Characterization of magnetoimpedance on polycrystalline and amorphous chromium oxides bilayered thin films

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The impedance of chemical vapor deposited CrO_2 and Cr_2O_3 bilayered thin films, composed of polycrystalline and amorphous structure, have been systematically studied in function of frequency and temperature. In the polycrystalline-CrO_2/amorphous-Cr_2O_3 bilayer, the real part of impedance at low frequency ($f<300$ kHz) demonstrates a sharp transition at temperature around 330 K, with a specific feature of positive temperature coefficient, similar to the variation of dc resistance occurs at ferroelectric-paraelectric transition in the $BaTiO_3$ ceramics. In contrast, the imaginary part of impedance, at frequency $f>300$ kHz, shows a characteristic of negative temperature coefficient. Further analysis of the frequency dependence of the impedance shows the contribution from the dynamics of both the dielectric and magnetic dipoles in the layers. Comparison of polycrystalline-CrO_2 and amorphous-Cr_2O_3 single layer with the CrO_2/Cr_2O_3 bilayer is discussed. [DOI: 10.1063/1.1455605]

I. INTRODUCTION

The ferromagnet chromium dioxide (CrO_2) is one of the technologically important transition-metal oxides, which is widely used as a particulate recording medium in storage applications. The theoretical band structure studies by Schwarz[1] and others[2] have predicated that CrO_2 is a half-metallic ferromagnet; that is, the majority electrons have a metallic band structure, while the minority ones have a semiconductor-like energy gap at the Fermi surface. The high degree of spin polarization of the electrons at the Fermi level suggests that CrO_2 could be an ideal material for the spin tunnel devices.[3,4] A large value of magnetoresistance has been reported in several experiments on polycrystalline films and powder compacts of CrO_2.[5–8]

Our recent study[9] on the high frequency transport properties of polycrystalline CrO_2 thin films has demonstrated specific feature of low-loss dielectrics distinct from the usual metallic ferromagnet. Interplay of complex magnetic dipoles and dielectrics dynamics may be responsible for the characteristics of impedance in the CrO_2/Cr_2O_3 multilayer. The aim of this work is to study the impedance of chemical vapor deposited CrO_2 and Cr_2O_3 bilayered thin film, in form of polycrystalline and amorphous structure, respectively, to disclose possible phenomenon by dynamic interaction in the composite films.

II. EXPERIMENT AND RESULTS

In this study, the chromium oxide thin films were prepared by the chemical vapor deposition method. The amorphous Cr oxide film was grown on quartz substrate by using $Cr(CO)_6$ as precursor and O_2 as the carrier gas. The deposition pressure is 4 Torr under 30 sccm flowing of O_2, and the deposition temperature is around 300 °C. The powder x ray data show the film to be amorphous. While in the same run, on the Si(100) substrate, which placed at the same deposition zone, the polycrystalline Cr_2O_3 was obtained. Based on this observation, it is assumed the film on the quartz should be amorphous Cr(+3) oxide. The second layer CrO_2 film was grown using CrO_3 as the precursor, with O_2 carrier gas in flowing of 50 sccm. The deposition temperature is 400 °C, while the precursor is placed in the 260 °C zone. The powder x ray has shown the film to be polycrystalline CrO2.

The magnetization of the film was measured with a vibration sample magnetometer (LakeShare VSM7307). Figure 1 shows the temperature dependent spontaneous magnetization $M_s(T)$ of a polycrystalline-CrO_2/amorphous-Cr_2O_3 bi-

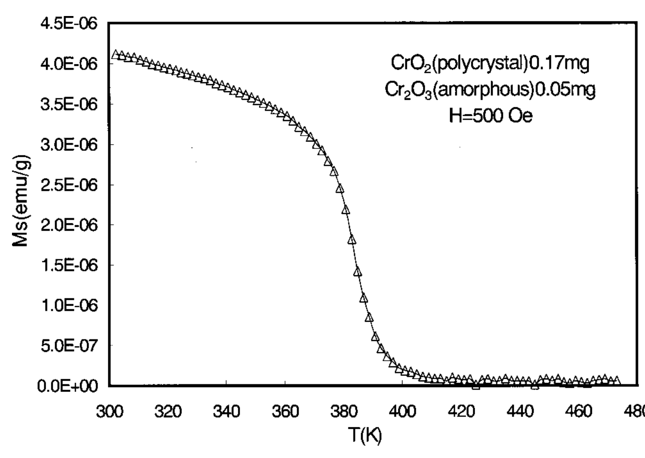

FIG. 1. The temperature dependence of spontaneous magnetization $M_s(T)$ of a polycrystalline-CrO_2/amorphous-Cr_2O_3 bilayer thin film. Experimental data are represented by triangular symbols with a solid guided line.

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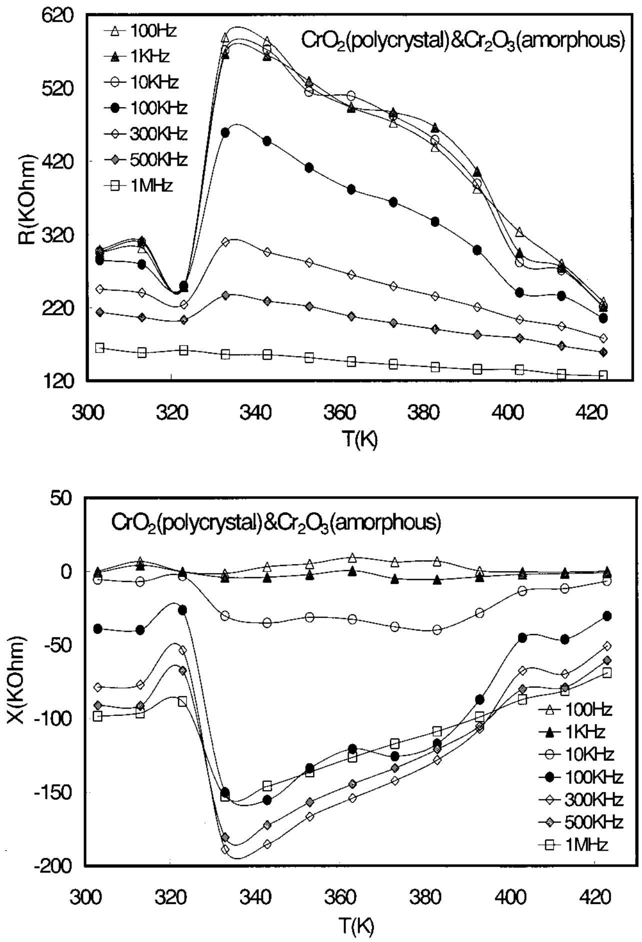

FIG. 2. The temperature dependence of the real and imaginary components of the impedance ($Z=R+iX$) of a polycrystalline-CrO_2/amorphous-Cr_2O_3 bilayer thin film.

layer thin film, (denoted as poly-CrO_2/α-Cr_2O_3). The poly-CrO_2/α-Cr_2O_3 film demonstrates a magnetic phase transition at temperature $T\approx 390$ K. The feature of $M_s(T)$ curve is similar to that of the purity phase poly-CrO_2 thin film with a ferromagnetic-paramagnetic phase transition at Curie temperature $T_C\approx 390$ K.[9] The Néel temperature of antiferromagnetic Cr_2O_3 is around 307 K. The antiferromagnetism is much lessened in the α-Cr_2O_3 layer. The behavior of magnetization of poly-CrO_2/α-Cr_2O_3 bilayer is mainly contributed by poly-CrO_2. By further analysis of $M_s(T)$ data near the transition, the $M_s(T)$ follows Bloch's law, and the calculated magnetic moment is about $2\mu_B$ per Cr atom in the part of poly-CrO_2 layer, designating a typical character of spin polarization in the half-metallic CrO_2 film.[9]

In order to study the high frequency transport properties on the poly-CrO_2/α-Cr_2O_3 bilayer, the impedance of the thin film was measured by a HP4284A impedance analyzer with a frequency range from 20 Hz to 1 MHz. Figure 2 shows the temperature dependence of the real and imaginary components of the impedance ($Z=R+iX$) for the poly-CrO_2/α-Cr_2O_3 thin film. The observed $Z(T)$ reveals several specific features in the real and imaginary components. First, at low frequency ($f<100$ kHz) and high temperature ($T>340$ K), the temperature dependence of the real components $R(T)$ decreases linearly with increasing temperature. Second, an abrupt change of R occurs at a temperature range between 320 and 340 K. The $R(T)$ increases steeply with increasing temperature starting from $T\approx 320$ K, and reaches a maximum value at $T\approx 335$ K. Further increasing of T, $R(T)$ decreases. Third, the specific variation of $R(T)$ is gradually smeared out as frequency is increased. At $f>1$ MHz, the $R(T)$ curve is almost linearly changed, in a slow negative slope with increasing temperature.

On the other hand, the temperature dependence of the imaginary components of the impedance $X(T)$ display a distinct feature. At high frequency $f>100$ kHz and high temperature $T>340$ K, the temperature dependence of the imaginary components $X(T)$ increases linearly with increasing temperature. A change of derivative $dX(T)/dT$, from negative to positive, occurs with increasing temperature from 320 to 340 K. The $X(T)$ curve exhibits a minimum at $T\approx 335$ K. The feature of abrupt variation of $X(T)$, appeared as a "V" shape, is less pronounced at low frequency ($f>300$ kHz).

It is noticed that the $R(T)$ and $X(T)$ transition temperature ($T\approx 330$ K) is far off the $M_s(T)$ transition of poly-CrO_2/α-Cr_2O_3 bilayer. While in a single poly-CrO_2 thin film reported earlier,[9] the $R(T)$ usually shows a smooth crossover in the slope at temperature near T_C, and the curve of $R(T)$ follows a simple model of $R(T)=R_0+\alpha T^2 \exp(-\Delta/T)$ at low frequency. We have also examined the feature of temperature dependent impedance $Z(T)$ of a single α-Cr_2O_3 thin film. Both real and imaginary components of the impedance are insensitively varied in a board temperature of 290–400 K [the $R(f)$ is shown in the inset of Fig. 3]. To understand the mechanism underlying the phenomena of the specific feature of impedance in the poly-CrO_2/α-Cr_2O_3 bilayer thin film, we have performed systematical measurement of the frequency dependence of the impedance.

Figure 3 displays the frequency dependence of the real and imaginary components of the impedance for the poly-CrO_2/α-Cr_2O_3 thin film, in a frequency range from 20 Hz to 1 MHz at various temperatures. The $R(f)$ decreases monotonically with frequency and exhibits negative slopes. On the other hand, the $X(f)$ show positive slopes with increasing frequency at low T (<330 K). The variation of impedance spectra in each temperature appears to be complicated, nevertheless, it is in good corresponding with the $Z(T)$ curves shown in Fig. 2. We have analyzed the impedance spectra in the poly-CrO_2/α-Cr_2O_3 thin film based on fundamental electrodynamics.[9] The impedance Z of electromagnetic fields propagating in a continuous media is expressed in function of frequency ($f=\omega/2\pi$), permittivity ϵ and magnetic permeability μ: $Z=[\iota\omega\mu/(\sigma+\iota\omega\epsilon)]^{1/2}$, where σ is the conductivity of the matter. In case of low loss dielectrics ($\sigma/\omega\epsilon\ll 1$), the expression of impedance can be rewritten as $Z\cong(\mu/\epsilon)^{1/2}$. It is assumed the poly-CrO_2/α-Cr_2O_3 bilayer to be a low loss dielectric media, in which the dielectric response and dynamical magnetization could be responsible for the impedance spectra. When the sample is subjected to an electromagnetic field, the dielectric constant can be represented by $\epsilon=\epsilon_0[(\kappa_0-1)/(1+\iota\omega\tau_E)+1]$, where κ_0 is the static dielectric constant and τ_E is the dielectric relaxation

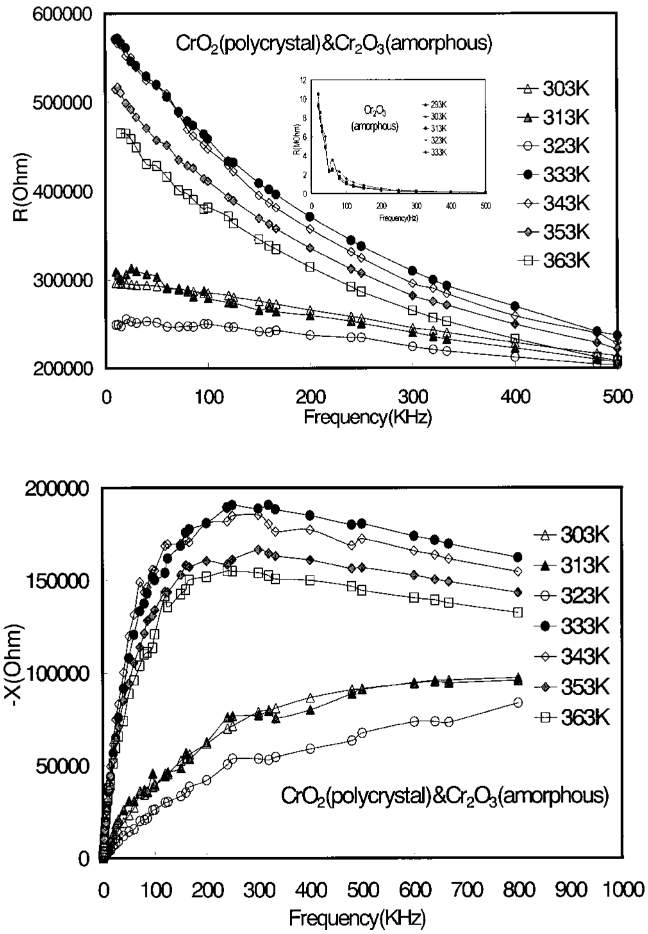

FIG. 3. The frequency dependence of the real and imaginary components of the impedance ($Z = R + iX$) of a poly-CrO_2/α-Cr_2O_3 thin film. [The axis of frequency is represented in different scales on $R(f)$ and $X(f)$ for clear view of feature.]

time. The magnetic permeability can be written as $\mu = \mu_0[\chi_0/(1+\iota\omega\tau_H)+1]$, where χ_0 is the initial susceptibility and τ_H is the relaxation time due to magnetization response. Thus, impedance Z is expressed by

$$Z = (\mu_0/\epsilon_0)^{1/2}\{[1+\chi_0/(1+\iota\omega\tau_H)]/[1+(\kappa_0-1)/(1+\iota\omega\tau_E)]\}^{1/2}. \tag{1}$$

With the earlier expression, we can obtain good agreement of the experimental data with the theoretical model for $T \approx 303-323$ K. The result implies that the impedance of the poly-CrO_2/α-Cr_2O_3 bilayer might be correlated to the dynamics of dielectric and magnetic dipoles in the film.

It is noticed that (Fig. 3) the impedance spectra at $T \approx 303-323$ K reveal different features compared to those at $T > 333$ K, and the latter cannot be well described by Eq. (1). Further inspection on single α-Cr_2O_3 film, at $T \approx 290-400$ K, has shown (inset of Fig. 3) that the $R(f)$ curves have collapsed into unique curvature, so too for the $X(f)$ curves. Thus, the anomalous variation of the $Z(T)$ and $Z(f)$ of poly-CrO_2/α-Cr_2O_3 bilayer may not be simply due to each single layer. The mechanism remains unclear so far. The interface between the poly-CrO_2/α-Cr_2O_3 could play a crucial role.

Furthermore, it is well known that donor-doped $BaTiO_3$ ceramics exhibit a rapid resistance rise at the Curie temperature, known as the positive temperature coefficient of resistivity (PTCR).[10] The PTCR effect is regarded as an increase in the grain boundary potential barrier height with a rapid drop in dielectric constant at the transition of the ferroelectric tetragonal phase to the paraelectric cubic one. In the poly-CrO_2/α-Cr_2O_3 bilayer, the real part of impedance demonstrates a specific feature of positive temperature coefficient, while the imaginary part of impedance shows a negative temperature coefficient, at $T \approx 320-340$ K. The observed phenomenon may be associated with the interfacial layer of complex coupling between polycrystalline and amorphous structures, or related with half-metal/insulator surface trap state and carries injection. Further investigation of electrical conduction and dielectric properties is needed to clarify underlying mechanism.

ACKNOWLEDGMENT

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