

# High-resolution dilatometry measurements of SrTiO<sub>3</sub> along cubic and tetragonal axes

Mao Liu and Trevor R. Finlayson

*Department of Physics, Monash University, Clayton, Victoria 3168, Australia*

T. Fred Smith

*Vice-Chancellor's Department, La Trobe University, Melbourne, Victoria 3083, Australia*

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High-resolution dilatometry measurements have been made for SrTiO<sub>3</sub> along the tetragonal-phase  $a$  and  $c$  axes and the cubic-phase [100] direction between 2 and 280 K. A moderate stress field was used to align the domains of the low-temperature tetragonal phase or to force a single domain. Anisotropy in the thermal strain at temperatures above the cubic to tetragonal transformation has been observed in the cubic phase for the measurements along the  $a$  and  $c$  directions. No structural change or strain anomaly has been detected at  $\sim 37$  K, the proposed transition temperature to the quantum-paraelectric regime. The expansion of the tetragonal  $c$  axis with decrease of temperature is arrested below 10 K. The implications of the observed anomalies are discussed. [S0163-1829(97)00706-6]

## I. INTRODUCTION

The perovskite, SrTiO<sub>3</sub>, is one of the classical systems exhibiting a second-order, displacive, structural phase transition that has been successfully described by the theory of phonon-mode softening.<sup>1</sup> The transformation in SrTiO<sub>3</sub> from perovskite cubic to tetragonal occurs at  $T_c \sim 105$  K on cooling. Near and below  $T_c$ , the order parameter is the rotational angle,  $\varphi$ , of the Ti-O<sub>6</sub> octahedra about the tetragonal  $c$  axis.<sup>2</sup> The tetragonal distortion is very small ( $c/a \sim 1.0009$  at 10 K as determined by neutron diffraction<sup>3</sup>) and the low-temperature structure was first determined using electron paramagnetic resonance<sup>4,2</sup> (EPR) which is very sensitive to structural symmetry. Subsequently, the lattice parameters of the tetragonal phase were determined by double-crystal neutron and x-ray diffraction.<sup>3,5</sup>

The "soft-phonon" mode responsible for the 105 K transformation<sup>6</sup> corresponds to the rotation of the oxygen octahedra about the  $\langle 100 \rangle$  axes. Accompanying the phonon softening, which occurs at the point  $[1/2, 1/2, 1/2]$  on the Brillouin zone boundary, there is quasi-elastic critical scattering, the central peak, above and near the critical temperature,  $T_c$ .<sup>7</sup> The mechanism of the cubic to tetragonal transformation is well understood in the context of phonon-mode softening,<sup>1</sup> but doubt remains as to whether the central peak has its origin in the dynamic anharmonicity of the soft mode,<sup>8</sup> or a static, defect-induced lattice strain.<sup>9</sup>

More recent interest in SrTiO<sub>3</sub> has been focused upon the quantum-paraelectric (QPE) state first postulated by Müller and Burkard.<sup>10</sup> The divergent temperature dependence of the dielectric constant would predict a ferroelectric transition at  $T_q \sim 35$  K, but saturation occurs below 4 K. The failure to form the Ti-O dipole through the static displacement of the Ti ions is attributed to the magnitude of their zero-point quantum fluctuations. Sharp anomalies in the EPR fine-structure magnetic-field splitting for Fe<sup>3+</sup> in SrTiO<sub>3</sub> have been observed<sup>11</sup> at  $T_q$ . Furthermore, it has been deduced from the EPR measurements that the rotation of the Ti-O octahedra, which is the order parameter for the tetragonal

phase, has an anomalous contribution below  $T_q$ . While no change in the spatial symmetry of the lattice occurs about  $T_q$ , it is deduced that the crystal does become more cubic.

New anomalies associated with the QPE state have been reported in inelastic neutron<sup>12</sup> and Brillouin<sup>12,13</sup> scattering and dielectric loss.<sup>14</sup> A brief review of the theoretical situation has been given by Tosatti and Martoňák.<sup>15</sup>

Such a wealth of phonon-related phenomena make SrTiO<sub>3</sub> an attractive candidate for the high-resolution dilatometry study of phase transformations that we are conducting. Previous thermal expansion measurements for the martensitically transforming materials, In-Tl,<sup>16</sup> Ni-Al,<sup>17</sup> and V<sub>3</sub>Si (Ref. 18) showed pretransformation anisotropy in the high-temperature cubic phase which we have attributed to pretransformation lattice distortion. These systems all exhibit significant  $[\zeta\zeta 0]$  TA phonon-mode softening above the phase-transition temperature.<sup>19-21</sup> In contrast, thermal expansion measurements for a Cu-Al-Ni single crystal showed no pretransformation anomaly,<sup>22</sup> and it is known that there is no significant softening for the corresponding phonon mode for this system.<sup>23</sup> This distinction raises the question: Is there a correlation between the observed absence/existence of the pretransformation anisotropy in the thermal expansion and phonon mode softening? SrTiO<sub>3</sub> with its displacive structural phase transition and distinct phonon-mode softening represents a nonmetallic counterpart of these systems. Furthermore, there is also the question of the possible link between the pretransformation strain anisotropy and central mode behavior.

## II. EXPERIMENTAL DETAILS

A cube of approximate dimensions  $6 \times 6 \times 6$  mm<sup>3</sup> was cut using a circular diamond saw from a single crystal of SrTiO<sub>3</sub>. The crystal was colorless and had a room temperature x-ray rocking curve with a full width at half maximum value of  $0.42^\circ$  for the 200 reflection using Cu  $K\alpha$  ( $\lambda = 1.54059$  Å) radiation. The rocking curve was taken from a nonetched, cut, and polished face of the sample. The

crystal was aligned by the x-ray Laue method so that the sample could be cut with face normals along  $\langle 100 \rangle$ , to an accuracy of better than  $1^\circ$ . The exact orientation of the sample was also confirmed by the Laue method after the cutting. The sample faces were then lapped and polished using  $25 \mu\text{m}$ , followed by  $6 \mu\text{m}$  diamond paste, on rotating polishing discs. The finished opposite faces were parallel to within 20 min of arc. For convenience we specifically identify three mutually perpendicular sides of the cube as  $[100]$ ,  $[010]$ , and  $[001]$ .

Thermal expansion measurements were performed using a three-terminal capacitance dilatometer.<sup>24</sup> Heating runs were made for the three  $\langle 100 \rangle$  directions of the sample from 4.2 K to room temperature and cooling runs for the 4.2–2 K range. A cooling run for the  $[010]$  from 300 K to 77 K was also done.

Similar to our previous measurements for In-Tl (Ref. 16) and  $\text{V}_3\text{Si}$ ,<sup>18</sup> the  $a$  axis in the tetragonal phase was aligned along the measurement direction,  $l$ , by applying a sufficiently large uniaxial stress along  $l$ , whereas the  $c$  axis was obtained along  $l$  by using biaxial stress perpendicular to  $l$  to force the crystal to be a monodomain in the low-temperature phase. In the latter case, a much larger stress level was needed than in the former.

### III. RESULTS

The initial thermal strain measurements along  $[100]$ ,  $[010]$  and  $[001]$  showed similar behavior from 4 to 300 K. Due to the small uniaxial compressive stress ( $\sim 1$  bar) along the  $l$  direction due to the sample mounting springs, all three axes follow tetragonal  $a$ -axis behavior as seen from their strain  $\epsilon = \Delta l/l_0$ , where  $l_0$  is the sample length not far above the critical temperature (we have chosen  $T = 160$  K as this reference point) vs temperature ( $T$ ) curves as discussed below.

A representative strain-temperature curve from later measurements along  $[001]$  with a larger uniaxial stress ( $\sim 10$  bars) is shown as curve  $\epsilon_a$  in Fig. 1. For  $[010]$ , the initial run with the smaller stress ( $\sim 1$  bar) produced a strain-temperature curve identical to curve  $\epsilon_a$ . For  $[100]$  and  $[001]$  the strain-temperature behavior is similar to curve  $\epsilon_a$  for the initial runs; they are predominantly  $a$ -axis-like, i.e.,  $d\epsilon/dT > 0$  below  $T_c$ , but extrapolate to a smaller  $T = 0$  thermal strain [ $\approx -8 \times 10^{-4}$  with respect to  $\epsilon(160 \text{ K}) = 0$ ] than that for curve  $\epsilon_a$  ( $\approx -10.2 \times 10^{-4}$ ). This is taken to indicate that the transformation phase is composed of domains that are only partially aligned with the  $a$  axis along  $l$ . It was found that increasing the stress to  $\sim 10$  bars gave results identical to curve  $\epsilon_a$ . Further increase of the stress level up to 20 bars did not change the results for all three directions, indicating the  $a$  axis could be fully aligned for the whole crystal with a stress level exceeding  $\sim 10$  bars. It is suggested that the smaller stress needed to align the  $a$  axis along  $[010]$  compared to that for  $[100]$  and  $[001]$ , is probably due to an anisotropic internal stress field in the as-grown crystal, compensating the applied stress field for the latter two directions.

Biaxial compressive stresses of about 40 bars were applied along the  $[100]$  and  $[010]$  while thermal expansion measurements were made for  $[001]$  direction. The measured

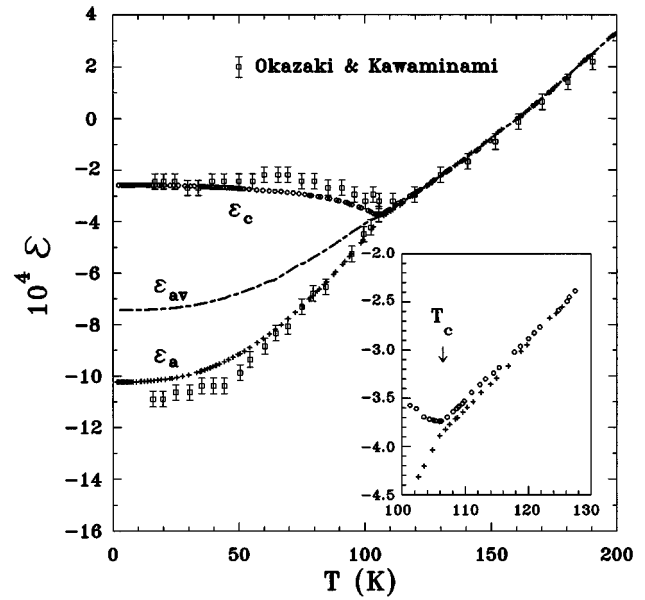


FIG. 1. Thermal strain ( $\epsilon$ ) vs temperature ( $T$ ) curves for  $\text{SrTiO}_3$  measured along cubic  $[001]$ . Under different stress conditions,  $[001]$  becomes either the tetragonal  $a$  ( $\epsilon_a$ ) or  $c$  ( $\epsilon_c$ ) axis below  $T_c$ . Curve  $\epsilon_a$  was measured with a uniaxial stress ( $\sim 10$  bars) along  $[001]$ ; curve  $\epsilon_c$  was measured with biaxial stresses of about 40 bars applied along  $[100]$  and  $[010]$ . The thermal strains derived from the x-ray data of Okazaki and Kawaminami (Ref. 5) are also included for comparison. The inset shows on a larger scale the deviation from the expected isotropic behavior for a cubic lattice just above  $T_c$ . The dot-dashed curve is the derived average strain,  $\epsilon_{\text{av}} = (2\epsilon_a + \epsilon_c)/3$ .

$\epsilon$  vs  $T$  data are shown as curve  $\epsilon_c$  in Fig. 1.

The stress level used here is far below that required for stress-induced ferroelectricity ( $\sim 1.6$  kbar along  $[100]$  at 4.2 K) (Ref. 25) and the tetragonal-to-trigonal phase transition ( $\sim 2.5$  kbars along  $[111]$  at 4.2 K).<sup>26</sup>

The thermal strains derived from the x-ray lattice parameters of Okazaki and Kawaminami (1973) (Ref. 5) in the range 15–300 K are also plotted in Fig. 1 for comparison. Their later (1974) (Ref. 5) experiments gave more accurate results in the range 80–120 K, but are within uncertainty limits of the 1973 data reproduced here. The neutron data of Heidemann and Wettengel<sup>3</sup> taken over the temperature range 10–300 K are essentially the same as the x-ray data.

It can be seen that curves  $\epsilon_a$  and  $\epsilon_c$  are in reasonable agreement with the temperature dependence of the thermal strains derived from the  $a$  and  $c$  lattice parameters, except that the magnitude of the dilatometry,  $\epsilon_a$ , is slightly smaller than that of the x-ray data below 60 K.<sup>27</sup> Thus, it is concluded that the uniaxial stress and the biaxial stresses have forced the transformed crystal into an aligned domain structure or into a single domain for the measurements shown as curve  $\epsilon_a$  and  $\epsilon_c$ , respectively.

Support for the above conclusion was obtained by examining the crystal with and without applied stress under an optical microscope after cooling through the transition to 80 K. In the absence of applied stress, the prepolarized light reflected from the back face of the sample revealed the twinning bands parallel to  $[110]$  on the  $(001)$  plane within the

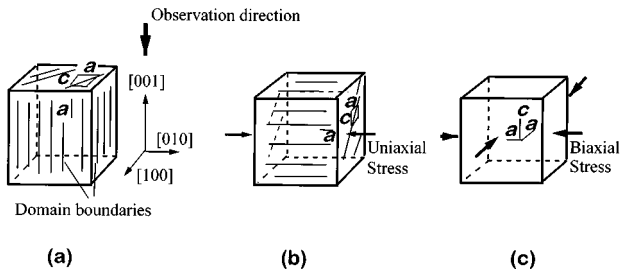


FIG. 2. Sketch of the low-temperature phase domains in (a) an external-stress-free sample, (b) with uniaxial stress applied along [010], or (c) with biaxial stresses along [010] and [100], while the crystal is viewed in polarized light along [001].

transparent crystal [see Fig. 2(a)]. The contrast between the banded domains disappeared (appeared) gradually when heating (cooling) the crystal above (below)  $T_c$ . When the crystal was uniaxially stressed along [010] and examined on the (001) plane again, no bands were observed at any depth in the crystal, consistent with [010] being constrained to the  $a$ -axis direction. In this condition, slab-shaped twinning domains would be parallel to [010] and could not be seen along the observation direction, [001], as the light path was blocked by the overlapped slabs seen along [001] [Fig. 2(b)]. The same situation arose (i.e., no bands were observed) when the crystal was biaxially stressed along the [100] and [010] and observed along [001], but in this case the crystal should be monodomain [Fig. 2(c)].

The linear thermal expansion coefficient,  $\alpha(T) = \Delta\epsilon/\Delta T$ , derived from successive data points is shown in Fig. 3. The structural transformation temperature,  $T_c$ , is determined to be  $105.5 \pm 0.5$  K from the sharp discontinuity in  $\alpha(T)$ .

A careful search for anisotropy in the expansion was made at temperatures above  $T_c$ . A detailed plot of the strain as a function of temperature between 100 and 130 K for the  $a$  and  $c$  directions is shown in the inset of Fig. 1, where the

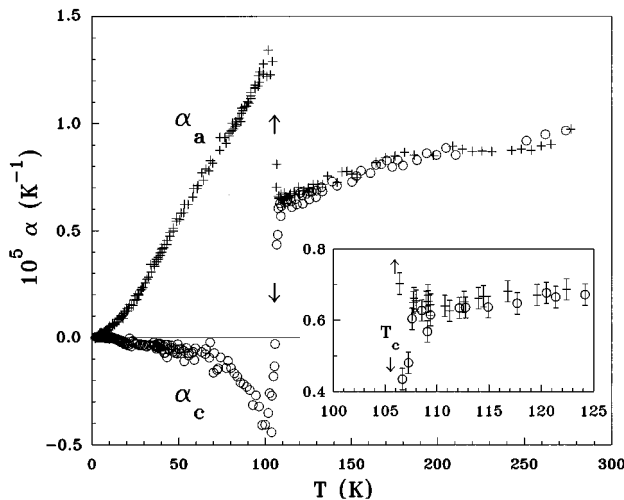


FIG. 3. Linear thermal expansion coefficients ( $\alpha$ ) of SrTiO<sub>3</sub> for the  $a$  and  $c$  axes derived from the strain curves in Fig. 1. Arrows indicate the critical temperature,  $T_c = 105.5 \pm 0.5$  K. The inset shows the data near and above  $T_c$ .

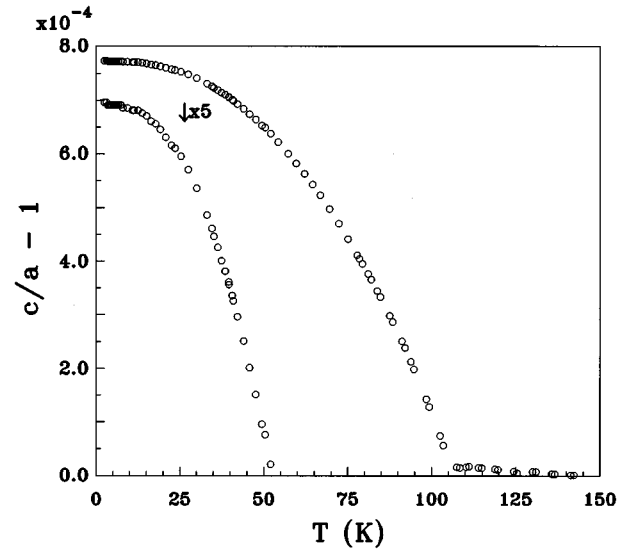


FIG. 4. Tetragonal distortion,  $c/a - 1$ , vs temperature,  $T$ , derived from the strain data in Fig. 1. The lower-left curve is an expanded view of the data from 2 to 50 K.

experimental uncertainty ( $\sim 2 \times 10^{-6}$ ) is smaller than the size of the data symbols. A small, but clearly resolved separation ( $\sim 10^{-5}$ ) of the two strain curves occurs at  $\sim 120$  K thus resulting  $c/a > 1$  above  $T_c$  (see Fig. 4). However, as the two strain curves then follow similar temperature dependences from 120 K down to  $T_c$ , there is no detectable anisotropy in  $\alpha(T)$  (as shown in the inset of Fig. 3).

The average strain,  $\epsilon_{av} = (2\epsilon_a + \epsilon_c)/3$ , below  $T_c$  joins smoothly with the curve through the data for the isotropic strain in the cubic state (Fig. 1) with no change in volume at  $T_c$ .

The tetragonal distortion,  $c/a - 1$ , derived from the strain data of Fig. 1 using  $c/a = (1 + \epsilon_c)/(1 + \epsilon_a)$  increases continuously on cooling from  $T_c$  down to 10 K (Fig. 4). There is no evidence of the sample becoming more cubic at  $\sim 37$  K, as inferred by Müller, Berlinger, and Tosatti.<sup>11</sup> Note the anisotropy in the strain above  $T_c$  is clearly seen.

An interesting feature is observed for  $\epsilon_c(T)$  at about 10 K, below which temperature the growth in the  $c$  direction shows an abrupt arrest and possibly a down turn. Data for four independent runs are shown in Fig. 5(a). There is no corresponding arrest in the thermal strain for the  $a$  axis as seen in Fig. 5(b).

#### IV. DISCUSSION

There are three significant features in the temperature dependence of the thermal strain; (i) the absence of any anomalous behavior at  $T_q$ , (ii) the abrupt arrest in the growth of the  $c$ -axis thermal strain below 10 K, and (iii) the anisotropy above  $T_c$ . We shall discuss the significance of (i) and (ii) in the context of the formation of the quantum paraelectric state and relate (iii) to the appearance of the central peak scattering.

The x-ray measurements of Okazaki and Kawaminami<sup>5</sup> show that the  $a$ -axis contraction in the tetragonal phase down to 75 K is due to the rotation of the Ti-O<sub>6</sub> octahedra. Thus,

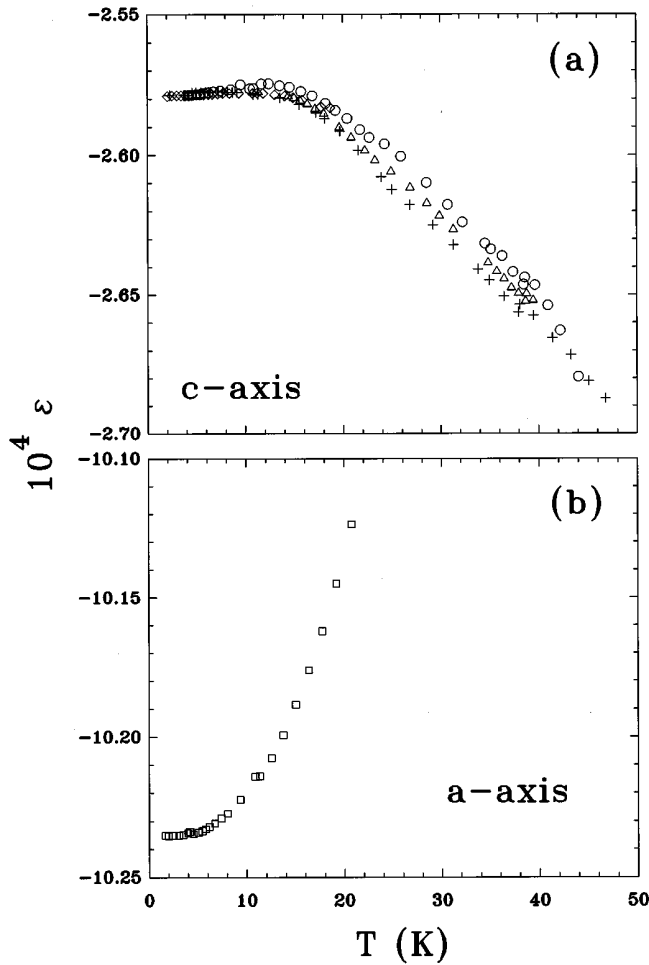


FIG. 5. Low-temperature data for (a)  $\epsilon_c(T)$  and (b)  $\epsilon_a(T)$  as for Fig. 1 shown on larger scales. The  $c$ -axis data are for four independent runs and are represented by different symbols. Symbol  $\circ$  identifies the data from the first run (Fig. 1). Other symbols are the data from subsequent runs for which the reference point for calculating  $\epsilon(T)$  was initially 4.2 K. These data points have then been shifted vertically to match the first-run data ( $\circ$ ) at 4.2 K.

the tetragonal distortion of the octahedra is due entirely to  $c$ -axis expansion, which is consistent with a tendency towards the formation of Ti-O dipoles due to the off-symmetry displacement of the Ti ions. Below 75 K the octahedra continue to expand in the  $c$  direction, but also contract in the  $a$  direction. There is no evidence of any singularity in the lattice thermal strain down to 15 K.

While noting the absence of any evidence of an anomaly in the lattice expansion, Tosatti and Martoňák<sup>15</sup> argue there is sufficient evidence to claim Ti-O dipoles form below  $T_q$ , but that on account of quantum fluctuations they do not assume a stable orientation and behave as quantum rotors. This being the case, it would be expected that there would be no further influence of the Ti-ion displacements on the lattice strain as the temperature is lowered, which is contrary to what is observed.

A more consistent explanation would be one in which the dipole formation continues to develop as the thermal fluctuations of the Ti ions are reduced by lowering the temperature and it is about 10 K where the quantum fluctuations become

dominant, resulting in the abrupt arrest in the growth of the  $c$ -axis thermal strain. Such an explanation is also consistent with the temperature dependence of the dielectric constant, which follows a smooth increase before saturating below 10 K.<sup>10</sup> The possible decrease in the  $c$ -axis thermal strain below 10 K would then be due to the normal behavior associated with the decrease in phonon excitation.

The present measurements provide further evidence for an association between phonon-mode softening and pretransformation anisotropy in the thermal expansion or thermal strain. The question that then arises is the possibility that central mode behavior is linked to pretransformation strain anisotropy.

There are two schools of thought on the origin of the elastic central peak that appears above  $T_c$  for a structural transformation associated with a soft-phonon mode. The earliest, and possibly most popular model, describes the central peak in terms of a dynamic process,<sup>8</sup> although increasing evidence has been put forward<sup>28</sup> to support the model based on defect-induced lattice strain.<sup>9</sup>

The cubic symmetry of the lattice prior to the transformation is implicit in both models. Yet, the present observation of strain anisotropy for SrTiO<sub>3</sub> up to 15 K above the transformation, contributes to the increasing evidence<sup>16–18</sup> that there is a departure from cubic symmetry well above  $T_c$  for soft-mode-driven systems. We propose that it is this static distortion of the lattice symmetry from cubic which gives rise to the formation of the reciprocal lattice associated with the new symmetry that is responsible for the Bragg reflection identified as the central peak.

There is already some experimental evidence to support this proposal. Riste *et al.*<sup>7</sup> have reported that for the central peak “its neutron intensity varies from superlattice point to superlattice point as the Bragg intensity below  $T_c$  does.”

Experimental evidence is also found in other systems. Shapiro *et al.*<sup>20</sup> have reported the development of a central peak at a wave vector close to that for which softening occurs in the  $[\zeta\zeta0]$  transverse acoustic mode for cubic (CsCl-type) Ni<sub>62.5</sub>Al<sub>37.5</sub> and that these peaks lie close to Bragg peaks in the pseudo-orthorhombic transformation phase. It has also been reported<sup>29</sup> for Ni<sub>2</sub>MnGa that “strong temperature-dependent elastic scattering was observed at wave vectors close to  $\zeta_0$ , which develops into a Bragg peak of an intermediate phase, as the martensitic transition temperature is approached.”

A more thorough test of this proposal that the central peak is a precursor to a Bragg reflection in the transformation phase may be made by a search in reciprocal space above  $T_c$  for other “forbidden” reflections associated with the transformation lattice structure in soft-mode systems. It is noted that off-zone-center, soft-mode systems will offer the most favorable conditions for such a search as zone-center softening, as occurs in Nb<sub>3</sub>Sn,<sup>8</sup> results in the central-mode scattering emerging from an existing Bragg peak. This presents a problem both with resolution and detection, as the intensity of the central-mode scattering is far less than that of the Bragg peak,<sup>7,30</sup> and only slightly displaced from it.

## V. CONCLUSION

From dilatometric measurements of thermal strain above and below the cubic  $\rightarrow$  tetragonal phase transformation in

SrTiO<sub>3</sub> it has been concluded that (i) the onset of the quantum-paraelectric state occurs at  $\sim 10$  K and not  $\sim 37$  K as previously suggested,<sup>11</sup> (ii) the quasielastic central peak observed in neutron, x-ray, and optical scattering above the phase transformation is due to the static tetragonal distortion of the lattice.

## ACKNOWLEDGMENTS

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- <sup>27</sup>The unit-cell dimension of SrTiO<sub>3</sub> is doubled at  $T_c$  as the phase transition is antistortive and the low-temperature phase unit cell consists of eight distorted unit cells of the cubic phase in order to conserve translational symmetry of the lattice (Ref. 4). For the convenience of relating the dimensions of the tetragonal phase to those of the cubic phase, here we use dimensions of a single subunit cell of the tetragonal phase as the tetragonal  $a$  and  $c$  parameters. This is also the case in the precision neutron and x-ray diffraction measurements quoted here (Refs. 3 and 5). In some other cases, different coordinates have been used for the tetragonal phase, e.g., the  $c$  direction is still along pseudocubic [001] but the  $a$  directions are along [110] and  $[\bar{1}\bar{1}0]$  as in the dielectric measurements [J. G. Bednorz and K. A. Müller, *Phys. Rev. Lett.* **52**, 2289 (1984)].
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