Spin-dependent tunneling in epitaxial Fe/Cr/MgO/Fe magnetic tunnel junctions with an ultrathin Cr(001) spacer layer

Rie Matsumoto,[1,2,3] Akio Fukushima,[1] Kay Yakushiji,[1] Shingo Nishioka,[2] Taro Nagahama,[1] Toshikazu Katayama,[1] Yoshishige Suzuki,[1,2] Koji Ando,[1] and Shinji Yuasa[1,*]

[1]*National Institute of Advanced Industrial Science and Technology (AIST), Nanoelectronics Research Institute, Tsukuba, Ibaraki 305-8568, Japan*

[2]*Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan*

[3]*Japan Society for the Promotion of Science, Sumitomo-Ichibancho FS Bldg., 8 Ichibancho, Chiyoda-ku, Tokyo 102-8472, Japan*

(Received 1 October 2008; revised manuscript received 13 April 2009; published 29 May 2009)

We fabricated fully epitaxial Fe/Cr/MgO/Fe(001) magnetic tunnel junctions (MTJs) with an atomically flat ultrathin Cr(001) layer grown below the MgO barrier layer and studied the spin-dependent transport to clarify scattering process of tunneling electrons. Because Cr does not have Bloch states with Δ_1 symmetry at the Fermi energy (E_F), Δ_1 evanescent states in MgO, which dominantly mediate the tunneling current, cannot couple with Cr Bloch states without a scattering process. The Fe/Cr/MgO/Fe(001) MTJs are therefore a model system for studying nonspecular scattering processes where the orbital symmetry of tunneling states is not conserved. The resistance-area (RA) product of the MTJs was found to not increase exponentially as a function of the Cr thickness (t_{Cr}), indicating that the Cr layer does not act as a perfect tunnel barrier despite of the absence of Δ_1 states at E_F. Moreover, the magnetoresistance ratio of the MTJs was seen to oscillate as a function of t_{Cr} with a period of 2 monolayers, reflecting the layered antiferromagnetic structure of Cr(001). Surprisingly, the MR ratio showed local maxima at the odd numbers of Cr monolayers and local minima at the even numbers of Cr monolayers, indicating that the tunneling current is oppositely spin polarized with respect to the interface magnetization. These results suggest that nonspecular scatterings mediate the coupling between evanescent states in MgO and certain non-Δ_1 Bloch states in Cr that have negative spin polarization, thereby inducing nonspecular tunneling current even at a low temperature and a small bias voltage. We also investigated, as a reference sample, Fe/MgO/Cr/Fe MTJs with a less-oxidized Cr/MgO interface by growing the Cr(001) layer on the MgO barrier layer and found that their RA product increased much more rapidly with increasing t_{Cr}. This indicates that partial oxidation of interface Cr atoms in the Fe/Cr/MgO/Fe MTJs is one of the major origins of nonspecular scatterings. Both an increase in temperature and the application of bias voltage were found to greatly enhance the electron scattering that gives rise to tunneling conductance. While the temperature dependence of the antiparallel conductance due to magnon scatterings follows the Bloch $T^{3/2}$ law, the conductance due to nonspecular scatterings deviates from the Bloch $T^{3/2}$ law. The present experimental results give us important clues to a clearer understanding of the tunneling process in MgO-based MTJs.

DOI: 10.1103/PhysRevB.79.174436 PACS number(s): 73.40.Gk, 73.40.Rw, 85.75.Dd

I. INTRODUCTION

A magnetic tunnel junction (MTJ) consisting of a thin insulating layer (the tunnel barrier) sandwiched between two ferromagnetic electrode layers exhibits the tunnel magnetoresistance (TMR) effect due to the spin-dependent electron tunneling.[1] The TMR effect is evaluated by the magnetoresistance (MR) ratio defined as $(R_{AP}-R_P)/R_P\times 100(\%)$, where R_P and R_{AP} are the tunneling resistances when the magnetizations of the two electrodes are aligned in parallel (P) and antiparallel (AP) states. An amorphous aluminum oxide (Al-O) has been widely used as a tunneling barrier since the discovery of the TMR effect at room temperature (RT) in 1995 (Refs. 2 and 3), but an incoherent tunneling process in the Al-O-based MTJs keeps their MR ratios at RT below 100% and seriously limits their applications in devices such as magnetoresistive random access memories and the read heads of hard disk drives. The amorphous tunnel barrier also makes it difficult to understand the physics of the TMR effect. In the simple model of spin-dependent tunneling (Julliere's model), the tunneling probability is assumed to be the same for all the Bloch states in the electrodes, and the spin polarization of tunneling electrons is expressed as a function of spin-dependent density of states (DOS) of the ferromagnetic electrode material.[1] In many cases, however, even the sign of the spin polarization predicted by Julliere's model is inconsistent with experimental results[4] because the actual tunneling probability depends on the Bloch states and the detailed tunneling processes. A rigorous calculation of the spin polarization, however, is impossible because of the amorphous structure of the Al-O barrier.

In epitaxial MTJs with a crystalline tunnel barrier, on the other hand, a rigorous first-principles calculation is possible. First-principles theories have predicted that epitaxial Fe(001)/MgO(001)/Fe(001) MTJs with a crystalline MgO(001) barrier, which has a fourfold in-plane crystalline symmetry, will have MR ratios over 1000%.[5,6] In the band gap of MgO, there are three kinds of evanescent states—totally symmetric Δ_1 states $(s+p_z+d_{2z^2-x^2-y^2})$, Δ_5 states $(p_x+p_y+d_{xz}+d_{yz})$, and $\Delta_{2'}$ states $(d_{xy}+d_{y^2-z^2})$—and the decay of the density of states in a MgO barrier is slowest for the Δ_1 evanescent states. When the fourfold crystal symmetry is conserved throughout the MTJ, the Fe-Δ_1 Bloch states, which are fully spin polarized at the Fermi energy (E_F) (i.e., only the majority-spin Δ_1 states exist at E_F.) for the (001) direction ($k_{\parallel}=0$ direction), couple efficiently with those slowly decaying MgO-Δ_1 evanescent states, and a large tunneling current flows in the P state as a result of the dominant tunneling of the majority-spin Δ_1 states. Although a small tunneling current flows even in the AP state because of the so-called *hot-spot* tunneling, the AP conductance is much smaller than the P conductance. Epitaxial MgO-based MTJs are therefore expected to show a giant TMR effect. In 2004, giant RT MR ratios up to about 200% have been experimentally obtained in epitaxial Fe/MgO/Fe MTJs (Refs. 7 and 8) and textured FeCo/MgO/FeCo MTJs.[9] We have also achieved even higher MR ratios of 271% and 410% at RT in epitaxial Co/MgO/Fe and Co/MgO/Co MTJs, respectively.[10,11] The highest MR ratios of MgO-based MTJs reported to date is about 600% at RT and slightly above 1000% at low temperature.[12] These MR ratios, however, are still lower than theoretical values over 10 000% expected for FeCo and Co electrodes.[13] Clarifying the reason for this orders-of-magnitude discrepancy between the theoretical MR ratios and those obtained experimentally is of great importance with regard to not only applications but also fundamental physics.

The suppression of the actual MR ratio should be due to various scattering processes not taken into account in the conventional first-principles calculations, the most important of which are thought to be magnon scattering and nonspecular scattering. Magnon scattering decreases the MR ratio by increasing the tunneling conductance for the AP state. In the AP state, magnon scattering can couple a majority-spin Δ_1 state in one electrode with a majority-spin Δ_1 state in the other by flipping the spin of tunneling electron. An influence of magnon scattering is enhanced by increasing a bias voltage or a temperature. Magnon scatterings are known to yield a characteristic structure in a tunneling spectrum at low bias voltages below 100 mV.[14–16] They also give the tunneling conductance a temperature dependence that follows the Bloch $T^{3/2}$ law.[17] Magnon scattering is not likely to be the major origin of the suppressed TMR at low temperature and a low bias voltage, however, because it should have little effect in the ground state. On the other hand, nonspecular scattering processes such as impurity scattering and defect scattering were theoretically formulated by Zhang *et al.*[18] In the present paper, nonspecular scattering is defined as a scattering where the orbital symmetry of a tunneling state is not conserved. The scattering may or may not be accompanied with a scattering of in-plane momentum vector ($k_{\parallel}$). A nonspecular scattering can give rise to the conductance in the AP state by opening an additional conductance channel because nonsymmetry-conserving scattering can couple a majority-spin Δ_1 Bloch state in one electrode with a minority-spin Bloch states without Δ_1 symmetry in the other. Although there have been a few experimental reports on scattering processes in MgO barrier,[19] there have been no systematic experimental studies on the nonspecular scattering processes. Little experimentally obtained information about the influence of nonspecular scattering on transport properties at the ground state and their bias voltage and temperature dependences is available.

To clarify the basic nature of the nonspecular tunneling process experimentally, in this study we focused on Fe/Cr/MgO/Fe MTJs with an ultrathin Cr(001) electrode layer. The main reason we used Cr is its special band structure. The band structure of Cr is similar to that of the minority-spin Fe, especially in terms of the absence of Δ_1 states at E_F.[20] The tunneling conductance in Fe/Cr/MgO/Fe MTJs is therefore similar to the AP conductance in Fe/MgO/Fe MTJs. It should be noted that the scatterings suppress the MR ratio mainly by increasing the AP conductance. For studying the nonspecular tunneling process, the conductance in Fe/Cr/MgO/Fe MTJs is more suitable than the AP conductance in Fe/MgO/Fe MTJs. One reason is that in the AP state of Fe/MgO/Fe MTJs not only nonspecular scattering but also spin-flip scattering such as magnon scattering can open conductance channels and give rise to tunneling conductance. In Fe/Cr/MgO/Fe MTJs, on the other hand, magnon scattering cannot open additional conductance channels because there are no Cr-Δ_1 states at E_F unless a large momentum ($k_{\parallel}$) scattering takes place. The tunneling current in Fe/Cr/MgO/Fe MTJs is therefore considered to be largely nonspecular tunneling current. Another reason we used Cr is due to the layered antiferromagnetic (LAF) structure of Cr(001).[21] The magnetic moments of Cr(001) are ferromagnetically aligned within each monolayer (ML), and the magnetization of each atomic layer is aligned opposite to the magnetization of the adjacent layers. This LAF structure of Cr(001) enables us to investigate the role of the electrode/barrier interface in tunneling conductance.

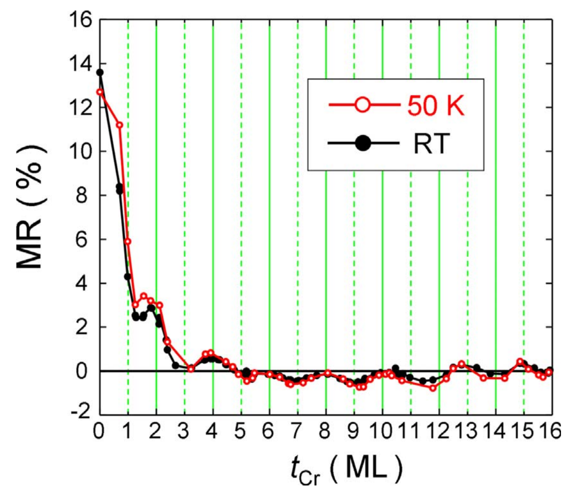

FIG. 1. (Color online) Dependence of MR ratio on Cr thickness (t_{Cr}) at 50 K (open circles) and room temperature (solid circles) for Fe/Cr/Al-O/FeCo MTJs [data from Ref. 20].

In a previous study, we used Fe/Cr/Al-O/FeCo MTJs with a conventional amorphous Al-O barrier. We expected the strongly disordered interface between crystalline Cr(001) and amorphous Al-O to enhance not only the nonspecular scatterings but also scatterings of the momentum ($k_{\parallel}$) of tunneling electrons, and we indeed observed evidence of strong momentum scatterings at the Cr/Al-O interface.[20] As seen in Fig. 1, the Fe/Al-O/FeCo MTJs [namely, the Fe/Cr/Al-O/FeCo MTJs with a Cr thickness (t_{Cr}) of 0 ML] have a MR ratio of 14%. When a Cr layer was inserted at the interface, the MR ratio rapidly decreased and was almost zero at t_{Cr} of 3 ML. This indicates that not only the orbital symmetry but also the momentum of tunneling electrons is strongly scattered at the disordered Cr/Al-O interface. When t_{Cr} was increased further, the small MR ratio oscillated as a function of t_{Cr} with a period of 2 ML. The MTJs with an even number of Cr MLs have a positive MR and those with an even number have a negative one. Here we define P and AP states according to the magnetic configurations of the top and bottom ferromagnetic layers (Fe or FeCo). It should be noted that the magnetization of the Cr monolayer at Cr/Al-O interface is parallel with that of the underlying Fe layer when the MTJ has an even number of Cr MLs and is antiparallel when the MTJ has an odd number of Cr MLs. Therefore, the results indicate that the MR ratio sensitively reflects the spin polarization of the interfacial Cr monolayer adjacent to the amorphous Al-O layer. It is therefore very meaningful to compare the interface scattering at the Cr/Al-O interface with that at the epitaxial Cr/MgO interface. Greullet *et al.* recently studied the tunneling properties in epitaxial Fe/Cr/MgO/Fe MTJs and reported that the Cr layer acts as a tunnel barrier because of the absence of Δ_1 states at the E_F in Cr.[22] In this paper, we report more systematic experimental results and a different interpretation of the tunneling mechanism in Fe/Cr/MgO/Fe MTJs and discuss the tunneling process from the viewpoint of the nonspecular scatterings.

II. EXPERIMENTAL

We fabricated a high-quality fully epitaxial Fe(001)/Cr(001)/MgO(001)/Fe(001) MTJ film with a wedge-shaped Cr layer by using molecular-beam epitaxy (MBE) growth and highly precise microfabrication techniques. The cross section of the film is shown schematically in Fig. 2(a). We optimized the growth conditions of Cr on Fe to ensure its layer-by-layer growth. An appropriate growth temperature is needed not only for layer-by-layer growth but also for preventing interdiffusion at the Fe/Cr interface. So to find the optimum growth temperature for Cr, we first fabricated Fe/Cr/Fe trilayer films [Fig. 3(a)] at various growth temperatures and plotted their coercive fields as a function of t_{Cr} [Fig. 3(c)]. The coercive fields of the upper Fe layer are strongly influenced by interlayer exchange coupling (IEC) energy J_s. In Fig. 3(c) a short-period IEC oscillation with a period of $t_{Cr}=2$ ML can clearly be seen, which is evidence of layer-by-layer growth of Cr(001) with little interdiffusion at the Fe/Cr interface. Although the optimum growth temperature for the Cr layer was reported by Pierce *et al.* to be 250 °C,[21] we found it to be 120 °C. This difference may be due to errors in calibrating the substrate temperature and/or to the different underlying Fe layers used in the two studies. In this study, the bottom Fe layer was grown on an MgO(001) substrate, while in the work reported by Pierce *et al.* a Fe(001) whisker substrate was used. We used the 120 °C growth temperature to fabricate the Fe/Cr/MgO/Fe MTJ film on an MgO(001) substrate in the present study because an Fe whisker substrate is too small for microfabrication processes. It should also be noted that the growth temperature used in the present study was the same as that used in our previous study of Fe/Cr/Al-O/FeCo MTJs,[20] although in this study we used a different MBE growth chamber. The growth conditions for the Fe and MgO layers were the same as in our previous studies.[7,23]

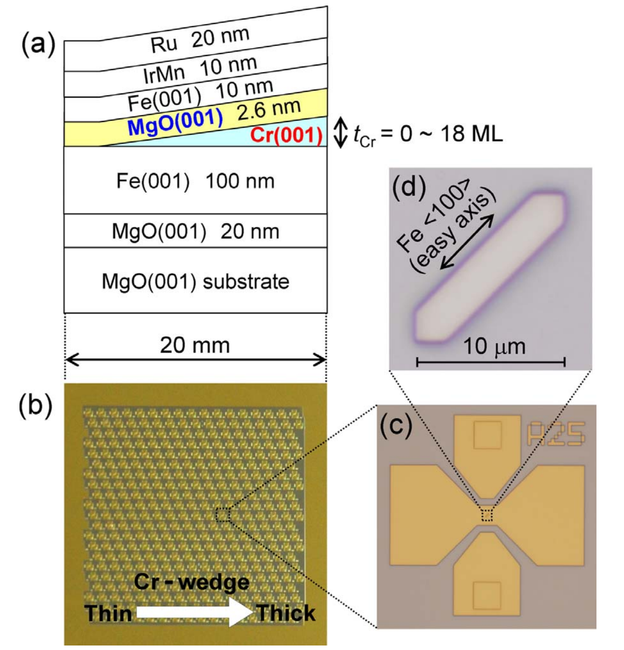

FIG. 2. (Color online) (a) Cross section of epitaxial Fe/Cr/MgO/Fe(001) MTJ film with wedge-shaped Cr(001) layer. [(b)–(d)] Top-view photographs of Fe/Cr/MgO/Fe MTJs prepared with highly precise microfabrication techniques: (b) overall view of all MTJs fabricated on identical substrate, (c) closeup of one MTJ with contact pads, and (d) optical microscopic image of MTJ with a junction area of 36 μm^2.

We also prepared, as a reference sample, a Fe/MgO/Cr/Fe MTJ film in which a wedge-shaped Cr(001) layer was grown on the MgO barrier layer. The Cr in this sample was not grown layer-by-layer because Cr grows three dimensionally on MgO. It is noted that Cr does not wet the MgO(001) surface very well in the initial growth stage. Here, the Cr and upper Fe layers were grown at room temperature to suppress roughness and interdiffusion as much as possible. Then, the film was postannealed at 200 °C. It should be noted that the qualities of the Cr/MgO interface also seemed to differ between Fe/Cr/MgO/Fe MTJ and Fe/MgO/Cr/Fe MTJ films. This point is discussed later.

The MTJ films were fabricated into MTJs with a junction area (A) of 36 μm^2 [Fig. 2(d)] by using the highly precise microfabrication process [Figs. 2(b)–2(d)] we used in a previous study.[23] Because the wedge-shaped Cr layers were grown on the same substrate, the relative experimental error in t_{Cr} between MTJs in the same lot was negligibly small. The dependence of the MR ratio, R_P and R_{AP} on t_{Cr}, and the bias-voltage dependence and temperature dependence were systematically measured with the dc four-probe method. Unless noted otherwise, in this paper positive bias means that electrons flow from the top electrode to the bottom electrode. This definition is the same as that in the report of our previous study of Fe/Cr/Al-O/Fe MTJs.[20]

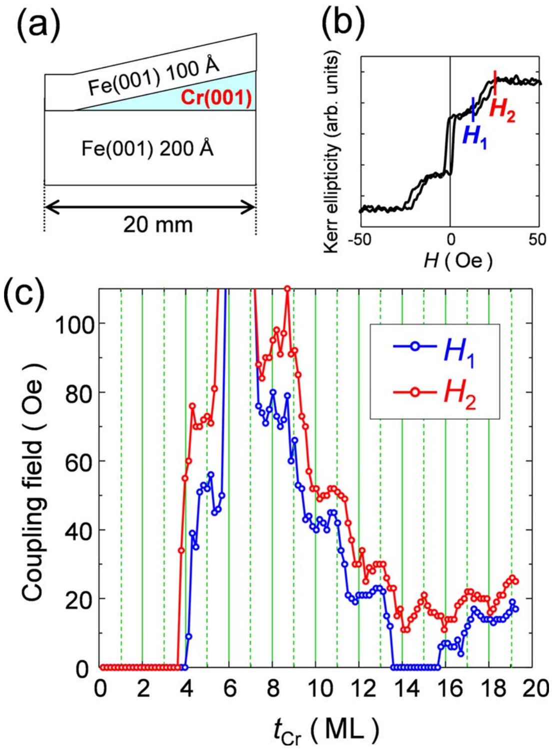

FIG. 3. (Color online) (a) Cross section of epitaxial Fe/Cr/Fe(001) trilayer film with wedge-shaped Cr(001) layer grown at 120 °C on a substrate and (b) its magnetic hysteresis loop at Cr thickness (t_{Cr}) of 13.6 MLs where the interlayer exchange coupling between Fe layers is antiferromagnetic. Magnetic hysteresis loop was measured by longitudinal magneto-optical Kerr effect at room temperature. (c) Coupling fields (H_1 and H_2) of the upper Fe layer of the structure shown in (b) as a function of t_{Cr}.

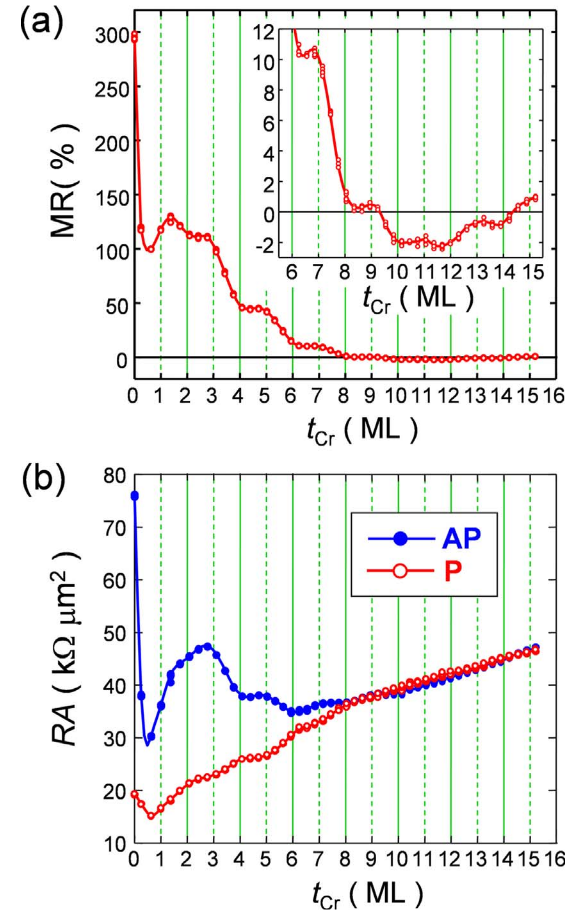

FIG. 4. (Color online) Cr thickness (t_{Cr}) dependence of (a) MR ratio and (b) RA product in parallel magnetic state (P state) (open circles) and antiparallel magnetic state (AP state) (solid circles) at 20 K for epitaxial Fe/Cr/MgO/Fe(001) MTJs.

III. RESULTS AND DISCUSSION

A. Cr thickness dependence of tunneling transport

We first report experimental results on the Fe/Cr/MgO/Fe MTJs, in which the Cr layer was grown on the bottom Fe layer. The t_{Cr} dependence of the MR ratio measured at 20 K and a bias voltage of −10 mV is plotted in Fig. 4(a). The MR ratio oscillates as a function of t_{Cr} with a period of t_{Cr} =2 ML for $t_{Cr} \geq 2$ ML, reflecting the LAF structure of Cr(001). In two respects, however, this oscillation differs substantially from that observed for the Fe/Cr/Al-O/FeCo MTJs (Fig. 1). The first is the phase of the oscillation: the MR ratio of the Fe/Cr/MgO/Fe MTJs has a local maxima at the odd numbers of Cr MLs and local minima at the even numbers, while that of the Fe/Cr/Al-O/FeCo MTJs has local maxima at the even numbers of Cr MLs and local minima at the odd numbers. The second difference is the decay length of the MR ratio in the thin-Cr region: the MR ratio of the Fe/Cr/MgO/Fe MTJs decays relatively slowly and nearly reaches zero around t_{Cr}=8 ML, while that of the Fe/Cr/Al-O/FeCo MTJs decays much more rapidly and nearly reaches zero at t_{Cr}=3 ML. These differences indicate that the electron-scattering process at the Cr/MgO interface differs from that at the Cr/Al-O interface. The slow decay of the MR ratio in the Fe/Cr/MgO/Fe MTJs indicates that the momentum scattering at the Cr/MgO interface is much weaker than that at the Cr/Al-O interface. An important question here is whether the electron transport through the Cr layer is tunneling or metallic. A good criterion to determine the transport mechanism is the dependence of junction resistance on t_{Cr}. If the electron transport through the Cr layer is perfectly tunneling, the tunneling resistance should increase exponentially with respect to t_{Cr}, which was not demonstrated experimentally by Greullet *et al.*[22] The resistance-area (RA) products for the P and AP states (R_PA and $R_{AP}A$) are plotted against t_{Cr} in Fig. 4(b). A 2-ML-period of RA-product oscillation, which corresponds to the TMR oscillation, was observed for both P and AP states. Each RA product also increases with t_{Cr}, but the increase is much slower than an exponential increase. This indicates that Cr does not act as a perfect tunnel barrier but acts as a metallic layer where a nonspecular current flows. It is considered that the Cr layer acts as a tunnel barrier for a specular current mediated by Δ_1 electrons, while it acts as a metallic layer for a nonspecular current mediated by non-Δ_1 electrons. Because the specular current decays exponentially with respect to the Cr thickness, the nonspecular current should be dominant over the specular current. As a result, the transport through the Cr layer becomes almost metallic. Judging from these results, we can conclude that the evanescent states in MgO couple with certain Cr-Bloch states that do not have Δ_1 symmetry. These Bloch states have a negative spin polarization at the Cr/MgO interface because the MR ratio has local maxima at the odd numbers of Cr MLs and local minima at the even numbers. The slow decay of the MR ratio for the thin-Cr region indicates that there is less momentum scattering at this interface than there is at the interface with an amorphous Al-O barrier. For $t_{Cr}<2$ ML, the t_{Cr} dependences of the MR ratio and RA products do not show 2-ML-period oscillation but instead show irregular behavior (Fig. 4). A similar irregular t_{Cr} dependence for $t_{Cr}<2$ ML is commonly observed in Fe/Cr/Fe trilayer systems and is said to be due to the interdiffusion at the Cr/Fe interface.[21] Thus, the irregular t_{Cr} dependence observed in Fe/Cr/MgO/Fe MTJs is attributed to this extrinsic origin.

It should be noted that a certain scattering process is necessary for the nonspecular current to flow. According to first-principles calculations,[22,24] the Cr layer of Fe/Cr/MgO/Fe MTJs with an ideal structure should act as a nearly perfect tunnel barrier because of the absence of Δ_1 Bloch states at E_F in Cr. Therefore, the nonspecular scattering in the ground state is considered to be induced mainly by structural defects that break the fourfold symmetry at the barrier/electrode interfaces. In the present MTJs, the most probable structural defect is the partial oxidation of interface Cr atoms. Partial oxidation of the interface has been shown theoretically to significantly influence tunneling transport.[25] The Cr surface has a strong getter effect on oxygen atoms and molecules, and it was exposed to an oxygen partial pressure of 1–2 $\times 10^{-7}$ Pa for more than 1 min when MgO was evaporated for deposition.[8] We therefore cannot exclude the possibility of partial oxidation of the interface Cr atoms. We investigated this possibility by measuring the tunneling transport of the Fe/MgO/Cr/Fe reference sample, in which an ultrathin Cr(001) layer was grown on the MgO(001) barrier [inset in Fig. 5(a)]. Because the Cr layer in this sample was deposited on a clean MgO surface under an ultrahigh vacuum (pressure $<5\times 10^{-8}$ Pa), the interface Cr atoms should have been less oxidized than those in the Fe/Cr/MgO/Fe MTJs. The t_{Cr} dependence of the MR ratio measured at 20 K for epitaxial Fe/MgO/Cr/Fe MTJs is plotted in Fig. 5(a). The 2-ML-period oscillation in MR ratio was not observed in the Fe/MgO/Cr/Fe reference sample. This is simply because the growth mode of Cr on MgO was not the layer-by-layer one. One sees in Fig. 5(b) that inserting a 20-ML-thick Cr layer increases the RA product by a factor of about 15. This rate of increase is much more rapid than that observed in the Fe/Cr/MgO/Fe MTJs. This difference between the Fe/Cr/MgO/Fe and Fe/MgO/Cr/Fe MTJs clearly indicates that partial oxidation of interface Cr atoms in the Fe/Cr/MgO/Fe MTJs is one of the major origins of the nonspecular scatterings. The scattering process is discussed more in detail in the following sections. It should be noted here that the scattering process should be sensitive to the details of structural defects. Therefore, MTJ samples having different types of structural defects may show different scattering process.

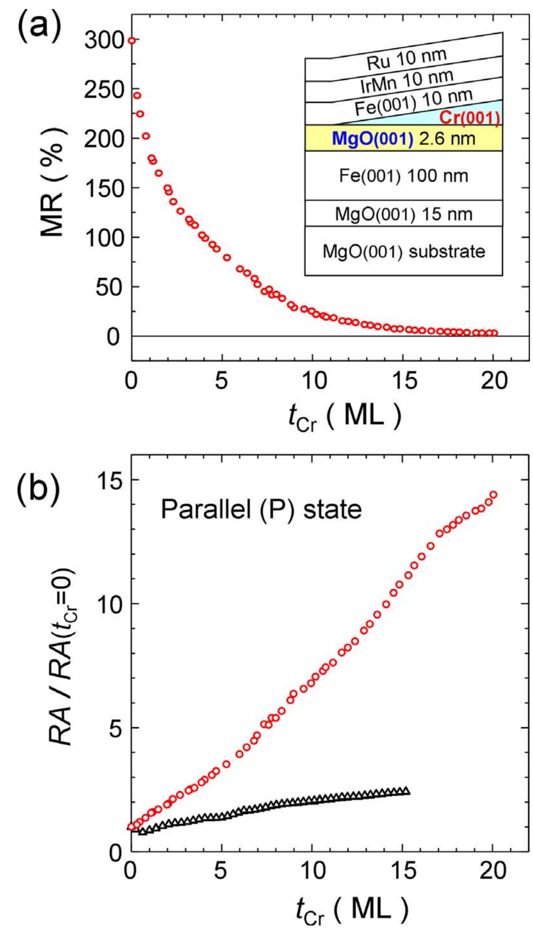

FIG. 5. (Color online) Cr thickness (t_{Cr}) dependence of (a) MR ratio and (b) normalized RA product in P state at 20 K for epitaxial Fe/MgO/Cr/Fe MTJs (open circles), in which a wedge-shaped Cr(001) layer was grown on the MgO(001) barrier layer. The inset in (a) schematically shows the cross section of the MTJs. RA product is normalized by the value at $t_{Cr}=0$ and the triangles in (b) show the normalized RA product for epitaxial Fe/Cr/MgO/Fe MTJs with the Cr layer grown under the MgO barrier layer [data from Fig. 4(b)].

B. Temperature dependence of tunneling transport

Scattering processes are generally reflected in the temperature and bias-voltage dependences of tunneling properties. To investigate the scattering process that induces nonspecular tunneling current, we measured the temperature and the bias-voltage dependences of tunneling conductance for the P and AP states. First, let us look at the temperature dependence. The dependence of the RA product on temperature is a good index for studying the scattering mechanisms.[17,18] The dependence of the R_PA and $R_{AP}A$ on temperature is plotted in Figs. 6(a) and 6(b) for Fe/Cr/MgO/Fe MTJs with various t_{Cr}. Each plotted RA product is normalized by that at 20 K. In the Fe/MgO/Fe MTJs ($t_{Cr}=0$ ML) for the P state, the RA product is nearly independent of temperature, which is a characteristic feature commonly observed in MgO-based MTJs. In the Fe/MgO/Fe MTJs for the AP state and Fe/Cr/MgO/Fe MTJs, on the other hand, the RA product decreases with temperature. For the P state, the rate of decrease increases with increasing t_{Cr} and saturates at around $t_{Cr}=8$ ML. It should be noted that $t_{Cr}=8$ ML corresponds to a thickness where the MR ratio is nearly zero [Fig. 4(a)], indicating that the dependence of R_PA on temperature

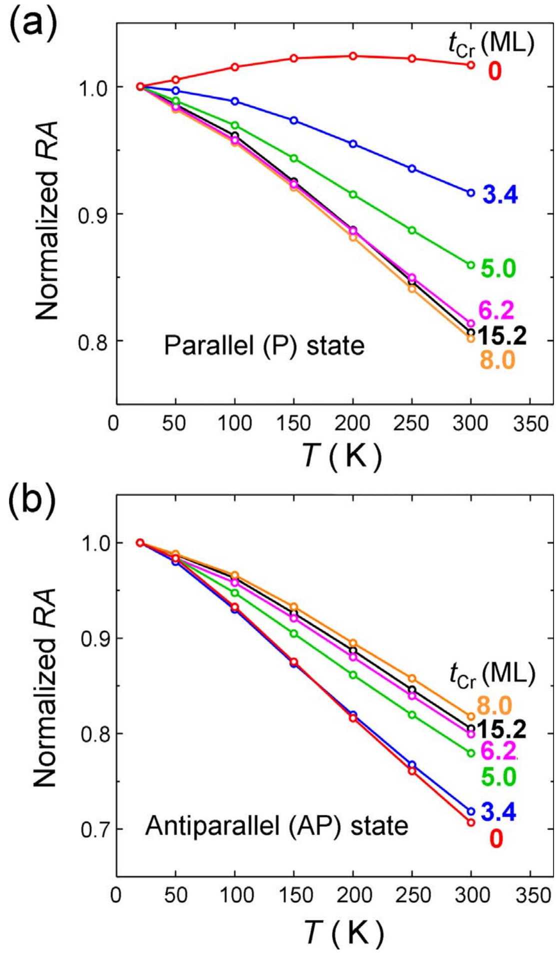

FIG. 6. (Color online) Dependence of RA product (R measured at bias voltage of -10 mV) on temperature (T) in (a) P state and (b) AP state for epitaxial Fe/Cr/MgO/Fe(001) MTJs with various Cr thicknesses (t_{Cr}). For each t_{Cr}, the RA product is normalized by that at 20 K.

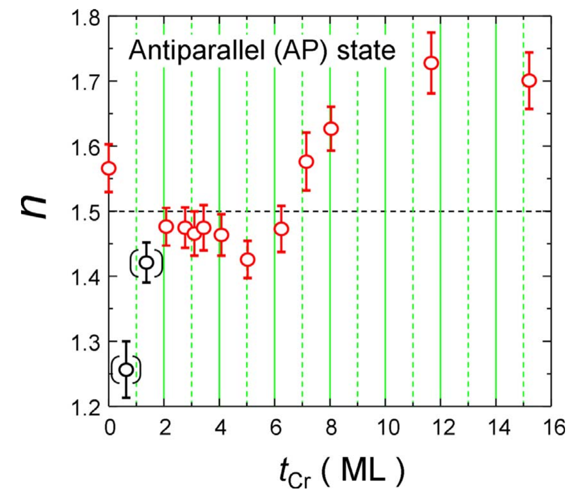

FIG. 7. (Color online) Cr thickness (t_{Cr}) dependence of exponent n in $\Delta G_{AP}(T)=C\bullet T^n$. Error bars indicate standard deviation (σ) in least-squares fitting.

is closely related to electron scattering in the Cr layer. Thermal spin fluctuations are thought to enhance scattering, opening additional conductance channels.

Here we discuss the scattering process more in detail by analyzing the temperature dependence of AP conductance: $\Delta G_{AP}(T)=G_{AP}(T)-G_{AP}(\sim 0\ \text{K})$. If the contribution of bulk magnon scattering is dominant, $\Delta G_{AP}(T)$ should be fitted to $C\bullet T^{3/2}$.[17] Here C is a coefficient. To find out whether or not the bulk magnon scattering is dominant, we fitted the $\Delta G_{AP}(T)$ for each t_{Cr} to a power-law form ($C\bullet T^n$) using the least-squares method. The dependence of the exponent n on t_{Cr} is plotted in Fig. 7. In the case of Fe/MgO/Fe ($t_{Cr}=0$ ML) and Fe/Cr/MgO/Fe with $t_{Cr}=2-7$ ML, $\Delta G_{AP}(T)$ was fitted relatively well to $C\bullet T^{3/2}$, indicating that magnon scattering is dominant. The plots for $0<t_{Cr}<2$ ML should be excluded from the discussion here because of the extrinsic issue of interdiffusion. For $t_{Cr}>7$ ML, on the other hand, the exponent n gradually deviates from 3/2 and seems to settle around 1.7. This indicates that the ferromagnetic magnon scattering is less dominant for the thicker Cr region. It should be noted that $t_{Cr}\sim 7$ ML where this deviation occurs approximately corresponds to $t_{Cr}\sim 8$ ML where the MR ratio goes nearly to zero [Fig. 4(a)]. Because for $t_{Cr}<7-8$ ML some of the tunneling electrons are transported between the two Fe layers coherently, the influence of ferromagnetic magnon scatterings is considered to be significant. Therefore the temperature dependence follows the Bloch $T^{3/2}$ law. For $t_{Cr}>7-8$ ML, on the other hand, the nonspecular tunneling current becomes dominant and the temperature dependence therefore deviates from the $T^{3/2}$ law.

C. Bias-voltage dependence of tunneling transport

In this section, let us look first at the bias-voltage dependence of second-derivative conductance (d^2I/dV^2) (i.e., the second-derivative tunneling spectrum) at 20 K for Fe/MgO/Fe MTJs [Figs. 8(a) and 8(b)] and Fe/Cr/MgO/Fe MTJs with $t_{Cr}=10$ ML [Figs. 8(c) and 8(d)]. The second-derivative tunneling spectrum contains information on the DOS structure[16] and inelastic-scattering processes in the downstream electrode.[14,15] In Fig. 8, the spectra are shown mainly for one bias polarity to focus the discussion on the scattering process at the Cr/MgO interface. The spectrum of Fe/MgO/Fe for the AP state has a large peak near 1.0 V, while the d^2I/dV^2 spectra of Fe/Cr/MgO/Fe for both the P and AP states have a faint peak near 0.9 V. These peaks are considered to reflect the bottom edge of the Fe minority-spin Δ_1 band in Fe (Ref. 16) and that of the Δ_1 band in Cr (Ref. 20) because specular tunneling current mediated by Δ_1 states begins to flow above these bias voltages. Because Cr is antiferromagnetic, there are peaks for both the P and AP states. In addition to these peaks, a large peak is seen at low bias voltages in the spectrum of Fe/MgO/Fe for the AP state and in the spectrum of Fe/Cr/MgO/Fe for both the P and AP states. The low-bias-voltage peak in the spectrum of Fe/MgO/Fe is attributed to magnon scattering enhanced by an application of bias voltage because it is seen only for the AP state.[14–16] In the spectrum of Fe/Cr/MgO/Fe, an even larger peak is seen near 50 mV for both the P and AP states, indicating that the scattering mechanism is independent of spins. This low-bias-voltage peak is considered to be due to the

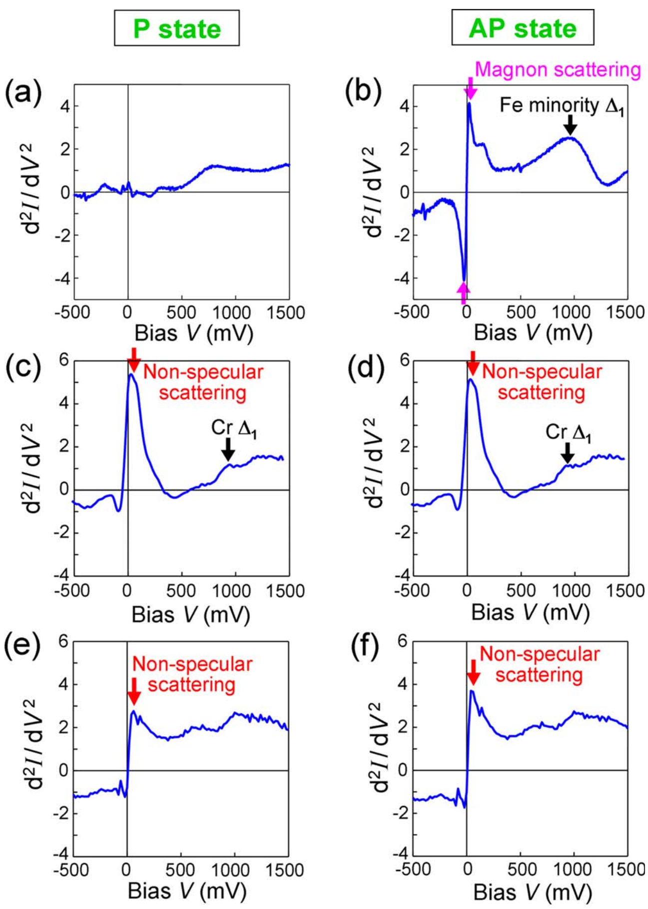

FIG. 8. (Color online) Bias voltage (V) dependence of second-derivative conductance (d^2I/dV^2) in P state (left-hand side) and AP state (right-hand side) for (a), (b) Fe/MgO/Fe MTJs and for (c), (d) Fe/Cr/MgO/Fe MTJs with $t_{Cr}=10$ ML and (e), and (f) Fe/MgO/Cr/Fe MTJs with $t_{Cr}=10$ ML. The vertical axis is the normalized second-derivative conductance $(d^2I/dV^2)/(dI/dV)$ in units of (A/V). In (a) and (b), the spectra were measured at 3 K with the conventional lock-in method and positive bias means that electrons tunnel from the top Fe layer to the bottom Fe layer. In [(c)–(f)], d^2I/dV^2-V characteristics were derived mathematically from the data of the I-V characteristics measured at every 20 mV measured at 20 K and positive bias means that electrons tunnel from the Fe layer to the Cr layer.

nonspecular scattering process, which mediates the coupling between MgO-Δ_1 evanescent states and certain Cr-Bloch states (including interface resonant states) because the tunneling current for this thick MgO barrier is predominantly carried by the MgO-Δ_1 evanescent states. There are two possible explanations for the peak position of 50 mV. One is that the nonspecular scattering is enhanced for hot electrons at 50 meV above E_F. The other is that the interface resonant states with non-Δ_1 symmetry have a peak of DOS 50 meV above E_F. In either case, an application of bias voltage of about 50 mV enhances the scattering and opens additional nonspecular tunneling channels, resulting in an increase in tunneling conductance.

We also measured the tunneling spectra for the Fe/MgO/Cr/Fe reference sample, and the second-derivative tunneling spectra at 20 K for Fe/MgO/Cr/Fe MTJs with $t_{Cr}=10$ ML are shown in Figs. 8(e) and 8(f). The low-bias peak is much smaller than that in Figs. 8(c) and 8(d). This is consistent with the fact that the nonspecular tunneling process in Fe/MgO/Cr/Fe MTJs having a cleaner Cr/MgO interface is weaker than that in Fe/Cr/MgO/Fe MTJs having a partially oxidized Cr/MgO interface.

IV. SUMMARY

Our experimental results on the epitaxial Fe/Cr/MgO/Fe MTJs show that their RA product does not increase exponentially with increasing t_{Cr}. This means that the Cr layer does not act as a perfect tunnel barrier despite the absence of Δ_1 states at E_F. Even at a low temperature and a small bias voltage, a nonspecular scattering process mediates the coupling between MgO-Δ_1 states and certain non-Δ_1 Cr-Bloch states with negative spin polarization (i.e., the tunneling current is oppositely spin polarized with respect to the interface magnetization), which induces nonspecular tunneling current. Comparison between the Fe/Cr/MgO/Fe MTJs and the Fe/MgO/Cr/Fe reference MTJs having a cleaner interface indicates that partial oxidation of interface Cr atoms in the Fe/Cr/MgO/Fe MTJs is one of the major origins of the nonspecular scatterings. Both an increase in temperature and the application of bias voltage enhance the electron scattering, opening additional conductance channels and giving rise to tunneling conductance. While the temperature dependence of the AP conductance in the Fe/MgO/Fe MTJs follows the Bloch $T^{3/2}$ law because of magnon scattering, the temperature dependence in the Fe/Cr/MgO/Fe MTJs deviates from the Bloch $T^{3/2}$ law as a result of nonspecular scattering. Bias voltages above about 50 mV greatly enhance the nonspecular scattering process.

Although the details of scattering process at the Cr/MgO interface are not clear at the present stage, we obtained various important clues. For example, the Cr-Bloch states that couple with MgO-Δ_1 states have negative spin polarization. Comparison between the present results and first-principles calculations on the MTJs with interface defects should clarify the details of nonspecular tunneling process in MgO-based MTJs. Because for the AP state the tunneling in Fe/Cr/MgO/Fe MTJs is similar to that in Fe/MgO/Fe MTJs, a clearer understanding of the tunneling process in Fe/Cr/MgO/Fe MTJs should provide us with clues to how we could control the AP conductance in Fe/MgO/Fe MTJs and obtain a higher TMR effect.

ACKNOWLEDGMENTS

This work was supported in part by NEDO spintronics nonvolatile functionality project and JSPS KAKENHI (Grant No. 19·01159). We would like to thank M. Yamamoto for her help with microfabricating the samples.

*Corresponding author; yuasa-s@aist.go.jp

[1] M. Jullière, Phys. Lett. **54A**, 225 (1975).

[2] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).

[3] T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).

[4] S. Yuasa and D. D. Djayaprawira, J. Phys. D **40**, R337 (2007).

[5] W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, Phys. Rev. B **63**, 054416 (2001).

[6] J. Mathon and A. Umerski, Phys. Rev. B **63**, 220403(R) (2001).

[7] S. Yuasa, A. Fukushima, T. Nagahama, K. Ando, and Y. Suzuki, Jpn. J. Appl. Phys., Part 2 **43**, L588 (2004).

[8] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nature Mater. **3**, 868 (2004).

[9] S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang, Nature Mater. **3**, 862 (2004).

[10] S. Yuasa, T. Katayama, T. Nagahama, A. Fukushima, H. Kubota, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **87**, 222508 (2005).

[11] S. Yuasa, A. Fukushima, H. Kubota, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **89**, 042505 (2006).

[12] S. Ikeda, J. Hayakawa, Y. Ashizawa, Y. M. Lee, K. Miura, H. Hasegawa, M. Tsunoda, F. Matsukura, and H. Ohno, Appl. Phys. Lett. **93**, 082508 (2008).

[13] X.-G. Zhang and W. H. Butler, Phys. Rev. B **70**, 172407 (2004).

[14] J. S. Moodera, J. Nowak, and R. J. M. van de Veerdonk, Phys. Rev. Lett. **80**, 2941 (1998).

[15] J. Murai, Y. Ando, M. Kamijo, H. Kubota, and T. Miyazaki, Jpn. J. Appl. Phys., Part 2 **38**, L1106 (1999).

[16] Y. Ando, T. Miyakoshi, M. Oogane, T. Miyazaki, H. Kubota, K. Ando, and S. Yuasa, Appl. Phys. Lett. **87**, 142502 (2005); S. Nishioka, R. Matsumoto, H. Tomita, T. Nozaki, Y. Suzuki, H. Itoh, and S. Yuasa, *ibid.* **93**, 122511 (2008).

[17] S. G. Wang, R. C. C. Ward, G. X. Du, X. F. Han, C. Wang, and A. Kohn, Phys. Rev. B **78**, 180411(R) (2008).

[18] X.-G. Zhang, Y. Wang, and X. F. Han, Phys. Rev. B **77**, 144431 (2008).

[19] G. X. Miao, Y. J. Park, J. S. Moodera, M. Seibt, G. Eilers, and M. Münzenberg, Phys. Rev. Lett. **100**, 246803 (2008).

[20] T. Nagahama, S. Yuasa, E. Tamura, and Y. Suzuki, Phys. Rev. Lett. **95**, 086602 (2005).

[21] D. T. Pierce, J. Unguris, R. J. Celotta, and M. D. Stiles, J. Magn. Magn. Mater. **200**, 290 (1999).

[22] F. Greullet, C. Tiusan, F. Montaigne, M. Hehn, D. Halley, O. Bengone, M. Bowen, and W. Weber, Phys. Rev. Lett. **99**, 187202 (2007).

[23] R. Matsumoto, A. Fukushima, T. Nagahama, Y. Suzuki, K. Ando, and S. Yuasa, Appl. Phys. Lett. **90**, 252506 (2007).

[24] I. Mertig (private communication).

[25] X.-G. Zhang, W. H. Butler, and A. Bandyopadhyay, Phys. Rev. B **68**, 092402 (2003).