Central peaks in $(NH_4I)_x(KI)_{1-x}$

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Abstract

Quasielastic measurements are reported on $(NH_4I)_x(KI)_{1-x}$. This mixed molecular system reveals a variety of structural phases and different glass states. Close to the structural phase transition, in constant Q-scans through the reciprocal lattice points at which superstructure reflections appear in the orientationally ordered phase, two central peaks were detected.

In plastic crystals the molecules occupy the sites of a regular center-of-mass lattice. At high temperatures most of these crystals reveal orientationally disordered states in which the molecules undergo fast reorientational motions. On cooling, these relaxations slow down and the pure systems reveal an orientationally ordered state at low temperatures. However, when the molecular units are statistically diluted by spherical ions, in most cases the phase transitions are suppressed and the mixed crystals reveal an orientational glass state. For many years these orientational glasses (OG) served as conceptual links between phase and glass transition phenomena [1]. In recent years the relaxation dynamics in OG has been studied in full detail. If the pure crystals show a doubling of the unit cell driven by a low-temperature phase transition and if in the mixed system this phase transition is almost or just avoided, central peak phenomena are expected which can be studied in full detail. $(NH_4I)_x(KI)_{1-x}$ is a mixed molecular system with a FCC structure at room temperature which reveals a variety of different low-temperature phases [2]. Central peak phenomena appear in the ε phase and in the glass phase.

Measurements were performed on the triple axis spectrometer E7 located on a thermal beam hole at the HMI Berlin. High-quality single crystals with ammonium concentrations $x = 0.55$, 0.65, 0.75 were mounted in a standard orange-type cryostat. Constant-Q and constant $E = 0$ scans were performed with an incident wave vector $k_i = 2.64$ Å^{-1} and collimator settings 20′/20′/20′/30′. Crystals with ammonium concentrations $x = 0.55$, 0.65 and 0.75 were investigated. In this short communication we will focus on the results for $x = 0.75$.

In $(NH_4I)_{0.75}(KI)_{0.25}$ the transition into ε phase appears at $T_c = 62$ K [3]. This phase reveals a complex orientational order with a slightly distorted FCC lattice of trigonal symmetry. We investigated the temperature dependence of the quasielastic scattered intensity at the (3 0 0) superlattice reflection which appears close to the structural phase-transition temperature. In Fig. 1 a representative constant-Q scan at the (3 0 0) reciprocal lattice point is shown for $T = 50.2$ K. In a careful analysis of these data we established that it is impossible to describe the shape of the quasielastic intensities with a single line (Lorentzian, squared

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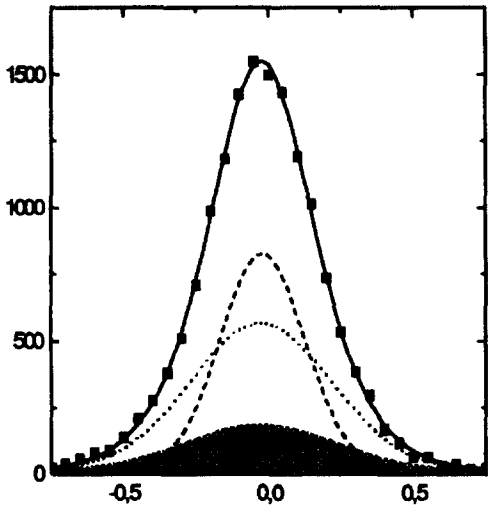

Fig. 1. Constant Q-scan in $(NH_4I)_{0.75}(KI)_{0.25}$ at the (3 0 0) reciprocal lattice point at $T = 50.2$ K. The filled area indicates the incoherent background. The measured line shape (dotted line) was fitted assuming two central components (dashed and dotted lines) in addition to the background contribution.

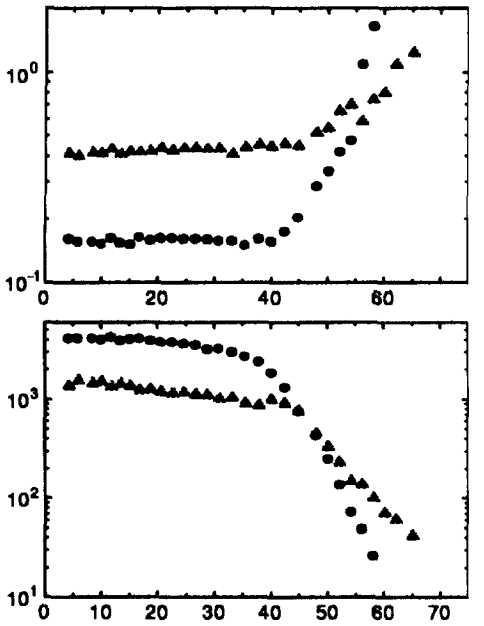

Fig. 2. Temperature dependence of the full-width at half-maximum (upper frame) and of the intensities (lower frame) of the two central components (triangles and circles) as measured at the (3 0 0) reciprocal lattice point in $(NH_4I)_{0.75}(KI)_{0.25}$.

Lorentzian or Gaussian). Instead it was necessary to introduce two lines of different width (dashed and dotted lines in Fig. 1). So far two central peaks in structural phase transitions have been observed in constant $E = 0$ scans in $SrTiO_3$ and were attributed to scattering from the surface and the bulk material [4]. Fig. 2 shows the evolution of the central peak intensities and of the full-width at half-maximum (fwhm) as a function of temperature in a semilogarithmic representation. One peak appears close to 60 K and saturates below 40 K (circles). Its half-width decreases continuously until it reaches the experimental resolution (quasielastic half-width of a Bragg peak). Below 40 K this intensity corresponds to the superstructure Bragg-reflections of the ε phase revealing static and long-range orientational order. The finite width of the quasielastic intensities close to 60 K reveals critical fluctuations at the structural phase transition. But in addition to this rather well-known phenomenon a second central peak appears just below 70 K. At low temperatures this intensity is by more than a factor 5 lower than the superstructure reflections. The width at low temperatures is limited by the energy resolution of the instrument as determined via vanadium scans (0.5 meV). The origin of this second central component is unclear at the moment. It indicates a second time scale of a relaxational process and may indicate rotations of the ammonium tetrahedrons arround the trigonal axis, which slow down with decreasing temperature. It is clear that critical fluctuations as well as disorder effects are relevant in the interpretation of the two time scales involved in $(NH_4I)_{0.75}(KI)_{0.25}$.

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