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Tuning the magnetoresistance symmetry of Pt on magnetic insulators with temperature and magnetic doping

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We present a comparison study of the temperature dependence of the intriguing magnetoresistance (MR) in Pt/YIG (yttrium iron garnet), Pt/YIGBB (the YIG substrate has been bombarded with Ar⁺), and Pt/SiO2 (with different Fe doping levels). With decreasing temperature, the MRs in Pt/YIG and Pt/YIGBB change symmetry from $R_z = R_x > R_y$ at room temperature to $R_x > R_z > R_y$ at low temperature. A similar behavior in both Pt/YIG and Pt/YIGBB implies that the underlying physics is due to magnetic scattering, instead of the pure spin current across the interface. By changing the Fe doping level in the SiO2 substrate, we can further systematically modulate the symmetry of MR in Pt/SiO2 (Fe doped). The doping level dependent symmetry can also qualitatively explain the controversy over the MRs of Pt/YIG and similar structures at low temperature. Published by AIP Publishing.

Heterostructures of heavy metals with strong spin-orbit coupling on magnetic insulators have been widely studied in various spin current related phenomena, such as spin Hall effect,1,2 spin pumping,3,4 spin Seebeck effect,5–7 and spin transfer torque.8,9 Among these structures, platinum (Pt)/yttrium iron garnet (YIG) is a typical system, where Pt acts as the spin current generator/detector, and YIG transports or produces spin current without accommodating charge current.5,7,10–15 Recently, an unconventional magnetoresistance (MR) has been reported first in Pt/YIG bilayer structures16–19 and subsequently in Pd/YIG,20–22 Ta/YIG,11,23,24 W/YIG,25 and Pt on other magnetic insulators.26–28 Interestingly, the resistance experienced by electrons passing only through the non-magnetic films reflects the magnetization orientation of the underlying insulating substrates, enabling the electronic detection of the magnetization of the ferromagnetic insulators. The resistance with in-plane field can be described as $R = R_x - \Delta R [m \cdot (z \times j)]^2$, where $\Delta R = R_x - R_y$, $R_x$ (or $R_y$) denotes the resistance with the magnetic field along the $x$-($y$-) axis, $m$ and $j$ are the unit vectors in the directions of magnetization $M$ and current $J$, and $z$ is the direction perpendicular to the film plane. This in-plane symmetry is identical to the well-known anisotropic magnetoresistance (AMR) in most ferromagnetic metals, with $R_x > R_y$ and a cosine square angular dependence. However, it exhibits an unconventional behavior under an out-of-plane magnetic field of $R_z = R_x > R_y$, in sharp contrast to the well-known behavior of $R_x > R_y = R_z$ for AMR.

The unconventional MR in Pt/YIG and similar system has drawn intense interest, both experimentally and theoretically. While most previous studies focused on the measurements at room temperature, the MR in Pt/YIG and similar systems at low temperature brings more interesting features. Lin et al., reported that the MR symmetry of Pd/YIG deviated from $R_z = R_x > R_y$ at room temperature to $R_y > R_x > R_z$ at low temperatures.21 They decompose the MR in Pd/YIG into two contributions: one from spin Hall magnetoresistance (SMR) $(R_x = R_z > R_y)$ and the other from the AMR of the polarized Pd layer $(R_y > R_z = R_x)$, which emerges only at low temperatures. A similar explanation is also adopted in the Pt/LaCoO3 structure.28 Vélez et al. observed $R_y > R_x > R_z$ in Pt/YIG at low temperatures, and they ascribed this to the weak anti-localization in Pt which would typically enhance the resistance of metal with strong spin-orbit coupling under the perpendicular magnetic field.29 However, Shiomi et al., found that the symmetry of $R_y = R_z > R_x$ was preserved for Pt/YIG even at low temperatures.30 Furthermore, weak anti-localization appears only after special surface treatment of the YIG surface before the Pt deposition.30 At present, different symmetries of MR, some without extensive data, have been reported in supposedly similar structures. Therefore, it is highly desirable to study the underlying mechanism that modulates the MR symmetry and explain the controversy between different observations.

In this work, we present detailed studies of the MRs in Pt films deposited on YIG, Ar⁺ bombarded YIG (YIGBB) and Fe-doped SiO2 substrates in a wide temperature range (10–300 K). We find that the MR symmetry is directly related to the magnetic scattering at interface. At room temperature, the MRs in these structures all have the same angular dependence of $R_z = R_x > R_y$, and the MR ratios increase with magnetic field. The MR of Pt/YIG at low temperature exhibits a behavior of $R_x > R_z > R_y$, which is significantly different from its unique angular dependence observed at room temperature. This phenomenon also persists in a similar system Pt/YIGBB (the YIG surface was purposely altered by Ar⁺ beam bombardment before deposition of Pt, see below), where the spin current has been intentionally blocked and

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only magnetic scattering exists. The similar MR symmetry observed in Pt/YIG and Pt/YIGTaN with or without pure spin current across the Pt-YIG interface, suggests the importance of magnetic scattering to the observed intriguing angular dependence change at different temperatures. By introducing Fe impurities with different doping levels into the amorphous SiO$_2$ substrate, we can further systematically tune the symmetry of MR in Pt/SiO$_2$(Fe) at low temperature from $R_x > R_y$ (clean SiO$_2$), through $R_x > R_y > R_z$ (low percentage Fe doping in SiO$_2$), and reach $R_x > R_z > R_y$ (high percentage Fe doping in SiO$_2$). Our finding emphasizes the importance of magnetic scattering at the interface and qualitatively explains the discrepancy over the symmetry of MRs in Pt/YIG and similar structures among different groups.

We use rf magnetron sputtering to deposit amorphous SiO$_2$ (with or without Fe dopants) onto thermally oxidized Si(001) substrates and dc magnetron sputtering to deposit 3-nm Pt onto polycrystalline YIG slab (SurfaceNet GmbH), YIGTaN, and Fe-doped SiO$_2$ substrates at room temperature. The thicknesses of all substrates are ~0.5 mm. The deposited thin films have been patterned into 0.2 mm wide Hall bars with one long segment (5 mm) and three short side bars 1.5 mm apart, as shown in Fig. 1(a). The 4-terminal method has been used to measure the MR with the current along the long segment and the voltage obtained from the two adjacent side bars. In order to access the angular dependence of MR, we rotate the magnetic field $B$ within the $xy$-, $xz$-, and $yz$-planes with angles $\phi_y$, $\phi_x$, and $\theta_y$ relative to $x$-, $x$-, and $z$-axes, respectively, where $x$-, $y$-, $z$-axes are parallel to the edges of the YIG substrate. For the thermal measurements, a perpendicular temperature gradient of $\nabla T \approx 20$ K/mm is established by placing the sample in between and in contact with two large Cu plates maintained at different constant temperatures. Under a vertical temperature gradient, the spin Seebeck effect in YIG drives a pure spin current flow along the $z$-direction and can be detected as a thermal voltage $V_{th}$ via the ISHE with $E_{ISHE} \propto J_z \times \sigma$ in the Pt layer, where $J_z$ and $\sigma$ are the flow direction of the spin current and spin index, respectively, with $\sigma$ parallel to the YIG’s magnetization direction. The distance between the two voltage leads for measuring the thermal voltage is around 4.2 mm.

Figure 1(b) presents the field dependence of $R_x$ (black circle) and $R_y$ (red square) for Pt(3 nm)/YIG with the in-plane field. The small plateau near the origin is commonly found in thick YIG substrates. This MR with a unique angular dependence with $R_x > R_y > R_z$ [see also Fig. 2(a)] has been widely discussed. In Fig. 1(b), the blue triangle (right scale) shows the thermal voltage $V_{th}$ for Pt (3 nm)/YIG under a vertical temperature gradient ($\nabla T \approx 20$ K/mm) and the field $B$ is applied along the $y$-axis ($\phi_y = 90^\circ$). The measured signal is of ~10 $\mu$V in magnitude and exhibits an asymmetric behavior in field, consistent with the symmetry required for ISHE ($E_{ISHE} \propto J_z \times \sigma$).

Before the deposition of the Pt film, we purposely alter the YIG surface by Ar$^+$ beam bombardment. The altered YIG surface greatly reduces the spin-mixing conductance at the Pt-YIG interface, thus blocking the spin current transmission. Bombardment on the YIG surface for 5 min (500 V, current density 0.4 mA/cm$^2$) essentially eliminates all spin current injection. Thus, both the MR ratio ($R_x > R_y$/$R_y$) at low field (100 mT) and thermal voltage $V_{th}$ disappear [Fig. 1(c)], which also corroborates that the MR at the low field is correlated with spin current, consistent with the SMR model proposed by Nakayama et al.. The negligible $V_{th}$ of Pt(3 nm)/YIGTaN (Fig. 1c inset) and considerable MR ratio at 1.5 T also evidence that the high field MR is not due to pure spin current. The observed MR was attributed to a combination of magnetic scattering and spin orbit coupling. In particular,

![Figure 1](image1.png)

**FIG. 1.** (a) Schematics for the measurements of the angular dependent magnetoresistance and the thermal voltage $V_{th}$ with temperature gradient along the $z$-axis. $\phi_y$, $\phi_x$, and $\theta_y$ denote the angle between the magnetic fields with respect to $x$-, $x$-, and $z$-axes within the $xy$-, $xz$-, and $yz$-planes, respectively. (b) Field dependent longitudinal MR $R_x$ (black circles), transverse MR $R_y$ (red squares), and thermal voltage $V_{th}$ (blue triangles, right scale) for Pt(3 nm)/YIG. (c) Field dependent $R_x$, $R_y$, and $V_{th}$ for Pt(3 nm)/YIGTaN. The inset of (c) presents the time evolution of thermal voltage $V_{th}$ of Pt(3 nm)/YIGTaN under different magnetic fields.

![Figure 2](image2.png)

**FIG. 2.** Angular dependent MR measured within $xy$-, $xz$-, and $yz$-planes with angles $\phi_y$, $\phi_x$, and $\theta_y$ relative to $x$-, $x$-, and $z$-axes, for (a) Pt(3 nm)/YIG at 300 K and (b) Pt(3 nm)/YIG at 10 K. (c) and (d) are the angular dependent MR within three planes for Pt(3 nm)/YIGTaN. The inset of (c) presents the time evolution of thermal voltage $V_{th}$ of Pt(3 nm)/YIGTaN under different magnetic fields.
the Ar⁺ bombarded YIG surface provides us the advantage to investigate the MR behavior related to the magnetic scattering only, since the spin current has been blocked at the interface.

Figure 2(a) presents the angular dependent resistance of Pt(3 nm)/YIG at room temperature within the xy-, xz-, and yz-planes with angles \( \phi_{xy} \) (red square), \( \phi_{xz} \) (black circle), and \( \phi_{yz} \) (blue triangle) relative to \( x \), \( x \), and \( z \)-axes, respectively. The magnetic field is fixed at 8.0 T which is sufficiently larger than the demagnetizing field of the YIG substrate. Rotating \( B \) within the \( xy \)- and \( yz \)-planes gives a cosine square angular dependence with respect to the \( x \)- and \( y \)-axes, respectively. The resistance essentially is a constant when the magnetic field is rotated within the \( xz \)-plane, i.e., \( R_z = R_x > R_y \). Although the resistance of Pt(3 nm)/YIG is isotropic when \( B \) is less than 100 mT [Fig. 1(c)], the difference among \( R_x \), \( R_y \), and \( R_z \) emerges and increases with increasing \( B \) also exhibiting \( R_z = R_x > R_y \) [Fig. 2(c)]. In both Pt/YIG and Pt/YIG, MRs follow \( R_z = R_x > R_y \) at room temperature and have the cosine square angular dependence; while both spin current and magnetic scattering (together with spin-orbit coupling) contribute to the MR in Pt/YIG at the high field, only magnetic scattering is involved in Pt/YIG. Pt/YIG is thus a suitable reference system with respect to Pt/YIG for further investigations, although the detailed reason why the joint effect of magnetic scattering and spin-orbit coupling gives the same symmetry to SMR is still elusive.

With decreasing temperature, the MR in Pt(3 nm)/YIG deviates from its symmetry observed at room temperature (\( R_z = R_x > R_y \)) and exhibits \( R_x > R_z > R_y \) at 10 K [Fig. 2(b)]. As shown in Fig. 2(d), the MRs in Pt(3 nm)/YIG exhibit similar behavior to Pt(3 nm)/YIG at 10 K and have \( R_x > R_z > R_y \). The similar behavior of MR in Pt/YIG and Pt/YIG, with or without pure spin current across the Pt-YIG interface, suggests that the magnetic scattering instead of spin current is the origin of the observed intriguing symmetry change at different temperatures. Figures 4(a) and 4(b) summarize the MR ratio \( \Delta R/R \) within three different planes for Pt(3 nm)/YIG and Pt(3 nm)/YIG at different temperatures. As the temperature decreases, MR symmetry in both Pt/YIG and Pt/YIG changes from \( R_z = R_x > R_y \) to \( R_x > R_z > R_y \). In Pt/YIG, both SMR and magnetic scattering contribute to the observed MR, where the peak behavior of \( \Delta R \) in Pt/YIG was explained by the spin Hall magnetoresistance theory with a constant spin Hall angle and temperature dependent spin diffusion length. For the Pt(3 nm)/YIG, only magnetic scattering exhibits and the MR ratios increase with decreasing temperature to 10 K [Fig. 4(b)].

In order to further demonstrate the importance of magnetic scattering, we performed the temperature dependent MR study of Pt on Fe-doped SiO₂ with different doping levels. As a control sample, Pt(3 nm)/SiO₂ exhibits isotropic feature when rotating the magnetic field within 3 different planes at room temperature, i.e., \( R_x = R_y = R_z \) [Fig. 3(a)], consistent with the previous study as pure Pt itself shows no observable feature. As temperature decreases, \( R_z \) would be enhanced with magnetic field exhibiting \( R_z = R_x = R_y \) due to weak antilocalization [Fig. 3(b)], which is commonly observed in nonmagnetic metals with strong spin-orbit coupling. Using the magnetron co-sputtering technique, we introduce Fe impurities into the SiO₂ substrate. When the SiO₂ is slightly doped, for instance, SiO₂(0.5% Fe), very small angular dependent MR is observed at room temperature [Fig. 3(c)]. Interestingly, with decreasing temperature, the contribution from magnetic scattering enhances, and an in-plane MR with \( R_x > R_z \) in Pt(3 nm)/SiO₂(0.5% Fe) emerges at 10 K. Meanwhile, the weak anti-localization still dominates, and therefore, Pt(3 nm)/SiO₂(0.5% Fe) exhibits an angular dependence with \( R_z > R_x > R_y \) at 10 K [Fig. 3(d)]. Further increasing the Fe doping level to 15.3%, sizable MR with \( R_z = R_x > R_y \) appears under 8.0 T even at room temperature, see Fig. 3(e). This MR, albeit with the same symmetry of the SMR model, is due to an interplay of magnetic scattering and strong spin-orbit coupling and is proportional to the Fe impurity in the substrate. At low temperature, \( R_y \) of Pt(3 nm)/SiO₂(15.3% Fe) is further suppressed compared with Pt(3 nm)/SiO₂(0.5% Fe), and the MR symmetry changes to \( R_x > R_z > R_y \) at 10 K [Fig. 3(f)]. Therefore, we demonstrate that the MR symmetry in Pt/Si!O₂(Fe doped) can be symmetrically modulated by only tuning the Fe doping level in the SiO₂ substrate, highlighting the importance of magnetic scattering at the interface. Figures 4(c) and 4(d) summarize the evolution of MR ratio within three different planes obtained at 8.0 T for Pt(3 nm)/SiO₂(0.5% Fe) and Pt(3 nm)/SiO₂(15.3% Fe), respectively. The MR ratios increase with decreasing temperature, which is expected as the magnetic scattering generally becomes stronger at low temperature. By summarizing the features of the temperature dependent and the doping-level dependent MRs, we attribute the peculiar change of MR symmetry at low temperature to the magnetic scattering.
which increases with increasing doping level and decreasing temperature.

As presented above, Pt/SiO$_2$(15.3%Fe) fully reproduces MR symmetry as Pt/YIG and Pt/YIG$_{\text{BB}}$ at both room temperature and low temperature. Due to the similarity, our findings can also serve as an explanation for large discrepancy among different groups for the temperature dependent symmetry of MR in Pt/YIG and similar systems. For instance, the angular dependence of $R_x > R_y > R_z$ was reported in Pt/YIG, 29 and $R_y > R_y > R_x$ was found in Pd/YIG, 21 and Pt/LaCoO$_3$ 28 at low temperature, respectively; whereas $R_y > R_x > R_y$ of Pt/YIG was preserved below room temperature in Ref. 30, and the authors note that the treatment of the YIG surface is important for the experimental observations. For Pt/SiO$_2$, no MR exists within the $xy$-plane and the extra enhancement of $R_x$ in 10 K is due to the weak anti-localization. In Pt/SiO$_2$(0.5%Fe), the weak anti-localization still dominates and the magnetic scattering already plays considerable contribution, thus it exhibits a behavior of $R_y > R_x > R_y$. This is similar to the case observed in Ref. 29. In Pt/SiO$_2$(15.3%), the magnetic scattering dominates and $R_x$ is further suppressed, resulting in a symmetry with $R_x > R_y > R_y$. This observed symmetry is similar to the one reported in Refs. 21 and 28. Thus, our findings also qualitatively explain why different angular dependences were observed by different groups and why the surface treatment was crucial to suppress weak anti-localization in Ref. 30 since the magnetic scattering at the interface could be very different with different preparation methods.

In summary, by tailoring the Pt-YIG interface, we studied the temperature dependent magnetoresistance in Pt/YIG with and without Ar$^+$ bombardment on the YIG surface in a wide temperature range. In both cases, the MR exhibits symmetry of $R_y = R_y > R_x$ at room temperature, and switches to $R_y > R_y > R_x$ at low temperature. The observed similar behavior in both Pt/YIG and Pt/YIG$_{\text{BB}}$ implies that the underlying physics is due to magnetic scattering, instead of the pure spin current across the interface. By introducing Fe impurities in SiO$_2$ substrate, we further demonstrate that different symmetries can be reproduced in Pt/SiO$_2$(Fe) by modulating the Fe doping level in the SiO$_2$ substrate only. Our findings evidence the importance of the magnetic scattering on the temperature dependent MR observed in Pt thin films on magnetic insulators. In addition, it also qualitatively explains the discrepancy over the angular dependences of MR in Pt/YIG and similar systems among different groups.

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