"Splitting" of the hypersound attenuation maximum in the weak ferroelectric TSCC

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A "splitting" of the attenuation maximum into a relaxation part and a fluctuation part for hypersonic phonons which are propagating along the polar axis has been observed near the ferroelectric phase transition in TSCC crystals. This effect is characteristic of a ferroelectric with a weakly polar soft mode (a weak ferroelectric).

The intrinsic absorption of sound upon structural phase transitions is determined by the joint effects of relaxation and fluctuation mechanisms.[1] The contributions of these two mechanisms reach peak at slightly different temperatures. However, it has not previously been possible to experimentally separate these contributions, primarily because of the pronounced difference in the magnitudes of the two components.

In TSCC crystals (uniaxial ferroelectrics; a transition not involving cell multiplication; $T_c \simeq 130.8$ K), two maxima have been observed in the attenuation near T_c during the propagation of longitudinal hypersonic phonons along the polar axis (Fig. 1). One maximum is symmetric with respect to T_c, while the other is asymmetric, shifted significantly into the ferroelectric phase. This behavior of the attenuation was found through high-precision measurements of Brillouin scattering in a well-cooled, three-pass, Burleigh Fabry-Perot interferometer, with signal accumulation and subsequent analysis by means of a computer. Previous studies[2–5] had revealed only a single (asymmetric) maximum in the attenuation, despite the use of excellent apparatus.

TSCC crystals are weak[3)] ferroelectrics, i.e., crystals with a weakly polar soft mode, and are characterized by unusual temperature anomalies in several physical properties.[3,6] One of these properties is the presence of an asymmetric maximum in the attenuation of longitudinal sound, propagating along the polar axis. This maximum is shifted into the ferroeletric phase. We were the first to detect this maximum. We identified it as a maximum of the relaxation absorption distorted by a crossover in the dynamics of the order parameter from a dipole type to an Ising type.[2,3] The maximum in Fig. 1, shifted with respect to T_c, should therefore be attributed to a relaxation absorption mechanism. We wish to stress again that TSCC should not be regarded as an ordinary uniaxial ferroelectric such as TGS. If one ignores the particular features of a weak ferroelectric, one is forced to resort to some extremely contrived assumptions in order to interpret data on hypersound absorption.[4,5]

The attentuation maximum at $T = T_c$ can, in our view, be attributed to a fluctuational mechanism in a natural way. Our analysis of the high-temperature wing of this maximum agrees with this assignment. The soft mode in the paraelectric phase of

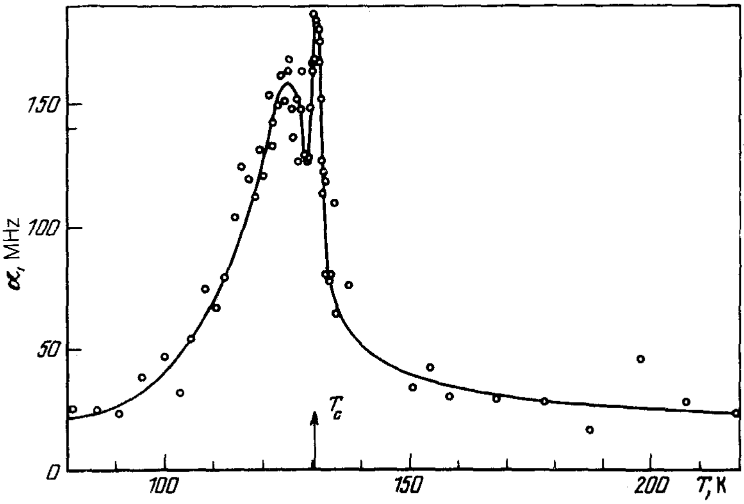

FIG. 1. Temperature dependence of the attenuation of longitudinal acoustic phonons which are propagating along the polar axis in TSCC. Shown here is the width at half-maximum of the Brillouin components of the phonon lines; the instrumental broadening has been subtracted.

TSCC is weakly attenuated,[7] at least to a temperature $T = T + 5$ K. We would thus expect to find the same temperature dependence for the fluctuation components of the imaginary and real parts of the elastic modulus.[8] Our calculations show[3] that the critical part of the elastic moduli in a weak ferroelectric in the region in which the Landau theory applies is given by $\mathrm{Re}\Delta c_{ii} \sim \ln(1 + \tau^{-0.5})$, where $\tau = (T - T_c)/T_k$, and T_k is the crossover parameter. This specific temperature dependence has indeed been confirmed experimentally in TSCC for $\mathrm{Re}\Delta c_{22}$ and $\mathrm{Re}\Delta c_{33}$. Several independent measurements revealed a parameter value $T_k \approx 7$ K in TSCC. Working from these representations, we checked the possibility of describing the critical part of the hypersonic attenuation coefficient $\Delta\alpha$ at $T > T_c$ by the same formula, i.e., by

$$\Delta\alpha(T) \sim \mathrm{Im}\Delta c_{22} \sim \mathrm{Re}\,\Delta c_{22} \sim \ln(1 + \tau^{-0.5}), \quad (1)$$

at $T_k = 7$ K. The results of this analysis are shown in Fig. 2. We see that by choosing the appropriate coefficient it is possible to fit (1) reasonably well to the experimental data over the temperature interval from $T - T_c \approx 100$ K down to $T - T_c \approx 2$ K.

As we move closer to the transition, $\Delta\alpha(T)$ initially increases substantially more rapidly, and then more slowly, than in (1), while the $\mathrm{Re}\Delta c_{22}(T)$ we immediately observe a slowing of the growth in comparison with (1). Let us compare the behavior in these cases. The deviation of $\mathrm{Re}\Delta c_{22}(T)$ from (1) is caused by two factors[3]: the dispersion of the first fluctuational correction[9] and the penetration into a region where the critical fluctuations are appreciable. Three factors are responsible for the deviation of $\Delta\alpha(T)$: the two just listed and also a transition from a resonant to a relaxational soft mode near T_c. The contributions of these factors are not additive, and it is difficult to quantitatively analyze their combined effects. It is possible, however, to offer a qualitative interpretation of the behavior $\Delta\alpha(T)$.

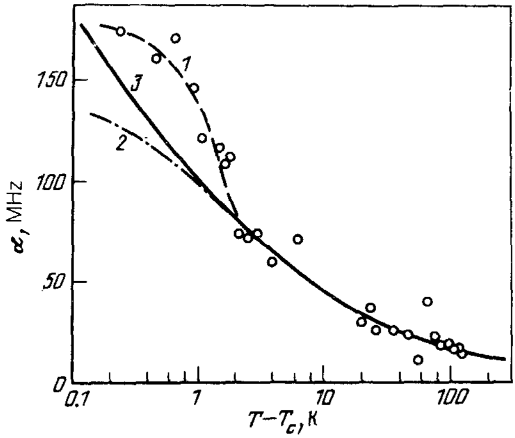

FIG. 2. 1—Temperature dependence of the critical part of the attenuation, $\Delta\alpha(T)$; 2—temperature dependence of the elastic modulus $\mathrm{Re}\Delta c_{22}(T)$ for the same phonons, from Ref. 3; 3—prediction of expression (1). The scale for curve 2 has been normalized in such a way that curves 1 and 2 coincide with 3 far from T_c.

The temperature dependence of the critical component of the attenuation can be estimated by means of the following integral for the case of the overattenuated soft mode in the region in which the Landau theory applies[9]:

$$\Delta\alpha \sim \int d^3k \ /(2\pi)^3\, \Omega_k^6 \ . \qquad (2)$$

For the region with a dipole dynamics of the order parameter, expression (2) yields $\Delta\alpha \sim (T - T_c)^{-1}$, i.e., a much more rapid change than would be expected from (1). It is thus logical to expect that the combined effects of three factors cause $\Delta\alpha(T)$ to increase more rapidly than $\mathrm{Re}\Delta c_{22}(T)$. In this case, only the first two of these factors are manifested (in agreement with experiment). We thus believe that the initial deviation of $\Delta\alpha(T)$ from (1) is due to a transition from a resonant mode to a relaxation soft mode, while the subsequent slowing is due to a dispersion, which leads to a finite attenuation at $T - T_c$.

The TSCC crystals proved to be convenient objects for a study of the "splitting" of the maximum in the attenuation of longitudinal hypersound propagating along the polar axis. In the other weak ferroelectrics which have presently been identified, e.g., $Li_2Ge_7O_{15}$, NH_4LiSO_4, $(NH_4)_2SO_4$, and $CsCoPO_4$, we would expect significantly smaller values of the parameter T_k on the basis of their dielectric properties.[6,10] In this case the "splitting" of the corresponding attenuation maximum should be considerably smaller. This small value would hinder, but in principle not rule out, the observations of this effect in these compounds.

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[3)]The term "weakly polar" was used in Refs. 3 and 6, but we now feel that is not the best term.

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