Anomalous Hall effects in amorphous $Ni_{74}Mn_{24}Pt_2$ film

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Hall resistivity and magnetic measurements for the amorphous $Ni_{74}Mn_{24}Pt_2$ film have been carried out as a function of magnetic field up to 120 kOe in a wide temperature range. The anomalous Hall coefficient, R_s, the ordinary Hall coefficient R_0, the total hysteresis width of the Hall resistivity ΔH are deduced for several temperatures in the temperature range of 1.5–150 K. The Hall voltage was observed in the zero external fields at the temperature below $T=10$ K for both zero field cooling (ZFC) and field cooling (FC) cases. The Hall resistivity hysteresis curves become completely symmetric with respect to the field axis at the temperatures above 15 K where the unidirectional fields lost its rigidity All these anomalous effects have been explained in terms of asymmetric spin-orbit scattering of the conduction electron, which are polarized to the direction of the unidirectional exchange field. It is concluded that the surface becomes dominant at low temperatures. This assertion has been supported by the susceptibility measurements.

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I. INTROTODUCTION

The Hall Effect in magnetic materials is commonly described by the phenomenological equation,

$$\rho_H = R_0[H_a + 4\pi(1 - N)M] + R_s 4\pi M, \tag{1}$$

where ρ_H is the Hall resistivity, H_a is the applied magnetic field , N is the demagnetization factor, R_0 is the ordinary Hall coefficient (OHE), R_s is the extraordinary (or anomalous) Hall coefficient (AHE). In the case of the perpendicular measurement, the value of N is unity. R_0 is related to the Lorentz force, acting on moving free charge carriers while R_s arises from the asymmetric scattering of the conduction electrons by the localized moments. It is widely accepted that R_s consists of two terms, namely the skew scattering[1,2] and side jump.[3] The former is proportional to the longitudinal resistivity, only being dominant in highly pure materials, whereas the latter is known as a side-jump effect, which is proportional to ρ^2 (ρ is the longitudinal resistivity), and becomes dominant in paramagnetic or ferromagnetic amorphous alloys[4] due to the large resistivity and the spin-orbit interaction of 3*d* or 4*d* electrons of transition-metal alloys. The slope of above saturation is given by the following equation:

$$\partial\rho_H/\partial H_a = R_0 + R_s 4\pi \ \partial M/\partial H_a = R_0 + R_s 4\pi\chi_{hf}, \tag{2}$$

where χ_{hf} is the high-field susceptibility.

The ferromagnetic $Ni_{1-x}Mn_x$ alloy system ($x<0.18$) provides a good example for the anomalous Hall effect of ferromagnetic metals, owing to a large spin-orbit interaction.[5] Olivier Jaoul[6–8] has demonstrated that the contribution of each mechanism mentioned above to AHE for the Ni based alloys depends on the impurity atoms. For example, for NiMn alloys, the contribution of the side-jump scattering is very small compared to that of the skew scattering, whereas in the contrary for NiRh the side-jump mechanism dominates over the skew scattering, even for the dilute alloys. We will not discuss the origin of the AHE. Rather, we present the anomalous part of the Hall effect that arises from the spin-orbit coupling between the conduction electrons and the localized moments in the resonant scattering by the virtual bound states.[9]

So far, a number of studies, such as ESR,[10] resistivity,[11,12] magnetoresistance,[13] have been realized on the amorphous $Ni_{74}Mn_{24}Pt_2$ film. The most characteristic features observed for this sample are the following: The resonance spectrum at $T=5$ K has a very broad line when the magnetic field is applied in the forward direction, but there is a noticeable peak for the reverse direction (see Ref. 10, Figs. 6 and 7). Another interesting result from the ESR study is that the surface unidirectional anisotropy field is of the order of or greater than that of the bulk exchange field, meaning that there is a very strong pinning on the film surface. The resistivity of this sample exhibits a shallow minimum at about 30 K following a sharp increases in the resistivity below $T=20$ K, indicating that the spin disorder within the mean free path of conduction electrons becomes severe (please see Ref. 12, Fig. 5). The other important result is concerned with the spontaneous resistivity anisotropy (SRA) observed at $T=4.2$ K for this sample such that SRA was found to be about 0.1%, owing to a large spin-orbit effect (please see Ref. 13, Fig. 6). Therefore, the Hall measurements made on the same sample will also enable us to interpret our previous (already mentioned) experimental findings at low temperatures.

II. EXPERIMENTAL

The amorphous $Ni_{74}Mn_{24}Pt_2$ film alloys were prepared by using the flash evaporation technique. The details of this technique were described elsewhere.[10] The thickness of these films is about 2500 Å. The film thickness was determined by means of a DEKTAL profilometer

The Hall resistivity ρ_{xy} ($\rho_{xy}=tR_H=tV_H/I$, where V_H is the Hall voltage, I is the current, and t is the thickness of the film) was measured using the ac-conventional four-terminal method with the applied field perpendicular to the film surface. The offset voltage due to the asymmetric Hall terminals

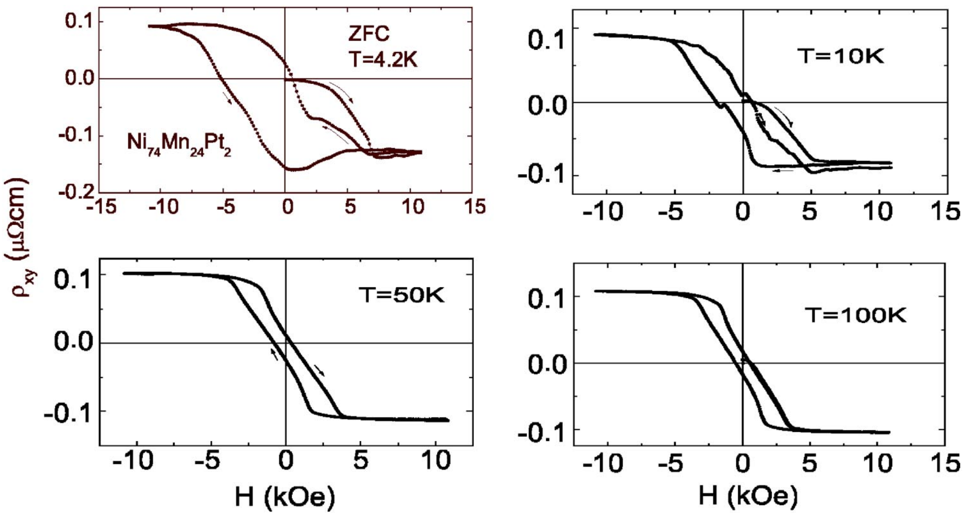

FIG. 1. The Hall resistivity ρ_{xy} as a function of applied field H at some selected temperatures. Note that the hysteresis loops become asymmetric at lower temperatures.

was compensated by an inductive voltage divider. Voltage accuracy was better than one part in 10^7. The magnetic field (0–16 T) was supplied using a superconducting magnet. The temperature was monitored and controlled via a carbon glass thermometer to within an estimated accuracy of 0.01 K below 40 K and 0.05 K above 100 K. The sign of the Hall coefficient was determined by a dc technique at room temperature.

The magnetic field which corresponds to the intersecting point of the initial slope with this linear extrapolation indicates the value of $4\pi M_s$. However, this value is uncertain because of anisotropy fields. The rigid component of the unidirectional anisotropy goes to zero at about 20 K. $4\pi M_s$ is estimated to be 3.75 kOe using the ρ_{xy} vs curve at 20 K. We have also determined the values of $4\pi M_s$ from the magnetic measurements, which are more or less the same as the estimated values from the Hall measurements. Thus, the value of $R_s 4\pi M_s$ is evaluated by extrapolation from the linear portion to zero magnetic fields. As for the R_0, from Eq. (2) its value involves the contributions from the spontaneous Hall coefficient R_s and high-field susceptibility χ_{hf}. However, the latter contribution is not remarkably large. We, therefore, have neglected this contribution, which is within the accuracy of the measurements.

All the magnetization measurements were done (at the Department of Physics, Texas A&M University) with a quantum design semiconducting quantum interference device magnetometer with a dc field perpendicular to the film surfaces.

III. RESULTS AND DISCUSSION

Figure 1 shows the Hall resistivity ρ_{xy} as a function of applied magnetic field H at some selected temperatures for ZFC cases. It is obvious from this figure that the ρ_{xy} vs H loops at 4 K and 10 K considerably differ from those at a higher temperature in some respects. The initial Hall resistivity curves increases slowly first up to about 3 kOe and 0.5 kOe at 4 K and 10 K, respectively and then increases rapidly and finally level off at just above 5 kOe. The hysteresis curves become asymmetric with respect to the field axis and broader at lower temperatures. Figure 2 is emphasized on the total hysteresis widths of these loops as a function of temperature. As seen in Fig. 2, the hysteresis continues to increase for the FC case as the temperature is further decreased, whereas it decreases drastically for the ZFC case and passes through a maximum at about 4 K. We have handled the ρ_{xy} vs H loops at 1.5 K (our lowest temperature) exclusively. Figure 3 shows the ρ_{xy} vs H loops at 1.5 K for both the FC and ZFC cases. As seen in the figure, the highly asymmetric behavior is the most characteristic and striking part of these curves. This anomalous Hall resistance (AHE) is proportional to the magnetization of the sample. In order to check the consistency of the AHE field dependence with the magnetization, the M vs H cycles (see Figs. 4 and 5) were recorded for both FC and ZFC cases at 2 K (which is the available lowest temperature) on the same film for perpendicular geometry (meaning that the applied field is perpendicular to the film surface as done for Hall effects measurements). All curves are completely symmetric. The emphasis was given to the low fields in the insets of these figures. The values of coercivity for forward field and re-

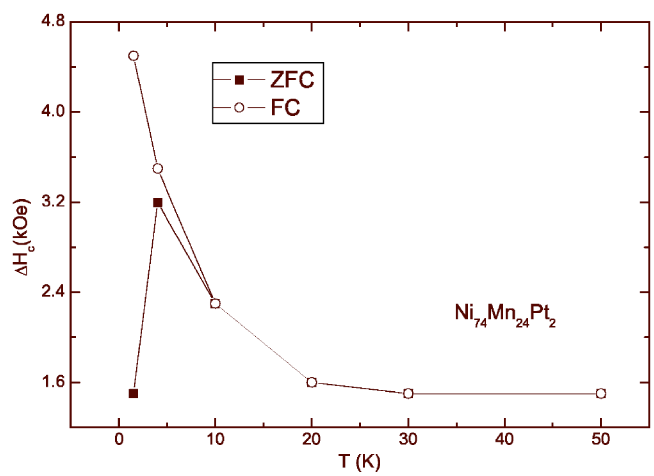

FIG. 2. The total hysteresis width of the ρ_{xy} vs H loops as a function of temperature. Note that the hysteresis for the FC case is significantly larger than that of the ZFC case.

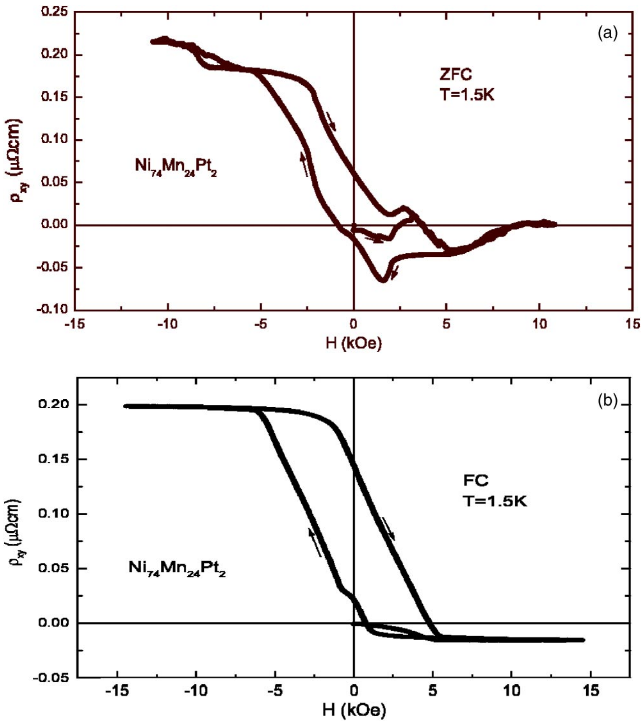

FIG. 3. (a) For the ZFC case, the Hall resistivity as a function of applied field at $T=1.5$ K. (b) For the FC case. The sample is cooled in a field of 8 kOe from 150 K to 1.5 K and then the Hall resistivity is recorded. Note that both ρ_{xy} vs H loops are asymmetric with respect to the field axis.

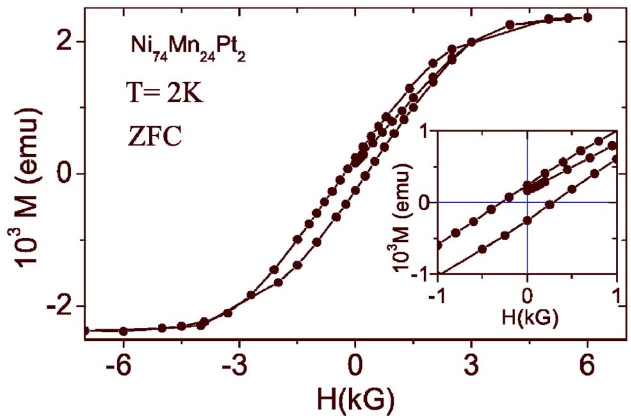

FIG. 4. Magnetic hysteresis of the amorphous $Ni_{74}Mn_{24}Pt_2$ film with an applied field perpendicular to the film surface for the zero field cooling case (ZFC) at 2 K. The inset shows the same hysteresis and the coercivity in the extended scale.

versed field directions, H_c^+ and H_c^-, are found to be 285 Oe and −295 Oe, respectively. The total hysteresis width, ΔH^c was obtained from the relation $\Delta H^c=\frac{1}{2}(H_c^+-H_c^-)$ and, therefore, $\Delta H^c=580$ Oe for the ZFC case. As for the FC case, H_c^+ and H_c are 140 Oe and −460 Oe, respectively. The rigid component of anisotropy H_a from the relation $H_a=\frac{1}{2}(H_c^+ +H_c^-)$ is found to be 300 Oe. It should be noted that the thermo-remnant magnetization for both cases is not considerably large enough to explain the asymmetry of the Hall voltage at this temperature. Keener and Weissman[14] have carried out a scanning electron microscopy (SEM) study on the partially ordered $Ni_{1-x}Mn_x$ films and observed ferromagnetic domains arranged antiferromagnetically. The average width of domains in different regions was constant (6.7 ± 0.2) nm. This structure accounts quite well for this small observed remnant when keeping in mind that the thickness of these films is about 250 nm. The displaced hysteresis cycle observed in the FC case can be attributed to the rigidity of the anisotropy field because turning strongly correlated spins' groups rotate as a whole against the local unidirectional anisotropy field coupled to the lattice due to anisotropic DM interactions . It has been recently shown that such a coupling would lead to a significant coercivity enhancement at low temperatures.[15] In fact, vector magnetization measurements,[16,17] torque measurements,[18] ESR studies[10] and also transverse susceptibility measurements[19] performed on NiMn samples showed that the anisotropy observed in these systems is purely unidirectional and rotates elastically at lower temperatures, but dissipatively at higher temperatures, resulting in larger hysteresis. However, Parker and Saslow[20] approached this subject by a different view (local mean field) and suggested that the irreversibility effects could arise from the onset of canting and spin disorder in a microscopic scale due to the interaction of defect bonds. I do not wish to discuss this subject further but it appears that the hysteresis observed in the Hall resistance is associated with the irreversibility in the microscopic scale while the hysteresis observed in the magnetization mainly stems from the macroscopic anisotropy fields. The temperature dependence of the low field susceptibility for the same perpendicular geometry was also represented in Fig. 6. The susceptibility passes a broad maximum at about 55 K where the irreversibility sets in. The ZFC branch stays below the FC branch as commonly seen in spin glasses. However, please note that both FC and ZFC curves have a tendency to turn upward presumably due to the strong surface unidirectional exchange field causing the spin order near the surfaces. We will turn again to this point below.

We wish to turn to the results of the Hall measurements. The total experimental Hall resistance consists of an anomalous Hall resistance and a normal Hall resistance as mentioned above. The two ferromagnetic Hall coefficients, R_s and R_0, have been measured in this film for several states of order using fields up to 120 kOe. Both R_s and R_0 are negative. It is not surprising because amorphous alloys due to a disordered structure are expected to have a spherical Fermi surface and therefore to be nearly free-electron like.[21] Since the normal Hall resistance is linear in the magnetic field [see Eq. (2)], the values of R_0 have been estimated from the slope of the ρ_{xy} vs H loops at higher fields. Some typical ρ_{xy} vs H curves at 1.5 K, 50 K, and 150 K are given in Fig. 7 allowing us to make a comparison between them. Indeed, one might expect that the normal Hall resistance should be temperature independent because the sample is amorphous, the Curie temperature (much above the room temperature) is much higher than our maximum measuring temperature ($T=150$ K). It would be satisfying if I could know with a high precision the values of $R_s 4\pi\chi_{hf}$ for each temperature. However, this second term on the right of Eq. (2)

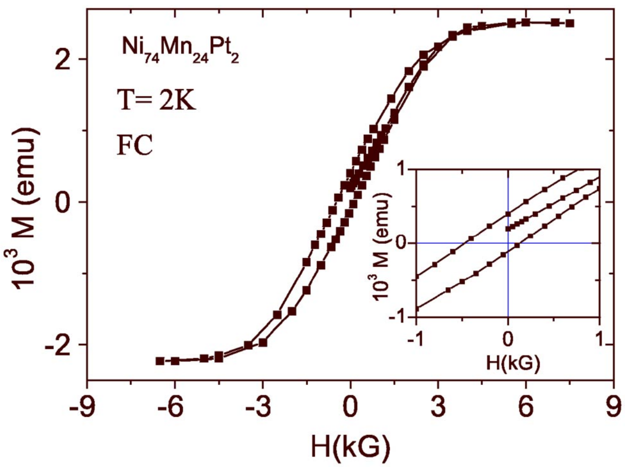

FIG. 5. Magnetic hysteresis of the amorphous $Ni_{74}Mn_{24}Pt_2$ film with an applied field perpendicular to the film surface for the field cooling case (ZFC) at 2 K. The sample was cooled in a field of 8 kOe from 150 K to 2 K before taking data. The inset shows the same hysteresis and the coercivity in the extended scale. Note that the hysteresis as a whole is shifted to the negative field side due to the unidirectional anisotropy field.

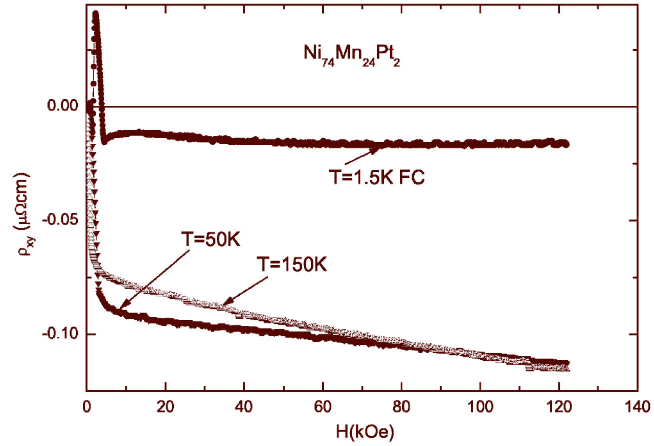

FIG. 7. The Hall resistivity ρ_{xy} as a function of applied field H up to 120 kOe at 1.5 K, 50 K, and 150 K. Note that Hall resistivity increases linearly with the applied field and the slope of its linear part decreases with decreasing temperature. At 1.5 K, the Hall resistivity becomes almost temperature independent at a higher field, meaning that the normal Hall coefficient goes to zero. Both R_s and R_0 are negative as indicated in the text.

was estimated from the magnetization data at 2 K to be of the order of $10^{-9}\mu\Omega$ cm/Oe, which is at least two orders smaller than those of R_0. The high field susceptibility contribution to the Hall resistance can be easily neglected in our temperature range. The absolute values of R_0 decreases gradually from $-4.3\times10^{-7}\mu\Omega$ cm/Oe at $T=150$ K to $-1.3\times10^{-7}\mu\Omega$ cm/Oe at $T=30$ K and then stay constant in between 10–30 K following a drastic drop in R_0 below $T=10$ K. Please note that the normal Hall resistance is almost field independent (see Fig. 7) at 1.5 K and a close inspection of this figure shows that it deviates slightly upward (to the negative direction of ρ_{xy}) at higher fields. But this deviation is too small to take it into account. However, R_s usually depends on the resistivity ρ. Karplus and Luttinger,[22] based on the band model for example, predicts $R_s\sim\rho^2$. As mentioned above, our recent resistivity measurements on the same sample exhibit a resistivity minimum at around 30 K. The increase in the resistivity below the resistivity minimum was about 1.5%. This slight deviation may be attributed to the second term on the right of Eq. (2) $(R_s 4\pi\chi_{hf})$. As for the drastic decrease in R_0 at low temperatures, it may be associated with the decrease *s*-like electron asymmetrically scattered from the orbits of localized moments into *d* states of conduction electrons.

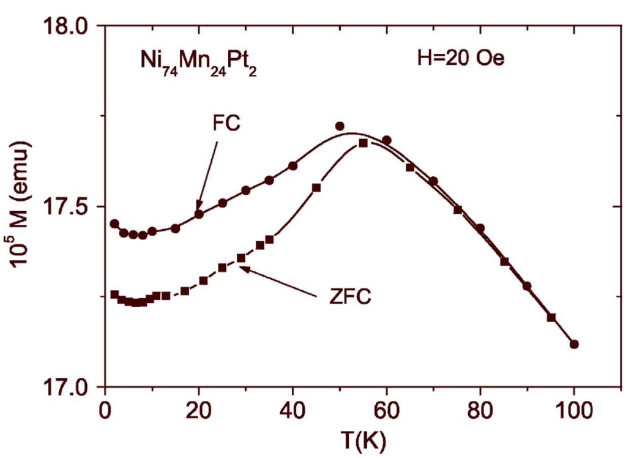

FIG. 6. Magnetization measurement data were taken with the measuring field of 20 Oe while heating and warming, after cooling the sample down to 2 K in the Earth's field. The warming and cooling curves are indicated by ZFC and FC, respectively. The magnetization data split into two branches at about 55 K setting on irreversibility effects. Note that both magnetization curves have a tendency turn upward at about $T=10$ K due to surface dominancy over the macroscopic magnetization.

The extraordinary Hall resistivity can be determined by a linear extrapolation of the data at high fields to $H=0$. Using this method, the saturated Hall resistivity were obtained. As mentioned above, it is necessary to determine $4\pi M_s$ from magnetic measurements. Since the Curie temperature is much higher than the room temperature, this value was supposed to be constant in our measuring temperature range. From magnetic data at 2 K and the ρ_{xy} vs H loops at 30 K where the rigidity of unidirectional anisotropy field almost disappears, the value of $4\pi M_s$ was estimated to be 3.75 kOe. The values of R_s were obtained from the saturated Hall resistivity divided by 3.75 kOe and lies between $-2.1\times10^{-5}\mu\Omega$ cm/Oe and $-2.3\times10^{-5}\mu\Omega$ cm/Oe in the temperature range of 20–100 K. This value decreases to $-1.9\times10^{-5}\mu\Omega$ cm/Oe at 150 K. At the temperatures below 10 K, the asymmetry of the ρ_{xy} vs H loops does not permit us to determine the values of R_s. The Hall voltage has a unidirectional component (meaning that the contribution along the cooling field) and the origin of this behavior will be discussed further under the light of the results presented briefly above.

Consider now the situation where the film is in the demagnetized state. In the absence of the external field, domains that are randomly arranged throughout the sample give no net skew scattering component on a macroscopic scale. We should also point out that the magnitude of the skew scattering reflects the combination of two features: the strength of the resonance between the virtual bound states[9] and the itinerant electrons within $\sim k_B T$ of the Fermi energy, and the degree of magnetic alignment produced in the spin systems by the applied field. At low temperatures where the unidirectional surface anisotropy field dominates the bulk magnetization for this sample, the unidirectional anisotropy fields polarize the conduction electrons and create an imbalance between spin-up and spin-down electrons over all surface or near the surface. Since the surface anisotropies for this sample are not identical at the both surfaces,[10] the two contributions do not exactly cancel each other and yields a Hall voltage even in the zero external magnetic field and, subsequently, an asymmetric Hall resistivity hysteresis loop. At higher temperatures, the bulk magnetization becomes dominants and we observe an usual symmetric ρ_{xy} vs H loop.

IV. CONCLUSION

A systematic study of the Hall resistance measurements on the amorphous $Ni_{74}Mn_{24}Pt_2$ film have been carried out up to the field of 120 kOe in the temperature range of 1.5–150 K. The ρ_{xy} vs H Hall resistivity loops become asymmetric below 10 K. This asymmetric behavior can be explained in terms of the generation of spin imbalance created due to the unidirectional anisotropy on and near the film surface and the existence of a skew scattering mechanism in this film. We suggest that the spin-orbit interaction, responsible for the unidirectional exchange anisotropy of spin-glass materials, may give rise to local unidirectional anisotropy on the surface sufficient in magnitude to polarize the conduction electrons. As the temperature is decreased, the surface becomes dominant over the transport properties. Since the surface anisotropies are not identical at the both surfaces in this film as pointed out in our previous ESR study,[10] the polarization of the conduction electrons on the surfaces due to the asymmetric surface anisotropy fields reflects itself on the Hall resistivity yielding an asymmetric ρ_{xy} vs H loop with respect to the field axis. Indeed, at the temperature below 10 K, the anomalous Hall coefficient for the reversed field direction increases, the ordinary Hall coefficient decreases drastically. The hysteresis in the Hall resistivity increases as well. This is supporting evidence for the existence of the strong microscopic irreversibility at lower temperatures, which is also responsible partly for the increase of the resistivity of this sample.

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