

to the deformation through nuclear forces, and hence should have a larger  $B(E1)$ . In this two-component model the observed  $B(E1)$  asymmetry would require the quantity  $V(\text{coupling})/\Delta E$  in  $^{13}\text{N}$  to be 60% of its value in  $^{13}\text{C}$ .

In a more detailed shell-model calculation the process is more complex since once the contribution from the large components cancel, weaker components of the wave functions also play a role. In particular, the  $E1$  decay of the  $(\frac{3}{2}^-, T = \frac{3}{2})$  state to the  $\frac{1}{2}^+$  state must proceed via components of the  $\frac{1}{2}^+$  state with  $T = 1$  parentage in the  $A = 12$  core. Experimentally these  $B(E1)$  values are about an order of magnitude weaker than those of the  $(\frac{1}{2}^+ \rightarrow \frac{1}{2}^-) \Delta T = 0$  transition and are at present impossible to calculate with sufficient reliability.

The prominent case of the  $(\frac{1}{2}^+ \rightarrow \frac{1}{2}^-)$  asymmetry exhibits great sensitivity to details of the nuclear structure, but it seems to be understandable in terms of the  $^{13}\text{C}$ - $^{13}\text{N}$  binding-energy difference without invoking charge-dependent nuclear interactions.

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## Behavior of $\text{SrTiO}_3$ near the [100]-Stress-Temperature Bicritical Point

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Good polydomain  $\text{SrTiO}_3$  samples show, for  $T \rightarrow T_a^+$ , paramagnetic resonance-linewidth broadening  $\Delta H(T)$  of the  $\text{Fe}^{3+}$ - $V_0$  center which is the average of  $\Delta H_{\parallel}(T)$  and  $\Delta H_{\perp}(T)$  of monodomain samples. Application of [110] and [100] stress on these samples enhances and reduces  $\Delta H(T)$ , respectively. This verifies the Ising and Heisenberg character of the transition in monodomain and annealed samples, respectively. In the Ising monodomain case the correction to the scaling exponent of the order parameter is found to be  $\chi > 0.5$  in agreement with renormalization group theory.

Following the discovery of critical behavior in the temperature dependence of the order parameter in  $\text{SrTiO}_3$ ,<sup>1</sup> calculations using the renormalization-group theory have been made to obtain the static critical exponents.<sup>2</sup> Of interest is the topology near the structural second-order phase transition  $T_a$ .<sup>3</sup> At  $T_a$  the cubic anisotropy is renormalized away and the fixed point is of the Heisenberg type.<sup>2,4</sup> Because the system is cubic, for  $T > T_a$  its symmetry can be lowered by the application of uniaxial stress  $P[\alpha\beta\gamma]$ , and a variety of phase boundaries  $T(P[\alpha\beta\gamma])$  can join in at  $T_a$ . The total hypercriticality at  $T_a \equiv T_c(P=0)$  including the various staggered fields of antiferrodistortive ordering has not yet been worked out. For compressive and tensile stress  $P[100]$  along a cubic [100] axis, the system has a topology<sup>3</sup> like the uniaxial antiferromagnet near the antiferromagnetic spin-flop bicritical point.<sup>5</sup> However, in  $\text{SrTiO}_3$ , the flop line is in common with the temperature axis  $T$ , whereas in the magnetic case there is a skewed one.<sup>6</sup> In the tetragonal

phase, the lattice is elongated along the  $c$  axis, i.e.,  $c/a > 1$ .<sup>7</sup> Therefore, application of a tensile stress along a [100] direction supports the elongation. This enhances  $T_c$  and one obtains a monodomain in the low-temperature phase. Because below  $T_c$  the direction of the rotational order parameter is fixed, tensile stress  $P[100] < 0$  yields an  $n = 1$  Ising-type phase boundary. For  $P[100] > 0$  two kinds of monodomains along [010] and [001] are formed and thus at  $T_a$  an  $n = 2$ ,  $XY$  second-order phase boundary originates, too.<sup>3</sup> Therefore, in the  $P[100]$ - $T$  plane  $T_a$  is a bicritical point,<sup>5</sup> and the transition temperature shift exponents  $\psi$  for the two boundaries are equal to the changeover exponent  $\varphi = 1.25$ , respectively.<sup>3,5</sup>

The earlier measurements of the temperature dependence of the order parameter have been carried out on monodomain samples<sup>8</sup> to achieve a better accuracy near  $T_c$ . The experimental value obtained gave  $\beta = 0.33 \pm 0.02$ .<sup>1</sup> Because of the uniaxial character of the sample, the Ising value  $\beta_1 \equiv \beta(1) = 0.315$  rather than the Heisenberg  $\beta_H$

$\equiv \beta(3) = 0.37$  is expected if the crossover from cubic behavior does not occur too close to  $T_c$ . Aharony and Bruce<sup>3</sup> have suggested from the anisotropy in EPR linewidth of the  $\text{Fe}^{3+}-V_0$  center that the crossover occurs sufficiently far away from  $T_c$ . Indeed, the linewidth anisotropy for rotations  $\bar{\varphi}$  around the monodomain<sup>8</sup> axis,  $\Delta H_{\parallel}$ , and the one perpendicular to it,  $\Delta H_{\perp}$ , extends about 15 deg above  $T_c$ .<sup>9</sup> The earlier interpretation of this observation has been in terms of a cubic anisotropy in the quadratic part of the Hamiltonian.<sup>9</sup> However, it has been shown that this part in the Hamiltonian is vanishing for sufficiently high order in  $\epsilon = 4 - d$  for  $T \rightarrow T_a^+$ .<sup>4</sup> Therefore, two types of experiments have been performed to check the new suggestion<sup>3</sup>: linewidth measurements on high-quality polydomain samples to obtain the Heisenberg behavior, and application of definite stresses on such samples to reproduce monodomain  $n = 1$  Ising and, in addition,  $n = 2$  XY-model behavior. All experiments were successful and are described below.

In the absence of stress or strain, the approach to  $T_a^+$  will yield isotropic fluctuations  $\Delta H$  due to the Heisenberg  $n = 3$  character. These are averages of  $\Delta H_{\parallel}$  and  $\Delta H_{\perp}$  of the  $n = 1$  or  $n = 2$  systems. The samples used were grown by National Lead Company with the Verneuil method in 1963. These 12-yr-old samples showed the smallest internal strain and spread in  $T_c$ . Thermal annealing of more recently produced crystals gave less satisfactory results and more spatial variation in  $T_c$ . Figure 1 shows the data points for a cylindrical sample of 1.45-mm length  $\parallel [001]$  direction and a diameter of 1.95 mm, for  $H \parallel [110]$  with  $P = 0$ . The resonance consisted of a single line at 2920 G using  $\nu = 19.2$  GHz. Away from  $T_a$ , cubic anisotropy is present. Therefore, a broad and a narrow line are expected due to "pancake"-type clusters with  $\Delta\omega_b$  and  $\Delta\omega_n$  if their mobilities are sufficiently slow.<sup>9</sup> As we see only one line, these mobilities are fast enough compared with the EPR linewidth of  $\Delta\omega(T) = \Delta H/g\beta\hbar$  that one averaged line is observed. On approaching  $T_a$ , a slowing down occurs and  $\Delta H(T)$  increases to a finite value at  $T_a$ . However, by then the cubic anisotropy has become small enough that  $\Delta\omega_b - \Delta\omega_n \ll \Delta\omega(T)$ . Finally, in a Heisenberg regime, of course, only one line is expected for  $T_a^+$ .

Extrapolation of  $\Delta H(T)$  to  $T_c = T_a$  yields  $\Delta H(T_a) = 10.5$  G which is below the monodomain value  $\Delta H_{\parallel}(T_c) = 17.5$  G but above  $\Delta H_{\perp}(T_c)$  of 6.7 G. The center-of-mass relation  $\frac{1}{3}(\Delta H_{\parallel} + 2\Delta H_{\perp}) = \Delta H(T_a) = 10.5$  G is found to hold, indicating that the value

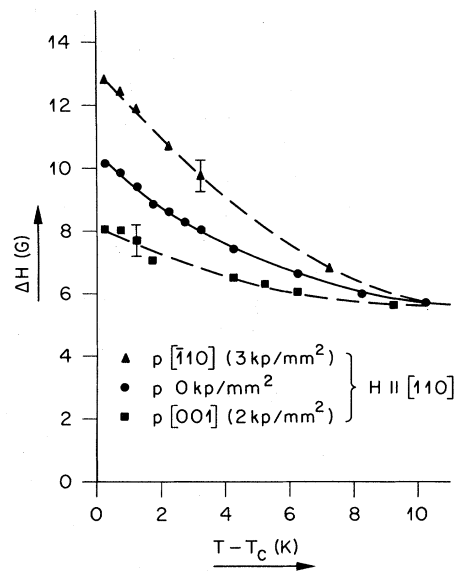


FIG. 1. EPR linewidth broadening of the  $\text{Fe}^{3+}-V_0$  center in polydomain samples for zero stress  $P$ , and  $[001]$  stress,  $n = 2$ , and  $[\bar{1}10]$  stress,  $n = 1$ , cases, respectively. 1 kp (kilopond) is equal to 9.80665 N.

obtained in the polydomain sample is indeed the average of the axial values measured in the monodomain sample. This result, although very satisfactory, should not yet be overrated because in the above relations, the critical and noncritical parts of the fluctuations  $\delta\varphi = \langle \delta\varphi^2(T = T_c) \rangle^{1/2}$  are present. We recall that in the slow-motion regime for  $H \parallel [110]$ ,  $\delta H = A\delta\varphi$  with  $A = 26$  G/deg holds and  $\delta H \approx \Delta H - \Delta H_b$ . Now the noncritical fluctuations contribute about  $\Delta H_b \approx 5$  G in the polydomain sample and  $\Delta H_b^{\parallel} = 2.9$  G and  $\Delta H_b^{\perp} = 3.5$  G in the monodomain, respectively. This difference of about 2 G is not understood. It could result from larger inhomogeneous static strains in the crystal due to its polydomain character. This static strain apparently has a smaller distribution in the uniformly strained monodomain samples. In the latter, a substantial fraction of the background width  $\Delta H_b \sim 3$  G is due to "white-noise" noncritical fluctuations extending up to  $11 \text{ cm}^{-1}$  in frequency and yielding a nonsecular relaxation.<sup>10</sup>

In order to substantiate the suggestion<sup>3</sup> further, uniaxial stresses were applied to induce the  $n = 2$  and  $n = 1$  regimes and with them to estimate the "equivalent stress" present in the monodomain samples. This could be accomplished in our experimental setup despite the fact that stress can only be applied perpendicular to the external mag-

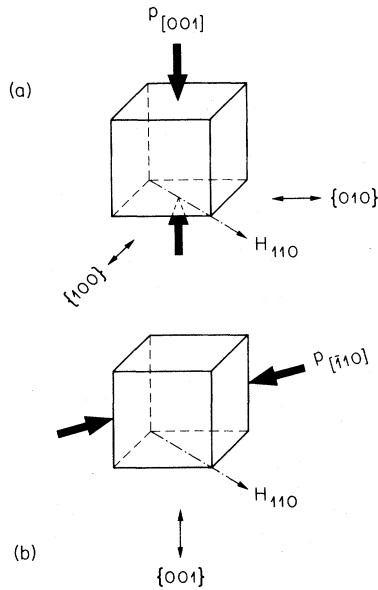


FIG. 2. Geometries for  $H \perp P$  of the magnetic field  $H \parallel [110]$  and stress  $P$  for (a)  $[001]$ ,  $n=2$ , and (b)  $[\bar{1}10]$ ,  $n=1$ , cases realized in the experiments of Fig. 1.

netic-field direction. Further, one should bear in mind that fluctuations  $\delta\varphi_\alpha$  are always probed around a direction  $\alpha$  which is perpendicular to the plane in which the magnetic field  $H$  is lying. Thus for  $H \parallel [110]$ , fluctuations around  $[001]$  are detected (see Fig. 2).

In a first experiment, stress was applied on the  $(001)$  faces of the cylindrical polydomain sample. The stress and magnetic-field orientations are shown in Fig. 2(a). Below  $T_c$ , evidently  $\{100\}$  and  $\{010\}$  domains are favored ( $c/a > 1$ ). This is the  $n=2$  case. The measured fluctuations  $\delta\varphi[001]$  occur around a *perpendicular* direction to these  $\{100\}$  and  $\{010\}$  domain orientations. Thus, one expects  $\Delta H_\perp$  to be *reduced*. This is indeed the case as the lower curve in Fig. 1 shows for  $P=2$   $\text{kp/mm}^2$  ( $1 \text{ kp} = 9.80665 \text{ N}$ ).

To reproduce the Ising  $n=1$  regime, a cylindrical sample with  $(\bar{1}10)$  faces of diameter 1.3 mm and length 1.6 mm, was carefully machined from the same 1963 boule. Application of stress to these faces [see Fig. 2(b)] induces a  $\{001\}$  domain below  $T_c$ .<sup>8</sup> Thus, this is an Ising transition. Figure 1 shows the linewidth data for  $T \rightarrow T_c^+$  for  $P=3$   $\text{kp/mm}^2$ . Because the  $[001]$  fluctuations are now chosen *around* the preferred Ising axis, they are *enhanced*. The compressive stress of 3  $\text{kp/mm}^2$  along  $[\bar{1}10]$  corresponds to an equivalent pulling stress of  $\frac{3}{2} = 1.5$   $\text{kp/mm}^2$  along the  $[001]$  axis. The enhancement observed of 2.8 G at  $T$

$= T_c'$  is the same as the depression at  $T = T_c$  for the  $n=2$ ,  $P[001]=2$   $\text{kp/mm}^2$  case. Thus the enhancement for  $n=1$  is  $1.4 \times 1.5 \approx 2.1$  times the depression for  $n=2$ , i.e., twice as large. This is expected and completes the verification of the suggestion<sup>3</sup> whose origin resulted from a discussion of Courtens<sup>11</sup> who measured the birefringence in the monodomain sample  $\Delta n = n_\parallel - n_\perp$ . These measurements gave, within experimental accuracy, the same fluctuation amplitudes  $\delta\varphi_\perp$  and  $\delta\varphi_\parallel$  as EPR in the slow-motion regime. Hata<sup>12</sup> has observed in recent specific heat measurements a clear difference especially in amplitude of strained and strain-free samples. For the former he obtained a positive critical exponent of 0.08 to 0.25 which brackets the Ising value of  $\alpha = 0.125$ .

From the enhancement of 2.8 G observed, we can estimate the "equivalent stress" present in the monodomain sample. Approximating  $\delta(\Delta H) \propto P$  near  $T_c$ , and noting that  $\Delta H_\parallel(T_c) - \Delta H(T_c) \approx 7$  G, the "equivalent stress" is of the order of 6–7  $\text{kp/mm}^2$ , an apparently high value. It is due to plastic deformation of the monodomain sample by grinding down the 0.3 mm-thick  $[110]$  platelets.<sup>8</sup>

With the Ising character of the monodomain samples established for  $T - T_c^+ \lesssim 15$  K we assume this to hold also for  $T_c^- - T \lesssim 7$  K. Thus we know  $\beta(1) = 0.315$  from high-temperature expansions and can analyze the crossover behavior of our accurate measurements<sup>1</sup> to classical behavior. We set, according to a proposition by Binder,<sup>13</sup>

$$\varphi(T) = \varphi_0 (1 - T/T_c)^{\beta(1)} \times [1 + b_1 (1 - T/T_c)^x + \dots], \quad (1)$$

where  $x$  is the correction exponent and  $b_1$  the non-universal amplitude. Then, putting  $\varphi_c(T) = \varphi_0 (1 - T/T_c)^{\beta(1)}$  for the critical part, simple algebra gives

$$\varphi^{1/\beta(1)} - \varphi_c^{1/\beta(1)} \approx 3b_1 \varphi_0^{1/\beta(1)} (1 - T/T_c)^{1+x}. \quad (2)$$

The exponent  $x$  has been calculated by Wegner<sup>14</sup> to be  $x = 0.5 \pm 0.05$  using renormalization theory and expansion to order  $\epsilon$ , and more recently by Saul, Wortis, and Jasnow<sup>15</sup> with a twelve-term high-temperature series. Thus plotting  $(\varphi^{1/\beta(1)} - \varphi_c^{1/\beta(1)})^{2/3}$  should give a straight line. This is done in Fig. 3 with the data of Ref. 1. It is seen that for  $T \rightarrow T_c^-$  there is a "hanging through" of the points near  $T_c^-$  indicating that  $x > 0.5$ . This would conform with the numerical solutions of recursion relations by Swift and Grover<sup>16</sup> who ob-

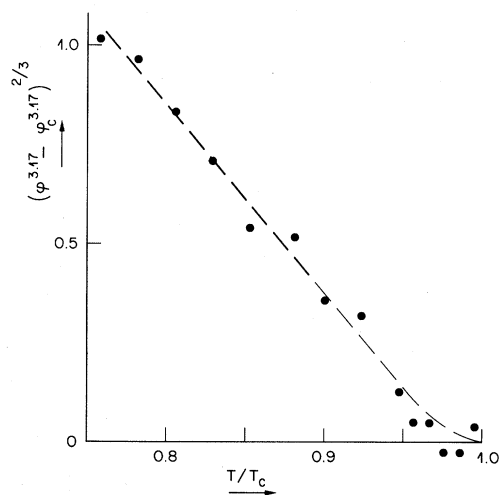


FIG. 3. Static changeover behavior of the order parameter  $\varphi(T)$  in the  $\text{SrTiO}_3$  monodomain,  $n = 1$  Ising samples measured in Ref. 1.

tained for  $n = 1$ ,  $d = 3$ ,  $x \approx 0.64$ .<sup>17</sup>

In summary, the topology and crossover behavior in the  $[001]$ -stress-temperature plane near the bicritical point of the  $\text{SrTiO}_3$  structural phase transitions has been elucidated and agrees with the present theoretical understanding and numerical estimates.

One of us (K.A.M.) has profited from illuminating discussions and correspondence with A. Aharony, A. D. Bruce, K. Binder, and E. Courtens and a critical reading of the manuscript by them and H. Beck.

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