Correlation of uniaxial magnetic anisotropy axes and principle resistivities in polycrystalline ferromagnetic films

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Abstract

In the present study, we demonstrate resistivity ($\rho$) measurement along the magnetic axes of a polycrystalline film of ferromagnetic permalloy. To this end, conventional Hall-bar and more recent extended van der Pauw methods were utilized for determining the 2D matrix of $\rho$ in the film plane. The samples were prepared by normal incidence sputter deposition within an in-situ magnetic field to induce in-plane uniaxial magnetic anisotropy in the film. Since $\rho$ measurements might be affected by the internal magnetization of the film, we preferred to use average resistivity obtained by rotation of saturating magnetic field in the film plane. Both methods indicate that the average resistivity is lower along the easy axis of the film compared to the hard axis. Since X-ray diffraction indicated no dominating texture in the film, we attributed the difference along hard and easy axes to the ordering of Ni-Fe pairs along the hard axis which causes higher scattering while Fe-Fe and Ni-Ni are aligned with the easy axis that presents limited scattering. Thus, measuring the in-plane resistivities enables determining the direction of magnetic anisotropy axes which can be applied to a wide range of compounds. In this regard developing fast and facile methods of resistivity measurement, such as extended van der Pauw utilized here, is of the prime of importance.

Keywords: Magnetic anisotropy, Atomic ordering, Resistivity tensor, van der Pauw, Hall-bar.

1. Introduction

It has been shown that magnetic anisotropy is a tunable parameter through the alloy composition \cite{1}, atomic order \cite{2} or in ultra-thin layers through epitaxial strain and interface mixing \cite{3}. Thus numerous methods have been developed to tune magnetic anisotropy in the ferromagnetic materials. However, the origin of induced uniaxial anisotropy, even in a popular ferromagnet such as permalloy Ni$_{80}$Fe$_{20}$ at. % (Py), has been a subject of huge debate over decades (cf. Refs \cite{4,2,5} and references therein). It has been thought that self shadowing and off-normal texture are responsible for the uniaxial anisotropy induced by in-situ deposition \cite{6,7}. We have already shown that uniaxial anisotropy can be achieved in very smooth (no-self shadowing) Py films with normal texture by in-situ deposition \cite{4}.

A more conventional way of inducing uniaxial anisotropy is by post annealing or growth in a magnetic field. Bozorth \cite{8,9} believed that applying an external magnetic field causes magnetostrictive deformation which during annealing in a field becomes permanent. The uniaxial anisotropy induced by in-situ magnetic field during the growth can also be explained by the same interpretation. It has been shown that both post annealing \cite{10} and growth \cite{11} in the magnetic field showing orientation dependency for the single crystal Py. In an effort to understand such a complication, turned out Py has a negligible magnetostriction i.e. its elastic energy is two order of magnitude smaller than anisotropic energy \cite{12}. Thus other effects such as the crystalline anisotropy can easily overcome magnetostriction. We have already shown that when the tilt deposition competes with a magnetic field the magnetic axes of polycrystalline film is dictated by tilt effect \cite{13}. More recently we showed that tilt effect can dictate magnetic axis along the [100] orientations of Py single crystal \cite{2} while both annealing and growth in the field failed to do so \cite{10,11}. We believe in both of single and polycrystalline Py, it is disordered arrangement of Ni and Fe that becomes dominant effect and leads to uniaxial anisotropy in Py. While in a ordered Py, crystalline anisotropy becomes dominant \cite{2}. Since Py exhibits a shrinkage upon ordering, the level of order in a single crystal can be analyzed by X-ray diffraction (XRD). In polycrystalline films, however, grain size and defect induced strain effectively contribute to the broadening of XRD peaks and thus limit detection of atomic order.

In the present study, we aim to understand the origin of uniaxial anisotropy induced by normal deposition with in-situ magnetic field during the growth. In order to minimize the contribution of magnetocrystalline anisotropy we grow nanocrystalline films. In this case, however, it is not trivial to detect atomic order by XRD as mentioned above. To solve this issue, we utilize resistivity measurement which strongly correlates with the atomic arrangement of Fe and Ni in Py. We show the principle resistivity axes are aligned with magnetic axes of the film which can be only explained by the atomic order.

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2. Experimental Method

The substrates used here were p-Si (001) with 100 nm thick layer of thermally grown oxide. After proper cleaning, the substrate were dehydrated at 140 °C and their surface modified with HMDS vapor to become hydrophilic prior spin coating of photoresist. Then the samples were exposed to deep UV with HMDS vapor to become hydrophilic prior spin coating.

The films were prepared using Py target in normal incidence deposition geometry and utilized a conventional method of applying an in-situ magnetic field (70 Oe) at the substrate to induce uniaxial anisotropy in the desired direction. The Hall-bar (0.4×1.6 mm²) and vdp (15x15 mm²) samples were grown simultaneously to make sure there is no difference between them from magnetic and microstructure viewpoint. Further detail on the preparation can be found in Ref. [4].

The film thickness and grain size were characterized by X-ray reflectivity (XRR) and grazing incidence X-ray diffraction (GIXRD), respectively. To obtain hysteresis loops, we used a high sensitivity magneto optical Kerr effect (MOKE) looper.

The resistance was measured by two conventional Hall-bar and vdp methods for comparison which is schematically shown in Fig. 1. The vdp method is simple and flexible technique to probe resistivity of a uniform, continuous thin films of arbitrary shape [14]. In the vdp method, four small contacts must be placed on the sample perimeter, not necessarily at the corners labeled A–D in Fig. 1. The isotropic resistivity value (ρiso) is obtained by the vdp formula:

\[ \exp\left(- \frac{\pi d}{\rho_{\text{iso}}} R_{\text{AB,CD}}\right) + \exp\left(- \frac{\pi d}{\rho_{\text{iso}}} R_{\text{AD,BC}}\right) = 1 \]  

(1)

where d is the film thickness and e.g. \( R_{\text{AB,CD}} \) is the resistance obtained by forcing current through AB and picking up the voltage at the opposite side between CD or vice versa.

Since (1) obtained by conformal mapping of a finite sample into an infinite half plane with contacts along the edge, it should be valid if e.g. an anisotropic rectangle with the lateral dimensions of \( a \times b \) is mapped into an isotropic one with \( a \times b' \). It has been shown that \( \rho_{\text{iso}} = \sqrt{\rho_{\parallel} \rho_{\perp}} \) is the geometric mean of the principle resistivities [15, 16]. We have already shown it is possible to determine the whole resistivity tensor in an arbitrary direction by extension of vdp [17]. Here, we use Price [16] extension for a rectangular sample with its sides cut along the \( \rho_1 \) and \( \rho_2 \), which better suits to be compared with Hall-bar method.

\[ \sqrt{\frac{\rho_1}{\rho_2}} = -\frac{b}{\pi a} \ln\left( \tanh\left( \frac{\pi d R_{\text{AD,BC}}}{16 \rho_{\text{iso}}} \right) \right) \]  

(2)

where \( b \) and \( a \) are the side lengths of a rectangular sample and \( R_{\text{AD,BC}} \) is resistance along the \( b \) sides as described above. The individual values of principle resistivities can subsequently be obtained using (2):

\[ \rho_1 = \rho_{\text{iso}} \sqrt{\frac{\rho_1}{\rho_2}} \]  

(3)

\[ \rho_2 = \rho_{\text{iso}} \sqrt{\frac{\rho_2}{\rho_1}} \]  

(4)

To decouple effect magnetization on resistivities we have rotated \( \mathbf{M}_{\text{sat}} \) to extract \( \rho_{\text{vec}} \). Thus the individual resistivities are expected to behave as below:

\[ \rho_{\text{long}} = \rho_{\perp} + \Delta \rho \cos^2 \phi \]  

(5)

here \( \phi \) stand for angle between current and saturated magnetization direction, \( \rho_{\text{long}} \) is the longitudinal resistivity with respect to current and \( \Delta \rho = \rho_{||} - \rho_{\perp} \), with \( \rho_{||} \) and \( \rho_{\perp} \) being resistivities with \( \mathbf{M}_{\text{sat}} \) parallel and perpendicular to the current direction, respectively.

The magnetoresistance measurements were done by rotation of a in-plane \( \mathbf{M}_{\text{sat}} \) of ~23 Oe. The strength of field is enough to saturate the magnetization as it is a few times of larger than both coercivity (\( H_c \)) and anisotropy field (\( H_k \)). All resistivity measurements were performed at room temperature using a low current density of 3 mA.

3. Results and discussion

The result of XRR measurements (not shown here) fitted according to the Parrat formalism [18] to determine film thickness, density and surface roughness. The thickness of the film, which is required to solve (1), is found to be 40 nm. We used Scherrer equation [19] to estimate the grain size from the (111) GIXRD peak (not shown here) which is found to be ~10 nm. Note that the grain size obtained by Scherrer equation from GIXRD has been shown to be in agreement with the TEM result for thin Py films [20]. We could not detect any peak shift due to order/disorder [2] since such a small grain size causes considerable peak broadening. Further, we studied texture in the film by polar mapping of (111) plains that indicates the lack of any texture in film. This is the case for very thin film in
which different crystal planes are still competing and none became dominant and thus grains are essentially equiaxed. Thus, the XRD characterization indicates the film is isotropic.

Fig. 2 shows the MOKE response of as deposited films before lift-off. It can be seen that the hard axis presents a completely linear behavior without hysteresis and $H_k$ of 5 Oe. The easy axis presents a square loop with slight rounding at the corners but still sharp switching and $H_c$ of 2.75 Oe. These indicate the film presents very well defined in-plane uniaxial anisotropy magnetization induced by applying an *in-situ* magnetic field during the growth. The dimensions of Hall-bars is chosen to be large enough to maintain induced magnetic anisotropic axes after lift-off.

Fig. 3 shows the variation of resistivity with the rotation of magnetization for Hall-bar patterns made with 0, 30 and 90° with respect to the easy axis. Here $\theta$ is defined as the angle between saturated magnetization and easy axis and should not be confused with $\phi$ i.e. the angle between magnetization and current directions (cf. Fig. 1). It can be seen that the absolute value of resistivity and consequently $\rho_{ave}$ increases as we rotate the Hall-bar from 0 to 90°. The anisotropic magnetoresistance (AMR) behavior is evident for all cases i.e. a maximum resistivity when magnetization is parallel to the current direction and decreases by rotation away from Hall-bar axis. The AMR measurement by Bozorth method [21], is performed by applying $M_{sat}$ parallel and perpendicular to the current direction which is expected to be independent of the the direction in a polycrystalline film. Comparing different hall-bars, we observe the difference in both AMR value and absolute value of resistivity. The result of Eq. (5) also shown for each Hall-bar as a solid line. It can be seen that Eq. (5) gives much better estimation along the easy (0) and hard (90°) axes while for the 30° Hall-bar the agreement is quite poor.

Note that the Hall-bars were fabricated large enough to make sure their geometry does not affect initial magnetic anisotropy. Further, we measured anisotropic resistivity using our extended

![Figure 2: The MOKE response of the as-deposited film along hard and easy axis.](image)

![Figure 3: Variation of $\rho_{long}$ with the rotation of $M_{sat}$ for Hall-bars patterned with (a) 0, (b) 30 and (c) 90° with respect to the easy axis. The solid and dashed lines indicate fitting with Eq. 5 and average of $\rho_{long}$, respectively.](image)
vdP method that can be applied to square-shaped samples \[4\]. Fig. 4 shows variation of different resistivities obtained using the vdP method by rotation of saturated magnetization. It can be seen that original vdP gives \(\rho_{iso}\), which does not change with the rotation of magnetization and thus it is not appropriate for AMR measurements on its own. On the other hand, \(\rho_1\) and \(\rho_2\) obtained by \[5\] and \[4\], change symmetrically around the \(\rho_{iso}\). This explains why the rotation of saturated magnetization does not change \(\rho_{iso}\). It is also clear that both \(\rho_1\) and \(\rho_2\) are characteristic AMR curves and can be used to determine AMR along the hard and easy axes, respectively. Similar to Hall-bars, different AMR and absolute value of resistivity is obtained along the hard and easy axis. Again, the \(\rho_{ave}\) along the hard axis is higher than that along the easy axis indicating existence of a correlation between magnetic anisotropy axes and principle resistivities.

In the semiconductor and superconductor community the resistivity tensor is commonly obtained by patterning and measurement of two Hall-bars along the principle resistivity axes. This is based on the fact that in an anisotropic film, off-diagonal elements of resistivity tensor become zero when measurements are performed along the principle resistivity axes. Once the principle resistivities (\(\rho_1\) and \(\rho_2\)) are determined one can determine resistivity in any direction using matrix rotation. For instance, we can apply this to calculate \(\rho_{ave}\) for the 30° Hall-bar from 0 and 90° ones which gives a value of 32.36 \(\mu\Omega\) cm. This is in close agreement with the \(\rho_{ave}\) obtained by resistivity measurement in our 30° Hall-bar. This indicates magnetic anisotropy axes of the sample are aligned with the principle resistivity axes. Note that \(\rho_1\) and \(\rho_2\) in our scheme are directions with highest and lowest \(\rho_{ave}\) obtained by rotation of \(M_{sat}\).

The latter indicates magnetostriction and internal magnetization cannot explain the alignment of magnetic anisotropy and principle resistivity axes. The only explanation left is that both of these are correlated with the atomic arrangement models. The latter includes atomic ordering and pair ordering those can be applied to single and polycrystalline films, respectively. We have recently shown that an increase in the atomic order towards L1\(_2\) Ni\(_3\)Fe superlattice changes uniaxial anisotropy into four-fold (biaxial) anisotropy and decreases resistivity \[2\]. In the pair ordering model, the direction of Ni-Ni, Fe-Fe and Ni-Fe pairs determines magnetic anisotropy \[22\]. Note that, in a fully ordered structure there exist only Ni-Fe pairs and moving away from order results Ni-Ni and Fe-Fe pairs accompanied with the development of uniaxial anisotropy. Since in-situ magnetic field during the growth aligns Ni-Ni and Fe-Fe pairs parallel to the field it gives more Ni-Fe pairs in perpendicular direction (cf. Fig. 5(a)). Thus, along the easy axis electrons are traveling through a more uniform medium while Ni-Ni and Fe-Fe sequence along the hard axis results more scattering.

A question that might arise here is why the result of measurement using the Hall-bars and vdP are not the same. Most of critics to vdP focus on the fact that it probes the whole plane of the sample not a specified direction. This has been already answered mathematically for the co-liner and square four-point-probes techniques and we refer interested reader to e.g. Ref \[23\]. We considered source of errors to be patterning in the Hall-bars and size and placement of contacts in the vdP method. Although, we have tried to pattern and grow all samples together, there might be little difference between different Hall-bars. But we reduced measurement error in the Hall-bars by utilizing 8-pad pattern. In such pattern two pads utilized for applying current and 6 remaining pads allows 6 combination for measurement. We measured all 6 combination and rejected those with more than 5% difference in averaging. Original vdP requires infinity small contacts for arbitrary plane \[14\]. Using four 2 mm contacts, spread into the film, a \(~2\%\) error was obtained compared to 0.2 mm contacts. The extended vdP is based on rectangle/square geometry that might be affected by contact displacement. For displacing one contact equal to \(a/2\) or \(b/2\) (cf. Fig. [1], i.e. having a contact at sides rather than corner of square, an error of 7 % was produced. Thus, we attribute the difference between Hall-bars and vdP measurement to the patterning.
4. Conclusion

Py film without any texture is prepared by sputtering in presence of a magnetic field to induce in-plane uniaxial magnetic anisotropy. It is shown that the $\rho_{ave}$ obtained by rotation of $M_{sat}$ using both Hall-bars and vdP methods is higher along the hard axis compared to that along the easy axis. This is due to the fact that magnetic anisotropy axes and anisotropic resistivity axes are aligned with each other. This is due to the fact that both of these are correlated with the atomic/pair order. In particular alignment of Ni-Fe pairs along the easy axis present lower scattering rate. While along the hard axis Ni-Ni and Fe-Fe pairs provide less uniform medium and higher $\rho$. Thus, it is possible to determine direction of magnetic axis by measuring the resistivity tensor. In the latter case we suggest using extended vdP as a versatile method that can be performed without patterning.

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