would not be surprising, since there the diffusive energy mode couples to the order parameter in complete analogy to the antiferromagnet MnF_2 [M. P. Schulhof, P. Heller, R. Nathans, and A. Linz, Phys. Rev. Lett. 24, 1184 (1970)]. Riste *et al.*, Ref. 3, invoked this coupling mechanism for the high-temperature side as well. This may be a contribution in the region $q/\kappa \gg 1$. However, in the hydrodynamic regime, i.e., for $q/\kappa \ll 1$, the fluctuations of the energy and of $\hat{\varphi}_{\vec{q}}$ are uncoupled.

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[18]These experiments rule out γ vanishing at $\vec{q}=0$, in accord with our assumption.

Fluctuations and Correlations in $SrTiO_3$ for $T \geqslant T_c$

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The linewidth broadening of the paramagnetic resonance of the Fe^{3+}-V_O center in $SrTiO_3$ near T_c has been measured accurately and analyzed for $T \to T_c^+$. It is found that the order-parameter susceptibility $\chi(\vec{q}, T)$ is two-dimensionally anisotropic with an anisotropy parameter $\Delta = \frac{1}{60}$. The critical exponent of the correlation length ξ is found to be $\nu = 0.63 \pm 0.07$. The EPR linewidth is cusp shaped for $T \to T_c^+$ and finite at $T = T_c$.

Recently it was found that the local fluctuations near the second-order phase transition in $SrTiO_3$ can be probed by the temperature-dependent linewidth broadening of the paramagnetic-resonance linewidth of the Fe^{3+}-V_O center,[1] i.e., an Fe^{3+} ion with a nearest-neighbor oxygen vacancy.[2] This broadening occurs approximately over the same temperature interval in which Blazey[3] observed simultaneously two electronic band transitions, and over which for $T \geqslant T_c$ Riste *et al.*[4] found a central mode with neutron diffraction. Schwabl[5] has developed a dynamical theory for $T \geqslant T_c$ which yields the central peak and the soft mode. He assumes the following form for the static order-parameter susceptibility for $\hat{\varphi}_{\vec{q}}{}^{\alpha}$ near $\vec{q}_R = a^{-1}(\pi, \pi, \pi)$:

$$\chi^{\alpha\alpha}(\vec{q}, \epsilon) = \chi_0 [\vec{q}^{\,2} - (1-\Delta) q_\alpha{}^2 + \kappa^2]^{-1+\eta/2}, \qquad (1)$$

where $\hat{\varphi}_{\vec{q}}{}^{\alpha}$ is the Fourier transform of the rotation angle $\varphi_{\vec{l}}{}^{\alpha}(t)$ at lattice site $\vec{l}$ of the (BO_6) octahedra around the direction $\alpha = [100]$, $[010]$, $[001]$; χ_0 is proportional to the single-particle susceptibility; and $\kappa(\epsilon) = \kappa_0 \epsilon^{\nu}$ is the inverse of the correlation length $\xi(\epsilon)$. The reduced temperature $(T - T_c)/T_c$ is denoted by ϵ; ν and η are critical exponents.[6] The temperature-independent parameter Δ describes the anisotropy of the dispersion of $\chi^{\alpha\alpha}$ near $\vec{q}_R$. Using Eq. (1) Schwabl has obtained an expression for the linewidth broadening for $T \to T_c^+$ for fast fluctuations,

$$\Delta H(\epsilon) \propto \frac{\kappa(\epsilon)^{-(1-2\eta)}}{\sqrt{\Delta}} \arctan\left(\frac{\pi\sqrt{\Delta}}{\kappa(\epsilon) a}\right), \qquad (2)$$

where $a = 3.9$ Å is the lattice constant.

In the present note we report on high-accuracy measurements of $\Delta H(T)$ which allowed us to determine ν and the anisotropy constant Δ which were so far unknown. The results obtained agree well with Eq. (2) from $T_c + 1°$ to $T_c + 46°$, where $\Delta H \simeq 0$. However, for $T \sim T_c^+$ there occurs a critical slowdown in the fluctuations and the linewidth does not diverge. We show theoretically and experimentally that $\Delta H(T)$ is cusp shaped,[6] behaving as $\Delta H^2(\epsilon) = \Delta H_{\max}^2 (1 - C\epsilon^{\nu})$ within 0.7° from T_c.

The influence of the rotation angle $\varphi_{\vec{l}}{}^{\alpha}$ on the resonance field H_r of the Fe^{3+}-V_O complex has been analyzed for all orientations of the center axis and directions of α and H_r.[7] It is found that for $\vec{H}$ in the (001) plane between $[100] + 10°$ and

$[100]+60°$ the resonance field of the center with axis parallel to [100] (high-field line) is linearly sensitive to the $\varphi^{[001]}$ component. The $\varphi^{[010]}$ component enters only quadratically, and its fluctuation contribution to the linewidth is of the order of 3% of the maximum linear $\varphi^{[001]}$ effect for $\vec{H}$ parallel to [110]. The quadratic $\varphi^{[010]}$ influence has been observed separately by the broadening of the low-field $g \sim 6$ line[7] with $\vec{H}$ parallel to [100] which shows no linear $\varphi^{[001]}$ contribution but is comparable in quadratic dependence. The result confirms the theoretical estimate. The measurements of the high-field resonance linewidth have been carried out with $\vec{H}$ in the (001) plane parallel to $[110]-0.3°$ [Fig. 1(a)]. The exact [110] direction was avoided because of overlap with centers pointing into the [010] direction.[2,7] Figure 1(b) exhibits the broadening of the low-field $g \sim 6$ resonance for $\vec{H}$ parallel to [110], which for this direction is independent[7] of φ and therefore shows no additional broadening near T_c. The large linewidth difference between these two lines of about 1:15 at T_c also excludes a T_1 relaxation process for the high-field line since even for Fe^{3+} ions in very anisotropic coordination no anisotropic T_1 effects have been reported.

The samples used for the K-band experiments have been described elsewhere.[8] To avoid disturbing influences of different domains for the measurements below T_c, plate-shaped crystals were used which below T_c transform into a monodomain sample.[8] Their domain axis was oriented parallel to the rotation axis of the scanning magnetic field.

For the analysis of the experimental effects we restricted ourselves to temperatures above T_c, since the experimental data below T_c are less reliable because of overlapping lines. Well above T_c the fluctuations are fast and cause the resonance lines to be homogeneously broadened.[9] They are thus Lorentzian as observed, with a width proportional to the fluctuation-correlation time. On approaching T_c from above, the fluctuations are critically slowed down, their correlation time increases and with it the width of the resonance lines. To isolate the influence of the critical rotational fluctuations on the measured linewidth, the background width resulting from different mode fluctuation must be subtracted. This background is by itself temperature dependent and has been interpolated as indicated in Fig. 1(c). The remaining critical linewidth is given by the time integral of the fluctuation-correlation function[9]:

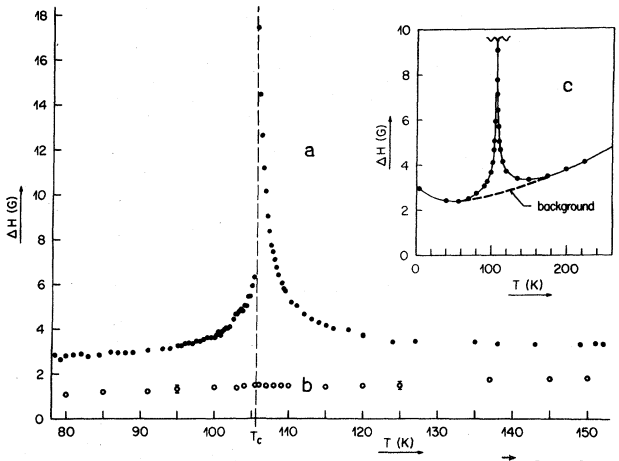

FIG. 1. Observed linewidth broadening for $\vec{H} \parallel [110] - 0.3°$, $\vec{c} \parallel [001]$ of the Fe^{3+}-V_O center at the structural phase transition of $SrTiO_3$ at 19.2 GHz. (a) For the high-field line; (b) for the low-field $g \sim 6$ line. (c) Determination of the background width.

$$\Delta H(\epsilon) \propto \int_0^\infty \langle \varphi^{[001]}(t)\varphi^{[001]}(0)\rangle_\epsilon \, dt = \int_0^{\pi/a} S(q, \epsilon, \omega = 0)\, d^3q, \quad (3)$$

where $S(q, \epsilon, \omega)$ is the temperature-dependent dynamical structure factor of the rotational fluctuations as derived by Schwabl.[5] For zero frequency his expression reduces to

$$S(q, \epsilon, \omega = 0) \propto T[\chi^{\alpha\alpha}(\vec{q}, \epsilon)]^2, \quad (4)$$

where $\chi^{\alpha\alpha}(\vec{q}, \epsilon)$ is given by Eq. (1). Using Eq. (4) in Eq. (3) yields Eq. (2).

Fitting the experimental points with expression (2) in the entire interval from 105.75 to 152°K would require $T_c = 104.5$°K. However, from experiment it is found that T_c is at 105.6°K where the linewidth is maximum and the order parameter vanishes.[10] Thus, for $T \to T_c^+$ the linewidth does not diverge as the fast-fluctuation formula (2) indicates. For the range of T between $T_c + 1°$ and $T_c + 46°$, an excellent fit is obtained (Fig. 2) using the following parameters in (2)[11]:

$$T_c = 105.6°\text{K}, \quad \nu = 0.63 \pm 0.07, \quad \Delta = 0.017 \pm 0.010.$$

η is set equal to zero as it is estimated to be of the order of the experimental error. $\kappa(115°\text{K}) = 0.05$ Å^{-1} was taken from the determination by neutron diffraction.[4] The value of the anisotropy parameter Δ means that sixty octahedral units in a (001) plane are correlated when one in a next (001) plane is correlated to them in the sense of

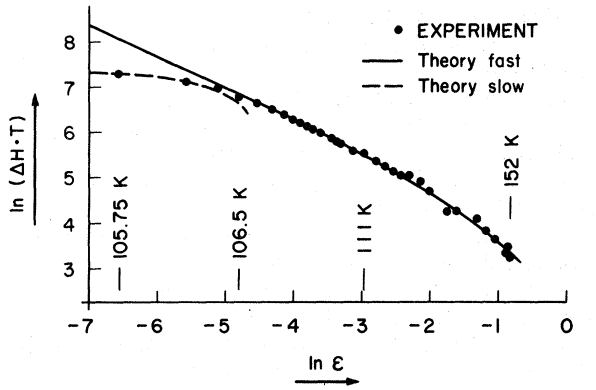

FIG. 2. Comparison between experimental and theoretical linewidths for the fast- and slow-motion regimes. For the latter, the theoretical curve shows $(\langle \Delta H_{\text{crit}}^2 \rangle)^{1/2}$; for convenience, it is adapted to the experimental points determined by linear subtraction of the background width, as in the fast-motion regime.

the *I*4/*mcm* structure. This is understood qualitatively by the strong in-plane coupling between the alternately rotating oxygen octahedra through the displacement of the common corners as compared to the weak coupling through the oxygen ions between the planes. The "pancake"-type correlations found here in the cubic phase may, to a certain extent, be compared with the behavior of the planar Heisenberg model as suggested by Stanley[12] although the comparison is limited by the fact that in our case the anisotropy is coupled with the direction of the "spins," i.e., the axis of rotation. This model yields critical exponents $\beta = \frac{1}{3}$, $\gamma = \frac{4}{3}$, and $\nu = 2\beta = \frac{2}{3}$ with $\eta = 0$. The experimentally deduced values for $SrTiO_3$ are $\beta = 0.333 \pm 0.010$ from EPR experiments[10] and $\nu = 0.63 \pm 0.07$ obtained here, as well as $\gamma = 1.29 \pm 0.10$ calculated by Schneider and Stoll[13] using the observed change-over from $\beta \simeq \frac{1}{2}$ outside the critical region, to $\frac{1}{3}$ near T_c.

Schwabl[5] also obtained from his theory an expression for the ultrasound absorption near T_c as carefully measured by Fossheim and Berre[14] for the propagation of ultrasound along a ⟨100⟩ direction. In view of the anisotropy in $\chi^{\alpha\alpha}(\vec{q}, \epsilon)$, the critical exponents of ultrasound absorption are also anisotropic, as observed by Rehwald.[15]

For temperatures below 106.5°K the fast-fluctuation regime is observed to be no longer valid. The nondivergent width is expected since it is a local variable not depending on the volume of the system considered, and the mechanical coupling between the rotating oxygen octahedra at most allows a maximum local value of $\varphi_{\vec{l}}^{\alpha}$ of the order of $\varphi_{\vec{l}}^{\alpha}(T = 0) \sim 2°$. The latter fact prevents $\langle \vec{\varphi}_{\vec{l}}^{\,2} \rangle$ from diverging for $T \to T_c$, where $\langle \vec{\varphi}_{\vec{l}}^{\,2} \rangle$ means the ensemble average of the square of the local fluctuations $\varphi_{\vec{l}}^{\alpha}$. On approaching T_c the line broadening changes over to a regime where the fluctuations are critically slowed down and their oscillator strength is more and more concentrated in the central peak[4,5] which contains only frequencies very close to zero. Its width narrows to zero for $T \to T_c^{+}$, and hence the fluctuations are concentrated at frequencies low compared to the characteristic spin precession frequency of the EPR experiment. The latter may be estimated in our experiment to be around 100 MHz.[9] In this temperature range therefore the spins "see" the fluctuations as static. Thus, the instantaneous resonance line is *inhomogeneously* broadened and is essentially characterized by a Gaussian line shape. The change from Lorentzian to more Gaussian-type resonances is found experimentally. The analysis yields 50% Gaussian at $T_c + 1°$. Then,

$$\langle \Delta H_{\text{crit}}^2 \rangle \equiv \langle \Delta H_{\text{expt}}^2 - \Delta H_{\text{background}}^2 \rangle \propto \langle \vec{\varphi}_{\vec{l}}^{\,2} \rangle = \langle \vec{\varphi}_{\vec{l}}(t) \cdot \vec{\varphi}_{\vec{l}}(t) \rangle = 2 \int_{q=0}^{\pi/a} \int_{\omega=0}^{\infty} S(q, \epsilon, \omega)\, d^3q\, d\omega, \tag{5}$$

where $\langle \vec{\varphi}_{\vec{l}}^{\,2} \rangle$ is the equal-time correlation function of $\vec{\varphi}_{\vec{l}}$ at site $\vec{l}$. Using Schwabl's expression for $S(q, \epsilon, \omega)$ this yields for the critical contribution ($\kappa \to 0$),

$$\langle \Delta H_{\text{crit}}^2(\epsilon) \rangle \propto \text{const} - [\kappa(\epsilon)/\sqrt{\Delta}] \arctan[\pi\sqrt{\Delta}/\kappa(\epsilon)a]. \tag{6}$$

The second term, which for $\kappa \to 0$ becomes linear in κ, vanishes at $T = T_c$ so that the first term describes the *finite* maximum width at the phase transition. Equation (6) is then rewritten for $T \gtrsim T_c$ as

$$\langle \Delta H_{\text{crit}}^2(\epsilon) \rangle = \Delta H_{\max}^2 (1 - C\epsilon^{\nu}), \tag{7}$$

with $\eta = 0$, and C a proportionality constant.[16] From the experiments we obtain $\Delta H_{\max} = 20.1$ G, $C = 17.7$. In this region the experimental uncertainties of T_c and $\Delta H_{\max}^{\text{expt}}$ do not allow an accurate independent determination of the exponent ν. Therefore $\nu = 0.63$ has been taken from the first analysis. The value of $\Delta H_{\max}$ derived from Eq. (7) lies well within the experimental error. This result shows that for $T \to T_c^{+}$ the linewidth is cusp shaped.[6] The theoretical results according to this regime are included in Fig. 2. The change-

over to the fast-fluctuation regime is found near 106.5°K. Strain inhomogeneities in the sample also cause a lowering of ΔH near T_c. However, such effects yield a zero derivative of ΔH with respect to T at T_c, whereas the experiments show a monotonic increasing derivative down to T_c +0.15° as predicted by the cusp-shape behavior. Hence the strain effects are limited to within 0.1°K in our experiment.

In agreement with our quantitative analysis yielding two-dimensional correlations are the critical anisotropic x-ray diffusive scattering data of Denoyer, Comes, and Lambert[17] in $KMnF_3$ and $NaNbO_3$ which undergo an analogous transition to $SrTiO_3$. We also note that the flat dispersion curve of the soft R-corner mode between the R and M points of the Brillouin zone found by Shirane[18] in $KMnF_3$ is indicative of a highly anisotropic correlation in q space. The M_3 point in $SrTiO_3$ lies at 8 meV,[19] about twice as high as in $KMnF_3$ at T_c, indicating a linear anisotropy twice as large in the latter. A recent nuclear magnetic T_1 relaxation study by Bonera, Borsa, and Rigamonti[20] in $NaNbO_3$ can only be interpreted by uncorrelated fluctuations occurring between equivalent and correlated (100) planes near T_c. Nearly two-dimensional correlations for $T \geq T_c$ in systems ordering three-dimensionally at T_c have recently also been found in magnetic systems like K_2NiF_4[21] and in superconducting layer-type compounds.[22] An extended account including the analysis of the linewidth for $T \leq T_c$ as well as its angular dependence in relation to the soft modes[19,23] below T_c will be published elsewhere.

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