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Effect of Force-Constant Changes on the Lattice Specific Heat of KCl:Tl

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A detailed study of the effects of force-constant changes produced by noninteracting Tl^+ impurity ions on the lattice specific heat of KCl has been made. Numerical computations have been performed for three impurity concentrations, 1, 3, and 5 mole % of Tl^+, and the results are compared with the experimental data of Karlsson. The effects of force-constant changes on the specific heat are seen to be quite large. The temperature dependence of the observed specific heat has been explained successfully by Green's-function theory in the temperature range 1–10 °K. The force-constant change due to Tl^+ in KCl is seen to be small and the impurity behaves essentially as a mass defect. Finally, the low-concentration theory is seen to be adequate for understanding the experimental results even at relatively higher concentration of impurities (5 mole % of Tl^+) present in a crystal.

I. INTRODUCTION

Quite striking changes in the phonon frequency spectrum of a crystal are obtained by introducing a finite concentration of impurities. The periodic symmetry of the crystal lattice is destroyed by the point defect. The normal modes of perfect crystal are thus modified. The modifications of the phonons are naturally large in the neighborhood of the defect. Essentially two new types of impurity modes may appear. The modes whose frequencies lie outside the range of those of the host crystal are called localized modes. This type of impurity mode is localized in the vicinity of the defect. The localized modes may appear owing to very light impurities such as *U* centers (H^-) and/or owing to impurities strongly bound to the host lattice. Another type of impurity mode is a resonance mode. The frequencies of these modes lie in the range of the phonon frequencies of the perfect lattice and they are characterized by a considerable enhancement of the amplitude over a narrow range of frequencies. The resonance modes may occur owing to the presence of very heavy impurities and/or owing to impurities interacting very weakly with the host lattice. Recently, detailed studies of impurity modes have been made by infrared-lattice-absorption,[1-3] inelastic-neutron-scattering,[4-8] and first-order Raman-scattering[9,10] experiments.

One of the most important properties of a crystal where the frequency spectrum plays a central role is the lattice specific heat. At low temperatures the high-frequency localized modes are not excited. At high temperatures a significant number of local modes are excited but all other phonons of the lattice are also excited. Thus the local modes will induce a relatively small change in the large total specific heat and the possibility of observing local modes in specific-heat measurements is very small. On the other hand, low-frequency resonance modes get excited even at low temperatures and can be observed in specific-heat experiments. Lehman and De Wames[11] and independently Kagan and Iosilevskii[12] have shown that the contribution of resonance modes to the lattice specific heat may be appreciable at low temperatures. Some numerical estimates for isotopic impurities have also been made by later groups of workers. Quite recently some combined experimental and theoretical studies[13-15] have been made for the temperature dependence of the lattice specific heat of crystal containing small concentrations of impurities.

Karlsson[13] has measured the specific heat of KCl crystals doped with 1-, 3-, and 5-mole% TlCl in the temperature range 1–30 °K. He has discussed the experimental results on the basis of a theory of Kagan and Iosilevskii[12] using a Debye frequency spectrum for the host lattice. He obtained reasonable fits in the temperature range 5–10 °K, but the agreement was poor below 5 °K. In Kagan and Iosilevskii's theory one assumes that the lattice is monatomic, which is certainly not true in the case of KCl. Further, in KCl there exist long-range Couloumbic and dipolar forces and, therefore, the use of a Debye theory may not be reliable.

In the present work we have determined the contribution of point defects to the lattice specific heat of a crystal having rock-salt structure. In the calculations we have employed realistic models both for the defect perturbation and the host KCl crystal. In the perturbation model mass change as well as nearest-neighbor central-force-constant change are considered. The results of the lattice dynamics of KCl in the deformation dipole model have been used in the calculations. Numerical computations for specific heat have been performed for the three impurity systems of KCl : Tl^+ containing 1, 3, and 5 mole% of Tl^+ ion experimentally studied by Karlsson.

In Sec. II A we briefly describe the Green's-function theory for the change in the lattice specific heat due to impurities in a crystal in the low-concentration limit. In Sec. II B, the nearest-neighbor perturbation model for the point defect is discussed. In Sec. III, we calculate the enhancement in the specific heat and also discuss the results based on an effective force constant. In Sec. IV the results are discussed and summarized.

II. THEORY

A. Lattice Specific Heat

Owing to the presence of impurities the enhancement of the vibrational specific heat of a crystal (per mole) at temperature T, can be expressed as

$$\Delta C_L(T) = \frac{\hbar^2}{4k_B T^2} \int_0^\infty \omega^2 \Delta N(\omega)\,\mathrm{csch}^2(\hbar\omega \,|\, 2k_B T)\,d\omega\ , \tag{1}$$

where k_B is Boltzmann's constant, ω is the phonon frequency, and other symbols have their usual meanings. $\Delta N(\omega)$ denotes the change in the phonon density of states $N(\omega)$ which is defined as the number of normal modes in the interval ω and $\omega + d\omega$ in the limit as $d\omega \to 0$. $N(\omega)$ is related to a similarly defined function $N(\omega)^2$, which is a more common function in the theory of crystal-lattice dynamics, as

$$N(\omega) = 2\omega N(\omega^2)\ .$$

The evaluation of $\Delta N(\omega^2)$ is very difficult for a general defect, but the problem is tractable if the perturbation caused by a defect exhibits some symmetry. For instance, one observes a point-group symmetry of the host lattice for a point defect substituted at a lattice site and considers only the impurity space of dimension $3b \times 3b$ where b is the total number of ions directly affected by a defect, including itself. A group-theoretical analysis may be performed for the change in the phonon density of states in such cases. For a crystal containing a single defect one may therefore write

$$\Delta N(\omega^2) = \sum_\nu \Delta N_\nu(\omega^2)\ , \tag{2}$$

where $\Delta N_\nu(\omega^2)$ is the contribution made by the symmetric motion ν and is given by

$$\Delta N_\nu(\omega^2) = -\frac{l_\nu}{\pi}\,\mathrm{Im}\left(\frac{1}{D_\nu(z)}\,\frac{dD_\nu(z)}{d\omega^2}\right)\ . \tag{3}$$

Here l_ν is the dimension of the irreducible representation ν, $z = \omega^2 + 2i\omega\xi$ is the complex squared frequency in the limit $\xi \to 0$, and $D_\nu(z)$ is the resonance denominator corresponding to the irreducible representation ν. It is defined by

$$D_\nu(z) = \left| \underline{\mathrm{I}} + \underline{\mathrm{g}}_\nu(z)\underline{\mathrm{P}}_\nu(\omega^2) \right|\ , \tag{4}$$

where $\underline{\mathrm{I}}$ is the unit matrix, $\underline{\mathrm{P}}_\nu(\omega^2)$ and $\underline{\mathrm{g}}_\nu(z)$ are the perturbation and Green's-function matrices projected onto the subspace of irreducible representation ν.

The frequency of an impurity mode, i.e., localized or resonance mode, in the symmetric motion ν may be determined by the condition

$$\mathrm{Re}D_\nu(z) = 0\ . \tag{5}$$

After introducing phase shifts defined by

$$\tan\delta_\nu = -\,\mathrm{Im}D_\nu(z)/\mathrm{Re}D_\nu(z)\ , \tag{6}$$

the contribution of modes of symmetry ν towards the change in the density of states can be written

$$\Delta N_\nu(\omega^2) = \frac{l_\nu}{\pi}\,\frac{d\delta_\nu}{d\omega^2}\ . \tag{7}$$

Substituting for $\Delta N(\omega)$ in Eq. (1) and integrating once by parts, the change in specific heat due to a single defect can be written

$$\Delta C_L(T) = \sum_\nu \Delta C_L^\nu(T)\ , \tag{8}$$

where the contribution of each irreducible representation is given by

$$\Delta C_L^\nu(T) = -\frac{2k_B B'^2}{3N\pi} \int_0^\infty \delta_\nu \omega\,\mathrm{csch}^2(B'\omega) \times [1 - B'\omega\coth(B'\omega)]\,d\omega\ , \tag{9}$$

where $B' = \hbar/2k_BT$ and N is the number of unit cells in a crystal.

In the low-concentration limit we assume that the impurities do not interact among themselves and, therefore, the change in the density of states that is due to a small concentration of impurities in a crystal may be determined by multiplying the change due to a single defect by the impurity concentration. The enhancement of specific heat due to a fractional concentration c of point defect is, thus, given by

$$\Delta^{\text{total}} C_L(T) = cN\Delta C_L(T) . \qquad (10)$$

B. Perturbation Model

Except for the cases of defects with off-center configurations or molecular impurities, the induced perturbation retains the full cubic point symmetry and both the matrices $\underline{\mathrm{P}}(\omega^2)$ and $\underline{\mathrm{g}}_0(z)$ can be easily diagonalized into blocks belonging to the irreducible representations A_{1g} (one dimensional), E_g (two dimensional), and F_{1u} (three dimensional) of the O_h point group. The nearest-neighbor perturbation model for a substitutional ion in a NaCl-type lattice has been described in previous works.[2,16,17] The resonance denominators for a cation impurity are seen to be

$$D_{A_{1g}}(z) = 1 + 2\lambda\chi(g_4 + 2g_5) , \qquad (11)$$

$$D_{E_g}(z) = 1 + 2\lambda\chi(g_4 - g_5) , \qquad (12)$$

and

$$D_{F_{1u}}(z) = 1 - \epsilon\omega^2 g_1 + 2\lambda(g_1 + \chi g_3 - 2\chi^{1/2} g_2) - 2\epsilon\omega^2\lambda\chi(g_1 g_3 - g_2^2) , \qquad (13)$$

where $\chi = M_+/M_-$ is the ratio of the masses of two ions of an unit cell, λ is the change in the mass-reduced nearest-neighbor central-force constant, and ϵ is the mass change parameter equal to $(M_+' - M_+)/M_+$. M_+' is the mass of the thallium ion.

If we define an effective force constant η, introduced earlier[18] in the framework of a nearest-neighbor rigid-ion model for a cubic crystal lattice, the resononce denominator in the F_{1u} irreducible representation simplifies to

$$D_{F_{1u}}(z) = (1 + \lambda/\eta)(1 - \epsilon\omega^2 g_1) + (3\lambda/\eta)(1 + \epsilon)(\omega^2/Z\eta)(1 + \omega^2 g_1) , \qquad (14)$$

where Z is the number of nearest neighbors of a lattice site and is equal to six for rocksalt structure. The various Green's functions appearing in Eqs. (11)-(14) are given in Ref. 19.

III. NUMERICAL COMPUTATIONS AND RESULTS

Equation (10) has been employed for calculating the change in the lattice specific heat of KCl doped with Tl^+ ions in the temperature range 1-30 °K. The force-constant change λ was taken as a parameter to obtain best fits with the experimental data of Karlsson at the three impurity concentrations 1, 3, and 5 mole% of Tl^+. In the calculation we have used the values of the five Green's function computed earlier by Benedek and Nardelli[19] in Hardy's deformation-dipole model for KCl at 4096 points in the first Brillouin zone.

In Figs. 1-3 we present the theoretical results for two different values of λ, i.e., $\lambda = -0.204 \times 10^{26}$ sec^{-2} and -0.408×10^{26} sec^{-2}, along with the experimental data. In these figures we have also shown results in the mass-defect approximation. We observe that a decrease in the nearest-neighbor force constant due to the impurity increases the specific heat while the reverse is true for an enhanced force constant. This result is in agreement with that drawn earlier by one of the present authors[20] in a scalar lattice model. In the present calculation a 5% change in force constant (see below) produces approximately a change of ~ 20% in the enhanced specific heat. An over-all good agreement with experiment takes place for $\lambda = -0.204 \times 10^{26}$ sec^{-2} at all the three impurity concentrations. However, in the case of 3-mole% Tl^+, the agreement is seen to be better for $\lambda = -0.408 \times 10^{26}$ sec^{-2}.

In Fig. 4, we have plotted the real part of the resonance denominator for F_{1u} symmetry modes

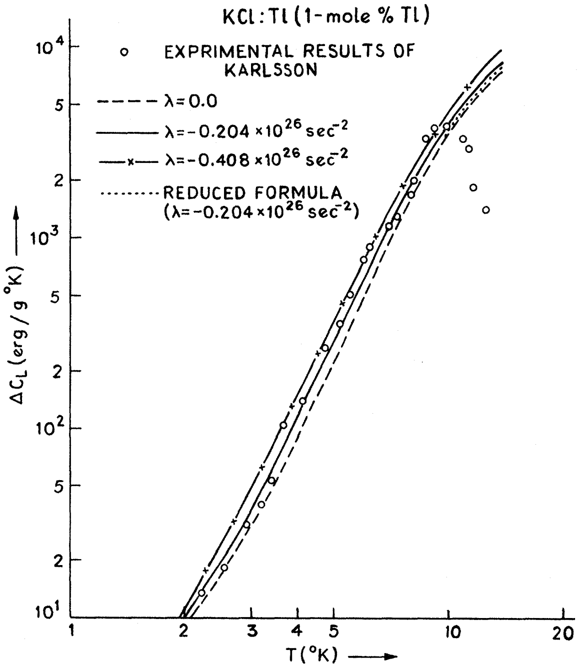

FIG. 1. Comparison of the predicted contributions to the specific heat with the experimental results in KCl: 1-mole% Tl^+.

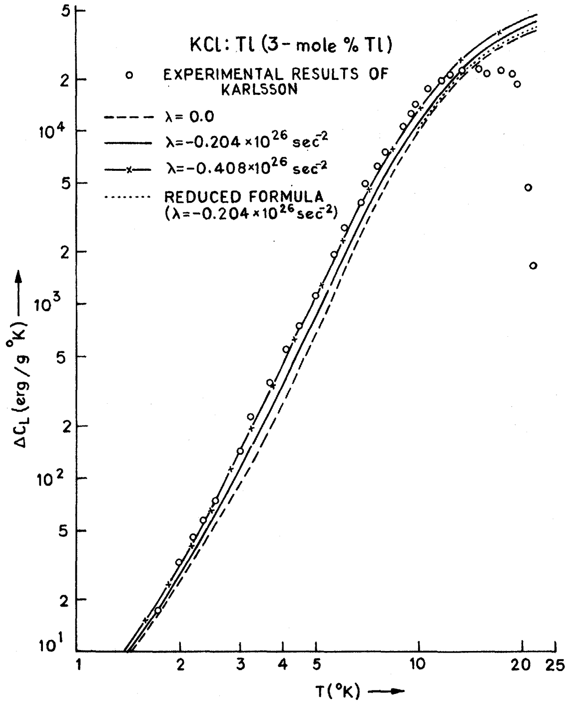

FIG. 2. Comparison of the predicted contributions to the specific heat with the experimental results in KCl: 3-mole% Tl^+.

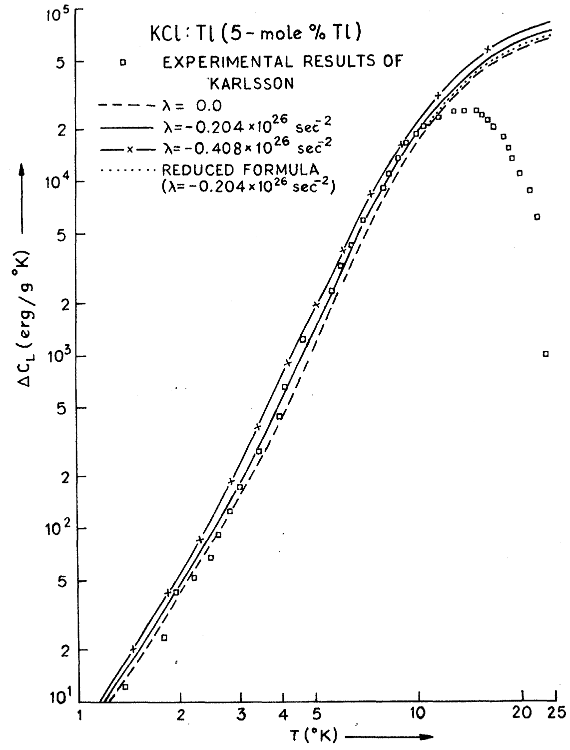

FIG. 3. Comparison of the predicted contributions to the specific heat with the experimental results in KCl: 5-mole% Tl^+.

for $\lambda = -0.204 \times 10^{26}$ sec^{-2}. A low frequency resonance appears at $\omega_r = 41.7$ cm^{-1}. This resonance is found at a frequency $\omega_r = 42.5$ cm^{-1} in the mass-defect approximation. The experimental information about the resonance frequency is not very consistent because on the one hand the infrared absorption measurements done by Kahan and Sievers[21] have revealed a weak line in KCl: Tl^+ at about 39 cm^{-1}, while on the other hand the thermal conductivity experiments[22] have detected a resonance at 45 cm^{-1}.

The phase shift in the F_{1u} irreducible representation is shown in Fig. 5. A phase shift of $\frac{1}{2}\pi$ appropriate at resonance is well produced in the calculation. The contributions of phase shift to the specific heat are seen to be negligible above the frequency 106 cm^{-1}.

The contributions of the two other irreducible representations A_{1g} and E_g are comparatively much smaller. At 4 °K, the contributions of E_g and A_{1g} symmetry motions to specific heat are only 0.5% and 0.2%, respectively, while the contribution of F_{1u} modes is 99.3%. At high temperatures the contributions of E_g and A_{1g} irreducible representations increase, e.g., at 25 °K, the contributions of E_g and A_{1g} irreducible representations are 1.8% and 0.7%, respectively.

The contribution of F_{1u} symmetry modes has also been determined in the effective-force-constant approximation by using Eq. (14). An effective-force-constant has been determined by Eq. (14) using $\lambda = -0.204 \times 10^{26}$ sec^{-2} and the resonance frequency 41.7 cm^{-1}. The calculated value of the effective nearest-neighbor central force constant η

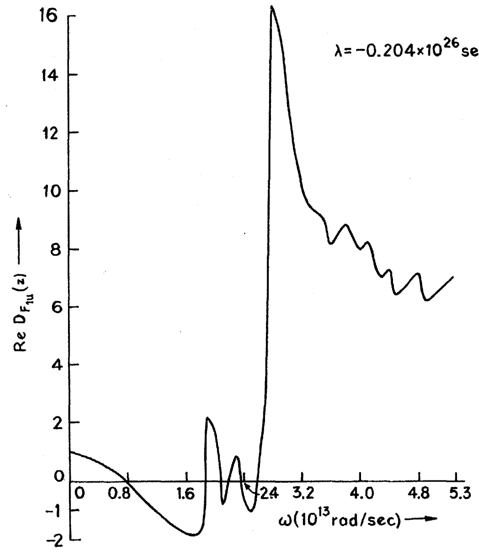

FIG. 4. Real part of the determinant of the F_{1u} symmetry modes for the force-constant change $\lambda = -0.204 \times 10^{26}$ sec^{-2}.

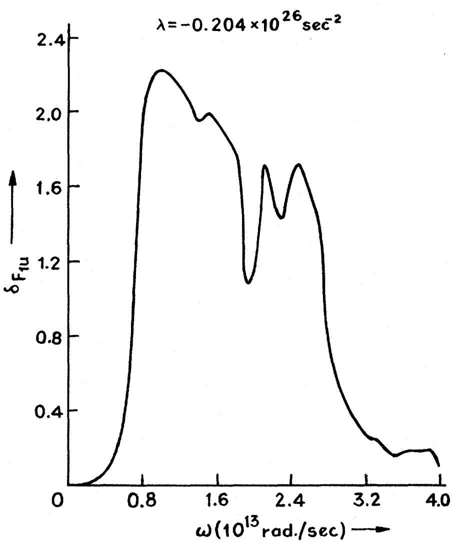

FIG. 5. Phase shifts of F_{1u} symmetry modes for $\lambda=-0.204\times10^{26}\ \mathrm{sec}^{-2}$.

is $4.334\times10^{26}\ \mathrm{sec}^{-2}$. This value of effective force constant is quite close to the value $4.09\times10^{26}\ \mathrm{sec}^{-2}$ calculated by Benedek and Nardelli.[19] Thus the percentage change in the force constant (λ/η) caused by a thallium ion is seen to be very small, i.e., about 5%.

The total contribution of the three irreducible representations using the effective-force-constant approximation for F_{1u} symmetry motions have been depicted in Figs. 1–3. It may be noted that these results are very similar to those obtained in a mass-defect approximation except at higher temperatures (above 10 °K) where there are some differences in the magnitudes of the two values. These discrepancies arise because at high temperatures the contributions of E_g and A_{1g} irreducible representations are appreciable. Finally, the specific-heat values in the effective-force-constant model differ in magnitudes from those in a realistic calculation [Eq. (13)] by about 24% at 4 °K, and by 6% at 25 °K. These discrepancies disappear as we go to higher temperatures.

IV. DISCUSSION AND CONCLUSIONS

The temperature dependence of the experimentally measured lattice specific heat of KCl doped with Tl^+ impurity ions has been explained successfully in the temperature range 1–10 °K. However, the drop in observed specific heat above 10 °K cannot be understood on the basis of the theory. The present low-concentration Green's-function theory is seen to explain the observed specific heat even at comparatively high concentration (5-mole% Tl^+).

The reported error in the measured specific heat is ± 2%, which gives rise to an error of ± 10% over most of the temperature and concentration ranges of the enhanced specific heat. In the present calculation we have found that the enhanced specific heat is very sensitive to force constant changes e.g., a 5% change in the force constant produces ~ 20% change in the increased specific heat. Our determined force-constant change has, therefore, an accuracy of about 2.5% and the evaluated change in force constant may be written as (5 ± 2.5)%.

The change in force constant induced by thallium ion is not seen to be very large. The percentage change is within 10%. It implies that Tl^+ impurity ion may be treated as a "mass defect." Similar conclusions have been drawn by Harley *et al.*[9] and Kravitz[10] after analyzing their measured data of first-order Raman scattering. These authors have noted very small changes in the force constants.

Harley *et al.* have, however, observed a 10% stiffening in the force constant as compared to a 5% softening in the present calculation. The enhancement in the specific heat is mainly due to the mass defect which, in turn, is determined by the Green's function g_1^+. The disparity in the calculated force-constant changes obtained from the specific-heat data and Raman scattering data may arise because of the different values of the Green's functions g_1^+ used in the two calculations. The resonance frequency calculated by Page[23] in the mass-defect model is 43 cm^{-1} (as compared to 42.5 cm^{-1} in the present calculation). Usually the real part of the Green's function g_1^+ remains constant in the low-frequency region. A value of $\omega_r=43\ \mathrm{cm}^{-1}$ is obtained if we decrease our value of g_1^+ by about 4%. This change in the Green's function will lower the enhanced specific heat by 6%. Consequently, with the use of g_1^+ calculated by Page, a further decrease in the force constant by about 1.5% will be observed. Thus, it is difficult to reconcile the different changes in the force constant obtained by the specific-heat data and the Raman scattering data.

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Zero-Sound Elastic Constants of Solid Krypton at $T = 114\,^{\circ}\mathrm{K}$[*]

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The phonon dispersion of a single crystal of krypton has been measured for the $[110]\mathrm{L}$, $[110]\mathrm{T}_1$, and $[100]\mathrm{T}$ modes at $T = 114\,^{\circ}\mathrm{K}$ in the region $0.03 < q < 0.15$ Å^{-1} by the inelastic scattering of neutrons. Analysis of the results based on a nearest-neighbor-force-constant fit gives for the zero-sound elastic constants $C_{11} = (289 \pm 4)$, $C_{12} = (185 \pm 4)$, and $C_{44} = (144 \pm 1) \times 10^8$ dyn cm^{-2}. This results in an anisotropy $A = 2C_{44}/(C_{11} - C_{12}) = 2.76 \pm 0.05$ and a departure from the Cauchy relation of $\delta = (C_{44} - C_{12})/C_{12} = -0.22 \pm 0.02$.

I. INTRODUCTION

Theoretical calculations of the properties of rare-gas solids has progressed considerably in the last decade. Computational capability has been increased and hence calculations need not be restricted to simple interaction potentials of limited validity, i.e., the Lennard-Jones potential. One property of importance is the elastic constants and the present paper is directed towards their measurement in krypton. Such measurements can give a measure of the importance of three-body forces by comparison of the results with calculations which have utilized a suitable two-body potential.[1,2] One indication of three-body effects is the deviation of the Cauchy relation $\delta = (C_{44} - C_{12})/C_{12}$ at liquid-helium temperatures, where departures due to quantum effects have been minimized by the heavy mass of the krypton atom. The three-body effect acts in opposition to the quantum effect and Hüller *et al.*[1] give for krypton, at $T = 0$, a value for δ of $+0.02$ and -0.07 for calculations using a Lennard-Jones potential without and with a three-body force, respectively.

While the measurement of elastic constants on single crystals through the use of ultrasonic transducers is well known, its direct application to the case of rare-gas solids has not been straightforward. The main difficulty is obtaining one oriented single crystal of sample to which transducers can be satisfactorily bonded; the bonding problem as the temperature changes is maximized by the large volume expansivity in these solids. Ultrasonic measurements have been made as a function of temperature for the transverse and longitudinal sound velocities in polycrystalline krypton,[3–6] which alone do not give the elastic constants. Korpiun *et al.*[6] have further used circuitous reasoning to justify the assumption of a measurement in two samples as being made in the [100] direction, thus obtaining C_{11}, C_{44}, and thence C_{12} from the polycrystalline measurements.

Recently Brillouin scattering has been used by Gornall and Stoicheff[7,8] and Stoicheff *et al.*[9] to measure the sound velocities of oriented single crystals