Field effect controlled magnetism and magnetotransport in low dimensions

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Chapter 4

Gate-Controlled Spin-Dependent Magnetoresistance of a Platinum | Paramagnetic Insulator Interface

Magnetoresistance (MR), the change of electrical resistance in response to an applied magnetic field, plays an indispensable role in developing data storage devices. A new type of MR is reported in this letter. We induced an atomic thin ferromagnetic layer on the surface of platinum (Pt) with an easy axis normal to the film plane by ionic gating. Both longitudinal and transverse transports can be modulated magnetically showing non-saturating characteristics. This phenomenon is ascribed to the paramagnetic nature of the ionic liquid being used and has been interpreted successfully by applying the spin-Hall magnetoresistance theory with perpendicular magnetic anisotropy. The present results indicate that paramagnetic ionic gating can serve as a versatile tool to control the magnetic properties of spintronics devices by electrical means.
4.1 Introduction

4.1.1 Spintronics

Spintronics, namely the electronics based on spin, emerged from a series of discoveries in 1980s. It studies the subject that how to inject, transport and detect the spins. In addition to charge, which is the primary characteristic of electrons, manipulating spin is of great interest for the reason that the spin transport is a non-volatile process with advantages in the data transfer efficiency and energy consumption.

The spin refers to the angular momentum \( S \) that is separated from the orbital motion \( L \) of an electron. The magnitude of the projection of the spin along an arbitrary axis is quantized as \( \hbar/2 \). There is an associated magnetic moment, with its magnitude of

\[
\mu = \frac{\sqrt{3}}{2} \frac{e}{m_e} \hbar,
\]

where \( e \) is the elementary charge of electrons, \( m_e \) is the effective mass of electrons. A wide variety of magnetic phenomena as well as other intriguing transport effects originate from the spin. The phase transition that many spins align with one another forms the ground state of ferromagnetism, whereas once two electrons with up and down spins form pairs, the coherent state of superconductivity emerges. In non-magnetic materials, the electrons are equally presented in the system with up and down spins. In ferromagnetic material, the chemical potential of spin up and spin down are shifted, leading to a net spin polarization and can be quantized as

\[
P = \frac{N^\uparrow - N^\downarrow}{N^\uparrow + N^\downarrow},
\]

where \( N^\uparrow \) and \( N^\downarrow \) represent the number of spin up and spin down electrons at \( E_F \).

The net spin polarization can be converted into a current through several approaches. If it is carried by a charge current, such as in a diffusive conductor, then we call it spin current. In another case, if the spin angular momentum propagates through an insulator, then we term this quasi-particle as magnon hence the flow of magnon is a spin wave.

Spin (or magnon) transports through media in a diffusive way. The period of time that such a non-equilibrium population can last without fully relaxed is known as spin (magnon) lifetime \( \tau \). Accordingly, the distance that spin population can propagate is defined as spin (magnon) diffusion length \( \lambda \). The decay mechanisms of spin (magnon) current are mainly due to spin-flip scattering and spin dephasing. In metals, especially heavy element with strong spin-orbit interaction (SOI), \( \lambda \) is very short and we call it spin sink. In light materials and semi-metals, e.g. graphene, \( \lambda \) can be as long as one hundred microns, which partly contributes to the enormous interests and vigorous researches on graphene recently.
4.1 Introduction

4.1.2 Molecular spintronics

The use of molecular spin state as a carrier for quantum information storage and computing has attracted growing interest in developing next generation spintronics devices. Molecules can respond actively to the external stimuli, such as light, electric bias voltage and magnetic field. So far, molecules have been used as magnets [1-3], spin filters [4], electric conductors [5, 6], electric switches [7] and spin valves [8]. For example, when a neutral planar molecule, zinc methyl phenalenyl are grown on ferromagnetic cobalt surface, interfacial charge transfer generates a hybridized organometallic supramolecular layer, which shows large magnetic anisotropy with spin filter properties. In turns, this layer creates a spin dependent resistance and causes an interface magnetoresistance effect [9].

The idea that using organic molecules as gating media dates back to 2004, when the group led by Yoshihiro Iwasa invented a method of controlling the surface carrier density by organic molecules in certain devices called electric double layer transistors (EDLT). Later, the gating media develop into organic electrolyte, e.g. KClO₄ dissolved in polyethylene oxide. The introduction of ionic liquids to the EDLT is a big step forward. Because of the wide chemical window and low melting temperature, the gating can be performed at lower temperature and higher voltage. Under these conditions, the chemical side-reaction is suppressed and induced carrier density can as high as triggering superconductivity in some band insulators [10].

The ionic gating technique offers a new scheme for controlling the electronic state of materials. However, so far it has not been developed into the field of spintronics. Adding the spin degree of freedom for controlling the electron/spin transport would be of great interests. With this regard, ionic liquids that contain transition metal elements with unpaired spins promise to serve as effective dopant that would be able to form considerable interaction with the channel materials via gating.

The flexible composition and diverse functionality of paramagnetic ionic liquids have enriched ample space to design proper systems for applications in spintronics. In chapter 2 and chapter 3, we have demonstrated how to induce novel ferromagnetic and Kondo states in normal metal films, respectively. In this chapter, we will mainly focus on the interfacial transport phenomenon from spin point of view.

4.1.3 Paramagnetic ionic gating induced magnetoresistance

Magnetoresistance (MR) of metals, the change of the resistivity tensor caused by application of external magnetic field \( B \), is the key of developing data storage [11, 12], sensors [13] and logic devices [14]. The rapid advancement in information technology relies on the discovery of novel types of MR. For example, the discovery of giant magnetoresistance that is a breakthrough in the field of spintronics triggers the revolution of many applications [11, 12]. In general, magnetoresistive devices
require the usage of ferromagnets, where the magnetization direction is crucial to the electrical transport. Paramagnets have often been regarded less interesting to new MR effect for lacking spontaneous magnetization and long range magnetic ordering. On the other hand, it is highly desirable to control the magnetization electrically from both fundamental and technological points of view [15-17]. Due to intrinsically large carrier densities and consequently short Thomas-Fermi screening lengths, metals are difficult to be manipulated electrically. Ionic gating, however, offers a unique approach that is able to alter the electronic state of the channel surface significantly by applying only a few volts [18-20]. In this chapter, we will discuss the observation of a new type of MR in a non-magnetic metal/paramagnetic insulator system with in-plane $B$ field that is gate-controllable. The magnitude of the induced MR is observed to be proportional to the magnetization of the proximate paramagnetic insulator. Contrary to the anisotropic magnetoresistance of ferromagnets that saturates after magnetization are fully aligned, the observed MR can be well described with the Langevin function of paramagnetism, which gives rise to a non-saturating spin-dependent characteristic.

4.2 Concepts

4.2.1 A brief history of the magnetoresistance

Magnetoresistance is the key of developing a variety of electronic and spintronic devices. Before we start exploring a new MR effect in the present system, let us first have a brief review on the history of the discovery of various kinds of MR.

![Figure 4.1](image_url) The family tree of the current known magnetoresistance and the corresponding discovering time.
Magnetoresistance was first discovered by William Thomson (Lord Kelvin) in 1856. He noticed that the resistance of ferromagnets decreased with magnetic field normal to the current direction and increased when they were in parallel. The difference $\Delta \rho = \rho_\parallel - \rho_\perp$ is the anisotropic magnetoresistivity (AMR) and the normalized quantity $\Delta \rho / \left( \frac{1}{3} \rho_\parallel + \frac{2}{3} \rho_\perp \right)$ is called the anisotropic magnetoresistivity ratio [21].

For non-magnetic metals (NMs), the ordinary magnetoresistance (OMR) is in general very small at low fields.

- In metals with closed Fermi surfaces, electrons are constrained to their orbit in $k$ space. Magnetic field $H$ increases the cyclotron frequency of the electrons, given by

$$\omega_c = \frac{eH}{2\pi m_e},$$

where $e$ and $m_e$ are the elementary charge and mass of the electrons. For small $H$, $\omega_c \tau \ll 1$, and MR increases with $H$ quadratically; whereas for large $H$, $\omega_c \tau \gg 1$, MR will saturate.

- For NMs with equal numbers of electrons and holes, such as semi-metals bismuth or some Dirac metal graphene, MR increases with $H$ linearly up to highest field measured.

- NMs that contain Fermi surface with open orbits show very large MR in these crystallographic directions while saturates quickly in other directions that orbits are closed.

The giant magnetoresistance (GMR) was discovered in 1988, first in Fe/Cr/Fe multilayer system [11, 22]. It was found that depending on the magnetization direction of the two ferromagnetic layers, the resistance difference $\Delta R = (R_{\perp} - R_{\parallel})/R_P$ can be as large as 80%. It has a great impact on the information industry. The data storage capacity increases dramatically afterwards and the dispute between the disk and the hard drive finally ended with triumph of the later.

The magnitude of GMR can even increase when exchanging the non-magnetic layer from conducting materials to insulating materials. This breakthrough was ascribed to the great advancement on the molecular beam epitaxial (MBE) technique, which was invented in 1960s. Thanks to MBE, the growth of ultra-thin insulating layer becomes possible, and electrons can tunnel through two ferromagnetic layers. It is much easier for electron with parallel magnetization to find free states to tunnel. Nowadays, this tunneling magnetoresistance (TMR) replaces GMR to be the most commonly used technique in industry and the related device is called magnetic tunneling junction (MTJ).
Figure 4.2 Schematic diagram of the various types of magnetoresistance. (a) Anisotropic magnetoresistance depends on the relative direction between magnetization $M$ and electrical current $I_e$. (b) Anisotropic interface magnetoresistance relies on the ultra-thin systems with perpendicular magnetic anisotropy. (c) Giant (tunneling) magnetoresistance requests a non-magnetic (insulating) layer sandwiched between two ferromagnetic layers. Depending on the relative magnetization direction of two ferromagnetic layers for anti-parallel and parallel structures, the perpendicular resistance reaches high and low states, respectively. (d) The spin-Hall magnetoresistance is an interface effect that requires a heavy metal with strong SHE and a ferromagnetic insulator. (e) The Hanle magnetoresistance depends on the external $B$ field direction instead of $M$. (f) The magnetic proximity effect is an alternative explanation of the resistance modulation of the non-magnetic conductor/ferromagnetic insulator interface, which assumes the non-magnetic conductor is magnetized by the proximate ferromagnet. The color code: green for the ferromagnetic conductor (FMC), blue for the paramagnetic conductor (PMC), orange for the ferromagnetic insulator (FMI). Red, green purple and dark blue arrows denote the electric current, spin current, magnetic field and magnetization directions, respectively.

The aforementioned two effects are related with magnetic multilayers. Colossal magnetoresistance (CMR), on the other hand, is an extremely large resistivity change in a single material with the active ions having mixed-valence state. The first reported system is Sr-doped LaMnO$_3$ in 1994 [23]. The matrix material is antiferromagnetic and insulating. By doping 0.2-0.4 Sr, the donor electrons become delocalized and the matrix materials become conducting and ferromagnetic. The conducting mechanism is presumed to be hopping through two adjacent Mn ions, which have valence states 3+ and 4+ alternatively. Applying external magnetic field facilitates the alignment of the spins the hopping, hence increases the conductivity.

Another approach without involving magnetic materials is geometric magnetoresistance. In general, from application point of view, applying magnetic
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Field results in low resistance state. Corbino, however, proposed a device that magnetic field could increase the MR significantly. The so-called Corbino disc comprises of a conducting annulus with the inner and outer rims connected to the power supplier. Without magnetic field, a radial current flows. When applying a magnetic field parallel to the axis of the annulus, due to the Lorentz force, a circular component of the current flows as well. This will lead to a change of the carrier velocity \( \nu \) that is inverse proportional to \( B \), hence the resistance will decrease. The higher the mobility of the disc material is, the larger this effect will be.

Ideal magnetic field sensor would require large dynamic range. Most of the commercial magnetic field sensors use Hall effect to correlate the \( B \) with responding signal \( V_{xy} \). For accuracy concern, the larger the value of \( |V_{xy}/B| \), the better the accuracy. This would require the use of materials with high mobility, such as two-dimensional electron gas. However, a drawback of this type of sensors is that at very large \( B \) field, it will show quantum Hall effect, when \( B \) is no longer proportional to the \( V_{xy} \). Therefore, materials with linear magnetoresistance without saturating would be of great interests in this application. So far, several families of materials have been discovered, such as doped silver chalcogenide [24-26] and tungsten ditelluride [27].

The rise of the spintronics after the discovery of GMR motivates the science community to look for new type of MR based on spin reorientation. Magnetic domain walls may cause an additional MR that can be moved by electrical current. When ultrathin ferromagnetic metal is sandwiched between two layers of heavy metal with strong SOI, such as Pt, perpendicular magnetic anisotropy is induced for the FM/Pt layer. Depending on the thickness of the FM layer, an additional interface MR contribution is generated in the same order of magnitude as the AMR, which is termed as anisotropic interface magnetoresistance [28].

The strong SOI in Pt also allows us to create and detect spin current in a way called spin-Hall magnetoresistance (SMR). It was first discovered in non-magnetic metal (NM)/ferromagnetic insulator (FMI) bilayer system [29]. We will explicitly discuss this MR effect later.

SMR refers to the effect with a geometry that external magnetic field \( B \) aligns the magnetization direction in the plane. For applying \( B \) out of the plane, another MR effect emerges, which is called Hanle magnetoresistance. The Hanle effect is due the spin dephasing arising from the simultaneous spin precession and diffusion. The spin accumulation at the interface is suppressed under \( B \) field, which will lead to a correction to the resistance.

Besides the strong SOI, Pt is also on the verge of the ferromagnetic excitation. When Pt is close to a ferromagnetic material, ferromagnetism may be induced. The magnetic proximity effect (MPE) is proposed to account for the induced AMR in Pt.
4.2.2 Anisotropic magnetoresistance

In chapter 2, we discussed the family of Hall effect, including the anomalous Hall effect (AHE) for the ferromagnetic materials. Besides AHE, another intrinsic effect found in ferromagnets is the **anisotropic magnetoresistance** (AMR), which depends on the orientation of the magnetization $M$ with respect to the electric current direction $I$.

The electric transport is described by the **Ohm's law** that the electric field $\vec{E}$ is proportional to the electric current density $\vec{j}$ with the resistivity tensor $\hat{\rho}$:

$$\vec{E} = \hat{\rho} \cdot \vec{j}$$

The electric field due to spontaneous magnetization is given by [30]

$$\vec{E} = \rho_\perp \vec{j} + (\rho_H - \rho_\perp) (\vec{j} \cdot \hat{m}) \hat{m} + \rho_H \hat{m} \times \vec{j},$$

where $\hat{m}$ is the unit vector along $M$, $\rho_\parallel, \rho_\perp$ are the resistivities parallel and perpendicular to $\hat{m}$, $\rho_H$ the Hall resistivity. Eq. 4.5 can be further expanded into

$$\vec{E}(\phi, \theta) =
\begin{pmatrix}
\rho_\perp + \Delta \rho m_x^2 & \Delta \rho m_x m_y - \rho_H m_z & \Delta \rho m_x m_z + \rho_H m_y \\
\Delta \rho m_x m_y + \rho_H m_z & \rho_\perp + \Delta \rho m_y^2 & \Delta \rho m_y m_z - \rho_H m_x \\
\Delta \rho m_x m_z - \rho_H m_y & \Delta \rho m_y m_z + \rho_H m_x & \rho_\perp + \Delta \rho m_z^2
\end{pmatrix} \begin{pmatrix}
J_x \\
J_y \\
J_z
\end{pmatrix},$$

where $\Delta \rho = \rho_\parallel - \rho_\perp$, $m_x = \sin \theta \cos \phi$, $m_y = \sin \theta \sin \phi$, $m_z = \cos \theta$. The $\theta$ defines the angle between the vector normal to the film plane and the $M$ and $\phi$ is the azimuthal angle between the projection of $M$ in the film plane and the $J$.

In Eq. 4.6, the AMR refers to the diagonal terms [30], the AHE corresponds to the first non-diagonal matrix elements (in blue), and the **planar Hall effect** (PHE) is attributed to the second terms of the non-diagonal matrix (in red) [31, 32].

If we apply a current along the $x$-axis, magnetization is forced to be lied within $xy$ plane, then $\theta = 90^\circ$, $J_y = J_z = 0$. The expressions of AMR, PHE and AHE in Eq. 4.7 can be simplified to

$$\rho_{AMR} = \rho_\parallel + \Delta \rho \cos^2 \phi,$$

$$\rho_{PHE} = \Delta \rho \sin \phi \cos \phi,$$

$$\rho_{AHE} = -\rho_H \sin \phi.$$

4.2.3 Spin-Hall magnetoresistance

In spintronics, electric transport and spin transport are often correlated in a way called **spin Hall effect** (SHE) that a flow of longitudinal charge current $I$ generates into a transverse spin current $I_s$ due to the spin-orbit interaction (SOI), where the directions of $I$, $I_s$ and spin polarization $\sigma$ are normal to each other.

In analog to the power supply in electronics, a spin source is indispensable in the spintronic field as well. **Spin pumping** is the technique that can generate the spin current. It relies on the microwave ferromagnetic resonance, or other optical
and electrical methods to cause magnetization motion that will induce a flow of spin, similar to that electric fan generate wind.

The reciprocal effect of spin pumping would be that spin currents cause magnetization motion, resulting in the spin-transfer torque $\tau$ [33]. The interaction between the spins and the magnetic layer is governed by the spin-mixing conductance, which describes how many spin transfer momentum can be transferred into the magnetic layer.

When an electric current $I$ flows in a heavy metal with strong SOI, such as Pt in our case, a spin accumulation $\mu$ transverse to the direction of $I$ will be generated towards the Pt/PIL interface. The generated $\mu$ can be absorbed or reflected depending on the relative angle $\phi$ between the direction of the magnetization $M$ and the spin polarization $\sigma$. When $M$ is perpendicular to $\sigma$, $I$ will be absorbed by $M$ as a spin-transfer torque. In contrast, when $M$ is parallel to $\sigma$, $I$ will be reflected by the $M$, and converted back into $I$ via inverse spin Hall effect (ISHE). This concerted manner between SHE and ISHE has been well established in a paradigm namely spin-Hall magnetoresistance (SMR) [29].

SMR was discovered in Pt/yttrium iron garnet (YIG) system. Later it was found to be an universal effect in other magnetic insulators, such as CoFe$_2$O$_4$ [34], CoCr$_2$O$_4$ [35] and Fe$_3$O$_4$ [36]; and the spin injector/detector has also been extended to Ta, W [37], Rh [38] for being used as to other systems.

The effects of SMR are present in both longitudinal and transverse transport, given by the following equation [39]:

$$\rho_L = \rho + \Delta \rho_0 + \Delta \rho_1 \left(1 - m_y^2\right)$$

$$\rho_T = \Delta \rho_1 m_x m_y + \Delta \rho_2 m_z$$

where $\rho$ is the measured electrical resistivity, $\rho_L$, $\rho_T$ the longitudinal and transverse resistivities, respectively; $m_x$, $m_y$, and $m_z$ the components of the magnetization in the x, y, z directions, respectively. $\Delta \rho_0$, $\Delta \rho_1$, $\Delta \rho_2$ are change of resistivity described as follows:

$$\frac{\Delta \rho_0}{\rho} = -\theta_{SH}^2 \frac{\lambda}{t_N} \tanh \frac{t_N}{2\lambda},$$

$$\frac{\Delta \rho_1}{\rho} = -\theta_{SH}^2 \frac{\lambda}{t_N} \text{Re} \frac{2\lambda G_{t1} \tanh^2 \frac{t_N}{2\lambda}}{\sigma + 2\lambda G_{t1} \cosh \frac{t_N}{2\lambda}},$$

$$\frac{\Delta \rho_2}{\rho} = -\theta_{SH}^2 \frac{\lambda}{t_N} \text{Im} \frac{2\lambda G_{t1} \tanh^2 \frac{t_N}{2\lambda}}{\sigma + 2\lambda G_{t1} \cosh \frac{t_N}{2\lambda}},$$

where $\rho$ and $\sigma$ are the intrinsic electric resistivity and conductivity of the bulk normal metal, $\theta_{SH}$ is the spin Hall angle, $\lambda$ is the spin diffusion length, $t_N$ is the Pt thickness, $G_{t1}$ is the spin mixing conductance at the NM/FM interface, given by

$$G_{t1} = G_t + iG_i$$
Figure 4.3 Schematic diagram of the spin-Hall magnetoresistance. (a) The high resistance state when the magnetization direction is perpendicular to the spin polarization. (b) The low resistance state when the magnetization direction is parallel to the spin polarization. The black, red, green, blue and purple arrows denote the direction of spin polarization, electrical current, spin current, magnetization and magnetic field, respectively.

4.3 Experiments

4.3.1 Sample preparation

The fabrication of the heavy metal/magnetic material heterostructure requires the use of physical deposition techniques, such as pulsed laser deposition, atomic layer deposition and magnetron sputtering, etc. Applying the paramagnetic ionic liquid on top of metal film, however, provides another simple method of fabricating metal/insulator interface.

Electric double layer transistor devices used for PIL gating were all fabricated by standard micro-fabrication. Using electron beam lithography (EBL), we defined the Hall-bar with length $l = 7 \, \mu m$, $w = 3.5 \, \mu m$. All metal channels (Pt, Pd, and Au) were prepared by DC magnetron sputtering (Kurt J. Lesker) after pumping the chamber below $1.0 \times 10^{-8} \, mbar$. Sputtering powers (50 to 200 W) and duration time (2 to 12 s) were optimized for preparing films with various thicknesses.

Separately, contact electrodes comprising bilayer Ti/Au (5/45 nm) were deposited onto the patterned Hall bars using e-beam evaporation (Temescal FC-2000) below $1.0 \times 10^{-6} \, mbar$. Afterwards, an $Al_2O_3$ isolation layer (30 nm) was deposited to cover all contact electrodes, limiting the gating effect only to the exposed channel surface. Figure 4.4 shows a typical device geometry and morphology measured by atomic force microscope.
4.3 Experiments

![Figure 4.4 Bird-eye view of the surface morphology of the Pt thin film (t = 12 nm). The yellow dash line indicates the place where the measurement of the film thickness was performed. The scale bar represents the height of the film.](image)

4.3.2 Electrical measurement method

Low temperature electrical transports were measured in a helium cryostat (PPMS, Quantum Design) under out-of-plane magnetic fields up to 6 T. All transport properties were measured by two lock-in amplifiers (SR830, Stanford Research) using a constant AC current excitation of 50 μA at 13.367 Hz. The voltage bias on BMIM[FeCl₄] (the PIL used in all gating experiments) was applied by a source measure unit (Model 2450, Keithley).

To characterize the magnetic and electrical properties of the devices, we investigated the magnetotransport of PIL-gated Pt with respect to different angle $\phi$ between the applied magnetic field $B$ and the electrical current $I$.

For applied in-plane magnetic field $B$, we performed angular dependent magnetoresistance (ADMR) and field dependent magnetoresistance (FDMR) measurements, where longitudinal and transverse voltages ($V_L$, $V_T$) were monitored simultaneously by passing a constant drain-source current $I_{DS} = 50$ μA.

In ADMR experiment, the device was rotated continuously within the $xy$ plane under a constant $B$ field, where the changes of longitudinal and transverse resistances were measured as a function of azimuthal angle $\phi$ between $B$ and $I$ (Fig. 4.5a). In FDMR experiment, the device was fixed at a particular $\phi$ and resistances were measured with sweeping the magnetic field. The corresponding resistivities were normalized to the dimensions according to:

$$\rho_L = \frac{V_L}{I} \frac{w t}{l},$$

and

$$\rho_T = \frac{V_T}{I} t.$$
Figure 4.5 (a) The in-plane measurement with $B$ field rotating with sample $xy$ plane. The out-of-plane measurement with $B$ field parallel (b) and normal (c) to the current direction $I$. The angles between the $B$ field and $I$ are defined as $\phi$ and $\theta$ for in-plane and out-of-plane measurements, respectively.

For out-of-plane measurement, there are two ways to rotate $B$ field respect to sample $xy$ plane, which are by fixing the $B$ field parallel or normal to the current direction (Fig. 4.5 b, c).

4.3.3 Paramagnetic ionic gating

For inducing the magnetic interaction at the metal/insulator interface, we first need to perform the paramagnetic ionic gating. Platinum (Pt) was studied in this chapter for the reason that it is the most commonly used materials in insulator spintronics as spin injector and detector.

The devices consist of a Pt Hall bar, which is covered by paramagnetic ionic liquid (PIL) as gate (Fig. 4.6). The low end of gate is connected to the source electrode and to the ground. During the gating, electrons were attracted from or depleted to the ground.

Figure 4.7 shows the transfer curve of PIL-gated Pt indicating that the longitudinal sheet resistance $R_L$ can be reversibly controlled by sweeping $V_G$. The red region illustrates the electron accumulation process that is induced by the absorption of cation on the Pt surface, driven by the positive $V_G$.

Figure 4.6 The device configuration for magnetoresistance measurement with paramagnetic ionic liquid as gate.
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4.3.4 Magnetic characterization of the PIL

We measured the magnetic properties of the PILs using a SQUID magnetometer (MPMS XL-7, Quantum Design), where the applied magnetic field ranges from -7 T to 7 T and the temperature from 2 to 350 K.

We measured the magnetization curve of the PIL used for gating at 5 K, namely butylmethylimidazolium tetrachloroferrate (BMIM[FeCl$_4$]). All five $d$-orbitals of Fe$^{3+}$ in the anions are unpaired and in high-spin state, giving a total spin quantum number of $S = 5/2$.

The effective magnetic moment of the anion can be calculated by

$$
\mu_{\text{eff}} = g \sqrt{J(J + 1)} \mu_B,
$$

where $g$ is the Landé g-factor, $J$ is the total angular momentum, and $\mu_B$ is the Bohr magneton, respectively. For 3$d$ metal ions, the crystal field is much stronger than the spin-orbit coupling, leading to so-called orbital quenching effect. Therefore, $\mu_B$ depends only on spin ($L = 0$, $J = S$, $g = 2$), which gives $\mu_{\text{eff}} = 5.92 \mu_B$. The crystal field of the magnetic FeCl$_4^-$ anion is shown in Figure 4.8.

Figure 4.9 shows the magnetization curve ($M$-$B$) of BMIM[FeCl$_4$] up to 7 T at 5 K. No hysteresis loop was observed, indicating there is no long range ferromagnetic ordering within PIL. $M$-$B$ curves show linear dependence despite the slight curvature, implying the paramagnetic (PM) nature. With the increase of the $B$ field, the magnitude of the magnetization increases, as illustrated by the red fitting line (Fig. 4.9). For small $y$, $L(y) = y$, as indicated by the dash line which is tangential to the curve near the origin.
Figure 4.8 The angular distribution of the $d$ orbitals crystal field of FeCl$_4^-$.

We analyze the magnetization curve at 5 K with the Langevin function [40],

$$L(y) = \coth y - \frac{1}{y}, \quad (4.19)$$

where $y$ is defined as

$$y = \frac{\mu_{\text{eff}} B}{k_B T}, \quad (4.20)$$

in which $\mu_{\text{eff}}$ is the effective magnetic moments and $B$ the external magnetic field, $T$ the temperature and $k_B$ Boltzmann constant.

By fitting the $M$-$B$ curve with the Langevin function, we can derive the saturation magnetization $M_s$ and effective magnetic moment $\mu_{\text{eff}}$ per anion based on:

$$\frac{M}{M_s} = \coth \left( \frac{\mu_{\text{eff}} B}{k_B T} \right) - \frac{k_B T}{\mu_{\text{eff}} B}, \quad (4.21)$$

in which,

$$\mu_{\text{eff}} = n \cdot \mu_B. \quad (4.22)$$

In GCS unit, $\mu_B = 9.274 \times 10^{-21}$ erg Gs$^{-1}$, $k_B = 1.38 \times 10^{-16}$ erg K$^{-1}$.

In order to confirm the paramagnetic nature of PIL, we performed temperature dependent magnetic susceptibility $\chi$ measurement. The measurement was carried out with $H = 100$ Oe.

For small $B$ field, applying Taylor expansion to Eq. 4.19, we have

$$\coth(y) = \frac{1}{y} + \frac{y}{3} + O(y^3), \quad (4.23)$$

so that Eq. 4.21 can be approximated to

$$\frac{M}{M_s} \approx L(y) \approx \frac{y}{3} = \frac{\mu_{\text{eff}} B}{3k_B T}. \quad (4.24)$$
At small $B$ field, $\chi \ll 1$, so that

\[ B = \mu_0(1 + \chi)H \approx \mu_0 H, \quad (4.25) \]

and

\[ \chi = \frac{M}{H} \approx \frac{\mu_0 M}{B}, \quad (4.26) \]

By taking Eq. 4.26 into Eq. 4.24., we have

\[ \chi = \frac{N\mu_0\mu_{\text{eff}}^2}{3k_B T}. \quad (4.27) \]

This means the magnetic susceptibility is inversely proportional to the temperature, which is known as the Curie's law.

For a particular material, the product of $\chi$ and $T$ is a constant, which is known as Curie constant:

\[ \chi T = \frac{N\mu_0\mu_{\text{eff}}^2}{3k_B}. \quad (4.28) \]

In CGS unit, $\mu_0 = 1$. Therefore, the effective magnetic moment can be derived according to

\[ \mu_{\text{eff}} = \frac{\sqrt{\frac{3k_B}{N\mu_B^2}}\sqrt{\chi_{\text{mol}} T}}{\sqrt{M_{\text{mol}}} \approx 2.828\sqrt{\chi_{\text{mol}} T}, \quad (4.29) \]

in which

\[ \chi_{\text{mol}} = \frac{M M_{\text{mol}}}{mH}, \quad (4.30) \]

where $m = 3.4$ mg, $M_{\text{mol}}(\text{BMIM}[\text{FeCl}_4]) = 336.87$ g mol$^{-1}$, $\mu_B = 9.724 \times 10^{-21}$ ergG$^{-1}$, $N_A = 6.02 \times 10^{23}$. 

**Figure 4.9** Magnetization curve of classical paramagnetic PIL measured at 5 K. The red line is the fitting described by the Langevin function. The black dash line is tangential to the curve near the origin.
The measured magnetic susceptibilities identify BMIM[FeCl₄] as a paramagnet with a large effective magnetic moment of \( \mu_{\text{eff}} = 5.77 \mu_B \) (Fig. 4.10a), which agrees well with the theoretical value of \( \mu_{\text{eff}} = 5.92 \mu_B \). The PIL remains paramagnetic down to 2 K, the lowest temperature considered. The weak antiferromagnetic interaction observed in terms of \( \chi T \) that deviated from Eq. 4.27 (Fig. 4.10b) is consistent with other reports [41, 42].

Similarly, in Figure 4.9, we fit the magnetization data at 5 K by Eq. 4.24 and extract \( M_s \) to be 5.82 \( \mu_B \). At 6 T, magnetic moments are not fully aligned and \( M \) is equal to 3.62 \( \mu_B \), slightly lower than the theoretical value, which agrees with the suppressed Fe moments at the lowest temperatures from susceptibility measurement (Fig. 4.10b).

In contrast to the ferromagnetic insulator \( \text{Y}_3\text{Fe}_5\text{O}_{12} \) (YIG), which is a ferrimagnetic insulator that possesses long-range magnetic ordering3,4; our results prove that PIL is paramagnetic. Hence, we may exclude a ferromagnetic proximity effect5,6 at the Pt|PIL interface.

### 4.4 Results and Discussion

#### 4.4.1 Temperature dependence of longitudinal resistance

A whole cycle of electrical transport measurement normally consists of five consequent steps (Fig. 4.11). The details of each step will be explained as follows.

**Step 1:** The paramagnetic ionic gating was performed at 220 K, which is slightly higher than the frozen temperature \( T_m \) of BMIM[FeCl₄]. The temperature was chosen based on two considerations. First, the temperature should be as low as possible in order to minimize possible chemical reaction. Second, the ionic mobility
of the PIL should be high enough so that it is possible to move the ions with electric bias voltage $V_G$.

**Step 2:** After observing an apparent $R_s$ drop (Fig. 4.11), the device was immediately cooled down rapidly to 180 K while keeping $V_G$. This is for the purpose of freezing the PIL as well as the gate induced electronics state of Pt. Afterward, the device was cooled down to 5 K at 2 K min$^{-1}$.

**Step 3:** The PIL-gated state of Pt is ferromagnetically polarized at surface. In chapter 2, we have discussed extensively the measurement of PIL gating induced two-dimensional ferromagnetic state, which we denote as ON in Figure 4.11. After the measurements, the sample was warmed up to 260 K with $V_G = 0$ V. In principle, $V_G$ can be retracted anytime below $T_m$, because once the ions are frozen, $V_G$ plays no role in terms of forming electric double layer.

**Step 4:** Above 200 K, $R_s$ slowly recovered revealing the retraction of the gating effect, which is due to PIL melting above its $T_m$. Those ions that were driven to the Pt surface by $V_G$ were relaxed back to their equilibrium position. This process was finished above $\sim$230 K, as the slope of the $R_s$-$T$ curve became the same as the part below 200 K.

**Step 5:** After reaching 250 K for fully ionic relaxation, the sample was cooled down to 5 K again. The $R_s$ at 220 K had exact the same value as the one before gating, suggesting that there was no sample deterioration and the Pt film had an identical electronic state. Consequently, there is no effect at low temperature, denoted as OFF state in Figure 4.11.

**Figure 4.11** Temperature dependence of sheet resistance $R_s$ for five consecutive sequences: gating (1), first cooling down with $V_G$ (2), warming up without $V_G$ (3), melting of PIL (4) and second cooling down without $V_G$ (5). The ON and OFF states correlate with the transfer curve as well as the ADMR measurements.
4.4.2 Modulation of the resistivities

With in-plane $B$ field geometry, we analyzed the states with and without $V_G$ at low temperature by performing the ADMR measurement.

The ADMR measurements were carried out at 5 K after temperature was stable. When the sample was cooled down with $V_G$ as indicated as step 2 in Figure 4.11, we found that both longitudinal and transverse resistivities ($\rho_L$ and $\rho_T$, respectively) showed modulations depending on $\phi$, denoted as ON state (Fig. 4.12). This implies an anisotropy effect that external magnetic field acting on the current, resulting differences in the probed voltages. When we released the gated state by warming up the sample up to PIL melting temperature and cooled down again without $V_G$, the modulation of resistivities vanished, showing no $\phi$ dependence on the transport and denoted as OFF state.

The direct correlation between the magnetoresistivities with the ON and OFF states in the transfer curve indicates that the observed effect is induced by paramagnetic ionic gating.

![Figure 4.12](image)

**Figure 4.12** Longitudinal (a) and transverse (b) resistance changes as a function of the angle $\phi$ between the current and magnetic field direction. The red and green curves represent the gate ON and OFF state.

4.4.3 Angle dependence of magnetoresistivity

To further investigate the observed magnetoresistivity modulation, we performed the field-dependent ADMR measurements at 5 K, where the longitudinal and transverse voltages ($V_L$ and $V_T$, respectively) were measured simultaneously by rotating the angle $\phi$ at various fixed $B$ from 0.5 T to 6 T.

Figure 4.13a shows the results of $\rho_L$ under various $B$ fields. All measurements display similar resistivity modulations in a $\cos^2 \phi$ dependence with a period of $\pi$, where the maximum and minimum values are observed for $B \parallel I$ and $B \perp I$, denoted as $\rho_\parallel$ and $\rho_\perp$, respectively. In the meantime, a $\sin 2\phi$ dependence with a period of $\pi$ was observed for the transverse resistivity $\rho_T$, where the maximum
and minimum appear at $\phi = 45^\circ$ and $\phi = 135^\circ$, respectively (Fig. 4.13b). At $\phi = 0$, $\rho_\parallel$ remains the same for all measured $B$ field. The amplitude of $\Delta \rho_L$ is determined by the difference of $\rho_L$ between $\phi = 0^\circ$ and $\phi = 90^\circ$, while the amplitude of $\Delta \rho_T$ is determined by the difference of $\rho_T$ between $\phi = 45^\circ$ and $\phi = 135^\circ$.

This behaves similar to the AMR effect and the corresponding PHE of ferromagnets, where $\rho_L$ and off-diagonal terms in the resistivity tensor $\rho_T$ are modulated by the angle $\phi$ between $I$ and $M$, given by

$$\rho_L(\phi) = \rho_L + \Delta \rho_L \cos^2 \phi \tag{4.31}$$
$$\rho_T(\phi) = \Delta \rho_T \sin \phi \cos \phi \tag{4.32}$$

where $\Delta \rho_L = \rho_\parallel - \rho_\perp$ and $\Delta \rho_T = \rho(\phi=45^\circ) - \rho(\phi=135^\circ)$.

Compared with AMR, despite of the similarity in shape, the amplitudes of present system differ significantly from AMR as a function of $B$ field. In the AMR system, $\Delta \rho_L$ ($\Delta \rho_T$) becomes a constant after $M$ saturates at $B$ larger than coercivity $H_c$ and shows a nonzero value at $B = 0$ T due to the finite remanence; while in the present system, $\Delta \rho_L$ ($\Delta \rho_T$) is zero without $B$ and the value increases with enlarging $B$, showing no hint of saturation.

![Figure 4.13](image_url)

**Figure 4.13** In-plane magnetic field dependence of the evolution of the longitudinal resistivity $\rho_L$ (a) and transverse resistivity $\rho_T$ (b) as a function of angle $\phi$, where $\phi$ is the angle between magnetic field $B$ and current direction $I$. All measurements were performed at 5 K.
Because of the highly insulating nature of the PIL, we can safely exclude any charge transfer contribution that comes from the PIL. In addition, the strong screening of the induced carrier by ionic gating confines the effect at the Pt/PIL interface. This raises the question: what is the reason causing this peculiar effect?

In section 4.3.4, we have studied the magnetic properties of the ionic gating medium we use in this paper. It is a low-melting-temperature ionic liquid butylmethylimidazolium tetrachloroferrate (BMIM[FeCl₄]), where all five d-orbitals of the Fe³⁺ in the anions are unpaired, giving high-spin state with total spin quantum number $S = \frac{5}{2}$. Magnetic susceptibility measurement shows that BMIM[FeCl₄] is paramagnetic with an effective magnetic moment $\mu_{\text{eff}} = 5.77 \mu_B$ derived from fitting to Curie’s law, which agrees well with the theoretical effective magnetic moment $\mu_{\text{eff}} = 5.92 \mu_B$. The PM nature of the BMIM[FeCl₄] is further demonstrated by magnetization measurement at 5 K and is described with the Langevin function

$$\frac{M}{M_s} = \coth\left(\frac{\mu_{\text{eff}} B}{k_B T}\right) - \frac{k_B T}{\mu_{\text{eff}} B}, \quad (4.33)$$

where $M_s$ is the saturation magnetization, $\mu_{\text{eff}}$ the effective magnetic moments and $B$ the external magnetic field, $T$ the temperature and $k_B$ Boltzmann constant.

Notably, the observed non-saturating behaviors for both $\rho_L$ (green) and $\rho_T$ (red) from ADMR measurement scale nicely with the measured magnetization curve of BMIM[FeCl₄] (blue) (Fig. 4.14). This correlation suggests that the paramagnetic ionic liquid is responsible for the observed non-saturating MR.

An alternative explanation can be given by adapting the Stoner-Wohlfarth model (which we will discuss explicitly in the chapter 5) to the present system with a fixed in-plane magnetic field. In Figure 4.15a, we show the schematic diagram

**Figure 4.14** Correlation between the magnetization of the BMIM[FeCl₄] and in-plane magnetoresistivity of the Pt thin film at $T = 5$ K. The magnetization curve of the BMIM[FeCl₄] (in blue) was measured by SQUID magnetometer.
4.4 Results and discussion

Illustrating directions of current \( I \), magnetic field \( H \), magnetization \( M \) and magnetic easy axis with respect to the Pt film. We consider the magnetocrystalline anisotropy term \( K \sin^2 \theta \), where \( K \) is the anisotropy constant; and the Zeeman energy, given by \( M_s H \cos(\gamma - \theta) \), in which \( \theta \) (\( \gamma \)) are the angles between \( M \) (\( H \)) and the magnetic easy axis, respectively.

Applying an in-plane \( H \) field to the gated film will gradually pull down the magnetization into the plane. For thin films, there is addition shape anisotropy leads to an energetic saving for keeping the magnetization in the plane of the film, which can be described by \( \frac{1}{2} \mu_0 M_s^2 \cos^2 \beta \), where \( \beta \) is the angle between the film normal and \( M \), \( \mu_0 \) the vacuum permeability. Therefore, the total energy \( E \) to be considered is as follow:

\[
\frac{1}{2} \mu_0 M_s^2 \cos^2 \beta
\]

\( \beta \) is the angle between the film normal and \( M \), \( \mu_0 \) the vacuum permeability. Therefore, the total energy \( E \) to be considered is as follow:

**Figure 4.15** Numerical modeling of the magnetic field dependence of the in-plane magnetization based on the Stoner-Wohlfarth model. (a) Schematic illustration of angles with respect to the film plane. \( H \), \( M \) denote the external magnetic field and magnetization directions. \( a \), \( b \), \( c \) are the three axes of the Cartesian coordinate system. \( \gamma \), \( \theta \) and \( \phi \) are the angle between \( H \) and easy axis, \( M \) and easy axis, current \( I \) and \( H \), respectively. (b) The angle \( \theta \) dependent in-plane magnetization \( M_{||} \) as a function of \( H \). (c) The correlation between \( M_{||} \) and the in-plane magnetoresistivities \( \rho_L \) and \( \rho_T \). (d) The influence of the shape anisotropy on \( M_{||} \) for \( \theta = 75^\circ \), where \( x \) represents the ratio of the shape anisotropy compared to the magnetocrystalline anisotropy.
\[ E = K \sin^2 \theta - M_s H \cos(\gamma - \theta) + \frac{1}{2} \mu_0 M_s^2 \cos^2(90^\circ - \gamma + \theta), \] (4.34)

where the first term denotes the magnetocrystalline anisotropic energy and the second term refers to the magnetostatic potential (Zeeman) energy, and the last term indicates the shape anisotropy energy.

The angle \( \theta \) is determined by magnetocrystalline anisotropy energy of the FM Pt layer under equilibrium states, where:

\[ \frac{\partial E}{\partial \theta} = 0, \] (4.35)

and

\[ \frac{\partial^2 E}{\partial \theta^2} > 0. \] (4.36)

By taking into account

\[ h = \frac{H}{H_s}, \] (4.37)

and

\[ H_s = \frac{2K}{M_s}, \] (4.38)

Then, Eq. 4.35 can be simplified to

\[ E = 2K \left[ \frac{1}{2} \sin^2 \theta - h \cos(\gamma - \theta) + \frac{M_s^2}{4K} \cos^2(90^\circ - \gamma + \theta) \right], \] (4.39)

where we define

\[ x = \frac{M_s^2}{4K}, \] (4.40)

which represents the degree of magnitude that in-plane shape anisotropy has on the out-of-plane magnetocrystalline anisotropy.

Figure 4.15 b and d show the numerical modeling results using Matlab. Without shape anisotropy \( (x = 0) \), the dependence of possible directions of easy axis with respect to the in-plane \( H \) is shown in Figure 4.15b. The in-plane projection of the magnetization has a finite value when the magnetocrystalline anisotropy of system is not perfectly perpendicular. On the other hand, including the effect of shape anisotropy further shifts the saturation field \( H_s \) to lower bound (Fig. 4.15d).

### 4.4.4 Field dependence of magnetoresistivity

In order to confirm the correlation between \( \Delta \rho_L (\Delta \rho_T) \) and \( M \) of PIL, we performed the FDMR measurements at the same temperature \( (T = 5 \text{ K}) \) at various angle \( \phi \). At \( \phi = 90^\circ \), where \( B \) is applied within the sample \( xy \) plane and perpendicular to the direction of \( I \), we observe a negative non-saturating MR with increase of \( B \) (Fig. 4.15). At \( \phi = 0^\circ \), i.e. \( B \) is parallel to the direction of \( I \), however, we observed no change of MR with increasing \( B \) (Fig. 4.16).
4.4 Results and discussion

Figure 4.16 In-plane field dependent magnetoresistivity for different $\phi$ measured at 5 K. (a) Longitudinal MR shows negative $\Delta \rho_L$ for $\phi = 90^\circ$ and no significant change for $\phi = 0^\circ$. (b) Transverse MR shows negative for $\phi \in (0, 90^\circ)$ and positive for $\phi \in (-90^\circ, 0^\circ)$. The black dash lines indicate the references at $B = 0$ T, which are the same for all measurements. The color dot lines show the theoretical values of $\Delta \rho_L$ and $\Delta \rho_T$, which are close to the $\rho_L$ and $\rho_T$ at $B = 6$ T, demonstrating the consistency with the ADMR measurement.

Figure 4.17 Reconstructed angular dependent magnetoresistance from the FDMR measurement.

By extracting the values of $\rho_L$ and $\rho_T$ for each $\phi$ at various $B$ and summarizing them as a function $\phi$ in the same figure, we are able to fully reconstruct the profile of ADMR measurement, where $\cos^2 \phi$ and $\sin 2\phi$ dependence with a period of $\pi$ are shown in Figure 4.17a and Figure 4.17b, respectively. It further demonstrates the consistency of these two independent experiments.

Table 4.1 Longitudinal saturation resistances $\rho_L$ derived from the fitting to Langevin function at each $\phi$ (unit: $\mu\Omega$ cm)

<table>
<thead>
<tr>
<th>$\phi$ (°)</th>
<th>90</th>
<th>60</th>
<th>45</th>
<th>30</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experimental</td>
<td>22.81</td>
<td>17.96</td>
<td>12.86</td>
<td>7.16</td>
<td>-2.5</td>
</tr>
<tr>
<td>Theoretical</td>
<td>23.16</td>
<td>17.74</td>
<td>13.28</td>
<td>7.92</td>
<td>-4.02</td>
</tr>
</tbody>
</table>
Figure 4.18 Longitudinal magnetoresistivity at 5 K fitted by Langevin equation.

For further demonstration of the paramagnetism related MR in PIL-gated Pt, we fitted the FDMR data for each individual angle $\phi$ from 90° to 0° with the Langevin function (Eq. 4.33), shown as Figure 4.18. The prefactor $\mu_{\text{eff}}/k_B T$ was fixed the same 0.468 as the result from fitting of magnetization curve for PIL, and longitudinal saturation resistances $\rho_L$ at each $\phi$ can be derived hereafter.

Figure 4.19 Schematic in-plane MR of the Pt|PIL interface. (a) Angular-dependent longitudinal MR with perpendicular magnetic anisotropy for various B fields. (b) Field-dependent longitudinal MR for $\phi = 0^\circ$ and 90°. (c) Angular-dependent transverse MR (planar Hall effect) with perpendicular magnetic anisotropy for various B fields. (d) Field-dependent transverse MR for $\phi = 45^\circ$, 90° and 135°.
4.4 Results and discussion

Considering there is a little error of measured angle from the real value causing by the backlash, we vary the nominal (read out from the rotator) by 10°, which is a reasonable approximation that is often seen from the ADMR measurement, and calculate the theoretical values comparing with the experimental values. The result is summarized in Table 4.1.

In literatures, a negative non-saturating MR has been reported in granular ferromagnets [43, 44], which is caused by spin-dependent scattering at the boundaries of nonaligned magnetic grains. Depending on either metallic or dielectric matrices, the resulted decrease of MR with increasing $B$ can be interpreted as GMR or TMR mechanism due to the alignment of magnetic moments. Below its percolation threshold, the granular ferromagnet is isotropic regardless of the direction of $I$ to $B$. However, the present system shows a significant difference between $B \parallel I$ and $B \perp I$. Therefore, further investigation is necessary for understanding the mechanism behind. Overall, the effects are summarized in Figure 4.19.

4.4.5 Temperature-dependent in-plane magnetoresistivity

The influence of thermal energy is studied by performing the temperature-evolved FDMR experiment at $\phi = 45°$. We chose this angle on purpose, because at this $\phi$ both longitudinal ($\rho_L$) and transverse magnetoresistivity ($\rho_T$) show considerable large non-zero field dependence (Fig. 4.13).

The magnitude of $\delta \rho_L = \rho_L(B) - \rho_L(B=0)$ at $\phi = 45°$ is $\sqrt{2}/2$ time of $\delta \rho_L$ at $\phi = 90°$ that is equal to the amplitude of $\Delta \rho_L = \rho_{||} - \rho_L$ in ADMR measurement. $\delta \rho_T = \rho_T(B) - \rho_T(B=0)$ at $\phi = 45°$ is the same as $\Delta \rho_T$ from ADMR measurement.

The signs of both $\rho_L$ and $\rho_T$ at 5 K were consistent with the AMR, which is negative for $\rho_L$ and positive for $\rho_T$. In order to understand the physical origin of the observed effect and its relation with AMR, we performed the identical FDMR experiment at various temperatures upwards. The angle $\phi$ was fixed at 45° during all the measurements for the reason that at this angle both $\rho_L$ and $\rho_T$ have.

The results are shown in Figure 4.20. We noticed that the sign of $\rho_T$ remained positive and the amplitude was roughly the same. In contrast, the amplitude of $\rho_L$ first decreased with increasing temperature, and continued to increase after sign reversal at $\sim 40$ K. This is quite extraordinary because it implies that the high temperature behavior is not consistent with AMR theorem anymore and even the low temperature behavior is of the other origin.
Figure 4.20 Temperature-dependent magnetoresistivity as a function of in-plane $B$. (a) Longitudinal magnetoresistivity $\rho_L$ shows a sign-reversal at $\sim 40$ K. (b) The sign of the transverse magnetoresistivity remains the same for all temperatures.

The absolute amplitude of $\rho_L$ and $\rho_T$ was equivalent at their maximum value, which was 5 K and 120 K. We did not measure at temperature higher than 160 K for the reason that the PIL-gated state can only exist below the melting temperature of the PIL.

In contrast to the pristine state of Pt, a positive MR with similar magnitude as the low temperature negative MR burgeons at $T > 40$ K for both in-plane and out-of-plane $B$ configurations (Fig. 4.20a, Fig. 4.23a). At this moment, we do not fully understand its origin. We temporarily refer it as unidentified magnetoresistance (UIMR) in the following paragraphs.
4.4 Results and discussion

**Figure 4.21** Schematic illustrations of the temperature-dependent in-plane MR. (a)–(c) and (d)–(f) represent the longitudinal and transverse MR, respectively. (a)–(d), (b)–(e) and (c)–(f) are at low, intermediate and high temperature, respectively. The changes of the measured MR are illustrated by the blue arrows. The magnitudes of the UPMR and SMR are indicated by green and red bars.

Now we try to explain the sign reversal phenomena in the present system. For the in-plane configuration at \( \phi = 45° \), the measured MR (blue arrows or dots in Fig. 4.21) is comprised of two contributions: the UIMR and the interface SMR. At low temperatures, the strong PMA dominates and causes the negative MR (Fig. 4.21a). With increasing temperature, both the weakening PMA and the additional UIMR shift the ADMR modulation to higher values. At \( \sim 40 \) K, the in-plane transport properties resembles the conventional AMR or SMR type of behavior with vanishing MR at \( \phi = 45° \) (Fig. 4.21b). Further increasing the temperature, the UIMR eventually dominates the ADMR modulation, and we measure a positive MR (Fig. 4.21c). The UIMR also participates in transverse transport. Since the \( \delta \rho_T \) at \( \phi = 45° \) is already positive at low temperatures (Fig. 4.21d), its increase does not change the sign. Moreover, the magnitude of the interface SMR decreases at higher temperatures, which partly compensates the UIMR, resulting in a roughly temperature-dependent result with relative similar magnitudes (Fig. 4.21e,f).
4.4.6 Temperature-dependent out-of-plane magnetoresistivity

For the reason of figuring out the mechanism of sign reversal observed for in-plane measurement $\delta \rho_L$, control experiment with out-of-plane geometry has been carried out, in which $B$ is applied perpendicular to the sample $xy$ plane and normal to the direction of $I$.

Pt device with $t = 16$ nm was gated positively to the same state when $R_s$ saturates. FDMR measurement shows the magnitude of a longitudinal magnetoresistance with perpendicular $B$ field growing $\delta \rho_p$ with the increase of temperature (Fig. 4.22a). This seems not to be consistent with the in-plane longitudinal results. However, it is noteworthy to mention that in different from in-plane FDMR measurements that $\delta \rho_L$ and $\delta \rho_T$ were zero at OFF state, pristine non-gated Pt film under out-of-plane $B$ field also showed positive MR especially at low temperature (Fig. 4.22b). Considering the fact that gating effect only happens at PIL/Pt interface, while the bulk of Pt film is screened out, additional data processing is necessary in order to obtain the absolute interface contribution.

Pt is a heavy metal with strong SOI, so quantum corrections to the electrical conductivity result in weak anti-localization and a positive MR [45]. On the other hand, the FM surface of Pt causes a negative MR due to the $s-d$ scattering [46]. Assuming that the MR is caused by the sum of the interface and the bulk scatterings (Matthiessen’s rule) $1/\tau = 1/\tau_{in} + 1/\tau_b$, the interface contribution $\rho_P$ (Fig. 4.22c) can be qualitatively extracted by subtracting the MR data measured for the gated state (Fig. 4.22b) from ones under the pristine state (Fig. 4.22a), in which the bulk Pt remains unchanged by PIL gating. On the other hand, for the transverse resistivity, the bulk contribution will not affect the AHE part, so that we show the $\rho_H$ as measured (Fig. 4.23b).

We found that the net surface contribution under out-of-plane geometry after extracting the bulk contribution looks similar to the in-plane magnetoresistivity results (Fig. 4.20a). It showed a sign-reversal at roughly the same temperature ($\sim 40$ K) and the magnitude of the $\delta \rho_p$ at 5 K and 120 K were also close in absolute value. This observation indicates that for longitudinal transport, as long as $B \perp I$, whatever $B$ is in the sample $xy$ plane or normal to it, the transport behaviors follow the same rules.

The sign-reversal in $\delta \rho_L$ with out-of-plane $B$ fields cannot be explained so easily. First, the interface and bulk contributions to the observed signals MR = $MR_{in} + MR_b$ must be disentangled. In Figure 4.22c the bulk contribution has been subtracted, which leaves three effects affecting the interface MR, i.e. the FM topmost Pt layer (FTMR), the interface SMR and the UIMR. Assuming that the FM Pt layer is a granular, multi-domain ferromagnet [43, 44], the negative MR can be interpreted by spin-dependent scattering at the boundaries of nonaligned magnetic grains, which is isotropic and independent of the relative directions of $I$ and $B$ [43].
Figure 4.22 Temperature dependence longitudinal magnetoresistivity $\rho_P$ under out-of-plane external magnetic field. (a) $\rho_P$ of pristine Pt without gating. (b) $\rho_P$ after PIL gating. (c) Changes of $\rho_P$ of the surface where the PIL gating effect occurs.

The interface SMR, on the other hand, is anisotropic and gives a positive correction to the resistivity with larger M coming from the combination effect of the alignment of the PM ions in the PIL under $B$ field and the FM Pt layer. The UIMR is also believed to be related to M, but causes a positive MR at higher temperature.

To sum up, the overall M of the system under a particular B field is suppressed by temperature, which weakens the FTMR and SMR and enhance the UIMR. The competition between these three effects exhibits a crossover from the negative MR at low temperatures to the positive MR at high temperature that is closely related to the variation of the B field-induced M of the interface.

### 4.4.7 Temperature-dependent anomalous Hall effect

Apart from observing similar sign-reversal in $R_p$ (Fig. 4.23a), similar phenomenon also appeared in the transverse voltage buildup $R_H$ that was perpendicular to both
B and I, namely the anomalous Hall effect (AHE). $R_H$ displays clear hysteresis loop at 5 K (Fig. 4.23b), indicating the emergence of a ferromagnetic (FM) state, which shows a perpendicular magnetic anisotropy (Section 3.4.2).

The AHE can be described by [47]

$$\rho_{xy} = \rho_0 + \rho_H = R_0 B + R_A \mu_0 M,$$

where $R_0$ and $R_A$ are the ordinary and anomalous Hall coefficient, respectively. For the whole temperature range from 5 K to 120 K, the hysteresis diminished rapidly, and the sign of the second term in Eq. 4.41 changed at ~60 K (Fig. 4.22), which can be caused by either $R_A$ or $M$.

The theory of AHE sign-reversal is profound despite that it has been observed in many materials experimentally [48-50]. In the present system, since the ordinal Hall effect remains electron-like, the sign reversal of the AHE cannot be ascribed to the global change of the Fermi surface topology.

Skew scattering [51] and side jump [52] are two mechanisms proposed to explain AHE. The former one dominates in the good-metal regime with low resistivity ($< 1 \mu\Omega \text{ cm}$) [53], while the later one is sensitive to the impurity type and concentration. In contrast, we obtained $\rho_L$ as high as $10^2 \mu\Omega \text{ cm}$, which excludes skew scattering as the origin accounting for AHE sign reversal [53].

![Figure 4.23](image)

**Figure 4.23** Temperature-dependent longitudinal magnetoresistance $R_L$ (a) and transverse Hall resistance $R_H$ (b) with out-of-plane $B$ field. It is worth noting that $R_H$ shows the emergence of anomalous Hall effect and hysteresis loop at 5 K, indicating the existence of ferromagnetic state.
Furthermore, in our experiment, we use the same PIL with Fe$^{3+}$ as spin dopants and control the concentration by gating to roughly the same voltage when $R_s$ saturates [54]. The sign of AHE reverses only at Pt films above certain thicknesses, which is difficult to reconcile with the side-jump mechanisms. It is worth noting that the AHE sign-reversal temperature is close to those found in longitudinal transport $\delta\rho_L$ and $\delta\rho_p$, implying a common physics background.

### 4.4.8 Magnetic phase diagram

We summarized all transport results of the PIL-gated Pt system in a magnetic phase diagram shown in Figure 4.24. For thinner films with $t = 8$ nm and $t = 12$ nm, $\delta\rho_H$ remains positive for all the measured temperature range. Because we found that PIL-gated Pt shows FM state at low temperature (in chapter 2), the saturation magnetization was determined by extrapolating the linear part of $\rho_{xy}$ at large $B$ field ($\rho_0$ in Eq. 4.41) and fitted the high temperature data ($T \geq 40$ K) with the Bloch equation [55]:

$$\delta\rho_H = \delta\rho_H^0 (1 - CT^\beta),$$

where $\delta\rho_H^0$ is the spontaneous magnetization at $T = 0$ K and $\beta$, the Bloch constant, equals 3/2 for three-dimensional (3D) system.

![Figure 4.24 Phase diagram of the PIL-gated Pt films. The red squares, green triangles, blue triangles and purple diamonds denote the in-plane longitudinal ($\delta\rho_L$), transverse magnetoresistivity ($\delta\rho_T$), and perpendicular magnetoresistivity ($\delta\rho_p$) and Hall resistivity ($\delta\rho_H$) of 16 nm Pt film, respectively, where $\delta\rho_i = \rho_i(6T) - \rho_i(0T)$. The orange circles and brown hexagons represent the Hall resistivities of 8 nm and 12 nm Pt samples. A sign reversal was observed in 16 nm sample for $\delta\rho_L$, $\delta\rho_p$ and $\delta\rho_H$ at roughly 40 K, while the sign of $\delta\rho_T$ remains the same. In thinner samples, a prominent up-turn feature was observed at low temperature. This deviation from the Bloch law suggests the onset of a paramagnetic contribution to the overall magnetization.](image)
Interestingly, the observed $\rho_H$ deviates from $T^{3/2}$ dependence at low temperature. Instead, it increases with $1/T$, which belongs to the Curie law for paramagnetism. This low temperature anomaly suggests that the PM contribution of the PIL becomes comparably large at low temperature on top of the FM interaction revealed by AHE. In addition, the temperature dependence of $\rho_H$ reveals the nature of the paramagnetism correlated MR from another point of view.

For thicker sample ($t = 16$ nm), besides the low temperature Curie type of up-turn feature, we observed the reversal behavior of the magnetoresistivity sign. For the longitudinal transport with $B \perp I$, both in-plane ($\delta\rho_L$) and out-of-plane ($\delta\rho_P$) geometry shows a similar sign-reversal temperature $\sim 40$ K, while the transverse behavior is more complicated. In contrast to thinner films, the sign of AHE ($\delta\rho_H$) reverses at $\sim 60$ K and magnitude continues to grow with increasing temperature.

In addition, in terms of the in-plane transverse magnetoresistivity ($\delta\rho_T$), both sign and magnitude hold for all the measured temperature range. This inconsistency implies there might be two mechanisms affecting the transverse transport [56]. We tentatively assign the AHE to the FM contribution of FM Pt layer and the low temperature PHE-like MR to the PM contribution of PIL.

4.4.9 Theory of spin-dependent magnetoresistance at Pt/paramagnetic insulator interface

Because of the highly insulating nature of the PIL that no electrical current can pass through ($I_G < 10^{-12}$ A at 5 K), resistivity mixing of PIL contribution is safely excluded. In addition, the formation of electric double layer under bias $V_G$ generates an extremely large electric potential ($E = V/d$) due to the very short capacitive distance, which is equal to the radius of PIL ions. The electric field attenuates rapidly with deep into the bulk of the channel, limiting the gating effect to the top-most layer of the Pt [57, 58]. Together with strong screening, paramagnetic ionic gating forms an interface with property distinct from the rest of the Pt. We have discussed this induced interface ferromagnetism with perpendicular magnetic anisotropy in chapter 3.

Moreover, since Pt is a heavy metal with strong spin-orbit interaction, large spin Hall effect (SHE) is expected that converts electrical current $I$ to a spin current $I_s$, leading to a spin accumulation $\mu_s$ towards the Pt/PIL interface with its direction normal to both $I$ and $I_s$. Depending on the magnetization direction, $\mu_s$ will be absorbed by the magnetization forming spin-transfer torque; or reflected back to the Pt channel. Because of the inverse spin Hall effect (ISHE), these reflected spins will convert into an electrical current again. Therefore, we attempt to interpret the observed phenomenon by the well-established spin-Hall magnetoresistance (SMR) theory with intrinsic perpendicular magnetization [29, 39].

In Figure 4.25b, when $B = 0$ T, the direction of $M$ at the FM Pt top-layer is out-of-plane and normal to the spin polarization $\sigma$ direction of $I_s$ coming from bulk
PM Pt. The absorption of $\mu_s$ by $M$ is therefore the strongest, leading to the high resistance state of $\rho_L$. Increasing $B$ field gradually pulls down $M$ to the in-plane direction. At $\phi = 0^\circ$, $\sigma$ keeps normal to $M$, so that the absorption of $\mu$ by $M$ remains constant, keeping $\rho_L$ at high resistance state (Fig. 4.25c).

At $\phi = 90^\circ$, however, the angle between $M$ and $\sigma$ changes from $90^\circ$ to $0^\circ$ with increasing $B$. This will drive the influence of $M$ on $\mu$ from maximum absorption to effective reflection, resulting in the decrease of the $\rho_L$ (Fig. 4.25d). We expect the hybridization of the magnetization from the PIL molecules and the FM Pt top-layer, which forms localized spin-ordered states at the Pt/PIL interface [9, 59-61]. Therefore, with further increase of $B$ field after magnetic moments of FM Pt have been fully aligned, $\rho_L$ continues to grow because of the PM contribution of PIL.

In contrast, for systems with spontaneous in-plane $M$, such as Pt/yttrium iron garnet (YIG) bilayer structure, decreasing $B$ to the amount that cannot hold the direction of $M$ will trigger the in-plane precession with a finite remanence through all angle $\phi$, which results in $\rho_L$ changing from the maximum to the minimum and back (Fig. S4.3c) [62]. In addition, due to the finite coercivity of YIG, $\rho_L$ becomes constant after $M$ is fully aligned to the direction of $B$, which is significantly different from the non-saturating PM feature of PIL.

Figure 4.25 Theory of the spin-Hall magnetoresistance at Pt/PIL interface with paramagnetic ionic gating. (a) Without PIL gating, Pt shows no resistivity modulation with respect to in-plane $B$ field. (b) After gated with PIL, surface Pt becomes ferromagnetic with perpendicular anisotropy. With applying an in-plane $B$ field, the system is in high (c) and low (d) resistance state with respect to $\phi = 0^\circ$ and $\phi = 90^\circ$, respectively.
4.5 Supplementary Information

4.5.1 Two-channel model of electrical transport with out-of-plane geometry

The short Thomas-Fermi screening length due to the intrinsically large carrier density in Pt limits the gating effect on to the top-most atomic layer [63], which leaves the bulk remain pristine.

In chapter 3, we discussed the coexistence of the Kondo effect in the presence of the ferromagnetism with the two-channel model. In this chapter, we observed the sign reversal signature of the Hall coefficient at \( \sim 60 \) K. However, the apparent longitudinal transport shows no sign reversal, presumably because of the mixing of the bulk-conducting channel. To find out the correlation between the transverse Hall signal and the surface contribution of the longitudinal magnetoresistance, we apply the two-channel model to separate the surface contribution from the overall experimental results.

Consider a two-channel in parallel model consisting of a surface gated layer that is more conducting and a bulk non-gated layer that has the normal resistance, we have

\[
\frac{1}{R} = \frac{1}{R_s} + \frac{1}{R_b}. \tag{4.43}
\]

Take \( \rho = Rt \) into equation (14), we have

\[
\frac{t_s + t_b}{\rho} = \frac{t_s}{\rho_s} + \frac{t_b}{\rho_b}, \tag{4.44}
\]

where \( t_s, t_b \) are the effective thickness of the surface gated Pt layer and the bulk non-gated layer, respectively. \( \rho_s, \rho_b \) and \( \rho \) are the surface, bulk and overall resistivities of the channels while \( \rho_b \) and \( \rho \) are obtained from the measurements without (Fig. 4.22a) and with PIL gating (Fig. 4.22b), respectively. Eq. 4.44 can be further converted to

\[
\rho_s = \frac{\rho_b t_s}{\rho_b (t_s + t_b) - \rho t_b}. \tag{4.45}
\]

We investigate \( \delta \rho_p = \rho_s(6T) - \rho_s(0T) \) and plot the evolution of \( \delta \rho_p \) with temperature \( T \) in Figure S4.1. The temperature dependent \( \rho_s(0T) \) can be drawn from analyzing the data from Figure 4.22 into Eq. 4.45. By plotting \( \rho_s(0T) \) versus \( T \), we are able to reconstruct the temperature dependence of sheet resistivity curve of Figure 4.11.
4.5 Supplementary information

Figure S4.1 Simulation of the sign reversal of the longitudinal magnetoresistivity \( \delta \rho_p \), given by \( \delta \rho_p = \rho_s(6T) - \rho_s(0T) \). The legend represents the nominal surface thickness \( t_s \) of the effective gated Pt layer. It is clearly seen that the sign of \( \delta \rho_p \) changes at 25 K, which is close to the experimental value 30 K.

Figure S4.2 Simulated temperature dependent resistivity curves for various surface layer thicknesses (labeled on the right). The black one indicates the \( R-T \) curve of pristine non-gated Pt film, which is consistent with Figure 4.11. The blue one with a resistivity change of \( \sim 4\% \) is correlated with the gating effect observed in transfer curve (Fig. 4.7) and the \( R-T \) curve of gated Pt film (Fig. 4.11).

In Figure S4.2, the nominal thickness of the surface gated layer \( t_s \) is labeled on the right of the figure. The black line is identical to the green line in Figure 4.11, which refers to the non-gated state. \( \rho_s(0T) \) decreases with increasing gate voltage \( V_G \) (Fig. S4.2). It is experimentally difficult to determine precisely the effective thickness of the gated layer. However, it is clearly seen that the thinner the sample is,
the larger the gating effect, which is reflected as a greater decrease of the sheet resistance $R_s$ (Fig. 4.7).

### 4.5.2 Comparison of the in-plane magnetoresistance of PIL gated Pt with other systems

In our case of applying an in-plane $B$ field to a material with spontaneous out-of-plane $M$, sweeping $B$ in principle only affects the in-plane component of $M$, so that the time reversal symmetry (TRS) will be preserved, where the profile of FDMR is symmetric with $B = 0$ T (Fig. S4.3a, d).

To demonstrate the significant difference from the AMR of ferromagnet, we fabricated Co thin film sample with thickness of 10 nm that is covered by Al$_2$O$_3$ capping layer to prevent Co from oxidation. The results of magnetoresistance measurement with in-plane $B$ field are shown in Figure S4.3b, d. A prominent $\rho_L$ drop at low field was observed for $B \parallel I$, while $\rho_L$ increase for $B \perp I$. At $\phi = 45^\circ$ or $135^\circ$, $\rho_L$ has no change at low field. For the transverse transport measurement, from Eq. 4.7 and Eq. 4.8, we know that the changes of $\rho_T$ is shifted by $45^\circ$ from $\rho_L$. At $\phi = 45^\circ$ or $135^\circ$, $\rho_T$ increases and decreases with increase of $B$, while at $\phi = 0^\circ$, $90^\circ$, $180^\circ$, $\rho_T$ shows no significant change at low temperature. All data has the same value at $B = 0$ T.

For the standard SMR system with an in-plane magnetization, such as Pt/YIG, the magnetization direction of the ferromagnetic insulator can be probed with electrical means, referred as the magnitude of the resistance of the proximate Pt layer (Fig. S4.3c, f). For each measurement, we first applied a $B$ field as large as saturating the magnetization of YIG. At $\phi = 0^\circ$ or $180^\circ$, with decreasing $B$ field and eventually reversing the direction of $B$ field, $\rho_L$ first decreases and then increases at low $B$ field range. At $\phi = 45^\circ$ or $135^\circ$, the behavior of $\rho_L$ is more complicated. $\rho_L$ changes first to the lowest value and then increases to the highest value. The observed feature may come from the subtle domain structure, which is interpreted as the in-plane procession of the YIG magnetization that experiences all angle $\phi$ between $B$ and $I$, resulting in the full-scale change of $\rho_L$. Similar to the AMR of Co, $\rho_T$ shifts $45^\circ$ from $\rho_L$. Nevertheless, despite the subtle details at low field, the sign of the resistivity changes with respect to $\phi$ is consistent in both AMR and SMR mechanisms, while in different from the reported PIL-gated Pt system.

In contrast to AMR or conventional SMR, the present non-saturating MR shows a difference in $\phi$ dependent symmetry. The formers show $4\pi$ symmetries for both $\rho_L$ and $\rho_T$, where $\rho_L(\phi) = -\rho_L(\phi+90^\circ)$ and $\rho_T(\phi) = -\rho_T(\phi+90^\circ)$; while the later has a 2-fold symmetry for $\rho_L$ but a $4\pi$ symmetry for $\rho_T$, where $\rho_L(\phi) = -\rho_L(\phi+180^\circ)$ and $\rho_T(\phi) = -\rho_T(\phi+90^\circ)$. 


4.5 Supplementary information

Figure S4.3 Comparison of the in-plane magnetoresistance of PIL gated Pt with other systems. (a), (b), (c) Longitudinal magnetoresistivities of Pt/PIL, cobalt thin film and Pt/YIG systems. (d), (e), (f) Transverse magnetoresistivities of Pt/PIL, cobalt thin film and Pt/YIG systems.

At small $B$ field, there is TRS breaking for AMR or SMR due to the $M$ switching or precession. However, as mentioned before, TRS is protected in the presented system, which leads to the magnitude of $\Delta \rho_T$ in FDMR half of the amplitude of $\Delta \rho_T$ in ADMR.

On the other hand, the TRS will be broken with sweeping $B$ perpendicular to the sample $xy$ plane (Fig. 4.23). Magnetization reversal process with finite coercivity can be illustrated by AHE with hysteresis of Hall resistivity $\rho_H$ at $\phi = 90^\circ$, where $\phi$ is the angle between the $B$ and film $xy$ plane.

4.5.3 Finite element modeling of the electrical transport

At low temperature, where the positive shift of ADMR modulation due to UPMR is minimized, the system can be simply described by the SMR theory with the PMA. In order to confirm the validity, we carried out finite element model calculations to identify the ratio of $R_{xx}/R_{xy}$ based on the sample geometry measured from by atomic force microscopy (AFM) (Fig. 4.4). Ideally, the ratio between $\Delta R_{xx}$ and $\Delta R_{xy}$ is given by the ratio between the separation of two Hall-bar electrodes ($l$) and the width of the Pt channel ($w$), assuming that the width of the Hall-bar ($w_{\text{bar}}$) leads are negligibly small. Experimentally, however, the Hall leads are often of certain finite widths. In this case, the exact ratio $\Delta R_{xx}/\Delta R_{xy}$ may not equal to the dimension of the channel $l/w$, whereas finite widths ($w_{\text{bar}}$) of the Hall electrodes will cause deviations. For $\rho_{xx} = \rho_{xy}$, where
\[ \rho_{xx} = \frac{R_{xx} \rho t}{l}, \]  

(4.46)

and

\[ \rho_{xy} = R_{xy} t, \]  

(4.47)

in which \( t \) is the film thickness, \( \Delta R_{xx}/\Delta R_{xy} \) depends on the Hall-bar geometry.

This effect is especially prominent in the situation when the width of the Hall-bar is close to the width of the channel. It is not straightforward to determine how big difference between \( l/w \) and \( \Delta R_{xx}/\Delta R_{xy} \) by assuming the current bypasses from the Hall-bar for a short depth, but rather more reliable based on finite element modeling (FEM). For the above reason, we used the value not exactly the one \( l/w \) measured from AFM, but based on the FEM result for Figure 4.14 and 4.15c.

We make use of a two-dimensional (2D) steady-state FEM to calculate the expected ratio \( \Delta R_{xx}/\Delta R_{xy} \) for the employed experimental geometry in this study. The electrical transport is anisotropic, where the longitudinal and transverse resistivities can be written as [30, 31]:

\[ \rho_{xx}(\phi) = \rho_{\perp} + \Delta \rho_{xx} \cos^2 \phi \]  

(4.48)

and

\[ \rho_{xy}(\phi) = \Delta \rho_{xy} \sin \phi \cos \phi. \]  

(4.49)

They are related to the longitudinal and transverse conductivities by [64]:

\[ \sigma_{xx} = \frac{\rho_{xx}}{\rho_{\perp}^2 + \rho_{xy}^2} \]  

(4.50)

and

\[ \sigma_{xy} = \frac{\rho_{xy}}{\rho_{\perp}^2 + \rho_{xy}^2}. \]  

(4.51)

The model solves the electrical transport equation

\[ \begin{pmatrix} \mathbf{J}_{xx} \\ \mathbf{J}_{xy} \end{pmatrix} = - \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{xy} & \sigma_{xx} \end{pmatrix} \begin{pmatrix} \nabla V_{xx} \\ \nabla V_{xy} \end{pmatrix} \]  

(4.52)

in the Hall-bar, where \( J_{xx} \) and \( J_{xy} \) are electrical current densities along the \( x \) and \( y \) directions, respectively. The nonzero off-diagonal elements in the conductance matrix reflect anisotropic transport properties.

In the model, a constant current is applied to the Hall channel, and the voltage drops in longitudinal \( (V_{xx} = V_1 - V_2) \) and transverse \( (V_{xy} = V_1 - V_3) \) configurations are evaluated for different \( \phi \). The longitudinal and transverse magnetoresistance, \( \Delta R_{xx} \) and \( \Delta R_{xy} \), can be subsequently calculated by:

\[ \Delta R_{xx} = \frac{V_{xx}(\phi = 0^\circ) - V_{xx}(\phi = 90^\circ)}{l} \]  

(4.53)

and

\[ \Delta R_{xy} = \frac{V_{xy}(\phi = 45^\circ) - V_{xy}(\phi = 135^\circ)}{l}, \]  

(4.54)

respectively, from which \( \Delta R_{xx}/\Delta R_{xy} \) can be obtained.
**Figure S4.4** Finite element model results for anisotropic electrical transport in a Hall-bar geometry. (a) The 2D Hall bar geometry used in this study. Color represents the voltage profile when sourcing a current through the Hall bar. Voltages are probed through $V_1$, $V_2$ and $V_3$ terminals. (b) The ratio $\Delta R_{xx}/\Delta R_{xy}$ calculated as a function of the width of Hall-bar ($w_{\text{bar}}$).

Therefore, increasing the width of the Hall-bar will decrease the ratio $\Delta R_{xx}/\Delta R_{xy}$. The reason that we designed a relatively wide Hall-bar was because it was more robust and made the device less likely to be burnt during measurements.

On the other hand, even though at 5 K $\Delta \rho_{xx}/\Delta \rho_{xy} \approx 1$ indicates the resistivity modulation is of the same origin for both longitudinal and transverse transports, the temperature-dependent sign reversal happens for $\rho_{xx}$, not $\rho_{xy}$ implies that the observed phenomena have more mechanism working simultaneous rather than a simple AMR type of behavior.
4.6 Summary

To sum up, magnetoresistive devices in general require the usage of ferromagnets, where the magnetization direction is crucial to the electrical transport. Paramagnets have often been regarded less interesting to new MR effect for lacking spontaneous magnetization and long range magnetic ordering. On the other hand, it is highly desirable to control the magnetization electrically from both fundamental and technological points of view. Due to intrinsically large carrier densities and consequently short Thomas-Fermi screening lengths, metals are difficult to manipulate electrically by gating.

Ionic gating, however, offers a unique approach that is able to alter the electronic state of the channel surface significantly by applying only a few volts. Our reported paramagnetic ionic gating technique combines the advantage of ionic gating with magnetic moments, which imparts a new degree of freedom to the controllable transport properties of electron: the spin.

In this chapter, we report the observation of a new type of MR in the non-magnetic metal (NM)/paramagnetic insulator (PMI) system by an in-plane $B$ field. The magnitude of this new MR is proportional to the magnetization built-up of the proximate paramagnetic insulator under $B$. Contrary to the AMR of ferromagnets that saturates after magnetization are fully aligned, the observed MR can be well described with Langevin function of paramagnetism, which gives rise to the non-saturating contribution to both the longitudinal and transverse MR.

Our experiment extends the concept and application of the SMR mechanism in a Pt/paramagnet insulator system with spontaneous perpendicular magnetization. The observed Langevin type of MR serves as an evidence of paramagnetism induced electrical transport. The emergent phenomena are gate tunable, which paves the path of developing new type of spintronic devices.
4.7 References


4.7 References

Chapter 4


