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Effect of surface scattering on the extraordinary Hall coefficient in ferromagnetic films

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The effect of surface scattering on the extraordinary Hall coefficient has been studied in thin films of nickel and granular Ni-SiO_2 mixtures. The surface scattering contributions to the Hall coefficient and longitudinal resistivity have been extracted from the respective total values. The temperature-independent linear relation between the two parameters has been found. Different scattering mechanisms need to be separated, and the applicability of the existing models for heterogeneous systems with spatially extended scattering centers should be reexamined.

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INTRODUCTION

The effects of surfaces and interfaces on magnetic and transport properties of heterogeneous and nanoscale magnetic systems are dramatically enhanced as compared to homogeneous bulk magnets. Giant magnetoresistance (GMR), a good example of "nonbulk" phenomena, is intimately associated with spin-dependent scattering in the presence of interfaces separating magnetic and nonmagnetic regions. Relatively to magnetoresistance in multilayers and granular heterogeneous ferromagnets, quite modest attention has been paid so far to another electric transport property: the Hall effect. Data reported are scarce and significantly different from those known in bulk homogeneous magnets.

The Hall effect in magnetic materials is commonly described by the phenomenological equation[1] $\rho_H = R_0 B + R_e \mu_0 M$, where ρ_H is the Hall resistivity, B the magnetic induction, and M the magnetization. R_0 is the ordinary Hall coefficient and is related to the Lorentz force acting on moving charge carriers. R_e, the extraordinary Hall coefficient, is associated with a break of right-left symmetry during spin-orbit scattering in magnetic materials and can be much larger than R_0. In these cases the Hall voltage can serve as a direct measurement of magnetization. In bulk magnets it has been established, both experimentally and theoretically, that there is a direct correlation between the extraordinary Hall coefficient and longitudinal resistivity in the form $R_e \propto \rho^n$, where n depends on the predominant scattering mechanism involved: $n=1$ for skew scattering and $n=2$ for side jumps.[2,3] Superposition of two effects is usually presented as $R_e = a\rho + b\rho^2$, where a and b are coefficients corresponding to the skew scattering and side jump, respectively. Skew scattering is assumed to be dominant in low-resistivity systems, and the only bulk materials where $n=1$ has been observed are low-resistivity dilute alloys at low temperatures.[4] The rest of the previously studied homogeneous ferromagnets with relatively high resistivity demonstrated $n \simeq 2$, and the side jump mechanism was claimed to be dominant. It is important to mention that in bulk ferromagnets the resistivity was varied either by temperature or by modest doping, low enough to avoid significant changes in the band structure of the material.

Remarkably different results have been reported recently in heterogeneous ferromagnetic systems, multilayers, and granular mixtures. $n=2$ was reported for molecular-beam-epitaxy-grown Co/Cu superlattices,[5] $n=2.6$ was found for electron-beam-evaporated Fe/Cr multilayers,[6] and n as high as 3.7 was reported for heterogeneous giant magnetoresistance films of Co-Ag.[7] Roughness of the interfaces has been found[8] to modify the relationship between the extraordinary Hall coefficient and longitudinal resistivity of Fe/Cr multilayers. Polarity of the extraordinary Hall coefficient in granular Co-Ag mixtures has been found[9] to change from negative in thick (200 nm) to positive in thin (10 nm) films. The latter was interpreted as evidence for competition between the bulk and surface scattering contributions. A nonmonotonic field dependence of R_e has been found in several GMR systems like Fe/Cr (Ref. 10) multilayers and Co-Ag granular mixtures.[7,11]

The theory of the extraordinary Hall effect in systems affected by geometrical constrains is far from being complete. Kogan and Ustinov[12] were probably the first to consider the surface scattering in calculations of the effect in ferromagnetic films. Their model is analogous to the "bulk" model of Luttinger.[13] The surface scattering is presented by impurities localized on the film boundaries, their surface concentration is different from that of the bulk. The final formulas are obtained in the effective mass limit. The calculated surface Hall contribution is qualitatively similar to the generalized bulk expression $R_{ess} = \alpha(\rho - \rho_b)\rho/\rho_b$, where R_{ess} is the surface contribution to the extraordinary Hall coefficient, and ρ_b and ρ are the bulk and effective thin-film resistivities, respectively. More work has been done since the discovery of the giant magnetoresistance phenomenon.

Zhang[14] used the side jump mechanism as a basis for the extraordinary Hall effect in magnetic multilayers. The principal conclusion of this work was that the commonly used scaling relation between the extraordinary Hall resistivity and longitudinal resistivity is not valid. Skew scattering has been treated within the quantum[15] and quasiclassical[16] size effects. Skew scattering in granular GMR alloys involving not one, but a few grains has been calculated by Vedyaev *et al.*[17] Unusual scaling power values, including $R_e \propto \rho^{3.8}$, have been found for a suitable selection of model parameters.

Our attempt to clarify the picture starts by a simple assumption that scattering by phonons, magnons, impurities, boundaries, and surfaces can affect resistivity and the extraordinary Hall effect differently. It is not evident, therefore, that the total resistivity is a good parameter to characterize the effect. Instead, the contribution of each scattering process should be isolated and determined. This work is focused on the surface scattering contribution to the extraordinary Hall coefficient.

SAMPLES

Two types of systems have been chosen. The first are thin films of nickel with electronic mean free path of the order of or shorter than their thickness. Following the original Fuchs size-effect theory,[18] external surfaces impose a boundary condition on the electron-distribution function, which enhances the thickness-independent bulk or intrinsic resistivity ρ_b, to the thickness-dependent total resistivity ρ. The difference between the two can be considered as the surface scattering resistivity ρ_{ss}. The samples used in this study were prepared by conventional high-vacuum deposition techniques in conjunction with standard lithographic procedures. Quartz substrates were ultrasonically cleaned in a sequence of diluted HCl and ethanol to ensure the removal of organic and inorganic surface contamination. After calibration of the thickness monitor of the deposition chamber, Ni films in the nominal thickness range 5–100 nm were deposited at room temperature in a vacuum of 10^{-8} Pa using a multisource *e*-beam evaporator at an electronically controlled deposition rate of 4–5 nm/s. Subsequently, SiO_2 films of thickness 200 nm were deposited to prevent contamination and degradation of the Ni films. The Ni/SiO_2 stack was patterned into Hall bars of 3×0.4 mm length and the appropriate three pairs of contacts $\sim$1 mm apart. To deposit standard Cr/Au pads for wire attachment, contact windows were etched into the SiO_2 using wet chemical etching with buffered HF.

The second group of samples consists of relatively thick Ni-SiO_2 granular films with a variable content of SiO_2. The total thickness of the series was about 200 nm. Due to the mutual immiscibility of the components, silica is distributed within the nickel matrix in the form of small nanoscale islands. By gradually increasing the SiO_2 content, the percolation threshold can be reached. This is the limit studied recently in a number of granular systems.[19–21] In our case we have limited the range of samples to relatively low silica volume concentrations (below 25%) to avoid complications of the fractal structure in the vicinity of the percolation threshold.

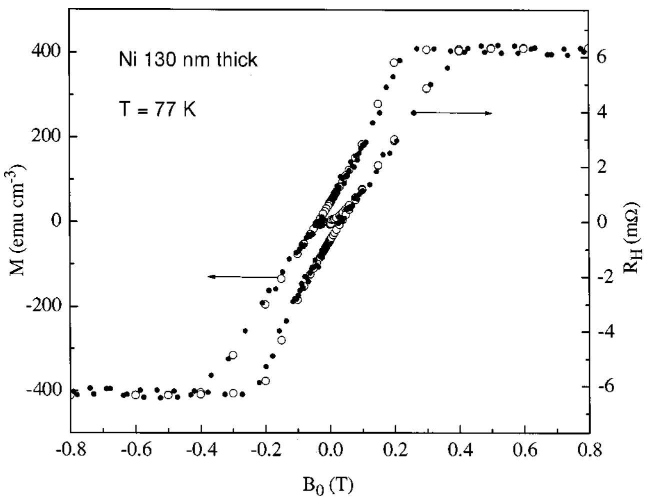

FIG. 1. Hall resistance (solid circles, right-hand axis) and magnetization (open circles, left-hand axis) of 130-nm-thick Ni film measured at 77 K with magnetic field B_0 perpendicular to the film plane.

RESULTS AND DISCUSSION

The transverse (Hall) and longitudinal resistance of all the studied samples were measured between room and liquid-helium temperatures up to fields much exceeding the field of magnetic saturation. A magnetic field was applied perpendicular to the plane of films. Every field-dependent measurement included upward and downward sweeps for both field polarities. The typical magnetic field dependence of the Hall resistance ($R_H = V_{xy}/I_{xx}$) of 130-nm-thick Ni films is plotted in Fig. 1, together with its magnetization measured by a superconducting quantum interference device (SQUID). The scales are normalized for the clarity of presentation. Both curves are practically identical, which points out an intimate correlation between the extraordinary Hall effect and magnetization. For the following discussion, the extraordinary Hall resistance R_{Hs} and resistivity ρ_{Hs} are defined by extrapolating the high-field linear slope of the measured signal $R_H(B_0)$ to zero field: $\rho_{Hs} \equiv R_{Hs} t$, where t is thickness of the film. The longitudinal resistivity ρ is defined as the zero-field resistivity in virgin state. Specification of the state here is of limited importance since the magnetoresistance of the studied films is less than 1%.

The Curie temperature and saturation magnetization of Ni films with thickness down to 2 nm was found[22] to be the same as in bulk Ni. Deviations of magnetic behavior, occasionally reported for very thin films, are rather related to the superparamagnetic character of weakly coupled grains and not to the intragranular magnetic changes. Correlation between ρ_{Hs} and the extraordinary Hall coefficient R_e is based, therefore, on the assumption that magnetization of all films in the saturated state is thickness independent. We also limit the present study to films thicker than 5 nm.

The resistivity of several Ni films is plotted in Fig. 2 as a function of their thickness for three temperatures: 294, 77, and 4.2 K. As expected for films with a mean free path of the order of the thickness, the resistivity is strongly enhanced in the thin-film limit, following the Fuchs-Sondheimer model.[18]

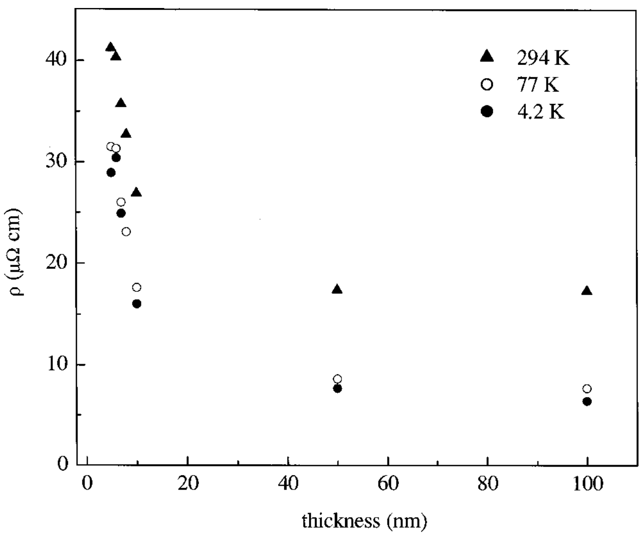

FIG. 2. Resistivity of Ni films as a function of their thickness at 294, 77, and 4.2 K.

The resistivity of our thinnest 5-nm film is about an order of magnitude higher than that of the 100-nm-thick film. The difference of the resistivity values between the room and helium temperatures is almost the same for all samples of the series, which indicates their similar bulk properties. The same can be concluded from comparison of the normal Hall coefficients [high-field linear slope of the $R_H(B_0)$ curves], which is constant within an accuracy of $\pm 10\%$ for the entire series. Nonepitaxial films, like those discussed here, deposited and measured at the same temperature are usually free of strain and its influence. We can, therefore, safely assume that the observed variation of resistivity with thickness is due to the enhanced surface scattering.

The extraordinary Hall resistivity ρ_{Hs} is shown in Fig. 3 as a function of thickness. The qualitative behavior is similar to that of the longitudinal resistivity: constant in thick films and strongly enhanced in the thin-film limit.

The standard analysis of the extraordinary Hall coefficient

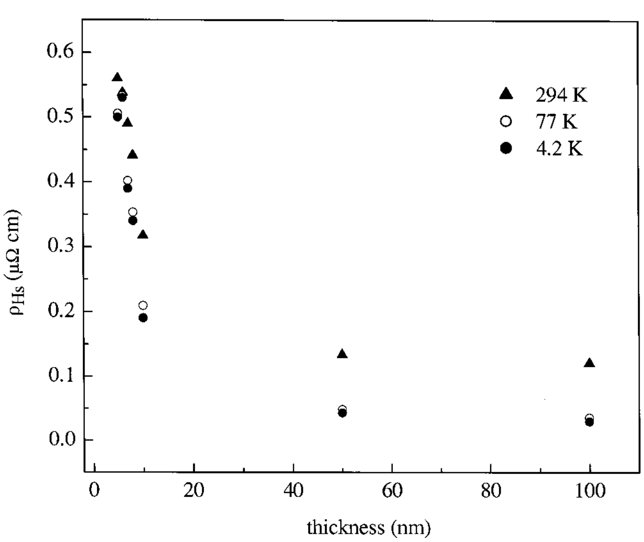

FIG. 3. Extraordinary Hall resistivity of Ni films as a function of their thickness.

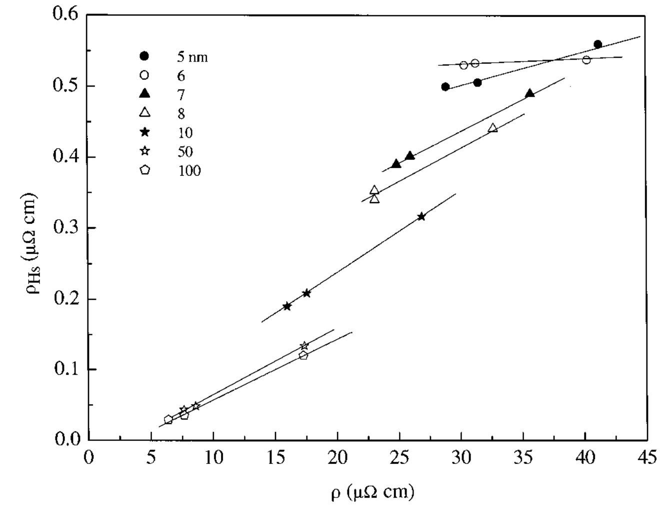

FIG. 4. Extraordinary Hall resistivity of Ni samples as a function of their total resistivity. Three data points per sample correspond to three temperatures 294, 77, and 4.2 K. Straight lines are guides for the eyes.

in earlier studies was based on the following relation between the longitudinal and transverse resistivities: $\rho_H = a\rho + b\rho^2$, where a and b are coefficients corresponding to the skew scattering and side jump. A similar analysis can be tried in this case when the resistivity is varied either by the temperature or by the thickness of the samples. ρ_{Hs} of all Ni films measured at three temperatures (294, 77, and 4.2 K) is plotted in Fig. 4 as a function of the respective longitudinal resistivity. Quite limited information can be extracted from this plot, but a surprising observation that ρ_{Hs} of the 6-nm film seems to be independent of its temperature-dependent resistivity. The coefficients a and b can be determined by plotting ρ_H/ρ as a function of ρ. Figures 5 and 6 present the same data, but analyzed following two different parameters separately: temperature and thickness. Figure 5 shows ρ_{Hs}/ρ as a function of ρ, where each symbol represents an isothermal measurement of different samples: triangles for room temperature, open circles for 77 K, and solid circles for

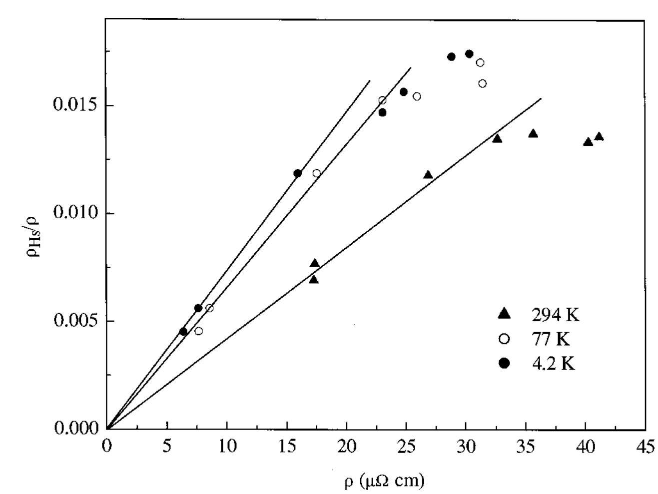

FIG. 5. The same data as in Fig. 4 plotted as ρ_{Hs}/ρ vs ρ. Each symbol corresponds to a different temperature.

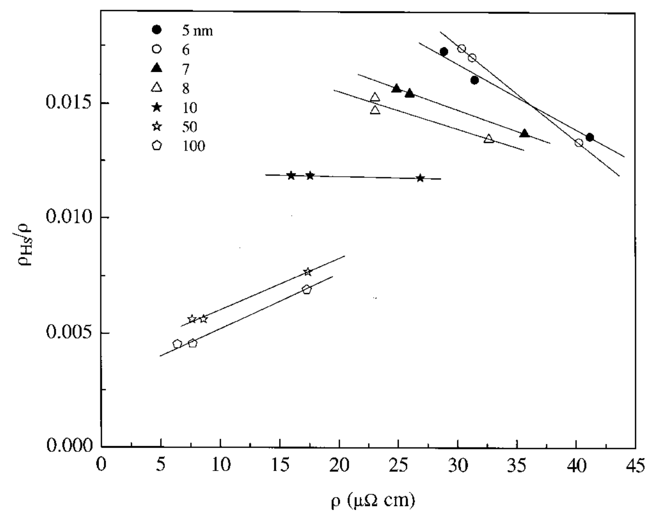

FIG. 6. The same data as in Fig. 4 plotted as ρ_{Hs}/ρ vs ρ. Each symbol corresponds to a different sample.

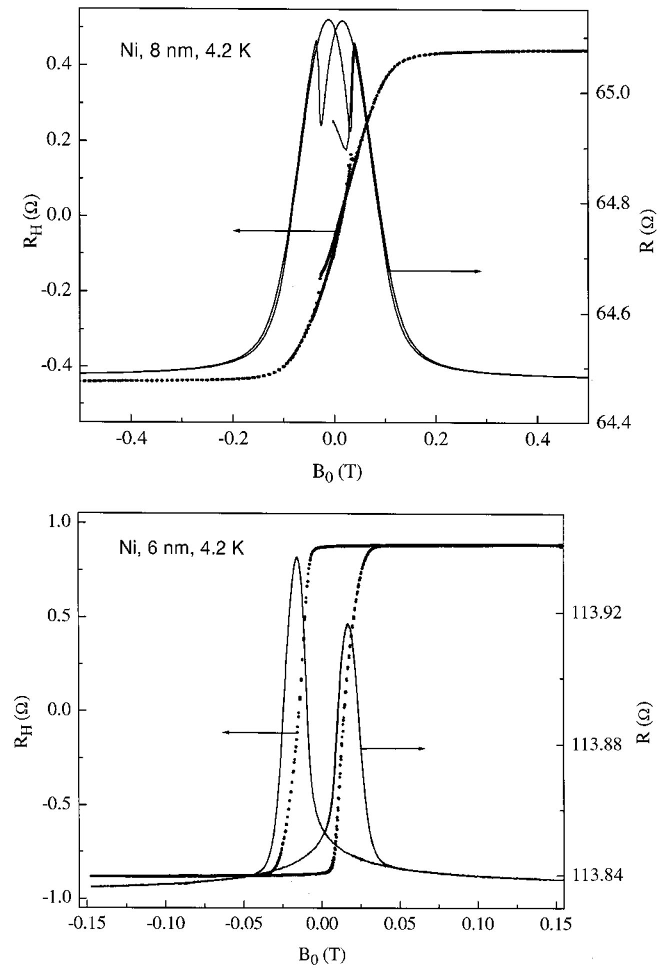

FIG. 8. Hall resistance (solid circles, left-hand axis) and longitudinal resistance (solid line, right-hand axis) as a function of applied magnetic field of (a) 8-nm- and (b) 6-nm-thick Ni films. $T = 4.2$ K.

4.2 K. Linear variation can be assumed for the low-resistivity (thick film) part of the plot, giving $a = 0$ and a temperature-dependent b. Following the previous discussion, that would mean that the side jump is the dominant mechanism of the extraordinary Hall effect. Coren and Juretschke,[23] Pichard *et al.*,[24] and Schad *et al.*[25] have drawn this conclusion from similar experimental results on thin Ni and Fe films. However, the high-resistivity (thin) samples deviate strongly from the linear variation and do not support this scenario.

Another presentation of the same data is shown in Fig. 6, where each symbol corresponds to a given sample measured at different temperatures. The same analysis would imply (i) a strong (dominant) influence of the side jump in thick (100 and 50 nm) films with $a \approx 0$ and positive b, (ii) dominant skew scattering ($b \approx 0$) in 10-nm-thick films and (iii) combination of both skew scattering and side jump in thin films ($t < 10$ nm) with negative coefficient b. We fail to propose any solid arguments to justify these conclusions, in particular the change of polarity of the coefficient b with thickness.

As mentioned earlier, the goal of this work is to extract the surface scattering contribution to the extraordinary Hall effect. Assuming that the 100-nm-thick film represents the properties of bulk Ni, the effect of surface scattering can be found by a simple subtraction of the bulk parameters from that of thinner films. The surface scattering contribution to the extraordinary Hall effect at a given temperature can be defined as $\Delta\rho_{Hs} = \rho_{Hs} - \rho_{Hsb}$, where ρ_{Hsb} is the extraordinary Hall resistivity of the 100-nm-thick film. Respectively, the surface scattering contribution to the longitudinal resistivity is defined as $\Delta\rho = \rho - \rho_b$, where ρ_b is the resistivity of the 100-nm-thick film. The results are shown in Fig. 7 for all the films of the series; different symbols relate to different temperatures. Evidently, all results collapse onto a single straight line.

Notably, the magnetic anisotropy of films and details of their magnetization reversal do not affect the variation of the extraordinary Hall coefficient. Figure 8 presents both the

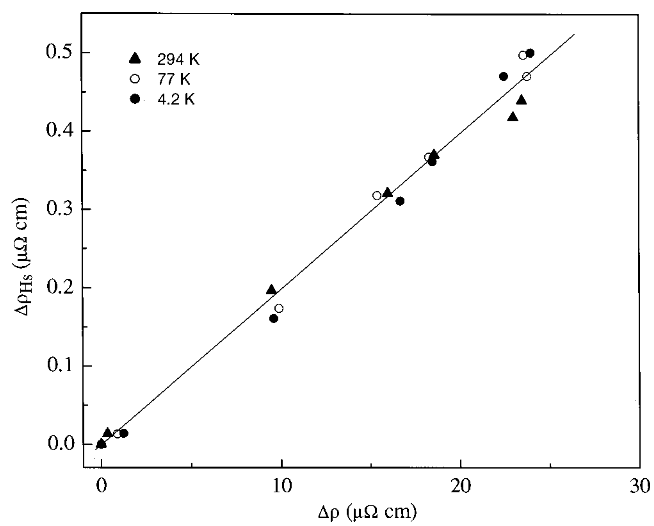

FIG. 7. Surface scattering component of the extraordinary Hall resistivity as a function of the respective resistivity term. $\Delta\rho_{Hs} = \rho_{Hs} - \rho_{Hsb}$, where ρ_{Hsb} is the extraordinary Hall resistivity of the 100-nm-thick film. The surface scattering contribution to the longitudinal resistivity is defined as $\Delta\rho = \rho - \rho_b$, where ρ_b is resistivity of the 100-nm-thick film. The results are shown for all films of the series; different symbols indicate different temperatures.

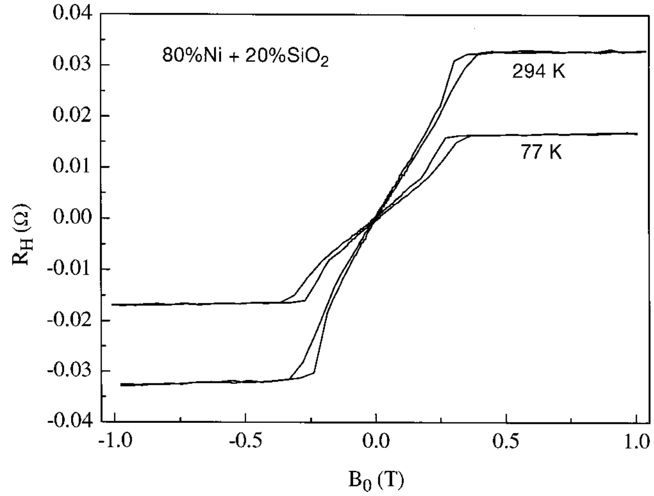

FIG. 9. Hall resistance of the Ni-SiO_2 granular film as a function of applied magnetic field. Ni volume concentration is 80%. Thickness is about 200 nm.

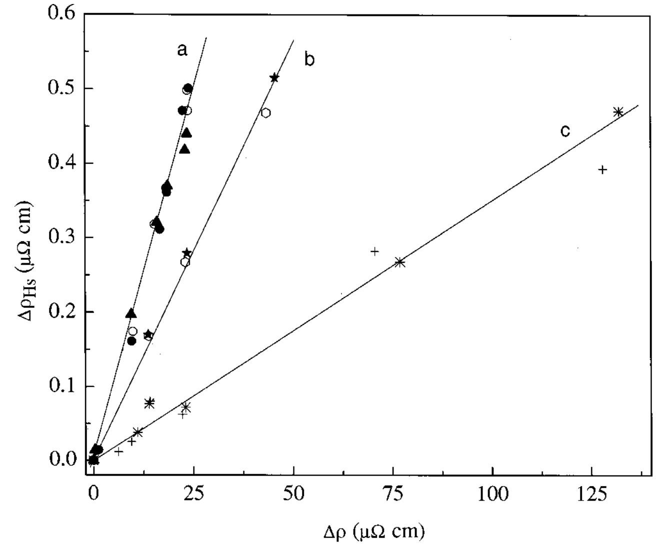

FIG. 10. Surface scattering component of the extraordinary Hall resistivity as a function of the respective resistivity term for (a) high-quality thin Ni films, (b) ''low''-quality Ni films, and (c) Ni-SiO_2 mixtures. Group (a) is shown for three temperatures: 294, 77, and 4.2 K. Groups (b) and (c) are shown for 294 and 77 K. Straight lines are guides for the eyes.

Hall resistance and magnetoresistance of two films: 6 and 8 nm thick. The magnetic properties and, respectively, magnetotransport characteristics are dramatically different: mostly reversible magnetoresistance with a characteristic low-field pattern and reversible Hall signal in the 8-nm film as compared with two profound peaks in magnetoresistance and a strong, almost rectangular hysteresis of the Hall signal in the 6-nm one. The difference between these two films can be related to the different orientation of the magnetic anisotropy: in plane in the 8-nm film and normal to the plane in the 6-nm one; the latter subject is out of the scope of this paper.

Two more sets of samples have also been studied. One is the series of Ni films prepared by electron beam deposition with ''inferior'' quality as compared with the first one. The longitudinal resistivity of this series is about 3 times higher: the room-temperature resistivity of the 100-nm-thick film of this series is 35 $\mu\Omega$ cm. The second is a series of codeposited Ni-SiO_2 granular samples of about 200 nm thick. Typical curves of the Hall resistance in Ni-SiO_2 measured as a function of applied field are shown in Fig. 9 for a sample with a 20% volume of SiO_2. The enhancement of the resistivity of these films with increasing SiO_2 content is mainly related to the scattering on interfaces of the insulating inclusions. The interface contribution to the extraordinary Hall effect and longitudinal resistivity can be extracted following the same procedure as for chemically uniform thin Ni films. We summarize the results for all three systems—high-quality thin Ni films, ''low'' quality Ni films, and Ni-SiO_2 mixtures—in Fig. 10. The surface scattering Hall term follows linearly the resistive one in all the studied cases. The slopes are different for each system, but the tendency is general.

CONCLUSIONS

The description of the extraordinary Hall effect in systems with multiple-scattering mechanisms requires the selection of proper macroscopic parameters. The traditionally used global resistivity seems not to be the best choice. Meaningful information can be extracted only by isolating contributions of different scatterers. Here the surface scattering term has been separated from the total resistivity and a linear ratio between the extraordinary Hall and longitudinal resistivities has been found. A similar linear dependence has been predicted for the skew scattering mechanism in bulk homogeneous ferromagnets. The latter is expected to dominate over the side jump in samples with very low resistivity and has been observed in clean alloys at low temperatures. The resistivity of ultrathin and granular films discussed here is in the range of tens to hundreds of $\mu\Omega$ cm and falls far away from the ''clean'' limit. The domination of the linear term in this limit contradicts, therefore, existing models developed both for bulk homogeneous materials with point scattering centers and for thin films and granular mixtures. The applicability of existing models for heterogeneous systems with spatially extended scattering centers, like surfaces and interfaces, should be reexamined.

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