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Brillouin-scattering study of the orientational glass transition in $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ mixed crystals

J. F. Berret, A. Farkadi, M. Boissier, and J. Pelous
Laboratoire de Sciences des Matériaux Vitreux, Université des Sciences et Techniques du Languedoc,
F-34060 Montpellier, France

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Using Brillouin-scattering experiments, we have measured the sound velocity and the inverse mean free path of longitudinal phonons in the $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ mixed crystals between 10 and 300 K. In these mixtures the incomplete softening of the longitudinal and transverse elastic constants have been analyzed with use of an orientational glass model based on the existence of a quenched random strain field. Static strains are responsible for the suppression of the ferroelastic transition below x_c. In support of the theoretical predictions based on the origin of random strains, our results confirm that the field is much larger in $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ than in $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$.

I. INTRODUCTION

The mixed alkali-halide–alkali-cyanide $(MX)_{1-x}$-$(M\mathrm{CN})_x$ crystals are one of the most attractive model systems for structural glasses. These compounds, where M is an alkali-metal ion (M=Na, K, or Rb) and X is a halogen ion, have been experimentally and theoretically investigated during the last several years in two aspects: First, the disorder-order transitions between the freely rotating and the frozen-in regimes of the cyanide dynamics,[1] and secondly, the low-temperature orientationally disordered phase. Concerning the latter point, it is well established that the thermal, acoustic, and dielectric properties below 1 K are similar to those of structural disordered solids.[2]

In this paper we are concerned with the former aspect of the model system, namely the transitional phenomena towards the orientational glass state in mixed crystals $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$. Although less studied than the well-known $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$, the KCl:KCN system exhibits a comparably rich (x,T) phase diagram.[3] Above a critical concentration $x_c \simeq 0.80$, the mixtures in a cubic phase at room temperature undergo structural (first-order) phase transitions at T_s to monoclinic or orthorhombic structures. These structural changes result in a bilinear translation-rotation (T-R) coupling between orientational degrees of freedom of the CN molecules and the translational T_{2g} modes (thermal phonons).[4] As a consequence of the T-R interactions, striking features are observed, such as a soft mode in the shear elastic constant c_{44}.[5] The $x>x_c$ crystalline structure is characterized by a long-range order of the CN orientations at low temperature.

A substitution of the dumbbell-shaped molecules by spherical Cl ions strongly lowers the transition temperature. Below x_c, pure structural changes are replaced by a gradual freezing of the CN dynamics. The slowing down of the cyanide reorientations yields a low-temperature state where the molecular motions are frozen out and the CN orientations randomly distributed. As the orientational glass phase establishes in the MX:MCN compounds, the elastic constants associated with the transverse- (TA) and the longitudinal- (LA) acoustic modes exhibit a minimum at $T_F(x,\omega)$, which shifts to lower temperature when both the frequency $\nu=\omega/2\pi$ and/or the concentration are decreased.

Recent low-frequency elastic response experiments performed on $(\mathrm{KBR})_{1-x}(\mathrm{KCN})_x$ crystals[6] provided a better understanding of the transition between crystalline and glasslike behaviors, and of the dynamics of the molecular motions in mixed cyanides. Following Volkmann *et al.*,[6] the orientational glass transition can be regarded as a pure relaxation mechanism. For crystals in the middle range of the phase diagram, it has been shown that two consecutive relaxation processes occur upon cooling. The primary relaxation is sensitive to acoustic measurements only, and thus related to the above defined freezing temperature T_F, whereas the secondary process, also named "β relaxation" in reference to amorphous polymers is detected in both acoustic and dielectric susceptibilities. More recently, the primary and secondary relaxations have been interpreted in terms of collective freezing and individual slowing down of the molecular reorientations, respectively.[7]

In the present paper we report Brillouin-scattering experiments on the KCl:KCN monocrystals, i.e., on the real and imaginary parts of the orientational susceptibility at gigahertz frequencies. Since both relaxations described above coincide at these frequencies,[7] we are essentially concerned here with a unique relaxation process. We have measured the longitudinal elastic constant $c_{11}(x,T)$ and the corresponding phonon mean free path $l_{11}(x,T)$ for the concentrations x=0.005, 0.05, 0.20, 0.30, 0.40, and 0.96, and for temperatures ranging from 10 to 300 K. Moreover, we have tested previously published KCl:KCN c_{44} data[8] with a theory recently proposed by Michel.[9] To account for the incomplete softening of the TA modes in cyanide crystals below x_c, Michel developed a theory of the orientational glass state, based on the existence of quenched random strain fields. According to this model, which deals with the so-called primary relaxation only, the strain field in the MX:MCN systems is due to the substitutional halogen ions and the suppression of the ferroelastic transitions for $x < x_c$ results from the competition between the *T-R* and the strain-rotation couplings. Thus, using Michel's predictions, we are able to fit the $(KCl)_{1-x}(KCN)_x$ c_{44} data down to $T \simeq 50-70$ K, i.e., well below the extremum of the acoustic susceptibility. Compared to a recent paper by Fossum and Garland,[10] who applied ultrasonic methods, we confirm that the amplitude of the random strain field is higher in chloride than in bromide cyanides, thus corroborating the basic assumption that the mean field originates from the volume mismatch between *X* and CN ions.

II. EXPERIMENTAL RESULTS

Single crystals of KCl:KCN investigated here have been obtained from Professor H. Ahrend, at Eidgenössische Technische Hochschule–Zürich. The temperature dependence of the longitudinal elastic and viscoelastic constants have been measured using a Brillouin spectrometer in the backscattering configuration. The experimental setup is described in Ref. 11.

Since the dumbbell-shaped molecules are assumed to point preferentially in the eight $\langle 111 \rangle$ equivalent directions, acoustic waves propagating along the $\langle 100 \rangle$ crystalline directions at sound velocity $v_l = (c_{11}/\rho)^{1/2}$, where ρ is the mass density,[12] interact most strongly with molecular excitations of E_g symmetry. This corresponds to a coupling of the LA waves with the librational modes of the CN molecules. In Fig. 1 we show the longitudinal elastic constant $c_{11}(x,T)$ as a function of the temperature for $x = 0.5\%$, 5%, 20%, 30%, 40%, and 96%. As a comparison, we include data obtained by means of Brillouin scattering for the pure KCl and KCN (Ref. 5) crystals.

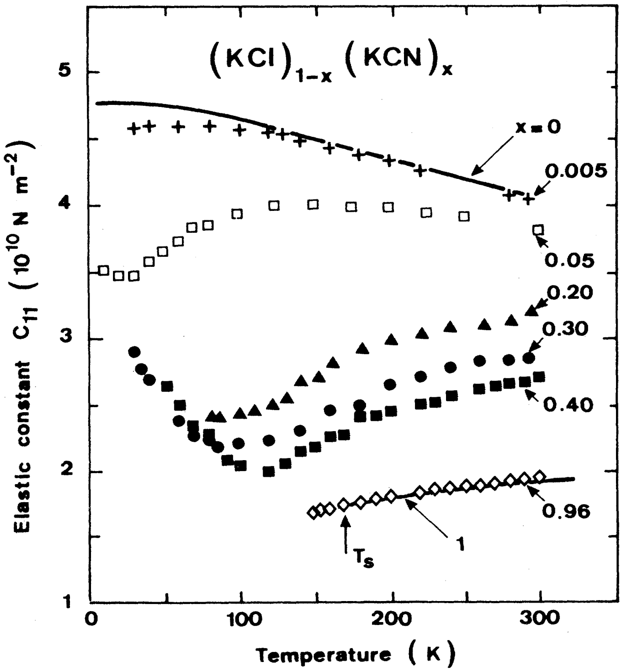

FIG. 1. Elastic constant c_{11} vs temperature measured by Brillouin scattering in $(KCl)_{1-x}(KCN)_x$ mixed crystals for several concentrations. c_{11} data for pure KCN are taken from Ref. 5. The arrow indicates the ferroelastic transition temperature.

As evidenced in Fig. 1, a 4% substitution of CN by Cl ions does not affect the sound velocity of thermal phonons. However, the critical temperature T_S is shifted from 168 K in KCN to 150±3 K. Due to the formation of macroscopic domains of aligned CN, the sample becomes opaque and no Brillouin lines could be detected any more below T_S, in accordance with previous reports on optical transparency.[1]

The absence of soft-mode behavior above x_c in the longitudinal elastic constant, analogous to what Wang *et al.*[8] observed in $c_{44}(x,T)$ in the same system, can be understood according to symmetry arguments. As already mentioned, LA waves couple to librational E_g modes of the cyanide molecule only. Such an interaction mechanism leaves the CN orientation unchanged, and thus should not participate in the strong lattice-mediated CN-CN quadrupolar coupling which triggers the transition at T_S. Compared to the T_{2g}-symmetry molecular excitations, the E_g translational and rotational modes are less involved and consequently less perturbed by the transition. Hence, $c_{11}(T)$ for $(KCl)_{0.04}(KCN)_{0.96}$ (and more generally for systems with $x \gtrsim x_c$) does not vanish as $T \rightarrow T_S$. We will return to this point later.

Below x_c the light-scattering experiments could be carried out down to the lowest temperatures. Instead of a discontinuity in the c_{11} behavior, the elastic constant passes through a minimum, which changes markedly with the composition, from $T_F = 110$ K at $x = 0.40$ to $T_F = 30$ K at $x = 0.05$. On the two less concentrated samples ($x = 0.5\%$ and 5%), the $c_{11}(T)$ data increase upon cooling from room temperature, indicating the weak role played by the *T-R* coupling at these compositions. No minimum could be evidenced for the $x = 0.005$ crystal. Moreover, both pure KCl and $x = 0.005$ samples exhibit a typical crystalline behavior: $c_{11}(T)$ increases linearly with decreasing temperature before reaching a plateau as $T \rightarrow 0$. In order to observe changes in the elastic properties of KCl due to diluted elastic quadrupoles, Brillouin experiments should be performed at a temperature such as $k_B T \simeq \Delta$, where Δ denotes the tunnel energy of an isolated impurity [for CN in KCl, $\Delta \simeq 1.2$ K (Ref. 13)]. Therefore, the $x = 0.5\%$ crystalline behavior can be ascribed in the present *T* range to anharmonic phonon-phonon interactions,[14] rather than to resonant coupling of acoustic waves with elastic defects.

The configuration of our Brillouin spectrometer enables us to measure the inverse mean free path of the thermal phonons, $(l_{11})^{-1} = \alpha_{11}$. Here α_{11} is the acoustic attenuation. Directly related by the half-width at half maximum (HWHM) of the Brillouin line Γ, $(l_{11})^{-1} = 4\pi\Gamma/v_l$ is determined with an experimental accuracy of 10% for widths larger than 10 MHz (it should be kept in mind that the Brillouin shifts currently range between 10 and 20 GHz). Among the different acoustic measurements available, the Brillouin experiments appear to be one of the most efficient tools to measure the phonon damping in systems like $(MX)_{1-x}(M\text{CN})_x$.

Figure 2 displays a plot of the inverse mean free path

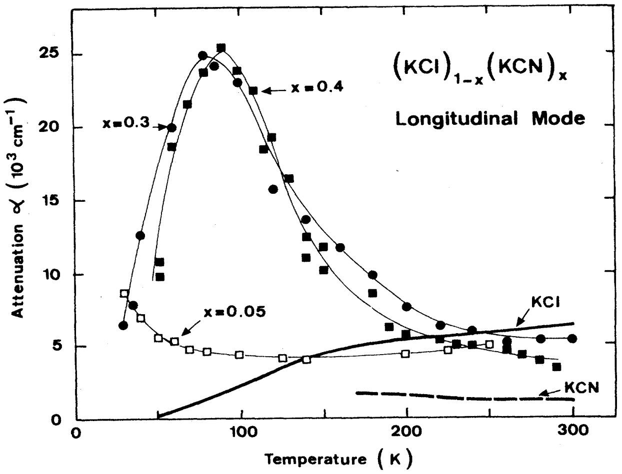

FIG. 2. Variation of the longitudinal-phonon damping α_{11} as a function of temperature for three KCl:KCN mixed crystals with compositions $x=5\%$, 30%, and 40%. The attenuation data for $x=0$ (Ref. 14) and (Ref. 5) are also shown. The solid lines are a guide to the eyes only.

of longitudinal phonons for the compositions $x=0$, 0.05, 0.30, 0.40, and $x=1$. With increasing CN concentrations room-temperature α_{11} values decrease approximately by a factor of 5 from pure KCl to KCN. However, the most conspicuous feature is the occurrence of a tremendous peak in attenuation at $x=0.30$ and 0.40, above the lattice contribution. Figure 2 clearly shows that the location of the peak shifts to higher temperature by increasing the composition. Centered at $T=86\pm3$ and 92 ± 3 K for $x=0.30$ and 0.40, respectively, their forms are asymmetric and their comparable heights are 10 times larger than the attenuation in pure KCl. As expected from the Kramers-Kronig relations, these absorption peaks are associated with the finite cusps observed in the real part of the acoustic susceptibility (Fig. 1). The freezing temperatures $T_F(x)$ defined as the temperatures at which $c_{11}(T)$ $[\alpha_{11}(T)]$ passes through a minimum (a maximum) are listed in Table I. Compared to the $\alpha_{11}(x=1)$ data, for which a weak increase of the phonon damping is observed at T_s (Ref. 5) (see the dashed curve in Fig. 2), this feature reinforces the idea that the freezing process in the mixed systems is a dynamical phenomenon. In passing, we disagree with recent experimental findings obtained on the KBr:KCN systems by Vanderwal *et al.*,[15] who claimed that the "critical" temperatures (given by the extrema in c_{ii} and α_{ii}, $i=1$ or 4) are essentially the same for each respective concentration. For x varying from 0.20 to 0.40 in Table I, we observe a maximum absorption always located below the c_{11} cusps by about 5–15 K.[16]

III. RANDOM-STRAIN-FIELD ANALYSIS OF THE RESULTS

As outlined in the the Introduction, it is now our purpose to provide a quantitative analysis of the $(KCl)_{1-x}(KCN)_x$ elastic data, as a function of composition and temperature. $c_{11}(x,T)$ and $c_{44}(x,T)$ are investigated. To this end, we reexamine previously published Brillouin-scattering results on the elastic shear constant obtained by Wang *et al.*,[8] and compare their T behaviors to a model of orientational glass state derived recently for mixed cyanides.[9]

We first focus our analysis on the high-temperature regime, where both c_{11} and c_{44} are decreasing with temperature. According to the initial T-R coupling theory applied to molecular crystals, the softening of the elastic constants can be described by a Curie-Weiss law of the form

$$c_{ii}(T)=c_i^0\frac{T-T_{ci}}{T-T_{0i}} \ . \tag{1}$$

Here, $i=1$ or 4 and c_i^0 is the bare elastic constant that would be observed in absence of T-R coupling. c_i^0 is taken to be temperature independent. T_{ci} is the Curie-Weiss temperature, at which $c_{ii}(T)$ vanishes for $x>x_c$ or would go to zero if the substitutional halogens at $x<x_c$ would not have frustrated the transition. T_{0i} is a negative temperature related to the CN-CN orientational interactions. Thus, treating c_i^0, T_{ci}, and T_{0i} as independent fitting parameters, both elastic constants $c_{11}(T)$ and $c_{44}(T)$ for $(KCl)_{1-x}(KCN)_x$ in its cubic structure could be conveniently fitted at all x according to Eq. (1).

Results of the fitting procedure are summarized in Fig. 3, where the longitudinal constant normalized to its room-temperature value, $c_{11}(T)/(c_{11})_{RT}$ is plotted against T. In Fig. 3, a remarkable behavior is evidenced: for $x=0.20$, 0.30, 0.40, 0.96, and 1, the ratio $c_{11}(T)/(c_{11})_{RT}$ follows a nearly identical T variation whatever the composition. The c_{11} data points above 100 K are found superimposed and are in agreement with Eq.

TABLE I. Temperatures of the extrema observed in the longitudinal elastic constant (Fig. 1) and attenuation (Fig. 2) in $(KCl)_{1-x}(KCN)_x$. As an indication, values for $x=0.56$ are included for which only the mixed elastic constant $c''=\frac{1}{2}(c_{11}+c_{12}+2c_{44})$ has been measured (see inset in Fig. 3), as well as the temperature of the ferroelastic transition for $x=0.96$ and 1. The relatively large uncertainty in the determination of $T_F(c_{11})$ is due to the flat minimum exhibited by the elastic constant at the orientational glass transition. (n.o. denotes not observed.)

	Concentration						
	0.05	0.20	0.30	0.40	0.56	0.96	1
$T_{F,S}(c_{11})$ (K)	30±5	85±5	90±5	110±5	125±5	150±3	168
$T_F(\alpha_{11})$ (K)	n.o.	~80	86±3	92±3	~110		

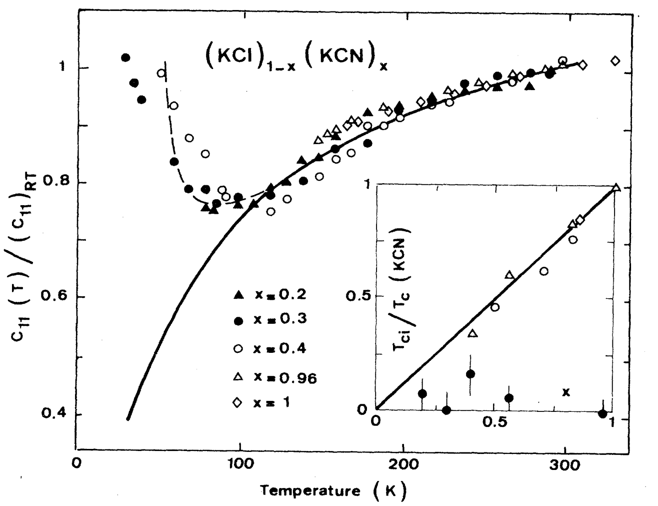

FIG. 3. Elastic constant c_{11} normalized to its room-temperature value $(c_{11})_{\mathrm{RT}}$ in $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ for five concentrations between $x=0.20$ and 1. The solid curve represents a Curie-Weiss law [Eq. (1)] with $T_{c1}=0$ and $T_{01}=-65\pm10$ K as adjustable parameters. For the dashed curve, see text. Inset: Normalized Curie-Weiss temperatures $T_{c1}/T_c(\mathrm{KCN})$ (solid circles) and $T_{c4}/T_c(\mathrm{KCN})$ (open circles) are plotted against the CN concentration [$T_c(\mathrm{KCN})=153$ K (Refs. 8, 10, and 17)]. Open diamonds, from Ref. 8; open triangles, from Ref. 17. For the TA modes only, a relation $T_{c4}\simeq xT_c(\mathrm{KCN})$ is evidenced, according to the mean-field theory (given by the straight line in the inset).

(1) using $T_{c1}=0\pm20$ K and $T_{01}=-65\pm10$ K as adjustable parameters. The corresponding Curie-Weiss law is represented in Fig. 3 by the smooth curve.

Contrary to this, Fig. 4 demonstrates that the softening of the transverse elastic constant depends on the composition. Again in this figure we plotted the constant normalized to its room-temperature value. The nominal concentrations are $x=0.30$, 0.50, 0.70, and $x=0.82$. As suggested above the cusps, the higher the composition, the faster c_{44} vanishes. The results of the best-fit calculations of the ratio $c_{44}(T)/(c_{44})_{\mathrm{RT}}$ using Eq. (1) are not displayed on the graph. However, in the inset on Fig. 3 we show the dependences of the critical temperatures T_{c1} and T_{c4} on cyanide composition. As already reported by Garland *et al.*,[17] and in accordance with the mean-field theory, $T_{c4}(x)/T_c(\mathrm{KCN})$ varies linearly with x (see the straight line in the inset). For the longitudinal mode, we receive from the same fitting procedure T_{c1} values roughly equal to 0 between $x=0.20$ and 0.96. The present findings (see solid circles in Fig. 3) obviously support the former observation of a unique behavior for the ratio $c_{11}(T)/(c_{11})_{\mathrm{RT}}$ at $T\geq T_{S,F}$ and attest again that the lattice-mediated CN-CN interactions via LA phonons do not play an important role in the various structural changes observed in the mixed cyanides.

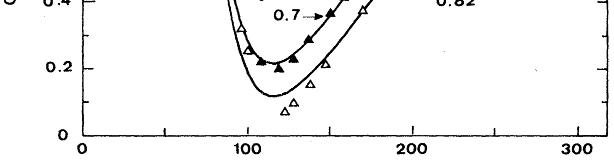

FIG. 4. Shear elastic constant c_{44} normalized to its room-temperature value $(c_{44})_{\mathrm{RT}}$ vs temperature in $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$. The Brillouin-scattering data originate from Ref. 8. The solid curves are results of best-fit calculations according to Michel's model of the orientational glass transition [Eq. (2)]. The corresponding fitting parameters are given in Table II.

In order to account for the freezing process of the CN reorientations below the crossover concentration, Michel has recently proposed a microscopic theory of the orientational glass.[9] The model assumes that static strains are present at all temperatures in such molecular crystals and act as an external random field. Following Michel, this strain field arises basically from the volume mismatch between the halogen ions and the CN molecules. As the anion radius is increased from Cl^- (1.81 Å) to Br^- (1.95 Å) one expects a decrease of the critical concentration. Coupled to both orientational and translational modes, the random strain field in cyanide mixtures [and in other systems like $(\mathrm{NaCN})_{1-x}(\mathrm{KCN})_x$ (Ref. 18)] is responsible for the disappearance of structural transitions below x_c and for the frequency-dependent cusp in the orientational susceptibility. Finally, constant-pressure molecular dynamics has stressed the importance of random strain fields in crystals like $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ and $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$.[19]

In order to test Michel's theoretical predictions, we will utilize the above reported elastic data. The shear elastic constant in presence of the random field can be derived within a mean-field treatment by replacing in Eq. (1) (for $i=4$ only) T_{c4} by $T_c[1-x(1-x)\xi h^2/T^2]$ and T_{04} by $T_0[1-x(1-x)\xi h^2/T^2]$. Here h represents the strength of the random strain field and ξ is a slightly varying temperature function. According to Michel, we take $\xi=0.07$ over the whole T range of interest. Like T_{c4} and T_{04}, T_c and T_0 are expected to be proportional to x. From the above considerations, one obtains

$$c_{44}(T)=c_{44}^{0}\,\frac{T-T_c[1-x(1-x)\xi h^2/T^2]}{T-T_0[1-x(1-x)\xi h^2/T^2]}\ . \tag{2}$$

Results of best-fit calculations using Eq. (2) are shown in Fig. 4 as solid lines for $x=0.30$, 0.50, 0.70, and 0.82. As in the previous analysis, c_{44}^0, T_c, T_0, and h are considered adjustable parameters. We note that Eq. (2) accounts well for the $c_{44}(x)$ cusps at the freezing. However, since the intensity of the random field, h^2/T^2, plays an increasing role as $T\rightarrow0$, we were able to achieve good fits only down to 30 K below T_F. Actually, when

TABLE II. Values of adjustable parameters c_{44}^0 (expressed in 10^9 N/m^2), T_c, and h received from best-fit calculations on the transverse elastic constant c_{44} (Fig. 4) using Eq. (2).

$(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$	c_{44}^0 (10^9 N/m^2)	T_c (K)	h (K)
$x=0.30$	6.4 ± 0.8	28 ± 5	290 ± 10
$x=0.50$	6.7 ± 1.3	105 ± 10	440 ± 10
$x=0.70$	4.5 ± 0.4	155 ± 2	540 ± 7
$x=0.82$	5.7 ± 1.9	162 ± 4	650 ± 13

$x(1-x)$, $\xi h^2/T^2$ becomes larger than 1, and c_{44} diverges at some lower temperature. That indicates that the present model is no longer appropriate to describe the elastic properties of $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ in its orientational glass state: Approaching the lowest temperature, a quantum-mechanical treatment is required.

The values of the fitting parameters c_{44}^0, T_c, and h are listed in Table II. Contrary to the bare elastic constant, the Curie-Weiss temperature strongly varies with x, but the predicted relation $T_c \propto x$ is not observed.[20] The strength of the strain field is found to increase linearly with the composition: h ranges from 300 to 650 K for $x=0.3$–0.8. Although this variation is unexpected within the initial microscopic theory, the overall value of h corresponds to Michel's estimations: $h(\mathrm{Cl})\simeq 450$ K.[9] The results in Table II differ from recent determinations of h(Cl) due to Fossum and Garland,[10] who utilized ultrasonic results on the TA sound velocity. These authors received for the present systems a value for the random field of about 400 K independent of x. According to us, this discrepancy originates from the fact that, due to the disappearance of the ultrasonic echoes well above T_F, their estimates of h (and T_c) are cast only from the softening regime, and not from the location of the minimum in the shear elastic constant. Both analysis of $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ data reveal, however, that the random strain field is larger in the chloride than in bromide cyanides, as expected from the model [$h(\mathrm{Br})\sim 150$ K].

There exists another experimental feature which deserves attention. The incomplete softening of the constants associated with the LA and TA modes occur at distinct freezing temperatures. As shown in Figs. 1 and 4 for $(\mathrm{KCl})_{0.7}(\mathrm{KCN})_{0.3}$, the minimum in c_{44} occurs nearly 20 K below that of c_{11}. Granted that Michel's model can be applied to E_g-symmetry translational modes, we achieved consistent fits of both real and imaginary parts of the corresponding orientational susceptibility. At each respective concentration, we obtain for LA modes stronger h values than those displayed in Table II for TA modes (this is the straightforward consequence of the analytical expression of Eq. (2) which shows a minimum at $T_F=h[3x(1-x)\xi]^{1/2}$ (Ref. 9)). Computed for $(\mathrm{KCl})_{0.7}(\mathrm{KCN})_{0.3}$ with a field value $h=370\pm20$ K, the dashed curve in Fig. 3 attests to the reliability of the fit on c_{11} data down to the freezing temperature. A theoretical verification of this outcome should represent a decisive test of the random-strain hypothesis.

IV. CONCLUSIONS

In summary, the transition between the crystalline and glasslike phases clearly appears in the elastic properties below the crossover concentration $x_c\simeq0.8$. This transition is a dynamical phenomenon, which is presently interpreted in terms of a quenched random strain field. The random strain operates as an external field with a Gaussian distribution at each molecular site.[9] The incomplete softening of $c_{11}(T)$ and $c_{44}(T)$ is analyzed quantitatively, providing an estimate for the strain field in agreement with theoretical predictions. However, the observed linear tendency for the variation of h with x remains an open question. As a support of Michel's ideas on the microscopic origin of the random field, we confirm that its strength is much larger in $(\mathrm{KCl})_{1-x}(\mathrm{KCN})_x$ than in $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$.

ACKNOWLEDGMENTS

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[12] The sound velocity of LA modes, v_l, is calculated from the shift $\Delta\nu_B$ of the Brillouin component with respect to the elastic scattered Rayleigh line through the relation $\Delta\nu_B/\nu_0=\pm2(n/c)v_l\sin(\theta/2)$. Here, n is the refractive index, c the light velocity, and θ the scattering angle (in the present

configuration, $\theta = 180°$). As far as the refractive index and the mass density are concerned, we assume their x dependences to be the weighted average between the corresponding values for KCl and KCN. Their T variations are also considered in computing v_l by the use of the Clausius-Mossotti relation.

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[20] The procedure fitting the c_{44} data according to Eq. (2) uses a Marquardt algorithm which allows the parameters freedom to adjust. Results of the calculation provide values for these quantities within a 90% confidence interval. While c_{44}^0, T_c, and h are obtained with a reasonable accuracy, the procedure fails to determine the remaining parameter T_0 unambiguously. As far as T_0 is concerned, one observes a large 90% confidence interval, indicating finally that this temperature has a weak influence on the overall T behavior of the elastic constant [in Eq. (2)].