Strong Bias Effect on Voltage-Driven Torque at Epitaxial Fe-MgO Interface

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Torque can be provided to magnetization in nanomagnets directly by electric current and/or voltage. This technique enables electric current (voltage)-to-spin conversion without electromagnetic induction, and has been intensively studied for memory device applications. Among the various kinds of torque, torque induced by spin-orbit splitting has recently been found. However, quantitative understanding of bulk-related torque and interface-related torque is still lacking because of their identical symmetry for current-in-plane devices. In this paper, we propose that a pure interface-related torque can be characterized by spin-torque ferromagnetic resonance with a current-perpendicular-to-plane tunnel junction. Epitaxial Fe-MgO-V tunnel junctions are prepared to characterize the interface-related torque at Fe-MgO. We find that the current-driven torque is negligible, and a significant enhancement of the voltage-driven torque is observed when the MgO barrier thickness decreases. The maximum torque obtained is as large as 2.8×10^{-5} J/(Vm²), which is comparable to the voltage-controlled magnetic anisotropy of 180 fJ/Vm. The voltage-driven torque shows strong dc-bias-voltage dependence that cannot be explained by conventional voltage-controlled magnetic anisotropy. Tunnel anisotropic magnetoresistance spectroscopy suggests that the torque is correlated to an interface state at the Fe-MgO. This surface-state-sensitive electric modulation of magnetic properties provides new insight into the field of interface magnetism.

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I. INTRODUCTION

Electric current or voltage provides torque to magnetization in nanomagnets. This enables electric current (voltage)-to-spin conversion without electromagnetic induction and has been intensively studied.

Current-driven spin-transfer torque [1,2] has been studied for memory device applications [3,4]. The spin-transfer torque can arise in a current-perpendicular-to-plane (CPP) magnetic tunnel junction following spin accumulation induced by spin injection from a ferromagnetic counter electrode [5–8]. The spin-transfer torque can also arise in current-in-plane (CIP) multilayers. In the CIP multilayers, spin accumulation can be induced by spin-dependent scattering in bulk nonmagnetic metals [9–16] or surface conductive states of topological insulators [17–19], which is termed the spin-Hall or Rashba-Edelstein effect. Spin-orbit splitting at a ferromagnet-nonmagnet interface can also induce current-driven torque [11], but the existence of such a torque is still under debate [15]. This is because both the bulk-related and interface-related torques have the same

symmetry in CIP multilayers with some torque measurement such as spin-torque ferromagnetic resonance (FMR) [20,21].

While electric current is needed to generate the aforementioned torque, torque can also be induced by voltage alone. For the voltage-driven torque, voltage-controlled magnetic anisotropy (VCMA) at a ferromagnet-dielectric interface [22,23] can have a significant influence on the experimentally observed torque vector in CPP magnetic tunnel junctions [24,25]. However, in such experiments, a ferromagnetic counter electrode inducing tunnel magnetoresistance (TMR) is indispensable for the torque measurement. Therefore, it is difficult to attain a pure voltage-driven torque vector free from spin-transfer torque induced by the counter electrode.

To characterize a pure torque vector originating from a ferromagnet-nonmagnet interface while avoiding the aforementioned issues, we employed a CPP tunnel junction with a single ferromagnetic electrode in the present study. First, the system enables us to characterize the interface-related torque free from the bulk-related one. An electric current flows in a rotational symmetry axis in the CPP tunnel junction; hence, there is no spin accumulation induced by the bulk-spin-dependent scattering. On the other hand, torque from ferromagnet-dielectric interface can be observed, which is understood as the inverse effect of tunnel anisotropic magnetoresistance (TAMR) [26]. Second, any influence of the spin-injection-induced spin-transfer effect could be

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excluded because of the nonmagnetic counter electrode. Therefore, the system enables us to discuss the interface-related torque over a wide current density range. Instead of TMR, spin-torque FMR could be performed using TAMR [27]. Moreover, while spin-torque FMR in a CIP multilayer only provides the torque vector direction [12,28], it is feasible to conduct quantitative characterization of the vector components by employing a CPP tunnel junction [29,30].

We prepared epitaxial Fe-MgO-V tunnel junctions with various MgO thicknesses, giving a wide range of current density. As a result, we found that the interface-related current-driven torque was negligible, but the voltage-driven torque increased as the MgO thickness decreased. The maximum voltage-driven torque was as large as 2.8×10^{-5} J/(Vm²). This is comparable to the VCMA at a Fe-MgO interface of 180 fJ/Vm, which is larger than any reported VCMA-induced voltage-driven torque [24,25,31–35]. The voltage-driven torque shows nonlinear dc bias voltage dependence, with a maximum around zero dc bias; thus, an electronic state in the vicinity of the Fermi level at a Fe-MgO interface is likely to be of importance to generate the large voltage-driven torque. An interface state at Fe-MgO was also found in the vicinity of the Fermi level from the TAMR spectroscopy.

II. EXPERIMENTAL

Figure 1(a) shows a schematic of the device structure. An epitaxial single crystalline multilayer of MgO(001)substrate-MgO buffer(5 nm)-V buffer(30 nm)-Fe(0.74 nm) - MgO($t_{\text{MgO}} = 0.8 - 1.8$ nm) - V(3 nm)-Au(30 nm) was fabricated by molecular beam epitaxy under ultrahigh vacuum. The V layer was employed because of its good lattice match with MgO and because it can induce perpendicular magnetic anisotropy in the Fe [36,37]. Because the counter electrode is nonmagnetic, it is possible to characterize the torque vector independently from the spin-injection-induced spin-transfer torque. During deposition of the MgO buffer and the V buffer, substrate temperature was maintained at 150 °C. The V buffer was post-annealed at 500 °C for 30 minutes. Other layers were deposited at room temperature. The multilayer was then patterned into 390-nm-diameter tunnel junctions. Figure 1(b) shows reflection high-energy electron diffraction images for the surfaces of the V (001) buffer, Fe (001), and MgO (001) barrier, where the incident electron beam was parallel to the [100] and [110] azimuths of the MgO substrate. Sharp streak patterns were observed in all layers, indicating the formation of epitaxial and flat interfaces.

Figure 2(a) shows the device resistance as a function of the MgO barrier thickness (t_{MgO}). The device resistance increased exponentially as the MgO barrier thickness increased, indicating tunneling conduction. The right axis of Fig. 2(a) shows the resistance-area product (RA), defined as the device resistance multiplied by the junction area. Figure 2(b) shows a typical magnetoresistance (MR) curve

FIG. 1. (a) Schematic illustration of the device structure. (b) *In-situ* reflection high-energy electron-diffraction images of surfaces of the MgO barrier, Fe, and V buffer.

of the device. The results measured from a tunnel junction with $t_{\text{MgO}} = 0.94$ nm are shown. When the magnetic-field angle is perpendicular to the film plane ($\theta_H = 90^\circ$), the device shows positive magnetoresistance. In an ultrathin Fe layer, shape anisotropy and interface anisotropy at a Fe-MgO interface can be comparable [37], and an external magnetic field easily changes its magnetizations from in-plane to perpendicular. The MR has the same symmetry as the anisotropic magnetoresistance in ferromagnetic metals [38]. Figure 2(c) shows the variation of the MR ratio, defined as the difference in the saturation resistances between perpendicular and in-plane magnetic fields, as a function of the MgO barrier thickness. The MR ratio is almost identical when the MgO barrier thickness increases. Hence, the observed MR in a Fe-MgO-V tunnel junction is not anisotropic magnetoresistance in the metallic Fe layer but TAMR at the Fe-MgO interface. Figure 2(c) shows that the MR ratio decreases for MgO-barrier thicknesses below 0.9 nm. It shows that for $t_{\text{MgO}} < 0.9$ nm, the resistance of the MgO barrier does not govern the device resistance, and the resistance of the metal electrode, which has no MR, significantly contributes to the total device resistance.

Figure 3(a) shows a schematic of the experimental setup for torque characterization, in which the spin-torque FMR combined with TAMR was employed [27].

FIG. 2. (a) Device resistance as a function of the MgO barrier thickness (t_{MgO}). (b) Typical TAMR, where t_{MgO} and resistance are 0.94 nm and 189 Ω, respectively. The magnetic-field angle definition (θ_H) is depicted in Fig. 3(a). (c) Magnetoresistance ratio as a function of t_{MgO} .

Microwave-frequency voltage was applied through a bias tee to the tunnel junction, and the dc voltage generated in the tunnel junction was measured with a lock-in amplifier. Figure 3(b) shows the definition of the torque vector. The blue arrow represents a unit spin vector of the Fe. The torque vector components ($\beta_{//}$, $\beta_{\perp}$) can be expressed as the following modified Landau-Lifshitz-Gilbert equation:

$$\frac{ds}{dt} = \gamma_0 \mathbf{s} \times \mathbf{H}_{\text{eff}} - \alpha \mathbf{s} \times \frac{ds}{dt} - \frac{A}{S} (\beta_{//} \mathbf{s} \times (\mathbf{z} \times \mathbf{s}) + \beta_{\perp} (\mathbf{z} \times \mathbf{s})) \sqrt{\eta} V_{\text{rf}}, \quad (1)$$

where $\mathbf{s}$ and $\mathbf{z}$ are the unit vectors of Fe and the out-of-plane direction, respectively. $\mathbf{H}_{\text{eff}}$ is the effective magnetic field applied to the Fe; γ_0 (<0); and α , S , and A are the gyromagnetic ratio, Gilbert damping constant, and total spin-angular momentum in the Fe layer and area of the tunnel junction, respectively. Here, η is the transmission efficiency of voltage due to the impedance mismatch between the tunnel junction and the transmission line, and V_{rf} is the amplitude of the voltage in the transmission line from the microwave generator. Hence, the amplitude of $\sqrt{\eta} V_{\text{rf}}$ is the applied microwave-frequency voltage in the tunnel junction. The definition and experimental method to obtain η are provided in our previous paper [24]. In a precise sense, γ_0 should be a tensor. However, during FMR measurements, the magnetization angle did not change significantly. Hence, we treated it as a constant. Note that $\mathbf{z}$ is the rotational symmetry axis if we do not take into account the fourfold crystal structure in the system. Therefore, $\mathbf{z}$ should be the reference vector in the system, and the coordinates of $\mathbf{s} \times (\mathbf{z} \times \mathbf{s})$ and $\mathbf{z} \times \mathbf{s}$ are employed to express the torque vector ($\beta_{//}$, $\beta_{\perp}$). Although there can be an in-plane symmetry breaking axis due to the crystal structure of Fe, the $\mathbf{z}$ reference axis is a standard and most suitable choice because we apply the current or voltage in the $\mathbf{z}$ direction. In all of the experiments in this study, the direction of in-plane projection of $\mathbf{s}$ is set to $\mathbf{x}$, which corresponds to [100] azimuth of the Fe. Figure 3(c) shows typical spin-torque FMR spectra of a tunnel junction with $t_{\text{MgO}} = 0.94$ nm and microwave power of 0.1 μW. A

magnetic field tilted 45° from the film plane was applied to maximize the dc voltage signals. To characterize the torque vector, the obtained spectra were fit by the following equation [20,21]:

$$V_L \frac{(\Delta H)^2}{(H - H_{\text{FMR}})^2 + (\Delta H)^2} + V_{\text{AL}} \frac{(\Delta H)^2}{(H - H_{\text{FMR}})^2 + (\Delta H)^2} \frac{H - H_{\text{FMR}}}{\Delta H} + aH + b. \quad (2)$$

Equation 2 consists of the linear combination of the Lorentzian function, the first term with the fitting parameter V_L and the anti-Lorentzian function, and the second term with the fitting parameter V_{AL} . Here, H is an external magnetic field, and V_L , V_{AL} , resonant field (H_{FMR}), linewidth (ΔH), and linear background (a , b) are taken as fitting parameters. From the fitting parameters of V_L and V_{AL} , torque vector components of $\beta_{//}$ and $\beta_{\perp}$ can be obtained using the following equation:

$$V_{L(\text{AL})} = -\frac{1}{2} \sin \theta \sin 2\theta \frac{R(\theta)(R_{\theta=0^\circ} - R_{\theta=90^\circ})}{R_{\theta=0^\circ} R_{\theta=90^\circ}} \times \frac{\beta_{//(\perp)}}{2\pi\gamma_0 \Delta H} \eta (V_{\text{RF}})^2. \quad (3)$$

To derive Eq. (3), we employed the analysis provided in Ref. [24] and angle dependence of the TAMR, $R^{-1}(\theta) = (R_{\theta=90^\circ}^{-1} + R_{\theta=0^\circ}^{-1})/2 + (R_{\theta=90^\circ}^{-1} - R_{\theta=0^\circ}^{-1})/2 \times \cos 2\theta$. Here, $R(\theta)$ is the device resistance when the spin-vector direction defined in Fig. 3(b) is θ . From the angle dependence of the TAMR, we can deduce the values of θ in the torque analysis.

III. RESULTS AND DISCUSSION

Figure 4(a) shows torque vector components in Fe-MgO-V tunnel junctions, which were obtained by spin-torque FMR spectra and Eqs. (2) and (3). Black

FIG. 3. (a) Schematic illustration of the measurement setup. (b) Definition of perpendicular ($\beta_{\perp}$) and in-plane ($\beta_{\parallel}$) torque. (c) Typical spectra for spin-torque FMR. The magnetic-field direction is $\theta_H = 45^\circ$, and the input power (P_{rf}) is $0.1 \mu\text{W}$.

and red circles represent perpendicular and in-plane components, respectively. The measurements were conducted under a magnetic-field angle of $\theta_H = 45^\circ$, and input microwave power and frequency were set to $P_{\text{rf}} = 0.1 \mu\text{W}$ and 5 GHz, respectively. First, a significant in-plane component was not observed in every device, and the perpendicular component was dominant. The theory predicted that the current-driven in-plane torque could be dominant because of interfacial spin-orbit splitting [26], but this is inconsistent with our observation. This does not mean that there are some problems with the theoretical estimation of the in-plane torque in Ref. [26]. The inconsistency might be because the simple model employed in Ref. [26] did not include the physics that can induce the large perpendicular torque in our experiment. Second, the perpendicular torque is voltage driven. The variation of the perpendicular torque in Fig. 4(a) is at most twice as large, whereas the current density of the tunnel junction with $t_{\text{MgO}} = 0.8 \text{ nm}$ is 140 times larger than that with $t_{\text{MgO}} = 1.4 \text{ nm}$. Therefore, the perpendicular component is not current driven but voltage driven. Note that the current-induced magnetic field cannot be the origin of the voltage-driven perpendicular torque. Any influence of the spin-Hall effect from the V underlayer is also excluded in the same manner.

We have also characterized the in-plane magnetization direction (φ) dependence of the torque, but significant change was not observed, which also supports our premise that the current-induced magnetic field and the spin-Hall effect are not the origin of the torque. The analysis above shows that the voltage-driven perpendicular torque originates from the Fe-MgO interface. The dashed curve in

Fig. 4(a) shows that the maximum torque was about $2.8 \times 10^{-5} \text{ J}/(\text{Vm}^2)$, which is the same as the voltage-driven torque induced by the large VCMA of 180 fJ/Vm at the Fe-MgO interface. To the best of our knowledge, this is larger than any previously reported VCMA-induced voltage-driven torque [24,25,31–35].

Figure 4(b) shows the input power dependence of peak-to-peak dc voltage in the spin-torque FMR spectra of devices with $t_{\text{MgO}} = 0.94 \text{ nm}$. The microwave frequency was 5 GHz. A linear relation between the peak-to-peak voltage and the input power is only confirmed in the small input power region, namely, less than 10^{-4} mW . This indicates that large spin precession is excited by only a small input power, which is consistent with the large torque vector components obtained in Fig. 4(a). More quantitatively, the magnetization precession angle (1.5° at $P_{\text{rf}} = 10^{-4} \text{ mW}$) is of the same order as that of the highly sensitive spin-torque diode device with low resistance-area product ($2.5 \Omega \mu\text{m}^2$, 3.6° at $P_{\text{rf}} = 10^{-4} \text{ mW}$) [8].

To characterize the VCMA at the Fe-MgO interface in the device, the dc-bias-voltage dependence of the resonant field was characterized as shown in Fig. 4(c). Microwave frequency and power were 5 GHz and $0.1 \mu\text{W}$, respectively. Figure 4(c) shows that the resonant field changes almost linearly as a function of the dc bias voltage. The resonant field change corresponds to a VCMA of about 20 fJ/Vm . Note that the VCMA of 20 fJ/Vm is too small to reproduce the observed large perpendicular torque component in Fig. 4(a).

Figure 4(d) shows a dc-bias-voltage dependence of the perpendicular torque component. The torque was

FIG. 4. (a) Torque vector components. Black and red circles represent perpendicular ($\beta_{\perp}$) and in-plane ($\beta_{\parallel}$) torque, respectively. The dashed curve shows the theoretical line of the torque induced by the voltage-controlled magnetic anisotropy of 180 fJ/Vm. (b) Input power dependence of peak-to-peak dc voltage in the spin-torque FMR spectra. (c) The dc-bias-voltage dependence of the resonant field. (d) The dc-bias-voltage dependence of the perpendicular torque. All data were obtained with a magnetic-field angle of $\theta_H = 45^\circ$ and an input microwave frequency of 5 GHz.

characterized by Eq. (3), where dc-bias-voltage dependence of the MR ratio is included. The torque shows nonlinear dc-bias-voltage dependence and takes its maximum around zero dc bias. The dependence is unique because the reported VCMA at a Fe-MgO interface is linear [24,25,31–33]; hence, VCMA-induced voltage-driven torque should be independent of the dc bias.

From the discussion above, the Fe-MgO-V tunnel junction shows large voltage-driven perpendicular torque. The torque originates from the Fe-MgO interface; however, it cannot be explained by conventional VCMA at the Fe-MgO interface. Namely, the spin torque is equivalent to a *dynamic* VCMA of 180 fJ/Vm; however, the Fe-MgO interface shows a *static* VCMA of only 20 fJ/Vm. In the previous studies of Fe(Co)-MgO interfaces, a static VCMA of 20–30 fJ/Vm was reported [24,37,39], which is consistent with the present study. In addition, dynamic VCMA was observed, as the voltage-driven torque was basically identical to the static VCMA [24,25,32,33]. Contrary to this, the dynamic VCMA is several times larger than that of the static VCMA in the present study.

Figure 4(d) shows that electronic states at the Fe-MgO interface around the Fermi level are likely to have a strong influence on the unique torque property. Hence, to characterize the electronic state of the Fe-MgO interface, the dc-bias-voltage dependence of the TAMR was characterized. For this purpose, a device with $t_{\text{MgO}} = 1.77 \text{ nm}$, with $10\text{-}\mu\text{m}^2$ -sized junctions, was fabricated. The magnetoresistance under 1.5 T at various magnetic field angles was

measured using the dc two-terminal method, and it is displayed in Fig. 5(a) as black circles. The data in Fig. 5(a) can be fitted using the following equations [40,41]:

$$R(\theta)^{-1} = \sum_{n=0}^4 G_{2n\theta} \cos 2n\theta, \quad (4)$$

$$\frac{\cos \theta}{\sin \theta} = \frac{H \cos \theta_H - H_d \cos \theta}{H \sin \theta_H}. \quad (5)$$

Here, H , θ , and θ_H are, respectively, the magnetic field strength, magnetization angle, and magnetic field angle defined in Fig. 3(a). Note that H_d is the saturation magnetic field in the perpendicular direction ($\theta_H = 90^\circ$), and $G_{2n\theta}$ are fitting coefficients. The red curves in Fig. 5(a) are the fit results. Fit coefficients of twofold ($G_{2\theta}/G_0$) and fourfold ($G_{4\theta}/G_0$) components are depicted in Figs. 5(b) and 5(c). The error bars in Figs. 5(b) and 5(c) are the results when H_d varies from 0.4 to 0.7 T.

The TAMR is correlated with electronic states at the Fe-MgO interface [40–42]. The origin of the twofold component in the TAMR is similar to the AMR. The twofold component can appear if one symmetry axis exists: that is, electric current direction and spin-dependent scattering due to the spin-orbit interaction at the Fe-MgO interface. The fourfold component cannot be explained in the same manner. It should be correlated with the crystal

FIG. 5. (a) The dc-bias-voltage dependence of TAMR. Black circles and red curves are experimental data and fitting curves using Eq. 4, respectively. (b) Twofold component of the TAMR. (c) Fourfold component of the TAMR.

structure of Fe and/or MgO that has fourfold symmetry. As shown in the previous study [40], the fourfold component in the TAMR indicates the existence of an interface state at the Fe-MgO interface. Figure 5(c) shows that the fourfold component is not significant under negative dc bias and that it is only significant under positive bias. This indicates that an interface state at the Fe-MgO interface exists around the Fermi level of the Fe as schematically shown in the inset of Fig. 5(c). An interface resonance state with a similar energy level at the Fe-MgO interface is also reported in previous studies [43,44]. Interestingly, the torque component in Fig. 4(d) takes its maximum when the energy level is almost identical to the interface state characterized by the angular dependence of the TAMR as shown in Fig. 5.

As discussed, the Fe-MgO-V tunnel junction in the present study shows unique properties as compared with previous studies [24,37,39]. Namely, the dynamic VCMA that induces voltage-driven perpendicular torque is several times larger than the static VCMA. Finally, we would like to discuss unique points in the device structure and possible origins of the large voltage-driven torque.

The V buffer layer is worth discussing because it is a unique feature of the device structure, and V can form an alloy with Fe. In addition, because the lattice constants of Fe, V, and MgO are 0.286, 0.303, and 0.298 nm, respectively, the in-plane lattice constant of the Fe can be expanded. It is reported that the underlayer material can change the VCMA coefficient and its dc-bias-voltage dependence at the Fe(CoB)-MgO interface [36,45–49], which can be induced by alloying with the underlayer materials [47] and/or lattice distortion [48,49]. However, in all the studies mentioned above, large modified VCMA was

obtained with post-annealing. Contrary to this, post-annealing was not conducted in the Fe-MgO tunnel junction in the present study. In addition, we have also conducted similar measurements with the same sample post-annealed at 250 °C for 30 minutes, and we found that the magnitude of the torque was almost identical to as-deposited devices, but the perpendicular magnetic anisotropy and ferromagnetic resonance linewidth of the Fe slightly increased. Therefore, it should be noted that the existence of the large torque is robust to the sample fabrication process. The only unique point in our sample is the small MgO barrier thickness (<1.1 nm). All of the previous studies concerning VCMA in magnetic tunnel junctions employed thicker MgO barriers than the present study.

The influence of electrochemical reactions [50–53] and/or charge traps [36] should be discussed as a possible origin of the large torque. However, in such cases, dynamic VCMA inducing torque should be smaller than static VCMA. This is because electrochemical reactions and/or charge trap inducing static VCMA have limited operation speed owing to thermal activation processes. Moreover, it is reported that there is no voltage-driven electrochemical reaction that substantially changes a valence state of d orbitals [54–56].

To explain the observed phenomena in a different way, they occur only when very high frequency ($>\text{GHz}$) or small (for instance, $\sqrt{\eta}V_{\text{rf}}$ under $P_{\text{rf}} = 10^{-4}$ mW is 3.2 mV) external voltage is applied; thus, a large voltage-driven perpendicular torque was induced. Hence, if the MgO barrier in the tunnel junctions has some localized state [57] and its capacitance shows dc-bias-voltage dependence, then

the nonlinearity in the bias dependence of the torque can be explained. In either case, nonlinear dc-bias dependence of the voltage-driven torque in Fig. 4(d) demonstrates the importance of the surface state at the Fe-MgO interface. Interestingly, the voltage-driven torque takes its maximum around the interface state at the Fe-MgO interface as shown in Fig. 5. For further discussion, a theoretical approach including a first-principles study would be indispensable.

IV. CONCLUSION

To conclude, the torque vector at an epitaxial Fe-MgO interface was characterized by spin-torque FMR in a CPP Fe-MgO-V tunnel junction. The current-driven torque, which can be induced by an interfacial spin-orbit splitting at Fe-MgO, was negligible; however, the voltage-driven perpendicular torque component increases as the MgO barrier thickness decreases. The obtained maximum torque is as large as 2.8×10^{-5} J/(Vm²) for a thin MgO barrier (<1.1 nm). The perpendicular torque is comparable to the VCMA at an Fe-MgO interface of 180 fJ/Vm. The voltage-driven torque component shows nonlinear dc-bias-voltage dependence, and it takes its maximum when the energy level is in the vicinity of the Fermi level, where the interface state at the Fe-MgO interface is also located. This surface-state-sensitive electric modulation of magnetic properties provides new insight in the field of interface magnetism.

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