ac impedance techniques to study oxidation process of tunnel barriers in CoFe–AlO\(_x\)–CoFe magnetic tunnel junctions

J. C. A. Huang\(^a\)

Department of Physics, National Cheng Kung University, Tainan 701, Taiwan, Republic of China, Department of Applied Physics, National University of Kaohsiung, Kaohsiung 811, Taiwan, Republic of China, and Taiwan SPIN Research Center, National Chung Cheng University, Chiayi 621, Taiwan, Republic of China

C. Y. Hsu

Department of Physics, National Cheng Kung University, Tainan 701, Taiwan, Republic of China

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The complex impedance spectra of CoFe–AlO\(_x\)–CoFe tunnel junctions with under-, proper-, and overoxidized tunnel barriers have been investigated by ac impedance techniques. Two sets of parallel resistance \((R)\) and capacitance \((C)\) elements and a \(R\) element in series, modeling the impedance contributions of the metal-oxide interfaces and bulk insulating layers, are employed to describe the impedance spectra of under- and proper-oxidized junctions. This model, however, reveals a discrepancy for overoxidized junctions. This discrepancy can be reconciled by including a third set of parallel \(RC\) element, which suggests the appearance of overoxidized CoFeO\(_x\) layer upon the bottom electrode. From further analysis of interfacial capacitance as a function of oxidation time, the bottom interfacial capacitance widely diverges from the top interfacial capacitance and can be related to the oxidation process of tunnel barrier. The analyzing results of impedance technique are also consistent with the results by complex capacitance technique. © 2005 American Institute of Physics. [DOI: 10.1063/1.2058177]

I. INTRODUCTION

Magnetic tunnel junctions (MTJs), composed of an ultrathin (typically 1–3 nm) insulating layer separated by two magnetic layers, have been intensively investigated due to advanced magnetic applications, such as magnetic random access memories and hard disk read heads in the next generation.\(^1\)–\(^5\) Aluminum oxide \((\text{AlO}_{\text{x}})\), the most popular candidate as the ultrathin insulating layer of MTJs, is generally created by subsequent exposure of the oxygen plasma after the metallic Al layer is deposited. The precise control of oxidation condition is therefore the key step to fabricate a reliable MTJ because underoxidation with superfluous Al layer and overoxidation with additional oxide layer upon the bottom electrode can significantly reduce the tunnel magnetoresistance ratio. The complex impedance techniques have recently been attempted to study the formation of plasma-oxidized tunnel barrier.\(^6\) The interfacial effect,\(^7\)–\(^9\) originating from the large contrast in resistivity at a metal-insulator interface, was demonstrated to be a predominant mechanism influencing the transport of electron flow from one electrode, across the insulating layer, to the other electrode. However, the contribution of the interfacial effect had not been included in the analyzing process of complex impedance method of under-, proper-, and overoxidized MTJs.

The aim of this work is to disclose the dependence of interfacial properties on the oxidation process of metallic Al layers by ac impedance technique. Equivalent circuit method, containing \(R\) and \(C\) components, has been used to describe the metal-oxide interfaces as well as bulk AlO\(_x\) contributions of CoFe–AlO\(_x\)–CoFe tunnel junctions with under-, proper-, and overoxidized tunnel barriers. The results of the analysis reveal that interfacial capacitance is greatly sensitive to the oxidation process of Al layers and can be further applied to optimize the high-frequency-related device of magnetic tunnel junctions.

II. EXPERIMENTAL DETAILS

The MTJs on glass substrates were prepared by a dual ion-beam sputter with a base pressure of \(5 \times 10^{-7}\) Torr. The structure of MTJs consisted of CoFe(25 nm)/AlO\(_x\) (2.5 nm)/CoFe(10 nm)/Ta(5 nm). The AlO\(_x\) layer was fabricated by exposure of the Al layer under O\(_2\)/Ar-ion-beam oxidation and the oxidation time \((t_{\text{ox}})\) was varied from 30 to 130 s. The O\(_2\)/Ar-ion-beam was processed with an acceleration voltage of 100 V and a gas flow rate of 6 SCCM (standard cubic centimeter per minute). The cross-patterns of the CoFe–AlO\(_x\)–CoFe MTJs obtained by shadow masks were 200×200 \(\mu\text{m}^2\) in area. The MTJs with under- \((t_{\text{OA}}=30 \text{ and } 50 \text{ s})\), proper- \((t_{\text{OA}}=80 \text{ s})\) and over- \((t_{\text{OA}}=100, 120, \text{ and } 130 \text{ s})\) oxidized tunnel barriers had been confirmed in our previous work.\(^10\) The complex impedance spectroscopy was carried out by a Hewlett-Packard 4294A impedance analyzer using two-point contact in a frequency range from \(10^2\) to \(10^5\) Hz with a fixed oscillating voltage of 30 mV. The purpose of choosing a 30 mV ac signal to measure the complex impedance spectra was to ensure the effectiveness for analyzing the spectra by equivalent circuit model because this amplitude of ac signal was located in the linear current-voltage interval of 0–120 mV for under-, proper-, and overoxidized MTJs.

\(^{a}\)Electronic mail: jcahuang@mail.ncku.edu.tw
III. RESULTS AND DISCUSSION

The complex impedance $Z(f) = R(f) + jX(f)$ spectra of under-, proper-, and overoxidized MTJs have been demonstrated in Figs. 1 and 2. The real parts of the complex impedance $R(f)$ for under-, proper-, and overoxidized MTJs initially remain a constant, then follow several intermediate steps in the frequency range of $10^4$–$10^7$ Hz. The dependence of $R(f)$ below $10^3$ Hz on $t_{ox}$ shows an increase with $t_{ox}$ rising from 30 to 130 s. The negative value of imaginary parts of the complex impedance $-X(f)$ further demonstrates more than one local maximum in the frequency range from $10^2$ to $10^7$ Hz. The lowest-frequency response, the local maximum in the frequency range from $10^3$ to $10^5$ Hz, shifts toward lower frequency as $t_{ox}$ increases from 30 to 130 s.

Based on the above descriptions, two types of electric element combination can be used to model the possible contributions of complex impedance in different frequency range: (i) One $R$ and $C$ element in parallel is used to describe an intermediate step of $R(f)$, corresponding to a local maximum of $-X(f)$. (ii) A single $R$ element in series to the other electric element combination characterizes the nonzero value of $R(f)$ at $10^7$ Hz. For the under- and proper-oxidized MTJs, two sets of parallel $RC$ components in series to a $R$ element, representing the contribution from metal-oxide interfaces, bulk insulating layer ($AlO_x$) and leads of crosspatterns, are employed to model the complex impedance spectra, as shown in Eq. (1):

$$Z_1 = \left( \frac{1}{R_i} + j\omega C_i \right)^{-1} + \left( \frac{1}{R_b} + j\omega C_b \right)^{-1} + R_L,$$

where the parameters of $R_i$, $C_i$, $R_b$, $C_b$, and $R_L$ in Eq. (1), respectively, represent the resistance and capacitance of metal-insulator interfaces and bulk insulating layer ($AlO_x$) as well as the resistance of leads. It is here noted that the contribution of lead of crosspatterns is attributed to two-point probe method. As demonstrated by the solid curves of Figs. 1(a)–1(c), this model shows great agreement with the experimental results. However, this model shows a discrepancy in high-frequency interval for overoxidized tunnel junctions, as
indicated by the dash curve of Fig. 2(a). We notice that \( R(f) \) below \( 10^3 \) Hz significantly increases from 68 to 128 k\( \Omega \) for \( t_{ox} \) increasing from 100 to 130 s, similar to the increasing tendency for \( t_{ox} \) increasing from 30 to 80 s. This result indicates that the bottom CoFe layer has likely been oxidized for \( t_{ox}=100, 120, \) and 130 s MTJs, which resembles the oxidation of the Al layer for \( t_{ox}=30, 50, \) and 80 s MTJs. In addition, the tunnel magnetoresistance ratio gradually decreases when \( t_{ox} \) exceeds 80 s.\(^1\) Therefore, this discrepancy can be recovered by including an additional set of parallel \( RC \) components to characterize the overoxidized layer, CoFeO\(_x\), i.e., three sets of parallel \( RC \) components and a \( R \) element in series are utilized to model the overoxidized tunnel junctions. The complete equivalent circuit model, describing the complex impedance spectra of the overoxidized junctions, can be expressed as

\[
Z_2 = \left( \frac{1}{R_i} + j\omega C_i \right)^{-1} + \left( \frac{1}{R_b} + j\omega C_b \right)^{-1} + \left( \frac{1}{R_o} + j\omega C_o \right)^{-1} + R_L,
\]

where the \( R_o \) and \( C_o \) components represent the parallel \( RC \) components of the overoxidized layer, CoFeO\(_x\). The complex impedance spectra of the overoxidized tunnel junctions can be well described by Eq. (2), as displayed by the solid curves of Figs. 2(a)–2(c). The fitting parameters of Eqs. (1) and (2) are separated into two groups, the interfacial parameters \((R_i, C_i)\) and bulk parameters \((R_b, C_b, R_o, \) and \( C_o)\), as discussed in the following.

The fitting parameters of the interfacial resistance and capacitance \((R_i, C_i)\) of the under-, proper-, and overoxidized MTJs are shown in Figs. 3(a) and 3(b). The fitting interfacial resistance \( R_i \) increases with \( t_{ox} \) increasing from 30 to 80 s, then remaining almost a constant until \( t_{ox} \) was raised to 100 s, and further increases with \( t_{ox} \) from 100 to 130 s, as shown in Fig. 3(a). The dc four-point probe resistance and interfacial resistance, analyzing by complex capacitance (CC) techniques,\(^10\) also follow the same tendency of \( R_i \), which suggests that the electric conduction behavior is dominated by the electronic transport associated with the metal–insulator interfaces. On the other hand, the interfacial capacitance \( C_i \), as indicated in the Fig. 3(b), increases from \( t_{ox} =30 \) to 50 s, reaching a peak maximum at 80 s, then slowly decrease with \( t_{ox} \) up to 130 s. The interfacial capacitance \( C_i \) includes the contributions of top and bottom metal–oxide interfaces. Hence we attempt to separate the contribution of top interfacial capacitance from the bottom one. The interfacial capacitance \((C_i)\) can be regarded as the series combination of top interfacial capacitance \((C_i)\) and bottom interfacial capacitance \((C_b)\), as indicated in Eq. (3):

\[
C_i^{-1} = C_u^{-1} + C_b^{-1}.
\]

What type of metal and oxide contact is the predominant factor influencing parameter \( C_i \) for MTJs with various oxidized tunnel barriers?\(^11\)–\(^13\) For the proper-oxidized MTJs, it is reasonable to suppose the parameter \( C_i \) to be two similar capacitances in series, i.e., \( C_i=C_i/2 \) or \( C_i=C_i/2 \), because both top and bottom interfacial capacitances contribute by the CoFe–AlO\(_x\) interface. The postoxidation process of metallic Al layer mainly influences the interfacial condition between the insulating layer and the bottom electrode, instead of that between the insulating layer and the top electrode. Hence \( C_i \) of the under- and overoxidized MTJs is almost identical to that of the proper-oxidized MTJs. Further, the bottom interfacial capacitance can be further calculated according to Eq. (3). The calculating results are demonstrated in the inset of Fig. 3(b). For the underoxidized MTJs \((t_{ox} =30 \) and 50 s), the bottom interfacial capacitance is mainly contributed from the Al–AlO\(_x\) interface, which is different from the interfacial capacitance of the top CoFe–AlO\(_x\) interface due to the additional Al layer upon the bottom CoFe layer. With the \( t_{ox} \) increases to 80 s (the proper-oxidized MTJs), the additional Al layer is gradually oxidized and the bottom CoFe layer almost entirely contacts the AlO\(_x\) barrier. The similar interfacial condition between top and bottom interfaces therefore leads to the crossover of \( C_i \) and \( C_b \) curves, as indicated in Fig. 3(b). For the overoxidized MTJs \((t_{ox} =100–130 s)\), the bottom interfacial capacitance demonstrates a slight decrease from 80 (crossover point) to 100 s and a further decrease from 100 to 130 s. This discrepancy between top and bottom interfacial capacitances above 80 s can be explained by the appearance of the overoxidized CoFeO\(_x\) layer. Thus, for the overoxidation condition, the bottom interfacial capacitance is resulted from the CoFe–CoFeO\(_x\) interface, different from the top CoFe–AlO\(_x\) interface. The analysis of the results indicates that the interfacial capacitance is very sensitive to the oxidation process of Al layer.

![FIG. 3.](image-url)
The bulk parameters of $R_b$, $C_b$, $R_o$, and $C_o$ analyzing from complex impedance (CI) technique are represented in terms of relaxation frequency, $f_j = 1/(2\pi R C)$, for the sake of comparison to the results of CC technique\textsuperscript{10}. The relaxation frequency, analyzing from the CI and CC techniques, as a function of $t_{ox}$ is demonstrated in the Fig. 4(a). The behavior of relaxation frequency of AlO$_x$ and CoFeO$_x$, $f_{AlO_x}$ and $f_{CoFeO_x}$ respectively, on $t_{ox}$ is indicated from the analysis of the CI and CC techniques to follow similar tendency. The $f_{AlO_x}$ demonstrates a clear step near $t_{ox} = 100$ s, then gradually approaches to saturation by both techniques. The results can be related to the oxygen incorporation into the metallic Al layer and the AlO$_x$ layer gradually transforms from initial to final oxidation stage near $t_{ox} = 100$ s. The $f_{CoFeO_x}$, on the other hand, decrease for $t_{ox}$ from 100 to 130 s and the order of $f_{CoFeO_x}$ from 100 to 130 s is also comparable to the $f_{AlO_x}$ from 30 to 50 s. This result suggests that the bottom CoFe layer still situate in the initial oxidation stage for $t_{ox} = 100–130$ s.

According to the above discussion, one can find that the impedance contribution of metal-oxide interfaces is the predominant term compared to that of the bulk layers. Properties of metal-oxide interfaces, like equivalently interfacial capacitance, can be clearly identified by the CI spectra. In the CC spectra,\textsuperscript{10} the capacitance contribution of metal-oxide interfaces mainly reflects on the imaginary part of complex capacitance due to the leakage characteristic and drastically decreases with increasing frequency, based on the formula of $C''(\omega) = \sigma'/(\omega R C)$, where $\sigma'$ is the real part of complex conductivity and $\omega$ is the angular frequency. Therefore, dielectric contribution of bulk oxide layers appears in the CC spectra, i.e., the arcs in the Cole-Cole plots, and characteristic of bulk oxide layers can be identified.

IV. CONCLUSIONS

To summarize, the under-, proper-, and overoxidized MTJs have been systematically studied by employing the equivalent circuit model, composed with $R$ and $C$ components, to describe the metal-oxide interfaces as well as the bulk insulating layer. The interfacial capacitance is indicated from the analysis of equivalent circuit method to be sensitive to the oxidation process of Al layer. The relaxation frequencies of AlO$_x$ and CoFeO$_x$ obtained by the CI and CC techniques are also compared and show similar tendency as a function of Al oxidation process. Both CI and CC spectra are useful to characterize the metal-oxide interfaces and bulk layers of MTJs. Besides, the interfacial roughness effect on electrical and magnetic properties has been observed in other systems.\textsuperscript{14,15} In our MTJ systems, the significant variation of electrical- and magnetotransport behaviors influencing by interfacial roughness also has been systematically observed. The results will be published elsewhere.

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