Dynamical Structure Factors in Quantum Many-Body Systems
from Quantum Monte Carlo Calculations

A. Roggero, F. Pederiva, and G. Orlandini

Dipartimento di Fisica, Università di Trento,
via Sommarive, 14 I-38123 Trento, Italy and
INFN - Istituto Nazionale di Fisica Nucleare,
Gruppo Collegato di Trento, I-38123 Trento, Italy

Abstract

An \textit{ab-initio} method for determining the dynamical structure function of an interacting many-body quantum system has been devised by combining a generalized integral transform method with Quantum Monte Carlo methods. As a first application, the coherent and, separately, the incoherent excitation spectrum of bulk atomic $^4$He has been computed, both in the low and intermediate momentum range. The peculiar form of the kernel in the integral transform of the dynamical structure function allows to predict, without using any model, both position and width of the collective excitations in the maxon–roton region, as well as the second collective peak. A prediction of the dispersion of the single–particle modes described by the incoherent part is also presented.

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Many important physical properties of matter, from viscosity to magnetic susceptibility, are closely related to the underlying microscopic dynamics. The computation of such quantities is routinely performed for classical systems. Moreover, thanks to the growing computational capabilities, we currently have a number of methods capable of a true ab-initio treatment of the ground state of quantum many-body systems. However, the detailed study of dynamical properties has been yet elusive.

In this letter we will focus on the problem of extending in a consistent and reliable way a class of Quantum Monte Carlo (QMC) algorithms to make it possible to determine the Dynamical Structure Function (DSF) for a generic quantum many-body system and a generic excitation operator. Such an extension is based on the use of the Integral Transform (IT) technique with generalized kernels. The amount and quality of information that can be extracted by the proposed scheme is illustrated in the application to the study of the coherent and incoherent density excitation spectrum in $^4$He.

At present, QMC calculations provide benchmark results for the study of a huge variety of many-body systems ranging from quantum chemistry, physics of ultra-cold gases, and nuclear physics. The most dramatic limitation of QMC methods is their inability to treat dynamical properties in a similarly reliable way. This failure is essentially due to the fact that QMC works in imaginary-time rather than in real time. This implies that quantities that do not directly translate into imaginary-time language, when analytically continued to real time, are affected by statistical noise that can be hardly reduced, making calculations unfeasible.

The mainstream approach to the problem is to attempt a numerical inversion starting from the imaginary-time autocorrelation function, i.e. the Laplace transform of the DSF. As is well known, such an inversion is an exponentially ill-posed problem [1] and sophisticated regularization techniques [2, 3] are needed to correctly extract the physical information. Due to the infinite range of the kernel function, the extraction of such informations is extremely delicate, and many details of the structure of the excitation spectrum can be missed. As an example, one of the most powerful inversion schemes, the Maximum Entropy Method [4, 5], cannot resolve the (measured) double peaked structure of $S(\mathbf{q}, \omega)$ in superfluid $^4$He [6], corresponding to a higher energy collective roton mode. In a recent paper this structure was eventually resolved inverting the imaginary time correlation function in a Path Integral Ground State calculation by using a falsification method based on a genetic algorithm [7].
For strongly interacting few-body systems the problem of computing various DSF of density and current excitations is solved by using a generalized integral transform approach, i.e. the Lorentz Integral Transform (LIT) method [8, 9]. The success of this approach, applied in nuclear physics, lays in the specific choice of the Lorentzian function as kernel of the IT. On the one hand this choice allows to calculate the transform with bound state techniques, even when the response is defined in the continuum, however avoiding its discretization. On the other hand, and most important, the fact that the kernel is a representation of the \( \delta \)-function allows for a reliable and stable inversion. So far, the application of this technique has been limited to a small number of particles (up to N=6 [10] and 7 [11]), due to the huge computational costs of the diagonalizations needed to calculate the LIT. In the following we will discuss how to extend the idea of the LIT to many-body systems, by developing a QMC equivalent.

At zero temperature the contribution to the response of a system of interacting particles due to a perturbative probe transferring momentum \( q \) and energy \( \omega \) to it, can be expressed using a spectral representation

\[
S_\hat{O}(q, \omega) = \sum_\nu |\langle \Psi_\nu | \hat{O}(q) | \Psi_0 \rangle|^2 \delta(E_\nu - \omega) \tag{1}
\]

\[
= \langle \Psi_0 | \hat{O}^\dagger(q) \delta(\hat{H} - \omega) \hat{O}(q) | \Psi_0 \rangle \tag{2}
\]

where \( | \Psi_0 \rangle \) is the ground state of the system, \( | \Psi_\nu \rangle \) are the final states of the reaction, \( \hat{O} \) is an excitation operator, \( \delta(\hat{H} - \omega) \) is the spectral-density of the hamiltonian and the summation is extended to all discrete and continuum spectrum states in the set.

The cost of a direct calculation of \( S_\hat{O}(q, \omega) \) becomes rapidly prohibitive as the number of particles or the energy transfer \( \omega \) increases. The latter problem is due to the fact that to account for continuum states one would need to solve the many-body scattering problem.

One can instead consider an integral transform of \( S_\hat{O}(q, \omega) \) with a generic kernel \( K(\sigma, \omega) \)

\[
\Phi(q, \sigma) = \int K(\sigma, \omega) S_\hat{O}(q, \omega) d\omega. \tag{3}
\]

The substitution of the expression (11) for \( S_\hat{O}(q, \omega) \) yields:

\[
\Phi(q, \sigma) = \sum_\nu \langle \Psi_0 | \hat{O}^\dagger(q) | \Psi_\nu \rangle K(\sigma, \omega) \langle \Psi_\nu | \hat{O}(q) | \Psi_0 \rangle \\
= \langle \Psi_0 | \hat{O}^\dagger(q) K(\sigma, \hat{H}) \hat{O}(q) | \Psi_0 \rangle \tag{4}
\]
Equation (4) can be viewed as a generalized sum rule which depends on a continuous parameter $\sigma$. Provided that the kernel and the excitation operator have suitable analytic properties, the right hand side of Eq. (4) can be calculated using bound-state type methods. This is the case both for the Stieltjes kernel \[17, 18\], and for the Lorentz kernel. However, while in the former case the inversion of the transform is as problematic as in the case of the Laplace kernel, in the latter case even a rather simple regularization procedure allows to obtain accurate and stable results. The reason can be easily understood. In the case of the Laplace or the Stieltjes kernels the information about $S_\delta(q, \omega)$ in the $\omega$ domain is spread in a large $\sigma$ domain. On the contrary the Lorentz kernel, as well as any function that is a $\delta$-function representation, keeps that information in an arbitrarily narrow $\sigma$ domain, governed by the width of the kernel. (In the $\delta$ function limit of the kernel no inversion would be needed!)

The number of $\delta$-function representations is large. However, very few have a practical implementation. In the past the use of Gaussian kernels has been investigated in different fields from condensed matter \[12, 13\] to non perturbative QCD \[14, 15\] with limited results. Here the idea is to recast one possible $\delta$-function representation in the imaginary-time propagation language, typical of QMC methods, in a similar way as was suggested in Ref. \[16\].

Consider the following family of integral kernels built out of the so-called Sumudu transform:

$$K(\sigma, \omega) = \frac{N}{\sigma} \left[ e^{-\mu \omega} - e^{-\nu \omega} \right]^P,$$

where

$$\mu = \frac{ln[b] - ln[a]}{b-a}a; \quad \nu = \frac{ln[b] - ln[a]}{b-a}b,$$

and the parameters $P, a, b$ are integer numbers with $b > a$. The normalization constant $N$ is a function of $P, a, b$ such that $\int d\omega K(\sigma, \omega) = 1$.

The efficiency of this transform is displayed by the fact that the kernel function converges to a delta function $\delta(\omega - \sigma)$ in the $P \to \infty$ limit, independent on the choice of $a$ and $b$. For a finite $P$ the kernel $\sigma K_P(\sigma, \omega)$ is still centered around $\omega = \sigma$ but has a finite width that depends on $P$. These properties make the choice of the resolution and the energy range of interest extremely flexible, similarly to the Lorentian kernel.

Using a binomial expansion, and rewriting powers as exponential functions, leads to a
more transparent form of the kernel:

\[ K_P(\sigma, \omega) = \frac{N}{\sigma} \sum_{k=0}^{P} {P \choose k} (-1)^k e^{-\ln(b/a) \frac{b-a}{b-a} P + k} \frac{\omega}{\sigma}. \quad (7) \]

By operating the substitution \( \omega \rightarrow \hat{H} \) according to Eq. (4), we are lead to a simple linear combination of imaginary-time propagators \((\hbar=1)\), taken at imaginary-time points \( \tau_{P_k} = \ln(b/a) \frac{a}{b-a} P + k / \sigma \). Projection QMC methods are all based on the implementation of such an imaginary-time propagation. The underlying idea is to solve the corresponding integral equation:

\[ \Psi(R, \tau) = \int dR' G(R, R', \tau) \Psi(R', 0). \quad (8) \]

This can be achieved, for instance, sampling a representation in coordinate space of the Green’s function \( G(R, R', \tau) \) to propagate a set of configurations representing in turn an expansion of the function \( \Psi(R, \tau) \) in eigenstates of the position operator (Diffusion Monte Carlo methods). Alternatively, it is possible to break up the Green’s Function in a product of short time propagators in coordinate space:

\[ \Psi(R, \tau) = \int dR' \ldots dR^n G(R, R^n, \Delta\tau) \times \]
\[ \times G(R^n, R^{n-1}, \Delta\tau) \cdots G(R', R', \Delta\tau) \psi(R', 0), \quad (9) \]

with \( \tau = n\Delta\tau \). This formulation is implemented in the so-called Path-Integral Ground State methods \[23\], and in the Reptation Monte Carlo (RMC) algorithm \[19\], where the whole path \( \{R, R', R'', \ldots, R^n\} \) is sampled from the product of the short-time propagators \( G \), possibly modified with the use of a suitable importance function \( \Psi_T \) to be determined in a variational calculation. The fact that estimating \( \Phi(\sigma) \) reduces to the computation of an imaginary time correlation function makes this second formulation more convenient and straightforward. In particular, path-based methods yield estimates that never depend on the (necessary) importance function used to improve the convergence of the calculation.

In order to evaluate the transform (3) within a QMC approach, we need to compute the imaginary-time correlation function, and then construct the corresponding linear combinations. A small width of the kernel might be achieved either using a large value of \( P \) or by reducing the value of the ratio \( b/a \). In both cases this would require the computation of the imaginary time correlation function for long imaginary time. This might indeed be a serious complication. However, a kernel that has a width smaller or comparable to the
distance between the typical structures of the DSF is in principle sufficient to extract useful informations from the transform. We have also confirmed this in numerical tests. In the application described below we used the kernel $\sigma K_P(\sigma, \omega)$ with the typical values of $P = 2$, $a = 1$, and $b = 2$.

As a first benchmark application to a realistic physical problem, we have considered the density excitation response in bulk atomic $^4$He at $T = 0$. The system is modeled as a box with periodic boundary conditions, containing $N = 64$ or $N = 125$ $^4$He atoms interacting via the HFDHE2 pair-wise potential ([22], [21]), which quantitatively reproduces the binding energy of bulk $^4$He up to the freezing point, effectively including three-body contributions. Calculations are performed at the experimental saturation density ($n_0 = 0.02186 \ \text{Å}^{-3}$). The density excitation operator is defined as:

$$\hat{O}(q) \equiv \hat{\rho} = \sum_{i=1}^{N} e^{i q \cdot r_i},$$

and the transformed DSF in Eq. (3) becomes

$$\Phi(\sigma) = \langle \Psi_0 | \sum_{i,j=1}^{N} e^{i q \cdot r_i} K_P(\sigma, \hat{H}) e^{-i q \cdot r_j} | \Psi_0 \rangle$$

As it is customary in neutron spectroscopy one can distinguish the contribution coming from the so-called coherent part, given by the terms with $i \neq j$, related to collective excitations, and an incoherent part with $i = j$ that essentially picks up contributions from single particle excitations. We have obtained results for both the full and for its incoherent part, in the most studied region of the spectrum: the low momentum phonon-maxon-roton part $q \approx 0.3 \div 3.5 \ \text{Å}^{-1}$. Computations have been performed by means of a Reptation Monte Carlo (RMC) algorithm, as described in Ref. [19]. The variational importance function includes two- and three-body correlations expanded in a basis set [20] and optimized using a variational Monte Carlo procedure. Ground state properties are well reproduced: the ground-state energy per particle is $\epsilon_{0}^{RMC} = -7.23 \pm 0.01 \ \text{K}$, in good agreement with previous calculations using the same potential [19], and with the experimental value $\epsilon_{0}^{exp} = -7.17 \ \text{K}$. The static structure factor $S(q)$ is consistent with experimental data and previous calculations ([19], [20]).

Turning to the result on $S(q, \omega)$, the striking difference between the estimate obtained inverting the transform with the Laplace kernel or the one in Eq. (5) can be seen in Fig. 1, where we compare the results of the inversion obtained from RMC data with both kernels.
FIG. 1: A typical result for the response function \((Q, \omega)\) in liquid \(^4\)He. Points with errorbars are experimental results at \(Q = 0.4\,\text{Å}^{-1}\) at \(T = 1.34\,\text{K}\). The continuous line is the result obtained using a Laplace kernel. The dotted line is the result computed by using the Sumudu kernel. Theoretical calculations refer to \(Q = 0.44\,\text{Å}^{-1}\), value determined by the size of the simulation box.

Apart from the small shift of the peak due to the 0.04 Å⁻¹ difference in the momentum transfer (the momenta are limited by the discretization imposed by the use of a finite simulation cell, here \(L = 14.306\,\text{Å}\)), the new kernel permits to retrieve the information on the second peak and gives a much more realistic height and width of the one-phonon peak.

A few comments are necessary concerning the methods used to invert the transform. We have used both the Entropy Maximization Maximum Likelihood (EMML) and the Simultaneous Algebraic Reconstruction Technique (SMART). The bars in the figures indicate the computed widths of the excitations. Both the peak position and the linewidth are robust with respect to the inversion method used. We found that for \(q \leq 2.4\,\text{Å}^{-1}\) both methods converge to the same solution. In Fig. 2 we have plotted the excitation spectrum obtained using the new transform. The experimental low-lying part is extremely well reproduced up to \(q \approx 2.6\,\text{Å}^{-1}\), where the dispersion does not bend over around \(2\Delta\) but continues to raise. In this region, however, the EMML and SMART give different results indicating that the statistical uncertainty in the QMC data is too large to allow a consistent reconstruction. The two-phonon branch is clearly visible and well resolved. As it happens in Ref. 7, it only qualitatively compares to the experiment. However, it should be noted that at \(T = 0\) the peak corresponding to the collective excitation should be substantially
FIG. 2: Dispersion of the collective modes in liquid $^4$He at equilibrium density and $T = 0$. Points with errorbars are the computed values. Errorbars are estimates of the width of the peaks. $+$ and $\times$ are the corresponding experimental data from Ref. [24] at $T = 1.1K$.

FIG. 3: Dispersion of the first and second peak of the incoherent DSF computed by means of the Sumudu integral transform. Empty squares are simulations performed in a simulation box with $N=64$ atoms, stars refer to a box with $N=125$ atoms. The dashed line is the free-particle excitation spectrum. $\Delta$ is the roton gap, and the lines at energy $\Delta$ and $2\Delta$ are drawn as reference.

An estimate of the intrinsic peak width is $\Delta \omega \approx 5 \times 10^{-4}K$ [27]. Therefore, the experimental width is essentially due to the resolution of the apparatus [26].

The dispersion of the peaks of the collective modes, displayed in Fig.2, was obtained combining two simulations at $\Delta \tau = 0.002K^{-1}$ and $0.001K^{-1}$ respectively obtaining a mesh separation less than 0.5 until about $40K$. In order to obtain meaningful results in the high energy regime, a large collection of RQMC data taken with different imaginary-time steps is needed in order to increase the sampling points. Due to this technical difficulty,
at present we have not performed an exhaustive research in the high momentum-transfer limit. However, some preliminary calculations show that the spectrum has the expected approximately free-particle like behavior, and that for $Q \approx 6 A^{-1}$ and above the incoherent part of the Dynamic Structure Factor accounts for the total scattering.

Indeed, the most useful feature of these calculations is that the resolution is good enough to allow for separately computing the incoherent part of the full response function, in order to study single-particle excitations.

In Fig.3 we have plotted the calculated excitation spectrum of single-particle excited states. The spectrum shows at least two distinct branches. A lower energy excitation starts from $Q \approx 0.5 A^{-1}$ and propagates with a velocity resulting slightly higher than the superfluid critical velocity $v_e/v_c \approx 1.57$. A second branch can be observed starting at an energy slightly below two times the roton energy, tending asymptotically to the free particle spectrum. Interestingly enough, the lower energy branch crosses the collective excitation spectrum exactly at the roton minimum, thereby reinforcing the picture of the roton as a single particle excitation of an atom exiting the superfluid. The behavior of these single-particle excitations might be significantly affected by the quantum many-body correlations induced by the particle-particle interaction. An extensive experimental analysis of the single particle excitations properties in superfluid $^4$He is unfortunately not available.

We have proposed a method to extract reliably well resolved spectra from numerical calculations implementing imaginary time propagation of an initial state, such as DMC or RMC. Computations might be easily extended to the $T \neq 0$ case by using standard PIMC methods. The application to the study of the collective and single-particle excitations in $^4$He shows the robustness and the higher resolution power of this technique. The limit to the accuracy of the spectra is in principle limited only by the available computer power available.

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