Kinetic Energies in Quantum Solids

L. K. Moleko
Department of Physics, University of Ottawa, Ottawa K1N 6N5, Canada

and

H. R. Glyde
Department of Physics, University of Delaware, Newark, Delaware 19716
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We show that the recently measured kinetic energy of atoms in solid helium is substantially larger than expected for even a moderately anharmonic solid. The large kinetic energy is rather an explicit measure of the highly anharmonic nature of solid helium. An explanation is proposed here in terms of the anharmonic one-phonon response functions which have large, high-frequency tails that contribute significantly to the kinetic energy.

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Recent measurements[1,2] of the atomic kinetic energy (KE) in solid helium by means of neutron scattering at high momentum transfer provide exciting new information on quantum solids. The KE has also been evaluated by Monte Carlo (MC) methods in the extensive studies of solid helium by Whitlock *et al.*[3] and more recently by Whitlock and Panoff.[4] These results are displayed in Fig. 1 where we see that the MC values lie slightly below the observed values. The aim of the present Letter is to show that the KE provides a quantitative measure of the highly anharmonic character of solid helium. Firstly, the observed KE is much larger than expected for even a moderately anharmonic solid. Secondly, we propose that the large KE can be explained in terms of the highly anharmonic one-phonon response functions found in quantum solids.[5-7]

At low temperature a moderately anharmonic solid can be described reasonably well by a Debye model.[8] In this model the KE per atom at $T=0$ K is $\frac{9}{16}\theta_D$, where θ_D is the Debye temperature. We take $T=0$ K since in solid helium $T \ll \theta_D$ and thermal energies should be less than 5%–10% of the zero-point energy at $T \approx 1$ K. In Fig. 1 we show the Debye KE of solid ^{4}He calculated with use of observed values of θ_{DW}, the Debye θ as obtained from the Debye-Waller factor. For example, we used $\theta_{DW}=25$ K at $V=21.1$ cm^3/mol,[9] and $\theta_{DW}=50$ K at $V=16.0$ cm^3/mol[10] for hcp ^{4}He. These empirical values of θ_{DW} are determined from observed values of the mean square vibrational amplitude, $\langle u\rangle^2 = 9\hbar^2/4Mk\theta_{DW}$, where M is the mass, via the Debye-Waller factor. The values of θ_D in bcc ^{4}He obtained from specific heat measurements[11] lie within 5% of θ_{DW}. From Fig. 1 we see that for hcp ^{4}He the Debye KE lies well below the observed values of Hilleke *et al.*[1] and significantly below the MC values. Thus an empirically adjusted Debye model cannot even approximately account for the observed or MC values of the KE. The failure of the Debye model is greatest at larger volumes where the atomic vibrational amplitudes and anharmonic effects are largest.

If we assume a Gaussian vibrational distribution of the atoms about their lattice points, the observed θ_{DW} can be used to set $\langle u^2\rangle$. The atomic KE predicted by a Gaussian distribution is $E_k = 9\hbar^2/8M\langle u^2\rangle = \theta_D/2$ which lies slightly below the Debye KE shown in Fig. 1. Thus an effective harmonic picture using Gaussian vibrational amplitudes also cannot be adjusted to explain both the observed θ_{DW} and the observed KE.

To develop an anharmonic model, we now relate the KE to averages over a full anharmonic, one-phonon

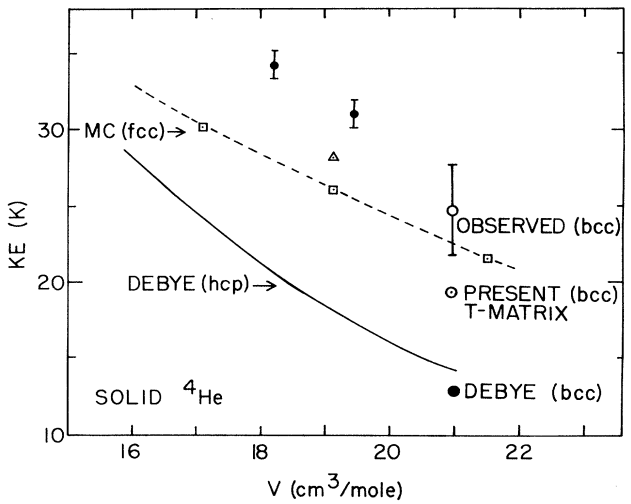

FIG. 1. Kinetic energy in solid ^{4}He. Solid circle with error bars, observed values in hcp ^{4}He (Refs. 1 and 2); open circle with error bar, bcc ^{4}He (P. Sokol, private communication); triangle, calculated Monte Carlo value in fcc ^{4}He (Ref. 4); squares, calculated MC values in fcc ^{4}He (Ref. 3) with dashed line as guide to eye; solid curve, Debye model KE $= \frac{9}{16}\theta_D$ in hcp ^{4}He; solid circle, Debye KE in bcc ^{4}He; open circle, present anharmonic T-matrix KE in bcc ^{4}He from Table I.

response function[8,12]

$$A(q\lambda,\omega)=\frac{8\omega_{q\lambda}^2\Gamma(q\lambda,\omega)}{[-\omega^2+\omega_{q\lambda}^2+2\omega_{q\lambda}\Delta(q\lambda,\omega)]^2+[2\omega_{q\lambda}\Gamma(q\lambda,\omega)]^2}. \quad (1)$$

Here $\omega_{q\lambda}$ is the basis one-phonon frequency for wave vector q and branch λ while Δ and Γ are the phonon frequency shift and inverse lifetime due to further anharmonic terms, respectively. The $A(q\lambda,\omega)$ is observed, for example, in the one-phonon dynamic form factor[7,8,12]

$$S_1(Q,\omega)=(2\pi)^{-1}|F(Q,q\lambda)|^2A(q\lambda,\omega)\Delta(\mathbf{Q}-\mathbf{q}), \quad (2)$$

where $F(Q,q\lambda)$ is the structure factor. If we make the usual expansion of the atomic displacements $u_\alpha(t)$ in terms of normal coordinates (here interacting), we have[12]

$$\sum_\alpha\langle u_\alpha(t)u_\alpha(t')\rangle=\frac{1}{N}\sum_{q\lambda}\frac{\hbar}{2M\omega_q}\int_0^\infty\frac{d\omega}{2\pi}e^{-i\omega(t-t')}A(q\lambda,\omega). \quad (3)$$

We may differentiate (3) with respect to t and t' to obtain the velocity correlation function. Taking the limit $t=t'$ after differentiation we obtain $\langle v_\alpha^2\rangle$ and

$$E_k=\frac{M}{2}\sum_\alpha\langle v_\alpha^2\rangle$$

$$=\frac{\hbar}{4N}\sum_{q\lambda}\frac{1}{\omega_{q\lambda}}\int_0^\infty\frac{d\omega}{2\pi}\omega^2A(q\lambda,\omega). \quad (4)$$

In (4) the KE is expressed as the second moment of $A(q\lambda,\omega)$ for each phonon $q\lambda$.

To display the basic character of (4) we examine two points. Firstly, if the phonons have infinite lifetimes, then

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

and (4) reduces to $E_k=(\hbar/4N)\Sigma_{q\lambda}\omega_{q\lambda}$ which is the familiar harmonic form. The $A(q\lambda,\omega)$ and KE also take this form in the self-consistent harmonic (SCH) approximation ($\omega_{q\lambda}=\omega_{q\lambda}^{\mathrm{SCH}}$). Since the $\omega_{q\lambda}^{\mathrm{SCH}}$ exceed observed values in bcc ^{4}He (see Fig. 2 and Ref. 12), the SCH KE also ought to be too large if the harmonic form is correct. Secondly, the $S_1(Q,\omega)$ satisfies the Ambegaokar-Conway-Baym sum rule[13]

$$\int_{-\infty}^{\infty}d\omega\,\omega S_1(Q,\omega)=\omega_{q\lambda}|F(Q,q\omega)|^2, \quad (5)$$

so that from (2)

$$\frac{1}{\omega_{q\lambda}}\int_0^\infty\frac{d\omega}{2\pi}\omega A(q\lambda,\omega)=1. \quad (6)$$

The first moment of $A(q\lambda,\omega)$ must be equal to $\omega_{q\lambda}$, the starting or basis frequency used to calculate $A(q\lambda,\omega)$. For example, the SCH frequencies may be used as basis frequencies ($\omega_{q\lambda}=\omega_{q\lambda}^{\mathrm{SCH}}$). If the cubic anharmonic term is added as a perturbation to calculate Δ and Γ, $A(q\lambda,\omega)$ in (1) will peak at a frequency below $\omega_{q\lambda}^{\mathrm{SCH}}$. This is illustrated in Fig. 3. The Ambegaokar-Conway-Baym sum rule tells us that the resulting $A(q\lambda,\omega)$ must have sufficient width and shape that, although it peaks below $\omega_{q\lambda}^{\mathrm{SCH}}$, the first moment remains equal to $\omega_{q\lambda}^{\mathrm{SCH}}$. Indeed in bcc He the $A(q\lambda,\omega)$ are found to have tails extending up to high frequency.[5-7] We expect high-frequency tails to contribute significantly to the second moment appearing in (4), making the anharmonic KE large and greater than the SCH value.

In our explicit calculations we begin with the SCH frequencies as a basis and include the cubic anharmonic term in $A(q\lambda,\omega)$. Using this $A(q\lambda,\omega)$ and following Horner,[6] we define frequencies

$$\hat{\omega}_{q\lambda}\equiv\int d\omega\,\omega A(q\lambda,\omega)/\int d\omega\,A(q\lambda,\omega).$$

These $\hat{\omega}_{q\lambda}$ are taken as the best infinite-lifetime basis frequencies to represent solid helium ($\omega_{q\lambda}=\hat{\omega}_{q\lambda}$) and are used to evaluate $A(q_\lambda,\omega)$ in final form (see Glyde and Hernadi[14] for details). In Fig. 3 we show a typical response function calculated using the $\hat{\omega}_{q\lambda}$. This has a

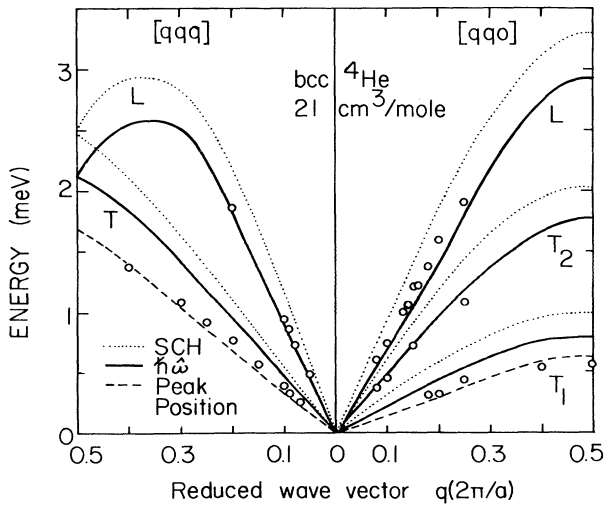

FIG. 2. The $\hat{\omega}$ (solid line) and SCH (dotted line) frequency dispersion curves in bcc ^{4}He ($V=21$ cm^3/mol). The points are observed values (Ref. 5). The dashed line shows the peak position of $A(q\lambda,\omega)$ for two transverse modes.

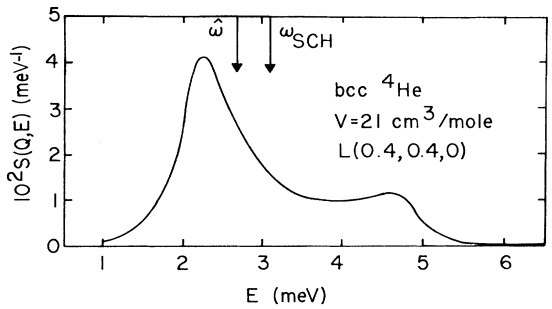

FIG. 3. $S_1(Q,\omega)$ calculated by use of the $\hat{\omega}_{q\lambda}$ frequencies and the cubic anharmonic term.

substantial width and a high-frequency tail. High-frequency tails in $S_1(Q,\omega)$ are due to the steep repulsive (anharmonic) core of the interatomic potential. High-frequency tails in $S(Q,\omega)$ are also expected[15] in liquid ^{4}He. These $A(q\lambda,\omega)$ were used in (4) to obtain the anharmonic KE and to verify the sum rule (6). We have used here the Beck potential[16] which represents the He-He potential well.[17] The repulsive core is the important part of the potential for the dynamics. While the potential is well known for the purpose here, there remains some debate[18] about the details of the core. In the example shown in Fig. 3 we used a T-matrix treatment[19] of the short-range correlations (SRC's) between atoms to calculate $\hat{\omega}_{q\lambda}$, Δ, and Γ in (1). To test the sensitivity of the results to the treatment of SRC's we have also used the original Nosanow-Jastrow[20] method.

The important contribution to the KE from the high-frequency tails of $A(q\lambda,\omega)$ is displayed in Table I. If an infinite lifetime model is adequate, then

$$E_k = (\hbar/4N)\sum_{q\lambda}\hat{\omega}_{q\lambda} \quad (7)$$

ought to agree with experiment, since the $\hat{\omega}_{q\lambda}$ are approximately equal to the observed values in Fig. 2. The full anharmonic KE obtained from (4) is substantially larger, especially in bcc ^{3}He. Also, the KE calculated by use of the T-matrix treatment[19] of the SRC's is larger than that obtained for the Nosanow[20]-Jastrow method. This is because the T-matrix SRC function does not cut off the anharmonic hard core of the interatomic potential as severely as does the Nosanow-Jastrow function. More of the anharmonic hard core remains in the T-matrix method, leading to higher $\hat{\omega}$ and a larger cubic anharmonic term in $A(q\lambda,\omega)$. The present T matrix should provide a better treatment of the hard core and a more reliable KE since in this method the SRC function is obtained by solution of a differential equation in the real potential. The present calculations are approximate, however, and depend upon the input frequencies to $A(q\lambda,\omega)$. Use of the peak position of the response (see Fig. 3) would give a somewhat higher KE ($\sim 20\%$) while use of SCH frequencies would give a lower KE ($\sim 5\%$). The chief aim is to illustrate the high-frequency tail contributions to the KE explicitly.

TABLE I. Kinetic energies (in kelvins) in bcc ^{4}He ($V=21$ cm^3/mol) and ^{3}He ($V=24$ cm^3/mol) calculated by use of the Nosanow-Jastrow (NJ) and T-matrix methods: $\hat{\omega}$, Eq. (7); SCH, Eq. (7) with SCH frequencies; anharmonic, Eq. (4).

	^{4}He		^{3}He	
	T matrix	NJ	T matrix	NJ
$\hat{\omega}$	16.0	13.9	14.5	13.5
SCH	18.6	15.1	18.0	14.3
Anharmonic	19.5	15.6	27.9	14.5
Debye[a]	12.7		11.0	

[a]With $\theta_{DW}=22.5$ K (^{4}He, Ref. 5) and $\theta_D=19.5$ K (^{3}He, Ref. 21).

The ratio of the T-matrix anharmonic KE to the Debye value in bcc ^{4}He is $R=1.5$. The same ratio applied to hcp ^{4}He at $V=19.45$ cm^3/mol predicts an anharmonic KE of 27 K. This lies slightly above the MC values shown in Fig. 1 but still below the observed[1] value of 31.1 ± 0.9 K. Inclusion of further anharmonic terms could increase R somewhat.

In bcc ^{3}He the ratio of anharmonic KE calculated by use of the T matrix to Debye KE is significantly larger, $R=2.5$. That is, we predict a substantially larger KE in ^{3}He than would be suggested by a comparison of the Debye temperatures in ^{3}He and ^{4}He. In bcc ^{3}He we have used the maximum value of θ_D observed by Greywall[21] which agrees well with the maximum θ_D observed earlier by Castles and Adams.[22]

The purpose here is to show that the observed KE in ^{4}He is nearly twice that expected for a moderately anharmonic solid. We propose that the large KE results from the high-frequency tails of the anharmonic one-phonon response function which contributes greatly to the KE but little to θ_{DW}. Measurements of the KE in bcc ^{3}He could test this hypothesis since we predict the tail contributions to be larger in ^{3}He than in ^{4}He.

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