

Microscopic dynamics of superfluid ^4He : a comprehensive study by inelastic neutron scattering

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The dynamic structure factor of superfluid ^4He has been investigated at very low temperatures by inelastic neutron scattering. The measurements combine different incoming energies resulting in an unprecedentedly large dynamic range with excellent energy resolution, covering wave vectors Q up to 5 \AA^{-1} and energies ω up to 15 meV. A detailed description of the dynamics of superfluid ^4He is obtained from saturated vapor pressure up to solidification. The single-excitation spectrum is substantially modified at high pressures, as the maxon energy exceeds the roton-roton decay threshold. A highly structured multi-excitation spectrum is observed at low energies, where clear thresholds and branches have been identified. Strong phonon emission branches are observed when the phonon or roton group velocities exceed the sound velocity. The spectrum is found to display strong multi-excitations whenever the single-excitations face disintegration following Pitaevskii's type a or b criteria. At intermediate energies, an interesting pattern in the dynamic structure factor is observed in the vicinity of the recoil energy. All these features, which evolve significantly with pressure, are in very good agreement with the Dynamic Many-body calculations, even at the highest densities, where the correlations are strongest.

I. INTRODUCTION

Understanding the dynamics of correlated bosons is a subject of general interest in several fields of physics. Bose-Einstein condensation and superfluidity^{1,2}, first found in ^4He , are fundamental phenomena that imprint remarkable signatures on the dynamics of these systems. Experimentally, superfluid ^4He is the simplest example of strongly correlated bosons. The interaction potential is particularly well known, and substantial effort has been devoted to develop a coherent theoretical framework able to describe and explain the extraordinary properties of this quantum fluid¹⁻¹¹. The theoretical methods can be generalized to other many-body problems, including for instance up-to-date approaches of the complex case of correlated fermions¹²⁻¹⁶.

The prediction by Landau³ of the phonon-roton excitation spectrum of superfluid ^4He and its direct observation in the dynamic structure factor $S(Q, \omega)$ using neutron scattering techniques^{4,17} are cornerstones of modern physics, at the origin of the present microscopic descriptions of matter^{5,18,19}. The dynamics of superfluid ^4He at very low temperatures, in the vicinity of the ground state, is dominated by the “phonon-maxon-roton” excitation branch. The corresponding excitations, extremely sharp, correspond essentially to poles of the dynamic density-density response function. They are referred to as “single-excitations” in the neutron scattering literature, and as “quasi-particles” in theoretical works. An effective description of the dynamics of such systems can be obtained in terms of these modes, allowing for instance a very accurate statistical evaluation of the low

temperature thermodynamic properties^{4,20}.

Sharp excitations are absent above twice the roton energy^{4,17,21}, and the dynamics at intermediate energies is described in terms of broad excitations, named “multi-excitations” for reasons described below. Multi-excitations still have a significant statistical weight in the dynamic structure factor^{4,17,22-30}. Their spectrum is known to display some structure since the early measurements of Svensson, Martel, Sears and Woods²³. More recent investigations²⁴⁻²⁹ showed that some features could be ascribed to multi-excitations. These were related to pairs of high density-of-states roton (R) and maxon (M) modes (noted hereafter 2R, 2M, and MR). The broad ridges observed in $S(Q, \omega)$ at SVP (see Figure 1 of Ref. 24), and at 20 bars (see Figure 1 of Ref. 29) were consistent with the calculated energies of the main combinations (2R, 2M and MR).

A much finer structure in the dynamic response was observed in our recent work at zero pressure³⁰, including sharp thresholds, narrow branches, and a new two-phonon decay process, the “ghost phonon”. Explaining this rich dynamic response, observed from the continuum limit to subatomic distances, constitutes a challenge and an opportunity for microscopic theories.

Finally, at high energies, the dynamic structure factor gradually approaches a quasi-free-particle behavior²² described by the impulse approximation^{4,17,31}.

Even though helium is one of the most intensively investigated physical substances, measurements covering a large kinetic range are scarce. The canonical results by Cowley and Woods²², Dietrich *et al.*³² or Svensson *et al.*²³ have a low resolution by modern standards, while

later measurements specialize in specific ranges^{27–29,33,34}. Our extensive high-resolution measurements, presented in Fig. 1, provide a detailed and complete map of the dynamics of superfluid ⁴He. In addition to its aesthetic merits, the picture shows new features which are the object of this manuscript.

Helium is highly compressible. Since the atomic correlations depend on the density, it is interesting to investigate the pressure dependence of the density excitations. Much of the earlier work has been focused on the effect of pressure on the single-excitation response, in order to determine, for example, the Landau parameters characterizing the dispersion curve. The multi-excitation spectrum has also been found experimentally^{27–29,32–35} and theoretically^{4,7,9,36} to be strongly modified by the pressure. It was therefore desirable to extend our recent high sensitivity measurements³⁰ to finite pressures, and more particularly near solidification, where theory⁹ predicted radical changes in the dynamics.

In this manuscript, we present a detailed investigation of the effect of pressure on the dynamics of superfluid ⁴He. We cover a large energy and wave vector range while preserving the resolution needed to observe the fine structure of the spectra. High resolution maps of the dynamic structure factor $S(Q, \omega)$ have been obtained at saturated vapor pressure (SVP) and at $P = 24$ bars, close to solidification, as shown in Fig. 1. Additional measurements have been made in a smaller dynamic range at the intermediate pressures 5 and 10 bars. We finally compare our data to microscopic calculations of $S(Q, \omega)$ within the Dynamic Many-Body theory⁹ performed at the densities corresponding to the experimental pressure conditions.

II. EXPERIMENTAL DETAILS

The measurements were performed on the IN5 time-of-flight spectrometer at the high-flux reactor of Institut Laue Langevin. Our previous work³⁰ at low temperatures and saturated vapor pressure used cold neutrons of energy $E_i=3.55$ meV. In the present work, we combine data taken using four different incident neutron energies, $E_i=3.55, 5.11, 8.00,$ and 20.45 meV, for which the energy resolution (FWHM) at elastic energy transfer was 0.070, 0.12, 0.23 and 0.92 meV, respectively. This allowed us to obtain a complete map of the dynamic structure factor at the most relevant pressures, *i.e.*, saturated vapor pressure (SVP) and near solidification ($P = 24$ bars). We also investigated a few intermediate pressures using $E_i=3.55$ meV.

The cylindrical sample cell was made out of aluminum 5083, with 1 mm wall thickness and 15 mm inner diameter³⁰. Cadmium disks of 0.5 mm thickness were placed inside the cell every centimeter to reduce multiple scattering. The cell was thermally connected to the mixing chamber of a very low temperature dilution refrigerator using massive OFHC copper pieces. Heat exchangers made out of sintered silver powder were used to

provide a good thermal contact with the helium sample. Care was taken to thermally anchor the filling capillary at several places along the dilution unit, in order to reduce heat leaks to the cell. The measurements were all performed at very low temperatures, well below 100 mK, *i.e.* essentially at zero temperature for the properties under investigation.

High purity (99.999 %) helium gas was condensed in the cell at low temperatures, using a gas handling system including a “dipstick” cold trap operated in a helium storage dewar. The dipstick was used to condense the gas and to pressurize the helium sample. The pressure in the system was measured with a precision of 6 mbars with a 0-60 bars Digiquartz gauge located at the top of the cryostat. The corresponding precision for the pressures inside the cell is 20 mbars, after applying helium hydrostatic head corrections. The actual pressures in the cell for the nominal 0, 5, 10 and 24 bars are essentially 0 (SVP at 100 mK), 5.01(2), 10.01(2) and 24.08(2) bars.

III. DATA REDUCTION

Standard time-of-flight data-reduction³⁷ was used to obtain the dynamic structure factor $S(Q, \omega)$ from the raw data. The contribution of the cell scattering was subtracted, as well as that of double scattering events of type “inelastic helium scattering plus elastic scattering from the cell”. This type of double scattering is essentially independent of wave vector.

The contribution of the multiple scattering within the helium was corrected using Monte Carlo simulations³⁸. Due to the small diameter of our sample cell and the presence of several cadmium plates, multiple scattering corrections are small (the ratio of double-scattered to single-scattered neutrons is on the order one percent³⁰), but may be comparable to the multi-excitation signal. It is therefore essential to verify that multiple scattering is not contaminating the spectra in the energy and wave vector regions of interest, and perform the corrections when necessary, in particular at low Q .

Since multiple scattering depends on the incident neutron energy, as shown in figure 2, while multi-excitations do not, Monte Carlo calculations can be used to select the most appropriate incident neutron energy for the experiments, and also to experimentally distinguish multi-excitations from multiple scattering.

The only input needed by the Monte Carlo simulations³⁸ in the present case is the initially measured scattering function $S(Q, \omega)$ after corrections for multiple scattering processes involving the cell, and the coherent scattering cross section of ⁴He, $\sigma_c = 1.34$ barns. We first calculate the total scattering cross section^{38,39} $\sigma_s(E_i)$:

$$\sigma_s(E_i) = \frac{N\sigma_c}{2k_i^2} \int QdQ \int S(Q, \omega)d\omega, \quad (1)$$

where N is the number of scatterers and k_i the incident

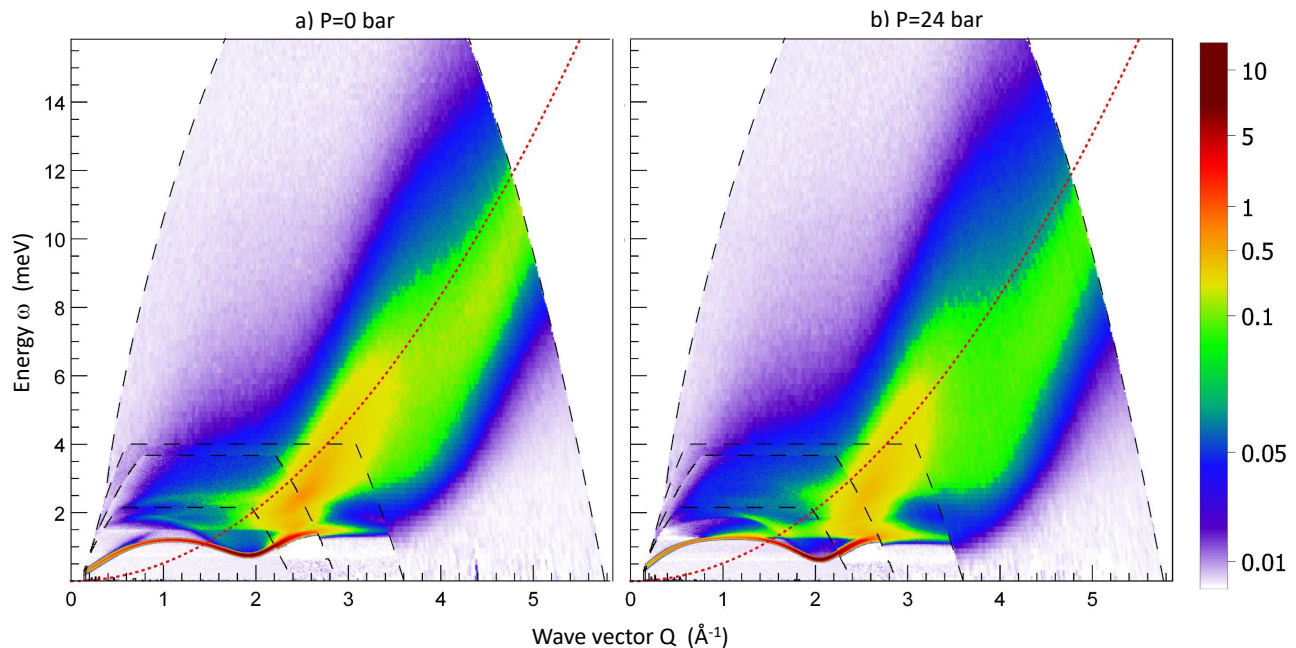


FIG. 1. $S(Q, \omega)$ of superfluid ${}^4\text{He}$ as a function of wave vector and energy transfer, measured at $T \leq 100$ mK at (a) saturated vapor pressure ($P \approx 0$) and (b) near solidification ($P = 24$ bars). The plots combine data measured at different incident neutron energies ($E_i=3.55, 5.1, 8.00$ and 20.45 meV) for an optimum energy resolution; the dashed black lines represent the limits of the corresponding kinetic ranges. The dotted red line is the free ${}^4\text{He}$ atom recoil energy $E_r = \frac{\hbar^2 Q^2}{2M}$. The color-coded intensity scale is in units of meV^{-1} .

neutron wave vector.

We find $\sigma_s(E_i = 3.55 \text{ meV}) = 0.64$ barns, about one half of the coherent scattering cross section σ_{coh} . The multiple scattering fraction is 0.8% for $E_i = 3.55$ meV, increasing slightly with pressure from 0.79% at SVP to 1.06% at 24 bars. This agrees well with calculations using the semi-analytical method developed by Sears⁴⁰, which give

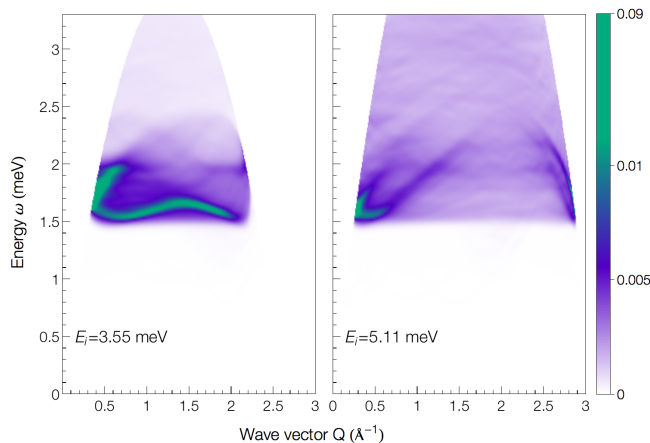


FIG. 2. Monte Carlo calculation of the contribution of double-scattering within the helium to $S(Q, \omega)$. Results are shown for two incident neutron energies, $E_i = 3.55$ and 5.11 meV. The color-coded intensity scale is in units of meV^{-1} .

values increasing from 0.93% to 1.09% for the same pressures. Multiple scattering can be seen in the experimental spectra at low wave vectors, thus providing a way to check the Monte Carlo calculations used to eliminate this effect. This is a crucial step in the data analysis, needed to ensure that all the features we report in $S(Q, \omega)$ do indeed correspond to multi-excitations.

The calculated contribution due to multiple scattering within the helium has been subtracted from the spectra measured using incident neutron energies $E_i = 3.55$ and 5.11 meV. This was found to be unnecessary for $E_i = 8.00$ and 20.45 meV, because multiple scattering processes are negligible in the corresponding regions of the “combined” spectra of Fig. 1.

An overall scale factor was applied to $S(Q, \omega)$ at SVP, so that the weight of the single excitation $Z(Q)$ agrees with that of Cowley and Woods²² near the roton, *i.e.*, $Z(Q = 2 \text{ \AA}^{-1}) = 0.93$ at SVP. At higher pressures, the same scaling factor was used, but corrected for the density ratio $\rho(P)/\rho(P = 0)$.

IV. EXPERIMENTAL RESULTS

A. Spectra at SVP and $P=24$ bars in a large dynamic range

Our comprehensive results on the dynamic structure factor $S(Q, \omega)$ at SVP and $P = 24$ bars are shown in

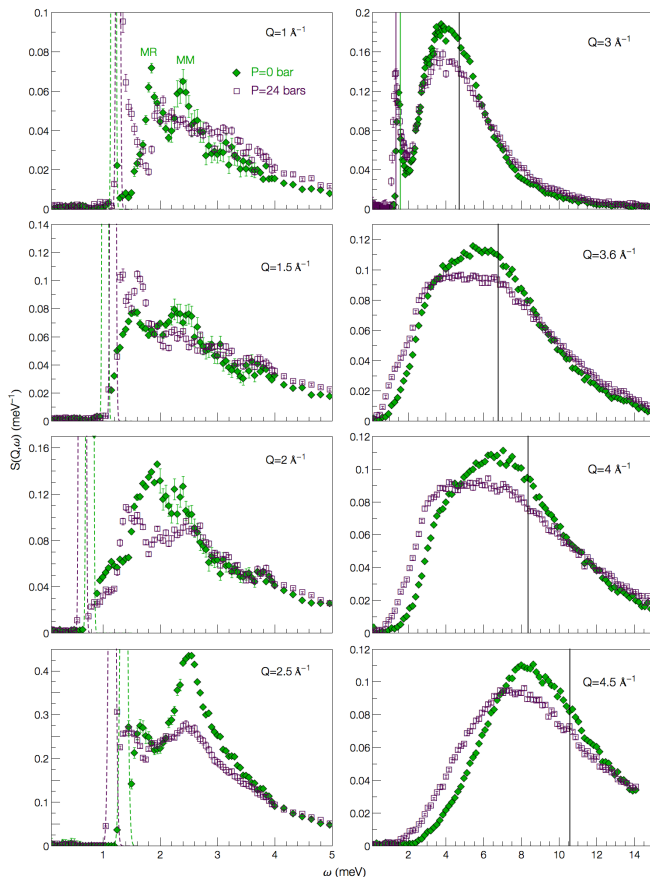


FIG. 3. Dynamic structure factor $S(Q, \omega)$ combining data at four incident neutron energies: spectra for different wave vectors Q at SVP (green diamonds) and $P = 24$ bars (purple squares). The dashed lines are Gaussian fits of the resolution-limited phonon-roton peaks (off scale). The black lines represent the helium recoil energy. At $Q = 3 \text{ \AA}^{-1}$, the purple and green lines represent the two-roton energy $2\Delta_R$ at SVP and $P = 24$ bars, respectively. At $Q = 1 \text{ \AA}^{-1}$, MR and MM are the energy positions at SVP of the maxon-roton and maxon-maxon multi-excitations, respectively.

Fig. 1. These maps were obtained by combining the four different neutron energies. Higher energies make a larger dynamic range accessible, but the instrumental energy resolution deteriorates rapidly (see section II). Since the corresponding dynamic ranges have a substantial overlap, we can select the most appropriate data set in terms of resolution, neutron counts or cleanest background for each region of the $Q - \omega$ plane. The $S(Q, \omega)$ maps are built in the following way: first, the spectrum measured at $E_i = 3.55$ meV is represented; outside its useful kinetic range, the data at $E_i = 5.11$ meV are added, then the data at $E_i = 8.00$ meV and finally, the data at $E_i = 20.45$ meV.

The constant wave vector scans presented in Fig. 3, obtained as particular “cuts” of Fig. 1, provide a complementary perspective on the data. The phonon-roton single-excitation mode is very narrow at the scale of Figs. 1 and 3, and the observed width is essentially a mea-

sure of the experimental energy resolution (with the remarkable exception of the maxon at high pressures, which is discussed in the next section). The influence of a finite energy resolution is clearly seen in Fig. 1 as a width discontinuity in the Pitaevskii plateau^{4,21}, between ranges corresponding to different incident neutron energies. It is important to note, however, that the experimental broadening effects are negligible in all the *multi-excitation* region investigated in the present work (except at the end of the Pitaevskii plateau).

Merging data measured with different resolutions has been successfully achieved, judging from the remarkable continuity in intensity between the different regions represented in Fig. 1. This is essentially due to the fact that the sharpest multi-excitations are found in the low energy and low wave vector sector, adequately covered by our high resolution data at $E_i = 3.55$ and 5.11 meV. Conversely, the spectra in the quasi-free particle region, at high energies and wave-vectors, are intrinsically broad, and adequately covered by our data at 8.00 and 20.45 meV, in spite of their lower resolution. Using optimized incident neutron energies reveals the complete evolution of the system, characterized by several multi-excitation branches merging progressively at high wave vectors to form a broad but rather intense feature. Intensity in this region was observed in early studies^{4,17}, but the data were either strongly truncated^{24,25,29}, or measured with low resolution²². This feature finally becomes, after a strong oscillation, a less intense branch progressively approaching the free particle parabolic dispersion.

B. High resolution spectra as a function of pressure

We present in this section the spectra obtained using an incident neutron energy of $E_i = 3.55$ meV, for wave vectors up to $Q = 2.5 \text{ \AA}^{-1}$ and energies up to $\omega = 2.22$ meV. The results are shown in Fig. 4(a), where we represent our earlier data³⁰ at SVP, the present data at 5 and 10 bars, and the data at $P = 24$ bars discussed in the previous section. One can readily note that both the single-excitation and the multi-excitation components of the dynamic structure factor are modified by the pressure.

Our results for the single-excitations dispersion measured at several pressures, shown in Fig. 5a, are in excellent agreement with previous works^{4,17,24,25,28,32,34,41,42}. The roton parameters at each pressure have been obtained from fits of the single-excitations dispersion relation $\epsilon_Q(Q)$ to the expression

$$\epsilon_Q = \Delta_R + \frac{\hbar^2}{2m\mu_R}(Q - Q_R)^2 + b(Q - Q_R)^3 + c(Q - Q_R)^4, \quad (2)$$

where Δ_R is the roton energy gap, Q_R the wave vector at the roton minimum, and μ_R the roton effective mass; b and c are additional adjustable parameters. Fits were

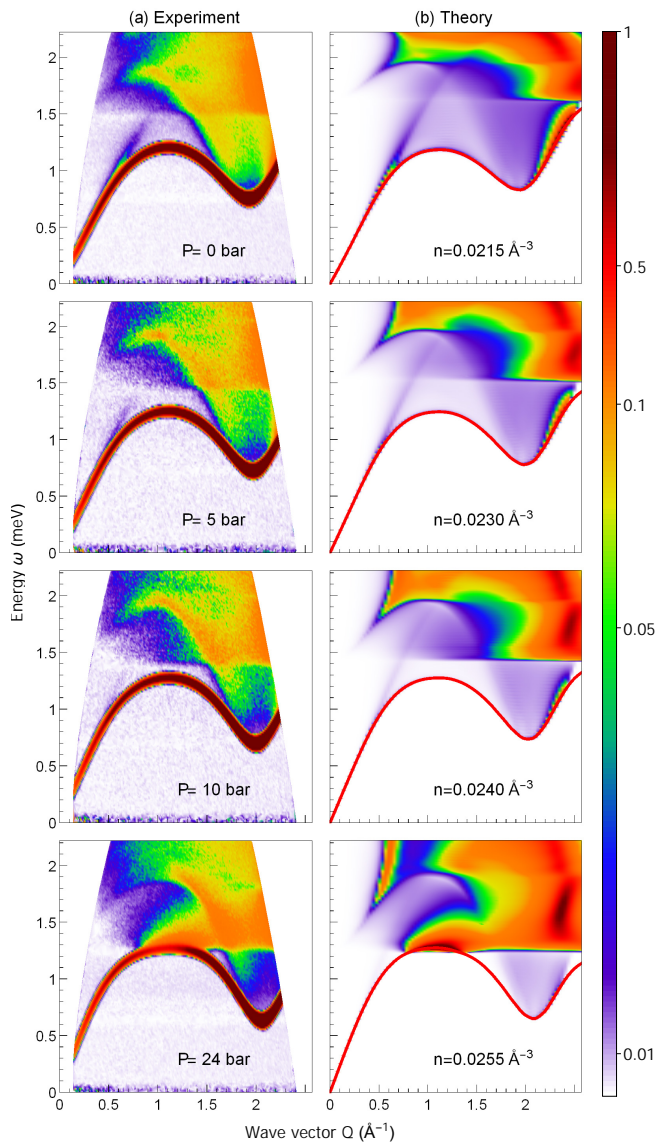


FIG. 4. (a) $S(Q, \omega)$ of superfluid ${}^4\text{He}$ measured as a function of wave vector and energy transfer, at $P = 0, 5, 10$ and 24 bars and temperature $T \leq 100$ mK. The incident neutron energy is $E_i = 3.55$ meV. (b) Dynamic many-body theory calculation of $S(Q, \omega)$ at corresponding densities ($n = 0.0215, 0.0230, 0.0240$ and 0.0255 \AA^{-3} , see text). Note that the main features of the experimental data are well reproduced. The color-coded intensity scale is in units of meV^{-1} . The intensity is cut off at 1 meV^{-1} in order to emphasize the multi-excitations region. The apparent width of the phonon-rotor excitations in the experimental plot is due to an energy resolution of 0.07 meV , while the calculated phonon-rotor dispersion curve has been highlighted by a thick red line.

made over a total wave vector range ΔQ up to 0.47 \AA^{-1} . Due to the large number of individual detectors and the high neutron rate of IN5, the statistical uncertainty of the fits is very good (see Table I). The roton mass determined in our work is lower than the one obtained by Andersen et al.^{24,25} using a parabolic fit of the roton minimum,

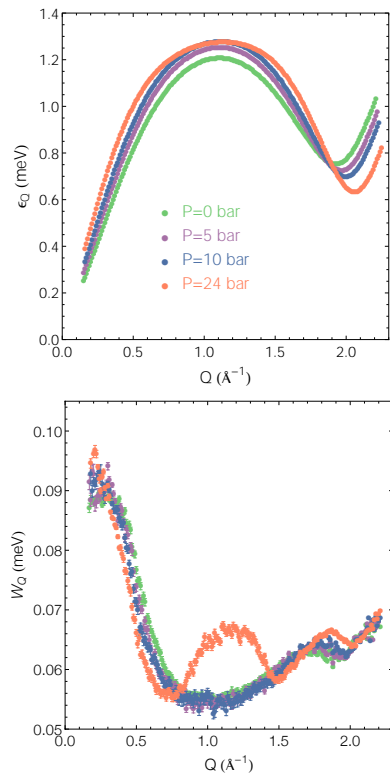


FIG. 5. a) Dispersion relation $\epsilon_Q(Q)$ of the single-excitations measured at 0, 5, 10 and 24 bars. Note the flattening of the curve at the maxon at high pressures. b) The wave vector dependence of the measured width (FWHM) of the single-excitation peaks. The measured width reflects the shape of the experimental resolution ellipsoid cut by the dispersion relation curve at different angles. At 24 bars, however, a physical broadening of the maxon is clearly observed.

but it agrees well with earlier measurements⁴¹ where the parabolic fit was limited to a very narrow wave vector range.

P (bars)	Δ_R (meV)	Q_R (\AA^{-1})	μ_R
0	0.7416(10)	1.9260(2)	0.1240(4)
5.01(2)	0.7143(10)	1.9655(2)	0.1096(4)
10.01(2)	0.6885(10)	1.9963(2)	0.1000(4)
24.08(2)	0.6254(10)	2.0579(2)	0.0879(4)

TABLE I. Roton energy gap Δ_R , wave vector of the roton minimum Q_R and roton effective mass μ_R ; values in parenthesis are one standard deviation errors from least-squares fits described in the text.

A similar analysis can be performed in the maxon region. The corresponding maxon parameters Δ_M , Q_M and μ_M (d and e are additional adjustable parameters) have been calculated by fits of ϵ_Q in the maxon region, over a wave vector range ΔQ on the order of 0.8 \AA^{-1} , to

the formula:

$$\epsilon_Q = \Delta_M - \frac{\hbar^2}{2m\mu_M} (Q - Q_M)^2 + d(Q - Q_M)^3 + e(Q - Q_M)^4. \quad (3)$$

The results are given in Table II.

P (bars)	Δ_M (meV)	Q_M (\AA^{-1})	μ_M	$2\Delta_R$
0	1.1966(10)	1.1073(2)	0.492(1)	1.4832(20)
5.01(2)	1.2422(10)	1.1089(3)	0.541(1)	1.4286(20)
10.01(2)	1.2668(10)	1.1150(3)	0.614(2)	1.3777(20)
24.08(2)	1.2662(10)	1.1336(4)	0.915(3)	1.2508(20)

TABLE II. Maxon energy Δ_M , wave vector Q_M and effective mass μ_M ; values in parenthesis are one standard deviation errors from least-squares fits described in the text. The last column gives twice the energy of the roton gap $2\Delta_R$, for comparison with the value of Δ_M .

As expected for a system approaching localization⁴³, the phonon and the maxon energies increase steadily with pressure, while the energy of the roton minimum decreases. The single-excitation data of Fig. 5a clearly show in addition a substantial flattening at the level of the maxon in the dispersion curve corresponding to a pressure of 24 bars. Earlier results at this pressure did not detect this effect^{32,34}, while more recent systematic results by Gibbs *et al.*²⁸ were limited to pressures below 20 bars. The data at 24 bars are qualitatively different from those at low pressures because the maxon energy exceeds twice the roton energy. At high pressures, the maxon excitation can therefore decay by phonon emission, exactly as in the case of higher wave vectors, at the Pitaevskii’s plateau²¹.

We also observe the corresponding broadening of the maxon single-excitation (Fig. 5b): the measured maxon total width of 0.012 meV, obtained after subtraction of the instrumental resolution, is substantial compared to typical phonon and roton widths (see Ref. 42 and references therein). The excitations in the maxon region broaden until they become unobservable in confined helium^{17,44,45}, where very high pressures can be reached before solidification.

We now concentrate on the multi-excitation region, shown in Fig. 4(a), which displays highly structured spectra for all pressures. The data for the pressures 5 and 10 bars are qualitatively similar to our previous results at saturated vapor pressure³⁰. The high resolution spectra display very clearly a threshold in energy at about 1.5 meV. This feature, which corresponds to the decay of an excitation into a pair of rotons, depends on pressure, since the roton energy depends on pressure. In addition, we observe several well-defined multi-excitation branches displaying substantial dispersion. Their gradual evolution reflects, as will be shown in section VI, the change with pressure of the single-excitation dispersion.

We also observe important *qualitative* changes at high pressures. We examine first the multi-excitation region

of the “ghost-phonon”. This multi-phonon excitation, observed in our previous work, appears as a linear extension of the phonon branch³⁰. We observe in the present work that the ghost-phonon intensity strongly decreases with pressure until it disappears at some pressure below 24 bars.

We also see very clearly in Fig. 1(a) a multi-phonon region just above the roton branch for wave vectors of the order of 2.2 to 2.4 \AA^{-1} . The high resolution spectra at $E_i=3.55$ meV only show part of this multi-excitation region, but the results have been completed by spectra taken at $E_i=5.11$ meV at SVP and 24 bars, shown in Fig. 1. The intensity of these multi-excitations, described in detail in Section VIC, decreases strongly with pressure, behaving similarly as the ghost-phonon.

The multi-excitation spectra are strongly modified at high pressures, as the maxon enters the multi-excitations continuum. Fig. 4 shows that substantial intensity develops at this pressure for energies just above the maxon. Similar effects were also observed by Graf *et al.*³³, Talbot *et al.*⁴⁶, and by Gibbs *et al.*^{27–29} at a lower pressure (20 bars). The present data benefit from a sharper resolution, as can be seen by directly comparing spectra at $Q \approx 1 \text{\AA}^{-1}$ around the maxon peak.

All these effects will be discussed in detail in section VI in the context of a comparison with theoretical calculations.

V. CALCULATIONS WITH THE DYNAMIC MANY-BODY THEORY

We present in this section our calculations of the dynamic structure factor of superfluid ⁴He at zero temperature obtained within the Dynamic Many-Body theory^{6,9}.

A. State of the art of Theory

Theoretical studies of the dynamic structure function in ⁴He began with the work of Feynman⁴⁷, and Feynman and Cohen⁴⁸. The Feynman theory of elementary excitations was developed in a systematic Brillouin-Wigner perturbation theory by Jackson and Feenberg^{49–51}. An important contribution was the identification of classes of theories for the dynamic structure function⁵² that satisfy the ω^0 and ω^1 sum rules exactly.

The most complete evaluation of the phonon-roton dispersion relation in terms of Brillouin-Wigner perturbation theory was carried out by Lee and Lee⁵³ who obtained an impressive agreement with the experimental phonon-roton spectrum up to the wave vector of 2.5 \AA^{-1} . The major drawback with these early calculations was that the required input, pair and three-body distribution functions, were poorly known.

Manousakis and Pandharipande^{36,54} used input states of the Brillouin-Wigner perturbation theory including “backflow” correlations in the spirit of Feynman and Cohen. Through the gradient operator acting on the wave

function, specific dynamic correlations are introduced to all orders. The “backflow-function” is, however, chosen per physical intuition rather than by fundamental principles, and the evaluation of the perturbative series becomes very complicated. Topologically, diagrams similar to those of Lee and Lee⁵³ were included. While the accuracy of the theoretical roton energy is comparable to that of Lee and Lee, one can clearly see an inconsistency since the energy of the Pitaevskii plateau²¹ lies below twice the energy of the roton gap.

The first theoretical descriptions of the multi-excitations^{36,54,55} were qualitatively in agreement with the early multi-excitations data^{22,32,33}. The simplest version of Correlated Basis Functions theory produces phonon, maxon and roton modes, as well as multi-phonons. In this approximation, the calculated multi-excitations decay into Feynman modes instead of the correct single-excitations; large gaps are found in the spectrum, and many predicted features are not seen in the experiments. Other features calculated in the multi-excitation region do indeed survive in recent theories, like the presence of intensity above the phonon branch and that of a well-defined 2-roton threshold (these effects are described below). These calculations, as well as many others addressing specific aspects of the multi-excitation dynamics, could not be quantitatively compared to the experimental results, but they motivated further investigations on multi-particle dynamics. Reviews can be found in Ref. 4 and 5.

More recent calculations⁹ used a hybrid approach of Brillouin-Wigner perturbation theory and equations of motion for time-dependent multi-particle correlation functions to derive a self-consistent theory of the dynamic density-density response of ⁴He. The self-consistency of this semi-analytic method allows the identification of mode-mode coupling processes that lead to observable features in the dynamic structure function. The underlying physical mechanisms, their relationship to the ground state structure, and the consequences on the analytic properties of the dynamic structure function, emerge directly from the theory.

A very different approach involves novel numerical methods^{7,8,10,15,56} that give access to dynamic properties of quantum fluids. These important algorithmic developments will reproduce, extend and complete the experimental data with the future development of computing power; their present accuracy and consistency, however, are still limited in the multi-excitations region investigated here.

B. Dynamic Many-Body Theory calculation

In order to calculate quantitatively both the single-excitation and the multi-excitation response, our calculations include up to three-body dynamic fluctuations in the correlation functions of the equations of motion⁹. We derive the self-consistent density-density response of ⁴He

$\chi(Q, \omega)$, expressed as

$$\chi(Q, \omega) = \frac{S(Q)}{\omega - \Sigma(Q, \omega)} + \frac{S(Q)}{-\omega - \Sigma(Q, -\omega)} \quad (4)$$

where $S(Q)$ is the static structure factor, and the self-energy $\Sigma(Q, \omega)$ is determined by the integral equation

$$\Sigma(Q, \omega) = \epsilon_0(Q) + \frac{1}{2} \int \frac{d^3p d^3k}{(2\pi)^3 n} \delta(\vec{Q} - \vec{p} - \vec{k}) \times \frac{|V^{(3)}(\vec{Q}; \vec{p}, \vec{k})|^2}{\omega - \Sigma(p, \omega - \epsilon_0(k)) - \Sigma(k, \omega - \epsilon_0(p))}. \quad (5)$$

In this expression, $\epsilon_0(Q)$ is the Feynman dispersion relation, and $V^{(3)}$ the three-body coupling matrix element. The simplest approximation for $V^{(3)}$, the so-called convolution approximation⁵¹, including static ground state triplet correlations⁵⁷, improves the density-dependence of the roton minimum visibly. The most advanced calculation⁶, which is taken here and in Ref. 9, sums an infinite series of diagrams, the so-called “fan-diagrams” which is the minimum set of diagrams that must be included to reproduce exact features of $V^{(3)}$ for both, long wavelength and short distances.

Linear response theory^{4,9} provides the relation between the experimental dynamic structure factor and the dynamic susceptibility calculated by the theory described above: the dynamic structure factor $S(Q, \omega)$ is proportional to the imaginary part of the dynamic susceptibility $\chi(Q, \omega)$, the linear response of the system to a density fluctuation.

Full maps of $S(Q, \omega)$ have been calculated for different atomic densities, see Fig. 10 in Ref. 9. The data shown in Figs. 1 and 4 correspond to $n = 0.0215, 0.0230, 0.0240$ and 0.0255 \AA^{-3} , values which provide the best overall agreement with the experiment. They turn out to be very close to the experimental results for $P = 0, 5, 10$ and 24 bars, $n_{\text{exp}} = 0.0218, 0.0230, 0.0239$ and 0.0258 \AA^{-3} . The small shift in density is within the expected accuracy of the theoretical calculations.

The calculations presented here have been performed using only the most relevant diagrams⁹. This approximation is sufficient to provide an excellent description of the dynamics, but minor discrepancies can still be seen. The most salient effect is that the roton energy is overestimated; at zero pressure, for instance, the calculated value is 0.83 meV while the measured value is $0.7416(10) \text{ meV}$. This discrepancy could be resolved by including additional diagrams, but it does not seem necessary to perform such a tedious calculation given the quality of the agreement already achieved at this stage.

The calculation provides absolute values for the structure factor. In our previous work³⁰, the calculated values were multiplied by an overall normalization factor of 1.28 in order to have $Z(Q = 2 \text{ \AA}^{-1}) = 0.93$ near the roton. Here, this normalization has not been applied. Given the finite number of diagrams involved in the calculations, a

factor of this order is within their estimated absolute accuracy.

C. Mode-mode coupling

Multi-excitations arise from the enhanced response of the system at particular energies and wave vectors corresponding to two or more single-excitations into which they can decay. The theory considers (see equation 5) the most relevant processes where a density fluctuation (\vec{Q}, ω) of wave vector \vec{Q} and energy ω decays into a pair of single-excitations with corresponding values (\vec{p}, ω_p) and (\vec{k}, ω_k) . The calculations have been shown to be in excellent agreement with experiment at saturated vapor pressure³⁰. Here we investigate the general pressure dependence of the dynamics, and several particularly intense mode-mode couplings. The latter were examined theoretically in Ref. 9, and additional calculations specialized to the main mode-mode couplings (phonon-phonon, phonon-roton, roton-roton, maxon-roton) can be found in Ref. 58. The next section provides a detailed comparison between the theory and the experimental data.

VI. IDENTIFICATION OF THE MULTI-EXCITATIONS

Above the sharp and intense phonon-maxon-roton dispersion curve, we observe a highly-structured multi-excitation region. Multi-excitations are relatively strong if they can decay into a pair of high intensity single-excitation modes. The energy and momentum of these pair combinations is directly related, by the conservation of energy and momentum, to those of the underlying elementary excitations. It is possible to determine the position of the main multi-excitation resonances in the dynamic structure factor map (2-Phonons, 2-Rotons, 2-Maxons and Maxon-Roton) from pure kinematic considerations, *i.e.* energy and momentum conservation. The challenge for microscopic theories is to predict the *intensity* of the multi-excitation spectrum, if possible in a large dynamic range. Obtaining the fine structure we observe requires a quantitative calculation of mode couplings.

We first present in this Section a brief description of the kinematic constraints for different pair-excitations, setting the framework for their identification. The following two subsections concentrate on new features observed in the multi-excitation spectrum, that we named “ghost-phonon” and “ghost-roton”. We then describe a different type of multi-excitations, associated to roton-roton coupling, which we observed in particular “above the maxon” and “beyond the roton”. We conclude this Section by a discussion on higher order multi-excitations, and the progressive evolution to the high energy regime.

A. Kinematic constraints for pair-excitations

The kinematic constraints calculated for the main low energy multi-excitations are shown in Fig. 6. We use below the notation R^- and R^+ to distinguish rotors on each side of the roton minimum.

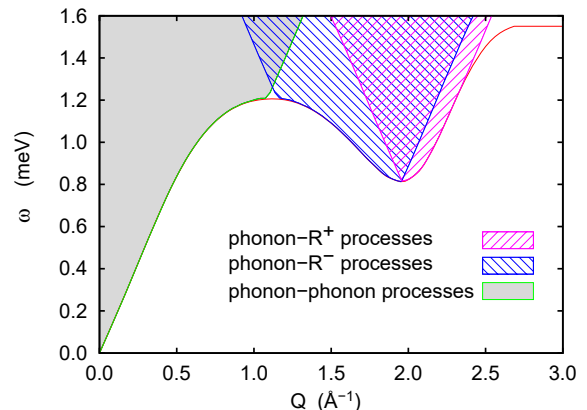


FIG. 6. Kinematically allowed regions for different multi-phonon processes: P-P (including P-M⁻), P-R⁻, and P-R⁺. See text for details.

The allowed regions are necessarily located above the single-excitations dispersion curve. The P-P region is found at low wave vectors. Beyond the maxon, P-R⁻ excitations are allowed in a large region delimited by the dispersion curve and two lines starting at the maxon maximum and at the roton minimum, with slopes equal to $-c$ and $+c$, respectively, where c is the speed of sound. P-R⁺ excitations occupy a region delimited by the dispersion curve and a line starting from the roton minimum with slope $-c$. There is a large overlap with the P-R⁻ region.

The case of 2R, not shown, is particularly simple, with a threshold at twice the roton energy, $2\Delta_R$. The situation for 2M processes is similar, with an upper limit equal to $2\Delta_M$. M-R combinations of excitations may lead to branches with substantial dispersion. The kinematic constraints are sufficient to determine unambiguously which are the dominant processes in some multi-excitations regions, in particular at low Q above the phonon dispersion, and inside the roton parabolic dispersion curve.

The evolution of the observed multi-excitations in a large energy range, for different pressures, is illustrated in Figs. 1 and 4. We can distinguish different types of multi-excitations. Several narrow branches are easily identified, as indicated in Fig. 7, as corresponding to 2P, 2R, 2M and M-R processes. The 2R feature is observed in Fig. 4 as a clear threshold, both in the theoretical and experimental data.

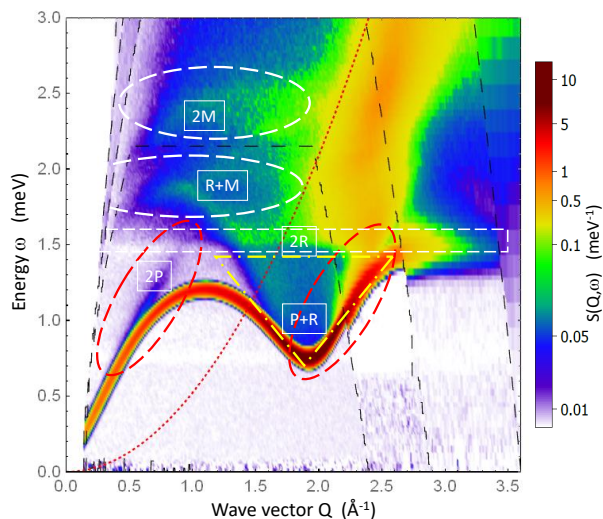


FIG. 7. Map of $S(Q, \omega)$ at SVP identifying remarkable mode-mode coupling regions: phonon-phonon (2P, with an ellipse around the “ghost-phonon”), phonon-roton (P+R, a region marked by a triangle, which includes an ellipse indicating more specifically a high-intensity “ghost-roton” region), roton-roton (2R, marked by a rectangle around 1.5 meV), and at higher energies the roton-maxon (R+M) and maxon-maxon (2M) regions. The description of the different lines is given in Fig. 1.

B. Phonon-phonon coupling: the ghost-phonon

The ghost-phonon^{9,30} (see Section IV B and Fig. 7) corresponds to a process where a high energy multi-excitation decays into a pair of phonons of lower energy. In the case of phonon *single-excitations*, anomalous dispersion opens the phase space needed for such processes. The anomalous character of the phonon dispersion strongly decreases with increasing pressure, and normal dispersion is recovered at high pressures^{4,17,59,60}. The ghost-phonon intensity follows this trend: the pressure dependence is strong, and the ghost-phonon is clearly suppressed at $P = 24$ bars, as shown in the experimental and theoretical results in Fig. 4, and in more detail in Fig. 8.

Cuts of $S(Q, \omega)$ at several wave vectors at the ghost-phonon level are presented for $P=0, 5$ and 10 bars in Fig. 9. The ghost-phonon peaks for the different wave vectors are clearly located on the extension of the linear part of the phonon branch. According to the calculations [see Eq. (6.4) of Ref. 9], the ghost-phonon remains visible until twice the wave vector up to which the dispersion relation is to a good approximation linear. Indeed, Fig. 9 shows that the energy, strength and shape of the calculated ghost-phonon are in excellent quantitative agreement with the experiment at all pressures.

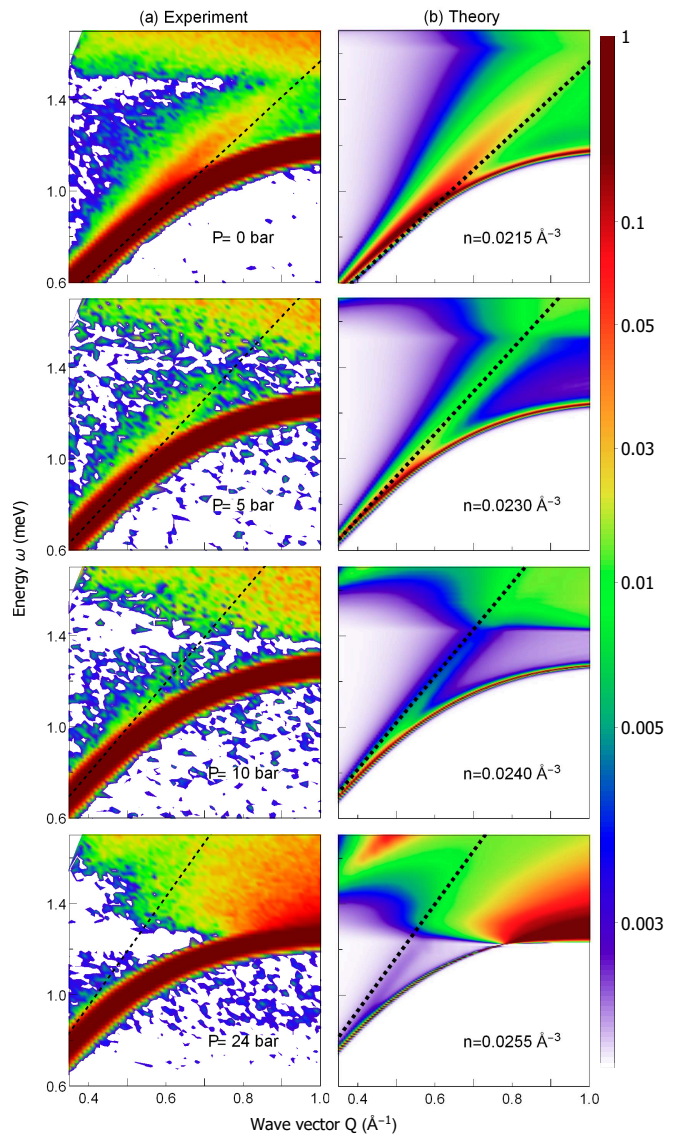


FIG. 8. Left: measured dynamic structure factor $S(Q, \omega)$ in the ghost-phonon region at $P = 0, 5, 10$ and 24 bars. The dashed straight lines correspond to the sound dispersion curve at each pressure, taken from direct measurements of the sound velocity⁶¹. Right: calculated dynamic structure factor at corresponding densities, $n = 0.0215, 0.0230, 0.0240$ and 0.0255 \AA^{-3} , respectively (see text). The dashed straight lines correspond to the calculated sound velocities. The color-coded intensity scale is in units of meV^{-1} .

C. Phonon-roton coupling and the emergence of the ghost-roton

One notes in Fig. 4, for all pressures, the presence of substantial intensity in the region within the roton parabola. Near the roton minimum, where P-R processes are expected to dominate, we observe that the intensity is not symmetric with respect to the roton minimum wave vector Q_R : a faint branch, clearly related to the kinematic limitation for P-R⁺ processes (see Fig. 6) is seen

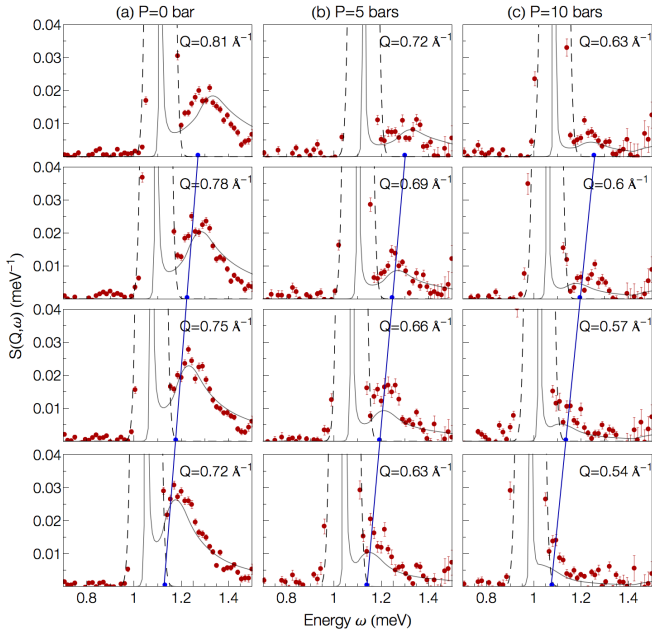


FIG. 9. Dynamic structure factor $S(Q, \omega)$ in the ghost-phonon region: spectra for different wave vectors Q at (a) $P = 0$ bar, (b) $P = 5$ bars and (c) $P = 10$ bars. Filled circles: Experimental $S(Q, \omega)$. Theoretical dynamic structure factor spectra shown as solid lines at densities $n = 0.0215$, 0.0230 and 0.0240 \AA^{-3} . Dashed lines: Intensity of the phonon-roton mode (cut off) calculated directly from the self energy⁹ and convolved with the instrumental resolution of 0.07 meV . The blue lines represent the linear phonon dispersion $\epsilon_Q(P)/\hbar Q = C_0(P)$, where $C_0(P)$ is the sound velocity at a given pressure⁶¹.

for $Q < Q_R$, while a strong branch is formed just above the dispersion curve for $Q > Q_R$. These new features, and in particular the one for $Q > Q_R$, provide a significant contribution to the multi-excitations weight at low pressures (Fig. 10). They appear as an extension of the roton parabolic dispersion towards higher energies, and by analogy with the ghost-phonon, we call these multi-excitations “ghost-rotors”.

It is remarkable that the intensity in this region of the P-R multi-excitations, as was the case for the ghost-phonon, is high at $P = 0$, but is suppressed in the 24 bars data, as shown in Figs. 10, 11 and 12. The origin of these effects is discussed below.

Spectra for several wave vectors in the region of the ghost-roton are shown in Fig. 12 at $P = 0$ and 24 bars (experiment), and in Fig. 13 for the corresponding densities $n = 0.0215$ and 0.0255 \AA^{-3} (theory). We observe a good agreement between theory and experiment, even at the highest densities, near solidification. Studies of mode-mode couplings^{58,62} can therefore be most conveniently performed in the ghost phonon and the ghost-roton regions, rather than looking for a very small broadening of single excitations.

Pitaevskii²¹ described the decay of *single-excitations* when their group velocity reaches the velocity of sound.

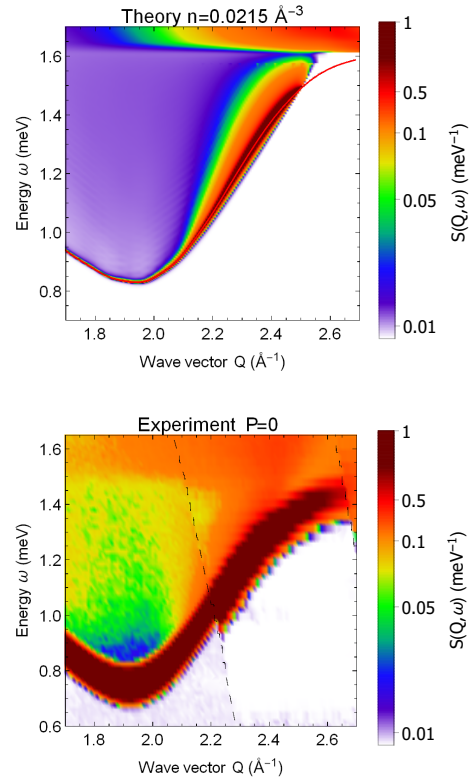


FIG. 10. Theoretical and experimental results for $S(Q, \omega)$ at saturated vapor pressure displaying enhanced multi-excitations (“ghost-rotors”) above the R^+ roton branch, in the supersonic rotors region. The dashed lines represent the limits of different neutron kinetic ranges, see Fig. 1. The small oscillations observed along some contours should be disregarded, they result from numerical discretization.

He named this mechanism of single-excitation broadening “type a”. The process considered here, however, is the emission of phonons by *multi-excitations* in the vicinity of nearly supersonic single-excitations. The generation of multi-excitations by neutron scattering in the R^+ rotors region by this mechanism was qualitatively predicted by Burkova⁶³. Here we show that the ghost-roton corresponds to this effect, that the ghost-phonon is a similar effect, involving supersonic phonons, and that both are correctly predicted by the Dynamic Many-Body Theory⁹.

It has been observed by Dietrich *et al.*³² and confirmed by several groups (see⁴² and references therein) that the R^+ rotors group velocity reaches the sound velocity for $Q \approx 2.2 \text{ \AA}^{-1}$ at low pressures, but remains below the speed of sound near the melting pressure. We show in Figs. 14 and 15 our measured and calculated curves for the group velocity of the single-excitations, for different pressures. Two regions of interest are clearly seen: the first one, at low wave vectors, corresponds to the anomalous dispersion region and gives rise to the ghost-phonon, while the second occurs for wave vectors somewhat above that of the roton minimum (and slightly below the roton

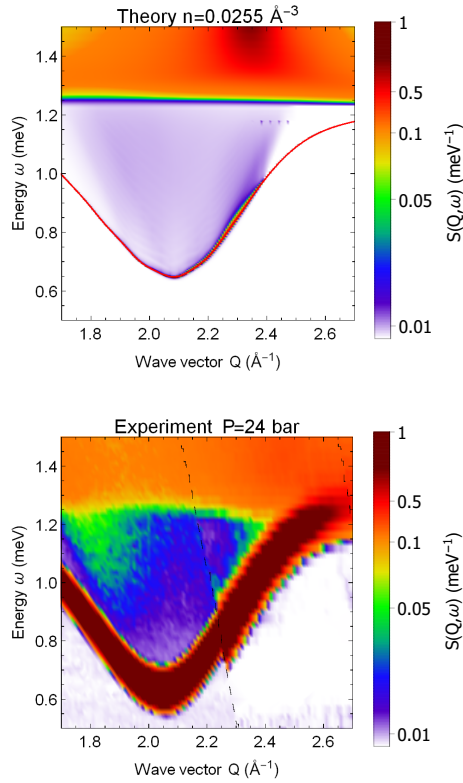


FIG. 11. Theoretical and experimental results for $S(Q, \omega)$ at $P = 24$ bars. A comparison with Fig. 10 shows that at high pressures, ghost-roton multi-excitations are strongly suppressed. They are masked by the finite energy resolution in the experimental graph, but still visible in the calculation.

minimum, but with a much smaller intensity), producing the ghost-roton.

According to the analytic calculations by Burkova⁶³, the neutron-scattering spectrum which corresponds to the production of one roton should have a linear wing on the high-energy side, with a slope which depends on the wave vector. This is not really observed, neither in the experimental data, nor in the Dynamic Many-Body calculation: the linear part, if any, is probably not visible at the scale of the graphs (see Figs. 10, 11, 12 and 13), or is buried inside a broadened single-excitations branch.

Several effects are thus observed when the roton single-excitations approach the speed of sound: a broadening of the roton branch, a downward bending of the dispersion curve, and the appearance of a multi-phonon region just above the distorted dispersion curve. These effects are large at low pressures; the rapid increase of the sound velocity with pressure is responsible for the suppression of the ghost-roton multi-excitations at 24 bars.

D. Roton-roton coupling

We discuss now a different type of *multi-excitations*, related to Pitaevskii's "type b" *single-excitations* decay

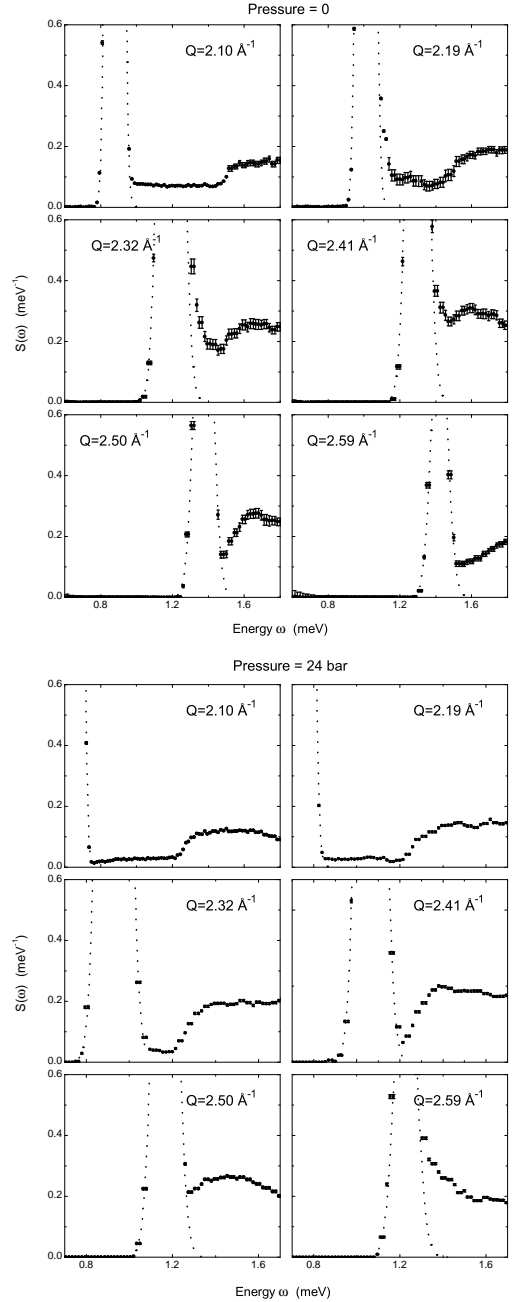


FIG. 12. $S(Q, \omega)$ measured in the region of the ghost-roton at $P = 0$ (upper graph). At $P = 24$ bars (lower graph) one observes the suppression of the ghost-roton. Dashed lines are Gaussian fits of the single-excitation peaks. A comparison with Figs. 10 and 11 clarifies the origin of the observed roton-peak asymmetry for some wave vectors.

processes where the disintegration of a single-excitation occurs as its energy exceeds twice the roton gap^{21,63}.

At high pressures, the maxon energy exceeds twice the roton gap, and a maxon can decay into two rotors. We described in Section IV B the broadening of the maxon as it enters the continuum. At 24 bars, the maxon is in the continuum of the multi-excitations for wave vectors

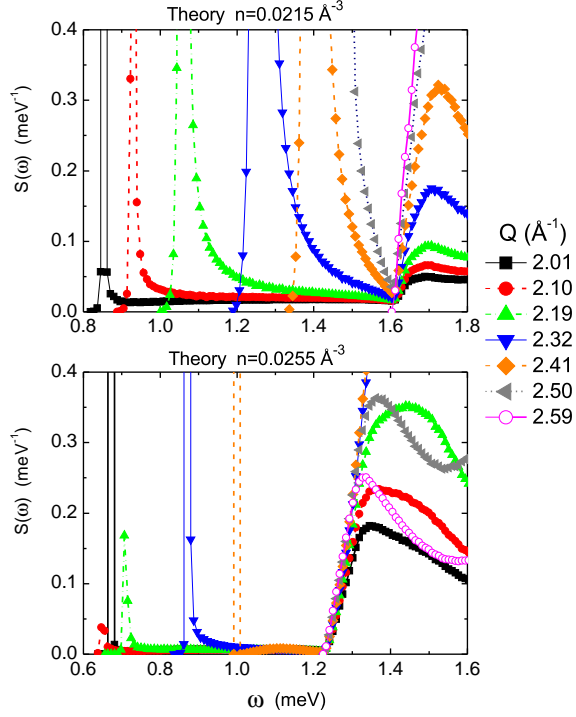


FIG. 13. $S(Q, \omega)$ calculated for wave vectors in the region of the ghost-roton, at densities $n = 0.0215$ and 0.0255 \AA^{-3} , associated to $P = 0$ and $P = 24$ bars respectively.

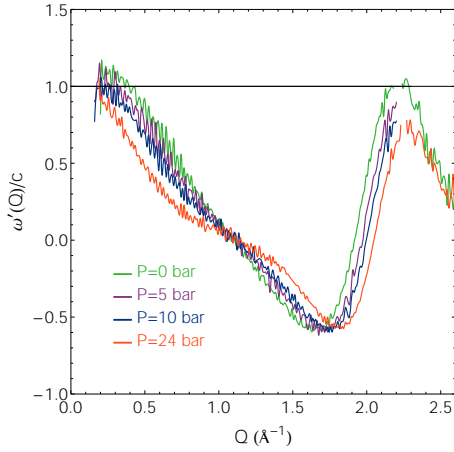


FIG. 14. The experimentally determined group velocity of single-excitations normalized by the sound velocity⁶¹, as a function of wave vector for several pressures.

between $Q = 0.8$ and $Q = 1.5 \text{ \AA}^{-1}$. Under these conditions, a strong multi-excitation intensity is observed above the maxon (Figs. 16 and 17). The very characteristic “rainbow-like” measured spectrum is in very good agreement with the theoretical calculation, showing in particular that the weight of the maxon is transferred to the two-roton excitations.

The multi-excitations discussed above, observed above the maxon at high pressure, are a special case of roton-

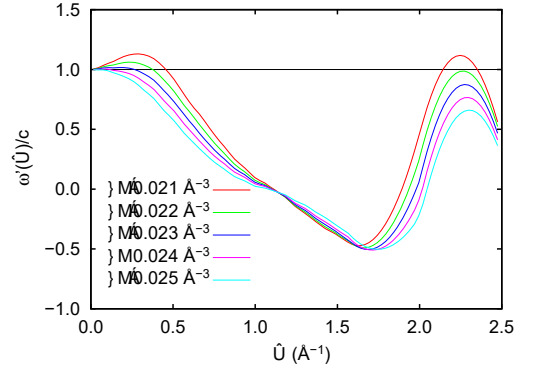


FIG. 15. The theoretically calculated group velocity of single-excitations normalized by the calculated sound velocity, as a function of the wave vector, for several densities. The experimental values of the densities for $P = 0, 5, 10$ and 24 bars are $n_{exp} = 0.0218, 0.0230, 0.0239$ and 0.0258 \AA^{-3} .

roton decay. In fact, a sharp roton-roton threshold is observed at all wave vectors (Figs. 1, 4 and 17), in regions located far from single-excitations. The roton-roton threshold is, in particular, observed at low Q in the present work. It is also clear, in fact, that the intensity of the RR threshold is enhanced in the vicinity of single-excitations, as was the case above the maxon at 24 bars, but also in the region above the Pitaevskii plateau. Theory and experiment display a similar shape of the spectra and intensity pattern around the roton-roton threshold, at all pressures (see Figs. 1 and 4).

E. Higher order multi-excitations

The sharp “branches” described above correspond to decay mechanisms into 2-excitations. Phase-space arguments show that the signal of higher order processes will be distributed in a rather featureless way in the energy-wave vector space, due to the vector addition of momenta. However, the data of Fig. 1 show that the multi-excitations region at wave vectors on the order of 1.5 \AA^{-1} extends to rather high energies, on the order of 4 meV . This last value constitutes a clear experimental demonstration that multi-excitations of higher order, related to 3 and 4 single-excitations (the energy of rotons and maxons is on the order of 1 meV), play a significant role in the dynamics of superfluid ^4He .

One can also examine the corresponding effect on the wave vector axis, beyond the end-point of the Pitaevskii plateau. The plateau could be expected to end at $2Q_R$, for a multi-excitation of energy $2\Delta_R$ decaying into two rotons of colinear wave vectors. Previous measurements^{35,64,65} have found that the plateau intensity vanishes at $Q = 3.6 \text{ \AA}^{-1}$, considerably below $2Q_R = 3.84 \text{ \AA}^{-1}$. This is also observed in the present work, as seen in Figs. 1 and 3. This effect has been attributed to the decay into two rotons with an attractive

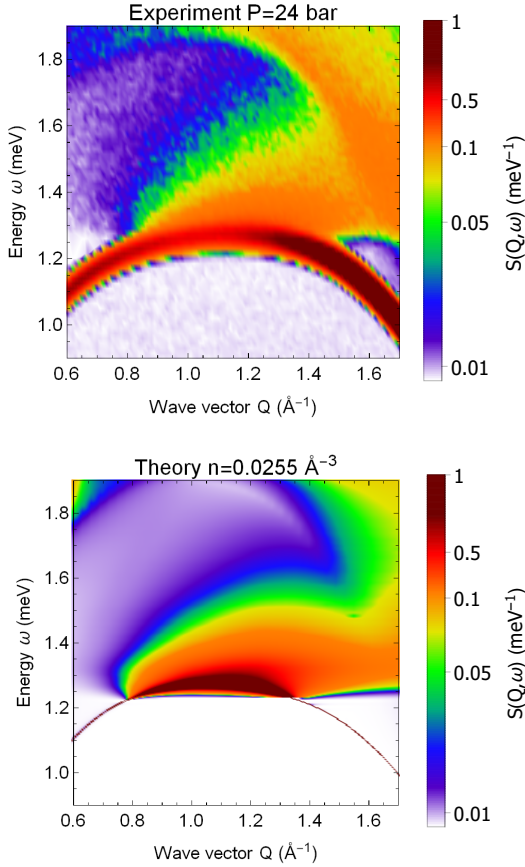


FIG. 16. $S(Q, \omega)$ in the region of the maxon at $P = 24$ bars (experiment) and at the corresponding density of 0.0255 \AA^{-3} (theory).

R-R interaction⁶⁶, but other possible interpretations of the data are presently debated. We also note that the intensity does not extend to higher Q -values at higher energies as expected for decays into 3- and 4-excitations processes, an effect which is probably related to the small phase-space available for colinear combinations of wave vectors. As discussed above, the energy, a scalar, is a better probe for detecting high order multi-excitation processes. The data at 24 bars display similar effects with a simple shift towards higher wave vectors, due to the larger value of $Q_R = 2.06 \text{ \AA}^{-1}$ at this pressure.

We now concentrate on the multi-excitations region located slightly below the free-particle dispersion curve, around 2.5 \AA^{-1} (see Fig. 1). Earlier studies^{22,35,67} observed a rather intense broad feature extending to higher energies. We find here a much finer structure than previously believed, and also that it depends rather strongly on the pressure. Multi-excitations in this region can only decay into 3 or more single-excitations, which is therefore of interest for mode-mode coupling theories. The fact that we observe a high intensity peak is probably related, at these relatively high energies, to an enhanced system response in the vicinity of the free-particle dispersion curve, which is the asymptotic behavior at higher

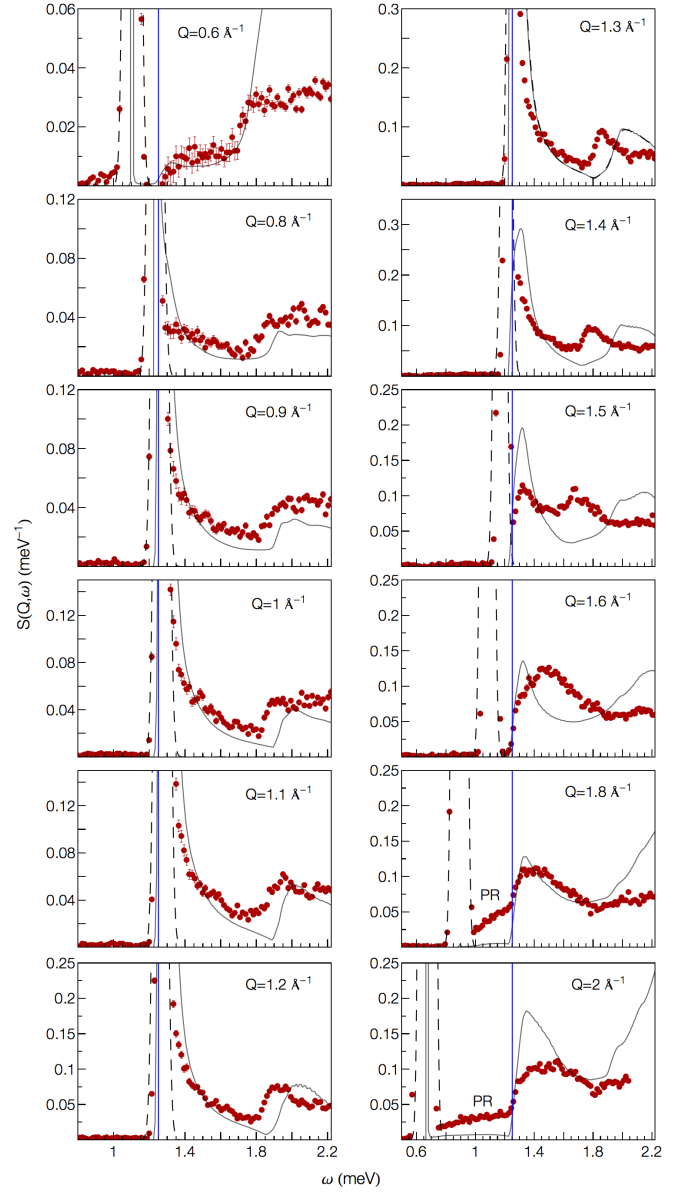


FIG. 17. Dynamic structure factor $S(Q, \omega)$: spectra for different wave vectors Q in the maxon region, at $P = 24$ bars. Filled circles: Experimental $S(Q, \omega)$. Solid lines: theory at the density $n = 0.0255 \text{ \AA}^{-3}$. Dashed lines: Intensity of the phonon-roton mode (cut off) calculated directly from the self energy⁹ and convolved with the instrumental resolution of 0.07 meV . Blue line: energy of the roton-roton threshold. PR indicates phonon-roton multi-excitations.

energies. The peak at 24 bars is less intense than the corresponding one at SVP, suggesting that the maxon, strongly reduced at this pressure, is involved in the corresponding decay processes.

Finally, at the highest energies explored here, $S(Q, \omega)$ progressively converges towards the free-particle parabola, remaining below it (see Fig. 1). The so-called “glory” oscillations seen as a function of Q , both in the peak position and the width, are well documented in the

literature⁶⁸. Directly related to the corresponding oscillations in the static structure factor $S(Q)$, they result from the hard core part of the ${}^4\text{He}$ - ${}^4\text{He}$ interaction potential and from quantum coherence effects. Earlier works could not fit the spectra of the first oscillation with a single peak. The highly structured multi-excitations seen in the present work show that this peak of unusual shape results in fact from the superposition of a few multi-excitation “branches” corresponding to decays into a few single-excitations. Again, the dynamic structure factor in this region depends on pressure, and the spectra for $Q \approx 3.5 \text{ \AA}^{-1}$ are strongly affected by the collapse of the maxon.

VII. CONCLUSION

A comprehensive understanding of the dynamics of interacting Bose systems, going from the Landau quasi-particles and multi-excitations regimes, up to the high-energy limit where the independent particle dynamics is recovered, emerges from our combined experimental and theoretical work. The up-to-now largely unexplored multi-excitations regime has been systematically investigated. Ghost-phonon and ghost-roton regimes have been observed, associated to phonon emission in the region of nearly supersonic multi-excitations, by a Cherenkov-like process qualitatively predicted by Burkova’s exten-

sion of Pitaevskii’s theory. Several other multi-excitation branches or thresholds have been observed and identified in the low energy sector, where an excellent quantitative agreement is found with the predictions of the Dynamic Many-Body theory. This agreement extends even to high pressures, near solidification, as shown for example for the remarkable case of the maxon disintegration into two rotons. The calculations including specific multiparticle fluctuations to all orders⁹ provide a good description of the dynamics for energies as high as 2 meV. Above this value, higher order processes dominate the dynamics. Our high energy/wave vector data call for further theoretical developments able to describe quantitatively the behavior observed at higher energies, above the simple multi-excitations region but still substantially below the quasi-free particle (impulse-approximation) sector.

VIII. ACKNOWLEDGEMENTS

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