Roton Excitation Spectrum in Liquid Helium II

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We derive a one-particle quantum equation for roton excitations by considering rotons as bound states of helium atoms. Our approach is based on a self-similar solution of the nonlinear Schrödinger equation with a self-consistent confining potential in a roton cluster surrounded by the condensate. The symmetric vibrational excitation of the roton determines the gap in the energy spectrum of the elementary excitations in liquid He II. Analysis of the scattering process of long-wavelength neutrons from the liquid on the basis of our theory leads directly to Landau's roton spectrum of excitations with an effective roton mass and energy spectrum gap that are in quantitative agreement with the experimental data.

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Sixty years ago, Landau [1,2] phenomenologically introduced his famous roton energy spectrum to explain the behavior of liquid helium II. Bogoliubov [3] developed a theory for the elementary excitations of a dilute Bose gas, and later there were several derivations to higher order of the ground state of a low density Bose gas by Lee, Yang, and Huang [4,5]. A rigorous consideration of the leading term in the energy of a low density Bose gas has been recently presented in Ref. [6]. The relation between the energy spectrum of the elementary excitations of liquid ^{4}He and the structure factor of the liquid was found by Feynman [7] and later improved by Feynman and Cohen [8]. Moreover, Feynman proposed a model of the roton excitation structure [9] as a vortex ring with characteristic size of the order of the mean atomic distance in the liquid helium II. Vortex rings carrying "quantum circulation" were observed experimentally by Rayfield and Reif [10], confirming that Feynman's mechanism is responsible for the breakdown of superfluidity in liquid helium II [11,12]. To date, however, there has been no experimental confirmation that the vortex ring and roton are the same excitations in liquid helium.

In this Letter we present a new model of the roton structure and on this basis develop a theory which leads to Landau's roton energy excitation spectrum in liquid helium II and is in agreement with the experimental data obtained by neutron scattering from the liquid. The full theory of the roton excitation spectrum should include both the many-body interactions in liquid helium II and an analysis of the scattering process of neutrons from the liquid [13,14]. Our treatment is based on the assumption that in liquid ^{4}He at temperature $0.6 \lesssim T \lesssim 1.2$ K there exist stable clusters which are bound states of some number N_c of atoms ^{4}He ($N_c \gg 1$). The number N_c of atoms in such a roton cluster can be found by minimizing the free energy, which constitutes the stability condition of the roton. Our estimates show that the stable cluster should have approximately spherical shape and $N_c = 13$. The mean radius $\overline{a}$ of the cluster can be found from $N_c = \rho V_c/m$, where $\rho = 145\ \mathrm{kg/m^3}$ is the density of liquid helium [15] and V_c is the cluster volume, giving $\overline{a} = [3mN_c/(4\pi\rho)]^{1/3} \simeq 5.22$ Å at $N_c = 13$. A necessary condition for the existence of a roton cluster is $\lambda_D \gtrsim 2\overline{a}$, where $\lambda_D = 2\pi\hbar(3mk_BT)^{-1/2}$ is the thermal wavelength, and hence $T \lesssim 1.2$ K [16].

We derive a nonlinear equation of motion for the mean field (order parameter) $\psi_R(\mathbf{x},t) = \langle\hat{\psi}_\alpha(\mathbf{x},t)\rangle$ of the roton by assuming that all atoms in the cluster are in the same quantum state and by using a density matrix of the form $\rho = \rho_R \otimes \rho_0$, where ρ_R and ρ_0 are the matrices of the roton and of the surrounding equilibrium condensate at fixed temperature [17]:

$$i\hbar\,\frac{\partial\psi_R}{\partial t} = \left(-\frac{\hbar^2\nabla^2}{2m} + \tilde{U}_0|\psi_R|^2 + U_A(\mathbf{x})\right)\psi_R\,. \quad (1)$$

Here $\int_{V_c}|\psi_R|^2 d^3\mathbf{x} = N_c$, $\tilde{U}_0 = (4\pi a_0\hbar^2/m)(N_c - 1)/N_c$ is the repulsive interaction constant multiplied by Hartree and normalization factors and $a_0 \simeq 2.2$ Å is the s-wave scattering length of ^{4}He atoms [12]. The confining potential $U_A(\mathbf{x})$ in Eq. (1) takes into account all forces between atoms of the cluster and those of the surroundings, and also long-range many-body attractive forces among atoms of the cluster. We assume that the forces acting on the atoms in the roton cluster have an oscillatory character; i.e., the time-averaged forces are equal to zero, which leads to the following equation for the self-consistent confining potential (valid when $\psi_R \neq 0$):

$$\tilde{U}_0\overline{|\psi_R(\mathbf{x},t)|^2} + U_A(\mathbf{x}) = \Lambda\,, \quad (2)$$

where the overline indicates time averaging and Λ is a constant. We should thus solve the self-consistent system of Eqs. (1) and (2) where Eq. (1) formally has the form of a Gross-Pitaevskii equation [18,19] and where Eq. (2) defines an unknown confining potential which depends on the spatial coordinate $\mathbf{x}$. This complicated problem has a self-similar solution when the dimensionless parameter $\varepsilon_c = \hbar^2(2\tilde{U}_0\rho\overline{a}^2)^{-1}$ is small, a condition that is in practice verified for the above parameters: $\varepsilon_c = \overline{a}[6(N_c - 1)a_0]^{-1} \simeq 0.033$. Using the self-similarity method for the nonlinear Schrödinger equation [20,21] generalized to 3D space we find the following solution

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of Eqs. (1) and (2) at $\varepsilon_c \ll 1$ for the amplitude $A(\mathbf{x}, t) = |\psi_R(\mathbf{x}, t)|$ of the roton mean field:

$$A(\mathbf{x}, t) = \left(\frac{15N_c}{8\pi a_1(t)a_2(t)a_3(t)}\right)^{1/2}\left(1 - \sum_{k=1}^{3}\frac{x_k^2}{a_k^2(t)}\right)^{1/2}, \quad (3)$$

when $\sum_{k=1}^{3}[x_k/a_k(t)]^2 < 1$ and $A(\mathbf{x}, t) = 0$ when $\sum_{k=1}^{3}[x_k/a_k(t)]^2 \geq 1$. The phase of the field $\psi_R(\mathbf{x}, t)$ can be written as

$$\phi(\mathbf{x}, t) = \phi_0(t) + \frac{m}{2\hbar}\sum_{k=1}^{3}\left(a_k^{-1}\frac{da_k}{dt}\right)x_k^2, \quad (4)$$

where

$$\phi_0(t) = \phi_0(0) - \frac{15(N_c - 1)a_0\hbar}{2m}\int_0^t \frac{dt'}{a_1(t')a_2(t')a_3(t')}, \quad (5)$$

$$\frac{d^2a_k}{dt^2} + \beta_k a_k = \frac{15(N_c - 1)a_0\hbar^2}{m^2}(a_1a_2a_3a_k)^{-1}. \quad (6)$$

This is an asymptotical solution as ε_c approaches 0, generalizing the Thomas-Fermi solution to the nonstationary case [20]. The solution implies that the confining potential $U_A(\mathbf{x})$ has the form

$$U_A(\mathbf{x}) = \frac{m}{2}\sum_{k=1}^{3}\beta_k x_k^2, \quad (7)$$

where

$$\beta_k = \frac{15(N_c - 1)a_0\hbar^2}{m^2\overline{a}_1\overline{a}_2\overline{a}_3\overline{a}_k^2}, \quad (8)$$

which really follows from Eqs. (2) and (3). We use this self-similar solution to quantize the many-body problem in the roton cluster in terms of elliptical coordinates. The Hamiltonian of the roton cluster described by Eqs. (1) and (2) for the mean field $\psi(\mathbf{x}, t) = \psi_R(\mathbf{x}, t)/\sqrt{N_c}$ (normalized to 1) has the form

$$\mathcal{H} = \int \psi^*\left(-\frac{\hbar^2\nabla^2}{2m} + U_A(\mathbf{x}) + (N_c - 1)\frac{2\pi a_0\hbar^2}{m}|\psi|^2\right)\psi\, d^3\mathbf{x}. \quad (9)$$

Using Eqs. (3)–(7) and calculating the integrals in Eq. (9) we find

$$\mathcal{H} = \sum_{k=1}^{3}\frac{\pi_k^2}{2m_0} + \frac{m_0}{2}\sum_{k=1}^{3}\beta_k a_k^2 + \frac{\mathcal{G}}{a_1a_2a_3}. \quad (10)$$

Here $\pi_k = m_0 da_k/dt$, and the renormalized mass m_0 and interaction constant $\mathcal{G}$ are given by

$$m_0 = \frac{m}{7}, \qquad \mathcal{G} = \frac{15(N_c - 1)a_0\hbar^2}{7m}. \quad (11)$$

One can check that the Hamilton equations $\dot{a}_k = \partial\mathcal{H}/\partial\pi_k$ and $\dot{\pi}_k = -\partial\mathcal{H}/\partial a_k$ lead to Eqs. (6), which means that a_k are the generalized coordinates and π_k are the appropriate canonical momenta defined by the Hamiltonian (10). The quantization of the Hamiltonian (10) yields a Schrödinger equation for the roton wave function χ:

$$i\hbar\frac{\partial\chi}{\partial t} = \left(-\frac{\hbar^2}{2m_0}\sum_{k=1}^{3}\frac{\partial^2}{\partial a_k^2} + V(a_1, a_2, a_3)\right)\chi, \quad (12)$$

where the potential $V(a_1, a_2, a_3)$ has the form

$$V(a_1, a_2, a_3) = \frac{m_0}{2}\sum_{k=1}^{3}\beta_k a_k^2 + \frac{\mathcal{G}}{a_1a_2a_3}. \quad (13)$$

The parameters β_k should satisfy the stationarity conditions $(\partial V/\partial a_k)_{a_k=\overline{a}_k} = 0$, which leads again to Eqs. (8). To find the oscillatory solutions of Eq. (12), we expand the potential (13) up to the second order in terms of the variables $y_k = (a_k/\overline{a}_k) - 1$, where $\overline{a}_k = \overline{a}$; i.e., we assume spherical symmetry for the averaged elliptical parameters. Diagonalization of this quadratic polynomial by the linear transformation $z_1 = \alpha y_1 + \beta y_2 + \gamma y_3$, $z_2 = \beta y_1 + \alpha y_2 + \gamma y_3$, and $z_3 = \gamma(y_1 + y_2 + y_3)$ with $\alpha = -[1/3 + \sqrt{3}/6]^{1/2}$, $\beta = [1/3 - \sqrt{3}/6]^{1/2}$, and $\gamma = 1/\sqrt{3}$ yields the following stationary eigenstates:

$$\langle z_1, z_2, z_3 \,|\, n\rangle = C_n \exp\left(-\frac{1}{2}\sum_{k=1}^{3}\xi_k^2\right) \times H_{n_1}(\xi_1)H_{n_2}(\xi_2)H_{n_3}(\xi_3). \quad (14)$$

Here $n = (n_1, n_2, n_3)$ and $n_k = 0, 1, 2, \ldots$; $H_n(\xi)$ are the Hermite polynomials, with $\xi_k = (m_0\omega_k/\hbar)^{1/2}z_k$, where ω_k are given by

$$\omega_1 = \omega_2 = \sqrt{2/5}\,\omega_3, \qquad \omega_3 = \frac{5[3(N_c - 1)a_0]^{1/2}\hbar}{m\overline{a}^{5/2}}. \quad (15)$$

The energy spectrum of these stationary solutions (14) is

$$E_n = \hbar\omega_\perp(n_1 + n_2 + 1) + \hbar\omega_3(n_3 + 1/2) + \overline{V}, \quad (16)$$

where $\omega_\perp = \sqrt{2/5}\,\omega_3$ and $\overline{V} = 5\mathcal{G}/(2\overline{a}^3)$. For s-wave scattering processes of neutrons by a roton cluster in its ground state $|000\rangle$, the lowest possible excitations of the roton are $|001\rangle$ and $|110\rangle$ because the transitions $|000\rangle \rightarrow |100\rangle$ and $|000\rangle \rightarrow |010\rangle$ are forbidden by the symmetry of the scattering problem under reflection $x_1 \rightarrow x_2$, $x_2 \rightarrow x_1$ [17]. So the lowest excitation state of a roton by a neutron is the symmetric vibration excitation $|001\rangle$, with transition vibrational energy $\hbar\omega_3$.

The measurement of the roton energy spectrum in liquid He II by scattering long-wavelength neutrons [13,14] is based on conservation laws that for our model of roton cluster have the form

$$\frac{\mathbf{p}_\mathrm{i}^2}{2m_\mathrm{n}} + K_\mathrm{i} + E_\mathrm{i} = \frac{\mathbf{p}_\mathrm{f}^2}{2m_\mathrm{n}} + K_\mathrm{f} + E_\mathrm{f}\,, \qquad \mathbf{p}_\mathrm{i} + \mathbf{P}_\mathrm{i} = \mathbf{p}_\mathrm{f} + \mathbf{P}_\mathrm{f}\,, \tag{17}$$

where $\mathbf{p}_\mathrm{i}$, $\mathbf{P}_\mathrm{i}$ and $\mathbf{p}_\mathrm{f}$, $\mathbf{P}_\mathrm{f}$ are the initial and final momenta of the neutron and roton clusters, K_i, E_i and K_f, E_f are the initial and final kinetic and vibrational energies of the roton cluster, m_n is the neutron mass, and the mass of the roton cluster is $M = N_\mathrm{c} m$. The definition of the energy of elementary excitation $E(p)$ [13,14] with momentum $p = |\mathbf{p}_\mathrm{i} - \mathbf{p}_\mathrm{f}|$ by Eq. (17) can be written

$$E(p) \equiv \frac{\mathbf{p}_\mathrm{i}^2}{2m_\mathrm{n}} - \frac{\mathbf{p}_\mathrm{f}^2}{2m_\mathrm{n}} = \hbar\omega_3 + K_\mathrm{f} - K_\mathrm{i}\,. \tag{18}$$

Considering a quadratic term in the vicinity of a local minimum $p = p_0$ of the roton energy spectrum with respect to variable $\delta = p - p_0$ we find by Eqs. (17) and (18) the average energy of roton excitation under initial momenta P_i is

$$\langle E(p)\rangle = \hbar\omega_3 + \frac{2N_\mathrm{c}\langle\mathcal{K}\rangle}{p_0^2}(p - p_0)^2. \tag{19}$$

Here $\mathcal{K}$ is the initial kinetic energy per particle of the roton cluster $K_\mathrm{i} = N_\mathrm{c}\mathcal{K}$. Writing $\langle E(p)\rangle = \varepsilon(p)$, the roton excitation spectrum is in Landau's form [9,15]

$$\varepsilon(p) = \Delta + \frac{(p - p_0)^2}{2\mu_\mathrm{R}}\,, \tag{20}$$

where we find from Eq. (19) the explicit results for the roton energy gap and effective roton mass

$$\Delta = \frac{5[3(N_\mathrm{c} - 1)a_0]^{1/2}\hbar^2}{m\overline{a}^{5/2}}, \qquad \mu_\mathrm{R} = \frac{p_0^2}{4N_\mathrm{c}\langle\mathcal{K}\rangle}\,. \tag{21}$$

In the general case, the average kinetic energy of a particle in the roton cluster is the sum $\langle\mathcal{K}\rangle = \langle\mathcal{K}_\mathrm{I}\rangle + \langle\mathcal{K}_\mathrm{T}\rangle$, where $\langle\mathcal{K}_\mathrm{I}\rangle$ and $\langle\mathcal{K}_\mathrm{T}\rangle$ are internal energy and center-of-mass kinetic energy, respectively. It follows from Eq. (12) that the internal kinetic energy can be written

$$\langle\mathcal{K}_\mathrm{I}\rangle = \int \frac{p^2}{2m_0} W(\mathbf{p}) d^3\mathbf{p} = \sum_{k=1}^{3} \frac{\hbar\omega_k}{4}\coth\left(\frac{\hbar\omega_k}{2k_\mathrm{B}T}\right), \tag{22}$$

where $W(\mathbf{p}) = \langle\mathbf{p}|\rho_\mathrm{R}|\mathbf{p}\rangle$ is the momentum distribution

$$W(\mathbf{p}) = C\exp\left(-\sum_{k=1}^{3}\frac{\tanh[\hbar\omega_k/(2k_\mathrm{B}T)]}{\hbar m_0\omega_k}p_k^2\right)$$

for the three-dimensional harmonic oscillator given by the wave functions (14) and energy spectrum (16) interacting with the surrounding equilibrium condensate; C is a normalization constant. At $T = 1.1$ K the internal kinetic energy given by Eqs. (15) and (22) with the above parameters has the value $\langle\mathcal{K}_\mathrm{I}\rangle/k_\mathrm{B} \simeq 4.95$ K. The center-of-mass kinetic energy of the atoms of the roton cluster may be approximated by the energy of an ideal Bose gas per particle [9]: $\langle\mathcal{K}_\mathrm{T}\rangle/k_\mathrm{B} \simeq 0.77(T/T_\mathrm{c})^{3/2}T \simeq 0.3$ K using the experimental value $T_\mathrm{c} = 2.17$ K. This yields the full kinetic energy $\langle\mathcal{K}\rangle/k_\mathrm{B} \simeq 5.25$ K.

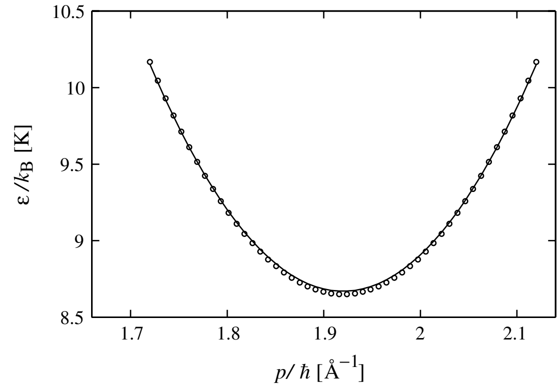

FIG. 1. Energy spectrum of roton excitations in liquid He II at a temperature $T = 1.1$ K. The circles represent the experimental data of Yarnell *et al.* [13], while the solid line is the theoretical prediction.

The simple calculation of the momentum $p_0 = \hbar k_0$ at the minimum of roton spectrum by $k_0 = 2\pi/L_0$ [9], where $L_0 = \overline{a}\sqrt{3/7}$ is the classical average distance between atoms in the cluster leads to $k_0 = 1.84$ Å^{-1}. We do not consider here the rigorous evaluation of this parameter but use instead in the calculations below the experimental value [13] $k_0 = (1.92 \pm 0.01)$ Å^{-1}, which is close to our classical estimate. Using Eqs. (21) and the parameters quoted above for $N_\mathrm{c} = 13$, we find $\Delta/k_\mathrm{B} \simeq 8.673$ K and $\mu_\mathrm{R}/m = 0.1638$, in excellent agreement with the experimental measurements of Yarnell *et al.* [13]: $\Delta/k_\mathrm{B} = (8.65 \pm 0.04)$ K; $\mu_\mathrm{R} = (0.16 \pm 0.01)$ m. Note that the only free parameter in Eqs. (21) is the number of atoms in the cluster. Choosing this to obtain the best fit to experimental values fixes $N_\mathrm{c} = 13$, the choice of $N_\mathrm{c} = 12$ or $N_\mathrm{c} = 14$ giving results in error by approximately 2%–3% for the gap in the energy spectrum. A cluster of 13 atoms has a natural classical model as a central atom surrounded by a shell of 12 atoms situated at the vertices of a regular icosahedron. The stability of this configuration is also favored by its having the greatest number (six) of nearest neighbors for each atom in a shell. We show in Fig. 1 the experimental spectrum of the roton excitations in liquid He II [13] and our theoretical prediction, given by Eqs. (20) and (21).

We also note that the (linear) phonon region of the elementary excitation spectrum turns into the roton region at a characteristic momentum $p_\mathrm{c} = \hbar k_\mathrm{c}$. In our cluster model it is natural to estimate this by $k_\mathrm{c} = 2\pi/d \simeq 0.6$ Å^{-1}, where $d = 2\overline{a}$ is the diameter of the cluster. This agrees with the experimental value [14].

In conclusion, we have proposed a new model of the roton excitation structure in liquid helium II. We have developed a theoretical treatment of the roton excitation spectrum which is in quantitative agreement with experimental data from neutron scattering.

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