ONE-PHONON SPECTRAL FUNCTION FOR SOLID HELIUM

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One-Phonon Spectral Function For Solid Helium*

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Abstract

Approximate numerical calculations are used in order to understand some of the differences in size and shape of secondary structure obtained in calculations of the one-phonon spectral function for solid helium which have been reported in the literature. It is seen that Horner's self-consistent treatment of the bubble diagram significantly increases the size of such structure over that obtained in perturbative calculations such as used by Glyde and co-workers. A computational procedure used by Horner in evaluating the spectral function may also further enhance the appearance of secondary peaks in the tail region. Previous explanations of these secondary peaks are reviewed and criticised. Other calculations of the one-phonon spectral functions for the solid heliums are briefly discussed.

I. Introduction

The interpretation by Horner¹ of recent inelastic neutron scattering experiments² on bcc ⁴He points to a pronounced effect of anharmonicity on the van Hove structure factor, via rather sizeable interference terms and distorted one-phonon spectral functions. Such effects are expected to be more severe in the case of the solid heliums than for other solids due to the small atomic mass and the weak binding forces in these solids - i.e. they are quantum crystals. A number of rather sophisticated calculations of the one-phonon spectral function for these solids exist,³⁻⁶ with somewhat conflicting results: The work of Horner³ has for some years yielded distorted one-phonon spectral functions, with well defined secondary peaks. Goldman et al.⁴ have encountered similar effects for longitudinal phonons near the zone boundary in their calculations for neon and fcc ⁴He. Koehler and Werthamer⁵ have not reported secondary peaks, although they have obtained rather broad and asymmetric profiles for many longitudinal phonons. The work of Glyde and co-workers⁶ has yielded relatively symmetric spectral functions with only small high frequency tails.

Secondary peaks in the one-phonon spectral function have been encountered in finite temperature calculations for other solids,⁷ and the qualitative manner in which they arise from the phonon self-energy has been demonstrated.⁷ Detailed discussions of the possible origin of the relevant features in the self-energy have been given by Leath and Watson⁸ and by Ruvalds.⁹ We feel that the secondary peaks seen by Horner³ and by Goldman et al.⁴ originate in completely different mechanisms. Thus while

Ruvalds' explanation may be appropriate to the work of Goldman et al., we feel that neither explanation is relevant to the work of Horner. It is the purpose of this paper to come to some understanding of the mechanisms responsible for the substantial distortion, and in particular the secondary peaks, obtained by Horner. Since the approach used by Glyde and co-workers⁶ is in many ways quite similar to that of Horner, we have based our discussion on an approximate numerical comparison of these two methods. We comment on the other papers in the light of these results, including a brief comment on Horner's analysis of the neutron scattering results.

II. Mathematical Background

The thermodynamic one-phonon Green's function is defined as usual¹⁰ by

$$G_{\alpha\beta}(\ell, \ell'; t) = -i \text{Tr} (T u_{\ell\alpha}(t) u_{\ell'\beta}(0) e^{-\beta H}) / \text{Tr} e^{-\beta H} \quad (1)$$

where H is the Hamiltonian of the system, $u_{\ell\alpha}$ is the α^{th} component of the displacement of atom ℓ from its lattice site, and T is the time ordering operator. It is customary to work with a transformation of this function which may be written $G_{jj}(\vec{k}, z)$, where $\vec{k}$ and j are the phonon wave vector and polarization indices, and z the complex frequency. For our purposes it will be sufficient to consider $G_{jj}(\vec{k}, z)$ diagonal in the polarization indices. Dyson's equation relates the Green's function to the self-energy, $\sigma_j(\vec{k}, z)$:

$$z^2 G_j(\vec{k}, z) = 1 + \sigma_j(\vec{k}, z) G_j(\vec{k}, z). \quad (2)$$

Lattice dynamical theory is usually conceived in terms of coupling parameters, or force constants, $\phi^{(n)}$, which are formally defined as the coefficients of a Taylor series expansion of the potential:

$$V = \sum_{n=0}^{\infty} \frac{1}{n!} \phi^{(n)}_{\alpha_1 \dots \alpha_n}(\ell_1 \dots \ell_n) u_{\ell_1 \alpha_1} \dots u_{\ell_n \alpha_n}. \quad (3)$$

The Einstein summation convention has been used. By the use of Eq. (3), the self-energy may be re-expressed in terms of the Green's function, thus completing the set of self-consistent equations. Some of the relevant diagrams for the self-energy are shown in Fig. 1. Finally, the one-phonon spectral function may be defined by

$$A_j(\vec{k}, \omega) = \text{Im} \lim_{\epsilon \rightarrow 0} G_j(\vec{k}, \omega - i\epsilon). \quad (4)$$

In the conventional approach applied to weakly anharmonic crystals, such as the alkali halides, it is presumed that the dominant contribution to the self-energy comes from the two-phonon vertex, Fig. 1a, with the leading anharmonic correction being the bubble diagram, Fig. 1d. The contribution of the bubble diagram is usually evaluated perturbatively, as for example in the paper by Cowley and Cowley.⁷ This procedure is unsatisfactory even at zero temperature for the light rare gas solids, because of their relatively large zero point motion. The self-consistent harmonic (SCH) approximation, used successfully for neon,¹¹ takes the leading contribution to the self-energy as the sum of all diagrams in the first line of Fig. 1. This sum serves to define a renormalized two-phonon vertex. The next most significant contribution is taken to be the bubble diagram, with similarly renormalized three-phonon vertices. Klein et al.¹² have pointed out, however, that the chain diagrams, Fig. 1h, and all similar diagrams with more links in the chain, are necessary to properly calculate the isothermal elastic constant.

In an equivalent point of view, one may consider the SCH approximation to be based on an expansion of the form of Eq. (3) in terms of the (renormalized) force constants, $\phi^{(n, \text{SCH})}$. These are obtained as thermal averages of the derivatives of the many-body potential, using the density matrix derived from a harmonic Hamiltonian with force constants $\phi^{(2, \text{SCH})}$. Even this approach fails for the highly anharmonic quantum crystals, e.g. the solids of helium. These averages diverge for potentials such as the Lennard-Jones, due to the inadequate treatment of

the short-range correlations between atoms in the averaging procedure. One apparent solution to this problem would be to perform the averaging with the true density matrix, which Meissner¹³ has shown leads to a renormalized two-phonon vertex summing all instantaneous diagrams, e.g. also those such as Fig. 1f omitted in the SCH renormalization. However, he has also noted that such renormalized quadratic force constants would lead to frequencies about a factor of two larger than experiment - not an optimal starting point if perturbative calculations are desired. This problem has been treated in a variety of ways which generally result in using derivatives of an effective potential in SCH averages. The precise nature of the resultant renormalized force constants, $\phi^{(n,R)}$, is not clear, although it is probable that some, but not all, of the instantaneous diagrams omitted by the original SCH resummation have been incorporated.

The work of Glyde and co-workers,⁶ Horner,³ and of Koehler and Werthamer⁵ are all essentially zero temperature calculations of the contribution, $\sigma_j^{(1)}(\vec{k}, z)$, of the bubble diagram - preceded, of course, by determination of the renormalized force constants. The calculations of Goldman et al.⁴ include all of the chain diagrams. Glyde and co-workers present a straight forward perturbation calculation ($\lambda = \vec{k}j$)

$$\sigma^{(0)}(\lambda, z) = w^{(R)}(\lambda)^2 = \phi_{jj}^{(2R)}(-\vec{k}, \vec{k}) \quad (5)$$

$$\sigma^{(1)}(\lambda, z) = \frac{\hbar\pi^2}{8} \sum_{\lambda_1, \lambda_2} \frac{|\phi^{(3R)}(\lambda, \lambda_1, \lambda_2)|^2}{w^{(R)}(\lambda_1)w^{(R)}(\lambda_2)} \left[\frac{1}{z - w^{(R)}(\lambda_1) - w^{(R)}(\lambda_2)} \right]$$

$$= \frac{1}{z + \omega^{(R)}(\lambda_1) + \omega^{(R)}(\lambda_2)} \Bigg] \quad (6)$$

$$\sigma^{(1)}(\lambda, \omega - i0^+) = \Delta(\lambda, \omega) + i\Gamma(\lambda, \omega) \quad (7)$$

$$A(\lambda, \omega) = \frac{\Gamma(\lambda, \omega)}{(\omega^2 - \omega^{(R)}(\lambda))^2 - \Delta(\lambda, \omega)^2 + \Gamma^2(\lambda, \omega)} \quad (8)$$

$$\text{where } \phi^{(nR)}(k_1 \cdots k_n) = \phi^{(nR)}(\ell_1 \cdots \ell_n) T_{\ell_1 \alpha_1, k_1 j_1} \cdots T_{\ell_n \alpha_n, k_n j_n} \\ j_1 \cdots j_n \quad \alpha_1 \cdots \alpha_n \quad (9)$$

$$T_{\ell \alpha, k j} = \frac{1}{\sqrt{mN}} \hat{e}_{\alpha, k j} e^{i \vec{k} \cdot \vec{R}_\ell} \quad (10)$$

with $\hat{e}_{k j}$, the polarization vectors; and $\vec{R}_\ell$, the ℓ^{th} lattice vector. For reasons which will become clear later on, we have multiplied the force constants $\phi^{(3R)}$ by a weight factor η in Eq. (6). For Glyde's work η would then be equal to one. The work of Horner approximates a self-consistent solution of the coupled Green's function equations including the bubble diagram. He solves essentially Eqs. (5)-(10) with the frequencies $\omega^{(R)}(\lambda)$ in Eq. (6) replaced by the line centers given by

$$\bar{\omega}(\lambda) = \int_0^\infty d\omega \omega A(\lambda, \omega) / \int_0^\infty d\omega A(\lambda, \omega). \quad (11)$$

Koehler and Werthamer⁵ use an approach which is formally rather different from either of those just mentioned. They arrive at an expression similar to Eq.(6) by diagonalizing the Hamiltonian in a rather special manifold of a variational ground state, and appropriate one-and two-phonon states. As far as the evaluation

of the spectral function $A(\lambda, \omega)$, Eq. (8), is concerned, they also take into account $\sigma^{(1)}(\lambda, z)$ in a perturbative way.

The rather special mathematical techniques necessitated for treating anharmonicity in the quantum crystals tend to obscure simple comparison to other solids. Some of this complexity signals only the inadequacy of a truncated Taylor series expansion to the potential as a theoretical starting point, and is not of physical consequence. The experimentalist, after all, does not see pathological differences between the solid heliums and heavier rare gas solids. It is perhaps more physical, then, to make a comparison at the level of the (renormalized) force constants defining an expansion of the form of Eq. (3). For all of the solids we have mentioned, the (renormalized) quadratic terms lead to frequencies in rough agreement with experiment, and the higher order terms are in some sense relatively small perturbations on this quadratic term. Furthermore, the relative size of $\phi^{(3,R)}$ compared to $\phi^{(2,R)}$ is about the same for the quantum crystals as for the heavier rare gas solids, as can be judged from calculations of the Grüneisen constant which yield about the same result in each case, i.e. on the order of two.⁶ It is the small atomic mass and weak binding forces (leading to small $\phi^{(2,R)}$) of the quantum crystals which result in even the ground state wave function "seeing" more of the anharmonic terms in the (renormalized) expansion in Eq. (3) than is the case for the non-quantum solids where both of these quantities are larger. This means, of course, that comparable anharmonicities in the non-quantum solids occur in general at much higher temperatures where

exited states of comparable "width" are statistically important.

III. Previous Explanations

The calculations of Cowley and Cowley⁷ have yielded a highly distorted spectral function for one of the optical modes in high temperature sodium iodide (See Figs. 7 and 9 of their paper). Consider the spectral function written as in Eq. (8), with the unrenormalized harmonic frequencies, $\omega^{(o)}(\lambda)$, replacing the $\omega^{(R)}(\lambda)$. They have observed that a large peak in the joint density of states would lead to a rapidly fluctuating $\Delta(\lambda, \omega)$ which might intersect the curve $\omega^2 - \omega^{(o)}(\lambda)^2$ to the high frequency side of the main peak. Depending on the size of $\Gamma(\lambda, \omega)$, this may give a secondary peak in $A(\lambda, \omega)$. A less restrictive statement is simply that a relatively strong minimum in the function $(\omega^2 - \omega^{(o)}(\lambda)^2 - \Delta(\lambda, \omega))^2$ will suffice (again conditional on the size of $\Gamma(\lambda, \omega)$), as is evident from the third peak in their Fig. 7. In addition to secondary peaks, some of the spectral functions calculated by Cowley and Cowley show rather asymmetric main peaks, having steep low frequency sides and relatively slowly decaying high frequency sides. This form of asymmetry is well known to occur when the peak overlaps the region of initial rapid increase of $\Gamma(\lambda, \omega)$. Weaker forms of either or both kinds of distortion can lead to a rather symmetric main peak, with a small tail to the high frequency side.

The above comments are generally applicable once the self-energy is given. The question then concerns the origin of sharp features in the self-energy. Leath and Watson⁸ have suggested that structure of sufficient sharpness to explain Horner's results might be due to singularities in the joint

phonon density of states arising from critical points, zeros of $\vec{v}_{\vec{q}} (\omega_j(\vec{q}) + \omega_j(-\vec{q}-\vec{k}))$, of the saddle-point variety. In three dimensional systems, critical points which lead to genuine divergence in the one-(or two-) phonon density of states are in some sense accidental, depending, for example, on special relationships between the force constants. These matters have been discussed in conjunction with the one-phonon density of states.^{14,15} Since Horner has found more or less the same type of subsidiary structure in the spectral function throughout large parts of the Brillouin zone, and for various densities and lattice types of both solid ³He and ⁴He, such pathological behavior can be ruled out. The more usual critical points, whose existence is guaranteed by symmetry, lead of course only to discontinuities in slope of density of state functions in three dimensions.^{16,17} In the absence of real, or near, divergences in the joint phonon density of states, the identification of sharp features with particular critical points as proposed by Leath and Watson would seem to lead to some ambiguity. For it is often the case that a relatively sharp peak will exhibit several discontinuities in slope. Thus it seems clear that one is concerned with the broad overall features of this function, rather than with the finer details. These details are apt to be obscured anyway given the relatively small number of points in $\vec{k}$ space customarily used in evaluating the self-energy.

Ruvalds⁹ has suggested an alternate possible source of sharp structure in the self-energy related to the idea of a two-phonon bound state. This approach requires the inclusion of all chain diagrams in the approximation to the self-energy.

Since Horner's calculations did not include the (renormalized) quartic interaction in any way, this explanation can not be relevant to his work. However, Ruvalds' idea may explain the secondary peaks seen by Goldman et al.⁴ in their calculations for neon, which did include the full sum of chain diagrams.

IV. Model Calculations

In this section we report numerical calculations which appear to explain some of the discrepancy between the work of Glyde and that of Horner with respect to the appearance of subsidiary structure in the spectral function. As our goal is an understanding of these features, and not so much quantitative accuracy, we have used a number of approximations to facilitate numerical computation. We have used the two-branch approximation^{6,18} in evaluating Eqs. (5)-(10), according to which the polarization vectors are taken to be strictly longitudinal or transverse, and the two transverse frequencies are required to be degenerate. The renormalized harmonic frequencies are given in this approximation by

$$m \omega_L^{(R)}(\vec{k})^2 = \sum_{\alpha, \beta} D_{\alpha\beta}(\vec{k}) k_\alpha k_\beta / |\vec{k}|^2 \quad (12a)$$

$$2m \omega_T^{(R)}(\vec{k})^2 = \sum_{\alpha} D_{\alpha\alpha}(\vec{k}) - m \omega_L^{(R)}(\vec{k})^2 \quad (12b)$$

where the dynamical matrix is

$$D_{\alpha\beta}(\vec{k}) = \sum_{\ell=\ell'} e^{i\vec{k} \cdot (\vec{R}_\ell - \vec{R}_{\ell'})} \phi_{\alpha\beta}^{(2R)}(\ell, \ell'). \quad (13)$$

Because the two transverse branches are degenerate throughout the Brillouin zone, Eq. (6) is independent of any ambiguity in choosing the transverse polarization vectors. We have used the renormalized force constants obtained by Glyde and Khanna⁶ (kindly given to us by Dr. H. R. Glyde) for bcc ³He at a volume of 24.0 cc/mole, which were used by these authors in arriving

at Figs. 1 and 4 of their paper. In determining the transformed quadratic and cubic force constants via Eq. (9), we retained only terms through third neighbors and first neighbors respectively. Finally, we have used a grid of 128 points in the full Brillouin zone in evaluating Eq. (6). While this small number of points is clearly not adequate if numerical accuracy is desired, it does suffice for our purposes. We have made selected tests with a grid of 432 points in order to estimate the effect of this approximation on the features of interest which we shall mention in the subsequent discussion.

We have evaluated Eqs. (5)-(10) using the above approximations. Following Glyde and Khanna,⁶ we have also used

$$\left(\omega - i\omega_1 + \omega_2\right)^{-1} \simeq \frac{\omega_1 \omega_2}{(\omega_1 + \omega_2)^2 + \gamma^2} + i \frac{\gamma}{(\omega_1 + \omega_2)^2 + \gamma^2} \quad (14)$$

where we have taken γ to be 0.088THz. The self-energy curves and spectral function for the zone boundary [100] longitudinal phonon, $\omega_L^{(R)} = 0.689\text{THz}$, are given in Fig. 2. The slight bump in the spectral function near 0.96THz, and the suggestion of a similar feature near 1.20THz, are due to local minima in $\left(\omega^2 - \omega_L^{(R)}(\vec{k})^2 - \Delta_L(\vec{k}, \omega)\right)^2$ as discussed in the previous section. The corresponding peaks in Γ are due to TT and LT intermediate phonons in the bubble diagram. The effect of the LL intermediate phonons is not noticeable for this $\vec{k}$ -vector. When the grid of 432 points is used in evaluating Eq. (6), two well defined peaks in Γ are no longer discernable, although the overall shape of this function remains about the same. The tail of the spectral function is consequently quite flat. When a smaller value of γ (0.042THz) is used with the grid of 432 points, the width of

the main peak in the spectral function is decreased significantly. However, since the low frequency behavior of Δ is much less sensitive to the value of γ , the position of the main peak is not noticeably shifted. The smaller γ appears to slightly adjust weight within the tail of the spectral function so as to restore the appearance of the slight bump at about 0.96THz. The overall size of the tail appears about the same, as is also evidenced by the fact that the line center given by Eq. (11) - about 9% higher than the peak position - is unchanged to within better than half a percent. It is on the basis of the facts that the peak position and the line center are not noticeably affected by these choices of grid size and γ that we have judged our original choices to be adequate for the subsequent calculations in this paper. Although the numerical agreement with the more accurate calculations of Glyde and Khanna⁶ (see their Fig. 4) is only about 60% for the self-energy curves, we feel that the qualitative features of their spectral function have been preserved here. That is, the distortion consists of a small and relatively flat tail to the high frequency side of the main peak.

Horner³ has noted that one of the more important differences between his work and that of Glyde and co-workers is his approximate self-consistent treatment of the bubble diagram as described on page 7. With this in mind, we took the results of the previous paragraph as a first iteration, using the line centers in place of the $w_j^{(R)}(\bar{k})$ in Eq. (6) for the second iteration, and so on. We were unable to obtain convergence. After the third iteration, the line centers had been reduced

so severely that the spectral function for the transverse mode for one $\vec{k}$ -vector did not exhibit a main peak, because $\omega^{(R)}(\lambda)^2 + \Delta(\lambda, 0) < 0$. Successful converge was obtained, however, when the renormalized cubic force constants were reduced slightly in size, corresponding to $\eta < 1$ in Eq. (6).

The sensitivity of Horner's self-consistent scheme to the size of the cubic force constants may be illuminated by the following somewhat artificial device: Let us assume that in the course of a given iteration, the line center of each mode is altered by the same fractional amount. One may then solve Eqs. (6), (8) and (11) self-consistently for the single parameter α , where $\tilde{\omega}_j(\vec{k}) = \alpha \omega_j^{(R)}(\vec{k})$. This calculation has been performed using the self-energy curves shown in Fig. 2, scaled by the weight factor η^2 . The results are shown in Fig. 3. Both the line center and peak position are indicated. For comparison, the equivalent curves for the perturbative, i.e. first iteration, solution are also shown. In general there appear to be either two or no solutions, although in the former case the smaller of the two solutions is computationally unstable and we shall not further consider it.¹⁹ Bearing in mind the rather artificial nature of this example, a number of observations may be made: (1) About a factor of three in size of the cubic force constants distinguishes between a regime in which the self-consistent and perturbative solutions are only negligibly different, and a regime where there is no self-consistent solution at all. (2) The self-consistent solution consistently gives a more distorted spectral function than the perturbative solution, as is evident from the difference between the line centers and peaks. (3) There is

no self-consistent solution for Glyde and Khanna's cubic force constants, which correspond to $\eta^2=1$. Horner has kindly given us his force constants for bcc ^3He at a volume of 23.8 cc/mole. His renormalized harmonic frequencies are about 10% larger than those of Glyde and Khanna, while his cubic force constants are about 30% smaller. Extending the above scaling arguments, these values correspond to $\eta^2 \sim 0.3$ in Fig. 3, and so there should be a self-consistent solution.

With the above ideas in mind, we repeated the full calculation with $\eta^2=0.55$. Spectral functions for some of the [100] longitudinal phonons are shown in Figs. 4 and 5 for the perturbative and self-consistent solutions respectively. It is evident that the self-consistent solution has enhanced the distortion of these profiles. For example, the fraction of the first moment sum rule used up by the main peak alone is 81% and 66% respectively for the zone boundary phonons in Figs. 4 and 5. Since the low frequency part of Δ is negative, peaks of the spectral functions will generally occur at lower frequencies than those which were used in place of the $\omega^{(R)}(\lambda)$ in Eq. (6). In order for Horner's self-consistent scheme to converge, it is necessary for there to be enough spectral weight to the high frequency side of the peak to make up this difference. The iterative procedure augments the self-energy in size, while shifting it downward in frequency closer to the curve $\omega^2 - \omega^{(R)}(\lambda)^2$. This permits stronger local minima, or simply smaller values, of the function $(\omega^2 - \omega^{(R)}(\lambda)^2 - \Delta(\lambda, \omega))^2$, which enhances subsidiary structure in the spectral function.

Some question can be raised about Horner's technique in that other moments could have been used in defining the $\tilde{w}(\lambda)$ used in Eq. (6). On the other hand it is quite plausible that the qualitative features seen here - enhancement of subsidiary structure in the spectral function, and the absence of convergence for overly large cubic force constants - are true for the fully accurate self-consistent treatment of the bubble diagram.

A second important difference between the work of Horner and that of Glyde concerns a matter of computational technique. In the preceding calculations, as in those of Glyde and co-workers, the spectral function has been approximated by

$$A(\lambda, \omega) \simeq \frac{\Gamma_Y(\lambda, \omega)}{\left(\omega^{2-\omega(R)}(\lambda)^2 - \Delta_Y(\lambda, \omega) \right)^2 + \Gamma_Y^2(\lambda, \omega)} \quad (15)$$

where $\Delta_Y(\lambda, \omega)$ and $\Gamma_Y(\lambda, \omega)$ are the real and imaginary parts of $\sigma^{(1)}(\lambda, \omega - iY)$, for a small but finite Y . Horner³ has used an alternate method. For a finite grid in $\vec{k}$ -space, the Green's function

$$G(\lambda, z) = \frac{1}{z^{2-\omega(R)}(\lambda)^2 - \sigma^{(1)}(\lambda, z)} \quad (16)$$

is analytic everywhere except for a finite number of poles at frequencies $z = \pm \Omega_i$ on the real axis. One may then write

$$G(\lambda, z) = \sum_i \frac{W_i}{z^2 - \Omega_i^2} \quad (17)$$

where

$$W_i^{-1} = 1 - \left(\frac{\partial}{\partial(z^2)} \sigma^{(1)}(\lambda, z) \right) \Big|_{z = \Omega_i} \quad (18)$$

since the right hand side of Eq. (16) less that of Eq. (17) is analytic everywhere and must therefore be a constant - which may be seen to be zero. From Eq. (17) the spectral function is given by

$$A(\lambda, \omega) = \sum_i \frac{W_i}{2\Omega_i} \left[\delta(\omega - \Omega_i) - \delta(\omega + \Omega_i) \right]. \quad (19)$$

Horner evaluates Eq. (19) folded by a Gaussian of appropriate width. When this technique is used to calculate the spectral function, the results of the self-consistent calculation with $\eta^2=0.55$ are as shown in Fig. 6. These results are to be compared to those in Fig. 5 obtained using the more conventional approximation to the spectral function given by Eq. (15).

Although the main-peak positions and the line centers of the profiles shown in Figs. 5 and 6 agree to better than a percent, an adjustment of spectral weight within the tail region, which enhances the appearance of secondary peaks, is evident in the latter. This enhancement is quite sensitive to the function used in the folding procedure. We have used a Gaussian, $\exp[-(\omega - \omega')^2/d^2]$, with a width $d=0.063\text{THz}$, consistent with that used by Horner. While Fig. 6 was calculated using a $\mathbf{k}$ -space grid of 128 points, the zone boundary phonon spectral function was also calculated with a grid of 432 points with essentially the same result. Horner used a grid of 512 points.

Some insight into the difference between the two computational techniques may be gained by folding Eq. (19) with a Lorentzian, $\gamma/[(\omega - \omega')^2 + \gamma^2]$. In order to get secondary structure of about the same sharpness and size as seen in Fig. 6, it was necessary to take γ as small as 0.02THz . In this case,

the source of the sharpness of the secondary structure is clear. One is evaluating $\text{Im } G(\lambda, \omega - i\gamma)$, or equivalently

$$A(\lambda, \omega) \simeq \frac{\Gamma_Y(\lambda, \omega) + 2\omega\gamma}{\left(\omega^2 - \omega^{(R)}(\lambda)^2 - \Delta_Y(\lambda, \omega) - \gamma^2\right)^2 + \left(\Gamma_Y(\lambda, \omega) + 2\omega\gamma\right)^2}. \quad (20)$$

The value of $\gamma = 0.02\text{THz}$ is too small for the grid size we have been using, so that the functions $\Delta_Y(\lambda, \omega)$ and $\Gamma_Y(\lambda, \omega)$ are rapidly fluctuating. This leads to rather sharp secondary peaks in the manner described previously. The breadth of the main peak is also understood in this calculation to be largely due to the additional term, $2\omega\gamma$, in Eq. (20).

We feel that the results shown in Fig. 6 are in sufficient qualitative agreement²⁰ with those reported by Horner³ for bcc ⁴He, to suggest that two factors contributing to the size and sharpness of the secondary peaks he has obtained in the one-phonon spectral function are (1) his self-consistent treatment of the bubble diagram and (2) the difference of computational technique just discussed. The effect of these factors on the real part of the Green's function may be seen in Fig. 7. We present in this figure the Hilbert transforms of the zone boundary profiles given in Figs. 4, 5, and 6.

V. Discussion

Calculations of the one-phonon spectral function for the solid heliums³⁻⁶ have generally yielded rather broad and asymmetric profiles for longitudinal phonons near the zone boundaries. The perturbative treatments of the bubble diagram used by Glyde and co-workers⁶ and by Koehler and Werthamer⁵ have led to spectral functions with relatively smooth high frequency tails extending, in the former case, to frequencies well beyond the position of the main peak. Our results suggest that the significantly larger structure obtained by Horner³ at these high frequencies is due mostly to his having treated the bubble diagram in a self-consistent manner.²¹ The basic effect of this procedure is to augment the self-energy in size, and to shift it to lower frequencies. This serves to increase the size of the tail, and may give weak secondary peaks corresponding roughly to peaks in the imaginary part of the self-energy. The self-consistent solution is extremely sensitive to the size of the cubic force constants, and in fact may not exist if these constants are too large. We feel that the sharpness of the subsidiary structure obtained by Horner, its appearance as well defined secondary peaks, is partially due to the manner in which he has used Eq. (19) to evaluate the spectral function. This technique has not affected the overall size of the high frequency tail, but appears to have adjusted spectral weight within it. We suggest that the secondary peaks reported by Goldman et al.⁴ are due to completely different mechanisms, since their work differs from that of Horner in both the matter of self-consistency and in the manner in which the

spectral function is evaluated. Since they have included all of the chain diagrams, Ruvalds' explanation may apply.

Horner has based his analysis of the experimentally determined Debye-Waller factor for bcc $^4\text{He}^2$ on the points that (1) the pure one-phonon spectral function has a tail besides the more or less sharp peak, and (2) that as far as the van Hove structure factor is concerned, the interference terms may be viewed as redistributing intensity within this pure one-phonon line including the tail. We have seen in this paper that his approximate self-consistent treatment of the bubble diagram increases the size of this tail. The factors which multiply both the real and imaginary parts of the one-phonon Green's function in a calculation of the interference terms can be expected to be increased by this self-consistent procedure in much the same way that the self-energy was increased.

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19. The lower frequency solution corresponds to the main structure of the self-energy having been shifted to sufficiently low frequencies that $\omega^2 - \omega^{(R)}(\lambda)^2$ intersects $\Delta(\lambda, \omega)$ to the high as well as the low frequency side of the region where $\Gamma(\lambda, \omega)$ is sizeable. Consequently, the spectral function in this case consists essentially of two sharp peaks at these frequencies.
20. In Ref. 3, Horner has plotted $\omega \cdot A(\lambda, \omega)$, which enhances the appearance of the secondary structure. This fact is not completely clear in the text of that paper. In Ref. 1, the neutron scattering work, Horner plots $A(\lambda, \omega)$. (Private communication, H. Horner).
21. It is also Horner's belief that his self-consistent treatment is the source of the large subsidiary structure he obtains in the spectral function. (Private communication, H. Horner).

Figure Captions

- Figure 1: Diagrammatic expansion of the self-energy, σ . The intermediate lines are full Green's functions, and the vertices are unrenormalized.
- Figure 2: The one-phonon spectral function (dashed line), $A(\lambda, \omega)$; self-energy curves (solid lines), $\Delta(\lambda, \omega)$ and $\Gamma(\lambda, \omega)$; and the function $\omega^2 - \omega^{(R)}(\lambda)^2$ (dash-dot line) for the zone boundary [100] longitudinal phonon. The bubble diagram has been treated perturbatively. $\eta^2 = 1.0$.
- Figure 3: Comparison of phonon frequencies as obtained by the approximate self-consistent and by the perturbative treatment of the bubble diagram. A simple scaling model for the frequencies has been used. Frequencies corresponding to the line centers (solid lines) and to the main peaks (dashed lines) are taken to be $\alpha \omega^{(R)}(\lambda)$, and given in terms of the parameter α . The parameter η indicates the relative size of the cubic force constants (see Eq. (6)) and is unity for the choice of Glyde and Khanna.
- Figure 4: The spectral functions for selected longitudinal phonons in the [100] direction. The bubble diagram has been treated perturbatively. The spectral function has been evaluated using Eq. (15). Arrows indicate the line centers. $\eta^2 = 0.55$.
- Figure 5: The spectral functions for selected longitudinal phonons in the [100] direction. The bubble diagram has been treated self-consistently. The spectral

function has been evaluated using Eq. (15). Arrows indicate line centers. $\eta^2=0.55$.

Figure 6: The spectral functions for selected longitudinal phonons in the [100] direction. The bubble diagram has been treated self-consistently. The spectral function has been evaluated using Eq. (19) folded by a Gaussian. Arrows indicate line centers. $\eta^2=0.55$.

Figure 7: The Hilbert transforms of the zone boundary profiles in Fig. 4 (dotted line), Fig. 5 (dashed line), and Fig. 6 (solid line).

$$\begin{aligned}
 \sigma = & \text{(a)} \text{---}\bullet\text{---} + \text{(b)} \text{---}\bullet\text{---} + \text{(c)} \text{---}\bullet\text{---} + \dots \\
 & + \text{(d)} \text{---}\bullet\text{---} + \text{(e)} \text{---}\bullet\text{---} + \text{(f)} \text{---}\bullet\text{---} + \text{(g)} \text{---}\bullet\text{---} + \dots \\
 & + \text{(h)} \text{---}\bullet\text{---} + \dots
 \end{aligned}$$

Fig. 2

Fig. 4

Fig. 6