Rotation-translation coupling in $(NH_4I)_{0.5}(KI)_{0.5}$

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Abstract

Inelastic neutron scattering experiments have been performed to study the temperature dependence of the LA-phonon frequency and the intensity of the quasielastic component in the orientational glass $(NH_4I)_{0.5}(KI)_{0.5}$. The results can be explained assuming a coupling of the translational phonon modes to the orientational degrees of freedom. Deviations of the experimental results from model predictions signal the onset of glassy freezing.

Keywords: Phonons; Glasses; Elastic constants

During the last few years glasses have attracted considerable attention. In order to investigate the dynamic character of the glass transition, model systems have been chosen. Here orientational glasses (OG) served as a conceptual link for an understanding of the dynamic processes in glasses [1]. In this communication we report on phonon properties of the dipolar glass $(NH_4I)_x(KI)_{1-x}$. Below critical concentration x_c, the mixed systems $(NH_4I)_x(KI)_{1-x}$ are representative of a large family of OG which show a collective freezing of the orientational degrees of freedom. Above x_c a rich structural polymorphism with complex long-range orientational order has been detected [2,3]. Central peak (CP) phenomena appear when approaching the low-temperature orientational glass phase [3–6]. The CP intensities showed anomalous behaviour and a minimum has been found in the temperature dependence of the TA phonon frequencies at the X-point at about $T \cong 100$ K. The results have been interpreted in terms of a rotation-translation (RT) coupling in which the lattice vibrations soften due to the coupling to rotational modes of the NH_4^+-tetrahedra.

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In this short communication we focus on the results of the measurements of the central peak and the longitudinal acoustic (LA) phonon at the reciprocal lattice point $\boldsymbol{Q} = (4.2\ 0\ 0)$. The measurements were performed on the triple-axis spectrometer IN8 located on a thermal beam hole at the ILL, Grenoble. The inelastic scans have been performed with an experimental resolution of about 0.85 meV in the constant $k_f = 4.1\ \text{Å}^{-1}$ mode. A single crystal with size $8 \times 15 \times 20\ \text{mm}^3$ has been investigated. The lattice parameters and the spectrometer configuration have been readjusted at each scan to improve the accuracy of the phonon position.

Fig. 1 shows the scattered intensities as a function of energy transfer at different temperatures. Focusing on the spectrum measured at $T = 287$ K one finds a contribution from the phonon excitation and the CP. On cooling, the intensities of the phonon contribution decreases in favour of the CP intensity. The spectra have been fitted by a superposition of the phonon mode and two further contributions for the CP. The CP intensity represents a convolution of a Lorentzian QE line which strongly increases on decreasing temperatures and a temperature-independent Gaussian line, which represents elastic incoherent scattering. The squared phonon energies and the CP intensity are shown in Fig. 2. The squared phonon energies of the LA mode decrease on cooling and subsequently reach a minimum at about $T \cong 90$ K (Fig. 2a). They can be described using an ansatz developed for crystals characterized by a RT

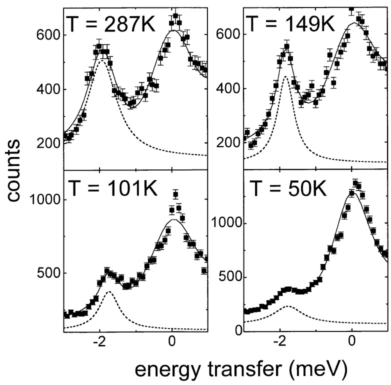

Fig. 1. Scattered neutron intensities at the reciprocal lattice point $\boldsymbol{Q} = (4.2\ 0\ 0)$ for several temperatures. The dashed line is a fit to the longitudinal acoustic phonon contribution and the solid line represents the sum of the fits of the phonon and the central peak.

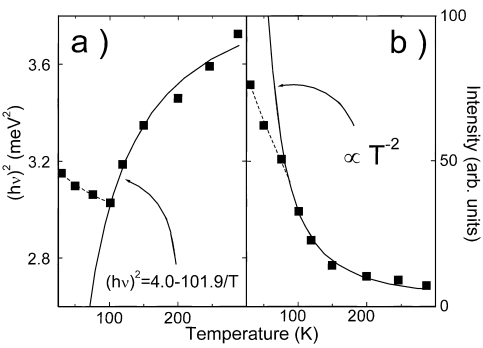

Fig. 2. Temperature dependence of the squared LA phonon energy (left plot) and of the intensity of the quasielastic contribution (right plot) versus temperature. Solid lines represent fits as described in the text. Dashed lines are drawn to guide the eye.

coupling [7]:

$$(h\nu)^2 \propto C_{11}(T) = C_{11}^0 (T - T_0)/(T - T_1),$$

where T_0 denotes the structural phase transition temperature if hypothetically the collective freezing would be suppressed. At this temperature the quadrupolar susceptibility diverges. T_1 characterizes the interactions between the NH_4^+-molecules and have been neglected in the fit. The softening of the LA frequencies can be reasonably described using this equation. However, significant deviations appear for $T < 90$ K where the reorientational motion of the molecules becomes much slower than the time window of the experiment and the phonons strengthen again in a lattice with quasi-frozen – in molecular reorientations. The intensity of the central peak strongly decreases on heating (Fig. 2b). OG glasses are dominated by random strains in which the CP intensity represents the characteristic order parameter and should follow a T^{-2} behaviour [8]. Deviations appear again below $T < 90$ K. We conclude that a coupling exists between the LA phonon and the orientational degrees of freedom of the ammonium tetrahedron. Below $T < 90$ K the cooperative freezing of the orientational degrees of freedom are responsible for the incomplete softening of the LA phonon.

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