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FERROELASTICITY AND GLASS-LIKE BEHAVIOR IN ALKALI HALIDE-ALKALI CYANIDE MIXED CRYSTALS

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Single crystal neutron diffraction studies in $(KBr)_{1-x}(KCN)_x$ are summarized. Mixed crystals with CN^- concentrations $x > 0.6$ exhibit ferroelastic phase transitions from a high-temperature plastic phase into a low-temperature elastically ordered phase in which the CN^- orientations show long range orientational order and the center of mass lattice exhibits shear distortions. For concentrations $x \leq 0.6$ orientational disorder is frozen-in and transitions into an orientational glass state occur. Close to the critical concentration $x_c \sim 0.6$ the diffraction profiles at the transition temperatures are dominated by diffuse-scattering contributions. These results are compared to model calculations from Mayer and Cowley predicting a two-dimensional melting transition in a crystal with a planar instability. In the spirit of this model we suggest that the glass state close to x_c can be described as a frozen-in two dimensional liquid. A schematic phase diagram is constructed which bears similarities with a phase diagram as proposed by Galam within the framework of a compressible ferroelastic model.

1 INTRODUCTION

Solid solutions of alkali halides and alkali cyanides have been studied intensively during the last decade (Lüty, 1981, 1990; Knorr, 1987; Loidl, 1985, 1989). These mixed crystals are model systems for the study of ferroelastic phase transitions and for transitions into an orientational glass state. Pure KCN exhibits a "plastic" high-temperature phase where the cubic symmetry is established and stabilized via a fast reorientational motion of the aspherical CN^- ions. The orientational distribution in this dynamically disordered phase has been determined by Rowe, Hinks, Price, Susman and Rush (1973) and by Loidl, Knorr, Rowe and McIntyre (1988). The maximum probability for orientations of the CN^- ions was found to be directed along [111] while the [110] directions exhibited a minimum probability. The orientational degrees of freedom of the dumb-bell shaped CN molecule are strongly coupled to the rotational degrees of freedom (Michel and Naudts, 1977). This rotation-translation coupling is responsible for an effective CN^-–CN^- interaction which is mediated via lattice strains. As a consequence, at $T_s = 168$ K KCN exhibits a ferroelastic phase transition into a low-temperature orthorhombic phase in which the CN^- ions are oriented along the former [110] axes. The phase transition is accompanied by a strong softening of the elastic shear constant c_{44} (Haussühl, 1973). Although the softening of the transverse elastic constant is almost complete, the phase transition is strongly of first order (Suga, Matsua and Seki, 1965). The CN^- ions carry, in addition to their elastic (quadrupole) moment, an electric dipole moment of the order of 0.3D. At $T_{AFE} = 81$ K the residual head-to-tail disorder of the orthorhombic phase is removed and the CN-dipoles order antiferroelectrically (Suga *et al.*, 1965).

Substitution of the aspherical CN^- molecules by spherically shaped Br^- ions reduces the ordering temperatures. At first glance this can be thought to result from the reduction of the rotation-translation coupling. However, the coefficient dT_s/dx is too large if only this effect is taken into consideration and finally, below a critical concentration x_c, long range orientational order is fully suppressed and the mixed crystals undergo an orientational glass transition (Höchli, Knorr and Loidl, 1990). In close analogy to the experimental observations in spin glasses (Binder and Young, 1986), the glass transition is characterized by a cooperative freezing-in of random CN^- orientations. At present it is unclear to what extent long range orientational order is suppressed by random fields which are introduced by the substitutional impurity atoms (Michel, 1986, 1987) or by the interplay of site disorder and anisotropic CN–CN interactions yielding a frustrated ground state (Binder, 1990; Galam, 1990). In the glass state the mixed cyanides exhibit low temperature thermal (DeYoreo, Knaak, Meissner and Pohl, 1986), ultrasonic (Berret, Doussineau, Levelut, Meissner and Schön, 1985) and dielectric properties (Moy, Dobbs and Anderson, 1984) which are similar to those found in canonical glasses and in amorphous solids. In amorphous systems the low temperature properties appear to be dominated by two-level tunneling centers (Anderson, Halperin and Varma, 1972; Phillips, 1972); in the glassy state of KBr:KCN an attempt was made to identify the tunneling centers with dipolar reorientations (180°-flips) of the CN^- ions (Sethna and Chow, 1985).

This article is organized as follows: after a short introduction into the static properties of $(KBr)_{1-x}(KCN)_x$ mixed crystals we focus on quasielastic neutron scattering results at the phase and glass transitions in mixed crystals close to the critical concentration. The experimental results are interpreted within a model of a two-dimensional melting transition which has been developed by Mayer and Cowley (1988). From these experimental results we deduce a schematic phase diagram which bears similarities with a phase diagram proposed by Galam (1990) resulting from a model of broken percolation.

2 PHASE DIAGRAM

A complete x,T-phase diagram of $(KBr)_{1-x}(KCN)_x$ on the basis of X-ray, neutron and specific heat measurements has been constructed previously by Knorr and Loidl (1985), Mertz and Loidl (1987) and Loidl *et al.* (1989). Figure 1 shows the phase diagram as published by Loidl *et al.* (1989). With decreasing CN^- concentration the ordering temperatures T_s and T_{AFE} are reduced significantly. But, astonishingly, the solid solutions exhibit a variety of different crystallographic low-temperature phases: with decreasing x the sequence orthorhombic, monoclinic and rhombohedral can be identified. Finally, below a critical concentration $x_c \sim 0.6$ cubic symmetry is, at least on the average, not broken down to the lowest temperatures. However, as we will

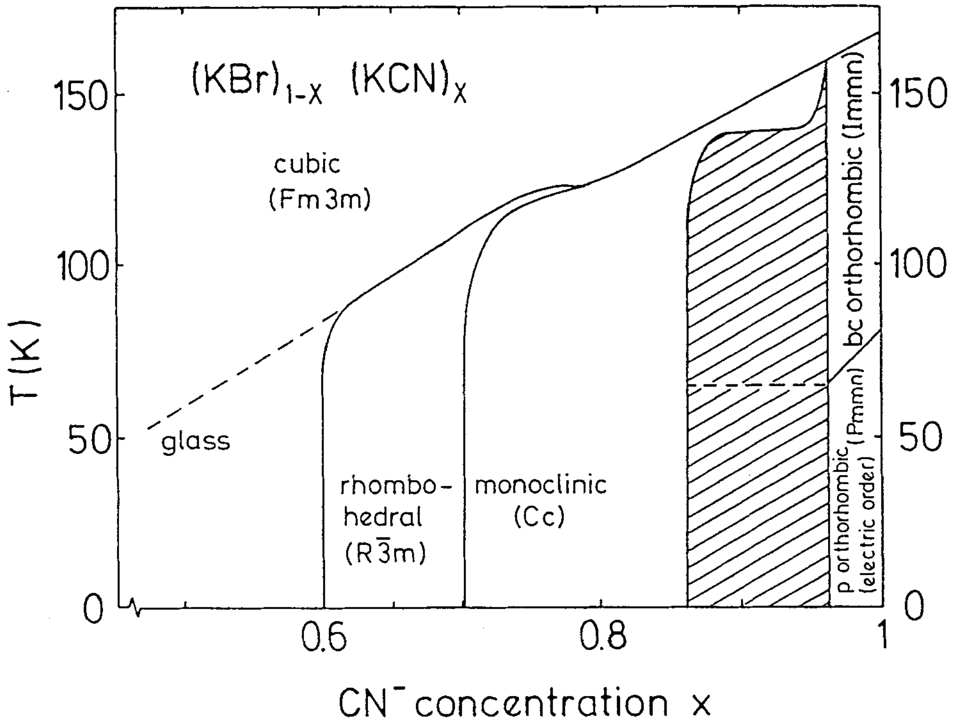

Figure 1 x–T phase diagram of $(KBr)_{1-x}(KCN)_x$. The solid lines give an estimate of the phase boundaries between different crystallographic phases. From Loidl *et al.* (1989).

see later, structural anomalies can be detected which reveal the existence of an orientational glass state below x_c and at low temperatures.

As can be seen from Figure 1, the orthorhombic and monoclinic phases are separated by a broad coexistence region. Figure 1 also reveals that electric order can only be established in the orthorhombic phase. All the other low-temperature states exhibit residual dipolar disorder down to the lowest temperatures. The polymorphism is a striking feature of the phase diagrams of all mixed cyanides (Höchli *et al.*, 1990) and is reminiscent of the polymorphism observed in a number of canonical glasses. It is however still an open question whether or not the tendency to form different structural phases is directly related to the tendency to form a glassy state. The ability of many covalent liquids to form a glass appears to be a direct consequence of the indifference of the crystalline network structure with respect to distortions (Jäckle, 1986).

The structural phase transition becomes almost second order close to the critical concentration x_c. This was concluded directly from the heat capacity anomalies at T_s. Figure 2 shows the heat capacity c_p in $(KBr)_{1-x}(KCN)_x$ for different concentrations vs temperature as determined by Mertz and Loidl (1987). The cubic to noncubic phase transitions show up as anomalies in the temperature dependence of c_p.

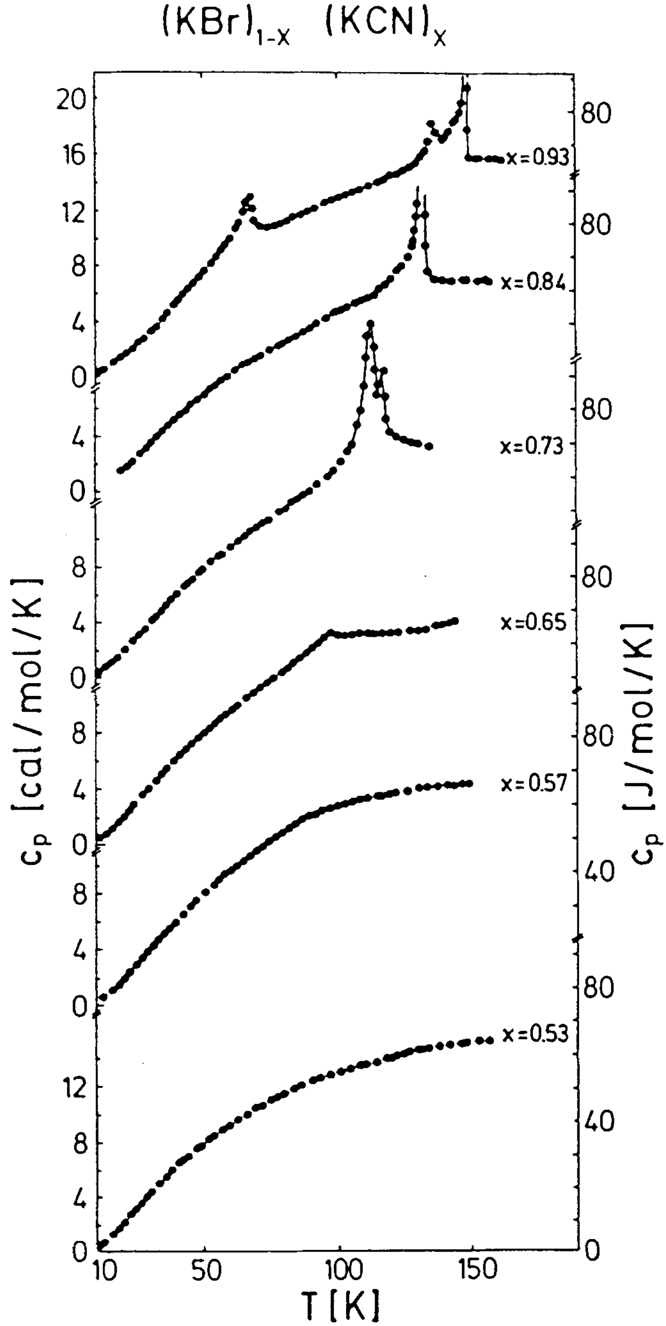

Figure 2 Heat capacity of $(KBr)_{1-x}(KCN)_x$ mixed crystals. From Mertz and Loidl (1987).

However, the entropy changes at the structural phase transitions dramatically decrease with decreasing concentration. Finally, for $x = 0.65$ as a function of T the heat capacity exhibits a change of slope only, which is located at the cubic rhombohedral phase transition temperature. For concentrations $x < x_c c_p$ varies smoothly with temperature and no characteristic glass transition temperature can be deduced from these quasi-adiabatic heat capacity experiments.

3 THE TWO-DIMENSIONAL MELTING TRANSITION

In $(KBr)_{1-x}(KCN)_x$ the strong coupling of the molecular reorientations to the phonon modes results in a soft elastic constant c_{44}. The shear constant c_{44} is proportional to the sound velocity of transverse waves of T_{2g}-symmetry. Figure 3 shows the temperature dependence of c_{44} in $(KBr)_{1-x}(KCN)_x$ for several concentrations: $x = 0$, 0.008, 0.035, 0.16 and 0.2 (from Feile, Loidl and Knorr, 1982); $x = 0.49$, 0.57 and 0.76 (from Garland, Kwiecien and Damien, 1982) and $x = 1$ (from Haussühl, 1973). The solid lines were calculated using a Curie–Weiss law for the quadrupolar susceptibility (Feile *et al.*, 1982). The crystals with $x = 1$ and 0.76 undergo structural phase transitions. For lower concentrations x the solid solutions undergo transitions into an orientational glass state. These mixed crystals stay (not locally but on the average) cubic down to the lowest temperatures. The freezing transition in orientational glasses is characterized by cusp-like anomalies in the appropriate susceptibilities. The cyanide glasses are of quadrupolar-type and the quadrupolar susceptibility of T_{2g}-symmetry reveals a frequency dependent cusp. Hence c_{44} is expected to pass through a minimum and to recover again for further decreasing temperatures. The temperature of the minimum elastic constant has operationally been defined as frequency dependent freezing temperature $T_f(\omega)$ (Loidl, Feile and Knorr, 1982; Volkmann, Böhmer, Loidl, Knorr, Höchli and Haussühl, 1986). In ultrasonic experiments the signal close to the minimum elastic constant often is lost due to heavy damping. Figure 3 reveals that for $x = 0.57$ and 0.49 the signals in the ultrasonic experiments are lost already far above the expected minimum. For the concentrations $x = 0.16$ and 0.2 however, the signals were recovered again at low T. In low frequency torsion pendulum experiments $c_{44}(T)$ can be followed down to the lowest temperatures (Knorr, Volkmann and Loidl, 1986) and for $x = 0.5$ a deep minimum close to 75 K has been detected. The elastic data reveal that for $x \rightarrow x_c c_{44}(T_s)$ approaches zero and the structural phase transition becomes second order. For $x = 0.73$ this has been demonstrated unambiguously in an inelastic neutron scattering study by Knorr, Loidl and Kjems, 1985).

Ferroelastic instabilities have been classified by Cowley (1976) and by Folk, Iro and Schwabl (1976, 1979) with renormalization group theory. For cubic systems T_{2g} modes set up a two-dimensional manifold (c_{44} occurs in planes of the reciprocal space). Hence, the structural instability in $(KBr)_{1-x}(KCN)_x$ is a planar one and belongs to the universality class $m = 2$ in the definition of the classification schemes by Cowley (1976) and Folk *et al.* (1976, 1979). A planar instability affects the thermodynamic averages and leads to diverging critical fluctuations of T_{2g} symmetry at T_s. As a consequence, the Debye–Waller factor is predicted to diverge logarithmic-

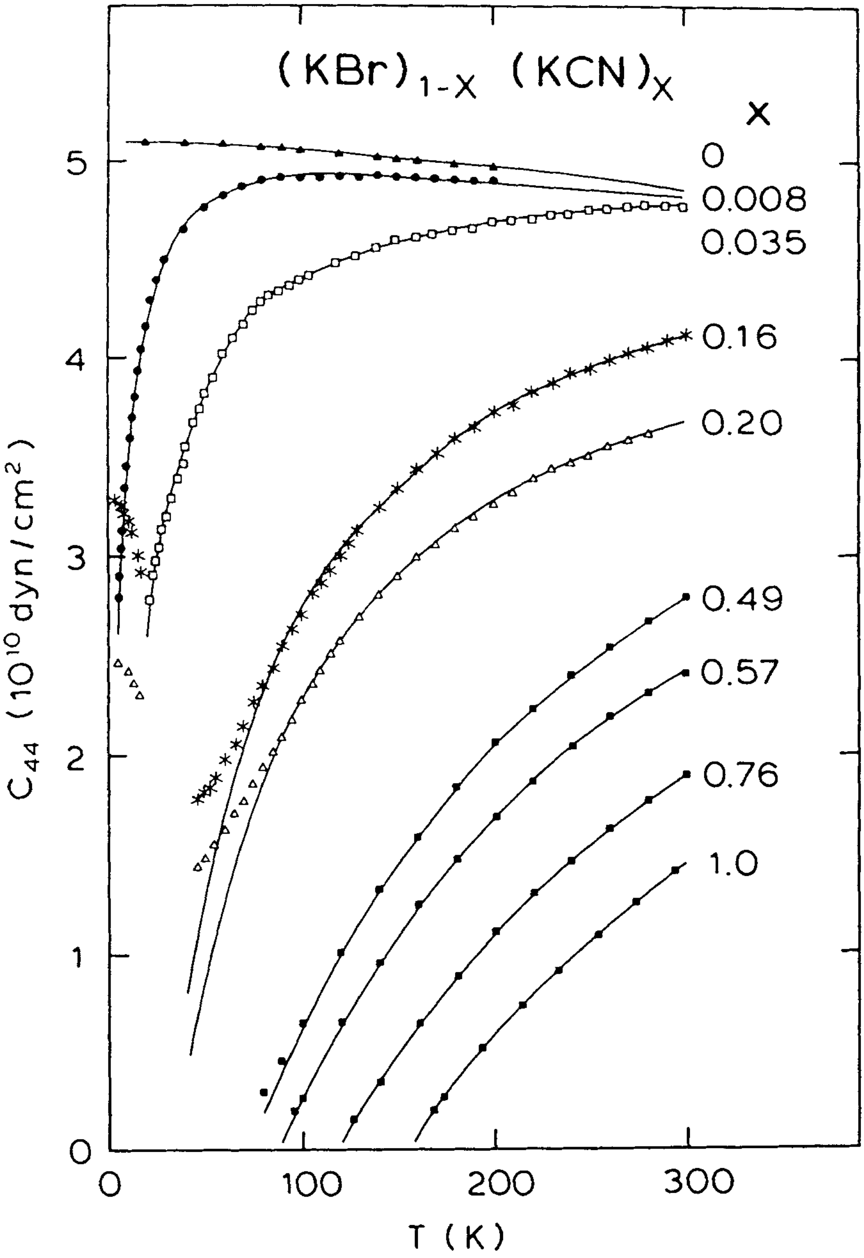

Figure 3 Elastic constant c_{44} vs. temperature for several concentrations x. Results from Feile *et al.* (1982) ($x = 0$, 0.008, 0.035, 0.16, 0.2), from Garland *et al.* (1982) ($x = 0.49$, 0.57, 0.76) and from Haussühl (1973) ($x = 1$). The solid lines were calculated assuming a Curie–Weiss law for the quadrupolar susceptibility. From Feile *et al.* (1972).

ally when the critical temperature is approached and hence, the Bragg intensities should disappear. Mayer and Cowley (1988) showed that this divergence destroys long range order and a continuous melting transition has been predicted. Particularly, they calculated the scattering cross section at T_s and found that the Bragg peaks disappear and are replaced by power law singularities. They derived quantitative results predicting $S(Q) \sim 1/q^{\delta}$, where $\delta = 2\,[2 - a(Q, T)]$ just above T_s. Here Q is the momentum transfer, q the phonon wave vector and $a(Q)$ a coefficient depending on Q and T.

4 EXPERIMENTAL RESULTS

Detailed diffuse and quasielastic neutron scattering experiments in $(KBr)_{1-x}(KCN)_x$ close to the critical concentration have been performed by Loidl, Müllner, McIntyre, Knorr and Jex (1985) and by Loidl *et al.* (1988, 1989). A single crystal X-ray investigation has been reported by Knorr and Loidl (1986). Some of these results can be compared directly to the theoretical predictions by Mayer and Cowley (1988). $(KBr)_{0.35}(KCN)_{0.65}$ reveals a structural phase transition from the high-temperature cubic into the low-T rhombohedral phase at 97.5 K. The temperature dependence of transverse scans through the (060) reflection is shown in Figure 4 (from Loidl *et al.*, 1988). The experimentally observed intensities were fitted assuming an exponential line shape for the diffuse scattered intensities (shaded area) and a Gaussian line shape for the "true" Bragg intensities (empty area). At 117 K only a small diffuse component appears below a well defined Bragg-like scattering. However, with decreasing temperatures the diffuse intensity rapidly increases, while at the same time the Bragg spike strongly decreases. Close to T_s the (060) Bragg reflection has almost disappeared and has been replaced by an anomalous scattering function which follows an exponential behavior $I \sim I_0 \exp(-\lambda q)$. Below T_s Bragg scattering recovers again and well defined rhombohedral satellites become apparent. However, even at temperatures 20 K below the ferroelastic phase transition a considerable fraction of scattered intensities shows up in the diffuse components. The temperature dependence of the Debye–Waller factor is shown in Figure 5 (from Loidl *et al.*, 1988). A strong increase of the mean square displacements seems to indicate a divergence at T_s and thus, indeed suggests a two dimensional melting transition.

Equal intensity contour plots around the reciprocal lattice points reveal that the local shear distortions are of T_{2g}-symmetry. The pioneering experiments have been performed by Rowe, Rush, Hinks and Susman (1979) and an interpretation was given by Michel and Rowe (1980). Figure 6 (from Loidl *et al.*, 1989) shows equal-intensity control lines of the diffuse scattered intensities around the (020) and the (440) reflections for $x = 0.65$ just above the structural phase transition. Two facts appear to be relevant: i) the contour lines exhibit an extreme anisotropy and ii) the (*hh*0) reflections show, in addition an asymmetry along the cube axis. Point i) implies that longitudinal scans are only less affected by contributions from diffuse scattered intensities and hence, that the lattice constant a of the cubic lattice is still well defined. The cubic lattice of the high temperature phase becomes disordered via a distribution

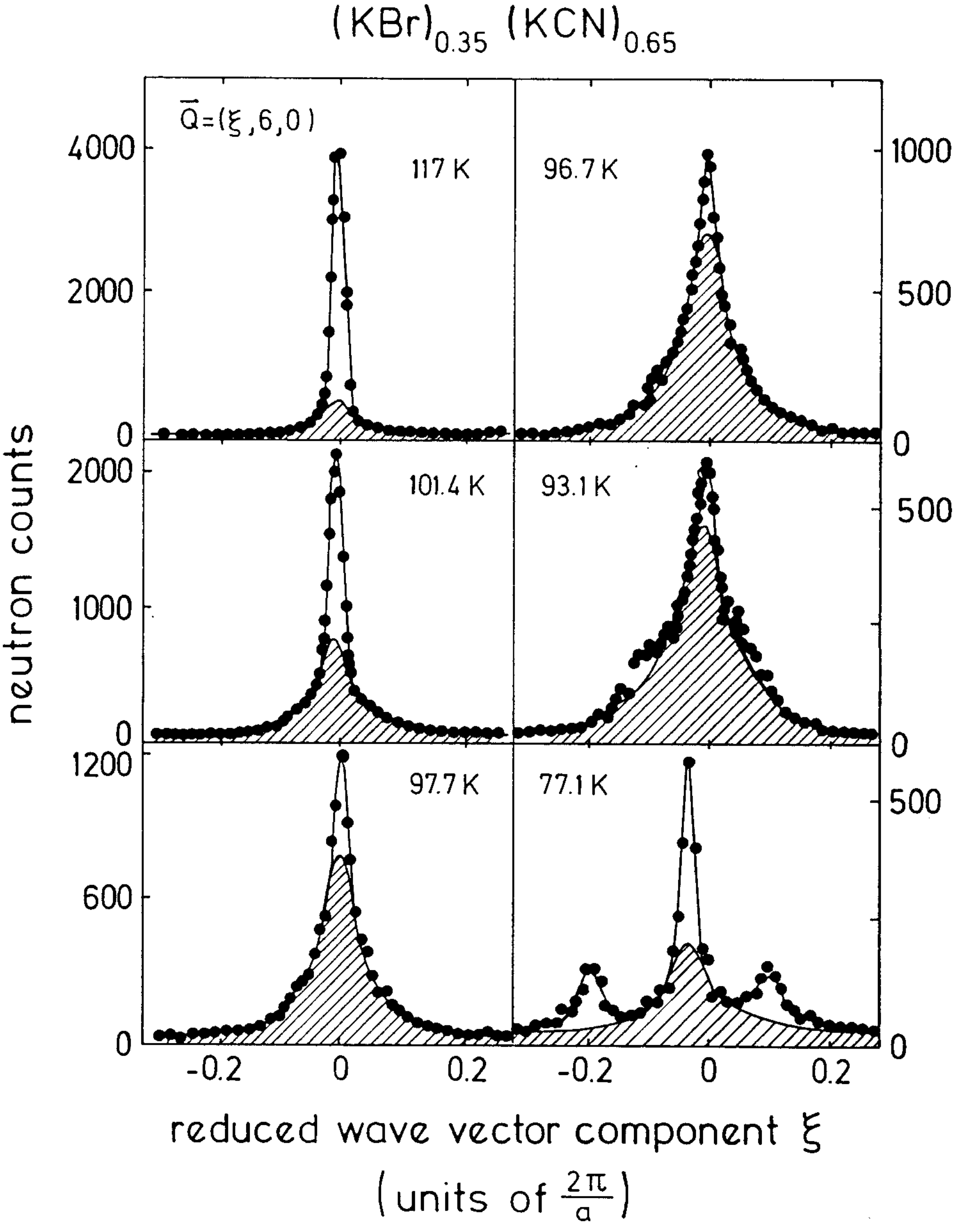

Figure 4 Transverse scans at zero energy transfer through the (060) reciprocal lattice point in $(KBr)_{0.35}(KCN)_{0.65}$ at various temperatures. From Loidl *et al.* (1988).

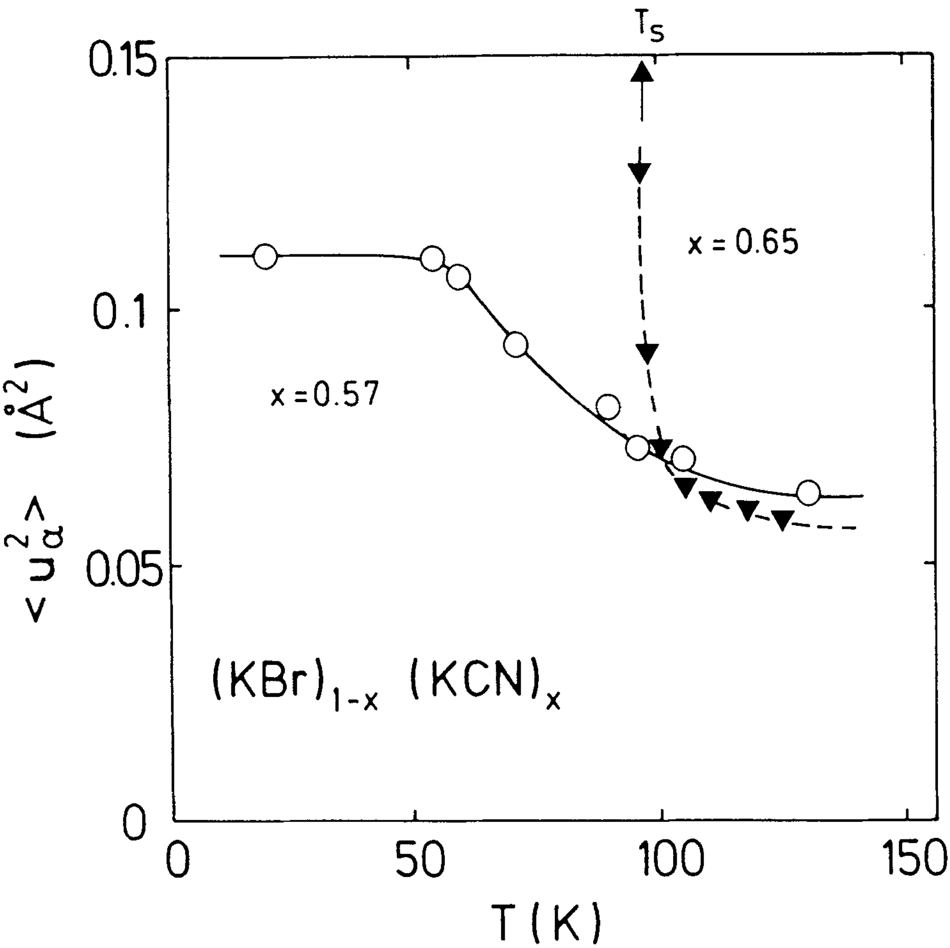

Figure 5 Temperature dependence of the mean square displacements in $(KBr)_{1-x}(KCN)_x$ for concentrations $x = 0.57$ and $x = 0.65$. T_s for $x = 0.65$ is indicated by an arrow. From Loidl *et al.* (1988).

of local shear distortions only. This effect has also been predicted by Mayer and Cowley (1988). The asymmetry ii) can be explained by the bilinear coupling of rotations and shear distortions (Michel and Rowe, 1980).

For comparison and to get some insight into the statics of the orientational glass state, similar experiments have been performed in $(KBr)_{1-x}(KCN)_x$ below the critical concentration (Loidl *et al.*, 1988, 1989). Figure 7 shows transverse scans at zero energy transfer through the (060) reflection in the (001) plane of $(KBr)_{0.43}(KCN)_{0.57}$ at various temperatures (from Loidl *et al.*, 1988). At 105 K only a small diffuse component is visible below a well-defined Bragg spike. With decreasing temperatures the diffuse contributions gradually increase while the Bragg scattering decreases.

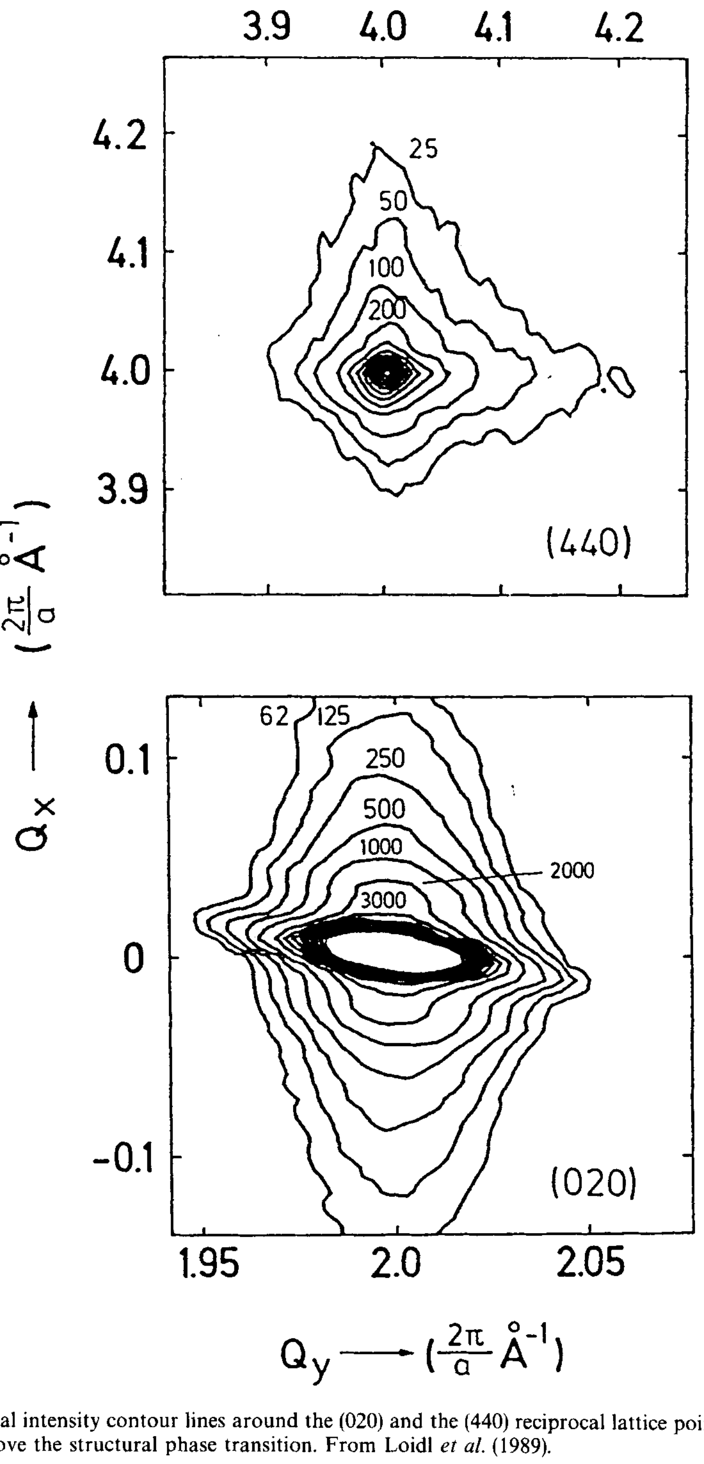

Figure 6 Equal intensity contour lines around the (020) and the (440) reciprocal lattice point in the 65% crystal just above the structural phase transition. From Loidl *et al.* (1989).

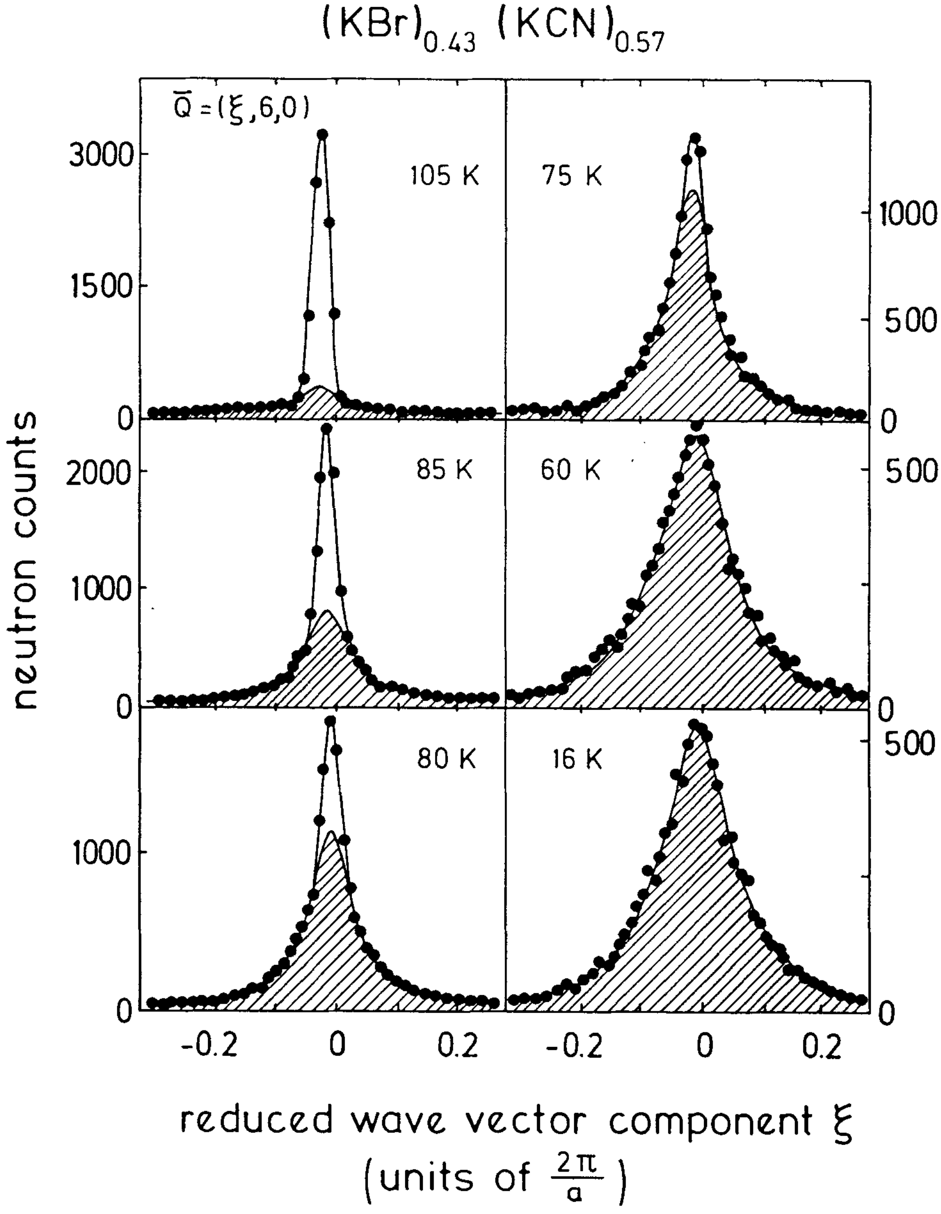

Figure 7 Transverse scans at zero energy transfer through the (060) reflection in $(KBr)_{0.43}(KCN)_{0.57}$. From Loidl *et al.* (1988).

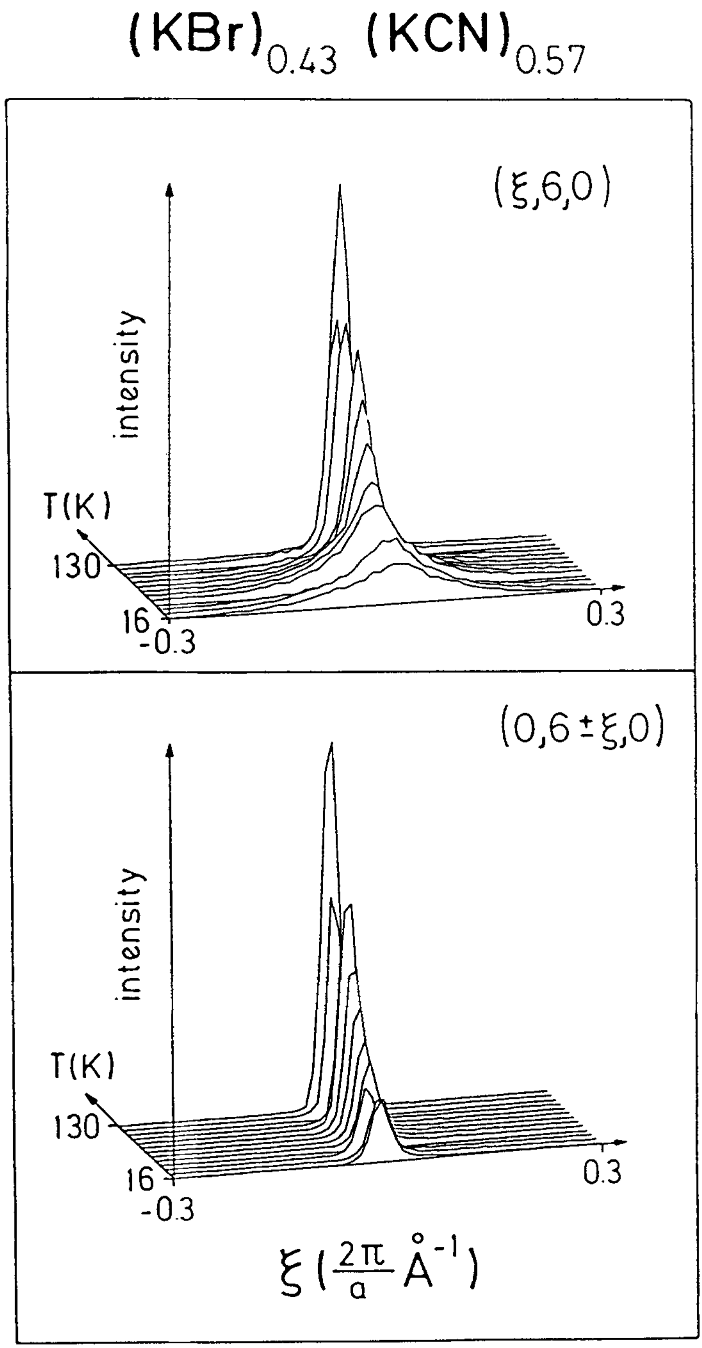

Figure 8 Three dimensional plots of the observed line shapes in $(KBr)_{0.40}(KCN)_{0.67}$. The intensities are shown vs. the reduced wave vector component ζ for series of temperatures between 16 K and 130 K. From Loidl *et al.* (1989).

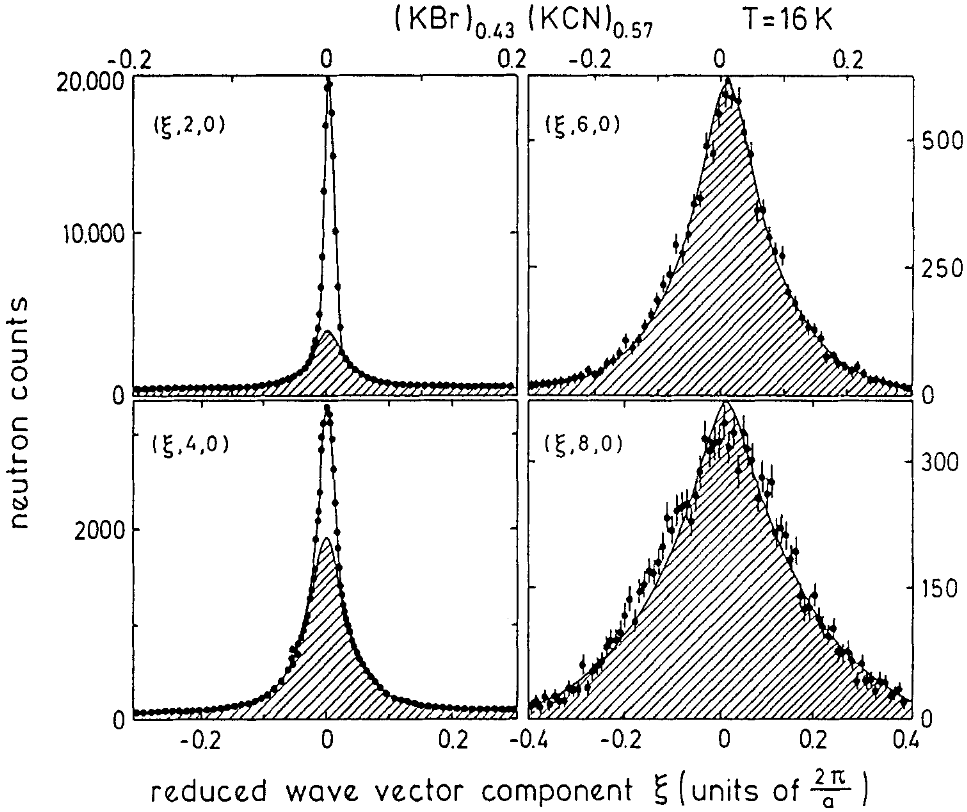

Figure 9 Transverse scans through a series of (0k0) reflections in $(KBr)_{0.43}(KCN)_{0.57}$ at 16 K. From Loidl *et al.* (1988).

Below 70 K the Bragg-like component has vanished completely and the local shear distortions become frozen-in.

The three-dimensional plots of line shapes of transverse and longitudinal scans through the (060) reflection in Figure 8 elucidate the idea that the glass state can be characterized by a supercooled two dimensional liquid. The upper part shows the variation of the scattered intensity in transverse scans and gives a clear impression of the break-down of true long-range order due to shear distortions. Despite the absence of Bragg-like intensities in the transverse scans the longitudinal scans which test the lattice constant are still resolution limited and point towards long correlation lengths.

Figure 9 illustrates the Q-dependence of Bragg scattering and anomalous diffuse scattering at 16 K. Here transverse scans through a series of (0*k*0) reflections are shown. True Bragg scattering is visible only for $h \leq 4$. The increasing half width of the diffuse component can readily be seen. To demonstrate quantitatively that the line shape of the diffuse component has an exponential form with a width that

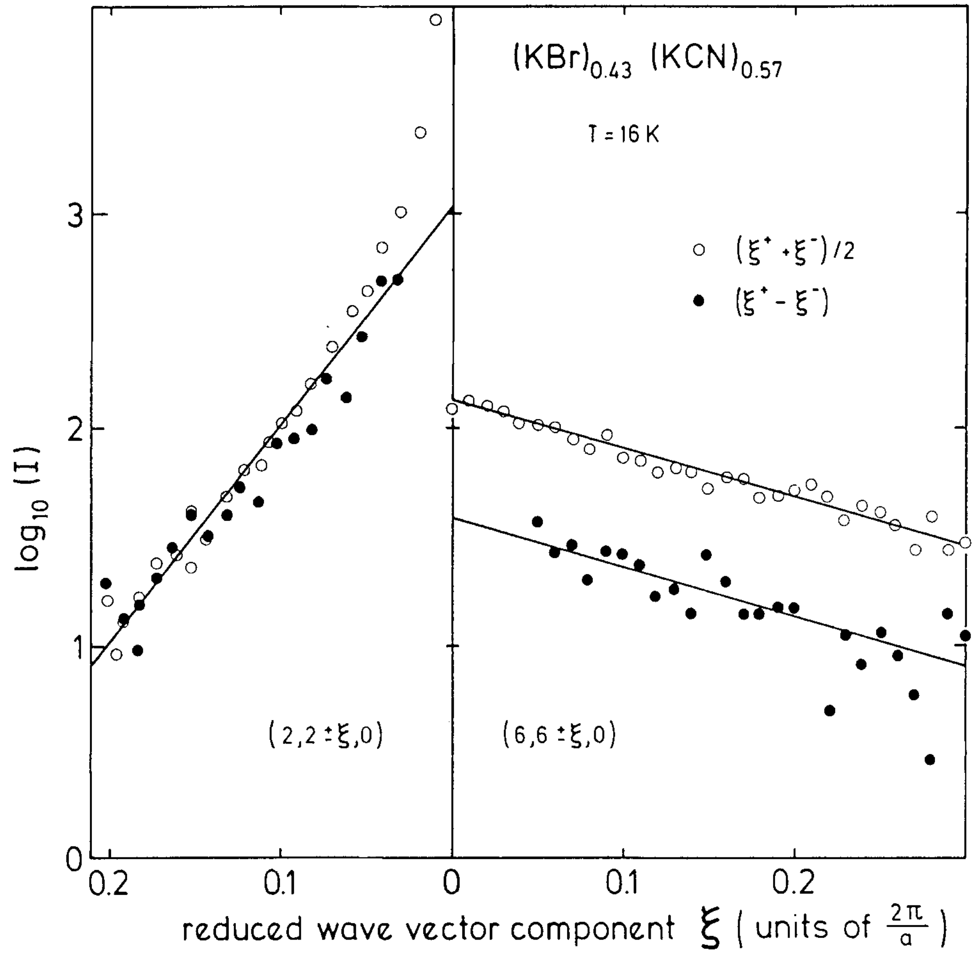

Figure 10 Symmetrized and antisymmetrized intensities of transverse scans through the (220) and (660) reciprocal lattice points in $(KBr)_{0.43}(KCN)_{0.57}$. From Loidl *et al.* (1988).

increases with increasing momentum transfer Q, Figure 10 shows the experimentally observed intensities at the (220) and (660) reciprocal lattice points in a semilogarithmic plot. To get an estimate of the characteristic asymmetries of the diffuse components (see Figure 6) we plotted symmetrized and antisymmetrized data separately. Figure 10 reveals that the line shape is exponential and the half width is strongly Q-dependent. At a given momentum transfer Q symmetrized and antisymmetrized data behave very similar. However, for the higher nodes of the (hh0) reflections the antisymmetric component is weaker almost by half an order of magnitude.

Loidl *et al.* (1988) determined the Debye–Waller factor also for the $x = 0.57$ sample. The resulting mean square displacements are shown in Figure 5 together with the results from the 65% crystal. For $x = 0.57$ the mean square displacements exhibit a gradual increase with decreasing temperatures. Saturation effects become dominant below 60 K.

5 DISCUSSION AND CONCLUSIONS

What are the conclusions that can be drawn from these elastic single crystal neutron diffraction studies in $(KBr)_{1-x}(KCN)_x$? For $x \rightarrow x_c$ the structural phase transitions become almost second order with a soft elastic shear constant c_{44}. In this concentration range the mixed crystals undergo a planar ferroelastic instability. At T_s the

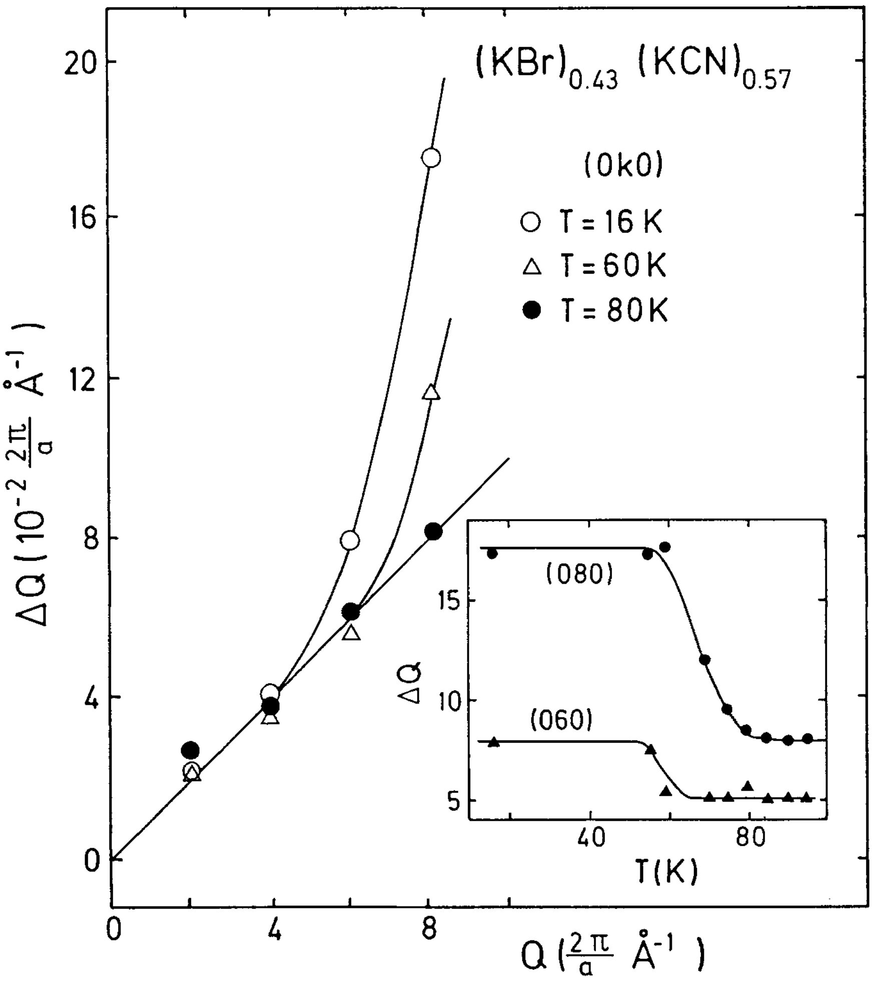

Figure 11 Half width of half maximum ΔQ of the diffuse scattered intensities vs. momentum transfer at different temperatures as measured in $(KBr)_{0.43}(KCN)_{0.57}$. The inset shows the temperature dependence of ΔQ for the (060) and the (080) reflections. From Loidl *et al.* (1988).

Bragg-like intensities are replaced by anomalous diffuse intensities which follow an exponential decay. The half widths strongly depend on the momentum transfer Q. Theoretically, power law singularities were predicted by Mayer and Cowley (1988) where the exponent depends on T and Q. For concentrations $x < x_c$ the same pattern of intensities is observed. However, this pattern is frozen-in at low T. We believe that the glass-like state just below the critical concentration can be described in terms of a frozen-in two dimensional liquid. And indeed, the diffuse patterns at the reciprocal lattice points exhibit a characteristic temperature and Q dependence which qualitatively supports this idea: Figure 11 shows the half width at half maximum of the diffuse intensities as observed at the (060) reciprocal lattice points as function of lattice vector and temperature. The half width ΔQ shows an increase which is much stronger than linear in Q. As can be seen from the inset of Figure 11 a significant increase of ΔQ appears with decreasing temperatures, with saturation effects appearing below 60 K.

For concentrations $x < 0.5$ the cubic symmetry of the center of mass lattice is preserved and the diffuse intensities can be explained in terms of Huang scattering which results from the presence of elastic defects. This concentration range has been intensively investigated by Wochner (1989) and Wochner, Burkel, Peisl, Zeyen and Petry (1988).

Guided by these observations it seems plausible to conclude that in $(KBr)_{1-x}(KCN)_x$ orientational order is established for $x > x_c$ at low T. The onset of

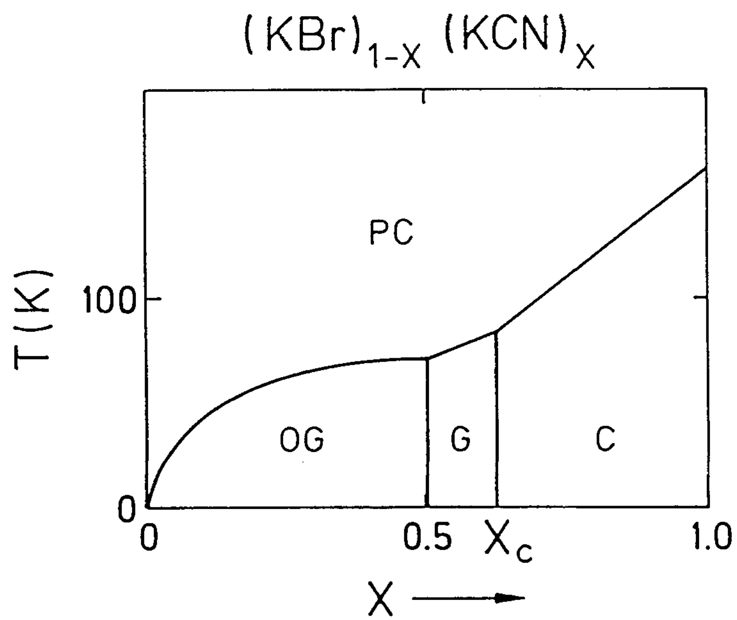

Figure 12 Schematic x–T phase diagram of $(KBr)_{1-x}(KCN)_x$: plastic crystal (PC), crystal (C), glass (G) and orientational glass (OG)

long-range orientational order is accompanied by long-range homogeneous shear distortions of the center of mass lattice. For $x \geq x_c$ the ordered low-temperature phase develops out of a two dimensional melt, which appears at the transition temperature. At T_s the Debye–Waller factors seem to diverge and no true Bragg-scattering can be detected. For $x \leq x_c$ the two dimensional liquid becomes supercooled and finally, at low T, frozen-in. This region of the phase diagram is a "true" glass state characterized by the absence of both orientational and translational long-range order. For further decreasing concentrations an orientational glass state appears. In the orientational glass state the reorienting moments (quadrupoles) undergo a collective freezing process devoid of long range order. The translational order is not affected. This is due to the fact that, like in spin glasses, at the glass transition the time scales for reorientations and diffusion processes are fully decoupled. The resulting phase diagram is shown in Figure 12. This phase diagram shows striking similarities with the phase diagram of KBr:KCN as proposed by Galam (1990) within the framework of a random anharmonic compressible ferroelastic model.

An alternative approach has been developed by Michel (1987) and by Bostoen and Michel (1988). For $x \leq x_c$ they predicted a "non-ergodic instability". A "non-ergodic instability" shows up when the system is close to a second order phase transition and, as T_s is approached, critical fluctuations are frozen-in by weak random fields. However, contrary to the experimental observations the diffuse patterns should be independent of the reciprocal lattice point.

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