Atomic kinetic energies in bcc ^{3}He

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The kinetic energy of atoms in solid ^{3}He is evaluated using self-consistent phonon methods. The results show that the high-frequency tails in the anharmonic response functions make most important contributions to the atomic kinetic energy. The high-frequency tails result from interatomic interactions via the steeply repulsive core of the interatomic potential.

I. INTRODUCTION

Helium atoms interact via a pair potential having a steeply repulsive core.[1] Since atoms in quantum crystals are not highly localized, two atoms can approach closely enough to be strongly repelled from each other by the repulsive core. This strong repulsion introduces high-energy components into the dynamics and high-energy tails into the dynamical response functions.[2] In a previous Letter,[3] we showed that these high-frequency tails, for example in the one-phonon response function $A(q,\lambda;\omega)$, contribute significantly to the kinetic energy, E_k, of atoms in solid ^{4}He.

For example, in a harmonic approximation such as the self-consistent harmonic (SCH) approximation,[2] the $A(q,\lambda;\omega)$ is a δ function at the SCH frequency $\omega_{q\lambda}$. Thus the SCH $A(q,\lambda;\omega)$ does not have high-frequency tails. Similarly, in an empirical Debye model the density of phonon states is cut off at $\omega=\omega_D$, where ω_D is the Debye frequency. Tails are introduced into $A(q,\lambda;\omega)$ when cubic and higher-order anharmonic terms are included in the dynamics.[2,4,5] For solid ^{4}He we showed[3] these tails double the E_k beyond that predicted by an empirical Debye model using the observed Debye temperature $\Theta_D=\hbar\omega_D/k$ and bring E_k close to the observed value. Thus the large E_k observed in solid ^{4}He may be regarded as a direct manifestation of the highly anharmonic nature of solid helium.

Sokol, Sköld, Price, and Kleb[6] and Carlson *et al.*[7] have come to similar conclusions in liquid ^{3}He. They show that the high-momentum (high-energy) components in the momentum distribution $n(p)$ contribute significantly to the kinetic energy of atoms in liquid ^{3}He. Indeed, the tail of $n(p)$ for momentum values $p\geq 2p_F$, where p_F is the Fermi momentum, contributes[6,7] approximately one-half of E_k. The existence of high-frequency tails in the dynamic structure factor $S(Q,\omega)$ of liquid helium and the relation of these tails to the repulsive core of the potential has been discussed by several authors.[8]

In this note we evaluate E_k in bcc ^{3}He at $T=0$ K and four volumes using the self-consistent phonon (SCP) theory. The calculations show that high-energy tails are again most important and that including them leads to E_k values close to those obtained using Monte Carlo (MC) methods.[9] The E_k values obtained using T-matrix short-range correlation functions[2,3] are significantly higher than those obtained using the Nosanow-Jastrow short-range correlation function.[2,10] Measurement of E_k could be used to distinguish between these two functions.

II. DYNAMICS OF SOLID HELIUM

To evaluate E_k we use the SCP theory of bcc ^{3}He as set out by Glyde and Hernadi.[5] This theory begins with the SCH approximation. Short-range correlations in the atomic motion are included in the SCH approximation here using both a T-matrix method[3] and the Nosanow-Jastrow method[10] to explore different treatments of these correlations. The SCH frequencies, $\omega_{q\lambda}$, are given by the usual harmonic expression with force constants obtained by averaging the second derivative of the potential over the vibrational distribution of the atoms. To evaluate this average we need the expectation values of the relative displacements of atoms, labeled i and j, of the form[2]

$$\langle \mathbf{u}_i\mathbf{u}_j\rangle=\frac{\hbar}{NM}\sum_{q,\lambda}e^{i\mathbf{q}\cdot\mathbf{R}_{ij}}\epsilon_{q\lambda}\epsilon_{q\lambda}\frac{1}{\omega_{q\lambda}}\int_0^\infty\frac{d\omega}{2\pi}A(q,\lambda;\omega)\,. \quad (1)$$

This expresses $\langle \mathbf{u}_i\mathbf{u}_j\rangle$ in terms of the one-phonon response function $A(q,\lambda;\omega)$ for $T=0$ K. Here $\mathbf{q}$ is the phonon wave vector, λ the branch index, and $\epsilon_{q\lambda}$ is the polarization vector. In the SCH limit, the one-phonon response function is

$$A(q,\lambda;\omega)=2\pi[\delta(\omega-\omega_{q\lambda})-\delta(\omega+\omega_{q\lambda})]\,, \quad (2)$$

and (1) reduces to the usual form,

$$\langle \mathbf{u}_i\mathbf{u}_j\rangle=\frac{\hbar}{NM}\sum_{q,\lambda}e^{i\mathbf{q}\cdot\mathbf{R}_{ij}}\epsilon_{q\lambda}\epsilon_{q\lambda}\frac{1}{\omega_{q\lambda}}\,. \quad (3)$$

The cubic anharmonic term is then added as a perturbation to the SCH theory.[5] With the cubic term included, the one-phonon response function is

$$A(q,\lambda;\omega)=\frac{8\omega_{q\lambda}^2\Gamma}{(-\omega^2+\omega_{q\lambda}^2+2\omega_{q\lambda}\Delta)^2+(2\omega_{q\lambda}\Gamma)^2}\,, \quad (4)$$

where $A(q,\lambda;\omega)$ and $\Gamma(q,\lambda;\omega)$ are the real and imaginary parts of the self-energy due to the cubic term. The cubic

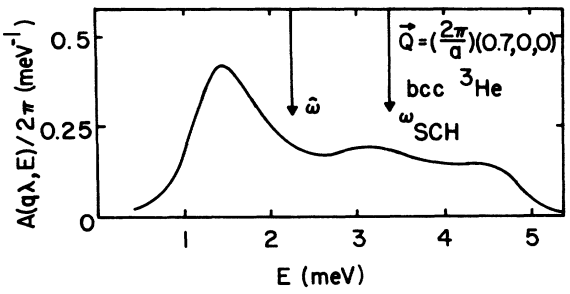

FIG. 1. The one-phonon response function $A(q,\lambda;\omega)$ for a longitudinal phonon having wave vector $Q=(2\pi/a)(0.7,0,0)$ in bcc ^{3}He at $V=24$ cm^{3}mol.

term can be incorporated in an iterative scheme, at least approximately, by substituting the anharmonic $A(q,\lambda;\omega)$ of (4) into (1). The resulting $\langle \mathbf{u}_i \mathbf{u}_j \rangle$ can be expressed in the usual SCH form (3) if we define a new frequency $\hat{\omega}_{q\lambda}$ as[11]

$$\frac{1}{\hat{\omega}_{q\lambda}} \equiv \frac{1}{\omega_{q\lambda}} \int_0^\infty \frac{d\omega}{2\pi} A(q,\lambda;\omega) . \tag{5}$$

The $\hat{\omega}_{q\lambda}$ are the mean frequencies of $A(q,\lambda;\omega)$, i.e.,

$$\hat{\omega}_{q\lambda} = \int d\omega\, \omega A(q,\lambda;\omega) \Big/ \int d\omega A(q,\lambda;\omega)$$
$$= \int d\omega\, \omega S_1(Q,\omega) \Big/ \int d\omega\, S_1(Q,\omega) . \tag{6}$$

The equivalence of (5) and (6) follows from the one-phonon dynamic structure factor

$$S_1(q,\omega) = \frac{1}{2\pi} [F(Q;q,\lambda)]^2 A(q,\lambda;\omega) \Delta(Q-q) ,$$

where $F(Q;q,\lambda)$ is the one-phonon form factor and the Ambegaokar, Conway, and Baym[12] sum rule,

$$\int d\omega\, \omega S_1(Q,\omega) = \omega_{q\lambda} [F(Q;q,\lambda)]^2 ,$$

which gives

$$\int d\omega\, \omega A(q,\lambda;\omega) = 2\pi \omega_{q\lambda} .$$

We may view the $\hat{\omega}_{q\lambda}$ as improved, infinite lifetime phonon frequencies given by the mean of the anharmonic response function $A(q,\lambda;\omega)$. The $\hat{\omega}_{q\lambda}$ are used[5] as new propagator frequencies to reevaluate $\langle u_i, u_j \rangle$, the $\Delta(q,\lambda;\omega)$, the $\Gamma(q,\lambda;\omega)$, and the response function $A(q,\lambda;\omega)$. This corresponds to the first iteration of an approximate iterative scheme including the cubic term. The resulting $A(q,\lambda;\omega)$ for a typical phonon is shown in Fig. 1 along with the original SCH frequency and the input $\hat{\omega}_{q\lambda}$. The $A(q,\lambda;\omega)$ clearly has high-frequency tails. These $A(q,\lambda;\omega)$ are used to evaluate E_k as discussed below.

III. THE KINETIC ENERGY

To obtain a general expression for E_k in terms of $A(q,\lambda;\omega)$ we begin with the time-dependent generalization of the displacement correlation function (1) for $i=j$,

$$\sum_\alpha \langle u_\alpha(t) u_\alpha(t') \rangle$$
$$= \frac{\hbar}{2NM} \sum_{q,\lambda} \frac{1}{\omega_{q\lambda}} \int_0^\infty \frac{d\omega}{2\pi} e^{-i\omega(t-t')} A(q,\lambda;\omega) . \tag{7}$$

By differentiating (7) with respect to t and t' we obtain the velocity correlation function, $\langle v_\alpha(t) v_\alpha(t') \rangle$. Taking the limit $t'=t$ we obtain the equal-time velocity expectation value of an atom, $\langle v_\alpha^2 \rangle$, and the atomic kinetic energy as

$$E_k = \frac{M}{2} \sum_\alpha \langle v_\alpha^2 \rangle = \frac{\hbar}{4N} \sum_{q,\lambda} \frac{1}{\omega_{q\lambda}} \int_0^\infty \frac{d\omega}{2\pi} \omega^2 A(q,\lambda;\omega) . \tag{8}$$

This is the basic relation we use.

In the SCH theory, the phonons have infinite lifetime and $A(q,\lambda;\omega)$ is a δ function given by (2). The corresponding SCH kinetic energy is then

$$E_k = \frac{\hbar}{4N} \sum_{q,\lambda} \omega_{q\lambda} , \tag{9}$$

where $\omega_{q\lambda}$ are the SCH frequencies. Similarly if we again approximate the full $A(q,\lambda;\omega)$ by a δ function, now at the mean frequency $\hat{\omega}_{q\lambda}$, given by (6), the corresponding kinetic energy is

$$E_k = \frac{\hbar}{4M} \sum_{q,\lambda} \hat{\omega}_{q\lambda} . \tag{10}$$

These two kinetic energies are shown in Fig. 2. The SCH frequencies lie significantly (approximately 20%) above the observed frequencies (for all branches except the T_2

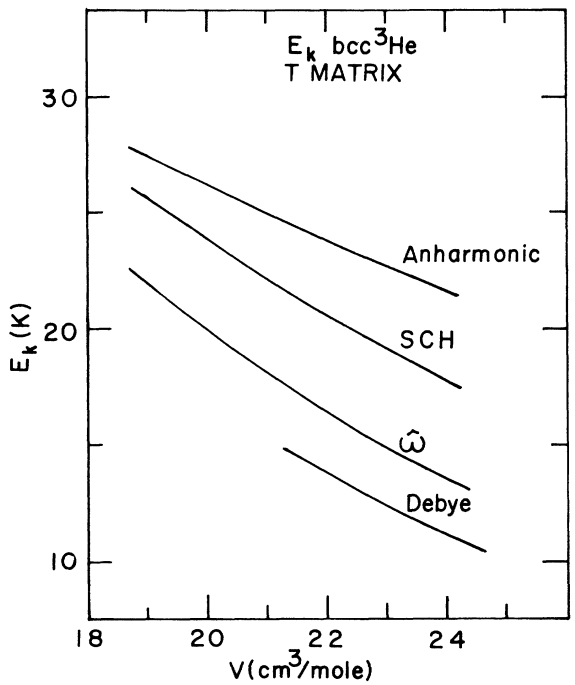

FIG. 2. Atomic kinetic energy calculated using (a) the full anharmonic response function in (8), (b) the self-consistent harmonic (SCH) frequencies in (9), (c) infinite lifetime phonons of frequency $\hat{\omega}$ in (10), and (d) the Debye model $E_k = \frac{9}{16}\Theta_D$ with Θ_D from Ref. 13.

branch in the bcc structure) in solid helium.[2] Thus if a δ function $A(q,\lambda;\omega)$ is reasonably valid we would expect the SCH E_k to lie above the observed value. In bcc ^{4}He the $\hat{\omega}_{q\lambda}$ lie close to the observed values.[4] Also shown in Fig. 2 is the kinetic energy in the Debye model

$$E_k = \tfrac{9}{16} R\Theta_D \tag{11}$$

obtained using observed values[13] of Θ_D. Since observed values of Θ_D are used, this should give a reasonable estimate of E_k if it is a reasonable approximation to cutoff the density of states [i.e., each $A(q,\lambda;\omega)$] at the Debye frequency $\omega_D = k\Theta_D/\hbar$. From Figs. 2 and 3 we see the Debye E_k is only one half the anharmonic value and the Monte Carlo (MC) value suggesting the high-frequency components of $A(q,\lambda;\omega)$ are most important.

In Figs. 2 and 3 we show the full anharmonic E_k evaluated using (8) and anharmonic response functions $A(q,\lambda;\omega)$ of the form given by (4). Since this E_k lies above those given by (9), (10), or (11), the high-frequency tails in $A(q,\lambda;\omega)$ depicted in Fig. 1 clearly make important contributions to E_k. The tails arise from including the cubic anharmonic term. From Fig. 3 we see that the E_k calculated using the T-matrix method lies close to the MC values. Thus the large E_k of atoms in bcc ^{3}He (above the Debye value, for example) may be viewed as a direct manifestation of the highly anharmonic nature of quantum solids.

The T-matrix E_k still lies below the MC value, suggesting that higher anharmonic terms[14] beyond the cubic term contribute further to the tails in $A(q,\lambda;\omega)$.

Similarly, solid hydrogen is a quantum solid but less anharmonic than solid helium. In solid H_2, observed values[15–17] of E_k lie approximately 50% above the corresponding Debye value given by (11). We expect that in solid H_2 the high-frequency tails of the anharmonic response function will also make significant contributions to E_k.

It is also interesting that the E_k calculated using the T-matrix short-range function lies above that obtained using the Nosanow-Jastrow function. In the T-matrix method, the short-range function is obtained by solving a differential equation in the interatomic potential. On the other hand, the Nosanow function is a postulated trial function.[10] The Nosanow function cuts off the pair wave function quite sharply when two atoms approach closely. This reduces the impact of the anharmonic hard core of the interatomic potential. As a result, the cubic anharmonic term is much smaller for the Nosanow-Jastrow function than for T-matrix function.[3,14] A measurement of E_k could distinguish between the two E_k values and the two short-range functions. Since the MC values[9] should be accurate the T-matrix E_k is favored. This does not say that the T-matrix method is better than the Nosanow-Jastrow method but rather that a short-range function obtained by solving a differential equation is more accurate than a postulated one. Such a function could be readily incorporated in the Nosanow-Jastrow method.

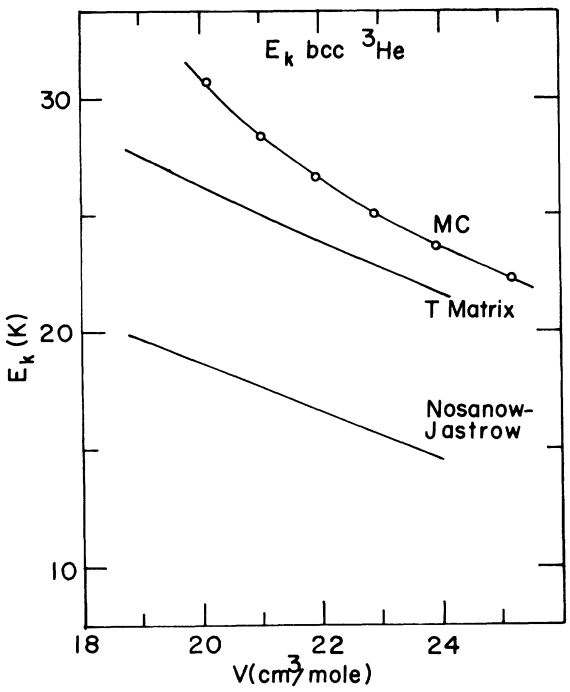

FIG. 3. Monte Carlo (MC) calculations of the atomic kinetic energy from Ref. 9 compared with present full anharmonic values from (8) using T-matrix and Nosanow-Jastrow short-range methods.

In summary, we find that the high-frequency tails of the response functions in quantum crystals make most important contributions ($\sim$50%) to the atomic kinetic energy. These high-energy (momentum) tails arise from interactions between atoms via the highly repulsive anharmonic core of the interatomic potential. In this sense the large kinetic energy (above the predicted Debye value) is a direct measure of the highly anharmonic nature of quantum crystals beyond that predictable by an effective harmonic model.

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