Magnetoelectric control of antiferromagnetic domain of Cr₂O₃ thin film toward spintronic application

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Cr₂O₃ is a magnetoelectric antiferromagnet, and its antiferromagnetic domain state is controllable by the simultaneous application of magnetic and electric fields. In the 2000s, that is, more than 50 years since the discovery of the magnetoelectric effect in Cr₂O₃, efforts were initiated to apply this effect to engineering applications. In this article, we review the recent progress of the magnetoelectric control of the antiferromagnetic domain state and the related phenomena of Cr₂O₃, in particular, in an all-thin film system, an essential step to the application.

Key words: magnetoelectric effect, antiferromagnetic domain, Cr₂O₃, thin film

1. Introduction

The interplay between magnetism and electricity has been known as the magnetoelectric (ME) effect, which is referred as the magnetization (M) induction caused by an electric field (E) and the electric polarization (P) induction caused by a magnetic field (H). The ME effect was predicted in the 19th century by Curie. In 1920s, Perrier and Staring investigated this effect using Fe and Ni and proposed the existence of the ME effect. Later on, however, they corrected their experiment was wrong. Meantime, Piccard suggested the impossibility of the effect on symmetry grounds. After these research, Debye who first used the term of “magnetoelectric” suggested that the ME effect was impossible and Van Vleck described in his book the reason why the ME effect was impossible. After 50 years later from the Curie’s proposal, Landau and Lifshitz dealt with this problem and showed that the ME effect should exit in magnetic crystal. Based on the prediction by Landau and Lifshitz, Dzyaloshinskii predicted that Cr₂O₃ was an actual candidate causing this effect. Soon after the prediction, the H-induced polarization P₀ and E-induced magnetization M₁ were experimentally confirmed independently. After the discovery of the ME effect in Cr₂O₃, the research on this effect progressed to the field of “multiferroics”. More details about the history of the ME effect can be found in textbook and the progress of the ME effect after the early development can be found in previous reviews.

As Cr₂O₃ exhibits a linear ME effect, the removing field disperses the ferroic feature. This could partly be the reason why efforts to apply the ME effect of Cr₂O₃ to engineering applications were not very active until the pioneering work by Borisov et al. Their idea is based on the fact that to complement the limitation of the linear ME effect, the ferromagnetic (FM) layer coupled with Cr₂O₃ was used as a marker of the ME-controlled antiferromagnetic (AFM) order parameter. The interfacial exchange coupling between FM and AFM spins results in the exchange bias and the exchange bias polarity is, in principle, determined by the interfacial AFM spin direction. Based on this fundamental, they demonstrated that the exchange bias polarity could be reversed by the so-called ME-field cooling (MEFC) method. In this scenario, to switch the exchange bias polarity, temperature increase was necessary, that is, the system was required to be initialized by heating the sample above the Néel temperature. In 2010, He et al. developed this technique in the isothermal mode. However, these two studies adopted the bulk Cr₂O₃ substrate, and thus the realization of the ME effect in the all-thin film system, which is essential to the device application, was challenging.

Before the achievement of the ME effect through the Cr₂O₃ thin film, some studies theoretically analyzed surface magnetization (or boundary magnetization) and it was supposed that the ME switching of the exchange bias polarity was due to the electrically controllable boundary magnetization that coupled with the AFM order parameter. In 2014, we presented the experimental evidence of the boundary magnetization on Cr₂O₃(0001) including the roughness-insensitive magnetization, absence of the training effect of exchange bias, and ME switching. Nowadays, owing to the achievement of both the MEFC and isothermal switching in an all-thin film system, we can access the details of the ME switching such as the energy condition of the switching and switching dynamics. In this paper, we review the recent progress of the ME control of the AFM domain state of Cr₂O₃, mainly based on our own results as the subject of the magnetic society of Japan (MSJ) outstanding research award 2017.

2. Magnetoelectric control of AFM Cr₂O₃ domain state

In Cr₂O₃, the Cr³⁺ spin lies along the c axis, and the spin direction is contradictory at the neighborhood c plane (Fig. 1). According to this spin alignment, Cr₂O₃(0001) thin film is suitable for inducing the perpendicularly-directed exchange bias that meets the recent requirement of the spintronic devices. In reality, a high perpendicular exchange magnetic anisotropy Jₖ above 0.4 mJ/m² was reported, which can be altered using...
different underlayers by alternating the crystal parameters (the lattice parameters and specific ion positions of Cr³⁺ and O²⁻ ion)⁹. Furthermore, in contrast to the fact that the coercivity enhancement is sometimes accompanied with the exchange bias, the coercivity enhancement of the FM/Cr₂O₃ stacked film can be suppressed using a suitable spacer layer at the FM/Cr₂O₃ interface³⁰). Details about the exchange bias can be found in some previous reviews¹⁶-¹⁸) and, in particular that about the perpendicular exchange bias using a Cr₂O₃(0001) film can be found in our previous review³¹), respectively.

According to the above-mentioned spin alignment, two magnetic domains with the opposite Cr³⁺ spins are energetically degenerated (Fig. 1). The free energy of the two domains under both electric field E and magnetic field H is expressed by³²)

\[
F = F_0 + \sigma_i H_i + \rho_i E_i + \frac{1}{2} \chi_{ij} H_i H_j + \frac{1}{2} \chi_{ij}' E_i E_j + \alpha_{ij} E_i H_j + \ldots
\]

(1)

where the first term \(F_0\) is a constant; \(\sigma_i\) and \(\rho_i\) are pyromagnetic and pyroelectric coefficients, respectively; \(\chi_{ij}\) and \(\chi_{ij}'\) are magnetic and dielectric susceptibility, respectively; and \(\alpha_{ij}\) is the ME coefficient. The second and third terms represent the pyromagnetism and pyroelectric polarizations, respectively, which can be eliminated for Cr₂O₃ because of its crystallographic symmetry.³²) The fourth and fifth terms represent the magnetization and electric polarization, respectively, and the sixth term represents the ME effect. The sign of the sixth term depends on the AFM order parameter of the two AFM domains. \(F^-\) and \(F^+\) denote the free energies of the two domains. Equation (1) indicates that the energy difference of

\[
\Delta F = 2\alpha_{ij} E_i H_j
\]

(2)

was generated for the two domains by simultaneously applying the electric E and magnetic H fields, i.e., the degeneration was broken. Consequently, two AFM domains become selectable. Since the exchange bias of the film is determined by the AFM domain state³³,³⁴) and is controllable by the ME effect. Till date, two types of ME-induced switching protocols have been proposed: MEFC and isothermal processes. In the following section, we describe the results for each process.

2.1 MEFC process

MEFC is a cooling method in which the magnetic and electric fields are simultaneously applied during the cooling from above the Néel temperature of Cr₂O₃ (~307 K)¹⁰,³⁵). In this process, the AFM domain state is determined by the energy competition between the interfacial exchange coupling and ME effect on the onset of the AFM ordering. When electric field E is below the threshold value, the interfacial exchange coupling dominates the AFM domain state, and the exchange bias polarity is then determined by the magnetic field (more precisely, the FM spin direction) during the cooling. This situation is similar to the conventional field-cooling (FC) process. In contrast, when electric field E exceeds the threshold value, the energy gain due to the ME effect, proportional to the product of \(E \times H\) (EH product, see eq. (2)) overcomes the interfacial exchange coupling energy. As a result, the exchange bias polarity is the opposite of that obtained in the previous case.

As a model system to assess the above-mentioned argument, we adopted the Pt/Co(Pt)/Cr₂O₃/Pt thin film exhibiting a perpendicular exchange bias. The details of the film fabrication and the structural details can be found in refs. 29 and 30. To detect the exchange bias polarity, we measured the magnetization curve based on the anomalous Hall effect (AHE) measurements using
the microfabricated device. The optical microscope image of the typical Hall device with the equivalent circuit is shown in Fig. 2. In this device, the Pt buffer layer and the FM layer acted as the bottom and the top electrodes, respectively. In this setup, the electric field was applied across the Cr2O3 layer. The positive directions of magnetic and electric fields were defined as the direction from the bottom electrode to the top electrode. For the identical device used for the AHE measurements, the leakage current was also evaluated. At 312 K and 2000 kV/cm, the leakage current was the order of 1 A/cm² which is sufficiently low to rule out the current-induced switching.

Fig. 3(a) shows the AHE loops after the MEFC. Note that the AHE loops were measured at the highest magnetic field \( H = \pm 2 \text{kOe} \) and zero electric field \( E \), and thus the AFM domain switching cannot occur during the AHE measurements. The negative exchange bias was observed for both the FC process with \( H = +10 \text{kOe} \) (black dotted) and the MEFC process with \( H = +10 \text{kOe} \) and \( E' = -1000 \text{kV/cm} \) (gray solid).

Considering that Co and Cr spins are antiferromagnetically coupled at the interface, the positive magnetic field favors the upward Co moment and the downward interfacial Cr moment. Since the interfacial Cr moment, i.e., as the boundary magnetization couples with the AFM order parameter, a negative \( EH \) also favors the downward Cr moment. Consequently, when \( EH \leq 0 \), the above-mentioned energy competition, and thus the switching of the exchange bias polarity, does not occur. However, when a positive \( EH \) was applied, the upward interfacial Cr moment is energetically favorable. When the value of \( EH \) is high enough to overcome the interfacial exchange coupling, the interfacial Cr moment should be upward, consequently leading to a positive exchange bias. As shown by the black solid line in Fig. 3(a), the AHE loop after the MEFC with \( H = +10 \text{kOe} \) and \( E' = +1200 \text{kV/cm} \) represents the positive exchange bias, which agrees with the above-mentioned arguments.

As long as the exchange bias polarity is determined by the energy competition, there should be a threshold condition to switch the exchange bias polarity. Fig. 3(b) shows the change in the exchange bias field \( H_{EX} \) with respect to the electric field \( E \) during the MEFC with a constant magnetic field \( H \). The sign of the exchange bias changes from negative to positive with increasing \( E \). The change in \( H_{EX} \) with \( E \) can be represented by the functional form of \( \tanh(\Delta G) \), where \( \Delta G \) represents the energy difference between negative and positive exchange-biased states. This agrees with the previous energetic interpretation to induce the positive exchange bias in Fe/FeF2 system by using the conventional FC process in which the energy competition between the interfacial exchange coupling and Zeeman energy in FeF2 layer was considered. This analogy can be understood through eq. (2), which represents the Zeeman energy of the \( E \)-induced magnetization (\( \alpha \)|\( E \)) based on the magnetic field \( H \).

When defining the threshold electric field \( E_{th} \) at which the exchange bias is zero, \( E_{th} \) increases with decreasing \( H \) during the MEFC. As shown in Fig. 3(c), \( E_{th} \) is inversely proportional to \( H \), as shown by eq. (2). The slope of the \( E_{th} \)–1/\( H \) relationship yields the required \( EH \) product to switch the exchange bias polarity, \( (EH)_{th} \). According to the above mentioned arguments, the \( (EH)_{th} \) value increases with the interfacial exchange coupling. Although the direct determination of the interfacial exchange coupling energy is difficult, \( J_K \) is a measure of the exchange coupling energy. Fig. 3(c) shows the \( E_{th} \)–1/\( H \) relationship for some films with different \( J_K \) values. The slope of the curve increases with \( J_K \), thus supporting the earlier discussion.
2.2 Isothermal process

Although the switching based on the MEFC process requires temperature hysteresis, the temperature change is not involved in the isothermal switching. In addition, we can obtain additional information, such as the reversibility of the switching\(^{27}\) and the dynamics of the ME-induced switching\(^{28}\), which could help with the full understanding of ME switching. This section presents the results for the Pt/Co/Au/Cr\(_{2}O_3/Pt\) stacked film, in which the Au spacer layer was deposited to tune the interfacial exchange coupling strength and interfacial magnetic anisotropy\(^{40}\).

2.2.1 Static switching using DC voltage

First, we discuss the reversible switching of the perpendicular exchange bias. Iyama and Kimura\(^{39}\) reported that Cr\(_2O_3\) showed a clear hysteresis in both the \(M\)-\(H\) curve under constant electric field \(E\) and the \(P\)-\(E\) curve under constant magnetic field \(H\); a ferromagnetic (ferroelectric) feature under constant \(B\). This implies that the magnetic domain state of Cr\(_2O_3\) is switchable in an isothermal manner. As the exchange bias polarity couples with the AFM domain state through boundary magnetization, hysteresis also occurs with the change in \(H\) and \(E\) under constant \(H\), as shown in Fig. 3(a). This is contrastive to the MEFC process in which the interfacial Cr\(^{3+}\) orientation is affected by the above-mentioned energy competition. We obtained different \(E_h\) values of \(H\) for the positive-to-negative (P-to-N) and the negative-to-positive (N-to-P) switchings. The difference in \(E_h\) should be due to the unidirectional nature of the interfacial exchange coupling at the FM/AFM interface. The sign of the switching direction denotes the exchange bias polarity. Here, we discuss the energy condition of the isothermal switching. The magnetic free energy of the oppositely directed AFM domains per unit area for the FM/Cr\(_2O_3\) exchange-coupled system can be expressed as

\[
F = K_{AFM}t_{AFM} \sin^2 \theta - \alpha_{33} E H t_{AFM} \cos \theta - M_{AFM} H \cos \theta - J_{AFM} S_{AFM} \cos \theta
\]

\[
= K_{AFM}t_{AFM} \sin^2 \theta
\]

\[
= (\alpha_{33} E t_{AFM} + M_{AFM}) H \cos \theta
\]

\[
- J_{AFM} S_{AFM} \cos \theta
\]

(3)

where \(K_{AFM}\) is the magnetic anisotropy energy density of the AFM layer, \(t_{AFM}\) is the AFM layer thickness, \(\alpha_{33}\) is the ME coefficient, \(J\) is the interfacial exchange coupling energy, \(S_{FM}\) and \(S_{AFM}\) are the FM and AFM spins, respectively, and \(M_{AFM}\) is the uncompensated AFM moment. Further, \(\theta\) is denoted by the angle between the interfacial AFM and FM spins (or \(B\)). The first, second, and third terms represent the magnetic anisotropy energy, Zeeman energy of the \(E\)-induced magnetization and uncompensated AFM spin, and the interfacial exchange coupling, respectively. \(M_{AFM}\) can be caused by the defect-induced finite magnetization\(^{40}\) and/or the interfacial uncompensated AFM moment, which was the dominant factor in our case.\(^{27}\) Note that in eq. (3), the FM spins are fixed to the direction of magnetic field \(H\) because the applied magnetic field \(H\) during the application of electric field \(E\) is high enough (\(> 40 \text{kOe}\)) to fix the FM spin. Thus, the Zeeman energy and magnetic anisotropy energy of the FM layer are not included in eq. (3) because these terms become constant when the FM magnetization is fixed. By minimizing eq. (3), the energy condition for ME switching is derived as\(^{27}\)

\[
\left(\alpha_{33} E + M_{AFM} \frac{t_{AFM}}{t_{FM}}\right) H = \pm 2K_{AFM} \frac{J_{FM} S_{FM} S_{AFM}}{t_{AFM}} \quad (4)
\]

Each sign of the first term corresponds to the N-to-P and P-to-N switchings. Fig. 3(b) shows the magnetic field dependence of \(E_h\), which is inversely proportional to \(H\), as shown in eq. (4). The unique feature that appeared in the all-thin-film system was (1) the different switching energy depending on the switching direction and (2) the appearance of the offset \(E\) in the \(E\)-\(1/H\) relationship. The former is found in the difference in the slope of two curves, i.e. N-to-P switching and P-to-N switching. Accordingly, the slopes of the two curves give \(K_{AFM}\) and \(J_{FM} S_{FM} S_{AFM}\), whose values are \(4.5 \pm 0.6 \times 10^3 \text{ J/m}^3\) and \(1.5 \pm 0.2 \times 10^{-2} \text{ mJ/m}^2\), respectively, assuming \(\alpha_{33} = 3-4 \text{ ps/m}\). The estimated \(K_{AFM}\) is approximately half of the \(K_{FM}\) of the bulk Cr\(_2O_3\) at the measurement temperature of 280 K. This underestimation is probably because of the above-mentioned estimation assumes the coherent rotation, while in reality, the nucleation and propagation of the reversed AFM domains should be involved\(^{28}\). In addition, the estimated value of \(J_{FM} S_{FM} S_{AFM}\) differs from that of the ideal model. That is, the estimated \(J_{FM} S_{FM} S_{AFM}\) was...
approximately twice of the exchange anisotropy energy density, \( J_c = H_{ex}M_s \). \( t_{FM} = 5.8 \times 10^{-3} \) mJ/m². The values of \( J_{S_{AFM}}S_{AFM} \) and \( J_c \) might be equal when the pinned spin model \( 1 \) is valid in our film. However, in actuality, the interfacial Cr moments were not perfectly pinned but canted from the original direction with respect to the FM interfacial Cr moments. The offset \( E \) in the \( E/1/H \) relationship, i.e., the nonzero \( E_0 \) interception in the limit of \( 1/H \) to zero is another characteristic of all-thin-film system. According to eq. \( 2 \), the offset \( E_0 \) is caused by the uncompensated AFM moment \( M_{AFM} \). In our system, \( M_{AFM} \) relevant to the offset \( E_0 \) is mainly the interfacial uncompensated AFM moment. This should be reasonable because the ME switching of the AFM domain detected through the exchange bias is an interfacial effect. In other words, by utilizing \( E_0 \) positively, the shift of the \( E/1/H \) curve from the origin becomes significant. In the practical use, \( E_0 \) at the certain \( H \) decreases with increasing \( E_0 \), i.e. increasing \( M_{AFM} \) or decreasing \( t_{AFM} \) which may become one solution to decrease the switching energy.

### 2.2.2 Dynamical switching using pulse voltage

The important advantage of isothermal switching is that we can access the switching dynamics by adopting the pulsed magnetic or electric fields. In general, the pulse width of the pulsed magnetic field above several tens of kOe is in the range of milliseconds.\(^{39} \) In contrast, the fast pulse \( E \) below microseconds can be easily generated using the general pulse generator. Thus, we can access fast dynamics by adopting the pulsed electric field and a constant magnetic field. Figure 4(a) shows the change in \( H_{ex} \) and \( M_{AFM}/M_s \) as a function of pulse width. These factors were collected from the AHE loops after applying \( H = 60 \) kOe and pulsed electric field with the amplitude of \(-333 \) kV/cm and width of \( 20 \) ns–\( 10 \) μs.\(^{29} \) As the DC \( E_0 \) value of this film was \(-275 \) kV/cm (\(-4.1 \) V; see reference 28), the above-mentioned condition is slightly above the DC threshold condition. With increasing pulse width, \( H_{ex} \) changes from negative (positive) to positive (negative) at the pulse width of 500 ns. The change in \( M_{AFM}/M_s \) is gradual compared with that in \( H_{ex} \).

This is because the coercivity \( H_c \) is lower than \( H_{ex} \), the magnetization reversal is sharp around \( H_c \), and the two-step magnetization process was observed at the intermediate state, as shows in the inset of Fig. 4(a). The slow switching time suggests that the switching process is dominated by the domain wall motion. In such a case, the switching time is dominated by the domain wall velocity and could be decreased by increasing the pulse amplitude. Figures 4(b) and 4(c) show the change of \( H_{ex} \) and \( M_{AFM}/M_s \) as functions of pulse amplitude and pulse width, respectively. The magnetic field during the application of an electric field was maintained at \(-60 \) kOe. The switching time decreases with increasing pulse amplitude, as expected earlier. As the ME-induced switching is triggered by the electric field, the microscopic origin of the domain wall motion might be different from the magnetic-field induced domain wall dynamics observed in the ordinal FM layer.\(^{44–46} \)

Nonetheless, as indicated by eqs. (1) and (2), the role of electric field \( E \) is to induce magnetization, and a change in \( E \) alters the energy gain by the Zeeman energy of the \( E \)-induced magnetization. Therefore, the driving force to induce the magnetic domain wall propagation should be analogous to the case of FM (or ferrimagnetic) domain wall. By assuming that \( Cr_2O_3 \) under a finite \( E \) behaves as a ferrimagnet,\(^{32} \) the domain wall velocity was calculated based on the simple theory discussed in refs. 44–47. The details of the calculation can be found in ref. 28. As shown by the solid line in Fig. 4(c), the switching time can be roughly reproduced using the simple model with some assumed values: domain wall width, \( 38 \) nm; ME

![Fig. 5](image-url) (a) Changes in \( H_{ex} \) and \( M_{AFM}/M_s \) as a function of pulse width. \( H_{ex} \) and \( M_{AFM}/M_s \) were collected from the AHE loops after applying pulsed \( E \) under the constant \( H \) (= \(-60 \) kOe). The pulse amplitude was \(-333 \) kV/cm. Inset of (a) represents the typical AHE loop at the intermediate state. Changes in (b) \( H_{ex} \) and (c) \( M_{AFM}/M_s \) as functions of pulse amplitude and width, with white/black circles representing positive/negative values and their color depth representing absolute values. Solid line in (c) represents the calculated switching time. Measurement temperature was 280 K.\(^{29} \)
coefficient at 280 K, 3.5 ps/m; Ginbert damping parameter of each sublattice, 0.05; and travel distance, 1 μm (half of the Hall device width). Despite the rough agreement in the switching times, some assumed values need further investigation. For instance, the assumed Ginbert damping parameter of each sublattice is the order of $10^{-2}$–$10^{-1}$, which may be larger than the predicted value. One reason may be the creep motion of the domain wall under our adopted experimental condition as indicated by the large deviation in the low amplitude regime. The direct observation of the ME-induced magnetization reversal process is beneficial for both the quantitative analysis and deeper understanding of the phenomena.

3. Element-specific magnetic domain observation

For the observation of the ME-induced magnetization switching process, it can be simply assumed that FM and AFM domains are coupled spatially, at least, at the remanent state. In other words, if this assumption is valid, we can obtain the AFM domain pattern by observing the FM domain. However, this is nontrivial. In the case of the in-plane exchange-biased film, the AFM domains can be imaged through X-ray linear dichroism (XMLD). Accordingly, the AFM domain just below the individual FM domains is not a single domain state, implying that FM and AFM domain patterns are not the same. Hence, as a first step in the investigation, the magnetic domain states of FM and AFM layers must be obtained independently.

The XMLD technique is not applicable for our system because the AFM spin orientation should be restricted to upward or downward directions (see Fig. 1), and such collinear spins are not distinguishable through XMLD. X-ray magnetic circular dichroism (XMCD) complements this limitation and is a powerful tool for the perpendicularly directed spin system. Based on the XMCD measurements by using the focused soft X-ray, we observed the spatial distribution of the XMCD intensity corresponding to the magnetic domains for FM Co and AFM Cr spins independently. In this work, we employed the scanning XMCD microscope equipped at BL25SU, SPring-8. The schematic drawing of the scanning XMCD microscope is shown in Fig. 6. In this microscope, the soft X-ray was focused on the sample using the Fresnel Zone Plate (FZP) and the order sorting aperture (OSA). By scanning the sample in x and y directions in keeping the focused state, the special distribution of the XMCD intensity was collected. In this microscope, the XMCD signal can be detected based on the surface-sensitive total electron yield method, suitable to detect the tiny signal from the interfacial uncompensated Cr spin. The details of this microscope can be found in refs. 33 and 52.

Figure 7 shows the spatial distributions of XMCD of Co and Cr for the Pt/Co/Cr2O3/Pt thin film measured at 205 K. The multidomain state was created through AC-demagnetization at room temperature, and then the

![Fig. 6 Schematic drawings of the scanning XMCD microscope.](Image)

![Fig. 7 Spatial distribution of XMCD measured at 205 K.](Image)
sample was cooled to retain the demagnetized state. Note that the XMCD from the Cr was performed using the interfacial uncompensated Cr moment. Two patterns were observed to be very similar indicating that the FM and AFM domains are spatially coupled. This spatial coupling of the magnetic domains is probably due to the strong interfacial exchange coupling and the collinear interfacial spin alignments of both Co and Cr. The sign of the XMCD is contradictory for Co and Cr indicating antiferromagnetic interfacial exchange coupling. From the spatial coupling of the magnetic domains, the exchange bias polarity can be assumed to be determined according to the domain-by-domain basis, implying that the exchange bias polarity is determined by the interfacial AFM moments, i.e., the boundary magnetization. This can be directly verified by measuring the local magnetization curve on the individual magnetic domains’ this is under investigation. For the direct observation of the ME-induced magnetization switching process, in addition to the imaging technique described previously, the applications of magnetic field above several tens of kOe and the electric field (of both DC and pulse) in the microscope instrument are necessary. These techniques are now in the developing stage and will be reported in the near future.

4. Summary

In this article, we reviewed the ME-induced magnetization reversal mainly based on our own results. Discovered in the 1950s in bulk Cr$_2$O$_3$, the ME effect was recently observed in an all-thin-film system. Owing of this development, we could address the various phenomena involving the ME effect, e.g., the control of the interfacial magnetization and switching dynamics. Many challenges still exist for the actual application of the ME effect to a storage/memory device, e.g., the reduction of the switching energy, fast switching, and thermal stability. To solve these challenges, a deeper understanding of the ME-induced phenomena is essential.

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References

1) P. Curie: J. Phys. (Paris), 3, 395 (1894).
2) A. Perrier: Arch. Sci. Phys. Nat., 4, 369 (1922).
3) A. Perrier and A. J. Starling: Arch. Sci. Phys. Nat., 4, 373 (1922), ibid, 5, 333 (1923).
4) A. Perrier and C. E. Borel: Arch. Sci. Phys. Nat., 7, 289 (1925).
5) A. Piccard: Arch. Sci. Phys. Nat., 6, 404 (1924).
6) P. Debye: Z. Phys., 36, 300 (1926).
7) J. H. Van Vleck: The theory of electric and magneto susceptibilities (Oxford University Press, London, 1932), pp.113-121 and pp. 279-281.
8) L. D. Landau and E. M. Lifshitz, Electrodynamics of continuous media (English Transl. Pregamon Press, Oxford, 1969), p. 115, p.119, pp.313-344.
9) I. E. Dzyaloshinskii: Sov. Phys. JETP, 10, 628 (1959).
10) N. Astrov: Sov. Phys. JETP, 11, 765 (1960).
11) V. J. Folen, G. T. Rado, and E. W. Stadler: Phys. Rev. Lett., 6, 607 (1961).
12) T. Arima: J. Phys. Soc. Jpn., 80, 053001 (2011).
13) T. H. O’Dell, The electrodynamics of magneto-electric media (North-Holland Publishing Company, 1970), Chapter 1.
14) M. Fiebig: J. Phys. D: Appl. Phys., 38, R123 (2005).
15) F. Borisov, A. Hochstrat, X. Chen, W. Kleemann, and C. Binek: Phys. Rev. Lett., 94, 117203 (2005).
16) N. Ogusu and I. K. Schuller, J. Magn. Magn. Mater., 192, 203 (1999).
17) A. E. Berkowitz and K. Takano: J. Magn. Magn. Mater., 200, 553 (2000).
18) R. L. Stamps: J. Phys. D: Appl. Phys., 33, R247 (2000).
19) X. He, Y. Wang, A. N. Caruso, E. Vosco, K. D. Bealschchenko, P. A. Dowben, and C. Binek: Nat. Mater., 9, 579 (2010).
20) A. F. Andreev, JETP Lett., 65, 758 (1996).
21) K. D. Belashchenko: Phys. Rev. Lett., 105, 147204 (2010).
22) K. Toyoki, Y. Shiratsuchi, T. Nakamura, C. Mitsumata, S. Harimoto, Y. Takechi, T. Nishimura, H. Nomura, and R. Nakatani: Appl. Phys. Express, 7, 114201 (2014).
23) A. F. Andreev, JETP Lett., 65, 758 (1996).
24) K. Toyoki, Y. Shiratsuchi, T. Nakamura, C. Mitsumata, S. Harimoto, Y. Takechi, T. Nishimura, H. Nomura, and R. Nakatani: J. Magn. Magn. Mater., 117, 19D902 (2015).
25) T. Ashida, M. Oida, N. Shimomura, T. Nozaki, T. Shibata, and M. Sahashi: J. Magn. Magn. Mater., 372, 504 (2015).
26) K. Toyoki, Y. Shiratsuchi, A. Kobane, C. Mitsumata, Y. Kotani, T. Nakamura, and R. Nakatani: J. Magn. Magn. Mater., 346, 117203 (2015).
27) Y. Shiratsuchi, Y. Nakano, N. Inami, T. Ueno, K. Ono, R. Kumai, R. Sagayama, and R. Nakatani: J. Appl. Phys., 123, 103903 (2018).
28) Y. Shiratsuchi, W. Kuroda, T. V. A. Nguyen, Y. Kotani, K. Toyoki, T. Nakamura, M. Suzuki, K. Nakamura, and R. Nakatani: J. Appl. Phys., 121, 073905 (2017).
29) Y. Shiratsuchi, Y. Shiratsuchi, A. Kobane, S. Yoshida, and R. Nakatani: J. Magn. Magn. Mater., 369, 2100 (2016).
30) Y. Shiratsuchi, Y. Shiratsuchi, S. Watanabe, H. Yoshida, N. Kishida, R. Nakatani, Y. Kotani, K. Toyoki, and T. Nakamura: to be submitted.
31) Y. Shiratsuchi, S. Watanabe, H. Yoshida, N. Kishida, R. Nakatani, Y. Kotani, K. Toyoki, and T. Nakamura: to be submitted.
32) T. J. Martin, and J. C. Anderson: IEEE Trans. Magn., 2, 446 (1966).
33) Y. Shiratsuchi, S. Watanabe, H. Yoshida, N. Kishida, R. Nakatani, Y. Kotani, K. Toyoki, and T. Nakamura: to be submitted.
34) Y. Shiratsuchi, S. Watanabe, H. Yoshida, N. Kishida, R. Nakatani, Y. Kotani, K. Toyoki, and T. Nakamura: to be submitted.
(2015).
39) A. Iyama, and T. Kimura: Phys. Rev. B, 87, 180408(R) (2013).
40) T. Kosub, M. Kopte, R. Hühne, P. Appel, B. Shields, P. Maletinsky, R. Hübner, M. O. Lièdeke, J. Fassbender, O. G. Schmidt, and D. Makarov: Nat. Comm., 8, 13985 (2017).
41) W. H. Meiklejohn, and C. P. Bean: Phys. Rev., 102, 1413 (1956), ibid 105, 904 (1957).
42) M. Al-Mahdawi, S. P. Pati, Y. Shiokawa, S. Ye, T. Nozaki, and M. Sahashi: Phys. Rev. B, 95, 144423 (2017).
43) T. Nakamura, Y. Narumi, T. Hirono, M. Hayashi, K. Kodama, M. Tsunoda, S. Isogami, H. Takahashi, T. Kinoshita, K. Kindo, and H. Nojiri: Appl. Phys. Express, 4, 066602 (2011).
44) T. Ono, H. Miyajima, J. Shigeto, N. Hosuito, and T. Shinjo: Science, 284, 468 (1999).
45) D. Atkinson, D. A. Allwood, G. X. Cooke, C. C. Faulkner, and R. D. Cowburn: Nat. Mater., 2, 85 (2003).
46) G. D. Beach, C. Nistor, K. Maxim, and J. K. Erskine: Nat. Mater., 4, 741 (2005).
47) R. Giles, and M. Mascuripur: J. Mag. Soc. Jpn., 18, 299 (1991).
48) K. D. Belashchenko, O. Tedemeshyov, A. A. Kovaliv, and O. A. Tretiakov: Appl. Phys. Lett., 108, 132403 (2016).
49) P. Borisov, T. Ashida, T. Nozaki, M. Sahashi, and D. Lederman: Phys. Rev. B, 93, 174415 (2016).
50) A. Scholl, J. Stöhr, J. Lüning, J. W. Seo, J. Frompeyrine, H. Siegwart, J. –P. Locquet, F. Nolting, S. Anders, E. E. Fullerton, M. R. Scheinfein, and H. A. Padmore: Science, 287, 1014 (2000).
51) F. Nolting, A. Scholl, J. Stöhr, J. W. Seo, J. Frompeyrine, H. Siegwart, J. –P. Locquet, S. Anders, J. Lüning, E. E. Fullerton, M. F. Toney, M. R. Scheinfein, and H. A. Padmore: Nature, 406, 767 (2000).
52) Y. Kotani, Y. Senba, K. Toyoki, D. Bikkington, H. Okazaki, A. Yasui, W. Ueno, H. Ohashi, S. Hiroasa, Y. Shiratsuchi, and T. Nakamura: J. Synchrotron Rad., 25 in press.

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