

Bose–Einstein quantum statistics and the ground state of solid ^4He

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Abstract. The ground state of solid ^4He is studied using the diffusion Monte Carlo method and a new trial wave function able to describe the supersolid. This wave function is symmetric under the exchange of particles and used as a guiding function in the method allows for reproducing the experimental equation of state. The use of this zero-temperature technique overcomes the conceptual ambiguity of finite-temperature methods in the search of a supersolid. Results for the one-body density matrix show the existence of off-diagonal long-range order with a very small condensate fraction $\sim 10^{-4}$, the specific value being not fully independent of the trial wave function due to the remaining bias in the extrapolated estimator. The superfluid density of the commensurate system is below our resolution threshold, $\rho_s/\rho < 10^{-5}$. This zero-temperature result is incompatible with recent experimental measures of superfluidity in solid ^4He showing that the origin of the experimental findings is not that of a supersolid in a perfect crystal. Introducing in the system a 1% concentration of vacancies the superfluid density is manifestly larger, $\rho_s/\rho = 3.2(1) \times 10^{-3}$.

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Contents

1. Introduction	2
2. Trial wave function and method	3
3. Results	4
4. Discussion and conclusions	9
Acknowledgments	9
References	9

1. Introduction

Solid ^4He is far from being a classical crystal as proved by its high degree of anharmonicity, large kinetic energy per particle ($T \sim 24\text{ K}$) and significant displacements, measured by the Lindemann ratio $\gamma \sim 0.26$ (to be compared with classical solids, with $\gamma \sim 0.03$ near melting). The counterintuitive possibility of simultaneous solid order and superfluidity in solid ^4He has attracted the attention of both theory and experiment for a long time, starting with the theoretical proposal of Andreev and Lifshitz [1] about a possible supersolid phase related to the presence of a finite concentration of vacancies. Finite values of the superfluid density and/or condensate fraction in solid ^4He would be a macroscopic effect induced by Bose–Einstein statistics. After several unfruitful experiments to detect superfluid signals in solid ^4He , Kim and Chan reported in 2004 the first evidence of non-classical rotational inertia (NCRI) both in a confined environment [2] and in bulk [3]. From then on, several other experimental groups have observed NCRI using different samples containing small or ultra-small ^3He concentrations, in a simple crystal or in a polycrystal, and using several annealing schemes [4]. There is an overall agreement of all the data concerning the onset temperature $T_0 = 75\text{--}150\text{ mK}$ at which the superfluid fraction becomes zero, the lowest value corresponding to ultra-pure samples (only 1 ppb ^3He). Much more controversial is the value of the superfluid density since the experimental values reported so far change by more than one order of magnitude ($\rho_s/\rho \simeq 0.03\text{--}0.5\%$) depending on the purity, annealing, conditions in which the crystal is grown, etc. This high dispersion has led one to think that the superfluid signal observed in solid ^4He is probably due to the presence of some defects in the crystal, which could be of a different nature: dislocations, vacancies or grain boundaries. In fact, superfluid flow has been detected when grain boundaries are present [5] and also recent measurements of the specific heat have shown a broad peak at the same onset temperature T_0 [6].

Theoretical calculations at very low temperatures, based on the path integral Monte Carlo (PIMC) method, have not been able to reproduce the experimental findings on the supersolid. PIMC results show that a commensurate perfect crystal does not exhibit either superfluid fraction [7] or condensate fraction [8]. Finite values of ρ_s/ρ have been observed only when disorder is introduced in the form of a glassy phase [9] or through dislocation lines [10], but puzzlingly no signal of the superfluid transition is obtained over the temperature range observed in experiments. As the critical temperature of a supersolid is not known, conceptually any finite-temperature method cannot provide the definite answer and one has to resort to a strictly zero-temperature calculation. In this paper, we propose a new trial wave function which allows simultaneously for spatial solid order and Bose symmetry, and with the benefit of a simple use for importance sampling in diffusion Monte Carlo (DMC) calculations.

The remainder of the paper is as follows. In section 2, we analyze the symmetrized trial wave function that is proposed and describe briefly the method of calculation. In section 3, we present the results that we obtain for the ground state of solid ^4He . We finish in section 4 by giving some discussion and the conclusions of this work.

2. Trial wave function and method

The structure and energetic properties of solid ^4He at zero temperature have been widely studied in the past using the Nosanow–Jastrow (NJ) trial wave function,

$$\Psi_{\text{NJ}}(\mathbf{r}_1, \dots, \mathbf{r}_N) = \prod_{i < j}^N f(r_{ij}) \prod_{i, I=1}^N g(r_{iI}), \quad (1)$$

where N is the number of particles and lattice sites (commensurate crystal), $f(r)$ a two-body correlation function and $g(r)$ a one-body localization factor which links every particle i to its site I . The wave function Ψ_{NJ} leads to an excellent description of the equation of state and the structure properties of the solid [11], but it cannot be used for estimating properties that depend directly on the Bose–Einstein statistics since it is not symmetric under the exchange of particles. The symmetry requirement can be formally written as

$$\Psi_{\text{PNJ}}(\mathbf{r}_1, \dots, \mathbf{r}_N) = \prod_{i < j}^N f(r_{ij}) \left(\sum_{P(J)} \prod_{i=1}^N g(r_{iJ}) \right), \quad (2)$$

with a sum over all the permutations $P(J)$ of lattice sites. This wave function has been used in the past in variational Monte Carlo (VMC) calculations by introducing an explicit sampling over the permutation space [12]. However, this sampling is inherently inefficient and difficult to incorporate in a DMC code. To avoid the complexity of random walks in the permutation space one can approximate Ψ_{PNJ} by [13]

$$\Psi_{\text{LNJ}}(\mathbf{r}_1, \dots, \mathbf{r}_N) = \prod_{i < j}^N f(r_{ij}) \prod_{i=1}^N \left(\sum_{J=1}^N g(r_{iJ}) \right). \quad (3)$$

This wave function fulfills the symmetry requirement but it is not well-suited for genuinely describing solid ^4He . The problem with using Ψ_{LNJ} as an importance sampling function in DMC is that the solid phase melts, arriving to liquid (L) or glassy configurations. This outcome can be understood by analyzing the behavior of the localization factor (second term in (3)) under the per-site occupancy number. Contrary to what is physically reasonable, multiple occupancy per site is only suppressed via the Jastrow part (first term in (3)). To preserve the solid structure it is necessary to take into account, already in the localization factor, a penalization accounting for the voids in the original sites created by multiple occupancy, a feature missing in the Ψ_{LNJ} wave function.

We introduce a new type of wave function Ψ_{SNJ} in which this feature is present and at the same time the necessary requirements of solid order and Bose symmetry are fulfilled. The key point is to use the site occupancy as the building block for the localization factor, thus voids are unequivocally taken into account. Our model takes the form

$$\Psi_{\text{SNJ}}(\mathbf{r}_1, \dots, \mathbf{r}_N) = \prod_{i < j}^N f(r_{ij}) \prod_{J=1}^N \left(\sum_{i=1}^N g(r_{iJ}) \right), \quad (4)$$

where the product in the second term runs over sites instead of particles, and the localization factor properly suppresses the voids arising due to double occupancy, a right behavior which is also present in the more general wave function Ψ_{PNJ} . There have been other proposals for the study of the supersolid which do not rely on the symmetrization of the NJ model: a Bloch-like function [12], inspired in the band theory for electrons, and the shadow wave function [14]. The first model was used in the past in the VMC simulations of Yukawa solids [12] and proved to be difficult to optimize and provided significantly worse energetic results than the NJ model. More successful has been the shadow wave function which has been applied recently in the VMC calculations of supersolid ^4He [14]. However, it has never been applied as an importance sampling wave function in a DMC calculation. Recently, it has been implemented in the path integral ground state (PIGS) method to eliminate the variational constraints and applied to the study of two-dimensional solid ^4He [15].

The ground state of solid ^4He has been studied by using the DMC method and Hamiltonian

$$H = -\frac{\hbar^2}{2m_{\text{He}}} \sum_{i=1}^N \nabla_i^2 + \sum_{i<j}^N V(r_{ij}), \quad (5)$$

where N is the number of atoms and the interaction between particles is modeled by the semi-empirical radial pair potential due to Aziz *et al* [16]. The DMC method solves stochastically the imaginary-time (τ) Schrödinger equation, providing exact results for boson systems within controllable statistical errors. When $\tau \rightarrow \infty$, the ground state dominates and one has a collection of walkers $\mathbf{R}_i \equiv \{\mathbf{r}_1, \dots, \mathbf{r}_N\}$ that follow the probability distribution function $(\Psi_0 \Psi)$, Ψ_0 and Ψ being the ground-state wave function and the trial wave function for importance sampling, respectively. The short-time Green's function according to which the walkers evolve is accurate to order $(\Delta\tau)^3$ [17] and internal parameters of the calculation such as the mean population of walkers and time step $\Delta\tau$ have been adjusted to eliminate any bias.

3. Results

We have carried out DMC simulations of solid ^4He using an hcp lattice with $N = 180$ and 448 atoms and a fcc one with $N = 108$; the range of densities analyzed starts slightly below melting, $\rho = 0.470\sigma^{-3}$ ($\sigma = 2.556 \text{ \AA}$) with pressure $P = 21$ bar, and ends at $\rho = 0.600\sigma^{-3}$ with $P = 160$ bar. The Jastrow factor in Ψ_{NJ} (1) and Ψ_{SNJ} (4) is of McMillan type, $f(r) = \exp[-0.5(b/r)^5]$, and the Nosanow term is in both cases a Gaussian, $g(r) = \exp(-0.5\beta r^2)$. The parameters b and β of Ψ_{SNJ} have been optimized using VMC, the optimal values being $b = 1.10\sigma$ and $\beta = 10.1\sigma^{-2}$, and we have neglected their slight density dependence. The variational energy at density $\rho = 0.491\sigma^{-3}$ is $E/N = -4.246(15)$ K, to be compared with the DMC value $E/N = -5.812(10)$ K. If one *measures* the quality of the trial wave function defining $(E_{\text{DMC}} - E_{\text{VMC}})/E_{\text{DMC}}$, one gets 30% for Ψ_{SNJ} and 70% for the liquid phase at the same density and using only the Jastrow term. In both cases and as expected, the DMC method arrives to the exact value for each phase with no problems. If the quantum statistics is ignored and one uses Ψ_{NJ} , the VMC energy is $E/N = -5.071(21)$ K with optimal parameters $b = 1.12\sigma$ and $\beta = 7.5\sigma^{-2}$.

In figure 1, we show DMC results for the energy per particle E/N as a function of the density for both the symmetric SNJ (4) and non-symmetric NJ (1) wave functions. The calculations used an hcp lattice with $N = 180$ atoms and size corrections to the energy were

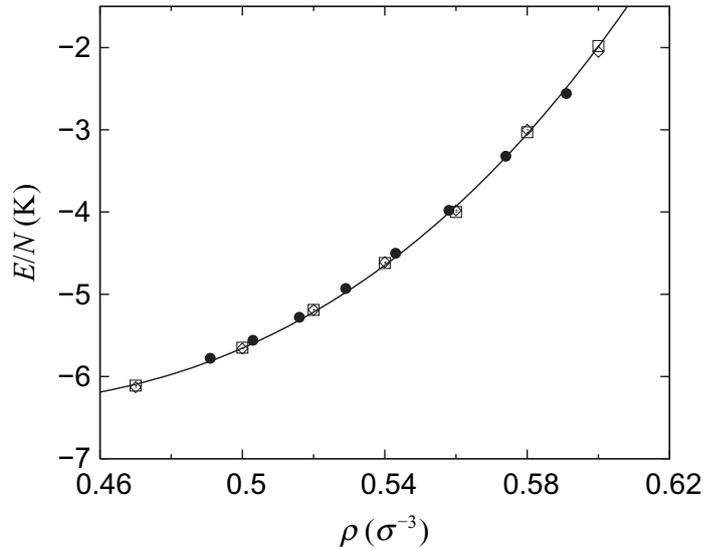


Figure 1. DMC results for the energy per particle of solid ^4He as a function of the density. Squares and diamonds stand for the symmetric (SNJ) and non-symmetric (NJ) wave functions, respectively. The solid line is a polynomial fit to the SNJ results. Experimental data from [18] is plotted with solid circles.

estimated using the N -dependence observed in VMC calculations of the same system. In a recent work [11], we have verified that this procedure improves the equation of state with respect to the standard approach of assuming that the system is homogeneous beyond $L/2$, with L being the length of the simulation box. As is known, the NJ model is able to reproduce the experimental equation of state with high accuracy; our present results confirm this feature as can be observed in figure 1. More importantly, the results obtained with the right symmetrization (SNJ) are statistically indistinguishable of the NJ ones and are in agreement with experiment. On the contrary, we have carried out some simulations with the LNJ model (3) and found that the energies are larger than the SNJ and NJ ones and the solid order is lost, the spatial structure resembling a glassy state.

One of the most clear signatures of the solid phase is drawn from the form of the static structure function $S(k) = 1/N \langle \rho_{\mathbf{k}} \rho_{-\mathbf{k}} \rangle$, with $\rho_{\mathbf{k}} = \sum_{i=1}^N e^{i\mathbf{k} \cdot \mathbf{r}_i}$. In figure 2, we show DMC results of $S(k)$ for hcp solid ^4He calculated using the pure estimator method [19]. The results presented correspond to a density $\rho = 0.491 \sigma^{-3}$ and have been obtained for the symmetric (SNJ) and non-symmetric (NJ) wave functions. Both results look rather similar, with the three main singular peaks corresponding to the reciprocal lattice points clearly visible. The height of these peaks for the symmetric system is slightly smaller than for the NJ one but these results confirm that the solid order is preserved by Ψ_{SNJ} . The small decrease in the strength of the main $S(k)$ peaks is probably related to the atomic diffusion we have observed along the simulations. By monitoring the distance of any particle to its initial position along the simulation one can know if there is any atomic diffusion in the crystal as a consequence of particle exchanges. Our results show that in a typical simulation $\sim 15\%$ of the atoms present displacements $r/a > 2$, with a being the lattice constant; obviously, the same measurement for the NJ case does not show any of them. This outcome is illustrated in figure 3. On the other hand, the relevance of these exchanges is not observed in the density profile around the lattice points $\mu(r)$. The results of $\mu(r)$ obtained

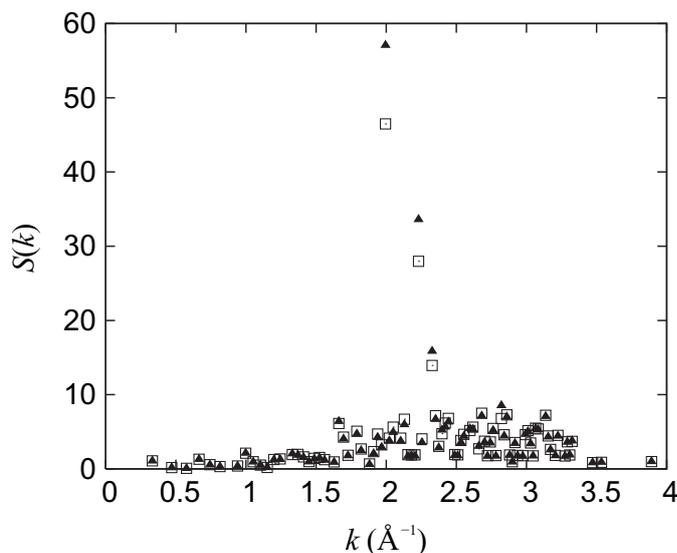


Figure 2. Static structure function $S(k)$ at density $\rho = 0.491 \sigma^{-3}$ ($N = 180$) calculated with the right symmetry (SNJ) (squares) and with the NJ model (triangles).

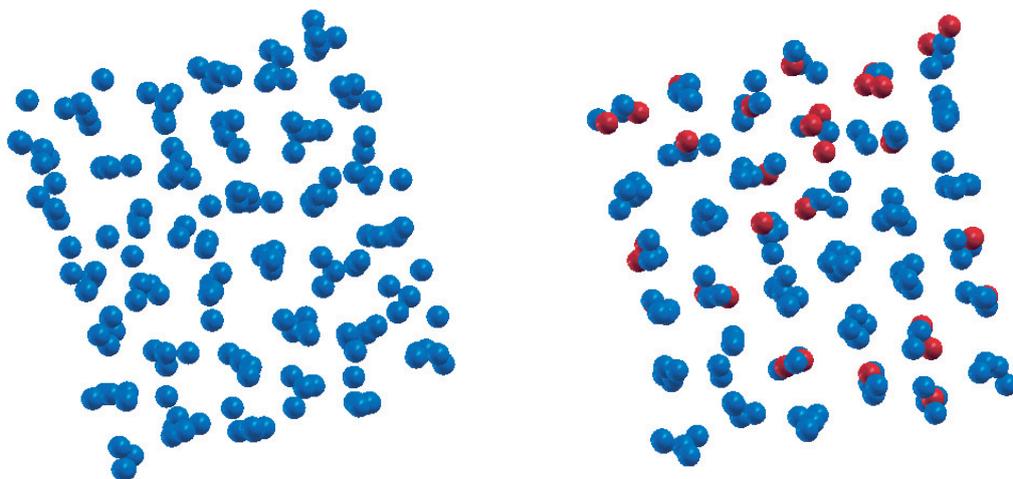


Figure 3. Instantaneous DMC configurations of solid ${}^4\text{He}$ obtained at density $\rho = 0.491 \sigma^{-3}$ by using trial wave function Ψ_{NJ} (left) and Ψ_{SNJ} (right) as importance sampling. Particles which move a distance larger than $2a$ with respect to their position at the beginning of the simulation are colored in red.

with SNJ and NJ are statistically indistinguishable and therefore both Lindemann ratios have a common value $\gamma = 0.26$, in agreement with experimental data.

A well-known drawback of the NJ model is the impossibility of answering the fundamental question about the possible existence of off-diagonal long range order (ODLRO) in solid ${}^4\text{He}$. Instead, the symmetrized model SNJ fulfills the right Bose–Einstein statistics and therefore is able to provide this information. Quantitatively, ODLRO is measured by the condensate fraction n_0 , that is estimated through the asymptotic behavior of the one-body density matrix $\rho(r)/\rho$,

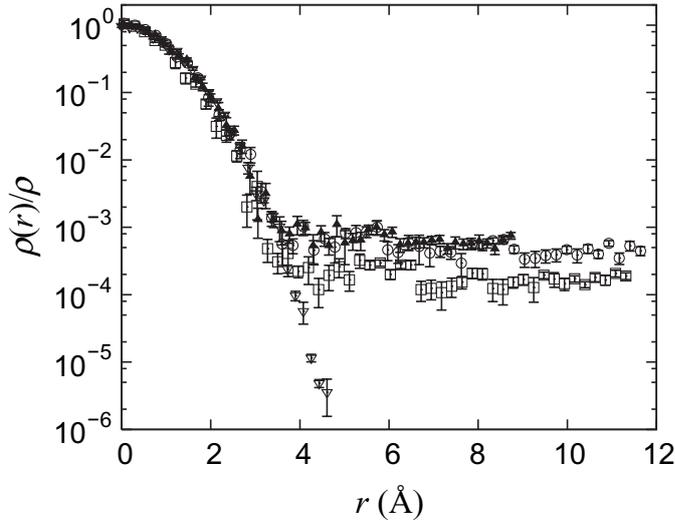


Figure 4. One-body density matrix $\rho(r)/\rho$ of solid ^4He at densities $\rho = 0.491$ (circles) and $\rho = 0.535\sigma^{-3}$ (squares). Full triangles stand for a calculation at $\rho = 0.491\sigma^{-3}$ with $N = 180$ particles and open triangles for the NJ result.

$n_0 = \lim_{r \rightarrow \infty} \rho(r)/\rho$. Results for $\rho(r)/\rho$ at densities $\rho = 0.491$ and $0.535\sigma^{-3}$, corresponding to pressures $P = 31$ and 68 bar, respectively, are reported in figure 4. At both densities, one can see unambiguously the existence of ODLRO, and from the asymptotic behavior we estimated the values of n_0 : $4.3(2) \times 10^{-4}$ and $1.7(4) \times 10^{-4}$ at $\rho = 0.491$ and $0.535\sigma^{-3}$, respectively (figures within parentheses correspond to the errors). For comparison, we show in the figure results for $\rho(r)$ obtained with the NJ model. In order to analyze the influence of the number of particles used in the simulation, we have carried out calculations of $\rho(r)$ with $N = 180$ and 448 . The results obtained are plotted in figure 4 at the lower density; within the statistical error both estimations lead to the same value of n_0 . All the results plotted in figure 4 have been obtained using extrapolated estimators fulfilling the condition that extrapolations of the same accuracy, namely $n_0 \simeq 2n_0^{\text{mix}} - n_0^{\text{var}}$ and $n_0 \simeq (n_0^{\text{mix}})^2/n_0^{\text{var}}$ (mix and var stand for mixed and variational estimators, respectively), coincide. This equality is achieved by slight variation of the parameters of the trial wave function (4) which otherwise does not modify the energy results. If the function $g(r)$ (4) is a Gaussian the agreement between both extrapolations at density $\rho = 0.491\sigma^{-3}$ is achieved with $\beta = 10.5\sigma^{-2}$. In order to estimate the remaining bias in the extrapolated estimator for n_0 we have carried out a DMC calculation at the same density but using a Padé-like function, $g_P(r) = \exp(-c_1 r^2 / (1 + c_2 r))$. Imposing the equality between the two extrapolations as in the Gaussian case, we obtain a condensate fraction $n_0 = 18(1) \times 10^{-4}$ corresponding to parameters $c_1 = 6.0\sigma^{-2}$ and $c_2 = 0.35\sigma^{-1}$ (the energy and superfluid density are not modified by the change of the function $g(r)$). Therefore, the extrapolated estimation of n_0 is not completely unbiased but both cases show unambiguously ODLRO. It is worth mentioning that we have made some VMC calculations removing the Jastrow part of the SNJ trial wave function and, in all cases, a finite value of the condensate fraction is observed. This result points to a property which seems to be inherent to the symmetrization of the NJ model.

From the different experimental measures carried out on the supersolid, it seems clear that the presence of vacancies, dislocations or other defects can be crucial to understand the

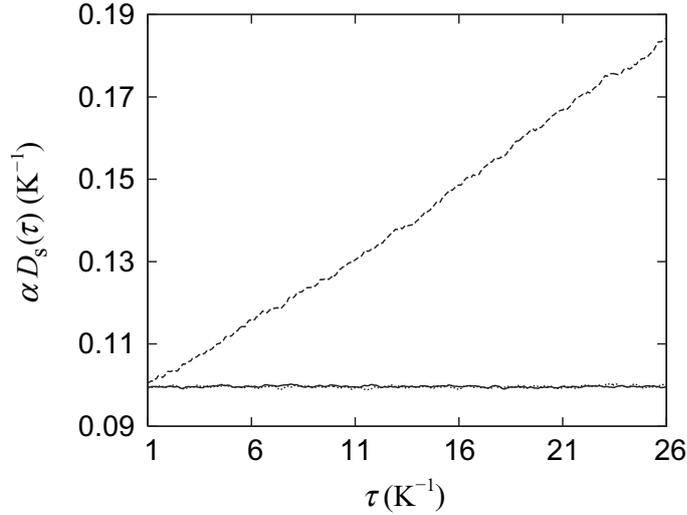


Figure 5. Diffusion of the center of mass $\alpha D_s(\tau)$ as a function of the imaginary time τ for solid ${}^4\text{He}$ at $\rho = 0.491\sigma^{-3}$. The slope of the lines is directly the superfluid density ρ_s/ρ . The solid line stands for the commensurate solid; the dotted line for the case with $N - 1$ sites; the dashed line for a solid with a $\sim 1\%$ concentration of vacancies.

dispersion on the superfluidity results. We have used the SNJ model to determine this influence in the condensate fraction and superfluid density for two particular cases: a vacancy, i.e. a system with N lattice sites and $N - 1$ particles, and the absence of a lattice site, i.e. N particles and $N - 1$ sites. Our simulations show that with $\sim 1\%$ vacancy concentration the condensate fraction raises nearly a factor two with respect to the commensurate case: at $\rho = 0.491\sigma^{-3}$, $n_0 = 8.7(4) \times 10^{-4}$. On the contrary, the effect of removing one of the sites in a 1% fraction does not modify the commensurate value $n_0 = 4.3(2) \times 10^{-4}$.

The superfluid density of a bosonic system can be calculated with DMC by extending the winding-number technique, originally developed for PIMC calculations, to zero temperature [20]. Explicitly

$$\frac{\rho_s}{\rho} = \lim_{\tau \rightarrow \infty} \alpha \left(\frac{D_s(\tau)}{\tau} \right), \quad (6)$$

where $\alpha = N/6D_0$ with $D_0 = \hbar^2/2m$, and $D_s(\tau) = \langle (\mathbf{R}_{\text{CM}}(\tau) - \mathbf{R}_{\text{CM}}(0))^2 \rangle$ with \mathbf{R}_{CM} the center of mass of the particles in the simulation box. In figure 5, results for the function $\alpha D_s(\tau)$ at a density $\rho = 0.491\sigma^{-3}$ are plotted as a function of the imaginary time τ . The slope of this function is directly the superfluid density ρ_s/ρ . Our results show that ρ_s/ρ for the commensurate solid is smaller than our precision limit, that we have established at 1×10^{-5} . We have carried out simulations with different number of particles with the same result. Also we have calculated $D_s(\tau)$ at higher pressure and at a pressure below melting where the solid is metastable obtaining identical conclusion. No significant difference with the regular system has been detected for the case with $N - 1$ sites, also shown in figure 5. On the other hand, when vacancies are present in the crystal with a $\sim 1\%$ concentration a superfluid signal clearly emerges; from the slope, $\rho_s/\rho = 3.2(1) \times 10^{-3}$. Finally, we have calculated the energy of formation of a single

vacancy [21],

$$\varepsilon = E(N - 1, 1, (N - 1/N)V) - \frac{N - 1}{N} E(N, 0, V) \quad (7)$$

with V the volume of the simulation box of the commensurate system. At density $\rho = 0.475\sigma^{-3}$, we obtain $\varepsilon = 17.7(1.6)$ K a value that is slightly larger than experimental estimations [22, 23] (14–15 K) and statistically compatible with a variational calculation using the shadow wave function [24] (15.6(3.8) K).

4. Discussion and conclusions

To summarize, we have introduced in the microscopic description of solid ^4He a new wave function (SNJ) which properly symmetrizes the well-known NJ model. Our results, based on the essentially exact DMC method, prove that the SNJ trial wave function used for importance sampling in the method is able to simultaneously reproduce the experimental equation of state (as the NJ does) and predict results for the condensate fraction and superfluidity due to its Bose symmetry. As the critical temperature of a possible supersolid is not known, the finite-temperature PIMC method suffers from the conceptual ambiguity of using a temperature that can be higher than the critical one. Therefore, only zero-temperature methods can provide a definite answer on the properties of the ground state. The extrapolated estimator of the condensate fraction is not able to remove completely the influence of the trial wave function but different functions show always ODLRO with a condensate fraction $n_0 \sim 10^{-4}$ near melting. A finite but smaller value has been obtained by Galli *et al* [14] using VMC with the shadow wave function. On the contrary, PIMC simulations at finite temperature [8] and a PIGS calculation in two-dimensional solid ^4He [15] do not observe ODLRO. An important conclusion of our work is that the zero-temperature upper-bound for the superfluid fraction $\rho_s/\rho < 1 \times 10^{-5}$ (a lower-bound to ρ_s has been prepared by Aftalion *et al* [25]) is incompatible with recent experimental measurements (3×10^{-4} – 5×10^{-3}) [4], ruling out an explanation of the superfluid signal as that of a supersolid in a perfect crystal. The introduction of 1% vacancies in the system increases the condensate fraction and a clear signature of superfluidity is detected. Work is in progress to understand microscopically the connection of other defects and/or disorder with both ρ_s and n_0 .

Acknowledgments

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