PCCP

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: Y. Zhang, W. Mi, X. Wang and X. Zhang, *Phys. Chem. Chem. Phys.*, 2015, DOI: 10.1039/C5CP01955A.

This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this *Accepted Manuscript* with the edited and formatted *Advance Article* as soon as it is available.

You can find more information about *Accepted Manuscripts* in the [Information for Authors](#).

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard [Terms & Conditions](#) and the [Ethical guidelines](#) still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.

Scaling of Anomalous Hall Effects in Facing-Target Reactively Sputtered Fe₄N Films

Y. Zhang¹, W. B. Mi^{1,*}, X. C. Wang^{2,*}, X. X. Zhang^{3,*}

¹*Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparation Technology, Faculty of Science, Tianjin University, Tianjin 300072, China*

²*Tianjin Key Laboratory of Film Electronic & Communicate Devices, School of Electronics Information Engineering, Tianjin University of Technology, Tianjin 300384, China*

³*PSE Division, KAUST, Thuwal 23955-6900, Kingdom of Saudi Arabia*

* Author to whom all correspondence should be addressed.

E-mail: miwenbo@tju.edu.cn (W.B. Mi); wangxccn@126.com (X.C. Wang);

xixiang.zhang@kaust.edu.sa (X.X. Zhang)

ABSTRACT

Anomalous Hall effect (AHE) in the reactively sputtered epitaxial and polycrystalline γ' -Fe₄N films is investigated systematically. The Hall resistivity is positive in the entire temperature range. The magnetization, carrier density and grain boundaries scattering have a major impact on the AHE scaling law. The scaling exponent γ in the conventional scaling of $\rho_{\text{AH}} \propto \rho_{\text{xx}}^\gamma$ is larger than 2 in both the epitaxial and polycrystalline γ' -Fe₄N films. Although $\gamma > 2$ has been found in heterogeneous systems due to the effects of the surface and interface scattering on AHE, $\gamma > 2$ is not expected in homogenous epitaxial systems. We demonstrated that $\gamma > 2$ results from residual resistivity ($\rho_{\text{xx}0}$) in γ' -Fe₄N films. Furthermore, the side-jump and intrinsic mechanisms are dominant in both epitaxial and polycrystalline samples according to the proper scaling relation.

Introduction

With the rapid development of spintronics, the iron nitrides have attracted considerable interest due to its potential applications in the spintronic devices.^{1–7} Among them, γ' -Fe₄N has been paid much attention as a ferromagnetic electrode. Highly enhanced negative spin polarization of transport electrons in γ' -Fe₄N has been theoretically predicted.⁸ The spin polarization of γ' -Fe₄N(100) lattice measured by point-contact Andreev reflection is 60%.⁹ The γ' -Fe₄N Curie temperature of 767 K makes it a suitable material for practical applications. Recently, the current-induced magnetization switching was reported in the magnetic tunnel junctions (MTJ) with the γ' -Fe₄N free layer.¹⁰ The structure, magnetic properties and tunnel magnetoresistance of the MTJ based on γ' -Fe₄N electrode were also investigated.^{2,3,11,12} Negative anisotropic magnetoresistance (AMR) has been observed in the epitaxial γ' -Fe₄N films due to the spin-down conduction electrons.^{13–15} Meanwhile, a rectangular-like AMR was reported in epitaxial γ' -Fe₄N/CoN bilayers.¹⁶ Up to now, no papers reported the anomalous Hall effect (AHE) of γ' -Fe₄N films.

The AHE in ferromagnetic materials has received intense renewed attention in recent years due to its intrinsic physics related to asymmetric scattering of spin-polarized electrons and technical applications.¹⁷ The Hall resistivity (ρ_{xy})

follows the phenomenological equation

$$\rho_{xy} = R_0 B + 4\pi R_s M \quad (1)$$

where the first term is the ordinary Hall resistivity from the Lorentz force and the second one is the anomalous Hall resistivity (ρ_{AH}) that is proportional to the out-of-plane magnetization of the ferromagnetic film.¹⁸ In order to identify the AHE mechanisms, the scaling law between ρ_{AH} and ρ_{xx} is usually given as¹⁹

$$\rho_{AH} \propto \rho_{xx}^{\gamma} \quad (2)$$

Skew scattering mechanism leads to $\gamma=1$,²⁰ while both scattering-dependent side jump and scattering-independent intrinsic mechanisms yield $\gamma=2$.^{21–23} The intermediate values $1 < \gamma < 2$ accepted as a superposition of these mechanisms. In the recent theoretical investigation on AHE,²⁴ the scaling law of the AHE is expected to depend on the longitudinal resistivity (ρ_{xx}) of the ferromagnetic materials. The magnitude of ρ_{xx} of the γ' -Fe₄N film is about $10^3 \mu\Omega \text{ cm}$ at room temperature, which lies in the middle region between the Fe ($10 \mu\Omega \text{ cm}$) and Fe₃O₄ ($10^4 \mu\Omega \text{ cm}$) films. The AHE of Fe and Fe₃O₄ films has been investigated in details, which is in good agreement with the unified theory of the AHE.^{25,26} However, the origin of AHE in

γ' -Fe₄N films has remained confusing. The clarification of the AHE in γ' -Fe₄N films is of fundamental importance for better understanding of AHE.

In this paper, the AHE scaling in the epitaxial and polycrystalline γ' -Fe₄N films fabricated by reactive facing-target sputtering is investigated in details. We study the effect of magnetization, carrier density and grain boundaries scattering on the AHE scaling in γ' -Fe₄N films. The scaling exponent $\gamma > 2$ has been found in both the epitaxial and polycrystalline γ' -Fe₄N films by the conventional scaling of $\rho_{\text{AH}} \propto \rho_{\text{xx}}^\gamma$, which can be attributed to the residual resistivity ($\rho_{\text{xx}0}$). The different contributions to AHE in γ' -Fe₄N films are also investigated.

Experimental section

γ' -Fe₄N films with different thicknesses t were fabricated on glasses, natural oxidized Si, and MgO(100) wafers by a DC reactive facing-target sputtering method from a pair of pure Fe (99.99%) targets. The sputtering pressure was kept at 1.0 Pa with an Ar (99.999%) and N₂ (99.999%) gas mixture. The Ar and N₂ gas flow rates were fixed at 100 and 20 sccm, respectively. The substrate temperature was set at 450 °C. The film thickness was determined using a Dektak 6M surface profiler and confirmed by the transmission electron microscopy (TEM). The deposition rate was 0.08 nm/sec. The structure was analyzed by X-ray diffraction (XRD) with Cu K_α

radiation (wavelength 1.5406 Å) and high-resolution TEM (HRTEM). The magnetic properties were measured by a Quantum Design superconducting quantum interference device (SQUID). Hall bars were fabricated by using the shadow masks that have five terminals, which can be used to measure transverse and longitudinal resistances simultaneously. The width of the films in the Hall bar geometry is 0.3 mm. Hall loops were measured with a Quantum Design physical property measurement system (PPMS-9) under a magnetic up to 5 T applied perpendicular to the film plane in the temperature range of 5–350 K. The Hall resistivity ρ_{xy} was calculated from $V_{xy}t/I$, where V_{xy} was the transverse voltage, t was the film thickness, and I was the applied DC current. For excluding the contact resistance, the real Hall resistivity was obtained by subtracting the Hall resistivity measured at the negative magnetic fields from that measured at the positive magnetic fields, then divided by 2 because the magnetoresistance is an even function of magnetic field.

Results and discussion

Fig. 1(a) shows the θ - 2θ XRD pattern of the polycrystalline γ' -Fe₄N films on natural oxidized Si wafer. The diffraction peaks located at 33.2° and 69.3° are from Si(200) and (400) lattices. Two strong peaks situated at 41.2° and 47.8° come from the γ' -Fe₄N(111) and (200). A weak peak located at 84.5° is attributed to

γ' -Fe₄N(311). The XRD results of the film on Si suggest that the film is composed of single phase γ' -Fe₄N, and no other phases of iron nitrides appear. Fig. 1(b) shows the XRD θ - 2θ patterns of the γ' -Fe₄N film on MgO(100). It is clear that only the peaks from γ' -Fe₄N(100) and (200) appear, and no other peaks from γ' -Fe₄N are observed, suggesting that the films grow with a preferred (100) orientation. In order to confirm that the preferred oriented films are epitaxial, X-ray φ scan was performed. For the (100)-oriented films, X-rays were collected at $2\theta=41.22^\circ$, $\alpha=5.26^\circ$, where no peaks from MgO appear, and only a peak from γ' -Fe₄N(111) is detected. In the left inset of Fig. 1(b), it is clear that for the (100)-oriented film, 4-fold symmetric peaks with identical intervals appear. The above X-ray θ - 2θ and φ scan suggest that the γ' -Fe₄N films on MgO(100) substrate are epitaxial and cubic. In order to further confirm the epitaxial growth of γ' -Fe₄N films, X-ray pole figure of γ' -Fe₄N films on MgO(100) substrate is recorded. In the right inset of Fig. 1(b), the relevant strong diffraction peaks indicate that all the films are epitaxial.

In order to observe the growth mechanism of the reactively sputtered polycrystalline γ' -Fe₄N films, the cross-section SEM image of the films on Si is given in Fig. 2(a). In Fig. 2(a), Fe₄N grains grow with a columnar structure, which is similar to the growth manner of Fe₃O₄ grains using reactive sputtering.²⁷ In Fig. 2(b), it is clear that the disordered grain boundaries exist between the grains. Meanwhile, the density of the grain boundaries increases with the decrease of film thickness.⁷

Fig. 2(c) shows the TEM images of γ' -Fe₄N films on MgO(100). It is clear that the film thickness is about 90 nm with a sharp interface. The high-resolution TEM image at the γ' -Fe₄N/MgO(100) interface in Fig. 2(d) shows the epitaxial growth of the film, further confirming the X-ray θ -2 θ , φ scans and pole figure results.

Fig. 3(a) shows the ρ_{xx} - T curves of the epitaxial γ' -Fe₄N films with different t . It is clear that the ρ_{xx} of the epitaxial γ' -Fe₄N films increases with the increase of temperature, showing a metallic conductance. The inset of Fig. 3(a) shows the ρ_{xx} - t curves at 350 K. The ρ_{xx} decreases with the increase of t , which is ascribed to the enhanced surface scattering due to the decreased t . Meanwhile, ρ_{xx} of epitaxial γ' -Fe₄N films is smaller than that of polycrystalline ones because of the enhanced grain boundary scattering in the latter, as shown in the inset of Fig. 3(b). For both the epitaxial and polycrystalline films, the decrease ratio of the ρ_{xx} with the decreased temperature increases as t increases, implying that the electron scattering increases with the decreased t . This phenomenon can be ascribed to the enhanced surface scattering for the epitaxial films and the increased surface and grain boundaries scattering for the polycrystalline ones as the grain size decreases with the decrease of t .⁷ Generally, at $T \geq \theta_D$, (θ_D is the Debye temperature) temperature-dependent ρ_{xx} from the electron-electron scattering follows $\rho_{ph}(T) \propto T$, and at $T \leq \theta_D$, the ρ_{xx} can be written as $\rho_{ph}(T) \propto T^5$. The electron-electron interaction is prominent at the low-temperature region where the electron-phonon scattering is

frozen out and the ρ_{xx} follows the $\rho_{ee}(T) \propto T^2$ law. The contribution to ρ_{xx} from the disordered localized magnetic moment is important in the entire temperature range and can be given as $\rho_m(T) \propto T^2$ for the ferromagnetic coupling, T^5 and T^4 for the antiferromagnetic coupling.⁷ In this case, the experimental data of both the epitaxial and polycrystalline films satisfies the T^2 law at the low temperatures, which suggests that the ρ_{xx} mainly comes from the thermal disordering of the localized ferromagnetic moments and/or electron–electron interaction. In the high-temperature range, the ρ_{xx} – T curves satisfy the T^n relation, where n is smaller than 1. This relation suggests that ρ_{xx} should be from the electron–phonon scattering and the slope of ρ_{xx} – T curve decreases with the increased temperature because if T^n ($n > 1$) phase is in ρ_{xx} – T curves at high temperatures, the slope of ρ_{xx} – T curves should increase with temperature, so only T phase can be considered in the ρ_{xx} – T curves.

Fig. 4 shows the ρ_{xy} – H of the epitaxial and polycrystalline γ' -Fe₄N films with different t . The ρ_{AH} can be extracted from the Hall loops by extrapolating the high-field data from positive field to zero field. Both the polycrystalline and epitaxial γ' -Fe₄N films show the positive ρ_{xy} at positive magnetic field in the entire range of t in contrast to the negative ρ_{xy} in Fe₃O₄.²⁶ In the temperature range from 5 to 350 K, ρ_{AH} increases with the increase of temperature. It should be mentioned that the $\rho_{AH}(T)$ show a strong dependence on temperature for both epitaxial and polycrystalline in contrast to Fe samples.²⁵ The value of room temperature ρ_{AH}

increases from 14.15 $\mu\Omega$ cm for $t=100$ nm up to 18.20 $\mu\Omega$ cm for $t=18$ nm in epitaxial γ' -Fe₄N films. The thicker the films are, the smaller ρ_{AH} is. Meanwhile, at the same temperature, ρ_{AH} in epitaxial films is larger than that polycrystalline ones.

Since AHE may originate from either intrinsic or extrinsic mechanisms characterized by different exponential factors γ in the scaling relationship of $\rho_{\text{AH}} \propto \rho_{\text{xx}}^\gamma$, it is informative to examine the relation between the ρ_{AH} and ρ_{xx} for determining the exponential factor γ . Fig. 5(a) shows the scaling relation of the epitaxial γ' -Fe₄N films with different t in a log-log scale. The $\rho_{\text{AH}}-\rho_{\text{xx}}$ curves can be fitted by a straight line with a slope of $\gamma=2.28$ below 125 K. However, the ρ_{AH} deviates from the linear ρ_{xx} dependence at high temperatures. In fact, varying temperature inevitably changes saturation magnetization M_s in samples. The M_s of γ' -Fe₄N films varies significantly with temperature, and satisfies the modified Bloch's spin wave theory.⁷ The insert of Fig. 5(b) shows the temperature-dependent normalized magnetization. The normalized magnetization m defined by

$$m(T)=M_s(T)/M_s(5\text{ K}) \quad (3)$$

In Fig. 5(b), we plot the $\rho_{\text{AH}}/m-\rho_{\text{xx}}$ curves in a log-log scale. We can see that all data have collapsed along a straight line with $\gamma=2.25$. Therefore, the nonlinear scaling relation of the epitaxial γ' -Fe₄N films at high temperatures results from the

temperature-dependent magnetization.

In order to clarify whether the AHE current in γ' -Fe₄N films is dissipationless, it is necessary to factor out the carrier density n . Based on the simple theoretical model proposed by Nozières and Lewiner,²⁸ AHE current can be expressed as

$$J_{\text{AH}} = 2ne^2\lambda E \times S \quad (4)$$

where λ is the enhanced spin-orbit parameter, e is the electronic charge, E is the crossed electric fields, S is the electron spin, the electron carrier density n was calculated using the formula $n = -1/R_0e$, where R_0 is the ordinary Hall coefficient. Fig. 5(c) shows the ρ_{AH}/n - ρ_{xx} curves in a log-log scale for all the epitaxial γ' -Fe₄N films at 5 K. By fitting the data with a straight line, we obtain a slope of $\beta = 2.41$. Conventionally, the quadratic dependence of ρ_{AH}/n on ρ_{xx} was considered as the dissipationless nature of AHE current.^{29,30} The slope of $\beta = 2.41$ in the epitaxial γ' -Fe₄N films indicates that the AHE conductivity is scattering-dependent.

In order to clarify the effect of the grain boundaries scattering on the scaling law, we perform the measurements on the polycrystalline films fabricated at the same conditions. Fig. 5(d) shows the scaling law of the polycrystalline γ' -Fe₄N films with different t . The ρ_{AH} - ρ_{xx} curves revealed significant influence of film thickness in contrast to epitaxial samples, which can be attributed to the different grain

boundaries varying with the film thickness. The scaling exponents $\gamma=2.10, 2.75, 2.56, 1.95$ below 125 K show a downward trend with increasing t . However, no linear relation can be observed in the high-temperature region. After considering the effect of magnetization, the $\rho_{\text{AH}}/m-\rho_{\text{xx}}$ curves can be linearly fitted in the entire temperature regions, and is also thickness-dependent, as shown in Fig. 5(e). Fig. 5(f) shows the scaling relation of $\rho_{\text{AH}}/n \propto \rho_{\text{xx}}^\beta$ of the polycrystalline γ' -Fe₄N films at 5 K. By fitting the data using a straight line, we obtain the slope of $\beta=2.61$, suggesting that a scattering-dependent AHE conductivity in the polycrystalline γ' -Fe₄N films.

We now focus on the scaling exponent $\gamma>2$ in γ' -Fe₄N films. The scaling exponent $\gamma>2$ was usually observed in heterogeneous ferromagnetic systems, e.g., $\gamma=3.7$ in the Co-Ag granular films,³¹ $\gamma=2.6$ in the Fe/Cr multilayers,³² $\gamma=5.7$ in the Co/Pd multilayers.³³ The spin-dependent interfacial and surface scattering on AHE were considered to account for $\gamma>2$ in the ferromagnetic heterogeneous systems.^{31,33} In Fig. 6, we plot the $\rho_{\text{AH}}-\rho_{\text{xx}}$ curves of both the epitaxial and polycrystalline γ' -Fe₄N films with various thicknesses below 125 K, where the influence of the temperature dependence of the magnetization on ρ_{AH} is negligible. One can find that the scaling exponents of most samples are larger than 2. Although $\gamma>2$ in the polycrystalline γ' -Fe₄N films can be attributed to grain boundaries scattering on AHE, $\gamma>2$ is not expected in the homogenous epitaxial γ' -Fe₄N films. It should be noted that the unconventional scaling exponents $\gamma>2$ in the epitaxial γ' -Fe₄N films are

thickness-independent, suggesting that the surface scattering contribution to the scaling of the AHE is insignificant. Recent studies on AHE in Fe and Co films have demonstrated a proper scaling of $\rho_{\text{AH}} = a'\rho_{\text{xx}0} + b\rho_{\text{xx}}^2$.^{34,35} The proper scaling relation excluded the contribution of phonons to skew scattering and showed that the extrinsic contributions to AHE are only related to impurity or defects scattering. In contrast, the skew scattering contributions from phonons and impurity or defects in the conventional scaling of $\rho_{\text{AH}} \propto \rho_{\text{xx}}^\gamma$ are treated on an equal basis. The neglect of the differences between phonon and impurity or defects contributions to AHE transport data maybe result in the unconventional scaling exponent $\gamma > 2$ in γ' -Fe₄N films. Based on this consideration, we propose a new scaling of

$$\rho_{\text{AH}} = c'\rho_{\text{xx}0} + b\rho_{\text{xx}}^\alpha, \quad (5)$$

which separates out the temperature-independent residual resistivity ($\rho_{\text{xx}0}$) from the different contributions to the measured AHE transport data. As shown in Fig. 7, the data fits quite well with the new scaling for both the epitaxial and polycrystalline γ' -Fe₄N films, which provides convincing evidence that the contributions from phonon and impurity or defects to AHE are distinctly different. The obtained new scaling exponents α are between 1 and 2 as shown in Fig. 7. The fitting results strongly suggest that the $\rho_{\text{xx}0}$ in γ' -Fe₄N films leads to the unconventional scaling

exponent $\gamma > 2$.

The new scaling of $\rho_{\text{AH}} = c'\rho_{\text{xx}0} + b\rho_{\text{xx}}^\alpha$ has the same form as the proper scaling of $\rho_{\text{AH}} = a'\rho_{\text{xx}0} + b\rho_{\text{xx}}^2$. We also consider that the temperature-dependent scattering leads to intrinsic mechanism.³⁴⁻³⁶ The $\rho_{\text{xx}0}$ were separated from various contributions to AHE in both new scaling and proper scaling relation. The scaling exponent α in the new scaling relation is a fitting result from the measured AHE transport data, while the scaling exponent in the proper scaling relation is a constant=2 representing the intrinsic contribution to AHE. The novel of the new scaling relation is that the scaling exponent α from the fitting result can give a direct evidence that the large scaling exponent $\gamma > 2$ in the conventional scaling of $\rho_{\text{AH}} \propto \rho_{\text{xx}}^\gamma$ is closely related to the $\rho_{\text{xx}0}$. It should be mentioned that $\alpha=1.92, 1.96, 2.01$ and 1.94 found in epitaxial samples with $t=40, 60, 80$ and 100 nm are nearly to 2, which indicates the same physics as the proper scaling relation. Unluckily, $\alpha < 2$ has been found in other samples, especially for polycrystalline ones. This is an imperfect result that the temperature-dependent scaling exponent corresponding to the intrinsic contribution to AHE are not a constant=2. At the present, we suggest that the deviation of the scaling exponent α from the constant=2 may be due to the thermal disorder from a relatively small skew scattering contribution.³⁷ However, comprehensive understanding of the phenomenon need to more efforts from both experimental and theoretical studies.

At last, we plot ρ_{xx}^2 - ρ_{AH} curves of both epitaxial and polycrystalline samples according to the proper scaling relation as shown in Fig. 8. It is clear that the proper scaling relation also shows an excellent agreement with the experiment data. The intrinsic contribution b from temperature-dependent scattering to AHE obtained by the proper scaling relation is plotted in Fig. 9(a). The intrinsic contribution is 276 and 196 S/cm for epitaxial and polycrystalline samples respectively. The large intrinsic contribution in epitaxial sample can be attributed to its homogenous structure.¹⁹ In order to clarify the effect of impurities scattering on AHE, Fig. 9(b) and (c) shows the ρ_{AH} - ρ_{xx} curves at 5 K with different t for both the epitaxial and polycrystalline samples respectively. By fitting the data to a straight line, we obtained the slope value $\gamma=2.01$ for epitaxial samples and $\gamma=1.87$ for polycrystalline samples indicating the less contribution from skew-scattering to AHE. Therefore, we suggest that the side-jump and intrinsic mechanisms are dominant in both epitaxial and polycrystalline samples.

Conclusion

In conclusion, the scaling of AHE in the epitaxial and polycrystalline γ' -Fe₄N films is investigated systematically. The ρ_{xy} is positive in the whole temperature range. The deviation of the scaling relation in high temperature region is due to the

magnetization that varies with temperature significantly. The scaling relation in the polycrystalline γ' -Fe₄N films shows a strongly dependence on film thickness, which can be attributed to different grain boundaries scattering in various films. The normalized anomalous Hall conductivity by carrier density is not a constant, indicating that the anomalous Hall conductivity is scattering-dependent in γ' -Fe₄N films. The scaling exponent γ is larger than 2 in both epitaxial and polycrystalline γ' -Fe₄N films by the conventional scaling of $\rho_{\text{AH}} \propto \rho_{xx}^\gamma$, which can be attributed to the ρ_{xx0} . The mechanisms of AHE in γ' -Fe₄N are mainly from side-jump and intrinsic contributions.

Acknowledgements

This work was supported by the National Natural Foundation of China (51171126), Key Project of Natural Foundation of Tianjin City (12JCZDJC27100), Program for New Century Excellent Talents in University (NCET-13-0409) and Scientific Research Foundation for the Returned Overseas Chinese Scholars, State Education Ministry.

References

- 1 W. Mi, Z. Guo, X. Feng and H. Bai, *Acta Mater.*, 2013, **61**, 6387.
- 2 Y. Komasaki, M. Tsunoda, S. Isogami and M. Takahashi, *J. Appl. Phys.*, 2009, **105**, 07C928.
- 3 K. Sunaga, M. Tsunoda, K. Komagaki, Y. Uehara and M. Takahashi, *J. Appl. Phys.*, 2007, **102**, 013917.
- 4 K. Yamaguchi, T. Yui, K. Yamaki, I. Takeya, K. Kadowaki and T. Suemasu, *J. Cryst. Growth*, 2007, **301**, 597.
- 5 Y. Chen, E. Jiang, Z. Li, W. Mi, P. Wu and H. Bai, *J. Phys. D: Appl. Phys.*, 2004, **32**, 1429.
- 6 X. Feng, W. Mi and H. Bai, *Appl. Surf. Sci.*, 2001, **257**, 7320.
- 7 W. Mi, X. Feng, X. Duan, H. Yang, Y. Li and H. Bai, *Thin Solid Films*, 2012, **520**, 7035.
- 8 S. Kokado, N. Fujima, K. Harigaya, H. Shimizu and A. Sakuma, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2006, **73**, 172410.
- 9 A. Narahara, K. Ito, T. Suemasu, Y. K. Takahashi, A. Ranajikanth and K. Hono, *Appl. Phys. Lett.*, 2009, **94**, 202502.
- 10 S. Isogami, M. Tsunoda, Y. Komasaki, A. Sakuma¹ and M. Takahasi, *Appl. Phys. Express*, 2010, **3**, 103002.

- 11 A. Narahara and T. Suemasu, *Jpn. J. Appl. Phys.*, 2007, **46**, L892.
- 12 A. Narahara, K. Ito and T. Suemasu, *J. Cryst. Growth*, 2009, **311**, 1616.
- 13 M. Tsunoda, Y. Komasaki, S. Kokado, S. Isogami, C. C. Chen and M. Takahashi, *Appl. Phys. Express*, 2009, **2**, 083001.
- 14 M. Tsunoda, H. Takahashi, S. Kokado, Y. Komasaki, A. Sakuma and M. Takahashi, *Appl. Phys. Express*, 2010, **3**, 113003.
- 15 Z. Li, X. Feng, X. Wang and W. Mi, *Mater. Res. Bull.*, 2015, **65**, 175.
- 16 Z. Li, W. Mi, X. Wang and X. Zhang, *ACS Appl. Mater. Interfaces*, 2015, **7**, 3840.
- 17 J. Inoue and H. Ohno, *Science*, 2005, **309**, 2004.
- 18 C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972).
- 19 N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald and N. P. Ong, *Rev. Mod. Phys.*, 2010, **82**, 1539.
- 20 J. Smit, *Physica*, 1955, **21**, 877.
- 21 L. Berger, *Phys. Rev. B: Solid State*, 1970, **2**, 4559.
- 22 R. Karplus and J. M. Luttinger, *Phys. Rev.*, 1954, **95**, 1154.
- 23 Z. Fang, N. Nagaosa, K. S. Tahakashi, A. Asamitsu, R. Mathieu, T. Ogasawara, H. Yamada, M. Kawasaki, Y. Tokura and K. Terakura, *Science*, 2003, **302**, 92.
- 24 S. Onada, N. Sugimoto and N. Nagaosa, *Phys. Rev. Lett.*, 2006, **97**, 126602.
- 25 S. Sangiao, L. Morellon, G. Simon, J. M. De Teresa, J. A. Pardo, J. Arbio and M.

- R. Ibarra, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2009, **79**, 014431.
- 26 A. Fernández-Pacheco, J. M. De Teresa, J. Orna, L. Morellon, P. A. Algarabel, J. A. Pardo and M. R. Ibarra, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2008, **77**, 100403(R).
- 27 J. Shen, W. Mi, Z. Li, P. Wu, E. Jiang and H. Bai, *J. Vac. Sci. Technol. A*, 2006, **24**, 390.
- 28 P. Nozières and C. Lewiner, *J. Phys.*, 1973, **34**, 901.
- 29 W. L. Lee, S. Watauchi, V. L. Miller, R. J. Cava and N. P. Ong, *Science*, 2004, **303**, 1647.
- 30 W. Xu, B. Zhang, Q. Wang, W. Mi, Z. Wang, W. Li, R. Yu and X. Zhang, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2011, **83**, 205311.
- 31 P. Xiong, G. Xiao, J. Q. Wang, J. Q. Xiao, J. S. Jiang and C. L. Chien, *Phys. Rev. Lett.*, 1992, **69**, 3220.
- 32 S. N. Song, C. Sellers and J. B. Ketterson, *Appl. Phys. Lett.*, 1991, **59**, 479.
- 33 Z. Guo, W. Mi, R. O. Aboljadayel, B. Zhang, Q. Zhang, P. Barba, A. Manchon and X. Zhang, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2012, **86**, 104433.
- 34 Y. Tian, L. Ye and X. Jin, *Phys. Rev. Lett.*, 2009, **103**, 087206.
- 35 D. Hou, Y. Li, D. Wei, D. Tian, L. Wu and X. Jin, *J. Phys.: Condens. Matter.*, 2012, **24**, 482001.
- 36 G. Su, Y. Li, D. Hou, X. Jin, H. Liu and S. Wang, *Phys. Rev. B: Condens. Matter*

Mater. Phys., 2014, **90**, 214410.

37 A. Gerber, A. Milner, A. Finkler, M. Karpovski and L. Goldsmith, *Phys. Rev. B:*

Condens. Matter Mater. Phys., 2004, **69**, 224403.

Figure captions

Fig. 1. The θ - 2θ XRD patterns of (a) the polycrystalline γ' -Fe₄N films on natural oxidized Si substrate and (b) the epitaxial γ' -Fe₄N film on MgO(100). The insets of Fig. 1(b) give X-ray ϕ scan and pole figure of γ' -Fe₄N films on MgO(100).

Fig. 2. (a) The cross-section SEM image of the polycrystalline γ' -Fe₄N film on Si substrates, (b) planar-view TEM image of the polycrystalline γ' -Fe₄N film, (c) cross-sectional TEM image of γ' -Fe₄N films on MgO(100), (d) the high-resolution TEM image at the γ' -Fe₄N/MgO(100) interface.

Fig. 3. The temperature-dependent ρ_{xx} of (a) the epitaxial γ' -Fe₄N films and (b) the polycrystalline γ' -Fe₄N films with different t . The insets give the relevant film-thickness dependent ρ_{xx} measured at 350 K.

Fig. 4. The ρ_{xy} - H curves of the epitaxial and polycrystalline γ' -Fe₄N films measured at different temperature, (a) $t=18$ nm epitaxial film, (b) $t=18$ nm polycrystalline film, (c) $t=100$ nm epitaxial film, and (d) $t=100$ nm polycrystalline film.

Fig. 5. The scaling relation of the epitaxial and polycrystalline γ' -Fe₄N films with different t , (a) $\rho_{AH}-\rho_{xx}$ curves for epitaxial films, (b) $\rho_{AH}/m-\rho_{xx}$ curves for epitaxial films, (c) $\rho_{AH}/n-\rho_{xx}$ curves for epitaxial films, (d) $\rho_{AH}-\rho_{xx}$ curves for polycrystalline films, (e) $\rho_{AH}/m-\rho_{xx}$ curves for polycrystalline films, (f) $\rho_{AH}/n-\rho_{xx}$ curves for polycrystalline films. The insert of Fig. 5(b) shows the temperature-dependent m of γ' -Fe₄N films.

Fig. 6. The $\rho_{AH}-\rho_{xx}$ curves below 125 K with different t in log-log scale for both epitaxial and polycrystalline γ' -Fe₄N films, (a) $t=18$ nm, (b) $t=26$ nm, (c) $t=40$ nm, (d) $t=60$ nm, (e) $t=80$ nm, (f) $t=100$ nm. The red and black line is a linear fit of the data.

Fig. 7. The $\rho_{AH}-\rho_{xx}$ curves below 125 K with different t for both epitaxial and polycrystalline γ' -Fe₄N films, (a) $t=18$ nm, (b) $t=26$ nm, (c) $t=40$ nm, (d) $t=60$ nm, (e) $t=80$ nm, (f) $t=100$ nm. The red and black line is fitting result with a new scaling of $\rho_{AH} = c' \rho_{xx0} + b \rho_{xx}^\alpha$.

Fig. 8. The $\rho_{AH}-\rho_{xx}^2$ curves below 125 K with different t for both epitaxial and polycrystalline γ' -Fe₄N films, (a) $t=18$ nm, (b) $t=26$ nm, (c) $t=40$ nm, (d) $t=60$

nm, (e) $t=80$ nm, (f) $t=100$ nm. The red and black line is a linear fit of the data.

Fig. 9. (a) The t dependence of the intrinsic contribution b to AHE obtained by the proper scaling relation for both epitaxial and polycrystalline samples. The $\rho_{\text{AH}}-\rho_{xx}$ curves at 5 K with different t for (b) epitaxial films, (c) polycrystalline films.

Fig. 1, Y. Zhang, *et al.*

Fig. 2, Y. Zhang, *et al.*

Fig. 3, Y. Zhang, *et al.*

Fig. 4, Y. Zhang, *et al.*

Fig. 5, Y. Zhang, *et al.*

Fig. 6, Y. Zhang, *et al.*

Fig. 7, Y. Zhang, *et al.*

Fig. 8, Y. Zhang, *et al.*

Fig. 9, Y. Zhang, *et al.*