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Nurut Agusutrisno, M., Marrows, C.H., Kamataki, K. et al. (2023) On-axis Sputtering Fabrication of $Tm_3Fe_5O_{12}$ Film with Perpendicular Magnetic Anisotropy. *Thin Solid Films*, 788. 140176. ISSN: 0040-6090

<https://doi.org/10.1016/j.tsf.2023.140176>

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1 **On-axis Sputtering Fabrication of $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ Film with Perpendicular**
2 **Magnetic Anisotropy**

3
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17
18 *Keywords:* spintronics, sputtering, Rare-earth iron garnet, Perpendicular magnetic
19 anisotropy

20 (Dated: 18 October 2023)

21

1 **ABSTRACT**

2

3 Thulium iron garnet, $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ with perpendicular magnetic anisotropy is
4 fabricated using an on-axis sputtering technique followed by annealing, whereas
5 previous reports have used unusual off-axis geometries. Stoichiometric
6 $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ is obtained after the modification of the deposition conditions
7 involving the position of the substrate relative to the cathode, which affects both
8 the chemical and structural properties. The effective perpendicular magnetic
9 anisotropy of 8.6 kJ/m^3 is well in line with the results of previous studies using
10 pulse laser deposition and off-axis sputtering. A maze domain pattern is
11 observed, and the domain-wall energy is evaluated as 0.69 mJ/m^2 .

12

1 **1. Introduction**

2 Rare-earth iron garnets have attracted much attention as a materials platform in spintronics
3 because of the rareness of ferrimagnetic insulators, which allow a long-range propagation of spin
4 waves [1–3]. Through the manipulation of the magnetic moment with spin-orbit torque [3], these
5 magnetic insulators are considered as candidates for energy-efficient magnetic memory and logic
6 devices [4]. Furthermore, the low magnetic damping constant of rare-earth iron garnet has the
7 potential to realize the domain-wall qubit [5,6].

8 For application to integrated systems, perpendicular magnetic anisotropy (PMA) is
9 beneficial, in addition to the weak magnetic moment stemming from the compensated
10 ferrimagnetism [7–11]. Kubota *et. al.* demonstrated a PMA thin film using very thin (46 nm in
11 thickness) thulium iron garnet, $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ (TmIG), on a $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG) (111) substrate by
12 taking advantage of the large negative magnetostriction constant of $\lambda_{111} = -5.2 \times 10^{-6}$ and the
13 epitaxial strain of +0.49% [7] followed by the demonstrations of the topological Hall effect [10],
14 spin-orbit torque switching [8], and the creation and transport of skyrmions [11]. Rosenberg *et al*
15 systematically studied the effect of Y-substitution on the magnetic anisotropy [12]. Wu *et. al.*
16 demonstrated the fabrication of TmIG using off-axis sputtering, and showed the saturation
17 magnetization of 99 kA/m, which is close to the bulk value [13]. They also disclosed that the
18 magnetic anisotropy largely depends on the longitudinal distance between the sputtering target
19 and the substrate because of the strain dependence on the chemical composition of the film [14].

20 The production of TmIG film has been limited to pulsed laser deposition (PLD) [8,12,15]
21 and off-axis sputtering methods [13,14]. Because PLD limits the size of the available wafers,
22 sputtering is more useful for manufacturing. Although off-axis sputtering provides an opportunity
23 to fabricate TmIG by reducing ion bombardment [14,15], the effect of the plasma damage is
24 poorly understood. An important challenge in advancing research in this field is exploring a

1 simple method to fabricate TmIG films: *on-axis* sputtering, which is usually applied to fabricate
2 oxide thin films on a large scale [16]. Here, we demonstrate the fabrication of TmIG with PMA
3 through on-axis sputtering followed by thermal annealing.

4 5 **2. Experimental details**

6 *2.1 Thin film fabrication*

7 TmIG thin films were grown on GGG (111) substrates through radiofrequency (RF)
8 magnetron sputtering with on-axis geometry. Different substrate positions in the holder and
9 different deposition times were examined. The layout of substrate holder and the sputtering target
10 is shown in Fig. 1a. The distance L between the surface normal of the target and the substrate
11 holder was 55.5 mm, while the angle formed between L and the surface normal of the substrate
12 holder was 20° . A commercially available sputtering target of TmIG, with a diameter of 50.8 mm
13 and a thickness 3 mm (purity of 99.9%, KOJUNDO CHEMICAL LABORATORY), was placed
14 on a magnetron cathode. Three different positions on a sample holder are labelled A, B, and C. A
15 top-view schematic of the sample holder is shown in Fig. 1b. The blue cross indicates the point
16 at which the line extending the center line intersects the substrate holder. The pink squares
17 represent GGG substrates ($10\text{ mm} \times 10\text{ mm}$) on the sample holder. $L_A = 64\text{ mm}$, $L_B = 93\text{ mm}$, and
18 $L_C = 98\text{ mm}$ denote center-to-center distances between the target and each substrate. $\phi_A = 15^\circ$, ϕ_B
19 $= 36^\circ$, and $\phi_C = 39^\circ$ were the angles formed with respect to the L . The flow rates of oxygen and
20 argon were set as 5 and 18 standard cm^3/min , respectively. The total pressure was kept at 3 Pa
21 during deposition which was performed by supplying RF power of 60 W to the cathode. After the
22 sputtering deposition, post-annealing at 800°C for 3 hours was carried out in an oxygen
23 atmosphere at ambient pressure. Deposition was conducted for 30, 90, 120, and 150 minutes (6,
24 18, 24, and 30 nm in thick) to confirm that the magnitude of the X-ray diffraction (XRD) peak of

1 TmIG increased with the thickness.

2

3 *2.2 Thin film characterization*

4 The structural analysis of TmIG was carried out adopting high-resolution XRD $2\theta-\omega$ scan
5 with Cu $k\alpha$ ($\lambda = 1.5406 \text{ \AA}$) and X-ray reflectivity (XRR) measurement techniques using a Bruker-
6 D8 Discover. The composition of TmIG was measured adopting the X-ray fluorescence (XRF)
7 (Rigaku Primus II), by utilizing the fundamental parameters calculation method available on the
8 XRF equipment. The magnetic moment was examined using a superconducting quantum
9 interference device (SQUID) (Quantum Design, MPMS3) with DC scan at 300K. The
10 ferromagnetic resonance (FMR) was measured using an electron spin resonance system (Bruker
11 EMX). A piece of substrate was cut into $4 \text{ mm} \times 2 \text{ mm}$ in size and pasted on a quartz stick. The
12 frequency of applied microwave was 9.6 GHz, and the power was 2.1 mW. Angle dependence
13 measurements were performed by changing the angle between the external magnetic field and the
14 substrate normal with a DC magnetic field scan at each angle. A Kerr microscope (Evico
15 microscope) was used to visualize the structure of the magnetic domains. A maze domain structure
16 at zero field was examined by an ac demagnetizing procedure [17]. The surface roughness was
17 examined using an atomic force microscope (AFM) (AFM5100N Hitachi).

18

19 **3. Results and discussion**

20 *3.1 Crystal structures*

21 XRD measurements were carried out to confirm the formation of the crystal of TmIG.
22 Figure 2a shows the results of XRD $2\theta-\omega$ scans of the samples deposited at position A for different
23 times (30, 90, 150 min) followed by annealing. All the samples showed a peak at 51.55° stemming
24 from TmIG (444) in addition to the peak of substrates at 51.06° . The peak same as that in a

1 previous study [7] implies that the TmIG film had the same lattice strain as films in the previous
2 study, which is expected to provide PMA. Thus, the feasibility of on-axis sputtering to fabricate
3 TmIG was confirmed by these XRD patterns. Thick films with $t = 13, 24,$ and 30 nm showed that
4 the peak from TmIG(444). The intensity of the TmIG peak increased with the deposition time at
5 on-axis position A.

6 To check the dependence of the attainability of TmIG on the position, three samples were
7 grown under the same condition with a film thickness of 24 nm and examined as shown in Fig.
8 2b. Interestingly a clear peak of TmIG(444) only appeared for a sample deposited at position A.
9 In addition, the thickness of the TmIG film t depended on the position, which was measured by
10 XRR. The thickness was $24, 13,$ and 7 nm at positions A, B, and C, respectively. As the thickness
11 t of the sample deposited at position B was greater than that of a thin sample ($t = 6$ nm) deposited
12 at position A, the attainability of TmIG did not depend only on the volume of the film. Rather, it
13 depended also on the chemistry, which is supported by the atomic density of the film evaluated
14 by XRF measurement.

15 The results of XRF are shown in Fig. 3. The densities of Fe and Tm were evaluated from
16 the XRF spectra using an analysis software equipped with our XRF system (ZSX Primus II,
17 Rigaku) based on Fundamental Parameter (FP) method [18]. Clear peaks are observed from these
18 samples. The ratio of $[\text{Tm}]/[\text{Fe}]$ was 0.6 (stoichiometric) for the sample fabricated at position A
19 and 0.7 for the other samples (shown in Fig. 2b).

20 A transportation of ion in material (TRIM) simulation was carried out to clarify the difference
21 in the spreading angles between the sputtered Tm and Fe. The composition versus substrate position
22 is shown with the result of the TRIM simulation in Fig. 4. The kinetic energy of Ar was assumed to be
23 100 eV or 150 eV, but no significant difference was observed in between these conditions. Sputtered
24 atoms from the target were measured 55 million times. The spreading angles ϕ are counted for each

1 sputtered atom. Histograms of ϕ of Fe and Tm were made to evaluate the Tm/Fe ratio. The positive
2 trend between ϕ and [Tm]/[Fe] is observed. The density ratio between Tm and Fe is close to 0.6
3 when the angle ϕ less than 18° . However, in the range of ϕ higher than 20° , the density ratio
4 exceeds 0.7. Hence, it could conceivably be hypothesized that the different sputtering angles between
5 Tm and Fe cause the different chemical compositions of the film in the different positions. A more
6 quantitative understanding of the growth condition dependence of chemical composition will be
7 provided by future research.

8 The surface morphology of the film displayed in Fig. 5 reveals that the RMS roughness is
9 sufficiently low at 0.4 nm and close to the results reported by other groups[13].

11 3.2 Perpendicular magnetic anisotropy

12 Figure 6a displays the hysteresis loop of the TmIG film at position A, which has a thickness
13 of 24 nm. When an applied field is directed in the in-plane direction, the film exhibits a larger
14 saturation field compared to the out-of-plane direction. The observed pattern indicates that the
15 easy axis of the film aligns with the out-of-plane direction, suggesting that the film exhibited
16 PMA properties. The thickness dependence of our film grown at position A is shown in Fig. 6b.
17 The saturation magnetization M_s of the sample thickness of 18, 24, 30 nm are 98, 108, and 55
18 kA/m, respectively. The decrease of M_s at $t = 30$ nm is also reported in previous research by Duong
19 *et al* [19], which is attributable to the stress relaxation.

20 The magnetometry of the samples of different composition is shown in Fig. 6c. All the
21 samples show the hysteresis in M - H curves under out-of-plane magnetic field. The magnitude of
22 M_s in growth position of B and C are 23 kA/m and 30 kA/m, which are significantly smaller than
23 that of position A. This decrease in M_s in Tm-rich films is consistent with those of Wu's [14].
24 They explained the decrease was the result of the Fe vacancies on the tetrahedral sites. Because

1 of this, in our slightly off-stoichiometric TmIG films the Fe vacancies on the tetrahedral sites
2 reduces magnetic moments.

3 The effective anisotropy energy K_{eff} was evaluated. FMR-measurements were carried out
4 using the TmIG sample ($t = 24$ nm) deposited at on-axis position A. An AC field h_{rf} was applied
5 to the sample under a static external field H from different angles relative to the film plane θ_H as
6 shown in Fig. 7a. Figure 7b shows a representative FMR spectrum at $\theta_H = 0^\circ$. Clear FMR signals
7 were observed. The θ_H dependence of the resonance field, H was obtained using the peak search
8 function of commercially available software after integrating the raw data. The angulare
9 dependence is shown in Fig. 7c. The black symbols represent the resonance field $\mu_0 H_{\text{FMR}}$. The
10 uniaxial magnetic anisotropy field h_u was evaluated using following equation [20]:

$$11 \quad \left(\frac{\omega}{\gamma}\right)^2 = [\mu_0 H_{\text{FMR}} \cos(\theta_H - \theta) - h_u \cos(2\theta)] \times [\mu_0 H_{\text{FMR}} \sin(\theta_H - \theta) + h_u \sin^2 2\theta] = 0 \quad (1),$$

$$12 \quad \mu_0 H_{\text{FMR}} \sin(\theta_H - \theta) + \frac{h_u}{2} \sin 2\theta = 0 \quad (2),$$

13 where $\omega = 2\pi f$ and $\mu_0 H_{\text{FMR}}$ are the angular frequency of the applied microwave and the resonance
14 field obtained from the FMR signal, respectively. θ_H and θ are the angles of the applied magnetic
15 field and saturation magnetization with respect to the film plane, respectively. The lowest value
16 detected at 100° supports that the sample possesses PMA. Equations (1) and (2) were solved self-
17 consistently by substituting in the measured resonance field, and h_u of the TmIG sample was
18 evaluated as -0.16 T. The corresponding $K_{\text{eff}} = h_u M_S / 2$ was then estimated to be 8.6 kJ/m³ by
19 considering $M_S = 108$ kA/m measured by the SQUID in an out-of-plane field. These values are
20 consistent with those reported by other groups using PLD and off-axis sputtering techniques
21 [13,14,19]. The results show that the oxygen ion bombardment of a substrate does not appreciably
22 diminish the magnetic anisotropy or magnetic moment in a magnetic insulator, although in the
23 case of semiconducting devices, it reduces the conductivity of the film [21].

1

2 3.3 Analysis and characterization of magnetic domain wall energy

3 The domain morphology was evaluated by examining samples of various thickness with a
4 polar magneto-optical Kerr effect microscope. Initially, an out-of-plane magnetic field was
5 applied. After AC demagnetization, the microscope image was captured as shown in Fig. 8. A
6 maze domain pattern, which also supports the appearance of PMA, was observed. In addition, we
7 confirmed the maze domain patterns for samples deposited with different thicknesses at position
8 A. All of these results support our main claim of the demonstration of the *on-axis* sputtering
9 fabrication of TmIG with PMA.

10 Two-dimensional fast Fourier transforms were processed, and the radial averaging of the
11 intensity was computed. The relation between the normalized intensity and radius is shown in Fig.
12 8d. The stripe spacing D_s , which was evaluated by fitting a Gaussian function, were 28, 11, and
13 8 μm for thickness 18, 24, 30 nm, respectively.

14 On the basis of these results, we applied the Kaplan-Gehring model and estimated the energy
15 density of the domain wall σ_{DW} , which is expressed as [12]:

$$16 \quad \sigma_{\text{DW}} = \frac{2K_d t}{\pi} \ln \left[\frac{D_s}{t \exp\left(\frac{\pi b}{z} + 1\right)} \right] \quad (3),$$

17 where $b = -0.666$ is a model-dependent constant, t is the film thickness, and $K_d = \mu_0 M_s^2/2$ is the
18 dipolar energy constant. The energy density of domain walls was 0.51 mJ/m² for 18 nm, 0.69
19 mJ/m² for 24 nm, and 0.2 mJ/m² for 30 nm. The smaller M_s resulted in a smaller domain wall
20 energy density (Fig. 8), leading to the smaller domain spacing. These values are on the same order
21 as those for other TmIG films manufactured by PLD or off-axis sputtering [12-14]. The exchange
22 stiffness of the 24 nm thick was evaluated as $A_{\text{ex}} = 3.4$ pJ/m using the relation $A_{\text{ex}} = \sigma_{\text{DW}}^2/16K_{\text{eff}}$.
23 The origin of the difference of A_{ex} might be attributable to the non-uniformity of t of the sample,
24 which had a root-mean-square roughness of 0.4 nm.

1

2 **4. Conclusions**

3 In summary, we demonstrated the on-axis sputtering deposition of PMA-TmIG.
4 Stoichiometric TmIG was obtained from the sample deposited on a suitable position of the
5 substrate relative to the target. The perpendicular magnetic anisotropy was confirmed by the
6 hysteresis loop patterns, the angle dependence of FMR, a polar magneto-optical Kerr effect
7 microscope. The effective anisotropy was 8.6 kJ/m^3 , which is in line with the results of previous
8 studies using PLD and off-axis sputtering [12-14]. Our results show the inertness of magnetic
9 anisotropy and the magnetic moment in the ferrimagnetic insulator to the ion bombardment during
10 deposition. This study showed the possibility of the industrial-friendly production of integrated
11 spintronic devices based on TmIG with PMA.

12

13 ACKNOWLEDGMENTS

14 The authors acknowledge support from JSPS KAKENHI in the form of an Early-Career Scientists
15 grant (No. 22K14292), and Grant-in-Aid for Scientific Research (A) (No. 20H00142), the Iketani
16 Science and Technology Foundation, the Yazaki Memorial Foundation for Science and
17 Technology, and Japan International Cooperation Agency. The authors are grateful to Mr. Kizuku
18 Ikeda for discussions on the analysis of the TRIM simulation. The authors thank Edanz
19 (<https://jp.edanz.com/ac>) for editing a draft of this manuscript.

20

21

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- 24

1 **FIGURE CAPTIONS**

2 Figure 1: The layout of the substrate holder and target

3 (a) Schematic side view of the sputtering chamber.

4 (b) Schematic top view of the substrate holder. The blue cross is the intersection mark between
5 the substrate holder and the normal target. The positions of the substrates are indicated by the
6 pink squares.

7 Figure 2: XRD patterns

8 (a) XRD patterns of TmIG films deposited at position A with different of deposition times (150,
9 90, 30 min) and a bare substrate from top to bottom. The thickness of each film was 30 nm, 18
10 nm, and 6 nm, respectively.

11 (b) XRD patterns of TmIG films at positions A, B, C, and substrate from top to bottom.

12 Figure 3: XRF spectra of TmIG films at position A (red), B (blue), and C (green). (a) Fe and (b)
13 Tm peaks are clearly observed.

14 Figure 4: TRIM simulation of the dependence of Tm/Fe ratio on angle. (a) Image of the model.

15 Ar ions have the energy of E_{Ar} in x direction and incident on a $Tm_3Fe_5O_{12}$ target. The spreading
16 angles of the sputtered Tm and Fe based on the x axis (ϕ) were measured. The histogram of the
17 spreading angles of Tm and Fe are obtained and the Tm/Fe ratio at each ϕ was calculated. (b)

18 Comparison of the ϕ -dependence of the Tm/Fe ratio. The filled circle and the open triangle
19 represent the results of simulations at E_{Ar} of 100 eV and 150 eV, respectively. The red inverted
20 triangles and the experimental results based on XRF measurement.

21 Figure 5: The surface morphology in a $2\ \mu m \times 2\ \mu m$ scan area for position A with a thickness 24
22 nm.

23 Figure 6: Hysteresis loop of TmIG film measured by SQUID MPMS3. The paramagnetic
24 background of the substrate has been subtracted. (a) Out of plane and in plane field applied at 24

1 nm. (b) Different thickness. The brown, the red, and the gold plots represent the results of the film
2 thickness of the 18 nm, 24 nm, and 30 nm, respectively. (c) Different position at A, B, and C. Red,
3 blue, and green plots represent the results of the film growth position of A, B, and C, respectively.

4 Figure 7: Angle dependence of the resonance field

5 (a) Schematic of the experimental set up. The symbols are defined in the main text.

6 (b) Representative FMR spectra at $\theta = 0^\circ$. The dashed line indicates the value of $\mu_0 H_{\text{FMR}}$ (=468
7 mT).

8 (c) Angle dependence of $\mu_0 H_{\text{FMR}}$. Black symbols and red lines represent experimental results and
9 values using a self-consistent method [20].

10
11 Figure 8: Observation of the domain space with different thickness by a Kerr microscope.

12 (a) 18 nm

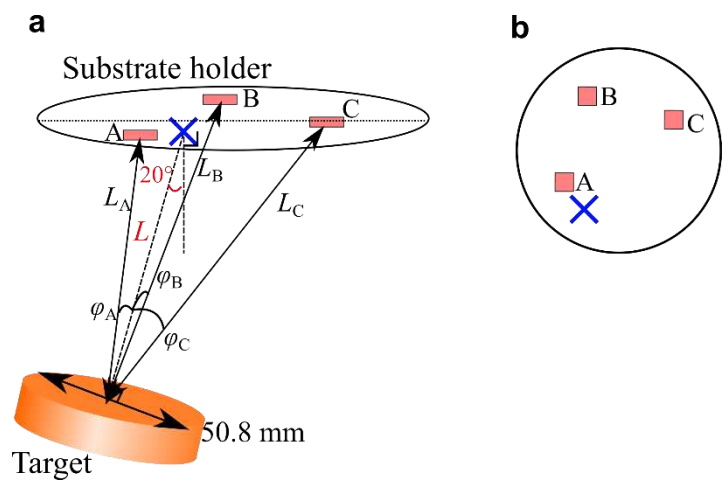
13 (b) 24 nm

14 (c) 30 nm

15 (d) Radially averaged intensity of the microscopic image after two-dimensional FFT. The brown,
16 the red, and the gold plots represent the results of the film thickness of the 18 nm, 24 nm, and 30
17 nm, respectively.

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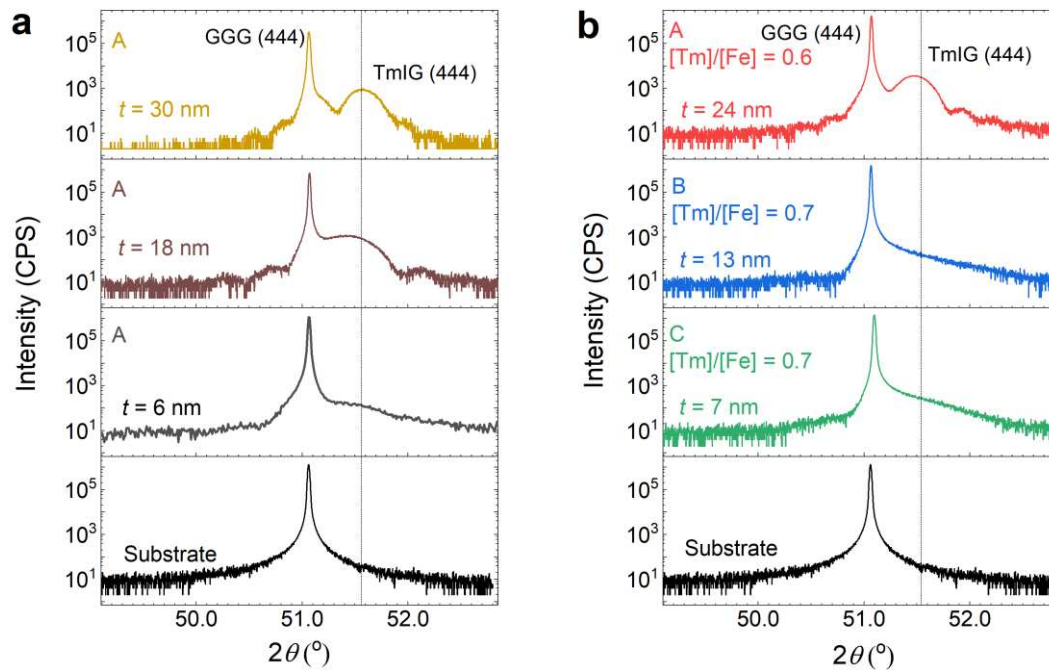
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Figure 1. Marlis Nurut Agusutrisno *et al.*

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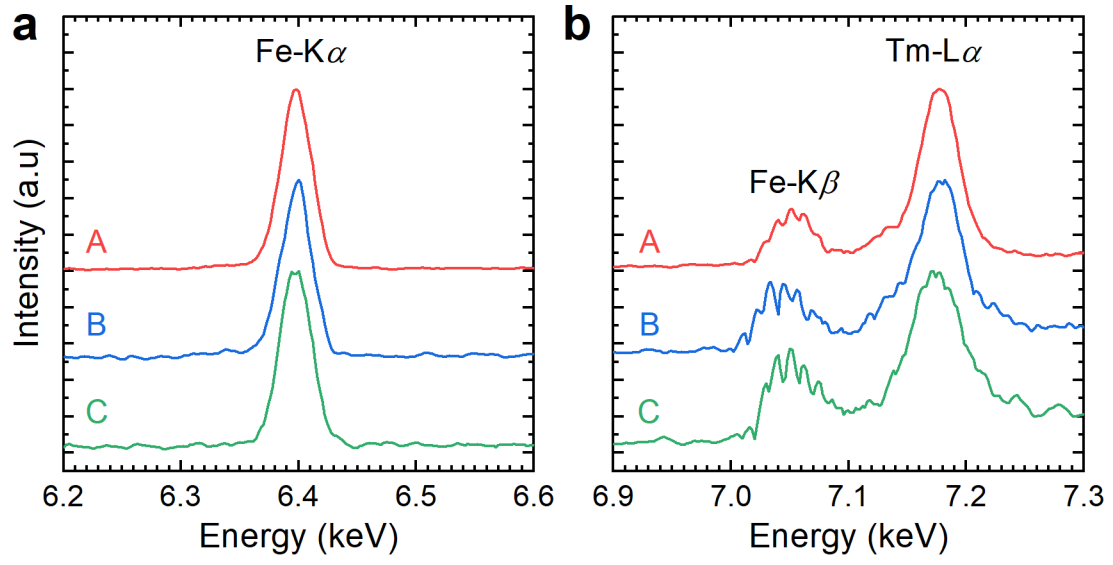
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Figure 2. Marlis Nurut Agusutrisno *et al.*

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Figure 3. Marlis Nurut Agusutrisno et al.

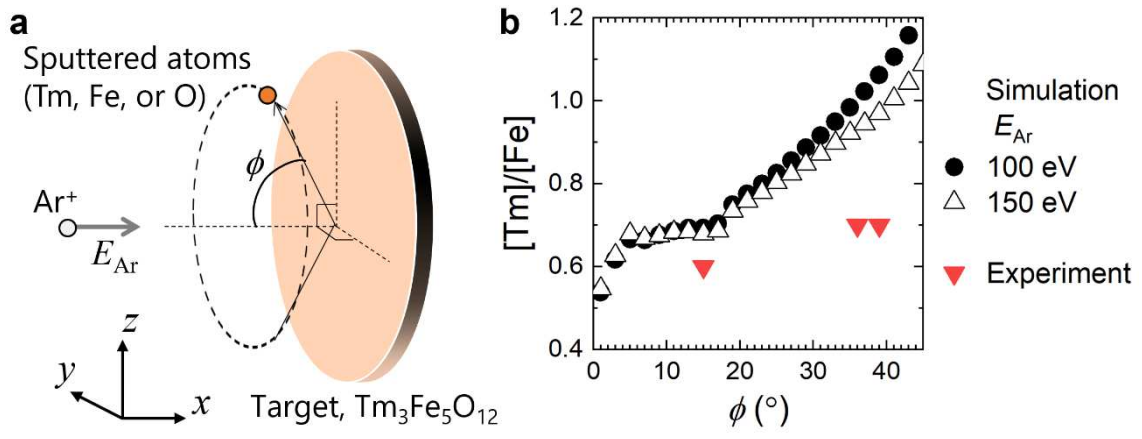
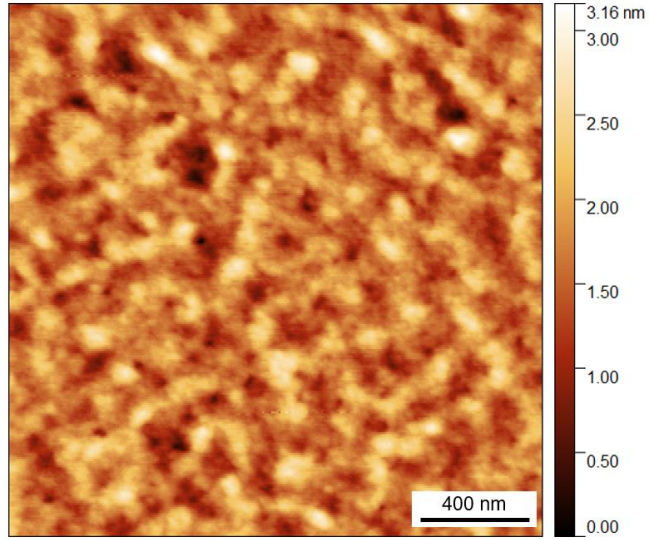


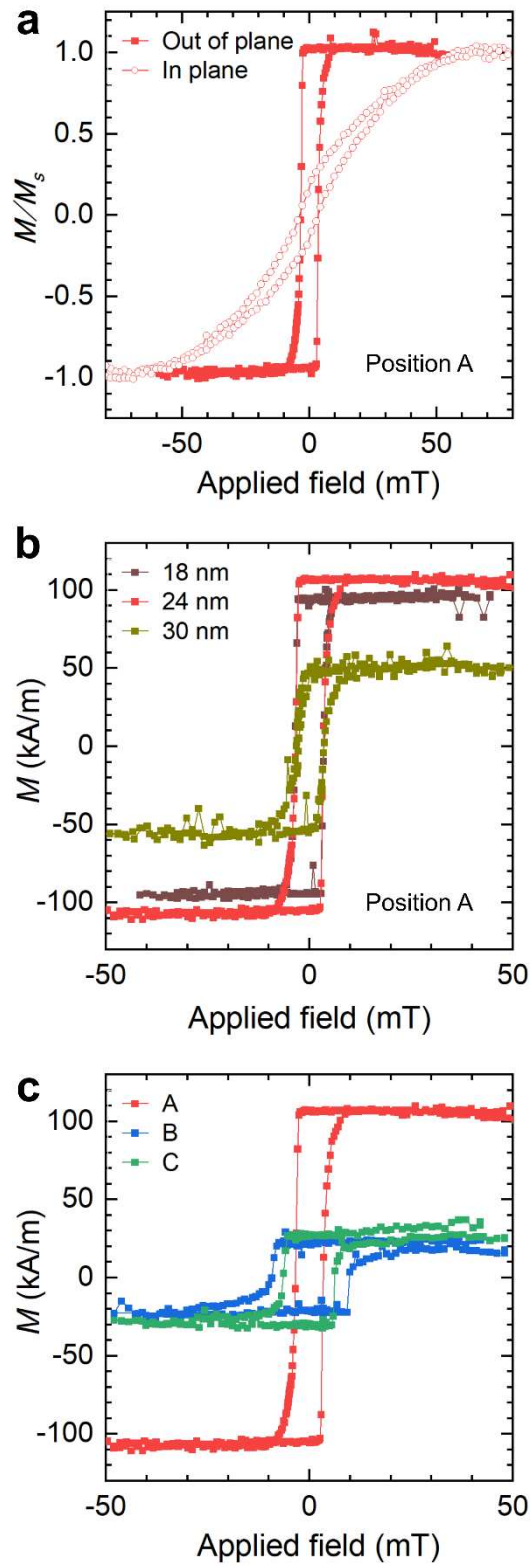
Figure 4. Marlis Nurut Agusutrisno et al.

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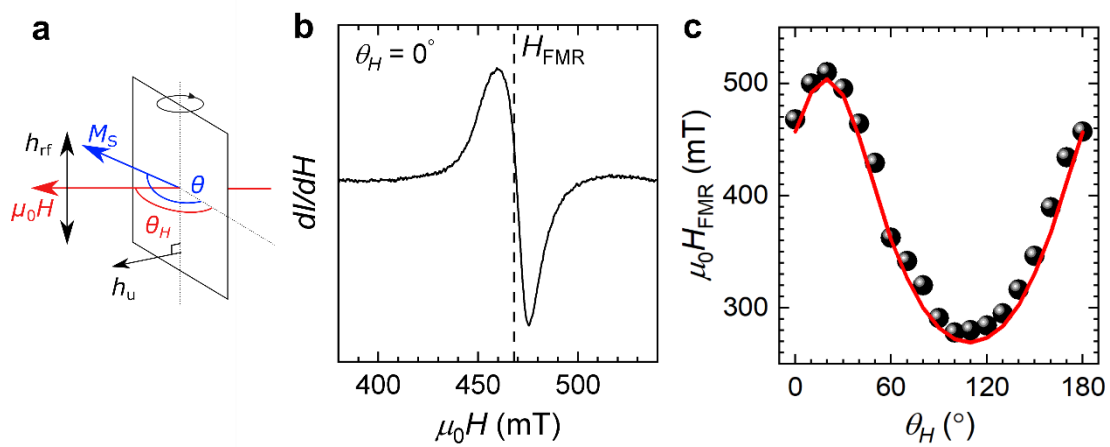
Figure 5. Marlis Nurut Agusutrisno et al.



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Figure 6. Marlis Nurut Agusutrisno *et al.*

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Figure 7. Marlis Nurut Agusutrisno *et al.*

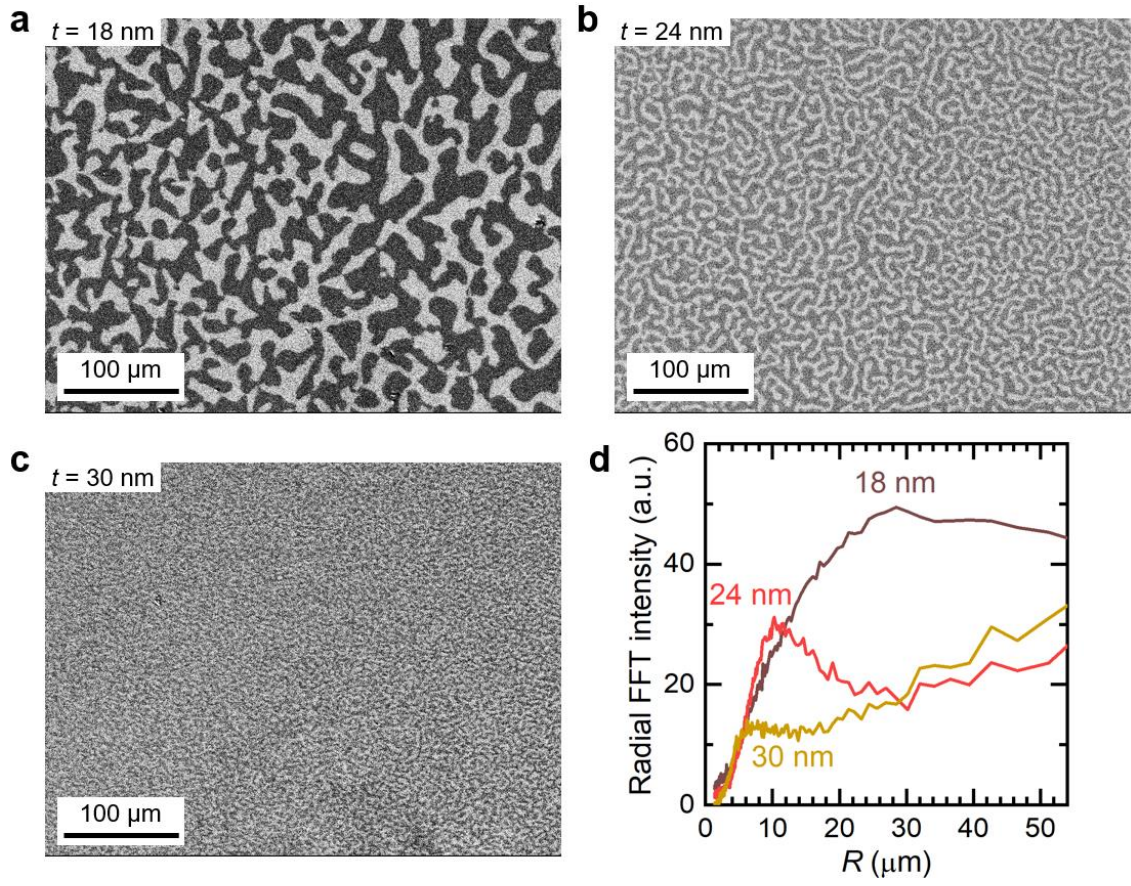


Figure 8. Marlis Nurut Agusutrisno *et al.*

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