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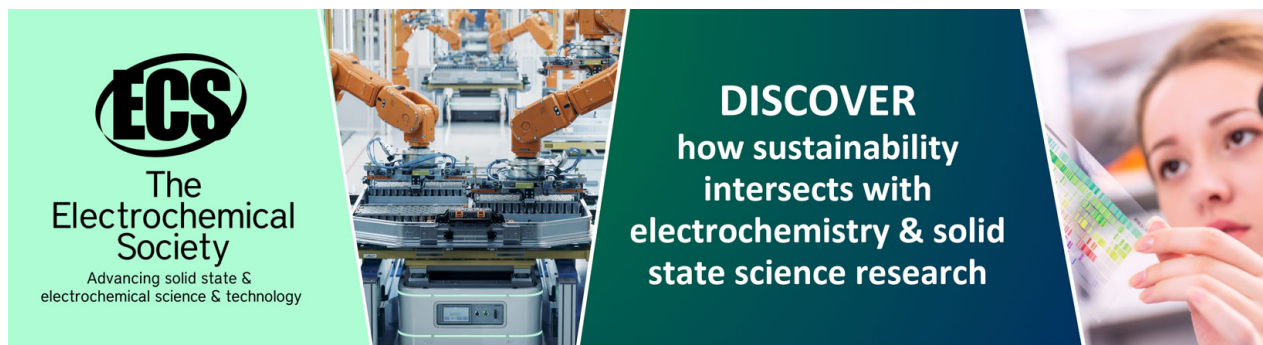
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A large quantum-well oscillation of the TMR effect

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Abstract

Magnetic tunnel junctions (MTJs) with ultrathin single-crystal electrodes were prepared, and the influence of the quantum-well (QW) states formed in the electrodes on the tunnel magnetoresistance (TMR) effect was studied. MTJs with an ultrathin Fe(001) electrode showed oscillations in the conductivity and the TMR ratio in tunnel spectra. MTJs with an ultrathin non-magnetic Cu(001) layer inserted between an Al–O tunnel barrier and a ferromagnetic Co(001) electrode showed an oscillation in the TMR as a function of the Cu-layer thickness. The period of the TMR oscillation coincides with the oscillation period of interlayer exchange coupling in a Co(001)/Cu(001)/Co(001) film, indicating that the TMR oscillation originates from the spin-polarized QW states in Cu(001). The amplitude of the oscillation was so large that even the sign of the TMR ratio alternated.

1. Introduction

Since the discovery of the giant magnetoresistance (GMR) effect [1, 2], the interactions between the electric charge and spin of conduction electrons have been utilized in solid-state devices made of metals, semiconductors, and oxides. A new field of electronics called spin-electronics has thus emerged [3]. The tunnel magnetoresistance (TMR) effect [4, 5] in a magnetic tunnel junction (MTJ), which consists of two ferromagnetic (FM) metal layers (electrodes) separated by a thin insulating layer (a tunnel barrier), is especially important because of the large magnetoresistance at room temperature and the application for magnetoresistive random-access-memory (MRAM). However, the mechanism of the TMR effect is still not very clearly understood.

The TMR effect is known to originate from the spin polarization of the electrodes, i.e. the difference between the densities of states (DOSs) of the up- and down-spin sub-bands in the two electrodes. According to Julliere's simplified

model [6], the TMR ratio is expressed as

$$\text{TMR ratio} = \frac{R_a - R_p}{R_p} = \frac{2P_1P_2}{1 - P_1P_2}, \quad (1)$$
$$P_{1(2)} = \frac{D_{1(2)\uparrow} - D_{1(2)\downarrow}}{D_{1(2)\uparrow} + D_{1(2)\downarrow}},$$

where R_p and R_a are the tunnel resistances when the magnetizations of the two FM electrodes are aligned parallel and antiparallel, respectively; and P_1 and P_2 are the spin polarizations of the two electrodes; and $D_{1(2)\uparrow}$ and $D_{1(2)\downarrow}$ are the DOSs of the electrodes at the Fermi level for the up-spin and down-spin sub-bands, respectively.

The TMR effect is also known to be sensitive to the barrier/electrode interface. De Teresa *et al* [7] studied MTJs with various barrier materials and found that even the sign of the TMR depends on the barrier materials. On the other hand, in our previous study [8], we investigated the effect of crystal anisotropy of the spin polarization by using MTJs with single-crystal Fe electrodes of various crystal orientations. We found a clear crystal-orientation dependence of the TMR, which might reflect the crystal anisotropy of the electronic

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states in the electrodes. Based on these findings, the spin polarization is apparently not an inherent property of the electrode material. It is also unclear how the coherence of the wave functions and the spins of the conduction electrons are conserved in the transport process. For example, the resonant tunnelling effect [9], in which the coherence of the electron wave functions is essential, has hardly been observed in spin-polarized systems. Spin-polarized resonant tunnelling is crucial in developing highly functional devices such as a resonant tunnelling spin-transistor [10].

To shed light on these problems, a study of the quantum-size effect in the TMR effect would be valuable. If bulk electronic states have some influence on the TMR effect, the TMR could be controlled by using the quantum-size effect to modulate the electronic states of the electrodes. When one of the two electrodes is made of an ultrathin single-crystal FM layer (figures 1(a) and (c)), the up-spin electrons would be confined in the ultrathin FM layer. If the coherence of the electron wave functions is conserved, the electrons can form spin-polarized quantum-well (QW) states, and the TMR effect can be modulated by the QW states.

On the other hand, when an ultrathin non-magnetic (NM) layer is inserted between the tunnel barrier and one of the FM electrodes (figures 1(b) and (d)), the down-spin electrons are confined in the NM layer and form spin-polarized QW states. Many researchers have studied this structure [11–18]. Theory predicts that spin-polarized resonant tunnelling will cause the TMR effect to oscillate as a function of the NM-layer thickness [17, 18]. However, no clear QW oscillations have been observed experimentally [11–16]. The TMR ratio either decreases monotonically with respect to the NM-layer thickness [11] or shows a small modulation and sign reversal [12–16]. These behaviours could be attributed to the nature of polycrystalline electrodes, in which hardly any QW states are formed. An ultrathin single-crystal layer is essential for the formation of well-defined QW states. In this paper, we

investigated the QW effect of TMR by fabricating MTJs with ultrathin single-crystal FM and NM electrodes. We obtained clear QW oscillations of the TMR effect due to spin-polarized resonant tunnelling.

2. Experimental

We prepared two kinds of film structures for MTJs (figures 2(a) and (b)). The films were prepared by molecular beam epitaxy (MBE) in an ultra-high vacuum chamber with a base pressure better than 2×10^{-10} Torr [8]. The single-crystal buffer layers and lower electrodes were formed by MBE on single-crystal MgO(001) substrates. The epitaxial growth was monitored by *in situ* RHEED observation. The amorphous Al–O tunnel barrier was formed by reactive deposition or natural oxidation in pure O₂ gas. The polycrystalline upper electrodes were deposited without exposure to the air.

Figure 2(a) shows a Fe(001)/Al–O/Fe₅₀Co₅₀ film structure. The lower electrode (an ultrathin single-crystal Fe(001) layer) was grown on Cr(001). The thickness of the Fe(001) layer was varied from 2 to 9 ML (monolayers) across the wafer by using a shutter moving linearly during the deposition of Fe.

Figure 2(b) shows MTJs with Co(001)/Cu(001)/Al–O/Ni₈₀Fe₂₀ film structure. The lower electrode (an ultrathin single-crystal Cu(001) layer) was grown on ferromagnetic Co(001). The thickness of the Cu(001) layer was varied from 0 to 29 Å across the wafer by the same method as above.

RHEED images for the lower electrodes of Fe(001) and Cu(001) are shown in figures 3(a) and (b), respectively. The streak patterns with specular reflection spots indicate that bcc Fe(001) and fcc Cu(001) structures were successfully grown with atomically flat single-crystal layers. These films were subsequently fabricated into tunnelling junctions with areas ranging from $2 \times 2 \mu\text{m}^2$ to $3 \times 10 \mu\text{m}^2$ by using various micro-fabrication processes (photolithography, Ar ion etching, SiO₂ sputtering deposition, lift-off, etc). An antiparallel magnetic configuration could easily be induced with the appropriate magnetic field by using different coercive fields in the upper and lower electrodes. In addition to these MTJs, we also prepared a Co(001)/Cu(001)/Co(001) film structure

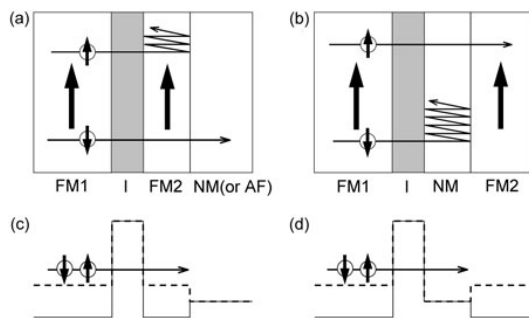


Figure 1. Schematic diagrams of MTJs with (a) a FM1/I/FM2(ultrathin)/NM(or AF) structure and (b) a FM1/I/NM(ultrathin)/FM2 structure, in which electrons with different spin directions tunnel from the left electrode to the right electrode. Here, FM1 and FM2 denote ferromagnetic layers, while I, NM, and AF denote an insulating layer (a tunnel barrier), a nonmagnetic layer and an antiferromagnetic layer, respectively. (c) and (d) represent schematic illustrations of the potential energy for the MTJs shown in (a) and (b), respectively. Here the solid lines are the potentials for up-spin electrons, while the broken lines are the potentials for down-spin electrons.

(a)	(b)	(c)
Au-cap	Au-cap	Au-cap
Fe ₅₀ Co ₅₀ 200 Å	Ni ₈₀ Fe ₂₀ 100 Å	
FM	FM	
Al–O 17 Å	Al–O 12 Å	Co(001) 50 Å
I	I	I
Fe(001) 2 ~ 9 ML	Cu(001) 0 ~ 32 Å	Cu(001) 0 ~ 45 Å
FM	NM	NM
Cr(001) 200 Å	Co(001) 200 Å	Co(001) 50 Å
	FM	FM
Buffer layers	Buffer layers	Buffer layers
MgO(001) substrate	MgO(001) substrate	MgO(001) substrate

Figure 2. (a) Film structure for an MTJ with an ultrathin single-crystal Fe(001) electrode. 1 ML of Fe(001) corresponds to 1.43 Å. (b) Film structure of the film for an MTJ with an ultrathin single-crystal Cu(001) electrode. (c) Interlayer exchange-coupled film structure.

(figure 2(c)) by the same methods. We investigated the interlayer exchange coupling through the Cu(001) layer. This interlayer exchange coupling is known to oscillate as a function of the Cu-layer thickness because of the QW states formed at the Fermi level in Cu [19].

To investigate the quantum size effect in TMR, we used tunnel spectroscopy and made the magnetoresistance measurements. The tunnel spectroscopy measurement is more sensitive than the usual I - V measurement. By using tunnel spectroscopy, we measured the bias dependence of the conductivity of the MTJs. According to [20], the derivative of an I - V curve (the differential conductivity) is expressed as

$$\frac{dI}{dV} = \left(\frac{2\pi e^2}{\hbar} \right) |t|^2 D_1(E_F) D_2(E_F + eV), \quad (2)$$

where $|t|^2$ is the tunnel probability, D_1 and D_2 are the DOSs of the two electrodes, E_F is the Fermi energy, and V is the bias voltage applied to electrode 2. By changing the bias voltage V , we can examine the features of the unoccupied DOS in electrode 2. The electric resistance was measured by the DC 4-probe method with a bias voltage of up to 700 mV. The bias voltage direction was defined with respect to the lower (single-crystal) electrode. The magnetoresistance measurements were carried out at 2 and 300 K in magnetic fields of up to 2 T. For the Fe(001)(2–7 ML)/Al–O/FeCo MTJs, the tunnel spectra were measured at 2 K by the conventional lock-in method, using an I - V converter for the differential conductivity and a bridge circuit for the second derivative of the I - V curve [21]. For the Co(001)/Cu(001)/Co(001) MTJs, the magnetization curves were measured at room temperature by making longitudinal magneto-optical Kerr effect (MOKE) measurements in magnetic fields of up to 1 kOe, with the field applied parallel to the film.

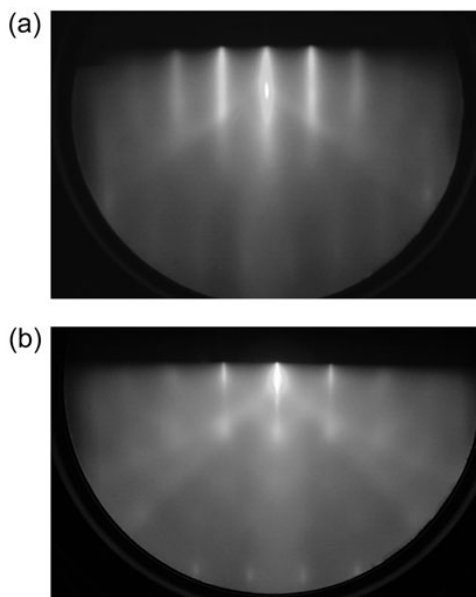


Figure 3. RHEED images for (a) an ultrathin bcc-Fe(001) layer ([110] azimuth) and (b) an ultrathin fcc-Cu(001) layer ([100] azimuth).

3. Results and discussion

3.1. MTJ with an ultrathin Fe(001) electrode: Fe(001)/Al–O/FeCo

The TMR ratio was measured as a function of the Fe-layer thickness, but no oscillation was observed probably. The oscillation amplitude may have been small compared to the accuracy of the measurements. On the other hand, the tunnelling spectra showed clear oscillatory behaviour. Figure 4(a) shows the differential conductivities, dI/dV , for typical Fe thicknesses. The parabolic background curve was subtracted from the raw data by using Simmons' I - V formula [22]. The differential conductivity clearly oscillates as a function of the bias voltage, and the phase of the oscillation shifts as the Fe-layer thickness is varied. This behaviour is characteristic of the QW effect [23, 24].

We should note that these oscillations were observed only for positive bias voltage. When a positive bias voltage is applied, electrons flow into the unoccupied states in the ultrathin Fe electrode. Therefore, the oscillations indicate that the electronic states in the electrode are modulated by the quantum-size effect. On the other hand, no oscillations were observed for negative bias voltage. This is because the FeCo layer is polycrystalline and 200 Å thick, so the QW states do not form in the FeCo electrodes. The differential MR, which is the magnetoresistance ratio obtained by using the differential resistance with the background (raw data), is shown in figure 4(b). The differential MR is expressed by

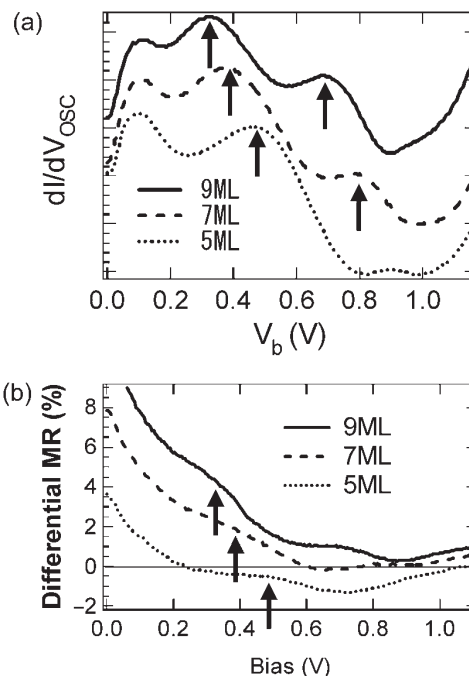


Figure 4. (a) Bias dependence of the oscillation component of conductivity (dI/dV_{osc}) for Fe(100)(n ML)/Al–O(17 Å)/FeCo. (b) Bias dependence of the differential TMR ratio for Fe(100)(n ML)/Al–O(17 Å)/FeCo. The peaks indicated by the arrows are located at the same bias voltages in both plots.

using the DOSs of the electrodes:

$$\text{Differential MR} = \frac{(dI/dV_{\uparrow\downarrow})^{-1} - (dI/dV_{\uparrow\uparrow})^{-1}}{(dI/dV_{\uparrow\downarrow})^{-1}} = \frac{(D_{1\uparrow} - D_{1\downarrow})(D_{2\uparrow} - D_{2\downarrow})}{D_{1\uparrow}D_{2\uparrow} + D_{1\downarrow}D_{2\downarrow}}. \quad (3)$$

This formula corresponds to Julliere's formula for conventional TMR. In figure 4(b), the differential MR shows clear oscillations only for positive bias voltage, and the phases of the oscillations are the same as those of the conductivity oscillations. The TMR oscillations originate from the difference in amplitude of the conductivity oscillations between the parallel magnetic configuration and the antiparallel configuration. These results clearly indicate that the TMR effect is modulated by the spin-polarized QW states in the ultrathin Fe(001) electrode.

According to band structure calculations [25], the Fe minority band structure is very similar to the band structure of Cr, and consequently, the QW states are formed only in the Fe majority sub-band. However, simple calculations of QW states based on the bulk bcc-Fe band dispersion fail to quantitatively explain the observed oscillations. More advanced calculation of the ultrathin Fe electronic structure and realistic transport calculation are thus necessary.

3.2. MTJ with an ultrathin Cu(001) electrode: Co(001)/Cu(001)/Al–O/NiFe

Figure 5 shows the TMR ratio at 300 K and a low bias voltage of +10 mV as a function of the Cu(001)-layer thickness, t_{Cu} . The TMR ratio clearly oscillates up to a t_{Cu} of 32 Å. We should note that the amplitude of the TMR oscillation is so large that even the sign of the TMR ratio alternates. The TMR oscillation can be fitted well to a damped oscillation function plus a small positive background curve (a simple damping curve). The period of the TMR oscillation is 11 Å. On the other hand, the tunnel resistance (R_p) itself also shows a slight oscillatory

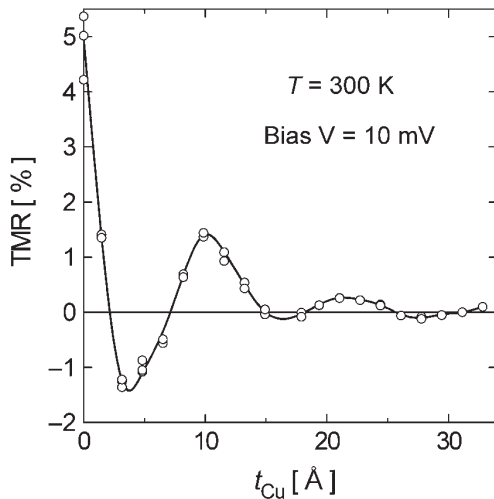


Figure 5. TMR ratio at 300 K and a bias voltage of +10 mV as a function of the Cu-layer thickness (t_{Cu}) for Co(001)/Cu(001)(0 ~ 32 Å)/Al–O(12 Å)/Ni–Fe junctions.

behaviour, although it is not as clear as the TMR oscillation. The tunnel resistance takes a maximum at $t_{\text{Cu}} = 4$ Å, where the TMR ratio takes a minimum (negative) value.

We now consider the origin of the TMR oscillation. Spin-dependent reflection of conduction electrons occurs at the Co/Cu interface. Tunnelling electrons with a spin parallel to the magnetization of the Co (up-spin) are easily transmitted into the Co layer, while those with a spin antiparallel to the magnetization of the Co (down-spin) have a higher probability of being reflected at the Co/Cu interface. Therefore, the down-spin electrons will form QW states in the Cu layer. Two kinds of QW states, corresponding to the scattering vectors q_1 and q_2 , are known to be formed in ultrathin Cu(001) (figure 6). According to band calculations, q_1 and q_2 are responsible for QW oscillations with periods of 10.6 Å and 5.9 Å, respectively [19]. Such oscillations are observed in the interlayer exchange coupling through the Cu(001) layer, although the short-period oscillation due to q_2 disappears easily because of the roughness of the films [19, 23, 26]. The observed period of the TMR oscillation (11 Å) agrees with the long-period oscillation due to q_1 .

On the other hand, the interlayer exchange coupling in a Co(001)/Cu(001)/Co(001) film prepared for this study also shows a QW oscillation with a period of 11 Å (figure 7). The maximums of H_s indicate that the interlayer exchange coupling is antiferromagnetic. The oscillation period of the interlayer exchange coupling agrees well with that of the TMR oscillation. This clearly suggests that both the TMR and the exchange coupling oscillations originate from the same QW states corresponding to q_1 . We thus successfully observed the QW oscillation of the TMR effect, although other researchers observed no oscillatory behaviour with a polycrystalline Cu layer [11, 13–15].

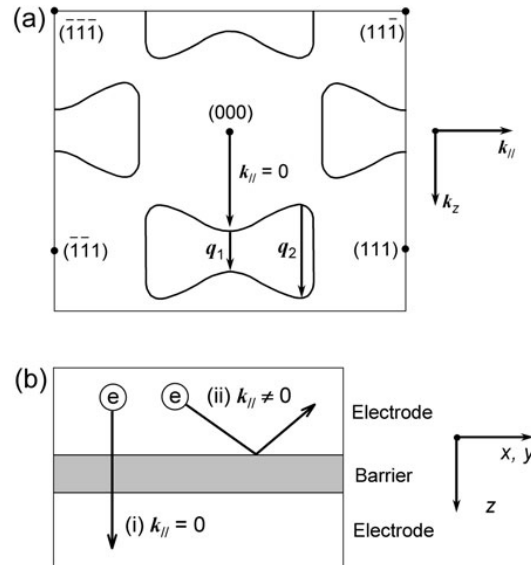


Figure 6. (a) Fermi surface of fcc Cu. When conduction electrons are confined in the [001] direction (z -direction), QW states with scattering vectors q_1 and q_2 can be formed [19]. (b) Schematic diagram of a tunnel junction, in which an electron with $k_{\parallel} = 0$ tunnels through the barrier while an electron with $k_{\parallel} \neq 0$ is reflected at the barrier.

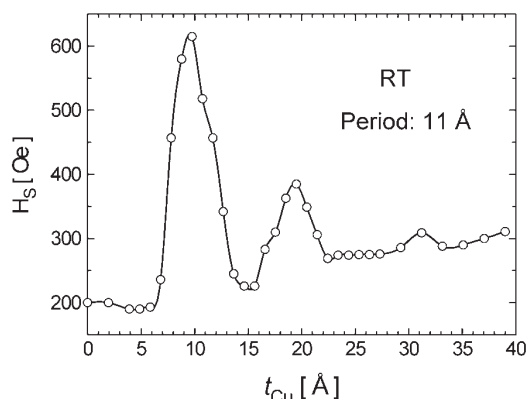


Figure 7. Saturation magnetic field (H_s) for a Co(001)/Cu(001)/Co(001) interlayer exchange-coupled film structure as a function of the Cu-layer thickness (t_{Cu}). H_s was determined from longitudinal MOKE hysteresis loops obtained at room temperature with the magnetic field applied parallel to the film.

We note here that the conduction electrons with a wave vector $\mathbf{k}$ normal to the tunnel barrier ($k_{\parallel} = 0$) are the main contributors to the tunnelling current in an ideal tunnel junction (figure 6(b)). This is because the tunnelling probability sharply decreases as the direction of $\mathbf{k}$ deviates from the barrier normal [8, 27]. As a consequence, the tunnelling electrons can be efficiently injected into the QW states located at $k_{\parallel} = 0$, i.e. the QW states of q_1 . The negative (inverse) TMR is considered to originate from the spin polarization of Cu(001), because the spin polarizations of Ni-Fe and Co are positive [28]. This may indicate that the QW states are formed in the down-spin sub-bands and have a negative spin polarization.

It should also be mentioned that the amplitude of the TMR oscillation in the Co(001)/Cu(001)/Al-O/NiFe junctions was much larger than that observed in the Fe(001)/Al-O/FeCo junctions. The difference can be attributed to the mean free path and spin diffusion length of the conduction electrons, which are both much longer in NM noble metals such as Cu than in FM metals such as Fe. Therefore, noble metals are superior for the formation of QW states.

4. Conclusions

We have studied MTJs with ultrathin single-crystal Fe(100) and Cu(001) electrodes. For the Fe(001)/Al-O/FeCo junctions, small but clear QW oscillations were observed in both the bias dependence of the conductivity and the TMR ratio. We have not yet identified the Fe sub-band in which the QW states are formed. For the Co(001)/Cu(001)/Al-O/NiFe junctions, the TMR ratio oscillates as a function

of the Cu-layer thickness up to 32 Å. The oscillation period agrees with that of the interlayer exchange coupling in Co(001)/Cu(001)/Co(001), suggesting that both the TMR oscillation and the interlayer exchange coupling oscillation originate from the same spin-polarized QW states in Cu(001). The amplitude of the TMR oscillation is so large that even the sign of the TMR ratio alternates, which may indicate a negative spin polarization of the QW states in the Cu(001) electrode.

Acknowledgments

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