

Utrip Report

Fabrication and characterization of Mn₄N epitaxial thin films
of different thickness by pulsed laser deposition

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Abstract

Materials with perpendicular magnetic anisotropy (PMA) are being investigated for magnetic random access memory (MRAM) and other spintronics applications. We report electric properties of perpendicular magnetized Mn₄N epitaxial films on the MgO substrate with various thicknesses. The X-ray diffraction results revealed the other kind of crystal whose peaks near MgO(200). And the result of the abnormal hall-effect fit the theory very well.

Introduction

Spintronics is gaining popularity because of the interesting physical phenomena that can be observed in these systems as well as due to the potential applications that can be derived from it. Thin films with perpendicular magnetic anisotropy (PMA) are one type materials featured with easy magnetization axes perpendicular to the film surface. Recently, research interest in application of PMA films in the field of spintronics has increased remarkably. Spin valves and magnetic tunneling junctions with perpendicular anisotropy have been demonstrated to allow the realization of low-dimensional and highly reliable spintronic devices, e.g., spin-transfer switching magneto-resistivity random access memory.

For applications in spintronic devices, the materials with large spin polarization and low saturation magnetization are strongly desired. And Mn₄N is a promising material for spintronics devices to possess such properties. In this structure (Fig.1), the ferrimagnetism of Mn₄N originates from its antiperovskite structure. And the presence of inequivalent Mn sites leads to a complex triangular spin configuration. The direction of their magnetic moments is aligned anti-parallel. And the magnetic moments of Mn atoms at sites I and II were around 3.85 μ B/u.c and 0.90 μ B/u.c., respectively. Consequently, a low M_s can be achieved at room temperature while the Neel temperature is as high as 738K. In order to comprehend the intrinsic properties of its material better and introduce it into application in spintronics, epitaxial Mn₄N crystal films are preferable.

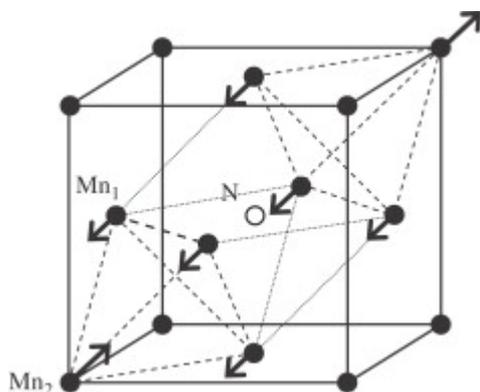


Fig. 1 The crystal structure of Mn₄N

Experiment

In this work, the Mn₄N films with different thickness were grown on MgO(100) single crystal substrate by pulsed laser deposition(PLD) with a nitrogen partial

pressure of 1.0×10^{-6} Pa and the pulsed frequency of 10 Hz. And the temperature of substrate during the deposition is 500 K. After deposition, the films were covered with Al_2O_3 layer of 3 nm thick to prevent oxidization. The deposition time of Mn₄N was controlled at 10 min, 20 min, 40 min and 80 min. The crystal structures and epitaxial relationships of the films were characterized by x-ray diffraction (XRD). The AHE resistivity was measured from 10 to 300 K without magnetic field.

Discussion

The XRD $\theta - 2\theta$ scan for the Mn₄N films is shown in Fig. 2. Apart from the peaks from MgO substrates, Only the Mn₄N(100), and Mn₄N(200) peaks can be observed. And the crystallinity of thicker Mn₄N films is better because the full width at half maximum of Mn₄N peak is short. The calculated MnO crystal parameter is 4.433 Å. If $n=2$, the calculated distance is 4.42 Å which is almost the same as MnO crystal parameter.

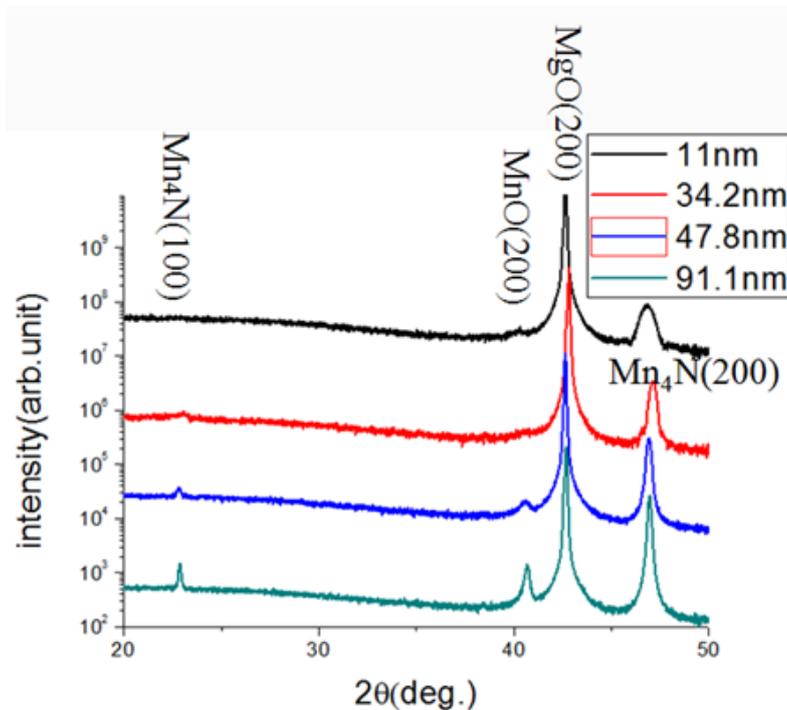


Fig. 2 XRD result of 1D detector under different thickness

And the XRD image by 2D detector is shown as below(Fig.3).

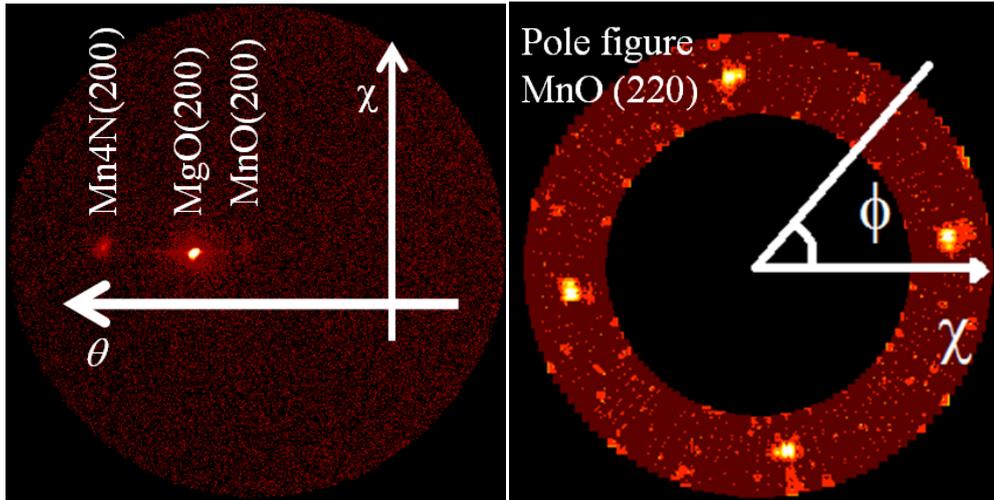


Fig. 3 XRD result of 2D detector(left) and phi dependence XRD result of the unknown peak(right)

The dot-like image by XRD 2D detector and the obvious peak showed by Fig. 2 shows that the high quality single crystal $Mn_4N(100)$ is obtained. And the small peak near the peak of $MgO(200)$ means that there exists impurity on the substrate except $Mn_4N(100)$. After measurement, it is supposed that the peak belongs to MnO . Fig. 3 (right) is the image which shows the ϕ dependence XRD result of the small unknown peak. It provides us the information that the impurity should be cubic cell which is totally fit our supposition. It is very possible that MnO is also epitaxially grown on MgO substrate.

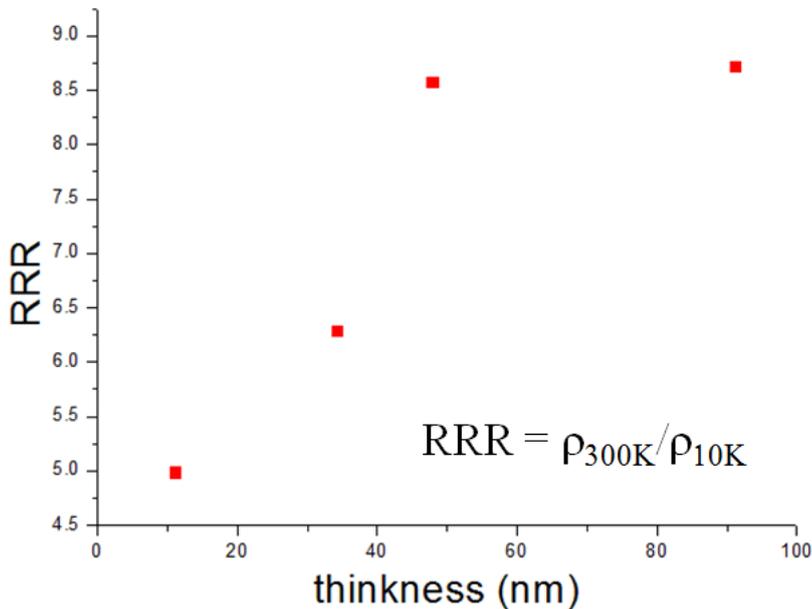


Fig. 4 the RRR (residual resistivity ratio) of films with different thickness

Fig. 4 shows the residual resistivity ratio of different thickness films. When the thickness is low, the residual resistivity is low which means the worse crystallinity. We can conclude that the resistivity of the thin film is decreasing because of better crystallinity.

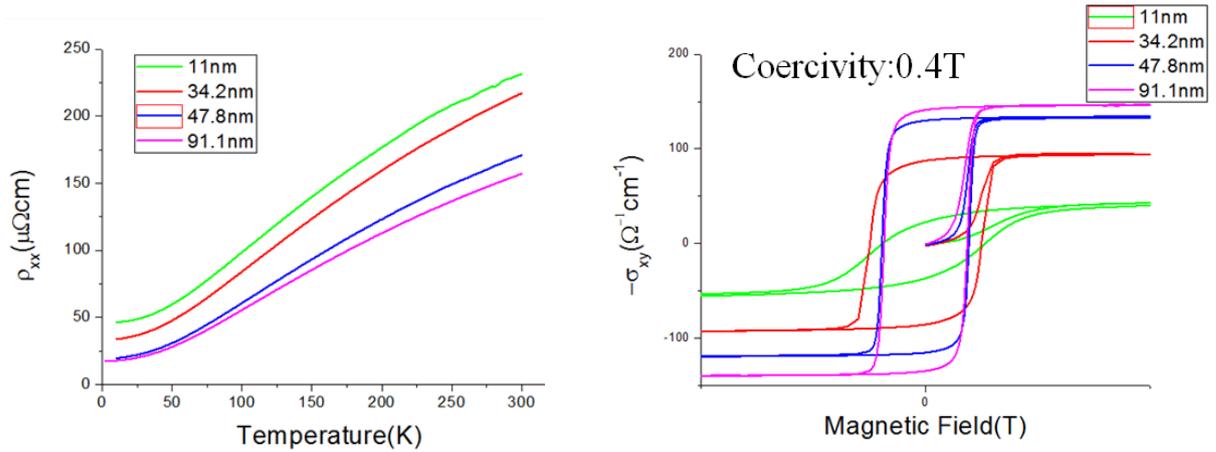


Fig. 5 ρ -T curves (left) and the abnormal hall conductivity-magnetic field curves for Mn_4N films with different thickness (right).

Also, the temperature property is another important factor to characterize the thin film, too. The temperature dependence resistivity of different thickness films is shown in Fig. 5. The resistivity has a positive relationship with the temperature while a negative relationship with the thickness. As the temperature increases, the structure of the film will become disorder which can cause the increase of scattering. In this way, the resistivity becomes higher. Fig. 5 shows the result of the longitudinal resistivity and anomalous Hall conductivity of samples with different thickness. It is clear to see that thicker film has lower longitudinal resistivity and high anomalous Hall conductivity. As previous conclusion, the quality of crystal becomes high when the film is thicker. In this way, the direction of magnetic moment of each cell becomes more consistent, which caused the increase of saturation magnetization. In the way the anomalous Hall effect is more obvious to see.

Here is the discussion about the better crystallinity. MnO has Anti-ferromagnetic property so the impurity weakens anomalous Hall effect of the thin film. MnO may generate from oxygen diffusion at interface of $\text{MgO-Mn}_4\text{N}$. Thicker the film, lower influence from interfacial MnO impurity (Fig. 6).

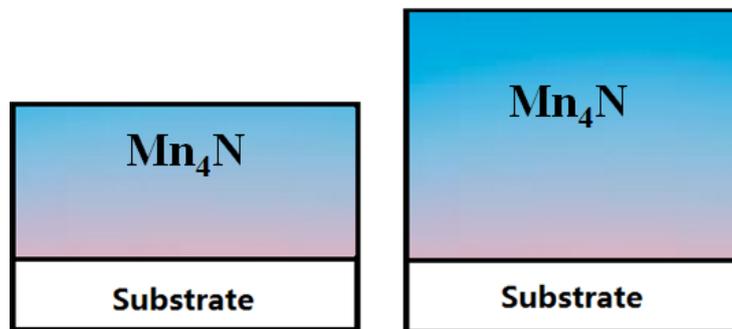


Fig. 6 scheme of oxygen diffusion at interface

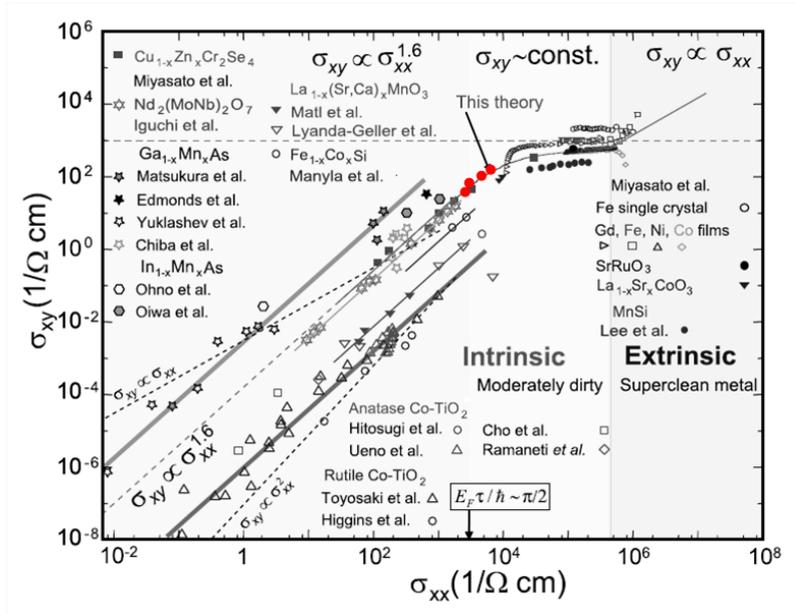


Fig. 7 the theory of AHE

In the theory of AHE (Fig. 6), it is said that there are three regions according to different value of σ_{xx} , which is shown in Fig. If $\sigma_{xx} > 1 \times 10^6 \Omega^{-1} \text{ cm}^{-1}$, it belongs to the extrinsic regime in the super clean metal and σ_{AHE} is proportional to σ_{xx} . If σ_{xx} is between $10^4 \Omega^{-1} \text{ cm}^{-1}$ to $10^6 \Omega^{-1} \text{ cm}^{-1}$, it belongs to intrinsic regime in the moderately dirty metal and σ_{AHE} is almost constant. If $\sigma_{xx} < 10^4 \Omega^{-1} \text{ cm}^{-1}$, it belongs to dephased intrinsic regime in the dirty metal and σ_{AHE} is proportional to $\sigma_{xx}^{1.6}$. The red points in the Fig. 7 are the results of experiment. It is almost consistent with the middle part which is moderately dirty metal. The conclusion can also provide the evidence of the existence the defects in the films (maybe it is caused by MnO impurity).

Conclusion

Mn₄N epitaxial single crystal films were successfully fabricated on MgO substrate. There exists MnO impurity which may generate from the interface of Mn₄N-MgO and it is epitaxially grown on MgO substrate. Mn₄N films show thickness dependence on crystallinity, resistivity and anomalous Hall effect. The origin of thickness dependence may come from interfacial impurity MnO. Furthermore, the result of anomalous Hall conductivity is consistent with the AHE theoretical study very much.

Reference

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