

Tunneling magnetoresistance of magnetic tunnel junctions using perpendicular magnetization $L1_0$ -CoPt electrodes

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Magnetic tunnel junctions (MTJs) using $L1_0$ -ordered CoPt electrodes with perpendicular magnetic anisotropy were fabricated. Full-epitaxial CoPt/MgO/CoPt-MTJs were prepared onto single crystal MgO-(001) substrate by sputtering method. X-ray diffraction analyses revealed that both bottom and top CoPt electrodes were epitaxially grown with (001)-orientation. The $L1_0$ -chemical order parameter of 0.82 was obtained for the bottom CoPt electrode deposited at substrate temperature of 600 °C. The transport measurements with applying magnetic field perpendicular to the film plane showed a tunnel magnetoresistance ratio of 6% at room temperature and 13% at 10 K. © 2008 American Institute of Physics. [DOI: 10.1063/1.2913163]

Tunneling magnetoresistance (TMR) effect in magnetic tunnel junctions (MTJs) is a typical phenomenon studied in the spintronics research field. Recent rapid enhancement of the TMR ratio because of the discovery of crystalline MgO barrier presents great possibilities to realize a gigabit class high-density magnetic random access memory (MRAM).¹⁻⁴ A remaining critical issue for developing gigabit MRAM is the reduction of thermal stability caused by miniaturization of the device below a few tens of nanometers. To avoid thermal instability, a minimal stability ratio of magnetic anisotropy energy $K_u V$ to thermal energy $k_B T$, $K_u V/k_B T$ of ~ 60 , is necessary. Conventional 3D ferromagnets such as Fe, CoFeB used in Refs. 1-4 cannot satisfy this requirement because of their small magnetic anisotropy energy (K_u), less than 10^6 erg/cm³. MTJs using ferromagnets with high perpendicular magnetic anisotropy (p MTJs) cannot only satisfy the thermal stability requirement but also have no limit of the cell aspect ratio. Therefore, exploration of the p MTJs is strongly desired.

To develop p MTJs, $L1_0$ -ordered alloys such as FePt and CoPt are good candidates because they have a large perpendicular magnetic anisotropy attributable to their high K_u ($\sim 10^7$ erg/cm³) with a (001)-easy axis. Additionally, they also have small lattice mismatching with the MgO(001) plane less than 10%, which is important to fabricate full-epitaxial MTJs with a (001)-oriented crystalline MgO barrier and to obtain a high TMR ratio. For the present study, we adopt $L1_0$ -ordered Co₅₀Pt₅₀ as ferromagnetic electrodes in the p MTJ. We chose CoPt because it has not only large K_u of 4.9×10^7 erg/cm³ but also smaller saturation magnetization (M_S of about 800 emu/cm³) than that for FePt (~ 1140 emu/cm³).⁵ Small M_S is important to reduce the critical current density of spin-transfer-driven magnetization reversal, which is a key writing technology for gigabit MRAM.⁶

Full-epitaxial p MTJs of MgO(001) substrate/Cr/Pt/CoPt/MgO/CoPt/Ta structure were fabricated using a UHV ($P_{\text{base}} < 2 \times 10^{-7}$ Pa) magnetron sputtering system

without breaking vacuum. The CoPt layers and MgO tunnel barrier were deposited from Co₅₀Pt₅₀ alloy and a sintered MgO sputtering target, respectively. The Cr layer was deposited at ambient temperature, with subsequent annealing at 600 °C for 1 h then the Pt layer was continuously grown after cooling the substrate temperature to 350 °C. In this lower stacking structure, the epitaxial condition of MgO(001)[100]/Cr(001)[110]/Pt(001)[100] / CoPt(001)[100] is obtainable. For this study, we first optimized substrate temperature (T_s) and Ar gas pressure (P_{Ar}). In fact, P_{Ar} is known as an effective parameter to obtain a high degree of $L1_0$ ordering, even at low T_s , as reported in several previous works.^{7,8} Structural analysis is performed using x-ray diffraction (XRD) measurements with Cu K_α radiation. The $L1_0$ order parameter S was defined as the probability of correct site occupation in the $L1_0$ lattice; it is given as the following:

$$S^2 = (I_{001}/I_{002})_{\text{meas}} / (I_{001}/I_{002})_{\text{calc}}, \quad (1)$$

where I_{001} and I_{002} represent the integrated intensity of the (001) superlattice peak and (002) fundamental peak, and $(I_{001}/I_{002})_{\text{meas}}$ and $(I_{001}/I_{002})_{\text{calc}}$, respectively, denote the measured and calculated diffraction intensity ratios. Magnetic properties were measured using a vibrating sample magnetometer in fields of 15 kOe. The film topographic images were observed using atomic force microscopy (AFM).

After optimization of the bottom CoPt electrode, samples with a p MTJ structure were fabricated. The junctions were patterned into an area of $8 \times 8 \mu\text{m}^2$ using conventional photolithography and Ar ion milling process. The TMR effect was measured using a four-point probe system with the magnetic field perpendicular to the film plane from 10 to 310 K.

Figure 1 shows the result of θ - 2θ XRD scan measurements for the MgO substrate/Cr(40)/Pt(3)/CoPt(20) samples (values in parentheses denote thickness in nanometers) with different P_{Ar} from 0.1 to 1.1 Pa. In fact, T_s for CoPt is fixed at 500 °C. Only the (001) and (002) peaks derived from CoPt layer are detected for the entire range of P_{Ar} . The ϕ scan measurement for (022) plane (not shown here) showed fourfold in-plane symmetry, indicating (001)-

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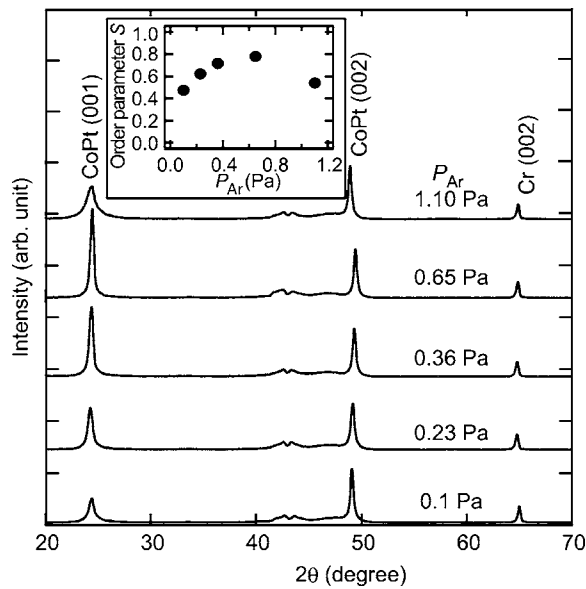


FIG. 1. θ - 2θ XRD scans of CoPt films as a function of P_{Ar} . The CoPt films were deposited at substrate temperatures of 500 °C. The inset shows the order parameter S for CoPt epitaxial films as a function of P_{Ar} .

epitaxial growth of CoPt layer. Intensity of (001) superlattice peak increases from 0.1 to 0.65 Pa and shows a maximum at 0.65 Pa, signifying the improvement of $L1_0$ -chemical ordering. The mechanism for enhancement of the chemical ordering with increasing Ar gas pressure is related to the lattice strain-induced preferred phase transformation because compressive stress along the c axis is preferred to transform the cubic to a tetragonal structure that appears in the $L1_0$ -ordering phase.⁷ The calculated chemical order parameter S using Eq. (1) is portrayed in the inset as a function of P_{Ar} . The sample with $P_{Ar}=0.65$ Pa shows maximum S of 0.78.

The M - H curves of perpendicular and parallel loops for corresponding samples are presented in Fig. 2; the component of diamagnetism was removed. Two M - H curves show almost identical behaviors in the sample with $P_{Ar}=0.1$ Pa, indicating small magnetic anisotropy. Perpendicular magnetic anisotropy is largely improved by increasing P_{Ar} . We estimated the magnetic anisotropy energy K_u from the M - H curves. Consequently, maximum K_u of about 1.2×10^7 erg/cm³ was obtained in the sample with $P_{Ar}=0.65$ Pa as well as order parameter S . This K_u is almost

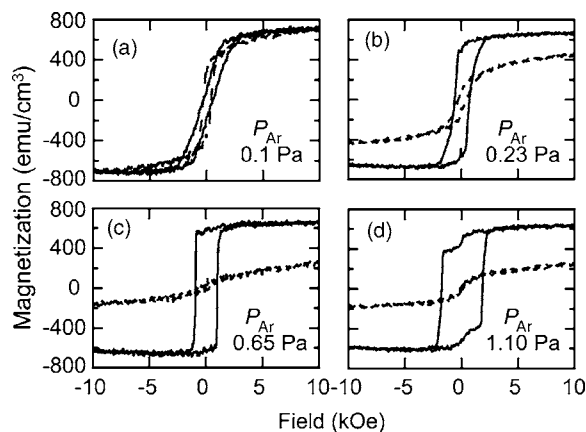


FIG. 2. (M - H) hysteresis loops for CoPt epitaxial films as a function of P_{Ar} . Solid and broken lines show perpendicular and parallel loops, respectively.

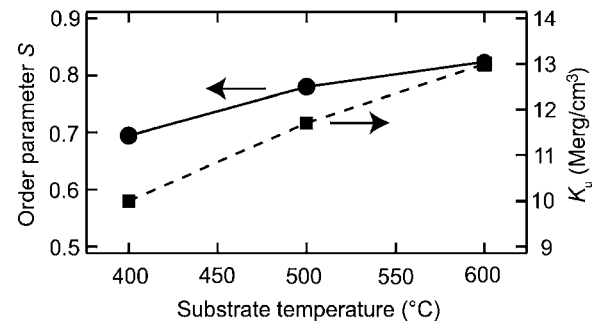


FIG. 3. Order parameter S and perpendicular magnetic anisotropy constant for CoPt epitaxial films as a function of substrate temperature.

equivalent to that in previous reports and slightly smaller than the bulk value of 4.9×10^7 erg/cm³.^{5,9,10} This small reduction of K_u is inferred to occur because of the partial imperfection of $L1_0$ chemical ordering ($S < 1$). Results further showed that coercive field gradually increased with P_{Ar} , which agrees with previous reports.⁸ Control of the coercive field merely by changing P_{Ar} is an important technique to form an antiparallel (AP) configuration of the two CoPt magnetizations in a p MTJ, as described below.

For further improvement of K_u , the substrate temperature T_s was optimized. Figure 3 shows the T_s dependence of the S and K_u , where P_{Ar} was 0.63–0.66 Pa. Both S and K_u gradually improved with T_s and reached 0.82 and 1.3×10^7 erg/cm³, respectively, at $T_s=600$ °C. In addition, AFM measurements showed that the average roughness of CoPt surface was less than 0.5 nm for all samples, which is sufficiently small to enable its use as the bottom electrode of MTJ.

We fabricated the p MTJs of MgO substrate/Cr(40)/Pt(3)/CoPt(20) $T_s=600$ °C, $P_{Ar}=0.65$ Pa/MgO(3)/CoPt(30)/Ta(5) structure (values in parentheses denote thickness in nanometers). To obtain a clear AP alignment of the two CoPt moments, we fixed P_{Ar} for the top CoPt electrode at 1.2 Pa, at which the H_c is much larger than that for the bottom CoPt electrode deposited at 0.65 Pa. Figure 4 shows M - H curves at room temperature of p MTJs with different T_s for the top CoPt electrode, (a) 300, (b) 400, and (c) 500 °C. We find a clear AP state and the smallest in-plane component of magnetization in $T_s=400$ °C, where the top CoPt electrode is magnetically harder. Therefore, we made

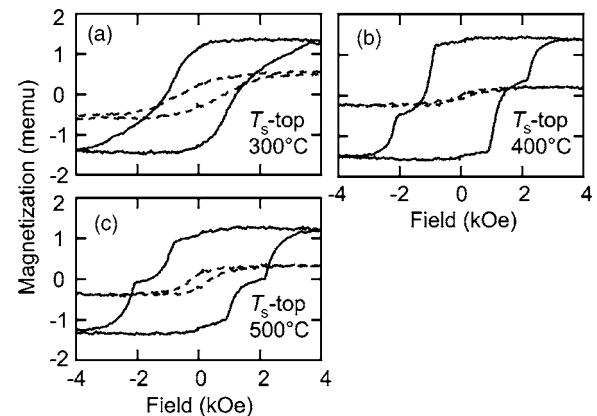


FIG. 4. (M - H) hysteresis loops at room temperature for p MTJs as a function of substrate temperature for the top CoPt electrode (T_{s-top}). The top CoPt electrode is deposited at Ar gas pressure of 1.2 Pa. Solid and dotted lines show perpendicular and parallel loops, respectively.

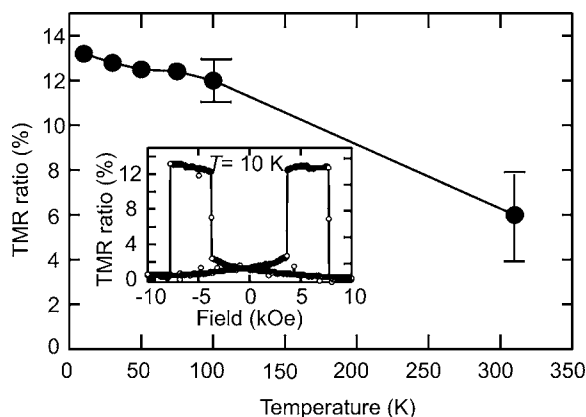


FIG. 5. Dependence of the TMR ratio on temperature for a *p*MTJ prepared with an epitaxial CoPt bottom and top electrodes. The inset shows the TMR curve at 10 K. All the measurements are conducted below 5 mV.

four-terminal devices in this MTJ using a microfabrication process and investigated the magnetotransport properties.

Results of TMR measurements show clear TMR behavior in the fabricated CoPt/MgO/CoPt-MTJs, as shown in the inset of Fig. 5. The observed TMR ratio is about 6% at 310 K and 13% at 10 K with applied bias voltage below 5 mV.

The possible reasons for the observed small TMR ratio are: (i) poor crystallinity of MgO barrier and interfacial dislocations between MgO barrier and CoPt electrode because of the lattice mismatching of 9.5%; and (ii) intrinsic reduction of effective spin polarization of tunneling electrons attributable to the energetic location of Δ_1 band, which is the most important to obtain giant TMR ratio, as predicted in Fe/MgO/Fe-MTJs.¹¹ However, these problems can be solved during future development by inserting a thin layer of other ferromagnetic material into both upper and lower inter-

faces of CoPt and MgO. For example, Fe has small lattice mismatching with both CoPt and MgO (−6.7% and 3.5%, respectively) and shows a large TMR ratio, as already demonstrated in Fe/MgO/Fe-MTJs.²

In conclusion, we fabricated a full-epitaxial MTJ with a $L1_0$ -CoPt electrode and MgO tunneling barrier. Large magnetic anisotropy energy K_u ($\sim 1.3 \times 10^7$ erg/cm³) and $L1_0$ -order parameter S (~ 0.82) for the bottom CoPt electrode were obtained by optimizing the substrate temperature and Ar gas pressure during CoPt deposition. Magnetotransport measurements showed clear TMR behavior because of the AP alignment of two CoPt magnetizations. We observed TMR ratio of about 6% at 310 K and 13% at 10 K.

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¹S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S. Hun Yang, *Nat. Mater.* **3**, 862 (2004).

²S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, *Nat. Mater.* **3**, 868871 (2004).

³D. D. Djayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yamagata, N. Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, *Appl. Phys. Lett.* **86**, 092502 (2005).

⁴J. Hayakawa, S. Ikeda, Y. M. Lee, F. Matsukura, and H. Ohno, *Appl. Phys. Lett.* **89**, 232510 (2006).

⁵T. Klemmer, D. Hoydick, H. Okumura, B. Zhang, and W. A. Soffa, *Scr. Metall. Mater.* **33**, 1793 (1995).

⁶K. Yagami, A. A. Tulapurkar, A. Fukushima, and Y. Suzuki, *Appl. Phys. Lett.* **85**, 5634 (2004).

⁷T. Suzuki, K. Harada, N. Honda, and K. Ouchi, *J. Magn. Magn. Mater.* **193**, 85 (1999).

⁸T. Suzuki, H. Muraoka, Y. Nakamura, and K. Ouchi, *IEEE Trans. Magn.* **39**, 691 (2003).

⁹K. Barmak, J. Kim, L. H. Lewis, K. R. Coffey, M. F. Toney, A. J. Kellock, and J.-U. Thiele, *J. Appl. Phys.* **98**, 033904 (2005).

¹⁰M. R. Visokay and R. Sinclair, *Appl. Phys. Lett.* **66**, 1692 (1995).

¹¹J. Mathon and A. Umerski, *Phys. Rev. B* **63**, 220403R (2001).