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Single-particle kinetic energies in solid neon

D. A. Peek,* I. Fujita, M. C. Schmidt,† and R. O. Simmons
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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Inelastic-eV-neutron-scattering experiments have been performed on solid natural neon at five temperatures between 4.7 and 26.4 K and at pressures between equilibrium vapor pressure and 17.6 MPa. Measurements of atomic-recoil line broadening were made within the impulse approximation. Average single-particle kinetic energies $\langle E_k \rangle$ have been obtained by assuming a Gaussian atomic momentum distribution. A ground-state kinetic-energy value of 49.1 ± 2.8 K is found and the temperature variation of $\langle E_k \rangle$ is determined. Comparisons are made to somewhat restricted deductions of $\langle E_k \rangle$ from previous experiments on neutron-phonon scattering and heat capacity. Comparisons with existing theoretical calculations show that none of them predicts the observed ground-state kinetic energy, although cell-model variational calculations and self-consistent harmonic calculations in the literature find that the ground-state kinetic energy is substantially larger than the ground-state potential energy, in agreement with our results.

I. INTRODUCTION

The ground-state energies (GSE's) of noble-gas solids have been of interest for a long time, because they constitute a significant fraction of the total cohesive energy of these crystals.[1] Generalizing results via laws of corresponding states,[2–4] through use of a short-range interatomic potential of common form, is another attraction of studies of this family.

In this family, neon is the lightest member to solidify under its own vapor pressure. Values of its lattice properties form a bridge between those of the helium isotope "quantum solids," to which conventional Born–von Kármán lattice theory simply does not apply and for which other methods have therefore been devised, and those heavier members argon, krypton, and xenon, to which anharmonic perturbation theories have been applied with some success at low temperatures.[4] Further, for neon at usual solid densities, the influence of multibody effects in the interatomic interactions is relatively small, which simplifies model calculations. Neon, therefore, has been the subject of calculations beyond cell models and anharmonic perturbation treatments, such as self-consistent phonon theories[5–8] and Monte Carlo simulations.[9,10]

A number of theoretical papers have taken up questions relating to neutron scattering by such anharmonic crystals as neon.[11–13] Some of the discussions are *ad hoc,* applied to a particular feature of the phonon spectra, e.g., the one-phonon response, without demanding that all features be reproduced in the model. The calculations have generally been restricted to small values of wave-vector transfer Q. As for x-ray scattering, in which up to now no direct phonon inelastic measurements on noble-gas solids have been possible, theoretical studies of the Debye-Waller factor or of the mean square atomic displacement $\langle u^2 \rangle$ have been made by a number of authors.[14–18] In any case, it appears that to some extent, quasiharmonic fits and spectral moments analyses can be used as consistency checks between diverse measurements on neon.[19]

Several experimental studies of neon isotope solids have yielded results showing the influence of the ground-state kinetic energy (GSKE). These include measurements of the lattice parameters[20,21] and their temperature dependences,[21] of the equilibrium vapor pressures,[22] and of the specific heats.[23] However, such results show this influence only indirectly. For example, interpretation of the lattice parameter data in terms of the GSE requires information about the compressibility, and interpretation of the vapor pressure and of the heat capacity data requires information about the heat of vaporization.

It might be thought that the GSE of solid neon could be unambiguously determined through the measurement of one-phonon dispersion curves by coherent inelastic neutron scattering[24–27] and of x-ray Debye-Waller factors.[19] Although more direct than thermodynamic property measurements, such studies are not without practical disadvantages in interpretation. For example, the neutron-measured phonon wave-vector and energy values (Q and E, respectively) are commonly parametrized by Born–von Kármán models in order to produce smoothed curves, consistent between the branches and directions. The models also are used to obtain values, by interpolation and extrapolation, for the dispersion of phonons in directions not directly measured. The choice of number of force constants and their symmetry is somewhat arbitrary, and details of phonon densities of states calculated from them may vary with the choice. It follows that the inferred GSE may also be dependent upon the choice. More important, there is also the question of how well a Born–von Kármán fit can be depended upon to represent the situation in an anharmonic crystal, in which various phonon frequencies are substantially renormalized.[25] These matters are discussed in Sec. III A below.

High-energy inelastic neutron scattering directly measures the single-particle kinetic energy, which at low temperature is the GSKE, in contrast to all the experimental methods mentioned above. In it, the broadened widths of scattering recoil peaks give this direct result, analogous to the historic use of x rays to determine electron momentum distributions.[28] We report here such measurements on a series of solid neon samples. A brief account of part of this work has already been published.[29]

II. NEUTRON SCATTERING

A. Principles

As noted in Sec. I, the lattice dynamics of neon has been extensively studied by coherent inelastic neutron scattering at wave-vector transfers, Q, corresponding essentially to the central unit cells of the reciprocal lattice, that is, for $Q < 50$ nm^{-1}. Here we are interested in the large-Q regime.

In the limit that Q is very large, the neutrons sample the atomic single-particle momentum distribution $n(\mathbf{p})$. This scattering is represented in the impulse approximation (IA), with a dynamic structure factor $S(\mathbf{Q},E)$ given by[30]

$$S(\mathbf{Q},E)=\int n(\mathbf{p})\delta(E-E_r-\hbar^2\mathbf{Q}\cdot\mathbf{p}/M)d\mathbf{p} \ , \tag{1}$$

where E is the energy transfer, M is the atomic mass, and $E_r=\hbar^2Q^2/2M$ is the recoil energy of the atom. The present measurements are at values of Q above 200 nm^{-1}. We have shown elsewhere[31] that the IA is sufficiently well satisfied in this range for neon, for data of the present statistical uncertainty.

For a momentum distribution of the spherical Gaussian form

$$n(p)=A\exp(-p^2/2\sigma_p^2) \ , \tag{2}$$

where p is the atomic wave vector, it can be shown that $S(Q,E)$ is a Gaussian of energy width σ_Q centered about the recoil energy, with

$$\sigma_Q=\hbar^2Q\sigma_p/M \tag{3}$$

and that the average single-particle kinetic energy is

$$\langle E_k\rangle=\tfrac{3}{4}\sigma_Q^2/E_r \ . \tag{4}$$

We assume that a Gaussian form for the neon $n(p)$ is sufficient.[9,32]

B. Experimental conditions

We used the High Resolution Medium Energy Chopper Spectrometer (HRMECS) of the Intense Pulsed Neutron Source (IPNS)[33] at Argonne National Laboratory with 30-Hz pulsed neutrons of incident energy near 498 meV and groups of ^{3}He time-of-flight detectors at mean scattering angles near 87°, 106°, 121°, and 136°, respectively. This yielded Q values for neon between 200 and 280 nm^{-1}.

Five samples were run at temperatures between 4.7 and 26.4 K. The four lowest temperature samples were at saturated vapor pressure, while the 26.4-K sample was at 17.6±0.1 MPa, in order to solidify the sample somewhat above the triple-point temperature. All temperatures and molar volumes are shown in Table I. In our experiments, aluminum sample cells were used which consisted of several thin-walled tubes arranged in a vertical plane facing the detectors, at an angle of 45° with respect to the incident neutron beam. Four of our measurements used a sample cell consisting of seven tubes of 0.953 cm inside diameter and 0.083 cm wall thickness. Our 11.4-K measurement used a cell of four tubes of 1.55 cm inside diameter and 0.02 cm wall thickness. All tubes exposed 10.1 cm height to the beam. Temperatures were measured by the use of platinum and germanium resistors in the dc four-terminal mode.

For the experiments at 11.4 K and higher, the sample cells were cooled by being attached to a Displex closed-cycle refrigerator. For the 4.7- and 9.4-K samples, the cell was cooled by being attached to a liquid- ^{4}He cryostat. The sample cell was carefully evacuated, cooled *in situ*, loaded with 99.999% pure natural neon,[34] then the solids were prepared by further cooling to the desired temperature.

Although no predictions of crystalline anisotropy in $n(p)$ have been published for neon, it is possible that such an effect might be present, having the point symmetry of the fcc crystal. In Eq. (1), one sees that the measured scattering is a component taken in the direction of the wave-vector transfer $\mathbf{Q}$, so that the scattering from a single crystal sample therefore might also be anisotropic.[35] However, because our sample cells consisted of separate cylinders of high thermal conductivity, it is likely that the samples were polycrystalline and therefore our $\langle E_k\rangle$ values are averages over crystallographic direction. Careful diffraction measurements over various parts of rather similar cells containing hcp ^{4}He have shown that a variety of crystal orientations are present.[36]

Data collection required about 1–2 days for each sample, and an additional 1–2 days for each background run. Temperatures given in Table I are time-average values. For the 11.4-K sample, the background run was simply an empty cell data collection run. For the other samples, background was determined by emptying the cell of neon and refilling it with a small amount of ^{3}He gas, in an amount having a total absorption cross section equal to the scattering cross section of the neon sample. (Since the ratio of scattering to absorption cross sections is very

TABLE I. Experimental values of the average single-particle kinetic energy, $\langle E_k\rangle$, for solid neon at several temperatures and molar volumes, as directly measured by high-energy neutron scattering. Note that there is little variation in the kinetic energy below 11.4 K, suggesting that ground-state values are being measured at these temperatures.

Temperature (K)	Molar volume (cm^3)	$\langle E_k\rangle/k_B$ (K)
4.7±0.1	13.39	49.2±2.8
9.4±0.1	13.42	49.1±4.0
11.4±0.2	13.45	49.0±2.4
17.8±0.2	13.65	51.2±2.8
26.4±0.2	13.92	57.9±2.0

small at these energies, and the scattering peaks of neon and ^{3}He are widely separated kinematically, helium scattering should be insignificant.)

Sample-dependent multiple scattering was minimized through use of a rather weakly scattering sample and use of neutron absorptive shielding placed in the vicinity of the sample.

C. Data analysis and results

In Eq. (1) the response is seen to be centered about the kinematic value of the recoil energy, which depends upon the atomic mass. As we have considered elsewhere for data having similar statistical precision,[31] for natural neon the appropriate mass to be used in Eq. (3) is the natural-abundance- and cross-section-averaged mass, 20.13 amu.

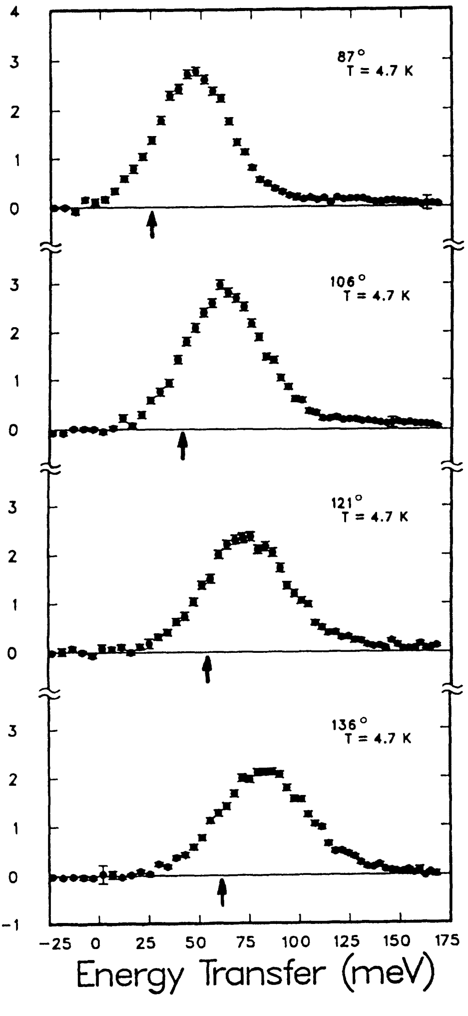

FIG. 1. Neutron double-differential scattering for solid neon at 4.7 K and scattering angles of 87°, 106°, 121°, and 136°, respectively. Arrows mark the kinematic center of the aluminum sample cell background scattering, which has been subtracted. Neon peak center is at the recoil energy, and weak multiple scattering appears in the apparent high-energy transfers around 125, 140, 150, and 160 meV, respectively. Note the increase in recoil energy and peak width with higher scattering angle, corresponding to higher momentum transfer.

Typical resolution-broadened data are shown in Figs. 1–3. In all cases, the residual background, presumably from multiple scattering, was small, broad, and centered in the apparent high-energy transfer regime. Various physical and instrumental effects vary in different ways over the observed range of 200 to 280 nm^{-1} in Q. The neon intrinsic peak width broadens directly with Q, its location moves to higher energy loss as Q increases, and the apparent multiple scattering has different magnitude and location for each of the different detector groups, as can be seen in Fig. 1. The relative location of the aluminum cell background scattering also changes with Q. Finally, HRMECS resolution is different for different detector groups. With all these different varying effects, there is the possibility to check for the presence of systematic error.

Elsewhere, we have considered HRMECS resolution for neon by two different methods, analytical[37] and numerical.[33,38] We found no detectable systematic differences between the two methods.[31] Here we report values using the numerical method.

After calculating the resolution function for each of the four angular groups of detectors, we fitted the data to a Gaussian function with polynomial background terms up to quadratic. In order to account for statistical variations and possible limitations of our fitting model, we fitted each data set over eight different fitting ranges, each range including that part of the Gaussian peak which was above the instrumental noise. We found that the different fitting ranges chosen for each angular group of detectors had little effect on the average value deduced for the kinetic energy. For different detector groups, the differences between average values was somewhat larger, although we could find no systematic variations. As our kinetic energy, $\langle E_k \rangle$, we quote the average of the resulting 32 values of $\langle E_k \rangle$ for each data set. As our estimates of uncertainty, we give the standard deviations from those values. Typical fitted data for one detector group are shown in Fig. 3.

As shown in Table I and Fig. 4, $\langle E_k \rangle / k_B$ of solid neon

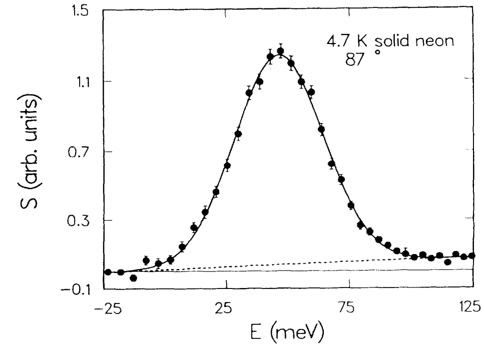

FIG. 2. Experimental data for 4.7 K and scattering angle of 87°, converted to S, fitted (solid line) with a Gaussian plus a small quadratic polynomial contribution used to account for multiple scattering (dashed line).

does not vary detectably below about 15 K. A confirmation of the consistency of our technique, particularly with respect to the handling of multiple scattering and background subtraction, is the fact that the 11.4-K sample was in a substantially thicker cell of somewhat different geometry, and that its measured $\langle E_k \rangle / k_B$ was nevertheless not different from the 4.7- and 9.4-K samples. Considering that the three lowest temperature values of $\langle E_k \rangle$ are so similar, we believe that our estimated uncertainties may be conservatively large. We quote the GSKE of solid natural neon to be 49.1 ± 2.8 K.

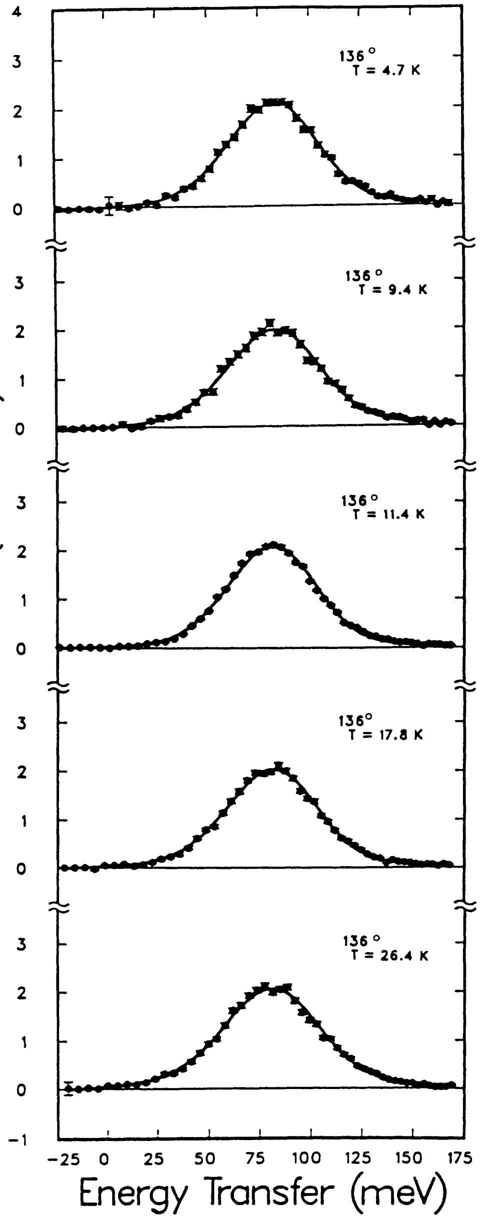

FIG. 3. Resolution-fitted scattering cross section for solid neon at scattering angle of 136° at temperatures of 4.7, 9.4, 11.4, 17.8, and 26.4 K, respectively. A model Gaussian $S(Q,E)$ with quadratic terms to account for multiple scattering has been convoluted with the instrumental resolution function and fitted to the data to determine the kinetic energy values stated in the text. Compared to lower scattering angles (see Fig. 1), the subtracted cell scattering for this peak has moved, and the multiple scattering has decreased in relative magnitude and also shifted in relative location. The intrinsic peak width is now 40% broader, whereas the instrument resolution has narrowed.

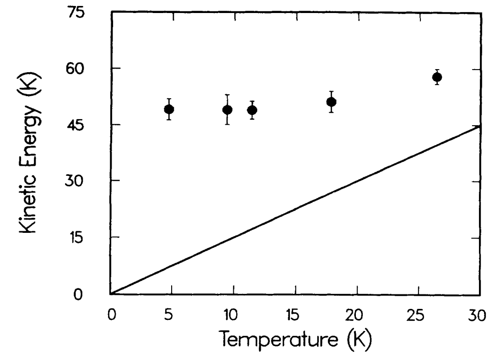

FIG. 4. Single-particle kinetic energy, $\langle E_k \rangle / k_B$, of solid neon as a function of temperature. Deviation from classical behavior is shown by comparison with the line shown of slope $\frac{3}{2}$. Volume changes over the entire temperature and pressure range measured amount to less than 5%. Correction to constant volume conditions, if desired, can be made by use of Eq. (6).

Figure 4 also shows that $\langle E_k \rangle / k_B$ rises as the temperature rises toward melting. It is important to note that the *relative* uncertainty of values of $\langle E_k \rangle / k_B$ at different temperatures is smaller than the absolute uncertainty of each point. In a given detector bank, the resolution function is constant, the multiple scattering varies only slightly as the density of the sample changes with thermal expansion, etc. Thus, while there is some limitation in measuring absolute kinetic energies, it is easier to tell differences between sets of data taken at different temperatures. Our three lowest temperature values of $\langle E_k \rangle / k_B$ differed by only 0.2 K, and we therefore estimate that the uncertainty in the *temperature dependence* of the kinetic energy for our five measurements is of that order. This conclusion is reinforced by another observation: similar sensitivity was found in $n(p)$ studies of liquid and solid argon, krypton, and xenon under classical conditions where equipartition applies to the values of $\langle E_k \rangle / k_B$.[39]

III. DISCUSSION

A. Comparison with deductions from other experiments

As noted in Sec. I, previous coherent inelastic neutron-scattering studies, for wave vectors within the central Brillouin zone, require the intervention of a model for the phonon dispersion in order to obtain the total phonon density of states and hence the GSE. In addition, specific heat measurements upon solid neon when viewed within a quasiharmonic framework can also yield information on the GSE. It is instructive to examine the results of such analyses and compare them with results of the present direct measurements of single-particle kinetic energies.

From a Gilat and Raubenheimer[40] frequency distribution program using a set of force constants determined by inelastic neutron scattering,[24–26] a phonon density of states, $g(\omega)$, can be generated. The temperature independent GSE is then identified as the first term inside the integral in the expression for the vibrational energy per particle, E_{vib},

$$E_{\text{vib}} = \int \hbar\omega \left[\frac{1}{2} + \frac{1}{e^{\hbar\omega/k_B T} - 1} \right] g(\omega) d\omega \ . \tag{5}$$

Eckert *et al.*,[27] used their experimentally determined quasiharmonic mode Grüneisen parameters to reanalyze earlier thermal neutron measurements of the phonon dispersion relations in low-density neon crystals. From a Born–von Kármán model employing general forces to third nearest neighbors plus axially symmetric forces to represent the interaction between an atom and its fourth to eighth nearest neighbors, Eckert *et al.* calculated a set of force constants for the $T=0$ K equilibrium density solid. E_{vib} was calculated as the resulting quasiharmonic GSE from this reanalysis; it is 78.0 K. In the harmonic approximation, $\langle E_k \rangle$ is taken to be one-half of E_{vib}, thus a GSKE of 39.0 K is obtained.

In order to test for the possibility that the inferred quasiharmonic GSE ($E_{\text{g.s.}}$) depends heavily upon the particular force constant model chosen to represent the actual experimental phonon dispersion measurements we utilized sets of force constants given by Leake *et al.* and Skalyo *et al.* for low-density neon crystals (molar volumes of 13.37 and 13.30 cm^3, respectively) in a Gilat and Raubenheimer routine in order to determine their associated $g(\omega)$'s. With these $g(\omega)$'s, GSE's were computed via Eq. (5). They differed from the result given by Eckert *et al.* by less than 1%, after correction of experimental volumes (V) to the $T=0$ equilibrium value, $V_0 = 13.39$ cm^3, with the expression

$$\frac{\Delta E_{\text{g.s.}}}{E_{\text{g.s.}}} \approx \left[\frac{V_0}{V} \right]^{\gamma} - 1 \ . \tag{6}$$

Here γ is the thermodynamic Grüneisen parameter chosen in the range of generally accepted experimental values (2.5–2.8). Thus, like scalar thermodynamic quantities involving integrals over $g(\omega)$, the GSE is not very sensitive to the finer details of the phonon spectrum.

Alternatively, instead of using an experimentally determined $g(\omega)$, the Debye spectrum, $g(\omega) = 3\omega^2/\omega_D^3$ can be substituted into Eq. (5) such that

$$E_{\text{vib}} = \frac{9}{8} k_B \Theta_C + 9 k_B \frac{T^4}{\Theta_C^3} \int_0^{\Theta_C/T} \frac{x^3}{e^x - 1} dx \ , \tag{7}$$

where $\Theta_C = \hbar\omega_D / k_B$ is the Debye characteristic temperature for the specific heat and ω_D the cutoff frequency. In this case, E_{vib} depends solely upon the parameter Θ_C which is explicitly temperature dependent for a quasiharmonic model and is commonly used to represent specific heat measurements.[41,42] In order to use Eq. (7) with $\Theta_C(T)$, the GSE term's Θ_C must be replaced[43] by its high-temperature limit Θ_C^∞. This is based on the assumption that the phonon spectrum is harmonic, even if it does not have the Debye form of the phonon density of states. If the crystal has anharmonicities at high temperature, then the value of the Debye temperature at large T is no longer a good measure of the GSKE. It has therefore been suggested that in the intermediate temperature range, where anharmonicities are small and can be explained by quasiharmonic models, an expansion for Θ_C as a function of temperature be developed that shows the high-temperature limiting behavior of Θ_C before severe anharmonic behavior sets in.[44,45]

In order to determine Θ_C^∞ from specific heat measurements, it is customary to model the temperature behavior of Θ_C with a quasiharmonic high-temperature expansion given by Domb and Salter:[43]

$$\Theta_C^2 = \Theta_C^\infty 1 - A(\Theta_C^\infty/T)^2 + B(\Theta_C^\infty/T)^4 - \cdots) \ . \tag{8}$$

The coefficients A and B depend upon the moments of $g(\omega)$ defined by

$$\mu_n = \int \omega^n g(\omega) d\omega \ . \tag{9}$$

Thus by plotting Θ_C^2 versus T^{-2} and then extrapolating from the appropriate intermediate temperature range to $T^{-2}=0$ a value for Θ_C^∞ can be determined. The commonly accepted range for this extrapolation is from $\Theta_C/6$ to $\Theta_C/3$. The lower bound is set by the fact that Eq. (8) is divergent for lower temperatures. The upper bound is chosen as the range in which anharmonicity in heavier noble-gas solids becomes too severe to be handled by a quasiharmonic expression. Plotting the extensive results of Fugate and Swenson[41] (Fig. 5) in this manner shows the inadequacy of such a quasiharmonic expression in order to determine the high-temperature limit of Θ_C^∞ for neon.

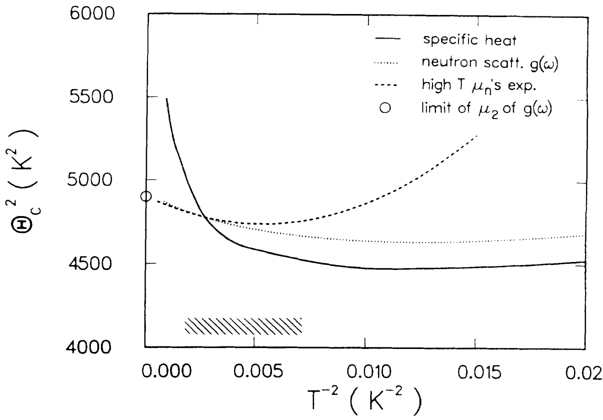

FIG. 5. Plot for the estimation of Θ_C for equilibrium vapor pressure neon. Solid curve represents specific heat data by Fugate and Swenson (Ref. 41). Dotted curve calculated from $g(\omega)$ given by neutron scattering by Eckert *et al.* (Ref. 27). Circle represents Θ_C via second-moment calculation of the Eckert *et al.* $g(\omega)$. Dashed line corresponds to a high-temperature moments expansion (up to the T^{-4} term) utilizing moments calculated from the $g(\omega)$ given by Eckert *et al.* It is shown on the graph to illustrate how one would expect the specific heat data to behave if neon were truly quasiharmonic in the high-temperature limit. The shaded region indicates the range from which extrapolation is conventional.

From a comparison of the plot of the specific heat data with the corresponding graph due to the quasiharmonic $g(\omega)$ determined by experimental phonon dispersion curves (also shown in Fig. 5), a low-temperature offset is visible. If one takes this offset along with the temperature dependence of the quasiharmonic curve, and thus ignores explicitly anharmonic effects, a value of 69 K can be given for the specific heat Θ_C^∞. The GSE in this case is 78 K with a corresponding harmonic GSKE of 39 K.

We note that anharmonicity for solid neon is severe enough that the quasiharmonic parameter Θ_C^∞ is not a useful quantity in its usual sense of being related to $\langle u^2 \rangle$ and thus a good approximation to the GSE according to Domb and Salter. In fact this is an indication that for $T > \Theta_C/6$ the shape of the density of states is changing from the one predicted by the quasiharmonic model. To model the frequencies of vibrations they must not only scale directly with volume for higher temperatures but also become explicitly temperature dependent. The inability to arrive at a consistent solution to the Debye temperature through these different analyses of phonon spectra and heat capacity work is a demonstration of the fact that anharmonicity is severe enough in neon to prevent any of these techniques from providing anything other than a qualitative statement of the GSE of solid neon.

In summary, it might be thought that a measurement of the phonon spectra would offer a simple calculation of the density of states, and thus a measurement of the GSE. However, in order to determine force constants from the data, quasiharmonic assumptions must be made. It might also be thought that a measurement of the heat capacity at large temperatures would allow a calculation of a correct Debye temperature to be used in calculating the GSE. However, severe anharmonicities in solid neon at high temperature prevent a good limiting value of Θ_C from being determined. In contrast, our data provide direct measurement of the GSKE, independent of any assumptions about the anharmonicity of solid neon.

B. Comparison with calculations

In spite of the large number of published theoretical papers about the lattice dynamics of solid neon, few give numerical results for $\langle E_k \rangle$, probably because to date this quantity has not been directly accessible through experiment. An early variational calculation by Bernardes[46] assumed uncorrelated single-particle wave functions, and arrived at a value of 48.0 K. In this model he found that the GSKE was 58.6% of the total ground-state vibrational energy, independent of the particular Lennard-Jones potential parameters chosen. In Table II we show several theoretical and experimental values of GSKE and GSE,

TABLE II. Comparison of theoretical calculations of the GSKE (GSE) and $-E_0$ with experimental values. σ and ε are the values of the pair-potential parameters Lennard-Jones Parameters in the respective papers. Numbers in square brackets are the GSKE in the harmonic approximation, in which the GSKE is one-half of the GSE. The Fugate and Swenson values are calculated from the quasiharmonic expansion of the Debye temperature at intermediate temperatures, and not simply from the Debye temperature as T becomes large. The Eckert *et al.* values are calculated from an integration of the first moment of the measured phonon density of states. See text Sec. III B for details.

Reference (method)	σ (nm)	ε/k_B (K)	GSKE$/k_B$ (K)	$-E_0/k_B$ (K)
Bernardes[a] (variational with single-particle product functions)	0.274	36.23	48.0	−211
Nosanow and Shaw[b] (Hartree)	0.274	36.23	42.8	−217
Koehler[c] (self-consistent harmonic)	0.274	36.23	42.6	−220
Hansen[d] (variational with correlations)	0.2786	36.76		−228
Fugate and Swenson[e] (specific heat)			[39] 78 (GSE)	
Eckert *et al.*[f] (neutron phonon density of states)	0.2818	36.81	[39.0] 78.0 (GSE)	
Present work (momentum distribution)			49.1±2.8	
McConville[g] (vapor pressure)				−232.5±1.0

[a] Reference 46.

[b] Reference 47.

[c] Reference 5.

[d] Reference 9.

[e] Reference 41.

[f] Reference 27.

[g] G. T. McConville, J. Chem. Phys. **60**, 4093 (1974).

and also harmonic values of the GSKE (one-half of the GSE) for several of the theoretical papers and experimental measurements. It is important to note that our measured GSKE is much larger than any values inferred from harmonic models; this shows that anharmonicity in solid neon is substantial.

Nosanow and Shaw[47] made a Hartree calculation which yielded a slightly lower GSKE, and a sublimation energy $E_0/k_B = 217$ K. Since the experimental value of the sublimation energy is 232.5 ± 1.0 K, and the Hartree model yields the lowest upper bound for a variational calculation, it became necessary to include particle correlations in order to obtain better agreement with experiment. Koehler[11] used a self-consistent harmonic theory to get a similar kinetic energy and a slightly lower sublimation energy. Hansen[9] made a variational calculation using a Jastrow cutoff function to account for correlations between atoms, and calculated a good value of the sublimation energy, as well as a value for $\langle u^2 \rangle$, the average atomic deviation from the equilibrium lattice site. Goldman[14] and Kanney[16] have also calculated values of $\langle u^2 \rangle$. Unfortunately, no value of the kinetic energy was quoted, and one cannot directly obtain $\langle E_k \rangle$ from a value of $\langle u^2 \rangle$ alone.

The presence of three-body interactions has also been invoked as having importance in properties of condensed neon.[48] Careful determinations of the neon pair potential now allow for the investigation of this possibility.[49] However, whether this element is a factor, either in the discrepancies in $\langle E_k \rangle / k_B$ values noted above, or implicit in the other calculations, one cannot say. It would have been helpful for the various publications depending upon extensive numerical calculations to have included a value for GSKE. Now that direct measurements have been made, we hope that future accounts of numerical work will do so.

IV. CONCLUSION

We have directly measured the single-particle scattering of solid natural neon over a broad range of temperature and determined its ground-state kinetic energy to be 49.1 ± 2.8 K, within the assumption that $n(p)$ is Gaussian. No published theory correctly predicts the GSKE, although calculations by both Bernardes[3] and Koehler[5] show that the kinetic energy should be substantially larger than one-half of the total E_{vib}.[50] This same conclusion arises experimentally, by comparison of our data with the neutron-scattering data of Eckert *et al.*[27] and with the specific-heat data of Fugate and Swenson.[41] There is a similar experimental result for another moderately quantum solid, hydrogen.[38]

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[*] Present address: Institute for Defense Analyses, 1801 N. Beauregard Street, Alexandria, VA 22311.

[†] Present address: Amoco Research Center, P.O. Box 3011, Naperville, IL 60566.

[1] V. Deitz, J. Chem. Phys. **2**, 296 (1934).

[2] J. DeBoer, Physica **14**, 139 (1948).

[3] N. Bernardes and C. A. Swenson, in *Solids Under Pressure,* edited by W. Paul and D. M. Warschauer (McGraw-Hill, New York, 1963), p. 101.

[4] *Rare Gas Solids,* edited by M. L. Klein and J. A. Venables (Academic, New York, 1976), Vol. I and Vol. II (1977).

[5] T. R. Koehler, Phys. Rev. Lett. **17**, 89 (1966).

[6] T. R. Koehler, in *Dynamical Properties of Solids,* edited by G. K. Horton and A. A. Maradudin (American Elsevier, New York, 1975), Vol. II, p. 148.

[7] N. S. Gillis, N. R. Werthamer, and T. R. Koehler, Phys. Rev. **165**, 951 (1968).

[8] V. V. Goldman, G. K. Horton, and M. L. Klein, J. Low Temp. Phys. **1**, 391 (1968).

[9] J. -P. Hansen, Phys. Rev. **172**, 919 (1968).

[10] J.-P. Hansen and M. L. Klein, J. Phys. (Paris) Lett. **35**, L-29 (1974).

[11] T. R. Koehler, Phys. Rev. Lett. **22**, 777 (1969).

[12] V. V. Goldman, G. K. Horton, and M. L. Klein, Phys. Rev. Lett. **24**, 1424 (1970).

[13] H. Horner, Solid State Commun. **9**, 79 (1971); L. Bohlin, *ibid.* **9**, 141 (1971); **10**, 1219 (1972).

[14] V. V. Goldman, Phys. Rev. **174**, 1041 (1968).

[15] G. A. Wolfe and B. Goodman, Phys. Rev. **178**, 1171 (1969).

[16] L. B. Kanney, J. Low Temp. Phys. **12**, 239 (1973).

[17] E. V. Zarochentsev, K. B. Tolpygo, and E. P. Troitskaya, Fiz. Nizk. Temp. **5**, 1324 (1979) [Sov. J. Low Temp. Phys. **5**, 626 (1979)].

[18] G. A. Heiser, R. C. Shukla, and E. R. Cowley, Phys. Rev. B **33**, 2158 (1986).

[19] F. N. Kabat, R. C. Windecker, and R. O. Simmons (unpublished).

[20] V. S. Kogan, B. G. Lazarev, and R. F. Bulatova, Zh. Eksp. Teor. Fiz. **40**, 29 (1961) [Sov. Phys. JETP **13**, 19 (1961)]; L. H. Bolz and F. A. Mauer, Adv. X Ray Anal. **6**, 242 (1963).

[21] D. N. Batchelder, D. L. Losee, and R. O. Simmons, Phys. Rev. **173**, 873 (1968).

[22] J. Bigeleisen, J. Chem. Phys. **34**, 1485 (1961); J. Bigeleisen and E. Roth, *ibid.* **35**, 68 (1961).

[23] K. Clusius, P. Flubacher, V. Piesbergen, K. Scheich, and A. Sperandio, Z. Naturforsch. **15a**, 1 (1960); E. Somoza and H. Fenichel, Phys. Rev. B **3**, 3434 (1971).

[24] J. A. Leake, W. B. Daniels, J. Skalyo, Jr., B. C. Frazer, and G. Shirane, Phys. Rev. **181**, 1251 (1969).

[25] J. Skalyo, Jr., V. J. Minkiewicz, G. Shirane, and W. B. Daniels, Phys. Rev. B **6**, 4766 (1972).

[26] Y. Endoh, G. Shirane, and J. Skalyo, Jr., Phys. Rev. B **11**,

1681 (1975).

[27] J. Eckert, W. B. Daniels, and J. D. Axe, Phys. Rev. B **14**, 3649 (1976).

[28] J. W. M. Dumond, Rev. Mod. Phys. **5**, 1 (1933).

[29] D. A. Peek, M. C. Schmidt, I. Fujita, and R. O. Simmons, Jpn. J. Appl. Phys. Suppl. **26-3**, 451 (1987).

[30] P. C. Hohenberg and P. M. Platzman, Phys. Rev. **152**, 198 (1966).

[31] D. A. Peek, M. C. Schmidt, I. Fujita, and R. O. Simmons, preceding paper, Phys. Rev. B **45**, 9671 (1992).

[32] V. F. Sears, Can. J. Phys. **63**, 68 (1985).

[33] C. K. Loong, S. Ikeda, and J. M. Carpenter, Nucl. Instrum. Methods A **260**, 381 (1987).

[34] Linde Division of Union Carbide Corporation.

[35] P. E. Sokol, D. A. Peek, R. O. Simmons, D. L. Price, and R. O. Hilleke, Phys. Rev. B **33**, 7787 (1986), found no measurable effects due to crystalline anisotropy in the GSKE of hcp ^{4}He.

[36] R. C. Blasdell (private communication).

[37] R. O. Hilleke, P. Chaddah, R. O. Simmons, D. L. Price, and S. K. Sinha, Phys. Rev. Lett. **52**, 847 (1984).

[38] K. W. Herwig, J. L. Gavilano, M. C. Schmidt, and R. O. Simmons, Phys. Rev. **41**, 96 (1990).

[39] D. A. Peek and R. O. Simmons, J. Chem. Phys. **94**, 3169 (1991).

[40] G. Gilat and L. J. Raubenheimer, Phys. Rev. **144**, 390 (1966).

[41] R. Q. Fugate and C. A. Swenson, J. Low Temp. Phys. **10**, 317 (1973).

[42] H. Fenichel and B. Serin, Phys. Rev. **142**, 490 (1966).

[43] C. Domb and L. Salter, Philos. Mag. **43**, 1083 (1952).

[44] T. H. K. Barron, W. T. Berg, and J. A. Morrison, Proc. R. Soc. London **A242**, 478 (1957).

[45] M. P. Tosi and F. G. Fumi, Phys. Rev. **131**, 1458 (1963).

[46] N. Bernardes, Phys. Rev. **112**, 1534 (1958).

[47] L. H. Nosanow and G. L. Shaw, Phys. Rev. **128**, 546 (1962).

[48] G. G. Chell and I. J. Zucker, J. Phys. C **1**, 35 (1967); G. G. Chell, *ibid.* **1**, 839 (1968); V. V. Goldman and M. L. Klein, J. Low Temp. Phys. **22**, 501 (1976); Yu. V. Petrov, Fiz. Tverd. Tela (Leningrad) **25**, 2399 (1983) [Sov. Phys. Solid State **25**, 1377 (1983)].

[49] R. A. Aziz, W. J. Meath, and A. R. Allnatt, Chem. Phys. **78**, 295 (1983).

[50] Long ago it was noted that for quantum solids, harmonic division of the GSE into equal kinetic and potential energy parts yields too low a value for the zero-point kinetic energy: J. de Boer and J. S. Blaisse, Physica **14**, 149 (1948).