Epitaxial integration of a perpendicularly magnetized ferrimagnetic metal on a ferroelectric oxide for electric-field control

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Abstract Ferrimagnets, which contain the advantages of both ferromagnets (detectable moments) and antiferromagnets (ultrafast spin dynamics), have recently attracted great attention. Here, we report the optimization of epitaxial growth of a tetragonal perpendicularly magnetized ferrimagnet Mn$_2$Ga on MgO. Electrical transport, magnetic properties and the anomalous Hall effect (AHE) were systematically studied. Furthermore, we successfully integrated high-quality epitaxial ferrimagnetic Mn$_2$Ga thin films onto ferroelectric 0.7PbMg$_{1/3}$Nb$_{2/3}$O$_3$–0.3PbTiO$_3$ single crystals with a MgO buffer layer. It was found that the AHE of such a ferrimagnet can be effectively modulated by a small electric field over a large temperature range in a nonvolatile manner. This work thus demonstrates the great potential of ferrimagnets for developing high-density and low-power spintronic devices.

Keywords Ferroelectric oxides; Ferrimagnetic metals; PMN-PT; Mn$_2$Ga; Anomalous Hall effect

1 Introduction

Contemporary mass storage for data centers predominantly relies on hard disk drives that are based on perpendicularly magnetized ferromagnetic granular films, the spin states of which can be easily manipulated by external magnetic fields generated by current coils due to remarkable macroscopic moments. However, as limited by the characteristic GHz spin dynamics, ferromagnetic materials could hardly be utilized for the static random-access memory technology and the in-memory computing, which needs a sub-ns response speed. Similar to ferromagnets, ferrimagnets possess large magnetic moments. Besides, they exhibit ultrahigh spin dynamics of THz as a result of antiferromagnetic exchange coupling [1–6] akin to antiferromagnets. Hence, ferrimagnets are rising star materials for new-generation sub-ns information devices [7–12].

Tetragonal D$_{022}$ Mn$_2$Ga with $a = 0.3905$ nm and $c = 0.7193$ nm, a classical ferrimagnet with a high Curie temperature of $\approx 710$ K, exhibits large magnetic anisotropy along its [001] crystallographic orientation. (001)-oriented single-crystalline or textured films are thus perpendicularly magnetized. In addition, D$_{022}$ Mn$_2$Ga is of high spin polarization at the Fermi level and low Gilbert damping constant [13], both of which are useful for spin valves, perpendicular magnetic tunnel junctions [14], narrow-band terahertz emission from coherently excited spin precession [15], and high-density spin-transfer-torque magnetoresistive random access memories (STT-MRAM) [16].

However, the information writing (corresponding to the spin state manipulation) of Mn$_2$Ga-based spintronic devices such as spin valves and STT-MRAM mostly depends on electrical-current-generated magnetic fields or electrical currents, which generates significant Joule heating and hence results in high power consumption. Alternatively, if one can effectively control the spin state of Mn$_2$Ga with a non-electrical-current manner, the energy for writing a bit could be substantially lowered.

An electric field applied onto a conductor yields an electrical current. In contrast, for highly insulating ferroelectric oxide materials, the application of an electric field
generates a negligible current. Instead, it induces piezoelectric strain [1–7, 17–23]. Assuming (001)-oriented D022 Mn2Ga could be epitaxially integrated onto ferroelectric oxides, electric-field-generated strain could harness the spin state of Mn2Ga as the electronic states of solid-state materials are all sensitive to the periodic lattice.

Nevertheless, epitaxial growth of intermetallic Mn2Ga on a ferroelectric oxide is quite challenging. The key reasons are: (1) highly ordered epitaxial films need high thermal energy to let the atoms diffuse into their equilibrium sites during thin film growth, and therefore, high growth temperatures are required; (2) intermetallic alloys are of strong chemical activity, which can easily be oxidized or form secondary alloys at high temperatures while landing on ferroelectric oxide substrates; (3) lattice mismatch between intermetallic alloys and ferroelectric oxides could break epitaxial growth. Accordingly, a proper oxide substrate or an oxide buffer layer with robust high-temperature stability and low oxygen diffusion coefficient could be crucial for realizing epitaxial growth of Mn2Ga on ferroelectrics. In this letter, we report the optimal epitaxial growth of D022 Mn2Ga on ferroelectric PMN-PT. Furthermore, electric-field control of its AHE has been achieved over a large temperature range, which paves the way for low-power Mn2Ga spintronic device applications.

2 Experimental

MgO, a transparent insulating oxide with a large bandgap of 7.8 eV, shows excellent high-temperature stability with a melting point of 2800 °C. It has a cubic lattice with \( \alpha = 0.4212 \) nm, which is close to the in-plane lattice constant of D022 Mn2Ga. Therefore, Mn2Ga films were firstly grown on single-crystalline (001)-oriented MgO substrates using the magnetron sputtering technique at different temperatures ranging from 30 to 550 °C. The base pressure and the direct current (DC) sputtering power were \( 1.0 \times 10^{-6} \) Pa and 60 W, respectively. During the deposition, the Ar pressure was kept at \( 4.0 \times 10^{-1} \) Pa. With X-ray reflectometry, the deposition rate was determined to be 3.5 nm·min\(^{-1}\) and the total thickness was first kept at 100 nm, which was later changed to 50 and 30 nm for thickness dependence studies. The crystal structure of Mn2Ga thin films was measured by a four-circle Bruker D8 Discover X-ray diffractometer (XRD). Magnetic characterization and electrical measurements were performed by a Quantum Design VersaLab with a vibrating sample magnetometer option. The standard linear four-probe method and the Hall geometry were used for longitudinal and Hall resistance measurements, respectively.

3 Results and discussion

Figure 1a shows single-crystal XRD patterns of Mn2Ga films grown on MgO substrates fabricated at different growth temperatures (\( T_G \)). For \( T_G \) below 450 °C, no thin-film peaks are seen, implying disordered polycrystalline films. At 450 °C, the (002) peak and a weak (001) peak of D022 Mn2Ga show up. With the growth temperature increasing to 550 °C, both the (001) and (002) peaks of D022 Mn2Ga become sharper, indicating enhanced crystallinity and chemical ordering. The growth temperature could not be further raised. That is because at higher temperatures, the surface energies of intermetallic Mn2Ga and ferroelectric oxide PMN-PT are largely different, and due to the non-wetting issue, Mn2Ga could not form continuous thin films but only separate islands.

The metallicity, which could be examined by the normalized resistivity relative to room-temperature values, is demonstrated in Fig. 1b. When the substrate temperature is lower than 450 °C, Mn2Ga/MgO films are semiconducting, which is contrary to its bulk metallic behavior. This suggests the existence of a large degree of chemical disorder including grain boundaries. The films fabricated at 450 and 550 °C are metallic, and the metallicity is improved with enhancing substrate temperature. All these electrical transport results are consistent with XRD patterns shown in Fig. 1a.

The out-of-plane and in-plane magnetic moments versus magnetic fields (\( M-H \)) are measured at 50 and 300 K for single-crystalline Mn2Ga/MgO films (Fig. 1c–f). Overall, both films exhibit the feature of perpendicularly magnetic anisotropy. The room-temperature saturation magnetization of Mn2Ga/MgO films is \( \sim 290 \) and \( \sim 335 \) kA·m\(^{-1}\) for growth temperature of 450 and 550 °C, respectively, which are comparable with the magnetization values of previously reported ferrimagnetic Mn2Ga films [24]. Similar to the XRD and electrical resistivity data, the increase in the growth temperature improves the squareness of the out-of-plane \( M-H \) loop, which is favorable for perpendicular spintronic device applications. Therefore, 550 °C is the optimized growth temperature for high-quality epitaxial and perpendicularly magnetized ferrimagnetic Mn2Ga films.

In addition, the room-temperature anisotropy field \( \mu_0 H_k \) is determined to be \( \sim 10 \) T for the 550 °C-fabricated Mn2Ga film, which corresponds to a uniaxial magnetocrystalline anisotropy energy (\( K_u \)) of \( \sim 1.68 \) MJ·m\(^{-3}\), the same order with the previously reported highest \( K_u \) (\( \sim 2.17 \) MJ·m\(^{-3}\)) [25] for molecular-beam-epitaxy-fabricated ferrimagnetic Mn-Ga films. For real applications, the duration of information storage needs to be more than 10 years, which requires the ratio of magnetocrystalline energy of a bit \( K_u V \) greater than 40 times room-temperature...
thermal energy of $40k_BT \sim 1.67 \times 10^{-19}$ J. Considering a bit cell consisting of a single Mn$_2$Ga layer with a cubic shape, the critical bit size could accordingly be reduced to $\sim 4.6$ nm. This thus implies great potential of our optimized Mn$_2$Ga ferrimagnetic thin films with perpendicular magnetized anisotropy for high-density data storage.

Longitudinal magnetotransport properties of Mn$_2$Ga thin films fabricated at various temperatures were examined for out-of-plane magnetic fields. It was found that for $T_G < 300$ °C, the magnetoresistance (MR) effect is rather weak and almost not affected by the growth temperature. Figure 2 shows MR curves collected at different temperatures ranging from 50 to 300 K for Mn$_2$Ga thin films with $T_G > 30$ °C. For $T_G = 150$ °C (Fig. 2a), the MR above 50 K is positive, which implies that the orbital scattering due to the Lorentz force is dominant. However, the MR turns into negative for 50 K, suggesting the important role of magnetic moments of Mn. For $T_G = 300$ °C (Fig. 2b), the room-temperature MR is negligible, while the low-temperature MR curves are interestingly linear. It is worth noticing that the positive MR at 150 K is the largest, reminiscent of the maximal magnetotransport properties at $\sim 200$ K for noncollinear antiferromagnets Mn$_3$Sn [26] and Mn$_3$Ge [4]. For crystallized epitaxial Mn$_2$Ga films, the butterfly-shape hysteresis MR curves (Fig. 2c, d) are clearly seen, characteristic of long-range ferrimagnetic/ferromagnetic order. In addition, the positive orbital scattering is more significant at low temperatures, leading to suppressed negative MR.

Systematic transverse magnetotransport properties, i.e., the Hall effect, of the Mn$_2$Ga/MgO films deposited below 450 °C, are demonstrated in Fig. 3. Similarly, the Hall curves for $T_G = 30$ and 150 °C (Fig. 3a, b) are comparable and are linear above 50 K. At 50 K, the Hall effect becomes nonlinear, signature of the magnetic-moment-related AHE, which is consistent with the negative MR at 50 K in Fig. 2a. For $T_G = 300$ °C, the AHE is obvious below 200 K, indicating the formation of magnetic order.

The Hall effect of epitaxial Mn$_2$Ga films is shown in Fig. 4a, b. The general shape of the Hall curves is in excellent agreement with that of the $M$-$H$ loops. Therefore, the Hall effect could serve as a sensitive electrical probe to magnetic properties of perpendicularly magnetized Mn$_2$Ga. Detailed scaling law analysis [7] (Fig. 4c) on the optimized film reveals that for low longitudinal resistivity range $280 \mu\Omega\text{cm} < \rho_{xx} < 330 \mu\Omega\text{cm}$, $\rho_{xy}^2 \approx \rho_{xx}^2$, the Berry curvature is the dominant origin for the AHE, which is a pseudo magnetic field in momentum space and determined by the topological bands interaction of Bloch electrons [27, 28]. While for the large resistivity region with $\rho_{xx} >$
330 μΩ-cm, skew scattering becomes more important for generating a transverse Hall voltage, leading to $\rho_{xy} \approx \rho_{xx}$ [29]. The excellent perpendicular magnetic anisotropy remains in thinner films such as 50 and 30 nm (Fig. 5), which, in turn, leads to significantly enhanced anomalous Hall resistance. The much larger anomalous Hall resistance in thinner Mn$_2$Ga films could facilitate the electrical read-out for memory devices.

Based on the experimental results mentioned above, 30-nm-thick Mn$_2$Ga films were further epitaxially integrated onto (001)-oriented PMN-PT ferroelectric oxides with a 25-nm-thick MgO buffer layer so as to manipulate its AHE or magnetism by electric-field-induced piezoelectric strain [1–6, 17–23, 30]. The MgO buffer layers were grown by a pulsed laser deposition system at 400 °C and post-annealed at 600 °C for 1 h, which was utilized to
prevent Pb element in PMN-PT substrates from diffusing into the chamber and Mn$_2$Ga films to form secondary alloys at high temperatures [18, 31]. As shown in Fig. 6a, XRD spectrum containing (002) peak of the MgO buffer layer and the (001) and (002) peaks of the Mn$_2$Ga thin film indicates the epitaxial growth of the Mn$_2$Ga on the MgO.

Fig. 4 Hall effect for a 100-nm-thick Mn$_2$Ga/MgO film fabricated at a $T_G = 450\, ^\circ C$ and b $T_G = 550\, ^\circ C$; c scaling law analysis for the Mn$_2$Ga film fabricated at $T_G = 550\, ^\circ C$

Fig. 5 Hall effect of Mn$_2$Ga/MgO films with smaller thickness fabricated at $T_G = 550\, ^\circ C$: a 50 nm; b 30 nm

Fig. 6 a XRD pattern of a Mn$_2$Ga/MgO/PMN-PT heterostructure; b Hall effect measurements of Mn$_2$Ga/MgO/PMN-PT heterostructure at different temperatures ranging from 50 to 400 K
buffer layer. The field-dependent Hall signals of an epitaxial Mn$_2$Ga/MgO/PMN-PT heterostructure (Fig. 6b) at different temperatures are in concert with that of Mn$_2$Ga/MgO in Fig. 4b, which suggests the excellent perpendicular magnetic anisotropy of the epitaxially integrated Mn$_2$Ga films on ferroelectric PMN-PT.

To explore the effect of piezoelectric strain on the AHE in the Mn$_2$Ga/MgO/PMN-PT heterostructure, an electric field $E_G$ of $-5$ kV/cm was perpendicularly applied across the PMN-PT substrate (Fig. 7a) to pole the ferroelectric substrate at room temperature. To examine any possible variation of the AHE, the Hall curves were re-measured after electric poling of the PMN-PT. It turns out that under such an electric-field excitation, the AHE is enhanced for all the temperatures (Fig. 7b–g). The relative electric-field-induced nonvolatile modulation of the zero-field anomalous Hall resistance is extracted and plotted in Fig. 7h, which reaches $\sim 16\%$ below 200 K and $\sim 14\%$ at 300 K.

To further confirm the nonvolatile nature of the electric-field-induced piezoelectric strain in PMN-PT, the room-temperature electric-field-dependent longitudinal resistance of the Mn$_2$Ga film in the Mn$_2$Ga/MgO/PMN-PT heterostructure is measured with the linear four-probe geometry (Fig. 8a). As shown in Fig. 8b, the positive and negative peaks in perpendicular gating current through the MgO buffer layer and PMN-PT clearly exhibit the reversible ferroelectric polarization switching feature. Correspondingly, the electric-field-dependent longitudinal resistance (Fig. 8c) shows an asymmetric and nonvolatile butterfly loop, which is similar to what we obtained in previous measurements [3, 23].

Empirically, the AHE in ferromagnetic materials is closely related to magnetization. Motivated by this understanding, we examined the out-of-plane magnetization change of the Mn$_2$Ga film for the Mn$_2$Ga/MgO/PMN-PT heterostructure upon electric-field poling (Fig. 9a) of the ferroelectric substrate PMN-PT. As shown in Fig. 9b, c, the perpendicular magnetization has been changed substantially. At 50 K, the electric-field poling of the PMN-PT alters the saturation magnetization of Mn$_2$Ga from $\sim 360$ to $\sim 423$ kA/m (Fig. 9b), which corresponds to an $\sim 17.5\%$ magnetization enhancement, similar to the anomalous Hall resistance increase ratio in Fig. 7b. For 300 K, the out-of-plane magnetization changes from $\sim 335$ to $\sim 382$ kA/m (Fig. 9c), well consistent with the $\sim 14\%$ anomalous Hall resistance variation in Fig. 7g. Thus, these experimental results clearly illustrate that the piezoelectric-strain-induced anomalous Hall effect modulation is predominantly caused by the strain-induced magnetization variation. For ferrimagnetic materials with two opposite unequal sublattices, the enlargement of the net magnetization would likely pertain to the weakening of the compensation of two sublattices in terms of the spin rotation, reminiscent of the scenario of noncollinear antiferromagnetic spin structure modulation by piezoelectric strain as theoretically described by Lukashev et al. [32].
In conclusion, we have fabricated epitaxial ferrimagnetic Mn$_2$Ga thin films with perpendicular magnetic anisotropy on MgO substrates. The mechanisms of the AHE were unveiled for different longitudinal resistivity ranges. When MgO is used as buffer layer, Mn$_2$Ga thin films with perpendicular anisotropy have been successfully integrated onto ferroelectric PMN-PT substrates which is useful for utilizing ferrimagnetic materials in high-density spintronic devices and could enable the fabrication of other exotic epitaxial heterostructures with ferrimagnets interfacing with some novel materials [33–46]. Via the defects engineering in thin films [47], the spin structure and the AHE of ferrimagnetic Mn$_2$Ga could further be modulated to realize the topological Hall effect. More importantly, the AHE of ferrimagnetic Mn$_2$Ga films is largely modulated by the electric-field-induced piezoelectric strain, which paves the way for magnetic-field-free low-power ferrimagnetic spintronic device applications. Furthermore, this type of multiferroic devices could be promising for mechanical energy harvesters, magnetic-field sensors, electromagnetic wave generators and other piezo/magnetic applications [48–52].

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**Declarations**

**Conflicts of interests** The authors declare that they have no conflict of interests.

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