

BRILLOUIN SCATTERING IN FERROELASTICS AND QUASI-ONEDIMENSIONAL FERROELECTRICS

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ABSTRACT

We have measured the Brillouin spectra of ferroelastic $\text{KH}_3(\text{SeO}_3)_2$ in both phases and under influence of the uniaxial stress. The scaled results collapse into the single universal curve thus confirming the validity of Landau model for this case. The Brillouin spectra of quasioonedimensional ferroelectric CsH_2PO_4 show no temperature dependence in the ferroelectric region. The critical mode thus seems to be of rather low frequency (< 1 GHz).

Brillouin laser light scattering is a powerful experimental technique for studying phase transitions in various crystals via the influence of driving mechanisms on the elastic constants. Basically, we can determine sound velocities of thermally generated hypersound (~ 10 GHz) waves as the function of temperature and/or external fields. Advantage of this method lies in the fact that one often does not need highly polished and parallel sides of a crystal, one can deal with small samples and one can obtain the information in the case that the fundamental dispersion region for the elastic constants lies above the ultrasonic range of frequencies. In this paper we are showing some recent results obtained by studying the ferroelastic $\text{KH}_3(\text{SeO}_3)_2$ (KTS) and quasioonedimensional ferroelectric CsH_2PO_4 (CDP). The full account of the results will be published elsewhere^(1,2).

KTS exhibits a structural phase transition at 121 K, going from orthorhombic group mmm to monoclinic $2/m$. Note that the transition does not destroy the centre of symmetry. By symmetry the transition is purely a ferroelastic one and indeed below the transition spontaneous shear strain x_6 appears, the magnitude of which goes like $(T_c - T)^{1/2}$ (3). Such a transition should also exhibit the vanishing of associated elastic constant, that is in our case c_{55} component of elastic tensor.

We have investigated by Brillouin scattering the behaviour of the c_{55} component of the elastic tensor in the immediate vicinity of the transition point and in the ferroelastic phase as well as the influence of external shear stress conjugate to spontaneous strain on the relevant elastic constant. Such an experiment is completely analogous to the dielectric constant measurements with applied electric

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field in ferroelectric material, but has to our knowledge not been made in ferroelastic. For the measurements we have constructed a special cell which enabled us to measure the Brillouin spectra under the uniaxial stress while maintaining mK stabilization of temperature on the sample embedded in the index matching fluid. The experimental details will be published elsewhere⁽⁴⁾.

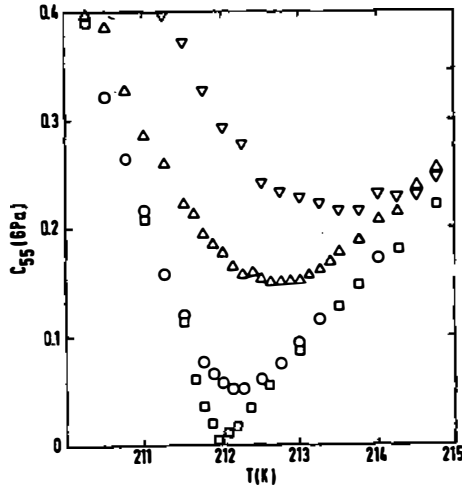


Fig. 1. c_{55} in KTS at different applied stresses.
 $\delta_5 = 0$ (\square), $= 163$ kPa (\circ), $= 325$ kPa (Δ), $= 442$ kPa (∇)

Fig. 1 shows measured elastic constant c_{55} of KTS at different applied stresses. As expected the elastic constants do harden under uniaxial stress. We have analysed the results by using the Landau type analysis of phase transitions⁽¹⁾.

The ferroelastic transition in KTS has been described in terms of a Landau free energy of the form⁽¹⁾

$$F = F_0 + \frac{1}{2} \alpha(T)Q^2 + \frac{1}{4} \beta Q^4 + \frac{1}{2} c_{55}^0 x_5^2 + \gamma x_5 Q + F_1(x_i, Q) \quad (1)$$

where Q is some internal coordinate, connected with hydrogen motion and driving the transition and $F_1(x_i, Q)$ represents all the other elastic terms and their interactions with the variable Q . $F_1(x_i, Q)$ has only a minor influence on the transition the essential point in the free energy (1) is that both Q and x_5 are of the same symmetry B_{2g} and can therefore be bilinearly coupled.

One interesting derived result is seen in Fig. 2. As we have scaled the results of Fig. 1 by using $c_{55}/\sigma_5^{2/3}$ (σ_5 is external stress) versus $(T - T_c)/\sigma_5^{2/3}$, all experimental points have collapsed on the single curve which can be calculated from the Landau theory, thus confirming it.

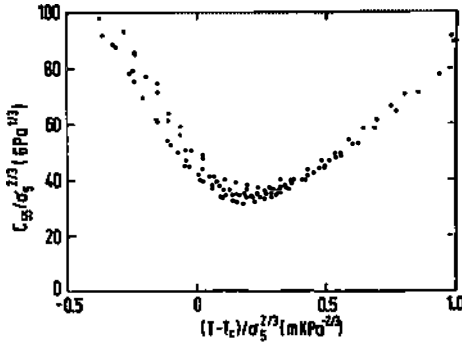


Fig. 2.
The data of Fig. 1, scaled with $\sigma_5^{2/3}$, as a function of scaled temperature

CsH_2PO_4 (CDP) undergoes a quasi-one-dimensional ferroelectric phase transition at 153 K. Unlike KH_2PO_4 where all hydrogen bonds are equivalent there are two different hydrogen bonds in the structure. The shorter bond links the phosphate groups into chains, is disordered above transition and goes over to an ordered asymmetric configuration below T_c . The longer bond crosslinks the chains into layers, is already ordered in the paraelectric phase and does not change essentially in the ferroelectric phase⁽²⁾. Quasi-one-dimensional ferroelectrics are currently the objects of intensive research. We have recently investigated a similar crystal of PbHPO_4 ⁽⁵⁾ and found a prominent dispersion region in the behaviour of c_{22} elastic constant below T_c , thus establishing the fact that the critical mode, responsible for the transition, must be of the millimeter wave frequency range. Here, we are presenting

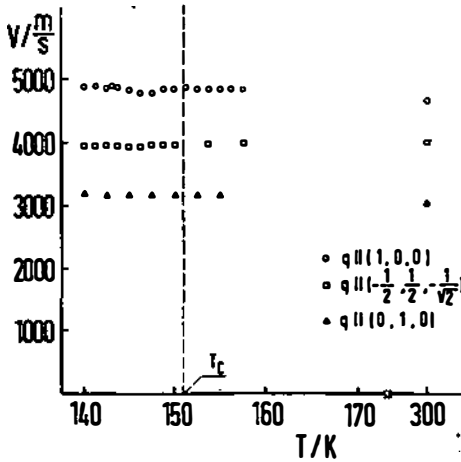


Fig. 3.
Brillouin spectra of CDP. The velocities for two longitudinal acoustic modes (\circ , $\rho v^2 = c_{11}$), (Δ , $\rho v^2 = c_{22}$) and quasi-longitudinal (\square) mode as function of temperature are shown

the Brillouin results for CDP, shown in Fig.3. Previously obtained Raman spectra have shown no soft mode in the ferroelectric phase. The obtained elastic constants c_{11} and c_{22} which should in principle undergo a jump at the phase transition following with a dispersion, show no such phenomena. This fact seems to indicate that the critical mode in CDP is of very low frequency (< 1 GHz).

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