Orientational Glass Behaviour of $KBr_{0.96}(CN)_{0.04}$[*]

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Ultrasonic measurements on the mixed crystal $KBr_{0.96}(CN)_{0.04}$ show a minimum of most of the elastic constants at 16 K. In addition we determined $c_{11}(T)$ at 10 MHz and at 50 MHz observing dispersion effects. These results and previous Brillouin and neutron scattering results of other authors on higher CN-concentrations are interpreted by an orientational glass behaviour. A semiquantitative description is given in terms of the mean random field approximation.

I. Introduction

KCN and the related pseudobinary compounds $KBr_{1-x}(CN)_x$ are model systems of molecular crystals because of the high symmetry of the lattice (NaCl-structure) and the simple dumb-bell shape of the CN-molecule. A large amount of work has been spent on the structure and lattice dynamics. At 168 K pure KCN undergoes a transition from the cubic to an orthorhombic low temperature phase. This transition is announced by a strong softening of the shear elastic constant c_{44} [1]. The temperature T_s where the extrapolated elastic constant would vanish is slightly lower, namely 151 K [2]. The partial substitution of CN by halide ions leads to a reduction of T_s linear with x as shown by ultrasonic measurements for $0.96 \leq x \leq 1$ [2] and Brillouin experiments for $x = 0.75$ [3]. At even lower concentrations the crystal remains pseudocubic (disordered with respect to the CN^- orientations) down to the lowest temperatures. Brillouin data for $x = 0.56$, 0.34 and 0.19 [3], neutron inelastic scattering results at $x = 0.5$ [4], and our own preliminary ultrasonic measurements at $x = 0.04$ and 0.14 [5] suggest that the softening of c_{44} is incomplete; c_{44} rather shows a minimum at a characteristic temperature T_F. The concentration dependence of T_s resp. T_F is shown in Fig. 1.
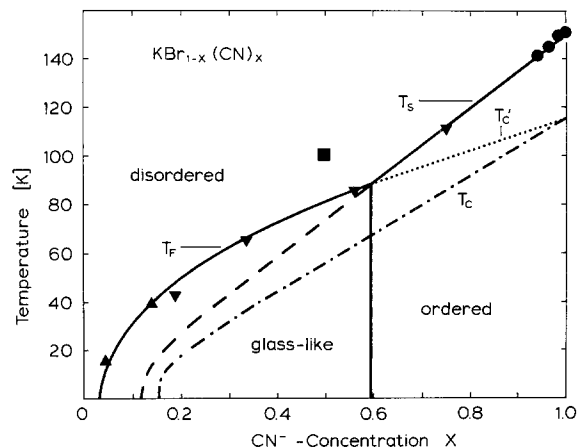
Fig. 1. The dependence of the characteristic temperatures T_s, T_F, T_c, T_c' (for definitions see text) on the concentration x. The symbols indicate experimental results: ● ultrasonic results from [2] (in this case pure KCN was diluted with Cl^- ions); ▼ Brillouin data from [3]; ■ neutron results from [4]; ▲ ultrasonic data from [5] and the present work. The lines present the theoretical results. (Below $x = 0.59$ T_c' runs very close to T_F and is not plotted in the phase diagramm)

In the following we will complement the results by detailed ultrasonic data for $x = 0.04$, by presenting results on the elastic constants and attentuations for all main symmetry modes and give a semiquantitative explanation in terms of a transition into a

pseudo-spin-glass phase. A quadrupolar glass phase was found to exist in the solid ortho-para-hydrogen mixtures [6] up to concentrations as high as 55 % of ortho hydrogen. Glass properties for CN⁻ defects in alkali halides were predicted by Fischer and Klein [7] and have also been suggested by Rowe et al. [4] to explain the neutron results in $KBr_{0.5}(CN)_{0.5}$. In a recent paper these experimental results have been qualitatively explained by an extension of the earlier work of Michel and coworkers [8] including an orientational glass state parameter in the frustrated phase [9]. In our calculations we use the model of a singlet ground state magnet [10] combined with the ideas of the mean random field approximation (MRFA) developed by Klein and coworkers [11]. With one free parameter these model calculations explain the general features of the experimental data over the entire concentration range, namely $T_s(x)$ and $T_F(x)$, the temperature dependence of the velocity and the damping of sound waves, and the onset of elastic intensity at T_F around reciprocal lattice points, which are related to the temperature dependence of the order parameter of the glass state.

II. Ultrasonic Experiments and Results

The ultrasonic experiments were carried out on single crystals of $KBr_{0.96}(CN)_{0.04}$ which were grown by F. Rosenberger in the Crystal Growth Laboratory of the University of Utah from zone refined KBr and KCN materials. The CN⁻ concentration was determined by chemical analysis. The lattice constant at room temperature, as measured by neutron diffraction, was 6.59 Å [5]. [100] and [110] surfaces were obtained by cleaving resp. cutting and subsequent polishing. The parallelity of opposed faces was better than 0.05 degrees. The thickness of the slabs was about 4 mm. The 10 MHz quartz transducers, X-cut for longitudinal and AC-cut for transverse excitation, were attached to the faces with hydrocarbon bonds at higher temperatures and solidified gases (N_2, C_2H_6, CF_2Cl_2) at lower temperatures. The sound velocities were determined using the pulse echo overlap method; the sound attenuation was obtained from the intensity ratio of the first and second echo. Some temperature scans were performed with an overtone of 50 MHz. The absolute values of the elastic constants are accurate to about 4 %, whereas the relative error was about 10^{-4}.

Figure 2 shows the results of the measurements at 10 MHz. The elastic constants except c_{44} show a minimum at about 16 K. The attenuation exhibits a maximum which is surprisingly at a slightly lower temperature. The signal of the c_{44} mode (T_{2g}-
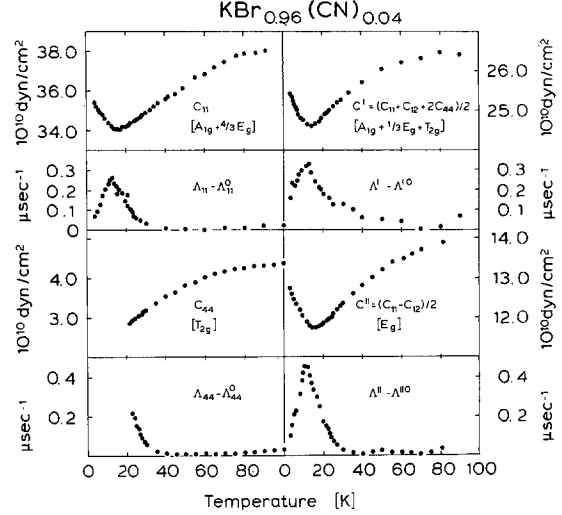

Fig. 2. The temperature dependence of the elastic constants and the sound wave attenuation in $KBr_{0.96}(CN)_{0.04}$. The attenuation is presented as a difference of the actual value minus the background value attained at about 60 K

symmetry) was lost at 22 K due to large damping but presumably c_{44} has the same qualitative temperature dependence as the other modes [5]. A radically different behaviour of c_{44} would have shown up in the mode $c' = (c_{11} + c_{12} + 2c_{44})/2$. When the bulk module ($A_{1g}$-symmetry) is derived from c_{11} and $(c_{11} - c_{12})/2$ (E_g-symmetry) one notes that it is practically temperature independent and does not show the characteristic minimum at 16 K. Formulated differently: the temperature dependence of c_{11} is entirely due to the E_g-contribution and the temperature dependence of c' is due to the combined E_g and T_{2g} contributions. Focusing on the elastic constants with the representations A_{1g}, E_g and T_{2g} one notes that the T_{2g}-mode c_{44} shows the strongest temperature effect. The relative changes of these three main symmetry elastic constants between 100 K and 20 K are about 0, 15 and 35 %, respectively.

The longitudinal sound waves in the [100]-direction have been measured additionally with the 50 MHz overtone. The comparison between the 10 MHz and the 50 MHz results is shown in Fig. 3. The insert gives the difference between the high and low frequency elastic constants where we approximated the data by smooth curves. The relative error between these measurements with different harmonics using an otherwise identical set up is about $3 \cdot 10^{-3}$. It is due to an uncertainty of one soundwave period in the overlap pattern. Around the freezing temperature the effect of dispersion is in the order of 0.5 %. As both minima are relatively broad the present data can only set an upper limit for a possible shift of the freezing temperature, namely 0.5 K or so.
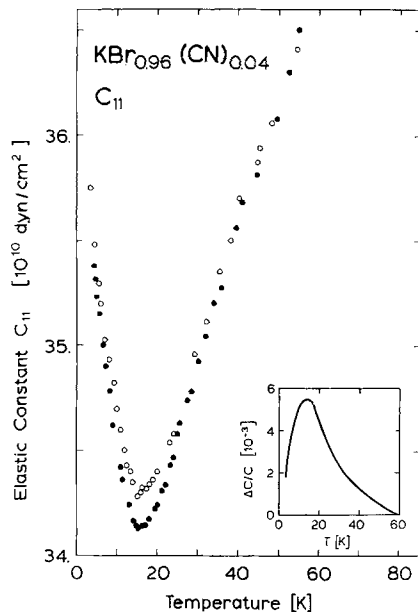
Fig. 3. The temperature dependence of the elastic constant c_{11} determined frequency of 10 MHz (●) and of 50 MHz (○). The insert shows the fractional difference $[C_{11}^{50}(T) - C_{11}^{10}(T)]/C_{11}^{10}(T)$ as a function of temperature

Summarizing the experimental facts, the temperature dependence of the elastic constants with the representations E_g and T_{2g} exhibit distinct minima at the same temperature. This coincidence would be purely accidental in terms of the three level scheme of [5]. The bulk modul is totally decoupled from the rotational excitations. One definitively knows that the center of mass lattice is stable down to the lowest temperatures as can be seen from the stable fcc-Bragg peaks in the elastic neutron scattering experiments and also from Brillouin measurements, where only crystals with concentrations $x \leq 0.56$ became opaque at 10 K on cooling, indicating the onset of a multi-domain phase transition [14]. Therefore the most reasonable explanation of the temperature dependence of the elastic constants is the onset of an orientational glass phase at T_F. This also explains the short range order elastic intensity around the Bragg points for $T \lesssim T_F$ [4]. The frequency dependence of the minima in the sound velocities reminds of the frequency dependent cusp in the susceptibilities of some real spin glasses.

For a quantitative explanation of the experimental results we follow the MRF approximation. Although this theory is known to be in poor shape [19] we use this approach, as it is easy to handle and we achieve a rough agreement with all experimental data available.

III. Theoretical Background

Molecular crystals are in some respect analogous to magnetic system like crystal field split $4f$-states in rare earth systems. In the case of CN-dumb-bells embedded in a host crystal these crystal field states have to be identified with the eigenstates of the CN-molecules, which are described by a single ion Hamiltonian consisting of a free rotator term in a hindering potential of Devonshire-type [12]. The coupling between the CN^--ions is regarded as an effective interaction between electric quadrupoles which may be either direct or indirect, e.g. mediated via lattice strains. The system of the CN-molecules will be disordered or paraelastic at high temperures and orientationally ordered below a 'Curie'-temperature T_C. However, in real crystals this transition is clamped by the structural transition temperature T_S. For concentrations x smaller than a critical concentration one expects an elastic-quadrupol glass below a temperature T_F, which is characterized by random frozen-in orientation of the molecules with no long range order left. This glass phase probably occurs as a consequence of the anisotropic nature of the effective multipole-multipole coupling between CN-pairs, into which not only the orientations of the multipoles, but also the bond vector enters. In the magnetic analog this situation is believed to exist in the spin glasses, where the frustration occurs because the Rudermann-Kittel interaction changes sign as a function of distance. Via quadrupolar coupling the effects of the molecular system are carried over onto the phonon modes.

In the calculations we use a two-level system which we treat in the concentrated KCN crystal in the framework of the RPA-MFA and in the glass state of the mixed system in the MRF-approximation, in which the strain on the quadrupoles is replaced by a random external strain field which may have either a Gaussian or Lorentzian probability distribution [11]. We are aware that the MFA and in particular the MRFA are rather crude approximations. Furthermore, the exact nature of the effective interaction between the CN-molecules is not known, which hampers any theoretical treatment. Thus we regard the following considerations as tentative and illustrative. The experimental facts establish the existance of the quadrupolar glass phase without relying on any theoretical evaluation.

The single CN-molecule in a cubic Devonshire potential is described by the dynamic susceptibility $u(\omega)$. We rigorously approximate the complex scheme of the molecular states [12] by a system of two levels $|0\rangle$ and $|1\rangle$ with an energy separation Δ and a transition matrix element $\alpha = \langle 1|\tilde{O}_\Gamma|0\rangle$ where

$\tilde{O}_\Gamma$ is an operator which transforms according to an irreducible representation of the symmetry group O_h. Since we are finally interested in the temperature dependence of c_{44} and $(c_{11}-c_{12})/2$ we specify $\tilde{O}_\Gamma$ as E_g or T_{2g} quadrupole operators. Of course α, Δ and all derived quantities might be different for the two representations. The susceptibility is of Van Vleck type [13]

$$u(\omega) = \frac{2\alpha^2\Delta}{\Delta^2 - \omega^2 + i\gamma\omega}\tanh\left(\frac{\hbar\Delta}{2k_BT}\right) \qquad (1)$$

$\gamma\omega$ is the width of the molecular excitation. The resonances have in fact been observed in alcali cyanides by optical spectroscopy [14] and by inelastic neutron scattering, in the latter case by their anti-crossing with the phonon branches [5, 15, 16]. Introducing an effective coupling J between the CN-molecules, which is analogous to the exchange coupling in magnetic systems the full wave vector dependent susceptibility of the molecular system is obtained as [13]

$$\chi(\mathbf{q},\omega) = \frac{u(\omega)}{1 - J(\mathbf{q})u(\omega)} \qquad (2)$$

This effective coupling has to be regard as an interaction between the CN^--electric quadrupoles, which may either the direct or indirect, e.g. mediated by lattice strains.

The pole of χ determines the dispersion relation $\omega_R(\mathbf{q})$ of the collective molecular excitations. The molecular system orders at the temperature T_c with the ordering wave vector $\mathbf{q}_0$ where the static susceptibility $\chi(\mathbf{q}_0,0)$ diverges and the excitation energy $\omega_R(\mathbf{q}_0)$ vanishes. T_c can be derived from

$$\tanh\left(\frac{\hbar\Delta}{2k_BT_c}\right) = \frac{\Delta}{2\alpha^2 J(\mathbf{q}_0)} \qquad (3)$$

The results in KCN indicate that the ordering wave vector $|\mathbf{q}_0| = 0$ [17], i.e. the molecular excitation softens at the zone center.

In the ordered phase the CN-molecules experience an additional mean field ε, a quadrupolar stress, which mixes the two states $|0\rangle$ and $|1\rangle$ of the disordered phase via the mixing angle Θ and induces a spontaneous quadrupole moment Q which has to be determined selfconsistently from the mean field equation [10]

$$Q = \alpha$$
$$\cdot \sin\Theta\tanh\left\{\frac{\hbar\Delta}{2k_BT}\cdot\left[\cos\Theta + Q\frac{J(0)\alpha}{\Delta}\sin\Theta\right]\right\} \qquad (4)$$

The susceptibility and the dispersion in the ordered phase can still be calculated from Eq. (2) when the correct temperature dependent values for the level splitting and the matrix elements Δ' and α' are used, where $\Delta' = \Delta/\cos\Theta$, $\alpha' = \alpha\cos\Theta$. The phononlike excitations of the coupled system of internal and external modes are obtained by inserting the molecular susceptibility weighted by the coupling coefficient g^2 into the phonon propagator. Considering an acoustic branch in the long wavelength limit, the elastic constant is given by

$$c + iA = c_0\{1 - g^2\cdot[\chi'(0,0) + i\chi''(0,\omega)]\}. \qquad (5)$$

The background elastic constant c_0 describes the decoupled phonon system. The static susceptibility $\chi'(0,0)$ is the real part of the full susceptibility in the limits $q\to 0$ and $\omega\to 0$. $\chi''(0,\omega)$ is the imaginary part with $q\to 0$. The elastic constant c vanishes at a temperature T_s with

$$\tanh\left(\frac{\hbar\Delta}{2k_BT_s}\right) = \frac{\Delta}{2\alpha^2(J(0)+g^2)}. \qquad (6)$$

The sound attenuation A is determined by the imaginary part of (5). The treatment so far is valid for the concentrated system KCN where all CN-sites are identical. In the mixed crystals every CN-molecule has its own environment, the effective coupling to the other molecules and the resulting field ε varies through the crystal. In the MRF-approximation the fields are given by a probability distribution centered around zero field which in the glass phase is of Gaussian shape at low temperatures, then changes to Lorentzian shape at higher temperatures and finally to a δ-peak in the paraelastic phase. The characteristic parameter of the distribution is the mean magnitude of the field which is proportional to the concentration x for the Lorentzian and proportional to $\sqrt{x}$ for the Gaussian case [7, 11]. The same dependencies hold for the exchange constant $J(0)$ [18], namely $J = x\cdot J_0(0)$ resp. $J' = \sqrt{x}\cdot J_0(0)$ where $J_0(0)$ is the $CN-CN$ coupling in pure KCN at the ordering wave vector $\mathbf{q}_0 = 0$. In addition we postulate a linear concentration dependence for the coupling constant, $g^2 = xg_0^2$, for the mixed crystals. The molecular system for $x < 1$ has now two ordering temperatures T_c and T_c' with $T_c < T_c'$ which are given from (4) with J and J' respectively. For $T_c < T < T_c'$ there is a competition between the disordered and the ordered phase. The stable state is determined by the smaller static susceptibility $\chi'(0,0)$ or, equivalently, by the higher energy of the collective excitation $\omega(\mathbf{q}_0)$. This condition leads to the characteristic cusp in the temperature dependence of
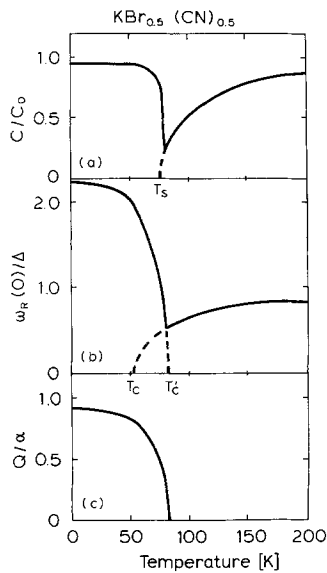
Fig. 4. The calculated temperature variation of the elastic constant normalized to the background value, of the energy of the collective molecular excitation and of the order parameter of the glass phase for $x=0.5$. All quantities are of T_{2g}-symmetry

$\chi'(0,0)$, $\omega(\mathbf{q}_0)$ and c which defines the freezing temperature T_F. Figure 4 shows the results of the temperature dependence of the elastic constant c (4a), of the soft rotational excitation $\omega_R(\mathbf{q}_0=0)$ (4b) and of the quadrupolar moment for $KBr_{0.5}(CN)_{0.5}$ (4c) within the MRF approximation. The latter quantity is not the spontaneous quadrupolar moment, but rather the configurational average of the squared moment, which is the new order parameter of the glass phase. The important features are that neither the rotational excitation, nor the elastic constant show a complete softening. At the freezing temperature they exhibit a minimum and recover in the elastic quadrupolar glass state.

IV. Comparison with the Experiment and Conclusion

The simple model presented has three parameters: $J_0(0)$, g_0^2 and the splitting of the two molecular states in the disordered phase. The matrix element α^2 can be set to unity since it appears only in combination with $J_0(0)$ and g_0^2. We first concentrate on T_{2g} quadrupolar properties since the elastic constant of this symmetry shows the strongest temperature dependence in KCN and the mixed crystals. From Brillouin data rotational transitions in T_{2g} symmetry were determined to increase from 10 K at the dilute limit reaching approximately 60 K at $x=0.3$ and remain constant to the highest concentrations [14].

Neutron scattering experiments revealed 3 K for the very dilute region [16] and 70 K for $x=1$ [17]. Though there are some doubts whether this mode is exclusively responsible for the temperature dependence of c_{44}-excitations with lower energy transfer might be involved, too – we set the splitting Δ to 35 K independent of x. Note that the actual value of Δ is of little importance as long as $k_B T \gg \hbar \Delta$. T_s of pure KCN was determined to be 151 K, which leads according to (8) to the relation $\alpha^2(g_0^2 + J_0(0)) = 151\,K$. Thus there is effectively one free parameter, say $J_0(0)$ or, what is equivalent, T_{c0}, the temperature where the CN-molecules – if they were decoupled from the lattice strains – would show spontaneous T_{2g} quadrupolar order in pure KCN. In order to describe the observed T_s and T_F we adjusted T_{c0} to 117 K; the theoretical dependence of the characteristic temperatures T_s, T_c, $T_{c'}$ and T_F on concentration are then obtained as shown in Fig. 1.

For $0.59 \leq x \leq 1$ the mixed crystals undergo a structural phase transition at a temperature T_s due to the softening of the elastic constant c_{44}. For $x<0.59$ a transition from the disordered phase into a spin glass phase takes place at the freezing temperature T_F. From Brillouin data this critical concentration was determined to be 0.56 [14]. The predictions of this model with one free parameter are in good agreement with the experimental result. In Fig. 4 the temperature dependence of the elastic constant, of the energy of the molecular excitation and of the order parameter of the glass state is calculated with the same parameters for $KBr_{0.5}(CN)_{0.5}$. We note that the last quantity should be directly related to the diffuse elastic intensity centered around the Bragg-peaks which was observed in a neutron scattering experiment on a sample of this concentration [4]; the measured softening of the TA phonon can be related to Fig. 4a. Figures 5 and 6 present the temperature dependence of the elastic constant and the attenuation of the mode which should be compared to the corresponding ultrasonic, Brillouin and inelastic neutron scattering data on c_{44}. The general features of the experiments are well reproduced by the calculation. Some discrepancies can be observed a) in the rise of c_{44} just below T_F which is smoother in the experiment and b) in the absolute variation of c_{44} at low concentrations. For $x=0.04$ e.g. we have observed a 35% reduction of c_{44} between 100 K and T_F whereas the model predicts only 15%. Presumably the proportionality of the coupling constant g^2 and x is not warranted at these low concentrations. The shortcoming a) indicates that either the level scheme assumed is too simple and should be replaced by a more general one – we remind: the mean field spontaneous magnetisation saturates less
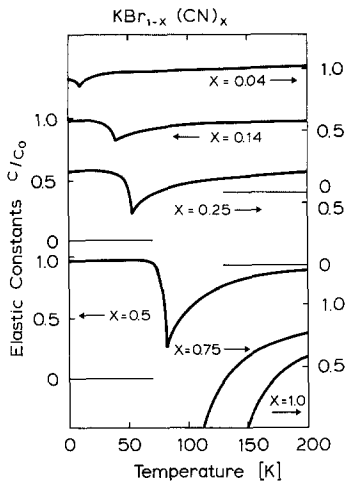
Fig. 5. The calculated temperature dependence of the normalized T_{2g}-elastic constant for various concentrations x
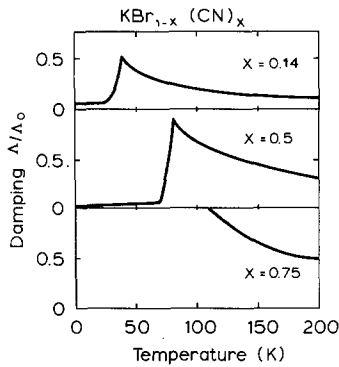

Fig. 6. The calculated temperature dependence of the ultrasonic (T_{2g}) attenuation for three concentrations x

rapidly for $S = \infty$ than for $S = 1/2$; or that the mean random field treatment is at all a poor approximation as in real spin glasses [19]. Nevertheless we think that the overall agreement between the experiment and the present model is a good description of the glass phase in $KBr_{1-x}(CN)_x$.

The ordering resp. freezing is governed by T_{2g}-modes. The E_g-modes seem to play a minor role. In the disordered phase the T_{2g}- and E_g-modes are decoupled. Thus the elastic constant $(c_{11} - c_{12})/2$ is described by an independent set of parameters. For pure KCN one knows from the ultrasonic data [1] that $T_s(E_g)$ is about 140 K which is lower than $T_s(T_{2g})$. In the present low concentration results the temperature variation of $(c_{11} - c_{12})/2$ is again smaller than that of c_{44}. We conclude that the ordering is driven by the T_{2g} rather than by the E_g quadrupolar excitations. In the ordered phase the two symmetry modes are coupled, since any T_{2g}-quadrupolar

field induces not only a T_{2g}- but also a E_g-quadrupole moment. This fact explains the experimental observation that the freezing temperatures of the different modes are identical as can be seen from Fig. 2.

We have previously given an alternative explanation for the occurrence of a minimum in the elastic constants where we postulated that the scheme of molecular states can be approximated by three levels. Of course, these assumptions yield the minima mentioned but it needs two very special sets of parameters for the minima of the T_{2g} and the E_g-mode to occur at the same temperature. The coincidence would be purely accidental whereas in the spin glass concept it emerges quite naturally.

Using (5) the dispersion effect near T_F of Fig. 3 results from a frequency dependent susceptibility in the MHz range. Looking at real spin systems we find contradictory results. In the $La_{1-x}Gd_xAl_2$ system one finds increasing freezing temperatures and smoother cusps of AC susceptibilities for higher frequencies (7.2 Hz to 1,142 Hz) [20], an observation which favours a model of a metastable glass state. In clear discrepancy to this work Dahlberg et al. [21] reported practically unchanged AC susceptibilities from 16 Hz up to 2.8 MHz. Recently these contradicting observations have been explained by a qualitative picture of Murani [22] in terms of a wide range of relaxation times. In the framework of the mean random field approximation different frequencies yield different exchange constants and result in shifted ordering temperatures T_C and T_C'. But especially around T_F this approach is probably inadequate and we desist from a quantitative explanation of these results.

In conclusion we recapitulate the experimental results which support the existence of a glass phase in $KBr_{1-x}(CN)_x$ for $x < 0.6$:

a) the incomplete softening of c_{44} which leads to a minimum at a temperature T_F

b) the fact that the elastic constant $(c_{11} - c_{12})/2$ shows a minimum at the same temperature

c) the existence of diffuse elastically scattered intensity around Bragg-points in the neutron scattering experiments

d) the frequency dependence of the elastic constants at T_F. Apart from $d)$, these features have been semi-quantitatively explained in this article.

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