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Express Letter

High Tunnel Magnetoresistance at Room Temperature in Fully Epitaxial Fe/MgO/Fe Tunnel Junctions due to Coherent Spin-Polarized Tunneling

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We fabricated fully epitaxial Fe(001)/MgO(001)/Fe(001) magnetic tunnel junctions (MTJs) and observed a magnetoresistance (MR) ratio of 88% at $T = 293$ K (146% at $T = 20$ K), the highest value yet reported. The origin of the high MR ratio is not the diffusive tunneling of Julliere's model but the coherent spin-polarized tunneling in epitaxial MTJs, in which only the electrons with totally symmetric wave functions with respect to the barrier-normal axis can tunnel. The bias-voltage dependence of the MR was very small, resulting in a high output voltage of 380 mV. This high voltage will help overcome problems in the development of high-density magnetoresistive random-access-memory (MRAM).
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KEYWORDS: tunnel magnetoresistance, TMR effect, magnetic tunnel junction, MRAM, epitaxial growth

Magnetic tunnel junctions (MTJs) consisting of two ferromagnetic electrodes separated by an insulating layer (tunnel barrier) exhibit magnetoresistance at room temperature.[1,2] This phenomenon, known as the tunnel magnetoresistance (TMR) effect, has applications for magnetoresistive random-access-memory (MRAM) and magnetic sensors.[3] MRAM is expected to be a unified memory device having desirable properties such as non-volatility, high speed, and high density. A new field of electronics that makes use of both the electric charge and spin of conduction electrons is called *spintronics*; it has developed rapidly over the last few years. The magnetoresistance (MR) ratio of an MTJ is defined as $\Delta R/R_p = (R_{ap} - R_p)/R_p$, where R_p and R_{ap} are the junction resistance when the magnetizations of the two electrodes are aligned parallel and antiparallel, respectively. According to Julliere's formulation,[4] the MR ratio at a low bias-voltage limit is expressed as

$$\text{MR ratio} = (R_{ap} - R_p)/R_p = 2P_1P_2/(1 - P_1P_2),$$
$$\text{where } P_\alpha = (D_{\alpha\uparrow}(E_F) - D_{\alpha\downarrow}(E_F))/(D_{\alpha\uparrow}(E_F) + D_{\alpha\downarrow}(E_F)),$$
$$\alpha = 1, 2. \qquad (1)$$

Here, P_α is the spin polarization of the electrodes, and $D_{\alpha\uparrow}(E_F)$ and $D_{\alpha\downarrow}(E_F)$ are the densities of states (DOS) at the Fermi energy (E_F) for the majority and minority spin bands, respectively. Since the spin polarization of ferromagnetic transitional metals and alloys is observed to be about 0.5 or less, Julliere's formula gives the highest expected MR ratio, about 70%. MTJs are usually fabricated using an amorphous Al-O tunnel barrier and poly-crystalline electrodes, and a magnetoresistance (MR) ratio as high as about 70% at room temperature has been reported.[5] This high MR ratio makes it difficult to obtain an output voltage higher than 200 mV, which limits the maximum capacity of the MRAM to about 64 Mb. This strictly limits potential applications of MRAM. Because the MR ratio of current MTJs is approaching the theoretical limit for transition metals, novel materials or methods are needed to achieve Gb-scale MRAMs, which require a much higher MR ratio and output voltage. While novel electrode materials with $P = 1$ (so-called *half metals*) have been synthesized in an effort to obtain a higher MR ratio, such highly spin-polarized materials have not yet been fabricated at room temperature.

Another way to obtain a higher MR ratio is to use fully epitaxial MTJs, in which coherent electron tunneling should occur. *Ab-initio* calculations for fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs have shown the likelihood of an extremely high MR ratio.[6,7] The mechanism of such a high MR ratio differs from that of Julliere's model. When the momentum of conduction electrons is conserved during tunneling, the tunneling current is dominated by the electrons with momentum vectors normal to the tunnel barrier ([001] direction for the Fe(001) electrode).[8] Based on the band structure of Fe in the [001] (Γ-H) direction (Fig. 1), the DOS at E_F is not highly polarized because both the majority and minority spin sub-bands have states at E_F. However, when the coherency of the electron wave functions is conserved during tunneling, only the conduction electrons with totally symmetric wave functions with respect to the barrier-normal axis are connected to the states in the

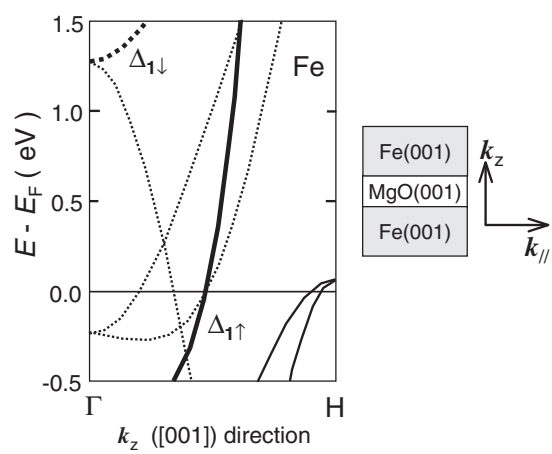

Fig. 1. Band dispersion of Fe in [001] (Γ-H) direction. Solid and dotted lines represent majority-spin and minority-spin sub-bands, respectively. The thick solid and dotted lines represent majority-spin and minority-spin Δ_1 bands, respectively.

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barrier region and have a finite tunneling probability. For the Fe(001) electrode, the Δ_1 band has totally symmetric characteristics. As shown in Fig. 1, the majority spin Δ_1 band exists at E_F while the minority spin Δ_1 band has no states at E_F. Because of this *half-metallic* feature of the Fe Δ_1 bands, an extremely high MR ratio should occur in coherent spin-polarized tunneling.

Fully epitaxial (single-crystal) MTJs are ideal for achieving coherent tunneling because electron scattering during tunneling can be suppressed. Coherent spin-polarized tunneling using a high-quality single-crystal Co/Cu electrode has been demonstrated.[9] There have been several reports on the fabrication of epitaxial Fe(001)/MgO(001)/Fe(001) MTJs,[10–13] and a relatively high MR ratio of 67% at room temperature has been reported.[12] However, the MR ratios observed for Fe(001)/MgO(001)/Fe(001) are still lower than the best values reported for conventional MTJs with an Al-O tunnel barrier.[5] In this letter, we report the highest MR ratio (88%) yet observed at room temperature for fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs; it was achieved apparently due to the coherent spin-polarized tunneling.

A high-quality single-crystal Fe(001) bottom electrode was prepared using molecular beam epitaxy. An MgO(001) seed layer was first deposited on a single-crystal MgO(001) substrate to improve the morphology of the substrate surface. A 500-Å-thick epitaxial Fe(001) bottom electrode was subsequently grown at room temperature followed by annealing at 350°C in an ultra-high vacuum (2×10^{-8} Pa). A RHEED image of the Fe surface (Fig. 2(a)) showed good crystallinity and flatness of the epitaxial Fe(001). Then, a 20-Å-thick MgO(001) barrier layer was epitaxially grown on the Fe(001) electrode at room temperature using electron beam evaporation of MgO. The RHEED pattern (Fig. 2(b)) showed high-quality epitaxial growth of the MgO(001) layer.

A high-quality single-crystal Fe(001) upper electrode (100 Å) was epitaxially grown on the MgO(001) tunnel barrier at room temperature, and a 100-Å-thick Co layer was subsequently deposited on the upper Fe electrode to obtain an antiparallel magnetic configuration by enhancing the coercive field of the upper electrode.

The sample was then patterned into tunnel junctions using micro-fabrication techniques (photolithography, Ar ion milling, etc.). We fabricated 160 MTJs with a junction size of $3 \times 12\,\mu$m on each substrate.

Typical magnetoresistance curves of the Fe(001)/MgO(001)/Fe(001) MTJs are shown in Fig. 3. MR ratios of 88% and 146% were observed at $T = 293$ and 20 K, respectively. To our knowledge, this is the highest MR ratios observed at room temperature. They cannot be explained by the spin polarization of the Fe(001) electrode,[8] so they must be due to the coherent spin-polarized tunneling. The variations in the MR ratio and junction resistance between MTJs fabricated on the same substrate were smaller than ±15%, and the yield rate of the MTJs was better than 90%. It should be noted that the resistance-area product (RA) of the MTJs was a few k$\Omega\cdot\mu$m^2, which is suitable for MRAM devices.

The bias-voltage dependence of the MR ratio at room temperature is shown in Fig. 4(a). Surprisingly, the bias-dependence was considerably small and asymmetric despite the symmetric structure of the Fe(001)/MgO(001)/Fe(001) MTJs. This asymmetry is possibly due to the interface structure. If the upper and lower Fe(001)/MgO interfaces have some asymmetry in their structure, asymmetric bias dependence may result. We cannot determine if this is the case because the detailed interface structure is still unclear. The bias voltage where the MR ratio reaches half the zero-bias value (V_{half}) was 1250 and 390 mV for the positive and negative bias directions, respectively. The 1250 mV is the highest V_{half} yet reported. Typical V_{half} values for MTJs with an Al-O tunnel barrier range from 300 to 600 mV.

The output voltage, V_{out} ($\equiv$ bias $V \times (R_{ap} - R_p)/R_{ap}$)), of

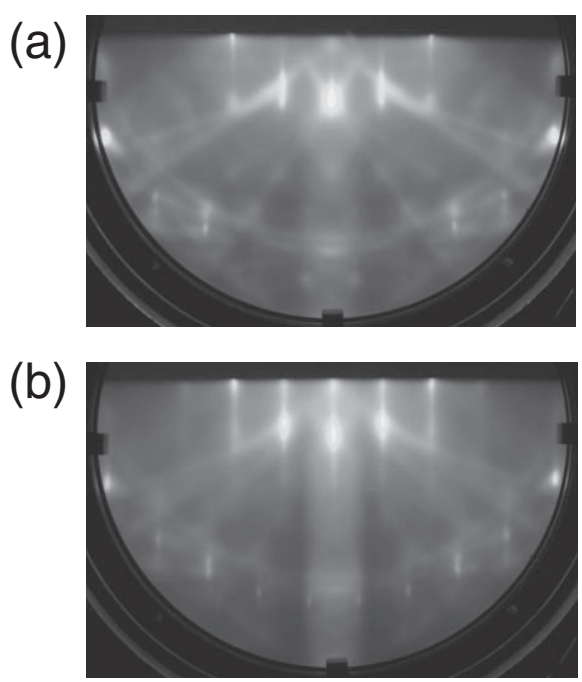

Fig. 2. RHEED images of (a) Fe(001) bottom electrode layer ([110] azimuth) and (b) MgO(001) tunnel barrier layer ([100] azimuth).

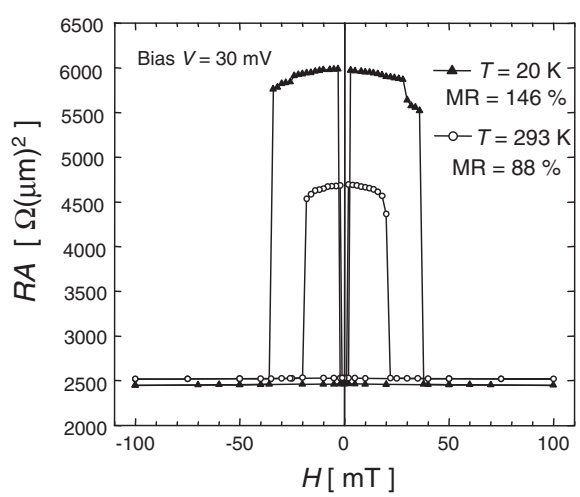

Fig. 3. Magnetoresistance curves for Fe(001)/MgO(001)(20 Å)/Fe(001) MTJ at $T = 293$ and 20 K. The MR ratios were 88% and 146%, respectively.

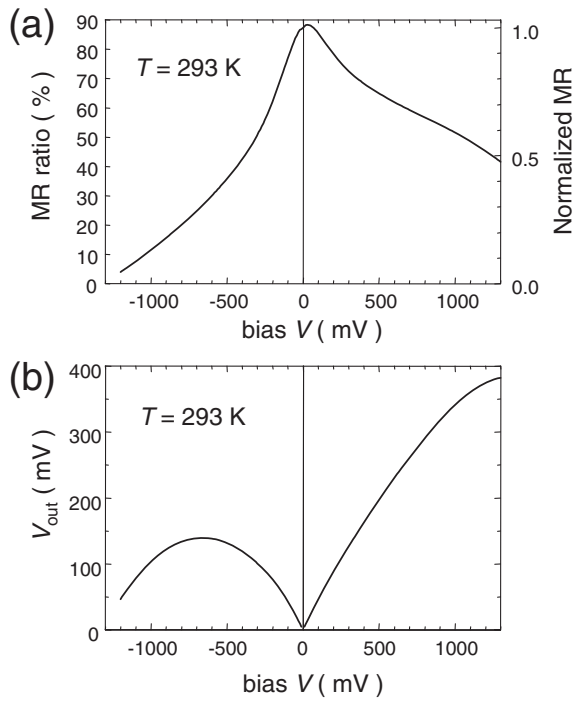

Fig. 4. (a) Bias-voltage (V) dependence of MR ratio for Fe(001)/MgO(001)(20 Å)/Fe(001) MTJ at room temperature. Direction of bias voltage is defined with respect to upper electrode. The V_{half} (bias voltage where MR ratio reaches half the zero-bias value) was 1250 and 390 mV for positive and negative bias directions, respectively. (b) Output voltage (V_{out}) of MTJ, defined as bias $V \times (R_{ap} - R_p)/R_{ap}$, as a function of bias V. Maximum V_{out} was 380 mV.

the MTJs is shown in Fig. 4(b). It reached a maximum of 380 mV for the positive bias. This is about twice that for conventional MTJs with an Al-O barrier (~200 mV)[14] and is high enough for developing next-generation high-density MRAMs.

In conclusion, we fabricated fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs with high reproducibility and observed an MR ratio of 88%, a V_{half} of 1250 mV, and a V_{out} of 380 mV at room temperature, the highest reported values at room temperature. The high MR ratio is attributed to the coherent spin-polarized tunneling. Those results demonstrate the feasibility of achieving a very high MR ratio for epitaxial MTJs and suggest that Fe(001)/MgO(001)/Fe(001) is a key device for future Gb MRAMs.

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