Current-induced butterfly shaped domains and magnetization switching in magnetic tunnel junctions

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Abstract

Patterned magnetic tunnel junctions (MTJs) with the layer structure of Ta (5 nm)/Ni\textsubscript{79}Fe\textsubscript{21} (5 nm)/Cu (20 nm)/Ni\textsubscript{79}Fe\textsubscript{21} (5 nm)/Ir\textsubscript{22}Mn\text sub{78} (10 nm)/Co\textsubscript{75}Fe\textsubscript{25} (4 nm)/Al (0.8 nm)-oxide/Co\textsubscript{75}Fe\textsubscript{25} (4 nm)/Ni\textsubscript{79}Fe\textsubscript{21} (20 nm)/Ta (5 nm) were fabricated using magnetron sputtering deposition and lithography. High tunnelling magnetoresistance ratios of 22 and 50\% were obtained at room temperature before and after annealing, respectively. The evolution of leaf shaped images was observed via Lorentz transmission electron microscopy (LTEM) on the MTJs, which were deposited on a patterned and carbon-coated transmission electron microscopy grid. These leaf-like LTEM images correspond to a butterfly shaped domain structure that was confirmed by a micromagnetics simulation. When a large DC current or bias voltage was applied across the MTJ, the butterfly-like vortex domain structures could be induced to form in the free layer of the MTJ, resulting in a significant decrease of magnetization in the free layer. The existence of these butterfly shaped domains could be one of the major causes of the bias voltage dependence of the TMR ratio.

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1. Introduction

Research in magnetic tunnel junctions (MTJs), no matter whether the focus is materials development or device fabrication has caught the attention of researchers in both academic and industrial sectors due to the promising application potential of MTJs for magnetoresistive random access memory (MRAM) and high-sensitivity field sensors. [1–5] Although considerable progress in both experimental and theoretical research in MTJ materials and the tunnel magnetoresistance (TMR) effect has been achieved [6–11], some magnetic and structure properties as well as spin-electron transport characteristics of MTJs have not yet been fully investigated. For instance, the dynamic domain structure of MTJs has not yet been characterized completely due to their complex structure, while the difference between experimental data and theoretical evaluations of the DC bias voltage dependence of the TMR effect is also an area of dispute. [6,12,13] Therefore, there are still pressing needs for further investigations on these unclear topics in order to improve the understanding of the MTJ structure, thereby to facilitate the development and enhancement of MTJ-based devices.

In this letter, we present the fabrication and the magneto-transport characteristics of the MTJs with the layer structures of Ta (5 nm)/Cu (20 nm)/Ni\textsubscript{79}Fe\textsubscript{21} (5 nm)/Ir\textsubscript{22}Mn\textsubscript{78} (10 nm)/Co\textsubscript{75}Fe\textsubscript{25} (4 nm)/Al (0.8 nm)-oxide/Co\textsubscript{75}Fe\textsubscript{25} (4 nm)/Ni\textsubscript{79}Fe\textsubscript{21} (20 nm)/Ta (5 nm). Direct observation of the magnetic domain structures in the free layer of the MTJ under a current-induced Oersted field was achieved using a Lorentz transmission electron microscopy (LTEM), while the domain structures and the magnetization reversal process were simulated based on micromagnetic calculations.

The MTJ films were first deposited on a Si/Si\textsubscript{2}O\textsubscript{2} wafer using an ULVAC TMR R&D Magnetron Sputtering System (MPS-4000-HC7) without breaking the vacuum. An in-situ
magnetic field of around 100 Oe was applied in-plane throughout the MTJ film deposition process. The Al-oxide barrier was formed by inductively coupled plasma oxidation with a mixture of 0.75 Pa oxygen and 0.25 Pa argon for 40 s. Then, the MTJ films were patterned using lithography combined with Ar ion-beam etching and CF$_4$ reactive etching. Finally, in order to obtain a high magnetoresistance ratio, the as-deposited MTJs were annealed in vacuum at 300 °C for one hour with an in-situ in-plane magnetic field of around 300 Oe. At room temperature (RT), we obtained TMR ratios of 22 and 50%; resistance times area products, RS, of around 3022 and 4364 Ωµm$^2$; and free layer coercivity of 25 and 23 Oe; for the as-deposited and annealed MTJs, respectively. Sample size was 8×8 µm$^2$, as shown in Fig. 1.

The MTJs fabricated for the LTEM experiment were deposited on patterned carbon thin film coated TEM substrate. The MTJ structure was the same as described above while the circularly patterned MTJ had a diameter of around 100 µm and approximately 100 patterned MTJs were formed on the TEM substrate. The Fresnel mode of LTEM was adopted in the observation, so the magnetic images appeared in high contrast. [14,15] The electron beam passed perpendicular to the plane of the MTJ, and the LTEM images were taken at different times. With the aid of the electron beam in the LTEM of the MTJ, a corresponding DC current can be considered passing through the MTJ perpendicular to the plane. We can therefore, directly observe the dynamic magnetic structure of the free layer of the MTJ under the effect of the DC current. (Note: It is assumed that the hard layer of the MTJ was strongly pinned by the antiferromagnetic layer. Thus no significant domain activity could occur due solely to a small DC current in the hard layer.)

Fig. 2 shows a typical LTEM image of the free layer after an electron beam exposure of around 2 min. (Note: In the first few seconds of the electron beam exposure, only magnetic ripples were observed. By ripple we mean a very narrow region where the magnetization vector has, on average, a positive z value along one edge and a complementary negative z value on the opposite edge.) This LTEM image, with a size of 20×20 µm, was taken from the central section of a circular MTJ image with the diameter of 100 µm, for a sample. Such LTEM images appeared all over each circular MTJ. We observed more than 50 MTJs and they all showed this kind of leaf-like pattern of magnetic contrast. In Fig. 2 five large leaf-like magnetic contrast images are visible, each with a size of around 5×5 µm, and a smaller one with a size of 2.4×2.4 µm appears at the top-left corner. This is probably the first time such leaf-like magnetic contrast images in MTJ have been reported in the literature. Besides the leaf-like magnetic contrast, magnetization ripples also appear in Fig. 2. The magnetization ripples resulted from the texture and the random distribution of the grains in the polycrystalline free layer. Magnetization ripples are always oriented perpendicular to the easy magnetization direction of a domain. [14] Therefore, the easy magnetization direction (EMD) of the free layer can be recognized in Fig. 2 (marked by the arrow). The free layer EMD was induced by the in-situ magnetic field applied in-plane during the MTJ film deposition process.

The distribution of the leaf-like LTEM images implies that the junction resistivity was different at different locations. Thus the potentials across the junction were also different in different areas. We observed a number of
leaf-like LTEM images on the whole circular MTJ with the junction diameter of 100 μm. There are two possible causes of different resistivity at different local regions in the MTJ. One is the barrier thickness inhomogeneity, which can lead to a large difference in current density or junction resistance, up to 3–5 orders of magnitude. Such an effect was confirmed by observing the current images on low-resistance MTJs using conducting atomic force microscopy (C-AFM).[16] Another possible cause is the formation of pin-holes that may lead to very low junction resistance as the pin-hole barrier width can be considered to be zero.

The largest diameter of leaf image is 5 μm as that shown in Fig. 2. The center of each leaf-like image corresponds to a very small region with a much thinner barrier and a much lower resistance, or even a pin-hole. Considering that the electron beam density of the LTEM was on the order of 100 pA/cm² and both Coulomb blockage and electron accumulation could occur on the MTJ during the electron beam exposure, a linear current of the order of 10 nA up to 100 μA could pass through the low resistance local regions of the MTJ. Therefore, a current-induced Oersted field that could lead to the magnetization switching may have appeared in the MTJ resulting in the formation of the leaf-like magnetic contrast images in the free layer. It is expected that such leaf-like images, which correspond to one kind of dynamic domain structure, can occur in the free layer whenever a DC bias voltage or current is applied over a critical value, depending on the MTJ structure.

In order to verify the LTEM images described above, micromagnetics simulations were done using the minimization of energy method.[17] The total energy of the MTJ system consists of the exchange interaction energy, the magnetocrystalline anisotropy energy, the Zeeman energy under a current-induced Oersted field, and the stray field energy.

Fig. 3(a) and (b) show two typical micromagnetics results simulating an MTJ with a size of 2.4×2.4 μm² when DC linear current of 8 and 14 mA, respectively, passes through its center. In the simulations, the bottom magnetic electrode of Co (4 nm) is pinned along the EMD. However, the top layer of Py (20 nm)/Co (4 nm) is free. The magnetocrystalline anisotropy constant, spontaneous magnetization, and exchange interaction constant of the free layer are $K_1=12 \times 10^3$ erg/cm³, $M_s(T=300 \text{ K})=800 \text{ Gs}$, and $A=1.0 \times 10^{-6}$ erg/m, respectively, and are taken from the parameters of the Ni 79Fe 21 alloy. The normal distribution of magnetic grain size in the free layer was adopted. The pinned boundary condition was adopted in simulation because the aim is to simulate the leaf-like images with the size of around 2.4×2.4–5.0×5.0 μm², which show patterns that developed in a much larger sample, a 100 μm diameter circular MTJ, and the magnetization direction of the magnetic grains located at the four outer edges for each leaf-like image basically remained in the parallel alignment state along the EMD in Fig. 2. The simulation calculated the linear current passing through the center of the leaf-like image to be between 0.8 and 13 mA, by the best fitting to the image under the pinned boundary condition. A novel butterfly shaped domain structure produced by the simulation reflects the main characteristics of the leaf-like LTEM image at certain amperages, rather than always forming a simple circular vortex domain structure.

It can be seen from both the experimental image and the theoretical simulations that the average magnetization direction in both the leaf-like image and the butterfly-like domain is oriented from the left side to the right side, aligned along the direction of the external magnetic field applied during the MTJ’s deposition (Figs. 2 and 3(a) and (b)). The black and white areas in Fig. 3(a) and (b) represent the magnetization direction of the grains upward and downward, respectively, relative to the MTJ surface, and the gray scale in between shows the relative out-of-plane projection of the grains’ average magnetization vectors in a three dimensional magnetization distribution. The bright leaf-stem line of the ellipse-marked leaf-like image in Fig. 2 consists of the 90° Néel wall and a long bright ripple line, and a similar pattern can be seen in Fig. 3(b) at
the top–middle area. The top–end area of the leaf-stem corresponds to a triangle of vortex domain structures, and in the butterfly, two similar domains appear, defining the butterfly’s two antennae in Fig. 3(b). A circular vortex domain, defining the core and tail of the butterfly body, formed in the center area of the butterfly-like domain due to the strong influence of the current-induced Oersted field. We can see that the black leaf-tip line in Fig. 2 consists of a black ripple line at the bottom and a white line in the upper part of the leaf-stem. In Fig. 3(b), the same sort of reversal can be seen in the distinct contrast between the black center line that defines the butterfly’s tail and the white ripple line extending above the head. Also white ripple lines, essentially the same region, define the butterfly’s wings. The large bright area with the shape of a horse’s hoof that constitutes most of the leaf’s body in Fig. 2 corresponds to the C-shaped areas that define the wings of the butterfly-like domains due to the distinct directional variation of the magnetization-vector along the z-axis on the whole MTJ surface in Fig. 3. In fact, the antennae, head, body, and double wings of such a butterfly-like domain also consist of more, smaller magnetic micro-structures. Overall, the key characteristics of the leaf-like LTEM image in Fig. 2 are quite consistent with those of the butterfly-like domain structure in Fig. 3(a) and (b). Hence, we feel that such micromagnetics simulations are effective and reasonable.

The dynamic butterfly-like domains and magnetization switching can occur in the free layer of an MTJ when a DC current passes through the MTJ on the order of 100 μA–10 mA, under a DC bias voltage of 10–500 mV. Our simulations show that the magnetization distribution of the MTJ free layer varies from an initial parallel-aligned magnetization state at 0 mA into a small butterfly-like domain structure that appears at around 1 mA, mutating into a large butterfly-like domain structure at around 10–20 mA, then degenerating into a circular vortex domain structure in response to current above 40 mA, all under a pinned boundary condition.

The spin-electron transport properties and TMR ratio can be dramatically affected by the magnetization switching via the current-induced Oersted field. The butterfly-like and circular vortex domain structures that form can result in a sharp decrease of magnetization in the free layer. We simulated two cases of the patterned MTJs under an unpinned boundary condition (i.e. after patterning, a typical MTJ is 8 × 8 and is independent of its neighboring MTJs, so the magnetic grains in the free layer that are located at the boundary can easily switch magnetization direction). One has an inhomogeneous barrier which is a small thinner region at the center or even having a small pin-hole in the center, which allows linear current to pass through the MTJ. (The simulation result presented in Fig. 3 is for the MTJ with a low TMR ratio in the as-deposited state.) The simulated case at the other extreme has a very flat barrier, which is pin-hole-free, thus a planar current passes through the MTJ (i.e. an MTJ with a high TMR ratio of 50% at RT after annealing). A workable MTJ is usually in some intermediate state between these two cases we simulated. Fig. 4(a) and (b) show the DC current I dependence of the normalized magnetization J/I/J, for the two MTJs, based on the simulations at RT. The open circles are simulated data and the solid lines represent the fitted curves based on an exponential decay function using an empirical formula as follows:

\[
\frac{J(T, I)}{J_S(0, 0)} = \left[ 1 - 0.1187 \alpha \left( \frac{T}{T_c} \right)^{1/2} \right] e^{-\frac{\beta I}{J}}, \text{ for } T < T_c. \tag{1}
\]

or

\[
\frac{J(T = 300 K, I)}{J_S(300 K, 0)} = e^{-\frac{\beta I}{J}}, \text{ for } T < T_c. \tag{2}
\]

Here, J_S(0, 0) is the spontaneous magnetization of the free layer at 0 K and zero current. Therefore, J(T = 300 K, I = 0) = J_S(0, 0)\{1 − 0.1187(\alpha T/T_c)^{1/2}\} = 800 Gs, \(S = 1\), and T_c = 900 °C for the Ni_{80}Fe_{21} alloy, and the maximum energy of magnon excitation E_m = 3k_BT_c/(S + 1) =121 meV. The α and β are the fitting parameters. Although the normalized magnetization of the free layer shows approximately the same exponential decay in both cases with increasing I, the MTJ with the flat barrier can maintain a higher magnetization value and higher TMR ratio than the MTJ with the inhomogeneous barrier.

Fig. 4. DC current I dependence of the normalized magnetization, simulated under an unpinned boundary condition for an MTJ with (a) an inhomogeneous barrier where a pinhole exists; and (b) a homogeneous barrier.

Fig. 5. (a) DC bias voltage dependence; and (b) the corresponding DC current dependence of TMR ratio for the same MTJ as that shown in Fig. 1 measured at RT.
Fig. 5(a) and (b) show the typical DC bias voltage $V$ dependence and the corresponding DC current $I$ dependence of the TMR ratio measured at RT for the same MTJ as that shown in Fig. 1. The open circles are experimental data and the dashed lines are guides for the eyes. The solid line in Fig. 5(a) represents the calculated values at RT, based on the spin-polarization electron tunnelling theory developed by Zhang et al. [6,12]

As can be seen from Fig. 5(a), the theoretical values agree well with the experimental data when the absolute values of the biased voltage is less than $200 \text{ mV}$ (i.e. $I \leq 4 \text{ mA}$) because the net magnetization value of the free layer remains unchanged in both parallel and antiparallel magnetization configurations of the free and pinned layers. [6,12] However, for applied DC bias voltage higher than $200 \text{ mV}$ (or $I \geq 4 \text{ mA}$), the experimental value of TMR ratio decreased more quickly than did the theoretical evaluation. Our micromagnetics simulation shows that the difference in the TMR values is due to the quick decrease of the net magnetization of the free layer; i.e. the decrease of the effective spin-polarization or the effective density of states (DOS) of the free layer due to the appearance of the butterfly-like vortex domain structures. Therefore, the Jullière’s formula—i.e. $\text{TMR} = \frac{R_{UP} - R_{DP}}{R_{UP}} = 2P_1 P_2 \left(1 - P_1 P_2\right)$—and the extended magnon—and phonon-assisted tunnelling theories hold only when no vortex domain structures appear in the ferromagnetic electrodes. [6,12,13] However, for more precise theoretical calculations, especially in the high bias voltage regime, the dynamic domain structure and magnetization switching due to the Zeeman interaction energy (i.e. the combination of a current-induced Oersted field and an external field) should be considered.

In conclusion, a current-induced Oersted field could lead to the formation of butterfly-like domain structures and local magnetization reversals in the ferromagnetic electrodes—especially in the free layer—of the MTJs that have inhomogeneous barrier layer structure (i.e. non-uniform thickness or the existence of a pin-hole). The micromagnetics simulation shows that the butterfly-like vortex domain structures can decrease the net magnetization values of the ferromagnetic electrodes under high voltage biasing, so the TMR ratios decrease significantly at high bias voltage (i.e. large dc current) regimes. Zeeman interaction energy should be considered for analyzing the magneto-electronic properties of the MTJs when large biasing voltages are applied across the junctions.

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