Gate-controlled magnetoresistance of a paramagnetic-insulator\-platinum interface


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We report an electric-field-induced in-plane magnetoresistance of an atomically flat paramagnetic insulator\-platinum (Pt) interface at low temperatures with an ionic liquid gate. Transport experiments as a function of applied magnetic field strength and direction obey the spin Hall magnetoresistance phenomenology with perpendicular magnetic anisotropy. Our results establish the utility of ionic gating as an alternative method to control spintronic devices without using ferromagnets.

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I. INTRODUCTION

Magnetoresistance (MR), the change of the electrical resistance by external magnetic fields, is the key functionality of data storage [1,2], sensors [3], and logic devices [4]. The evolution of information technology relies on the discovery of new types of MR. For example, the giant magnetoresistance [1,2] and tunnel magnetoresistance [5,6] in magnetic multilayers have been breakthroughs in the field of spintronics that triggered technological revolutions. Conventional magnetoresistive devices contain ferromagnetic elements with stray fields that cause undesirable cross-talk energy loss. Paramagnets that lack spontaneous magnetization play only passive roles, e.g., as spacer layers [7]. The magnetization of ferromagnets cannot be simply switched off; even the physically important and its technologically desirable electric control [8–10] is difficult due to the intrinsically large carrier density and consequently short Thomas-Fermi screening length of metallic magnets. These drawbacks can be overcome by ionic gating, which can generate very large electric fields by applying only a few volts [11–13].

Platinum (Pt) is an essential material for spintronics. It is widely employed as spin injector and detector due to its strong spin-orbit interaction (SOI) and hence large spin Hall angle [14]. According to the Stoner criterion, the large density of state at the Fermi energy puts Pt very close to the ferromagnetic (FM) phase transition. Recently, ferromagnetism was induced in Pt by electrostatic gating using paramagnetic ionic liquid (PIL) [15], a special type of ionic liquid containing paramagnetic ions. The gate-induced carriers are confined on an atomic length scale to the Pt surface. The PIL on top of Pt forms an atomically flat interface to Pt and becomes an electrical insulator below its melting point ($T_m$). In contrast to conventional magnetic thin film multilayers, the physical properties of the present interface can be tuned by varying the voltage of the PIL gate in its liquid phase. Here, we report the observation of a novel gate-controllable MR in the PIL\-Pt system for an in-plane magnetic field $B$. We find that the gating induces a resistance that depends on the direction of $B$. The symmetry is distinctively different from the conventional anisotropic magnetoresistance (AMR) of ferromagnets or the spin Hall magnetoresistance for Pt\-magnetic insulator bilayers with in-plane magnetizations [14,16,17]. On the other hand, the observations can be well explained by the spin Hall magnetoresistance when the conduction electrons in the bulk Pt interact with the interface with perpendicular magnetization. The results illustrate the unique tuning option provided by our system that adds functionalities to spintronic devices such as easy reprogrammability.

We organize the manuscript by first exposing the paramagnetic liquid gate field effect transistor with thin-film Pt channel and the experimental technique in Sec. II. The experimental results including several control experiments are summarized in Sec. III. We analyze our results by analytical and numerical model calculations in Sec. IV (with a related Appendix). We conclude the paper with Sec. V.

II. MATERIALS AND EXPERIMENTS

A. Magnetic properties of paramagnetic ionic liquid

Our device consists of a Pt Hall bar ($t = 12$ nm) covered by a PIL gate [Fig. 1(a)]. The PIL used in our experiment is butylmethylimidazolium tetrachloroferrate (BMIM[FeCl4]) [Fig. 1(b)]. The d shell of Fe$^{3+}$ in the magnetic anions is half-filled with spin quantum number $S = 5/2$, a high spin state [Fig. 1(c)] [18]. The magnetic properties of the PILs are measured by a SQUID magnetometer (MPMS XL-7, Quantum Design). The magnetization curves (M\-B) were taken at various temperatures for $B$ fields up to $7$ T, showing no hysteresis loop even at the lowest temperature [Fig. 1(d)]. The temperature dependence of magnetic susceptibility ($\chi$\-T) is measured at $B = 0.1$ T during warming up after zero-field

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cooling down to 2 K [Fig. 1(e)]. The magnetic susceptibility \(\chi\) of BMIM[FeCl\(_4\)] follows Curie’s law indicating the paramagnetic (PM) nature of it. For small magnetic field \(H\ll 1\), \(B \approx \mu_0 H_c\), we have (in SI units) \(\chi = M/T \approx \frac{M}{B} = \frac{n\mu_B}{k_B T}\), where \(k_B = 1.38 \times 10^{-23} \text{JK}^{-1}\), \(n\) is the number of magnetic atoms, and the molar susceptibility \(\chi\) is in units of \(\text{m}^3\text{mol}^{-1}\). We find a large effective magnetic moment \(\mu_{\text{eff}}\) of BMIM[FeCl\(_4\)] of 5.77 \(\mu_B\), where \(\mu_B\) is the Bohr magneton. Assuming orbital quenching, this value agrees well with the theoretical value for a half-filled 3d atomic shell of 5.92 \(\mu_B\) calculated from \(g\sqrt{\chi(S+T)}\), where \(g = 2\) is the Landé factor.

### B. Device fabrication

The transport properties are measured in a PIL-gated transistor shown schematically in Fig. 1(a). The device consists of a Pt Hall bar with length \(l = 7 \mu\text{m}\) and width \(w = 3.5 \mu\text{m}\) that is patterned by electron beam lithography and characterized by atomic force microscopy as shown in Fig. 2(a). A Pt thin film is deposited on a SiO\(_2\)/Si substrate by dc magnetron sputtering. The electrical contacts and the side gate for PIL gating are made of Au|Ti (45/5 nm) deposited by electron beam evaporation.

### C. Electrical transport properties of paramagnetic ionic gated-Pt

The electrical transport is measured by two Stanford SR830 Lock-in amplifiers for the longitudinal and transverse voltages (\(V_L\), \(V_T\)) simultaneously under constant ac current excitation \(I = 50 \mu\text{A}\) at \(\sim 13\) Hz. The longitudinal and transverse resistivities were calculated according to \(\rho_L = \frac{V_L}{I} l w\) and \(\rho_T = \frac{V_T}{I} l t\), where \(l\), \(w\), \(t\) are the length, width, and thickness of the Pt Hall bar. During the PIL gating process, a dc gate voltage \(V_G\) is applied between the Pt channel and the side gate electrode through a Keithley 2450 source meter. Depending on the polarization of the voltage bias, by applying a positive or negative gate voltage, cations or anions are driven towards the Pt surface, which collect or depletes electrons in the top-most layer, respectively. The gating experiment with scan rate of 50 mV s\(^{-1}\) is carried out at 220 K.

To characterize the magnetic and electrical properties of the PIL-gated device, we carry out angular-dependent magnetoresistance (ADMR) and field-dependent magnetoresistance (FDMR) experiments at 5 K. In the ADMR experiments, the rotation is along the axis perpendicular to the plane of the film with angle \(\phi\) between \(\mathbf{I}\) and \(\mathbf{B}\). The \(\rho_L\) and \(\rho_T\) are measured
under a constant applied $B$ field. In the FDMR experiment, the device is fixed at a particular $\phi$ and resistivities are measured as a function of applied $B$ field. We have also performed FDMR measurements for different temperatures. For the in-plane $B$ configuration, the device is placed with $\phi = 45^\circ$; for the out-of-plane $B$ configuration, the device is placed with film plane normal to the $B$.

**D. Gate voltage dependent longitudinal resistance as a function of temperature**

The PIL-gating experiment is initiated at 220 K. This temperature is chosen based on the following criteria: (1) the temperature should be as low as possible to suppress possible chemical reactions and (2) the ionic mobility of the PIL should be high enough so that ions still can move in an applied electric field.

The $V_G$ dependence of the longitudinal resistance $\rho_L$ shows reversible control with negligible leakage current $I_G$ [Fig. 2(b)], which indicates a change of the electronic surface state of Pt. At positive $V_G$, $\rho_L$ decreases with the increase of Fermi level in the band structure of Pt [15] and saturates after $V_G > 2$ V. After observing the resistance drop in $\rho_L$ [step 1 in Fig. 2(e)], we fix this low resistance state at $V_G = 2.2$ V (“ON” state) by rapidly cooling the device below the $T_m$ of the PIL [step 2 in Fig. 2(e)]. ADMR measurements at 5 K show a clear modulation of $\rho_L$ [red curves in Figs. 2(c) and 2(d)], indicating a dramatic change of the magnetic properties of the Pt channel after switching to the “ON” state. The sample was subsequently warmed up to 260 K with $V_G = 0$ V [step 3 in Fig. 2(e)]. Below 210 K, PIL remains frozen so as to the induced-electronic state of Pt. Above 210 K, $R_L$ gradually recovers [step 4 in Fig. 2(e)], revealing a relaxation of the gate-induced resistance change by the equalization of the ion distribution that is caused by the melting of the PIL. This process is complete above $\sim 230$ K, as the reduction rate of the sheet resistance becomes the same as the one below 200 K, indicating the sample had returned to the completely relaxed state, i.e., the pristine state of Pt. The sample was cooled down to 5 K again for comparison [step 5 in Fig. 2(e)]. At 220 K, $\rho_L$ has exactly the same value as the one before gating; suggesting that thermal cycling does not deteriorate the sample quality and the electronic state of the Pt film remains the same. This aforementioned effect disappears as we see no resistivity changes as a function of $\phi$ even at $B = 6$ T after cooling down with $V_G = 0$ V (“OFF” state) [blue curves in Figs. 2(c) and 2(d)]. This direct correlation of $\Delta \rho_T$, $\Delta \rho_L$ as a function of $\phi$ [Figs. 2(c) and 2(d)] with $V_G$ proves that the observed effect is induced by the PIL gating. We also find that gating by a nonparamagnetic liquid...
of measurements display harmonic modulations with a period at \( T = 45^\circ \) [Fig. 3(c)]. This dependence mimics the AMR and planar Hall effect (PHE) of ferromagnets, in which \( \rho \) reference values at \( B = 0 \) T lead to such a state [15].

Bis(trifluoromethanesulfonyl) imide: DEME-TFSI) does not (e.g., N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl) imide: DEME-TFSI) does not lead to such a state [15].

III. RESULTS AND DISCUSSIONS

A. Angular-dependent magnetoresistivity for different in-plane field \( B \)

Figure 3(a) shows the geometry of ADMR measurements. The longitudinal resistivity \( \rho_L \) at 5 K for the “ON” state under \( B \) field strengths from 0.5 to 6 T is shown in Fig. 3(b). All measurements display harmonic modulations with a period of \( \pi \), where the maximum and minimum values for \( B \parallel I \) (current) and \( B \perp I \) are denoted as \( \rho_{\parallel} \) and \( \rho_{\perp} \), respectively.

A similar angle dependence is observed for the transverse resistivity \( \rho_T \), in which the maxima and minima are shifted by 45° [Fig. 3(c)]. This dependence mimics the AMR and planar Hall effect (PHE) of ferromagnets, in which \( \rho_L \) and \( \rho_T \) are governed by the angle \( \phi \) between \( I \) and magnetization \( M \) as

\[
\rho_L(\phi) = \rho_{\perp} + \Delta \rho_L \cos^2 \phi, \quad (1)
\]

\[
\rho_T(\phi) = \Delta \rho_T \sin \phi \cos \phi. \quad (2)
\]

Despite the similarity in shape, our field-dependent amplitudes \( \Delta \rho_L = \rho_{\parallel} - \rho_{\perp} \) and \( \Delta \rho_T = \rho(\phi = 45^\circ) - \rho(\phi = 135^\circ) \) are significantly different from the AMR. The field-dependent modulations in the present system show the same values of \( \rho_L \) at \( \phi = 0^\circ \), indicated by the dashed lines [Fig. 3(b)], whereas \( \rho_L \) would be constant at \( \phi = 45^\circ \) for the latter case. Moreover, if the effect is caused by AMR, \( \Delta \rho_L \) and \( \Delta \rho_T \) must saturate for fields exceeding the coercivity \( H_C \) and remain finite even at \( B = 0 \) T due to magnetic remanence. We, however, find \( \Delta \rho_L \) and \( \Delta \rho_T \) to vanish without \( B \) and increase with field strengths without saturating even at 6 T [Fig. 3(d)]. This dependence resembles the magnetization curve of Pt at 5 K, which is well-described by the Langevin function of paramagnetism,

\[
L(x) = \coth x - 1/x, \quad (3)
\]

where \( L(x) = \frac{M(x)}{M_s} \), \( x = \frac{\mu B r}{k_B} \), \( M_s \) is the saturation magnetization, \( \mu \) the magnetic moment, and \( k_B \) the Boltzmann constant.

We further characterize \( \Delta \rho_L \) and \( \Delta \rho_T \) by the field-dependent magnetoresistance (FDMR) at the same temperature (\( T = 5 \) K) and at various angles \( \phi \). At \( \phi = 90^\circ \), when \( B \) is in the sample \( xy \) plane and perpendicular to the current direction \( I \), we observe a negative MR that does not saturate at the maximum \( B \) field of 6 T [Figs. 3(e) and 3(f)]. At \( \phi = 0^\circ \), when \( B \) is in-plane and along \( I \), however, the MR vanishes for all \( B \) fields. Although in the ADMR measurement, the \( \rho_L \) profile at a fixed \( B \) strength shows an AMR-like modulation \( \sim M \cdot I \), the observed anisotropic FDMR firmly excludes the AMR.

With increasing temperature, the magnetic susceptibility of the PIL decreases significantly [Fig. 1(e)] and the induced magnetization of the Pt surface becomes weaker. Both effects are expected to affect the interface MR. We therefore carry out FDMR measurements at various temperatures under both in-plane and out-of-plane \( B \) fields as shown in Sec. II C.
B. Field-dependent magnetoresistivity for different in-plane angles \( \phi \)

Figure 4(a) shows the in-plane longitudinal and transverse magnetoresistivities as a function of \( B \). The increase in \( \rho_L \) and \( \rho_T \) scales nicely with the magnetization of the PIL, from which we can derive a saturation \( \rho_L \) of 23.16 \( \mu \Omega \) cm with a prefactor \( \mu_{\text{eff}}/k_B T = 0.468 \) taken from the fit of the \( M-B \) curve of the pure PIL [Fig. 4(b)].

The in-plane magnetoresistivities also scale with the susceptibility of the PIL for the FDMR data measured at each individual angle \( \phi \) from 90° to 0° [Fig. 4(a)]. Like the PIL magnetization, the FDMR of both \( \rho_L \) and \( \rho_T \) does not saturate at magnetic fields up to 6 T. We obtain the longitudinal saturation resistivity \( \rho_L \) at each angle \( \phi \) from FDMR measurements by extrapolating the Langevin function to a high magnetic field (Table I).

The correlation of the magnetotransport experiments with the susceptibility as sketched above is tantalizing, suggesting a paramagnetic origin of the observed phenomena. However, we are not aware of a microscopic mechanism that would explain this behavior. More importantly, the angle dependences of \( \rho_L \) and \( \rho_T \) do not agree with a paramagnetic phase as illustrated in Figs. 3(e) and 3(f): the ADMR \( \rho_L \) shows a sinusoidal behavior pinned at \( \phi = 0° \) [Fig. 3(b)]. This differs from the conventional anisotropic magnetoresistance (AMR) or spin-Hall magnetoresistance (SMR) of the Pt|YIG system [14,16,19] with small anisotropies as well as an MR from a paramagnet, for which the field-independent point is at \( \phi = 45° \) (and 135°). FDMR measurements at \( \phi = 0° \) and 90° are therefore not symmetric with respect to the reference line at \( B = 0 \) T [Fig. 3(e)], in stark contrast to the AMR of a magnetic film or SMR of the Pt|YIG system. \( \rho_T \) shows sinusoidal behavior as well, but is shifted by 45° as expected from the resistivity tensor [Fig. 3(c)]. In this case, \( \rho_T \) at 90° shows \( B \) independent behavior and \( \rho_T(45°) \) and \( \rho_T(135°) \) are mirror symmetric as a function of \( B \) [Fig. 3(f)].

These features are strong evidence for a spontaneous perpendicular magnetization at the interface.

C. Temperature-dependent magnetoresistivities

In order to understand the temperature contribution to the observed effect, we performed FDMR measurement at various temperatures. Figures 5(a) and 5(b) show the evolution of the in-plane MR with increasing temperature. All data were collected at \( \phi = 45° \), at which angle both \( \rho_L \) and \( \rho_T \) depend on \( B \). By symmetry, \( \delta \rho_L(45°) = \rho_L(0) \) should be \( \sqrt{2/2} \) times \( \delta \rho_L(90°) \), which is equal to \( \Delta \rho_L = \rho_{||} - \rho_{⊥} \) in the ADMR measurement; moreover, \( \delta \rho_T(45°) = \rho_T(0) \) should equal \( \Delta \rho_T \) from the ADMR. For comparison, we also measured the longitudinal (magnetococonductivity \( \sigma_T \)) and transverse signals (Hall resistivity \( \rho_H \) as a function of perpendicular \( B \) in the out-of-plane geometry at different temperatures, as shown in Figs. 5(c) and 5(d). Similarly, the changes of the interface \( \delta \rho_T \) are defined as \( \delta \rho_T = -(\sigma_T(B) - \sigma_T(0)) \) after subtracting the parallel bulk contribution; whereas \( \delta \rho_H \) is extracted by extrapolating the linear Hall response from high to zero magnetic field [red lines in Fig. 5(d)].

A crossover of \( \delta \rho_L = -\delta \rho_T \) and \( \delta \rho_H \) from negative values at low temperature to positive ones at high temperature causes a sign change at roughly 40 K [Figs. 5(a), 5(c), and 5(d)]. In contrast, \( \Delta \rho_T > 0 \) up to the highest measured temperatures [Fig. 5(b)].

The \( B \) dependence of the \( M \) of the PIL show no hysteresis even at the lowest temperatures [Fig. 1(d)], indicating the absence of long-range FM ordering. Therefore the \( M \) of the PIL increases with increasing magnetic field or decreasing temperature. Once being frozen, PIL becomes highly insulating and no electrical current can enter. In addition, the strong Thomas-Fermi screening limits gate-induced changes in the electronic state of the Pt to the top-most atomic layers [20–22]. Therefore our paramagnetic gating-induced magnetoresistance (PMR) effect must originate from the Pt side of the PIL/Pt interface.

In a conductor with spontaneous \( M \), Ohm’s relation between the electric field \( E \) and the electric current \( \mathbf{I} \) reads [23]

\[
E = \rho_{||} \mathbf{I} + (\rho_{||} - \rho_{⊥})(\mathbf{M} \cdot \mathbf{I})\mathbf{M} + \rho_H \mathbf{M} \times \mathbf{I}.
\]
When $B$ is perpendicular to the sample $xy$-plane (hence the direction of $I$), we detect an anomalous transverse voltage (also at 5 K) [Fig. 5(d)], i.e., the last term in Eq. (4), where $\rho_H$ is the Hall resistivity. The observation is consistent with our recent report of a PIL gating-induced FM state in Pt with perpendicular magnetic anisotropy (PMA) [15]. The effect of an in-plane $B$ on the intrinsic perpendicular $M$ can be described by the Stoner-Wohlfarth model of coherent magnetization rotation and will be discussed later.

**D. Temperature-dependent anomalous Hall effect**

The anomalous Hall effect (AHE) [24,25] can be parameterized by [26]

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

where $\rho_0(\rho_H)$ and $R_0 (R_H)$ are the ordinary (anomalous) Hall resistivities and coefficients, respectively. The ordinary part of $\rho_{xy}$ at higher $B$ shows that the conducting carriers are negatively charged and the anomalous part demonstrates that the direction of the spontaneous magnetization is parallel to $B$. We determine the anomalous Hall resistivity $\delta \rho_H$ by extrapolating the linear dependence of the measured $\rho_{xy}$ at large $B$ to zero field [see Fig. 5(d)].

In the present system, the ordinary Hall effect remains electronlike, which implies that the sign reversal of the AHE cannot be a result of the global change of the Fermi surface topology. However, the energy of singular hot spots at the Fermi surface determined by the FM exchange interaction is sensitive to subtle environment changes, such as the temperature variation [25]. Moreover, extrinsic mechanisms such as impurity skew [27] and side-jump [28] scattering may also explain the sign change of the AHE; clarification of the exact mechanism requires investigations beyond the scope of the present article.

**IV. MODELLING**

**A. Stoner-Wohlfarth model for the perpendicular magnetic anisotropy in PIL-gated Pt**

Materials with perpendicular magnetic anisotropy (PMA) are of great importance for spintronics [29–35]. The PMA in
our material become evident by the square-shaped hysteretic $\rho_{xy}$ under a $B$ field normal to the film plane [36], which is a measure for the hysteresis loop of the magnetization. Along the ferromagnetic easy axis, the magnetization saturates rapidly, which is characteristic for a strong PMA.

The magnetic field dependence of the in-plane magnetization can be interpreted by the Stoner-Wohlfarth model. In Fig. 6(a), we show a schematic illustrating the directions of current $I$, magnetic field $B$, magnetization $M$, and magnetic easy axis with respect to the Pt film. We consider a magnetocrystalline anisotropy term $K \sin^2 \theta$, where $K$ is the anisotropy constant, and the Zeeman energy $M_s B \cos(\gamma - \theta)/\mu_0$, in which $\theta$ (or $\gamma$) are the angles between $M$ (or $B$) and the magnetic easy axis, respectively. The thin film shape anisotropy is described by $\frac{1}{2} \mu_0 M_s^2 \cos^2 \beta$, where $\beta$ is the angle between the film normal and $M$ and $\mu_0$ the vacuum permeability. The total magnetic energy $E$ then reads

$$E = K \sin^2 \theta - M_s B \cos(\gamma - \theta)/\mu_0 + \frac{1}{2} \mu_0 M_s^2 \cos^2(90^\circ - \gamma + \theta).$$  

The equilibrium angle $\theta$ is governed by energy minimization:

$$\frac{\partial E}{\partial \theta} = 0$$  

and

$$\frac{\partial^2 E}{\partial \theta^2} > 0.$$  

With

$$h = \frac{B}{B_s}$$  

and

$$B_s = \frac{2K\mu_0}{M_s},$$

Eq. (6) can be simplified to

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

We also define the parameter

$$x = \frac{M_s^2}{4K},$$

which measures an in-plane shape anisotropy relative to the out-of-plane magnetocrystalline anisotropy.

Figures 6(b) and 5(c) show the numerical results as a function of applied magnetic field. The equilibrium magnetization...
Pristine Pt is a normal metal so an in-plane $\mathbf{B}$ field should not generate a significant MR [Figs. 2(c), 2(d), and 7(a)]. The spontaneous magnetization $\mathbf{M}$ at and perpendicular to the PIL/Pt interface when cooled down to low temperatures with $V_G$ is normal to the polarization of the spin motive force $\mathbf{\sigma}$ over the remainder of the Pt film. Without an external $\mathbf{B}$ field, $\mathbf{\sigma} \perp \mathbf{M}$ and the generated spin current is efficiently absorbed as a spin transfer torque, leading to the high resistance state of $\rho_L$ [Fig. 7(b)]. Increasing the in-plane $\mathbf{B}$ gradually pulls $\mathbf{M}$ into the plane. At $\phi = 0^\circ$, $\mathbf{\sigma}$ is still normal to $\mathbf{M}$, the absorption of the spin current remains constant, and $\rho_T$ remains at the high resistance state [Fig. 3(e) and 7(c)]. At $\phi = 90^\circ$, on the other hand, $\mathbf{M}$ is pulled into the plane with increasing $\mathbf{B}$ until $\mathbf{M} \parallel \mathbf{\sigma}$, which is when the spin current generated by the SHE is mostly reflected, resulting in a decrease of $\rho_L$. According to this SMR mechanism, the maximum amplitude of the longitudinal and transverse ADMRs should be the same, $\Delta \rho_L = \Delta \rho_T$, when the sample geometry is factored in [Fig. 2(a)], in good agreement with low temperature data. We adopt the highest SMR value $\Delta \rho/\rho = 0.027\%$, obtained with an external field of 6 T at 5 K, in the equation

$$\frac{\Delta \rho}{\rho} = \frac{\theta_{\text{SH}}}{t} \frac{4\lambda^2 G_s \tanh^2 \theta_{\text{SH}}}{\pi} \frac{2GM_s \coth^2 \frac{1}{2}}{\lambda},$$

where the thickness $t = 12 \text{ nm}$, resistivity $\rho = 59.4 \mu\Omega \text{cm}$, spin Hall angle $\theta_{\text{SH}} = 0.044$, and spin diffusion length of Pt $\lambda = 3.5 \text{ nm}$ [38], and find that $G_s = 2.88 \times 10^{14} \text{ S/m}^2$. This value is roughly in the same order of magnitude as that of the prototypical YIG/Pt systems.

C. Finite element modeling of the electrical transport

The magnetoresistivities are calculated from the measured voltages that need to be normalized to the geometric factor.
In the limit of vanishing width of the Hall contacts, this ratio is governed by the ratio between the separation of two Hall-bar electrodes (l) and the width of the Pt channel (w), whereas finite widths (w_{bar}) of the Hall electrodes will cause deviations. For \( \rho_{xx} = \rho_{yy} \),

\[
\rho_{xx} = \frac{V_{xx}w_{l}}{I_{xx}}
\]

(14)

and

\[
\rho_{xy} = \frac{V_{xy}w_{l}}{I_{xx}},
\]

(15)

where \( t \) is the film thickness and \( \Delta V_{xx}/\Delta V_{xy} \) depends on the Hall-bar geometry.

However, these conditions are not valid once \( w_{bar} \) is not negligible compared to \( w \). We carried out finite element model (FEM) calculations in order to confirm the validity of this prediction by computing the ratio \( \Delta V_{xx}/\Delta V_{xy} \) based on the sample geometry measured by atomic force microscopy (AFM) [Fig. 2(a)].

Here we use a two-dimensional steady-state FEM to calculate the ratio \( \Delta V_{xx}/\Delta V_{xy} \) for the experimental geometry sketched in Fig. 8(a). The longitudinal and transverse resistivities can be written as [23,39]

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

(16)

and

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

(17)

\( \rho_{xx} \) and \( \rho_{xy} \) are related to the longitudinal and transverse conductivities by [40]

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

(18)

and

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

(19)

The model solves the electrical transport equation

\[
(J_{xx}, J_{xy}) = -\begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{xy} & \sigma_{xx} \end{pmatrix} \begin{pmatrix} \nabla V_{xx} \\ \nabla V_{xy} \end{pmatrix}
\]

(20)

in a Hall-bar geometry, where \( J_{x} \) and \( J_{y} \) are electrical current densities along the x and y directions, respectively. A constant current \( I \) is applied to the Hall channel, and the longitudinal \( (V_{xx} = V_{1}-V_{2}) \) and transverse \( (V_{xy} = V_{1}-V_{2}) \) voltage drops are evaluated for different \( \phi \). The longitudinal and transverse magnetoresistances \( \Delta R_{xx} \) and \( \Delta R_{xy} \) can be subsequently calculated by

\[
\Delta R_{xx} = \frac{V_{xx}(\phi = 0\degree) - V_{xx}(\phi = 90\degree)}{I}
\]

(21)

and

\[
\Delta R_{xy} = \frac{V_{xy}(\phi = 45\degree) - V_{xy}(\phi = 135\degree)}{I},
\]

(22)

respectively, from which \( \Delta R_{xx}/\Delta R_{xy} \) follows.

Figure 8(b) summarizes the results. With the increase of \( w_{bar} \), the bypass effect through the Hall-bar leads to a decrease of \( \Delta R_{xx}/\Delta R_{xy} \). For \( w_{bar} = 2.0 \mu m \) from the AFM measurement, \( \Delta R_{xx}/\Delta R_{xy} = 1.7 \), which leads to a renormalized resistivity from 7/3.5 based on the length and width of the conducting channel to a smaller value (\( \approx 6/3.5 \)). By renormalizing the \( \Delta R_{xx} \) and \( \Delta R_{xy} \) values from the ADMR experiments at 5 K [Figs. 3(b) and 3(c)], we find that \( \Delta \rho_{xx} \approx \Delta \rho_{xy} \) [Fig. 3(d)], which agrees with the SMR mechanism.

D. Interpretation of temperature-dependent magnetoresistivities

The MR as a function of temperature is governed by the competition between an MR that is diminished with the reduced spontaneous magnetization and an enhanced paramagnetism that generates magnetic order along an applied magnetic field. With increasing temperature, a positive MR effect evolves for both in-plane and out-of-plane B configurations in Figs. 5(a) and 5(c), which possibly has a paramagnetic origin [41]. For the in-plane configuration and at an angle of \( \phi = 45\degree \), the FDMR (blue arrows or dots in Fig. 9) consists of the PMR effect on top of a positive shift of the background. At low temperatures, the strong PMA dominates and causes a negative MR [Fig. 9(a)]. With the increase of temperature, the PMA weakens and the background that contributes to the observed signal increases. At \( \sim 40 \) K, the in-plane transport properties resemble the conventional AMR or SMR type.
FIG. 9. Schematic of the temperature dependence of in-plane MR. (a)–(c) and (d)–(f) represent the longitudinal and transverse MR, in which $\Delta \rho/\rho = \rho(\phi, B)/\rho(\phi, B = 0) - 1$. (a)–(d), (b)–(e), and (c)–(f) are at low, intermediate, and high temperatures, respectively. The changes of the measured FDMR signals and the magnitudes of the PMR are illustrated by the blue arrows and red bars with respect to other effects.

of behavior with vanishing MR at $\phi = 45^\circ$ [Fig. 9(b)]. At even higher temperatures, the aforementioned positive MR eventually overwhelms the contribution of PMR, resulting in a positive FDMR signal [Fig. 9(c)].

The transverse transport [Figs. 9(d)–9(f)], on the other hand, remains almost unchanged at different temperatures, which is consistent with the following scenario. With increasing temperature, the PMA and $M$ of the gate-induced FM layer weakens, leading to a gradual transition of the in-plane component of magnetization from a FM interface with PMA to one with enhanced PM susceptibility. Consequently, the PMR changes into the conventional in-plane SMR [Fig. 9(f)]. When the in-plane $B$ field forces the perpendicular magnetization into the plane at low temperature and that of polarizing the paramagnet at high temperatures to be the same, the transverse magnetoresistance does not depend on temperature, as observed.

V. CONCLUSION

In conclusion, our study introduces a tunable spintronic system that employs a liquid paramagnetic insulator. At low temperatures, we observe a spin-dependent in-plane MR effect that can be explained by extending the SMR model to a PIL|Pt interface with spontaneous perpendicular magnetization. The physics underlying the rich transport features as a function of temperature remains to be fully understood. The versatile gate-tunable magnetic phenomena lay the foundation for reprogrammable spintronic devices.

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APPENDIX: SEPARATION OF THE SURFACE AND BULK CONTRIBUTIONS TO THE MAGNETORESISTIVITIES

With $B$ applied perpendicular to the plane, the measured MR contains additional contributions to the Pt|PIL interface response, such as a positive MR from the bulk of Pt and a negative MR from the surface FM Pt due to magnetic ordering.

In the Sommerfeld model of metals with the relaxation-time approximation, the equation of motion of electrons in a $B$ field reads

$$m^* \left( \frac{d\mathbf{v}}{dt} + \frac{\mathbf{v}}{\tau} \right) = -e\mathbf{E} - e\mathbf{v} \times \mathbf{B}, \quad (A1)$$

where $\mathbf{v}$ is the drift velocity, $e$ the elementary charge, $m^*$ the effective mass, and $\tau^{-1}$ the relaxation rate. When the electrons drift parallel to $B$, i.e., for the in-plane $B$ configuration, the longitudinal MR vanishes. Without gating ($V_G = 0$) we find indeed a negligibly small in-plane angular-dependent MR
FIG. 10. Temperature dependent longitudinal magnetoresistivity $\rho_p$ under an out-of-plane $B$. (a) $\rho_p$ of pristine Pt without gating. (b) $\rho_p$ after PIL gating. (c) Changes in the conductivities of the surface contribution that reflect the effects of PIL gating. The changes are defined as $-[(\sigma_p(G) - \sigma_p(Pr))]$.

In order to extract the Pt|PIL interface contribution in the perpendicular configuration, the interface and bulk contributions to the observed signals MR should be disentangled. To this end, we measured the FDMR of pristine Pt without gating for several temperatures [Fig. 10(a)]. The positive ordinary MR at low temperatures saturates at large $B$ and vanishes at $T > 40$ K. We then applied the PIL gating on the same device and observed an almost vanishing MR at 5 K that increases with rising temperatures [Fig. 10(b)].

In the absence of a proper transport theory, we can still estimate the bulk and interface contributions to the resistivity of thin films in the limit of a bulk mean-free path $\lambda$ that is smaller than the film thickness. First, we analyze the thickness-dependent transport in pristine Pt. We find a higher conductivity than Fischer et al. [22] but comparable with Althammer et al. [14], presumably due to differences in the

FIG. 11. The thickness-dependent conductivities of Pt films [14,15,43].
fabrication technique (sputtered films have higher conductivity than evaporated ones) (Fig. 11). The mean-free path $\lambda$ of Pt can be estimated from the conductivity in the free electron model,

$$\frac{\sigma}{\lambda} = \left( \frac{8\pi}{3} \right) \frac{e^2}{h} n^2,$$

(A2)

where the conduction electron density $n$ is $1.6 \times 10^{22} \text{ cm}^{-3}$ for Pt. With $\sigma (5 \text{ K}) = 3.4 \times 10^4 \text{ S cm}^{-1}$, we find $\lambda = 6.9 \text{ nm}$ and it becomes only shorter at higher temperature. Assuming that the bulk metal is a short for the electric conductance, viz. $\sigma = \sigma_{\text{int}} + \sigma_{\text{bulk}}$, then only the interface contribution is modulated by the gating, while $\sigma_{\text{bulk}}$ remains unmodified due to the good screening. The difference in interface scattering can then be obtained by subtracting $\sigma$ (nongated) from $\sigma$ (gated) as plotted in Fig. 10(c).

We observe a change of sign around 30 K that is likely to be caused by the temperature-dependent competition of different interface scattering processes such as the MR of the top-most ferromagnetic Pt layer, an interface PIL-gating induced spin Hall magnetoresistance (PMR), and a positive magnetoresistance as a background that is yet identified. Assuming that the FM Pt layer is a multidomain ferromagnet [36,42] below a critical temperature $\sim 30 \text{ K}$ from the AHE, see Fig. 5(d)], a negative MR can be interpreted by magnetic field-induced suppression of spin-dependent scattering at the boundaries of nonaligned magnetic grains. The latter does not depend on the relative directions of $\mathbf{I}$ and $\mathbf{B}$ [36]. The PMR exists only in the ferromagnetic phase and should be very small as long the magnetization is spontaneously oriented perpendicular to the interface.


