

## Low-temperature thermal expansion and specific heat of KBr containing CN

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Thermal expansion coefficients and specific heats are reported for orientationally disordered KBr samples containing CN concentrations ranging from 0.03 to 50 mol %, and within a temperature range of 0.08–10 K. With increasing CN concentration, a broad spectrum develops in the coupling parameter between the tunneling excitations and lattice strains. As with other properties reported previously, the thermal expansion for 50 mol % CN in KBr is similar to that of an amorphous solid.

### I. INTRODUCTION

We report a study of the contribution by substitutional CN molecules to the thermal expansion and specific heat of KBr:CN crystals. The work is directed to a better understanding of the low-temperature behavior of glasses and other disordered solids.

The substitution of a rod-shaped CN molecule into a KBr crystal creates a localized  $\langle 111 \rangle$ -orientational-tunneling site.<sup>1</sup> This isolated tunneling state gives rise, for example, to a Schottky peak in the specific heat near 1 K. With increasing CN concentration, however, elastic interactions between the CN ions perturb the tunneling states. One result of these interactions is that, for moderate concentrations and temperatures  $T < 10$  K, the rodlike CN freeze into random directions forming thereby an "orientational glass."<sup>2</sup> At CN concentrations greater than  $\approx 55\%$ , the CN molecules form<sup>3,4</sup> phases having long-range orientational order. The present paper is concerned only with the glassy phase at CN concentrations  $\leq 50$  mol %.

An interesting feature of this orientational glass is that the low-temperature properties<sup>5,6</sup> are similar to those found in amorphous solids.<sup>7</sup> For example, the specific heat is essentially linear in  $T$ , the thermal conductivity is quadratic in  $T$ , and the dielectric susceptibility exhibits a frequency-dependent minimum near 0.1 K. In KBr:CN, first suggestions of these features appear near a CN concentration of  $\approx 2$  mol % and become fully developed at 50 mol %. In amorphous solids the low-temperature behavior is ascribed to two-level tunneling states (TLS) having a broad spectrum of excitation energies  $E$ , although there is no widely accepted microscopic model for the TLS. The controllable development of TLS in crystals such as KBr:CN, however, should be more amenable to a theoretical understanding. Some effort has been directed to this problem.<sup>8,9</sup>

The tunneling states in amorphous solids, and in KBr:CN, experience a strong elastic interaction with their environment. As a convenient representation of this interaction, we will use the deformation potential<sup>7</sup>  $D = dE/de$  where  $e$  is strain.<sup>10</sup> Evidence that  $D$  is large in amorphous solids is found in measurements of ultrasonic attenuation ( $\propto D^2$ ) and thermal conductivity

( $\propto D^{-2}$ ). Yet the measured thermal-expansion coefficient ( $\propto D$ ) of glasses is small.<sup>11</sup> This difference (small  $D$ , but large  $D^2$ ) is explained if there occurs a broad distribution<sup>12</sup> in  $D$  which is centered near  $D=0$ . If  $p(D)$  represents this distribution, then  $\langle D \rangle = \int Dp(D)dD$  can be small while  $\langle D^2 \rangle = \int D^2p(D)dD$  remains large. Therefore the TLS in amorphous solids occur with broad spectra both in energy  $E$  and in strain coupling  $D$ .

It has been possible, through specific-heat measurements on KBr:CN, to observe the broad spectrum in  $E$  develop as the CN concentration is increased.<sup>5,6</sup> In the present paper we report the development of a spectrum in  $D$  with increasing CN concentration.

### II. EXPERIMENTAL TECHNIQUE AND RESULTS

One sample of KBr containing 0.034 mol % CN was grown and provided by S. Susman of the Argonne Nation-

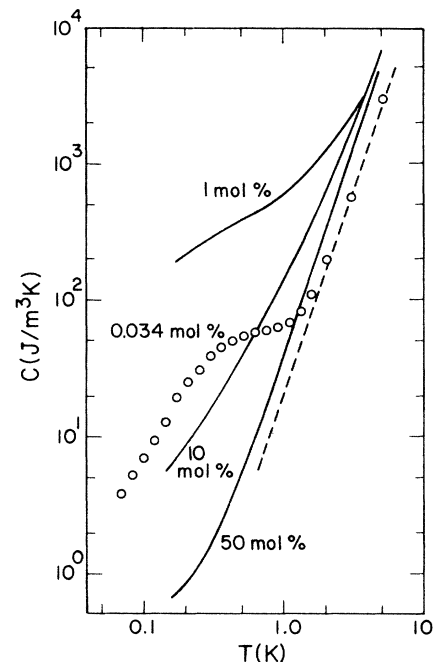


FIG. 1. Specific heat  $C$  of KBr:CN versus temperature  $T$  for four concentrations of CN. Data for the solid lines were obtained from Ref. 6. The dashed line represents the  $T^3$  Debye phonon contribution for the 0.034 mol % CN sample.

al Laboratory. Information concerning this sample may be found in Ref. 13. A significant fact is that no unwanted impurities were detected in this sample other than OCN, which was present at less than  $10^{-4}$  ppm. The 1, 10, and 50 mol % CN samples were those used in previous low-temperature measurements and are described in Ref. 6. They had been purchased from the Crystal Growth Laboratory of the University of Utah.

In measuring the specific heat, the sample was clamped between dry sapphire plates to provide a long equilibration time ( $\approx 10$  sec) between sample and refrigerator. A  $\approx 5$ -msec heat pulse  $Q$  was applied via an electrical heater, and the temperature excursion  $\Delta T$  was measured with a carbon-resistance thermometer. The signal-to-noise ratio was enhanced by a signal-averaging technique.<sup>14</sup> The specific heat was then computed using the relation  $C = Q/(V\Delta T)$ , where  $V$  is volume. The addenda heat capacity was negligible ( $< 1\%$ ).

The carbon-composition resistance thermometers used for the specific-heat (and thermal-expansion) measurements were calibrated *in situ* against three germanium resistance thermometers (for reliability). The germanium thermometers had been calibrated to an accuracy better than 1% on the EPT76 temperature scale against National Bureau of Standards superconducting fixed points using a cerium magnesium nitrate magnetic thermometer for interpolation. The absolute accuracy of a specific-heat measurement was  $\approx 5\%$ .

The total specific heats for the four CN concentrations are shown in Fig. 1. The dashed line represents the  $T^3$  phonon contribution to the specific heat of the 0.034 mol % CN sample. The corresponding Debye temperature is  $165 \pm 4$  K. Since 0.034 mol % CN should have little effect on the Debye temperature, this value may be compared with an earlier measurement<sup>5</sup> of 160 K for pure KBr, and with a value of 171 K computed from the elastic constants for pure KBr. An apparent  $T^3$  regime is also present for the other three samples of Fig. 1. However, the elastic constants at  $T=0$  are not known accurately, so it is not certain as yet that this  $T^3$  term arises entirely from thermal phonons, especially for the 50 mol %

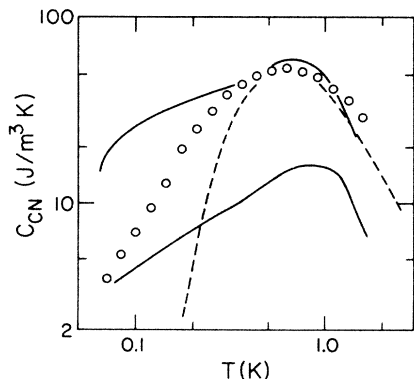


FIG. 2. The contribution  $C_{CN}$  by CN molecules to the specific heat of the 0.034 mol % CN sample. Circles represent present data. The solid curves are from Ref. 15 for 0.043 (top) and 0.021 mol % (bottom) CN concentration. The dashed curve represents a Schottky specific heat computed for a 1-3-3-1 tunneling state, see text.

CN sample.<sup>6</sup>

Subtracting the Debye phonon contribution from the total specific heat for the 0.034 mol % sample provides the contribution,  $C_{CN}$ , from the CN molecules alone. This contribution is shown in Fig. 2 along with earlier data.<sup>15</sup> The present data exhibit a more narrow peak than the earlier data, possibly indicating a smaller concentration of lattice defects or unwanted impurities in the present sample.

The linear thermal expansion was measured using a dilatometer incorporating a superconducting quantum interference device (SQUID) detector. The dilatometer, which has been described previously,<sup>16</sup> was improved by the replacement of the earlier quartz-disk transducer with a small piezoelectric ceramic tube.<sup>17</sup> This change considerably improved the reproducibility of the transducer package which retains the calibration of the dilatometer between runs. The transducer was calibrated periodically in separate runs against the known thermal expansion of high-purity copper at  $T \geq 2$  K. Since the output of this transducer is independent of temperature, the calibration could be reliably transferred to lower temperatures. The absolute accuracy<sup>18</sup> was still limited by the calibration procedure to  $\approx 10\%$ . Within a given run, the relative accuracy was 5%. All thermal-expansion data were measured along a  $\langle 100 \rangle$  crystalline axis.

The internal equilibrium time at  $T < 1$  K for the 1 mol % CN samples was an order of magnitude longer than for other concentrations. Also the response to a heat pulse was nonexponential in character which gave rise to a time-dependent specific heat<sup>6</sup> as well as to a time-dependent thermal expansion. For the 1 mol % sample, the thermal expansion was measured over a time interval

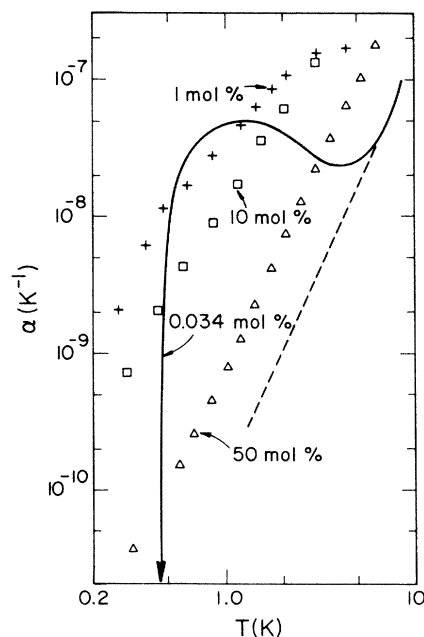


FIG. 3. Linear thermal expansion coefficient  $\alpha$  of KBr:CN vs  $T$  for four concentrations of CN. The solid line represents data for the 0.034 mol % CN sample, see also Fig. 4. The dashed line is the  $T^3$  phonon contribution for the 0.034 mol % sample.

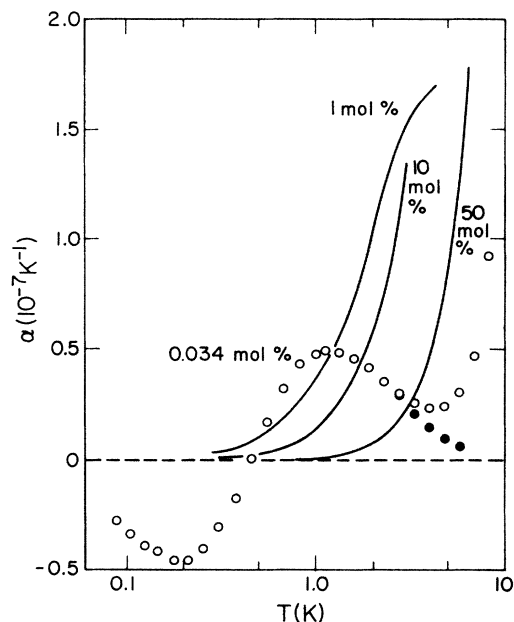


FIG. 4. The expansion coefficient  $\alpha$  for KBr:CN plotted on a linear vertical scale so that negative data below 0.5 K may be included. The filled symbols at  $T > 3$  K indicate the CN contribution to the expansion after the phonon contribution has been subtracted.

of 64 sec, following a change in temperature of  $\leq 10\%$ , at which time the expansion of the sample had become essentially constant. Likewise the specific-heat values were obtained long after each heat pulse was applied.<sup>6</sup> Therefore our measurements provide the "total" thermal-expansion coefficient or specific heat.

For convenient comparison with the specific-heat data of Fig. 1, the linear thermal-expansion coefficients are shown on a log-log plot in Fig. 3. However, since the expansion of the 0.034 mol % sample is negative below 0.45 K, a linear vertical scale is used in Fig. 4 so as to include

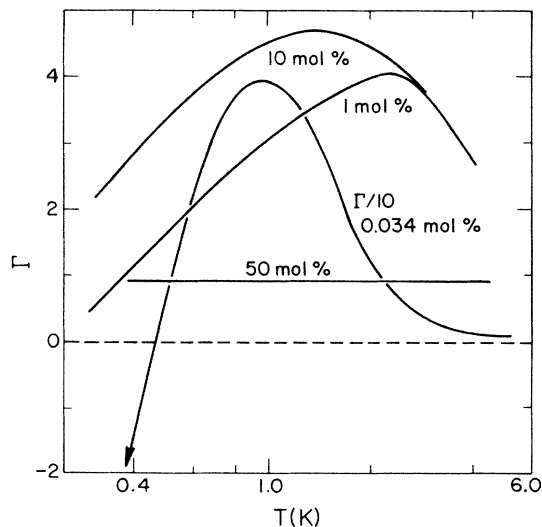


FIG. 5. Grüneisen parameters  $\Gamma$  for KBr:CN vs  $T$ . Note that  $\Gamma$  for the 0.034 mol % CN sample has been divided by a factor of 10. The  $\Gamma$  of the 50 mol % sample is influenced by tunneling states at least for  $T < 0.5$  K.

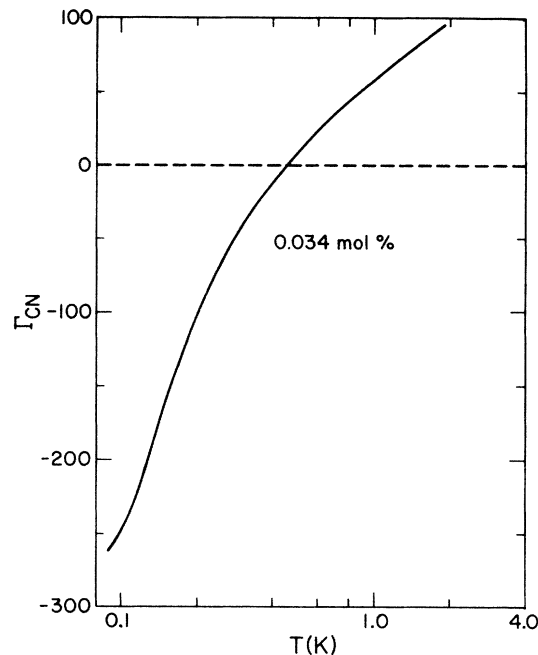


FIG. 6. The Grüneisen parameter  $\Gamma_{\text{CN}}$  for the 0.034 mol % sample.  $\Gamma_{\text{CN}}$  is related only to the CN tunneling states.

all data. In Fig. 3, the dashed line represents the  $T^3$  phonon contribution to the thermal expansion coefficient of the 0.034 mol % sample. As with the specific-heat data, we cannot be certain what the phonon contribution is for the other three samples. Subtraction of the phonon contribution for the 0.034 mol % sample produces the filled circles in Fig. 4 at  $T > 3$  K. At lower temperatures the CN contribution dominates the thermal expansion of this sample.

In a discussion of thermal expansion, it can be more informative to focus on the average expansion contributed by each excitation. This normalized expansion is conveniently represented by the Grüneisen parameter  $\Gamma$  given by an equation of the form<sup>19</sup>  $\Gamma = 3\alpha B/C$ , where  $B$  is the bulk modulus of the crystal.<sup>20,21</sup>

The Grüneisen parameters obtained from the measured specific heat and thermal expansion are shown in Fig. 5. Note that the line representing the 0.034 mol % sample has been divided by a factor of 10 on this graph. For this 0.034 mol % sample the total Grüneisen parameter may be separated into two parts: that contributed by thermal phonons alone ( $\Gamma_D = 3B\alpha_D/C_D$ ) and that contributed by the CN ions ( $\Gamma_{\text{CN}}$ ). Using the data of Figs. 1 and 3 for  $C_D$  and  $\alpha_D$  gives  $\Gamma_D = +0.34 \pm 0.05$ , which is in agreement with an earlier value<sup>22</sup> of  $\Gamma_D = +0.29 \pm 0.03$  found for pure KBr. The Grüneisen parameter  $\Gamma_{\text{CN}}$  related to the tunneling CN molecules in the 0.034 mol % sample is shown in Fig. 6. The parameter  $\Gamma_{\text{CN}}$  is remarkably temperature dependent, ranging from  $-250$  at 0.1 K to  $+100$  at 2 K.

### III. DISCUSSION

The intent of the above measurements was to obtain the thermal expansion coefficient and Grüneisen parameter associated with isolated CN sites, then monitor the

changes that occur as the CN-CN interactions are increased, with increasing CN concentration, until glasslike behavior is obtained.

The influence of CN-CN interactions as a function of CN concentration can be seen in Fig. 7. This figure shows estimates for those portions of thermal properties contributed by CN alone. The + symbol represents  $C_{CN}$ , the difference between the total specific heat and the phonon contribution estimated from ultrasonic velocities. The  $\circ$  symbols represent  $\alpha_{CN}$ , the difference between the measured expansion and the phonon contribution estimated by assuming  $\Gamma_D$  is independent of CN concentration. The  $\times$  symbols represent the inverse phonon mean free path deduced from thermal-conductivity measurements.

All data<sup>5,6,15,20,23</sup> in Fig. 7 were obtained at 1 K. This observational "window" at 1 K monitors the energy  $\Delta_0$  of isolated CN sites in the dilute limit. Ions having strong CN-CN interactions are removed from this observational window. Therefore the magnitude of these thermal properties at 1 K should be proportional to the concentration of those CN molecules which have little or no interactions with other CN molecules. For low CN concentrations, this magnitude is simply proportional to the concentration and is represented by the solid line in Fig. 7. The dashed line indicates the behavior expected if CN-CN neighbors separated by  $\leq 10$  Å interact strongly enough to exclude them from this observational window. Only CN sites which have no neighbors within  $\approx 10$  Å are counted here. It will be noted that the excess specific heat (+ symbol) roughly follows this dashed line. Near a concentration of  $x = 0.1$ , for example, about 2% of the CN continue to have  $\Delta_0 \approx 1$  K. On the other hand,  $l^{-1}$  appears to fall away from the solid line somewhat more rapidly than  $C_{CN}$ , while  $\alpha_{CN}$  is strongly suppressed even at  $x = 10^{-2}$ .

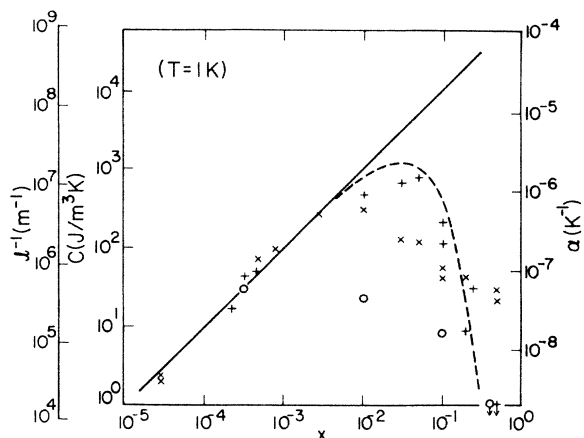


FIG. 7. Estimate of that portion of the specific heat (+), thermal expansion ( $\circ$ ), and inverse phonon mean free path ( $l^{-1}$ , obtained from thermal conductivity) ( $\times$ ) contributed by CN tunneling states having an energy near 1 K vs CN concentration  $x$ . Data are from Figs. 1 and 3, and from the literature. The solid line is the expected behavior with no CN-CN interactions present. The scales for  $C_{CN}$ ,  $\alpha_{CN}$ , and  $l^{-1}$  were shifted vertically so as to agree with this line for small  $x$ . It is likely that the expansion data should lie even lower on this figure. The dashed line represents a possible behavior for finite CN-CN interactions.

In brief, while the specific-heat data show that the CN-CN interactions do indeed reduce the density of tunneling states having energies  $\approx 1$  K, the thermal-expansion and conductivity data indicate that these interactions influence the deformation potential even more profoundly.

We now turn to the sample having the smallest CN concentration, 0.034 mol %. As mentioned in Sec. I, at small concentrations the CN behave as isolated  $\langle 111 \rangle$  tunneling sites. A simple computation suggests that the tunnel-split ground state should have four equally-spaced energy levels with degeneracies of 1-3-3-1. However, the specific heat contributed by the CN for a concentration of 0.034 mol % is broader than expected for a 1-3-3-1 Schottky peak. The dashed line in Fig. 2 represents a 1-3-3-1 Schottky peak for an energy splitting of  $\Delta_0 = 1.5$  K and, at 0.1 K, for example, the measured  $C_{CN}$  is larger than the computed value by a factor of  $\approx 50$ . Either an isolated CN ion in KBr should not be represented by a simple 1-3-3-1 tunneling state, or some fraction of the CN sites interact with neighboring defects. A similar problem is encountered with the thermal-expansion results for the 0.034 mol % sample, which are presented most conveniently in Fig. 6. A simple  $\langle 111 \rangle$  tunneling state (such as an isolated Li in KCl) gives rise to a temperature-independent Grüneisen parameter,<sup>24</sup> whereas that for CN in Fig. 6 is strongly temperature dependent. This behavior again could be explained in two ways. Either the thermal expansion of Fig. 6 is indicative of isolated CN ions which should not be represented as simple 1-3-3-1 tunneling states, or the positive expansion near 1 K is related to isolated CN ions whereas the negative expansion near 0.2 K is contributed by CN sites interacting with neighboring defects.

One obvious source of interactions is with neighboring CN ions. Figure 7 suggests that CN-CN interactions should not appreciably perturb the specific heat of a 0.034 mol % CN sample, but might influence the thermal expansion. If the broad peak in the measured specific heat, Fig. 2, is attributed to CN-CN interactions, a rough estimate of its magnitude can be calculated as in the case<sup>24</sup> of Li-Li interactions in KCl. Since the CN concentration is small for Fig. 2, it may be assumed that pair interactions dominate. That is, the elastic interaction energy is of the form  $U \propto R^{-3}$ , where  $R$  is the separation between the two CN sites. The specific heat of a 1-3-3-1 tunneling state is identical to that of a properly normalized two-level state. Therefore we approximate the levels of two interacting CN ions as an appropriately weighted set of two-level states.<sup>17</sup> The tunneling splitting is then of the form<sup>25</sup>  $\Delta = [(U^2 + 4\Delta_0^2)^{1/2} \pm U]/2$ , where  $\Delta_0$  is the splitting in the absence of interactions. It is further assumed that the probability distribution in  $U$  is governed by the random siting of CN ions on the KBr lattice. The net specific heat is therefore a spectrum of Schottky peaks having energies  $\Delta$ . Summation of this spectrum<sup>17</sup> gives the solid line in Fig. 8. The observed temperature dependence near 0.1 K has been reproduced.

Three adjustable parameters have been employed in Fig. 8. One is the CN concentration  $n$ . The fit of Fig. 8 requires  $n = 320$  ppm which is close to the values  $n = 340$  ppm reported from spectrochemical analysis,<sup>13</sup> and

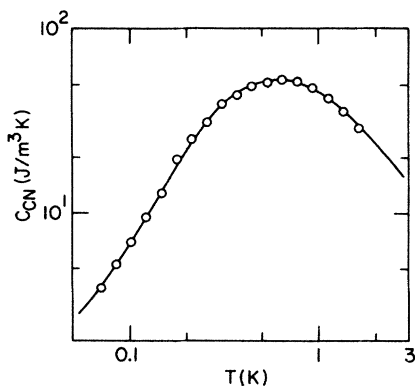


FIG. 8. Specific heat contributed by CN ions in the 0.034 mol % sample. The solid line represents a computation, discussed in the text, involving CN-CN interactions.

$n = 300$  ppm estimated from the measured specific heat using  $nk(\ln 8) = \int (C_{\text{CN}}/T)dT$ . The second parameter is  $\Delta_0 = 1.30$  K, which agrees with that obtained from infrared measurements.<sup>26</sup> The third parameter is  $U_0 = U(R=a) = 420$  K, where  $a$  is the Br-Br distance in KBr. This value for  $U_0$  is an order of magnitude larger than other estimates.<sup>27,28</sup> Therefore, if defect interactions are responsible for the broad peak of Figs. 2 or 8, either our crude assumptions concerning CN-CN pair interactions are inadequate, or some other unknown but randomly sited defect must be broadening the Schottky peak in the 0.034 mol % sample. It was noted in Sec. II, however, that this sample was of high purity. In addition, if defect interactions were responsible for the unusual temperature dependence of  $\Gamma_{\text{CN}}$  in Fig. 6, then it might be expected that the 1 mol % CN sample would exhibit an even stronger negative expansion near 0.1 K, which it does not. In brief, we find no convincing evidence that the thermal properties of the 0.034 mol % CN sample are in fact strongly perturbed by defect interactions.

On the other hand, if the broad peak in Fig. 8 or the strong temperature dependence of Fig. 6 are ascribed to a tunneling state other than a simple 1-3-3-1, the lower-energy tunneling levels of the ground-state manifold must respond to pressure in a very different manner from the higher levels in this manifold to produce the behavior found in Fig. 6. Perhaps this effect could arise because the top of the manifold (at an energy of  $\approx 5$  K) is rather close to the top of the tunneling barrier at 36 K. For Li in KCl by contrast, the tunneling states lie deeper in the  $\langle 111 \rangle$  potential-energy wells. It is clear that, unfortunately, we cannot be certain if the thermal expansion of our low-concentration sample exhibits behavior representative of an isolated CN, or includes a significant contribution from CN interacting with neighboring defects.

As the CN concentration approaches 50 mol %, the behavior of the thermal expansion becomes similar to that of a glass. That is,  $(\langle D^2 \rangle)^{1/2}$  remains large,  $\approx 100$ –200 K, as deduced from the thermal conductivity,<sup>10,29</sup> while  $\alpha$  and therefore  $\langle D \rangle$  become small with  $\langle D \rangle \lesssim 1$  K for the 50 mol % CN sample.<sup>30</sup> These facts indicate a broad distribution in  $D$  centered near  $D = 0$ . Therefore, as the tem-

perature rises, half of the excitations which appear in the specific heat expand the lattice while half allow the lattice to contract. Any model of the CN-related excitations, or TLS, in the orientational glass phase of KBr:CN must ultimately explain the development of this broad spectrum in strain coupling  $D$  as well as the broad spectrum in excitation energy  $E$ . The proposed mean-field model<sup>8</sup> does not provide this detail.

Within the tunneling-states model of a glass, the deformation potential is defined as

$$D = dE/de = (\xi/E)(d\xi/de) + (\Delta/E)(d\Delta/de).$$

Here  $E = (\Delta^2 + \xi^2)^{1/2}$ , where  $\Delta$  is again the tunnel splitting and  $\xi$  is the asymmetry of the potential-energy double well.<sup>7</sup> For glasses, the term  $d\Delta/de$  is negligible<sup>31</sup> whereas, for isolated CN in KBr, *only*  $d\Delta/de$  is relevant provided the crystal remains cubic.<sup>32</sup> Therefore, if the orientationally disordered phase of KBr:CN is indeed analogous to a glass, then a term proportional to  $d\xi/de$  must come to dominate  $D$  with increasing CN concentration. In this case, the observed similarity in magnitude<sup>29</sup> between  $\langle D^2 \rangle$  in dilute KBr:CN and  $\langle D^2 \rangle$  at 50 mol % CN concentration, as deduced from thermal conductivity measurements, would be coincidental.

In conclusion, it has been shown that the average coupling constant  $\langle D \rangle$ , between lattice strains and the low-energy excitations associated with CN molecules in KBr, decreases by a factor of  $\leq 100$  as the CN concentration increases to 50 mol % in this orientationally disordered solid. A more quantitative statement is not possible because of the unusual temperature dependence of the thermal expansion coefficient in the 0.034 mol % sample. It was noted that the specific heat of this sample also has an unexpected temperature dependence. These anomalies may be related either to CN interactions with neighboring defects or to the shallow nature of the CN tunneling site. Although there is a large reduction in  $\langle D \rangle$  with increasing CN concentration, thermal conductivity measurements show that the magnitude of  $\langle D^2 \rangle$  is roughly concentration independent. This indicates that, as the CN concentration increases, a broad spectrum in  $D$  develops concurrently with the development of the broad spectrum of energy splittings seen in specific-heat measurements. At 50 mol % CN in KBr, the thermal expansion is, as for other properties, similar to that of an amorphous solid. These results provide an additional test of any microscopic model proposed for the low-energy excitations in KBr:CN.

#### ACKNOWLEDGMENTS

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