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# Glass transition in $KBr_{1-x}KCN_x$ alloys

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It is shown that one can account for the temperature and frequency dependence of the dielectric and quadrupolar relaxations in $KBr_{1-x}$ $KCN_x$ with a single interaction energy between nearest neighbors, and a model based on the theory for hierarchically constrained relaxation of Palmer *et al.* [Phys. Rev. Lett. **53**, 958 (1984)]. In particular, the small change in the glass transition temperature with probe frequency can be accounted for. For $0.43 \leq x \leq 0.53$, the interaction energy deduced from fitting the data is 103 K. A Gaussian distribution of nearest neighbors has been assumed; the width of the distribution, and the orientation of the ion in the unit cell, are left as adjustable parameters.

## INTRODUCTION

A clear distinction can be made between a glass and a crystalline solid on the basis of their relaxational behavior: A perfect crystal cannot display anelastic phenomena; a glass, on the other hand, must, and the anelasticity of a glass is present, at least in principle, at all temperatures.

The glass transition is characterized by a slowing down of the relaxation of structural elements of the system. The system is considered frozen when the relaxation time exceeds the observation time. Thus, unlike, say, the liquid-solid transition, whether or not the system is considered to be frozen depends on the time scale of the observation.

The change in $T_g$, the perceived temperature of the glass transition, with the characteristic measurement time is usually very slow. An Arrhenius plot of the logarithm of this time against reciprocal temperature often yields very large activation energies and unphysically large prefactors, indicating that cooperative phenomena are involved. In addition, another relaxation process is often apparent at lower temperatures and is referred to as the beta relaxation. The relaxation process, which takes place at temperatures near the glass transition, is called the alpha relaxation. While the beta process does not display the characteristic temperature and frequency dependence of single Debye relaxation, the activation energies and prefactors derived for it are usually physically reasonable.

The freezing of a system of anisotropic defects in a crystalline matrix displays many of the features of the glass transition. The presence of a crystalline lattice, however, simplifies the situation. It will be shown that, as a consequence, a model that accounts for many features of the freezing process can be constructed.

The alloys of KCN with other alkali halides are materials in which the CN ions can enter an "orientational glass phase" at certain concentrations and temperatures. Of these, the KBr-KCN has been the most studied, and it will be to this system that we will devote most of our attention. The CN ion has both an electric dipole moment (referred to here as the "dipole"), and an elastic moment, often referred to as an elastic quadrupole moment (which we will call the "quadrupole"). An electric quadrupole moment also exists but is usually neglected. It is generally agreed that the elastic quadrupolar interactions are responsible for a glassy phase at concentrations $x$ between about 0.2 and 0.6. This manifests itself by a rapid slowing down of the quadrupolar relaxations as the temperature is lowered.

At temperatures below the glass transition temperature the dipoles are still able to "flip," head to tail,[1-3] leading to strong dielectric relaxation phenomena.[1] The dielectric relaxation peak is very broad in frequency. Its temperature and frequency dependence are fitted very well by a Gaussian distribution of energy barriers,[1-3] which neglects any interaction between the dipoles. Unfortunately, in order to obtain satisfactory agreement, a temperature dependence of the mean and the width of the distribution must be introduced *ad hoc.*

The barriers responsible for the dielectric relaxation are provided by the elastic interaction between the CN ions, i.e., the quadrupoles. These barriers are established when the quadrupoles freeze. Thus the quadrupolar relaxation plays the role of the alpha process, and the dipolar relaxation that of the beta process.

In a recent letter[4] a model for the relaxation, and freezing of the quadrupoles was described. The dipole relaxation was not considered. It is the purpose of this paper to develop that model further, and show that it not only accounts for the quadrupolar freezing, but also results in a set of energy barriers, which account for the dielectric relaxation phenomena.

As pointed out above, in order to obtain satisfactory agreement with experiment, previous models for the dielectric relaxation[1,2] found it necessary to make the mean activation energy, and the width of the Gaussian distribution, temperature dependent. The model for dielectric relaxation introduced here is similar in that a CN ion finds itself in a potential well that is produced by its interaction with a Gaussian distribution of neighbors. It differs from them in that no temperature dependence of the mean and width of the distribution is introduced. Instead, it is assumed that the depth of the potential well is proportional to the fraction of nearest neighbors, which are suitably oriented. This fraction is temperature dependent and is calculated using the same interaction $J$ used to model the quadrupolar freezing.

The rapid slowing of structural relaxations, which is a characteristic of glass transitions, can be explained by the interactions between the defects: as the relaxation time of one defect increases, it afffects a neighbor, which in turn influences another, and so on. But, if interactions are important, and the interaction depends on the relative configuration of the molecules, it is the net number of suitably oriented neighbors that controls the relaxation time. Now it is easy to see that if the temperature is on the order of the interaction energy the net number of neighbors must be small for relaxation to occur quickly. Therefore, those molecules locked in a particular configuration by large numbers of suitably oriented neighbors must wait for some of the neighbors to relax before they can themselves relax. The neighbors in turn will have to wait until their neighbors relax, and so on. Thus the relaxation process is hierarchical at these temperatures. Because the distribution of molecules is nonuniform, concentration gradients will connect regions of high concentration to those of lower concentration, with the result that relaxation of molecules with relatively few neighbors can relax molecules further up the gradient, and the hierarchies will consist of a series of decreasing numbers of neighbors.

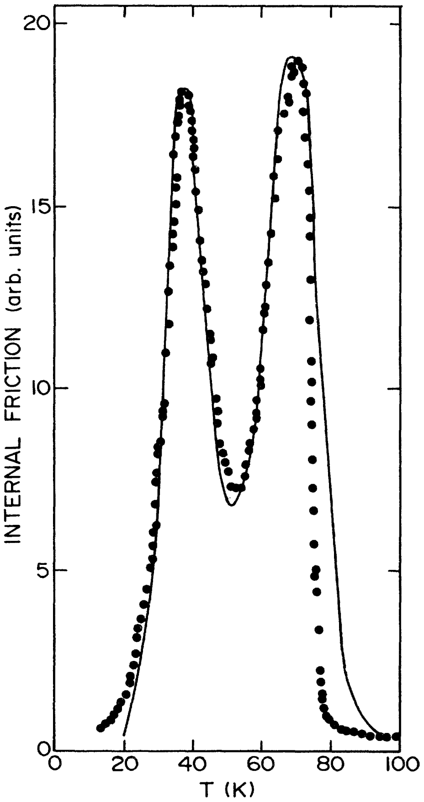

FIG. 1. The temperature dependence of the internal friction for a concentration of 0.53 determined by Knorr, Volkmann, and Loidl (Ref. 8). The solid line was calculated, as described in the text.

The relaxation time spectrum for a hierarchically relaxing system can be obtained using an approach based on the theory of Palmer *et al.*,[5] PSAA. We will use the PSAA theory in conjunction with the nearest-neighbor interactions to account for the quadrupolar freezing. We will then show that the resulting distribution of barriers accounts for the dielectric relaxation.

The relaxation of the quadrupoles is evident via their broadening of acoustic phonons[6,7] and in torsion pendulum results.[8] The torsion pendulum results, reproduced in Fig. 1, show two peaks in the temperature dependence of the internal friction. The lower temperature secondary peak has been associated with dipole relaxation,[8] while the higher-temperature primary peak is due to the quadrupoles. Ultrasonic results do not reveal the attenuation because the echoes are lost for the concentrations of interest here. However, Brillouin data clearly show a strong broadening, which is large enough to be easily measured. The width of the phonon increases to a sharp maximum[6,7] at a temperature $T_g$, which is slightly lower than that of the minimum in the velocity of sound, Figs. 2 and 3.

What is particularly interesting is how little the temperature of the quadrupole peak, $T_g$, changes for a change in frequency of seven orders of magnitude: for $x = 0.5 T_g$ is 78 K from Brillouin data,[6,7] whereas from the torsion pendulum results $T_g$ is 70 K. The dipole peak, on the other hand, moves from about 70–38 K. The data from

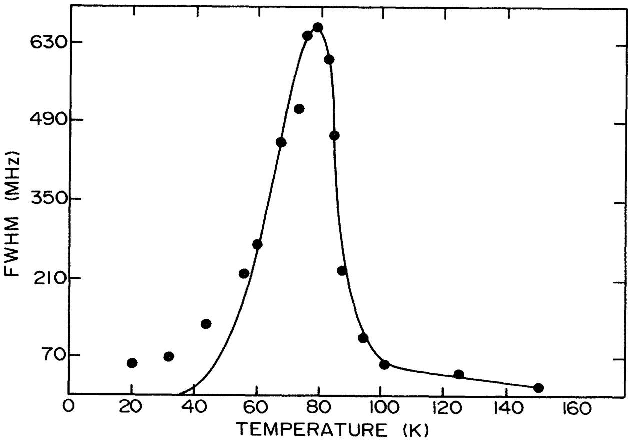

FIG. 2. The temperature dependence of the phonon width determined by Hu, Walton, and Vanderwal (Ref. 11) for a concentration of 0.5. The solid line was calculated as described in the text.

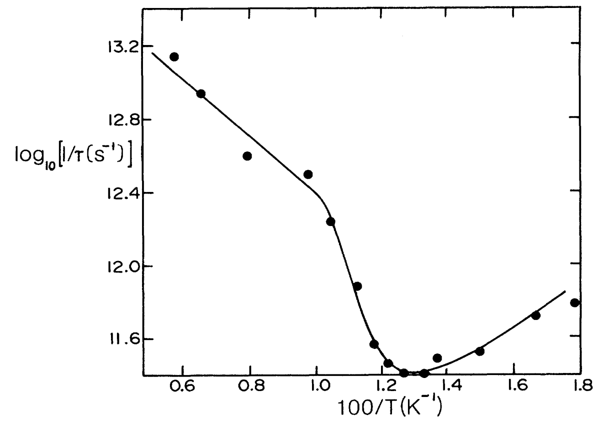

FIG. 3. The reciprocal relaxation time deduced from the data in Fig. 2 assuming a single relaxation process. The solid line is calculated as described in the text.

Ernst *et al.*,[1] reproduced in Fig. 4 also display the relatively much larger shift in the temperature of the dipole peak with frequency.

The purpose of this paper is to show that a model based on the theory of hierarchically constrained relaxation of Palmer *et al.*[5] can account for the quadrupole relaxation, and that the resulting distribution of barrier heights accounts for the dielectric relaxation.

The quadrupolar results also show a peak due to the dipolar relaxation. However, no clear evidence is available for a quadrupole peak in the dielectric data. The experimental situation is not clear in this regard, since either the data does not extend to the temperature of the peak, or it is for CN concentrations toward the low end of the glass range. Therefore, in fitting the dielectric data a possible quadrupolar peak will not be included.

The paper is organized as follows, first the theory for hierarchically constrained relaxation will be summarized. Then the experimental results will be briefly described. The analysis of the data in terms of the theory will be considered next, and finally the results of the analysis will be discussed.

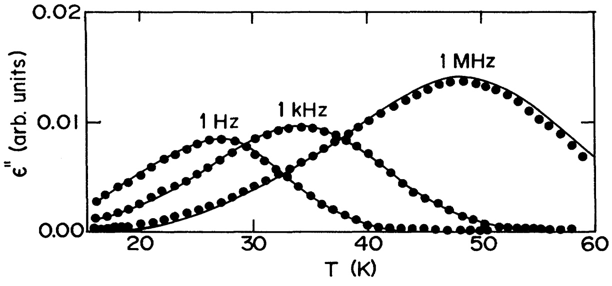

FIG. 4. The temperature dependence of the imaginary part of the dielectric constant. The data are from Ernst *et al.* (Ref. 1) and the solid line is calculated as described in the text.

## THEORY

### Quadrupolar relaxation

The elastic properties are conveniently described in terms of a complex elastic compliance $S$ such that the velocity $v$ of a sound wave of frequency $\omega$ is determined by the real part and its attenuation $\alpha$ by the imaginary part[9] of $S$:

$$1/v - i\alpha/\omega = [\rho(\mathrm{Re}S + i\mathrm{Im}S)]^{1/2}. \tag{1}$$

In general[9] the compliance S measures the strain produced when a stress is applied. There is an instantaneous response followed by a slower change, as the system relaxes.

If the system has a single relaxation time $\tau$,

$$S(t) = S_0 + \delta S[1 - e^{-t/\tau}]. \tag{2}$$

In this expression $S_0$ accounts for the instantaneous strain, and the second term for the subsequent relaxation.

In the frequency domain Eq. (2) becomes

$$S(\omega) = S_0 + \delta S/(1 - i\omega\tau), \tag{3}$$

and using Eq. (3) and values for the change in sound velocity and attenuation $\tau$ can be calculated.[9,10]

If there is a spectrum of relaxation times, Eq. (2) becomes

$$S(t) = S_0 + \sum_n \delta S_n(1 - e^{-t/\tau_n}). \tag{4}$$

In the frequency domain, the Fourier transform yields

$$S(\omega) = S_0 + \sum_n \delta S_n/(1 - i\omega\tau_n). \tag{5}$$

It is, of course, possible to replace the sum by a suitable average, and recover the expression for a single relaxation time.

$$S(\omega) = S_0 + \text{const} \times \frac{1}{1 - i\omega\tau_{\text{av}}}. \tag{6}$$

Since the velocity of sound is proportional to the real part, and the attenuation to the imaginary part, the constant can be eliminated, and $\tau_{\text{av}}$ obtained, as described above. If this is done with the data shown in Figs. 1 and 2 it is found that[11] the relaxation of the CN exhibits Arrhenius behavior at high temperatures, suggesting that a single relaxation-time approximation is valid. But at lower temperatures, just above $T_g$, it is found that the temperature dependence of the relaxation time leads to a high value for the activation energy and an unphysical magnitude for the pre-exponential, suggesting that more than one CN is involved.[11] These results are shown in Fig. 4.

Nuclear magnetic resonance has been used to obtain the relaxation time of the CN ion in NaCl-CN directly.[12] In this case the relaxation time $T_1$ is just the average discussed above.[20] The results are strikingly similar to those shown in Fig. 3, obtained from the Brillouin data.

All experimental results sample an average relaxation time. In order to compute this average correctly the relaxation time spectrum must be obtained.

### *The relaxation-time spectrum*

The PSAA theory postulates an assembly of $N$ Ising spins distributed over a number of levels such that each spin in level n+1 is only free to change its state if $\mu_n$ spins in level n attain one particular state of their $2^n$ possible ones. The relaxation times in the theory of Palmer *et al.* are related by ,

$$\tau_{n+1} = 2^{\mu_n}\tau_n \tag{7}$$

leading to

$$\tau_n = \tau_0 2^{\sum_{k=0}^{n-1}\mu_k} \tag{8}$$

and

$$S(\omega) = S_0 + \sum_n \delta S_n/(1 - i\omega\tau_0 2^{\sum_{k=0}^{n-1}\mu_k}). \tag{9}$$

The PSAA model is abstract, and does not specify any model for the levels of the system, except the requirement that spins in one level constrain spins in the level immediately above it. The CN ions constrain neighboring CN ions via their elastic strain fields; so, for a simple model it is logical to associate the levels with the number of near neighbors. This means that a CN in say, level 6, with six nearest neighbors cannot relax until $\mu_5$ CN ions in level 5 with five nearest neighbors have relaxed. In turn this implies that those spins with fewest neighbors relax first, and those with most relax last.

It will be assumed that the relaxing ions are all in hierarchies of nearest neighbors, such that the levels $1$–$n$ are all occupied. This model is very much a first approximation and, for instance, neglects any "gaps" in the hierarchy, regions of uniform concentration, the fact that hierarchies may end on a level higher than 1, and correlations between hierarchies.

The PSAA theory implies that some short-range correlation exists between the spins, i.e., that the temperature is below some ordering temperature. At high temperatures, the constraints must disappear, and a single relaxation time must be recovered.

The CN can take up various possible orientations in the unit cell, and the Ising model is not appropriate. Indeed Michel and Rowe[18] find that interacting ions are not constrained to any particular orientation. Lewis and Klein's[19] molecular dynamics calculations, show that in the glass phase there is a preference for the ⟨111⟩ directions on the average but also show substantial departures from that orientation.

In the absence of any firm experimental evidence for a particular orientation of the CN in the unit cell, this will be left as an adjustable parameter. In order to simplify the situation, only the following possibilities will be considered: the molecule could be oriented along one of the six possible ⟨110⟩ directions, and, because the molecule is not perfectly symmetrical, there may be an elastic interaction when the CN flips head to tail, leading to 12 possible configurations. Thus $\mu_n$ spins have $p^{\mu_n}$ possible states where $p$ can be 12, corresponding to orientation along a ⟨110⟩ axis, eight if the CN lies along a ⟨111⟩ axis, or six for a ⟨100⟩ orientation.

It is inconsistent to carry the relaxation process to CN ions with no nearest neighbors, so the hierarchy must end on an ion with one nearest neighbor. The first spins to relax are those in level 1, which have effectively one nearest neighbor. Their relaxation time is $\tau_1$. Next those in level 2 relax, and their relaxation time will be $\tau_2 = p^{\mu_1}\tau_1$, where $\mu_1$ is the average number of spins in level 1, which must relax for one spin in level 2 to relax, etc.

Following the argument in PSAA outlined above,

$$\tau_n = p^{\mu_{n-1}}\tau_{n-1} \tag{10}$$

and

$$\tau_n = \tau_1 p^{\sum_{k=1}^{n-1}\mu_k}. \tag{11}$$

To proceed further, the dependence of $\mu_n$ on $n$ is required. The elastic strain fields from the neighbors deepen the potential well in which the ion finds itself. Assume that when they are in one of the $p$ available configurations they will make a positive contribution to the energy barrier, whereas one of the other orientations leads to a negative contribution. Therefore, enough neighbors must take up one of their possible negative orientations to cancel the effect of the others. The simplest assumption is that both positive and negative contributions have an equal magnitude which will be equal to $J$, in which case, if half the neighbors flip they will cancel the effect of the other half, and $\mu_n = (n+1)/2$ at $T = 0$. At finite $T$ some of the neighbors will have flipped via thermal activation and this will reduce $\mu_n$, thus $\mu_n = (n+1)F(T)/2$. Also, the probability of an ion relaxing to that particular configuration will be reduced by $e^{-J/T}$, and

$$\tau_n = \tau_1[pe^{J/T}]^{\sum_{k=1}^{n-1}(k)F(T)/2}. \tag{12}$$

The simplest way to specify F(T) is with a mean-field theory. However, here we are dealing with clusters in which the concentration is manifestly nonuniform. It may be anticipated that the degree of order will increase as the number of nearest neighbors increases, i.e., towards the center of the cluster. Thus $F(T)$ must also be a function of the number of neighbors, and should be written $F(T,n)$. $F(T,n)$ can be estimated as follows.

### *The self-consistent field*

Due to the fact that we have assumed that the ions are in hierarchies of nearest neighbors, an ion with $n$ neighbors will have $n-1$ of them in level $n-1$ (i.e., with $n-1$ neighbors), and one neighbor in level n+1. On the average it will find itself in a field $B(n)$, due to its neighbors, which will be

$$B(n) = [(n-1)F(T,n-1) + F(T,n+1)]J. \qquad (13)$$

The probability that a spin will be "up" will be $P(+) = q^{-1}\exp[B(n)/T]$, where $q$ is a normalization factor. The probability that it will be "down" will be $P(-) = q^{-1}\exp[-B(n)/T]$. There are $p$ possible orientations, and the probability that it will not interact will be $P(0) = q^{-1}(p-2)$. $q$ may be determined from the fact that the sum of the probabilities must equal 1, and

$$F(T,n) = n[P(+) - P(-)] = \{2\sinh[B(n)/T]\}\{p - 2 + 2\cosh[B(n)/T]\}^{-1}. \qquad (14)$$

Given that $B(0) = 0, F(T,n)$ may be obtained by starting with an approximation to $F(T,n)$, and then refining it by iterating Eqs. (14) and (13). The process converges quite rapidly, and the final result is not at all sensitive to the initial guess.

Results of the calculation for $J = 103$ K are shown in Fig. 5 for a series of temperatures.

### *The relaxation time*

Equation (5) can be written

$$S(\omega) = S_0 + \frac{\sum_n \delta S_n}{1 - i\omega\tau_1[pe^{J/T}]^{\sum_{k=1}^{n-1}[kF(T,k)+F(T,k+2)]/2}}, \qquad (15)$$

where $F(T,k)$ is calculated numerically, as outlined in the preceding section.

It is necessary to specify $\delta S_n$: assume that each CN contributes an equal amount $C$, so that

$$\delta S_n = (N_n/N)C. \qquad (16)$$

Where $N_n$, is the number of CN ions in level $n$ (i.e., with $n$ nearest neighbors). $N_n$ is approximated by a Gaussian distribution about the most probable number of neighbors, which is equal to $12x$, where $x$ is the CN concentration.

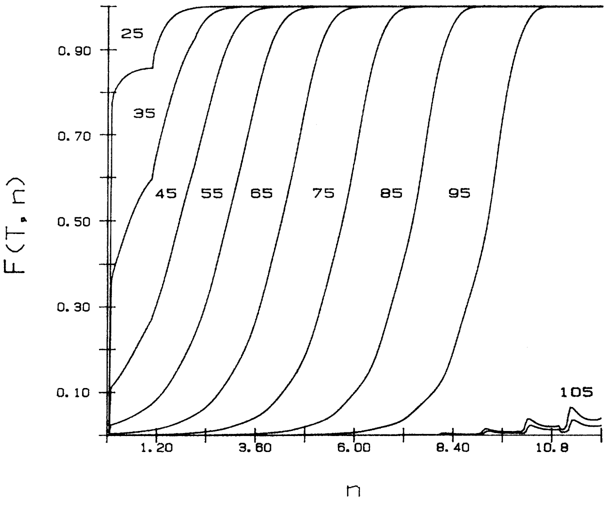

FIG. 5. The change in the order parameter $F(T,n)$ with level $n$ in a hierarchical cluster at a series of temperatures. The temperatures in K are to the left of the corresponding line, except for the line corresponding to 115 K, the line showing the lowest values at large n. The values of $F(T,n)$ were calculated numerically as described in the text.

Palmer *et al.* arguing that $n$ is large, replace the sums over $n$ by integrals. Zwanzig[13] has questioned the accuracy of this approximation; however, Palmer *et al.*, in their reply[14] argue that the integral has greater physical significance, and that the sum is only a device to introduce the model.

In our case the maximum number of nearest neighbors is not large; it is in fact 12. However, we also replace the sums by integrals: this has the effect of including the next-nearest-neighbor interactions in a very crude fashion:

$$S(\omega) = A\frac{\int_1^{12} dn\, e^{-[(n-12x)/\sigma]^2}}{1 - i\omega\tau_1[pe^{J/T}]^{\int_1^n[(x-1)F(T,x-1)+F(T,x+1)]/2}}, \qquad (17)$$

where $A$ is a constant, and $\sigma$ is the width of the distribution.

### Dielectric relaxation

Dielectric relaxation will be assumed to be due to 180° flips of the CN. If the energy barrier is taken to be due to nearest-neighbor interactions, the relaxation time will be

$$\tau_d(n) = \tau_0\exp[(n-1)F(T,n-1) + F(T,n+1)]J/T. \qquad (18)$$

$\tau_0$ is the relaxation rate in the absence of any neighbors; thus, since $\tau_1$ is the relaxation rate of an ion with one nearest neighbor, $\tau_0 = \tau_1\exp(-J/T)$.

The complex dielectric constant can be expressed as a sum of Debye equations:

$$\epsilon(\omega) = \epsilon_{\text{inf}} + (\epsilon_0 - \epsilon_{\text{inf}})\sum_n N(n)/[1 + i\omega\tau_d(n)]. \qquad (19)$$

## COMPARISON WITH EXPERIMENT

When fitting the data it was soon found that the number of possible orientations for the ion $p$ had to be as large as possible, namely 12. Lower values for $p$ resulted in too large a shift in the quadrupolar peak with frequency.

Choices for $\tau(1)$ are constrained by the high-temperature Brillouin results, and

$$\tau(1) = 10^{(-13.5\pm0.2)}\exp[(340 \pm 20 \text{ K})/T]. \qquad (20)$$

The only remaining adjustable parameters are $J$ and $\sigma$, and, of course, the ratio of the dipolar to the quadrupolar coupling constant, but the latter is simply a scaling factor and is of no interest in this calculation.

There are concentration differences between the samples used for the Brillouin experiments, the pendulum results, and the dielectric measurements. Accordingly, $\sigma$ was allowed to change with concentration, but, since the nearest-neighbor interactions would not be expected to be temperature dependent, $J$ was not. The value of $J$ that gave the best fit was 103 K.

First consider the torsion pendulum results shown in Fig. 1. The points are the data, the solid line was calculated using the imaginary parts of Eq. (19) for the secondary peak at 38 K and Eq. (17) for the primary peak at 70 K the value of $\sigma$ was 2.0.

Next consider the Brillouin data for $x = 0.5$, shown in Fig. 2. The solid line was calculated with $\sigma$ equal to 2.3.

The data in Fig. 3 are equivalent to those in Fig. 2, and the solid line was obtained with the same parameters.

Finally, the dielectric relaxation data of Ernst *et al.*[1] for a quoted concentration of 0.7 were fitted, with the results shown in Fig. 4. In this case the width $\sigma$ was 2.5. It appears that the concentrations quoted by Ernst *et al.* correspond to values in the melt, and the actual concentration would be less in the crystal. Thus the concentration was treated as an adjustable parameter and was found to be 0.43. It may be noted, for instance, that Volkmann *et al.*[14] find that the peak in the dielectric relaxation occurs at 40 K for x=0.5 and a frequency of $10^3$ Hz; so, if for the same frequency the peak occurs at 34 K, as it does in the data of Ernst *et al.*, the concentration must be lower.

## DISCUSSION

The agreement between theory and experiment shown in Figs. 1–4 is not perfect, but considering the elementary nature of the theory employed and the small number of adjustable parameters, it is far better than could be expected. What are the major approximations in the treatment presented?

The most important of these is the assumption that all the ions are in hierarchies in which the number of neighbors change by one in each shell. An improvement would be to consider a probability distribution for each shell of neighbors in the hierarchy. This would not be difficult to incorporate into the analysis, but would complicate the numerical computation.

The neglect of correlations between hierarchies is valid when the relaxation time is long on the scale of some characteristic experimental time. Unfortunately the more interesting region is close to the glass transition where this requirement is clearly not fulfilled. On the other hand the levels responsible for the correlations are the ones with numbers less than the mean, and they are not condensed, hence, no longer influencing the hierarchy at these temperatures.

The self-consistent field approach used here to describe the progressive freezing of the hierarchy neglects the effect of next-nearest neighbors. This probably means that the freezing process takes place more gradually with temperature than would otherwise be the case. This could be responsible for the fact that, as shown in Fig. 1, the experimental relaxation time above the glass transition clearly falls more quickly with temperature than the calculated one.

One of the clear results of fitting the data was that $p$ had to be equal to 12. Anything less yielded too large a temperature change with frequency for the quadrupolar relaxation peak. Another was that of the $p$ possible orientations of the CN; $p-2$ led to no interaction with a neighbor, whereas one of the remaining possibilities yielded a positive, and the other a negative interaction. A model was also tried where only a positive interaction was possible, and the remaining $p-1$ orientations yielded no interaction but was not satisfactory.

A recent paper by Hessinger and Knorr[15] quotes results for the decay of a frozen-in shear stress at various temperatures. In principle, it should be possible to account for their results with Eq. (5). However, on removing the shear stress there is an instantaneous elastic recovery [due to $S_0$ in Eq. (5)]. This can be calculated from a knowledge of the stress and values of $S_0$, but, unfortunately, the stress was not quoted.

The analysis presented here is entirely devoted to the time-dependent anelastic properties of glassy KBr-KCN alloys. As stated in the Introduction, it is believed that these are intimately connected with, and define, the glassy state. There is a corresponding effect on $S_0$, the elastic constant. This should, in principle, be combined with the temperature dependence of $S_0$ due to translational-rotational coupling between the CN rotational modes and the normal modes of the lattice. The translational-rotational coupling has been explored in detail by Michel,[16] and the elastic constants of KBr-KCN alloys are reviewed by Hochli, Knorr, and Loidl.[17]

## CONCLUSIONS

Using the theory for hierarchically constrained relaxation of Palmer *et al.*,[11] it has been possible to account for the temperature and frequency dependence of both the quadrupolar and dipolar relaxation of the CN ion in KBr for concentrations in the glass-forming range.

## ACKNOWLEDGMENTS

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