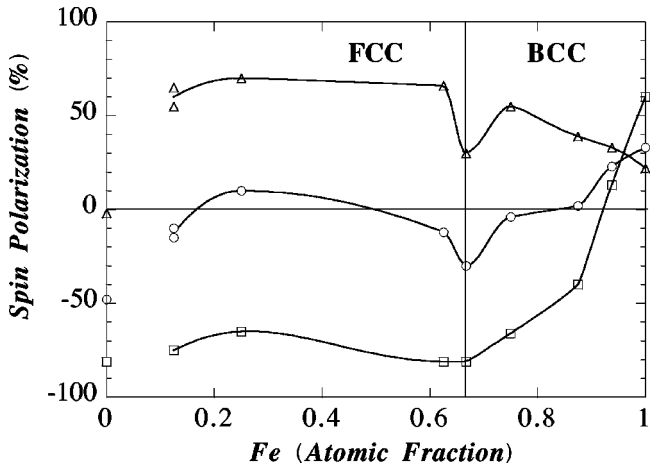

FIG. 3. The results of the band-structure calculations for P_N, P_{Nv}, and P_{Nv^2}: (□), P_N; (○), P_{Nv}; (△), P_{Nv^2}. The lines are guides to the eye. The results for pure Ni are shown for completeness.

to compare the experimental results with the calculations performed in the appropriate lattice structure. For Ni content $x<0.35$, the calculations were carried out in an average bcc lattice, for $x>0.35$ an average fcc lattice was used.[19] Several ordered Ni-Fe supercells with the compositions $NiFe_{15}$, $NiFe_7$, $NiFe_3$, $NiFe_2$, Ni_3Fe, and Ni_7Fe were used. The results of the calculations of the spin polarization (P_N, P_{Nv}, and P_{Nv^2}) are shown in Fig. 3. First of all, we observe that the three polarizations are dramatically different, which emphasizes once again the importance of the correct definition of the spin polarization for a given experiment. These differences are due to the strong variation of the kinematic properties between *s*-like and *d*-like electrons. Specifically, the Fermi velocity anisotropy between the different sheets of the Fermi surface, as well as the angular anisotropy, have to be taken into account for a quantitative description of any spin-transport experiment. While ''heavy'' parts of the Fermi surface dominate the DOS spin polarization, ''light'' parts determine the spin polarization relevant for transport and tunneling phenomena.[6] There is good agreement between the experimental data for the Ni-rich and Ni-poor alloys and band-structure calculations for the diffusive limit, P_{Nv^2} (except for pure Fe where P_{Nv} agrees with the experiment better than P_{Nv^2}). This result is quite reasonable because the electron mean free path, l_e, of these alloys (but not necessarily for the pure components) is typically very short (compared to the size of the contact) even at low temperatures, especially for minority spins ($l_e \sim 5-10$ Å).[20] This is also consistent with the agreement between PCAR and tunneling spin polarizations;[4] as mentioned above, the latter is also defined by $\langle Nv^2 \rangle$.

We could not perform reliable calculations for pure Ni. This reflects a well-known problem in conventional band-structure theory, which is unable to completely account for electronic correlation effects in the 3*d* states of metallic oxides and, to a lesser extent, of Ni. The correlation effects in Ni are known to reduce the exchange splitting by a factor of 2 which, in turn, should affect the spin polarization. For a different reason, we exclude the theoretical calculations for the compounds close to the 50:50 Ni:Fe composition. At relevant lattice parameters the fcc phase of Fe is antiferromagnetic, so close to its solubility limit in the fcc Ni (ap-

proximately 60–65 %) the corresponding Ni-Fe alloys must have Fe clusters which are sufficiently large to develop antiferromagnetic order. On the other hand, Fe-Ni and Ni-Ni interactions are ferromagnetic. This creates frustration leading to noncollinear spin ordering.[21]. A theory of spin-polarized transport in such systems is yet to be developed.

In summary, we have presented the band-structure calculations of the transport spin polarizations in the Ni-Fe system and the experimental measurements of the same system using the PCAR technique. Overall, the spin polarization measured by PCAR technique agrees reasonably well with the band-structure calculations for $P = P_{Nv^2}$. It is also in surprisingly good agreement with the most recent tunneling results.[4] Furthermore, our results repudiate the idea of a direct relationship between the spin polarization and the magnetic moment. At the same time we show that the spin polarization in electronic transport is determined by the delicate balance of the density of states and the kinematics of the *s* and *d* electrons (the variation of the Fermi velocity over the Fermi surface) and, therefore, dependent on the measurement technique and the transport process in question. In particular, our calculations give a quantitative explanation for a longstanding problem of the positive values of tunneling spin polarization observed for the Ni-Fe system, which has important implications both for fundamental issues of spin transport and for magnetoelectronics applications.

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[1] G.A. Prinz, Phys. Today **48** (4), 58 (1995); Science **282**, 1660 (1998).

[2] H. Ebert, P. Strange, and B.L. Gyorffy, J. Phys. F: Met. Phys. **18**, L135 (1988), and references therein.

[3] J.B. Gadzuk, Phys. Rev. **182**, 416 (1969); M.B. Stearns, J. Magn. Magn. Mater. **5**, 167 (1977).

[4] J.S. Moodera *et al.* (unpublished).

[5] R.J. Soulen *et al.*, Science **282**, 85 (1998).

[6] I.I. Mazin, Phys. Rev. Lett. **83**, 1427 (1999).

[7] S.K. Upadhyay *et al.*, Phys. Rev. Lett. **81**, 3247 (1998).

[8] R. Meservey, D. Paraskevopoulos, and P.M. Tedrow, Phys. Rev. Lett. **37**, 858 (1976).

[9] ''Companion'' samples of the same composition made under the same conditions (see R. van der Veerdonk, Ph.D. thesis, Eindhoven University, Eindhoven, 1999) were also measured by Moodera *et al.* (Ref. 4) using standard tunneling spectroscopy (Ref. 22).

[10] Based on the signal-to-noise ratios of the strongest peak in our diffraction patterns, it is estimated that a second phase could be detected if present in quantities greater than 5%.

[11] A.J. Bradley and A. Taylor, Philos. Mag. **23**, 545 (1937).

[12] M. Konno and H. Konno, J. Magn. Magn. Mater. **118**, 381 (1993).

[13] M.J.M. de Jong and C.W.J. Beenakker, Phys. Rev. Lett. **74**, 1657 (1995).

[14] G.E. Blonder, M. Tinkham, and T.M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).

[15] Note that our analysis is based on an assumption [used both in the BTK paper (Ref. 14) and in our modified version (Ref. 5)] of a ballistic contact, for which the mean free path l_e is much larger than the size of the contact, *d*. It is difficult to assess the effect of violation of the condition ($l_e \gg d$). Preliminary calculations show, however, that it mainly affects the values of *Z* in our fits, while the changes in the resulting spin polarization are relatively small.

[16] For some junctions we could obtain better fits by using a higher temperature. This is most likely due to the fact that we did not explicitly introduce any gap smearing, as is customary in superconducting tunneling [i.e., R.C. Dynes *et al.*, Phys. Rev. Lett. **53**, 2437 (1984)]. Fitting with an increased temperature is to a large extent equivalent to an additional gap smearing. We emphasize that even when increasing the temperature parameter impoves the quality of the fits, it hardly changes the resulting spin polarization.

[17] D. Paraskevopoulos, R. Meservey, and P.M. Tedrow, Phys. Rev. B **16**, 4907 (1977). In these measurements, a two-step junction fabrication process was used, while the more recent experiments (Ref. 4) used *in situ* fabrication. Tunneling spectroscopy is a more surface sensitive technique (Ref. 22) than PCAR and, therefore, may be susceptible to surface conditions of nickel, which are clearly improved by using an *in situ* process (Ref. 4).

[18] The LMTO-ASA Stuttgart code was used. See, e.g., O.K. Andersen and O. Jepsen, Phys. Rev. Lett. **53**, 2571 (1984).

[19] Using the correct structural phase in these calculations is important (replacing fcc by bcc and vice versa yields significantly different results).

[20] D.Y. Petrovykh *et al.*, Appl. Phys. Lett. **73**, 3459 (1998).

[21] Y. Wang *et al.*, J. Appl. Phys. **8**, 3873 (1997).

[22] P.M. Tedrow and R. Meservey, Phys. Rep. **238**, 173 (1994).