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Bias-controlled spin memory and spin injector scheme in the tunneling junction with a single-molecule magnet*

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A bias-controlled spin-filter and spin memory is theoretically proposed, which consists of the junction with a single-molecule magnet sandwiched between the nonmagnetic and ferromagnetic (FM) leads. By applying different voltage pulses $V_{\rm write}$ across the junction, the spin direction of the single-molecule magnet can be controlled to be parallel or anti-parallel to the magnetization of the FM lead, and the spin direction of SMM can be "read out" either by the magneto-resistance or by the spin current with another series of small voltage pulses $V_{\rm probe}$. It is shown that the polarization of the spin current is extremely high (up to 100%) and can be manipulated by the full-electric manner. This device scheme can be compatible with current technologies and has potential applications in high-density memory devices.

Keywords: single-molecule magnet, spin-transfer torque, spin-dependent electron tunneling

PACS: 72.15.Jf, 75.50.Xx, 75.60.Jk, 85.75.−d **DOI:** 10.1088/1674-1056/abd9b1

1. Introduction

Exploring the spin degree of freedom in quantum systems, such as single-molecule magnets (SMMs) and single-atom magnets, is an emerging field of promising applications, from nano-spintronics[1] to quantum information processing.[2] Conventionally, the smallest cell of the magnetic memory consists of 2–12 atoms.[3–5] Very recently, a single holmium atom on the thin film of the magnesium oxide (MgO) is demonstrated as the first example of a stable single-atom magnet.[6] The long relaxation time of the magnetic bit gives rise to the remanent magnetization, causing the limited size of the magnetic bit. Moreover, the experimental work in Ref. [7] shows that the nuclear spin of holmium atom can be reading and writing by a series of electric pulses with the help of spin-polarized scanning tunneling microscope (SP-STM)[8] and single-atom electron spin resonance (ESR) effect on an adjacent iron atom. The observed long lifetime (tens of seconds) and relaxation characteristics of the nuclear spin at the single-atom scale open the way to a completely new world of devices by which quantum logic may be implemented.

Owing to the recent development of material science and technology, there is increasing interest in the investigation of SMM[9] as the candidate for novel electronic devices,[10–12] such as random-accessing memories and spin injectors. Their properties depend on the combination of a large molecular spin and an easy axis magnetic anisotropy,[13,14] which results in a double-well energy potential resisting to the reversal of the magnetization. Controlling these molecules on different surfaces has attracted considerable attention as a prerequisite to single-molecule addressing.[15] In such studies, it was demonstrated that magnetic bistability persists and can be enhanced on some surfaces.[16] Especially efficiency of boosting the memory effect of both molecules and individual atoms can be achieved by depositing them on the thin insulating layer (*e.g.*, MgO) rather than directly on the metal surface.[17,18] Despite these remarkable results, how to electrically control the magnetic bistability of SMM between electrodes remains open. This is in part due to the limited experimental techniques that are sensitive enough to detect the magnetic properties of SMM.

In this work, we propose a novel type of spin filter, by a single magnetic atom coupled to an SP-STM or fabricating the junction with an SMM sandwiched between the normal metal and ferromagnetic (FM) lead. It is predicted that the spin direction of SMM can be readily read out and written by electrical bias pulse applied across the junction, and the highly spin-polarized current can be generated and controlled by this structure. Similar to the experimental work in Ref. [4], the manipulation of SMM's spin states needs a two-step scheme. First, a relative larger bias pulse is applied to obtain enough large spin-polarized currents to control a spin orientation of the SMM. The spin direction of SMM can be parallel or anti-parallel to the magnetization of the FM lead, depending on the direction of the bias pulse. And the measurement process is using a relatively low bias pulse to detect the magneto-resistance or spin-polarization coefficient of tunneling current.

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2. Model and Hamiltonian

The system we consider here consists of a single magnetic atom coupled to an SP-STM or sandwiching the junction with an SMM between two leads of normal and ferromagnetic metals as shown in Figs. 1(a) and 1(b), respectively. The Hamiltonian for both systems can be written as[19,20]

$$\begin{aligned} H &= \varepsilon_0 \sum_{\sigma} c_{\sigma}^{\dagger} c_{\sigma} + U c_{\uparrow}^{\dagger} c_{\uparrow} c_{\downarrow}^{\dagger} c_{\downarrow} - \mathcal{D}(S^z)^2 - J\boldsymbol{s}\cdot\boldsymbol{S} \\ &\quad -\Delta B(s^z + S^z) + \sum_{\boldsymbol{k},\sigma,\alpha} \varepsilon_{\boldsymbol{k}\sigma} a_{\alpha\boldsymbol{k}\sigma}^{\dagger} a_{\alpha\boldsymbol{k}\sigma} \\ &\quad + \sum_{\boldsymbol{k},\sigma,\alpha} (t_{\alpha} a_{\alpha\boldsymbol{k}\sigma}^{\dagger} c_{\sigma} + t_{\alpha}^{*} c_{\sigma}^{\dagger} a_{\alpha\boldsymbol{k}\sigma}). \end{aligned} \tag{1}$$

Here, ε_0 is the on-site energy of the lowest unoccupied molecular orbital (LUMO) of the SMM, which can be tuned by the gate voltage applied to the SMM, $c_{\sigma}^{\dagger}$ (c_{σ}) is the electron creation (annihilation) operator with σ as the spin index. The second term is the Coulomb interaction, the third term describes the magnetic uniaxial anisotropy, and J is the exchange coupling between the SMM and an electron in the LUMO. Since we assume the easy axis of the molecule as z axis in the spin space, $\Delta B(s^z + S^z)$ describes the Zeeman splitting by the external magnetic field applied along the easy axis of the molecule, where the g factor and Bohr magneton $\mu_{\rm B}$ have been absorbed into ΔB. $a_{\alpha\boldsymbol{k}\sigma}^{\dagger}$ ($a_{\alpha\boldsymbol{k}\sigma}$) is the creation (annihilation) operator for the electron of momentum $\boldsymbol{k}$, spin σ, and energy $\varepsilon_{\boldsymbol{k}\sigma}$ in lead $\alpha = \{\mathrm{L},\mathrm{R}\}$. The coupling between the SMM and two leads is described by t_{α}, which is assumed to be independent of momentum $\boldsymbol{k}$ and spin σ.

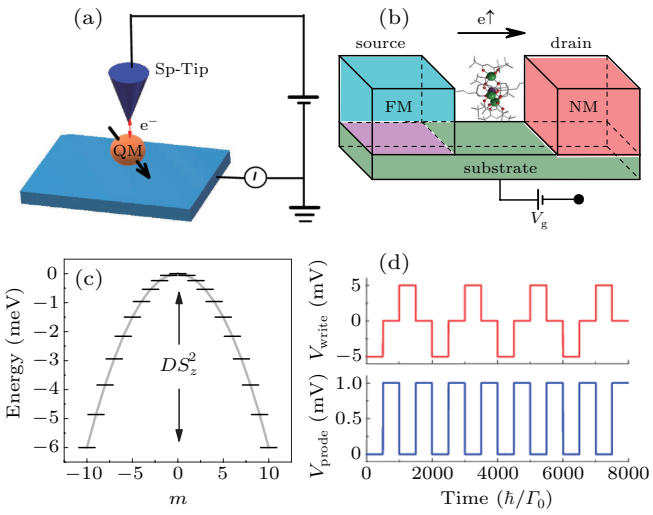


Fig. 1. The schemes of spin memory by a single-atom magnets coupled to an SP-STM tip and an SMM sandwich between the FM and NM leads are shown in panels (a) and (b), respectively. (c) Energy spectrum of the molecule with a tunneling barrier $\mathcal{D}S_z^2 \approx 60$ K for $\mathrm{Mn}_{12-\mathrm{ac}}$. (d) The time varying bias pulses of $V_{\rm write}$ (red) and $V_{\rm probe}$ (blue) for "writing" and "reading" processes.

The Hamiltonian of the individual SMM is described by the first five terms in Eq. (1). By defining $\boldsymbol{S}_T = \boldsymbol{s} + \boldsymbol{S}$, the eigenvalue m of S_T^z is a good quantum number due to commutation relation $[S_T^z, H] = 0$. In the following, $|n,m\rangle$ represents n electrons occupying the SMM with the total spin as m. The eigen-energies can be obtained as[21]

$$\begin{aligned} \varepsilon_{|0,m\rangle} &= -\mathcal{D}m^2 - \Delta B m, \\ \varepsilon_{|1,m\rangle} &= \varepsilon_0 - \Delta B m + J/4 - \mathcal{D}(m^2 + 1/4) \pm \varepsilon'(m), \\ \varepsilon_{|2,m\rangle} &= 2\varepsilon_0 + U - \mathcal{D}m^2 - \Delta B m, \end{aligned} \tag{2}$$

where $\varepsilon'(m) = \sqrt{\mathcal{D}(\mathcal{D}-J)m^2 + (J/4)^2(2S+1)^2}$.

In order to read out the magnetization of the SMM by a small bias voltage pulse, we need a magnet molecule with relative large anisotropy. In this paper, we use SMM of $\mathrm{Mn}_{12-\mathrm{ac}}$ with parameters as $\mathcal{D} = 0.06$ meV, $S = 10$, $J = 0.2$ meV, and $U = 25$ meV.[22,23] This SMM has two degenerate ground states of $S_z = \pm 10$ and a relative large tunneling barrier $\mathcal{D}S_z^2 \approx 60$ K. Additionally, we assumed the molecule is weekly coupled between two electrodes, so that the transport process is dominated by the sequential tunneling through the SMM energy level, while the cotunneling and direct tunneling can be neglected safely at low temperature. For the weak coupling between the SMM and two leads, the master equation approach is valid for our analysis. The SMM's magnetization can be defined as $M_{\rm SMM} = \sum_i m_i P_i$, and the current of spin σ flowing through the SMM can be written as $I_{\sigma} = (I_{\rm L}\sigma - I_{\rm R}\sigma)/2$, where $I_{\rm L}\sigma$ ($I_{\rm R}\sigma$) stands for the spin current flowing from the left (right) lead to the SMM as

$$I_{\alpha\sigma} = -(e/h)\sum_{i,f}(n_i - n_f) R_{\alpha\sigma}^{f\to i} P_f, \tag{3}$$

where P_i is the probability of finding the SMM in state $|i\rangle$, and $R_{\alpha\sigma}^{f\to i} = \Gamma_{\alpha\sigma}[f(\varepsilon_i - \varepsilon_f - \mu_{\alpha})\langle i|c_{\sigma}^{\dagger}|f\rangle^2 + f(\varepsilon_i - \varepsilon_f + \mu_{\alpha})\langle f|c_{\sigma}^{\dagger}|i\rangle^2]$ denotes the rate of transition between $|i\rangle$ and $|f\rangle$. $\Gamma_{\alpha\sigma} = 2\pi D_{\alpha\sigma}|t_{\alpha}|^2$ is the line-width function for lead α with $D_{\alpha\sigma}$ as the density of states at Fermi energy and f_{α} as the Fermi distribution function of lead α with temperature T_{α} and Fermi energy μ_{α}. The left electrode is the ferromagnetic metal with spin polarization defined as $\mathcal{P}_{\rm FM} = (D_{L\uparrow} - D_{L\downarrow})/(D_{L\uparrow} + D_{L\downarrow})$. We assume the line-width functions as $\Gamma_{L\uparrow} = \Gamma_0(1 + \mathcal{P}_{\rm FM})$ and $\Gamma_{L\downarrow} = \Gamma_0(1 - \mathcal{P}_{\rm FM})$. We take $\mathcal{P}_{\rm FM} > 0$ representing the major spin-up electrons. Following the numerical method suggested in Refs. [19,24], the time dependence of probability $P_i(t)$ and steady state probability $P_i(t \to \infty)$ can be obtained by solving a series of equations. The total current is equal to $I = \sum_{\sigma}(I_{\rm L}\sigma - I_{\rm R}\sigma)/2$ and the spin polarization coefficient of spin current is defined as

$$\eta = \frac{I_{\alpha\uparrow} - I_{\alpha\downarrow}}{I} \times 100\%, \tag{4}$$

by which the totally-polarized spin current has the coefficient as $\eta = \pm 100\%$.

3. Result and discussion

First, we demonstrate how the voltage pulse controls the magneto-resistance switch and spin-polarization in a FM/SMM/NM junction. At the low temperature, the SMM

must be in one of the two bistable ground states $M_{\rm SMM} = \pm S$. For the sake of definition, we set $M_{\rm SMM} = +S$ parallel to the magnetic polarization of the left FM lead. As shown in Fig. 1(d), an electric pulse $V_{\rm write}$ of the amplitude ± 5 mV is applied to the device, which changes its sign at every $1000\hbar/\Gamma_0$ ($\cong 20$ ns for $\Gamma_0 = 8$ GHz[24]) and drive the SMM's polarization to $\pm S$ periodically as shown in Fig. 2(d). It is noteworthy that the pulse signal only lasts $500\hbar/\Gamma_0$ ($\cong 10$ ns) every time it reaches ± 5 mV, and then returns to zero quickly. During the time of $V_{\rm write} = 0$, another series of the pulse $V_{\rm probe}$ denoted by the blue line in Fig. 1(d) is applied to read out the SMM's magnetization. The total voltage pulse $V = V_{\rm write} + V_{\rm probe}$ is shown in Fig. 2(a) and the magnetization response of the SMM to the applied pulse is shown in Fig. 2(d). Obviously, the magnetization of the SMM turns to $+S$ direction under the "-5 mV" pulse, while the "$+5$ mV" pulse will reverse the magnetization to $-S$. As shown in Figs. 2(b) and 2(c), the "$+5$ mV" pulse will drive the spin-polarization coefficient $\eta_{\rm probe}$ from 100% to -100%, indicating that only spin-down electron can tunnel from the FM lead to the NM lead. A high magneto-resistance is obtained as a result of the anti-parallel magnetic configuration between FM and SMM. Then, after the "-5 mV" pulse is applied, the electron current driven by $V_{\rm probe}$ rises quickly and polarization coefficient is increased to $+100\%$, which means only spin-up electron can tunnel through the junction and the low magneto-resistance is obtained. In addition, we show the different $\mathcal{P}_{\rm FM}$ of the FM lead in Fig. 2. Although the current signal $I_{\rm probe}$ will be suppressed or enlarged by the FM lead's magnetization $\mathcal{P}_{\rm FM}$, the polarization coefficients of the spin current in both spin directions are robustly fixed at $\eta_{\rm probe} = \pm 100\%$. As a result, the two bistable ground states $M_{\rm SMM} = \pm S$ can be read out by $V_{\rm probe}$ with the different spin-polarization coefficients and the magneto-resistances.

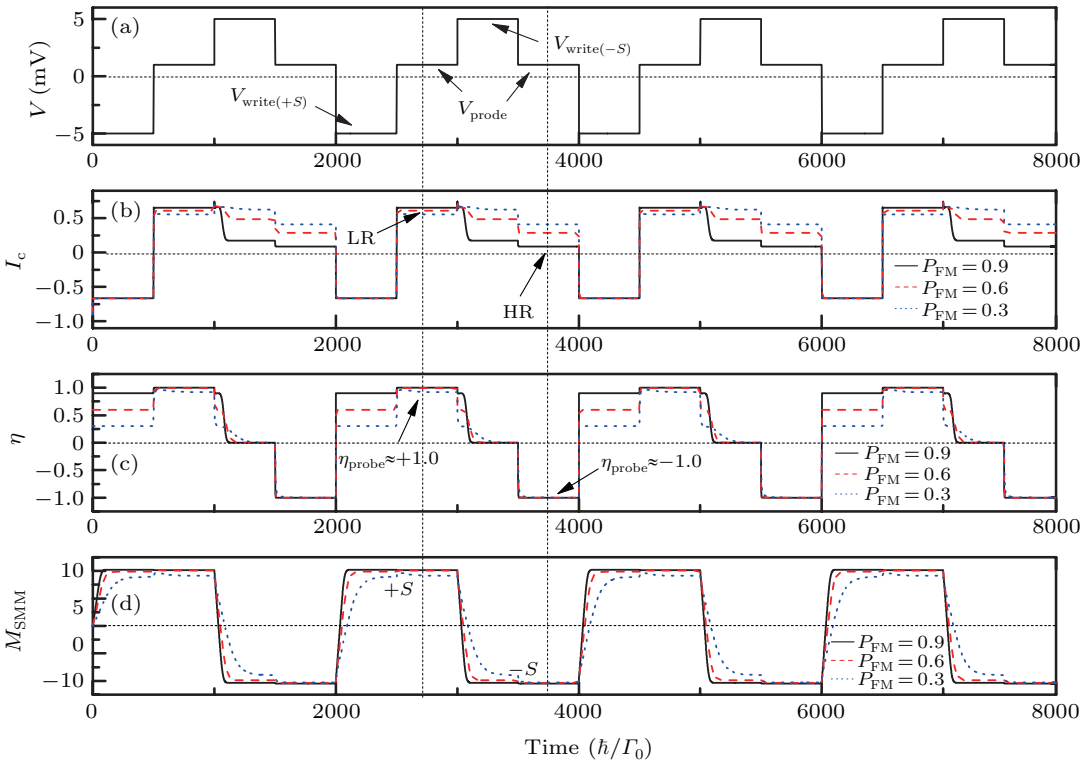


Fig. 2. (a) The voltage pulse of $V = V_{\rm write} + V_{\rm probe}$. (b) The tunneling currents (scaled by $e\Gamma_0/\hbar$) for different polarizations of FM leads $\mathcal{P}_{\rm FM}$. (c) The spin-polarization coefficient η. (d) The polarization of SMM $M_{\rm SMM}$. The initial state is $P_{|0,\pm S\rangle} = 0.5$ with $\varepsilon_0 = 0.5$ meV and the temperature $T = 0.5$ K.

In order to clarify the underlying mechanism, we plot the probabilities of molecular states for $\mathcal{P}_{\rm FM} = 0.95$. In Fig. 3, the probabilities of the states except $|0,\pm S\rangle$, $|1,S+1/2\rangle$, and $|1,-S-1/2\rangle$ are almost vanishing. In Fig. 3(a), it is shown that before "-5 mV" pulse is applied, $P_{|1,-S-1/2\rangle} + P_{|0,-S\rangle}$ is equal to 1, which means the SMM's spin is fixed at $-S$ direction. After $V_{\rm write} = -5$ mV is applied, $P_{|1,-S-1/2\rangle}$ and $P_{|0,-S\rangle}$ decrease rapidly while $P_{|0,S\rangle}$ and $P_{|1,S+1/2\rangle}$ increase, indicating SMM's magnetization is reversed from $-S$ to $+S$ by the spin–torque interaction.[25] The reversing process takes less than $100e\Gamma_0/\hbar$ (≈ 2 ns), and then the electron tunneling process is dominated by the transition between $|0,S\rangle$ and $|1,S+1/2\rangle$. In Fig. 3(b), we find that although the small voltage plus $V_{\rm probe} = 1$ mV gradually changes the probabilities of $|0,S\rangle$ and $|1,S+1/2\rangle$, we still have $P_{|0,S\rangle} + P_{|1,S+1/2\rangle} \approx 1$, indicating the tunneling process is still dominated by the transporting between these two states. Only spin-up electrons can tunnel between the states $|0,S\rangle$ and $|1,S+1/2\rangle$, which results in $\eta \approx 100\%$. In Fig. 3(c), when the "$+5$ mV" pulse is applied, $P_{|1,-S-1/2\rangle}$ and $P_{|0,-S\rangle}$ increase quickly, which means the SMM's spin direction is reversed from $+S$ to $-S$. Even though the small voltage plus $V_{\rm probe} = 1$ mV will affect $P_{|1,S+1/2\rangle}$ and

$P_{|0,S\rangle}$, the tunneling current is still dominated by the transition between $|1,S+1/2\rangle$ and $|0,S\rangle$. As a result, only spin-down electron can tunnel through the junction with $\eta_{\text{probe}} \approx -100\%$. Due to the high spin-polarization of the FM lead, the spin-up electrons are more easier to occupy the LUMO levels and the spin-up current $I_{|0,S\rangle\leftrightarrow|1,S+1/2\rangle}$ will be much larger than spin-down electron current $I_{|0,-S\rangle\leftrightarrow|1,-S-1/2\rangle}$, resulting in an HR–LR switch. No mater how the tunneling current is suppressed or enlarged by the FM lead, the extremely high spin-polarization coefficients with $\eta = \pm 100\%$ remain robustly.

In Fig. 4, we plot the magnetization of the SMM, spin-polarization coefficient η and spin currents against the amplitude of the voltage pulse. In Figs. 4(a) and 4(b), the arrows indicate the scanning directions of the voltage pulse, while the scanning process is assumed to be slow enough to allow the system to relax to the steady state. Figures 4(a) and 4(b) show that both η and the magnetization of the SMM exhibit hysteresis loop when scanning back and forth. χ_+ (χ_-) represents the critical point when SMM's magnetization is reversed from $+S$ ($-S$) to $-S$ ($+S$). The electron transport process in the regime $V_{\chi_-} < V < V_{\chi_+}$ is quite different, compared with the the regime $V < V_{\chi_-}$ and $V > V_{\chi_+}$. As shown in Fig. 4(d), if we choose two points $V_{\eta_-} = -5$ mV and $V_{\eta_+} = +5$ mV faraway from the critical points, the spin-polarization η can be obtained as

$$\eta_{\text{write}} = \begin{cases} \eta_- = \mathcal{P}_{\text{FM}}, & (V \ll V_{\chi_-}), \\ \eta_+ = 0, & (V \gg V_{\chi_+}). \end{cases} \tag{5}$$

In this regime, the spin-polarization η can be up to $\mathcal{P}_{\text{FM}}$ or suppressed to zero according to the bias direction. Similar effect has also been reported in other nano MTJ structures in Refs. [26–29], which is also called the spin-diode effect and experimentally demonstrated in Ni/InAs/Au sandwich structure.[30] But in the regime $V_{\chi_-} < V < V_{\chi_+}$, it is find that the SMM can keep its original polarization. Therefore, η in this regime can be observed as

$$\eta_{\text{probe}} \approx \begin{cases} +100\%, & (V \text{ scans from } V_{\eta_+} \text{ to } V_{\eta_-}), \\ -100\%, & (V \text{ scans from } V_{\eta_-} \text{ to } V_{\eta_+}). \end{cases} \tag{6}$$

According to Figs. 4(b) and 4(c), purely spin-polarized current can be generated by the SMM-MTJ, even with the FM lead of lower polarization $\mathcal{P}_{\text{FM}} = 0.4$. These highly polarized currents are originated from the spin selective property of the SMM. As discussed in Fig. 3, the magnetization of the SMM can be manipulated in one of the two metastable spin states at low temperature. The small V_{probe} can not reverse the magnetic directions of SMM so that only electrons with spin parallel to the SMM can flow through the junction,[10] which leads to the purely polarized current. As a result, to read out the spin direction of the SMM and magneto-resistance, the small bias pulse V_{probe} should be chosen in this bias regime $V_{\chi_-} < V_{\text{probe}} < V_{\chi_+}$.

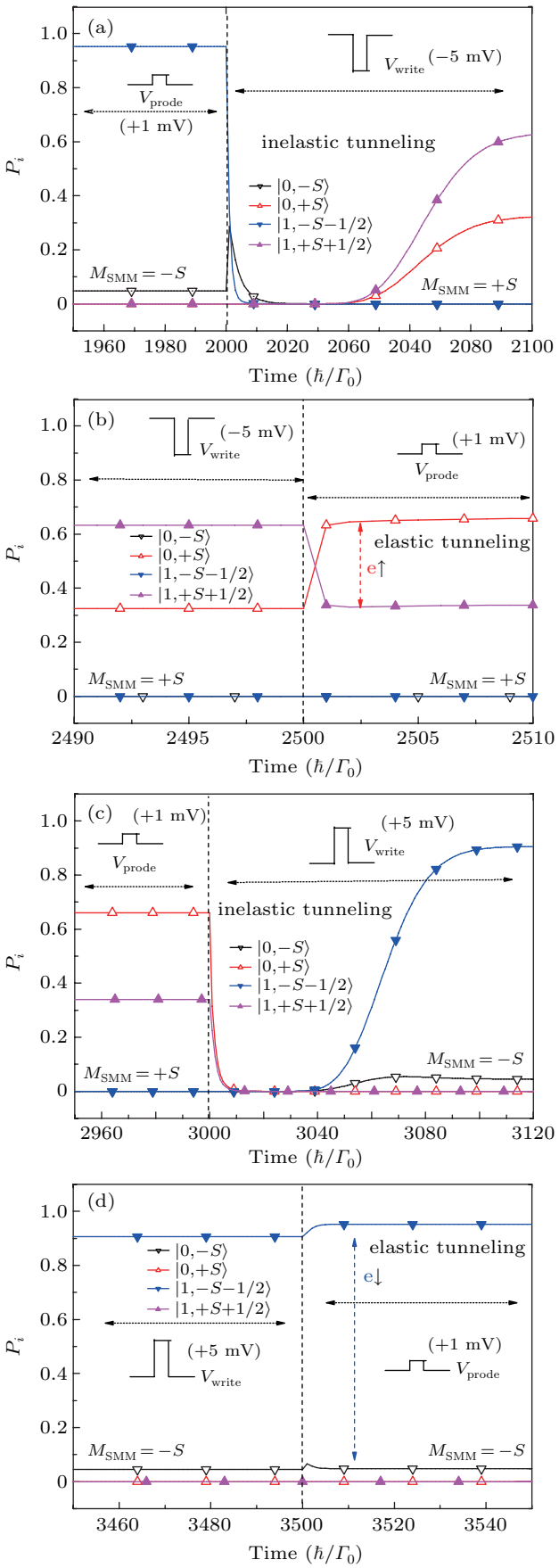


Fig. 3. The time-dependent probabilities of SMM states from V_{probe} to $V_{\text{write}(+S)}$, from $V_{\text{write}(+S)}$ to V_{probe}, from V_{probe} to $V_{\text{write}(-S)}$, and from $V_{\text{write}(-S)}$ to V_{probe} are shown in panels (a)–(d), respectively. The dashed lines (red or blue) in panels (b) and (d) represent the spin tunneling processes between states $|1,\pm S\pm 1/2\rangle$ and $|0,\pm S\rangle$.

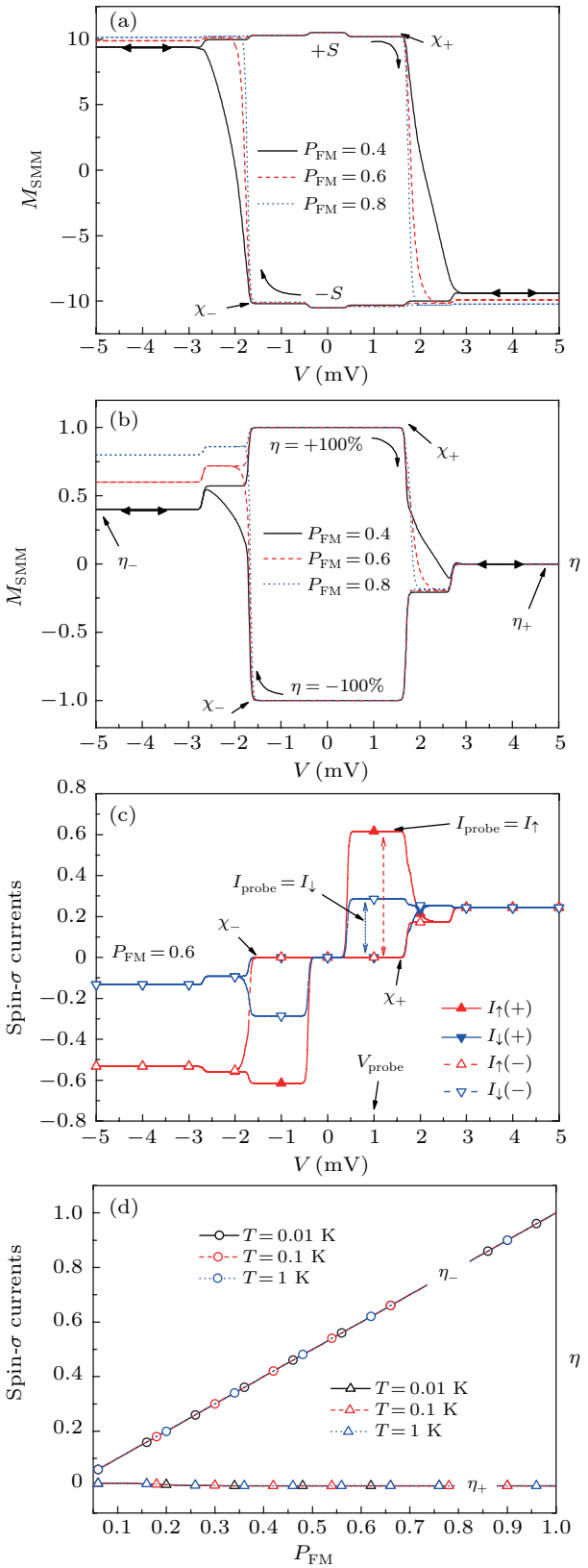


Fig. 4. The hysteresis loops of the SMM's polarization and spin-polarization coefficient η with the bias voltage scanning back and forth are plotted in panels (a) and (b), respectively. (c) The spin-σ currents (scaled by $e\Gamma_0/\hbar$) with the bias scanning from -5 mV to $+5$ mV (solid triangle) and from $+5$ mV to -5 mV (hollow triangle). (d) Spin-polarization coefficient $\eta_\pm$ in the large bias regime against the spin-polarization of FM lead $\mathcal{P}_{FM}$ for different temperatures.

In Fig. 5, we explored the magnetic reversal process of SMM in the presence of an external magnetic field ΔB. It is shown that both of reversal point χ_+ (χ_-) can be shifted to higher or lower voltage regime according to the different direction of ΔB. As discussed in Fig. 4, the suitable "read out" voltage V_{probe} should be chosen in the regime $V_{\chi_-} < V_{probe} < V_{\chi_+}$, while a relatively large external magnetic field ΔB will shift this bias windows to a higher or lower voltage regime. In Fig. 5(b), we also scan ΔB back and forth slowly enough to allow the system relax to steady state where Λ_+ (Λ_-) represents the critical point when the magnetization of the SMM from $+S$ ($-S$) to $-S$ ($+S$). Λ_+ and Λ_- can also be shifted to a higher or lower ΔB regime, according to the different bias directions. This result is similar with the traditional spin-transfer torque effect in MTJ structure. For the "$-$" bias pulse with the electrons tunneling from FM to NM, the polarization of SMM favors the direction parallel to the polarization of FM. And for "$+$" bias pulse, the polarization of SMM will favor the direction anti-parallel to the the magnetization of FM lead. Similar phenomenon of magnetic reversal point shifted by spin current has also been experimentally demonstrated in Fe atoms nano-structure,[4] where the bias pulse and FM lead are accomplished with SP-STM. However, if the external magnetic field is so small that the given $V_{probe} = 1$mV is still in the regime $[V_{\chi_-}^{\Delta B}, V_{\chi_+}^{\Delta B}]$, the pulse scheme we proposed can still work.

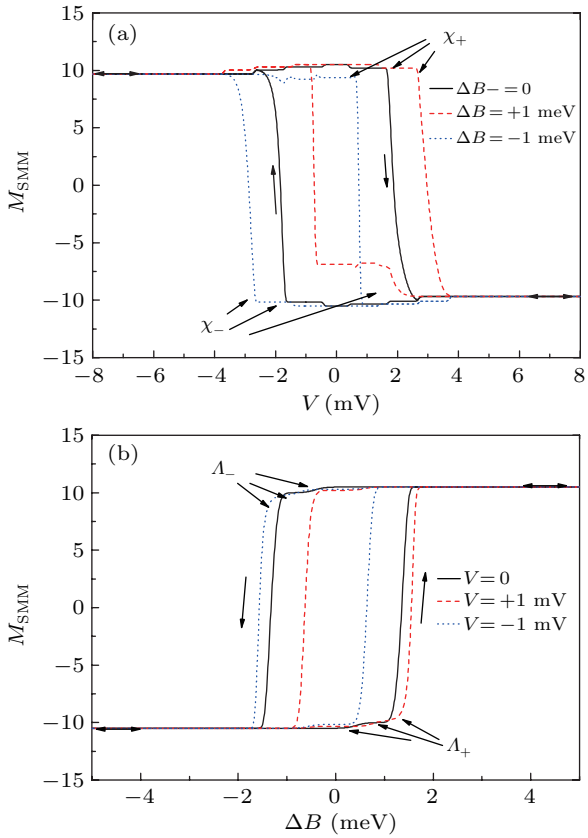


Fig. 5. (a) Magnetic hysteresis loops of SMM for different external magnetic fields ΔB, with bias voltage scanning back and forth. (b) Magnetic hysteresis loops of SMM for different bias voltages, with ΔB scanning back and forth.

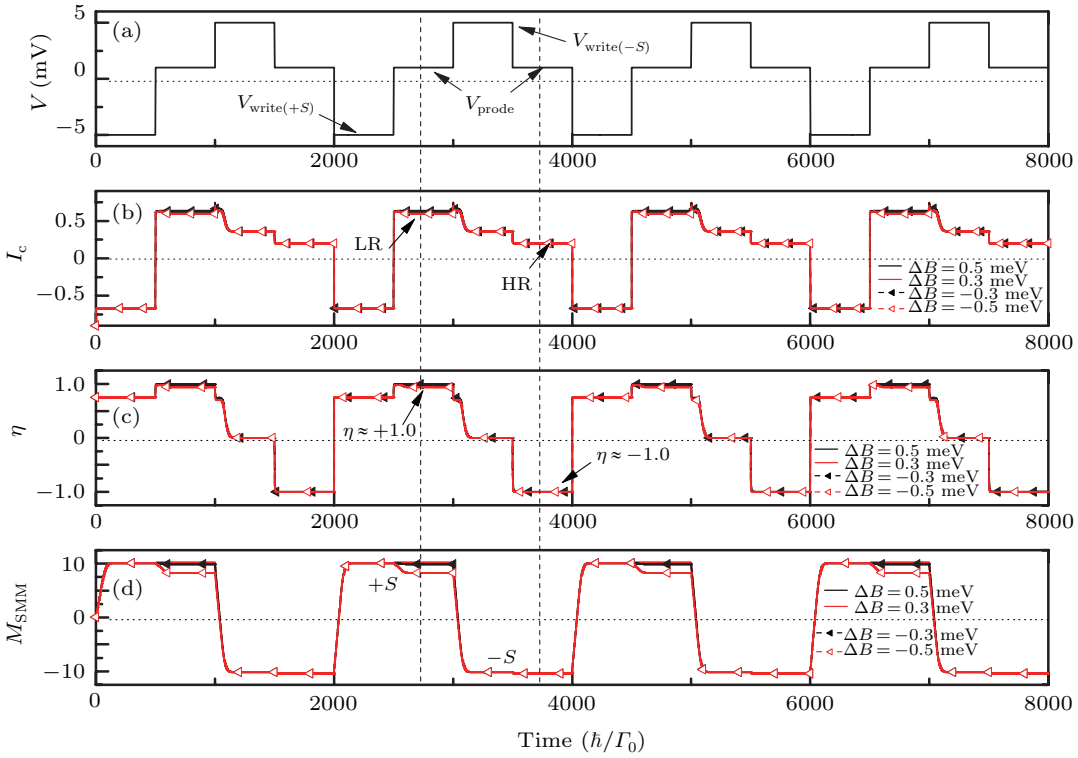


Fig. 6. Panel (a) shows the applied voltage pulse. The tunneling currents (scaled by $e\Gamma_0/\hbar$), spin-polarization coefficient η, and the polarization of SMM responding to the voltage pulse with different external magnetic fields and $\mathcal{P}_{\rm FM} = 0.7$ are shown in panels (b)–(d), respectively. The temperature is taken at $T = 0.5$ K.

In Fig. 6, it is demonstrated the process of writing and reading out spin states of an SMM-MTJ under small magnetic fields. When ΔB changes from $+0.5$ meV to -0.5 meV, the high/low magneto-resistance switch as well as extremely high spin-polarized current can still be detected by $V_{\rm probe} = 1$ mV. Note that, for the $B = -0.5$ meV in Fig. 6(c) the polarization coefficient η cannot reach 100%, which means purely spin-polarized current cannot be obtained at this regime. It is because the magnetic field $\Delta B = -0.5$ meV will make SMM's magnetization more easily turns to $-S$ direction. In Fig. 6(d), we can see the magnetization of SMM at this regime cannot keep exactly at $M_{\rm SMM} = 10$ but approximately at $M_{\rm SMM} \approx 8$. As a result, besides the transition between $|0, S\rangle$ and $|1, S+1/2\rangle$, other states such as $|1, S-1/2\rangle$ and $|0, S-1\rangle$ will take part in the transition process and the spin-down electrons can tunnel through the junction in this ΔB regime.

4. Conclusion

In summary, we theoretically investigate the tunneling process in an FM/SMM/NM heterostructure. By applying different bias pulse, the polarization of SMM can be controlled parallel or anti-parallel to the polarization of the FM layer, and the high and low magneto-resistances can be obtained. Such HR–LR switching can be read out by a relatively lower bias voltage $V_{\chi_+} < V_{\rm probe} < V_{\chi_+}$. Furthermore, extremely high spin-polarized currents with $\eta = \pm 100\%$ can be readily generated in a suitable window of $V_{\rm probe}$, no matter what the external magnetic field and the polarization of FM lead are chosen. This scheme is fully electrically controlled and is robust against the external magnetic field or FM lead. Our scheme demonstrates that the polarization of SMM can be reversed by the spin-transfer torque. The present device is a good candidate for future spintronic devices, *e.g.*, the bias-pulse-controlling spin memory or spin generator.

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