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Temperature-independent ferroelectric property and characterization of high-T$_C$ 0.2Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-0.8PbTiO$_3$ thin films

Linxing Zhang (张林兴), Jun Chen (陈骏), Hanqing Zhao (赵翰庆), Longlong Fan (樊龙龙), Yangchun Rong (戎阳春), Jinxia Deng (邓金侠), Ranbo Yu (于然波), and Xianran Xing (邢献然)

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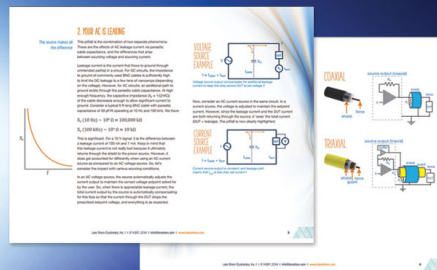Applied Physics Letters **86**, 262905 (2005); 10.1063/1.1978980

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Temperature-independent ferroelectric property and characterization of high-T_C 0.2Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-0.8PbTiO$_3$ thin films

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Ferroelectric property stability against elevated temperature is significant for ferroelectric film applications, such as non-volatile ferroelectric random access memories. The high-T_C 0.2Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-0.8PbTiO$_3$ thin films show the temperature-independent ferroelectric properties, which were fabricated on Pt(111)/Ti/SiO$_2$/Si substrates via sol-gel method. The present thin films were well crystallized in a phase-pure perovskite structure with a high (100) orientation and uniform texture. A remanent polarization ($2P_r$) of 77 μC cm^{-2} and a local effective piezoelectric coefficient d_{33}^* of 60 pm/V were observed in the 0.2Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-0.8PbTiO$_3$ thin films. It is interesting to observe a behavior of temperature-independent ferroelectric property in the temperature range of room temperature to 125 °C. The remanent polarization, coercive field, and polarization at the maximum field are almost constant in the investigated temperature range. Furthermore, the dielectric loss and fatigue properties of 0.2Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-0.8PbTiO$_3$ thin films have been effectively improved by the Mn-doping. © 2013 AIP Publishing LLC.
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Thin films of ferroelectric materials with a perovskite (ABO$_3$) structure have been arousing widespread concerns, because they exhibit remarkable achievements in the technology of ferroelectric and their potential applications, such as non-volatile ferroelectric random access memories (FeRAM).[1–3] To date, a great need of the industry for memories has been raised for ferroelectrics for modern usage at temperatures as high as possible. High Curie Temperatures (T_C) could guarantee operation at high temperature. The relative low T_C of the conventional ferroelectrics materials (usually lower than 386 °C of Pb(Zr$_{1-x}$Ti$_x$)O$_3$ (PZT)) limits their applications.[4] For applications with stability of structure and performance under specific temperature conditions, the search for bismuth-based perovskite ferroelectric materials has stimulated much interest due to the desirable behavior of high T_C, such as promising system of BiMeO$_3$-PbTiO$_3$, where Me could be single trivalent cation (e.g., Sc^{3+}, In^{3+}, and Fe^{3+}) or a mixed cation combination with an average trivalence (e.g., Zn$_{1/2}$Ti$_{1/2}$ and Mg$_{1/2}$Ti$_{1/2}$).[5–16] The improved ferroelectric performances in the BiMeO$_3$-PbTiO$_3$ are ascribed to the strong hybridization between oxygens and A-site Pb/Bi or B-site cations which have a strong ferroelectric activity, such as Ti and Zn.[17,18] Bi substitution plays a unique role in considerable enhancement on both large polarization and high T_C.[7] For instance, BiScO$_3$-PbTiO$_3$ (BS-PT) solid solutions exhibit an excellent high temperature performance (T_C = 450 °C and P_r = 74 μC cm^{-2}), which would be the best high temperature ferroelectric materials so far.[5,19–21] However, in the case of high cost scandium sources, the broad and potential device applications of BS-PT is severely restricted.

A significant alternative system of BiMeO$_3$-PbTiO$_3$ is involved in the Bi(Mg$_{1/2}$Ti$_{1/2}$)O$_3$-PbTiO$_3$ (BMT-PT), which has being considerably studied due to its good performances such as high T_C, low cost, zero thermal expansion, and other good properties.[9–13] Especially, it was found that the ferroelectric performance of BMT-PT ceramics is enhanced at elevated temperatures, which benefits applications of devices. It is ascribed to improved domain switching mobility which is associated with reduced lattice distortion.[11] Furthermore, BMT-PT exhibits zero thermal expansion over a wide temperature range (RT $\sim$ 500 °C), which benefits to reduce thermal shock and enhance the reliability of devices during application with thermal fluctuation.[9] For the consideration as high-T_C ferroelectric films, the temperature dependence of ferroelectric properties is most crucial for applications. However, most studies on thin films have only been relevant to structure and ferroelectric properties at room temperature or lower temperature.[12,15,22] Studies on high temperature performance are very rare. Therefore, it is necessary to investigate the temperature dependence of ferroelectric property of thin films. The present study selects $(1-x)$BMT-xPT to fabricate high-T_C thin films. The present adopted compositions of $(1-x)$BMT-xPT ($x = 0.8$) is focused not at the MPB compositions ($x = 0.37$) as previous studies normally did,[12] but at the tetragonal phase, since the composition of $x = 0.8$ is at the highest T_C zone. The composition of 0.2BMT-0.8PT would show higher thermal stability of ferroelectric properties.[10]

In this work, (100)-oriented 0.2BMT-0.8PT films were fabricated on Pt(111)/Ti/SiO$_2$/Si substrates by a sol-gel deposition method. The dielectric, ferroelectric, piezoelectric, leakage current, and fatigue properties were investigated. Particular attention was paid to study the temperature dependence of ferroelectric property. 0.2BMT-0.8PT

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films show comprehensive electric properties and especially the temperature independent remanent polarization property.

The 0.2BMT-0.8PT thin films exhibit pure perovskite phase, when annealed at 650 °C (Fig. 1). It was found that the 0.2BMT-0.8PT thin films can be crystallized in a pure perovskite structure by sintering at a temperature range of $600 \sim 700$ °C. This result shows that the 0.2BMT-0.8PT thin films, consisting of two end members of tetragonal $PbTiO_3$ and metastable perovskite orthorhombic $Bi(Mg_{1/2}Ti_{1/2})O_3$,[23] are stable in the perovskite structure. In the present study, PT thin films are taken as a reference for the 0.2BMT-0.8PT thin films.

The thin films of 0.2BMT-0.8PT have a highly preferential texture. The first peak at $2\theta = 22.54°$ of thin film should be indexed as (100) profile, since it is close to the (100) peak of bulk powders with the same composition. Due to the fact that the intensity maximum of the films appears at the (100) peak, only a weak (101)/(110) peaks and absence of (001) peak, it indicates that a highly (100)-oriented film was obtained (Fig. 1). Furthermore, the first peak in the XRD of 0.2BMT-0.8PT thin films is also close to the (100) peak of PT thin films, also indicating the (100) orientation property of the 0.2BMT-0.8PT thin films. The level of orientation can be described by the Lotgering factor (*f*).[24] In view of a large value of *f* (0.82), the 0.2BMT-0.8PT thin films possess a good texture. It is known that there are a series of factors resulting in preferential orientation, such as, lattice match,[25] surface energy of planes,[26] temperature of dry and pyrolysis, and interface layer of PbO.[25,27,28] The (100) preferential orientation of the present films under the present treatment process could be due to the lower surface energy of the (100) planes resulted from the difficult formation of *c*-axis perpendicular to the growth face with a large tetragonality (*c/a*) of 0.2BMT-0.8PT and PT films by RTP. The highly orientation of films could improve ferroelectric and mechanical properties. Additionally, in terms of the sharp profile of (100) peak, the 0.2BMT-0.8PT films should be well crystallized.
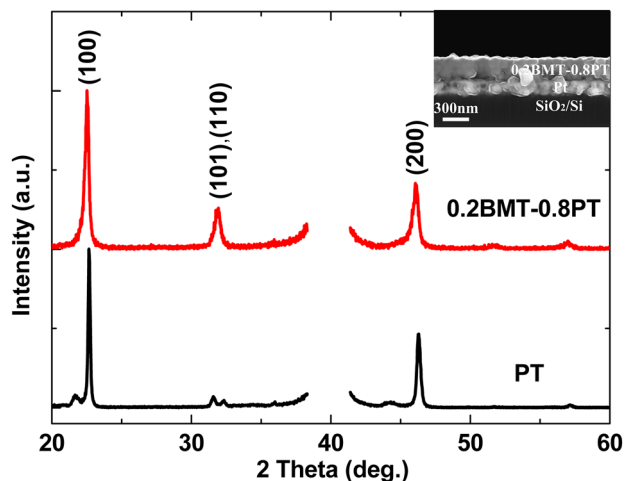

FIG. 1. XRD patterns of 0.2BMT-0.8PT and PT films. XRD of PT film is adopted for a comparison. The inset shows the cross-sectional image of the 0.2BMT-0.8PT thin films.

The change in the *c/a* of poly-domain 0.2BMT-0.8PT thin films could be observed in the XRD patterns (Fig. 1), according to the movement of (100) profile. Only one diffraction profile could be observed for the tetragonal diffraction index of {100}, and it has a small movement to the lower angle by comparison with bulk powders, indicating the *a(b)* axis has a small increase in the thin film. It should be expected that the (001) profile shifts to high 2θ position, even though it is not shown in the XRD pattern. Thus, the *c/a* of 0.2BMT-0.8PT should be reduced in the thin film which should be smaller than the value of *c/a* = 1.059 in the bulk powder state. A similar result was also revealed in the PT thin films, which is confirmed by uplifted (001) peaks and descending (100) peaks in the XRD patterns. The PT thin films have a reduced *c/a* compared with the PT bulk powders. The reduction in *c/a* could arise from size effect and interfacial stress.[29,30] The film stress would exist in the 0.2BMT-0.8PT thin films, deriving from thermal expansion mismatch and incomplete relaxation of lattice mismatch between the films and the substrates. Additionally, the residual films stress is reported to be more pronounced in the *a*-axis domains than *c*-axis domains.[29] So the present highly (100)-oriented films with more *a*-axis domains would exhibit a reduced *c/a*. A similar phenomenon was also observed in $PbTiO_3$ nano-powders and thin films due to the effect of small grain.[30]

The inset of Fig. 1 shows the cross-sectional image of the 0.2BMT-0.8PT thin films. The SEM micrograph indicates that the different layers between substrate and film are clearly visible except Ti layer with a thickness ∼220 nm of the present films. The SEM surface micrographs exhibit a uniform grain size with approximately ∼100 nm,[31] which would be easy to polarization switch due to the existing of predominantly multi-domains in the grain.[32]

According to lattice dynamical theory, Raman scattering spectra of the present films could be associated with rapidly determining the change of spontaneous polarization (P_S), as shown in Fig. 2. The four peaks of A_1(1TO), E(2TO), $B_1 + E$, and A_1(2TO) are identified corresponding to the one reported for PT single crystals with those lying at 148 cm^{-1}, 220 cm^{-1}, 290 cm^{-1}, and 360 cm^{-1},[33] which indicates that the local structure of the 0.2BMT-0.8PT thin films are the same to tetragonal perovskite. Note that there are three peaks at 110 cm^{-1}, 123 cm^{-1}, and 143 cm^{-1} below 150 cm^{-1} in Raman spectra of PT films. It was reported that anharmonicity of the double-well potential can lead to line shapes composed of multiple subpeaks of A_1(1TO) in ferroelectric PT.[34] The main peak is A_1(1TO) at 143 cm^{-1}, and the other two are the subpeak of A_1(1TO) at 110 cm^{-1} and the peak of E(1LO) at 123 cm^{-1}, respectively.[35] The positions of Raman peaks for the PT films with A_1(1TO), E(2TO), $B_1 + E$, and A_1(2TO) located at 143 cm^{-1}, 208 cm^{-1}, 287 cm^{-1}, and 340 cm^{-1}, respectively, shift to the low frequency side from those observed for PT single crystals. This suggests that the Raman peaks shift would be ascribed to stress effect in films confined by substrates. Similar phenomenon has been observed in crystal samples with the experiment of hydrostatic pressure.[29,36,37] Hence, the *c/a* should be changed by substrate induced strain in the films due to lattice mismatch and thermal expansion
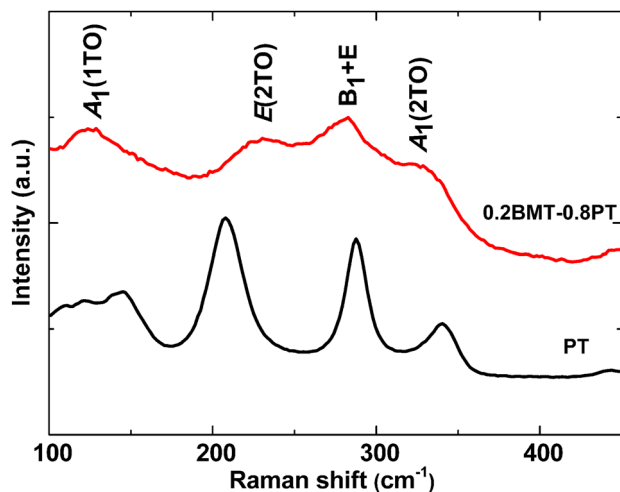
FIG. 2. Raman scattering spectra of 0.2BMT-0.8PT and PT films.

mismatch between the films and the substrates. It should be also true in the 0.2BMT-0.8PT films. The Raman peaks shift of PT films may also be partly due to the simultaneous contribution of size effect on the Raman frequency.[38] These are consistent with the XRD discussion as described previously. Taken together, the reduced c/a of PT and 0.2BMT-0.8PT films is attributed to both stress effect and size effect by Raman and XRD measurements. It is well known that the movement and switching of non-180° walls involve a significant change in c/a. In 0.2BMT-0.8PT thin films with high (100) orientation, the dominant domain would be the non-180° domain, notably 90° domain. Hence, the reduced c/a could enhance the active switchable ferroelectric domains, which would improve the remanent and maximum polarization.[11,39]

The A_1(1TO) mode consists of the vibration of BO_6 octahedra relative to A-site ions parallel to the P_S direction (c-axis). The A_1(1TO) is a useful vibration mode to quickly indicate the variation in P_S of PT-based compounds not only with normally reduced but also enhanced c/a.[8] In the A_1(2TO) mode, B-site ions move in the c-axis direction relative to A-site and oxygen ions, which is sensitive to the B-site P_S displacement.[40] To better understand the P_S variation of the 0.2BMT-0.8PT films, detailed comparisons of the frequency of the soft mode were carried out with PT thin films. The Raman active modes of A_1(1TO) and A_1(2TO) of the 0.2BMT-0.8PT films become soft. The downward shift reveals a reduced P_S according to the lattice dynamical theory. The decrease would be due to the strong negative quadratic effect of A-B coupling with the inclusion of Mg

ions in the 0.2BMT-0.8PT films.[10] From the above, we believe that the 0.2BMT-0.8PT films should have both reduced c/a and P_S.

With the characteristic analysis above, it is interesting to investigate the electric properties of 0.2BMT-0.8PT film. The ferroelectric, conductive, dielectric, and piezoelectric properties were characterized by polarization-electric field (P-E), leakage current-electric field (J-E), permittivity (dielectric loss)-frequency, and displacement-voltage measurements, respectively. Well-saturated hysteresis loops for the highly (100)-oriented 0.2BMT-0.8PT films were measured at room temperature (Fig. 3(a)). It can be clearly seen the growth of P-E loop at a typical low-frequency of 100 Hz with increasing applied electrical fields. A saturated polarization up to 54 μC cm^{-2} was observed. A remanent polarization ($2Pr$) of 77 μC cm^{-2} and a coercive field (Ec) of about 121 kV cm^{-1} were calculated for the 0.2BMT-0.8PT films, which are comparable with 0.25Bi(Ni$_{1/2}$Ti$_{1/2}$)O$_3$-PbTiO$_3$ thin films ($Pr \sim 40.9 \mu$C cm^{-2} and $Ec \sim 117$ kV cm^{-1}) with the same orientation and tetragonal composition.[41] The high Ec of the 0.2BMT-0.8PT films would be due to the factors of small grain size, interface stress, the (100) preferential orientation with many 90°-domains, and the relatively large c/a when compared with those compositions at MPB. As mentioned, well-saturated hysteresis loops and good polarization could be ascribed as follow: First, the (100) preferential orientation indicates very good texture and homogeneous microstructure; Second, more importantly, the high-T_C 0.2BMT-0.8PT thin films have a reduced c/a compared with bulk powders, which would improve domain wall mobility. Note that the 0.63BMT-0.37PT thin films near the MPB composition did not show saturated hysteresis loop owing to the leakage current, and exhibit a relatively low P_r (17.8 μC cm^{-2}).[12] However, the property of saturated hysteresis loops of present tetragonal 0.2BMT-0.8PT thin films exhibits a considerable high insulation resistivity, which is consistent with the low leakage current property.[31]

Simultaneously, the local piezoelectric response dependence of applied voltage is shown in Fig. 3(d). The measurement of local piezoelectric displacement was carried out by keeping the SPM tip fixed on the interesting grain with a DC voltage from −9 to 9 V to record piezo-response signal. A typical well-shaped "butterfly" loop is observed with a maximum displacement of 0.35 nm at 8.0 V. Through the observation of the piezoelectric hysteresis loop, the local effective piezoelectric coefficient d_{33}^* is estimated to be approximately 60 pm/V at 2.5 V. It is comparable to the piezoelectric response of highly (100)-oriented BS-PT thin films
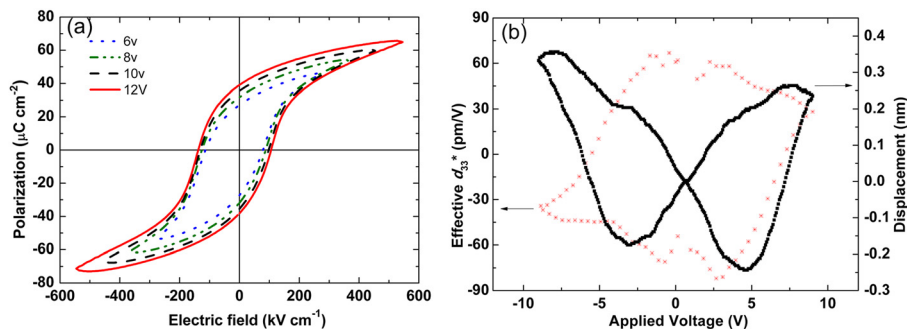

FIG. 3. Electric performances of the 0.2BMT-0.8PT films at room temperature; (a) The growth of ferroelectric hysteresis loop measured at 100 Hz at different applied fields; (b) Local piezoelectric response versus applied voltage.
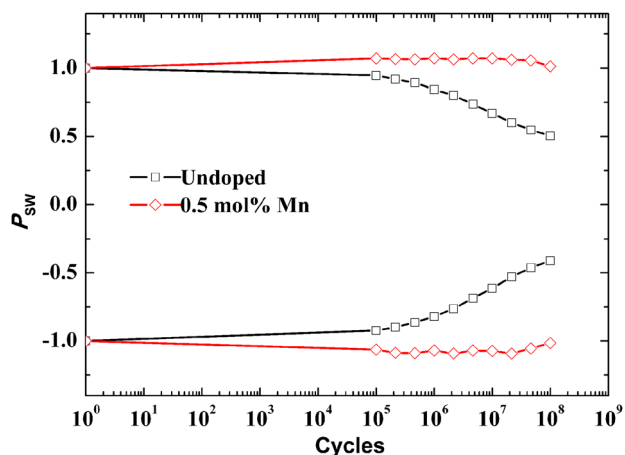
FIG. 4. Normalized polarization (P_{SW}) versus switching cycles for the undoped and 0.5 mol. % Mn-doped 0.2BMT-0.8PT thin films.

(70 pm/V).[25] The fine piezoelectric response of the 0.2BMT-0.8PT thin films probably arises from the (100) preferential orientation with many 90°-domains.

Fatigue is one of the most important factors in determining the lifetime and reliability of ferroelectric devices. The fatigue characteristic of the 0.2BMT-0.8PT thin films is displayed in Fig. 4. The fatigue measurement was performed according to the normal electric fatigue process with a fatigue signal of 8 V (about $3E_C$) in amplitude and 100 kHz in frequency. It can be seen that the polarization decreases by nearly 45% of the starting values after about 10^8 cycles, which is comparable to the PZT thin films with the same electrodes of Pt but inferior to the oxide electrodes,[42] and chemical doping. In addition, the 0.2BMT-0.8PT thin films exhibit a high dielectric constant of about 620 at 100 kHz. It needs to note that a large dielectric loss of 0.08 was observed, which should be reduced for the applications.[31] To improve the fatigue and dielectric performances, 0.5 mol. % Mn-doping was carried out in the present 0.2BMT-0.8PT thin films. As shown in the Fig. 4, the polarization decreases slightly after 10^8 cycles, indicating the enhanced fatigue stability. The dielectric loss has also been reduced from 0.08 to 0.044 by the Mn-doping.[31] The improved performances would be due to the reduction of hole and defect concentration resulting in lower leakage current density by the Mn-doping.[16] Furthermore, Mn-doped films possess the same remanent polarization with the undoped 0.2BMT-0.8PT films.

It is worth to noting that local temperature of an operating computer is easy to be higher than 100 °C for the application. A significant fraction of the computer memory industry, such as FeRAM, is involved in the performance stability under elevated temperature. In the present study, the ferroelectric hysteresis loops of the 0.2BMT-0.8PT films were measured at different temperature at 1 kHz (Fig. 5(a)). It is interesting to find that P_r, E_C and polarization at maximum field (P_{max}) are observed to remain constant with increasing temperature (Fig. 5(b)). At temperature higher than 100 °C, the slightly increase in the polarization is contributed by an increased conductivity due to easy thermal activation. It is interesting to note that the 0.2BMT-0.8PT thin films show a much different behaviour of temperature-polarization to the fundamental relationship in normal ferroelectric materials. Commonly, with increasing temperature, the cation P_S displacement should be reduced and polarization values decrease continuously.[43] Here, the temperature-independent polarization property of the 0.2BMT-0.8PT thin films should be due to the fact of high T_C (about 550 °C).[9,13] The investigated temperature (125 °C) in the present study is sufficiently lower than its T_C, and the P_S should slightly decrease in the investigated temperature range from RT to 125 °C. It is well known that polarization of ferroelectric is ascribed to not only the intrinsic polarization from cation displacement but also the extrinsic contribution from domain wall motion. Hence, the identical polarization values as function of temperature would be also ascribed to the fact that domains with the same dipoles become easier to switch with an increase in the amount of switchable non-180° domains. By taking account both facts of slightly decreasing P_S and easier domain wall motion at elevated temperatures, the property of temperature-independent polarization is achieved in the 0.2BMT-0.8PT thin films. The reliable temperature-independent polarization could be an important criterion for applications of FeRAM. For further improvement on application at higher temperature, the exploration would be encouraged to focus on handing the leakage characteristics at more elevated temperatures.[16,31]

In summary, the high-T_C 0.2BMT-0.8PT thin films with a thickness of 220 nm were deposited on Pt(111)/Ti/SiO$_2$/Si substrates *via* sol-gel method. The films show high (100) orientation and uniform microstructure. The piezoelectric and ferroelectric properties of the present low cost films exhibit d_{33}^* of 60 pm/V and $2P_r$ of 77 μC cm^{-2}, which is better than the BMT-PT thin films with 35.6 μC cm^{-2} near the MPB composition. The good thermal stability of polarization makes it interest to further exploration for ferroelectric material applications. Furthermore, Mn doping effectively reduced dielectric loss and enhanced fatigue reliability.
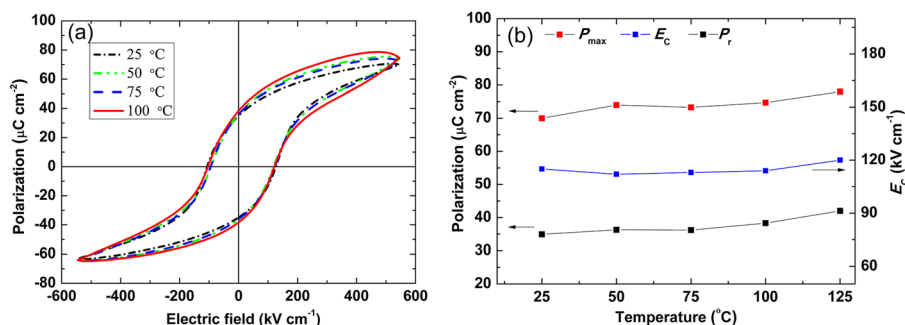

FIG. 5. Temperature dependence of (a) ferroelectric hysteresis loop, (b) P_r (remanent polarization), E_C (coercive field) and P_{max} (polarization at maximum field) for 0.2BMT-0.8PT films measured at 1 kHz.

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