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Manuscript to Journal of Physics: Condensed Matter

**Skew scattering dominated anomalous Hall effect in
 $\text{Co}_x(\text{MgO})_{100-x}$ granular thin films**

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ABSTRACT

We investigated the mechanism(s) of the anomalous Hall effect (AHE) in magnetic granular materials by fabricating 100-nm-thick thin films of $\text{Co}_x(\text{MgO})_{100-x}$ with a Co volume fraction of $34 \leq x \leq 100$ using co-sputtering at room temperature. We measured the temperature dependence of longitudinal resistivity (ρ_{xx}) and anomalous Hall resistivity (ρ_{AHE}) from 5 K to 300 K in all samples. We found that when x decreases from 100 to 34, the values of ρ_{xx} and ρ_{AHE} respectively increased by about four and three orders in magnitude. By linearly fitting the data, obtained at 5 K, of anomalous Hall coefficient (R_s) and of ρ_{xx} to $\log(R_s) \sim \gamma \log(\rho_{xx})$, we found that our results perfectly fell on a straight line with a slope of $\gamma = 0.97 \pm 0.02$. This fitting value of γ in $R_s \propto \rho_{xx}^\gamma$ clearly suggests that skew scattering dominated the AHE in this granular system. To explore the effect of the scattering on the AHE, we performed the same measurements on annealed samples. We found that although both ρ_{xx} and ρ_{AHE} significantly reduced after annealing, the correlation between them was almost the same, which was confirmed by the fitted value, $\gamma = 0.99 \pm 0.03$. These data strongly suggest that the AHE originates from the skew scattering in Co-MgO granular thin films no matter how strong the scattering of electrons by the interfaces and defects is. This observation may be of importance to the development of spintronic devices based on MgO.

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2 **Keywords:** granular thin films, ferromagnetism, anomalous Hall effect,
3 skew scattering

4

1. Introduction

The anomalous Hall effect (AHE) in ferromagnetic materials has been investigated extensively in an effort to discover both the underlying physics [1–3] and potential applications in magnetic field sensors [4,5]. The total Hall resistivity (ρ_{xy}), including ordinary Hall resistivity (ρ_{OHE}) and anomalous Hall resistivity (ρ_{AHE}), are generally expressed as

$$\rho_{xy} = \rho_{\text{OHE}} + \rho_{\text{AHE}} = R_0 H + R_s 4\pi M \quad (1)$$

where R_0 is the ordinary Hall coefficient, R_s is the anomalous Hall coefficient, H is the applied magnetic field, and M is the magnetization perpendicular to the film plane. Although it is well known that the mechanism of the ordinary Hall effect is purely classical (i.e., it is a Lorentz force acting on moving carriers), the mechanism(s) leading to the AHE, believed to be of quantum mechanical origin, are still under debate.

Several theoretical models based on intrinsic and extrinsic mechanisms have been proposed to interpret the origin of the AHE. Karplus and Luttinger [1] first suggested that the AHE originates from interband coherence due to spin-orbit coupling (SOC) of polarized conduction electrons in perfect ferromagnetic crystals. They predicted a quadratic dependence of R_s on the longitudinal resistivity (ρ_{xx}), i.e., $R_s \propto \rho_{xx}^2$, based on this mechanism. Since they considered only band structures and completely ignored the effect of

impurities in the materials, their model has then been referred to as the intrinsic mechanism of the AHE. In contrast to this model, Smit [2] argued that the AHE should vanish in perfect crystals and proposed an extrinsic mechanism model based on asymmetric scattering of electrons at impurities, again caused by SOC. This asymmetric scattering has been then referred to as skew scattering. In this model, the transverse current is proportional to the longitudinal current driven by the electrical field and consequently a linear dependence of R_s on ρ_{xx} , $R_s \propto \rho_{xx}$, is predicted. Later, Berger [3] proposed another model based on the extrinsic mechanism, the side-jump model. In this model, the trajectories of scattered electrons at impurity sites shift to one side due to the existence of SOC. Berger derived the quadratic dependence of the AHE on the longitudinal resistivity as $R_s \propto \rho_{xx}^2$ based on the side-jump mechanism. It should be noted that the scaling derived based on the extrinsic side-jump mechanism was identical to that obtained based on the intrinsic mechanism.

After the proposal of the Berry phase [6], anomalous Hall conductivity (AHC) was re-examined and ascribed to the sum of the Berry curvatures of the occupied Bloch states [7,8]. Intrinsic AHC was obtained by first-principles calculations within a generalized gradient approximation in bcc Fe [9], fcc Ni [10] and polycrystalline Co [11], which quantitatively agree with experimental

1 results [12 – 14]. To date, experimentally distinguishing the intrinsic
 2 mechanism and the side-jump mechanism in the AHE has been difficult, in
 3 addition to the unanswered question of whether the skew-scattering or the
 4 side-jump mechanism dominates the AHE.

5 To distinguish the mechanisms of the AHE experimentally, one usually
 6 plots experimental data based on the scaling relations $\rho_{\text{AHE}} \sim \rho_{xx}^{\gamma}$ to find the
 7 value of the exponent. However, various scaling dependences of ρ_{AHE} on ρ_{xx}
 8 have been reported for the well-known and simple ferromagnetic transition
 9 metals (TMs), Fe, Co and Ni, by different groups. The relation $\rho_{\text{AHE}} \propto \rho_{xx}^2$ [15]
 10 was reported for Fe, $\rho_{\text{AHE}} = a\rho_{xx} + b\rho_{xx}^2$ [13] for Co and $\rho_{\text{AHE}} \sim \rho_{xx}$ [16] or
 11 $\rho_{\text{AHE}} \sim \rho_{xx}^n$ ($1 < n < 2$) [14,17] for Ni. It has clearly been a great challenge to verify
 12 with experimental evidence the mechanisms of the AHE even for these simple
 13 TMs.

14 Understanding the mechanisms of the AHE could offer guidance for
 15 designing materials to achieve large anomalous Hall resistivity. Also, such
 16 understanding could be beneficial to general understanding of the spin Hall
 17 effect, which has the same mechanisms as the AHE, since the large spin Hall
 18 effect plays a critical role in spin transfer torque applications [18,19]. The
 19 giant anomalous Hall effect has been studied in last two decades. In
 20 ferromagnetic metal-insulator systems, a scaling relation of $\rho_{\text{AHE}} \propto \rho_{xx}^{\gamma}$ with

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$\gamma < 1$ was observed [20–22]. It is apparent that this scaling cannot be ascribed to any of the current models for the AHE. Zhang [23] suggested that the conventional scaling law is invalid when the layer thickness is of the same order or smaller than the mean free path in multilayers. Zhang considered only the side-jump effect of the electron scattering in the delta impurities but neglected interfacial scattering. However, in metal-insulator granular systems, the electrical transport properties must be dominated by the interfacial scattering due to their nanostructured nature. The theoretical interpretation of the observed scaling law, $\rho_{\text{AHE}} \propto \rho_{xx}^\gamma$, with $\gamma < 1$ remains elusive.

In metal-insulator thin films, SiO_2 is the commonly used insulating material to form percolated nano-granular materials with TMs for the study of the giant Hall effect. The ideal case is that the metals and insulators are completely separated by sharp interfaces and uniformly distributed in the materials. However, interface mixing and reactions [24] between metals and SiO_2 can not easily be avoided in the process of co-sputtering deposition. Achieving an ideal percolation system with minor interface mixing and reactions in experiments would improve the study of magneto-transport properties.

MgO is usually used in magnetic tunnel junctions as a barrier layer because of its good electrical isolation property and the minor diffusion between MgO and other magnetic materials (*e.g.*, CoFeB). Compared to SiO_2 , MgO has better

thermal stability and does not react as easily with TMs even at high deposition or annealing temperatures [15]. MgO is therefore a better choice to form a granular system that would improve the study of interfacial scattering effects on the AHE.

In this work, we studied the AHE of $\text{Co}_x(\text{MgO})_{100-x}$ granular thin films in which the interfacial scattering between Co and MgO dominated the electrical properties. The magneto-transport properties of all samples were measured in a magnetic field range of $-50 \text{ kOe} \leq H \leq 50 \text{ kOe}$ and a temperature range from 5 K to 300 K. The density of interfacial scattering was adjusted by changing the volume fraction of Co. Accordingly, the longitudinal resistivity and anomalous Hall resistivity varied in a wide range. By plotting $R_s \sim \rho_{xx}^y$ at 5 K for both as-prepared and annealed samples, we could observe a linear relation between R_s and ρ_{xx} , suggesting that interfacial scattering induced skew-scattering mechanism dominates the AHE in this $\text{Co}_x(\text{MgO})_{100-x}$ granular system.

2. Experiment

We prepared 100-nm-thickness $\text{Co}_x(\text{MgO})_{100-x}$ granular films with various volume fractions of Co ($34 \leq x \leq 100$) on oxidized silicon wafers by co-sputtering Co and MgO targets in a magnetron sputter system (ESCRD4, Equipment

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Support Company). The deposition chamber equipped three magnetron cathodes (ONYX 2", Angstrom Science) in which three targets, Co (99.99% purity), MgO (99.95% purity) and SiO₂ (99.995% purity), with 2-inch diameter were mounted. The distance between the Co (MgO) target and the substrate was about 25 cm (15 cm). The base pressure of the chamber was lower than 6.0×10^{-8} Torr. During the deposition, Ar pressure was maintained at 5.0 mTorr and the substrates were maintained at room temperature. DC power supply (MDX 1.5 kW, Advanced Energy) and RF power supply (R601-13, Seren IPS) were used for Co and MgO sputtering, respectively. The sputtering power of MgO target was fixed to 100 W and that of Co target varied from 12W to 113 W to adjust the volume fraction of Co. The volume fraction of Co was confirmed later by an energy dispersive x-ray spectrometer (EDX) on a scanning electron microscope (Quanta 600, FEI). After the deposition, ~20-nm-thick SiO₂ thin films were deposited on the samples without breaking the vacuum to prevent oxidation. Thermal annealing was performed at 300 °C for one hour under vacuum. The film thickness was determined using a surface profiler (Dektak 150, Veeco). The microstructure of the films was characterized by x-ray diffraction (XRD) (D8 ADVANCE, Bruker) and scanning transmission electron microscopy (STEM) (Titan 80–300, FEI). The magneto-transport properties were measured by a physical property

measurement system (Dynacool, Quantum Design). Mechanical masks were used to pattern the samples into a five-contact geometry for simultaneously measuring the Hall resistivity and the longitudinal resistivity on the same sample. The magnetic properties were measured by a magnetic property measurement system (MPMS3, Quantum Design).

3. Results and discussion

A. Microstructure of the as-prepared samples

STEM analysis was performed to investigate the microstructure and morphology of the $\text{Co}_x(\text{MgO})_{100-x}$ samples. The samples for STEM characterization were deposited directly on Cu grids at the same time that samples were deposited for other analyses to ensure that we maintained the same fabrication conditions. Figure 1 shows the STEM high-angle annular dark-field (HAADF) images of the samples with $x = 81$ and 66. The images show the clear contrast of black and white areas, a feature of granular structured materials. The bright regions correspond to the metallic Co clusters and the dark regions correspond to the MgO clusters. As expected, the area of the bright regions in the images reduced as the volume fraction of Co decreased from $x = 81$ to $x = 66$. For electrical transport, the electrons are much more scattered in the samples with the lower Co fractions than in the

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samples with higher Co fractions. The mean free path should therefore be shorter in the samples with lower Co fractions and limited to the size of the Co clusters, which would be reflected in the electrical properties.

B. Magneto-transport and magnetization of the as-prepared samples

To understand the electrical properties and explore the electrical transport mechanisms, we measured the temperature-dependent longitudinal resistivity ($\rho_{xx} - T$) curves for all samples at temperatures ranging from 5 K to 300 K with zero magnetic field, as shown in figure 2(a). The pure Co sample ($x=100$) had a positive temperature coefficient of resistivity (TCR, $d\rho/dT$) across the full temperature range, indicating metallic electrical transport. The TCR for samples with $47 \leq x \leq 81$ was positive at high temperatures, indicating a metallic conduction mechanism. Interestingly, at low temperatures, the TCR became negative, which can be ascribed to the electrical conduction being dominated by a two-dimensional weak localization effect (see figure S1 and the corresponding analysis in the Supplemental Material). The TCR for the samples with $34 \leq x \leq 42$ was negative across the full temperature range, indicative of semiconducting or insulating conduction behaviors. Using TCR to determine the transport regimes is not completely reliable because TCR of disordered alloys could be positive or negative [25,26]. Zabrodskii *et al* [27]

proposed a model to analyze temperature-dependent resistivity:

$$\rho_{xx}(T) = BT^{-m}\exp(T_0/T)^n, \quad (2)$$

where B , m , T_0 and n are constants and n is a critical factor to characterize the conduction mechanisms. If $n < 0$ ($n > 0$), the conduction of the samples is in the metallic (insulating) regime. By fitting the $\rho_{xx} - T$ curves of the samples with $34 \leq x \leq 42$ using equation (2) as shown in figure S2, we obtained the constant n as 0.084, 0.080, 0.076 and 0.21 for the samples with $x=42, 39, 36$ and 34, respectively. Based on this classification, we classified the conduction of the four samples as in the insulating regime. Overall, the electrical properties of the samples evolved from metallic to insulating as the Co fraction decreased. To capture the scattering effect, we plot the normalized temperature-dependent longitudinal resistivity in figure 2(b) for the samples with $47 \leq x \leq 100$. The resistivity ratio $\rho_{xx}(T)/\rho_{xx}(300 \text{ K})$ is a useful empirical parameter for quantifying the extent of disorder [28]. As shown in figure 2(b), the $\rho_{xx}(T)/\rho_{xx}(300 \text{ K})$ increases as x decreases, which clearly demonstrates that there is more interfacial scattering in samples with lower x . At low temperatures (*e.g.*, 5 K) when phonon/magnon scattering can be ignored, the interfacial scattering between Co and MgO dominates the electrical transport properties and leads to a four-order increase in resistivity from sample $x = 100$ to $x = 34$.

We now turn to examining how interfacial scattering affects anomalous Hall resistivity. The field-dependent Hall resistivity and magnetization for all samples were measured with a magnetic field applied perpendicularly to the film plane in the range of $-50 \text{ kOe} \leq H \leq 50 \text{ kOe}$ and at temperatures ranging from 5 K to 300 K. Figures 3(a) and (c) show the field-dependent Hall resistivity of selected samples as indicated in the caption. ρ_{xy} increases sharply at low fields and shows a weak and linear dependence on H at high fields, which are typical behaviors of magnetic materials. We ascribe the weak and linear field dependence of the AHE at high fields mainly to the ordinary Hall resistivity where magnetization is saturated. The sign of ρ_{AHE} for all samples is positive across the whole measured temperature range, which is consistent with previous reports [29,30]. The sign of ρ_{OHE} is negative, suggesting that electrons dominate transport properties. Figures 3(b) and (d) give the field-dependent magnetization for the same samples presented for the Hall resistivity measurements. The linear decrease in magnetization at high fields is due to the diamagnetic contribution from the silicon substrates or/and sample holders. The saturation magnetization (M_s) measured at 5 K decreases with x . Shown in figure 3(d) are the $M-H$ curves measured at different temperatures for the sample $x = 66$. M_s is clearly weakly dependent on temperature from 5 K to 300 K. This weak dependence is due to the high Curie

temperature (~ 1400 K) of cobalt. Although M_s is weakly temperature dependent, ρ_{xy} is strongly temperature-dependent.

By comparing figures 3(a) and (b), we see that the field-dependent Hall resistivity and magnetization curves follow similar patterns that are consistent with equation (1). Hysteresis loops are observed in both $\rho_{xy} - H$ and $M - H$ curves for samples with lower x and the coercivity (H_C) increases as x decreases. However, the coercivity in $\rho_{xy} - H$ is different from that in the corresponding $M - H$ curves at 5 K, which seems to violate the relation established by equation (1). We measured the total magnetization of the materials (film and substrate), i.e., the magnetization of the film, $M(\text{film})$ and the contribution of the substrate, $M(\text{sub}) = \chi H$, where χ is the magnetic susceptibility. Under a positive magnetic field, $M(\text{film}) > 0$, but $M(\text{sub}) < 0$, since $\chi < 0$ for the diamagnetic contribution of the substrates. Therefore, we expect a slightly larger H_C at which $M(\text{film}) + M(\text{sub}) > 0$, in comparison with a pure film without a substrate. In addition, in the $M \sim H$ measurement, we include all the Co clusters, no matter if they are involved in the electrical transport. Isolated small Co clusters normally have a much higher coercive field than large clusters or bulk Co have due to their high surface anisotropy [31,32]. Thus, the H_C extracted from the $M - H$ curves should be larger than that from the AHE measurement where only large, percolated clusters

contribute. The inhomogeneity-induced H_C difference in the $M - H$ and $\rho_{xy} - H$ curves was also reported in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ films [33].

C. Anomalous Hall effect scaling analysis

To explore the AHE mechanism in these samples, we extracted ρ_{AHE} from the field-dependent Hall resistivity curves by extrapolating the high-field data to the zero field. The relations $\rho_{\text{AHE}} \propto \rho_{xx}^y$ or $\rho_{\text{AHE}} = a\rho_{xx} + b\rho_{xx}^2$ are usually employed to verify the mechanism. The premise for using these two equations is that M_s should be kept the same for different ρ_{AHE} in the scaling laws because anomalous Hall resistivity is not only related to the longitudinal resistivity but also to the magnetization of the samples as suggested by equation (1). In materials in which the Curie temperature is high, M_s is almost the same below room temperature. Using the scaling law between ρ_{AHE} and ρ_{xx} is appropriate for Fe [15] and Ni [14]. It is even appropriate for spinel $\text{CuCr}_2\text{Se}_{4-x}\text{Br}_x$ with different x . If M_s is insensitive to x , $\rho_{\text{AHE}}/n \propto \rho_{xx}^2$ is good enough to demonstrate that the mechanism is intrinsic [34]. In our Co-MgO system, M_s measured at 5 K decreases with x as shown in figure 3(b) and the variation in M_s cannot be ignored in the scaling relations. We can therefore plot of $\rho_{\text{AHE}}/M_s \sim \rho_{xx}^y$ for our system. Using 5 K data to study the AHE mechanism excludes the phonon effect on the AHE, because photon scattering

may increase exponent γ from 1 to 2 [35], which would make the system too complicated to identify the mechanisms in our samples. We focus only on the effect of impurities on the AHE in this study. We plotted the curve of R_s versus ρ_{xx} logarithmically in figure 4 for all samples measured at 5 K. By linearly fitting this curve, we obtained the exponent $\gamma = 0.97 \pm 0.02$ in a very broad range of ρ_{AHE} (three orders) and ρ_{xx} (nearly four orders). This perfect linear dependence of R_s on ρ_{xx} across the several orders of magnitude in both ρ_{AHE} and ρ_{xx} strongly suggests that skew scattering dominates the AHE in the $\text{Co}_x(\text{MgO})_{100-x}$ system and contributions from intrinsic and/or side-jump mechanisms could be easily excluded.

D. The effect of annealing on the scaling relation of the AHE

As is well known, the conductivity of metal-insulator granular materials is improved by annealing. This improved conductivity can be mainly ascribed to the improved crystallinity and the enlarged size of the metal clusters [36]. Modification of the interfaces between the metal and insulator phases may also play an important role in improving the electrical conductivity. After annealing, the interfaces normally become sharper than those in the as-prepared samples due to the better separation of the two immiscible phases and to the improved crystallinity of the two phases, particularly the metal phase. The enlarged

1 metal clusters may also improve the metallic contacts between the metal
2 clusters. A reduced Hall effect in addition to improved conductivity has been
3 observed in Cu-SiO₂ granular materials after annealing under proper
4 conditions [37]. Therefore, we annealed the Co_x(MgO)_{100-x} samples to explore
5 how the AHE and its scaling relation with longitudinal resistivity are affected
6 by variations in interfaces and microstructure changes.

7 We measured the temperature-dependent longitudinal resistivity of the
8 annealed samples at temperatures ranging from 5 K to 300 K with zero
9 magnetic field, as shown in figure S3. Similar to those of the as-prepared
10 samples, the electrical properties of the annealed samples changed from
11 metallic to insulating conduction when x decreased from 100 to 34. All
12 samples became more conductive, i.e., their resistivity decreased after
13 annealing, indicating that the annealing process modified the electron
14 scattering mainly at the interfaces. The field-dependent Hall resistivity and
15 magnetization of all annealed samples were measured as in the as-prepared
16 samples. The results are shown in figure S4. Interestingly, although the
17 saturation magnetization of each sample increased after annealing, the Hall
18 resistivity decreased, indicating that the AHE is more closely associated with
19 the longitudinal resistivity rather than with the magnetization.

20 To gain a deeper understanding of how the annealing affects the transport

properties of the samples, we plotted in figure 5(a) the 5 K data of the anomalous Hall coefficient versus the longitudinal resistivity of the annealed samples. Remarkably, the correlation of R_S and ρ_{xx} is almost the same as that obtained with the as-prepared samples with a slightly larger exponent of $\gamma = 0.99 \pm 0.03$, even though both the AHE and longitudinal resistivity decreased significantly after annealing. We then plotted all 5 K data of R_S versus ρ_{xx} obtained from both as-prepared and annealed samples in figure 5(b). As we expected, all data fall into the same curve with a slope of 0.96 ± 0.02 . It is evident that skew scattering dominates the AHE in the $\text{Co}_x(\text{MgO})_{100-x}$ system. The slightly larger exponent, $\gamma = 0.99 \pm 0.03$, in the annealed samples is closer to 1 to indicate more obvious skew scattering. This might be ascribed to the fact that the quality of the interfaces was improved after annealing.

We also note the scaling between R_S and ρ_{xx} across more than four orders in magnitude, suggesting that the scaling is demonstrable without any ambiguity in determining the exponent of the scaling. The giant changes in both ρ_{xx} and ρ_{AHE} are critical and essential in reliable scaling to explore the mechanism(s) of the AHE in all materials. Actually, in many insulator/metal granular systems, it is quite difficult to achieve such extensive changes in both the longitudinal resistivity and the anomalous Hall resistivity. Therefore, exploring more about the electrical transport in this particular system would

be very interesting.

Overall, in both the as-prepared and annealed samples, the magnitude of ρ_{xx} and ρ_{AHE} respectively increases by nearly four and three orders as x decreases from 100 to 34 as shown in figures 6(a) and (b). Since these samples clearly belong to a percolation system, it would be interesting to explore if these ρ_{xx} and ρ_{AHE} data can be described by percolation theory. Based on percolation theory [38,39], both the longitudinal resistivity and Hall resistivity scale with the volume fraction of the metallic components, i.e.,

$$\rho_{xx} \propto (x - x_c)^{-t}, \quad x > x_c \quad (3)$$

and

$$\rho_{AHE} \propto (x - x_c)^{-g}, \quad x > x_c, \quad (4)$$

where x_c is the classical percolation threshold and $t(g)$ is a critical exponent.

We least-squares fitted the data in figures 6(a) and (b) to equations (3) and (4) and replotted them in logarithmic scales. As shown in figures 6(c), (d), (e) and (f), the data show good linear dependence, confirming the values of x_c and $t(g)$ for both as-prepared and annealed samples. The obtained parameters of x_c and $t(g)$ are summarized in Table 1. The values of x_c are higher than the value (≈ 15 [38,39]) in the ideal three-dimensional (3D) continuum model, possibly due to the immiscibility of the two phases not being perfect, i.e., the separation between Co and MgO is not atomically sharp, which is different from the

theoretical model in which the two phases are absolutely separated. In addition, there must be some individual Co atoms and small Co clusters embedded in the MgO, which also increase the percolation thresholds. However, the percolation threshold obtained in this Co–MgO system is already much smaller than the percolation thresholds in other granular systems [37,40], which is an advantage of this system. Moreover, our samples are ~ 100 nm thick and may not be considered as a 3D system. The fitted values of exponents t and g are larger than those ($t \approx 2$ and $g \approx 0.4 \sim 0.45$) in an ideal 3D percolation system but lower than those ($t = 4.5$ and $g = 3.5$) reported in a quasi-two-dimensional $(\text{NiFe})_x(\text{SiO}_2)_{100-x}$ percolation system [41].

By comparing the data of the annealed samples with that of the as-prepared samples, we find that both the longitudinal resistivity and anomalous Hall resistivity decreases after annealing. The decreased longitudinal resistivity strongly indicates that the scattering strength or density decreased after annealing. This decrease in resistivity may result from the following several factors: 1) the growth of the metal granules due to the separation/precipitation of the diluted Co atoms/small clusters from the MgO matrix. 2) the coalescence of the clusters that also increases the size of the metal granules. These two factors increases the number of conduction channels or the size of the cross-section of the conductors. 3) the metal clusters becoming cleaner due to the

precipitation of MgO from the metal granules and the deduction of the grain boundaries due to the improved crystallinity. 4) the improved interfaces due to the better separation of two materials and better crystallization of clusters. The interfaces become smoother and sharper, which certainly reduces the electron scattering in the cross-sections. Due to above factors, the percolation threshold (x_c) may decrease after annealing. For ρ_{xx} , the x_c remains the same after annealing, which may indicate that the effective volume fraction of the metal did not increase although the conductivity of the metal clusters increased. The x_c decreases from 26 to 20 in ρ_{AHE} data after annealing, which suggests the possible structure changes mentioned above.

E. The effect of cobalt oxides on the scaling relation of the AHE

It is natural to suspect that cobalt oxides exist in the samples because of the reaction at the interface between the Co and MgO. Since cobalt oxides, such as CoO and Co₃O₄, are antiferromagnetic materials, we may observe an exchange bias in the magnetic hysteresis loops after field cooling (FC) if such cobalt oxides are present in the samples. We measured the $M - H$ curves at 2 K after zero-field cooling (ZFC) and the FC process. The measurement procedures were as follows. First, we cooled the sample from 350 K to 2 K under a zero applied field. In this case, the hysteresis loop (ZFC $M - H$ curve)

was measured at 2 K in a field range of $-70 \text{ kOe} \leq H \leq 70 \text{ kOe}$. Vertical shift
 or exchange bias should not be evident in this hysteresis loop. The exchange
 bias (H_E) is defined as $H_E = (|H_{c-}| - |H_{c+}|)/2$, where $H_{c+(-)}$ is the positive
 (negative) coercivity field when magnetization is zero. We then cooled the
 sample from 350 K to 2 K under an applied field of 70 kOe. In this case, the
 hysteresis loop (FC $M-H$ curve) was measured at 2 K in a field range of
 $-70 \text{ kOe} \leq H \leq 70 \text{ kOe}$. If there were no exchange bias, the obtained loop
 would be the same as the one measured with ZFC cooling. Otherwise, the
 hysteresis loop would shift to the left. Figure 7 shows the ZFC and FC $M-H$
 curves for as-prepared and annealed samples at $x = 34$ as an example. The
 magnetic field was parallel with the sample planes during the measurement.
 One important feature is that all $M-H$ curves are irreversible under a high
 applied field (up to 70 kOe), which could be due to the existence of the spin
 glass phase [42,43]. We observed a vertical shift in FC hysteresis loops,
 indicating the existence of pinned phases whose moments could not be
 reversed even at 70 kOe. As seen in figure 7, H_E is almost zero in the ZFC $M-H$
 curves whereas there is an exchange bias in the FC $M-H$ curves for both the
 as-prepared and annealed samples. The exchange bias shown in the FC $M-H$
 curves indicates the existence of exchange-coupled phases. The cobalt oxides
 that are present and the spin glass phase may both contribute to H_E . However,

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cobalt oxides are antiferromagnetic materials that do not contribute to the anomalous Hall current and magnetization. The cobalt oxides are insulating materials and consequently would play the same role as MgO in the magneto-transport properties. These cobalt oxides, if they do exist in the samples, would therefore not affect the scaling relation of $R_s \propto \rho_{xx}$ or the conclusion that skew scattering dominates the AHE in this $\text{Co}_x(\text{MgO})_{100-x}$ system.

F. Discussion

Experimentally, the linear relation of $R_s \propto \rho_{xx}$ has usually been reported in high-conductivity metals, such as Ni with diluted impurities [16] and Fe with light doping of Co, Cr, Mn and Si [44,45]. It has been theoretically reported that skew scattering dominates the AHE in the high-conductivity regime ($\sigma_{xx} > 10^6 \text{ } \Omega^{-1}\text{cm}^{-1}$) [46]. In our $\text{Co}_x(\text{MgO})_{100-x}$ system, the longitudinal conductivity spans the moderately dirty regime and the bad metal regime. Indeed, skew scattering was still found to dominate the AHE across the whole range.

As reported in the literatures, the intrinsic mechanism was significantly suppressed in ultrathin epitaxial Fe films [15,47] because the intrinsic mechanism originates from the band structure of materials, which is related to the long-range order of the crystallinity. Since our $\text{Co}_x(\text{MgO})_{100-x}$ samples

have a highly disordered structure as confirmed by XRD patterns (not shown), we could completely neglect the interband coherence effects. Furthermore, when the size of the clusters becomes small enough, the energy bands can be greatly altered (the finite size effect). Therefore, it is easy to understand why skew scattering dominated the AHE from the perspective of traditional Boltzmann transport theory. However, to our best knowledge, this is the first time that the linear dependence of R_s on ρ_{xx} has been observed in granular thin films across a wide range in longitudinal resistivity.

4. Conclusions

$\text{Co}_x(\text{MgO})_{100-x}$ thin films with Co volume fraction x from 100 to 34 were prepared by cosputtering of Co and MgO. The microstructure, temperature-dependent longitudinal resistivity and Hall resistivity were investigated systematically. The STEM HAADF images verified that the samples are inhomogeneous granular thin films. The electrical properties evolved from metallic to insulating as x decreased. The linear scaling relation between the anomalous Hall coefficient and the longitudinal resistivity measured at 5 K was obtained for the as-prepared samples. The annealing process modified the scattering potential/possibility and reduced the longitudinal resistivity and the anomalous Hall resistivity. The scaling relation of $R_s \propto \rho_{xx}$ at 5 K holds in the

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annealed samples. This is the key result of this work which suggests a skew scattering dominated mechanism in $\text{Co}_x(\text{MgO})_{100-x}$ granular thin films, which requires further theoretical investigation.

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1 **TABLE**

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3 TABLE 1. The parameters obtained by fitting data using equations (3) and (4).

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	$x_c (\rho_{xx})$	$t (\rho_{xx})$	$x_c (\rho_{AHE})$	$g (\rho_{AHE})$
As-prepared samples	27	3.74	26	3.05
Annealed samples	27	3.38	20	3.42

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27 **Figure 1.** STEM HAADF images of the samples with (a) $x=81$ and (b)
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1 **FIGURE 2**

Figure 2. (a) The temperature-dependent longitudinal resistivity for the as-prepared samples with different Co fractions (x). (b) Normalized $\rho(T)/\rho(300 \text{ K}) \sim T$ curves for the as-prepared samples with $47 \leq x \leq 100$.

1 **FIGURE 3**

Figure 3. Field-dependent (a) Hall resistivity and (b) magnetization curves measured at 5 K for the as-prepared samples with $47 \leq x \leq 100$. Field-dependent (c) Hall resistivity and (d) magnetization curves obtained at different temperatures for the sample with $x=66$.

1 **FIGURE 4**

Figure 4. The anomalous Hall coefficient (R_s) versus longitudinal resistivity (ρ_{xx}) for all as-prepared samples measured at 5 K. The solid straight line is the least-squares fit to the data.

1 **FIGURE 5**

Figure 5. (a) The anomalous Hall coefficient (R_s) versus longitudinal resistivity (ρ_{xx}) for the annealed samples measured at 5 K. (b) the $R_s \sim \rho_{xx}$ curve for the as-prepared and annealed samples. The solid straight lines are the least-squares fits to the data.

1 **FIGURE 6**

Figure 6. (a) Longitudinal resistivity and (b) anomalous Hall resistivity measured at 5 K as a function of x for as-prepared and annealed samples. The legend is also applied to (c), (d), (e) and (f). The data in (a) [(b)] were replotted in logarithmic scales in (c) and (e) [(d) and (f)]. The solid straight lines are least-squares fits to the data.

1 **FIGURE 7**

Figure 7. Normalized $M \sim H$ curves after zero-field cooling and 70 kOe applied field cooling measured at 2 K for the (a) as-prepared and (b) annealed sample with $x=34$.