Anomalous temperature dependence of the damping of the soft mode in an incommensurate phase in the particular cases of $Cd_2Nb_2O_7$ and Rb_2ZnBr_4

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The anomalously large and slightly temperature-dependent damping of the soft mode in the Raman spectrum in an incommensurate phase suggests the presence of a phason. As $T \rightarrow T_i^-$, the temperature dependence of this damping is determined by an amplitudon.

Systems with an incommensurate phase have recently been the subject of active research by the methods of the Raman effect, neutron scattering, x-ray scattering, and nuclear magnetic resonance (NMR and nuclear quadrupole resonance).[1–8] One of the current problems in this research is to observe a low-frequency mode corresponding to a phason, and another is to study the behavior of a phason in the incommensurate phase. Research on an incommensurate phase by NMR and NQR[4-8] has reliably demonstrated the presence of a phason in this phase and the existence of solitons near T_c (the temperature corresponding to the transition from an incommensurate phase to a commensurate one). In the present letter we report the first observation of a phason in an incommensurate phase based on measurements of the anomalous damping of the soft mode in the Raman spectrum.

For the experiments we selected $Cd_2Nb_2O_7$ crystals. In the temperature interval $T_c = 46$ K $< T < T_i = 80$ K these crystals exhibit an anomalous thermal hysteresis and an anomalous temporal instability of ϵ and $\tan \delta$ which have been attributed to the presence of a phason[9] (T_i is the temperature corresponding to the normal-incommensurate phase transition). The soft mode, which appears in the Raman spectrum below T_i, is intense and well defined between 4 and ~77 K (Ref. 10), so that its damping $\Gamma(T)$ can be studied over a broad temperature range.

Figure 1 shows the temperature dependence of the damping of the soft mode in $Cd_2Nb_2O_7$. Below T_i the damping decreases with decreasing temperature, exhibiting the behavior observed in any structural phase transition in which a soft mode appears. Over the greater part of the incommensurate phase, however, the $\Gamma(T)$ behavior in $Cd_2Nb_2O_7$ is anomalous. Instead of monotonically decreasing with decreasing temperature, the soft-mode damping remains anomalously large and slightly temperature-dependent. As we go from the incommensurate phase to the commensurate one ($T < T_c$), the damping $\Gamma(T)$ decreases, and below T_c it behaves as the damping of the ordinary soft mode does.

We have also analyzed the Raman spectra of Rb_2ZnBr_4, recorded by other investigators.[11,12] From this analysis we determined the temperature dependence of the damping of the soft mode, which appears in the spectrum of this crystal below $T_i = 355$ K (Fig. 2). From Figs. 1 and 2 we see that the damping of the soft mode behaves identically with decreasing temperature in Rb_2ZnBr_4 and $Cd_2Nb_2O_7$. Further-

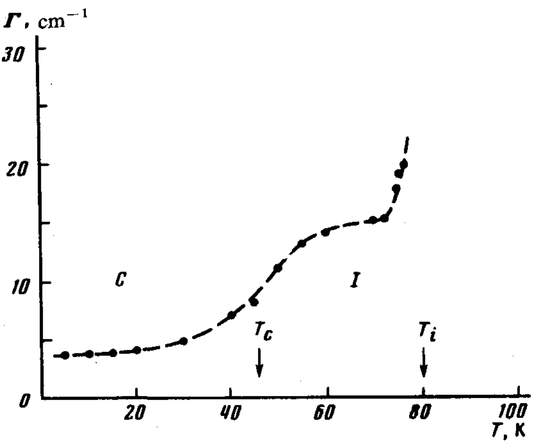

FIG. 1. Temperature dependence of the damping of the soft mode in $Cd_2Nb_2O_7$ in the commensurate phase (C) and in the incommensurate phase (I).

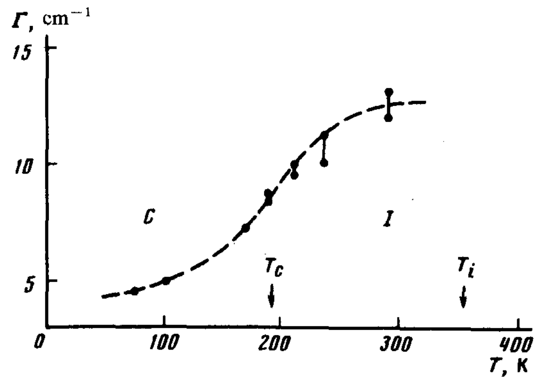

FIG. 2. Temperature dependence of the damping of the soft mode in the C and I phase in Rb_2ZnBr_4.

more, the temperature dependence of the damping in these crystals is similar to that of the rate of nuclear spin-lattice relaxation in Rb_2ZnBr_4 (Ref. 5) and Rb_2ZnCl_4 (Ref. 6) at $T < T_i$. It has been shown[5–8] that the decrease in the rate of nuclear spin-lattice relaxation with decreasing temperature near T_i is determined by an amplitudon and that the anomalous behavior of the spin-lattice relaxation rate in the incommensurate phase is due to a phason.

Pursuing the analogy between the behavior of the soft-mode damping in the Raman effect and the behavior of the rate of spin-lattice relaxation, we suggest that the anomalous behavior of the damping of the soft mode in the incommensurate phase is also due to a phason.

We know from the theory of incommensurate phase transitions[13,14] that two quasinormal modes, an amplitudon and a phason, appear in the excitation spectrum below T_i. Their frequencies, ω_A and ω_ϕ, are determined by the dispersion relations

$$\omega_\varphi^2(\mathbf{k}) = bk^2, \qquad T_c < T < T_i,$$

$$\omega_A^2(\mathbf{k}) = 2a(T_i - T) + bk^2, \quad T < T_i,$$

where $\mathbf{k}$ is the wave vector, and b and a are constants.

It can be seen from these relations that in the limit $\mathbf{k}\to 0$ the frequency of the amplitudon remains finite in the incommensurate phase, and it increases in proportion to $(T_i - T)^{1/2}$ with decreasing temperature. In the limit $\mathbf{k}\to 0$ the phason frequency approaches zero (if the phason is not pinned by a defect), and it does not depend on the temperature in the "plane-wave limit." In the many-soliton limit ($T \to T_c^+$) the phason frequency, which is related to a change in the soliton density,[6,13] depends on the temperature. As we go from the incommensurate phase to a commensurate one ($T < T_c$), the phason frequency increases abruptly, while the frequency of the amplitudon changes monotonically.

In summary, in the incommensurate phase, $T_c < T < T_i$, we can distinguish qualitatively different regions in the temperature dependence of the soft-mode damping.

These different regions reflect changes in the frequencies of the amplitudon and the phason with decreasing temperature. Near T_i the soft-mode damping, which decreases with increasing value of $(T_i - T)^{1/2}$, is determined by the amplitudon, while in the greater part of the incommensurate phase this damping depends weakly on the temperature and is determined by a phason. The decrease in $\Gamma(T)$ in the many-soliton limit $(T \to T_c^+)$ is apparently due to a decrease in the soliton density.

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