Evidence for asymmetric rotation of spins in antiferromagnetic exchange-spring

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Abstract
We demonstrate an asymmetric rotation of the antiferromagnetic (AFM) spins in the exchange-spring driven by perpendicularly magnetized Co/Pt. The static and dynamic behaviors of the twisted spin structure are directly revealed by a combination of element specific soft-x-ray absorption spectra and magnetoresistance measurements. X-ray magnetic linear dichroism spectra as a function of AFM thickness clarify the features of the whole exchange-spring, while the interfacial uncompensated spins are identified by the x-ray magnetic circular dichroism spectra. Moreover, the observed tunneling anisotropic magnetoresistance (TAMR) in AFM-based junctions based on this asymmetric rotation provides an electrical approach to monitoring the dynamic twist of the AFM spins. These investigations not only provide a deep insight into the spin structure of the exchange coupling layers but would also advance the development of AFM spintronics.

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The exchange coupling between the ferromagnetic (FM) and antiferromagnetic (AFM) layers is a long-standing issue in fundamental physics and modern spintronics focusing on the spin-valve effect. Recently, the understanding of this intriguing phenomenon has been renewed by the emerging AFM spintronics, which promote the transition of AFM from static supporting materials to functional materials. Because of the superior features of AFM, such as negligible ferromagnetic stray fields and the rigidity to magnetic field perturbations compared with FM [1–3], much effort has been made in manipulating the AFM spins to realize AFM-based memory resistors and tunneling anisotropic magnetoresistance (TAMR) [3–8]. A deep insight into the AFM spin structures is crucial for the control of AFM spins, which are hard to probe since they exhibit no net magnetic moment in a single layer. Although experimental techniques, such as nuclear magnetic resonance and neutron diffraction, have been adopted to identify the antiferromagnetism and magnetic structures of AFM [9, 10], they are constrained by the sample thickness, which should be thick enough to generate visible signals. Besides the earlier theories of exchange bias that focused on the interface of FM/AFM [11], x-ray magnetic linear dichroism (XMLD) and x-ray magnetic circular dichroism (XMCD) are preferable to detect the arrangement of interfacial spins simultaneously in FM and AFM, taking advantage of the element specificity and shallow probing depth for the total electron yield (TEY) mode [12–15]. Nevertheless, previous XMLD experiments were generally focused on the collinear AFM, such as NiO and FeRh [3, 13], regardless of the noncollinear AFM, due to their complex spin axes.

As the common AFM materials used widely in magnetic tunnel junctions, chemically-disordered FeMn and IrMn (L12-IrMn3) have a noncollinear magnetic structure in the bulk, making the spins hard to identify, let alone manipulate [11, 16–18]. It is worth pointing out that beyond the current passive role of AFM in pinning FM, spins in ultrathin AFM layer could also be motivated by the neighbored FM, leading to a twisted exchange-spring in AFM, accompanied by uncompensated moments at the interface [12, 13, 19, 20]. Direct recognition of the exchange-spring is in great demand because it is essential for manipulating AFM moments as well as realizing the functionality of AFM layers [21]. Perpendicular exchange coupling between FM and AFM offers a good opportunity to monitor the AFM spins in the exchange-spring, where the AFM spins are motivated towards out-of-plane orientations in the usual case [22]. Especially for FeMn and IrMn processing six-fold symmetry of in-plane spins owing to 3Q spin density wave, out-of-plane component of spin orientations could make the spins possible to be detected using XMLD.

In our work, we observe an asymmetric rotation of FeMn spins triggered by perpendicularly magnetized Co/Pt, and provide compelling evidence for these asymmetric spin structures at opposite remanent states combining XMLD and XMCD. We further demonstrate the FeMn-based TAMR effect on the basis of this asymmetric behavior. The comparison between the TAMR comprising of FeMn and IrMn reveals different characters of the twisted spin structure, providing an electrical means to directly recognizing the dynamic rotation procedure of the AFM exchange-spring.

Samples were deposited using magnetron sputtering on thermally oxidized Si substrate with a Ta buffer layer at room temperature. Pt(10)/[Co(0.5)/Pt(1)]4/Co(0.5)/Pt(0.6)/FeMn(6)/AlOx(2)/Pt(5) (unit in nanometers) stacks with an insert layer of 0.6 nm-thick Pt (marked as sample A) were prepared and fabricated into tunnel junctions of dimensions 20 × 12 μm² using photolithography and ion milling. Figure 1(a) shows the magnetoresistance curve of the junction at vertical H, where a hysteresis window with a stable high-resistance state (HRS) at +H and low-resistance state (LRS) at −H indicates a partial rotation of FeMn moments from...
in-plane [right inset of figure 1(a)] to out-of-plane (left inset) direction [6]. This observed asymmetry between the two states (HRS/LRS) is induced by the downward component of interfacial FeMn spins, which is an imprint of the domain pattern of as-deposited Co/Pt [6, 23, 24]. To be specific, the magnetic flux leakage from magnetron guns is able to make downward spins favorable in as-deposited Co/Pt, which is confirmed by its remanence. The initial tilt makes FeMn spins prefer to rotate downward at $-H$ while they align almost in-plane at $+H$. Note that the measured TAMR here indicates the existence of asymmetric rotation of FeMn spins at $+H/-H$, laying the foundation for the following detection of asymmetric spin structures in the FM/AFM exchange coupling system.

In order to determine the twisted spin structure of FeMn, soft-x-ray absorption spectroscopy (XAS) and XMLD in the TEY mode, which are element-specific techniques, were utilized. Since XMLD is featured by different absorptions of light with its linear polarization vector $E$ orientated parallel or perpendicular to the spin axis, it stands out as a unique method to identify the spin orientations in AFM, despite the absence of net moment [13]. It is noteworthy that in TEY mode the probing depth is limited by the escape length ($\sim 5$ nm) of the photogenerated electrons. To detect the Mn $L$-edge signals in FeMn, multilayers of substrate/Pt(10)/[Co(0.5)/Pt(1)]$_t$/Co(0.5)/Pt(0.6)/FeMn($t_1$)/Pt(2) ($t_1 = 2, 6, 15, 30$ nm) were adopted to carry out the XMLD experiment, where the core [Co/Pt]/FeMn structure and interface features are identical to that of sample A. As shown in figure 1(b), the incident angle was fixed at 60°, while the linear polarization of the photon was rotated by 90° to obtain the in-plane and out-of-plane (60° from the surface) $E$. In this scenario, the XMLD signals primarily
arise from the spin arrangement in the antiferromagnetic exchange-spring of FeMn away from the [Co/Pt]/FeMn interface. Since XMLD data is very sensitive to the stability of the photon energy, analogous experiments have been repeated to confirm the results. An alternative approach that rotates the sample while fixing the linear polarization direction was adopted to obtain XMLD spectra, showing similar signals. The details are discussed in figure S1 in the supplementary data.

The Mn L-edge XMLD spectra as a function of FeMn thicknesses $t_1$ are shown in figure 2(a), which are obtained by the differences in absorption intensities ($I_{\text{in-plane}} - I_{\text{out-of-plane}}$) of each sample at the two remanent states after magnetization (left column: $-1 \text{ kOe}$, right column: $+1 \text{ kOe}$): (a) substrate/Pt(10)/[Co(0.5)/Pt(1)]$_4$/Co(0.5)/Pt(0.6)/FeMn($t_1$)/Pt(2) ($t_1 = 2, 6, 15, 30 \text{ nm}$); (b) substrate/Pt(10)/[Co(0.5)/Pt(1)]$_4$/Co(0.5)/Pt(0.6)/IrMn ($t_2$)/Pt(2) ($t_2 = 6, 10 \text{ nm}$); (c) and (d) show the arrangement of spins in the multilayers when $\delta_{\text{critical}} < t < \delta_W$ and $t > \delta_W$, respectively. The bracketed parts denote the detection ranges of the XMLD experiments. The [Co/Pt]/FeMn interface marked by the grey arrow is described in the text.

![Figure 2. Mn L-edge XMLD difference in absorption intensities ($I_{\text{in-plane}} - I_{\text{out-of-plane}}$) of each sample at the two remanent states after magnetization (left column: $-1 \text{ kOe}$, right column: $+1 \text{ kOe}$): (a) substrate/Pt(10)/[Co(0.5)/Pt(1)]$_4$/Co(0.5)/Pt(0.6)/FeMn($t_1$)/Pt(2) ($t_1 = 2, 6, 15, 30 \text{ nm}$); (b) substrate/Pt(10)/[Co(0.5)/Pt(1)]$_4$/Co(0.5)/Pt(0.6)/IrMn ($t_2$)/Pt(2) ($t_2 = 6, 10 \text{ nm}$); (c) and (d) show the arrangement of spins in the multilayers when $\delta_{\text{critical}} < t < \delta_W$ and $t > \delta_W$, respectively. The bracketed parts denote the detection ranges of the XMLD experiments. The [Co/Pt]/FeMn interface marked by the grey arrow is described in the text.](image-url)
fingerprint at the Mn L3-edge are detected at opposite remanent states, again indicating the same arrangement of FeMn moments.

To clarify this t-dependent tendency, firstly we try to estimate the domain-wall width (δW₁) for FeMn that is calculated to be less than 28 nm [25], according to δW₁ = πA₁/K₁/2, where the typical values of exchange stiffness A₁ and anisotropy constant K₁ for FeMn are 4.1 × 10⁻¹² J m⁻³ and 1.3 × 10⁴ J m⁻³, respectively. It should be noted that a relatively large scatter of δW₁ between 9 nm and 50 nm was reported [19, 25–27]. The exact value is difficult to obtain, and strongly depends on the film quality and interface properties. The value of <28 nm estimated here could be treated as a general range, which is convinced by the XMLD measurements in thick FeMn. Accordingly, this means that the thick FeMn layer could be divided into two parts: the exchange-spring (<28 nm) and the bulk [6]. Since the moments in bulk FeMn, as sketched in figure 2(d), are aligned in-plane and hardly rotated by Co/Pt both at +H and −H, similar XMLD signals for 30 nm-thick FeMn could be interpreted. Accordingly, the main ‘plus/minus’ structure at the Mn L₃ peak (~639 eV) reflects the in-plane orientations of the IrMn spins, regardless of the first ‘minus’ fingerprint, which was observed experimentally but not in correspondingly theoretical spectra [28]. This is also evidenced by the similar ‘plus/minus’ feature of XMLD for a single 6 nm-thick FeMn layer without Co/Pt FM, the moments of which are aligned in-plane and are hardly rotated by the external fields (figure S3 in the supplementary data available at stacks.iop.org/NJP/16/123032/mmedia). Thereby, we can conclude that when t₁ is less than the domain-wall width, such as t₁ = 6 nm and 15 nm, the moments in the exchange-spring are in-plane at +H, according to the similar XMLD spectra with ‘plus/minus’ structure for t₁ = 6 nm, 15 nm, and 30 nm in the right column of figure 2(a). Nevertheless, the opposite signals of ‘minus/plus’ between t₁ ≤ 15 nm and 30 nm (left column of figure 2(a)) indicate a partial rotation of FeMn moments (t₁ ≤ 15 nm) towards out-of-plane direction at −H (figure 2(c)). Admittedly, this is a qualitative analysis about the shape of XMLD spectra, as shown by previous studies in other AFM systems [3, 13], while further theoretical calculations on these noncollinear AFM remain to be carried out.

When the thickness of FeMn decreases to 2 nm, the stable exchange-spring cannot be formed in an ultrathin layer, leading to the canted spins towards out-of-plane direction at both remanent states. This means that a critical thickness (tcritical) is needed to realize the asymmetric rotation of spins in AFM. This is convinced by the vanished TAMR effect at room temperature when the AFM thickness decreases to 2 nm, as described in Ref. [6]. In addition, the unstable exchange-spring in 2 nm-thick FeMn also provides evidence for the traditional thickness of FeMn that is adopted in the exchange-biased AFM/FM system, which is usually ~10 nm [11], much thicker than 2 nm.

As another type of the chemically-disordered γ-AFM, IrMn has a different spin structure with a shorter exchange-spring. That is, δW₂ is calculated to be ~8 nm owing to the higher K₂ (1.8 × 10⁵ J m⁻³) of IrMn compared with that of FeMn. In order to prove the different features of AFM exchange-spring, control experiments on substrate/Pt(10)/[Co(0.5)/Pt(1)]₄/[Co(0.5)/Pt(0.6)/IrMn(t₂)/Pt(2) multilayers are performed, where the typical thicknesses of IrMn (t₂) are 6 nm and 10 nm, distributed at both sides of the critical thickness of 8 nm. As shown in figure 2(b), the spins of the bulk part in the 10 nm-thick IrMn remain in-plane at the two remanent states, generating an analogical Mn L₃-edge XMLD signal. Similar to that of FeMn, when t₂ (e.g., 6 nm) is less than δW₂, the opposite signals observed in the left and right column verify a partial rotation of IrMn spins towards the out-of-plane direction. To sum up, these element specific XMLD results confirm the asymmetric rotation of FeMn and IrMn spins when
they are coupled with perpendicular FM at opposite $H$, which supports the graphics of moment arrangements in the AFM-based tunnel junctions (figure 1(a)). This enables experimental observations of different lengths of the exchange-spring in FeMn and IrMn, which is consistent with the earlier theoretical predictions [25].

After clarifying the spin arrangements in the whole AFM exchange-spring, we now turn towards the interfacial behaviors, which are also significant for the coupling between FM/AFM. In contrast to XMLD experiments, which concentrate on the spin directions in the exchange-spring, XMCD could be used to characterize the ferromagnetism in FM, together with the uncompensated moments in AFM at the interface [12, 20]. Tiny magnetic signals of the interfacial Fe and Mn spins could be separated from large signals of the Co FM layer [12, 20], taking advantage of the element specificity. In order to detect the uncompensated FeMn spins at the interface of [Co/Pt]/FeMn using XMCD, multilayers of substrate/Pt(10)/FeMn(6)/Pt(0.6)/[Co(0.5)/Pt(1)]$_2$ were prepared (figure 1(c)). It is noteworthy that Pt(0.6)/[Co(0.5)/Pt(1)]$_2$ multilayers were placed at the surface to ensure that the detected Mn signals arise mostly from the interface, and the structure here could be regarded as an inverted one of that adopted in the XMLD experiments (figure 1(b)). Thus, the interfacial features of the samples for XMLD detection is essentially identical to that in the XMCD experiments, as denoted in figure 2(c) (marked by the grey arrows). Meanwhile, the interfaces of the inverted samples behave magnetically similarly, taking the subtle difference of the perpendicular magnetic anisotropy (PMA) of the samples for XMLD and XMCD measurements into account (see figure S4 in the supplementary data). The x-ray absorption spectra were obtained with left and right elliptically x-rays that were perpendicular to the film plane (figure 1(c)), so as to acquire the perpendicularly magnetized signals.

The element specific XAS and corresponding XMCD $(I_{\text{left}}-I_{\text{right}})$ spectra are displayed in figure 3. The opposite signals of Co $L$-edge XMCD in Co/Pt between the $-H$ and $+H$ remanent states (figure 3(a)) reflect the downward and upward magnetizations exerted by the external fields, respectively. In contrast to the sizable XMCD of the perpendicularly magnetized Co with large ferromagnetic moments, only the uncompensated moments of FeMn AFM at the interface contribute to the Fe and Mn $L$-edge signals. The weak and nearly negligible signals for Mn indicate mostly compensated moments with little residual ferromagnetic spins. Nevertheless, the same polarity between the Fe (figure 3(c)) and Co $L$-edge peak indicates that the uncompensated Fe spins rotate following the ferromagnetic Co spins. These similar features of XMCD between Co FM and FeMn AFM demonstrate a parallel or ferromagnetic coupling between them. Remarkably, the XMCD experiments here point out the out-of-plane component of the interfacial uncompensated FeMn moments, where the weaker intensity of Fe signals at $+H$ than that at $-H$ indicate a slighter rotation (see the graphics in figure 2(c)), which can probably be ascribed to the pinning by in-plane spins in the exchange-spring at $+H$. It is found that a partial rotation of the whole exchange-spring in AFM from in-plane (at $+H$) to out-of-plane (at $-H$) direction has been clarified by means of XMLD in figure 2. Although a ferromagnetic sub phase possibly causes XMLD signals [28], it should be noted that the clear ferromagnetic signal of FeMn XMCD, originating from out-of-plane interfacial uncompensated spins, does not contribute to the XMLD spectra in the present case. Meanwhile a limited detecting depth with TEY mode simplifies the detection of XMLD and XMCD signals in the samples with inverted FM/AFM structures, where XMLD and XMCD signals would not mix together. As a combination of XMCD and XMLD measurements, it is evidenced that the
interfacial AFM spins are easily tilted by Co/Pt FM, in contrast to the asymmetric alignment of the spins in the whole exchange-spring at $+H$ and $-H$.

Having established the spin structures of FeMn both from the asymmetric behavior of $t$-dependent exchange-spring and interfacial uncompensated spins, we will further probe into the influence of spin structures on the TAMR effect. In the following we focus primarily on the FM/AFM interface, which has been proved to be crucial in obtaining the strong PMA, as well as the corresponding magnetization and magnetoresistance [29–31]. Apart from the sample structure mentioned in figure 1(a) (sample A), another two multilayers with different features of interface were prepared and fabricated into tunnel junctions to make a comparison, marked as B: Pt/[Co/Pt]$_4$/Co(0.5)/FeMn/AlO$_x$/Pt, where 0.5 nm-thick Co directly attaches to FeMn without Pt insert layer, and C: Pt/[Co/Pt]$_4$/Co(0.5)/Pt(0.6) (oxidation)/FeMn/AlO$_x$/Pt, where oxidation means that the sample was transferred into another chamber with 1 mTorr O$_2$ for 20 min and then transferred back [32]. Figure 4(a) shows the comparison of magnetic hysteresis loops at vertical $H$ among the three multilayers. The obviously superior PMA of sample A than both B and C emphasizes that two factors are critical. One is the insert of an ultrathin Pt layer to avoid the interdiffusion between Co and Fe, as well as Co and Mn [30]. The other is the prevention of
the oxidation at the interface. However, the scenario differs dramatically for the TAMR measurements. Despite the slight oxidization at the interface of sample C, the insert of Pt in both samples A and C promotes the sharp reversal between the HRS and LRS, and produces a higher TAMR ratio compared with that of sample B. This indicates that a 0.6 nm-thick Pt layer with strong spin–orbit coupling at the FM/AFM interface plays a significant role in enhancing the TAMR effect [33].

The superior PMA and TAMR for sample A are related to the interfacial spin structures, as proven by the XMCD measurements. Taking the detection depth into account, multilayers of Pt(10)/FeMn(6)/[Co(0.5)/Pt(1)]2 without Pt insert layer at the interface were prepared to perform the XMCD measurements. We point out that the interfacial characters here are consistent with sample B, so as to make a comparison with sample A with the Pt insert. Remarkably, clear Mn L-edge XMCD signals are observed in figure 4(c), in contrast to the relatively weak intensity for the sample with Pt insert layer [figure 3(b)]. It could be inferred from this that the diffusion between Mn and Co at the interface induces the uncompensated spins of Mn, bringing about the out-of-plane ferromagnetic signals in XMCD. The same polarity of the Mn and Fe XMCD spectra in figures 4(c) and (d) confirms the chemically-disordered structure, with parallel Fe and Mn moments predicted by the previous first-principles investigation [16, 34]. Nevertheless the Mn moments were reported to be aligned antiparallel to Co and Fe in some other systems [35, 36]. Our observation of the parallel alignment is not in conflict with the antiparallel case because the magnetic state of the Mn atoms is sensitive to the magnetic environment in which they are located [35]. One may expect that differences in film composition or on the interfacial roughness would induce a locally different environment for the Mn atoms, changing the local interaction of Mn with the Fe and Co atoms. In the present case,
we conclude that the induced uncompensated Mn spins at the interface are unfavorable for the asymmetric rotation of FeMn exchange-spring, according to the slow reversal between HRS and LRS for sample B (figure 4(b)).

The element specific XAS characterizations have revealed an asymmetric rotation of spins at the FM/AFM interface from the static view. We now address the question of whether or not an electrical approach can be used to dynamically detect the rotation. A comparison between the magnetization loop of the multilayer and the TAMR curve based on FeMn at vertical $H$ is presented in figure 5(a). Since the TAMR effect originates from the anisotropy of tunneling density of states when a partial rotation of AFM spins is triggered, the reversal of the resistance in TAMR curves qualitatively uncovers the rotation process of FeMn spins triggered by perpendicularly magnetized Co/Pt. As has been discussed in figure 1(a), a partial rotation of FeMn moments from in-plane to out-of-plane direction is induced with $H$ sweeping from $+600$Oe to $−600$Oe. It should be noted that the precise calculation of the rotation angle of AFM spins would require some independent calibration measurements of the transport anisotropy and the original spin angles, which are hard to detect in these noncollinear AFM.

It has been verified that the domain-wall width and the exchange-spring of IrMn is much shorter than that of FeMn. Here, the dynamic rotation process of IrMn moments is also found to be dramatically distinguished from that of FeMn moments. The magnetization loop of the IrMn-based junction and the corresponding TAMR curve are shown in figure 5(b), where the rotation process could be estimated in the same way as that of FeMn. Although the FeMn- and IrMn-based multilayers exhibit similar squareness of the magnetic hysteresis curves, reflecting the

\begin{figure}[h]
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\includegraphics[width=\textwidth]{figure5.png}
\caption{(a) A comparison of the magnetization loop of the Pt/[Co/Pt]/FeMn/AlO$_x$/Pt multilayers and the corresponding TAMR curve of the junctions. (b) The magnetization loop and the TAMR loop of Pt/[Co/Pt]/IrMn/AlO$_x$/Pt at vertical $H$. Inset of (b): a comparison of the effects of rotating FeMn- and IrMn-based junctions on resistances in fixed fields of 5 kOe. The device was rotated from $\theta=90^\circ$ (vertical $H$) to $0^\circ$ (in-plane $H$), and continued to $−90^\circ$.}
\end{figure}
strong PMA of both Co/Pt FM, their TAMR counterparts undergo slow and sharp rotation process for FeMn and IrMn spins, respectively. For the junctions comprising FeMn AFM (figure 5(a)), the gradual reversal between the HRS/LRS reflects the stepwise rotation of FeMn spins compared with the sharp reversal of Co/Pt spins. In contrast, a swift twist of IrMn spins is triggered by Co/Pt, as indicated by the sharp transition in the magnetoresistance.

In general, the rotation angle of AFM spins decreases with increasing distances from the [Pt/Co]/AFM interface [13, 19]. In addition, the rotation angle of AFM contributed to TAMR could be considered as the average rotation of all the moments within the whole exchange-spring. Accordingly, we can conclude that the FeMn spins near Co/Pt are easily motivated, even at a lower field than the coercivity of Co/Pt, and gradually finish the rotation of the whole exchange-spring. In contrast, the IrMn exchange-spring could be twisted by Co/Pt sharply (figure 5(b)), indicating a more stable and coherent structure of the exchange-spring. This could be proved by comparing the resistance when rotating the devices in a constant magnetic field, as shown in the inset of figure 5(b). A relatively higher resistance for IrMn-based junctions at vertical $H (\theta = 90^\circ)$ reveals that there are more stabilized spins in the exchange-spring, since the resistance is manipulated by the angles between AFM and Co/Pt. Furthermore, the differences between the motions of FeMn and IrMn exchange-springs could be explained by the different anisotropy constant $K$ [25]. The higher $K$ for IrMn indicates a more stable exchange-spring, together with a stronger exchange coupling between IrMn and Co/Pt.

In summary, we have observed an asymmetric rotation of FeMn spins driven by perpendicularly magnetized Co/Pt at opposite magnetic fields. A combination of the element specific XMLD and XMCD experiments enables us to identify the arrangement of the spins in the whole exchange-spring and the uncompensated AFM moments at the interface from the static view. In particular, different lengths of exchange-spring are verified in FeMn and IrMn using thickness-dependent XMLD, which are both featured with a twist of the exchange-spring at negative fields. Moreover, the TAMR effect based on the asymmetric behavior of AFM spins provides an electrical approach to dynamically detect the rotation behaviors of the AFM exchange-spring. The identification and manipulation of AFM spin structures would advance the application of AFM in spintronic devices.

Acknowledgments

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