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Strong nonlinearity and hysteresis of Hall resistance versus magnetization in nickel thin films

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ABSTRACT

The anomalous Hall effect (AHE) in ferromagnetic materials is perhaps one of the oldest unresolved mysteries in physics. First observed in 1881, its mechanism is still a controversial topic today. The question remains whether AHE is caused by intrinsic (Berry phase and band structure) or extrinsic (defect scattering) effects or a combination of both. Here we present experimental observation in nickel thin films that seems to add to the mystery, but may in fact provide crucial clues for ultimately resolving the controversy. The key observation is that the Hall resistivity of nickel films is a strongly nonlinear function of the magnetization and displays clear hysteresis with respect to M. Specifically, at low temperatures, the anomalous Hall coefficient switches between two saturated values under the magnetic field with a narrow transition region, but with a strong hysteresis, in contrast to the slow saturation of the magnetization. The nonlinearity and the hysteresis become more apparent with decreasing temperature or film thickness. Despite the simplicity of the lattice and magnetic structure of nickel films, these results are outside our current understanding of AHE, whether using intrinsic or extrinsic mechanisms of AHE. It presents a challenge for these models, and may be used as a test of validity for both types of theories.

Not until some 70 years after the observation of the AHE in ferromagnetic materials by Hall [1], was the first attempt made of a quantitative explanation of the phenomenon. In 1954, Karplus and Luttinger [2] proposed as the origin of AHE an intrinsic transverse current due to a correction in the group velocity of electrons when the electrons in a given band were coupled to other bands through spin–orbit interaction. The picture was questioned by Smit [3] whose analysis led to a nonvanishing contribution from skew or asymmetrical scattering of electrons only when the scattering probability was calculated in the second Born approximation. For decades, the extrinsic mechanism (including the skew scattering and the side jump proposed later by Berger [4] had dominated the field [5]. The situation was reversed when the intrinsic mechanism was reinterpreted with the concept of Berry curvature [6,7], which found support by a series of theoretical and experimental works [8–14]. The latest progress is a model which combines both the extrinsic and intrinsic mechanisms to get a unified picture [15]. And soon the experimental verification of the model was reported [16].

The AHE is closely connected to the magnetic structure of the material. The phenomenological relation [17] for the Hall resistivity expresses it in terms of the applied magnetic field and the spontaneous magnetization both along the z direction,

$$\rho_{xy} = \rho_0 + \rho_a = R_0 B + 4\pi R_s M \tag{1}$$

where R_0 and R_s are the normal and the anomalous Hall coefficients, respectively. Experimentally, R_s is usually obtained by linearly extrapolating the value in saturation magnetization region to zero field. At saturation, the Hall resistivity varies linearly with the magnetic field, with a slope R_0. Extrapolating back to zero field yields

$$\rho_{xys} - R_0 B = 4\pi R_s M_s = \rho_{as} \tag{2}$$

This step separates the ordinary Hall resistance from the anomalous Hall resistance. Usually, H is measured in an experiment, which is related to the inductance by $B = H + (1-N)M$ where N is the demagnetization factor. In thin films, because $N = 1$, the magnetic inductance B is identical to the field H. This allows a clean

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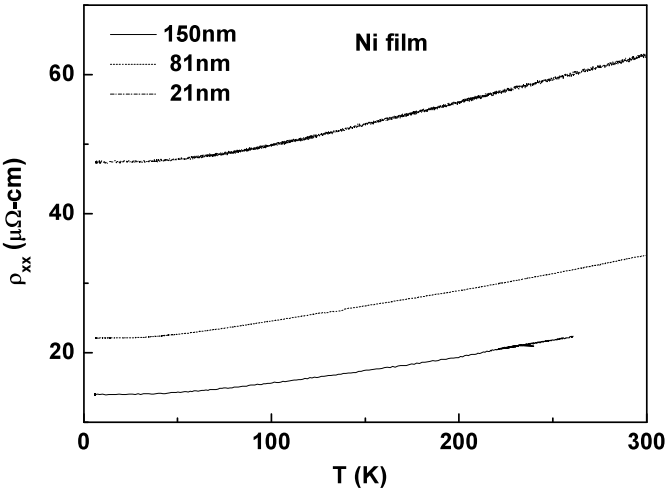

Fig. 1. The temperature dependences of Ni films with different thicknesses.

and unambiguous separation of the anomalous Hall resistance in thin films.

Our study shows that there is still a lot to learn even for a material as simple as pure nickel. As an elementary substance, pure nickel is free from any possible change or smearing of the chemical potential due to composition fluctuations. The cubic lattice structure also provides the simplest form of the magnetic crystalline anisotropy. Thus chemical, structural, and magnetic inhomogeneities are minimized. The pure nickel moment is rather robust in the bulk as well as in films. The Ni films were deposited onto Si(111) substrates by dc magnetron sputtering at room temperature with Ni target of 99.99% purity. The analysis of the structure, electronic, and magnetic properties of these samples have been given in a previous work [18]. The structure of the films was shown to be face-centered cubic (fcc) with mostly (111) texture. The average grain size was estimated to be about 30 nm. The bulk electron mean-free-path was estimated to be 30–50 nm from the thickness dependence of the resistivity. The temperature dependences of the resistivity for all the samples are nearly parallel (Fig. 1), showing similar electron–phonon scattering and phonon spectrum for different samples. From this, we conclude that there is no significant variation in the lattice structure among the samples [18].

Up to now, with a few exceptions [19,20], most works have focused on the regime where the magnetization is saturated. The results of our measurements for Ni films of three thicknesses in the saturated regime are in general agreement with previous data [19, 20]. Namely, the ratio ρ_{as}/ρ_{xx} increases with the decrease of film thickness, but the temperature slope changes sign in the process (Fig. 2a). The data do not show a constant Hall conductivity either (Fig. 2b). In the figure Hall conductivity is normalized to M_s [21]. Although these results are not strictly in agreement with either the skew scattering model or the Berry phase model for homogeneous bulk systems, it has been easy to attribute the discrepancies in terms of the difference between the surface scattering and the bulk scattering [20].

As the expression (1) is applicable to both below and above saturation [17,22,23], the process to saturation reveals much more information than the saturated values alone. Some unexpected features are revealed when we carefully trace the process to saturation. First, we notice that under an increasing magnetic field, ρ_a and M do not saturate at the same field, as one would expect from Eq. (1). The Hall resistivity ρ_a always reaches saturation before the magnetization M does. The difference increases with the decrease of temperature and film thickness, and becomes too big

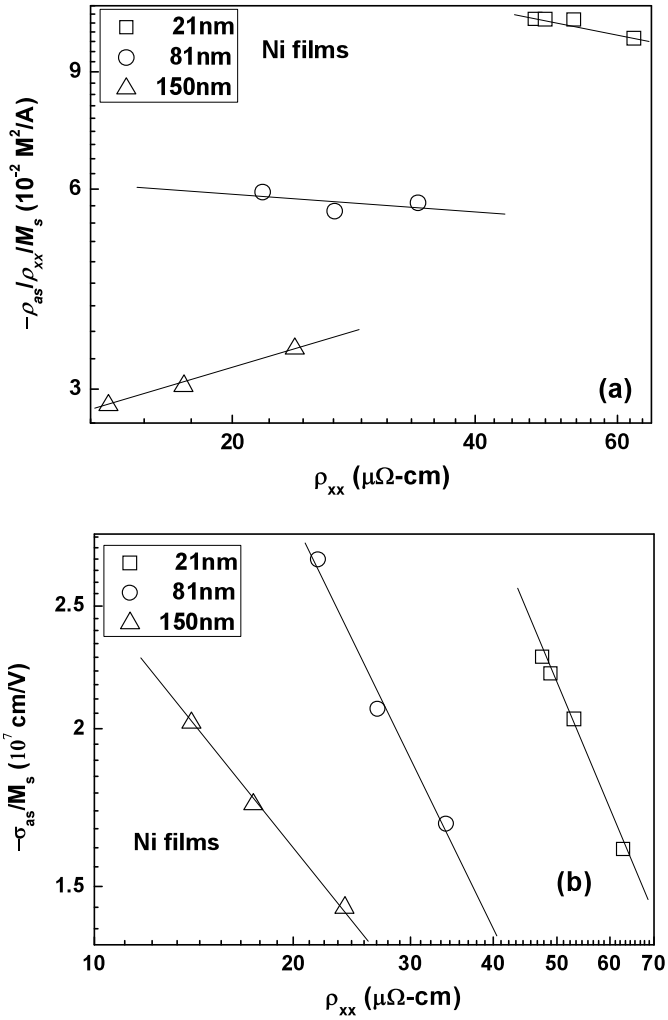

Fig. 2. Variation of the Hall resistivity (a) and the Hall conductivity (b) with temperature, plotted against the longitudinal resistivity, for different film thicknesses. The skew scattering model expects a constant ratio of ρ_{as}/ρ_{xx} for each sample. The Berry phase picture expects a constant Hall conductivity σ_{as}. The deviation of the experimental data from theory in the past has been attributed to surface scattering.

to neglect for thin films at low temperatures (Fig. 3). In addition, although the Hall resistivity approaches saturation faster than the magnetization near the saturation field, we found that for some of the samples under low fields the hysteresis loop for ρ_a can be much smaller than the loop for M, as shown in Fig. 3b.

The difference in the hysteresis loop is also reflected in very different coercive fields for ρ_a and M. Indeed, we find that the coercive field for M in most cases is larger than that for ρ_a (Fig. 4). The difference becomes significant with the increase of film thickness. For the film of 150 nm, even the temperature dependence appears to be opposite of each other (Fig. 4c).

These observed features demonstrate strong nonlinearity between ρ_a and M. To see how the deviation from linearity develops, we plot $(1-\rho_a/\rho_{as})$ as a function of $(1-M/M_s)$ on log–log scale in Fig. 5. This plot accentuates the part of the data close to saturation, when either ρ_a/ρ_{as} or M/M_s are close to 1. The straight line indicates where a linear relationship $\rho_a/\rho_{as} = M/M_s$ would be. In all cases there is a significant deviation from the linearity. The deviation becomes larger with either the decrease of temperature (Fig. 5a), or the decrease of film thickness (Fig. 5b), both having almost the same effect. This is quite different than the case for the longitudinal resistivity ρ_{xx}, where lowering the temperature has the opposite effect than decreasing the film thick-