Role of interfacial roughness on bias-dependent magnetoresistance and transport properties in magnetic tunnel junctions

J. C. A. Huang\textsuperscript{a} and C. Y. Hsu
Department of Physics, National Cheng Kung University, Tainan, Taiwan, Republic of China, Department of Applied Physics, National University of Kaohsiung, Kaohsiung, Taiwan, Republic of China, and Taiwan Spin Research Center, National Chung Cheng University, Chiayi, Taiwan, Republic of China

Y. F. Liao, M. Z. Lin, and C. H. Lee
Department of Engineering and System Science, National Tsing-Hua University, Hsinchu, Taiwan, Republic of China

(Received 19 July 2005; accepted 5 October 2005; published online 17 November 2005)

The effects of metal-insulator interfacial roughness, modulated by Ar\textsuperscript{+} irradiation, on bias dependence of tunnel magnetoresistance (TMR) and electrical transport of CoFe–AlO\textsubscript{x}–CoFe magnetic tunnel junctions (MTJs) have been studied. Reduction of TMR ratio and asymmetric TMR falloff curves as a function of dc bias have been observed for Ar\textsuperscript{+}-irradiated MTJs. The results are analyzed by x-ray reflectivity together with complex impedance techniques, indicating interfacial roughness which likely results in a proportional rising trap state density (TSD). Increasing TSD for Ar\textsuperscript{+}-irradiated MTJs increases an unpolarized current which decreases TMR ratio. The asymmetric TMR falloff curves are attributed to the different TSDs of bottom and top CoFe–AlO\textsubscript{x} interfaces in tunneling process. © 2005 American Institute of Physics. [DOI: 10.1063/1.2132096]

I. INTRODUCTION

Giant tunnel magnetoresistance (TMR) effect in magnetic tunnel junctions (MTJs) has been extensively studied because of potential applications in nonvolatile magnetoresistive random access memory, high-density magnetic recording, and advanced magnetic devices in the next generation.\textsuperscript{1,2} From the application viewpoint, one of the key issues on how MTJs can be used is the dc bias voltage dependence of TMR. The dramatic decrease in TMR ratio with increasing bias voltage (or so-called TMR falloff curves) has been experimentally and theoretically investigated and was attributed to the possible origins of spin excitation localized at metal-insulator (M-I) interfaces, the impurity-assisted tunneling in tunnel barrier, and bias-dependent density of states of ferromagnetic electrodes.\textsuperscript{3–5} Asymmetric TMR falloff curves have been observed in MTJs with formation of improper-oxidized and composite tunnel barriers, Heusler alloys ferromagnetic electrodes, and MTJs subjected to annealing process.\textsuperscript{5–8} The asymmetric TMR falloff curves generally were believed to be due to different interfacial conditions of top and bottom M-I interfaces in MTJs.\textsuperscript{5,6} However, the origin of this phenomenon remains unclear so far.

We would like to understand the impact of interfacial roughness on TMR falloff curves and transport properties of MTJs. First, precise control of oxidation process for tunnel barrier has been achieved to exclude improper oxidation effects on the TMR falloff curves and transport properties of MTJs. Based on the interfacial modulation technique,\textsuperscript{9} we design a study to only modulate the M-I interfacial roughness under different Ar\textsuperscript{+} irradiation times (t\textsubscript{Ar}). X-ray reflectivity together with complex capacitance (CC) and bias-dependent complex impedance (CI) techniques\textsuperscript{10} are utilized to probe the M-I interfacial roughness and barrier quality. The mechanisms of the observed reduction of TMR ratio, asymmetric TMR falloff curves, and variation of ac transport properties of MTJs are investigated.

II. EXPERIMENTAL DETAILS

MTJs of CoFe(25 nm)/AlO\textsubscript{x}(~3 nm)/CoFe(10 nm) were prepared on glass substrates by a dual ion-beam sputter system with a base pressure of 5 × 10\textsuperscript{−7} Torr. The Ar\textsuperscript{+} irradiation was processed with a fixed acceleration voltage of 100 V on the bottom CoFe electrodes before the initial Al layer deposition. The t\textsubscript{Ar} was varied from 0 to 300 s. The AlO\textsubscript{x} layer was fabricated by exposing the Al layer under O\textsubscript{2}/Ar ion-beam irradiation with the proper-oxidized condition.\textsuperscript{10} X-ray reflectivity was performed at the wiggler BL17A beamline of the National Synchrotron Radiation Research Center in Hsinchu, Taiwan.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the selected TMR curves of CoFe–AlO\textsubscript{x}–CoFe MTJs with t\textsubscript{Ar}=0, 75, and 300 s. Indeed we observe TMR ratio gradually decreases from 7.5% to 1.3% when t\textsubscript{Ar} increased from 0 to 300 s. The decrease of TMR ratio, induced by Ar\textsuperscript{+} irradiation, generally can be correlated to the change of barrier quality and/or the interfacial roughness. The CC spectroscopy has been demonstrated to be a convenient tool in probing the barrier quality of MTJs.\textsuperscript{10} The CC spectra of the 0, 75, and 300 s Ar\textsuperscript{+}-irradiated MTJs display a concave at intersection of the rising tail and the arc, as shown in Fig. 1(b). This specific feature indicates that all the tunnel barriers situate in the optimized oxidation condition, i.e., the improper oxidation possibility on decreasing TMR ratio of the MTJs can be excluded. Therefore, the de-
crease of TMR ratio is solely due to the variation of the M-I
interfacial roughness. To characterize the interfacial rough-
ness of Ar+-irradiated MTJs, x-ray reflectivity spectra of the
0, 75, and 300 s Ar+-irradiated MTJs are presented in Fig. 1(c).
For increasing $t_{Ar}$, the x-ray reflectivity peaks broaden
and shift toward higher $q$ values. The x-ray reflectivity data
are further simulated and the fitting curves show good agree-
ment with the experimental results, as illustrated in Fig. 1(c).
The analyzing results of x-ray reflectivity, as shown in Fig.
1(d), indicate that the roughness profiles for the bottom and
top M-I interfaces all increase with rising $t_{Ar}$. However, the
roughness of the bottom M-I interface is larger than the top
one and their difference enhances with increasing $t_{Ar}$. As
induced by Ar+ irradiation, the increasing M-I interfacial
roughness is responsible for the reduction of TMR ratio.

The dc bias dependence of TMR ratio is critical to opti-
mize the performance of MTJs in application. The normal-
ized TMR falloff curves of 0, 75, and 300 s Ar+-irradiated
MTJs are demonstrated in Fig. 2. Here the normalized TMR
ratio is defined as the normalization of $(R_0-R_s)/R_0$, where
$R_0$ and $R_s$ are, respectively, the junction resistance of zero
and saturated field. The normalized TMR falloff curves show
two specific features. First, symmetric normalized TMR fal-
lloff curves have been observed for $t_{Ar}=0$ MTJs, but they
become apparently asymmetric with increasing $t_{Ar}$. Secondly,
the normalized TMR falloff curves for $t_{Ar}=0$ MTJs demon-
strate a slow decrease with increasing dc bias ($V_{dc}$), in con-
trast with a faster decrease for increasing $t_{Ar}$. To explain the
above phenomena, advanced analyses by bias-dependent CI
spectra are further employed to probe the M-I interfacial
transport properties for the Ar+-irradiated MTJs. In the mea-
surement of bias-dependent CI spectra, a suitable choice of
$V_{dc}$ amplitude is essential to probe the interfacial conditions.
The dc four-point-probe $(4p)$ current-voltage $(I-V)$ curves
provide useful information for the choice of bias amplitude.
Figure 3(a) displays the dc $4p$ $I$-$V$ curves of 0, 75, and 300 s
Ar+-irradiated MTJs. The $I$-$V$ curves of the 75 and 300 s
Ar+-irradiated MTJs almost overlap in low-bias-voltage re-
region, but these two $I$-$V$ curves become distinguishable when
$V_{dc}$ exceeds about 100 mV. Therefore, we apply an addi-
tional $V_{dc}$ of 400 mV [typical for magnetic random access
memory (MRAM) operation] in the measurement of CI spe-
ctra. Based on the above design, the bias-dependent CI spectra
can be used to probe the variation of the top and bottom M-I
interfacial conditions perturbed by Ar+ irradiation on the bot-
tom electrode.

Figure 3(b) demonstrates the bias-dependent CI spectra
(at 400 mV dc bias from bottom to top electrodes) in type of
log-log Nyquist plots. The bias-dependent CI spectra can be
analyzed by equivalent circuit model, including the contribu-
tions for CoFe–AlO$_x$ interfaces ($R_i$ and $C_i$), bulk AlO$_x$
layer ($R_p$ and $C_p$), and lead of cross patterns ($R_l$). The fitting
curves in Fig. 3(b) demonstrate great agreement with experi-
mental results. Among these parameters, $R_i$ and $C_i$ are the
dominant factors related to the M-I interfacial transport prop-
erties discussed below. First, we observe that the interfacial
capacitance ($C_i$) increases with rising $t_{Ar}$, as shown in Fig.
3(c). It is noted that $C_i$ can be influenced by the charge redis-
tribution inside the tunnel barrier near the interface or by vari-
ation of interfacial traps. The former process, similar to
the formation of $p$-$n$ junctions, occurs only at a larger
length scale ($>100$ nm), thus, the possibility can be obvi-
atated. The latter process enhances the $C_i$ in proportion to the

FIG. 2. Normalized TMR falloff curves as a function of $V_{dc}$ ranged from
-400 to 400 mV for Ar+-irradiated CoFe–AlO$_x$–CoFe MTJs with irradiation
time $t_{Ar}=0, 75$, and 300 s. The inset indicates the patterns of normalized
$R_0-R_s$ vs $V_{dc}$.
The authors would like to thank Professor Y. H. Lee for the kind assistance of using the impedance analyzer. This work has been supported by the National Science Council of the ROC under Grant No. NSC 94-2112-M-006-003, Taiwan Spin Research Center of the National Chun Cheng University under Grant No. 93-EC-17-A-01-S1-026, and National Synchrotron Radiation Research Center under Project No. 2004-3-032-1.

ACKNOWLEDGMENTS

Finally, we attempt to clarify the underlying mechanism for the asymmetric TMR falloff curves. The asymmetric TMR falloff curves of the 75 and 300 s Ar⁺-irradiated MTJs can be due to the variation of junction resistance \( R_0 \), \( R_0 - R_s \), or their mixed contributions. We notice that the junction resistance of all MTJs decrease with increasing \( V_{dc} \), but these curves (not shown here) are in marked difference with the patterns of Fig. 2. The possibility of junction resistance variation causing the asymmetric properties can be eliminated. We also plot the normalized \( R_0 - R_s \) vs \( V_{dc} \). As shown in the inset of Fig. 2, the curves are almost the same as the normalized TMR falloff curves. The asymmetric properties hence are due to the spin-dependent contribution as further interpreted in the following. MTJs with increasing top and bottom interfacial roughnesses, shown in Fig. 1(d), show increasing TSD at bottom and top interfaces. When spin-polarized (SP) carriers are injected from the bottom FM electrodes (positive bias region), the SP carriers first encounter the bottom interface with a higher TSD. These trap states behave as unpolarized channels, thus, the SP carriers decrease more dramatically during tunneling through the bottom M-I interface. On the contrary, SP carriers injected from top FM electrodes (negative bias region) first encounter the top interface with a relatively lower TSD, i.e., more SP carriers tunnel through the top M-I interface compared to the reverse bias process. When \( V_{dc} \) increases, the difference of spin-independent currents become apparent and lead to asymmetric TMR falloff curves in the positive and negative regions.

IV. CONCLUSIONS

In summary, asymmetric TMR falloff curves have been observed for Ar⁺-irradiated MTJs. CI and x-ray reflectivity techniques have been employed to characterize the Ar⁺-irradiated M-I interfaces. An interpretation, based on the distribution of TSD at M-I interfaces, has been proposed to disclose the underlying mechanism for the asymmetric TMR falloff curves. This work also demonstrates that the control of interfacial roughness can be utilized to modulate the transport properties of MTJs.