I. INTRODUCTION

Eckert et al. and Thomlinson et al. have recently made an interesting and detailed study of the lattice dynamics of fcc $^4$He at high pressure by means of coherent inelastic neutron scattering. In this paper we present calculations of the dynamic structure factor $S(Q, \omega)$ and the one-phonon properties of fcc $^4$He observed in these experiments employing the self-consistent phonon (SCP) theory without short-range calculations. The purpose is first to test by comparison with experiment how well the standard SCP theory can describe this moderately anharmonic crystal. This includes a comparison of $S(Q, \omega)$ itself and the one-phonon properties such as the phonon frequencies and lifetimes and the change of these with temperature. It is well known, for example, that the bcc and hcp phases of helium, which occur at lower pressure, cannot be well described without taking account of the short-range correlations in the atomic vibrations. The reader is referred to substantial review of this interesting additional problem, for example, by Horner, Koehler, Glyde, and Varma and Werthamer.

The second purpose is to test the usual expansions of $S(Q, \omega)$ as a power series in scattering from single phonons, $S_1(Q, \omega)$, from two phonons, $S_2(Q, \omega)$, and from higher multiples of phonons as well as possible interference between these contributions. This expansion is useful if $S_1(Q, \omega)$ dominates all the other terms and is readily identifiable, sharply peaked function of frequency with the remainder forming on a generally frequency-independent background. The frequency and lifetime of the single phonon selected in the scattering can then be readily determined. The aim here is to identify the contributions that $S_1(Q, \omega)$, $S_2(Q, \omega)$, and interference make to the total observed $S(Q, \omega)$. We shall see that even at moderate momentum transfers $\hbar Q$ the $S_1(Q, \omega)$ is large and has a significant frequency dependence right in the frequency range of the higher-frequency longitudinal phonons of fcc $^4$He. This makes the scattering from single and pairs of phonons difficult to separate and explains why Eckert et al. and Thomlinson et al. were unable to determine the frequency of the higher-energy longitudinal phonons.

The paper is divided into six parts. Section II outlines the usual expansion of $S(Q, \omega)$ and the SCP theory in the form employed for the present calculations. Section III presents the results for $S(Q, \omega)$ in order to identify the one-phonon, the higher-order, and the interference contributions to it. The calculation of the one-phonon properties alone and the interference between the one- and two-phonon parts of $S(Q, \omega)$ are presented in Sec. IV and V, respectively. The results are discussed in Sec. VI.

II. THEORETICAL BACKGROUND

A. Dynamic form factor

In this section we briefly survey the form of $S(Q, \omega)$ used in the present study. $S(Q, \omega)$ is defined as the time Fourier transform

$$S(Q, \omega) = \int dt \, e^{i \omega t} \overline{S(Q, t)}$$

of the correlation function

$$\overline{S(Q, t)} = \frac{1}{N} \langle \rho(Q, t) \rho(-Q, 0) \rangle$$

of the particle density

$$\rho(Q, t) = \sum \exp[-iQ \cdot \mathbf{x}(t)].$$
The sum is over the positions \( r_i \) of all \( N \) atoms in the crystal. The observed coherent inelastic scattering cross section in which a neutron transfers a momentum \( h \mathbf{Q} \) and energy \( h \omega \) to (or from) the solid is directly proportional to this \( S(Q, \omega) \).

In a solid the atomic motions leading to correlations in density arise chiefly from localized vibrations of each atom \( l \) about its lattice point \( \mathbf{R}_l \).

(5)

Classically, a diffusive jump occurs once for approximately \( 10^3 \) vibrations in a solid near its melting point. We then write the atomic position \( \mathbf{R}_l = \bar{\mathbf{R}}_l + \mathbf{u}_l \) in terms of \( \mathbf{R}_l \) plus a displacement \( \mathbf{u}_l \) that we expect on average to be small. In terms of \( \mathbf{u}_l \),

\[
S(Q, t) = \frac{1}{N} \sum_{r} e^{i \mathbf{Q} \cdot \mathbf{u}_r} e^{-i Q \cdot R} e^{-i t \mathbf{Q} \cdot \mathbf{u}_0}. \tag{1}
\]

\( S(Q, \omega) \) is then expanded in powers of \( \langle Q \cdot \mathbf{u} \rangle \) giving \( Q \)

\[
S(Q, \omega) = S_0(Q) + S_1(Q, \omega) + S_{12}(Q, \omega) + \cdots. \tag{2}
\]

Here \( S_0(Q) \) is the elastic Bragg scattering of no interest here, while all higher orders are chiefly inelastic. The \( S_1(Q, \omega) \), proportional to \( \langle Q \cdot \mathbf{u} \rangle \), describes the scattering process in which a single phonon having wave vector \( \mathbf{Q} = \mathbf{q} \) is created or destroyed by the neutron. For this process, we expect a large resonance in the scattering when the energy \( (h \omega) \) transferred by the neutron equals the energy \( (h \omega_{\mathbf{q}}) \) of the single phonon involved. The \( S_1(Q, \omega) \), proportional to \( \langle Q \cdot \mathbf{u} \rangle \), describes the process in which two phonons are created or destroyed and is usually an uninteresting, uniform function of \( \omega \). \( S_{12}(Q, \omega) \), proportional to \( \langle Q \cdot \mathbf{u} \rangle \), is an interference term between the one- and two-phonon scattering processes due to anharmonic coupling between phonons.\(^{12,14,15}\) This term, since it involves a single phonon, often has an interesting frequency dependence near the single-phonon frequency. Ambegaokar et al.\(^ {12}\) proposed combining \( S_1(Q, \omega) \) and the interference terms involving a single phonon into one term labeled \( S_p(Q, \omega) \). At the present level of approximation,

\[
S_p(Q, \omega) = S_1(Q, \omega) + S_{12}(Q, \omega). \tag{3}
\]

The higher-order processes, \( S_{12}(Q, \omega) + \cdots \), should be uniform functions of \( \omega \) and we hope that higher-order interference terms will be relatively small.\(^ {16}\)

An aim here is to identify the terms in this expansion by comparing the calculated \( S(Q, \omega) \) with the scattering intensity observed by Eckert et al.\(^ {3}\) This should indicate at what point higher-order terms become large and the expansion ceases to be useful. To estimate the expansion parameter \( \langle Q \cdot \mathbf{u} \rangle^2 = \frac{1}{2} Q^2 \langle u^2 \rangle \), we express \( Q \) in reduced units \( Q = 2\pi/a_0 = \alpha \cdot \xi \) where \( \alpha \) is the lattice constant, and \( \langle u^2 \rangle^{1/2} \) as a fraction of \( \delta \) of the interparticle spacing \( R = a/\sqrt{2} \). Then

\[
\langle (Q \cdot \mathbf{u})^2 \rangle = \frac{1}{2} (\langle 2\pi \rangle \langle \xi \rangle^2) \sim \delta (\xi)^2.
\]

For bcc \( ^4 \)He, \( \delta \approx 0.30 \) so that even for \( \xi \sim 1 \), that is, scattering vectors at the edge of the first Brillouin zone, we expect higher-order terms beyond \( S_1(Q, \omega) \) to be important. For fcc \( ^4 \)He, and typical metals near their melting points, where \( \delta = 0.15 \), the higher-order terms may be expected to contribute significantly for \( \xi \gg 2 \).

**B. Phonon dynamics**

The one-phonon dynamic form factor is \( S_1(Q, \omega) \)

\[
S_1(Q, \omega) = \frac{4 \pi}{v} [\delta(\omega) + 1] d^2(Q) \sum_{\lambda, \mathbf{q}} \text{ } S_{\mathbf{q}}(Q) \frac{2 \lambda}{\omega_{\mathbf{q}}^2} \text{ } A(\mathbf{q}; \omega) \text{ } \Delta(Q - \mathbf{q}). \tag{4}
\]

Here \( n(\omega) \) is the Bose function, \( d(\mathbf{q}) \) is the Debye-Waller factor, \( x = \hbar/2m\omega_{\mathbf{q}} \), \( x \) and \( \omega_{\mathbf{q}} \) are the polarization and frequency of the phonon having branch \( \lambda \), and \( m \) is the atomic mass. \( A(\mathbf{q}; \omega) \) is the one-phonon response function.

In the simplest, quasiharmonic (QH) model the phonon frequencies are given by the usual harmonic expression in terms of force constants evaluated as derivatives of the interatomic potential at the lattice points. These phonons have infinite lifetime and the corresponding

\[
A(\mathbf{q}; \omega) = 2 \pi \delta(\omega - \omega_{\mathbf{q}}) \delta(\omega + \omega_{\mathbf{q}}) \text{ } (\omega_{\mathbf{q}}^2 / 2\pi)^2. \tag{5}
\]

The self-consistent harmonic (SCH) frequencies \( \omega_{\mathbf{q}} \) are also given by the usual harmonic expression but not the force constants are evaluated as derivatives of the interatomic potential averaged over the vibrational distribution of the atoms about their lattice points. The vibrational distribution is described by a Gaussian function of width \( \Delta \) consistent with the SCH frequencies. The SCH phonons also have infinite lifetime so that \( A(\mathbf{q}; \omega) \) is again given by (5). In the SCH theory in which the cubic anharmonic term is added as a perturbation \( \Gamma(\mathbf{q}^2 / 2\pi)^2 \) (SCH + cubic),

\[
A(\mathbf{q}; \omega) = 8 \omega_{\mathbf{q}}^2 \Gamma(\mathbf{q}; \omega) \times \left[ -\omega_{\mathbf{q}}^2 + 2 \omega_{\mathbf{q}} \Delta(\mathbf{q}; \omega) \omega_{\mathbf{q}}^2 \right]^{1/2} + [2 \omega_{\mathbf{q}}^2 \Gamma(\mathbf{q}; \omega)]^{1/2}. \tag{6}
\]

Here the \( \omega_{\mathbf{q}} \) are the SCH frequencies, and \( \Delta \) is the frequency shift and \( \Gamma \) the inverse lifetime \( (\tau^{-1} = \Gamma) \) due to the cubic anharmonic term. The SCH + cubic frequency is defined here by the maximum
in $S_1(\bar{Q}, \omega)$ with $A(\omega \lambda, \omega)$ given by (6). If $\Delta$ and $\Gamma$ depend little on frequency, then $A(\omega \lambda, \omega)$ is a simple Lorentzian function having a maximum at $\Omega^2 = \omega^2 \lambda + 2 \omega \omega \Delta(\omega \lambda)$ and an intrinsic full width at half maximum $W = 2 \Gamma(\omega \lambda)$.

For harmonic-like phonons having infinite lifetime the two-phonon dynamic form factor is

$$S_2(\omega, \omega) = [n(\omega) + 1] Z(\omega)$$

$$\times \left[ \sum_{n=1}^{\infty} J(1, 2, \omega) \Delta(\Omega - (n \omega))^2 \right],$$

where

$$J(1, 2, \omega) = n(n+1)$$

$$\times \left[ \delta(\omega - (I \omega + I \omega)) \right]$$

$$+ \eta \delta(\omega - (I \omega + I \omega)) \right] \delta(\omega - (I \omega + I \omega)).$$

Hence, the sum is over a dense enough mesh of phonons $\Omega = \Omega \lambda$ and $\Lambda = (\Omega \lambda - \Omega \lambda)$ so that the two-phonon density of states $J(1, 2, \omega)$ is independent of this density. $J(1, 2, \omega)$ is a general function of $\omega$ since a sum of all frequencies in the crystal is involved. However, at low temperature, where $\hbar \omega \lambda / kT - \Theta / kT \ll 1$ for most phonons, the Bose factors are small. In this case only the first two terms in $J(1, 2, \omega)$ contribute and in, addition, only "energy-loss" ($\omega > 0$) scattering is measured, only the first term contributes. When $S_1(\bar{Q}, \omega)$ is significant we have used the SCH + cubic frequencies, $\Omega_1$ and $\Omega_2$, in (7) ignoring the lifetime effects.

The $S_1(\bar{Q}, \omega)$ including the dominant interference terms may be written in the form

$$S_1(\bar{Q}, \omega) = S(\bar{Q}, \omega) \left[ 1 - 2 \left( A - B \right) \right],$$

where $\Omega_1$ is the one-phonon frequency including anharmonic terms. The $A$ and $B$ are proportional to $\bar{Q}$ and largely frequency independent. The interference thus makes two types of contributions, one largely proportional to $S_1(\bar{Q}, \omega)$, while the second vanishes at the peak of $S_1(\bar{Q}, \omega)$. The sign of both $A$ and $B$ is given by the sign of the reduced wave vector $\vec{q} = \bar{Q} - \bar{F}$, where $\bar{F}$ is the nearest reciprocal-lattice vector. Thus $A$ and $B$ change sign as $\bar{Q}$ passes from being just less than to just greater than the nearest reciprocal-lattice vector. Further discussion of $S_p(\bar{Q}, \omega)$ can be found elsewhere.

Throughout the calculations we have employed the Beck potential to represent the He-He interaction.

### III. CONTRIBUTIONS TO $S(\bar{Q}, \omega)$

In this section we make a direct comparison of the calculated $S(\bar{Q}, \omega)$ with the observed scattering intensity. The aim is to identify the relative importance of the one-phonon $S_1(\bar{Q}, \omega)$, the two-phonon $S_2(\bar{Q}, \omega)$, and interference contributions $S_1(\bar{Q}, \omega)$, given in Eqs. (1) and (2). Figure 1 shows the calculated $S(\bar{Q}, \omega)$ of fcc $^4$He at 9.97 cm$^{-3}$/mole and $T = 38$ K. The $S(\bar{Q}, \omega)$ is clearly dominant in the low-energy ($\omega \omega$) transfer range shown. The difference between $S_p(\bar{Q}, \omega)$ and $S_1(\bar{Q}, \omega)$ suggests a small interference contribution but $S_2(\bar{Q}, \omega)$ is negligible. Each $S_p(\bar{Q}, \omega)$ is convoluted with a Gaussian function of full width at half maximum (FWHM) indicated to simulate the instrument resolution width. The FWHM quoted in Fig. 1 is the total FWHM after the convolution while $W(\omega \omega)$ is the intrinsic width of the one-phonon response given by (4) and (6).

Figure 2 shows $S(\bar{Q}, \omega)$ for phonons having larger reduced wave vectors $q$, but for which the one-phonon frequency is still not large, now for fcc $^4$He at 9.93 cm$^{-3}$/mole and $T = 38$ K. The $S(\bar{Q}, \omega)$ is again clearly identifiable in this frequency range. Comparing the $S(\bar{Q}, \omega)$ with the corresponding observed scattering intensities in Figs. 1 and 2 shows that the data are generally well reproduced by the calculations. Hence, for phonons having low and intermediate energies and observed at total wave-vector transfers of $\bar{Q} = \bar{Q} / a^*$

$=$ $(q, \xi, \xi)$ with $\xi \leq 2$, the scattering is predominantly from single phonons. The observed lifetime of the phonons is also not significantly affected by interference.

Figure 3 shows $S(\bar{Q}, \omega)$ for three longitudinal phonons along the [111] direction having reduced wave vector near the edge of the Brillouin zone. In this case the corresponding phonon frequencies are higher than those phonons displayed in Figs. 1 and 2. In Fig. 3 we note particularly the size and frequency dependence of $S_1(\bar{Q}, \omega)$ in this higher frequency range. At an energy transfer of $\hbar \omega = 15-18$ meV, $S_1(\bar{Q}, \omega)$ increases sharply becoming much larger than $S_1(\bar{Q}, \omega)$. It is really the sharp increase in $S_1(\bar{Q}, \omega)$ which is important since otherwise $S_1(\bar{Q}, \omega)$ could be simply subtracted away as an uninteresting, uniform background even though it is large. A step of this kind in $S_1(\bar{Q}, \omega)$ will lead to observed phonon groups which look like normal one-phonon groups on the low-energy side but have a plateau of intensity near or just below the peak value on the high-energy side. The corresponding observed phonon groups shown in Fig. 3 show this
FIG. 1. Calculated $S(Q, \omega)$ for the transverse phonon at $Q^* = (0.3, 2.0, 2.0)$ and the longitudinal phonon at $Q^* = (2.5, 0, 0)$ in fcc $^4$He at 9.97 cm$^3$/mole and $T = 28$ K compared with the corresponding observed phonon groups of Thomlinson et al. (Ref. 2). The $S(Q, \omega)$ is convoluted with a Gaussian of FWHM of 0.55 meV (1.06 meV) in the transverse (longitudinal) example.

FIG. 2. Same as Fig. 1 for the wave vectors $Q^*$ indicated in fcc $^4$He at 9.03 cm$^3$/mole and $T = 38$ K. The calculated longitudinal (transverse) $S(Q, \omega)$ is convoluted with a Gaussian of FWHM of 1.3 meV (1.0 meV).
FIG. 3. Calculated $S(\mathbf{Q}, \omega)$ and the scattering intensity observed by Thomlinson et al. (Ref. 2) for longitudinal phonons [111]. The peaked, dotted line in the upper right-hand figure shows $S_p(\mathbf{Q}, \omega)$ alone. Each calculated $S(\mathbf{Q}, \omega)$ is convoluted with a Gaussian of resolution width equal to 1.5 meV.

FIG. 4. Right-hand side shows $S(\mathbf{Q}, \omega) = S_p(\mathbf{Q}, \omega) + S_i(\mathbf{Q}, \omega)$, convoluted with a Gaussian of width 1.5 meV and in units of $Q^2d^2(\mathbf{Q})$ for the longitudinal phonons at $\mathbf{Q}^* = (1.8, -1.8, 0)$ and $\mathbf{Q}^* = (2.2, -2.2, 0)$ demonstrating the difference due to interference contributions to $S_p(\mathbf{Q}, \omega)$. The left-hand side shows the observed phonon groups.
effect as well as indicating further multiphonon scattering which further increases $S(Q, \omega)$ on the high-energy side. The comparison of $S_p(Q, \omega)$ with the total in Fig. 3 also shows that both the apparent one-phonon frequency and group width is increased by $S_p(Q, \omega)$. Hence the frequency and lifetimes of high-frequency phonons will be difficult (or impossible) to measure accurately except at very low $Q$.

To display the interference contributions more explicitly, we show in Fig. 4 the $S(Q, \omega)$ for two longitudinal phonons having wave vectors $Q = (2\pi/a)(1, 1, 0)$ and $Q = (2\pi/a)(2, 2, 2, 0)$ which are equal distances away from the reciprocal lattice point $\pi = (2\pi/a)(2, 0, 2, 0)$. For these phonons the reduced vector $\vec{q} = Q - \pi$ is equal in magnitude but opposite in sign. The $S(Q, \omega)$ is shown in units of $Q^2d^2(q)$. Since $A(q\lambda, \omega)$ depends only on $|q|$ and $|q\lambda|$, $S(Q, \omega)$ is, from (4) and (6), identical for the two phonons in these units. The interference term $S_{13}(Q, \omega)$, however, changes sign with the sign of $\vec{q}$ so that the difference in intensity in Fig. 4 is due solely to this interference. Interference is clearly important even for phonons having small reduced wave vectors and long lifetimes. The peak position of the group and its width is not, however, affected so that strong interference here will not impede the measurement of phonon frequencies and lifetimes. In general, interference alters the peak position and full width at half maximum of the group only for phonons having a large intrinsic width $W^{14,15}$. A similar interference contribution is shown in Fig. 5.

In summary the strong frequency dependence of $S(Q, \omega)$ will make measurement of phonon frequencies and lifetimes for the high-frequency phonons difficult. Otherwise the observed phonon groups can be used to obtain one-phonon frequencies and inverse lifetimes.

IV. ONE-PHONON PROPERTIES

Having identified the contributions to $S(Q, \omega)$ in Sec. III, we turn here to the one-phonon properties. Figure 6 shows the phonon frequency dispersion curves of fcc $^4$He at 9.03 cm$^{-1}$/mole and 38 K calculated in the QH, the SCH, and the SCH + cubic approximations. The SCH approximation includes all the even anharmonic terms (quartic, sixth-order, ... ) as they would appear in a first-order perturbation correction to the QH approximation but calculated in a self-consistent manner. From Fig. 6 we see these anharmonic terms (mainly the quartic) substantially increase all the phonon frequencies above the QH values. At this stage the phonons have infinite lifetime with $A(q\lambda, \omega)$ given by (5). In the SCH + cubic approxi-
mation the cubic anharmonic term is added as a perturbation (of second order) to the SCH theory. The SCH + cubic frequencies are identified with the maximum of $A(q\lambda, \omega)$ in (5). The cubic anharmonic term reduces the frequencies to somewhat below half way between the SCH and QH values. Clearly, the SCH + cubic frequencies are not the final word and remaining higher anharmonic terms and different methods of including the cubic term, such as iterating it with SCH approximation, remain to shift these values somewhat. Detailed comparisons of different methods and extensions have been discussed elsewhere.$^{14,17,20}$

In Fig. 7 we compare the SCH + cubic frequencies with the observed values of Eckert et al.$^1$ for fcc $^4$He at 9.03 cm$^3$/mole and 38 K. The solid line is the Born von Kármán fit to the observed values constructed by Eckert et al. The SCH + cubic frequencies lie somewhat below but in reasonable agreement with the observed values. Goldman has included, in addition, those anharmonic terms of the form $V_2 V_4 V_3 + V_4 V_5 V_3 V_4 + \cdots$ and obtained frequencies which lie somewhat above the observed values and in somewhat better agreement with experiment.

Figure 8 compares the calculated and observed intrinsic one-phonon linewidths $W$ for fcc $^4$He at 9.03 cm$^3$/mole and 38 K. The calculated width is given by the full width at one-half the maximum height of $S_1(q, \omega)$ in (4). This calculated width here is due entirely to the cubic anharmonic term. The observed $W$ is obtained in the same way from the one-phonon part of $S(q, \omega)$ with the instrument resolution removed. Since we saw that $S_2(q, \omega)$ alters this width little and $S_3(q, \omega)$ affects the phonons of high energy ($\hbar \omega > 14$ meV) only, this comparison should be reliable for the phonons shown in Fig. 8. The comparison shows that the calculated linewidths are never greater than the observed values, are often in reasonable agreement and neverless than one half of the observed values.

In Fig. 9 we compare the computed one-phonon frequency and intrinsic linewidths $W$ with the ob-

FIG. 6. Phonon frequency dispersion curves of fcc $^4$He calculated in the QH and the SCH approximations, and from the peak of $S_1(q, \omega)$ when the cubic anharmonic term is added to the SCH theory (SCH + cubic). Here $a^* = 2\pi/a$.  

FIG. 7. Comparison of the calculated SCH + cubic (dashed lines) dispersion curves with the observed values of Eckert et al. The solid line is a Born-von Kármán fit to the observed values.
Self-consistent phonons in fcc $^4$He

The phonons in fcc $^4$He at 9.97 cm$^3$/mole and 28 K. Figure 9 shows that the inverse lifetime $\tau^{-1} = T = \frac{1}{2} W$ is always small compared with the phonon energy. The longitudinal phonon at $q^* = (0.5, 0.5, 0.5)$, which has the shortest lifetime, vibrates ~10 times before it decays. For comparison, the shortest-lived phonon in bcc $^4$He vibrates ~4–6 times before it decays. Again the SCH + cubic frequencies lie somewhat below but in reasonable agreement with the observed values.

The percentage change of the phonon frequencies with temperature at the two fixed volumes of $V = 9.03$ cm$^3$/mole and $V = 9.97$ cm$^3$/mole are shown in Figs. 10 and 11, respectively. Since the crystal volume remains constant during the temperature changes, there would be no frequency shift in a QH approximation. In the SCH approximation we find a positive shift of 1.0%–1.2% for all frequencies between 22 and 38 K at 9.03 cm$^3$/mole. The higher SCH frequency at $T = 38$ K results from the larger rms vibrational amplitude at 38 K than at 22 K. The negative shifts in the SCH + cubic frequencies shown in Fig. 10 thus results from the cubic shift, which is always negative and larger at $T = 38$ K than at $T = 22$ K. In Fig. 10 the maximum shift is observed to be ~15% at low $\xi$ for the $T[110]$ branch. While the SCH + cubic shifts show the correct wave vector and branch dependence, the maximum calculated shift is only

![Figure 8](image8.png)  
**FIG. 8.** Intrinsic phonon widths $W$ as calculated (lines) and as extracted from the observed phonon groups of Eckert et al. (Ref. 1) (points with error bars).

![Figure 9](image9.png)  
**FIG. 9.** Calculated phonon frequency and intrinsic one phonon widths $W$ of fcc $^4$He at $V = 9.97$ cm$^3$/mole and $T = 28$ K. The dots are the phonon energies observed by Thomlinson et al. (Ref. 2).

![Figure 10](image10.png)  
**FIG. 10.** Percentage shift in phonon energy $E = h\omega$ as the temperature is increased from $T = 22$ K to $T = 38$ K in fcc $^4$He at 9.03 cm$^3$/mole. The lines are the present SCH + cubic calculations and the points the observed values of Thomlinson et al. (Ref. 2).

![Figure 11](image11.png)  
**FIG. 11.** Same as Fig. 10 for fcc $^4$He at 9.97 cm$^3$/mole.
~ 8%. This discrepancy implies that outstanding anharmonic terms which produce negative frequency shifts may be important.

V. INTERFERENCE CONTRIBUTIONS

In Figs. 1–3 we noted the presence of the interference contributions to \( S(\mathbf{Q}, \omega) \) through the difference between \( S_p(\mathbf{Q}, \omega) \) and \( S_l(\mathbf{Q}, \omega) \). In addition, the difference in intensity between the pairs of phonon groups shown in Figs. 4 and 5 is due solely to interference. The interference contribution to \( S(\mathbf{Q}, \omega) \) may be made more quantitative by comparing the static form factors, \( S_p(\mathbf{Q}) \) and \( S_l(\mathbf{Q}) \), with and without interference. For example, the one-phonon form factor without interference

\[
S_l(\mathbf{Q}) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S_l(\mathbf{Q}, \omega)
\]

(8)

is for a harmonic crystal

\[
S_l(\mathbf{Q}) = d^2(\mathbf{Q})(\mathbf{Q} \cdot \mathbf{e}_\lambda)^2(\hbar/2M\omega_\lambda)[n(\omega_\lambda) + 1].
\]

(9)

In (9) we have included only the "energy-loss" scattering in which the neutron loses energy to the crystal [so that \( h\omega \geq 0 \) and the first delta function in (5) only is retained] which is the usual restriction employed experimentally. Thus,

\[
\frac{S_p(\mathbf{Q})}{S_l(\mathbf{Q})} \left[ \frac{(\mathbf{Q} \cdot \mathbf{e}_\lambda)^2(\hbar/2M\omega_\lambda)[n(\omega_\lambda) + 1]}{\omega_\lambda} \right] = d^2(\mathbf{Q}) - e^{-\mathbf{Q}^2}\]

(10)

is just the Debye-Waller factor. The observed intensity in the one-phonon peak region, with the multiphonon background subtracted, gives an \( S(\mathbf{Q}) \) which includes interference contributions. This is,

\[
S_p(\mathbf{Q}) = \int_{\text{one-phonon peak region}} \frac{d\omega}{2\pi} S_p(\mathbf{Q}, \omega)
\]

(11)

Thus the ratio of the observed \( S_p(\mathbf{Q}) \) to the \( S_l(\mathbf{Q}) \) expected for a harmonic crystal provides a convenient measure of the interference contribution. This ratio is usually plotted as

\[
\frac{S_p(\mathbf{Q})}{S_l(\mathbf{Q})} \left[ \frac{(\mathbf{Q} \cdot \mathbf{e}_\lambda)^2(\hbar/2M\omega_\lambda)[n(\omega_\lambda) + 1]}{\omega_\lambda} \right] = e^{-\mathbf{Q}^2}\]

(12)

to avoid an explicit determination of the Debye-Waller factor. The static form factor is a more reliable measure of the interference than is the first moment of \( S(\mathbf{Q}, \omega) \) since the \( S_p(\mathbf{Q}) \) in (11) is less sensitive to limits of the integration.

Figure 12 shows a plot of (12) as calculated using the SCH + cubic theory and as observed by Eckert et al. for \( \mathbf{Q} \) along the [111] and [110] directions. Since \( S_{\text{cal}}(\mathbf{Q}) \) is an odd function of the reduced wave vector \( \mathbf{Q} - \mathbf{G} \), we expect \( S_{\text{cal}}(\mathbf{Q}) \) to vanish when \( \mathbf{G} = 0 \) (when \( \mathbf{Q} \) equals a reciprocal lattice vector) and to show oscillatory behavior between the reciprocal lattice points. This symmetry also requires \( S_{\text{cal}}(\mathbf{Q}) \) to vanish when \( \mathbf{Q} \) is midway between two reciprocal-lattice points. This standard oscillatory behavior\(^\text{13, 14, 15}\) is shown in the [111] direction with \( S_p(\mathbf{Q})/S_l(\mathbf{Q}) = 1 \) for \( \mathbf{G} = (2\pi/a)(1, 1, 1) \), \( \mathbf{Q} = (2\pi/a)(2, 2, 2) \), and for \( \mathbf{Q} \) at midpoint (S). The magnitude of the calculated \( S_p(\mathbf{Q})/S_l(\mathbf{Q}) \) agrees well with the observed value.

Along the [110] direction we note an additional accidental zero (A) in the calculated \( S_{\text{cal}}(\mathbf{Q}) \) in addition to the symmetry-required zeros. This zero, where \( S_p(\mathbf{Q})/S_l(\mathbf{Q}) = 1 \), is also observed experimentally but occurs at a slightly smaller \( \mathbf{Q} \) value. Since the position of the accidental zeros has been found quite sensitive to the form of the cubic force constants, it is not surprising that the
calculated and observed positions do not coincide exactly. It is interesting that the calculation is able to reproduce this unanticipated accidental zero. Such accidental zeros were also observed\textsuperscript{15,21} in bcc \textsuperscript{4}He.

VI. DISCUSSION

A central aim of this paper is to examine how well the standard self-consistent theory of lattice dynamics can describe fcc \textsuperscript{4}He at high pressure. This was motivated particularly by the recent neutron scattering study\textsuperscript{11,12} of fcc \textsuperscript{4}He which provides a detailed account of the dynamics which any theory must properly describe. By standard SCP theory we mean the theory which describes the vibrational distribution of an atom about its lattice point by a Gaussian function, the function appropriate for a harmonic potential. The vibrational distribution then takes no direct account of the short-range correlations in the atomic motion induced by the steep repulsive core of the interatomic potential. It is well known, for example, that the bcc and hcp phases of solid helium require an explicit account of these short-range correlations, which is one reason why these phases are often referred to as quantum crystals.\textsuperscript{22}

The general good agreement of the present calculations with the observed dynamics suggests that compressed fcc \textsuperscript{4}He can be well described by the standard SCP theory. Given there remain outstanding anharmonic contributions, the calculated one-phonon frequencies agree well with the observed values. The calculated one-phonon linewidths \( W \) also agree quite well and are never less than one-half of the observed value. Given that the calculated width arises solely from the cubic term, this amount of error is to be expected. For example, a detailed comparison of the SCH + cubic linewidths with molecular dynamic determinations in solid potassium, which has a much softer core potential, shows a similar discrepancy.\textsuperscript{23} Also, the present results describe the relative contributions to the total scattering from one- and two-phonon processes and the interference effect well. To this extent, fcc \textsuperscript{4}He appears more like a classical crystal.

This does not say that including short-range correlations would not change the results, only that a reasonable description can be obtained without them. Horner\textsuperscript{4} and Kanney and Horton\textsuperscript{24} have found that incorporating short-range correlations in the heavier rare-gas crystals makes a significant difference to the dynamics. Here the intrinsic phonon linewidth may be some guide to the magnitude that can be expected in fcc \textsuperscript{4}He. When short-range correlations are incorporated the magnitude of the cubic anharmonic term is reduced, particularly the linewidth \( W \) is substantially reduced. For example, in bcc \textsuperscript{4}He using the \( T \) matrix method for treating short-range effects, \( W \) is typically \( -\frac{1}{2} \) of the observed value.\textsuperscript{25} When the Nosanow-Jastrow function method\textsuperscript{23} is used, which weights the short-range correlations more strongly with the Beck potential,\textsuperscript{25} the linewidth is \( -\frac{1}{10} \) of the observed value in bcc \textsuperscript{4}He. The implication here is that the short-range correlations are perhaps over weighted, even in the bcc phase, and should be only a subtle effect in fcc \textsuperscript{4}He if the correct phonon linewidth is to be obtained. This is also suggested by the temperature dependence of the phonon frequencies.

In this regard we note that we have attempted to use both Nosanow-Jastrow and the \( T \)-matrix methods to treat the short-range correlations in fcc \textsuperscript{4}He. In the Nosanow-Jastrow method, in which the parameter \( K \) in the short-range function, \( \exp[-Kv(r)] \), is determined by minimizing the energy, we found the energy minimum at \( K=0 \), no correlations at all. In the \( T \) matrix approach, we obtain reasonable values for the SCH frequencies if the equation for the short-range function was solved with the standard SCP theory parameters as input to this equation. However, when the solutions for the short-range function and the SCH equations were solved iteratively, the self-consistent vibrational amplitude of the atoms always increased without limit. This unphysical result was not changed by including the moment conditions introduced by Horner.\textsuperscript{4} This, we believe, is caused by the large cut or "wound"\textsuperscript{26} which the short-range correlation function makes in the Gaussian functions in fcc \textsuperscript{4}He. When the "wound" integral is large, the pair-correlation approximation to the energy used in the \( T \)-matrix method is a poor approximation to the total energy and this pair part can then decrease without limit. The "wound" integral increases with the density of the system and this problem is well known in \( T \)-matrix studies of dense nuclear matter.\textsuperscript{26} This suggests that short-range correlations require a subtle treatment in dense crystals and further work is clearly required to establish their importance.

Second, we examined the expansion of \( S(Q,\omega) \) and found that terms beyond the one-phonon \( S_1(Q,\omega) \) cannot be ignored for reduced wave vector \( \xi \geq 1.3 \) or \( Q \geq 3 \text{\AA}^{-1} \) at high energy transfers. Particularly important here is the sharp frequency dependence of the two-phonon \( S_2(Q,\omega) \). For example, \( S_2(Q,\omega) \) is small at low \( \hbar\omega \), rises rapidly in the region \( \hbar\omega \sim 14-18 \text{meV} \), and thereafter the plateau decreases slowly until it vanishes at \( \sim 32 \text{meV} \) (see Fig. 3). The rapid rise takes place just
in the range of the higher single-phonon energies and hence makes it difficult to separate \( S_1(Q, \omega) \) for these phonons from \( S_2(Q, \omega) \). This means that the one-phonon energies and lifetimes for high-energy longitudinal phonons can be measured accurately only using small wave-vector transfers.

The rapid rise in \( S_1(Q, \omega) \) takes place because the crystal is cold \( (kT < \Theta_F = 154 \text{ K}) \). Then in (7), \( n_1 \approx n_2 < 1 \) and, with \( \Theta_0 > 0 \), only the first term in \( J(1, 2, \omega) \) will be important. At small \( \omega \) this first term will be small since there are few pairs of phonons whose sum \( \omega_1 + \omega_2 \) is small. There will be a large number of such pairs when \( \omega \) reaches the sum \( \omega_1 + \omega_2 \) for two phonons both of which come from near the end of a dispersion curve where the density of phonon states is high. From Fig. 6 we see this occurs at \(-15 \text{ meV} \) as expected. The rapid rise in the two-phonon density of states can also be inferred from the one-phonon density of states \( g(\omega) \) calculated by Eckert et al. which rises rapidly to a broad peak in the range \( 6-8 \text{ meV} \). At high temperatures \( S_1(Q, \omega) \) is found to be a rather uniform function of \( \omega \).

To conclude, we note that Stassis et al. have just reported a most interesting neutron scattering study of \( ^4 \text{He} \) at the larger volume of 11.7 cm\(^3\)/mole. For low-frequency phonons having reduced wave vectors near the Brillouin-zone center, they observe sharply peaked one-phonon groups although the intensities show contributions from one-phonon and multiphonon interference. For phonons having intermediate wave vectors, the one-phonon groups broadened considerably but were still clearly separable from the multiphonon scattering. For the high-frequency phonons, however, Stassis et al. find that one-phonon and multiphonon scattering merge with the multiphonon scattering contributing mainly on the high-energy side. This is just as expected from the form of the two-phonon scattering shown here in Fig. 3.

Stassis et al. find that the one-phonon frequencies and lifetimes extracted from their measurements are generally very well described by Horner's one-phonon theory \(^4\) which includes short-range correlations. The one-phonon frequencies calculated by Goldman et al. without short-range correlations but including higher-order anharmonic contributions also agree well. Since Goldman et al. show only one branch and no lifetimes it is difficult to assess the role of short-range correlations, but Horner's results appear to agree best. Presumably the remaining higher-order anharmonic terms would also contribute to Horner's frequencies which might remove some of the good agreement.

It is also interesting that one-phonon frequencies extracted from the observed scattering and those calculated by Horner agree particularly well at low frequency, but the observed values lie above Horner's results for the high-frequency longitudinal phonons. As discussed above, at high frequency the two-phonon scattering is important and tends to shift the apparent one-phonon frequency to higher values. Hence, as noted by Stassis et al., the discrepancy between the observed and calculated one-phonon frequencies at high frequency is almost certainly due to the two-phonon component in the observed groups. Hence, the agreement between Horner's one-phonon theory and the one-phonon results is even better than suggested in Fig. 4 of Stassis et al.

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