Spin-Orbit Torque Switching of Noncollinear Antiferromagnetic Antiperovskite Manganese Nitride Mn$_3$GaN

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Noncollinear antiferromagnets have promising potential to replace ferromagnets in the field of spintronics as high-density devices with ultrafast operation. To take full advantage of noncollinear antiferromagnets in spintronics applications, it is important to achieve efficient manipulation of noncollinear antiferromagnetic spin. Here, using the anomalous Hall effect as an electrical signal of the triangular magnetic configuration, spin–orbit torque switching with no external magnetic field is demonstrated in noncollinear antiferromagnetic antiperovskite manganese nitride Mn$_3$GaN at room temperature. The pulse-width dependence and subsequent relaxation of Hall signal behavior indicate that the spin–orbit torque plays a more important role than the thermal contribution due to pulse injection. In addition, multistate memristive switching with respect to pulse current density was observed. The findings advance the effective control of noncollinear antiferromagnetic spin, facilitating the use of such materials in antiferromagnetic spintronics and neuromorphic computing applications.

I. INTRODUCTION

Noncollinear antiferromagnetic (AFM) materials have attracted significant attention in basic and applied science because in addition to having the excellent properties of collinear AFM—such as fast dynamics, suitability for high-density integration, and stability against external perturbations—noncollinear AFM materials can overcome the weakness of collinear AFM materials—namely the small electrical signal—by the anomalous Hall effect (AHE) [1–4]. Thus, efficient control of noncollinear AFM spin is essential for the application of such materials in AFM spintronics. In the past decade, electrical manipulation via spin–transfer torque (STT) and spin–orbit torques (SOTs) has become one of the most promising techniques in the field of ferromagnet (FM)–based spintronics, not only to take the place of dynamic random access memory in the current computer memory hierarchy [5] but also to instantiate multistate magnetoresistive random access memories for neuromorphic computing [6, 7].

Recent studies demonstrate that like collinear AFM materials, noncollinear AFM materials can be controlled via SOT in the same way as FM materials. Collinear AFM materials consisting of NiO/Pt bilayers and Pt/NiO/Pt trilayers show a critical current density ($J_c$) on the order of $10^7$–$10^8$ A/cm$^2$ [8–10], similar to that in typical FM/heavy metal (HM) bilayers. In contrast, $J_c$ values one to two orders of magnitude smaller $J_c$ have been observed in noncollinear AFM Mn$_3$GaN (MGN)/Pt bilayers (1.5 × 10$^6$ A/cm$^2$) [12] and Mn$_3$Sn/W bilayers (5 × 10$^6$ A/cm$^2$) [13]. In the case of a collinear AFM system, 90$^\circ$ switching of the Néel vector is required because the electrical signal, such as spin Hall magnetoresistance and anisotropic magnetoresistance, is maximal. To achieve 90$^\circ$ switching of the Néel vector, diagonal current flow using an eight- or four-terminal device with complex electrical write/read operation is required [8–10], except for CuMnAs using two terminal writing device [11]. A noncollinear AFM system, by contrast, needs 180$^\circ$ switching of each spin of triangular magnetic configuration, which can be accomplished using a simple four-terminal Hall device with no external magnetic field in Mn$_3$GaN [12], or with an external magnetic field in Mn$_3$Sn [13]. Therefore, in addition to their large electrical signals due to non-zero Berry curvature [14], noncollinear AFM systems have a distinct advantage in spintronic applications.

In the antiperovskite manganese nitrides Mn$_3$AN (where $A = \text{Ni}$, Ga, Sn, etc.), the Mn atoms form a kagome lattice in the (111) plane. The noncollinear AFM Mn$_3$AN with a nonzero Berry curvature has been predicted to exhibit a large anomalous Hall effect (AHE) and an anomalous Nernst effect even with a quite small canted magnetization of the order of 0.001–0.01 $\mu$B per atom [15–17]. The AHE has been established in Mn$_3$Ni$_{1-x}$Cu$_x$N films [18, 19], strained Mn$_3$NiN films [20], and strained Mn$_3$SnN films [21]. Although electrical current switching of both Hall resistance with no external magnetic field and nonlinear Hall resistance with respect to an external magnetic field have been reported in MGN/Pt bilayers, no clear evidence of AHE has yet been presented [12]. In addition, a thermal contribution due to pulse current injection such as a thermal activation effect, a joule heating effect, or an electromigration effect can change the Hall resistance [22–24]. Hence, as has been pointed out, both the low-electrical-current writing and reading operations of MGN/Pt bilayers could be of nonmagnetic thermal origin [25].

In this investigation, we studied the magnetotransport properties of MGN films and performed systematic switching operations of strained MGN/HM (HM = Pt, Ta) bilayers. At room temperature, no AHE was obtained in relaxed MGN films, whereas the AHE appeared

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with respect to the ratio between the lattice constants \(c\) and \(a\) (\(c/a\) ratio). The temperature dependence of the magnetization and the AHE suggests that skew scattering is dominant below 200 K, whereas another origin, probably noncollinear AFM order with nonzero Berry curvature, is dominant above 200 K. To estimate the effect of thermal contributions on write/read operations, the pulse-width dependence and relaxation after pulse injection were measured. We show that the thermal activation effect, joule heating effect, and electromigration effect play a minor role. The existence of multistate signal amplitude with respect to pulse current with no external magnetic field was demonstrated.

### II. EXPERIMENTAL DETAILS

MGN films were grown by reactive magnetron sputtering on MgO(001) substrates using a Mn$_3$Ga target at 400 °C. As details of the film growth were reported in our previous work, the \(c/a\) ratio was controlled by precise control of N$_2$ partial pressure during film growth [26]. The crystal structure was analyzed using both in-plane and out-of-plane X-ray diffraction (XRD) measurements with Cu K\(\alpha\) radiation. Magnetic properties were characterized using superconducting quantum interference device magnetometry. Transport properties were characterized by the standard DC four-terminal method. For write/read operations, a layer of Pt or Ta (3nm) was deposited by magnetron sputtering at room temperature after film growth. 20 µm-width Hall bars with Ti/Cu contact pads were prepared by a conventional photolithographic process. All SOT measurements were performed with no external magnetic field at room temperature (300 K). The sequence of write/read operations was the same as described in our previous report [12].

### III. RESULTS AND DISCUSSIONS

#### A. Film Characteristics and Magnetotransport Properties

Typical out-of-plane \(2\theta-\omega\) and in-plane \(2\theta-\phi\) X-ray diffraction (XRD) patterns for the 35-nm-thick MGN films are shown in Fig. 1(a). Only the MGN (002) and (200) planes exhibit Bragg peaks, in the out-of-plane and in-plane XRD patterns, respectively. In addition, epitaxial growth is confirmed by the results of \(\phi\)-scan measurement as shown in Fig. 1(b), showing that their epitaxial relationship is MgO(001)[100]/MGN(001)[100]. The lattice constants \(c\) and \(a\) are 0.38817 nm and 0.38965 nm, respectively, giving \(c/a = 0.9962\). The temperature dependencies of the resistivity \(\rho_{xx}\) and magnetization \(M\)
are shown in Fig. 1(c). In the $\rho_{xx}$ curve, there is a clear anomaly at 200 K. Likewise, an FM-like transition is observed at 200 K in the magnetization curve. These temperature dependencies were also observed in our previous switching study of MGN/Pt films [12]. The ground state of MGN at room temperature is well known to exhibit $\Gamma_{5g}$ spin structure [27]. The coexistence of $\Gamma_{5g}$ and M-1 phase below the FM-like transition temperature has also been previously reported [28]. As the MGN/FM bilayers exhibit an exchange bias at 4 K [29], it is concluded that $\Gamma_{5g}$ order and M-1 phase coexist below 200 K.

Figure 1(d) and 1(e) show the magnetic $H$ hysteresis and anomalous Hall resistivity $\rho_{xy}$ loops for the MGN films measured along the (001) direction at various temperatures. In the $\rho_{xy}$ loops, linear contribution from the ordinary Hall effect has been subtracted. The $\rho_{xy}$ loops before subtract the ordinary Hall effect and a summary of the Hall coefficient values are given in Fig. 6 of Appendix A. In both loops, hysteresis is clearly exhibited not only in the coexistence phase below 200 K but also in the $\Gamma_{5g}$ single phase. Both the magnetization and the $\rho_{xy}$ loops of the MGN films have similar coercive field ($H_c$) values, indicating that the AHE is directly related to the MGN magnetic order. The temperature dependencies of the magnetization and $\rho_{xy}$ at 2 T are shown in Fig. 1(f). Above 200 K, the magnetization remains nearly constant at ~ 0.02 $\mu_B$/Mn, and below 200 K, it increases monotonically with decreasing temperature. $\rho_{xy}$, on the other hand, increases slightly with decreasing temperature above 200 K and increases dramatically with decreasing temperature below 200 K, indicating that the increase in $\rho_{xy}$ is strongly linked to the increase in magnetization. In contrast to the magnetization, however, $\rho_{xy}$ begins to decrease below 75 K. As $\rho_{xy}$ is found to be proportional to $\rho_{xx}$ below 75 K, $\rho_{xy}$ below 200 K would be dominated by a net magnetization.

According to theoretical studies on piezomagnetism of MGN with $\Gamma_{5g}$ order, a net magnetization can appear by the induction of strain [30, 31], and the magnitude of the net magnetization increases linearly with respect to the strain $\epsilon$, with a coefficient of 0.013 $\mu_B$/Mn/[%] [30]. Our MGN films show $\epsilon$ and remnant magnetization of approximately 0.3 % and 0.004 $\mu_B$/Mn at 300 K, respectively. As the net magnetization observed in our MGN films is similar to the value calculated theoretically, it is considered that the canted $\Gamma_{5g}$ order is realized above 200 K. The $\Gamma_{5g}$ order does not show the AHE because it has mirror symmetry, and the symmetry operations make the Berry curvature vanish after integration over the entire Brillouin zone [15]. In antiperovskite nitride films with $\Gamma_{5g}$ order, however, the anomalous Hall conductivity (AHC) tensor is reported to be highly sensitive to strain. Although strain-free Mn$_3$N thin films show no AHE [18], strained Mn$_3$N thin films do show AHE [20]. This is explained by the reduction of the symmetry of a space group, under which nonzero Berry curvature is induced when a finite strain is applied [20]. Strained Mn$_3$SnN films likewise show a large AHE, but it is suggested that the biaxial strain induces $\Gamma_{5g}$ order from $\Gamma_{5g}$ order [21]. These past findings indicate that AHE cannot be accounted for by either extrinsic scattering processes or changes in magnetization; hence, nonzero Berry curvature plays an important role. In our MGN films, $\rho_{xy}$ is observed to increase with decreasing $c/a$ ratio at 300 K as shown in the inset of Fig. 1(f), highlighting the fact that appearance of AHE at 300 K in MGN films is also strongly related to the film strain and/or reduced magnetic space group. Therefore, although we cannot experimentally separate the contributions from canted net magnetization and nonzero Berry curvature due to non-collinear AFM order above 200 K, we can state that part of the AHE comes from noncollinear AFM order. From the view point of SOT, we will discuss the possible origin of AHE in the later section.

B. Dependence of Reversible Electrical Switching on HM

For the electrical write/read operations, typical Hall bar devices of bilayers consisting of strained-MGN (20 nm) with either Pt or Ta (3 nm), with 20 $\mu$m width were prepared. From a parallel circuit model, the $\rho_{xx}$ values for both the MGN/Pt and MGN/Ta bilayers can be derived using $\rho_{xx}$ of each single-layer film, which enable us to estimate current density through the Pt and Ta layers ($J_{e,HM}$). The $\rho_{xx}$ values at 300 K for MGN, Pt, and Ta single-layer films and MGN/Pt and MGN/Ta bilayers are given in Table. 1. Using these bilayers, sequential write/read operations were performed, with the results as shown in Fig. 2. Here, the top and bottom axes are the current densities derived from the Pt/Ta layer alone and the bilayer total thickness, respectively, and the left and right axes are the $\rho_{xy}$ values derived from the full bilayer and the MGN layer alone, respectively. In addition, the constant offset probably due to geometrical imperfections of the Hall bar and/or thermoelectric voltage was subtracted. $\rho_{xy}$ changes with respect to $J$, and a clear hysteresis loop is observed in both bilayers.

| Film          | $\rho_{xx}$ ($\mu\Omega$ cm) |
|---------------|-------------------------------|
| Mn$_3$GaN     | 1271                          |
| Pt            | 112                           |
| Ta            | 972                           |
| Mn$_3$GaN/Pt  | 504 (exp.), 541 (calc.)       |
| Mn$_3$GaN/Ta  | 1261 (exp.), 1222 (calc.)     |
presents the width for MGN/Ta bilayers. Although the switching ρ is the thermal stability factor, and τ amplitudes remain nearly the same, the hysteresis loops of Appendix B. Besides, the switching width for MGN/Pt and MGN/Ta devices is probably due to local variations in the quality and/or strain of the thin films, which is discussed in Fig. 7 of Appendix B. Besides, the comparison of ρxy width between field-sweep and SOT measurements using the same device is presented in Fig. 8 of Appendix C.

To evaluate the effect of joule heating by applying pulse current, the pulse-width dependence was investigated. Figure 3(a) shows ρxy as a function of J with several pulse widths for MGN/Ta bilayers. Although the switching ρxy amplitudes remain nearly the same, the hysteresis loops become slightly narrower with increasing pulse width. Jc is plotted as a function of pulse width in Fig. 3(b). Here, Jc is fitted by the thermal activation model [33]:

\[ J_c = J_{c0} \left[ 1 - \frac{1}{\Delta} \ln \left( \frac{\tau}{\tau_0} \right) \right], \tag{1} \]

where \( J_{c0} \) is the critical current density at 0 K, Δ is the thermal stability factor, and \( \tau_0^{-1} \) is the thermally activated switching frequency, for which we assume a frequency of 1/\( \tau_0 \) = 1 THz. It can be seen that Jc can be fitted by the thermal activation model with \( J_{c0} = 7.97 \times 10^6 \text{ A/cm}^2 \) and \( \Delta = 58.7 \). These results highlight the finding that a Jc value of the order

10^6 A/cm^2 originates intrinsically in spin torque whereas the thermal activation, through it exists, plays a minor role.

C. Thermal Contribution to Write/Read Operations

Because heating and electromigration effects can affect ρxy signal amplitudes [23], the relaxation behavior after switching was investigated. Figure 4 presents the continuous write/read operation using \( I_{pulse} = \pm 13 \times 10^6 \text{ A/cm}^2 \) with a 5 µs pulse width and subsequent relaxation measurements for MGN/Ta bilayers at 300 K. Figure 4(b) is an enlargement of the continuous ±Ipulse write part shown in Fig. 4(a). As discussed for synthetic AFM materials [34] and in our previous MGN/Pt study [12], the observed asymptotic ρxy behavior can be fitted by an exponential decay function \( y = y_0 + A \exp \left(-\left(x + x_0\right)/\tau\right) \) with time constants \( \tau \) of 14.9 pulse number (197.0 s) for +Ipulse and 15.7 pulse number (207.6 s) for −Ipulse with continuous write/read operations in approximately 13.25 s cycles. The relaxation behavior after switching was measured after three cycles of the continuous ±Ipulse write operations.

Figure 4(c) presents the ρxy relaxation as a function of time after continuous ±Ipulse write operations. The
The relaxation behavior is characterized by fit to a double exponential function [23]:

\[ d = d_0 + d_1 \exp\left(-\frac{t}{\tau_1}\right) + d_2 \exp\left(-\frac{t}{\tau_2}\right), \tag{2} \]

where \(d_0\) is a base line and is the value reached when attenuated, \(d_1, d_2\) are amplitude parameters, and \(\tau_1, \tau_2\) are the relaxation times. Both relaxations after \(\pm I_{\text{pulse}}\) write operations fit well with \(d_0 = -0.0467 \, \mu\Omega\,\text{cm}, d_1 = -0.0219 \, \mu\Omega\,\text{cm}, d_2 = -0.0003 \, \mu\Omega\,\text{cm}, \tau_1 = 8.63 \, \text{h}, \tau_2 = 11.67 \, \text{h}\) for after the \(+I_{\text{pulse}}\) write operation, and \(d_0 = 0.0331 \, \mu\Omega\,\text{cm}, d_1 = 0.0100 \, \mu\Omega\,\text{cm}, d_2 = 0.0194 \, \mu\Omega\,\text{cm}, \tau_1 = 0.25 \, \text{h}, \tau_2 = 3.89 \, \text{h}\) for after the \(-I_{\text{pulse}}\) write operation. With regard to the change in Hall resistance due to the effects of annealing and electromigration, the short and long decay times are reported to be approximately 4 min and 50 min, respectively [23]. Compared with these values, both the short and long decay times observed here are five to ten times longer. In addition, for the change in Hall resistance due to annealing and electromigration effects, the Hall resistance asymptotes to the initial value before pulse injection is a few minutes to a few hours [23, 35], in contrast with our MGN/Ta bilayers, for which \(d_0\) is a rather large value. Whereas our electrical measurements do not enable us to fully distinguish SOT and other possible contributions such as thermal activation and electromigration effects, the relaxation behavior and pulse-width behavior in the MGN/Ta bilayers highlight the fact that SOT plays an important role in the present switching behavior. In the case of collinear AFM materials such as NiO, 180° switching of the Néel vector is required, which can be achieved by the flow of current in two orthogonal directions. However, current flow in orthogonal directions causes inhomogeneous current density due to the current crowding effect, which induces a change in Hall resistance due to annealing and electromigration effect [23]. In the case of noncollinear AFM materials, in contrast, 180° switching of each non-collinear spin is required, which can be achieved by the flow of current in a straight line; this implies that the current crowding effect in this study can be considered small.

D. Memristive Switching with Respect to Pulse Current Density

Finally, here we discuss the memristive behavior of MGN/Ta bilayers. Figure 5 shows \(\rho_{xy}-J\) loops at 300 K, where \(J_{\text{max}} = 13.0 \times 10^8 \, \text{A/cm}^2\) was first applied and subsequently scanned from \(J_{\text{max}}\) to \(J_{\text{min}}\) and from \(J_{\text{min}}\) to \(J_{\text{max}}\). \(\rho_{xy}\) exhibits multiple stable signal amplitudes according to the magnitude of \(J_{\text{min}}\). The same behavior has been observed in Mn\(_3\)Sn/Pt bilayers [13] and AFM/FM bilayers [37], suggesting that the phenomenon of multiple stable magnitudes of the Hall resistance originates in the multi-AFM domain character, which allows us to tune the signal amplitude in an analog manner. In contrast to Mn\(_3\)Sn/Pt bilayers [13], MGN/Ta bilayers show a memristive behavior with no external magnetic field, highlighting the advantage of Mn\(_3\)AN systems for use in neuromorphic computing.

According to theoretical study of SOT of \(\Gamma_{4j}\) order, the noncollinear spins rotate in the (111) plane of the kagome lattice, where the injected spins are directed perpendicular to the kagome lattice. Therefore, \(J_c\) is determined...
by an in-plane anisotropic energy of the kagome lattice and the injected spin direction. Further improvements in SOT efficiency in noncollinear Mn$_3$AN systems may be attained using (110)-oriented films of low-anisotropy materials. Theoretically, Mn$_3$Ga$_{1-x}$Ni$_x$N [36] has a low anisotropy energy, implying that it would be worthwhile to investigate the dependence of SOT efficiency on $A$ atoms. On the other hand, if the AHE comes from only net magnetization (for simplicity, assume a net magnetization in the perpendicular direction like Ferrimagnet), the magnetic field parallel to the current direction is generally needed to realize the SOT switching. In contrast, SOT of noncollinear AFM theoretically satisfies even though with no external magnetic field [32, 36]. Indeed, no switching has been observed with no external magnetic field at low temperatures where the Ferrimagnetic M1 phase is dominant in the AHE [12]. Although the Joule heating can affect to the Hall resistivity, the pulse-width measurement and relaxation measurement show a heating effect plays a minor role. From these results, we can conjecture that AHE is possibly related to noncollinear AFM order from the view point of SOT at 300 K.

IV. CONCLUSION

In this study, we have shown the AHE and SOT switching of noncollinear AFM MGN at room temperature. By tuning the $c/a$ ratio, two origins of AHE were observed: one for AHE above 200 K, possibly related to noncollinear AFM order, and one for AHE below 200 K, dominated by magnetization. Using MGN/HM bilayers, we have demonstrated the SOT switching of noncollinear AFM spin in MGN at room temperature with no external magnetic field. The effects of thermal activation on $J_c$ and the effects of heating and electromigration on $\rho_{xy}$ were excluded by pulse-width measurements and relaxation measurements after pulse injection. In addition, multilayer memristive switching with respect to pulse current density was demonstrated. These results show that, efficient SOT can be attained in MGN/HM bilayers with memristive functionality with no external magnetic field, and demonstrate the potential application in AFM spintronics and neuromorphic computing.

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V. APPENDIX

Appendix A: Ordinary Hall effect of MGN films

Figure 6(a) shows the Hall resistivity $\rho_{xy}$ as a function of external magnetic field for MGN films ($c/a = 0.9962$) at various temperature before subtract the ordinary Hall effect. The temperature dependence of the Hall coefficient $R_H$ is summarized in Fig. 6(b).

![Figure 6](image)

FIG. 6. (a) Hall resistivity $\rho_{xy}$ as a function of external magnetic field for MGN films ($c/a = 0.9962$) before subtract the ordinary Hall effect. (b) Temperature dependence of the Hall coefficient $R_H$.

Appendix B: Sample dependence of $\rho_{xy}$ switching width in SOT measurements

Figure 7 shows the sample dependence of $\rho_{xy}$ switching width in SOT measurements as a function of $c$ lattice constant. The $\rho_{xy}$ obtained by the field sweep measurements of unpatterned films is also plotted. While no $\rho_{xy}$ switching is observed for samples with $c$ lattice constants longer than 0.3890 nm, $\rho_{xy}$ switching is observed in samples with shorter $c$ lattice constants, which is consistent with the results of AHE measurements of unpatterned films. On the other hand, although no large difference in $c$ lattice constant between 0.3882 and 0.3887 nm among SOT films in which $\rho_{xy}$ switching was observed, $\rho_{xy}$ switching width shows sample dependence. Since the amplitude of $\rho_{xy}$ is strongly related to $c/a$ ratio as discussed in the main text, the sample dependence of $\rho_{xy}$ switching width probably due to local variations in the quality and/or strain of the thin films.

Appendix C: Comparison of $\rho_{xy}$ width between field-sweep and SOT measurements using same device

Figure 8 shows the results of the Hall and SOT measurements using the same SOT device. Unfortunately, since our superconducting magnet has limited bore sizes,
FIG. 7. Sample dependence of $\rho_{xy}$ switching width in SOT measurements for MGN(20 nm)/HM(3 nm) bilayers. MGN/Pt and MGN/Ta results presented in the main text are the devices Pt#1 and Ta#1, respectively. For Pt#2 device, $\rho_{xy,MGN}$ was derived by assuming that the current flows through MGN and Pt in the same proportion as in Pt#1 device.

FIG. 8. Hall and SOT measurements using the same SOT device. (a) $R_{xy}$ after subtract estimated ordinary Hall effect vs external magnetic field of MGN/Pt bilayers. (b) $R_{xy}$ vs pulse number of MGN/Pt bilayers. All measurements were performed at room temperature using the same MGN/Pt Hall device. The data are reproduced from Appl. Phys. Lett. 115, 052403 (2019) [12], with the permission of AIP Publishing.

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