Scattering Effect on Anomalous Hall Effect in Ferromagnetic Transition Metals

Dissertation by
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ABSTRACT

Scattering effect on anomalous Hall effect in ferromagnetic transition metals

Qiang Zhang

The anomalous Hall effect (AHE) has been discovered for over a century, but its origin is still highly controversial theoretically and experimentally. In this study, we investigated the scattering effect on the AHE for both exploring the underlying physics and technical applications.

We prepared Co$_x$(MgO)$_{100-x}$ granular thin films with different Co volume fraction ($34 \leq x \leq 100$) and studied the interfacial scattering effect on the AHE. The STEM HAADF images confirmed the inhomogeneous granular structure of the samples. As $x$ decreases from 100 to 34, the values of longitudinal resistivity ($\rho_{xx}$) and anomalous Hall resistivity ($\rho_{AHE}$) respectively increase by about four and three orders in magnitude. The linear scaling relation between the anomalous Hall coefficient ($R_s$) and the $\rho_{xx}$ measured at 5 K holds in both the as-prepared and annealed samples, which suggests a skew scattering dominated mechanism in Co$_x$(MgO)$_{100-x}$ granular thin films.

We prepared $(Fe_{36}/Au_{12})_n$, $(Ni_{36}/Au_{12})_n$ and $(Ta_{12}/Fe_{36})_n$ multilayers to study the interfacial scattering effect on the AHE. The multilayer structures were characterized by the XRR spectra and TEM images of cross-sections. For the three serials of multilayers, both the $\rho_{xx}$ and $\rho_{AHE}$ increase with $n$, which clearly shows interfacial scattering effect. The intrinsic contribution decreases with $n$ increases in the three serials of samples, which may be due to the crystallinity decaying or the finite size effect. In the $(Fe_{36}/Au_{12})_n$ samples, the side-jump contribution increases with $n$, which suggests an interfacial
scattering-enhanced side jump. In the $\left(Ni_{36}/Au_{12}\right)_n$ samples, the side-jump contribution decreases with $n$ increases, which could be explained by the opposite sign of the interfacial scattering and grain boundary scattering contributed side jump. In the $\left(Ta_{12}/Fe_{36}\right)_n$ multilayers, the side-jump contribution changed from negative to positive, which is also because of the opposite sign of the interfacial scattering and grain boundary scattering contributed side jump. The interfacial scattering effect on the AHE is much more complicated than surface scattering in thin films or scattering by delta-impurities in bulk-like samples.
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<td>AHC</td>
<td>anomalous Hall conductivity</td>
</tr>
<tr>
<td>AHE</td>
<td>anomalous Hall effect</td>
</tr>
<tr>
<td>AMR</td>
<td>anisotropic magnetoresistance</td>
</tr>
<tr>
<td>EDX</td>
<td>energy dispersive x-ray spectrometer</td>
</tr>
<tr>
<td>FIB</td>
<td>focused ion beam</td>
</tr>
<tr>
<td>GMR</td>
<td>giant magnetoresistance</td>
</tr>
<tr>
<td>HAADF</td>
<td>high-angle annular dark-field</td>
</tr>
<tr>
<td>MFP</td>
<td>mean free path</td>
</tr>
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<td>MPMS</td>
<td>magnetic property measurement system</td>
</tr>
<tr>
<td>MR</td>
<td>magnetoresistance</td>
</tr>
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<td>OHE</td>
<td>ordinary Hall effect</td>
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<td>PPMS</td>
<td>physical property measurement system</td>
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<td>QHE</td>
<td>quantum Hall effect</td>
</tr>
<tr>
<td>SEM</td>
<td>scanning electron microscope</td>
</tr>
<tr>
<td>SHE</td>
<td>spin Hall effect</td>
</tr>
<tr>
<td>SOC</td>
<td>spin-orbit coupling</td>
</tr>
<tr>
<td>TEM</td>
<td>transmission electron microscopy</td>
</tr>
<tr>
<td>XRD</td>
<td>x-ray diffraction</td>
</tr>
<tr>
<td>XRR</td>
<td>x-ray reflectivity</td>
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<table>
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<tr>
<td>$\rho_{\text{AHE}}$</td>
<td>anomalous Hall resistivity</td>
</tr>
<tr>
<td>$\rho_{xx}$</td>
<td>longitudinal resistivity</td>
</tr>
<tr>
<td>$\rho_{xy}$</td>
<td>Hall resistivity</td>
</tr>
<tr>
<td>$\sigma_{\text{AHE}}$</td>
<td>anomalous Hall conductivity</td>
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<tr>
<td>$\sigma_{xx}$</td>
<td>longitudinal conductivity</td>
</tr>
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Chapter 1 Introduction and Background

1.1 Introduction to Hall effect

Hall effect has a big family that includes ordinary Hall effect, anomalous Hall effect, spin Hall effect, quantum Hall effect, et al. In this part, brief introduction about the concept of these Hall effect will be given.

1.1.1 Ordinary Hall effect (OHE)

In 1879, Edwin H. Hall [1] observed that when a current-carrying conductor is kept in magnetic field, the electrons are pressed to the direction perpendicular to both the current and magnetic field and a voltage appears in this direction. This is called ordinary Hall effect (OHE) if the conductor is non-magnetic material. The measurement schematic diagram is shown in Fig. 1.1. It was quite difficult to understand this phenomenon because electron hadn’t been discovered at that time. Now it’s well know that the electrons experience Lorentz force when moving in the direction perpendicular to the magnetic field and are pressed to the transverse side. This tremendous discovery provides a simple way to

Fig. 1.1 The measurement schematic diagram of ordinary Hall effect.
measure the carrier concentration and mobility in nonmagnetic conductors which plays a very important role in the birth of semiconductor electronics.

1.1.2 Anomalous Hall effect (AHE)

In 1881, Edwin H. Hall found the similar effect in ferromagnetic materials which is ten times larger than that in nonmagnetic material [2]. Because of the stronger effect than OHE, this is called anomalous Hall effect (AHE). The anomalous Hall resistivity directly depends on the instantaneous magnetization. That means even without magnetic field, anomalous Hall voltage can still be available if the material has spontaneous magnetization. The phenomenon of OHE and AHE are quite similar but the underlying physics are much different. The mechanisms of AHE are still under controversy and no universal explanation has been given. This is the key topic of this work. More details of the AHE will be present later.

1.1.3 Spin Hall effect (SHE)

Spin Hall effect (SHE) was predicted by M. I. Dyakonov et al. [3] in 1971. In 1999, J. E. Hirsch [4] predicted both the SHE and inverse SHE. It is proposed that when a charge current circulates in a paramagnetic metal, a transverse spin imbalance will be generated, giving rise to a “spin Hall voltage”. This is called SHE. Similarly, it is proposed that when a spin current circulates, a transverse charge imbalance will be generated, giving rise to a Hall voltage, in the absence of charge current and magnetic field. This is inverse SHE. In fact, the SHE and AHE are essentially the same and originate from the same mechanisms. They both are spin-dependent phenomenon and have spin imbalance in transverse direction. The only difference is that the AHE has transverse Hall voltage due to the spin
polarization of the ferromagnetic materials, while there is no transverse Hall voltage in SHE for non-magnetic metals.

1.1.4 Quantum Hall effect (QHE)

The integer quantization of Hall conductance was originally predicted by T. Ando et al. [5] in 1975. In 1980, K. Klitzing et al. [6] made the unexpected discovery of the quantum Hall effect at low temperature and strong magnetic field. In two-dimensional electron systems, with the increase of magnetic field, the Hall conductance $\sigma_{xy}$ exhibits a series of quantized plateaus, $\sigma_{xy} = \nu \frac{e^2}{h}$, and at the same time, the longitudinal conductivity becomes zero. That means the sample shows insulating property when the Hall conductivity is quantized.

Quantum spin Hall effect (QSHE) which is quantized spin Hall effect was proposed by Kane and Mele in graphene [7]. The Kane-Mele model has zero charge Hall conductance but a spin Hall conductance of exactly $\sigma_{xy}^{spin} = 2 \times \frac{e}{4\pi}$. It is quite difficult to observe the QSHE experimentally in graphene because the energy gap opened by spin-obit interaction is extremely small [8]. Later, Bernevig et al. [9] proposed theoretically that QSHE could be realized in inverted band-gap semiconductors, such as CdTe-HgTe quantum well. Then the QSHE was observed experimentally in CdTe-HgTe quantum well [10].

Quantum anomalous Hall effect (QAHE) is a special kind of the quantum Hall effect that occurs in ferromagnetic materials without a magnetic field. Base on first-principle calculations, Yu et al. [11] predicted that the tetradymite semiconductors Bi$_2$Te$_3$, Bi$_2$Se$_3$ and Sb$_2$Te$_3$ with two dimension and magnetically doping (by Cr and Fe) could exhibit
QAHE with a quantized Hall resistance of $\frac{h}{e^2}$. This prediction was realized experimentally in thin films of (Bi,Sb)$_2$Te$_3$ [12].

In summary, the six types of Hall effect were introduced above. Fig. 1.2 gives the schematic diagram about the concepts of all Hall effects [13]. Among these Hall effects, the debate of mechanisms in AHE remains unsettled. AHE is the main topic of this work. We will give a review about the theoretical and experimental studies in AHE.

1.2 Mechanisms of anomalous Hall effect

Although the AHE has been discovered for more than a century, the mechanisms of AHE are still under controversial experimentally and theoretically. In ferromagnets, the Hall resistivity increases sharply with applied field at low field and shows weak field-dependence at high field. This is quite different from the behaviors of OHE. Kundt [14]
first noticed that the saturate Hall resistivity is roughly proportional to the magnetization in Fe, Co, Ni materials. In 1930’s, an empirical relation between Hall resistivity ($\rho_{xy}$), applied field ($H$) and magnetization ($M$) was established as following [15,16],

$$\rho_{xy} = \rho_{\text{OHE}} + \rho_{\text{AHE}} = R_0 H + 4\pi R_S M,$$

(1.1)

where $\rho_{\text{OHE}}$ is ordinary Hall resistivity, $\rho_{\text{AHE}}$ is anomalous Hall resistivity, $R_0$ is the ordinary Hall coefficient and $R_S$ is the anomalous Hall coefficient. The first item is well known as ordinary Hall effect caused by Lorentz force. However, the underlying physics of second item was still poorly understood at that time. Only from 1950’s, the intrinsic and extrinsic mechanisms were proposed to explain the AHE.

1.2.1 Intrinsic mechanism (KL model)

In 1954, Karplus and Luttinger (KL) [17] proposed that the AHE in ferromagnetic materials could be understood as a consequence of the spin-orbit interaction of polarized conduction electrons. As a result of the spin-orbit interaction the stationary states of the system acquire a left-right asymmetry. When an external electric field is applied, the electrons have an anomalous velocity perpendicular both to the electric field and to the mean direction of magnetization. In ferromagnetic materials, the sum of the anomalous velocity over all occupied band states can be nonzero, giving a contribution to the Hall conductivity $\sigma_{xy}$. This anomalous velocity only depends on the Bloch functions of the electrons and are not very sensitive to the temperature and impurity content. Therefore, this has been referred to intrinsic mechanism of AHE. This intrinsic mechanism is illustrated in Fig. 1.3(a) [18].

The anomalous Hall conductivity is dependent only on the band structure of the perfect crystal. Thus, anomalous Hall conductivity should keep constant for one material.
Resistivity and conductivity in two-dimensional transport are tensors which could be expressed as

\[
\begin{pmatrix}
  j_x \\
  j_y
\end{pmatrix}
= \begin{pmatrix}
  \sigma_{xx} & \sigma_{xy} \\
  \sigma_{yx} & \sigma_{yy}
\end{pmatrix}
\begin{pmatrix}
  E_x \\
  E_y
\end{pmatrix}
\]  

(1.2)

and

\[
\begin{pmatrix}
  \rho_{xx} & \rho_{xy} \\
  \rho_{yx} & \rho_{yy}
\end{pmatrix}
\begin{pmatrix}
  j_x \\
  j_y
\end{pmatrix}
= \begin{pmatrix}
  E_x \\
  E_y
\end{pmatrix},
\]

(1.3)

where \( j \) is the current density, \( E \) is the electric field. Therefore, anomalous Hall conductivity could be expresses as

\[
\sigma_{xy} = \frac{\rho_{xy}}{\rho_{xx}^2 + \rho_{yy}^2} \approx \frac{\rho_{xy}}{\rho_{xx}^2}.
\]

(1.4)

If \( \sigma_{xy} \) keep constant, we should obtain \( \rho_{xy} \propto \rho_{xx}^2 \) which is used to examine intrinsic mechanism experimentally.
When KL model of intrinsic mechanism was proposed, there was no concept of Berry phase. After the proposal of Berry phase [19], the relationship between the anomalous velocity and Berry phase was analyzed by Chang et al. [20] and Sundaram et al. [21] They found that the anomalous velocity of magnetic Bloch electrons could be modified by Berry phase and the anomalous Hall conductivity can be given by the integral of the Berry curvature over all occupied Bloch states. Motivated by the new insight into Berry phase effects on Bloch electrons, the KL mechanism was re-examined by researchers. Intrinsic anomalous Hall conductivity has been evaluated in bcc Fe [22], ferromagnetic semiconductors [23] and oxides [24] using first-principles calculations, which gave quantitative agreement between theory and experiment.

1.2.2 Skew scattering

KL model only considered the AHE in perfect crystal without scattering from disorders or impurities. Soon after KL model, Smit [25,26] argued that AHE should vanish in a perfectly periodic lattice and proposed alternative mechanism, skew-scattering. As shown in Fig. 1.3(c), scattering of a carrier from an impurity introduces a momentum perpendicular to both the incident momentum and magnetization due to spin-orbit interaction. This leads to a transverse current proportional to the longitudinal current driven by electric field. Consequently, the Hall conductivity and longitudinal conductivity are both proportional to the transport lifetime. Equivalently, the anomalous Hall resistivity \( \rho_{AHE} \) is linearly dependent on longitudinal resistivity, \( i.e. \rho_{AHE} \propto \rho_{xx} \). Smit also discussed the contribution of phonon scattering and suggested that, in the skew scattering theory, \( R_S \propto \rho_{xx}^2 \) should hold for AHE due to phonon scattering above the Debye temperature [26].
Kondo [27] also proposed a finite-temperature skew-scattering model in a high-purity monodomain ferromagnet containing dilute magnetic impurities embedded in a nonmagnetic host (the dilute Kondo system). This model considered the asymmetric scattering of nonmagnetic $s$ electrons from spin-wave excitations of ordered $d$-band (or $f$-band) local moments. This skew scattering has been proved in silver, gold and aluminum with low concentration of rare-earth impurities [28].

### 1.2.3 Side jump

Berger [29] also focused on the scattering of impurities or disorders and proposed side jump mechanism. When considering the scattering of a Gaussian wave packet from a spherical impurity with spin-orbit interaction, a wave packet with incident wave vector will suffer a displacement in transverse direction as illustrated in Fig. 1.3(b). This side jump predicted quadratic dependence, $\rho_{AHE} \propto \rho_{xx}^2$, which has the same scaling law with that of intrinsic mechanism. It is quite confusing that the side jump mechanism caused by scattering at impurities does not depend on the density or strength of scatters. Also, it is difficult to distinguish the side-jump from intrinsic mechanism experimentally because of the same scaling law. Actually, the side jump and intrinsic contributions have quite different dependences on more specific system parameters, particularly in systems with complex band structures [30].

### 1.2.4 Universal scaling

A unified theory [31] including intrinsic and skew scattering mechanisms has been proposed recently to explain the AHE in multiband ferromagnetic metals with dilute impurities. Three broad regimes were distinguished depend on the longitudinal conductivity. In clean regime with high conductivity ($\sigma_{xx} > 10^6 \Omega^{-1}\text{cm}^{-1}$), skew scattering
dominates AHC which gives linear dependence $\sigma_{\text{AHE}} \propto \sigma_{xx}$. In intrinsic or scattering-independent regime ($10^4 \, \Omega^{-1}\text{cm}^{-1} < \sigma_{xx} < 10^6 \, \Omega^{-1}\text{cm}^{-1}$), $\sigma_{\text{AHE}}$ is constant. In bad metal regime ($\sigma_{xx} < 10^4 \, \Omega^{-1}\text{cm}^{-1}$), $\sigma_{\text{AHE}} \propto \sigma_{xx}^{1.6}$ is predicted due to the damping of intrinsic contribution. This theory seems to have resolved the controversy on mechanisms of AHE in a whole region. This unified theory has been experimentally proved in itinerant ferromagnets [32], epitaxial bcc Fe(001) thin films [33], low conductivity materials [34] and half-metal oxides [35,36], et al.

1.3 Experiments progress in anomalous Hall effect

Edwin H. Hall [2] first observed this AHE experimentally although he cannot explain the phenomenon. During the study of field-dependent Hall resistivity, Kundt [14] noted that the saturation value is roughly proportional to the magnetization in Fe, Co and Ni, and it has a weak anisotropy when the field direction is rotated with respect to the crystal. After the proposal of the three mechanisms, researchers start to measure the relationship between anomalous Hall resistivity and longitudinal resistivity and obtain the exponential from the scaling law, $\rho_{\text{AHE}} \propto \rho_{xx}^{0.94}$.

1.3.1 Pure Fe, Co and Ni

The scaling law for Fe has been plotted using experimental data by Karplus and Luttinger [17] to validate their intrinsic mechanism theory. The Hall resistivity and longitudinal resistivity were tuned by measuring temperature. Scaling relation $R_S \propto \rho_{xx}^{1.94}$ was obtained which confirmed the intrinsic mechanism in Fe [17]. Kooi [37] studied the scaling law in Fe with Si doping. Low concentration of Si doping could tune the resistivity of samples by large range and the samples could still be regarded as homogenous material. The scaling law was obtained as $R_S \propto \rho_{xx}^{1.9}$ which is quite close to that proposed by Karplus
and Luttinger [17]. Similar scaling law was also observed in dilute Cr doped Fe [38] which, however, was explained by side jump mechanism. While, the AHE was also explained by skew scattering in pure iron or low concentration Si, Co doped Fe [38,39]. In cobalt, the relation $\rho_{AHE} = a\rho_{xx} + b\rho_{xx}^2$ was usually observed [40]. This may give multiple mechanisms coexisting in the studied materials. For nickel, the scaling becomes complicated and various results have been reported. Karplus and Luttinger [17] first give a scaling relation $R_S \propto \rho_{xx}^{1.42}$ for pure Ni. Fert et al. [41] observed linear relation between $\rho_{AHE}$ and $\rho_{xx}$ in Ni doped with ultralow concentration of Mn, Cr or Cu which gives skew scattering dominated AHE. For Ni with several percent impurity doping, the exponential $\gamma$ in $R_S \propto \rho_{xx}^\gamma$ varied from 0.6 to 1.97 [26,42]. Even for one material, Ni with 99.9% purity, the exponential $\gamma$ is not constant and highly depends on the measurement temperature [42] which can hardly be explained by the present mechanisms.

Since the skew scattering and side jump mechanisms do give rise to linear and quadratic dependence of AHE on longitudinal resistivity which have been observed experimentally, the extrinsic mechanisms were accepted [43] to be responsible for the AHE and the intrinsic mechanism had been disregarded for a long time. However, due to a new insight into Berry phase effect, intrinsic mechanism was reconsidered by researchers. Yao et al. [44] evaluated the intrinsic anomalous Hall conductivity (AHC) of bcc Fe by first-principle calculation and the obtained AHC value is 751 $\Omega^{-1}cm^{-1}$ at zero temperature and 734 $\Omega^{-1}cm^{-1}$ at room temperature. The calculated values differed only by 30% from that reported in iron whiskers [45]. For hcp Co, because of the large magnetocrystalline anisotropy, the intrinsic Hall conductivity obtained from the first-principle calculation changes from 481 $\Omega^{-1}cm^{-1}$ along easy axis to 116 $\Omega^{-1}cm^{-1}$ on hard axis and the angular average value is 226
Ω⁻¹ cm⁻¹ [46] which is quite close to the measured value 205 Ω⁻¹ cm⁻¹ [40] in polycrystalline Co. For Ni, Wang et al. considered the intrinsic AHC as Fermi-surface property and computed intrinsic AHC by first-principle method [47]. The calculated value of intrinsic AHC is 2203 Ω⁻¹ cm⁻¹ for fcc Ni. Fuh et al. [48] calculated the intrinsic AHC based on the generalized gradient approximation (GGA) plus on-site electron-electron interaction. The obtained AHC value is 1066 Ω⁻¹ cm⁻¹ which is larger than the previous experimental data −646 Ω⁻¹ cm⁻¹ [42] but in great agreement with the latest measurement result −1100 Ω⁻¹ cm⁻¹ [49] at low temperatures. Now researchers accept the existence of intrinsic mechanism in AHE, but the intrinsic AHC values obtained in first-principle calculations still differ from that in experiments. For theoretical calculation, the calculated value of intrinsic AHC highly depends on the approximation methods which cannot give us the exact value. For experimental results, because of the same scaling law for side-jump and intrinsic mechanism, it is quite difficult to separate the two contributions experimentally. Up to now, the exact value of intrinsic AHC is still unachieved, which needs further investigation.

Recently, a very important progress on AHE has been made in Fe [50,51], Ni [49], and Co [52]. The effect of phonon on skew scattering was proved to be negligible in epitaxial Fe [50] experimentally as suggested in theory [43]. Tian et al. [50] proposed the empirical relation for AHE in Fe:

\[
\rho_{\text{AHE}} = (\alpha \rho_{xx0} + \beta \rho_{xx0}^2) + b \rho_{xx}^2
\]

(1.5)

or, in the form of conductivity,

\[
\sigma_{\text{AHE}} = (\alpha \sigma_{xx0}^{-1} + \beta \sigma_{xx0}^{-2}) \sigma_{xx}^2 + b
\]

(1.6)
where $\alpha$, $\beta$ and $b$ represent the contribution from the skew-scattering mechanism, side-jump mechanism and intrinsic mechanism, respectively. The variables $\rho_{xx0}$, $\sigma_{AHE}$ and $\sigma_{AHE0}$ are, respectively, the residual longitudinal resistivity, the anomalous Hall conductivity and the residual Hall conductivity. The subscript “0” indicates that the data were measured at low temperature where phonon scattering was excluded. Since the thermal effect on transport properties can be neglected at low temperatures, Eq. (1.6) can then be simplified to $\sigma_{AHE0} = \alpha\rho_{xx0} + (\beta + b)$ by taking the following approximations $\sigma_{AHE} \approx \sigma_{AHE0}$ and $\sigma_{xx} \approx \sigma_{xx0}$. By plotting $-\sigma_{AHE0} \sim \sigma_{xx0}$ curve for epitaxial Fe films with different thickness, [50]

![Graph](image)

**Fig. 1.4** (a) $-\sigma_{AH0}$ versus $\sigma_{xx0}$ curve measured at 5 K; (b) $-\sigma_{AH}$ versus $\sigma_{xx}^2$ curves for samples with different thickness. [50]
the value of $\alpha$ (skew scattering contribution) and $\beta + b$ (the sum of AHC caused by side jump and intrinsic mechanism) could be obtained, as shown in Fig. 1.4(a). By linear fitting $-\sigma_{\text{AH}} \sim \sigma_{xx}^2$ curve for each sample as shown in Fig. 1.4(b), the intercepts converge to one point which is the value of $b$ (intrinsic AHC). The intrinsic AHC for bcc Fe is about 1100 $\Omega^{-1} \text{cm}^{-1}$ [50] which is larger than the calculated value ($\sim 750 \Omega^{-1} \text{cm}^{-1}$) [44,47]. For fcc Co(001) films, the intrinsic AHC is 727 $\Omega^{-1} \text{cm}^{-1}$ [52] which is three times as large as the calculated value 249 $\Omega^{-1} \text{cm}^{-1}$ [46]. For fcc Ni, the intrinsic AHC is temperature dependent and changes from 1100 $\Omega^{-1} \text{cm}^{-1}$ at 5 K to 500 $\Omega^{-1} \text{cm}^{-1}$ at 300 K [49], which is comparable with the calculated value 1066 $\Omega^{-1} \text{cm}^{-1}$ [48].

In these literatures [49–52], they investigated epitaxial Fe, Co and Ni films with different thickness. Surface scattering was used to tune the longitudinal resistivity and Hall resistivity. Actually, the three mechanisms [17,25,26,29] were proposed theoretically based on bulk samples with/without impurities which may not be used in surface scattering dominated thin films. Also, Zhang [53] suggested that the scaling laws are not valid in structures where the mean free path (MFP) of carriers is limited by geometry. Therefore, the new scaling relation in Eqs. (1.5) and (1.6) is not that convincing and the validity needs further exploration.

1.3.2 Granular thin film with Fe, Co and Ni

During the long-running controversy, one of the main issues is that if the AHE comes from scattering of impurities or disorders. To verify this experimentally, material doping was used as scattering center in the study of AHE. When the doping level is very low (dilute impurities), we can still regard the material as homogenous system. If the concentration of doped material is high enough, we should consider the material as inhomogeneous system.
We will give a brief review about the granular systems with Fe, Co and Ni matrix which have relatively high doping level.

Giant Hall effect was first observed in percolating Ni-SiO$_2$ thin films which could have application in high sensitivity Hall sensor [54]. Similar results have been reported in Fe-SiO$_2$ [55], Co-SiO$_2$ [56], and Ni$_{80}$Fe$_{20}$-SiO$_2$ [57], et al. This granular structure could also be used for AHE mechanism exploration. The interfaces between Fe (Co or Ni) and nonferromagnetic materials could play as scatter center. Various scaling relations have been reported. In Co−Ag granular samples [58,59], a scaling relation $\rho_{AHE} \propto \rho_{xx}^{3.7}$ was reported which cannot be explained by any of the three mechanisms above. In insulating Fe−SiO$_2$ granular samples [60], the exponents $\gamma$ in relation $\rho_{AHE} \propto \rho_{xx}^\gamma$ are about 0.5 which are much lower than predicted values in the three mechanisms. Interfacial scattering in Ni-SiO$_2$ granular thin films lead to linear relation $\Delta\rho_{AHE} \propto \Delta\rho_{xx}$ [61] which could be explained by skew scattering mechanism. While quadratic relation $\rho_{AHE}/n \propto \rho_{xx}^2$ was reported in Fe−SiO$_2$ granular thin films and suggested scattering-independent AHE [62].

As shown above, the scaling relations are quite complicated in granular samples. And also, the scattering strength/potential is less controllable. The interface area does not only depend on the composition but also the grain size in granular samples, which may lead to the complicated scaling relations.

### 1.3.3 Multilayers

Ferromagnet/nonferromagnet multilayers are another kind of inhomogeneous system besides granular samples. Multilayers are more controllable to study the surface/interface scattering and explore the mechanisms of AHE.
For classical GMR metallic system, AHE in Fe/Cr multilayers gives a scaling relation \( R_s \propto \rho_{xx}^{2.6} \), which was explained by spin-dependent interface scattering [63]. Similar deviations from the commonly used scaling relation \( \rho_{AHE} \propto \rho_{xx}^{\gamma} \) \((1 < \gamma < 2)\) were also reported in Co/Pt [64], et al. Skew scattering and side jump are claimed to dominate the AHE in Co/Pd [65] and Co/Cu [66] multilayers, respectively, according to the scaling relations. The scaling laws in multilayers are also quite complicated and controversial. Zhang [53] analyzed the AHE theoretically and suggested the commonly used scaling relation is not valid in magnetic multilayers when the layer thickness is thinner than the MFP of carriers. This means it is not that meaningful to use scaling relation to explore the mechanisms of AHE in multilayers. This could also be applied to granular systems when the MFP is limited by the distance between nonmagnetic clusters.

Researchers also studied the effect of surface and interface on AHE. It has been reported the sign of surface scattering induced AHE is opposite to that of interface scattering in Co/Pd multilayers [67,68]. Interfacial contribution to AHE has an opposite sign to the bulk contribution in Co/Pd bilayers [69]. Surface scattering was found to dominate the AHE in Ni films [70,71], Co/Pd [65] multilayers, et al. Interface scattering was also found to dominate the AHE in Fe/Cr [63], Co/Pt [64,72], Co/Pd [73], et al. Furthermore, artificial interface could enhance the AHE in MgO/(Co/Pt)_n/MgO [74], (Pt/Co)_s/Ru/(Co/Pt)_s [75] and SiO\(_2\)/FePt/SiO\(_2\) [76] films. Obviously, the effect of surface/interface scattering on AHE is still controversial which may need further investigation.

In multilayers, because of the broken symmetry of surfaces/interfaces, the scatterings are quite different to that from delta-impurities in bulk samples. As suggested by Zhang [53], the spin-dependent scattering may lead to the invalidity of scaling laws. Therefore,
the AHE mechanism exploration using scaling laws in multilayers is still in doubt. Qualitatively analysis is possible to demonstrate the surface/interface scattering effect on AHE in multilayers.

1.4 Open questions and research in this work

From the review of AHE above, great progress has been achieved. However, there are still some open questions in studying the AHE as following:

(1) In granular thin films, the nonmagnetic clusters were added to provide scattering center and adjust the scattering potential/strength. However, the interfaces of ferromagnetic/nonmagnetic materials are quite difficult to be controlled experimentally. The interface diffusion may change the transport properties. Especially, reaction happened between ferromagnetic materials (such as Fe, Co, Ni) and nonferromagnetic materials (e.g. SiO₂) may change the magnetic properties and thus the scaling relation of AHE. The scaling relation $\rho_{\text{AHE}} \propto \rho_{xx}^\gamma$ with $\gamma < 1$ has been reported [54,57,60], but its theoretical interpretation remains elusive. Also, the validity of scaling laws in this inhomogeneous system is still in doubt as suggested by Zhang [53].

(2) In multilayer studies, the short-circuit and shunting effects of nonmagnetic layers could be a problem for the scaling relation [77,78]. Special structure design may be needed to avoid this problem. Also, as suggested by Zhang [53], the commonly used scaling law is not valid when the MFP is limited by layer thickness. Usually, in order to study the interfacial scattering, the thickness of each layer in multilayers is quite low. Spin-dependent scattering happened at interfaces may lead to abnormal exponents in scaling relations [58,63]. Obtaining a complete picture of scattering is necessary to avoid invalid scaling exploration.
(3) The role of interface and surface scattering in AHE is still controversial in multilayers. Recent experiments successfully separated the contributions of the three mechanisms in Fe [50], Ni [49], Co [52] using Eqs. (1.3) and (1.4). In their work, they only used surface scattering to tune the longitudinal resistivity and Hall resistivity. If using the interface scattering, the validation of the empirical relation of Eqs. (1.3) and (1.4) is in doubt, which may need further clarification.

In this study, we did the following works to investigate the interfacial scattering effect on the AHE:

(1) We prepared Co_x(MgO)_{100-x} granular thin films with volume fraction x and study the scattering effect on AHE. We choose MgO as the insulate material because of its good electrical isolation and less diffusion between MgO and Co. MgO is more difficult to react with TM materials even at high deposition or annealing temperature [50]. Annealing process was done to adjust the scattering strength/potential to study the scattering effect on the AHE.

(2) To better control the interfaces, we prepared (Fe_{36n}/Au_{12n})_n, (Ni_{36n}/Au_{12n})_n and (Ta_{12n}/Fe_{36n})_n (thickness in nanometers) multilayers to study the interfacial scattering effect on the AHE. In these samples, the thickness of all samples and the thickness ratio of Fe/Au, Ni/Au and Fe/Ta are the same. The only difference is the numbers of the interfaces, which is beneficial to the study of the interfacial scattering effect on the AHE.
Chapter 2 Experiments

In this chapter, the experimental procedures of thin film deposition and sample treatment are described. The characterization techniques for the samples are also introduced.

2.1 Sample preparation and treatment

The Co$_x$(MgO)$_{100-x}$ granular samples were prepared by co-sputtering. The Fe(Ni)/Au multilayers were prepared by e-beam evaporator. The Ta/Fe multilayers were fabricated by Singulus sputter system. The techniques with specifications for sample preparation are introduced below. Sample annealing process is also briefly described.

2.1.1 Granular thin film deposition

The granular thin films, Co$_x$(MgO)$_{100-x}$, with 100-nm thickness were deposited by a magnetron sputter system (ESCRD4, Equipment 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 SiO$_2$ (99.995% purity), with 2-inch diameter were mounted. The granular thin films were prepared on oxidized silicon wafer by co-sputtering Co and MgO targets. 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 $6.0 \times 10^{-8}$ 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 volume fraction ($x$) of Co was controlled by varying the
sputter power ratio of Co and MgO targets. After the deposition, ~20 nm SiO₂ thin films were deposited on the samples without breaking the vacuum to prevent the oxidation.

### 2.1.2 Multilayer thin film deposition

\[(\text{Fe}_{36}/\text{Au}_{12})_n\] and \[(\text{Ni}_{36}/\text{Au}_{12})_n\] (\(n = 1, 2, 3, 4, 5, 6, 8, 10, 12\)) (thickness in nanometer) multilayers were deposited using an e-beam evaporator (EXPLORER 14, Denton Vacuum). High purity Fe(Ni) (99.95%) and Au (99.995%) pallets were used as source materials. The samples were deposited, at room temperature, on substrates of oxidized silicon wafers with the base pressure lower than 2.0\times10^{-7} \text{Torr}. The deposition rate (1.0 Å/s) and thickness were monitored by a quartz deposition controller (XTC/3S, Inficon). The distance between the material sources and the substrate is about 51 cm. During the deposition, the substrate was rotating at a speed of 60 r/min. The Fe(Ni) and Au were alternatively deposited on the substrates and the top layer of each sample was always an Au layer to prevent oxidation.

\[(\text{Ta}_{12}/\text{Fe}_{36})_n\] (\(n = 1, 2, 3, 4, 5, 6, 8, 10, 12\)) (thickness in nanometer) multilayers were prepared by a magnetron sputter system (Rotaris, Singulus). The samples were deposited, at room temperature, with the base pressure lower than 8.0 \times 10^{-9} \text{mbar}. The deposition of SiO₂ with 5 nm-thickness as a top layer for each sample was to prevent sample oxidization. The diameter of target is 100 mm for each material deposited. The deposition power of Ta and Fe layers is 800 W and that of SiO₂ is 500 W. During deposition, the gas atmosphere was argon with flow rate of 55 sccm and process pressure was 3 \times 10^{-3} \text{mbar}.

### 2.1.3 Sample annealing

Granular thin film samples, Coₓ(MgO)₁₀₀₋ₓ, were annealed in a tube furnace (OTF−1200X, MTI Corporation) with high vacuum system. Before annealing process, the
tube furnace was pumped to vacuum below $1 \times 10^{-4}$ Torr. Then the temperature increased from room temperature to 300 °C by rate of 10 °C/min and stayed at 300 °C for one hour. After that, the temperature decreased to room temperature naturally under vacuum.

2.2 Sample characterization

Several techniques were used for sample characterization including structure, composition, electro-/magneto-transport and magnetic properties. These techniques are introduced below respectively.

2.2.1 X-ray diffraction (XRD)

The high angle $\theta–2\theta$ scan was performed by X-ray diffraction (XRD) (D8 Advance, Bruker) for the granular samples and the multilayer samples. During the scanning, the high voltage and current of X-ray tube are set to 40 kV and 40 mA, respectively. The increment step is 0.01° and the scan speed is 1.0 sec/step.

The X-ray reflectivity was measured by XRD (D8 Discovery, Bruker) for multilayer samples to characterize the multilayer structure. During the scanning, the high voltage and current of X-ray tube are set to 40 kV and 40 mA, respectively. Knife edge was used on the samples to avoid the direct X-ray beam. $\theta–2\theta$ scan was performed from 0° to 6°. The increment step is 0.002° and the scan speed is 2.0 sec/step. After the measurement, the curves were fitted using LEPTOS software suite (V7.02, Bruker) to get the thickness of each layer and interfacial roughness.

2.2.2 Transmission electron microscopy (TEM)

The microstructure and morphology of the $\text{Co}_x(\text{MgO})_{100-x}$ granular samples was characterized by a scanning transmission electron microscopy (STEM) (Titan 80–300,
FEI). A Schottky hot field emission electron gun, equipped in the TEM, was operated at 300 kV during imaging. The Co$_x$(MgO)$_{100-x}$ samples for STEM characterization were deposited directly on Cu grids with ultrathin carbon films. The STEM high-angle annular dark-field (HAADF) images of the samples were acquired to show the phase separation of Co and MgO.

The planar-view bright field TEM images of the cross sections of the multilayer specimens were acquired to demonstrate the multilayer structure. Elemental profiles of the multilayer samples were obtained by line scanning of energy-dispersive x-ray spectroscopy (EDX) in a STEM.

2.2.3 Scanning electron microscope (SEM)

The volume fraction of Co for Co$_x$(MgO)$_{100-x}$ granular samples was confirmed by an energy dispersive x-ray spectrometer (EDX) on a scanning electron microscope (SEM) (Quanta 600, FEI). During the EDX data collection, the SEM electron gun was operated at 30 kV.

The multilayer specimens for TEM characterization were lifted out from the macroscopic samples deposited on silicon substrates by a focus ion beam (FIB) equipped in a scanning electron microscope (SEM) dual beam system (Helios NanoLab 400s). The prepared specimens are less than 100-nm-thick.

2.2.4 Physical property measurement system (PPMS)

The electrical transport properties of samples were measured by a physical property measurement system (Dynacool 14 T, Quantum Design). The measurement temperature ($T$) range and magnetic field ($H$) range are $5 \text{ K} \leq T \leq 300 \text{ K}$ and $-50 \text{ kOe} \leq H \leq 50 \text{ kOe}$. The samples were patterned using mechanical masks during the deposition. Five-contact
geometry was made for simultaneously measuring the Hall resistivity and longitudinal resistivity on the same piece of sample. We applied DC current and then measured the longitudinal voltage ($V_{xx}$) and Hall voltage ($V_{xy}$) as shown in Fig. 2.1. The longitudinal resistivity was calculated using $ho_{xx} = \frac{V_{xx}}{I} \times \frac{w \cdot t}{L}$, where $w$ is the width (1.00 mm) of samples, $t$ is the thickness and $L$ is the length (2.00 mm) between the electrodes of longitudinal Voltage. Hall resistivity is calculated by $\rho_{xy} = \frac{V_{xy}}{I} \cdot t$. The samples were connected to DC sample holder of PPMS with copper wires and the electric contacts are indium disks.

2.2.5 Magnetic property measurement system (MPMS)

The magnetic properties were measured by a magnetic property measurement system (MPMS3, Quantum Design). The measurement temperature ($T$) range and magnetic field ($H$) range are $5 \text{ K} \leq T \leq 300 \text{ K}$ and $-50 \text{ kOe} \leq H \leq 50 \text{ kOe}$. The samples deposited on

![Diagram of samples and measurement configuration](image)

**Fig. 2.1** Schematic of samples patterned by mechanical masks and measurement configuration.
Si were cut to pieces with dimensions ~3 mm×3 mm. Brass sample holder was used and the measurement mode is VSM.
Chapter 3 Anomalous Hall effect in $\text{Co}_x(\text{MgO})_{100-x}$ granular thin films

3.1 Anomalous Hall effect of as-prepared $\text{Co}_x(\text{MgO})_{100-x}$ thin films

3.1.1 Compositions and structural properties

EDX analysis was used to characterize the compositions of $\text{Co}_x(\text{MgO})_{100-x}$ thin films. We selected 13 samples with different deposition conditions for EDX analysis. The preparation conditions and the composition obtained by EDX are listed in Table 3.1.

XRD characterization was done for all samples deposited on glass substrates. Figure 3.1 shows the $\theta$–2$\theta$ scan from 30° to 80°. For pure Co sample ($x = 100$), we can see three peaks which are hexagonal structure of Co (100), (002) and (110). The peaks are broad and intensity is quite low, which indicates the poor crystallinity of Co film. With Co fraction decreases, the peaks are even broader with lower intensity. For the samples with lower Co fraction, there is no diffraction peak, which indicates worse crystallinity or even amorphous structure. There is no peak shown for MgO in all samples because MgO has low fraction and may have small cluster size with amorphous structure in the samples.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Co power (W)</th>
<th>MgO power (W)</th>
<th>Co volume fraction ($x$)</th>
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<tbody>
<tr>
<td>1</td>
<td>100</td>
<td>0</td>
<td>100</td>
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<tr>
<td>2</td>
<td>113</td>
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</table>
TEM imaging is a direct way to demonstrate the granular structure in metal-insulator composite thin films. Usually, the planar-view bright field TEM images are done to investigate the microstructure and morphology of granular samples. However, the bright and dark contrast is not only due to the elements with different atomic numbers but also due to the diffraction contrast if the samples are polycrystalline, which may give unreliable conclusions of the granular structures. To avoid this diffraction contrast, we did STEM analysis with high-angle annular dark-field (HAADF) images for the selected Co\textsubscript{x}(MgO)\textsubscript{100-x} samples. The HAADF images are formed by scattered electrons at high angle, which are only sensitive to the variation of the atomic numbers of the elements. The samples for STEM characterization were deposited directly on Cu grids with ultrathin carbon films at the same time with that for other analysis to assure the similar structure and composition. Figure 3.2 shows the STEM HAADF images of the samples with x = 81 and x = 66. The images show a clear contrast of black and white areas, a feature of granular 

Fig. 3.1 XRD patterns for all as-prepared Co\textsubscript{x}(MgO)\textsubscript{100-x} samples.
structured materials. The bright regions correspond to the metallic Co 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 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 electrical properties.

3.1.2 Longitudinal resistivity

To understand the electrical properties and explore the electrical transport mechanisms, we measured the temperature-dependent longitudinal resistivity ($\rho_{xx} \sim T$) curves for all as-prepared samples at temperatures ranging from 5 K to 300 K with zero magnetic field, as shown in Fig. 3.3. The pure Co sample ($x = 100$) had a positive temperature coefficient of resistivity (TCR, $d\rho_{xx}/dT$) 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. Normally, this negative TCR could be explained as weak localization in metallic samples. In weak localization theory, the resistivity-temperature relation depends on the dimensions of the samples. In thin-film samples, if the diffusion length, $L = \sqrt{D\tau_{ln}}$ (where $D$ is the electron diffusion constant and $\tau_{ln}$ is the mean free time of the inelastic

\[ \begin{align*}
\rho_{xx}(\Omega \text{ cm})
\end{align*} \]

\[ \begin{align*}
T(\text{K})
\end{align*} \]

**Fig. 3.3** The temperature-dependent longitudinal resistivity for the as-prepared samples with different Co volume fraction ($x$).
scattering) is larger than the sample thickness, we can regard this sample to be a two-dimensional system; otherwise it is a three-dimensional system. In two-dimensional systems, the resistivity-temperature relation satisfies $\rho_{xx} \propto \ln T$; in three-dimensional system, this relation satisfies $\rho_{xx} \propto \sqrt{T}$. We applied the two relations to the data at low temperatures.

<table>
<thead>
<tr>
<th>$T$ (K)</th>
<th>$\rho_{xx}$ (µΩ cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4</td>
<td>3900.0</td>
</tr>
<tr>
<td>1.6</td>
<td>249.10</td>
</tr>
<tr>
<td>1.8</td>
<td>1236.0</td>
</tr>
<tr>
<td>2.0</td>
<td>6400.0</td>
</tr>
<tr>
<td>2.2</td>
<td>1736.0</td>
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<tr>
<td>2.4</td>
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<td>2.6</td>
<td>1736.0</td>
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<td>2.8</td>
<td>6400.0</td>
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<tr>
<td>3.0</td>
<td>1236.0</td>
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<tr>
<td>3.8</td>
<td>1736.0</td>
</tr>
<tr>
<td>4.0</td>
<td>6400.0</td>
</tr>
</tbody>
</table>

**Fig. 3.4** $\rho_{xx} \sim \ln T$ curves for the samples with $47 \leq x \leq 81$ at low temperatures. The solid red lines are least-squares fits to the data.
temperatures and found that $\rho_{xx} \propto \ln T$ fits the curves very well, indicating the likelihood of the two-dimensional weak localization effect at low temperatures. Figure 3.4 presents the plots $\rho_{xx} \sim \ln T$ for the samples with $47 \leq x \leq 81$ at low temperatures and indicates the linear dependency.

Figure 3.5 presents the normalized $\rho_{xx}(T)/\rho_{xx}(300 K) \sim T$ curves for the samples with $47 \leq x \leq 100$ to capture the scattering effect. The resistivity ratio $\rho_{xx}(T)/\rho_{xx}(300 K)$ is a useful empirical parameter for quantifying the extent of disorder. Generally, as the disorder increases, the resistivity decreases more slowly upon lowering the temperature [79]. As shown in Fig. 3.5, the $\rho_{xx}(T)/\rho_{xx}(300 K)$ increases as $x$ decreases, which clearly demonstrates that there is more interfacial scattering in the samples with lower $x$.

For the samples with low Co volume fraction ($x \leq 42$), the TCR 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

![Fig. 3.5](image.png)

Fig. 3.5 Normalized temperature-dependent longitudinal resistivity curves for the samples with $47 \leq x \leq 100$. 

disordered alloys could be positive or negative \[80, 81\]. Zabrodskii et al. \[82\] has developed one method by analyzing the temperature dependence of the reduced activation energy of the conductivity

$$w \equiv \frac{\varepsilon}{kT} = \frac{1}{T} \frac{\partial \ln \rho_{xx}}{\partial 1/T}. \quad (3.1)$$

The variation of $\rho_{xx}(T)$ could be expressed as

$$\rho_{xx}(T) = BT^{-m} \exp\left(\frac{T_0}{T}\right)^n, \quad (3.2)$$

where $B$, $m$, $T_0$ and $n$ are constants. $n$ 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. The $\rho_{xx}\sim T$ curves of the samples with $34 \leq x \leq 42$ were fitted by Eq. (3.2) as shown in Fig. 3.6. We obtained the constant $n$ as 0.084, 0.080, 0.076 and 0.210 for the samples with $x = 42$, 39, 36 and 34, respectively. The activation energy decreases with temperature if $0 < n < 1$ and increases with temperature decreases if $n > 1$. $n$ is positive for the four curves, which indicates that the conduction is in the insulating regime \[80, 83\]. The value of $n$ can be used to distinguish the conduction mechanism. $n = 1$ indicates thermal activation conduction, $n = 1/2$ tunneling and $n = 1/4$ Mott variable range hopping (VRH) \[81\]. However, only the sample $x = 47$ with $n = 0.21$ can be assigned to the VRH mechanism and other samples can’t be explained by any of the mechanisms mentioned above. This may because of the inhomogeneous structure and variable mechanisms co-exist in the samples.

Overall, the electrical properties of the samples evolved from metallic to insulating as the Co volume fraction decreased from 100 to 34. 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 longitudinal resistivity from sample $x = 100$ to $x = 34$.

3.1.3 Field-dependent Hall resistivity and magnetization

We now turn to examining how interfacial scattering affects anomalous Hall resistivity. The field-dependent Hall resistivity ($\rho_{xx} \sim \mathcal{R}$) and magnetization ($M \sim \mathcal{H}$) for all samples were measured with a magnetic field applied perpendicularly to the film plane in a range of $-50 \text{kOe} \leq \mathcal{H} \leq 50 \text{kOe}$ and at temperatures ranging from 5 K to 300 K. Figure 3.7(a) and (c) show the field-dependent Hall resistivity of selected samples as indicated in the caption. The $\rho_{xy}$ increases sharply at low field and shows a weak and linear dependence on $\mathcal{H}$ at high fields, which are typical features for ferromagnetic materials. We ascribe the weak and linear field dependence of the Hall resistivity at high fields mainly to the ordinary
Hall resistivity where the magnetization is saturated. The sign of $\rho_{\text{AHE}}$ for all samples is positive across the whole measured temperature range, which is consistent with the

Fig. 3.7 Field-dependent (a) Hall resistivity and (b) magnetization curves for selected samples with different $x$. Field-dependent (c) Hall resistivity and (d) magnetization curves obtained at different temperatures for sample with $x = 66$.

Hall resistivity where the magnetization is saturated. The sign of $\rho_{\text{AHE}}$ for all samples is positive across the whole measured temperature range, which is consistent with the
previous reports [56,84]. The sign of $\rho_{OHE}$ is negative, suggesting that electrons dominate transport properties. Figure 3.7(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 field is due to the diamagnetic contribution from the silicon substrates or/and sample holders. The magnetization curves have similar behaviors to that of the Hall resistivity. The saturation magnetization ($M_s$) measured at 5 K decreases with $x$. Shown in Fig. 3.7(d) are the $M\sim H$ curves measured at different temperature 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, $\rho_{xy}$ is strongly temperature-dependent.

Both Hall resistivity and magnetization curves have hysteresis loops and the coercivity ($H_c$) increases as $x$ decreases. However, the $H_c$ in $\rho_{xy}\sim H$ curves is different from that in the corresponding $M\sim H$ curves at 5 K, which seems to violate the relation established by Eq. (1.1). We will explain the coercivity difference in details later.

3.1.4 Explore the mechanism of anomalous Hall effect

To explore the AHE mechanisms in these samples, we extracted $\rho_{AHE}$ from the field-dependent Hall resistivity curves by extrapolating the high-field data to zero field. Figure 3.8(a) and (b) shows the $\rho_{xx}$ and $\rho_{AHE}$ as a function of $x$ measured at 5 K. Overall, the magnitude of $\rho_{xx}$ increases by nearly four orders as $x$ decreases from 100 to 34 while that of $\rho_{AHE}$ increases by three orders. This giant change in $\rho_{xx}$ and $\rho_{AHE}$ is essential for us to make a reliable scaling for the mechanism exploration. Since these samples clearly belong to a percolation system, it would be interesting to explore if these $\rho_{xx}$ and $\rho_{AHE}$ data can be described by percolation theory. Based on percolation theory [85,86], both the $\rho_{xx}$ and
\( \rho_AHE \) scale with the volume fraction of the metallic components, i.e.

\[
\rho_{xx} \propto (x - x_c)^{-t}
\]  \hspace{1cm} (3.3)

and

\[
\rho_{AHE} \propto (x - x_c)^{-g}
\]  \hspace{1cm} (3.4)

where \( x_c \) is the classical percolation threshold and \( t(g) \) is a critical exponent. We least-squares fitted the data in Fig. 3.8(a) and (b) to Eq. (3.3) and (3.4) and replotted them in logarithmic scales. As shown by solid lines in Fig. 3.8(c) and (d), we obtained the exponent value \( t = 3.74 \) for longitudinal resistivity and \( g = 3.05 \) for anomalous Hall resistivity.

The data show good linear dependence when \( x_c = 27 \) and 26 for \( \rho_{xx} \) and \( \rho_{AHE} \), respectively. The values of \( x_c \) are higher than the value (\( \approx 15 \) [85,86]) in the ideal 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 [87,88], 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 [86,89] ($t \approx 2$ and $g \approx 0.4 - 0.45$) in an ideal 3D percolation system but lower than those ($t = 4.5$ and $g = 3.5$) reported in quasi-2D (NiFe)$_x$(SiO$_2$)$_{100-x}$ percolation system [90].

Now we explore the mechanisms of AHE for our data. Usually, the relations

$$\rho_{\text{AHE}} \sim \rho_{xx}^y$$  \hspace{1cm} (3.5)

or

$$\rho_{\text{AHE}} = a\rho_{xx} + b\rho_{xx}^2$$  \hspace{1cm} (3.6)

are employed to verify the mechanisms. The premise for using the two equations is that $M_s$ should be kept the same for different $\rho_{\text{AHE}}$ 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 Eq. (1.1). In materials in which the Curie temperature is high, $M_s$ is almost the same below room temperature. Using the scaling law between $\rho_{\text{AHE}}$ and $\rho_{xx}$ is appropriate for Fe [50] and Ni [49]. It is even appropriate for the spinel CuCr$_2$Se$_4$-$_x$Br$_x$ with different $x$. If $M_s$ is insensitive to $x$, $\rho_{\text{AHE}}/n \sim \rho_{xx}^2$ ($n$ is the carrier density) is good enough to demonstrate that the mechanism is intrinsic [91]. In our Co$_x$(MgO)$_{100-x}$ system,
\(M_s\) measured at 5 K decreases with \(x\) as shown in Fig. 3.7(b) and the variation in \(M_s\) cannot be ignored in the scaling relations. We can therefore plot of \(\rho_{AHE}/M_s \sim \rho_{xx}^\gamma\) for our system. Using the 5 K data to study the AHE mechanism excludes the phonon effect on the AHE, because photon scattering may increase the exponent \(\gamma\) in Eq. (3.5) from 1 to 2 [18], which would make the system too complicated to verify the mechanisms in our samples. We focused only on the effect of impurities on the AHE in this study. We plotted the curve of \(R_s\) versus \(\rho_{xx}\) logarithmically in Fig. 3.9 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 \(\rho_{AHE}\) (three orders) and \(\rho_{xx}\) (nearly four orders). This perfect linear dependence of \(R_s\) on \(\rho_{xx}\) across the orders of magnitude in both \(\rho_{AHE}\) and \(\rho_{xx}\) strongly suggests that skew scattering dominates.

\[
\begin{align*}
\rho_{xx} (\Omega \text{ cm}) & \quad R_s (\Omega \text{ cm/G}) \\
10^{-1} & 10^{-9} \\
10^{-2} & 10^{-8} \\
10^{-3} & 10^{-7} \\
10^{-4} & 10^{-6} \\
10^{-5} & 10^{-5} \\
10^{-6} & 10^{-4} \\
10^{-7} & 10^{-3} \\
10^{-8} & 10^{-2} \\
10^{-9} & 10^{-1}
\end{align*}
\]

\(x=100 \quad x=66 \quad x=58 \quad x=50 \quad x=47 \quad x=42 \quad x=39 \quad x=36 \quad x=34 \quad x=32 \quad x=29 \quad x=26 \quad x=23 \quad x=20 \quad x=17 \quad x=14 \quad x=11 \quad x=8 \quad x=5 \quad x=2 \quad x=0.5 \quad x=0.2 \quad x=0.1 \quad x=0.05 \quad x=0.01 \quad x=0.001 \quad x=0.0001 \quad x=0.00001 \quad x=0.000001 \quad x=0.0000001 \quad x=0.00000001 \quad x=0.000000001 \quad x=0.0000000001 \quad x=0.00000000001 \quad x=0.000000000001 \quad x=0.0000000000001 \quad x=0.00000000000001 \quad x=0.000000000000001 \quad x=0.0000000000000001 \quad x=0.00000000000000001 \quad x=0.000000000000000001 \quad x=0.0000000000000000001 \quad x=0.00000000000000000001 \quad x=0.000000000000000000001 \quad x=0.0000000000000000000001 \quad x=0.00000000000000000000001 \quad x=0.000000000000000000000001 \quad x=0.0000000000000000000000001 \quad x=0.00000000000000000000000001 \quad x=0.000000000000000000000000001 \quad x=0.0000000000000000000000000001 \quad x=0.00000000000000000000000000001 \quad x=0.000000000000000000000000000001 \quad x=0.0000000000000000000000000000001 \quad x=0.00000000000000000000000000000001 \quad x=0.000000000000000000000000000000001 \quad x=0.0000000000000000000000000000000001 \quad x=0.00000000000000000000000000000000001 \quad x=0.000000000000000000000000000000000001
\]

**Fig. 3.9** The anomalous Hall coefficient \((R_s)\) versus longitudinal resistivity \((\rho_{xx})\) for all as-prepared samples measured at 5 K. The solid red straight line is the least-squares fit to the data.
the AHE in the Co$_x$(MgO)$_{100-x}$ system and contributions from intrinsic and/or side-jump mechanisms could be easily excluded.

3.1.5 Conclusion

In this part, Co$_x$(MgO)$_{100-x}$ thin films with Co volume fraction $x$ from 100 to 34 were prepared by co-sputtering of Co and MgO. The microstructure, temperature-dependent longitudinal and anomalous 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 isolating 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. This is the key result of this work which suggests a skew scattering dominated mechanism in Co$_x$(MgO)$_{100-x}$ granular thin films.

3.2 Anomalous Hall effect of annealed Co$_x$(MgO)$_{100-x}$ thin films

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 [92]. 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 metal clusters may also improve the metallic contacts between the metal clusters. A reduced Hall effect in addition to improved conductivity has been observed in Cu–SiO$_2$ granular materials after annealing under proper conditions [87]. Therefore, we annealed
the Co$_x$(MgO)$_{100-x}$ samples at 300 °C to explore how the AHE and its scaling relation with longitudinal resistivity are affected by variations in interface and microstructure changes.

### 3.2.1 Longitudinal resistivity

Temperature-dependent longitudinal resistivity of the annealed samples was measured at temperatures ranging from 5 K to 300 K with zero magnetic field. Figure 3.10 gives the $\rho_{xx} \sim T$ curves for all annealed samples. For sample $x = 100$, $\rho_{xx}$ decreases with temperature, which shows metallic conduction. At low temperatures, there is no weak localization effect, which indicates that this sample is quite clean (with less scattering) after annealing. For the sample with $36 \leq x \leq 81$, at high temperatures, the $\rho_{xx} \sim T$ curves show metallic conduction. At low temperature, $\rho_{xx}$ increases with temperature decreases, which shows localization effect. We defined the $T_{min}$ as the temperature where the $\rho_{xx}$ is minimum. $T_{min}$ increases with $x$ decreases, which shows a stronger localization effect in the samples with lower $x$. To explore the conduction mechanisms at low temperatures, we replotted the $\rho_{xx} \sim T$ curves below $T_{min}$ as $\rho_{xx} \sim \ln T$ and $\ln \rho_{xx} \sim T^{-1/4}$. The relations $\rho_{xx} \propto \ln T$ and $\ln \rho_{xx} \propto T^{-1/4}$ represent the mechanisms of 2D weak localization and Mott variable range hopping (VRH). However, we found that both $\rho_{xx} \sim \ln T$ and $\ln \rho_{xx} \sim T^{-1/4}$ show linear dependence for each sample below $T_{min}$ (The fittings are not shown here). It is therefore difficult to identify which mechanism dominates the conduction. Usually, the two mechanisms can’t coexist in one sample because weak localization happens in samples with metallic conduction while VRH with semiconducting/insulating conduction. The value of $n$ in Eq. (3.2) can be used to identify if the samples are in metallic, critical or insulating regimes. From the Eq. (3.2), $\log(w) \approx \log(nT_0^n) - n\log T$ can be obtained where $w$ is defined by Eq. (3.1) and we can easily find the $n$ by plotting $\log(w) \sim \log T$. 
Figure 3.11 shows the $w \sim T$ curves logarithmically for samples with $36 \leq x \leq 54$. The solid lines are the linear least-squares fits to the curves at low temperatures. We can find that $n$ is negative for samples $x = 54, 50$ and $47$, which indicates metallic conduction. $n$ is positive for the samples with $x = 42, 39$ and $36$ and these samples are in the insulating regime. Therefore, the $\rho_{xx} \sim T$ behavior at low temperatures for the samples with $47 \leq x \leq 81$ can
be explained by 2D weak localization. And the VRH mechanism should be applied for the samples with $34 \leq x \leq 42$. The $\rho_{xx} \sim \ln T$ curves for samples with $47 \leq x \leq 81$ and $\log \rho_{xx} \sim T^{-1/4}$ curves for samples with $34 \leq x \leq 42$ are presented in Fig. 3.12.

To clarify the extent of disorder, Fig. 3.13 gives the normalized $\rho_{xx}(T)/\rho_{xx}(300 \text{ K}) \sim T$ curves for all annealed samples. The resistivity ratio $\rho_{xx}(T)/\rho_{xx}(300 \text{ K})$ increases as $x$
Fig. 3.12 $\rho_{xx} \sim \ln T$ curves for samples with $47 \leq x \leq 81$ and $\log \rho_{xx} \sim T^{-1/4}$ curves for samples with $34 \leq x \leq 42$. The red solid lines are guides to the eyes.

decreases, which shows more disorder in the samples with lower $x$. Comparing to that of as-prepared samples in Fig. 3.5, the resistivity ratio decreases after annealing for each sample, which indicates reduced disorder after annealing. The absolute value of the resistivity decreases after annealing for each sample, which also implies that the extent of disorder decreases after annealing. From the analysis of longitudinal resistivity, we can conclude that the scattering because of disorder has been changed for all samples after...
annealing. Less disorder would reduce the scattering strength or possibility, which may affect the AHE.

3.2.2 Field-dependent Hall resistivity and magnetization

The field-dependent Hall resistivity and magnetization of all annealed samples were measured with magnetic field in the range of $-50 \text{kOe} \leq H \leq 50 \text{kOe}$ at temperatures ranging from 5 K to 300 K. The magnetic field was perpendicular to the sample plane for all measurements. Figure 3.14 gives the field-dependent Hall resistivity and magnetization curves for selected samples measured at 5 K. As shown in Fig. 3.14(a), the $\rho_{xy}$ increases sharply at low fields and follows almost linear dependency up to the magnetic saturation field. Beyond this magnetic saturation field, the curves show much weaker field-dependence, which could be ascribed to contributions of the ordinary Hall effect and the AHE due to a slightly change in magnetization. The total magnetization still increases
above the saturation field due to the field forced rotation of spins against the thermal agitation (except 0 K) and non-ferromagnetic couplings. For cobalt which has high Curie temperature, the linear dependence of Hall resistivity at high fields is caused mainly by the ordinary Hall effect. In this case, the $\rho_{AHE}$ can be obtained by extrapolating the linear part to zero field. Figure 3.14(b) gives the field-dependent magnetization curves for corresponding samples. The linear dependency at high fields mainly comes from the diamagnetism of sample substrates and sample holders in MPMS. The magnetization of samples decreases with $x$.

Comparing Fig. 3.14(a) and (b), the field-dependent Hall resistivity and magnetization curves have similar behaviors which is consistent with Eq. (1.1). Hysteresis loops have
been observed in both $\rho_{xy}\sim H$ and $M\sim H$ curves for samples with smaller $x$ and the coercivity increases with $x$ decreases. However, the coercivity in $\rho_{xy}\sim H$ is different from that in corresponding $M\sim H$ curves at 5 K, which has been observed in as-prepared samples. Similar phenomenon has been reported in Ti$_{1-x}$Co$_x$O$_{2-\delta}$ [93], SmFe alloy [94], Ga$_{1-x}$Mn$_x$As [95], Mn$_{3-x}$Ga [96], et al. For magnetization measurement, we used large size of samples (~4 mm × 4 mm) in MPMS and the measurement results should reflect the properties of the whole sample. Therefore, the coercivity from $M\sim H$ curves is quite convincing. While the coercivity in Hall resistivity could be affected by some factors. If the samples were patterned to Hall bar for Hall resistivity measurement and the Hall bar is too thin, the available paths for domain wall propagation would be suppressed and the domain walls were force to propagate through defects, which will induce larger coercivity [97]. Gate voltage, if applied during Hall resistivity measurement, could change anisotropy energy [98] or magnetic domain wall velocity [99], which also affects the coercivity. Nevertheless, above discussion can’t explain the coercivity difference in our samples because we didn’t apply gate voltage and the width of Hall bar is quite large (1.0 mm), which could exclude the suppression of domain wall propagation. The coercivity is always smaller in $\rho_{xy}\sim H$ curves than that in $M\sim H$ curves for our samples whether they are as-prepared or annealed samples. A reasonable explanation is as follows: $\rho_{xy}\sim H$ curves were measured when current was applied in the samples. Because our samples are inhomogeneous granular thin films, spins in carrier-rich regions where the resistivity is relatively lower contribute to both $\rho_{xy}$ and $M$, whereas spins in isolated regions didn’t contribute to $\rho_{xy}$ but do contribute to $M$ [95]. In other words, the current in Hall resistivity measurement tended to flow in low resistivity regions that has lower anisotropy which contribute more to $\rho_{xy}$ than high
resistivity regions. However, both regions equally contribute to $M$ in MPMS measurement. Joule heating in Hall resistivity measurement is another possible reason to reduce the coercivity if the applied current is too high [100]. In our measurement, the current we applied was quite low, so Joule heating effect, if it exists, is insufficient to influence the coercivity.

Figure 3.15 presents the field-dependent Hall resistivity and magnetization measured at different temperatures for the sample $x = 66$. At all temperatures, $\rho_{xy}$ follows linear field dependence up to the magnetic saturation field at which the strong magnetic field-dependent $\rho_{xy}$ transfers to a much weaker field dependence, as shown in Fig. 3.15(a). This

---

Fig. 3.15 Field-dependent (a) Hall resistivity and (b) magnetization at different temperatures for the annealed sample $x = 66$. 

---
behavior is commonly observed in magnetic thin films that have an easy plane, e.g. the magnetization is lying in film plane at zero applied magnetic field. When the magnetic field is applied perpendicular to the film plane, the magnetic saturation occurs at the demagnetizing field and shows no hysteresis loops. For this sample, the saturation field is around 1.3 Tesla. The field-dependent magnetization curves in Fig. 3.15(b) show similar behaviors with $\rho_{xy} \sim H$ curves. The magnetization at 5 K and 300 K are almost the same due to the high Curie temperature of cobalt.

3.2.3 Explore the mechanism of anomalous Hall effect

Figure 3.16 presents the longitudinal resistivity and anomalous Hall resistivity measured at 5 K as a function of $x$ for the as-prepared and annealed samples. For annealed samples, with $x$ decreases from 100 to 34, the $\rho_{xx}$ increases by almost four orders and the $\rho_{AHE}$ increases by three orders. 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 increase 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.

By plotting \( \rho_{xx} \sim (x - x_c) \) and \( \rho_{AHE} \sim (x - x_c) \) logarithmically for the data of the annealed samples, we got good linear dependence when \( x_c = 27 \) and \( 20 \) for \( \rho_{xx} \) and \( \rho_{AHE} \), respectively, as shown in Fig. 3.16(c) and (d). By linear fitting the two curves, the exponents in Eqs. (3.3) and (3.4) could be extracted as \( t = 3.38 \) and \( g = 3.42 \) which are quite close to the values obtained in the as-prepared samples. The obtained parameters of \( x_c \) and \( t (g) \) for the as-prepared and annealed samples are summarized in Table 3.2. For \( \rho_{xx} \), the \( x_c \) remains the same after annealing, which may indicate that the effective volume

![Graphs showing longitudinal resistivity and anomalous Hall resistivity measured at 5 K as a function of x for as-prepared and annealed samples.](image)
fraction of the metal did not increase although the conductivity of the metal clusters increased. The $x_c$ decreases from 26 to 20 in $\rho_{AHE}$ data after annealing, which suggests the possible structure changes mentioned above.

Annealing process modified the scattering strength or possibilities in the samples. We will see how the annealing process affects the scaling law. Shown in Fig. 3.14(b), the magnetization decreases with $x$. As discussed in section 3.1.4, we still used $R_s \sim \rho_{xx}^\gamma$ instead of $\rho_{AHE} \sim \rho_{xx}^\gamma$ to analyze the mechanism of AHE in the annealed samples. The $R_s \sim \rho_{xx}^\gamma$ for the annealed samples measured at 5 K was plotted logarithmically in Fig. 3.17(a). Remarkably, the correlation of $R_s$ and $\rho_{xx}$ 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 $\rho_{xx}$ and $\rho_{AHE}$ decreased significantly after annealing. This $R_s \sim \rho_{xx}$ relation was again confirmed in the annealed samples, which indicates the skew scattering dominated mechanism of AHE. Evidently, the intrinsic/side-jump mechanisms are excluded based on the above analysis.

We are wondering if the relation $R_s \sim \rho_{xx}$ is universal in this Co$_x$(MgO)$_{100-x}$ granular system. We then plotted all 5 K data of $R_s$ versus $\rho_{xx}$ obtained from both as-prepared and annealed samples in Fig. 3.17(b). As we expected, all data fall into the same curve with a slope of $0.96\pm0.02$. It is evident that skew scattering dominates the AHE in the

<table>
<thead>
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<th>$x_c$ ($\rho_{xx}$)</th>
<th>$t$ ($\rho_{xx}$)</th>
<th>$x_c$ ($\rho_{AHE}$)</th>
<th>$g$ ($\rho_{AHE}$)</th>
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<tbody>
<tr>
<td>As-prepared samples</td>
<td>27</td>
<td>3.74</td>
<td>26</td>
<td>3.05</td>
</tr>
<tr>
<td>Annealed samples</td>
<td>27</td>
<td>3.38</td>
<td>20</td>
<td>3.42</td>
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Fig. 3.17 (a) The anomalous Hall coefficient ($R_s$) versus longitudinal resistivity ($\rho_{xx}$) 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 red lines are the least-squares fits to the data.

Co$_x$(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. Thus, a firm
conclusion has been reached that $R_s \sim \rho_{xx}$ is a universal relation in Co$_x$(MgO)$_{100-x}$ granular samples and skew scattering dominated AHE in this system.

Experimentally, the linear relation of $R_s \propto \rho_{xx}$ has usually been reported in high-conductivity metals, such as Ni with diluted impurities [41] and Fe with light doping of Co, Cr, Mn and Si [38,39]. It has been theoretically reported that skew scattering dominates the AHE in the high-conductivity regime ($\sigma_{xx} > 10^6 \, \Omega^{-1} \text{cm}^{-1}$) [31]. In our Co$_x$(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 [50, 101] because the intrinsic mechanism originates from the band structure of materials, which is related to the long-range order of the crystallinity. Since our Co$_x$(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 $\rho_{xx}$ has been observed in granular thin films across a wide range in longitudinal resistivity.

### 3.2.4 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 Co$_3$O$_4$, 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 $M \sim H$ 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 $M \sim H$ 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 $M \sim H$ 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 3.18 shows the ZFC and FC $M \sim H$ 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 $M \sim H$ curves are irreversible under a high applied field (up to 70 kOe), which could be due to the existence of the spin glass phase [102,103]. 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 Fig. 3.18, $H_E$ is almost zero in the ZFC $M \sim H$ curves whereas there is an exchange bias in the FC $M \sim H$ curves for both the as-prepared and annealed samples. The exchange bias shown in the FC $M \sim H$ 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, 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.

### 3.2.5 Conclusion

The annealing process modified the scattering potential/possibility and reduced the longitudinal resistivity and the anomalous Hall resistivity. The linear relation $R_s \sim \rho_{xx}$ is valid in the annealed samples. When combining all the data of as-prepared and annealed
samples, the relation $R_s \sim \rho_{xx}$ remain valid, which gives the universal scaling law in
Co$_x$(MgO)$_{100-x}$ system. This is the key result of this chapter which suggests a skew
scattering dominated mechanism in Co$_x$(MgO)$_{100-x}$ granular thin films, which requires
further theoretical investigation.
Chapter 4 Anomalous Hall effect in Fe(Ni)/Au, Ta/Fe multilayers

In the last chapter, we prepared Co$_x$(MgO)$_{100-x}$ granular thin films and study the interfacial scattering effect on the AHE. In the granular systems, the interfacial scattering depends not only on the composition of metal and insulator, but also on the grain sizes of metal and insulator. Therefore, it is less controllable to study the interfacial scattering effect comparing to multilayer systems. In this chapter, we prepared Fe/Au, Ni/Au and Ta/Fe multilayers and study the interfacial scattering effect on the AHE.

4.1 Anomalous Hall effect in Fe/Au multilayer

In the present work, we investigated the interfacial scattering effect on the AHE in $(\text{Fe}_{36 / \pi} \text{Au}_{12 / \pi})_n$ multilayers. We designed this structure based on the following two considerations: first, the total thicknesses of the Fe and Au layers were kept unchanged in all samples and the current distribution in the Fe and Au layers was also the same during the transport measurements, including the Hall resistivity, longitudinal resistivity and magnetoresistance measurements. The possibility that various current distributions lead to different Hall resistivities can easily be excluded. Second, the total thickness of all the samples was the same, such that the scattering contributions from both surfaces would be the same. The only difference in the samples was the number of Fe/Au interfaces which is beneficial to the study of the impact of interfacial scattering on the AHE.

4.1.1 Structural characterizations

To characterize the multilayered structure of the samples, we ran the XRR measurements on all samples. Shown in Fig. 4.1(a) are the XRR patterns of all samples. Characteristic oscillations in the XRR spectra are evident, indicating relatively sharp and
flat interfaces. More peaks arising from the superlattice structure are observed in samples with more Fe/Au periods (larger $n$). To obtain quantitative information about both the periods and individual layer thicknesses of the samples, the XRR spectra were fitted with the LEPTOS software suite (V7.02, Bruker). The fitted thicknesses of each layer were very

![Image of XRR spectra and high-angle XRD patterns](image)

**Fig. 4.1** (a) XRR spectra and (b) high-angle XRD patterns of multilayered samples from $n = 1$ to $n = 12$. The peaks of FeAu alloys are labeled as (V)AuFe(111) and (◊)AuFe(111) in (b).
close to the preset thicknesses for sample preparation, confirming our ability to control the sample fabrication. Figure 4.1(b) shows the high-angle, $\theta$–$2\theta$ XRD patterns for all samples. The samples with few periods or small $n$ exhibited Fe(110) and Au(111) peaks, which are the strongest peaks for the polycrystalline Fe and Au, indicating good crystallinity of the Fe and Au layers. As $n$ increased, both peaks became broader and weaker, suggesting that the crystallinity of the samples was decaying. When $n = 8$ to 12, two weak peaks, appearing between the Fe(110) and Au(111) peaks, could be due to the FeAu alloy phase formed in the Fe/Au interfaces. Because these samples had a clearly layered structure, as suggested by the XRR spectra in Fig. 4.1(a), the alloy phases could exist only at the interfaces and the interface content in these samples would be quite small.

To further characterize the structure of the multilayer samples and the interfaces between Fe and Au layers, we analyzed the cross-section of selected samples by HRTEM. Figure 4.2 shows HRTEM images of the cross-sections of the multilayers and the corresponding elemental profiles. The HRTEM images clearly exhibit layered structures with relatively sharp interfaces. The rippled structures of the cross-sections of multilayers with $n = 6$ and 12 could be due to the release of strain, when the ~50-nm-thick specimens were extracted from the macroscopic samples using focused ion beam (FIB). The strain was generated at the Fe/Au interfaces during growth due to the different lattice constants and thermal expansion coefficients of Fe and Au [104,105]. This strain release is not evident in samples with small $n$, indicated in $n = 2$ sample. The rippling of the multilayers caused by strain release does not occur in the macroscopic samples; if it did, we would not observe the low-angle oscillations in the XRR spectra as shown in Fig. 4.1(a). The measured thickness of each layer and the total thickness of the samples in the HRTEM
images are consistent with the preset values, suggesting that the layered structure and thickness of each layer are well controlled during deposition. Elemental profiles were obtained by line-scanning of energy-dispersive x-ray spectroscopy (EDX) using a scanning transmission electron microscopy (STEM) as shown in Fig. 4.2(d), (e) and (f). As expected, these elemental profiles featuring the desired periodicity demonstrate the high quality of the multilayer samples.

Fig. 4.2 HRTEM images of cross-sections of samples (a) $n = 2$, (b) $n = 6$, (c) $n = 12$ and EDX depth profiles of Fe and Au elements in corresponding samples (d) $n = 2$, (e) $n = 6$, (f) $n = 12$. 
4.1.2 Longitudinal resistivity

To understand electrical transport in the multilayered materials, we measured the temperature-dependent longitudinal resistivity at zero magnetic field from 10 K to 310 K. Figure 4.3(a) shows the temperature-dependent $\rho_{xx}$ curves for all samples. The temperature coefficient of resistivity (TCR, $d\rho/dT$) was positive for all samples across the full temperature range, indicating the metallic nature of the samples. As $n$ increases from 1 to 12, the $\rho_{xx}$ curves shift monotonically to high values, although the total thicknesses of the Au and Fe layers remain the same in all samples. The resistivity flattens off below 50 K in all samples, indicating that the thermal contribution to the total resistivity is less important than the residual resistivity caused by electron scattering by structural defects in both individual layers and the interfaces, particularly at temperatures below 10 K. To better understand the effect of electron scattering by the interfaces on the measured resistivity, we replotted the data of Fig. 4.3(a) in different forms, as shown in Fig. 4.3(b)–(d). One interesting feature in Fig. 4.3(b) is that the curves of $\rho_{xx}(n)/\rho_{xx}(n = 1)$ as a function of temperature might be classified into two groups. The curves with $n \leq 4$ are flat across the full temperature range, while the curves for $n \geq 5$ are dependent on temperature, which becomes stronger as $n$ increases. This temperature dependence might suggest that dominant mechanisms of the electron scatterings are different for the two groups of samples. In the $n \leq 4$ samples, the contribution from scattering by two-dimensional (2D) interfaces is much weaker than that in the $n \geq 5$ samples, although zero-dimensional (0D, relative to the 2D interfaces), nanometer-sized defects in the individual layers of Fe and Au exist in all samples. Because the thickness of the individual layers of Fe and Au decreases as $n$ increases, the crystallinity of the samples decays and, consequently, the density of the 0D
defects, such as grain boundaries, increases. The decaying quality of the crystallinity of the samples with the decreasing thickness of individual layers is reflected in the high-angle XRD spectra shown in Fig. 4.1(b). The combined electron scattering from the 2D interfaces and 0D defects leads to an increase in the contribution of residual resistivity to the total resistivity as reflected in Fig. 4.3(c) in which the $\rho_{xx}(10 \text{ K})/\rho_{xx}(310 \text{ K})$ ratio increases with $n$. These plots also give information about the relative contribution of both phonons and structural imperfections to the electron scattering. Generally, as the disorder increases, the resistivity decreases more slowly as the temperature decreases [79]. It is therefore

Fig. 4.3 (a) Temperature dependent longitudinal resistivity curves for all samples; (b) $\rho_{xx}(n)/\rho_{xx}(n = 1)$ as a function of temperature for all the samples; (c) normalized $\rho_{xx}(T)/\rho_{xx}(310 \text{ K})$~$T$ curves for all the samples. (d) $\rho_{xx}(n)/\rho_{xx}(n = 1)$ as a function of number of period $n$ for selected temperatures.
evident that the samples with larger $n$ have greater scattering due to the interfaces and poor crystallinity. Due to size effect, the interfacial scattering plays more important role in resistivity than grain boundary scattering does for samples with $n \geq 5$.

Similar conclusions can be drawn from the plots in Fig. 4.3(d). The $\rho_{xx}(n)/\rho_{xx}(n = 1)$ ratio for low-temperature data ($T=10$ K and $50$ K) in which the phonon scattering can be neglected depend nearly linearly on $n$, which reveals the impact of interfacial scattering. The $\rho_{xx}(n)/\rho_{xx}(n = 1)$ ratio for high-temperature data in which the contribution from phonons becomes significant is markedly lower.

4.1.3 Field-dependent Hall resistivity

Our observations of the relationship between resistivity behaviors and temperature suggest that the Fe/Au interface plays a crucial role in electrical transport. We now turn our attention to the AHE to examine how the scattering processes affect anomalous Hall resistivity. The Hall resistivity ($\rho_{xy}$) for all samples was measured with a magnetic field applied perpendicularly to the plane of the films in a range of $-50$ kOe $\leq H \leq 50$ kOe and at temperatures ranging from 10 to 300 K. Figure 4.4(a) shows the field-dependent Hall resistivity of all samples at 10 K. As expected, $\rho_{xy}$ increases sharply with low applied magnetic field and follows a linear dependence up to magnetic saturation field at which the strong magnetic-field-dependent Hall resistivity suddenly changes to a much weaker field dependence. This behavior is commonly observed in magnetic thin films that have an easy plane, e.g., the magnetization is lying in film’s plane at zero applied magnetic field. When the magnetic field is applied perpendicularly to the film’s plane, magnetic saturation occurs at the demagnetizing field and no hysteresis loops are observed. In Fe thin films, magnetic saturation field occurs at about 20 kOe and remains almost constant from 10 K to 300 K,
due to the very high Curie temperature of pure Fe. Therefore, the behaviors of field-dependent Hall effect and field-dependent magnetization should be very similar, because anomalous Hall resistivity is proportional to the magnetization component in the field direction as described by Eq. (1.1). At magnetic fields higher than saturation field, the weakly, field-dependent Hall resistivity should be ascribed to the contributions from the

**Fig. 4.4** Field-dependent Hall resistivity of (a) all samples at 10 K and (b) the samples $n = 5$ at temperature ranging from 10 K to 300 K, (c) Temperature-dependent anomalous Hall resistivity for all samples.
normal Hall effect and from the slight change in the AHE caused by the improved alignment of the spins. The total magnetization continues to increase in this regime due to the field-forced rotation of the spins against the thermal agitation (except at 0 K) and non-ferromagnetic couplings. In Fe, Co and other ferromagnetic materials that have high Curie temperatures, the slope of the measured Hall resistivity in this region mainly originates from the ordinary Hall effect. In this case, $\rho_{\text{AHE}}$ could be obtained by extrapolating the linear part to zero field.

The sign of $\rho_{\text{AHE}}$ for all samples is positive across the full temperature range of the measurements, which is consistent with sign of $\rho_{\text{AHE}}$ observed in pure Fe films [50] and Fe/Si multilayers [106]. The sign of $\rho_{\text{OHE}}$ is negative, suggesting that electrons dominate the transport properties in all samples. Figure 4.4(b) shows the field-dependent $\rho_{xy}$ of the sample with $n=5$ measured at different temperatures as an example to illustrate the effect of temperature on AHE. As the longitudinal resistivity increases with temperature, $\rho_{xy}$ increases. Figure 4.4(c) presents the temperature-dependent $\rho_{\text{AHE}}$, which presents the same tendency as $\rho_{xx}(T)$. It clearly shows that $\rho_{\text{AHE}}$ depends very weakly on temperature below 50 K, indicating a quite weak contribution of phonons to AHE. Remarkably, at 10 K, $\rho_{\text{AHE}}$ increased by 40 times when $n$ increased from 1 to 12, although the longitudinal resistivity increased only by 4 times.

4.1.4 Anomalous Hall effect scaling analysis

To understand the origin/mechanisms of AHE experimentally, Eq. (3.3) is usually employed and the exponent, $\gamma$, is then extracted by fitting the experimental data to the power law. As discussed above, $\gamma = 1$ indicates that skew-scattering is the dominant mechanism, whereas $\gamma = 2$ may suggest that an intrinsic mechanism, an extrinsic side-
jump mechanism, or both dominate the AHE. The curve of $\rho_{\text{AHE}} \sim \rho_{xx}$ was plotted logarithmically in Fig. 4.5 for all samples measured at 10 K at which electron-photon scattering could be neglected. Apparently, not all data can be fitted to a single straight line. A closer analysis reveals that the data in Fig. 4.5 can be fitted to two linear functions as indicated by the blue and red lines. Fitting the data obtained from the $n = 1$ to $n = 4$ samples gave $\gamma = 2.65 \pm 0.10$, whereas $n = 4$ to $n = 12$ samples gave $\gamma = 1.90 \pm 0.04$. Normally, $\gamma = 1.90$ (~2) could be regarded as an intrinsic mechanism, a side-jump mechanism, or both, but it is difficult to distinguish between the two mechanisms conclusively based only on this scaling. When $\gamma$ is larger than 2, attributing this relation to any of the three mechanisms is difficult. A large exponent has previously been observed in other metallic samples, such as Fe/Cr multilayers [63] and Co–Ag granular thin films [58,107].

![Fig. 4.5](image)

**Fig. 4.5** The logarithmic plot of anomalous Hall resistivity ($\rho_{\text{AHE}}$) versus longitudinal resistivity ($\rho_{xx}$) measured at 10 K for all samples. The red line and blue line are linear fits to the data.
It could be possible that the different exponential components are due to the shunting effect of Au layers in the samples. After re-examining the structures of these samples, this possibility has been excluded in these multilayer structures based on the model shown in Fig. 6 of ref. 78. Because anomalous Hall voltage can be generated only from the Fe layer, the Hall voltage of the multilayers is therefore proportional to that of single Fe layers according to Kirchhoff’s law. This proportion remains the same in all samples because the ratio of the longitudinal resistivity \( \rho_{xxFe}/\rho_{xxAu} \) and the thickness \( t_{Fe}/t_{Au} \) between Fe and Au remains constant in all samples. The scaling law, Eq. (3.5), is therefore not affected by the Au layers.

Whereas the compositions of Fe and Au are exactly the same here, the \( \rho_{xx} \) and \( \rho_{AHE} \) are completely different in all samples. The scaling law, Eq. (3.5), gives different \( \gamma \) values for samples from \( n = 1 \) to \( n = 4 \) and for samples from \( n = 4 \) to \( n = 12 \). This means that the AHE is directly related to the density and strength of the scattering from the interfaces and grain boundaries. In inhomogeneous systems, like granular thin films, both the composition and the particle size of the non-magnetic material doped in the samples affect the AHE, which explains why various scaling relations have been reported for very similar granular thin films, e.g. Fe–SiO2 films, by different groups [62,108,109]. The samples used in this study were free from the complexity caused by the differences in composition.

In our previous study [110], giant magnetoresistance (GMR) and anisotropic magnetoresistance (AMR) effect were used to explore the surface/interface scattering in Co/Pd\(_{1-x}\)Ag\(_x\) multilayers. To understand the scattering on transport properties, MR curves were measured with magnetic field in the sample plane parallel and perpendicular to the current. The GMR effect is based on the two-current-model [111] that comes from the spin-
dependent scattering both within the ferromagnetic layer and at the interface between ferromagnetic and nonferromagnetic layers. [112,113] It depends on the relative direction of spins and is not related to the angle between magnetization and current. The AMR effect is caused by anisotropic scattering probability of carriers due to spin-orbit coupling. Generally, the longitudinal resistivity depends on the relative orientations of the magnetization and the current in polycrystalline ferromagnetic metals, which could be expressed as [114]

\[
\rho(\varphi) = \rho_\perp + (\rho_\parallel - \rho_\perp) \cos^2 \varphi
\]  

(4.1)

where \(\rho_\perp\) and \(\rho_\parallel\) are the resistivity when \(\varphi = 0^\circ\) and \(90^\circ\) for a magnetically single domain sample, \(\varphi\) is the angle between current and magnetization. Fig. 4.6 shows the MR curves of all samples measured at 10 K. The magnetic field parallel (perpendicular) to the current is referred to \(H_\parallel(H_\perp)\) in the figures. The corresponding magnetoresistance is defined as \(MR_\parallel(MR_\perp)\). MR is defined as \(MR = [R(H) - R(H_S)]/R(H_S)\), where \(H_S\) is the saturation field at which all the magnetic moments are aligned to the field direction. Several interesting features are observed in the field-dependent MR curves: 1) \(MR_\perp\) is always positive for all the samples, being in agreement with the definition of AMR; 2) the sign of \(MR_\parallel\) changes with \(n\). For \(n = 2, 3\) and \(4\), \(MR_\parallel > 0\), for other samples, \(MR_\parallel < 0\); 3) peaks of both \(MR_\parallel\) and \(MR_\perp\) for each sample are at the same field, suggesting a close correlation between \(MR_\parallel\) and \(MR_\perp\), although the peak field changes with \(n\); 4) the behaviors of MR curves at both \(H_\parallel\) and \(H_\perp\) vary with \(n\) strongly.

Apparently, when \(H_\parallel\) and \(H_\perp\) are at 100 Oe or larger, the resistivity is saturated for both directions and their values correspond \(\rho_\parallel\) and \(\rho_\perp\) in Eq. (4.1). AMR always gives \(\rho_\parallel > \rho_\perp\) and the maximum AMR can then be defined as, \(AMR_{\text{max}} = (\rho_\parallel - \rho_\perp)/\rho_\perp\). In the
application of AMR, the sample must be biased using a magnetic field to keep the sample magnetically saturated in order to reach maximum sensitivity.

For a multi-domain film which is unbiased as in our experiments, the field-dependent MR at $H_{\parallel}$ and $H_{\perp}$ are quite complicated and multi-valued, depending on the magnetic history. The resistance of a multi-domain ferromagnetic film is the sum of the resistance of each magnetic domain, which can be calculated using Kirchhoff’s circuit laws in principle. The resistance of each domain is determined by Eq. (4.1). Therefore, the behavior of $MR_{\parallel}$ and $MR_{\perp}$ should be very different when the magnetic field is lower than $H_{S}$. If only AMR effect exists in the samples, in principle, should we always observe

**Fig. 4.6** MR curves of the samples from $n = 1$ to $n = 12$ measured at 10 K. Magnetic field parallel to the sample plane during the measurement.
$MR_{||} < 0$ and $MR_{\perp} > 0$. Generally, the saturation and coercivity fields are the same at $H_{||}$ and $H_{\perp}$ for each sample without large geometric aspect-ratio. Below the saturation magnetic field, the sample is at the multi-domain state, the resistance should be between $R_{||}$ and $R_{\perp}$ ($R_{||} > R_{xx} > R_{\perp}$), where $R_{||}$ ($R_{\perp}$) is the resistance when magnetic field is parallel (perpendicular) to the current. However, the resistance of the sample depends on the domain configuration or the magnetic history.

The small saturation magnetic field indicates that our samples are soft ferromagnet and the magnetizing process is most likely through the domain wall motion. This has been confirmed by the field dependent magnetization data.

Now let’s turn to the experimental results of MR shown in Fig. 4.6. For samples with $n = 1$ and $n > 4$, the behavior of the MR curves can be mainly ascribed to AMR effect. When the sample was saturated magnetically at $H_{||}$ and $H_{\perp}$, the samples become single domain and the measured resistance should be $R_{||}$ and $R_{\perp}$. When the magnetic field reduces from $H_{S}$, the single domain state will gradually transfer to a multi-domain state through the nucleation and then domain wall motion. At the coercivity field, the total magnetization along the field direction become zero. However, infinite number of domain configurations can give us zero magnetization which could induce totally different resistance. Figure 4.7 shows three extremely simple domain configurations at coercivity field that could give zero magnetization but very different resistance for both $R_{||}$ and $R_{\perp}$.

In Fig. 4.7(a) and (b), $R_{||}$ and $R_{\perp}$ are almost the same for each configuration because only one domain wall is inserted in the original single domain sample. For configuration in Fig. 4.7(c), the resistance should be different with that in Fig. 4.7(a) and (b), because the current pass through magnetic domains with different resistivity ($\rho_{||}$ and $\rho_{\perp}$). For Fig.
In Fig. 4.7(c) and (d), both $MR_\parallel$ and $MR_\perp$ reach the maximum at coercivity field with $MR_\parallel < 0$ and $MR_\perp > 0$. If the domain wall scattering is also considered, different domain configurations will certainly lead to different values of $R_\parallel$ and $R_\perp$. Therefore, the measured $MR_\parallel$ and $MR_\perp$ should not be unique and uncontrollable, although they should have peaks at coercivity field. Based on above discussion, we can easily understand the behaviors of $MR_\parallel$ and $MR_\perp$ shown in Fig. 4.6 for $n=1, 5, 6, 8, 10,$ and $12$: 1) $MR_\perp$ is always positive and $MR_\parallel$ is always negative; 2) both $MR_\parallel$ and $MR_\perp$ have peaks at the same fields but the resistance at the peaks are not unique. This indicated that the coupling between the
magnetic moments of adjacent Fe layers are ferromagnetic, which leads to that the films behave as a ferromagnet with some non-ferromagnetic inclusions. The Au layers could be considered as defects in the films.

Now let’s turn to the MR data for samples with \( n = 2, 3 \) and \( 4 \) in Fig. 4.6. Both \( M_{MR\parallel} \) and \( M_{MR\perp} \) are positive which is sharply different from that of other samples with AMR effect. Particularly for \( n = 2 \) and \( 3 \), the amplitude of \( M_{MR\parallel(\perp)} \) is significantly larger than the \( AMR_{\text{max}} \). Most importantly, the sign of MR doesn’t depend on the relative angle between the current and magnetization. Therefore, the \( M_{MR\parallel(\perp)} \) is not dominated by the AMR effect. This positive MR with two peaks might be of the similar origin as for GMR observed in the multilayers in which there exist antiferromagnetic couplings between the adjacent ferromagnetic layers separated by a non-magnetic layer \([115,116]\), i.e. the magnetic moments of the adjacent ferromagnetic layers point to opposite directions at zero field. Normally, GMR is much larger than AMR as reported in multilayered systems. For sample \( n = 1 \), only one ferromagnetic layer in the sample and no GMR is expected. For samples with \( n = 2 \) and \( n = 3 \), GMR in both directions is evidently larger than the AMR (For \( n = 2 \), GMR=0.92% and AMR=0.21%; for \( n = 3 \), GMR=0.86% and AMR=0.31%). For sample with \( n = 4 \), GMR is comparable to the AMR (GMR=0.39% and AMR=0.41%). The peak of \( M_{MR\parallel} \) and \( M_{MR\perp} \) for each sample are at the same field which is related to the coercivity field. The relatively small GMR could be due to the very weak antiferromagnetic coupling between the adjacent Fe layers because of the relatively thick Au layers (6 nm, 4 nm and 3 nm for samples with \( n = 2, 3, \) and 4). Another possibility is that the small GMR comes from the spin-dependent scattering between the adjacent magnetic domains with different magnetization direction within one Fe layer.
As reported in Fe/Cr multilayers [63] and Co-Ag granular films [58,59] which are classical GMR systems, spin-dependent scattering which is responsible for GMR phenomenon plays an important role to the large exponential $\gamma (>2)$ in AHE scaling law. This could explain the linear scaling relation with large exponential ($\gamma = 2.65 \pm 0.10$) from sample $n = 1$ to $n = 4$ in Fig. 4.5. Sample $n = 1$ is an exception that it didn’t show GMR effect because this sample only contains one layer of Fe and the interfacial spin-related scattering induced GMR would vanish. The pretty thick layer of Fe (~36 nm thickness) would show the bulk property which may lead to prominent AMR effect. For the samples from $n = 5$ to $n = 12$, AMR dominated the electrical transport and it is related to the spin-orbital coupling which is also the origin of AHE (whatever intrinsic or extrinsic AHE). The exponential $\gamma = 1.90 \pm 0.04$ could be explained by the intrinsic/side-jump mechanisms.

4.1.5 Parsing of intrinsic, skew-scattering and side-jump contributions

To distinguish the contributions of the intrinsic mechanism and the side-jump mechanism in the measured AHE, we analyzed our data using the recently proposed scaling [50] as shown in Eqs. (1.5) and (1.6). To avoid spin-dependent scattering dominating the transport properties as in GMR materials, we chose the samples with $n = 4$ to 12 for this analysis. Since the thermal effect on transport properties can be neglected at low temperature (10 K), Eq. (1.6) can then be simplified to $\sigma_{AHE0} = \alpha \sigma_{xx0} + (\beta + b)$ by taking the following approximations $\sigma_{AHE} \approx \sigma_{AHE0}$ and $\sigma_{xx} \approx \sigma_{xx0}$. We then plotted $\sigma_{AHE0}$ as a function of $\sigma_{xx0}$ for data obtained at 10 K in Fig. 4.8(a). By fitting the data linearly, we extracted the slope $\alpha = (7.69 \pm 4.84) \times 10^{-4}$ and the intercept $\beta + b = (4.02 \pm 0.15) \times 10^2 \ \Omega^{-1}\text{cm}^{-1}$. The relatively small value of $\alpha$ may suggest that the skew-
scattering, if it exists, contributes very little to the measured AHE. Following the procedure used in Ref. 50, we then plotted the $\sigma_{\text{AHE}}$ curves as a function of $\sigma_{xx}^2$ for data obtained across the full temperature range (10 to 300 K) for $n \geq 4$ samples as shown in Fig. 4.8(b). By fitting the data to a linear dependence, we obtained the value of $b$ (the intercepts) for

Fig. 4.8 (a) $\sigma_{\text{AHE}0} \sim \sigma_{xx0}$ curve measured at 10 K and (b) $\sigma_{\text{AHE}} \sim \sigma_{xx}^2$ curves for samples $n = 4$ to 12. Inset gives the intercept values ($b$ values) for different $n$. (c) the $b \sim \rho_{xx0}$ and $\beta \sim \rho_{xx0}$ curves. The solid lines are the linear fittings to each curve.
each sample. Apparently, these curves do not intercept at the same point, differing from the results reported in Ref. 50. The inset of Fig. 4.8(b) gives the values of $b$ for different samples. Interestingly, the value of $b$ decreases linearly with increasing $n$. Using the value of $\beta + b$ from Fig. 4.8(a), the value of $\beta$ was easily obtained for each sample. Obviously, $\beta$ is almost zero for $n = 4$, suggesting that the intrinsic mechanism dominates AHE in this sample. Since $\beta + b$ is constant, as $n$ increases from 4 to 12, the value of $\beta$ increases and, consequently the value of $b$ decreases (as shown in the inset of Fig. 4.8(b)), which is indicative of the relative contributions from the intrinsic mechanism and the side-jump mechanism to AHE, depending on $n$. To understand how both the intrinsic mechanism and side-jump mechanism vary with the scattering properties (density and strength), we plotted both $b$ and $\beta$ in relation to residual longitudinal resistivity in Fig. 4.8(c). It is interesting that both $b$ and $\beta$ show a perfectly linear dependence on $\rho_{xx0}$, although they have opposite trends. This is in sharp contrast to the results reported in Ref. 50 where both $b$ and $\beta$ remain almost unchanged in single crystalline Fe films with different thicknesses. In their work, the Fe films have high quality and the mean free path of electrons could be quite large. The surface scattering plays very important role in electrical transport which largely tuned the longitudinal resistivity and Hall resistivity by changing the thickness of Fe films. In our work, interfacial scattering at Fe/Au interfaces plays a more important role than surface scattering does as indicated by resistivity data in Fig. 4.3. Original theories suggested that intrinsic mechanism happens in the perfect crystal [17] and side-jump contribution comes from impurity scattering [29]. We found that the relative contributions of the intrinsic mechanism and the side-jump mechanism depend on $\rho_{xx0}$ which mainly comes from interfacial scattering, suggesting that interfacial scattering suppresses the intrinsic
contribution and gives rise to side-jump contribution. Therefore, interfacial scattering plays a different role than surface scattering. Another interesting feature is that although the ratio of the intrinsic mechanism to the side-jump mechanism, $b/\beta$, decreases with increasing $\rho_{xx0}$ (or scattering), the contribution of the intrinsic mechanism dominates the entire longitudinal resistivity range and the full temperature range, which agrees with the results reported in Ref. 50.

4.1.6 Conclusion

In summary, we designed and fabricated a family of multilayered $(\text{Fe}_{36n}/\text{Au}_{12n})_n$ samples to study the AHE. We performed transport property measurements under different magnetic fields in a wide temperature range from 10 K to 310 K. We observed different scaling relations in different samples. Our quantitative analysis showed that the interfacial scattering suppresses the contribution of intrinsic mechanism and gives rise to side-jump contribution. In the race for large spin Hall and anomalous Hall effects, significant attention has been brought to the intrinsic [117] and skew-scattering [118,119] mechanisms. In comparison, side-jump has received only little attention lately. Our finding suggests that interfacial scattering can promote side-jump mechanism which is the source of AHE and opens interesting avenues for the design of this phenomenon in multilayered structures.

4.2 Anomalous Hall effect in Ni/Au multilayer

In this part, we studied the effect of interfacial scattering on the magneto-transport properties using the $(\text{Ni}_{36n}/\text{Au}_{12n})_n$ multilayers with different numbers of interfaces. This is the continue study of the AHE of $(\text{Fe}_{36n}/\text{Au}_{12n})_n$ multilayers. The aim of the continue study is to understand: (1) how does the quality of the interfaces affect the AHE? In the Fe/Au
multilayers, both the crystal structure and lattice constant of the two materials are different: Fe has body-center-cubic structure with lattice constant $a=0.2866$ nm and Au has face-center-cubic structure with lattice constant $a=0.4079$ nm. Because of the large lattice mismatch, microscopically rough interfaces and high stress are expected in the multilayers. Whereas in the Ni/Au multilayers, Ni and Au both have face-center-cubic structure with lattice constant $a=0.3524$ nm and $a=0.4079$ nm, respectively. Therefore, we expect the quality of the interfaces would be improved in Ni/Au multilayers. (2) How does the sign of spin polarization affect the AHE in multilayers? The sign of the AHE is related to that of spin polarization in bulk-like ferromagnetic materials. Fe has positive and Ni has negative spin polarization. As stated in Section 4.1 of Fe/Au multilayers, we designed the Ni/Au multilayers with the same total thickness and the same thickness ratio of Ni and Au. This specific structure design keeps the same shutting effect and surface scattering.

4.2.1 Structural characterization

We performed XRR measurements to analyze the multilayer structures of all samples. As shown in Fig. 4.9(a), the periodic oscillations were clearly seen, indicating relatively sharp and flat interfaces. More oscillation periods could be observed for the samples with more layers. Comparing to the XRR spectra of Fe/Au multilayers, Ni/Au multilayers have clearer oscillation peaks especially at larger $\theta$, although they have exactly the same structure. This indicates that the Ni/Au interfaces are flatter and sharper. To obtain the individual layer thickness of the multilayers, we fitted the XRR spectra by LEPTOS software (V 7.02, Bruker). The fitted layer thickness is quite close to the preset values, which confirmed the stability of thickness control during the deposition. Fig. 4.9(b) presents the high angle $\theta$–$2\theta$ XRD patterns for all samples. For samples with fewer periods
The peaks of Ni(111) and Au(111) could be observed, which show good crystallinity in Ni and Au layers. As the period increases, the two peaks become broad and the intensity decreases, which indicates crystallinity decaying in samples with \( n \) increases. The same trend was also observed in Fe/Au multilayers. Moreover, for samples \( n = 5 - 12 \), several satellite peaks were shown at \( 2\theta \) from 34° to 45°, which is consistent with a previous report.

**Fig. 4.9** (a) XRR spectra and (b) high-angle XRD patterns of multilayered samples from \( n = 1 \) to \( n = 12 \).
The peak position deviation of Ni and Au could be due to the interfacial strain/stress caused by different lattice constants and thermal expansion coefficients of Ni and Au. In addition, the intermixing of Ni and Au at the interfaces may give some alloy phases. This composition modulation (alloys with different composition) may also cause peak position deviation in XRD patterns.

Fig. 4.10 HRTEM images of cross-sections of samples (a) $n = 2$, (b) $n = 6$, (c) $n = 12$ and EDX depth profiles of Fe and Au elements in corresponding samples (d) $n = 2$, (e) $n = 6$, (f) $n = 12$. 
To further demonstrate the multilayer structure and the interfaces between Ni and Au layers, we characterized the cross-section of selected samples by HRTEM. Figure 4.10 shows the HRTEM images of the multilayer cross-sections and the corresponding elemental profiles for samples $n = 2, 6$ and $12$. The images clearly exhibit layered structures of the samples without any rippling, which indicate the stress due to the lattice mismatch and different thermal expansion coefficients is much weaker than that in the Fe/Au multilayers. The relatively sharp and flat interfaces may give less contribution to the resistivity of the samples compared to that in the Fe/Au multilayers, which will also affect the anomalous Hall resistivity. The measured thickness of each layer and the total thickness of the samples from the HRTEM images are consistent with the preset values, which indicate again that the layered structures and thickness can be well controlled during deposition. The element profiles were obtained by the line-scanning of energy dispersive x-ray spectroscopy (EDX) using scanning transmission electron microscopy (STEM), as shown in Fig. 4.10(d), (e) and (f). As expected, these elemental profiles featured with desired periodicity, demonstrating again the multilayer structures in the samples.

4.2.2 Longitudinal resistivity

The temperature-dependent longitudinal resistivity was measured in the temperature range of $5 \text{ K} - 310 \text{ K}$ at zero magnetic field. Figure 4.11(a) presents the $\rho_{xx} \sim T$ curves for all samples. All samples show positive temperature coefficient of resistivity (TCR, $d\rho_{xx}/dT$), which indicates metallic conduction in the measurement temperature range. With $n$ increases from 1 to 12, the $\rho_{xx} \sim T$ curves shift to high resistivity values monotonically, although the total thickness of each sample was kept the same. This resistivity enhancement could be partially attributed to the scattering effect by the increased
interfaces. Below 50 K, the resistivity flatted off for each sample, which indicates the thermal contribution to the resistivity could be neglected, especially at temperatures below 10 K. To better demonstrate the scattering effect, normalized resistivity was plotted in Fig. 4.11(b). The resistivity ratio $\rho_{xx}(T)/\rho_{xx}(310 \text{ K})$ is a useful empirical parameter for

![Fig. 4.11](image-url)

**Fig. 4.11** Temperature-dependent longitudinal resistivity ($\rho_{xx}$) curves for all samples; (b) normalized $\rho_{xx}(T)/\rho_{xx}(310 \text{ K})$ curves for all samples; (c) $\rho_{xx}(n)/\rho_{xx}(n = 1)$ as a function of number of period $n$ for selected temperatures.
quantifying the extent of disorder [79]. As shown in Fig. 4.11(b), the $\rho_{xx}(T)/\rho_{xx}(310 \, \text{K})$ increases with $n$, which clearly demonstrates more scattering effect in samples with larger $n$. Figure 4.11(c) shows the $\rho_{xx}(n)/\rho_{xx}(n = 1)$ ratio at several temperatures. With $n$ increases from 1 to 12, the resistivity increases by 2.7 times at 5 K and by 2.2 times at 300 K. This resistivity enhancement is smaller than that in the Fe/Au multilayers, although the multilayer structure design is exactly the same. At low temperatures ($T = 10$ and 50 K) where the electron-phonon interaction could be neglected, the $\rho_{xx}(n)/\rho_{xx}(n = 1)$ ratio is almost linearly dependent on $n$, which clearly shows interfacial scattering effect. The resistivity dependence on $n$ is relatively weaker, mainly being due to the sharper and flatter interfaces, than that in the Fe/Au multilayers, which demonstrates weaker interfacial scattering effect on resistivity in the Ni/Au multilayers. Actually, this resistivity variation with $n$ may originate from two mechanisms, (a) interfacial scattering and (b) the variation of crystallinity within layers of the samples with different $n$. The crystallinity of the samples decreases with decreasing the thickness of individual layers, which was confirmed by XRD $\theta$–$2\theta$ patterns, as shown in Fig. 4.9(b). However, due to size effect, the interfacial scattering plays a much more important role in resistivity than does grain boundary scattering, especially in samples with large $n$.

4.2.3 Anomalous Hall effect scaling analysis

Based on the temperature-dependent resistivity data, we understand that the interfacial scattering plays quite an important role in electrical transport. We then studied the interfacial effect on anomalous Hall resistivity. The Hall resistivity ($\rho_{xy}$) was measured with magnetic field perpendicular to the film plane in a range of $-50 \, \text{kOe} \leq H \leq 50 \, \text{kOe}$ and at temperatures ranging from 5 K to 300 K. Figure 4.12(a) shows the field-dependent
Hall resistivity curves for all samples measured at 5 K. The Hall resistivity increases sharply with low magnetic field and follows a linear dependence up to the magnetic saturation field. Beyond the saturation magnetic field, the Hall resistivity shows a much weaker linear dependence on magnetic field, which mainly comes from the contribution of normal Hall effect. Therefore, the anomalous Hall resistivity (\( \rho_{AHE} \)) could be extracted by

![Graph showing Hall resistivity curves](image)

**Fig. 4.12** (a) Field-dependent Hall resistivity (\( \rho_{xy} \)) of all samples at 5 K; (b) field-dependent Hall resistivity (\( \rho_{xy} \)) of the \( n = 5 \) sample at different temperatures; (c) temperature-dependent anomalous Hall resistivity (\( \rho_{AHE} \)) for all samples.
extrapolating the linear part to zero field. The Hall resistivity increases as $n$ increases from 1 to 12. The sign of $\rho_{AHE}$ for all samples is negative, which is consistent with that of experimental [42] and theoretical [48] results in Ni. The slope of Hall resistivity at high field gives negative normal Hall effect, indicating that electrons dominated transport properties in all samples. Figure 4.12(b) shows the field-dependent Hall resistivity of sample $n=5$ measured at different temperatures, as an example, to illustrate the temperature effect on the AHE. The Hall resistivity decreases with temperature and remains almost the same below 50 K, because the thermal effect on Hall resistivity is quite weak. Figure 4.12(c) presents temperature-dependent anomalous Hall resistivity for all samples. The $\rho_{AHE} \sim T$ curves show the same tendency as $\rho_{xx} \sim T$ curves shown in Fig. 4.11(a). Below 50 K, the anomalous Hall resistivity shows very weak temperature dependence, which indicates insignificant contribution of phonons to $\rho_{AHE}$. As $n$ increases from 1 to 12, the $\rho_{AHE}$ increases by 6.4 times at 5 K and by 4.2 times at 300 K.

To explore the mechanism(s) of the AHE experimentally, Eq. (3.5) is usually employed. The premise of using Eq. (3.5) is that the saturation magnetization of the samples is the same for different $\rho_{AHE}$, because anomalous Hall resistivity is not only related to the longitudinal resistivity but also the magnetization ($M$) of the samples, as suggested by Eq. (1.1). In other words, if the saturation magnetization is different for each $\rho_{AHE}$, we cannot apply the relation, Eq. (3.5), to our data. Based on the structure design of our samples, the composition of all samples (thickness ratio of Ni/Au) is exactly the same, which gives the same saturation magnetization for each sample. Therefore, we can use Eq. (3.5) safely to examine the mechanism(s) of the AHE. The curve $\rho_{AHE} \sim \rho_{xx}$ was plotted logarithmically in Fig. 4.13 for all samples measured at 5 K where the contributions of phonons could be
neglected. By linearly fitting this curve, we obtained the exponent $\gamma = 1.85 \pm 0.03$. The obtained exponent $\gamma$ is close to 2, which could be ascribed to intrinsic/side-jump mechanisms. Comparing to the two different scaling relations in the Fe/Au multilayers, we obtained only one scaling relation in Ni/Au multilayers that have exactly the same structure. In our Fe/Au multilayers, a large exponent ($\gamma > 2$) was observed in the samples that exhibit GMR-dominated magneto-transport properties. Therefore, it is necessary to check if there is GMR effect in the Ni/Au multilayers. To this end, we measured the magnetoresistance (MR) curves with magnetic field applied in the film plane. With magnetic field parallel or perpendicular to the current, the MR curves measured at 5 K for all samples are shown in Fig. 4.14. As is well known, GMR depends only on the relative direction of the spins and is not related to the angle between the magnetization and the current. Anisotropic magnetoresistance (AMR), generally depends on the relative

![Fig. 4.13](image)

**Fig. 4.13** The logarithmic plot of anomalous Hall resistivity ($\rho_{AHE}$) versus longitudinal resistivity ($\rho_{xx}$) measured at 5 K for all samples. The red line is a linear fit to the data.
orientations of the magnetization and the current in polycrystalline ferromagnetic materials. Therefore, as can be seen in Fig. 4.14, AMR dominated the electrical transport properties for all samples. The samples with \( n = 3 - 6 \) showed some contribution of GMR which, however, did not dominate the MR behavior. We then verified that the scaling relation \([\text{Eq. (3.5)}]\) is valid for all samples with exponent \( \gamma = 1.85 \pm 0.03 \).

The exponent is lower than 2, indicating that the contribution of skew scattering may not be neglected. Separation of intrinsic and side-jump contribution is also a key issue, because both mechanisms are predicted to give the same scaling law. Recently, significant progress was achieved by proposing a new scaling relation, \([\text{Eq. (1.5)}]\) \([\text{Eq. (1.6)}]\) to separate the skew-scattering, side-jump, and intrinsic mechanisms. Here, we analyzed our data by

Fig. 4.14 Magnetoresistance curves of all samples measured at 5 K. \( H/ (H\perp) \) indicates that the magnetic field was parallel (perpendicular) to the measurement current.
following the same method [50]. At low temperatures (5 K), Eq. (1.5) can then be simplified as

$$\frac{\rho_{AHE}}{\rho_{xx}} = \alpha + (\beta + b)\rho_{xx}, \quad (4.2)$$

by taking the following approximations $\rho_{AHE} \approx \rho_{AHE0}$ and $\rho_{xx} \approx \rho_{xx0}$. By plotting $\rho_{AHE}/\rho_{xx0}$ versus $\rho_{xx0}$ and linearly fitting this curve, the slope ($\beta + b$) and intercept ($\alpha$) could be obtained. However, as shown in Fig. 4.15(a), a significant deviation from the linear relation between $\rho_{AHE}/\rho_{xx0}$ and $\rho_{xx0}$ is observed. The premise of the linear dependence of $\rho_{AHE}/\rho_{xx0}$ on $\rho_{xx0}$ is that the $\alpha$ and $\beta + b$ both remain constant for all samples. Therefore, the $\alpha$ and $\beta + b$ cannot be regarded as constants for different samples and their values depend on the samples. We recall that the $\alpha$, $\beta$ and $b$ keep constant for one material with different contents of delta-impurity in original theories [17,25,26,29] derived for bulk diffusive transport. However, the interface/surface scattering effect on the values of $\alpha$, $\beta$ and $b$ is still missing in theory. In our samples, the interfacial scattering dominated the electrical transport especially in samples with larger $n$ due to the finite-size effect. Therefore, it is necessary to study the interfacial scattering effect on the mechanisms.

Previously, we found that both $\beta$ and $b$ vary linearly with the residual longitudinal resistivity in the Fe/Au multilayers, as shown in Fig. 4.8(c). Here, we can first obtain the value of $b$ using Eq. (1.6). By linearly fitting each curve in Fig. 4.15(b), the intercepts ($b$ value) could be obtained. As reported in epitaxial Ni thin films [49], the $\sigma_{AHE} \sim \sigma_{xx}^2$ curves cannot be fitted linearly, because of the temperature-dependent intrinsic contribution. However, in our samples, the linear fitting is quite good for each curve. Therefore, the scaling relation, Eq. (1.5), could be used to obtain the intrinsic contribution in our
multilayer samples. It is evident that the $b$ value is not constant and decreases with $n$ increases. The relation $b \sim \rho_{xx}$ in Fig. 4.15(c) shows good linear dependence, which is consistent with the observation in the Fe/Au multilayers. Therefore, we assume both $\beta$ and

**Fig. 4.15** (a) $\rho_{AHE} \sim \rho_{xx}$ curve measured at 5 K. The red line is the polynomial fit by Eq. (4.3) to the data. (b) $\sigma_{AHE} \sim \sigma_{xx}^2$ curves for all samples. The solid lines indicate the linear fittings to each curve. (c) $\alpha \sim \rho_{xx}$, $b \sim \rho_{xx}$ and $\beta \sim \rho_{xx}$ curves. The solid lines are guides to eyes.
$b$ linearly depend on $\rho_{xx0}$, as suggested in Fig. 4.8(c) and Eq. (4.2) can then be rewritten as

$$\frac{\rho_{\text{AHE0}}}{\rho_{xx0}} = \alpha + (C\rho_{xx0} + D)\rho_{xx0},$$

(4.3)

where $C\rho_{xx0} + D = \beta + b$. We then use this equation to fit the curve in Fig. 4.15(a) and the fitting result is quite good. It is noted that Eq. (4.3) gives a cubic term in the scaling of $\rho_{\text{AHE0}}$ versus $\rho_{xx0}$, which seems beyond the current theoretical prediction. Actually, the $C$ and $D$ in expression $C\rho_{xx0} + D = \beta + b$ have no physical meanings and this expression is only a mathematic operation to show the variation of $\beta + b$. Since the curve in Fig. 4.15(a) could be well fitted, we can obtain the $\alpha$ and $\beta + b$ by Eq. (4.3), i.e. the slope of the tangent line of the fitting curve is $\beta + b$ and the intercept of this tangent line is $\alpha$. Apparently, the $\alpha$ and $\beta + b$ are not constant and depend on the residual longitudinal resistivity. The $\beta$ value could then be easily obtained by using the $b$ values from Fig. 4.15(b). We then plotted $\alpha$, $\beta$ and $b$ in relation to the residual longitudinal resistivity in Fig. 4.15(c). It was found that $\alpha$ increases with $\rho_{xx0}$, while both $\beta$ and $b$ decrease linearly with increasing of $\rho_{xx0}$, which is different from that in the Fe/Au multilayers in which $\alpha$ keeps constant, $b$ decreases but $\beta$ increases with $\rho_{xx0}$ increases.

In the Ni/Au multilayers, $\alpha$ increases with $\rho_{xx0}$, which indicates an enhanced skew scattering effect by interfacial scattering. Because the intrinsic mechanism comes from the band structure of crystals, the decrease of the $b$ value with $n$ increases could be easily understood as the crystallinity decaying, which was verified by Fig. 4.9(b). Similarly, intrinsic contribution reduction has been reported in ultrathin Fe films due to the finite-size effect [50,101]. However, we have to understand why side-jump contribution decreases with increasing interfacial scattering. Since the residual longitudinal resistivity includes
two contributions: interfacial scattering between layers and grain boundary scattering inside each layer, both scatterings should contribute to side-jump mechanism. The possible interpretation is that the two contributions have opposite signs, as observed previously in Co/Pd bilayers [69]. That is, the decreases of side-jump contribution could be regarded as that the interfacial scattering contributing to side jump oppositely to intralayer scattering. The overall side jump therefore decreases with residual longitudinal resistivity increases. Actually, the sign of AHE in bulk-like samples with delta-impurity was well established, which depends on the spin polarization of the materials. However, the theory about the sign of interface/surface-scattering-induced AHE is still missing. As reported in Co/Pd multilayers [67,68], the sign of surface-scattering-induced AHE is opposite to that of interfacial scattering. The sign of interfacial-scattering-induced side jump may depend on the materials, which form the interfaces with broken symmetry. Therefore, the interfacial scattering effect on AHE still needs further investigation. Our findings may pave the way to enhance the interfacial-scattering-induced skew-scattering and side-jump contributions in applications of magnetic sensors and spin transfer torque.

4.2.4 Conclusion

In this work, we designed and prepared a series of \((\text{Ni}_{36}\text{Au}_{12})_n\) multilayers and studied the scattering effect on the AHE. The XRR spectra and TEM images of cross-sections show that the interfaces between the Ni and Au layer are sharper than that in Fe/Au multilayers. The transport properties were measured under magnetic fields of \(-50 \text{ kOe} \leq H \leq 50 \text{ kOe}\) in a wide temperature range from 5 K to 310 K. Interfacial scattering between Ni and Au plays quite an important role in longitudinal resistivity. Sharper interfaces lead to lower scattering strength/potential and thus the increments of \(\rho_{xx}\) and \(\rho_{AHE}\) from \(n =1\) to \(n =12\).
are lower than that in the Fe/Au multilayers. Scaling relation $\rho_{\text{AHE}} \propto \rho_{\text{xx}}^\gamma$ with $\gamma = 1.85 \pm 0.03$ was observed for low temperature data. Intrinsic and side-jump contribution were separated successfully and both decrease with residual longitudinal resistivity increases. The decrease of intrinsic mechanism with $\rho_{\text{xx0}}$ increases could be regarded as the crystallinity decaying. The interfacial scattering and intralayer scattering both contribute to side jump in the AHE, but with opposite sign and, consequently, the overall side jump was suppressed. Although Fe and Ni have opposite polarization, the sign of side-jump contribution induced by Fe/Au and Ni/Au interfacial scatterings is the same. This conclusion may enrich the understanding of the interfacial scattering effect on the AHE.

4.3 Interfacial scattering effect on the AMR and AHE in Ta/Fe multilayers

As shown in Fe/Au and Ni/Au multilayers, the interfacial scattering effect plays an important role in the AHE. This multilayer design can also be used to enhance the AHE. Since the AHE and AMR both are related to spin-orbit coupling and Au has relatively smaller SOC comparing to Ta, we prepared $(Ta_{12}/Fe_{36})_n$ multilayers which have the same structure with that of Fe/Au and Ni/Au multilayers to study the interfacial scattering effect on the AMR and the AHE.

4.3.1 Structural characterization

The XRR measurements were performed to characterize the multilayer structures of all samples. The XRR spectra, as seen in Fig. 4.16(a), show clear periodic oscillations for all samples, suggesting periodic multilayer structures with relatively sharp and flat interfaces. More oscillation peaks could be observed for the samples with more periods. To obtain the individual layer thickness of the multilayers, we fitted the XRR spectra by Leptos software
Fig. 4.16 (a) The XRR spectra and (b) high-angle XRD patterns of the multilayer samples.

(V 7.02, Bruker). The fitted layer thickness is quite close to the preset values, which confirmed the stability of thickness control during the sample deposition. Figure 4.16(b) presents the high-angle, $\theta - 2\theta$ XRD patterns for all samples. Only one peak, indexed as Fe(110), was seen for each sample. No peak of Ta layers was observed, suggesting the amorphous structure of Ta layers. As $n$ increases from 1 to 12, the Fe(110) peaks became
broader and weaker, indicative of the decaying crystallinity of the Fe layers. The Fe(110) peaks shift towards lower angles as \( n \) increases from 1 to 12. This peak shift may be due to the following reasons: (1) stress due to different thermal expansion coefficients of Ta and Fe; (2) stress because of the lattice mismatch between Ta and Fe (lattice constant \( a = 0.3306 \) nm for Ta and \( a = 0.2866 \) nm for Fe); (3) interfacial mixing of Ta and Fe (Ta doping in Fe with bcc structure will have larger lattice constant).

To further demonstrate the multilayer structures, we imaged the cross-sections of selected samples by HR-STEM. Figure 4.17 shows the STEM high-angle annular dark-field (HAADF) images of the multilayer cross-sections and the corresponding elemental profiles for samples \( n = 2, 6, \) and 12. The HAADF images show clear contrast of white and grey strips, demonstrating the multilayer structures. The contrast in STEM HAADF images is only sensitive to the atomic numbers of the elements. The white and grey strips, therefore, correspond to the Ta and Fe layers, respectively. As seen in Fig. 4.17(a), the second layer of Ta is much rougher than the Au layer in the Fe/Au multilayers. In our Fe/Au and Ni/Au multilayers, the interfaces are relatively flat because of the immiscibility of the Fe(Ni) layers and Au layers. However, in the Ta/Fe multilayers, the miscibility of the Ta and Fe layers may cause the rough interfaces and the interfacial mixing of Ta and Fe, which would be reflected in the electrical properties. The element profiles were obtained by the line scanning of energy dispersive x-ray spectroscopy (EDX) using scanning transmission electron microscopy, as shown in Fig. 4.17(d)–(f). As expected, these elemental profiles featured with the desired periodicity, demonstrating again the multilayer structures in the samples.
4.3.2 Longitudinal resistivity and interfacial scattering effect on the AMR

The temperature-dependent longitudinal resistivity was measured at temperatures ranging from 5 K to 300 K in a zero-magnetic field. The $\rho_{xx} \sim T$ curves for the samples from $n = 1$ to $n = 12$ were presented in Fig. 4.18(a). All curves show a positive temperature coefficient of resistivity (TCR, $d\rho_{xx}/dT$) across the full temperature range,
which suggests metallic electrical transport in all samples. Below 50 K, the $\rho_{xx}$ shows very weak temperature dependence, indicating negligible contribution of photon scattering to the resistivity. With $n$ increases from 1 to 12, the magnitude of $\rho_{xx}$ shifts to higher values monotonically across the whole measured temperature range. From $n = 1$ to $n = 12$, the $\rho_{xx}$ increases by 2.6 times at 300 K and by 6.4 times at 5 K. This resistivity enhancement clearly indicates the increased scattering effect, including Ta/Fe interfacial scattering and grain boundary scattering within each layer. At 5 K where the impurity scattering dominates the resistivity, the ratio of $\rho_{xx}(n = 12)/\rho_{xx}(n = 1)$ in Ta/Fe multilayers is

**Fig. 4.18** (a) Temperature-dependent longitudinal resistivity for all samples; (b) normalized $\rho_{xx}(T)/\rho_{xx}(300 \, K)\sim T$ curves for all samples.
higher than that in Fe/Au and Ni/Au multilayers, which suggests stronger scattering in Ta/Fe multilayers. Since the multilayer structure is the same for the Fe/Au, Ni/Au and Ta/Fe multilayers, the stronger scattering in Ta/Fe multilayers could be ascribed to the rough interfaces including the intermixing of Ta and Fe at the interfaces. To better demonstrate the scattering effect, we plotted the normalized resistivity curves, $\rho_{xx}(T)/\rho_{xx}(300 \text{ K}) \sim T$, for all samples in Fig. 4.18(b). As shown in Fig. 4.18(b), the resistivity ratio, $\rho_{xx}(T)/\rho_{xx}(300 \text{ K})$, increases with $n$. The values of $\rho_{xx}(5 \text{ K})/\rho_{xx}(300 \text{ K})$ increases from 0.28 to 0.71, which apparently demonstrated more scattering effect in samples with larger $n$. Comparing to grain boundary scattering inside the Ta or Fe layers, the Ta/Fe interfacial scattering plays much more important role in electrical properties because of the size effect, especially in samples with large $n$. Comparing to grain boundary scattering inside the Ta or Fe layers, the Ta/Fe interfacial scattering plays much more important role in electrical properties because of the size effect, especially in samples with large $n$. It is noted that the interfacial scattering enhanced longitudinal resistivity doesn’t depend on the spin-orbit coupling.

The interfacial scattering could also have crucial impact on magneto-transport properties. Spin-dependent interfacial scattering induced giant magnetoresistance has been reported in Fe/Cr multilayers [115]. Theoretical work has reported that interfacial spin-orbit scattering could induce anisotropic magnetoresistance [121]. In section 4.1, interfacial scattering results in anisotropic magnetoresistance in Fe/Au multilayers due to spin-orbit coupling of Au layers and the AMR was enhanced for the samples from $n = 1$ to $n = 12$. Since Ta has larger spin-orbit coupling, we then studied the interfacial spin-orbit scattering effect on the magnetoresistance. The magnetoresistance was measured with
magnetic field in the film plane in a range of $-1000 \text{ Oe} \leq H \leq 1000 \text{ Oe}$ at 5 K. With magnetic field parallel or perpendicular to the current, the $\text{MR} \sim H$ curves were measured at 5 K for all samples, as shown in Fig. 4.19(a)–(i). Generally, AMR depends on the relative orientations of the magnetization and the current in polycrystalline ferromagnetic materials, whereas GMR depends only on the relative direction of the spins. As seen in Fig. 4.19(a)–(i), AMR dominated the magneto-transport properties for all samples and minor GMR signals were shown in the samples with $n = 1 - 4$. The magnitude of AMR could be extracted from the MR curves and was plotted with $n$ in Fig. 4.19(j). For comparison, the AMR values of Fe/Au samples were also plotted. The AMR value is defined as $\text{AMR} = \frac{R_\parallel(1000 \text{ Oe}) - R_\perp(1000 \text{ Oe})}{R_\perp(1000 \text{ Oe})} \times 100\%$, where $R_\parallel(1000 \text{ Oe})$ and $R_\perp(1000 \text{ Oe})$ is the resistivity when magnetic field parallel (perpendicular) to the measurement current at 1000 Oe. As seen in Fig. 4.19(j), the AMR increases with $n$ in Ta/Fe and Fe/Au samples. Since the AMR effect is caused by anisotropic scattering of carriers due to spin-orbit coupling and is not related to the crystallinity of Fe layers, the enhancement of AMR values from $n = 1$ to $n = 12$ could be ascribed to the interfacial spin-orbit scattering. Comparing the AMR values of the Ta/Fe multilayers to that of the Fe/Au multilayers, we found that the AMR increases more slowly in Ta/Fe multilayers with $n$, which seems to exclude the spin-orbit coupling effect. The interfacial spin-orbit scattering does not only depend on the spin-orbit coupling but also depend on the quality of the interfaces between Fe and Ta(Au) layers. If the interfaces are too rough or intermixing happens in the interfaces, the scattered carriers may easily lose their spin coherence and the spin-dependent scattering will be suppressed. As analyzed in the images of the multilayer cross-sections, the interfaces between Ta and Fe are worse than that in Fe/Au samples. Therefore, the worse interfaces may suppress the
spin-dependent scattering and affected the AMR values. Another phenomenon is that shoulders exist in the MR curves especially for the samples with $n = 5 - 12$. The

**Fig. 4.19** (a)-(i) MR curves of all samples measured at 5 K. $H_\parallel (H_\perp)$ indicates that the magnetic field was parallel (perpendicular) to the measurement current; (j) the AMR values of Ta/Fe (black solid square) and Fe/Au (red open circle) multilayers as a function of the number of period $n$ at 5 K.
shoulders may correspond to another magnetic phase which may be TaFe alloy. Because of the miscible property of the Ta and Fe layers, the TaFe alloy may exist at the interfaces, as stated in XRD analysis.

4.3.3 Anomalous Hall resistivity and its scaling analysis

We then studied the interfacial scattering effect on anomalous Hall resistivity. The Hall resistivity for all samples was measured with magnetic field perpendicular to the film plane in a range of $-50 \text{kOe} \leq H \leq 50 \text{kOe}$ and at temperatures ranging from 5 K to 300 K. Figure 4.20(a) presents the field-dependent Hall resistivity for all samples measured at 5 K. Each curve shows a strong linear field dependence at low field until the magnetic saturation field ($H_{sat}$). Beyond the $H_{sat}$, the Hall resistivity shows a much weaker linear field dependence. No hysteresis loops are observed in all Hall resistivity curves. These are the typical behaviors for ferromagnetic films with in-plane magnetic moments. The weakly field-dependent Hall resistivity above $H_{sat}$ could be ascribed to three factors: (1) ordinary Hall effect due to classical Lorentz force on carriers in both Ta and Fe layers; (2) field-forced magnetic moments rotation against thermal agitation (except at 0 K) in Fe layers; (3) field-forced nonferromagnetic coupled spins at interfaces or defects inside Fe layers. The three factors have minor effect on Hall resistivity since the slope of Hall resistivity ($d\rho_{xy}/dH$) at field beyond the $H_{sat}$ is much smaller than that below the $H_{sat}$. Therefore, the Hall resistivity curves are dominated by the anomalous Hall resistivity ($\rho_{AHE}$) and the $\rho_{AHE}$ could be extracted by extrapolating the $\rho_{xy}$ data above the $H_{sat}$ to zero field. To illustrate the temperature effect on the Hall resistivity, we measured the field-dependent $\rho_{xy}$ of all samples at different temperatures. Figure 4.20(b) presents the $\rho_{xy} \sim H$ curves of the sample with $n = 5$ as an example. The curves at different temperatures show similar
behaviors to that observed at 5 K. The $\rho_{xy}$ increases monotonically with temperature. Figure 4.20(c) shows the temperature-dependent anomalous Hall resistivity for all samples. The $\rho_{AHE}$ increases with temperature for each sample across the whole measured

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**Fig. 4.20** (a) Field-dependent Hall resistivity of all samples at 5 K; (b) field-dependent Hall resistivity of the sample with $n = 5$ at different temperatures; (c) temperature-dependent anomalous Hall resistivity for all samples.
temperature range, which shows the same tendency as the $\rho_{xx} \sim T$ curves. Below 50 K, each curve flattens off, which suggest the minor thermal effect on the $\rho_{AHE}$ at low temperatures. With $n$ increases from 1 to 12, the $\rho_{AHE}$ shifts to higher value in the full measured temperature range, which clearly demonstrates the interfacial scattering effect on the anomalous Hall resistivity.

To understand the mechanism(s) of the AHE experimentally, the traditional scaling relation, Eq.(3.5), is usually applied to the data of $\rho_{AHE}$ and $\rho_{xx}$ and the exponent $\gamma$ could be extracted by linearly fitting the curve of $\log(\rho_{AHE}) \sim \log(\rho_{xx})$. $\gamma = 1$ indicates that skew-scattering mechanism dominates the AHE, whereas $\gamma = 2$ may suggest that an intrinsic mechanism, an extrinsic side-jump mechanism, or both dominate the AHE. To avoid the photon scatterings, we plotted the data of $\rho_{AHE} \sim \rho_{xx}$ measured at 5 K logarithmically, as shown in Fig. 4.21. By linearly fitting this curve, we obtained the

![Image of Fig. 4.21](image)

**Fig. 4.21** The logarithmic plot of anomalous Hall resistivity ($\rho_{AHE}$) versus longitudinal resistivity ($\rho_{xx}$) measured at 5 K for all samples. The red line is the linear least-squares fit to the data.
exponent $\gamma = 1.98 \pm 0.08$ which is almost 2. Therefore, the AHE was dominated by intrinsic/side-jump mechanisms or both. Here, we didn’t find the two different scaling relations as observed in the Fe/Au multilayers. As shown in the MR measurement in Fig. 4.19, AMR dominated the MR behaviors and only minor GMR signals were shown in the samples with $n = 1 - 4$. Therefore, the scaling relation, Eq.(3.5), is valid for the samples with $\gamma = 1.98 \pm 0.08$.

The scaling relation $\rho_{AHE} \propto \rho_{xx}^2$ could be ascribed to intrinsic/side-jump mechanisms. Experimentally separating of intrinsic and side-jump contribution is also a challenge in the AHE study. The newly proposed scaling relation [50], Eq. (1.5), completely excluded the contribution of phonon scattering to the mechanisms of skew scattering or side jump. Therefore, the contribution of the side-jump and intrinsic mechanisms were successfully separated. Here, we applied Eq. (1.5) to our data to obtain the contributions of skew-scattering, side-jump and intrinsic mechanisms. At low temperatures (e.g. 5 K), Eq. (1.5) could be simplified to Eq. (4.2). By plotting $\rho_{AHE0}/\rho_{xx0}$ versus $\rho_{xx0}$ and fitting this curve linearly, as shown in Fig. 4.22(a), the slope $\beta + b$ and intercept $\alpha$ were obtained. The obtained $\alpha$ value is $(1.24\pm0.51)\times10^{-3}$, which gives the skew-scattering contribution. By plotting the data, $\rho_{AHE}(T)$ versus $\rho_{xx}(T)$ (not shown), of each sample and linearly fitting each curve, the slope $b$ could be obtained for each sample. Taking the value of $\beta + b$ obtained in Fig. 4.22(a), the value of $\beta$ could also be obtained. Figure 4.22(b) presents the values of $b$ and $\beta$ for all samples. As seen, the intrinsic contribution decreases with $n$ increases except the sample $n = 1$, which may be due to the crystallinity decaying or finite size effect [50,101]. Similar results were also obtained in Fe/Au and Ni/Au multilayers. Remarkably, the sign of the side-jump contribution ($\beta$) changed from negative to positive.
with $n$ increases from 1 to 12. The scattering-induced side-jump contribution may have two origins: interfacial scattering between Ta and Fe and grain boundary scattering inside each Fe layer. The two contributions to side-jump mechanism may have opposite signs, as reported previously in Co/Pd bilayers [69]. This was also concluded in our Ni/Au multilayers, as analyzed in Section 4.2. For samples with less periods, the grain boundary scattering induced side jump dominated, which give a negative $\beta$. As $n$ increases, the

![Graphs](https://via2.mediachase.com/dm/u/11765/395568456.png)
interfacial scattering played a more important role in side jump, which gives a positive $\beta$. It must be noted that, the intrinsic contribution is much larger than the side-jump contribution to the AHE, showing an intrinsic mechanism-dominated AHE in Ta/Fe multilayers. This is consistent with the results in Fe/Au multilayers. As seen in Fig. 4.22(a), $\rho_{\text{AHE}0}/\rho_{xx0}$, denoted as anomalous Hall angle, increases with $n$ increases, which clearly suggests that the interfacial scattering enhanced the anomalous Hall angle. This enhancement may be of great benefit to the development of the AHE-based sensors.

4.3.4 Conclusion

In this part, we prepared $(\text{Ta}_{12}/\text{Fe}_{16})_n$ multilayers and studied the interfacial scattering effect on the AMR and the AHE. The XRR spectra and STEM images of cross-sections confirmed the multilayer structures. The longitudinal resistivity increased by 2.6 times at 300 K and by 6.4 times at 5 K from sample $n = 1$ to $n = 12$, indicative of the interfacial scattering effect. The interfacial spin-orbit scattering enhanced the AMR measured at 5 K. The enhancement of AMR is lower than that in Fe/Au multilayers because of the rough interfaces and the intermixing of Ta/Fe, although the SOC of Ta is larger than that of Au. The Hall resistivity was measured under magnetic field of $-50 \text{ kOe} \leq H \leq 50 \text{ kOe}$ and at temperatures ranging from 5 K to 300 K. The skew-scattering, side-jump and intrinsic contributions were separated successfully. As $n$ increases from 1 to 12, the intrinsic contribution decreases because of the decaying crystallinity or finite size effect and the intrinsic contribution dominated the AHE for all samples, which are consistent with the results in Fe/Au multilayers. The interfacial scattering and intralayer scattering both contribute to side jump in the AHE, but with opposite sign and, consequently, the overall side jump changed from negative to positive.
Chapter 5 Main results and remaining questions

5.1 Anomalous Hall effect in Co$_x$(MgO)$_{100-x}$ granular thin films

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

Metal-insulator granular thin films are typical percolation systems, which are beneficial to study the interfacial scattering effect on anomalous Hall effect. However, how to precisely control the microstructure and realize the sharp interfaces without any interfacial reaction is still a big issue because the interfacial scattering plays a crucial role in the electrical transport properties.

5.2 Interfacial scattering effect on anomalous Hall effect in the multilayers

5.2.1 Anomalous Hall effect in Fe/Au multilayers

We designed and fabricated a family of multilayered (Fe$_{36}$/$\text{Au}_{12}$)$_n$ samples to study the AHE. We performed transport property measurements under different magnetic fields
in a wide temperature range from 10 K to 310 K. We observed different scaling relations in different samples. Our quantitative analysis showed that the interfacial scattering suppresses the contribution of the intrinsic mechanism and gives rise to side-jump contributions. In the search for large spin Hall and anomalous Hall effects, significant attention has been brought to the intrinsic and skew-scattering mechanisms. In comparison, the side-jump mechanism has received only little attention lately. Our findings suggest that interfacial scattering can promote the side-jump mechanism which is the source of the AHE and open interesting avenues for the design of this phenomenon in multilayered structures.

5.2.2 Anomalous Hall effect in Ni/Au multilayers

We prepared a series of \((\text{Ni}_{36n} / \text{Au}_{12n})_n\) multilayers and studied the scattering effect on AHE. The XRR spectra and TEM images of cross-sections show that the interfaces between Ni and Au layer are sharper than that in Fe/Au multilayers. The transport properties were measured under magnetic fields of \(-50 \text{ kOe} \leq H \leq 50 \text{ kOe}\) in a wide temperature range from 5 to 310 K. Interfacial scattering between Ni and Au plays a quite important role on longitudinal resistivity. Sharper interfaces lead to lower scattering strength/potential and thus the increments of \(\rho_{xx}\) and \(\rho_{AHE}\) from \(n = 1\) to \(n = 12\) are lower than that in Fe/Au multilayers. Scaling relation \(\rho_{AHE} \propto \rho_{xx}^{\gamma}\) with \(\gamma = 1.85 \pm 0.03\) was observed for low temperature data. Intrinsic and side-jump contribution were separated successfully and both decreases with residual longitudinal resistivity increases. The decrease of intrinsic mechanism with \(\rho_{xx0}\) increases could be regarded as the crystallinity decaying. The interfacial scattering and intralayer scattering both contribute to side jump in AHE but with opposite sign and consequently, the overall side jump was suppressed. Although the Fe and Ni have opposite polarization, the sign of side-jump contribution
induced by Fe/Au and Ni/Au interfacial scatterings is the same. This conclusion may enrich the understanding of interfacial scattering effect on the AHE.

5.2.3 Anomalous Hall effect in Ta/Fe multilayers

We prepared \((\text{Ta}_{12}/\text{Fe}_{36})_n\) multilayers by magnetron sputter system and studied the interfacial scattering effect on the AMR and the AHE. The XRR spectra and STEM images of cross-sections confirmed the multilayer structures. The longitudinal resistivity increased by 2.6 times at 300 K and by 6.4 times at 5 K from sample \(n = 1\) to \(n = 12\), indicative of the interfacial scattering effect. The interfacial spin-orbit scattering enhanced the anisotropic magnetoresistance measured at 5 K. The enhancement of AMR is lower than that in Fe/Au multilayers because of the rough interfaces and the intermixing of Ta/Fe, although the SOC of Ta is larger than that of Au. The Hall resistivity was measured under magnetic field of \(-50 \text{ kOe} \leq H \leq 50 \text{ kOe}\) and at temperatures ranging from 5 K to 300 K. The skew-scattering, side-jump and intrinsic contributions were separated successfully. As \(n\) increases from 1 to 12, the intrinsic contribution decreases because of the decaying crystallinity or finite size effect and the intrinsic contribution dominated the AHE for all samples, which are consistent with the results in Fe/Au multilayers. The interfacial scattering and intralayer scattering both contribute to side jump in the AHE, but with opposite sign and, consequently, the overall side jump changed from negative to positive. The overall side jump shows the same tendency as that in Fe/Au multilayers.

5.2.4 Open questions

We could realize better interface control and study the interfacial scattering effect on anomalous Hall effect in multilayer samples. However, the intermixing of the two materials
at the interfaces still exist in our samples. Furthermore, the crystallinity of the layers will
decay when the layer thickness decreases, which may also influence the anomalous Hall
effect. Preparing epitaxial multilayer thin films with atomic sharp interfaces may be a
solution to the above issues. Also, the interfacial scattering strength/potential has not been
quantitatively obtained. How to quantitatively control and analyze the interfacial scattering
effect is still a great challenge.
PUBLICATIONS


BIBLIOGRAPHY


