Giant spin torque efficiency in single-crystalline antiferromagnet Mn_2Au films

Shaohai Chen[†], Xinyu Shu[†], Jing Zhou, Chenghang Zhou, Qidong Xie, Tieyang Zhao, Liang Liu, Weinan Lin and Jingsheng Chen^{*}

ABSTRACT Antiferromagnetic (AFM) materials have attracted wide attention in spin-orbit torque (SOT)-based spintronic due to its abundant spin-dependent properties and unique advantage of immunity against external field perturbations. To act as the charge-to-spin conversion source in energy-saving spintronic devices, it is of great importance for the AFM material to possess a large spin torque efficiency (ξ_{DL}). In this work, using the spin torque ferromagnetic resonance (ST-FMR) technique and a $\text{Mn}_2\text{Au}/\text{NiFe}(\text{Py})$ bilayer system, we systemically study the ξ_{DL} of AFM Mn_2Au films with different crystal structures. Compared with polycrystalline Mn_2Au with effective $\xi_{\text{DL}} < 0.051$, we show a much larger ξ_{DL} of ~ 0.333 in single-crystal Mn_2Au , which arises from the large spin Hall conductivity instead of electrical resistivity. Moreover, with a further contribution of interfacial effects, the effective ξ_{DL} of single-crystalline $\text{Mn}_2\text{Au}/\text{Py}$ system increases to 0.731, which is more than two times larger than the value of ~ 0.22 reported for the $\text{Mn}_2\text{Au}/\text{CoFeB}$ system. By utilizing the large ξ_{DL} of Mn_2Au in a perpendicularly magnetized $\text{MnGa}/\text{Mn}_2\text{Au}$ system, energy-efficient deterministic magnetization switching with a current density at $\sim 10^6 \text{ A cm}^{-2}$ is achieved. Our results reveal a significant potential of Mn_2Au as an efficient SOT source and shed light on its application in future AFM material-based SOT integration technology.

Keywords: antiferromagnetic, spin torque efficiency, electrical magnetization switching, spin torque ferromagnetic resonance

INTRODUCTION

In recent studies, antiferromagnets (AFMs) have been revealed to be a promising class of materials for developing next-generation spintronic applications [1–5]. In a spin-orbit torque (SOT) magnetic random-access mem-

ory (SOT-MRAM), owing to the negligible stray magnetic fields and the resilience to external magnetic field perturbations, AFM materials can effectively suppress the crosstalk between stored information and thereafter help reduce the recording bit size [6]. In the recording bit of the SOT-MRAM device with a damping-like spin Hall channel/ferromagnetic (FM) free layer structure, it is of fundamental importance for the spin Hall channel materials to possess a high and controllable SOT efficiency ξ_{DL} , because the writing current I_{write} is proportional to $1/\xi_{\text{DL}}$ [7]. Compared with the nonmagnetic (NM) metallic materials with $\xi_{\text{DL}} > 0.5$, such as β -W and Pt-derived Pt/Ti multilayer structure [7–12], a lot of studies show that the metallic AFM materials, such as PtMn, IrMn and IrMn₃, can provide comparable or even larger ξ_{DL} , as shown in Table 1 [13–17]. This encourages us to explore AFM materials as a spin current source and realize energy-efficient electrical magnetization switching in an AFM/FM structure with perpendicular magnetic anisotropy (PMA) [13,18].

Among potential AFM materials, the collinear Mn_2Au material has a great possibility of providing large ξ_{DL} since the gold atoms result in a strong spin-orbit coupling (SOC) [19,20]. Regarding previous studies on the ξ_{DL} of Mn_2Au film, Singh *et al.* [21] reported a large $\xi_{\text{DL}} = 0.22$ in a $\text{Mn}_2\text{Au}/\text{CoFeB}$ bilayer system. However, from the dual Mn_2Au (101) and (200) peaks shown in the X-ray diffraction (XRD) spectrum, it can be noticed that their Mn_2Au film is not fully chemically ordered. It has been demonstrated that the ξ_{DL} in a single-crystalline IrMn/FM system can be ten times larger than that of the polycrystalline counterpart [14–16]. At the same time, studies on the Néel vector switching in the Mn_2Au film suggest

Department of Materials Science and Engineering, National University of Singapore, Singapore 117576, Singapore

[†] These authors contributed equally to this work.

^{*} Corresponding author (email: msecj@nus.edu.sg)

Table 1 Summary of the resistivity (ρ_{xx}), spin Hall conductivity (σ_{SH}) and effective SOT efficiency (ξ_{DL}) of the AFM materials with single-crystalline and polycrystalline structures of our work and recently reported results

Material	ρ_{xx} ($\mu\Omega$ cm)	σ_{SH} ($10^3(\hbar/2e)\Omega^{-1}\text{cm}^{-1}$)	ξ_{DL}	Refs
Mn ₂ Au (single-crystalline)	176	>3.10	0.731	This work
Mn ₂ Au (polycrystalline)	370	<0.23	0.051	This work
Mn ₂ Au (single-crystalline)	150	1.46	0.22	[22]
IrMn (single-crystalline)	~85	~7	0.6	[14,15]
IrMn (polycrystalline)	173	0.58	0.09	[14,16]
IrMn ₃ (single-crystalline)	160	2.19	0.3	[16]
IrMn ₃ (polycrystalline)	183	0.55	0.10	[16]
PtMn (single-crystalline)	180	0.29	0.052	[36]
PtMn (polycrystalline)	~140	~1.5	0.24	[13]
PdMn (single-crystalline)	270	0.12	0.032	[36]

that the pure (002)-oriented Mn₂Au can be switched by a smaller current density than Mn₂Au with multiple crystal orientations [1,22–26]. These results indicate that single-crystalline Mn₂Au film may possess a larger ξ_{DL} . Furthermore, it has been reported that the (002)-oriented single-crystalline Mn₂Au possesses an exceptionally higher Néel temperature ($T_N \sim 1500$ K) [27]. Combined with the body-centered tetragonal (bct) lattice structure, these features will facilitate the integration of the Mn₂Au material into novel spin valves that incorporate a low symmetry tetragonal FM layer with PMA, such as L1₀-Mn_xGa [28–30].

In this work, we fabricate (002)-oriented single-crystalline and polycrystalline Mn₂Au films and measured corresponding spin torque ferromagnetic resonance (ST-FMR) spectra in Mn₂Au/NiFe(Py) bilayer structures. For the single-crystalline Mn₂Au film-based Mn₂Au/Cu/Py and Mn₂Au/Py systems, the effective ξ_{DL} values are obtained as >0.333 and 0.731, respectively, whose difference is mainly attributed to the strong interface contribution at the Mn₂Au/Py interface. For the polycrystalline Mn₂Au film, the effective ξ_{DL} value without the interface contribution is obtained as <0.051. Unlike the attribution of Singh *et al.* [21], we show this relatively small effective ξ_{DL} is because the spin Hall conductivity of polycrystalline Mn₂Au film is an order of magnitude smaller than that of single-crystalline Mn₂Au film. By utilizing Mn₂Au as the spin current source in a perpendicularly magnetized MnGa/Mn₂Au structure, we verify that the large ξ_{DL} of the Mn₂Au film can effectively reduce the critical switching current density J_c to an order of magnitude smaller than the reported $\sim 10^7$ A cm⁻² in MnGa/NM(Ta or Pt) systems [31,32]. Our results show that the Mn₂Au

material is a promising candidate as a spin current source in AFM material-based spintronic devices.

EXPERIMENTAL SECTION

All samples were fabricated by a magnetron sputtering system with base vacuum pressure smaller than 1.33×10^{-6} Pa. The sample structures are (i) substrate/Mn₂Au (25 nm)/Py (10 nm)/SiO₂ (4 nm) and (ii) substrate/MnGa (20 nm)/Mn₂Au (30 nm)/SiO₂ (4 nm), respectively. For samples of structure (i), to obtain different crystal textures in the Mn₂Au film, single-crystal SrTiO₃ (001) and naturally oxidized Si(001) substrates with substrate temperature $T_s = 600$ and 320°C were respectively utilized. After the Mn₂Au layer was cooled to room temperature, a 10-nm Py layer and a 4-nm SiO₂ layer were subsequently deposited as the detecting layer for ST-FMR measurement and the capping layer to protect the sample from oxidation, as shown in the inset of Fig. 1b. For simplification, the two samples are named as STO-sub sample and Si-sub sample in the discussion. For the sample of structure (ii), the MnGa layer was deposited on single-crystal STO(001) substrate with $T_s = 560^\circ\text{C}$. The magnetic hysteresis (M - H) loops and the crystalline textures of all thin film samples were measured by a vibrating sample magnetometer (VSM) system and a high-resolution XRD system, respectively. To measure the angle-dependent ST-FMR spectrum, the STO-sub and Si-sub samples were patterned into $30\text{ }\mu\text{m} \times 50\text{ }\mu\text{m}$ micro-strip matrix by standard photo-lithography processes, where the long edge of the strip rotated from $\varphi_m = 0^\circ$ to 360° corresponding to the substrate edge, as shown in Fig. 2c and the inset of Fig. 3a. The angle between the external magnetic field H_{ext} and the long edge of the

Figure 1 (a, b) XRD spectra of the STO-sub and Si-sub samples, respectively. The schematic in (b) shows the sample structure.

device strip was $\varphi_H = 45^\circ$. To carry out the harmonic Hall voltage response measurements, Hall bar strip with a size of $65 \mu\text{m} \times 10 \mu\text{m}$ was patterned on all three samples. The anomalous Hall effect (AHE) and harmonic Hall voltage response measurements were carried out by a physical property measurement system (PPMS-Quantum Design).

RESULTS AND DISCUSSION

The XRD spectra of the STO-sub and Si-sub samples are shown in Fig. 1a and b, respectively. For the STO-sub sample, standard Mn_2Au (002), (004) and (006) diffraction peaks are observed in Fig. 1a, which verifies a high crystallinity for the Mn_2Au film layer. For the Si-sub sample, in addition to the Si substrate, the θ - 2θ spectrum in Fig. 1b does not show any obvious diffraction peak, indicating a polycrystalline structure for its Mn_2Au film layer. In Fig. S1 of the Supplementary information, we show the in-plane M - H loops of the two samples after the field cooling process. The exchange bias phenomenon in the M - H loops of both samples indicates the anti-ferromagnetism in their Mn_2Au layer.

To study the effective ξ_{DL} of the STO-sub and Si-sub samples, in the ST-FMR measurements, the frequency f is varied from 4 to 12 GHz. The corresponding output voltage signal (V_{mix}) against the external magnetic field (H_{ext}) curves are shown in Fig. 2a and b, respectively. All

curves are fitted by the following equation [33]:

$$V_{\text{mix}} = V_s \frac{\Delta H^2}{\Delta H^2 + (H_{\text{ext}} - H_{\text{res}})^2} + V_A \frac{\Delta H(H - H_{\text{res}})}{\Delta H^2 + (H_{\text{ext}} - H_{\text{res}})^2}, \quad (1)$$

where ΔH and H_{res} are the linewidths and the resonant field, respectively. The V_s and V_A are the amplitudes for the symmetric and antisymmetric voltage components, respectively. In the ST-FMR model, V_s and V_A are associated with the magnitudes of in-plane torque ($\tau_{\parallel}$) and the out-of-plane torque ($\tau_{\perp}$), respectively. We show the decomposition of both symmetric and antisymmetric components for each sample at $f = 9$ GHz in Fig. 2d and e, respectively. The effective ξ_{DL} can be obtained by [15,33]

$$\xi_{\text{FMR}} = \frac{V_s}{V_A} \frac{e\mu_0 M_s t_{\text{Mn}_2\text{Au}} t_{\text{Py}}}{\hbar} \sqrt{1 + \frac{4\pi M_{\text{eff}}}{H_{\text{res}}}}, \quad (2)$$

and

$$1/\xi_{\text{FMR}} = \left(1 + \frac{\hbar}{e} \frac{\xi_{\text{FL,eff}}}{4\pi M_s t_{\text{FM}} t_{\text{Mn}_2\text{Au}}} \right) / \xi_{\text{DL}}, \quad (3)$$

where e , μ_0 , $\hbar$, $t_{\text{Mn}_2\text{Au}}$, t_{Py} , M_s and M_{eff} are the electron charge, permeability of free space, reduced Planck constant, the thicknesses of Mn_2Au and Py layer, the saturation magnetization of the Py layer and its effective value in the ST-FMR measurement, respectively. Based on the in-plane harmonic Hall voltage response results [7,15], as shown in the Fig. S2 of the Supplementary information, we show the field-like SOT efficiency ξ_{FL} of the STO-sub sample is nearly two orders of magnitude smaller than its ξ_{DL} . By combining Equations (2) and (3), the effective ξ_{DL} value in our work is represented by the ξ_{FMR} value.

According to the M - H loops, the M_s values of the two samples were both obtained at $\sim 600 \times 10^3 \text{ A m}^{-1}$. At the same time, as shown in the Fig. 2f, the M_{eff} values of the two samples were obtained by fitting the f versus the extracted H_{res} curves with the in-plane Kittel equation:

$$f = \frac{\gamma}{2\pi} \sqrt{(H_K + H_{\text{res}})(H_K + H_{\text{res}} + 4\pi M_{\text{eff}})}, \quad (4)$$

where H_K is the in-plane magnetic anisotropy. The fitted M_{eff} are 585×10^3 and $596 \times 10^3 \text{ A m}^{-1}$ for the STO-sub and Si-sub samples accordingly. With the obtained parameters, the saturated effective ξ_{DL} values at $f \geq 9$ GHz of the STO-sub and Si-sub samples were calculated as 0.731 and 0.051, respectively. This agrees well with our expectation that the crystal structure of the Mn_2Au film can greatly affect its ξ_{DL} . In IrMn- and IrMn₃-based systems, besides the spin-dependent scattering caused by the crystal structure, it can be noted that the interface exchange coupling is another reason for obtaining a large effective ξ_{DL} [14–16,34].

Figure 2 (a, b) ST-FMR spectra of the STO-sub and Si-sub samples with f ranging from 4 to 12 GHz, respectively. (c) Schematic and image of the device for the ST-FMR measurements. (d, e) The V_{mix} (circles) versus the H_{ext} curves with $f = 9 \text{ GHz}$ and corresponding fitting results for the Si-sub and STO-sub samples, respectively. The dotted and dashed lines correspond to the contributions of V_A and V_S , respectively. (f) The f versus H_{res} curves and corresponding fitting lines of the two samples with different substrates.

To further understand the influence of the magnetic structure on the ξ_{DL} of the Mn_2Au film, we measured the dependences of the effective ξ_{DL} on the in-plane device rotation angle φ_m of the STO-sub and Si-sub samples. As shown in Fig. 3a, the effective ξ_{DL} of the STO-sub sample shows a fourfold anisotropy, of which the maximum (~ 0.731) and the minimum (~ 0.499) values locate at $\varphi_m = 45^\circ$ (135°) and 0° (90° and 180°), respectively. This is consistent with our previous results in Ref. [14], which indicates the magnetic easy axis of the Mn_2Au film in the STO-sub sample is along the $[110](45^\circ)$ and $[1\bar{1}0](135^\circ)$ orientations. As shown in the inset in Fig. 3b, the dual in-plane magnetic easy axes results are well in

line with the predicted magnetic structure of the (002)-oriented single-crystal Mn_2Au [20,21,35]. By carrying out the in-plane harmonic Hall voltage response measurement in a reference sample with the structure of STO substrate/ Mn_2Au (25 nm)/Cu (1 nm)/Py (10 nm)/ SiO_2 (4 nm), as shown in the Fig. S3 of the Supplementary information, the obtained effective ξ_{DL} value is slightly larger than 0.333. Compared with the STO-sub sample, the effective ξ_{DL} of the $\text{Mn}_2\text{Au}/\text{Cu}/\text{Py}$ system is smaller due to the suppression of the interface contributions, such as interfacial spin transmission and interfacial exchange coupling. For the Si-sub sample, its ξ_{DL} does not show a clear variation with different φ_m . This is under-

Figure 3 (a) The dependence of ξ_{DL} on the in-plane angle ϕ_m of the STO-sub and MgO-sub samples. The insert schematic shows the relation between the ϕ_m and the [100] orientation of the substrate. The x -axis is parallel to the long side of the device strip. (b) The resistivities of the Mn_2Au films deposited on different substrates. The insert schematic shows the dual magnetic easy axes directions of the (002)-oriented single-crystal Mn_2Au film.

standable due to the distribution of disordered domains in the Mn_2Au layer leads to disordered magnetization arrangements. What should be noticed here is that the interfacial exchange coupling maybe still exists in the Si-sub sample, as evidenced by the center offset M - H loop in Fig. S1. Therefore, the effective ξ_{DL} value of polycrystalline Mn_2Au without the interface contribution is <0.051 .

The effective ξ_{DL} of the single crystalline Mn_2Au film without the interface contribution is one order of magnitude larger than that of the polycrystalline Mn_2Au film. To understand this, based on the four-probe technique, we measured the resistivity $\rho_{xx, \text{Mn}_2\text{Au}}$ of the Mn_2Au film deposited on the STO and Si substrates, which are 176 ± 5

and $370 \pm 5 \mu\Omega \text{ cm}$, respectively, as shown in Fig. 3b. According to the relation of $\xi_{\text{DL}} = (2e/h)T_{\text{int}}\rho_{xx}\sigma_{\text{SH}}$ [7] and by utilizing the same spin transparency $T_{\text{int}} \approx 0.61$ [21], the spin Hall conductivity σ_{SH} of the single-crystalline and polycrystalline Mn_2Au films without the interface contributions was calculated as >3.10 and $<0.23 \times 10^3 (\hbar/2e) \Omega^{-1} \text{ cm}^{-1}$. For our Mn_2Au films with different crystalline structures, therefore, it can be known that the spin Hall conductivity instead of the electrical resistivity makes a greater contribution to determining the ξ_{DL} . The σ_{SH} of the single-crystalline Mn_2Au film is an order of magnitude larger than that of the polycrystalline Mn_2Au film. This can be attributed to the highly ordered gold atoms in the single-crystalline Mn_2Au leading to a stronger spin Hall effect, which causes a larger electron trajectory bending for the up and down spins [36]. On the other hand, as shown in Table 1, compared with the reported $\rho_{xx, \text{Mn}_2\text{Au}} = 150 \mu\Omega \text{ cm}$ and $\sigma_{\text{SH}} = 1.46 \times 10^3 (\hbar/2e) \Omega^{-1} \text{ cm}^{-1}$ from Singh *et al.* [21], our single-crystalline Mn_2Au film has both larger $\rho_{xx, \text{Mn}_2\text{Au}}$ and σ_{SH} , resulting in the larger ξ_{DL} .

To verify the large ξ_{DL} of Mn_2Au can lead to an energy-efficient electrical magnetization switching, in a perpendicularly magnetized system with the structure of STO substrate/ MnGa (20 nm)/ Mn_2Au (30 nm)/ SiO_2 (4 nm), as shown in the Fig. 4a, we studied the electrical magnetization switching properties. We choose MnGa as the FM layer because it possesses not only a strong PMA but also a neglectable lattice constant mismatch (0.5%) with the single-crystal STO(001) substrate [29,30]. The squareness of the AHE loop, as shown in the Fig. 4c, confirms that the MnGa layer has a good out-of-plane magnetic easy axis. In the electrical magnetization switching measurements, as shown in the Fig. 4b and d, deterministic magnetic switching phenomenon is observed with an external magnetic field H_x along the current direction. Identical measurements were carried out in a reference sample with the structure of STO substrate/ MnGa (20 nm)/ SiO_2 (4 nm), in which no switching phenomenon is observed. This verifies that the switching is not derived from the FM layer itself [37]. With $H_x = 0.5 \times 10^6/4\pi \text{ A m}^{-1}$, the critical switching current was obtained at $J_c = 6.1 \times 10^6 \text{ A cm}^{-2}$, which is an order of magnitude smaller than the reported $J_c \sim 5.0 \times 10^7 \text{ A cm}^{-2}$ with $H_x = 5 \times 10^6/4\pi \text{ A m}^{-1}$ and $\xi_{\text{DL}} \sim 0.07$ in other $\text{MnGa}/\text{NM}(\text{Pt or Ta})$ systems and $J_c \sim 2.0 \times 10^7 \text{ A cm}^{-2}$ with $H_x = 0.2 \times 10^6/4\pi \text{ A m}^{-1}$ and $\xi_{\text{DL}} \sim 0.23$ in an AFM $\text{PtMn}/\text{Hf}/\text{CoFeB}$ system [13,31,32]. Based on the out-of-plane harmonic Hall voltage response measurement [38–40], as

Figure 4 (a) Schematic showing how the Mn₂Au layer induces a spin current and forms torques on the MnGa layer. (b) The AHE measurement result of the sample. (c) The current induced magnetization switching results of the sample with different external magnetic fields H_x along the current direction. (d) The extracted critical switching current density J_c versus H_x curve from (c).

shown in Fig. S4 of the Supplementary information, the ξ_{DL} and ξ_{FL} were calculated as 0.784 and 0.077, respectively. Therefore, it can be known that the reduction of J_c is derived from the larger ξ_{DL} of the Mn₂Au layer, which again validates our expectations. In other perpendicularly magnetized AFM/FM systems, it was reported that weakening the “spin memory loss” by inserting a thin Hf layer at the AFM/FM interface could help increase the effective ξ_{DL} [13]. This provides a further method to increase the effective ξ_{DL} of the Mn₂Au-based system.

CONCLUSION

In summary, based on the ST-FMR technique, we studied how the crystal structure can influence the ξ_{DL} of the AFM Mn₂Au film. Due to the combined effect of large spin Hall conductivity and interfacial contributions, the effective ξ_{DL} of the (002)-oriented single-crystalline Mn₂Au/Py system increases to 0.731, which is more than ten times larger than $\xi_{DL} = 0.051$ of the polycrystalline Mn₂Au/Py system. Our largest effective ξ_{DL} is comparable to the reported effective $\xi_{DL} > 0.6$ in single-crystalline

IrMn/Py system [16–18]. In a perpendicularly magnetized MnGa/Mn₂Au system, we verify the large ξ_{DL} of the Mn₂Au film can help to reduce the critical switching current density J_c to the order of $\sim 10^6 \text{ A cm}^{-2}$. Our results show that the Mn₂Au material is a promising candidate as spin current sources and will promote the application of AFM materials in future energy-efficient spintronics devices.

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Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Experimental details and supporting data are available in the online version of the paper.

Shaohai Chen is a research fellow of the National University of Singapore. He received a PhD degree in 2016 from Fudan University. During 2014–2015, he was a joint-training PhD student at the National University of Singapore. His research interest focuses on high-anisotropy magnetic materials, 2D materials and spintronics.

Xinyu Shu received his Bachelor degree from Harbin Institute of Technology and Master degree from Beijing University of Technology. He is now a PhD candidate at the Department of Material Science and Engineering, National University of Singapore. His current research interest is spintronics in metallic, oxide and low dimension heterostructures.

Jingsheng Chen received his PhD degree from Lanzhou University, China, in 1999. He joined Nanyang Technological University as a post-doctor in 1999–2001 and the Data Storage Institute, A*STAR as research scientist in 2001–2007. In the year of 2007, he joined the National University of Singapore as an assistant professor and was promoted to associate professor in 2013. His research interest includes high-anisotropy magnetic materials, spintronics, multiferroic materials, and nanostructure magnetic materials.

单晶反铁磁 Mn_2Au 薄膜的高自旋矩转换效率

陈绍海[†], 舒新愉[†], 周靖, 周成航, 谢奇东, 赵铁扬, 刘亮, 林伟南, 陈景升

摘要 反铁磁(AFM)材料因其丰富的自旋相关特性和独特的抗外部磁场扰动的优势, 在基于自旋轨道转矩(SOT)的自旋电子学研究中引起了广泛关注. 反铁磁材料能够成为低能耗自旋电子器件中电流-自旋流的高效转换源的一个重要标准是具有较大的自旋矩转换效率(ξ_{DL}). 在这项工作中, 我们基于自旋转矩铁磁共振(ST-FMR)技术, 系统地研究了 $\text{Mn}_2\text{Au}/\text{Py}$ (坡莫合金)双层膜结构中具有不同晶体结构反铁磁 Mn_2Au 薄膜的 ξ_{DL} . 我们发现多晶 Mn_2Au 薄膜的有效 ξ_{DL} 值低于0.051, 而单晶 Mn_2Au 薄膜的有效 ξ_{DL} 值高于0.333, 相较于薄膜电阻率的改变, 这一增长被归因于薄膜自旋霍尔电导率的改变. 此外, 单晶 $\text{Mn}_2\text{Au}/\text{Py}$ 系统中由于存在界面效应的贡献, 其有效 ξ_{DL} 值为0.731, 比所报道的 $\text{Mn}_2\text{Au}/\text{CoFeB}$ 系统的最大 ξ_{DL} 值(~ 0.22)高两倍以上. 利用 Mn_2Au 的高 ξ_{DL} , 我们在具有垂直磁化结构的 $\text{MnGa}/\text{Mn}_2\text{Au}$ 系统中, 实现了翻转电流密度为 $\sim 10^6 \text{ A cm}^{-2}$ 的电控磁翻转, 此结果比其他传统金属材料所需电流密度小了一个量级. 本文揭示了 Mn_2Au 材料作为高效自旋矩转换源的良好潜力, 并有助于推进反铁磁材料在基于自旋矩效应集成器件中的研发及应用.