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Giant oscillations in spin-dependent tunneling resistances as a function of barrier thickness in fully epitaxial magnetic tunnel junctions with a MgO barrier

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Giant oscillations in spin-dependent tunneling resistances as a function of MgO barrier thickness \(t_{\text{Mgo}}\) were observed for fully epitaxial magnetic tunnel junctions with Heusler alloy \(\text{Co}_2\text{Zn}_{0.3}\text{Al}\) electrodes and a MgO barrier. The oscillations in tunneling resistances were well approximated by a superposition of an exponential function of \(e^{at_{\text{Mgo}}+b}\) and a periodic function of \(1+C\cos[(2\pi/V)_{\text{Mgo}}+\varphi]\) with significantly large amplitudes \(C \approx 0.16 \pm 0.01\) even at 293 K for both parallel and antiparallel magnetization orientations.

The period was found to be almost independent of temperature and bias voltage \((V)\). The amplitudes \(C\) showed only weak dependence on \(V\) at least up to 0.2 V. These features should be a key to understand the origin of the pronounced oscillations.

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I. INTRODUCTION

Spin-dependent tunneling in magnetic tunnel junctions (MTJs) consisting of epitaxial, single-crystalline ferromagnet/insulator/ferromagnet trilayers has received intensive theoretical and experimental investigation in this decade.\(^1\)–\(^8\) The importance of the symmetry of electronic states in the electrodes and of the evanescent states in a single-crystal barrier for spin-dependent tunneling in epitaxial MTJs has been clarified.\(^1\)–\(^3\) This concept combined with atomic-level controlled tunnel barrier preparation technologies resulted in unprecedentedly high tunnel magnetoresistance ratios in fully epitaxial Fe/MgO(001)/Fe MTJs (hereafter, Fe MTJs) (Ref. 7) and related MTJs with a highly oriented MgO barrier and also with highly oriented transition-metal electrodes of Co\(_{1-x}\)Fe\(_x\) (Ref. 8) and CoFeB.\(^9\)

For the complex energy bands in a single-crystal MgO barrier, Butler et al. theoretically predicted a quantum interference effect between the evanescent states in the single-crystal MgO barrier.\(^2\) Yuasa et al. reported an oscillation in the tunneling magnetoresistance (TMR) ratio as a function of the MgO barrier thickness \(t_{\text{Mgo}}\) for epitaxial Fe MTJs at 20 and 293 K.\(^7\) Later, Matsumoto et al. reported oscillations in the tunneling resistances \(R_p\) and \(R_{\text{AP}}\) for the parallel (P) and antiparallel (AP) magnetization orientations between the lower and upper electrodes, respectively, as a function of \(t_{\text{Mgo}}\) at 20 K for Fe MTJs.\(^10\) They reported an oscillation with a short period of 0.32 nm for \(R_p\) and an oscillation expressed as a superposition of a short period of 0.32 nm and a long period of 0.99 nm for \(R_{\text{AP}}\). Matsumoto et al. discussed a possible origin based on the quantum interference effect of evanescent states in a MgO barrier predicted by Butler et al., in particular, through possible contributions of hot spots for both P and AP but the origin has not been understood yet.\(^10\)

Our purpose in the present study has been to investigate possible oscillations in \(R_p\) and \(R_{\text{AP}}\) in MgO-based MTJs with different electrodes and clarify the factors that affect the oscillations. The investigation and clarification are essential not only for the understanding of the physics behind the oscillations in MTJs but also for the creation of future-generation spintronic devices based on a novel operating principle. A possible candidate is a combination of Co-based Heusler alloys (Co\(_y\)YZ, where \(Y\) is usually a transition metal and \(Z\) is a main group element) (Ref. 11) and a MgO barrier. We recently proposed and developed fully epitaxial MTJs with Co\(_y\)YZ electrodes\(^12\)–\(^16\) and a MgO barrier to thoroughly utilize the potentially high spin polarization of this material system.\(^17\)–\(^20\) We have observed oscillations in the TMR ratio as a function of \(t_{\text{Mgo}}\) for fully epitaxial MgO-based MTJs with Heusler alloy Co\(_{28}\)Mn\(_{32}\)Si thin films as both lower and upper electrodes, Co\(_{28}\)Mn\(_{32}\)Si/MgO/Co\(_{32}\)Mn\(_{32}\)Si MTJs (Co\(_{32}\)Mn\(_{32}\)Si MTJs), with a period of 0.28 nm.\(^21\) The oscillation period of 0.28 nm was close to the short oscillation period of 0.32 nm for the oscillatory \(t_{\text{Mgo}}\) dependence of \(R_p\) and \(R_{\text{AP}}\) observed for Fe MTJs. However, we could not extract the oscillatory components of \(R_p\) and \(R_{\text{AP}}\) for the fabricated Co\(_{32}\)Mn\(_{32}\)Si MTJs because possible oscillations in \(R_p\) and/or \(R_{\text{AP}}\) were veiled by the scattering of the junction area. On the other hand, the junction area scattering does not affect the TMR ratio because the TMR ratio calculated as \((R_{\text{AP}}-R_p)/R_p\) is independent of the junction area.

Among several combinations of Co\(_2\)YZ and a MgO barrier, Co\(_2\)Zn\(_{0.3}\)Fe\(_{0.7}\)Al (CCFA) features a smaller lattice mismatch with MgO(001) of −3.7% for a 45° in-plane rotation within the (001) plane,\(^12\) which is a contrast to that of −5.1% between Co\(_{50}\)Mn\(_{50}\)Si(001) and MgO and also lower than that of −3.9% between Fe(001) and MgO. We have demonstrated extremely smooth and abrupt interfaces in (from the lower electrode side) CCFA/MgO(001)/Co\(_{50}\)Fe\(_{50}\) (CoFe) fully epitaxial MTJ trilayers and measured a relatively high TMR ratio of 317% at 4.2 K (109% at room temperature) for CCFA/MgO/CoFe MTJs.\(^14\)

Given this background, we investigated possible oscillations in fully epitaxial MTJs with a single-crystal MgO(001) barrier and Heusler alloy CCFA as both lower and upper electrodes CCFA/MgO(001)/CCFA MTJs (hereafter, CCFA MTJs). We found pronounced oscillatory behaviors for \(R_p\) and \(R_{\text{AP}}\) for CCFA MTJs with large oscillation amplitudes that were about eight times greater even at 293 K than those observed for Fe MTJs at 20 K.\(^10\) Thus, we unambiguously demonstrated the existence of oscillations in \(R_p\) and \(R_{\text{AP}}\) as a function of the barrier thickness.
II. EXPERIMENTAL

The fabricated CCFA-MTJ layer structure was as follows: (from the substrate side) MgO buffer (10 nm)/lower CCFA (50 nm)/MgO (0.8–3.4 nm)/upper CCFA (10 nm)/Ru (0.8 nm)/Co8Fe10 (2 nm)/IrMn (10 nm)/Ru cap (5 nm), grown on MgO(001) substrates. The preparation of the fully epitaxial CCFA MTJs is described in detail elsewhere. The nominal thickness of the MgO tunnel barrier ($t_{\text{MgO}}$) was varied from 0.8 to 3.4 nm on each 20 $\times$ 20 mm$^2$ substrate by a linearly moving shutter during the deposition by electron-beam evaporation. We fabricated densely arranged MTJs with the fully epitaxial layer structure through photolithography. The fabricated junction size was 10 $\times$ 10 $\mu$m$^2$ or 8 $\times$ 8 $\mu$m$^2$. The magnetoresistance was measured using a dc four-probe method with a magnetic field applied along the [110] axis of the CCFA. The bias voltage ($V$) was defined with respect to the lower electrode. For comparison, we also identically fabricated densely arranged fully epitaxial CCFA/MgO/CoFe MTJs with the nominal $t_{\text{MgO}}$ ranging from 0.8 to 3.4 nm. The fabricated CCFA/MgO/CoFe MTJ layer structure was essentially the same as the CCFA-MTJ layer structure described above except that the upper electrode was replaced by a CoFe electrode (3 nm). We measured $R_P$ and $R_{\text{AP}}$ as a function of $t_{\text{MgO}}$ for CCFA MTJs at 4.2 and 293 K and for CCFA/MgO/CoFe MTJs at 293 K.

III. RESULTS AND DISCUSSION

A cross-sectional high-resolution transmission electron microscope lattice image of a fabricated CCFA/MgO/CCFA MTJ layer structure is shown in Fig. 1. This image clearly shows that all the layers of the CCFA-MTJ trilayer were grown epitaxially and were single crystalline. It also confirms that extremely smooth and abrupt interfaces were formed.

We will now describe the $t_{\text{MgO}}$ dependence of $R_P$ and $R_{\text{AP}}$. Figures 2(a) and 2(b) show $R_P$ and $R_{\text{AP}}$ as a function of $t_{\text{MgO}}$ at 4.2 K and 293 K, respectively, along with the corresponding TMR ratios in (c), for fully epitaxial CCFA MTJs, where $A$ represents the nominal junction area. Pronounced oscillatory dependences were observed in both $\ln R_P$ vs $t_{\text{MgO}}$ and $\ln R_{\text{AP}}$ vs $t_{\text{MgO}}$ plots at 4.2 and 293 K, as shown in Figs. 2(a) and 2(b). These clear periodic behaviors in $\ln R$ vs $t_{\text{MgO}}$ plots for both P and AP for both 4.2 and 293 K are characterized by the alternate appearance of two regions in each period: one with a gentle slope $\alpha_L$ and the other with a steep slope $\alpha_H$ in both $\ln R$ vs $t_{\text{MgO}}$.

To extract characteristic features of these distinct periodic behaviors, we first approximated the baseline dependence of $R_P$ ($R_{\text{AP}}$) by an exponential function $\exp(a t_{\text{MgO}} + b)$ for $t_{\text{MgO}}$ range from 1.7 to 3.0 nm for 293 K and from 1.9 to 2.85 nm for 4.2 K by the least-squares method. Then, we analyzed the reduced tunnel resistance $r_P$ ($r_{\text{AP}}$), which is tunnel resistance $R_P$ ($R_{\text{AP}}$) divided by each baseline function $\exp(a t_{\text{MgO}} + b)$.
We then approximated \( r_p \) and \( r_{AP} \) at 4.2 and 293 K by

\[
  r = 1 + C \cos[(2\pi/T)_{MgO} + \varphi]
\]

with three parameters: oscillation amplitude \( C \), period \( T \), and phase \( \varphi \) (the first step of the approximation for \( r_p \) and \( r_{AP} \)). The periods thus obtained for \( P \) and \( AP \) and for 4.2 and 293 K agreed well each other and were 0.30 nm \( \pm 2\% \). The difference of \( \pm 2\% \) is negligible. Thus, we conclude that the periods are common for \( P \) and \( AP \) and independent of temperature, at least up to 293 K. The period of 0.30 nm obtained for CCFA MTJs is in good agreement with the period of 0.32 nm observed for the short-period oscillations in \( r_p \) and \( r_{AP} \) for Fe MTJs. Although both \( r_p \) and \( r_{AP} \) were well approximated by the first-step approximation with the three fitting parameters for each \( r_p \) or \( r_{AP} \), we again fitted the oscillations in \( r_p \) and \( r_{AP} \) with Eq. (1) with the thus-obtained common period of 0.30 nm (the second step of the approximation for \( r_p \) and \( r_{AP} \)) to obtain more certain phase difference values of \( \varphi_p - \varphi_{AP} \). (The amplitudes \( C_p \) and \( C_{AP} \) obtained by the second-step approximation were almost equal to those obtained by the first-step approximation.) The solid lines for \( r_p \) and \( r_{AP} \) shown in Fig. 3 are curves fitted in this manner. Both \( r_p \) and \( r_{AP} \) were well approximated by Eq. (1) at both 4.2 and 293 K.

Note that the oscillations in both \( r_p \) and \( r_{AP} \) for CCFA MTJs were well approximated by a single period of 0.30 nm. This is a contrast to the more complex oscillatory dependence on \( r_{AP} \) for Fe MTJs expressed by a superposition of a short-period oscillation and a long-period oscillation described above. The period of 0.30 nm observed for CCFA MTJs is close to that of 0.32 nm for oscillations observed in Fe MTJs. The period of 0.30 nm is independent of the MgO(001) interplanar distance (0.21 nm), as in the case for Fe MTJs.

Notably large oscillation amplitudes for \( P \) and \( AP \), \( C_p \) and \( C_{AP} \), respectively, were observed for both 4.2 K and 293 K for CCFA MTJs. The values of \( C_p \) and \( C_{AP} \) obtained by fitting were 0.17 \( \pm 0.02 \) (\( C_p=0.154 \) and \( C_{AP}=0.193 \)) at 4.2 K and 0.16 \( \pm 0.01 \) (\( C_p=0.149 \) and \( C_{AP}=0.168 \)) at 293 K. Thus, we conclude that (1) the oscillation amplitudes \( C \) in the reduced tunnel resistances \( r_p \) and \( r_{AP} \) were almost independent of temperature, at least up to 293 K, and (2) there was no significant difference in \( C_p \) and \( C_{AP} \) though \( C_{AP} \) was 1.25 and 1.13 times larger than \( C_p \) at 4.2 and 293 K, respectively. Note that these values of \( C_p \) and \( C_{AP} \) are significantly large, and are about eight times those of about 0.02 observed for oscillations in \( r_p \) and \( r_{AP} \) for Fe MTJs at 20 K.

Note that in Fig. 3 there is no background variation in the \( r_p \) vs \( t_{MgO} \) and \( r_{AP} \) vs \( t_{MgO} \) plots from the oscillatory dependences expressed as \( r = 1 + C \cos[(2\pi/T)_{MgO} + \varphi] \) at both 4.2 and 293 K, which is a contrast to significant background variations in the \( r_p \) vs \( t_{MgO} \) and \( r_{AP} \) vs \( t_{MgO} \) plots observed for Fe MTJs.

This means that \( R_p \) (\( R_{AP} \)) is well approximated by the simple product of two terms without any corrections: the first is the baseline function \( \exp(at_{MgO} + b) \) and the second is the periodic function \( 1 + C \cos[(2\pi/T)_{MgO} + \varphi] \). Thus, \( R_p \) and \( R_{AP} \) are well approximated by

\[
  R_{AP} = \exp(at_{MgO} + b) [1 + C \cos[(2\pi/T)_{MgO} + \varphi]].
\]

at both 4.2 and 293 K [Figs. 2(a) and 2(b)]. This expression provides approximate slopes \( a_t = a - (2\pi/T)C \gamma \) and \( a_t = a + (2\pi/T)C \gamma \) for the gentle and steep slope regions, respectively, in the \( ln(R) \) vs \( t_{MgO} \) plots for both \( P \) and \( AP \). Here, \( \gamma \) is a numerical factor of about 0.78 associated with the approximation. Thus, the pronounced periodic dependences observed for \( ln(R) \) vs \( t_{MgO} \) Plots for \( P \) and \( AP \) [Figs. 2(a) and 2(b)] are due to the large \( C_p \) and \( C_{AP} \) values for CCFA MTJs.

Oscillations in \( r_p \) and \( r_{AP} \) showed phase differences \( \varphi_p - \varphi_{AP} \) of \( \sim 0.16 \) and 0.07 times the period of 0.30 nm at 4.2 and 293 K, respectively. This temperature dependence of \( \varphi_p - \varphi_{AP} \) was mainly caused by the temperature dependence of \( \varphi_p \).

Closed and open circles in Fig. 2(c) show the corresponding experimental TMR ratios defined as \( (R_p - R_{AP})/R_p \) as a function of \( t_{MgO} \) at 4.2 and 293 K calculated from the experimental \( R_p \) and \( R_{AP} \) values shown in Figs. 2(a) and 2(b), respectively. Because of the oscillations in \( R_p \) and \( R_{AP} \), the TMR ratios \( \Delta \) showed clear oscillations with typical peak and valley \( \alpha \) values of \( \alpha_{peaks} = 238\% \) and \( \alpha_{valleys} = 136\% \) at 4.2 K (\( \alpha_{peaks} = 1.057 \) or \( \alpha_{valleys} = 1.75 \)) and \( \alpha_{peaks} = 60\% \) and \( \alpha_{valleys} = 38\% \) at 293 K (\( \alpha_{peaks} = 1.063 \) or \( \alpha_{valleys} = 1.58 \)). The solid curves in Fig. 2(c) are the TMR ratios calculated using the approximate functions for \( r_p \) and \( r_{AP} \) as a function of \( t_{MgO} \) with Eq. (2). Of course, the curves thus calculated well reproduce the experimental TMR ratios. This is because the experimental TMR ratios and the curves were both basically derived from the same experimental \( R_p \) and \( R_{AP} \) values: the experimental TMR ratios were calculated using the experimental \( R_p \) and \( R_{AP} \) directly, while the curves were calculated using the approximate functions of experimental \( R_p \) and \( R_{AP} \) as a function of \( t_{MgO} \) with Eq. (2).

Figures 4(a) and 4(b) show \( r_p \) and \( r_{AP} \) as a function of \( t_{MgO} \) at 293 K for various positive \( V \) up to 0.2 V for a different set of CCFA MTJs from those shown in Figs. 2 and 3 but fabricated in the same preparation run on the same 20
times 20 mm MgO substrate. Clear oscillations in $r_p$ and $r_{AP}$ were observed for $V$ up to 0.2 V for both polarities. The solid lines in Fig. 4 are curves fitted using Eq. (1). The fitting results showed periods $T - 0.29$ nm $\geq 0.3\%$ for both P and AP for both polarities up to $\pm 0.2$ V, which were in good agreement with $T - 0.30$ nm $\pm 2\%$ observed for the MTJs shown in Fig. 3 (the slight difference in $T$ of 3.0% may suggest a small inhomogeneity in $t_{MGO}$ along the direction perpendicular to the wedge direction on a substrate). Figure 4(c) shows the oscillation amplitudes $C_P$ and $C_{AP}$ as a function of $V$ from $-0.2$ to 0.2 V, obtained by fitting, which demonstrates that $C_{AP}$ is slightly larger than $C_P$. Importantly, both $C_P$ and $C_{AP}$ showed weak dependence on $V$ up to $\pm 0.2$ V. However, in more detail, they decreased by 20% and 15% with increasing $V$ from $-0.2$ V to 0.2 V, respectively. This asymmetric dependence on the bias polarity suggests that the possible difference in the bulk states or interface states of the lower and upper CCFA electrodes affects the oscillation amplitude.

We also found clear oscillations in $R_P$ and $R_{AP}$ as a function of $t_{MGO}$ for identically fabricated CCFA/MgO/CoFe MTJs at 293 K [Fig. 5(a)]. The oscillations in the reduced resistances $r_p$ and $r_{AP}$ [Figs. 5(b) and 5(c)] were also well approximated by Eq. (1) with respective parameters and without any background correction, as in the case of CCFA MTJs. The periods, $T_P$ and $T_{AP}$, are also nearly equal and are 0.31 nm $\pm 2\%$. Thus, the periods were also close to those observed for CCFA MTJs and Fe MTJs. Note that the amplitudes, $C_P$ and $C_{AP}$, observed for CCFA/MgO/CoFe MTJs were 0.053 $\pm 12\%$. These values are about 1/3 of those observed for CCFA MTJs but still about three times those observed for Fe MTJs. Oscillations in $r_p$ and $r_{AP}$ showed a phase difference $\varphi_p - \varphi_{AP}$ of $-0.09$ times the period of 0.31 nm at 293 K for CCFA/MgO/CoFe MTJs. This phase difference is close to that of $-0.07$ times the period of 0.30 nm at 293 K for CCFA MTJs.

We will now discuss whether our experimental findings can be explained by the quantum interference of evanescent states in the MgO barrier proposed by Butler et al. This model takes into account quantum interference of two wave functions having respective complex perpendicular wave-vector components $k_z$ with finite real-part values $k_{z1} \neq k_{z2}$ and the same imaginary part $\kappa$, resulting in a transmittance given by $\exp(-2\kappa t_{MGO})$, a damped oscillatory function with respect to $t_{MGO}$. This function provides a period $T = 2\pi/(k_{z1} - k_{z2})$ for a given pair of $k_{z1} = k_{z1} + i\kappa$ and $k_{z2} = k_{z2} + i\kappa$. However, the model cannot provide a uniquely determined period for the tunneling current oscillations. In addition, our experimental finding of the constant period with respect to temperature and $V$ up to $\pm 0.2$ V for both P and AP is hard to attribute to the model proposed by Butler et al. In particular, the constant single period observed even for the increased $V$ up to $\pm 0.2$ V is hard to discuss on the basis of this model.
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Furthermore, this model considers only the quantum interference effect within the MgO barrier. However, the experimental findings clearly demonstrate that (1) the oscillation amplitude is strongly dependent on the choice of ferromagnetic electrode materials and (2) it is also influenced by the possible difference in the bulk states or interface states of the lower and upper electrodes. Therefore, the bulk and/or interface states should be taken into consideration along with the complex energy band in the MgO barrier. Although it is beyond the scope of the present study, the investigation of the electronic and magnetic states and microscopic structural properties for the interfacial regions between ferromagnetic electrodes and a MgO barrier is particularly important to reveal the origin of the observed pronounced oscillations. Our experimental findings that (1) the periods and amplitudes are almost identical for both P and AP, (2) the periods are almost independent of temperature and V, and (3) the oscillation amplitudes show only weak dependence on V suggest that all electrons contributing to tunneling are influenced by a certain effect, which varies periodically with $t_{\text{MgO}}$ with the period of $\sim 0.30$ nm.

IV. SUMMARY

In summary, we unambiguously demonstrated pronounced oscillations in $R_p$ and $R_{AP}$ with $t_{\text{MgO}}$ for fully epitaxial MgO-based MTJs with Heusler alloy Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al as both the lower and upper electrodes. The period of oscillations was almost independent of temperature up to 293 K and bias voltage $V$ up to 0.2 V and the amplitude showed only weak dependence on $V$ up to 0.2 V. These experimental findings should be a key to understand the origin of the oscillations.

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