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Ionic liquid gating control of planar Hall effect in $Ni_{80}Fe_{20}/HfO_2$ heterostructures*

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We report a tunable transverse magnetoresistance of the planar Hall effect (PHE), up to 48% in the $Ni_{80}Fe_{20}/HfO_2$ heterostructures. This control is achieved by applying a gate voltage with an ionic liquid technique at ultra-low voltage, which exhibits a gate-dependent PHE. Moreover, in the range of 0-V to 1-V gate voltage, transverse magnetoresistance of PHE can be continuously regulated. Ferromagnetic resonance (FMR) also demonstrates the shift of the resonance field at low gate voltage. This provides a new method for the design of the electric field continuous control spintronics device with ultra-low energy consumption.

Keywords: ionic liquid gating, planar Hall effect, electric field regulation

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1. Introduction

The planar Hall effect (PHE) in ferromagnetic materials, meaning the appearance of an in-plane transverse voltage when the co-planar electric current and magnetic fields are not perfectly aligned to each other, can be considered as a promising candidate for applying in metallic magnetic devices such as magnetic sensors and magnetic random access memory,[1–4] which is resulted from the s–d mixing generated by spin–orbit coupling.[5,6] NiFe alloy with low-cost and availability is one of the common materials for PHE sensors,[1,2] which is based on the planar Hall magnetoresistance, and NiFe alloy has various magnetic properties with the change of Ni content, therefore it has high research value.[7] It is well known that the planar Hall magnetoresistance can be effectively tailored by changing the magnitude of an applied magnetic field, the angle of applied electric current and magnetic field, and the width of Hall bar. However, from a device application perspective, tuning should be reversible and continuous as far as possible. As metallic magnetic devices develop rapidly, the voltage-controlling magnetoelectric coupling in ferromagnetic/ferroelectric heterostructure can not only continuously mediate magnetic properties but also realize the nonvolatile behavior of that. For example, Laukhin *et al.*[8] regulate the exchange bias of the FeNi/$YMnO_3$ heterostructures by an electric field at very low temperatures, which achieved a one-way magnetization switch. Liu *et al.*[9] controlled the AMR of the $Ni_{80}Co_{20}$/PZN-PT film and the GMR of the FeMn/$Ni_{80}Fe_{20}$/Cu/Co/PZN-PT film by an electric field. Nevertheless, the piezo strain-mediated magnetoelectric coupling causes large energy consumption due to high ferroelectric gating voltage.[10–12] Recently, the ionic liquid gating with an electric double layer (EDL) has been widely used for voltage-driven material properties modification, which greatly reduces energy consumption. For instance, Nakano *et al.*[13] reached a VO_2 first-order metal–insulator transition using an ionic liquid at a gate voltage of around 1 V. Zhao *et al.*[14] controlled the spin reorientation transition of the Pt/$(Co/Pt)_2$/Ta multilayer film by ionic liquid, which accomplished the magnetic moment from the out-of-plane to the in-plane. At the same time, the magnetoresistance is easily tuned by the ionic liquid N, N-diethyl-N-(2-methoxyethyl)-N-methylammonium bis (trifluoromethylsulphonyl) imide (DEME-TFSI).[15–17] Therefore, in this work, the ionic liquid gating-controlling PHE is investigated in $Ni_{80}Fe_{20}/HfO_2$ at room temperature. When the gate voltage is 1 V, the change rate of the transversal reluctance R_{xy} is as high as 48%, and continuous, reversible and repeated tuning is achieved in the voltage range of 0 V–1 V. At a gate voltage of 4 V, the resonance field H_r drifts by 4 Oe (1 Oe = 79.5775 $A{\cdot}m^{-1}$), further indicating that the electric field can result in the magnetic change of the $Ni_{80}Fe_{20}$ film. This provides an important idea for designing continuously adjustable spintronic devices.

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2. Experimental details

$Ni_{80}Fe_{20}$ with 10-nm thickness was deposited on Si substrate by magnetron sputtering and then was covered by a cap layer HfO_2 (7 nm). The measurement of PHE was completed by a self-built transport test platform within an external magnetic field of 1000 Oe. The electric field-controlled PHE device is shown in Fig. 1(a), which uses an ionic liquid DEME-TFSI as the electrolyte. The $Ni_{80}Fe_{20}/HfO_2$ heterostructures thin film was fixed on a printed circuit board. A drop of ionic liquid was dropped on the top surface of HfO_2. Finally, a support structure was made to fix the top electrode. Ferromagnetic resonance (FMR) was measured using a JEOL, JES-FA 300 spectrometer with a test power of 1 mW.

3. Results and discussion

The measurement result of PHE is shown in Fig. 1(b). The inset in Fig. 1(b) shows the relationship between the test current $\boldsymbol{I}$ and $\boldsymbol{H}$. The starting point of the test was $\boldsymbol{H}||\boldsymbol{I}$. When the magnet was rotated, an angle φ was formed between $\boldsymbol{H}$ and $\boldsymbol{I}$. The transverse electric field strength of the ferromagnetic film is $E_{xy} = (\rho_{||} - \rho_{\perp}) j_x \sin\varphi\cos\varphi$.[18,19] Therefore, the transverse magnetoresistance $R_{xy} = 1/2(R_{||} - R_{\perp})\sin 2\varphi$,[20] where $\rho_{||}$ ($\rho_{\perp}$) is a resistivity in which the current density j_x in the x direction is parallel (vertical) to the magnetization $\boldsymbol{M}$. $R_{||}$ ($R_{\perp}$) is parallel (vertical) magnetoresistance of j_x and $\boldsymbol{M}$. Clearly, there is a functional relationship $R_{xy}\sin 2\varphi$, which results are in accordance with the theory.

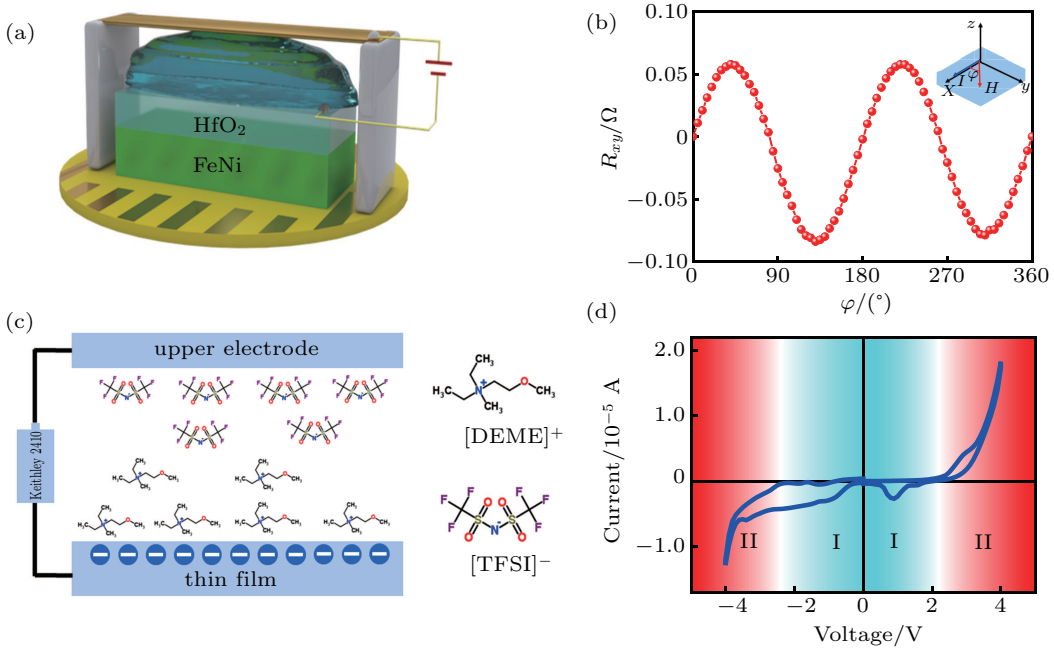


Fig. 1. (a) A schematic diagram of the transport test after the addition of the ionic liquid. (b) The measured planar Hall effect (PHE) curve at a magnetic field of 1000 Oe. The inset shows the relationship between the current and external magnetic fields. (c) Schematic diagram of the directional movement of anions and cations after application of a voltage by a voltage source Keithley 2410. (d) The *I*–*V* curve representing the amount of charge accumulation at the interface at different gate voltages.

PHE comes from the ferromagnetic layer $Ni_{80}Fe_{20}$ in the heterostructures, but HfO_2 also plays a pivotal role. HfO_2 can separate ionic liquid and $Ni_{80}Fe_{20}$ thin film, preventing direct chemical reactions between them. To achieve a large degree of regulation at a low voltage, it is necessary to increase the carrier density by other methods. The carrier density n is positively correlated with the capacitance C of the dielectric layer, that is, $n \sim CV_g/e$,[21] where V_g is the gate voltage and e is the electron charge. When V_g is constant, the carrier concentration is increased only by increasing the capacitance C. However, $C = \kappa\varepsilon_0/d$,[21] where κ is the permittivity of the dielectric, ε_0 is the vacuum permittivity, and d is the thickness of the dielectric layer. Therefore, increasing the permittivity of the dielectric layer can give rise to capacitance C, so that the carrier density is enhanced. However, HfO_2 has a high permittivity κ,[22–24] which can act as a role in increasing the carrier density. When a positive V_g is applied to the ionic liquid using the voltage source Keithley 2410, the cations and anions in the ionic liquid undergo directional movement. The cation $[DEME]^+$ moving toward the negative electrode attracted electrons and O^{2-} to move toward the top surface of the film, forming an EDL at the interface between the ionic liquid and the thin film. As shown in Fig. 1(c). This EDL is equivalent to a nano-gap capacitor, which creates a large electric field at the interface. The electric field generated by the 1-V gate voltage is approximately 10 MV/cm.[25] To characterize the extent of ion accumulation at different gate voltages, we tested the current–voltage (*I*–*V*) curve for the ionic liquid shown in Fig. 1(d). In the gate voltage range of -2.35 V $< V_g < 2.21$ V, the curve is very flat. In the gate voltage range of $V_g \leq -2.35$ V or $V_g \geq 2.21$ V, there is an obvious steep curve. Therefore, the *I*–*V* curve is divided into two parts.

The flat portion (I) is an electrostatically doped region, and the steep part (II) is the electrochemical reaction region.[26,27] The accumulation of ions at the interface of the latter is greater than that of the former.

Figure 2 shows the dependence of PHE on the electric field. When a 1-V gate voltage was applied to the ionic liquid, an EDL was formed at the interface of ionic liquid and film, defining this electric field as a positive electric field. The positive electric field causes a decrease of R_{xy}, as shown by the red curve in Fig. 2(a). When the angles between j_x and $\boldsymbol{M}$ are 45°, 135°, 225°, and 315°, the change rate of R_{xy} can reach 44%, 45%, 48%, and 44%, respectively. Under the removal of the positive V_g, the PHE curve was re-measured after at least 30 minutes, as shown by the blue curve in Fig. 2(a). The EDL disappears after removing the positive V_g, which leads to the curve almost returns to its original form. In other words, the electric field effect was eliminated. As shown in Fig. 2(a), the two curves are not completely coincident, which may be caused by a very small residual charge at the interface after applying a positive V_g. Under a negative V_g, the PHE curve moves upward, as shown by the pink curve in Fig. 2(a). It is difficult to control PHE in the negative electric field. Therefore, we have focused on the electric field regulation PHE at positive V_g. When the V_g is 0 V and 1 V, the effect of the electric field regulation PHE curve was very obvious. PHE was tested under different gate voltages as shown in Fig. 2(b). The meaning of A is to test when the gate voltage increases. At different gate voltages of 0.2 V-A, 0.4 V-A, 0.6 V-A, 0.8 V-A, and 1 V, the change rate of the transverse resistance R_{xy} is 7%, 9%, 20%, 32%, and 44% when the angle between j_x and $\boldsymbol{M}$ is 45°. Therefore, the electric field can continuously manipulate the PHE. The PHE curve can be restored to its original shape after removing the 1-V gate voltage for at least 30 minutes. It is preliminarily speculated that the effect of the electric field on PHE is non-volatile. To further investigate the variation of PHE when the V_g is removed, the PHE curves under different electric fields are also tested at a V_g interval of 0.2 V. The test results are shown in Fig. 2(c). The meaning of B is the curve when the V_g is removed. In the V_g range of 1 V and 0.2 V-B, the change of the PHE curve is relatively small. However, after waiting for 30 minutes, the PHE curve measured at +0 V returned to its original state. This indicates that the regulation of the electric field to the PHE is non-volatile. The PHE curve has biaxial symmetry. Under a saturated external magnetic field, the R_{xy} exhibits extreme values at 45°, 135°, 225°, and 315°.[13] The R_{xy} at these angles with different gate voltages is plotted in Fig. 2(d), which indicated the R_{xy} of the PHE curve to be regulated continuously by the electric field.

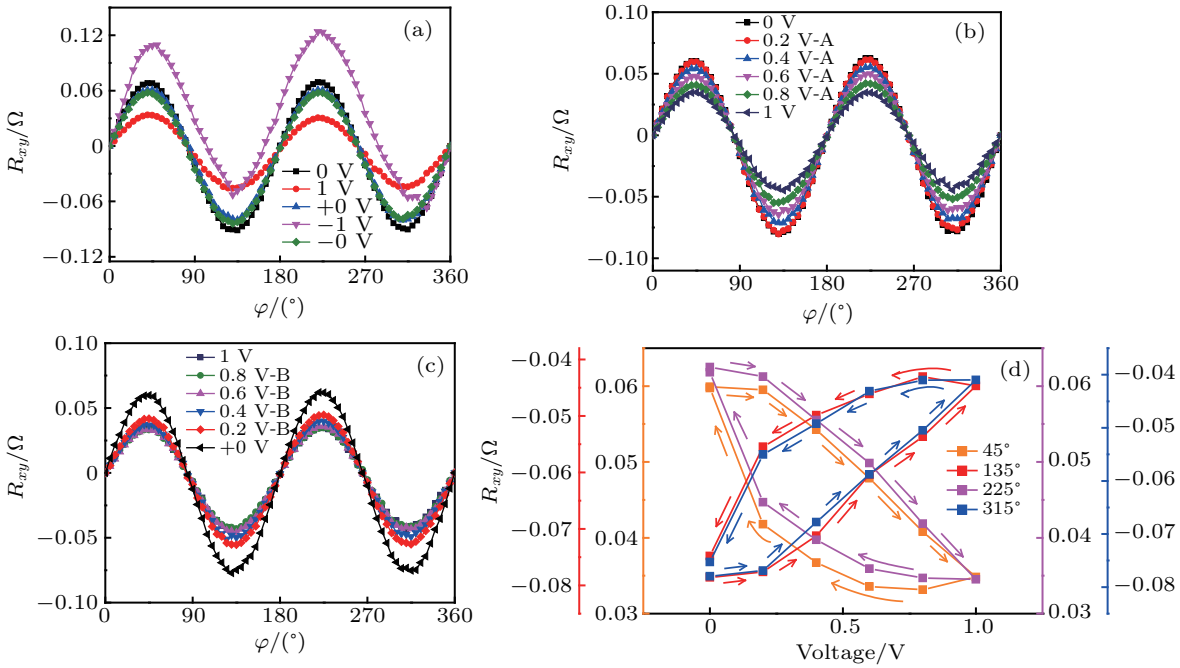


Fig. 2. (a) The measured PHE curve at different gate voltages. (b) At a voltage interval of 0.2 V, the gate voltage is gradually increased from 0 V to 1 V measured PHE curve. (c) At a voltage interval of 0.2 V, the PHE curve measured when the gate voltage is gradually reduced from 1 V to +0 V. (d) The curve of the transverse magnetoresistance R_{xy} at the extremum with the gate voltage.

In order to confirm further that the electric field can not only continuously regulate the PHE but also can achieve repeated regulation at the pulse gate voltage, the $Ni_{80}Fe_{20}$ film was repeatedly tested for PHE curves at gate voltages of 0 V and 1 V. When the V_g was increased or reduced, the waiting time to restart the test became different. After increasing the gate voltage from 0 V to 1 V, a stable electric field will be formed at the interface between ionic liquid and thin film after at least 5 min. After the gate voltage was reduced from 1 V to +0 V, it took at least 30 minutes to restart the test. As

mentioned above, the PHE regulation of $Ni_{80}Fe_{20}$ thin films by the electric field was non-volatile. The test results are shown in Fig. 3, suggesting that the data of the 1 V and 0 V repeated tests fit well, and the regulated amplitude is almost unchanged. This indicated that the electric field repeated the regulation of PHE. Similarly, the R_{xy} at the peak was extracted to further analyze the repeatability of electric field regulation. Figure 4 reflects the fact that the electric field has repeated regulation of the transverse magnetoresistance.

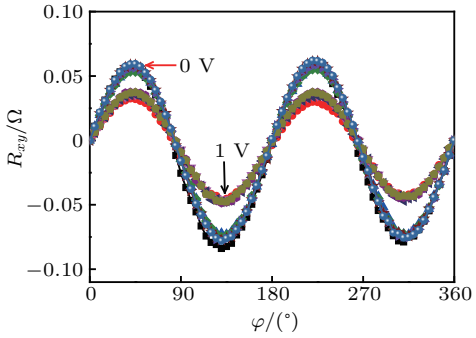


Fig. 3. The PHE curve was measured when the 1-V gate voltage was applied and removed repeatedly.

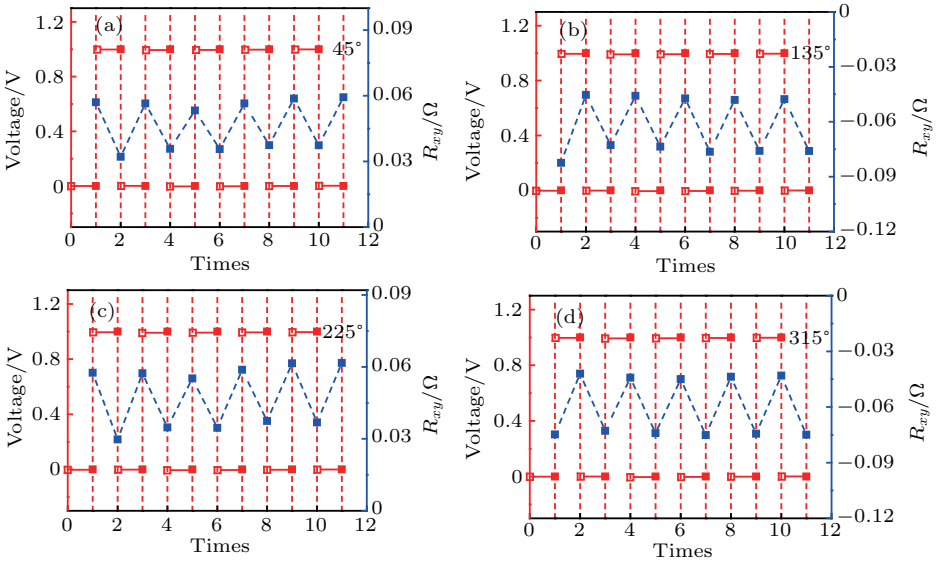


Fig. 4. (a)–(d) The control diagram of the transverse magnetic resistance R_{xy} at the extreme point when the 1-V gate voltage was repeatedly applied and removed.

Generally, the ionic liquid gate tuning magnetic properties is attributed to EDL. However, due to the screening effect, the thickness of the electric field-regulated magnetic film was limited. Especially at the gate voltage of 1 V, the R_{xy} of the 10-nm thick $Ni_{80}Fe_{20}$ film changes by 48% when the angle between j_x and M is 225°. Therefore, we speculate that in addition to EDL, there may be other mechanisms causing transverse magnetoresistance change. It had been reported that when a magnetic film was covered with oxide as an electrolyte gate, an inevitable oxidation process would exist in the magnetic layer.[28,29] Therefore, it is possible that the $Ni_{80}Fe_{20}/HfO_2$ heterostructures also have an O^{2-} migration process under the action of an electric field. When the positive V_g was applied, $[TFSI]^-$ accumulated on the top surface of HfO_2 would pump out O^{2-} in the $Ni_{80}Fe_{20}$ thin film. As shown in Fig. 5(a), the red solid circle represents O^{2-}, and the arrow indicates its moving direction. The content of O^{2-} in the $Ni_{80}Fe_{20}$ thin film was decreased. The weak oxidation state of the $Ni_{80}Fe_{20}$ thin film causes the transverse magnetoresistance to decrease.[17] This is the same as the test results.

The results of the electrical transport test have demonstrated that the electric field can regulate the PHE of the $Ni_{80}Fe_{20}$ thin film. In addition, the FMR *in situ* electric field control test results also prove that the electric field can regulate the magnetic properties of the $Ni_{80}Fe_{20}$ thin film. Transfer the test device shown in Fig. 1(a) to the FMR cavity, as shown in Fig. 5(b). The positive and negative electrodes were taken out with a 0.1-mm diameter Au wire. The DC magnetic field and the microwave magnetic field were in-plane of film. The test results are shown in Fig. 5(c). At the 4-V gate voltage, the signal of the FMR shifted to the left. After the V_g was removed, it returned to its original state. The differential absorption curve measured by the FMR is integrated as shown in the inset of Fig. 5(c). An asymmetric Lorentz function $\xi(H) = A[\Delta H \cos\delta + (H - H_r)\sin\delta]/[\Delta H^2 + (H - H_r)^2]$ was used to fit,[30] where H is an external magnetic field, A is the integral coefficient, ΔH is the half-width at half-maximum of the linewidth, δ is the phase difference between the real part and the imaginary part of the dynamic magnetic susceptibility, and H_r is the resonance field. After fitting, the resonance field H_r changed by 4 Oe at a gate voltage of 4 V. Besides, with applying gate voltage repeatedly, the shift of H_r is repetitive, as shown in Fig. 5(d). This illustrates further that the electric field can manipulate the magnetic properties of the $Ni_{80}Fe_{20}$ film.

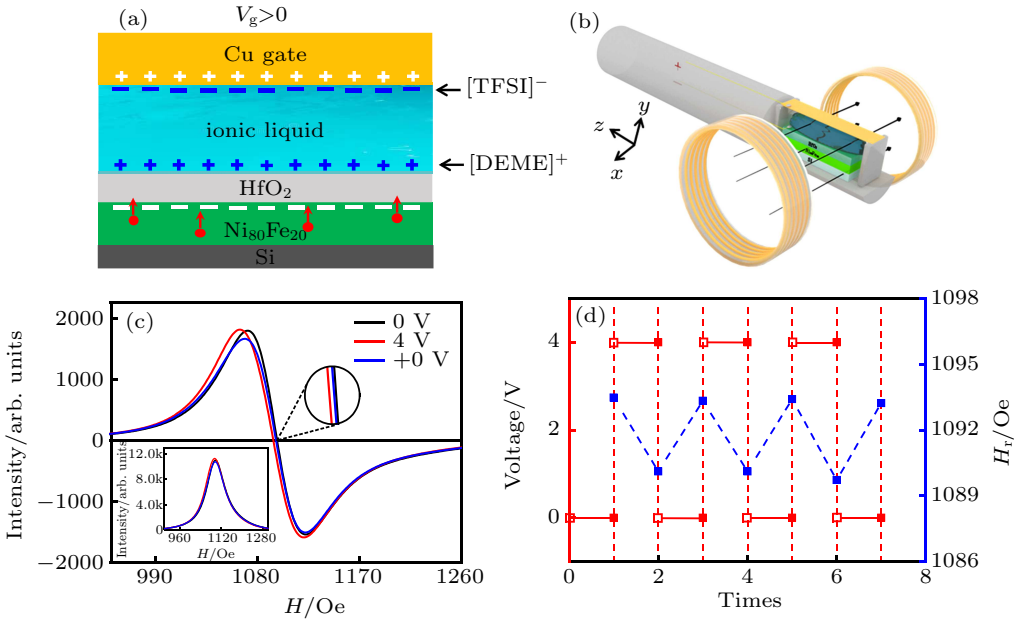


Fig. 5. (a) Schematic diagram of O^{2-} migration under positive gate voltage. O^{2-} is represented by a red solid circle, and the arrow indicates the direction in which it moves. (b) A schematic diagram of measuring ferromagnetic resonance (FMR). (c) The FMR differential spectrum measured by FMR at different gate voltages, the inset is the FMR integral spectrum. (d) The regulation diagram of the resonance field H_r when the 4-V gate voltage is applied and removed repeatedly.

4. Conclusions

In conclusion, the regulation of PHE of 10-nm thick $Ni_{80}Fe_{20}$ thin films has been achieved through the ionic liquid gate at ultra-low gate voltage, and this manipulation can be continuous within the 0-V to 1-V gate voltage. In particular, when the angle between j_x and M is 225°, the rate of change of R_{xy} reaches 48%. Furthermore, the results of the FMR test also illustrate that the ionic liquid gate can manipulate the magnetic properties of the $Ni_{80}Fe_{20}$ thin film. This provides a new idea for the design of spintronic devices with continuous electric field control under low energy consumption.

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