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Magnetic field modulated dielectric relaxation behavior of Pt/BiScO₃-PbTiO₃/La_{0.7}Sr_{0.3}MnO₃ heterostructure in metal-insulator transition region: An equivalent-circuit method

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Utilizing temperature and magnetic field dependent dielectric spectroscopy, two different dielectric relaxation processes, both accompanied with prominent positive magnetodielectric (MD) effects, were observed in Pt/BiScO₃-PbTiO₃/La_{0.7}Sr_{0.3}MnO₃ (Pt/BSPT/LSMO) heterostructure. An equivalent circuit model is established to quantitatively describe the relaxation data well. The simulation results elucidated that one relaxation process can be attributed to magnetic order associated BSPT/LSMO interfacial relaxation with abnormal dynamics feature. Another one is due to the external contact interface contributions. Both relaxation dynamics can be tuned, via magnetic field controlled localization of free interfacial charges, to induce MD effects in BSPT/LSMO interfaces and nonmagnetic Pt/BSPT contacts. © 2011 American Institute of Physics. [doi:10.1063/1.3667289]

I. INTRODUCTION

Multiferroic films have currently attracted considerable scientific interests for the prospect of magnetoelectric (ME) effect, fuelled by both the underlying new fundamental physics and the potential technological applications in magnetic sensors and multi-state memories, etc.¹ Because there is mainly scanty single-phase multiferroics at room temperature in nature due to mutually excluding of forming traditional ferromagnetic (FM) and ferroelectric (FE) orders in a single compound,² numerous research has focused on various artificial heterostructures, such as multilayers,³ superlattices,⁴ and nanocomposites,⁵ which are simultaneously constructed by individual FE and FM compounds. In general, the main intrinsic mechanisms that can give rise to ME and/or magnetodielectric (MD) effects in the heterostructures are elastic coupling in piezoelectric/magnetostrictive systems,⁶ hybridization between *3d* orbitals of interfacial atoms in FM metal/FE superlattices,⁷ and coupling of magnetic spins and electric dipole in FM nanoparticle/FE nanocomposites.⁵ Otherwise, an extrinsic mechanism was proposed that a MD effect also can be yielded by the combination of magnetoresistance (MR) and Maxwell–Wagner (MW) interfacial relaxation without intrinsic ME coupling.⁸ Moreover, this extrinsic MD mechanism driven by localization of free charges at the interfaces has been extended to a broad range of inhomogeneous systems, which some times is even free from magnetic composites.⁹ Hereby, such MD effects could be achieved readily and may be more practical for techno-

logical applications.^{9,10} Although there are also many experimental investigations on these interfacial polarization dominant MD effects,^{4,5,11} further quantitative correlations between dynamics processes of MW relaxation and magnetic related behaviors are required to be established, providing important insights on the details of interfacial free charge localization processes under different magnetic order and associated phase transitions and competitions.

Recently Zhang *et al.* roughly discussed the MD characteristics in a new FE-FM system of BiScO₃-PbTiO₃/La_{0.7}Sr_{0.3}MnO₃ (BSPT/LSMO) heterostructure at room temperature.¹² In this study, we present the temperature and dc magnetic field dependent dielectric spectroscopy of BSPT/LSMO heterostructure. A careful quantitative analysis on dielectric characteristics over metal-insulator (M-I) transition range of LSMO under various magnetic field was performed, utilizing an equivalent circuit model. Interestingly, two types of magnetic-field-modulated dielectric relaxation feature were observed in different frequency region. The related physical mechanisms behind these dielectric relaxation features were addressed in detail.

II. EXPERIMENT

BSPT thin films were synthesized using LSMO as bottom electrodes on silicon substrates via chemical solution deposition (CSD).¹² Before the dielectric properties measurements were carried out, capacitor structures were fabricated with sputtered Pt top electrodes (diameter of ~150 μm), which were led out by ultrasonic welding. The temperature and the magnetic field variation in dielectric properties as function of frequency were performed using a broadband dielectric

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spectrometer (BDS40, Novocontrol, Germany) combined with physical property measurement system (PPMS-9, Quantum Design, USA) from 50 to 300 K. The BSPT/LSMO was placed parallel to the direction of magnetic field mounted on PPMS sample puck. dc transport characteristics for LSMO/Si films were measured using standard four-point technique.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the frequency dependence of complex dielectric permittivity on BSPT/LSMO thin film at various temperatures. The real part of permittivity ϵ' illustrated in Fig. 1(a) exhibits obvious dielectric dispersion and a large temperature dependence, viz. ϵ' decreases with the increase of frequency and enhances on the elevated temperature. Meanwhile, when the temperature was cooled down to

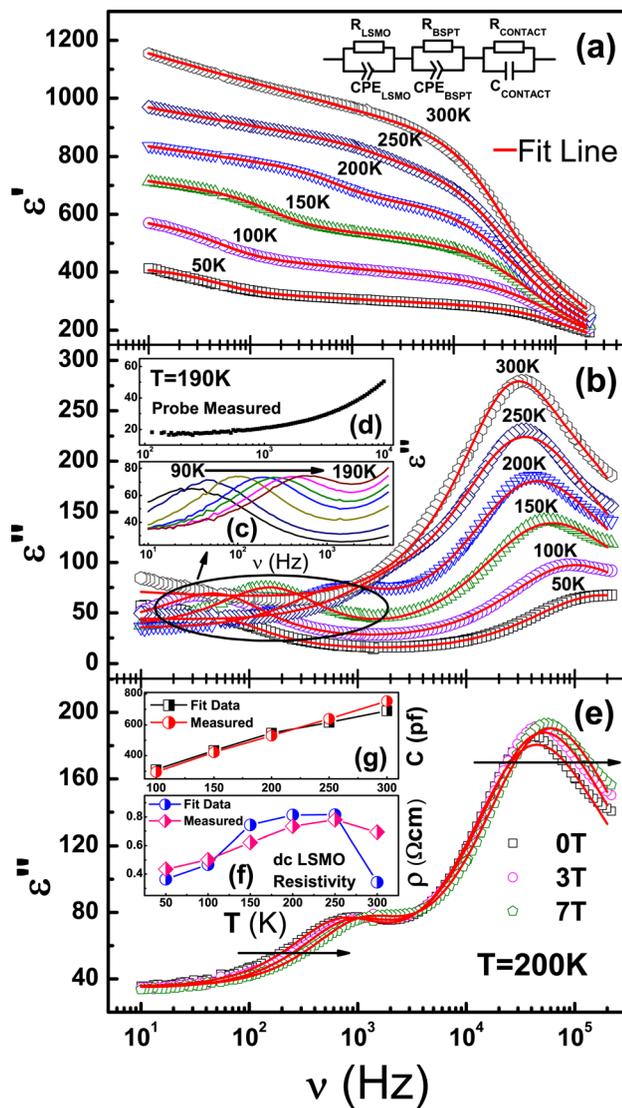


FIG. 1. (Color online) The (a) real part (ϵ') and (b) imaginary part (ϵ'') of permittivity as function of frequency for BSPT/LSMO heterostructure at various temperatures. The inset shows the CPE equivalent circuit. Solid lines: overall response fitted by equivalent circuit; (c) frequency-dependent of ϵ'' in low-frequency region from 90 to 190 K; (d) the dielectric spectrum with probe contact at 190 K; (e) the relaxation characteristic under magnetic field at 200 K; (f) dc resistivity characteristic of LSMO; and (g) capacitance of BSPT/LSMO for fitting and measured data at elevated temperature.

approximate 200 K, two successive steps in the ϵ' spectrum could be observed at high and low frequency regions, respectively. These step-like features in ϵ' are accompanied by two broad relaxation peaks in imaginary spectrum ϵ'' (Fig. 1(b)): the first low frequency region extends over range of 10 Hz to 3 kHz, and the second high frequency region spreads from 10 to 200 kHz. For the former region, when $T < 200$ K, the relaxation frequency corresponding to the peak sites shifts monotonously toward higher frequencies with increasing temperature (Fig. 1(c)), but the relaxation peaks fade gradually with further increasing temperature. In the latter region, compared with the relaxation behavior in the first region, more distinct high-frequency peaks appear in the ϵ'' spectrum and throw out a different frequency dependence as a function of temperature, shifting toward lower frequencies with an increase in temperature but moving to the opposite direction when $T > 250$ K. To check these relaxation behaviors subjected to magnetic field, Fig. 1(e) is plotted as a typical relaxation characteristic as the function of magnetic field at 200 K. It is indicated that the above mentioned two different dielectric relaxations conduct a similar type of frequency dependence as peak sites shifting toward higher frequencies totally upon increasing magnetic field from 0 to 7 T. According to the above experimental evidences, the kinetics of low-frequency relaxation is totally different from the high-frequency counterpart, but both yield the same magnetic-field-dependent behaviors. What is physical mechanism behind these dielectric relaxations?

In general, the presence of abundant heterogeneous interfaces is a distinctive feature of heterostructures. And these interfaces are always constructed by electrical heterogeneity of two different phases or media that are connected in series. When ac electrical current passes through inhomogeneous interfaces, the free charged carriers would be localized and accumulate at the interfaces between two media with different conductivities. This process could give rise to a Debye-like relaxation, termed Maxwell-Wagner interfacial relaxation.^{13,14} Assuming an ideal two-phase system, the relaxation time of heterostructure $\tau \approx C_1 R_2$, where C_1 and R_2 are the larger capacitance and smaller resistance in two components.¹⁴ Usually, because C_1 has weak temperature dependence and R_2 displays semiconducting temperature characteristics, i.e., decreasing exponentially with temperature, the relaxation dynamics is mainly determined by R_2 , and therefore typical frequency-dependent relaxation features (ϵ' steps and ϵ'' peaks) undergo a prominent shift to low frequency when temperature is reduced.¹⁴ In the present case, Pt/BSPT and BSPT/LSMO interfaces are the primary interfaces in this system and possibly play a crucial role in dielectric relaxations. As for the former, because of the obvious diversity of the work functions between metal and ultrasonic-point-welt-altered surfaces of ferroelectric films, the prominent Schottky barriers or diodes would be induced, leading to depletion layers at Pt/BSPT interfaces and then result in low-frequency MW relaxation, which has been reported in many colossal dielectric constants (CDCs) materials.¹⁵ This depletion layer always exhibits thermal-activated conductivity, and the above referred normal relaxation dynamics occurs, as low-frequency relaxation does here. We also checked the dielectric spectrum with probe

contact, and this low-frequency relaxation disappeared in same temperature region (Fig. 1(d)). Hence, Pt/BSPT interfacial polarization should correspond with the reasonable origin of low-frequency relaxation. Because high-frequency-relaxation behaves totally different than relaxation dynamics compared with that of the normal mode of MW relaxation, at first glance, it appears that it cannot contribute to MW relaxation of BSPT/LSMO interfaces. However, keep in mind that as lower resistivity is part of BSPT/LSMO interfaces, the temperature characteristic of LSMO resistivity mainly dominates the frequency dependent shifts of relaxation, and it is sensitive to magnetic order, associated magnetic order transitions, and phase competitions in LSMO rather than simple thermal-activated kinetics.¹⁶ As shown in Fig. 1(f), the dc resistivity behavior of polycrystalline LSMO films climbs up at the temperature achieved 250 K and exhibits a drop for the further increasing temperature, corresponding to the transition between the double exchange of the $\text{Mn}^{3+}\text{-O}^{2-}\text{-Mn}^{4+}$ configuration caused FM metallic order and Jahn–Teller-distortion-associated electron-phonon coupling governed polaron PM insulator order at 250 K.¹⁶ From Fig. 1, it is found that the temperature dependent frequency shifts of high-frequency relaxation rightly fall coincident with the charge transport variation of LSMO in M-I transition region. Thus, it is highly possible that the observed high-frequency relaxation can be regarded as MW relaxation of BSPT/LSMO interfaces.

To further clarify the essence of dielectric relaxations systematically, a modified constant phase element (CPE) equivalent circuit model (ECM) was introduced to fit dielectric spectroscopy.¹³ The model contains three parallel RC elements in series, representing the responses of electrode contact, BSPT and LSMO films, respectively, as illustrated in the inset of Fig. 1(a). CPE, viz. $Z_{\text{CPE}}^* = 1/A(j\omega)^n$, describes the deviation from ideal capacitance, where A and n are constants.^{17,18} Herewith, the total impedance of model is as follows: $Z^* = (1/R_c + jC_c\omega)^{-1} + (1/R_{\text{BSPT}} + A_1(j\omega)^\alpha)^{-1} + (1/R_{\text{LSMO}} + A_2(j\omega)^\beta)^{-1}$, and the total capacitance is obtained by $C^* = (j\omega Z^*)^{-1}$. It can be found in Fig. 1 that the experimental data without/with magnetic field are fitted quite well in the whole measurement range. From the simulation results, the estimated capacitance of BSPT/LSMO thin film is consistent with direct measured value of Pt/BSPT/Pt thin film (Fig. 1(g)). α is estimated to be 0.94–0.97 as Morrison *et al.* reported in PZT films.¹⁸ From the fitting, calculated values of ρ_{LSMO} were presented in Fig. 1(f). Compared with zero field transport measure values, both show a similar temperature dependence of transport and a M-I transition at 250 K. Meanwhile, in Fig. 2(a), the effect of applied magnetic field on high-frequency relaxation dynamics at 250 K is also well depicted by ECM. The fitting value of R_{LSMO} reduces from 5995 Ω at 0 T to 4468 Ω at 7 T, corresponding to distinguished negative MR effects of LSMO systems, rising from the suppression of spin fluctuation and/or spin-polarized tunneling under magnetic field.¹⁹ The values of MR, defined as $(\rho_H - \rho_{0T})/\rho_{0T}$ (%), from the fitting (Fig. 2(b)) are comparable with the direct measured values (e.g., calculated MR $\sim -12.33\%$ versus measured MR $\sim -14.37\%$ at 3 T). Trivial deviation may originate from elastic coupling in piezoelectric/magnetostrictive systems here. Therefore, the high-

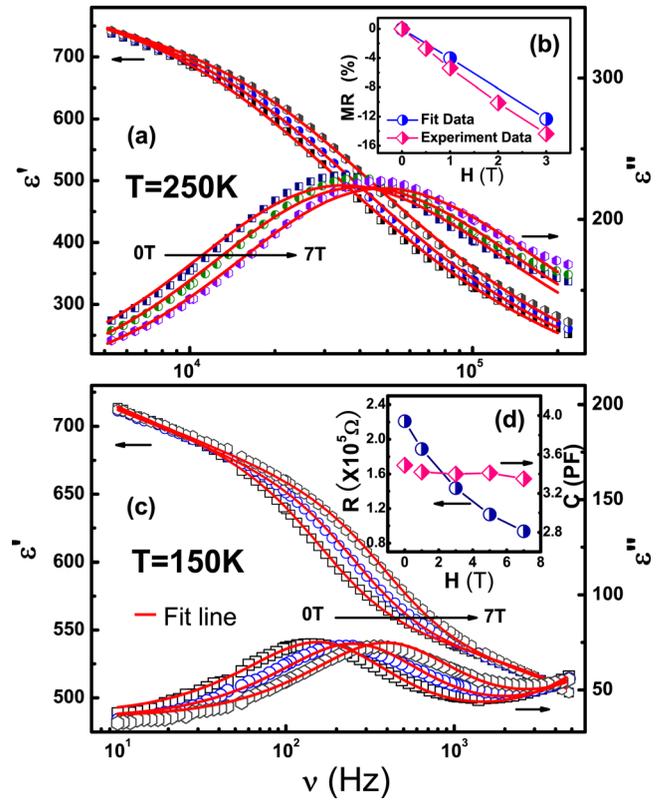


FIG. 2. (Color online) (a) Frequency and magnetic field dependence of dielectric behavior measured from 10 000 to 200 000 Hz at 250 K; (b) the magnetic field variation in MR for fitting and experiment data, respectively; (c) the dielectric behavior under magnetic field in low-frequency region at 150 K; (d) variation of the fitted resistance and capacitance under magnetic field.

frequency relaxation dynamics indeed have a close correlation with the magnetic transport behavior of LSMO, and a combination of negative MR and MW relaxation in BSPT/LSMO interface is the predominate reason for the observed high-frequency MD effect (maximum $\sim 11\%$ at 250 K).

As for the low-frequency MD effect here, magnetic field tuned relaxation dynamics are similar to that of high-frequency relaxation, and good fitting results are also obtained as illustrated in Fig. 2(c), so as to further validate Pt/BSPT interfacial polarization. This type of MD only comes from nonmagnetic Schottky barriers, just as reported MD in Au/GaAs:Si junctions.²⁰ A general concept of interface driven MD has been extended to nonmagnetic interface systems, such as diodes, grain boundaries, and even FE domain walls, by Maglione.⁹ Here, it also can be attributed to interactions between the free charges accumulated at the barrier interface and magnetic field.⁹ The dynamical motion of these interfacial charges is dominated by an external magnetic field via certain interactions, e.g., Lorentz force controlled charge trajectories, so that the resistance and/or thickness of barrier depletion layers are accordingly altered as fitting resistance displaying in Fig. 2(d). And in turn, the MW relaxation time and kinetics is tuned by magnetic field to consequently induce this relaxation related MD effect. However, a detailed process of interactions between interfacial charge and magnetic field is necessary to further investigate systematically.

IV. CONCLUSIONS

To summarize, two types of dielectric relaxation processes were observed in the Pt/BSPT/LSMO heterostructure. An equivalent circuit model is well established to describe the relaxation data and attributed one relaxation process to magnetic order associated BSPT/LSMO interfacial relaxation. Another one could be due to the external contact contributions. Both relaxation dynamics can be tuned by magnetic field to induce prominent positive MD effects.

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