

Superfluid ^4He dynamics beyond quasiparticle excitations

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The dynamics of superfluid ^4He at and above the Landau quasiparticle regime is investigated by high precision inelastic neutron scattering measurements of the dynamic structure factor. A highly structured response is observed above the familiar phonon-maxon-roton spectrum, characterized by sharp thresholds for phonon-phonon, maxon-roton and roton-roton coupling processes. The experimental dynamic structure factor is compared to the calculation of the *same* physical quantity by a Dynamic Many-body theory including three-phonon processes self-consistently. The theory is found to provide a quantitative description of the dynamics of the correlated bosons for energies up to about three times that of the Landau quasiparticles.

Liquid ^4He is the prime example of a strongly correlated quantum many-body system. It has been studied for decades and still offers surprises that lead to new insights. Understanding the helium fluids, due to their generic nature, lies at the core of understanding other strongly correlated many-particle systems, and is therefore of interest not only for the quantum fluids community. The description of the elementary excitations of superfluid ^4He in terms of phonon-roton quasiparticles is a cornerstone of modern physics, with profound implications in condensed matter physics, particle physics and cosmology. The empirical dispersion relation of these excitations proposed by Landau [1, 2] to explain thermodynamic data found support in the microscopic theory of Feynman and Cohen [3], initiating a fruitful development of the field theoretical description of correlated quantum particles.

From an experimental point of view, neutron scattering techniques allowed the direct observation of very sharp excitations in superfluid ^4He at low temperatures. The density fluctuations displayed, as predicted, a continuous phonon-maxon-roton dispersion curve: a linear phonon part at low wave vectors followed by a maximum (“maxon”) and then a pronounced “roton” minimum at a finite wave vector of atomic dimensions. Phonons naturally arise as the Goldstone mode associated with the continuous symmetry of the interacting system, whereas rotons are a direct consequence of strong correlations. Roton-like excitations have been proposed in cold atomic gases [4], in one-dimensional ^4He [5] and in two-dimensional fermionic systems [6]. Superfluidity emerges phenomenologically as a natural consequence of

the dynamics, while the knowledge of the dispersion relation allows the calculation of low temperature thermodynamic properties of superfluid ^4He [7].

The relation between theory and experiment, however, is not straightforward [8, 9]. The excitations considered by Landau, Feynman and others correspond to the single-particle response function associated with the description of an effective vacuum - the superfluid ground state - and non-interacting quasiparticle excitations. Neutron scattering, in turn, gives access to the dynamic structure factor $S(Q, \omega)$, a quantity related to the dynamic susceptibility, *i.e.*, the linear response of the system to a density fluctuation. The latter has a strong weight along the phonon-roton dispersion curve, but it also contains additional contributions, already observed in early neutron scattering experiments [10–13]. Much of the work on superfluid ^4He has been focused on the single-particle response function, attempting to extract the quasiparticle dispersion relation [12] and life-time [14, 15] from neutron data. In parallel, considerable effort has been devoted to the development of an accurate theoretical description of the dynamics of superfluid ^4He using various techniques [9, 16–20].

In this Letter, we investigate in detail the multi-excitation region of the dynamic structure factor $S(Q, \omega)$ of superfluid ^4He . We present new, high-precision neutron scattering measurements of $S(Q, \omega)$ at very low temperatures. We find several new features, in particular a “ghost phonon”, but also multi-particle thresholds that are much sharper than in earlier experimental work. These features are in agreement with a quantitative microscopic calculation of the density fluctuations $S(Q, \omega)$

within a recent Dynamic Many-Body theory [20]. Theoretical and experimental results for $S(Q, \omega)$ in a broad sector of the spectrum can be compared directly, leading to an unprecedentedly accurate description of the dynamics of superfluid ^4He .

The inelastic neutron scattering measurements were performed on the neutron time-of-flight spectrometer IN5 at the Institut Laue-Langevin using an incoming energy of 3.55 meV (wavelength 4.8 Å) and an energy resolution at elastic energy transfer of 0.07 meV. The high-purity superfluid ^4He sample was contained in a thin-walled cylindrical aluminum container of inner diameter 15 mm. The effective sample height in the beam was 50 mm. Cadmium disks were placed inside the cell at intervals of 10 mm to reduce multiple scattering, an important experimental artifact discussed below. The cell was connected to the mixing chamber of a dilution refrigerator via a copper piece equipped with silver sinter to ensure good thermal contact, thereby allowing measurements to be done at very low temperatures, $T < 100$ mK. The measurements were performed at saturated vapor pressure.

The quantity measured by a neutron spectrometer – the inelastic differential scattering cross section per target atom – is proportional to the dynamic structure factor:

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \hbar \omega} = \frac{b_c^2 k'}{\hbar k} S(Q, \omega)$$

where b_c is the bound atom coherent scattering length, k and k' the neutron wave vector before and after the scattering process, Q the wave vector transfer and $\hbar \omega$ the energy transfer [9]. Standard data reduction routines [21] were used to obtain the dynamic structure factor from the neutron raw spectra. The magnitude of $S(Q, \omega)$ was normalized by requiring that the single quasiparticle strength $Z(Q) = 0.93$ for $Q = 2.0 \text{ \AA}^{-1}$, a value obtained from previous works [9, 10, 20]. Fig. 1a displays essentially the raw data, after the usual corrections. The aluminum cell elastic background, measured before introducing the helium in the cell, was subtracted from the raw spectra. This led to the noisy region seen in Fig. 1a near zero energy. We also subtracted the inelastic signal originating from scattering events involving the aluminum cell and the helium sample. Rotons, due to their high density of states, dominate these processes, and this contribution is only significant at the roton energy. Since it is essentially Q -independent, it can be easily identified and removed. The subtraction of this contribution spoils the accuracy of the data in a small range around the roton energy in regions of the spectrum where the signal is small. The effect can be seen if the intensity scale is considerably expanded, for instance as in Fig. 2.

While earlier neutron scattering experiments [10–13] revealed the presence of broad, rather featureless multi-particle excitation regions above the single-particle dispersion curve, the improved precision (and possibly the much lower temperature) in the present experiment al-

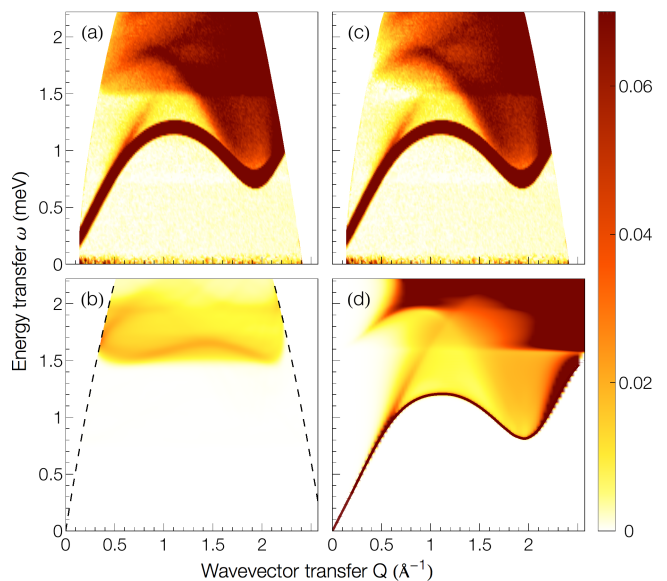


FIG. 1. (color online) (a) $S(Q, \omega)$ of superfluid ^4He measured as a function of wave vector and energy transfer, at saturated vapor pressure and temperature $T \leq 100$ mK. Contributions involving scattering with the aluminium cell have been subtracted, but not multiple scattering within the helium. (b) Helium multiple scattering contribution (numerical simulation); note that its magnitude is comparable to the multi-particle intensity seen in panels (c) and (d), and in Fig. 3. The dashed lines show the limits of the instrumental range, also valid for figures a and c. (c) Experimental dynamic structure factor $S(Q, \omega)$ after correction for multiple scattering. (d) Dynamic many-body theory calculation of $S(Q, \omega)$. Note that all the detailed features of the experimental data are reproduced. The units of the contour plots scale are meV^{-1} . The intensity is cut off at 0.07 meV^{-1} in order to emphasize the multi-excitations region. The apparent width of the Landau excitations in the experimental plot is due to the experimental resolution of 0.07 meV, while the calculated Landau dispersion curve has been highlighted by a thick line.

lowed us to observe a very rich structure in this region, with increasing weight at large wave vectors, as seen in the measured $S(Q, \omega)$ shown in Fig. 1a.

It is particularly important to distinguish the multi-particle excitations under investigation, which are an intrinsic property of helium, from multiple scattering. The former arise when a neutron creates in a single process a high energy perturbation which can decay into two or more excitations, while the latter is a spurious effect, dependent on the sample size, where a single neutron creates two or more excitations in successive scattering events. Since the two kinds of processes fulfill the same kinematic conservation rules, and their contributions have similar intensity for typical sample sizes, subtracting multiple scattering from the raw data is essential when dealing with the multi-particle region of the spectrum.

It is difficult in practice to determine this contribution

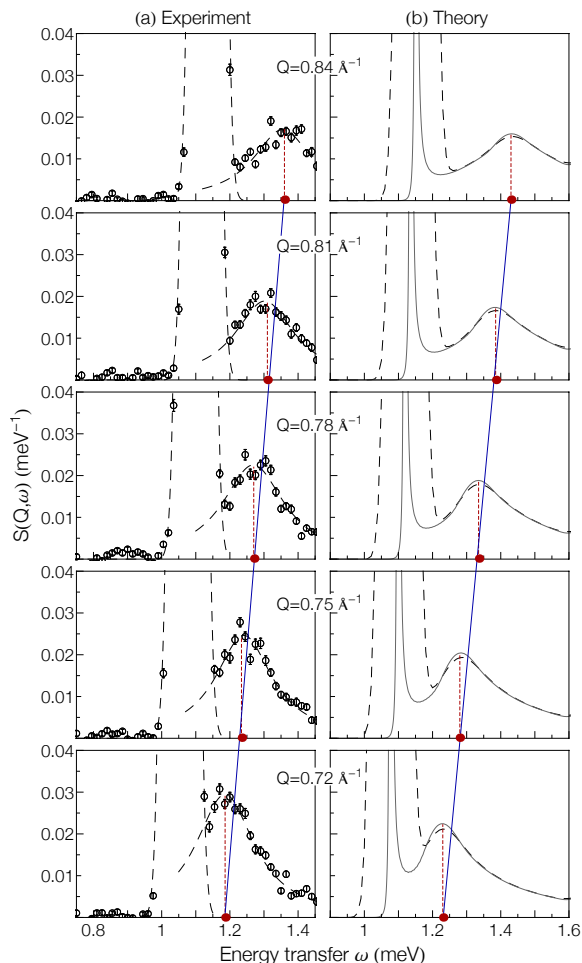


FIG. 2. (color online) (a) Experimental dynamic structure factor $S(Q, \omega)$: spectra at different wave vectors Q in the phonon region. The dashed lines are Gaussian fits of the phonon peak (cut off) and the much smaller “ghost phonon”. The maxima of the “ghost phonon”, indicated by vertical dashed lines, are located on the linear extension of the phonon dispersion (solid line). (b) Calculated spectra before and after convolution with the experimental resolution of 0.07 meV, shown as solid and dashed lines, respectively. The experimental data are very well reproduced by the mode-mode coupling calculation.

experimentally [22], for instance by using samples of different diameters, as one could naively believe. For this reason, we instead chose to perform an accurate calculation of the multiple scattering contribution using simulation software [23], and we verified the results with two other simulation programs [24, 25]. All three Monte Carlo programs track successive neutrons in a sample of given geometrical and physical characteristics, in particular its scattering function $S(Q, \omega)$. They have been adapted to calculate multiple scattering in helium. This contribution, shown in Fig. 1b, is found to be of the same order of magnitude as multi-particle excitations in all the region above 1.5 meV. The ratio of the multiple scatter-

ing intensity over the total intensity is 1.8%, a value in agreement with the calculation using the well-known semi-analytical method developed by Sears [22]. Multiple scattering was subtracted from the raw data, obtaining the corrected results for $S(Q, \omega)$ shown in Fig. 1c. The results shown in the figures clearly demonstrate that the new features we observe in this work are *not* caused by a multiple scattering artifact.

Having obtained reliable data for the multi-particle region of $S(Q, \omega)$, we can claim the unambiguous experimental observation of three new features in $S(Q, \omega)$: (i) A feature that we have named “ghost phonon” for obvious reasons, since it appears as scattering strength extrapolating linearly from the phonon for wave vectors $0.6 < Q < 0.9 \text{ \AA}^{-1}$; (ii) A prominent branch-like feature that appears at about twice the maxon energy; and (iii) A sharp *threshold* at twice the roton energy that extends to low wave vectors. Previous experimental works, including ours, show traces of the second feature, at the limit of the experimental resolution. Comparing the plots of Ref. [10–13] with the results shown in Fig. 1c illustrates the magnitude of the improvement in neutron techniques over the last decade.

The kinematic possibility of the decay of a mode into two low-lying quasiparticle excitations under energy and momentum conservation is a necessary condition for these effects to occur, and the observed features can be ascribed to modes decaying into pairs of excitations of large spectral weight. Even in the absence of a theory, it is possible to combine pairs of single-particle excitations to obtain the *position* of the main multi-excitation resonances in the dynamic structure factor (2-Phonons, 2-Rotons, 2-Maxons and M+R resonances). Obtaining the fine structure we observe, however, requires a quantitative calculation of mode couplings. Theoretical calculations [26, 27] based on early versions of the CBF method gave a highly structured multi-excitation region in the dynamic structure factor, which at first sight looks similar to the present experimental results. The similarity does not resist a more thorough inspection, since even the single particle modes were only very qualitatively reproduced. In particular, the calculated multi-excitations decay into Feynman modes instead of the true single-particle excitations, leaving large gaps in the spectrum. Also, the calculations predicted several additional features at high energies that have not been found in experiments. They provided nevertheless an appealing example of the effects that could be expected, and hence a motivation for further investigation of multi-particle dynamics.

The challenge to the theory imposed by the present data was to obtain the magnitudes of the corresponding coupling matrix elements. They have been calculated in this work using Dynamic Many-body theory [20, 28], which can describe excitations with wavelengths comparable to the interparticle distance. This requires an ap-

appropriate treatment of correlations at atomic scales. In their pioneering work, Feynman and Cohen introduced pair fluctuations [3], which allowed them to take into account “backflow” effects. In order to calculate mode-mode couplings in a microscopic and quantitative manner, we include here n -particle fluctuations for all n .

The dynamic structure factor calculated for a number density of $\rho = 0.022 \text{ \AA}^{-3}$ is shown in Fig. 1d. It is in quite satisfactory agreement with the experimental data. In addition to the well-known features of superfluid ^4He , namely, the linear phonon dispersion relation, the maxon, and the roton part of the spectrum turning eventually into the Pitaevskii plateau [29], the calculated $S(Q, \omega)$ also shows the finer features revealed by our high-precision experiment (see Fig. 1c).

Our Dynamic Many-body calculations predict a “ghost phonon” which, as seen in Fig. 1, extends to about twice the wave vector up to which the dispersion relation is essentially linear. This can be shown explicitly by the calculation of the three-phonon interaction [20]. Fig. 2 shows that the energy, strength, and shape of the calculated “ghost-phonon” agree very well with the experiment, even for wave vectors at atomic scale, of the order of 1 \AA^{-1} .

The mode above the maxon stems from interactions between rotons and maxons. Modes with that frequency and a wave vector close to that of the maxon decay predominantly into maxon and roton excitations with momenta close to those of rotons and maxons, but directed parallel and anti-parallel to the initial wave vector, respectively. A discussion of the kinematic situation, estimated by approximating the dispersion relation in the maxon and roton regions, is given in Ref. [20]. This feature is clearly present both in the experiments and the theory, and Fig. 1 c and d are very similar for energies as high as 2 meV.

A more quantitative comparison can be made on the spectra shown in Fig. 3. The experimental spectrum at $Q = 0.5 \text{ \AA}^{-1}$ is indeed very close to its theoretical analogue. At higher wave vectors, $Q = 0.8 \text{ \AA}^{-1}$, however, it is clear that only the low energy side of the experimental feature is reproduced by the theory. Contrarily to the situation encountered in the phonon region, where a very detailed calculation could be made, here the number of possible intermediate channels renders the calculation particularly demanding. The obtained semi-quantitative agreement is already quite gratifying.

The decay of an excitation of energy $2\Delta_r$ into two rotons with increasing angle between them explains the plateau or threshold that we observe down to low wave vectors. The effect, which has been extensively investigated [29], is discussed in detail in Ref. [20]. The noteworthy observation is that this structure is observable, with comparable strength in both experiments and theory (see Fig. 3), even below the maxon wave vector.

In conclusion, the dynamic structure factor of super-

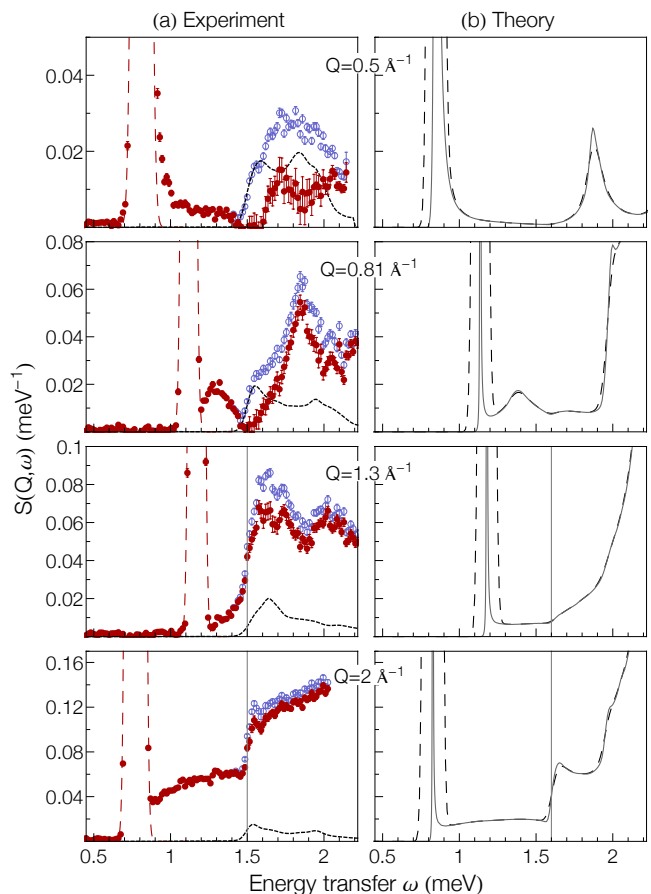


FIG. 3. (color online) (a) Experimental dynamic structure factor $S(Q, \omega)$: spectra for different wave vectors Q . Open circles: raw data; short-dashed line: calculated multiple scattering contribution; filled circles: $S(Q, \omega)$ corrected data (see text). The dashed lines are Gaussian fits of the phonon-roton peaks (cut off). (b) Theoretical dynamic structure factor spectra for the same wave vectors Q , before and after convolution with the experimental resolution of 0.07 meV, shown as solid and dashed lines, respectively. The plots have been chosen to emphasize differences between theory and experiment not readily observable in Fig. 1. Thin vertical lines: roton-roton threshold.

fluid ^4He , a canonical model system for correlated boson physics, exhibits a rich behavior at intermediate energies. We observe thresholds and fine structures delimited by kinematic constraints. The results are in remarkable agreement with the microscopic Dynamic Many-body calculation. There are still some quantitative differences, observable above the roton energy for wave vectors smaller than the roton one, between 1 and 2 \AA^{-1} . The experiments display a sizable response, which is present but weaker in the theory, as seen in Figs. 1 and 3. The origin of the observed enhancement of this interesting mode remains an open question for theorists. There are also undeniable discrepancies with the theory at energies above 2 meV. These can however be easily ascribed to higher

order processes [20]; they are mostly structureless and do not lend themselves to a clear identification. The combined experimental and theoretical work reported herein represents significant progress in the understanding of the effects of correlations on the dynamics of quantum fluids.

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