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Computation of Phonon Spectral Functions and Ground-State Energy of Solid Helium. I. bcc Phase

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Computations incorporating the three-phonon interaction process are presented of phonon spectral functions, yielding frequency and lifetime dispersion curves, for the bcc solid phase of both He^3 and He^4. Comparison of the compressibilities obtained from ground-state energy and elastic constants as functions of molar volume quantify the effects of certain analytical approximations. A model calculation shows the sensitivity of elastic anisotropy and long-wavelength dispersions to the precise form of the three-phonon vertex.

I. INTRODUCTION

In a recent publication,[1] we have presented a theory of the damping and frequency shift of phonons and of ground-state energy corrections due to interactions between phonons in quantum crystals with singular forces. We here turn to a numerical implementation of this theory and present computations applicable to solid helium (both He^3 and He^4) in the bcc phase.

Section II reviews the theoretical approach and summarizes the resulting formulas.[1] Some additional developments are given in order to arrive at a formula for the phonon spectral function which is computationally satisfactory. In Sec. III we exhibit numerical results: phonon self-energies, dispersion curves, linewidths, and spectral shapes for bcc He^3 at a molar volume of 21.5 cm^3 and bcc He^4 at 21.0 cm^3. Results are also given for phonon interaction corrections to the free energy, and for sound velocities as a function of molar volume in bcc He^3. Section IV compares this approach and its results with previous treatments by other authors, particularly Horner[2] and Glyde and co-workers.[3] Some schematically parametrized computations are presented which help to understand and reconcile certain differences between the conclusions of these authors.

II. REVIEW OF THEORETICAL APPROACH AND SUMMARY OF FORMULAS

The theoretical approach which we utilize[1] begins with the adoption of a trial ground-state wave function Ψ_0 of the Jastrow form, involving products of short-range pair functions f_{ij} and a phonon Gaussian:

$$\Psi_0(\vec{r}_1, \ldots, \vec{r}_N) = \prod_{i<j} f_{ij}(\vec{r}_{ij}) \exp(-\tfrac{1}{4}\vec{u}_{ij} \cdot \overleftrightarrow{\Gamma}_{ij} \cdot \vec{u}_{ij}), \tag{2.1}$$

where $\vec{r}_{ij} \equiv \vec{r}_i - \vec{r}_j$ and $\vec{u}_{ij} \equiv \vec{r}_{ij} - \vec{R}_{ij}$ are differences of displacements from equilibrium position $\vec{R}_i = \langle \vec{r}_i \rangle$. Trial excited-state wave functions are also constructed, to represent one-, two-, and three-phonon excitations, by multiplying Ψ_0 by mutually orthogonalized polynomials in the displacement variables. In the absence of short-range correlations, these polynomials reduce to the usual Hermite polynomials in the normal coordinates. Matrix elements of the Hamiltonian

$$\mathcal{H} = \sum_i \frac{p_i^2}{2M_i} + \sum_{i<j} v_{ij}(\vec{r}_{ij}) \tag{2.2}$$

can then be evaluated with these wave functions.

If we restrict attention to the submatrix of $\mathcal{H}$ in the zero-, one-, and two-phonon states, then it is possible[1] to diagonalize the submatrix and to minimize the lowest eigenvalue with respect to the variational parameters $\overleftrightarrow{\Gamma}_{ij}$, or more properly the combinations

$$\overleftrightarrow{G}_{ij} \equiv \sum_{i',j'} \tfrac{1}{2}(\delta_{i,i'} - \delta_{i,j'})(\delta_{j,i'} - \delta_{j,j'})\overleftrightarrow{\Gamma}_{i'j'} . \tag{2.3}$$

Simple expressions emerge because the minimization of the lowest submatrix eigenvalue is equivalent to the vanishing of matrix elements between the zero-phonon (ground) state and all two-phonon states, so that the submatrix is factorizable.

The many-body matrix elements are further reduced to computationally manageable two-body form by making the first-order van Kampen–Nosanow cluster expansion, which for our purposes is expressed by the approximation for the two-body distribution function,

$$\begin{aligned} g_{ij}(\vec{r}) &\equiv \langle 0 \,|\, \delta^3(\vec{r} - \vec{r}_{ij}) \,|\, 0 \rangle \\ &\cong \rho_{ij}(\vec{r}) \tilde{f}_{ij}{}^2(\vec{r}), \end{aligned} \tag{2.4}$$

where

$$\rho_{ij}(\vec{r}) \equiv \exp(-\tfrac{1}{2}\vec{u}\cdot\overleftrightarrow{\gamma}_{ij}\cdot\vec{u}) \Big/ \int d^3r \exp(-\tfrac{1}{2}\vec{u}\cdot\overleftrightarrow{\gamma}_{ij}\cdot\vec{u}), \tag{2.5}$$

with $\vec{u} \equiv \vec{r} - \vec{R}_{ij}$, and where

$$\begin{aligned}\tilde{f}_{ij}{}^2(\vec{r}) &\equiv f_{ij}{}^2(\vec{r}) \Big/ \int d^3r\, \rho_{ij}(\vec{r}) f_{ij}{}^2(\vec{r}) \\ &\equiv f_{ij}{}^2(\vec{r}) / \langle f_{ij}{}^2 \rangle_\gamma .\end{aligned} \tag{2.6}$$

Before specifying the matrix $\overleftrightarrow{\gamma}_{ij}$, it proves convenient to introduce the definition of the modified potential

$$\tilde{v}_{ij}(\vec{r}) \equiv \tilde{f}_{ij}{}^2(\vec{r})[v^*_{ij}(\vec{r}) - \langle \tilde{f}_{ij}{}^2 v^*_{ij} \rangle_\gamma], \tag{2.7}$$

where

$$v^*_{ij}(\vec{r}) \equiv v_{ij}(\vec{r}) - (\hbar^2/2M_{ij})\nabla^2 \ln f_{ij}{}^2(\vec{r}) \tag{2.8}$$

and where

$$M_{ij} \equiv [(M_i^{-1} + M_j^{-1})]^{-1} \tag{2.9}$$

is an average mass for a pair of particles. If we further introduce the effective spring constants $\overleftrightarrow{\Phi}_{ij}$ given by

$$\overleftrightarrow{\Phi}_{ij} \equiv \sum_{i',j'} \tfrac{1}{2}(\delta_{i,i'} - \delta_{i,j'})(\delta_{j,i'} - \delta_{j,j'}) \langle \nabla\nabla \tilde{v}_{i',j'} \rangle_\gamma , \tag{2.10}$$

which diagonalize into effective self-consistent frequencies Ω_Λ and polarization vectors $\vec{e}^{\,i}_\Lambda$, ($\Lambda = 1, \ldots, 3N$) via

$$\Omega^2_\Lambda \vec{e}^{\,i}_\Lambda = \textstyle\sum_j (M_i M_j)^{-1/2} \overleftrightarrow{\Phi}_{ij} \cdot \vec{e}^{\,j}_\Lambda , \tag{2.11}$$

then the $\overleftrightarrow{G}_{ij}$ which make the lowest eigenvalue of the Hamiltonian submatrix stationary are given by

$$\overleftrightarrow{G}_{ij} = \textstyle\sum_\Lambda [\hbar/2(M_i M_j)^{1/2}\Omega_\Lambda]^{-1} \vec{e}^{\,i}_\Lambda \vec{e}^{\,j*}_\Lambda , \tag{2.12}$$

and the $\overleftrightarrow{\gamma}_{ij}$ are defined through their three-dimensional tensor inverse,

$$\begin{aligned}\overleftrightarrow{\gamma}_{ij}{}^{-1} &= \sum_{i',j'} (\delta_{i',i} - \delta_{i',j})(\delta_{j',i} - \delta_{j',j}) \overleftrightarrow{G}_{i'j'}{}^{-1} \\ &= \sum_\Lambda \frac{\hbar}{2\Omega_\Lambda} \vec{E}^{ij}_\Lambda \vec{E}^{ij*}_\Lambda ,\end{aligned} \tag{2.13}$$

with the notation

$$\vec{E}^{ij}_\Lambda \equiv \textstyle\sum_l (\delta_{l,i} - \delta_{l,j}) M_l^{-1/2} \vec{e}^{\,l}_\Lambda . \tag{2.14}$$

The lowest submatrix eigenvalue is then just equal to $\langle 0 | \mathcal{H} | 0 \rangle$,

$$E_0 \equiv \langle 0 | \mathcal{H} | 0 \rangle = \tfrac{1}{4}\sum_\Lambda \hbar\,\Omega_\Lambda + \sum_{i<j} \langle \tilde{f}_{ij}{}^2 v^*_{ij} \rangle_\gamma . \tag{2.15}$$

The one-phonon states, denoted as $|\Lambda\rangle$, can be characterized by diagonalizing the matrix

$$\begin{aligned}\overleftrightarrow{D}_{ij} &\equiv \overleftrightarrow{G}_{ij}{}^{-1} + \sum_{i',j'} \overleftrightarrow{G}_{ii'}{}^{-1} \sum_{l,l'} \tfrac{1}{2}(\delta_{i',l} - \delta_{i',l'})(\delta_{j',l} - \delta_{j',l'}) \langle \nabla\nabla \tilde{f}_{ll'}{}^2 \rangle_\gamma \cdot \overleftrightarrow{G}_{j'j}{}^{-1} \\ &= \sum_{\Lambda,\Lambda'} (M_i M_j)^{-1/2} \vec{e}^{\,i}_\Lambda \left(\frac{\hbar}{2\Omega_\Lambda} \delta_{\Lambda,\Lambda'} + \frac{\hbar^2}{4\Omega_\Lambda \Omega_{\Lambda'}} \sum_{i',j'} \tfrac{1}{2} \vec{E}^{i'j'*}_\Lambda \cdot \langle \nabla\nabla \tilde{f}_{i'j'}{}^2 \rangle_\gamma \cdot \vec{E}^{i'j'}_{\Lambda'} \right) \vec{e}^{\,j*}_{\Lambda'}\end{aligned} \tag{2.16}$$

into

$$\overleftrightarrow{D}_{ij} = \textstyle\sum_\Lambda (M_i M_j)^{-1/2} \vec{\epsilon}^{\,i}_\Lambda D_\Lambda \vec{\epsilon}^{\,j*}_\Lambda . \tag{2.17}$$

The states $|\Lambda\rangle$ are then defined with the aid of the eigenvectors $\vec{\epsilon}^{\,i}_\Lambda$,

$$|\Lambda\rangle \equiv u_\Lambda |0\rangle = (\textstyle\sum_i M_i^{1/2} \vec{\epsilon}^{\,i*}_\Lambda \cdot \vec{u}_i) |0\rangle , \tag{2.18}$$

and the one-phonon submatrix of the Hamiltonian in this basis is diagonal,

$$\begin{aligned}\langle \Lambda | (\mathcal{H} - E_0) | \Lambda' \rangle &= (\hbar^2/2D_\Lambda) \delta_{\Lambda,\Lambda'} \\ &\equiv \hbar\omega_\Lambda \delta_{\Lambda,\Lambda'} .\end{aligned} \tag{2.19}$$

The phonon excitation frequencies ω_Λ are not equal to the self-consistent frequencies Ω_Λ; the latter serve merely to define the Gaussian averages $\langle \cdots \rangle_\gamma$.

Matrix elements involving orthonormalized two-phonon states work out to

$$\begin{aligned}&\langle \Lambda | \mathcal{H} | \Lambda_1, \Lambda_2 \rangle \\ &\quad \cong D_\Lambda^{-1/2} \sum_{i<j} \left\langle \prod_\mu \left[\left(\frac{\hbar}{2\Omega_\mu} \right)^{1/2} \vec{E}^{ij*}_\mu \cdot \nabla \right] \tilde{v}_{ij} \right\rangle_\gamma ,\end{aligned} \tag{2.20}$$

where the product runs over the three values $\mu = \Lambda^*, \Lambda_1, \Lambda_2$, and to

$$\begin{aligned}\langle \Lambda_1, \Lambda_2 | (\mathcal{H} - E_0) | \Lambda_1', \Lambda_2' \rangle &\cong \delta_{\Lambda_1\Lambda_2, \Lambda_1'\Lambda_2'} \hbar(\omega_{\Lambda_1} + \omega_{\Lambda_2}) \\ &\quad + \sum_{i<j} \left\langle \prod_\mu \left[\left(\frac{\hbar}{2\Omega_\mu} \right)^{1/2} \vec{E}^{ij*}_\mu \cdot \nabla \right] \tilde{v}_{ij} \right\rangle_\gamma ,\end{aligned} \tag{2.21}$$

where here $\mu = \Lambda_1^*, \Lambda_2^*, \Lambda_1', \Lambda_2'$. The eigenvalues of the coupled one- and two-phonon submatrices, giving the phonon frequencies as modified by interaction with two-phonon states, are the roots λ of the secular equation,

$$\begin{aligned}\det \Big| (\hbar\omega_\Lambda - \lambda)\delta_{\Lambda,\Lambda'} - \sum_{\Lambda_1 \leq \Lambda_2} \sum_{\Lambda_1' \leq \Lambda_2'} &\langle \Lambda | \mathcal{H} | \Lambda_1, \Lambda_2 \rangle \\ &\times \langle \Lambda_1, \Lambda_2 | (\mathcal{H} - E_0 - \lambda) | \Lambda_1', \Lambda_2' \rangle^{-1} \\ &\times \langle \Lambda_1', \Lambda_2' | \mathcal{H} | \Lambda \rangle \Big| = 0 .\end{aligned} \tag{2.22}$$

If it is argued[1] that the second line of Eq. (2.21), which represents an interaction between the two phonons of a phonon pair state, is not sizable and as a first approximation can be neglected when inserted into Eq. (2.22), then the secular equation simplifies to

$$\begin{aligned}\det \Big| (\hbar\omega_\Lambda - \lambda)\delta_{\Lambda,\Lambda'} - \sum_{\Lambda_1 \leq \Lambda_2} &\langle \Lambda | \mathcal{H} | \Lambda_1, \Lambda_2 \rangle \\ &\times (\hbar\omega_{\Lambda_1} + \hbar\omega_{\Lambda_2} - \lambda)^{-1} \langle \Lambda_1, \Lambda_2 | \mathcal{H} | \Lambda' \rangle = 0 .\end{aligned} \tag{2.23}$$

Enlarging the Hamiltonian submatrix by including three-phonon states in the basis provides the leading correction to the value E_0 for the ground-state energy. Approximations analogous to those used with the two-phonon states lead to the expressions for matrix elements involving three-phonon states,

$$\langle \Lambda_1, \Lambda_2, \Lambda_3 \,|\, (\mathcal{H}-\lambda) \,|\, \Lambda_1', \Lambda_2', \Lambda_3' \rangle \cong \hbar(\omega_{\Lambda_1}+\omega_{\Lambda_2}+\omega_{\Lambda_3})\delta_{\Lambda_1\Lambda_2\Lambda_3,\Lambda_1'\Lambda_2'\Lambda_3'}\,, \tag{2.24}$$

in which mutual interaction among the phonons is neglected, and

$$\langle 0 \,|\mathcal{H}|\, \Lambda_1, \Lambda_2, \Lambda_3 \rangle \cong \sum_{i<j} \left\langle \prod_{\mu} \left[\left(\frac{\hbar}{2\Omega_\mu} \right)^{1/2} \vec{E}_\mu^{ij*} \cdot \nabla \right] \tilde{v}_{ij} \right\rangle_{\gamma} , \tag{2.25}$$

where $\mu = \Lambda_1, \Lambda_2, \Lambda_3$. If we also neglect matrix elements of $\mathcal{H}$ between three-phonon states and all one- and two-phonon states which are of only minor significance in affecting the ground-state energy, then the improved value E_0' is given by the simple result

$$E_0' = E_0 - \sum_{\Lambda_1 \leqslant \Lambda_2 \leqslant \Lambda_3} |\langle 0 | \mathcal{H} | \Lambda_1, \Lambda_2, \Lambda_3 \rangle|^2 \times (\hbar\omega_{\Lambda_1} + \hbar\omega_{\Lambda_2} + \hbar\omega_{\Lambda_3})^{-1} . \tag{2.26}$$

The solution of a very large-sized secular equation such as Eq. (2.23), which arises naturally in a matrix approach to quantum mechanics, is not the most convenient method for numerical evaluation. It is preferable to make use of the well-established body of computer techniques developed around the more traditional formulas for anharmonic corrections to phonon spectral functions based on time-dependent methods. Some care is necessary in order to establish proper contact between the matrix and the time-dependent approaches. In systems for which the latter approach is naturally appropriate, the Hamiltonian can be written in terms of creation and destruction operators. Even though it is not usually made explicit certain characteristic reciprocal relationships then exist between various matrix elements of the bare and perturbation parts of the Hamiltonian. For example in the phonon case the element $\langle k_1 | \mathcal{H} | k_2 k_3 \rangle$ has a definite connection to $\langle 0 | \mathcal{H} | k_1 k_2 k_3 \rangle$, which obtains if the bare Hamiltonian is quasiharmonic. If matrix elements of a Hamiltonian evaluated in some arbitrary basis set obey these reciprocal relationships, then the frequency-dependent results of a field-theoretic development can be taken over just by identifying the factors which would enter the field-theoretic Hamiltonian.

The Hamiltonian matrix constructed in Ref. 1 and reviewed here does not precisely obey all the necessary reciprocal relationships, although they are obeyed in the limit $f^2 \to 1$. However with the approximations which lead to the secular equation (2.22) and matrix elements (2.20) and (2.21), a form is arrived at which would obtain in a time-dependent matrix development of the self-consistent phonon theory,[4] but with the replacement $v \to \tilde{v}$ and the distinction between $(\Omega_\Lambda, \vec{e}_\Lambda^{\,i})$ and $(\omega_\Lambda, \tilde{e}_\Lambda^{\,i})$. Thus we argue that a time-dependent propagator formulation of the present problem with short-range correlation would, to a good approximation, produce the result that the phonon spectral function takes the same form as in self-consistent phonon theory but with the modifications as cited.[5] Thus the spectral function $\mathcal{A}_{\Lambda\Lambda'}(\omega)$ is given by

$$\mathcal{A}_{\Lambda\Lambda'}(\omega) = \operatorname{Im}\,[\omega^2 \delta_{\Lambda,\Lambda'} - \Phi_{\Lambda\Lambda'}(\omega + i0^+)]^{-1} , \tag{2.27}$$

where the self-energy is of the usual form but with the matrix elements (2.20)

$$\Phi_{\Lambda\Lambda'}(\omega) = \omega_\Lambda^2 \delta_{\Lambda,\Lambda'} - \sum_{\Lambda_1 \leqslant \Lambda_2} \langle \Lambda | \mathcal{H} | \Lambda_1, \Lambda_2 \rangle \langle \Lambda_1, \Lambda_2 | \mathcal{H} | \Lambda' \rangle \times \hbar^{-1} [(\omega_{\Lambda_1} + \omega_{\Lambda_2} - \omega)^{-1} + (\omega_{\Lambda_1} + \omega_{\Lambda_2} + \omega)^{-1}] . \tag{2.28}$$

Note added in proof: A more rigorous justification of Eqs. (2.27) and (2.28) has been constructed by one of us [T.R. Koehler (unpublished)].

Considerable further simplification is possible in the present case of a periodically regular Bravais lattice in which $\Lambda \to (\vec{k}, \lambda)$, where $\vec{k}$ is a wave vector in the first Brillouin zone and λ is a polarization index, and in which

$$\vec{e}_\Lambda^{\,i} \to \vec{e}_{\vec{k}\lambda} N^{-1/2} e^{i\vec{k}\cdot\vec{R}_i} .$$

Then with further notation as per Ref. 4,

$$\begin{aligned}\mathcal{A}_{\Lambda\Lambda'}(\omega) \to \mathcal{A}(\vec{k}\omega) &\equiv \operatorname{Im}\{\omega^2 - \omega_{\vec{k}\lambda}^2 - 2\omega_{\vec{k}\lambda}[\Delta_\lambda(\vec{k}\omega) + i\Gamma_\lambda(\vec{k}\omega)]\}^{-1} \\ &= \operatorname{Im}\Bigg\{(\omega^2 - \omega_{\vec{k}\lambda}^2) - \tfrac{1}{2}N^{-1} \sum_{\vec{k}_1\lambda_1} \sum_{\vec{k}_2\lambda_2} \Delta(\vec{k} - \vec{k}_1 - \vec{k}_2) \\ &\qquad \times \left| \frac{1}{2} \sum_{\vec{\tau}} \left(\frac{\hbar}{2\omega_{\vec{k}\lambda}} \right)^{-1/2} \left\langle \prod_\mu \left(\left(\frac{\hbar}{2M\Omega_\mu} \right)^{1/2} (1 - e^{-i\vec{k}_\mu\cdot\vec{\tau}})\, \vec{e}_\mu^{\,*} \cdot \nabla \right) \vec{v}(\vec{\tau} + \vec{u}) \right\rangle_\gamma \right|^2 \\ &\qquad \times \hbar^{-1}[(\omega_{\vec{k}_1\lambda_1} + \omega_{\vec{k}_2\lambda_2} - \omega)^{-1} + (\omega_{\vec{k}_1\lambda_1} + \omega_{\vec{k}_2\lambda_2} + \omega)^{-1}] \Bigg\}^{-1} .\end{aligned} \tag{2.29}$$

The product runs over $\mu = (-\vec{k}\lambda, \vec{k}_1\lambda_1, \vec{k}_2\lambda_2)$, and we neglect polarization mixing along high-symmetry directions. Similarly the ground-state energy equation (2.26) simplifies to

$$E_0' = E_0 - \frac{1}{3!} N^{-1} \sum_{\vec{k}_1\lambda_1} \sum_{\vec{k}_2\lambda_2} \sum_{\vec{k}_3\lambda_3} \Delta(\vec{k}_1 + \vec{k}_2 + \vec{k}_3) \times \left| \frac{1}{2} \sum_{\vec{\tau}} \left\langle \prod_{\mu} \left[\left(\frac{\hbar}{2M\Omega_\mu} \right)^{1/2} (1 - e^{-i\vec{k}_\mu \cdot \vec{\tau}}) \vec{e}_\mu^{\,*} \cdot \nabla \right] \tilde{v}(\vec{\tau} + \vec{u}) \right\rangle_\gamma \right|^2 \hbar^{-1} (\omega_{\vec{k}_1\lambda_1} + \omega_{\vec{k}_2\lambda_2} + \omega_{\vec{k}_3\lambda_3})^{-1} . \quad (2.30)$$

It is these formulas, (2.29) and (2.30), which we implement numerically in the work reported here.

III. NUMERICAL RESULTS

The primary quantities generated by the computation are the complex self-energy and the spectral weight as functions of frequency ω. An example of typical results is illustrated in Fig. 1, for bcc He^3 at a molar volume of 21.5 cm^3. For each of the three phonon branches at the point $\vec{q} = (0.8, 0.8, 0)(2\pi/a)$ is shown the real and imaginary parts of the self-energy $\Delta + i\Gamma$ and the spectral function $\mathcal{A}$. A significant feature of these results is that whereas the two transverse phonons have narrow symmetric line shapes, the longitudinal phonon is quite broad and asymmetric. The asymmetry is characteristically in the form of a steep rise on the low-frequency side of the maximum and a more gradually sloping tail on the high-frequency side.

Because of this asymmetry, which occurs over a large region in the Brillouin zone, ambiguity arises on how to parametrize the line shape in order to present dispersion curves. We chose to evaluate as functions of $\vec{q}$ both the position of the maximum in $\mathcal{A}$ as well as the mean of the two half-maxima. Significant differences between these two alternative quantities are an indication of substantial line-shape asymmetry, or equivalently of strongly frequency-dependent damping. Another aspect of the sizable variation of the self-energy over a frequency comparable to its magnitude is that the maximum and width of $\mathcal{A}$ do not coincide with the values that would have been obtained from evaluating Δ and Γ at the unshifted frequency. The latter procedure is what would arise from a strict application of Rayleigh-Schrödinger perturbation theory, and is a prevalent one in more traditional perturbation treatments of weak anharmonicity. The examination of $\mathcal{A}(\omega)$, however, is much closer to Brillouin-Wigner perturbation theory, and is the more appropriate procedure in the present case of sizable shifts and broadening of phonon modes.

Figures 2 and 3 show parametrizations of Δ and Γ as functions of $\vec{q}$ along symmetry directions in the Brillouin zone, for bcc He^3 at 21.5 cm^3/mole and bcc He^4 at 21.0 cm^3/mole, respectively. Illustrated for each of the three branches are the values of Δ and Γ at the unshifted frequency, at the mean of the half-maxima, and at the maximum of $\mathcal{A}$. Regions of the Brillouin zone where these three values are substantially different are the regions where the three-phonon process is strong and significantly frequency dependent. It can also be seen that phonon damping, as measured by Γ, has dramatic variation with $\vec{q}$, even across a Brillouin zone face. Figures 4 and 5, again for He^3

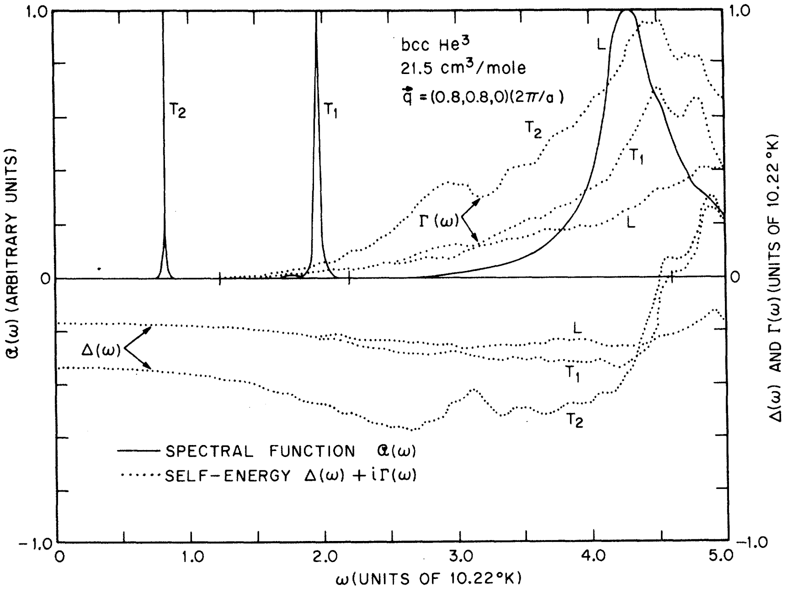

FIG. 1. Phonon spectral function $\mathcal{A}(\omega)$ (solid curves) and complex self-energy $\Delta(\omega) + \Gamma(\omega)$ (dotted curves) as a function of ω for the three $\vec{q} = (0.8, 0.8, 0)$ modes of bcc He^3 at a molar volume of 21.5 cm^3.

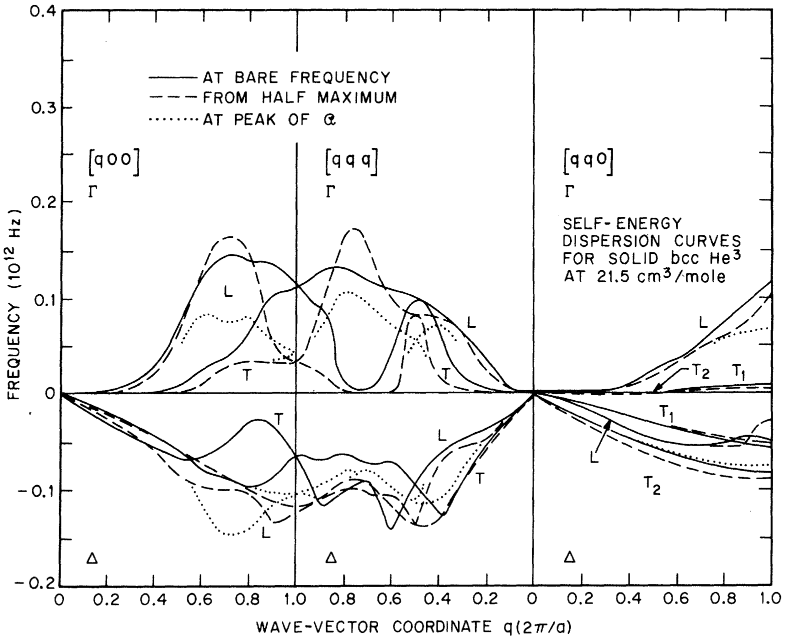

FIG. 2. Several parametrizations of the real and imaginary parts of the phonon self-energy, as a function of wave vector along the principal symmetry directions, for bcc He^3 at 21.5 cm^3/mole.

and He^4, respectively, plot dispersion curves for the phonon frequencies along symmetry directions. The three-phonon process always lowers the frequencies from their base unshifted values. Particularly in the vicinity of $\vec{q}=(1,0,0)(2\pi/a)$ there is spectral-function asymmetry, as measured by the differences between the positions of the maximum of 𝒜 and the mean of the half-maxima. Also in these regions the Rayleigh-Schrödinger result ("bare frequency + Δ") is not the same as that of Brillouin-Wigner ("peak of 𝒜") result.

The ground-state energy, as computed from formula (2.30), is plotted in Fig. 6 as a function of molar volume for bcc He^3. Both the unshifted energy E_0 ("first order") as well as the three-phonon-corrected energy E_0' ("second order") are shown. Because we use throughout the Lennard-Jones 6-12 interatomic potential, which is now believed not to be a sufficiently accurate representation of the true helium potential, we chose not to

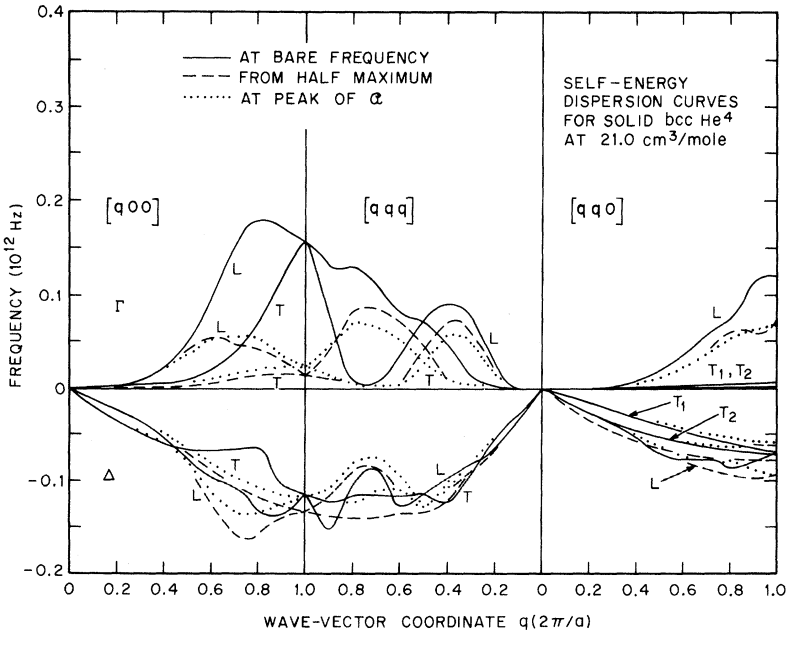

FIG. 3. Self-energy dispersion curves similar to Fig. 2, but for bcc He^4 at 21.0 cm^3/mole.

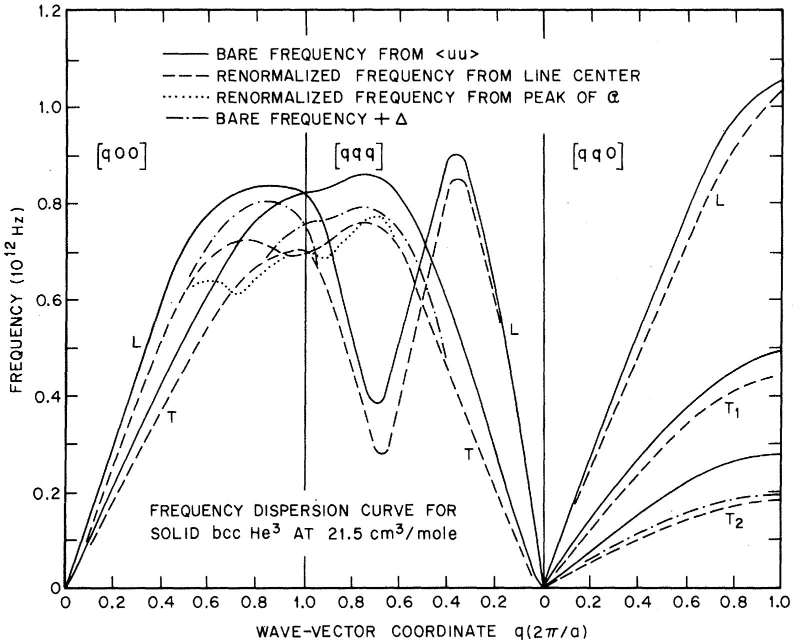

FIG. 4. Phonon frequency dispersion curves, defined in several differing ways, along the principal symmetry directions for bcc He^3 at 21.5 cm^3/mole.

compare these computations with the experimental values. Instead we plot the computer result of Hansen,[6] based on a variational wave-function treatment with Monte Carlo evaluation of the Hamiltonian expectation. Since such a result must be an upper bound to the exact ground-state energy with the Lennard-Jones 6-12 potential, it provides a more meaningful test of out theoretical approximations. Although the energy lowering due to the inclusion of three-phonon processes is sizable (order of 1 °K per particle) it is evident that this lowering does not fully compensate for making the two-body cluster approximation. It would seem that the most significant step that must be taken in the future to improve the theoretical treatment of quantum crystals such as helium is to improve on or avoid cluster expansions in the evaluation of Hamiltonian matrix elements.

The elastic constants are quantities of considerable current experimental interest which can be

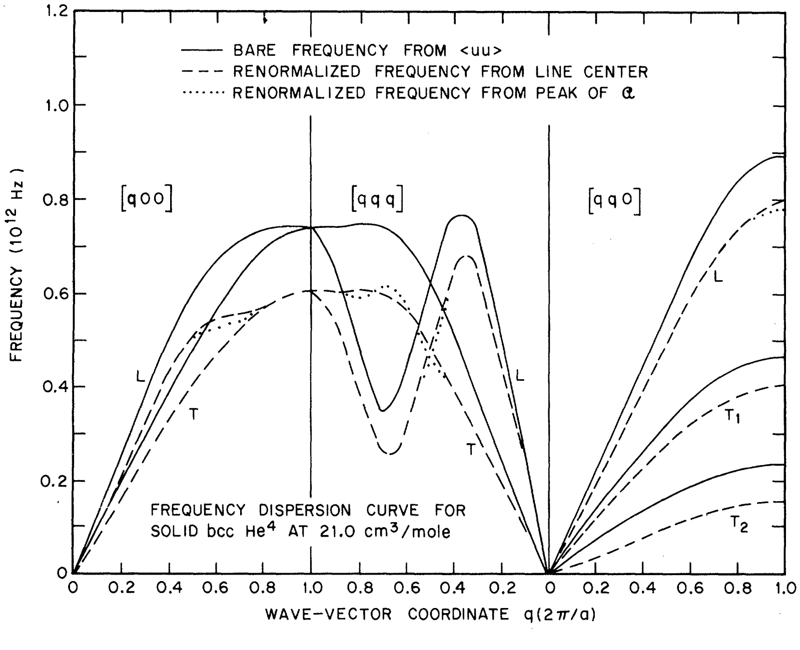

FIG. 5. Phonon frequency dispersion curves similar to Fig. 4, but for bcc He^4 at 21.0 cm^3/mole.

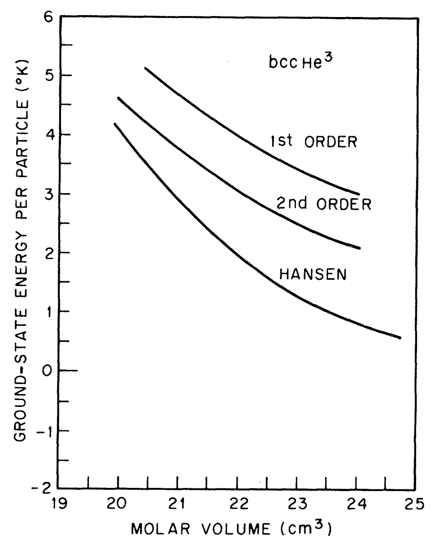

FIG. 6. Ground-state energy per particle for bcc He^3 as a function of molar volume, using the present theory to first and to second order, and a comparison with the Monte Carlo results of Hansen (Ref. 6).

extracted from our computations with little additional effort. Long-wavelength extrapolation of our renormalized phonon frequencies leads to elastic constants shown in Fig. 7, as a function of molar volume for bcc He^3. Here we compare our results with experiment, despite possible inadequacy of the Lennard-Jones potential, in the belief that this will not be the primary cause of discrepancy. The squares give the measurements of Greywall[7] at 21.6 cm^3/mole, while the circles show the results inferred by Wanner.[8] Agreement both in magnitude and in trend with molar volume may be considered as satisfactory (within order of 15%) considering the approximations involved. Further insight into the effect of these approximations may be gained from the compressibility, shown in Fig. 8, as calculated in two different ways: from the ground-state energy of Fig. 6 and from the elastic constants of Fig. 7. Experimental data equivalent to the elastic constants of Greywall[7] and of Wanner,[8] are also shown. The order of 30% inconsistency between the calculations can be traced to two sources, first the neglect of interactions between different two-phonon states [as represented by the second line of Eq. (2.21)], and second to the manner in which short-range correlations have been superimposed, by Jastrow functions of the Nosanow form,[9] onto the long-range phonon correlations. The latter cause of inconsistency is related to the differences which arise in our treatment between the self-consistent frequencies and the phonon frequencies, which has been discussed at some length by Horner.[10]

IV. DISCUSSION

One of the most unusual features of the phonon spectrum of bcc helium is the extraordinarily low frequencies of the lower transverse branch (T_2) in the [110] direction. This situation has been deduced from experimental observations both by Greywall[7] and by Wanner,[8] and also has been found in previous theoretical work by Horner[2] and by Glyde *et al.*[3] However, the existing theoretical calculations have been in considerable quantitative disagreement with each other, as well as with the present work. Although this disagreement might be due to differences in details of the numerical techniques employed, it is our belief that the predominant source is the differences in effective po-

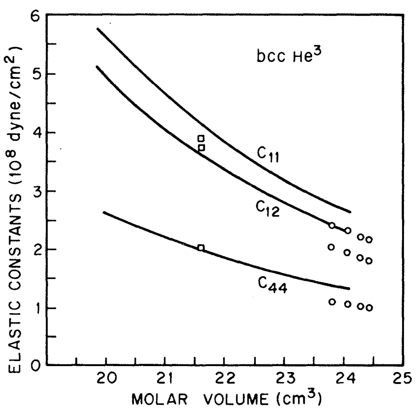

FIG. 7. Elastic constants for bcc He^3 as a function of molar volume and a comparison with the experimental values of Greywall (Ref. 7, squares) and Wanner (Ref. 8, circles).

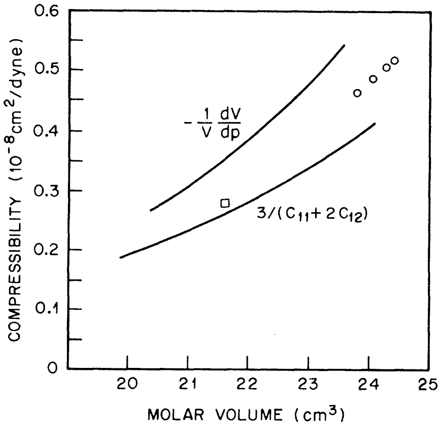

FIG. 8. Compressibility of bcc He^3 as a function of molar volume, calculated both from the elastic constants of Fig. 7 as well as from differentiations of the ground-state energy of Fig. 6, together with experimental values.

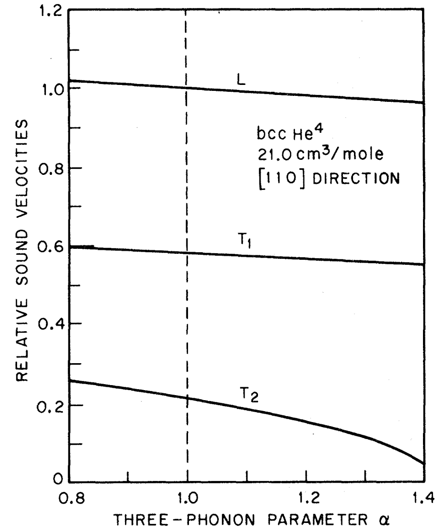

FIG. 9. Sound velocities of bcc He^4 at 21.0 cm^3/mole in the [110] direction, as a function of a three-phonon process parameter α, scaled relative to the longitudinal velocity at $\alpha = 1$.

tentials which are used to evaluate the three-phonon vertex. Rather than to review the analytical formulas employed by each of the respective authors or to reproduce their computations, we choose to study this question by inserting a phenomenological factor α multiplying the three-phonon part of the self-energy in Eq. (2.28) and by varying α away from the nominal value of unity. The way in which the parameter α is introduced corresponds to simulating changes in matrix elements $\langle \nabla\nabla\nabla v\rangle$ via an over-all scaling by a factor $\alpha^{1/2}$. Sound velocities in the [110] direction for bcc He^4 are shown in Fig. 9 as a function of α, with the velocities scaled relative to the longitudinal velocity at $\alpha = 1$. It is remarkable how sensitive is the T_2 velocity to the three-phonon interaction, an increase of 20% in $\langle \nabla\nabla\nabla v\rangle$ producing a drop in the T_2 velocity by more than a factor of 2. We believe that variations of this size are more than sufficient to explain the differences between the theoretical treatments.

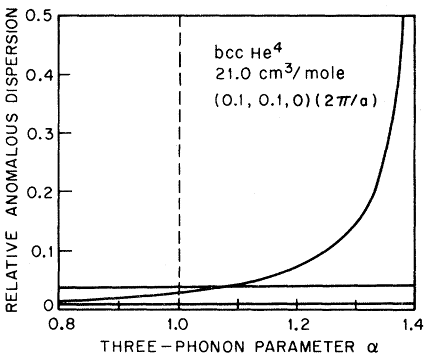

FIG. 10. Relative anomalous dispersion of phonons in the [110] direction of bcc He^4 at 21.0 cm^3/mole, parameterized by the quantity δ defined in the text, as a function of the three-phonon parameter α.

The earlier investigations, particularly that of Horner,[2] also emphasize anomalous dispersion of the T_2 branch in the [110] direction and the significance of this dispersion for understanding the unusual temperature dependence observed for bcc He^3 in the low-temperature specific heat. We parametrize this dispersion by a deviation δ from linearity at small wave vector,

$$\delta \equiv [\omega(0.2, 0.2, 0)/2\omega(0.1, 0.1, 0)] - 1;$$

δ is plotted vs α for each of the three branches in Fig. 10. Whereas the dispersion is light for the L and T_1 branches, the T_2 branch develops dramatic upward dispersion for those $\alpha > 1$ where the T_2 velocities are substantially reduced. Again the results of Horner[2] can be understood on the basis of a slightly larger three-phonon vertex than in the present work. It would seem that a good quantitative understanding of the unusual low-temperature thermodynamic properties of bcc helium, as well as of the large elastic anisotropy, depends critically on a reliable calculation of the three-phonon vertex, even though most of the phonon modes in this structure are rather insensitive to this process.

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