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Theory of the orientational glass state in mixed crystals $M(\mathrm{CN})_x X_{1-x}$. II. Dynamics

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On the basis of a microscopic model, the dynamic properties of orientationally disordered crystals with substitutional impurity atoms are investigated. The temperature and concentration dependence of the acoustic phonon frequencies is studied. The anomalous damping of phonons is explained. The resonances of the dynamic response function are studied as a function of frequency. The central peak is obtained as a static phenomenon; its temperature dependence is derived. A theoretical foundation for NMR line shapes and x-ray linewidths is presented, and the relation of these data with the central peak is established.

I. INTRODUCTION

In the preceding paper,[1] to be referred to as I, we have presented a microscopic model for the description of mixed crystals[2] $M(\mathrm{CN})_x X_{1-x}$, where M is an alkali-metal and X a halogen ion. The theory takes into account the random strain fields which are due to the substitutional halogen ions. Above a certain critical concentration of halogen ions, the random strain fields suppress the transition to a phase with long-range orientational order. While the overall structure of the crystal stays cubic, cubic symmetry is broken on a local scale. Deformations of the lattice are accompanied by a local freezing in of orientations. Experimental evidence from neutron scattering[3,4] suggests such a picture[5] of the orientational glass state.

The onset of the glass state is characterized by interesting experimental phenomena, in particular, the passage of the transverse-acoustic-phonon frequencies through a minimum[6–8] and the concomitant appearance of an elastic central peak.[3,4,6] A straightforward extension of the dynamic theory of translation-rotation coupling[9] is inadequate to account for the experimental phenomena. A discussion of the shortcomings is given in Refs. 10 and 5. In particular the description of dynamic phenomena in Ref. 9 is unsatisfactory since the statics which is used there is inadequate. The results of I give a new description of statics in mixed crystals and the purpose of the present paper is to extend this description to dynamics.

In Sec. II we discuss the static displacement-displacement susceptibility. Next (Sec. III) we study the dynamic response functions. In particular, we investigate the dependence of the phonon resonances on the frequency of the experimental method. The temperature dependence of phonon damping is discussed. In Sec. IV we study the central peak which appears in the neutron scattering law.

II. DISPLACEMENTS SUSCEPTIBILITY

In order to calculate the phonon frequencies, we first need the static displacements-displacements susceptibility. We start from Eq. (4.6) of I for the free energy,

$$\bar{F}=\bar{F}_W+\sum_{\mathbf{k}}\{\tfrac{1}{2}\bar{\mathbf{s}}^{\dagger e}(\mathbf{k})\underline{M}(\mathbf{k})\bar{\mathbf{s}}^{e}(\mathbf{k})+\bar{\mathbf{Y}}^{e\dagger}(\mathbf{k})\underline{v}(\mathbf{k})\bar{\mathbf{s}}^{e}(\mathbf{k})$$
$$+\tfrac{1}{2}[(\underline{\chi}^0)^{-1}+\underline{J}(\mathbf{k})+\underline{1}C^s]\bar{\mathbf{Y}}^{e\dagger}(\mathbf{k})\bar{\mathbf{Y}}^{e}(\mathbf{k})\}\ . \tag{2.1}$$

Here $\mathbf{Y}^e$ and $\mathbf{s}^e$ are the orientational and translational variables, respectively. The first term $\bar{F}_W$ represents the single particle orientational free energy. The terms within curly brackets account for the elastic interaction energy with the bare dynamical matrix $\underline{M}$, for the bilinear T-R coupling with interaction $\underline{v}$, and for the orientational interaction J. The quantity $\underline{\chi}^0$ denotes the single particle orientational susceptibility and is given by Eq. (4.9) of I:

$$\chi^0=x(y_W/T)[1-x(1-x)\xi_W h^2/T^2]\ . \tag{2.2}$$

Here y_W and ξ_W are single particle correlation functions, and h represents the amplitude of the random field. For further details about expressions (2.1) and (2.2), we refer to I.

For a given configuration of displacements $\{\bar{\mathbf{s}}^e(\mathbf{k})\}$, we minimize $\bar{F}$ with respect to the orientations $\bar{\mathbf{Y}}^e(\mathbf{k})$ and obtain

$$\bar{\mathbf{Y}}^e(\mathbf{k})=-[(\underline{\chi}^0)^{-1}+\underline{1}C^s+\underline{J}(\mathbf{k})]^{-1}\underline{v}(\mathbf{k})\bar{\mathbf{s}}^e(\mathbf{k})\ . \tag{2.3}$$

Inserting this result into Eq. (2.1), we get $\bar{F}$ as a functional of the displacements

$$\bar{F}=\tfrac{1}{2}\sum_{\mathbf{k}}\bar{\mathbf{s}}^{e\dagger}(\mathbf{k})\{\underline{M}(\mathbf{k})-\underline{v}^{\dagger}(\mathbf{k})$$
$$\times[(\underline{\chi}^0)^{-1}+\underline{1}C^s+\underline{J}(\mathbf{k})]^{-1}v(\mathbf{k})\}\bar{\mathbf{s}}^e(\mathbf{k})\ . \tag{2.4}$$

Using a standard method of statistical mechanics,[11] we then obtain for the displacement-displacement susceptibility

$$\begin{aligned}\underline{D}^{-1}(\mathbf{k})&=T^{-1}\langle \overline{\mathbf{s}}^{e\dagger}(\mathbf{k})\overline{\mathbf{s}}^{e}(\mathbf{k})\rangle \\ &=\underline{M}^{-1}(\mathbf{k})[\underline{1}+\underline{v}^{\dagger}(\mathbf{k})\underline{\chi}(\mathbf{k})\mathbf{v}(\mathbf{k})\underline{M}^{-1}(\mathbf{k})] \ . \end{aligned} \tag{2.5}$$

Here $\underline{\chi}(k)$ is the collective orientation-orientation susceptibility, given by Eq. (5.1) of I:

$$\underline{\chi}(\mathbf{k})=\chi^0[\underline{1}-\chi^0\underline{L}(\mathbf{k})]^{-1} \ , \tag{2.6a}$$

with

$$\underline{L}(\mathbf{k})=\underline{v}(\mathbf{k})\underline{M}^{-1}(\mathbf{k})\underline{v}^{\dagger}(\mathbf{k})-\underline{1}C^s-\underline{J}(\mathbf{k}) \ . \tag{2.6b}$$

Taking $\mathbf{k}$ along one of the cubic axes, say $\mathbf{k}=(0,0,k)$, we find that all matrices are diagonal. In the long-wavelength limit, $\underline{\chi}(\mathbf{k})$ is given by Eq. (5.3) of I, and from Eq. (2.5) we find

$$D_{11}(k)=M_{11}(k)\left[\frac{1-\chi^0\delta}{1+(J+C^s)\chi^0}\right] \ . \tag{2.7a}$$

Here $J=J_{22}$ $(k=0)$, and δ is the largest eigenvalue of $\underline{L}(k)$ given by Eq. (5.4) of I. The bare dynamical matrix $\underline{M}$ is proportional to the elastic constants c_{ij}^0 in absence of T-R coupling, while $\underline{D}$ is proportional to the renormalized elastic constants. In particular

$$D_{11}(k)/M_{11}(k)=c_{44}/c_{44}^0 \ , \tag{2.7b}$$

and the temperature dependence of c_{44} is governed by Eq. (2.7a). The temperature dependence of c_{44} is shown in Fig. 1 for several values of the concentration x of CN$^-$ ions. The most remarkable feature is the appearance of the rounded minimum if x is smaller than the critical concentration x_c which is given by Eq. (5.6c) of I. We recall that a minimum in D_{11} or in c_{44} corresponds to a rounded maximum in the collective orientational susceptibility $\chi_{22}(k)$. The latter has been investigated in I. From Fig. 1 we see that the position $T=T_f$ of the minimum shifts to lower values with decreasing concentration x. This effect has been discovered by Brillouin scattering experiments.[7] The concentration and temperature dependence of the elastic constants is a reflection of the phase diagram, Fig. 3 of I. The appearance of the minimum has been found first by inelastic neutron measurements of transverse-acoustic-phonon frequencies[6] in K(CN)$_x$Br$_{1-x}$. The position T_f of the minimum depends also on the experimental method.[10,12,13] Since ultrasound, Brillouin scattering, and neutron scattering are characterized by different frequency regimes, we shall investigate the dynamic response functions.

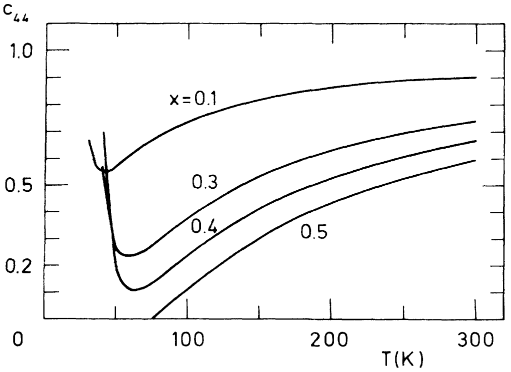

FIG. 1. Temperature dependence of elastic constants c_{44}/c_{44}^0 for $h=300$ and CN$^-$ concentration $x=0.1$, 0.3, 0.4, and 0.5.

III. DYNAMIC RESPONSE

We start from the formulation of the dynamic response functions which has been given previously for systems with bilinear translation-rotation coupling.[14] We transcribe the expressions of the dynamic correlation functions by taking into account the expressions of the static susceptibilities for mixed crystals $M(\mathrm{CN})_x X_{1-x}$.

Dynamic equations are most conveniently formulated in terms of Kubo's relaxation function $\Phi^{AB}(z)$, $z=\omega+i\epsilon$, $\epsilon\to 0$. The imaginary part $\Phi''(\omega)$ is related to the dynamic correlation function by

$$S^{AB}(\mathbf{k},\omega)=-\omega[1-\exp(-\omega/T)]^{-1}\Phi^{AB''}(\mathbf{k},\omega) \ . \tag{3.1}$$

We are interested in the dynamic displacement-displacement correlation function $\underline{\Phi}^{ss}(\mathbf{k},\omega)$ and take $\mathbf{k}=[0,0,k]$. Then $\underline{\Phi}^{ss}$ is diagonal and the relevant element Φ_{11}^{ss} refers to the s_x-s_x correlation. The corresponding equation of motion reads[14] as

$$[\omega^2-D_{11}(k)-\omega\sigma(\omega)]\Phi_{11}^{ss}(k,\omega)=[\omega-\sigma(\omega)][\underline{D}^{-1}(k)]_{11} \ . \tag{3.2}$$

Here D_{11} is given by expression (2.7a) and the function $\sigma(\omega)$ accounts for the orientational relaxation

$$\sigma_{11}(\omega)=\Delta_{11}/(\omega+i\lambda) \ , \tag{3.3a}$$

with

$$\Delta_{11}(k)=M_{11}(k)-D_{11}(k) \ . \tag{3.3b}$$

Expression (3.3a) for $\sigma_{11}(\omega)$ has been obtained under the assumption that the orientational dynamics corresponds to a relaxation process with relaxation frequency $\hbar\lambda$. We expect that the relaxation approximation is correct in the case of weak potentials and not too low temperature.[15] As has been discussed in I, the static susceptibilities of the present theory have been derived in the classical regime and we expect that for $T \gtrsim 30$ K, also the relaxation approximation for λ is valid. We estimate that $\hbar\lambda$ is of the order of 1 THz at 240 K which corresponds to the fast relaxation regime and is in agreement with neutron scattering results.[16] Since[15] $\lambda \propto \sqrt{T}$, we approximate the temperature dependence of λ by the expression $\lambda^2=\lambda_0^2(T/240)$, where $\lambda_0=1$ THz. The resonances of the scattering function $S_{11}^{ss}(\omega)$ are determined from the imaginary part of $\Phi_{11}^{ss}(\omega)$. We obtain from Eq. (3.2)

$$\Phi_{11}^{ss''}(\omega)=\frac{-\Delta_{11}f(\omega)}{[\omega^2-D_{11}(k,\omega)]^2+[\omega\Delta_{11}f(\omega)]^2} \ , \tag{3.4}$$

where we have defined

$$f(\omega)=\frac{\lambda}{\omega^2+\lambda^2} \ , \tag{3.5}$$

$$D_{11}(k,\omega)=M_{11}(k)-\Delta_{11}(k)\frac{\lambda^2}{\omega^2+\lambda^2} \ . \tag{3.6}$$

We notice that in the limit $\omega \to 0$, $D_{11}(k,\omega)$ reduces to $D_{11}(k)$. The quantity $D_{11}(k,\omega)$ determines the real part of the resonance frequencies. We have calculated $D_{11}(k,\omega)/M_{11}(k)$ as a function of temperature for several concentrations and for several values of ω. As is shown in Fig. 2, the minimum is more shallow for higher values of ω; in addition, the position of the minimum becomes frequency dependent. $T_f(\omega)$ shifts to higher values for larger frequencies ω. These results are in agreement with experimental observations. We remark that Δ_{11} plays the role of an effective translation rotation coupling. From Eq. (3.3b) it follows that the increase of $D_{11}(k)$ at temperatures $T<T_f$ appears as a decrease of the effective coupling Δ_{11} below T_f. Such a behavior has been inferred from neutron scattering results.[17]

We next consider the frequency-dependent damping. In KCN, the phonon linewidth increases with decreasing temperature in the cubic phase and reaches a maximum at the 168-K phase transition.[18] This anomalous behavior is accounted for by the theory of bilinear translation-rotation coupling.[15,18] On the other hand, in $K(CN)_xX_{1-x}$, with $x<x_c$, the phonon damping first increases with decreasing $T>T_f$, passes through an anomalous large maximum which is located near T_f and then decreases with decreasing $T<T_f$. The large damping near $T \approx T_f$ has been confirmed by various techniques, especially by ultrasonic experiments.[10,19] Most recently, the temperature variation of the phonon linewidth has been investigated by Brillouin spectroscopy.[20] As is discussed by the authors of Ref. 20, the previous dynamic theory,[9] although it takes into account some effects of phonon dispersion, is not able to describe qualitatively the temperature dependence of the phonon linewidth. From the denominator of Eq. (3.4), we see that the frequency-dependent phonon damping is given by

$$\Gamma(\omega)=\omega\Delta_{11}f(\omega) . \tag{3.7}$$

The temperature dependence of $\Gamma(\omega)$, for a fixed value of the frequency ω, is also shown in Fig. 2. The maximum of $\Gamma(\omega)$ occurs at a temperature which is slightly below the corresponding $T_f(\omega)$, where $D_{11}(k,\omega)$ has its minimum. Both the overall temperature dependence of Γ and the slight shift of the maximum with respect to $T_f(\omega)$ are in qualitative agreement with experimental results.[20] The shift of the maximum can also be deduced[21] from the data on the elastic properties.[19]

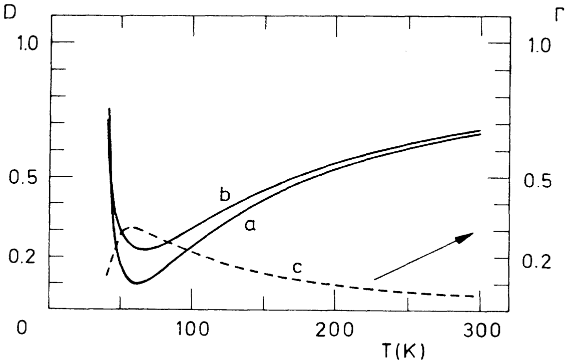

FIG. 2. Temperature dependence of $D_{11}(k,\omega)/M_{11}(k)$ for $\omega=0.2$ THz (curve b); $\omega=0.0$ (curve a). Dashed curve shows damping $\Gamma(\omega)/M_{11}(k)$, $\omega=0.2$ THz. In all cases, $x=0.4$ and $h=300$.

IV. CENTRAL PEAK

A characteristic feature which is associated with the orientational glass state is the appearance of a central peak in the neutron scattering law.[3] The width of the peak is below the resolution of neutron spectroscopy.[3,13] Therefore Rowe *et al.* have proposed[5] that the central peak is a static phenomenon which is due to the freezing in of orientational modes on a local scale. Following Ref. 5, we write for the instantaneous value of the orientational mode Y_α at lattice site l

$$Y_\alpha(l,t)=\langle Y_\alpha(l)\rangle+\delta Y_\alpha(l,t) . \tag{4.1}$$

The first term on the right-hand side represents a local average and is time independent, the second term accounts for time-dependent fluctuations around the equilibrium value with $\langle \delta Y_\alpha(l,t)\rangle=0$. The separation of $Y_\alpha(l,t)$ in a frozen in local part and in a fluctuating part is similar to the point of view taken by Halperin and Varma[22] for the description of displacement fields in the presence of frozen interstitial defects.

While in Ref. 5, $\langle Y_\alpha(l)\rangle \neq 0$ was taken as a working hypothesis, we will show now that the first term on the right-hand side of Eq. (4.1) follows directly from the microscopic model of I. Indeed the random strains which are generated by the substitutional halogens couple to the orientational modes of the residual CN^- molecular ions. The corresponding interaction is given by Eq. (3.5) of I:

$$V^{SR}(\{\sigma\})=\sum_{\mathbf{n}} \sigma(\mathbf{n})Y_\alpha(\mathbf{n})h_\alpha(\mathbf{n},\{\sigma\}) . \tag{4.2}$$

Here $\sigma(\mathbf{n})$ accounts for the occupation of the CN^- ions on the lattice and equivalently $\{\sigma\}$ determines the given configuration of substitutional impurities. The random strain field h_α has been explicitly discussed in I. Applying linear-response theory, we obtain for the frozen in orientation which is induced by V^{SR} at lattice site l

$$\langle Y_\alpha(l)\rangle=-\sigma(l)(y_W/T)h_\alpha(l,\{\sigma\}) . \tag{4.3}$$

Here y_W is the single particle expectation value, given by Eq. (4.10a) of I. From property (3.6) of I, we obtain for the configurational average (denoted by an overbar),

$$\overline{\langle Y_\alpha(l)\rangle}=0 . \tag{4.4}$$

On the other hand, Eq. (3.8a) of I implies

$$\overline{\langle Y_\alpha(l)\rangle\langle Y_\beta(l')\rangle}=\delta_{ll'}\delta_{\alpha\beta}x\Psi , \tag{4.5a}$$

where the glass order parameter Ψ is defined by

$$\Psi=x(1-x)y_W^2(h^2/T^2) . \tag{4.5b}$$

The strength of the random field is given by Eq. (3.8b) of I. It has been shown in Ref. 5, that the separation of the orientational mode in a frozen in part and in a fluctuating part leads to the dynamic structure function for orientational motion

$$S^{YY}_{\alpha\beta}(\mathbf{q},\omega)=x\delta_{\alpha\beta}\Psi\delta(\omega)+\sigma^{YY}_{\alpha\beta}(q,\omega)\ . \qquad (4.6)$$

The first term on the right-hand side gives rise to the central peak, while the second term accounts for the dynamical part of the scattering law. The intensity of the central peak increases as T^{-2} with decreasing temperature as follows from Eq. (4.5b) for Ψ. We recall that y_W is only weakly temperature dependent. Here again we should observe that the present theory is not valid at very low T where a quantum-mechanical treatment is required (see remarks of I, Sec. IV). While a T^{-2} behavior of Ψ leads to a divergence at $T=0$, a quantum-mechanical treatment should yield a saturation of Ψ at $T=0$. A saturation is indeed observed by experiment.[3,13] The T^{-2} dependence is the result of a lowest-order treatment in h^2/T^2, it describes qualitatively the increase of Ψ below T_f.

As has been discussed previously,[5] a local freezing in of orientations entails a freezing in of lattice displacements. From Eq. (3.10) of Ref. 5, we have

$$\langle s_j(\mathbf{n})\rangle=-\sum_{\mathbf{l}} G_{ij,p}(\mathbf{n},\boldsymbol{l})P^{(\alpha)}_{pi}\sigma(\boldsymbol{l})\langle Y_\alpha(\boldsymbol{l})\rangle\ . \qquad (4.7)$$

Here $G_{ij,p}$ is the spatial derivative $[\partial X_p(\mathbf{n})/\partial]\ G_{ij}(\mathbf{n},\boldsymbol{l})$ of the elastic Green's function

$$G_{ij}(\mathbf{n},\boldsymbol{l})=\frac{1}{NM}\sum_{\mathbf{k}}[\underline{M}^{-1}(\mathbf{k})]_{ij}\exp\{i\mathbf{k}\cdot[\mathbf{X}(\mathbf{n})-\mathbf{X}(\boldsymbol{l})]\}\ . \qquad (4.8)$$

The matrix $P^{(\alpha)}_{jp}$ is related to the bilinear coupling matrix $\underline{v}$, Eq. (3.2) of I, by the long-wavelength relation

$$v_{\alpha j}(\mathbf{k})=-(i/\sqrt{m}\,)k_pP^{(\alpha)}_{pj}\ . \qquad (4.9)$$

The locally frozen in orientation $\langle Y_\alpha(\boldsymbol{l})\rangle$ which occurs in Eq. (4.7) is determined by Eq. (4.3). Therefore the concomitant lattice displacements (4.7) are temperature dependent. At this point we should insist on the difference with the static lattice displacements $s_i(\mathbf{k},\{\sigma\})$, which have been derived in I, Eq. (2.12a). The latter are static, temperature-independent lattice displacements, which are generated by the presence of the substitutional halogen impurities. From Eqs. (4.7) and (4.4) we deduce

$$\overline{\langle s_i(n)\rangle}=0\ . \qquad (4.10)$$

Using Eqs. (4.5a) we find

$$\overline{\langle s_i(\mathbf{n})\rangle\langle s_j(\mathbf{n}')\rangle}=\delta_{\mathbf{nn}'}\delta_{ij}Q^{(\alpha)}_{ii}x\Psi\ , \qquad (4.11\text{a})$$

where

$$Q^{(\alpha)}_{ii}=\frac{1}{Nm^2}\sum_{\mathbf{k}}k_pP^{(\alpha)}_{pi'}[\underline{M}^{-1}(\mathbf{k})]_{i'i}k_{p'}P^{(\alpha)}_{p'j'}[\underline{M}^{-1}(\mathbf{k})]_{j'i}\ . \qquad (4.11\text{b})$$

The temperature dependence is contained in the factor Ψ. In obtaining this expression, we have neglected correlations between sites $\mathbf{n}$ and $\mathbf{n}'$. Cubic symmetry of the configuration averaged expression implies that all three components α of $Q^{(\alpha)}_{ij}$ are equal and that $i=j$. The result (4.11a) and (4.11b) is equivalent to expressions that have been derived in Ref. 5 in order to explain the elastic contributions to the neutron scattering law. In particular, it was shown that the coupling of frozen in orientational modes to frozen in lattice strains accounts for the asymmetric shape[3,4] of the elastic intensity contours in $\mathbf{Q}$ space. The asymmetric intensity contours are the clear signature of the fact that translational and orientational modes of T_{2g} symmetry are frozen in.

The temperature dependence of the frozen in acoustic displacements has further consequences for the interpretation of experiments. X-ray diffraction experiments in $K(CN)_xBr_{1-x}$ show a drastic increase of the linewidth of the powder diffraction lines at temperatures below T_f in the nonordering compounds.[23] As has been suggested by the authors of Ref. 23, the line shapes can be translated into a distribution of inhomogeneous T_{2g} shear strains, centered around zero strain, and the width can be directly interpreted as the mean random field order parameter of the glass state. These properties are reflected in our results (4.7) and (4.11a). Additional information about the local deformations of the lattice is provided by NMR experiments.[24] In $Na(CN)_xCl_{1-x}$ the quadrupole moment of the ^{23}Na nucleus interacts with the static part of the electric-field-gradient (EFG) tensor. Deviations from cubic symmetry on a local scale in the lattice are reflected in a distribution of EFG's which should lead to an inhomogeneous broadening of the central line and to a wide spread of satellite transitions. The distribution width of the EFG's is a measure of the mean local deviation from cubic symmetry at the ^{23}Na sites. Experimentally one finds[24] a drastic increase of the distribution width with decreasing temperature. These results reflect the increase of local deformations in the lattice with decreasing T. Combining Eqs. (4.3) and (4.7), we obtain the temperature-dependent local displacements

$$\langle s_j(\mathbf{n})\rangle=(y_W/T)\sum_{l} G_{ij,p}(\mathbf{n},\boldsymbol{l})P^{(\alpha)}_{pi}\sigma(\boldsymbol{l})h_\alpha(\boldsymbol{l},\{\sigma\})\ , \qquad (4.12)$$

with property (4.10) and distribution width (4.11a). It has been pointed out as an experimental fact, that the width of the $(220)_c$ x-ray diffraction line and the distribution width of the electric-field gradient at the ^{23}Na sites in $Na(CN)_xCl_{1-x}$ have the same T dependence.[25] Our theory leads to an understanding of these results. In addition we find that the intensity of the central peak has the same temperature dependence since it is also proportional to the quadrupolar glass order parameter Ψ, Eq. (4.5b).

V. CONCLUDING REMARKS

We have applied a microscopic model[1] of $M(CN)_xX_{1-x}$ to the description of elastic and inelastic scattering laws. In particular we have studied dynamic effects such as phonon resonances and their linewidth as a function of temperature and CN^- concentration. The theory accounts in an unified way for many experimental facts: occurrence of a rounded minimum in the phonon frequency at T_f, shift of T_f to lower values with decreasing CN^- concentration, frequency dependence of the position of the minimum. In particular the theory reflects the differences which are related to the specific frequency regimes of experimental methods: ultrasound,[10,19] Brillouin[7,20] and neutron scattering.[3,13] The anomalous temperature dependence of the phonon linewidth[20] is obtained.

The central peak, which has been discovered by neutron scattering,[3] is derived as a static phenomenon, in agreement with a previous interpretation.[5] We calculate the temperature dependence of its intensity which increases strongly below T_f.[3,13] In addition to the static, temperature independent strains which are due to the substitutional impurities, we investigate the temperature-dependent static strain fields which are a consequence of the bilinear translation-rotation coupling.[5] The temperature dependence of the width of the distribution of strain fields, Eq. (4.11a), reflects experimental results of x-ray profile linewidths[23] and NMR line-shape distributions.[24]

In the present paper we have only investigated phenomena of T_{2g} symmetry, i.e., associated with the quadrupolar glass state. Many interesting phenomena in mixed crystals are related to dipolar symmetry.[2] In particular dielectric measurements[26,27] provide relevant information. It is a subject of further work to extend the microscopic model of I to the study of the dipolar glass state.

The present theory has been based on classical statistical mechanics (see also discussion in I). An extension to a quantum-mechanical version is required if one wants to understand low temperature properties of orientational glasses.[28] Such an extension is also necessary if one wants to investigate possible relations between the quadrupolar glass state in $K(CN)_xBr_{1-x}$ and in ortho-parahydrogen mixed crystals.[29]

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