

Structural and magnetic properties of epitaxial Mn_3Ga thin films

Quanzheng Tao^{1,2}, Andrejs Petruhins², Rui Shu², Alexis Papamichail², Iurii Kibalin³,
Bachir Ouladdiaf³, Fabio Orlandi⁴, Dmitry Khalyavin⁴, Pascal Manuel⁴, Vanya Darakchieva^{2,5},
Johanna Rosen², and Andrew T. Boothroyd¹

¹*Department of Physics, University of Oxford, Clarendon Laboratory, Oxford OX1 3PU, United Kingdom*

²*Department of Physics, Chemistry, and Biology (IFM), Linköping University, SE-581 83 Linköping, Sweden*

³*Institut Laue-Langevin, 71 avenue des Martyrs, CS 20156, 38042 Grenoble cedex 9, France*

⁴*ISIS Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Oxfordshire OX11 0QX, United Kingdom*

⁵*Terahertz Materials Analysis Center, THeMAC, Solid State Physics and NanoLund, Lund University, S-22100 Lund, Sweden*



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We report on the synthesis and characterization of Mn_3Ga thin films with controlled phase and orientation using DC magnetron sputtering. High-quality Mn-deficient hexagonal Mn_3Ga films with C-plane and M-plane orientations were achieved on various single-crystal substrates, and their structural properties were systematically investigated by x-ray diffraction. Magnetization, Hall effect, and neutron diffraction measurements confirm the formation of antiferromagnetic order below $T_N = 460\text{--}470$ K and reveal its characteristic magnetic behavior. A sizable anomalous Hall effect is observed coexistent with weak net magnetization, consistent with the 120° noncollinear spin structure. The ability to tune both the crystal phase (hexagonal vs tetragonal) and orientation through composition and thermal treatment provides a platform for exploring the intrinsic magnetic and topological properties of Mn_3Ga , with promising implications for antiferromagnetic spintronic applications.

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I. INTRODUCTION

Hexagonal Mn_3X ($X = \text{Sn}$, Ge , and Ga) are proposed to be magnetic Weyl semimetals [1,2] and are promising for antiferromagnetic spintronics [3,4]. These materials also show an intricate correlation between electronic and magnetic structure. They show a large anomalous Hall effect at room temperature, which can be switched with a small magnetic field [5–9]. The unique properties can be useful for next-generation data storage applications, as the stray field in a conventional ferromagnet is the bottleneck to further increasing storage density. Antiferromagnets, on the other hand, have no or negligible stray field, which in principle can afford much higher storage densities compared to ferromagnets. An additional advantage of antiferromagnets is that they support ultrafast spin dynamics [10].

Hexagonal Mn_3Sn and Mn_3Ge with space group $P6_3/mmc$ have been widely studied in both single-crystal and thin-film form. Below $T_N \approx 420$ K (Mn_3Sn) and 380 K (Mn_3Ge) they form a noncollinear antiferromagnetic order known as the anti-chiral triangular spin structure [11,12], which has an associated small residual net magnetization. The symmetry of the magnetic structure permits the formation of Weyl points

in the electronic band structure giving rise to unconventional magnetotransport properties, and the weak ferromagnetism makes it possible to control the properties with an applied magnetic field.

In contrast, the closely related compound hexagonal Mn_3Ga (space group $P6_3/mmc$) is less explored [13,14]. Only a limited number of studies have investigated Mn_3Ga thin films [8,15]. Until recently, single-crystal samples of Mn_3Ga were lacking, making the study of its intrinsic properties difficult [16]. *Ab initio* electronic structure calculations predict that Mn_3Ga has a larger anomalous Hall effect than either Mn_3Sn or Mn_3Ge [17]. Moreover, the magnetic ordering temperature $T_N \approx 460\text{--}470$ K of Mn_3Ga is higher than that of the Sn and Ge compounds. In principle, these characteristics make Mn_3Ga a promising member of the Mn_3X family for technological applications.

On the other hand, Mn_3Ga exhibits complex structural behavior. The hexagonal phase is metastable and of interest for its topological and magnetic properties, but the thermodynamically stable phase at ambient temperature and pressure is the tetragonal phase. A cubic phase has also been reported at high Mn content [18]. Controlling the crystal structure of Mn_3Ga is thus essential for tailoring its properties and enabling device applications.

In this work, we investigate the growth, phase formation, and magnetic properties of Mn_3Ga thin films. Using DC magnetron sputtering, we demonstrate the controlled synthesis of both hexagonal and tetragonal Mn_3Ga phases by tuning the composition and thermal processing conditions. We report

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the successful growth of high-quality *c*-axis (C-plane) and *a*-axis (M-plane)-oriented hexagonal Mn_3Ga films on various single-crystal substrates, including GaN/SiC, MgO, and Ru/ Al_2O_3 . Structural and magnetic characterization confirms the antiferromagnetic nature of the hexagonal phase, while the tetragonal phase exhibits ferrimagnetism. Furthermore, we reveal a large anomalous Hall effect in (100)-oriented hexagonal Mn_3Ga thin films, consistent with its predicted topological character. Finally, neutron diffraction measurements confirm the noncollinear magnetic structure in the thin film, supporting its role as a candidate material for topological antiferromagnetic spintronics.

II. EXPERIMENTAL DETAILS

Thin Mn_3Ga films were deposited on different single-crystal substrates by direct current (DC) magnetron sputtering from elemental targets of Mn and Ga. An ultra-high vacuum chamber was used at a pressure of 3 mTorr Ar, with a base pressure of 10^{-7} Pa. The preparation of a liquid gallium target in a stainless steel holder is described in detail in Ref. [19]. Sputtering targets were operated in constant power mode. The power for the Mn target was tuned between 35 W and 55 W, while the Ga target was maintained at 5 W. The ratio 53:5 W corresponds to a stoichiometry close to 3:1.

For the growth of hexagonal Mn_3Ga , thin films were deposited at a substrate temperature of 400 °C. After deposition, the substrate temperature was increased to 500 °C and the sample was annealed for 10 min. After annealing, the sample was cooled to room temperature in vacuum before removal from the chamber. For the growth of tetragonal Mn_3Ga , thin films were deposited at a substrate temperature of 400 °C, without subsequent annealing. Cubic Mn_3Ga films were deposited on MgO(111) single-crystal substrates by direct current (DC) magnetron sputtering from a compound Mn-Ga target (ACI alloys) with Mn : Ga = 3 : 1 at 400 °C and a pressure of 3 mTorr Ar.

A W seed layer was deposited by magnetron sputtering. The substrate temperature is 200 °C. After deposition, the substrate is heated to 600 °C and annealed for 10 min. The W seed layer was used for growing Mn_3Ga with (100) orientation. A Ru seed layer was deposited by magnetron sputtering. The substrate temperature is 200 °C. After deposition, the substrate is heated to 500 °C and annealed for 10 min. The Ru seed layer was used for growing Mn_3Ga with a (002) orientation.

The crystal structure of the films was investigated by x-ray diffraction with a Rigaku SmartLab diffractometer and a Panalytical Empyrean diffractometer. Magnetization measurements were performed with a Magnetic Properties Measurement System (Quantum Design). Hall effect measurements were performed with a Physical Properties Measurement System (Quantum Design). The Hall effect was measured with a four-terminal geometry, with the magnetic field applied perpendicular to the thin film. The measurement was performed on a 5 mm × 2 mm × 30 nm Mn_3Ga sample with a (100) orientation on W (10 nm)-seeded MgO(110).

Neutron diffraction data were measured on the WISH instrument at ISIS, UK, and the D10+ instrument at the Institut Laue-Langevin (ILL), Grenoble. In both experiments,

the sample consisted of a 100-nm-thick thin film on a $10 \times 10 \text{ mm}^2 \times 0.5 \text{ mm}$ single-crystal GaN-on-SiC substrate. For the WISH experiment, the sample was mounted to access the H0L plane in a furnace [20]. For the D10+ experiment, the sample was mounted in a furnace capable of reaching temperatures close to T_N to separate the crystal-structure contribution from the magnetic diffraction measured at lower temperature. D10+ was used in a four-circle configuration which made it possible to access most of the reflections of interest. We performed crystal rotation (ω) scans for a set of reflections and their Friedel-pair partners. We also performed scans at several forbidden reflections to check for possible magnetic peaks. Structure refinement was performed with the Mag2Pol software [21].

Composition analysis was performed by Rutherford backscattering spectrometry (RBS) and time-of-flight elastic recoil detection analysis (ToF-ERDA) measurements at the Tandem Laboratory of Uppsala University. RBS measurements employed 2 MeV $^4\text{He}^+$ primary ions, which were detected at a scattering angle of 170°. ToF-ERDA measurements were performed using a 40 MeV $^{127}\text{I}^{9+}$ beam at 67.5° incidence relative to the surface normal and a 45° recoil angle [22].

III. RESULTS

Figure 1(a) shows an x-ray diffraction θ -2 θ scan from a 100-nm-thick hexagonal Mn_3Ga grown on GaN (002)/SiC substrate. GaN was grown by MOCVD on 4H-SiC substrate with AlN nucleation layer [23]. As shown in Fig. 1(a), only (002) and (004) reflections of Mn_3Ga can be seen besides those from the substrates and the GaN layer, indicating single-crystalline epitaxial film.

Figure 1(b) shows the azimuthal scans of the GaN (102), and the (201) and (202) peaks of hexagonal Mn_3Ga . Based on the azimuthal scans of both the film and substrate, the epitaxial relationship for the hexagonal Mn_3Ga on GaN is (002) $\text{Mn}_3\text{Ga} \parallel$ (002) GaN out of plane and $[100] \text{Mn}_3\text{Ga} \parallel [100] \text{GaN}$. At the bottom of Fig. 1(b), we also scanned for tetragonal Mn_3Ga (200), which is not present.

Of the three known polymorphs with the nominal Mn_3Ga composition, namely hexagonal (space group 194), tetragonal (space group 139), and cubic Mn_3Ga (space group 221) [18,24,25], we found that the formation of each phase can be readily controlled by tuning the composition and growth temperature. We noticed that when the Mn:Ga ratio is higher than 3:1 and the films are deposited from a compound target, cubic Mn_3Ga forms over a wide temperature range, see the Appendix for details. On the other hand, when the Mn:Ga ratio approaches 2:1, MnGa and Mn_3Ga coexist. In between ~2:1 and 2.5:1, either hexagonal or tetragonal Mn_3Ga is the main phase depending on the growth and annealing temperature. Figure 10 in Appendix shows the evolution of the Mn_3Ga (002) peak as a function of Mn:Ga power ratio. In addition, the crystal quality also degrades when the Mn:Ga ratio approaches 3:1. We carefully adjusted the ratio close to 3:1 while maintaining good crystal quality, as judged by the rocking curve. We found that good sample quality with high Mn content was achieved with the power of Mn and Ga powers of 50 W and 5 W, respectively, as shown in

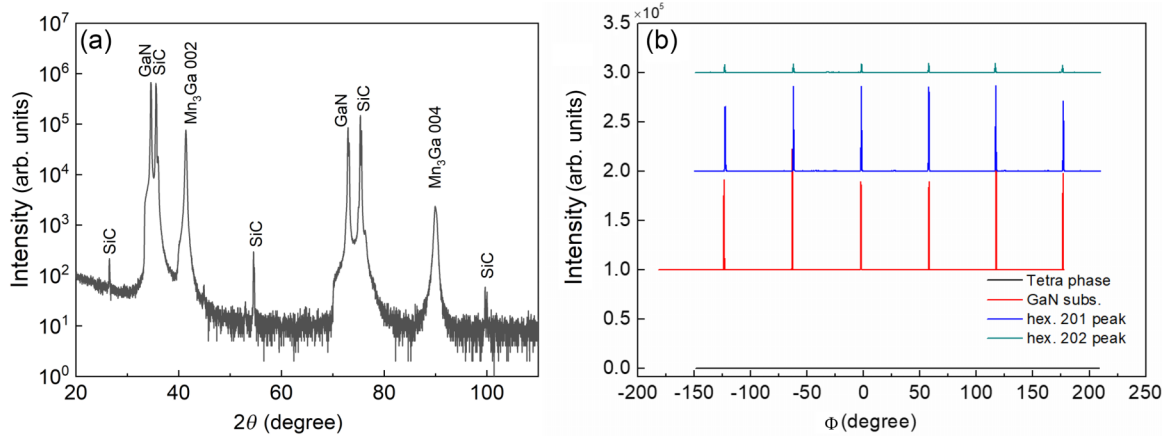


FIG. 1. XRD of hexagonal Mn_3Ga with 002 orientation. (a) θ - 2θ scan. (b) Azimuthal scans of the hexagonal Mn_3Ga (201) and (202) and the GaN (102) reflections, as well as the tetragonal Mn_3Ga (200) reflection.

Fig. 1(a). The composition is determined to be $\text{Mn}_{2.5(1)}\text{Ga}_1$ by RBS.

In addition to GaN/SiC substrates, we investigated the growth of Mn_3Ga on other single-crystal substrates, as shown in Fig. 2. The choice of substrates is discussed in Appendix. These include 4H-SiC, MgO(111), and Ru/ Al_2O_3 (002). The diffraction patterns indicate that hexagonal Mn_3Ga grows epitaxially on all these substrates. Only the Mn_3Ga (002) and (004) reflections are observed, aside from those originating from the substrates. These results demonstrate the versatility of Mn_3Ga epitaxy with respect to substrate selection.

Reciprocal space maps of Mn_3Ga (002) grown on different substrates are shown in Fig. 3. Because the GaN reflections are located far from those of Mn_3Ga in reciprocal space, only the SiC(107) reflection is visible in Fig. 3(a). Owing to the very large lattice mismatch between GaN and Mn_3Ga , the Mn_3Ga film grown on GaN is fully relaxed. Similarly, Mn_3Ga grown directly on SiC is also relaxed. In contrast, Ru exhibits a very

small lattice mismatch with Mn_3Ga , resulting in a slightly strained film. Notably, Mn_3Ga grown on GaN shows a sharp and intense (203) reflection in reciprocal space, indicating high crystalline quality, which is essential for the subsequent neutron scattering experiments.

When studying hexagonal Mn_3Ga , we also noticed that tetragonal and cubic Mn_3Ga can be synthesized under very similar conditions. With the same composition as for hexagonal Mn_3Ga , but annealing at a lower temperature, namely, 400 °C tetragonal Mn_3Ga can be obtained. By using a Mn_3Ga compound target, cubic Mn_3Ga can be synthesized at 400 °C. Hexagonal, tetragonal, and cubic Mn_3Ga appear identical in two theta-theta scans but can be differentiated by azimuthal scans. A brief discussion can be found in the Appendix.

After optimizing the growth of (002)-oriented Mn_3Ga , we studied the growth of Mn_3Ga with (100) surface orientation, also referred to as M-plane Mn_3Ga . Previously, W was successfully used as a seed layer for the growth of (100) Mn_3Sn [26]. Considering the very small lattice mismatch between W and Mn_3Ga , we attempted to use W as a seed layer for the growth of Mn_3Ga . First, a 10-nm W thin film was deposited on an MgO (110) substrate as seed layer, and then Mn_3Ga was deposited.

Figure 4(a) shows the θ - 2θ scan of the M-plane Mn_3Ga on W-seeded MgO. Only Mn_3Ga (020) and (040) reflections are observed as well as W (211) and MgO peaks. Figure 4(b) shows the azimuthal scan of the Mn_3Ga (201), W (110), and MgO (200) peaks, respectively. W and MgO show twofold symmetry as expected. Mn_3Ga also shows a peak at every 180°, rather than the expected sixfold symmetry, which is likely caused by epitaxial strain as observed in the case of Mn_3Sn on W^{28} , also see Appendix for the estimated epitaxial strain. The additional four peaks associated with the sixfold symmetry likely lie just outside the current measurement window. Based on the XRD scans in Figs. 3(a) and 3(b), the epitaxial relationship is $\text{MgO}(110)$ [001] $\parallel$ $\text{W}(211)[110]$ $\parallel$ Mn_3Ga (010)[001].

Figure 5 shows measurements of the magnetization of hexagonal and tetragonal Mn_3Ga thin films at room temperature. In Fig. 5(a), the hexagonal Mn_3Ga shows a very small net magnetic moment of 0.02 $\mu\text{B}/\text{Mn}$, consistent with the

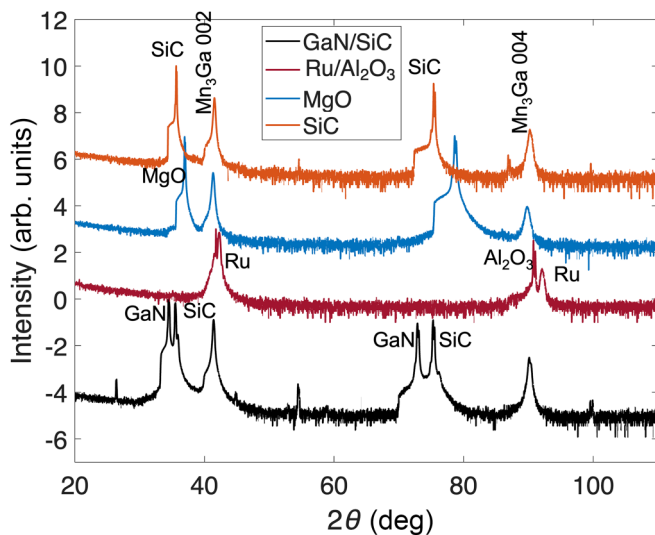


FIG. 2. XRD θ - 2θ scans of hexagonal Mn_3Ga films with 002 orientations on different substrates, GaN/SiC(002), Ru/ Al_2O_3 , MgO(111), SiC(002).

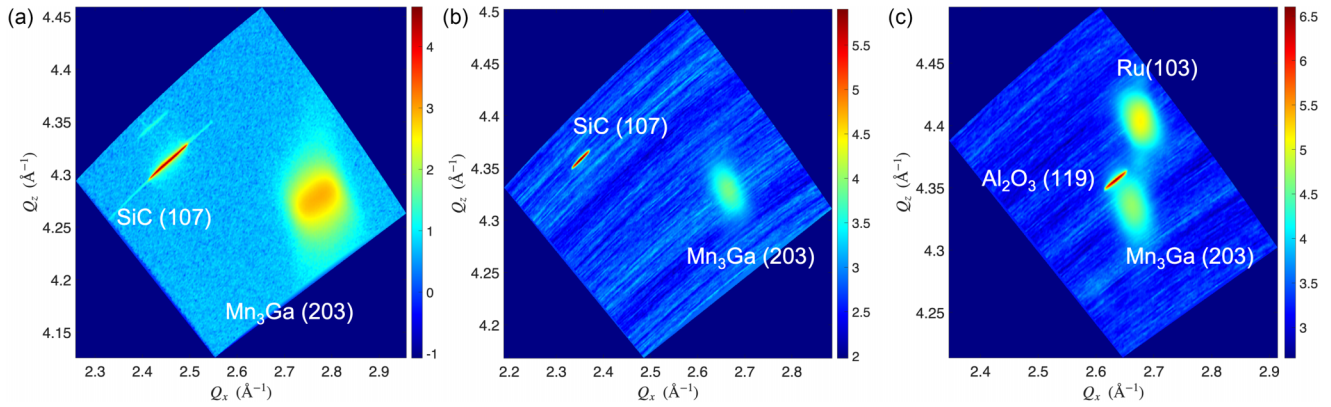


FIG. 3. RSM for Mn₃Ga films with 002 orientations on different substrates. (a) GaN on SiC(002), (b) SiC(002), and (c) Ru/Al₂O₃(002).

expected antiferromagnetic structure and known weak ferromagnetism. The small magnetization is a natural consequence of the inverse triangular spin structure [28]. The magnetization of the tetragonal Mn₃Ga thin film is shown in Fig. 5(b) for comparison. It displays a large hysteresis, with a remnant magnetization of 0.65 $\mu\text{B}/\text{Mn}$ as expected for ferrimagnetic tetragonal Mn₃Ga.

Hall measurements were performed on a (100)-oriented hexagonal Mn₃Ga thin film sample grown on MgO(110)/W(211). As shown in Fig. 6(a), a large anomalous Hall effect up to 0.02 $\mu\Omega\text{ cm}$ is observed despite the negligible magnetization. The sign of the anomalous Hall effect is consistent with the report by Ref. [8]. It is worth noting that the coercivity is only ~ 0.2 T, indicating high sample quality, which is comparable to the molecular-beam-epitaxial-grown Mn₃Sn thin films [29]. Figure 6(b) shows the magnitude of the anomalous Hall effect as a function of temperature. The magnitude of the anomalous part is largest at around 250 K and decreases with decreasing temperature. The thin W seed layer is expected to exhibit only an ordinary Hall effect on the order of $\sim 10^{-9} \Omega\text{ cm}/\text{T}$ [30] and therefore contributes negligibly to the measured Hall response. It does not influence the anomalous Hall behavior observed in the Mn₃Ga film.

To study the magnetic structure of the thin-film sample, neutron diffraction was performed on the WISH instrument at ISIS and the D10+ instrument at ILL. As the sample on GaN(002)/SiC showed the best crystal quality, it was chosen for the neutron experiment. Based on a previous neutron-powder study, the spin structure is expected to be commensurate with the propagation vector $k = (000)$; thus, magnetic reflections are superimposed on structurally allowed positions.

Using the WISH instrument, the sample was oriented to access the H0L family of reflections. In addition to the (00L) reflections, we were able to measure the (100), (101), (102), (103), and (300) reflections. The temperature-dependent integrated intensities of the (100) and (101) reflections are shown in Fig. 7. The (101) intensity remains nearly constant from 150 K to 400 K, but decreases significantly as the temperature approaches the magnetic ordering temperature, indicating that the Néel temperature is close to 450 K. A guide-to-the-eye curve using the functional form $I(T) = (T_N - T)^{2\beta}$ is included in the figure, with $\beta = 0.15$. In addition, there is no sign of any additional reflection attributable to Mn₃Ga, confirming the expected propagation vector $k = (000)$.

With the four-circle instrument, we could study 12 reflections at two temperatures, room temperature (RT) and 460 K

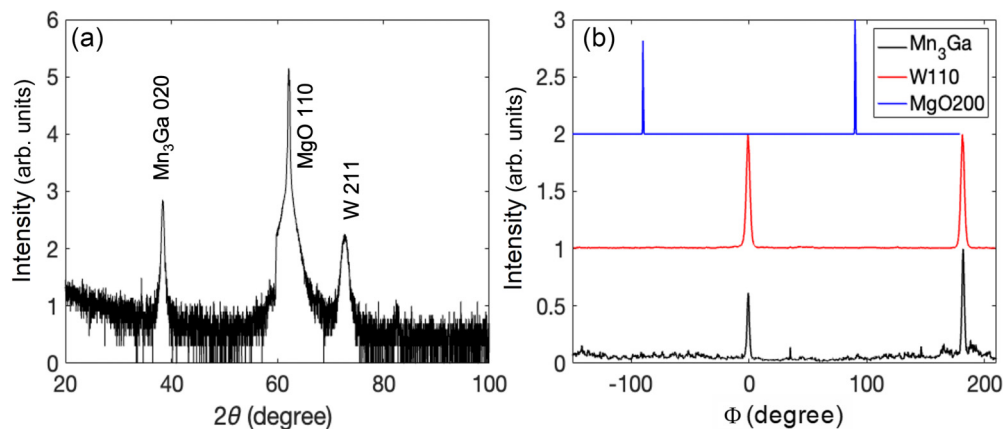


FIG. 4. (a) XRD θ - 2θ scan of hexagonal Mn₃Ga with (100) orientation on W/MgO. (b) Azimuthal scan of Mn₃Ga (201), W (110), and MgO (200) reflections, showing the epitaxial relationship.

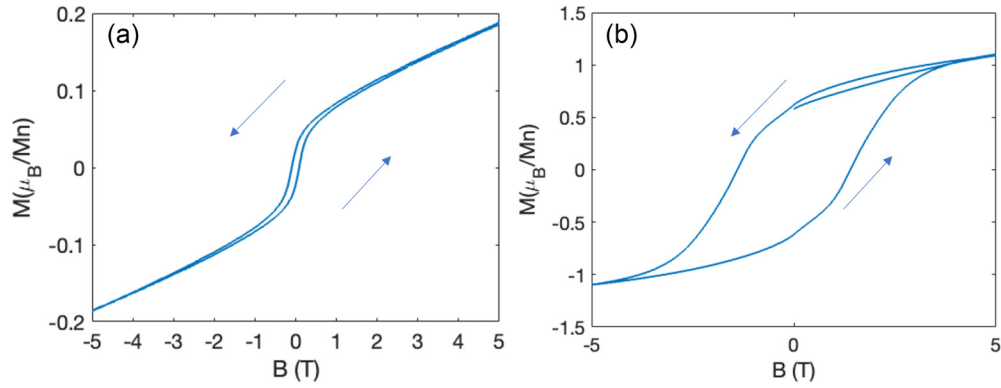


FIG. 5. Magnetization (M) as (a) function of field (B) for a 30-nm antiferromagnetic hexagonal Mn_3Ga thin film on 10 nm $\text{W/MgO}(110)$ with applied field in plane of the thin film, and (b) 30-nm ferrimagnetic tetragonal Mn_3Ga thin film on $\text{MgO}(001)$ with applied field in plane of the thin film. The diamagnetic contribution from substrate is subtracted. Both measurements are performed at 300 K. Arrows indicate scan direction.

(HT). Most of the reflections are sharp and can be well fitted with a Gaussian function. The (101) and (110) reflections are of particular interest for the magnetism of Mn_3Ga . Figures 8(a) and 8(b) show the (110) and (101) reflections at RT and HT, respectively. The intensity difference is caused by the greater contribution from magnetic scattering at RT.

First, the HT integrated intensities, which are assumed to be mainly nonmagnetic, were refined. Parameters include the atomic coordinate of $\text{Mn}[x = 0.18(1), 0.36, 0.25]$, a scale factor, isotropic extinction parameters, and the thermal factor $B[B_{\text{Mn}} = 1.3(3), B_{\text{Ga}} = 3.8(2)]$. The calculated and observed intensities are plotted in Fig. 8(c). The refinement gives a $\chi^2 = 1.6$. For the RT data, we assumed the same structure as at HT, and fixed all the structural parameters to the HT results. The magnetic symmetry considerations for Mn_3Ga are the same as those for Mn_3Ge and Mn_3Sn [11,12]. There are four symmetry-allowed magnetic structures. The observation of a weak in-plane ferromagnetic moment rules out the two structures with B_{1g} and B_{2g} symmetry. The other two structures have E_g symmetry and cannot be distinguished by unpolarized neutron diffraction. For the refinement of the RT data, we assumed the model IV, as found in Ref. [12] for Mn_3Ge as shown in the schematic in Fig. 9. In addition, the magnetic ground state allows six orientational domains; however, only three are distinct, since the neutron scattering cross section is

invariant under a 180° rotation of all spins. As the experiment was performed in zero field, we assumed equal population of each domain. The presence of magnetic contributions to both (110) and (101) reflections indicate that the spins in the a - b plane have a general orientation instead of being aligned along a certain axis. Thus, two parameters, the moment size and the orientation in the a - b plane, were refined. The model was refined with a $\chi^2 = 2.7$, and the results are plotted in Fig. 8(d). The refined magnetic moment is $1.5(1) \mu_B/\text{Mn}$, and the moment is aligned close to the $[210]$ direction.

Comparing our results with those obtained by neutron diffraction on a powder Mn_3Ga sample [13], the powder study gave a larger magnetic moment of $2.4 \mu_B/\text{Mn}$ at 300 K. The reason why our value for the ordered moment is smaller is that the HT data were collected slightly below T_N , so there remains a magnetic contribution to the HT peaks. As with the powder study, the magnetic contribution to the (002) peak is not detectable, so it is not possible to confirm the size of the weak ferromagnetic moment from neutron diffraction. We also note that the experiments presented here were performed based on the high-crystalline-quality Mn_3Ga on GaN/SiC . The present results highlight the feasibility of single-crystal neutron-diffraction technique for studying antiferromagnetic spintronic thin films, particularly within the Mn_3X family. For example, future experiments on the effect of epitaxial strain on

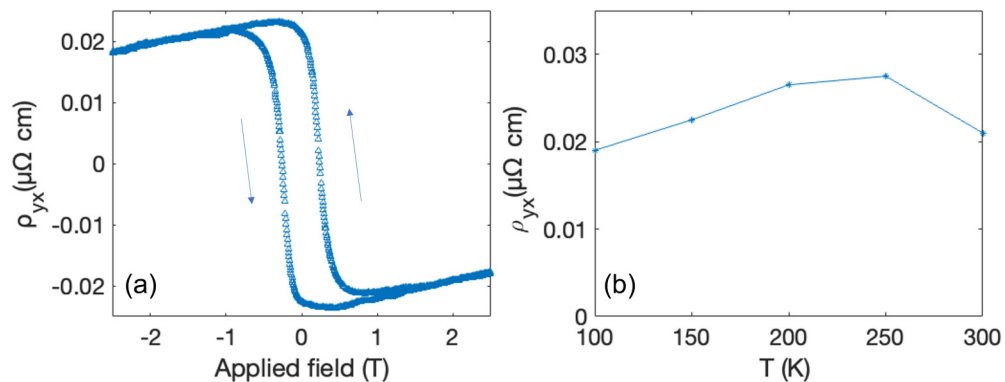


FIG. 6. (a) Anomalous Hall effect of 30-nm antiferromagnetic hexagonal Mn_3Ga thin film on 10-nm $\text{W/MgO}(110)$. (b) Temperature evolution of the anomalous Hall effect.

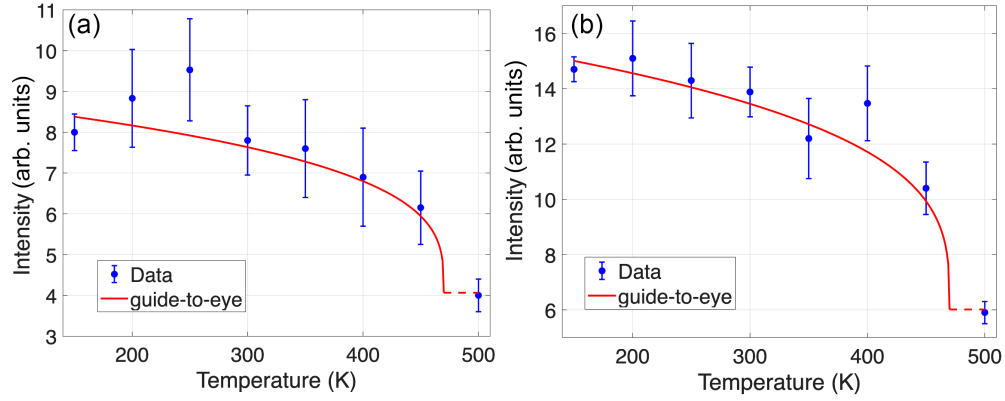


FIG. 7. Integrated peak intensity of neutron diffraction of (a) the (100) peak and (b) (101) peak of hexagonal Mn_3Ga on GaN at different temperature. The red curve is a guide-to-the-eye.

the spin structure are planned on hexagonal Mn_3Ga grown on W/MgO(110).

IV. CONCLUSIONS

In summary, we have investigated the growth and crystal structure of hexagonal and tetragonal Mn_3Ga thin films

synthesized by DC magnetron sputtering. High-quality C-plane hexagonal Mn_3Ga films were successfully grown on GaN/SiC, SiC, Ru/ Al_2O_3 , and MgO(111) substrates, with the GaN/SiC sample exhibiting the best crystalline quality. Furthermore, M-plane-oriented hexagonal Mn_3Ga films were realized using W(211)/MgO(110). By systematically varying the Mn:Ga ratio and postdeposition annealing conditions, we

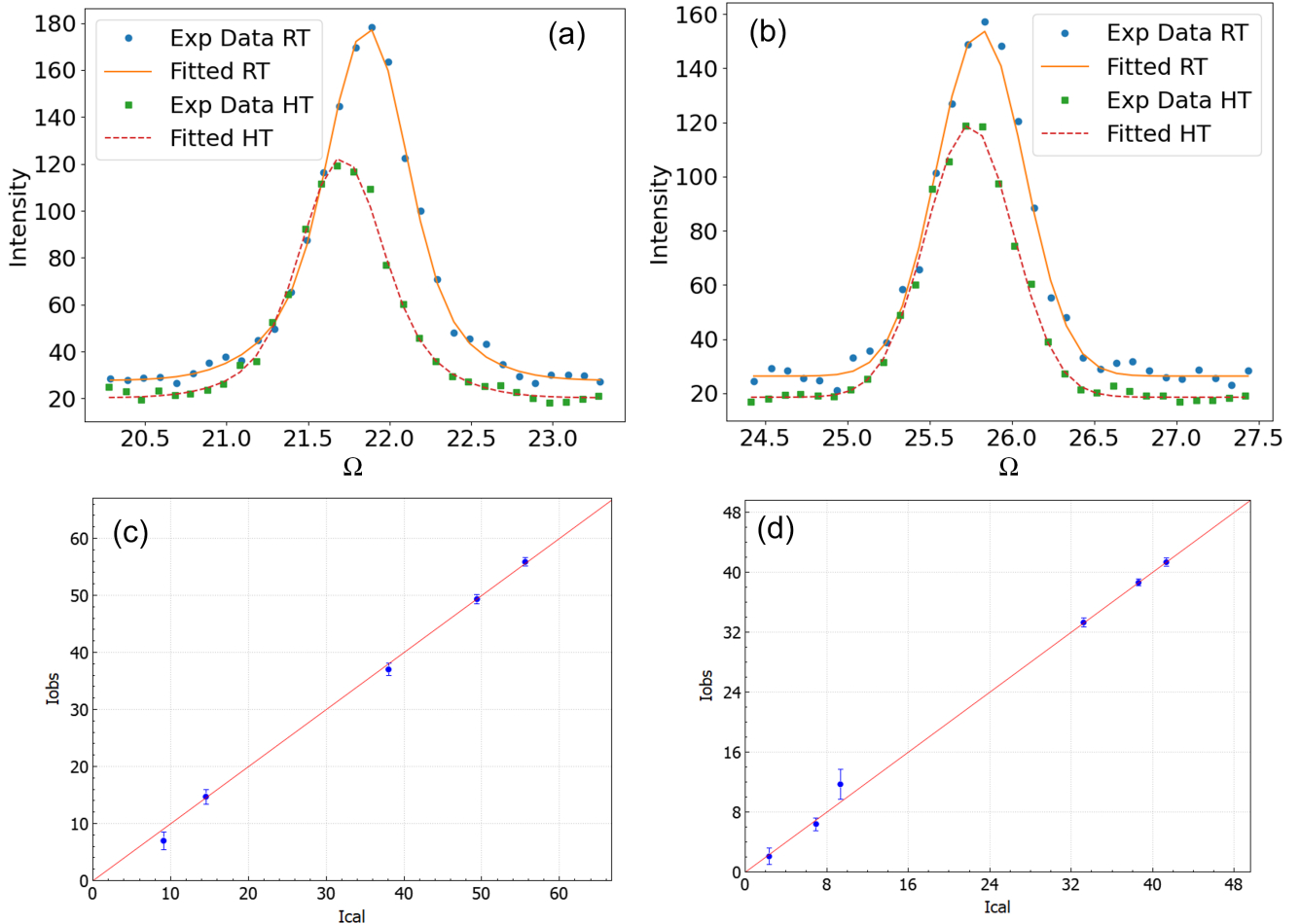


FIG. 8. (a) Neutron diffraction of the (110) peak of hexagonal Mn_3Ga on GaN at HT and RT. (b) Neutron diffraction of the (101) peak at HT and RT. (c) Refinement of the RT data. (d) Refinement of the HT data.

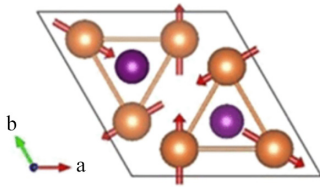


FIG. 9. Schematic of the magnetic structure of hexagonal Mn_3Ga .

established clear control over phase formation, enabling selective synthesis of either the metastable hexagonal or the thermodynamically stable tetragonal phase.

Magnetization measurements confirmed the antiferromagnetic nature of the hexagonal phase and the ferrimagnetism of the tetragonal counterpart. A pronounced anomalous Hall effect was observed in the (100)-oriented hexagonal films despite their negligible net magnetization, highlighting the role of Berry curvature in transport. Neutron diffraction measurements further confirmed the noncollinear magnetic structure of the hexagonal phase and yielded a refined moment of $1.5(1) \mu\text{B}/\text{Mn}$.

These results demonstrate the feasibility of engineering Mn_3Ga thin films with tailored structures and orientation, opening pathways to explore their topological transport and magnetoelectric properties for future antiferromagnetic spintronic applications.

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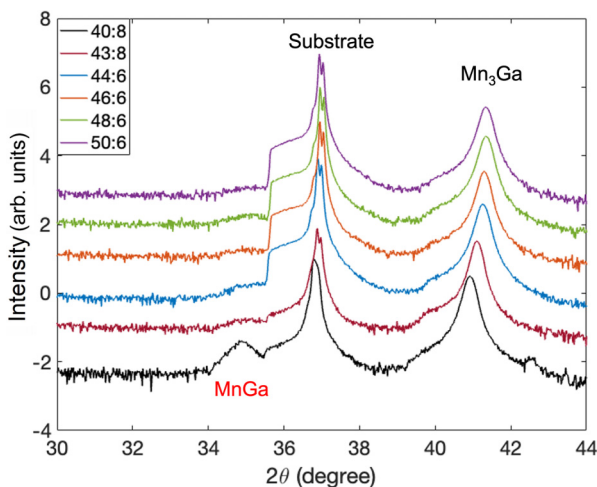


FIG. 10. Evolution of Mn_3Ga (002) peak as Mn:Ga power ratio increase from Mn : Ga = 40 : 8 to 50 : 6 W.

time on the WISH diffractometer at ISIS under proposal RB2310242 [31]. The authors thanks for the beam time on the D10+ instrument at ILL under proposal 5–54-394 [32].

DATA AVAILABILITY The data that support the findings of this article are openly available [31,32].

APPENDIX

Figure 10 shows the two theta-theta scan of thin films deposited at the same temperature but with different powers applied to the Mn and Ga targets. When the power is set to Mn 40W and Ga 8 W, the sample contains both Mn_3Ga and MnGa. As the Mn:Ga ratio increases, Mn_3Ga becomes the only phase, and the peak is shifted to higher angle. Further increasing the Mn power leads to a decrease in peak intensity.

It is worth mentioning that the quality of the W seed layer is important for the growth of Mn_3Ga . We optimized the epitaxial growth of W on MgO (110) substrate. Figure 11(a) in Appendix shows the XRD pattern of W thin film as a function of annealing temperature. Higher annealing temperatures lead to better crystalline quality of the W thin film. However, at temperatures above 750°C , a new peak appears, presumably attributable to the formation of MgWO_4 [27]. Figure 11(b) in Appendix shows the effect of Ar pressure on the W thin film. The (211) peak is shifted with Ar pressure. This effect might be useful for controlling the strain during the growth of Mn_3Ga . It could be of significance considering that the strain in $\text{Mn}_3\text{Ga}/\text{Mn}_3\text{Sn}$ is important for electrical switching. This topic will be explored further in future work.

Substrate selection and lattice matching. Based on the measured positions of the (002) and (201) reflections, we estimate the lattice parameters of Mn_3Ga grown on GaN to be $a = 5.43 \text{ \AA}$ and $c = 4.37 \text{ \AA}$. It is difficult to compare them with the reported bulk values, which often vary slightly owing to stoichiometric deviations and preparation methods. Using this estimate, we calculate the in-plane lattice mismatch to be approximately -0.4% with Ru, -1.7% with GaN, and -1.9% with SiC, assuming a 30° in-plane rotation. The small lattice mismatch motivated us to grow Mn_3Ga on these substrates. The epitaxial growth and small lattice mismatch are confirmed on the Ru seed layer. However, the 30° rotation on either GaN or SiC is observed. Instead, the Mn_3Ga lattice aligns directly with the underlying GaN or SiC without rotation. Despite the large lattice mismatch, the crystalline quality is very high for Mn_3Ga grown on GaN. This highlights the potential of GaN-based platforms for high-quality epitaxial antiferromagnetic films, particularly for experiments requiring large coherence volumes, such as neutron scattering or advanced spectroscopies.

The (100)-oriented Mn_3Ga has lattice constants of $a = 5.41 \text{ \AA}$ and $c = 4.26 \text{ \AA}$, calculated from the positions of the (020) and (201) reflection. Comparing these values with the relaxed lattice constants obtained from Mn_3Ga on GaN ($a = 5.43 \text{ \AA}$ and $c = 4.37 \text{ \AA}$), (100)-oriented Mn_3Ga has an out-of-plane compressive strain about -0.4% and an in-plane strain of about -2.4% .

Symmetry analysis. The crystal structure of hexagonal Mn_3Ga is the same as hexagonal Mn_3Ge . The symmetry analysis can be performed in the same way as for Mn_3Ge , which is detailed in the Soh *et al.* in Ref. [12]. Briefly, we

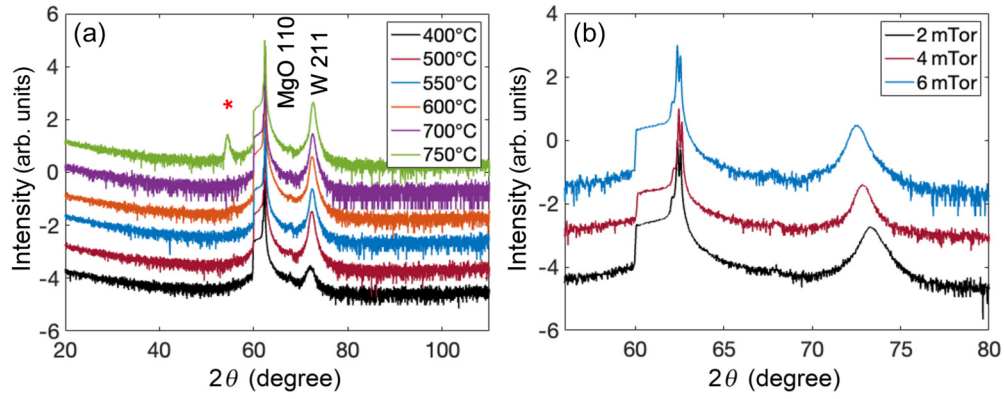


FIG. 11. (a) XRD θ - 2θ scans scans of W thin film after annealing at different temperatures. (b) XRD θ - 2θ scans of W thin films deposited at different Ar pressure.

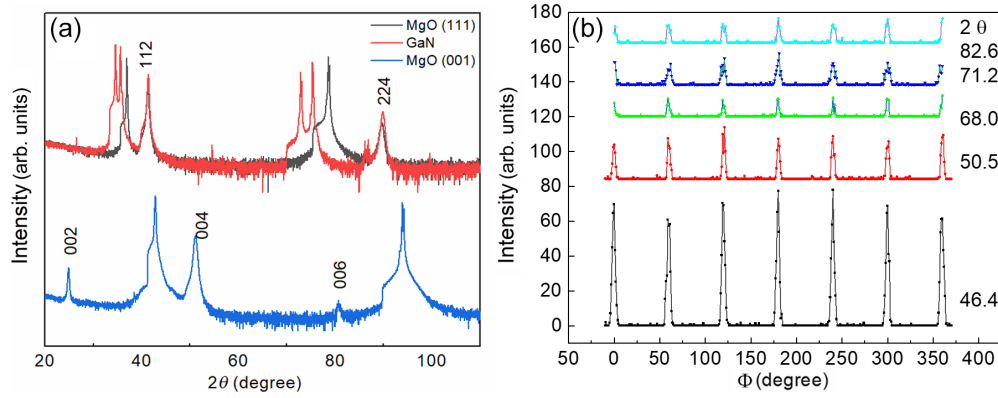


FIG. 12. (a) XRD θ - 2θ scans of tetragonal Mn_3Ga with (112) orientation on GaN(002)/SiC(002), MgO(111), and 002 orientation on MgO(001). The labeled peaks are Mn_3Ga peaks, and the remaining peaks are from the substrate. (b) azimuthal scans of tetragonal Mn_3Ga grown on GaN/SiC. The number on the right-hand side shows the 2θ value of each scan, corresponding to the tetragonal Mn_3Ga (200), (004), (220), (204), and (312) reflections, respectively.

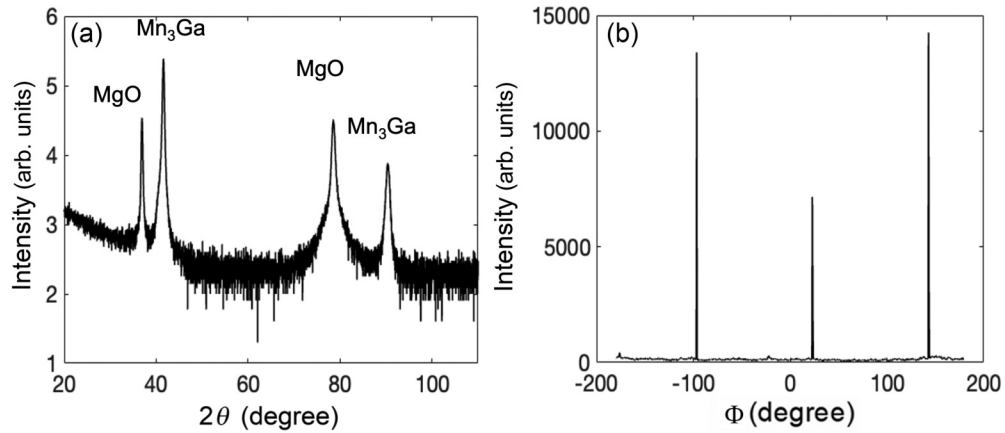


FIG. 13. (a) XRD θ - 2θ scans scans of cubic Mn_3Ga on MgO(111). (b) Azimuthal scan of cubic Mn_3Ga (200), measured at $2\theta = 48.2^\circ$, $\chi = 53^\circ$.

TABLE I. Character table for Mn_3Ga .

Irrep.	D_{6h}	E	$2C_{6z}$	$2C_{3z}$	C_{2z}	$3C_{2y}$	$3C_{2x}$	I	$2S_6$	$2S_3$	σ_h	$3\sigma_d$	$3\sigma_v$
Γ_3^+	B_{1g}	1	-1	1	-1	1	-1	1	-1	1	-1	1	-1
Γ_4^+	B_{2g}	1	-1	1	-1	-1	1	1	-1	1	-1	-1	1
Γ_5^+	E_{1g}	2	1	-1	-2	0	0	2	1	-1	-2	0	0

performed the symmetry analysis of hexagonal Mn_3Ga with the $P6_3/mmc$ space group for the $6h$ Wyckoff site with a magnetic propagation vector of $\mathbf{k} = (000)$. There are twelve irreducible representations (irreps) of the point group D_{6h} of $P6_3/mmc$. Because of the absence of the (111) reflection, the inversion center is preserved; thus, those irreps that change sign under inversion (I) are excluded. In addition, those irreps with a ferromagnetic moment along the c axis can also be excluded based on the magnetization data. Thus, the irreps are reduced to $\Gamma = \Gamma_3^+ + \Gamma_4^+ + 2\Gamma_5^+$, with B_{1g} , B_{2g} , and E_{1g} symmetry, respectively, as shown in Table I.

Tetragonal and cubic Mn_3Ga . When studying hexagonal Mn_3Ga , we also noticed that tetragonal Mn_3Ga can be synthesized under very similar conditions. With the same composition as for hexagonal Mn_3Ga , but annealing at a lower temperature, namely, 400 °C, tetragonal Mn_3Ga can be synthesized. Figure 12 shows the XRD θ - 2θ scans of tetragonal Mn_3Ga grown on different substrates. The upper panel in Fig. 12(a) shows the (112)-oriented Mn_3Ga grown on $\text{MgO}(111)$ and $\text{GaN}/\text{SiC}(002)$. The lower panel in Fig. 12(a) shows the (002)-oriented Mn_3Ga grown on $\text{MgO}(001)$. We note that (112)-oriented tetragonal Mn_3Ga can be wrongly identified as (002)-oriented hexagonal Mn_3Ga , as they appear to be identical in θ - 2θ scans. However, azimuthal scans (φ scans) can readily distinguish between the two phases. Here, χ is the tilt angle between the sample surface and the diffraction plane. Each crystallographic plane has a unique 2θ and χ value. For example, for the tetragonal phase, the angle

between the (112) and (200) planes is 56° and the calculated 2θ for the (200) reflection is 46.5° . To locate the (200) peak, we performed φ scan after setting $2\theta = 46.5^\circ$ and rotating the sample stage to $\chi = 56^\circ$. Once the peak was detected, the 2θ , χ , and ω were optimized to find the exact peak position, which was found at $\chi = 55^\circ$ and $2\theta = 46.4^\circ$. This procedure was repeated for a series of reflections characteristic of both the tetragonal and hexagonal phases. Figure 12(b) shows the φ scan of a list of peaks of the (112)-oriented tetragonal Mn_3Ga on $\text{GaN}/\text{SiC}(002)$. The observed peaks are (200), (004), (220), (204), and (312) from bottom to top, respectively. The patterns are shifted vertically for clarity. They are observed at their expected 2θ and χ values, consistent with the tetragonal phase. No peaks associated with the hexagonal phase were detected.

Figure 13 shows the two theta-theta scan of cubic Mn_3Ga on $\text{MgO}(111)$. The film was deposited with a single Mn_3Ga compound target with an $\text{Mn} : \text{Ga} = 3 : 1$ at a power of 55W. However, the $\text{Mn}:\text{Ga}$ ratio deposited on the substrate is slightly higher than the ratio in the target, which is quite common for compound targets. The $\text{Mn}:\text{Ga}$ ratio is estimated to be $\text{Mn}_{3.2(2)}\text{Ga}_1$, from EDX in the SEM. The peaks are indexed as the (111) and (222) reflections of cubic Mn_3Ga , which can be wrongly indexed as either tetragonal Mn_3Ga or hexagonal Mn_3Ga . As we explained in detail for the case of tetragonal Mn_3Ga or hexagonal Mn_3Ga , an azimuthal scan can differentiate these phases. Figure 13(b) shows the φ scan of the (200) peak of the cubic phase, confirming the cubic structure.

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