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Density dependence of the roton spectrum in liquid ^{4}He[†]

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The density dependence of the roton spectrum in liquid ^{4}He is calculated using Brillouin-Wigner perturbation theory. The trial ground-state wave function is an extended Jastrow function, including three-body factors. The three-body factors produce an improved density dependence over the pure Jastrow trial function. We note, however, that the amount of improvement depends upon the particular approximation chosen for the three-particle structure function, $S^{(3)}(\vec{k}_1, \vec{k}_2, \vec{k}_3)$. Specifically, better agreement with experiment is obtained by using an approximation for $S^{(3)}$ which has been obtained recently in the density-phase variable theory of the weakly interacting Bose gas, when compared to the convolution approximation for $S^{(3)}$.

The study of the density dependence of the elementary excitation energy in liquid ^{4}He has received much attention in the past few years. Dietrich *et al.*[1] have studied the elementary excitation spectrum in liquid ^{4}He as a function of pressure by inelastic neutron scattering. A theoretical study of the density dependence of the roton parameters has been done by Bartley *et al.*[2] using the Brillouin-Wigner theory first employed by Jackson and Feenberg.[3] Similarly, Padmore and Chester[4] have extended the backflow calculation of Feynman and Cohen[5] to finite pressure. Both of these calculations[2,4] have obtained a weaker density dependence of the roton gap Δ and a stronger density dependence of the roton curvature μ than observed experimentally, where the roton spectrum is parametrized as

$$\epsilon_{\mathrm{rot}}(k) = \Delta + \hbar^2(k-k_0)^2/2\mu . \tag{1}$$

In this note we investigate several possible explanations for this discrepancy.

The Feynman-Cohen[5] and Jackson-Feenberg[3] theories of the elementary excitation spectrum are both based on a trial excited-state wave function $\Psi_{\vec{k}}$ in the form of an admixture of single-phonon and two-phonon states:

$$\Psi_{\vec{k}} = |\vec{k}\rangle + \frac{1}{2}\sum_{\vec{l}\neq 0,\vec{k}} A_{\vec{k},\vec{l}}\,|\vec{k}-\vec{l},\vec{l}\rangle , \tag{2}$$

where single-phonon and two-phonon states are defined as

$$|\vec{k}\rangle = \rho_{\vec{k}}\Psi_0/[NS(k)]^{1/2} , \tag{3}$$

$$|\vec{k}-\vec{l},\vec{l}\rangle = \rho_{\vec{k}-\vec{l}}\,\rho_{\vec{l}}\,\Psi_0/[N^2S(|\vec{k}-\vec{l}|)S(l)]^{1/2} , \tag{4}$$

where

$$\rho_{\vec{k}} = \sum_{i=1}^{N} e^{i\vec{k}\cdot\vec{r}_i} \tag{5}$$

represents the density fluctuation operator for the N-particle system, Ψ_0 is the ground-state wave function,

$$H\Psi_0 = E_0\Psi_0 \tag{6}$$

and $S(k)$ is the ground-state liquid structure,

$$S(k) = (1/N)\langle\Psi_0|\rho_{\vec{k}}\,\rho_{-\vec{k}}|\Psi_0\rangle . \tag{7}$$

The single-phonon state is the well-known Feynman wave function with Bijl-Feynman excitation energy $\epsilon_0(k)$, where

$$\langle\vec{k}|H|\vec{k}\rangle = E_0 + \epsilon_0(k) , \tag{8}$$

$$\epsilon_0(k) = \hbar^2k^2/2mS(k) . \tag{9}$$

Feynman and Cohen choose a form of $A_{\vec{k},\vec{l}}$ in Eq. (2) which is motivated by a dipolar back flow model.[5] Jackson and Feenberg use the Brillouin-Wigner perturbation theory form for $A_{\vec{k},\vec{l}}$.

$$A_{\vec{k},\vec{l}} = \frac{\langle\vec{k}-\vec{l},\vec{l}\,|\delta H|\vec{k}\rangle}{\epsilon(k)-\epsilon_0(|\vec{k}-\vec{l}|)-\epsilon_0(l)} , \tag{10}$$

where

$$\delta H = H - E_0 - \epsilon_0(k) . \tag{11}$$

The corresponding Brillouin-Wigner energy $\epsilon(k)$ is given by the solution of the transcendental equation

$$\epsilon(k) = \epsilon_0(k) + \frac{1}{2}\sum_{\vec{l}\neq 0,\vec{k}} \frac{|\langle\vec{k}-\vec{l},\vec{l}\,|\delta H|\vec{k}\rangle|^2}{\epsilon(k)-\epsilon_0(|\vec{k}-\vec{l}|)-\epsilon_0(l)} . \tag{12}$$

The interaction matrix element in this equation can be expressed as

$$\langle\vec{k}-\vec{l},\vec{l}\,|\delta H|\vec{k}\rangle = \frac{\hbar^2}{2m}[NS(k)S(|\vec{k}-\vec{l}|)S(l)]^{-1/2} \times\left(\vec{k}\cdot(\vec{k}-\vec{l})S(l) + \vec{k}\cdot\vec{l}\,S(|\vec{k}-\vec{l}|) - \frac{k^2}{S(k)}S^{(3)}(\vec{k}-\vec{l},\vec{l},-\vec{k})\right) , \tag{13}$$

where

$$S^{(3)}(\vec{k}-\vec{l},\vec{l},-\vec{k})=(1/N)\langle\Psi_0|\rho_{\vec{k}-\vec{l}}\,\rho_{\vec{l}}\,\rho_{-\vec{k}}|\Psi_0\rangle \quad (14)$$

is the three-particle structure function which is kinematically related to the ground-state wave function.

The original calculations of the energy were carried out at equilibrium density, $\rho_0 = 0.0218$ Å^{-3}. The three-particle structure function $S^{(3)}$ (and in the Feynman-Cohen calculation the four-particle structure function) was approximated in terms of the liquid structure function $S(k)$, which is available from experiment. In particular, Jackson and Feenberg used the convolution approximation

$$S^{(3)}(\vec{k}-\vec{l},\vec{l},-\vec{k})\approx S(|\vec{k}-\vec{l}|)S(l)S(k)\,. \quad (15)$$

Currently available experimental information about $S(k)$ at finite pressure is inadequate for the purpose of carrying out these calculations at higher density. Consequently Bartley *et al.*[2] and Padmore and Chester[4] employ a Jastrow trial function of the form

$$\Psi_J=\prod_{i<j}^{N} e^{u(r_{ij})/2} \quad (16)$$

in place of the ground-state wave function Ψ_0. The most commonly used single-parameter trial function

$$u(r) = -(b/r)^5 \quad (17)$$

was used in both calculations,[2,4] with b chosen at each density to minimize the expectation value of the Hamiltonian in Ψ_J.

The poor density dependence of the roton spectrum may be attributed in part to the inadequacy of the trial ground-state wave function. That conclusion is supported by the fact that Padmore and Chester carried out their calculations by using a Monte Carlo procedure, thereby avoiding the necessity of approximating the multiparticle correlation functions in terms of $S(k)$. To investigate the effect of improving the wave function, we take the extended Jastrow function as the trial function, which has the form[6,7]

$$\Psi=\prod_{i<j}^{N} e^{u_2(r_{ij})/2}\prod_{i<j<k}^{N} e^{u_3(\vec{r}_i,\vec{r}_j,\vec{r}_k)/2}\,. \quad (18)$$

We determine u_2 and u_3 by a functional variational procedure described elsewhere.[7-9] In brief summary, we first let u_3 be zero and carry out the self-consistent paired-phonon analysis in conjunction with the HNC approximation to determine the optimum Jastrow function and the corresponding liquid structure function $S_J(k)$. Since this procedure is independent of any simple parametrization such as Eq. (17), it is a more sensitive indicator of the density-dependent structure. $S_J(k)$ has a higher and sharper major peak than that obtained from the simply parametrized Jastrow function, in better agreement with experiment at ρ_0. This lowers the roton energy, as can be seen most readily by noting the close association between the major peak in $S(k)$ and the roton minimum [viz. Eq. (9)].

Once $u_2(r)$ is determined, it is fixed, and u_3 is obtained by minimizing the expectation value with respect to u_3.[7] The formal results are presented in Ref. (7), where it is also seen that the resulting $S(k)$ calculated at equilibrium density ρ_0 has a higher and sharper major peak than $S_J(k)$, improving agreement with experiment still further. More importantly for the purposes of this note, this effect becomes more significant as density is increased.[9]

In order to solve Eq. (12) for $\epsilon(k)$, one still needs to know the three-particle structure function $S^{(3)}(\vec{k}-\vec{l},\vec{l},-\vec{k})$ with u_3 included in the ground-state wave function. One could simply take the convolution approximation of Eq. (15). Since the uncertainty involved in using this approximation is hard to estimate, we use another approximation obtained from the linked cluster expansion of $S^{(3)}$ in terms of $S_J^{(n)}$ and the Fourier transform of u_3.[10] Taking only the first two terms in the expansion, we find

$$S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)\approx S_J^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) + S_J(k_1)S_J(k_2)S_J(k_3)C_3(\vec{k}_1,\vec{k}_2,\vec{k}_3)\,,\qquad \vec{k}_1+\vec{k}_2+\vec{k}_3=0 \quad (19)$$

where

$$u_3(\vec{r}_1,\vec{r}_2,\vec{r}_3)=\frac{1}{N^2}\sum_{\vec{k}_1,\vec{k}_2,\vec{k}_3}\delta_{\vec{k}_1+\vec{k}_2+\vec{k}_3,0}\,e^{i\vec{k}_1\cdot\vec{r}_1}e^{i\vec{k}_2\cdot\vec{r}_2}\times e^{i\vec{k}_3\cdot\vec{r}_3}C_3(\vec{k}_1,\vec{k}_2,\vec{k}_3)\,.$$

If we then use the convolution approximation for the Jastrow three-particle structure function $S_J^{(3)}$, an approximation which has some support in numerical studies of Jastrow functions[11] and cluster analysis of Jastrow functions,[12] we obtain

$$S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)\approx S_J(k_1)S_J(k_2)S_J(k_3)[1+C_3(\vec{k}_1,\vec{k}_2,\vec{k}_3)]\,,\qquad \vec{k}_1+\vec{k}_2+\vec{k}_3=0\,, \quad (20)$$

a result first obtained by Feenberg and Kilic based upon an analysis of the Schrödinger equation for the extended Jastrow function.[13] Using the expression for C_3 from Ref. 7, Eq. (20) becomes

$$S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3) \approx -\{\vec{k}_1\cdot\vec{k}_2 S_J(k_3)[1+S_J(k_1)S_J(k_2)] + \vec{k}_2\cdot\vec{k}_3 S_J(k_1)[1+S_J(k_2)S_J(k_3)] + \vec{k}_1\cdot\vec{k}_3 S_J(k_2)[1+S_J(k_1)S_J(k_3)]\}\Big/\left(\frac{k_1^2}{S_J(k_1)}+\frac{k_2^2}{S_J(K_2)}+\frac{k_3^2}{S_J(k_3)}\right), \quad \vec{k}_1+\vec{k}_2+\vec{k}_3=0. \tag{21}$$

Several authors have obtained this form of $S^{(3)}$ in the density-phase variable theory of the ground state of a weakly interacting Bose fluid.[14,15] Particularly noteworthy is Berdahl's observation that Eq. (21) has the correct long-wavelength behavior (consistent with Landau's theory of quantum hydrodynamics) while the convolution approximation [Eq. (15)] fails this test.[14] We should emphasize here, however, that our derivation of the approximation as it appears in Eqs. (19) and (20) is purely kinematical, having nothing to do with the Hamiltonian.

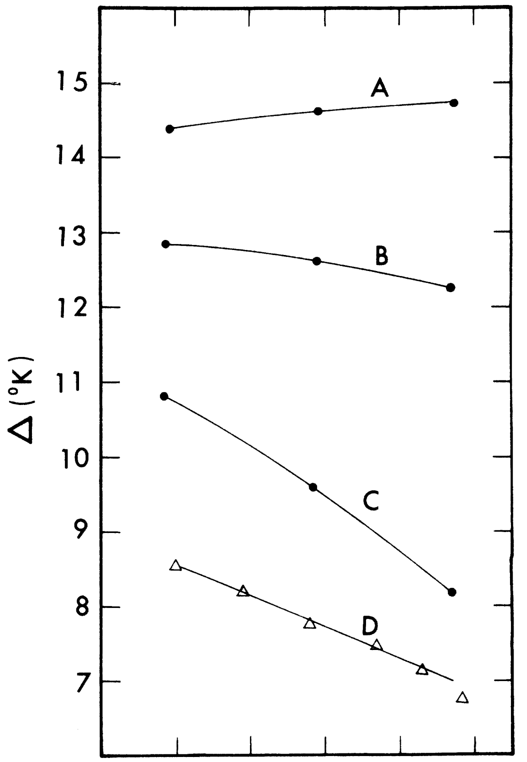

FIG. 1. Density dependence of the roton gap Δ. Curve A is obtained by using optimal Jastrow wave function for the ground-state wave function and using convolution approximation for $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$. Curve B is obtained by including U_3 in the ground-state wave function and using the convolution approximation for $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$. Curve C is obtained by including U_3 in the ground-state wave function. However, the approximation given in Eq. (21) is used for $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$. Curve D is the experimental results of Dietrich *et al.* (see Ref. 1). The theoretical values are at the densities of the solid circles. The curves are simply guides to the eye.

Our calculation is done at three densities, $\rho = 0.02185$, 0.02388, and 0.02571 Å^{-3}. At a given density we first carry out the self-consistent paired-phonon analysis to obtain the optimum Jastrow wave function and the corresponding liquid structure factor $S_J(k)$. The convolution approximation is used for the three-particle structure function $S_J{}^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$. The excitation energy is obtained by solving Eq. (12). The density dependence of the roton parameters Δ, k_0, and μ are plotted as curve A in Figs. 1, 2, and 3 respectively. Curves B in these three figures are obtained by including u_3 in the ground-state wave function and using the convolution approximation for $S^{(3)}(\vec{k}_1,\vec{k}_2,\vec{k}_3)$. In curves C, however, the approximation given in Eq. (21) is used for $S^{(3)}$. Curves D of the three figures are the experimental results of Dietrich *et al.* at low temperature.[1]

It is clear in these figures that the inclusion of three-particle factors in the trial ground-state wave function improves the density dependence of the roton spectrum. Note from the comparison of curves B and C that the roton parameters depend sensitively on the choice of approximation used for $S^{(3)}$, especially at high densities. This suggests the need for further assessment of these approximations by a more accurate procedure

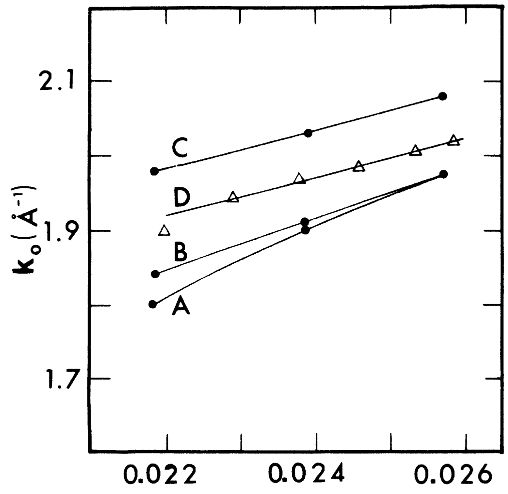

FIG. 2. Density dependence of the roton momentum k_0. Curves A, B, C, and D as in Fig. 1.

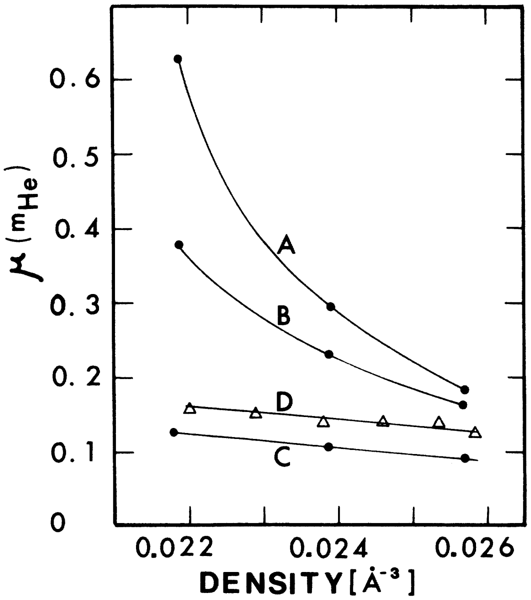

FIG. 3. Density dependence of the roton curvature parameter μ. Curves A, B, C, and D as in Fig. 1.

such as a Monte Carlo integration as in the treatment of the pure Jastrow trial function by Padmore and Chester.[4] We note, however, that the approximation in Eq. (21) produces better over-all agreement with experiment than the convolution approximation.

In the present work we have simplified the calculation of the roton parameters by three approximations: First, we neglect the slight nonorthogonality of the single-phonon and two-phonon states; second, we consider only the three phonon vertex [Eq. (13)]; and, third, we neglect third- and higher-order terms in the Brillouin-Wigner energy series. In this regard we note that Lee and Lee have recently done an extensive calculation at equilibrium density by including four-phonon vertices and considering the nonorthogonality of single-phonon, two-phonon, and three-phonon states.[16] They calculate the excitation spectrum to fourth order and find a roton gap which is only one degree above the experimental value. They begin with the optimum Jastrow function and then include three-phonon contributions to the ground state by perturbation theory. In that manner they have included some of the effects of three-body factors which we include in this note. They also include corrections to the convolution approximation for $S^{(3)}$ when it appears in lowest order. Consequently their procedure includes some of the important features of our present calculation, and we expect that if their calculation were carried out as a function of density it would provide further improved agreement with experiment. We note, however, that an intermediate procedure whereby one begins with the optimum extended Jastrow function including three-body factors [Eq. (18)] would simplify their calculation somewhat and would put their roton spectrum at ρ_0 even closer to the experimental value.

Finally we note that all of the recent theoretical calculations[2,4,16] including the present one use the Lennard-Jones 6-12 potential in the Hamiltonian. We cannot exclude the possibility that a more realistic interaction would bring about further improvements.

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