Collective States of ^{3}He Clusters

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The monopole ($L=0$) and quadrupole ($L=2$) strength distributions in normal ^{3}He clusters are calculated within the random-phase approximation. We use a phenomenological, zero-range ^{3}He-^{3}He interaction to generate the Hartree-Fock single-particle spectrum and the residual particle-hole interaction. The evolution of the collective modes with the number of atoms in the cluster is discussed.

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Neutron-scattering experiments have provided a powerful tool for studying the elementary excitations of liquid helium. Collective modes (zero sound, paramagnons, rotons, etc.) have been experimentally identified [1,2] and, at present, their structure is theoretically rather well understood [3,4]. An important question is to know whether collective excitations such as multipole modes do exist in finite-size clusters. To this end, inelastic neutron scattering leading to the response functions of clusters might also be a possible way to the answer.

In spite of the fact that the experimental information of helium drops is still rather scarce [5–7], their theoretical description has attracted some interest in recent years [8–14]. Previous theoretical studies on ^{3}He clusters have mainly dealt with the description of ground-state properties following two different approaches: a Monte Carlo method using the Aziz potential [12], and a density-functional method with a phenomenological, zero-range interaction [13,14]. In general, overall good agreement between both methods has been found.

In a previous paper [15] we have estimated the energies of multipole modes in normal ^{3}He clusters using a random-phase approximation (RPA) sum-rule method. The main conclusion from that work is that the average energies show a smooth, well-defined N and L dependence. For cluster sizes $N \geq 150$, they decrease with N, qualitatively following an $N^{-1/3}$ law. For small clusters the behavior changes, especially in the monopole case for which the average excitation energy increases when going from $N=40$ to 112.

In this paper we present the first detailed calculation of the density-density response function of ^{3}He clusters. We use a zero-range, density- and momentum-dependent ^{3}He-^{3}He interaction $v(1,2)$ [13], whose parameters have been adjusted to reproduce some bulk ^{3}He properties, namely, the saturation density, the binding energy per atom, the incompressibility coefficient, the variation with pressure of the specific heat, and the liquid surface tension. This phenomenological interaction must be considered as an effective force simulating the highly complicated interaction between quasiparticles embedded in the medium. It is simple to use but nevertheless it contains important features of a finite-range interaction (for instance, it gives rise to an effective mass) through its velocity dependence. This effective interaction reads

$$\begin{aligned} v(1,2) = {} & [t_0 + t_0' \rho^\gamma]\delta(\mathbf{r}_{12}) + \overleftarrow{\mathbf{k}}[t_2 + t_2' \rho]\delta(\mathbf{r}_{12})\overrightarrow{\mathbf{k}} \\ & + \overleftarrow{\mathbf{k}}^2[\tfrac{1}{2}t_1 + \tfrac{1}{2}t_1' \rho]\delta(\mathbf{r}_{12}) \\ & + \delta(\mathbf{r}_{12})[\tfrac{1}{2}t_1 + \tfrac{1}{2}t_1' \rho]\overrightarrow{\mathbf{k}}^2, \end{aligned} \tag{1}$$

where $\overrightarrow{\mathbf{k}}$ ($\overleftarrow{\mathbf{k}}$) is the relative momentum acting to the right (left) in the matrix element and ρ is the density at point $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$. The parameters of the force are given in Table I. Using interactions of this form, it has been shown [16] that the monopole and quadrupole RPA energies depend essentially on the values of the incompressibility coefficient and the effective mass, respectively. Thus, there are good reasons to think that the present interaction is reliable for predicting $L=0$ and $L=2$ energies. With this phenomenological interaction the Hartree-Fock (HF) problem for the cluster ground state can be solved and, thus, the self-consistent mean-field and single-particle levels are obtained [14]. As in Refs. [12,

TABLE I. Parameters of the effective ^{3}He-^{3}He interaction.

t_0	-1.3638×10^3 K Å^3	t_0'	2.9034×10^6 K Å$^{3(1+\gamma)}$
t_1	1.7091×10^4 K Å^5	t_1'	6.4889×10^3 K Å^8
t_2	-6.7510×10^3 K Å^5	t_2'	1.0815×10^4 K Å^8
γ	2.1090		

14], we have found that ^{3}He clusters smaller than a certain critical size (about 30 atoms) are not bound and that the magic numbers are those of the harmonic oscillator (40, 70, 112, 168, ...). The reader is addressed to these references for details on ground-state properties.

From the effective interaction (1), the total energy of the cluster can be written as a local-density functional. The residual particle-hole interaction V is defined [17] as the second functional derivative of the cluster energy with respect to the particle density. This generalizes the definition of the particle-hole interaction to the case where the starting effective interaction between particles is density dependent. The present RPA model is self-consistent in the sense that the same interaction generates the HF mean field and the residual interaction. One gets

$$\begin{aligned} V(1,2) = & [\tfrac{1}{2}t_0 + \tfrac{1}{4}t_0'(\gamma+1)(\gamma+2)\rho^\gamma + \tfrac{1}{4}(t_1'+3t_2')\tau - \tfrac{1}{16}(3t_1'-12t_2')\Delta\rho]\delta(\mathbf{r}_{12}) \\ & - (\overleftarrow{\nabla}_1^2+\overrightarrow{\nabla}_1^2+\overleftarrow{\nabla}_2^2+\overrightarrow{\nabla}_2^2)\{\tfrac{1}{16}[t_1+3t_2+(2t_1'+6t_2')\rho]\delta(\mathbf{r}_{12})\} \\ & + (\overrightarrow{\nabla}_1+\overleftarrow{\nabla}_1)(\overrightarrow{\nabla}_2+\overleftarrow{\nabla}_2)\{\tfrac{1}{16}[t_1-9t_2+(t_1'-15t_2')\rho]\delta(\mathbf{r}_{12})\} \\ & + (\overrightarrow{\nabla}_1-\overleftarrow{\nabla}_1)(\overrightarrow{\nabla}_2-\overleftarrow{\nabla}_2)\{\tfrac{1}{16}[t_1+3t_2+(t_1'+3t_2')\rho]\delta(\mathbf{r}_{12})\}, \end{aligned} \tag{2}$$

where $\tau \equiv \sum|\nabla\phi_i|^2$ is the kinetic-energy density at point $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$. We have used the convention that the gradient operators do not act on the quantities enclosed within curly brackets.

The calculation of the excitations of ^{3}He clusters that we will present below closely follows those carried out to describe giant resonances in nuclei [18] or plasma resonances in metal clusters [19]: A discrete particle-hole basis is constructed and the Hamiltonian $H=H_0+V$, the sum of the HF Hamiltonian H_0 and the residual particle-hole interaction V, is diagonalized using the RPA. The RPA equations [20]

$$\begin{pmatrix} A & B \\ B^* & A^* \end{pmatrix}\begin{pmatrix} X^{(n)} \\ Y^{(n)} \end{pmatrix} = E_n \begin{pmatrix} X^{(n)} \\ -Y^{(n)} \end{pmatrix} \tag{3}$$

are written in terms of angular-momentum-coupled matrix elements of the residual interaction [19].

In terms of the ground state $|0\rangle$, excited states $|n\rangle$, and excitation energies E_n, the strength function $S(\omega)$ corresponding to a multipole operator Q_L is given by

$$S(\omega) = \sum_n \delta(\omega - E_n)|\langle n|Q_L|0\rangle|^2 . \tag{4}$$

We have used $Q_L = f(r)Y_{L0}$ with $f(r)=r^L$, except for the monopole mode where $f(r)=r^2$. Within RPA the transition probability from the ground state to an excited state $|n\rangle$ is

$$\langle 0|Q_L|n\rangle = \frac{1}{2L+1}\sum_{\mathrm{p,h}}\langle \mathrm{p}||Q_L||\mathrm{h}\rangle(X_{\mathrm{ph}}^{(n)} - Y_{\mathrm{ph}}^{(n)}) , \tag{5}$$

and the reduced matrix element is written in terms of the single-particle radial wave functions R_{nl} as

$$\langle \mathrm{p}||Q_L||\mathrm{h}\rangle = \langle Y_{l_\mathrm{p}}||Y_L||Y_{l_\mathrm{h}}\rangle \int_0^\infty R_{n_\mathrm{p}l_\mathrm{p}} f(r) R_{n_\mathrm{h}l_\mathrm{h}} r^2 dr . \tag{6}$$

Once the HF mean field has been calculated in coordinate space, we construct the set of single-particle states with radial wave functions R_{nl} by diagonalizing H_0 on a large harmonic-oscillator basis. The results discussed below have been obtained by using a basis of 16 oscillator shells and a harmonic-oscillator parameter $\hbar\omega = 0.64$ K. We have checked that RPA results are stable enough against variations of these choices.

To build the space of particle-hole configurations corresponding to a given L mode with parity $(-1)^L$, we keep for each hole state $(n_\mathrm{h}, l_\mathrm{h})$ the lowest m particle states of all possible l_p compatible with total angular momentum and parity. The results presented below have been obtained with $m=10$ for the monopole ($L=0$), and $m=4$ for the quadrupole ($L=2$). In the case of the quadrupole we use a smaller m because of the big number of particle-hole configurations that become available for $L=2$. We have checked that the results are essentially unchanged by increasing m from these values.

A good check of the numerical accuracy and space truncation is to evaluate the energy-weighted sum rule, which can be independently calculated by RPA sum-rule methods [15]. We have verified that this sum rule is satisfied with less than 5% error in both the monopole and the quadrupole results.

Figure 1 displays the monopole strength distributions $S(\omega)$ obtained for the three magic clusters $N=40$, 70, and 112. On the left-hand side of this figure we have plotted the unperturbed (HF) strength distributions and on the right-hand side the RPA strength distribution. The vertical lines represent the discrete states and their height gives the corresponding transition probability. For the sake of presentation, we also display the curves obtained by folding the discrete lines with Lorentzian functions having an artificial width (solid lines). The widths are taken as $\Gamma=0.2$ K for $L=0$ and $\Gamma=0.1$ K for $L=2$.

One can notice that the RPA strength of the monopole mode is more fragmented than the HF strength. This is due to the strong repulsive effect of the residual interaction which shifts the strength above the particle emission threshold and produces a resonance in the continuum, similarly to the giant resonances in atomic nuclei. The distribution of collective states is broad (around 0.6 K). This width is larger than the parameter Γ and therefore it is a genuine width reflecting the combined effects of particle escape and Landau damping. Regarding the evolution with the number N of atoms in the cluster, the cen-

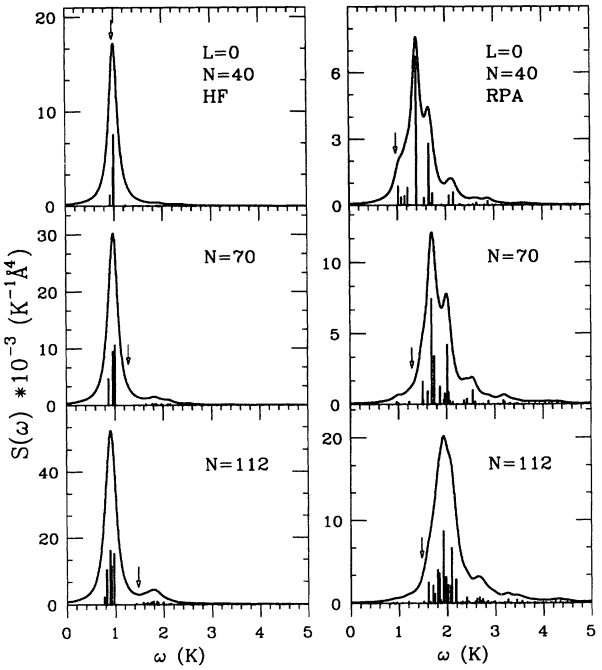

FIG. 1. Monopole strength distributions for the $N=40$, 70, and 112 clusters. The left-hand side displays the unperturbed (Hartree-Fock) distribution and the right-hand side the RPA distribution. Vertical lines give the position and strengths of the discrete states. The solid lines have been obtained by folding the discrete states with Lorentzians of width $\Gamma=0.2$ K. The strength of the discrete states has been divided by $\frac{1}{2}\pi\Gamma$. The arrows indicate the position of the continuum threshold in each cluster.

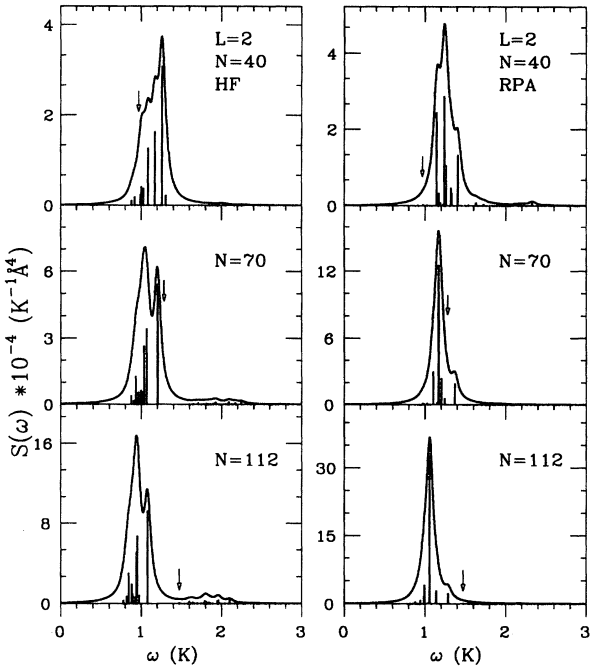

FIG. 2. Same as in Fig. 1 for the quadrupole mode. We have used $\Gamma=0.1$ K.

troid of the strength $\bar{E}$ moves to higher energies with increasing N. The N dependence of $\bar{E}$ is in good agreement with estimations based on sum rules [15]. In fact, using them one can find exact upper (E_3) and lower (E_1) bounds to the RPA strength centroid [16]. Table II collects the numerical values of $\bar{E}$, E_1, and E_3.

Figure 2 displays the quadrupole results. In this case the RPA distributions are narrower than the HF ones. This is especially marked for the $N=70$ and 112 clusters, for which a single line exhausts 60% and 72% of the quadrupole strength, respectively. We have found that the quadrupole mode for $N \geq 70$ lies in the discrete part of the spectrum. This explains why this mode is less Landau damped than the monopole mode. Also, the evolution with N of the collective mode is opposite to the monopole case, now slowly decreasing in energy with increasing size (Table II).

Another difference between the $L=0$ and $L=2$ modes is that the monopole mode involves volume oscillations of the cluster while the quadrupole mode is a surface vibra-

TABLE II. Centroid $\bar{E}$ of the calculated RPA strength and exact upper (E_3) and lower (E_1) bounds (in K).

		$L=0$		$L=2$	
N	E_1	$\bar{E}$	E_3	$\bar{E}$	E_3
40	1.5	1.6	1.8	1.3	1.3
70	1.9	2.0	2.2	1.2	1.2
112	2.0	2.1	2.3	1.1	1.1

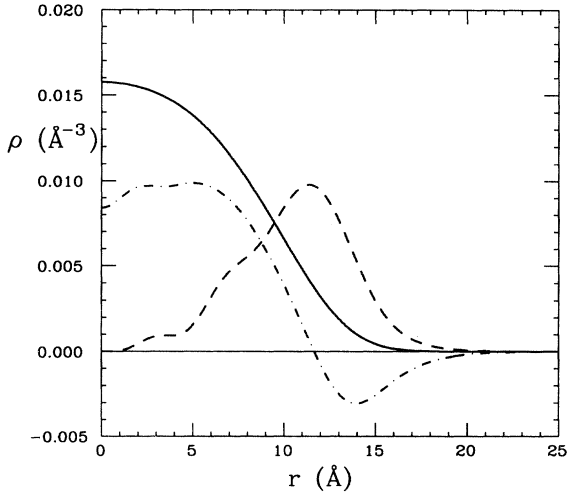

FIG. 3. $N=70$ single-particle density (solid line) and transition densities (in arbitrary units) for the monopole (dash-dotted line) and quadrupole (dashed line) corresponding, respectively, to the discrete state at 1.69 K that carries 30% of the strength and to the state at 1.17 K that carries 60% of the strength.

tion. This can be seen in Fig. 3 where we have plotted for the $N=70$ cluster the radial part $\delta\rho(r)$ of the transition density $\langle n|\hat{\rho}(\mathbf{r})|0\rangle$, where $\hat{\rho}$ is the density operator, corresponding to the discrete states that contribute the most to the $L=0$ and $L=2$ strengths.

Our results for the collective modes are not greatly modified if the interaction is arbitrarily changed to not reproduce the effective mass of the bulk liquid, but consider this to be the bare mass, while still reproducing all the other physical properties as before. The quadrupole energy of the $N=112$ cluster with such interaction increases by 0.2 K with respect to the previous results, thus still remaining well below particle emission threshold. On the other hand, the monopole energy reduces by only 0.1 K in the same cluster. These results are easy to understand if one considers that the effective mass at the cluster surface, which is the relevant region for surface modes, is rather close to the bare mass [14].

In conclusion, we have presented a detailed calculation of the response function of ^{3}He clusters. These clusters exhibit well-defined monopole and quadrupole collective modes. The monopole mode lies in the continuum and it presents an appreciable fragmentation and sizable Landau damping, whereas the quadrupole mode is much concentrated in energy and in big clusters it is almost exhausted by a single state. Similar calculations for multipoles higher than two indicate that the response function is strongly fragmented over a wide energy range, thus indicating that the collective character of the excitation is lost.

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