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LETTER TO THE EDITOR

Fluctuation quenching of thermal focusing in $Ba_2NaNb_5O_{15}$

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Abstract. Near $T_C = 846$ K barium sodium niobate exhibits a strong thermal focusing of laser light polarized along its optic c axis (focal length $F = +2$ to $+3$ cm for 1 W in a 0.45 mm beam waist). The beam convergence $\theta(T)$ is proportional to $K^{-1}(T)\, \mathrm{d}n_c(T)/\mathrm{d}T$, where K is the thermal conductivity and n_c is the c-axis index of refraction. For zero applied field, $\theta(T)$ far from T_C is dominated by the isotropic $\mathrm{d}n/\mathrm{d}T$ term, which varies as $t^{2\beta-1}$, where t is the reduced temperature and β is the coexistence curve exponent (approximately $\frac{1}{4}$ in this tricritical system). However, within a degree or two of T_C, $\theta(T)$ is dominated by $K(T)$, which becomes highly anisotropic and has a cusp-like dip for thermal diffusion along the polar c axis. In the present experiment we apply a DC electric field along c and show that a few kV cm^{-1} is sufficient to suppress the cusp.

The use of external electric fields to quench the fluctuations near T_C in ferroelectrics was exploited most successfully by Courtens and Gammon [1] in KH_2PO_4. They observed the field suppression of microscopic polarization fluctuations [1a] and their linear coupling to entropy fluctuations [1b] very near T_C. In the present letter we apply this technique to study, via thermal focusing, the analogous phenomena near T_C in barium sodium niobate ($Ba_2NaNb_5O_{15}$), which in an external electric field along the polar c axis is, like KH_2PO_4, an example of a type-zero (Cowley's notation [2]) phase transition very near [3] a tricritical point; that is, it is not a symmetry-breaking transition and involves 'softening' at only a point at $q = 0$ in the Brillouin zone. T_C in barium sodium niobate specimens varies from about 840 to 852 K, depending upon stoichiometry.

The phenomena of interest to us involve thermal focusing of laser light in barium sodium niobate near its Curie temperature. The simplest theory [4], based upon focusing in isotropic media, yields a focusing angle

$$\theta(T) = AK^{-1}(T)\, \mathrm{d}n_c/\mathrm{d}T \qquad (1)$$

where K is the thermal conductivity, n, the index of refraction, and A, a constant proportional to optical absorption, laser power, and path length, and inversely proportional to the square of the beam diameter. Far from T_C the beam angle of convergence $\theta(T)$ is dominated by the isotropic $\mathrm{d}n_c/\mathrm{d}T$ terms, which varies as $t^{2\beta-1}$, where t is the reduced temperature, and β is the critical exponent characterizing the coexistence curve

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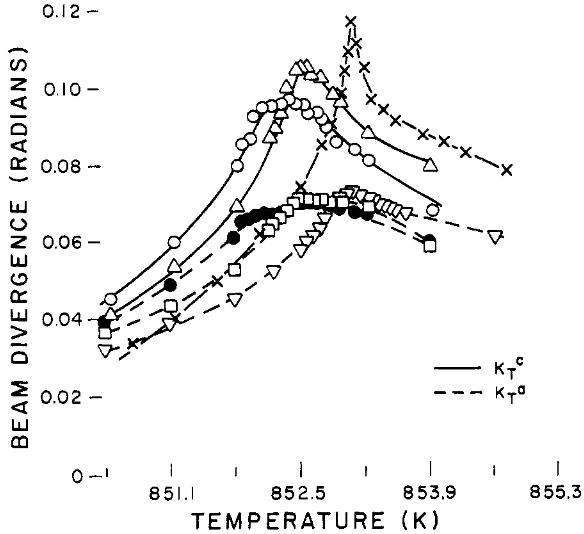

Figure 1. Beam convergence angle θ versus sample temperature T in barium sodium niobate at several values of applied electric field E_c along the polar c axis. The upper three curves are for heat propagation along the c axis (crosses, triangles, and open circles at $E = 0$, 625, and 847 V cm^{-1}, respectively); the lower three curves are for heat propagation along the a axis (inverted triangles, squares, and solid circles at $E = 0$, 625, and 847 V cm^{-1}, respectively). The curves are merely guides to the eye.

($\beta = \frac{1}{4}$ at tricritical points). However, within a few degrees of T_C, $\theta(T)$ exhibits a cusp-like increase (figure 1) in addition to the smooth $t^{2\beta-1}$ divergence, and becomes highly anisotropic [5, 6] (elliptical far-field images rather than circular are observed). We assume in what follows that this cusp is caused by critical fluctuations in polarization near T_C and that increasing the external field suppresses (or 'quenches') these polarization fluctuations and eliminates the cusp. The anisotropy (eccentricity) of the beam convergence angle $\theta(T)$ shows that $K(T)$ is dominant, since the factor dn/dT is isotropic for all thermal propagation directions, depending upon light polarization but not propagation angle.

Unfortunately, as discussed initially by Nettleton [7], there is no good microscopic theory of $K(T)$ near T_C in real ferroelectrics. In some crystals there is a large increase [8] in $K(T)$ at T_C; in most there is a cusp-like decrease [7]. So even the sign of the anomaly varies and is unpredictable. One might expect that in a second-order displacive phase transition the large density of low-frequency 'soft' optical phonons would contribute to phonon heat diffusion and hence to a sharp anomaly in $K(T_C)$. However, in ferroelectrics such a phonon density will occur for very long wavelengths ($q = 0$) and hence make no contribution, to lowest order, to the Umklapp processes responsible for thermal conductivity. This wave vector restriction may be relaxed by defects, including domain walls, and hence the anomaly in $K(T_C)$ may be extrinsic. In analogy with the Kondo problem, this possibility increases the complexity of any microscopic analysis. In the present letter we assume only that sharp anomalies in $K(T_C)$ are empirically typical of ferroelectrics and do not address their general origin or sign. We infer from the experimental data below that they are due at least in part to polarization fluctuations.

Note that the largest thermal conductivity anomaly reported [8] at a Curie temperature thus far (about 100%) is in $LiKSO_4$, where it was shown that the electrical conduction is almost entirely ionic. Near T_C, barium sodium niobate is also an ionic conductor, which may be important in this context.

The experimental details of the thermal focusing set-up in zero field have been given elsewhere [5]. A single-moded argon-ion laser operating at powers of 0.1–1.0 W at 514.5 nm in a 0.45 mm beam waist was incident upon a poled, detwinned $Ba_2NaNb_5O_{15}$ specimen contained in an alumina tubular furnace. The polar c axis was vertical, with both incident and scattered light polarized along c. Silver electrodes were painted on the c faces, which were approximately 4×5 mm^2. Fields from zero to 1 kV cm^{-1} were applied across the 1 mm c thickness, which produced a maximum current flow of 0.3 mA. Either pulsed DC or AC (60 Hz) could be used without serious sample degradation.

The far-field patterns 0.9 m from the sample were measured both visually and with a digitalized video system. Sample temperature was controlled by adjusting oven ambient to ± 0.5 K and then fine-tuning laser power to set internal specimen temperature (an adjustment of 10 mW gave a temperature change of 0.07 K near T_C). The parameter called $\theta(T)$ is the beam divergence measured at the screen, 0.92 m from the sample and 0.90 m from the focal point just beyond the sample. $\theta(T)$ is measured to the outermost bright ring. Because of self-induced phase shifts in the scattering, there is always a particularly bright outer ring in the pattern, which may include more than 20 concentric elliptical rings [9].

The data for θ versus T are shown in figure 1 at applied fields $E_c = 0$, 625 V cm^{-1}, and 847 V cm^{-1}. At higher fields we observed filamentary shorting near the edges of the electrodes. Also shown in figure 1 are the values of $\theta(T)$ for heat propagation along the non-polar a axis. In this case $\theta_a(T) = K_a^{-1}(T)\, \mathrm{d}n_c/\mathrm{d}T$ and there is no divergence at T_C; $K_a(T_C)$ is apparently only very weakly coupled to polarization fluctuations along c. Visual inspection of figure 1 indicates that the $\theta_c(T)$ dependence approaches that of $\theta_a(T)$ at high fields. In fact the eccentricity of the far-field ring pattern decreases from 1.55 at T_C for zero field to 1.44 at 847 V cm^{-1}. A crude linear extrapolation suggests an isotropic $K(T_C)$ near $E_c = 4$ kV cm^{-1}.

The curves in figure 1 show that an electric field of 847 V cm^{-1} decreases θ(maximum) by 15% at T_C. There is also a small shift in temperature for this maximum of 1.5 K at this field, which we assume is simply heating by the applied field (we independently measured a current flow of 0.3 mA). We note parenthetically that the current–voltage relationship in barium sodium niobate in this temperature range is known to be chaotic [10, 11]. This is another macroscopic measure of fluctuation effects in this system and may also involve domain wall motion in its microscopic description.

The lower three curves in figure 1 are for heat propagation along the a axis. Note that unlike the c-axis data they show no cusp-like divergence at the Curie temperature even in zero field, and they exhibit a negligible field dependence. The ratio θ_c/θ_a gives the eccentricity e of the observed elliptical far-field pattern at each value of T and E_c; we find e(max) = 1.55 at zero field, 1.52 at 625 V cm^{-1}, and 1.44 at 847 V cm^{-1}. Since K is the only quantity in (1) that is significantly anisotropic, we assume that this implies $K_a/K_c = 1.55$ in zero field at T_C. Moreover, since K_a seems independent of field, we infer that $K_c(T_C, E_c)$ increases with increasing field at about 10% per kV cm^{-1}. There are few experimental values with which we can compare these results; however, at 3 K in $SrTiO_3$ (somewhat above but near the extrapolated Curie temperature, which is at negative absolute temperatures) Sievers finds [12] that 5.4 kV cm^{-1} increases $K(T)$ by 100%, i.e., 18% per kV cm^{-1}, in close agreement with the present results.

It would obviously be desirable to test this hypothesis by direct, conventional thermal measurements of thermal conductivity near the Curie temperature in barium sodium niobate. It is known [13] that K is highly isotropic in this material up to 500 K. The fact that this material is a good ionic conductor at such temperatures may make $K(T)$ difficult to model; domain walls, mobile ions, and P_z lattice polarization are all presumably coupled. The chaotic current behaviour observed by Martin *et al* [10, 11] and our thermal lens effects are probably related, and presumably involve both fast-ion conduction and domain wall effects. We note that Courtens [1b] found that K is anomalous near T_C in KH_2PO_4 and also invoked defects as a microscopic explanation; it seems likely that the phenomena observed in $Ba_2NaNb_5O_{15}$ and KH_2PO_4 are related.

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