Review

Phase transition and random-field induced domain wall response in quantum ferroelectrics SrTi\textsuperscript{18}O\textsubscript{3}: review and perspective

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Abstract

The oxygen isotope exchange of $^{16}$O by $^{18}$O in SrTiO\textsubscript{3} causes a drastic change from paraelectric to ferroelectric, due to the suppression of quantum fluctuation. The phase transition and origin of the huge domain wall response were evaluated by dielectric, magnetic (NMR), and optical measurements (SHG, light scattering). The results obtained corroborate (1) smeared ferroelectric transition at $T_c$ due to quenched random field, (2) a quite large dielectric contribution from domain walls, (3) incomplete softening of the transverse optic mode, and (4) a large contribution from the relaxational mode to the phase transition. Quantitative explanations, given to individual results, may give hints to grasp the mechanism for the evolution of ferroelectricity, in which quantum fluctuation and random fields are dominant perturbations. Finally, some of the news studies on the SrTiO\textsubscript{3} were also introduced.

Keywords: Quantum paraelectric; Quantum ferroelectric; Ferroelectricity; SrTiO\textsubscript{3}; Isotope effect

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1. Introduction

In the last decade, remarkable new results have emerged from studies of SrTiO\textsubscript{3}, these results continue to attract a great deal of attention to this extensively studied perovskite-type crystal. SrTiO\textsubscript{3} is a typical example of quantum paraelectricity [1–3]. Its dielectric constant saturates at a high level, $\varepsilon' \approx 20000$ [1], associated with the softening of a long-wavelength transverse optic phonon mode. The application of uniaxial stress and proper substitution of the A-site ions can significantly destabilize the lattice, and induce a ferroelectric phase transition [4–6]. In addition, a crossover from quantum ferroelectric to relaxor behavior may occur as observed, e.g., in (Sr\textsubscript{1-1.5}\textsubscript{x}Bi\textsubscript{x})TiO\textsubscript{3} for $x > 0.027$ [7]. In this case, the relaxor behavior is attributable to a ferroelectric domain state induced by random fields [8].

Recently, our group demonstrated that ferroelectricity can be induced by an oxygen isotope exchange [9–11]. A dielectric peak near $T_m = 24.5$ K was detected for 99% $^{18}$O-isotope-exchanged SrTiO\textsubscript{3} (STO18). The Curie point of STO18 appeared in the very low-\textit{T} region, unlike in other ferroelectrics. For this reason, quantum mechanical
fluctuations are expected to decrease the polarization. In addition, the spontaneous or remnant polarization which appears may be suppressed, due to the formation of an random field (RF) induced domain wall state [12,13]. In the heavily strained STO18, it was shown that the external bias increases the permittivity below the Curie point [14] and such an unusual behavior could be attributed to the dynamics of domain walls. Since the relaxation of the field-induced remnant polarization is best explained by a generalized power law [12,13], it has been argued that the ferroelectric domain state might be induced by quenched RFs [12].

In this paper, we focus on the role of quantum fluctuation in SrTiO₃, and discuss theoretical achievements in the evolution of ferroelectricity and the local symmetry of ferroelectric nanodomains as well as offering a perspective for the future researches and applications.

2. Results and discussion

Details of the sample preparation, dielectric measurements [9,10,12,13], light scattering measurements [15–18], and SHG measurements [19] are given in the literatures. For all the measurements, single crystal plate of SrTiO₃ with a dimension 7 mm[001] × 2 mm[110] × 0.3 mm[110], was used.

2.1. Dielectric properties

Fig. 1 [11] shows the temperature dependence of the dielectric constant $\varepsilon'$ for the STO18-100x and STO18-100x/17-100y systems, where 17 and 18 represent ¹⁷O and ¹⁸O, respectively. It can be clearly seen that quantum paraelectric STO16 turns to show a rounded peak with an increase in the amount of heavy isotopes when it exceeds the critical composition. Maximum substitution (99%) still gives a smeared rounded peak at around 24.5 K.

Fig. 2 shows the variation of $T_c$ with the average mass of oxygen for STO18-100x and STO18-100x/17-100y. The figure clearly shows that $T_c$ follows the equation by the mean field expression

$$T_c = 29.6(m_{av}^{O} - 16.62)^{1/2}$$

where $m_{av}^{O}$ is the average mass of oxygen. Irrespective of the kind of oxygen isotopes, ¹⁶O, ¹⁷O, and ¹⁸O and their mixing state, $T_c$ is only a function of the oxygen mass. Thus Eq. (1) expresses a universal relation between $T_c$ and the oxygen mass for SrTiO₃. In order to describe the phase transition of STO18, anharmonic electron-phonon interaction [20] and single sublattice modes [21] have been used to explain the isotope exchange effect in SrTiO₃ in terms of a displacive soft-mode mechanism. However, recent Raman scattering study [22] shows that the ferroelectric soft mode reveals only incomplete softening of the $F_{1u}$ type polar optic mode at the Curie point. Similarly, as in the related impurity system Sr₁₋ₓCaₓTiO₃ (SCT) [23], the softening stops at an energy of about 3 cm⁻¹, and the result is completely consistent with the data obtained on light scattering [18], hyper-Raman scattering [17], and neutron inelastic scattering [24]. These results suggest that the ferroelectric phase transition is not a purely displacive one, but becomes in part order-disorder controlled in the vicinity of $T_c$. Yamada et al. [25] proposed a new model for the ferroelectric phase transition of STO18. The essential feature is described by a 3-state quantum order-disorder system characterizing the degenerated excited states in addition to the ground state of the TiO₆ cluster. The effect of isotope replacement was taken into account through the tunneling frequency between the excited states. The dielectric properties analyzed by the mean field approximation were found to qualitatively agree with the model through out the whole
range of composition. Following the treatment of phase transitions in the SCT system [23,26], Zhang et al. [14] adopted an appropriate order-disorder model to STO18. The transverse Ising model [27] has been recently used to explain the phase transitional properties of quantum paraelectrics and quantum ferroelectrics. [25,26,28]. RFs were additionally introduced in order to describe the observed domains state properties [29]. The result clearly provides evidence that the local disorder field gives rise to the existence of a ferroelectric multi-domain state, and that the dielectric behavior of STO18 under a bias field has an intimate relationship with the domain wall motion.

Fig. 3 shows the temperature dependence of $\varepsilon'$ for STO18-99 as measured under bias. With an increase in bias, the $\varepsilon'$ value is depressed, and the $T_c$ increases as expected. The insets (a) and (b), respectively, show $\varepsilon'$ and $\varepsilon''$ for heavily distorted STO18-93 [14]. Contrary to the less-distorted sample of STO18-99, the distorted STO18-93 sample shows a curious bias dependence and an appearance of peak below $T_c$ with an increase in bias. For the heavy distortion that occurs due to mechanical damaging at the surface layers and possibly due to multidomaining below antiferroditortive transition temperature 107 K, a secondary peak of $\varepsilon'$ below $T_c$ can sometimes be observed. Measurement of the dielectric constant under bias discriminates the contribution from the domain wall motion. In order to treat the interactions of the dipolar moments as well as quantum mechanical effects in the unified framework, Zhang et al. [14] used the Hamiltonian

$$H = -\Omega \sum_i S^z_i - \frac{1}{2} J_{ij} S^z_i S^z_j - 2\mu \sum_i (E_i + E) S^z_i$$

where $S^z_i$ are the $x(z)$ components of the pseudo-spins, and $i$ and $j$ sum overall pseudo-spins, which are proposed to correspond to the displacement of titanium with respect to the oxygen octahedra. $\Omega, J = \sum J_{ij}$, and $\mu$ are the tunneling frequency, the interaction constant, and the effective dipole moment, respectively. $E$ and $E_i$ are the homogeneous external field and the site-dependent RF. Since the numerical solution for Eq. (2) is difficult, some of the simplifying process was introduced avoiding that it was not an unrealistic description [14]. Fig. 4(a) and (b), respectively, show the hypothetical cross-section through two adjacent ferroelectric nano-domains under $E = 0$ used for the numerical calculation and $x$ dependence of $T_m, \varepsilon'(T_m)$, and $\varepsilon'(T = 0)$ under $E = 0$. We defined region (+) as the region with a positive frozen field and region(-) as the region with a negative frozen field. Numerical calculations based on some assumptions could reproduce the bias dependence of the dielectric constant, the enhancement of $\varepsilon'$ by the existence of the frozen field $E_{\text{frozen}}$, enhancement of the zero-field dielectric constant, and the depression of $T_c$ at the domain wall. Applying the bias beyond the frozen field causes the domain wall motion spurring the growth of the ferroelectric domains along the external field. A detailed explanation of

![Fig. 3. Temperature dependence of dielectric constant $\varepsilon'$ for STO18-99 under dc bias. Insert (a) and (b), respectively, show the real and imaginary parts of dielectric constant for heavily distorted STO18-93 [14].](image)
the explanation for the domain wall motions will be also given in the future [30].

Measurement of the dielectric response of STO18 measured under the dc bias field at fixed temperatures enabled us to acquire a great deal of information on the behavior of nano-domains. At this stage, we judged that the smeared peak of \( \varepsilon' \) was coming from relaxor-type or fluctuation-controlled quantum ferroelectric phase transition. In order to decide between a ferroelectric and glassy phase transition, we took advantages of the nonlinear coefficient \( a_3 = x_3/x_1 \) [31–33], where \( x_1 \) and \( x_3 \), respectively, are the linear and nonlinear susceptibilities for the equation

\[
\chi(E) = \chi_1 - 3\chi_3 E^2 + 5\chi_5 E^4
\]

for \( T > T_c \) [34]. Fig. 5(a) shows the bias dependence of dielectric constant measured at fixed temperatures and Fig. 5(b) shows the temperature dependence of the linear susceptibility \( \chi_1 \) nonlinear susceptibility \( \chi_3 \), and \( B = \chi_3/(\varepsilon_0 x_1^2) \). The behavior of \( B \) is similar to that described in our previous study [35], except for the gradual decrease for \( T > 30 \text{ K} \). Since both \( \chi_1 \) and \( \chi_3 \) stay finite when approaching \( T_c \), probably as a consequence of RF smearing, \( B(T_c) \) does not vanish as it would show in thermodynamic equilibrium. This result suggests that STO18 does not belong to the family of relaxors. However, in order to draw a definite conclusion concerning this, further discussion is needed based on other kind of data.

2.2. Local symmetry of ferroelectric nano-regions

The distribution of ferroelectric nano-domains was observed using an SHG microscope, and the images are given in Fig. 6 [36]. Each of the bright spots directly do not correspond to the real ferroelectric nano-regions, but to the distortion-coupled area around them. The azimuth dependence of the SH intensity for STO18 under the bias field revealed that the local symmetry of the ferroelectric nano-region should be orthorhombic \( mm2(C_{2v}) \), with the polar axis along the \([1\overline{1}0]_{\text{cubic}} \) direction. Although our previous study [37] revealed that locally variant mixtures of eight triclinic polar domains with \( d_{22} \) being the coupling...
The possible coexistence of displacive and order-disorder components in the phase transition mechanism is still an open question among perovskite titanates, as suggested for STO18 in the former section. This problem can be solved by measuring the electric field gradient (EFG) tensor by Ti NMR [39]. Fig. 7(a) shows the Ti spectrum for STO18 at 200 K with $B||[001]$ [40]. We observed two central peaks of $1/2 \to -1/2$ for $^{47,49}$Ti accompanied by a broad background. The presence of this satellite background and its angular dependence demonstrate that a dynamical local tetragonal symmetry breaking in cubic symmetry (Fig. 7(b)). Such a behavior is, in fact, expected if the order-disorder component is present in the phase transition mechanism. When the temperature was decreased from RT to 15 K, basically similar spectra were observed, i.e. marked broadening of the central peaks and splitting of the peaks could not be detected below $T_c$. The result of the NMR spectra can be summarized as follows.
(1) The existence of unresolved first order quadrupole satellites in the $^{47,49}$Ti NMR spectra demonstrate that the EFG tensor at the Ti sites is non-zero. This means that the Ti ions in SrTiO$_3$ are not at the center of the oxygen cage, but disordered between several off-center sites.

(2) The angular dependence of the second moment $M_2$ shows that the deviation from cubic symmetry is of tetragonal feature. The value of $T_2$ demonstrates that the Ti site disorder is dynamic rather than static. Although further experiments are needed to draw a definitive conclusion, we can tentatively conclude that spontaneous polarization in the unit cell is too small (an order of magnitude smaller than that of BaTiO$_3$) to detect the Ti 1/2 → -1/2 line splitting.

The dynamical disorder of Ti observed in cubic STO18 as well as in STO16 might have a close relation with their dielectric properties, i.e. residual induced polarization up to 200 K after poling at low temperatures [12,13], growth of polar nano-clusters below 100 K under zero- and bias-fields [15,16,36], other phenomena caused by a quite polar nature of the lattice.

2.3. Perspective for future researches applications

One of the topics related to ferroelectrics at present is the UV light-induced effect, which causes an apparently huge dielectric constant [41,42]. This effect is closely related to the quantum fluctuation; i.e. only quantum paraelectric SrTiO$_3$ shows this effect. Recently, Ishikawa et al. [43] measured photoconductivity for both SrTiO$_3$ and STO18 and found that only the former is a good photoconductor. Nasu [44] has theoretically concluded that UV light irradiation generates an itinerant and fluctuating superparaelectric large polaron in the lattice, and consequently causes an increase in the static dielectric constant and high photoconductivity.

Although there have been many reports on the dielectric properties of epitaxially grown thin films of perovskite-type titanates, almost all of the dielectric constant data are much depressed compared to those for single crystals. Progress in the thin film fabrication technique of SrTiO$_3$ has been reported by Takashima et al. [45]. By combining the laser MBE technique and mechanical polishing, these researchers succeeded to obtaining SrTiO$_3$ film with a thickness of 300 ~ 600 nm sandwiched between YBCO electrodes showing a single crystal-like behavior of dielectric constant, which exceeded 20000 below 4 K. Unlike bulk crystal, this thin film was found to be very stable to the applied bias. This microshort-free insulating film may aid in the development of oxide superconductor multilayer devices, such as superconducting quantum interference devices, three-terminal transistors, and field effect transistors.

3. Conclusions

SrTiO$_3$ itself is an old, well-known material, but an understanding of its physical properties has only quite recently been gained with the aid of information obtained from the experiments conducted by our group. The isotope substitution technique employed by our group has promoted many studies in various fields, because the variable parameter in this technique is mass only, which has enabled us to make the physics easier. A unified interpretation of the quantum effect in ferroelectric systems could provide insights of value for the many kinds of applications that will be developed in the future.

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