Anomalous Hall magnetoresistance in single-crystal Fe(001) films

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Keywords: anisotropic magnetoresistance, anomalous Hall magnetoresistance, angular dependent magnetoresistance

Abstract

The angular-dependent magnetoresistance (MR) in epitaxial Fe(001) films grown on MgO(001) is systematically investigated at room temperature. The resistivities with in-plane magnetic fields parallel ($\rho_x$) and transverse ($\rho_y$) to a current and a perpendicular field ($\rho_z$) show a correlation of $\rho_z \approx \rho_x > \rho_y$ for Fe film thickness ($d_{Fe}$) between 3 and 50 nm, which is distinctly different from the conventional anisotropic MR relation of $\rho_x > \rho_y \approx \rho_z$. The dependence of such unusual MR on $d_{Fe}$ is quantitatively explained by the competition between the anomalous Hall MR, intrinsic anisotropic MR, and the MR induced by the geometrical size effect in Fe films. Our results also reveal the strong four-fold symmetric terms in the measured angular-dependent MR with a linear dependence of $1/d_{Fe}$.

1. Introduction

Magnetoresistance (MR) in ferromagnetic (FM) materials, depending on its magnetization direction, plays pivotal roles in both the fundamental understanding of spin-dependent transport and technological applications [1]. Anisotropic magnetoresistance (AMR) [2–5] is a fundamental MR effect exhibited by magnetic materials, which describes the dependence of the longitudinal electric resistivity ($\rho_x$) on the magnetization (M) direction relative to the electric current in FM materials. The intrinsic mechanism of AMR is usually linked to the anisotropic s–d scattering stemming from the spin–orbit coupling [2–5]. Recently, the discovery of several types of MR effects with different origins has triggered renewed interest in spin-dependent MR. These MR effects include spin Hall magnetoresistance (SMR) in FM/heavy metal bilayers [6–8], Rashba–Edelstein magnetoresistance (REMR) in Bi/Ag/CoFeB [9], spin–orbit magnetoresistance (SOT-MR) in a YIG/Pt/Cu system [10], Hanle magnetoresistance (HMR) in HMs [11], anisotropic interfacial magnetoresistance (AIMR) in Pt/Co/Pt [12, 13], and anomalous Hall MR (AHMR) in single FM layers [14–16]. In a particular magnetic system, several types of MR could coexist. Thus, to separate the contribution from each type of MR, it is necessary to carefully measure the magnetization orientation correlation of MR.

Considering intrinsic AMR, the isotropic approximation in polycrystalline FM materials results in a characteristic dependence of $\rho_x > \rho_y = \rho_z$, where $\rho_x$ is the longitudinal resistivity for M along the current direction, $\rho_y$ is the resistivity for M aligned also in the plane but perpendicular to the current direction, and $\rho_z$ is the polar resistivity for M aligned perpendicular to the film plane. Usually, the AMR in thin polycrystalline films follows the same angular dependence as that of isotropic bulk samples, which always assumes $\rho_y = \rho_z$ [2]. However, there are a few violations reported that show angular dependence of $\rho_x > \rho_y > \rho_z$ in thin Co [17], Ni [18], and permalloy (Py) [19] films, which are attributed to the geometrical size effect (GSE). The GSE has been explained as an anisotropic effect of the spin–orbit interaction on the s–d scattering of the minority spins caused by film texture [17]. In 2011, Kobs et al [12]...
reported a new type of angular correlation of MR with $\rho_x > \rho_z > \rho_y$ in Pt/Co/Pt sandwiches, which was named as AIMR. Although AIMR was explained by the anisotropic specular scattering at the Pt/Co interface [12, 13], the microscopic mechanism still remains controversial, because the proximity effect and spin Hall effect from the neighboring Pt layer are difficult to separate. Recently, Yang et al. [16] reported that the anomalous Hall effect (AHE) in FM films can break the symmetry between $\rho_x$ and $\rho_y$, which is named as AHMR. AHMR has been explained by the spin accumulation, subsequent spin backflow, and spin–charge conversion inside the magnetic film [14–16]. The AHE is a common spin-dependent transport property in FM materials, but it still requires further experimental investigation to confirm whether AHMR is a general phenomenon in metallic FM materials. Besides the MR effect in single FM layers, in magnetic bilayers with Pt and ferrimagnetic insulators, such as YIG, the SMR effect with the characteristic relation of $\rho_x = \rho_z > \rho_y$ has been reported [6], and was interpreted by the spin–charge conversions caused by the spin Hall effect and inverse spin Hall effect. The REMR in Bi/Ag/CoFeB [9] and SOT-MR in YIG/Pt/Cu [10] have a similar relation of $\rho_x = \rho_z > \rho_y$, which is caused by the interfacial spin–orbit coupling.

Therefore, four distinct types of MR correlation have been observed in different FM films or multilayers to date, which are $\rho_x > \rho_y = \rho_z$ (conventional AMR), $\rho_x > \rho_y > \rho_z$ (GSE), $\rho_x > \rho_z > \rho_y$ (AIMR, AHMR), and $\rho_x = \rho_z > \rho_y$ (SMR, REMR and SOT-MR). In polycrystalline Fe thin films prepared by sputtering, Zou et al. [20] reported that all four types of MR correlations were observed because of the competing effects originating from thickness, texture, interface, and morphology. However, the underlying mechanisms of these MR correlations in a single Fe layer, especially for the relation of $\rho_x = \rho_z > \rho_y$, are not well understood, and it is better to perform the MR measurement in single crystalline Fe films.

In this paper, we systematically study the angular-dependent magnetoresistance (ADMR) effects in single-crystalline Fe films as a function of film thickness ($d_{Fe}$). The MR in Fe films shows a correlation of $\rho_x \approx \rho_z > \rho_y$, similar to that of SMR. Such an unusual MR correlation can be well understood by AHMR, which can readily match the thickness-dependent MR ratios. The AHMR in Fe films is determined to be larger than the intrinsic AMR. Moreover, the measured ADMR with the field rotating in the $xy$, $yz$, and $xz$ planes clearly shows four-fold symmetry, which is inversely proportional to $d_{Fe}$. Therefore, our results demonstrate that symmetry breaking at the interface can influence the AMR in FM thin films.

2. Experimental

The body-centered-cubic Fe films were prepared by molecular beam epitaxy in an ultra-high vacuum (UHV) system with a base pressure of $2 \times 10^{-10}$ Torr. The MgO(001) single-crystal substrates were first annealed at 600 °C for 30 min in the UHV system and then a 10 nm MgO seed layer was grown at 300 °C to decrease the surface roughness. The Fe layers were deposited at room temperature (RT) and then annealed at 350 °C for 10 min to improve their surface morphology. Fe/MgO(001) is one of few metal on insulator systems with high quality epitaxial growth [21–27], and becomes a model system to investigate the magnetic [24, 25] and transport properties [26, 27] of ultrathin magnetic films. The Fe film grown on MgO(001) has the well-known epitaxial relationship of Fe(001)[100]/MgO(001)[110] with a 3.5% tensile lattice mismatch. The stripe patterns in figure 1(a) and (b) measured by reflection high-energy electron diffraction (RHEED) confirm the high-quality epitaxial growth of the Fe films. Figure 1(c) shows the x-ray diffraction spectrum from a 10 nm Fe film, which presents the single Fe(002) peak from the film. From the diffraction peak of 66.2°, the vertical lattice parameter of Fe film can be calculated as 2.825 Å, which is ~1.4% less than the bulk Fe lattice constant of 2.866 Å due to the tensile strain from the MgO substrate. The samples were capped with a 4 nm MgO protection layer before being taken out of the growth chamber.

To systematically investigate the effect of the interface on AMR, the Fe film was grown into a wedge shape with a varying $d_{Fe}$ from 3 to 30 nm. The film was patterned into Hall bars with a width of 0.1 mm and length of 0.6 mm. Current was applied along the Fe(100) direction but perpendicular to the wedge direction (figure 1(d)). To study the AMR in a thicker film, an Fe film with a uniform $d_{Fe}$ of 50 nm was grown and patterned into similar Hall bars. Angular-dependent magnetotransport measurements were performed using a physical property measurement system equipped with a sample rotator.

3. Results and discussion

Figure 1(e) shows the thickness-dependence of the zero-field resistivity of the epitaxial Fe films. Because of increasing interface scattering in thinner films, the resistivity increases sharply with decreasing $d_{Fe}$. The inset in figure 1(e) shows that the resistivity has a linear dependence on $1/d_{Fe}$ when $d_{Fe} > 5$ nm.
According to the Fuchs–Sondheimer model [28, 29], the longitudinal resistivity can scale with thickness as
\[
\rho = \rho_b \left( 1 + \frac{3(1-p)}{8l_e} \right),
\]
where \(\rho_b\) is the bulk resistivity, \(p\) is the specular reflectivity, and \(l_e\) is the electron mean free path in Fe film. The fitted \(\rho_b\) of the Fe film on MgO(001) is 11.8 \(\mu\)\(\Omega\)\(\text{cm}\), which is larger than the bulk resistivity of single-crystalline bulk Fe of 9.7 \(\mu\)\(\Omega\)\(\text{cm}\) [30]. The larger bulk resistivity in thin film is usually attributed to the electron scattering by the structural defects and the interfaces. The fitted value of \(l_e\) is 11.4 nm. By assuming \(p = 0\), we can obtain the lower limit of the electron mean free path \(l_e\).

Figure 1(f) shows typical field-scan MR curves of an Fe film with \(d_{Fe} = 5.3\) nm for the field along the \(x\), \(y\), and \(z\) directions, which show the unusual MR correlation of \(\rho_x > \rho_y > \rho_z\). The \(\rho-H\) curves indicate that the saturation field for the field normal to the film is about 2 T. Above the saturation field, all the \(\rho-H\) curves show linear field dependence with a negative slope due to the suppression of the electron–magnon scattering by the high field [31].

To further understand the thickness-dependent AMR of Fe films, we measured ADMR with the field rotating in the \(xy\) plane (\(\alpha_{xy}\) scan), \(xz\) plane (\(\beta_{xz}\) scan), and \(yz\) plane (\(\gamma_{yz}\) scan), using the measurement geometries shown in figures 2(a), 2(b), and 2(c), respectively. All measurements were performed at RT with a rotating field of 5 T. The negative slopes of the \(\rho-H\) curves for the three field directions in figure 1(e) are similar, thus the MR relations measured at high field will not change with the strength of the applied field. Figures 2(d)–2(f) show typical ADMR curves as a function of magnetization orientation angle \(\theta_M\) in the \(\alpha_{xy}\), \(\beta_{xz}\), and \(\gamma_{yz}\) scans, respectively. In the \(\alpha_{xy}\) scan (figure 2(a)), \(\theta_M\) is expected to be the same as the field orientation angle \(\theta_M\) because the applied field of 5 T is much stronger than the in-plane anisotropic field of Fe film. In the \(\beta_{xz}\) scan (figure 2(b)) and \(\gamma_{yz}\) scan (figure 2(c)), because of the strong shape anisotropy, \(\theta_M\) can deviate from \(\theta_M\) even in the 5 T applied field. However, \(\theta_M\) can be calculated by minimizing the free energy \(E = -M_H \cos(\theta_M - \theta_M) + \frac{2\pi M_H^2 \cos^2(\theta_M)}{l_e^2} \) which consists of Zeeman energy and shape anisotropy energy calculated from the bulk Fe magnetization \(M_s\).

From the ADMR curves, we can obtain the anisotropic resistivity \(\Delta\rho\) and AMR ratio \(\Delta\rho/\rho\). Here, \(\Delta\rho\) in each measurement geometry is defined as \(\Delta\rho_{\alpha_{xy}} = \rho_x - \rho_y\) in the \(\alpha_{xy}\) scan, \(\Delta\rho_{\beta_{xz}} = \rho_z - \rho_x\) in the \(\beta_{xz}\) scan, and \(\Delta\rho_{\gamma_{yz}} = \rho_z - \rho_y\) in the \(\gamma_{yz}\) scan, as indicated in figure 2(e). We found that when \(d_{Fe} > 3.8\) nm, \(\Delta\rho_{\alpha_{xy}}\) and \(\Delta\rho_{\beta_{xz}}\) show similar values, and \(\Delta\rho_{\gamma_{yz}}\) has a small positive value, resulting in an unusual MR correlation of \(\rho_z \gtrsim \rho_x > \rho_y\), which is obviously different from the ordinary correlation of \(\rho_x > \rho_y > \rho_z\) for the conventional AMR in polycrystalline thin films [2]. It should be noted that the \(\rho_z > \rho_x\) relation has never been reported in single FM layers [2, 20], and was only recently reported in Co/Pt bilayers with a very thin Co layer [32]. Here, it should be noted that the observed \(\rho_z \gtrsim \rho_x\) relation could exist in Fe films up to 50 nm.

Another unusual feature in the ADMR curves in figure 2 is that the \(\beta_{xz}\) scan always shows a dominant four-fold contribution, different from the dominant two-fold contribution in the \(\alpha_{xy}\) scan. Moreover, the \(\gamma_{yz}\) scan shows the superposition of two- and four-fold contributions. The four-fold contribution of the ADMR curve cannot exist in the polycrystalline film, and has only been reported for single-crystalline films.
such as Fe₃O₄ [33, 34], (Ga,Mn)As [35], Ni [36], Fe₄N [37], and Co₂MnSi [38]. The ADMR curves of epitaxial Fe films in all αₓᵧ, βₓ𝒛, and γₓ𝒛 scans can be expressed by the following equation:

\[ \rho = \rho_{0} + \Delta \rho \cos^{2} \theta_{M} + \Delta \rho_{x} \cos 4 \theta_{M}. \]  

(1)

Here, \( \rho_{0} \) is a constant that is independent of the magnetization directions, the second term represents the two-fold contribution of AMR with \( \Delta \rho \) defined in figure 2(e), the third term denotes the four-fold contribution with the magnitude \( \Delta \rho_{x} \). Equation (1) is consistent with the phenomenological theory in which \( \rho_{0}, \Delta \rho_{x}, \) and \( \Delta \rho_{y} \) can be expressed by the galvanomagnetic tensors [2]. Figure 3(a) shows the \( d_{Fe} \)-dependence of \( \Delta \rho_{x}, \Delta \rho_{y}, \Delta \rho_{z} \) fitted from the ADMR curves. \( \Delta \rho_{y} \) and \( \Delta \rho_{z} \) always have similar values and quickly decrease with \( d_{Fe} \). \( \Delta \rho_{z} \) is one order of magnitude smaller than \( \Delta \rho_{y} \) and \( \Delta \rho_{z} \). All \( \Delta \rho_{x}, \Delta \rho_{y}, \Delta \rho_{z} \) are clearly inversely proportional to \( d_{Fe} \), as indicated in the inset of figure 3(a), which suggests an interfacial AMR contribution. Because \( \rho_{0} \) has a linear \( 1/d_{Fe} \) dependence as well (figure 1(e)), the measured AMR ratio \( \Delta \rho/\rho_{0} \) decreases with \( d_{Fe} \) but does not follow a highly linear \( 1/d_{Fe} \) dependence.

We will now discuss the origin of the observed ADMR relations as a function of \( d_{Fe} \). In conventional AMR theory [2], \( \Delta \rho_{y} \) is expected to be zero based on the symmetry analysis. However, substantial \( \Delta \rho_{y} \) in the \( \gamma_{xyz} \) scan has been reported for thin Co [17], Ni [18], and Py [19] films because of the GSE. Gil et al [17] pointed out that the GSE originates from the anisotropic effect of the spin–orbit interaction on the s–d scattering caused by the symmetry breaking between the in-plane direction and polar direction of magnetization. Thus, it is expected that the GSE will induce an additional AMR with a correlation of \( G \bullet m_{z} \), where \( m_{z} \) is the magnetic component of the unit magnetization. Usually, the symmetry breaking in a film is induced by texture or strain during growth, so the observed GSE has little dependence on film thickness [12, 39]. Conversely, the interface can break the symmetry, and induce a slight difference of the electronic structures for M aligned along the in-plane or polar direction. The GSE theory indicates that such interface symmetry breaking could induce the ADMR effect in the \( \gamma_{xyz} \) scan, which could be inversely proportional to the FM layer thickness. As discussed in reference [17], the sign and magnitude of the GSE are related to the anisotropic effect of the spin–orbit effect and the band splitting between 3d orbitals, so it is still possible to induce a positive \( \Delta \rho_{y} \) (\( G > 0 \)) in some FM systems depending on their electronic structures [20], although the observed GSE in Co [17], Ni [18], and Py [19] only shows negative \( \Delta \rho_{y} \) (\( G < 0 \)). However, the GSE will not induce the difference of resistance for \( M \) aligned along the x or y directions and the intrinsic AMR effect should always exist in the \( \alpha_{xyz} \) scan for FM materials. Thus, the GSE theory alone is insufficient to explain the experimental result that \( \Delta \rho_{xyz} \) is always similar to \( \Delta \rho_{x} \) in the Fe films with \( d_{Fe} \) up to 50 nm.

Recently, Yang et al [16] discovered that the AHE could also induce an additional substantial MR effect in the \( \gamma_{xyz} \) scan and confirmed this AHMR effect in the polycrystalline Fe₃Mn₁₋₋ₙ-based alloy systems. We found that the ADMR results observed for our Fe films can be well explained by AHMR. Including the

Figure 2. Schematic diagrams of ADMR measurements with the magnetization rotating in (a) \( xy \) plane, (b) \( xz \) plane and (c) \( yz \) plane. Angular-dependent resistivities for the field rotating in \( \alpha_{xyz}, \beta_{xsz} \) and \( \gamma_{yxz} \) scans with different Fe film thicknesses of (d) 3.8 nm, (e) 11.3 nm, and (f) 30 nm. All measurements were performed at room temperature with a rotating field of 5 T. The solid lines are the fitting curves from equation (1).
conventional AMR effect [2–5], GSE [17–19], and the AHMR effect [14–16] gives a general MR form of

\[ \rho (m) = \rho_0 \left\{ 1 + r_A m_x^2 + r_G m_z^2 + \left( \frac{\theta_{AH}}{\beta} \right)^2 m_z^2 + \left( 1 - \frac{2l_s}{d} \tanh \left( \frac{d}{2l_s} \right) \right) m_y^2 \right\}. \]  

(2)

Here, \( d \) and \( l_s \) are the thickness and spin diffusion length of the FM layer, respectively, \( m \) is the magnetization direction, \( \beta \) is the polarization of the longitudinal conductivity, \( \theta_{AH} \) is the anomalous Hall angle, \( r_A \) is the intrinsic AMR ratio, and \( r_G \) is the AMR ratio induced by GSE. Then, for the ADMR measurement in \( \alpha_{xy}, \beta_{xz} \) and \( \gamma_{yz} \) scans, we can obtain

\[ \frac{\Delta \rho_{xy}}{\rho_0} = r_A - \left( \frac{\theta_{AH}}{\beta} \right)^2 + \left( \frac{\theta_{AH}}{\beta} \right)^2 \frac{2l_s}{d} \tanh \left( \frac{d}{2l_s} \right), \]  

(3)

\[ \frac{\Delta \rho_{yz}}{\rho_0} = r_G + \left( \frac{\theta_{AH}}{\beta} \right)^2 \frac{2l_s}{d} \tanh \left( \frac{d}{2l_s} \right), \]  

(4)

\[ \frac{\Delta \rho_{xz}}{\rho_0} = r_G + \left( \frac{\theta_{AH}}{\beta} \right)^2 - r_A. \]  

(5)

Equations (3) and (4) can explain why \( \frac{\Delta \rho_{xy}}{\rho_0} \) and \( \frac{\Delta \rho_{yz}}{\rho_0} \) have the same thickness dependence, \( \frac{\Delta \rho_{yz}}{\rho_0} \) and \( \frac{\Delta \rho_{xz}}{\rho_0} \) can have similar magnitude if \( r_A - \left( \frac{2l_s}{d} \right) \tanh \left( \frac{d}{2l_s} \right) \beta \) have similar values. Because of the symmetry breaking at both Fe/MgO interfaces and different electron scattering behavior at the interfaces and interior of the Fe films, all the parameters, including \( r_A, r_G, \theta_{AH}/\beta, \) and \( l_s \), could depend on \( d_{Fe} \). However, we found that, if assuming the fixed values of \( r_A, r_G, \theta_{AH}/\beta, \) and \( l_s \), the experimental curves of the \( d_{Fe} \)-dependent \( \frac{\Delta \rho_{xy}}{\rho_0} \) and \( \frac{\Delta \rho_{yz}}{\rho_0} \) can be well fitted by equations (3) and (4), as shown in figures 3(b) and (c), respectively. The fitted parameters for the \( \frac{\Delta \rho_{xy}}{\rho_0} \) curve are \( l_s = 2.3 \pm 0.2 \) nm, \( \frac{\theta_{AH}}{\beta} = 0.081 \pm 0.001 \), and \( r_A = 0.0085 \pm 0.0004 \). The fitted parameters for the \( \frac{\Delta \rho_{yz}}{\rho_0} \) curve are \( l_s = 2.5 \pm 0.1 \) nm, \( \frac{\theta_{AH}}{\beta} = 0.082 \pm 0.001 \),
and \( r_G = 0.0020 \pm 0.0001 \). The fitted values of \( \frac{\Delta \rho_{xy}}{\rho_0} \) from the \( \frac{\Delta \rho_{xz}}{\rho_0} \) and \( \frac{\Delta \rho_{yz}}{\rho_0} \) curves are almost the same. It should be noted that the GSE-induced AMR has to be included to explain the finite value of \( \frac{\Delta \rho_{xy}}{\rho_0} \) for the thick Fe film. The fitted value of \( \bar{l} \) is within the range of those reported for other FM materials [16, 40]. Utilizing the giant magnetoresistance measurement, Gurney et al [41] found that the spin-dependent mean free path of Fe is 1.5 nm for the spin-up electron and 2.1 nm for the spin-down electron. The AHE in MgO/Fe/MgO has been well studied [27], and the measured \( \theta_{\text{AHE}} \) varies from 0.025 to 0.02 as the film thickness increases from 8 to 33 nm. Using the point-contact method, Nadgorny et al [42] showed that \( \beta \) of an Fe film is \( \sim 43\% \). Then, \( \theta_{\text{AHE}} \) can be calculated to be 0.035 from the fitted value of \( \frac{\Delta \rho_{xy}}{\rho_0} \), which is close to the value measured by AHE [27].

Equation (5) shows that the small value of \( \frac{\Delta \rho_{xy}}{\rho_0} \) results from the competition between \( r_A, r_G, \) and \( \left( \frac{\beta_{\text{AH}}}{\beta_{\text{st}}} \right)^2 \), and should be a constant if \( r_A, r_G, \theta_{\text{AHE}}, \) and \( \beta \) are all independent of \( d_{Fe} \), in contrast to our results in figure 3(d). Experimental studies have shown that \( \theta_{\text{AHE}} \) decreases with \( d_{Fe} \) [27, 43]. The GSE-induced AMR, i.e., \( r_G \), could also decrease with \( d_{Fe} \) because of its interfacial contribution, so it is reasonable that \( \frac{\Delta \rho_{xy}}{\rho_0} \) decreases with \( d_{Fe} \). In fact, the measured \( \frac{\Delta \rho_{xy}}{\rho_0} \) for \( d_{Fe} > 5 \text{ nm} \) can be well fitted by a simple function of \( a + b/d_{Fe} \), where \( a \) and \( b \) are fitting parameters, as indicated by the red line in figure 3(d). Although the possible thickness dependence of \( r_A, r_G, \theta_{\text{AHE}}, \) and \( \beta \) may impede the determination of their accurate values from the fitting using equations (3) and (4), the same thickness dependence of \( \frac{\Delta \rho_{xz}}{\rho_0} \) and \( \frac{\Delta \rho_{yz}}{\rho_0} \) provides strong evidence of the dominant contribution from AHMR in the Fe film. Equation (3) also shows that the AMR ratio \( \frac{\Delta \rho_{xy}}{\rho_0} \) in the \( \alpha_{xy} \) scan in a bulk material is the competing result between \( r_A \) and \( \left( \frac{\beta_{\text{AH}}}{\beta_{\text{st}}} \right)^2 \). This indicates that the intrinsic AMR ratio caused by the bulk s–d scattering should be larger than the values obtained in the ordinary \( \frac{\Delta \rho_{xy}}{\rho_0} \) measurements. It should be noted that the thickness-dependent decay of AHMR is caused by the spin accumulation near the interface within the short \( l_s \). Thus, it is reasonable that the \( \Delta \rho \) values in figure 3(a) show \( 1/d_{Fe} \) dependence for the films thicker than \( 2l_s \).

Recently, Zhang et al [44] predicted that interfacial spin–orbit scattering could also generate an additional ADMR effect, which can explain the substantial thickness-dependent AMR in the \( \beta_{xz} \) and \( \gamma_{yz} \) scans. However, \( \frac{\Delta \rho_{xy}}{\rho_0} \) estimated based on this theory was one order of magnitude smaller than our experimental values [44]. Therefore, the contribution from interfacial spin–orbit scattering should be much smaller than that from AHMR.

The thickness-dependent four-fold anisotropic resistivity \( \Delta \rho_{s} \) can be also determined by fitting the experimental ADMR curves using equation (1), as presented in figure 4(a). It is clear that the fitted \( \Delta \rho_{s}^{xy} \) is almost independent of \( d_{Fe} \), but \( \Delta \rho_{s}^{xz} \) and \( \Delta \rho_{s}^{yz} \) show a similar \( 1/d_{Fe} \) dependence. AMR with four-fold symmetry has been observed in single-crystal FM materials such as Fe3O4 [33, 34], (Ga,Mn)As [35], Ni [36], FeN [37], and CoMnSi [38], but all these studies focused on the measurements in \( \alpha_{xy} \) scans. Our measurements showed that four-fold symmetry could exist in all \( \alpha_{xy} \), \( \beta_{xz} \), and \( \gamma_{yz} \) scans. In the past, the four-fold AMR was just discussed based on the phenomenological theory, and only recently it has been studied theoretically based on the electronic band structure. Yahagi et al [45] theoretically investigated the four-fold symmetric AMR effect in a cubic single-crystal ferromagnet, and attributed the four-fold AMR to the s–d impurity scattering with cubic crystal fields and spin–orbit coupling. Kokado et al [46, 47] also theoretically investigated the four-fold symmetric AMR effect in a tetragonal single-crystal ferromagnet system, and attributed the four-fold AMR to the localized d states with spin–orbit interaction, the exchange field and the crystal field of tetragonal symmetry. Therefore, the four-fold AMR should

![Figure 4](image-url)
reflect the symmetry of the electron density distribution in the impurity d-band. Figure 4(b) shows the electron density distribution of all five d states. The $d_{x^2-y^2}$ and $d_{xy}$ states show four-fold symmetry in the $xy$ plane, and the $d_{xz}$ and $d_{yz}$ states present four-fold symmetry in the $xz$ and $yz$ planes, respectively. Thus, the spin–orbit coupling may cause the different electron scattering for $\rho_z$ rotating in $xy$, $xz$, or $yz$ planes, which should contain the four-fold symmetry. Because of the in-plane four-fold structural symmetry of the Fe/MgO(001) system with the tensile strain, it is reasonable that should contain the four-fold symmetry. Because of the in-plane four-fold structural symmetry of the spin–orbit coupling may cause the different electron scattering for $\rho_z$

4. Summary

Unusual ADMRs were observed in single-crystalline Fe/MgO(001) systems with an SMR-like correlation of $\rho_x \approx \rho_y > \rho_z$ for $d_{3z^2}$ up to 50 nm. This unconventional ADMR behavior is attributed to the competition between the intrinsic AMR, AHMR, and GSE-induced MR effect. The MR ratios in both $\alpha_{xy}$ and $\gamma_{xy}$ scans show a similar dependence on $d_{3z^2}$, which can be quantitatively fitted with the AHMR theory. Our results suggest that AHMR always exists in FM materials; thus, the effect of spin current on the AMR measurements induced by AHE is not limited to thin films, but also exists in bulk systems. It is important to separate the contributions from the intrinsic AMR, AHMR, and GSE-induced MR effect to understand the spin-dependent transport properties in FM materials. Four-fold symmetric terms in the ADMR measurements in $\beta_{xy}$ and $\gamma_{xy}$ scans with linear $1/d_{3z^2}$ dependence were observed, confirming that AMR could be influenced by interface scattering. Our experiments call for further theoretical investigation to better understand the relationship between the four-fold symmetric term in ADMR and electronic states of FM films.

Acknowledgments

We acknowledge helpful discussions with Prof. Shulei Zhang and Prof. Shufeng Zhang. This work was supported by the National Key Research and Development Program of China (Grant No. 2016YFA0300703), National Natural Science Foundation of China (Grant No. 11974079, 11734006, and 11434003), and the Program of Shanghai Academic Research Leader (No. 17XD1400400).

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