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Variational methods for ^{4}He using a modern He-He potential

S. A. Vitiello
Instituto de Física, Universidade Estadual de Campinas, 13083 Campinas–SP, Brazil

K. E. Schmidt
Department of Physics and Astronomy, Arizona State University, Tempe, Arizona 85287

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The two-body potential HFD-B3-FCI1 for He-He interactions proposed by Aziz and collaborators is analyzed using the variational Monte Carlo method. Complementary work using the Green's function Monte Carlo method is presented as well. The importance of three-body effects in the description of the interactions are considered and good evidence is presented for using a potential that takes into account these effects even at the equilibrium density of systems of ^{4}He atoms. An examination of the most usual trial functions forms employed in the studies of these systems, some experiments with other correlation factors, and some methodological notes complete this investigation. [S0163-1829(99)02741-1]

I. INTRODUCTION

Until the beginning of the 1980s the most popular potential used to investigate the properties of ^{4}He systems was the Lennard-Jones potential. From that time on, theoretical studies have often used a potential of the Hartree-Fock dispersion form, the HFDHE2 potential of Aziz and collaborators.[1] Today this is one of the most used potentials. Its widely acceptance was due in part to Green's function Monte Carlo (GFMC) results[2] that has shown how useful it was in computing properties in the condensed phase of the helium systems. Small inconsistencies with theoretical results and experimental data were detected sometime ago. Quantum Monte Carlo calculations[3] of the Born-Oppenheimer interaction energy have shown that the potential barrier was too high. It was also shown to be deficient in low temperature predictions of the second virial coefficients and the transport properties at high temperatures. In fact these properties are determined primarily by the overestimated repulsive wall.

This situation prompted revisions of the HFDHE2 potential. Since an early attempt[4] to address these difficulties, many improvements have been made by the Aziz group itself, for a review see Ref. 5. The series of revisions has led to the so-called HFD-B3-FCI1 potential.[6]

It is our intent in this paper to discuss the HFD-B3-FCI1 potential in relation to variational Monte Carlo calculations for systems of ^{4}He atoms. For completeness, and comparison, we give some GFMC results. The GFMC method for a boson system gives the correct ground-state energy subject only to statistical uncertainties and on the assumed interacting potential. This raw data, in the sense that it reveals all the properties and difficulties of this many-body system, allows one in principle to assess the accuracy of the employed potential. However, our focus here is on developing good trial wave functions for variational Monte Carlo calculations. These functions, in turn allow us to systematically improve and understand the physical correlations produced by the interaction potential. Another of our aims is to see the importance of the many-body forces involved. Since our results are model dependent, they might help us better understand the role of three-body and possible higher interactions. We have gone beyond simple energy minimizations, and have studied the implementation of boundary conditions, analyzed in some details the basis set method for constructing two-body correlations factors and introduced an analytical form for the pseudopotential in these factors. In the triplet term, together with this analytical form, due to fixed-phase arguments we have considered a functional form that relies on an exponential form rather than the usual Gaussian. For the system at equilibrium and for a solid at a density about 20% higher than the melting one, we give variational parameters for many of the trial functions we have used. We believe that such technical knowledge will prompt the adoption of the HFD-B3-FCI1 potential in future work. If optimal trial functions are needed for variational and GFMC calculations at other densities, our parameters can be interpolated and used as an initial values for the minimization procedure that will then become easier and faster.

In the next section we give a very simple overview of the HFD-B3-FCI1 interacting potential. In Sec. III we describe the trial functions used in this paper, along with a brief discussion of the basis set method of optimization of two-body correlations factors. We present the functional forms used in the triplet term and the motivation to use an exponential form for this term. In addition, an analytical form for the two-body pseudopotential is introduced. Section IV presents results of our calculations using the HFD-B3-FCI1 potential along with values of the variational parameters. To make comparisons, both in liquid and solid phases, we include results from the literature for the HFDHE2 potential as well. In the last section we discuss our results and draw some conclusions.

II. THE HFD-B3-FCI1 INTERACTING POTENTIAL

In most studies, the helium systems are assumed to be described by the Hamiltonian

$$H=\frac{-\hbar^2}{2m}\sum_{i=1}^{N}\nabla_i^2+\sum_{i<j}V(r_{ij}), \tag{2.1}$$

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where explicit three-body interactions are neglected. The model potential V we want to investigate, the HFD-B3-FCI1 potential, is based on the representation

$$E_{\mathrm{int}}=\Delta E_{\mathrm{scf}}+\Delta E_{\mathrm{corr}}, \tag{2.2}$$

where the interaction energy of a dimer is decomposed in a self-consistent field or Hartree-Fock part and in a correlation energy for the interaction. This last part is given by

$$\Delta E_{\mathrm{corr}}=\epsilon F(r)\sum_{j=0}^{2}\frac{C_{2j+6}}{r^{2j+6}} \tag{2.3}$$

with

$$F(r)=\begin{cases}\exp\left[-\left(\dfrac{Dr_m}{r}-1\right)^2\right] & \text{if } r\leqslant Dr_m,\\ 1, & \text{otherwise.}\end{cases} \tag{2.4}$$

The Hartree-Fock repulsion is represented by fitting *ab initio* calculations to

$$\Delta E_{\mathrm{scf}}=\epsilon A\exp(-\alpha r-|\beta|r^2). \tag{2.5}$$

In the HFDHE2 modern potential only the linear term in r was present in the exponential. In subsequent versions of the He-He potential, a term dependent on a parameter β was introduced to assure more flexibility in the fit.

The HFD-B3-FCI1 potential uses the overall damped Hartree-Fock dispersion form (HFD-B) and mimics very precise full configuration-interaction (FCI) calculations at intermediate separations. Retardation effects in this potential can be neglected at the accuracy level we perform our studies. The position of the energy minimum is $r_m=0.29683$ nm with a well depth of $\epsilon/\kappa_B=10.956$ K. For comparison the values for the HFDHE2 potential are $r_m=0.29673$ nm, and $\epsilon/\kappa_B=10.80$ K. Since the new potential is slightly deeper and has a large value for r_m, the well is shifted outwards by a small amount, σ(HFD-B3-FCI1)$=0.26413813$ nm while σ(HFDHE2)$=0.2639$ nm.

III. VARIATIONAL WAVE FUNCTIONS

Trial functions used to investigate quantum liquids and solids can be written in the general Feenberg[7] form as

$$\psi_T(R)=\prod_{i<j}f_{ij}\prod_{i<j<k}h_{ijk}\cdots\phi(R). \tag{3.1}$$

In this form, a wave function is written in terms of explicit n-body correlations factors, where n goes up to the number of particles in the system. In this work we consider two-body f and three-body h factors only. For the liquid state, we assume a model state $\phi\equiv 1$, where all particles are in a uniform condensate. For the solid phase, the model state ϕ is the traditional mean field factor, a product of one-body Gaussian terms, that localizes the particles around a given lattice.

A. The pair-product wave function

The simplest wave function that we consider is of the Jastrow form

$$\Psi_J(R)=\prod_{i<j}f(r_{ij}). \tag{3.2}$$

It explicitly correlates only pairs of particles through the factor $f(r)=e^{-(1/2)\,u(r)}$.

A simple form for the correlation factor was introduced by McMillan through a pseudopotential $u_M(r)$ of the form

$$u_M(r)=\frac{1}{2}\left(\frac{b}{r}\right)^5. \tag{3.3}$$

This pseudopotential u_M depends on a single variational parameter b.

For a Lennard-Jones 6–12 two-body potential, $f_M(r)=e^{-(1/2)\,u_M(r)}$ is an approximate solution of the two-particle Schrödinger-like equation

$$\frac{-\hbar^2}{m}\nabla^2 f(r)+[V(r)-\lambda]f(r)=0, \tag{3.4}$$

in the limit of small pair separation. The energy eigenvalue is discarded and only the dominant term r^{-12} is satisfied. Some of the effect of the remaining particles is taken into account in a variational calculation by tuning the parameter b of Eq. (3.3). As reported in Ref. 27, the zero-energy numerical solution of Eq. (3.4) is able to produce a radial distribution function $g(r)$ on par with a slightly generalized McMillan pseudopotential. The variational energy obtained with this pseudopotential was not reported.

The basis set method for quantum liquids and solids,[8] subsequently also called as the Euler Monte Carlo method in the literature, allows more freedom in simulation studies of these systems. The constraint of working with a fixed functional form of the pseudopotential was eliminated by defining

$$f(r)=\sum_{n=1}^{p}c_n f_n(r), \tag{3.5}$$

where the $f_n(r)$ are the spherically symmetric eigenfunction solutions of the two-particle Schrödinger-like equation (3.4), and the c_n variational parameters. Of course, any suitable basis could be used as well. Originally the solutions f_n were required to go smoothly to zero and to have a continuous derivative at a healing distance d. This distance can be treated as a variational parameter or eventually made equal to half the side of the simulation cell. Since we also require $f(r)$ of Eq. (3.5) to satisfy the continuity conditions, we have constrained the c_n to satisfy

$$1=\sum_{n=1}^{p}c_n. \tag{3.6}$$

This implies that for a basis set with $p+1$ elements (BS_{p+1}), due to the constraint of Eq. (3.6), the number of free variational parameters for the two-body correlation factor is only p. In other words, the number of elements in the basis is greater than the number of variational parameters by 1.

There are some advantages in using the basis set method having as basis elements solutions of the two-particle Schrödinger-like equation. In addition to having a physical motivation, in the limit of small pair separations, the resulting Jastrow function f of Eq. (3.5) is automatically a solution of the two-body Schrödinger-like equation.

Despite the well known importance of having the correct functional form for the two-body correlation factor at small pair separation, this fact is largely neglected in the literature when He-He modern interaction potentials are used. Perhaps because of its very simple form, and because it contains the basic physical requirements, namely a strong suppression of any overlap between any pair of particles and which goes smoothly to an uncorrelated pair of as the separation distance increases, often a pseudopotential of the McMillan form continues to be used. However, it is not hard to use the basis set method or to find an analytical pseudopotential that satisfies the two-particle Schrödinger-like equation in the limit of small pair separation when a modern He-He interaction potential is used.

In this work we have considered the analytical correlation factor obtained by finding the approximate two-particle Schrödinger's like equation in the limit of small pair separations for the HFD-B3-FCI1 potential. To take into account the effect of the remaining particles in the system, the potential amplitude and distances are rescaled by the variational parameters $\mathcal{A}$ and $\mathcal{S}$, respectively. The pseudopotential obtained in this way is given by

$$u_e(r)=\mathcal{A}\left(\frac{m}{\hbar^2}\right)\epsilon A e^{-\alpha r/\mathcal{S}}\left(\frac{r}{\alpha\mathcal{S}}+\frac{1}{\alpha^2}\right). \tag{3.7}$$

B. Explicit three-body terms

Accurate trial wave functions for the investigation of quantum liquids usually include three-body correlations. In Monte Carlo calculations a very useful form of such correlations is given by

$$h_{ijk}=\exp\left(-\frac{\lambda}{2}\sum_{\text{cycl}}\xi_{ij}\xi_{jk}\mathbf{r}_{ij}\cdot\mathbf{r}_{jk}\right), \tag{3.8}$$

where λ is a parameter and $\xi(r)$ is chosen variationally, cf. Schmidt *et al.*[9]

In this work we have used two different functional forms for ξ. One has a slightly simpler Gaussian form $(m=2)$ than that used in Ref. 9

$$\xi(r)=\exp\left[-\left(\frac{r}{w}\right)^m\right]. \tag{3.9}$$

The other has an exponential form, i.e., $m=1$. The motivation for this form comes from a fixed-phase[10] argument that implies

$$\xi(r)=\frac{u_e'(r)}{r}, \tag{3.10}$$

where $u_e(r)$ is the two-body pseudopotential of the exponential form. This same form was used previously in Refs. 11, 12. In addition to the amplitude λ in Eq. (3.8), there is only one more variational parameter introduced in the triplet terms: the width w in Eq. (3.9). We denote the Gaussian form of ξ by (T), and use (T_e) for the exponential form.

TABLE I. Comparison between two different ways of imposing the continuity of a pair-wise trial function and its first derivative. Results for the average total energy $\langle E\rangle/N$ and for the kinetic energy $\langle T\rangle/N$ per particle at the given number of particles N obtained at the equilibrium density and the given potentials. In the rows with the symbol u_p we have imposed the boundary conditions by a polynomial fit in the pseudopotential (PP) of the McMillan form, while in the rows with the symbol u we make a reflection of this PP at the edge of the simulation cell [see Eq. (4.1)]. In all cases the variational parameter b is equal to 0.307 nm.

PP	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)	N
	HFDHE2		
u_p	-5.717 ± 0.021[a]	15.099 ± 0.016	108
u	-5.7279 ± 0.0041	15.0892 ± 0.0027	64
u	-5.7246 ± 0.0078	15.1081 ± 0.0072	108
u	-5.7216 ± 0.0075	15.1209 ± 0.0073	256
	HFD-B3-FC11		
u_p	-5.848 ± 0.009	15.102 ± 0.006	108
u	-5.8512 ± 0.0079	15.1081 ± 0.0072	108
u	-5.8480 ± 0.0076	15.1209 ± 0.0073	256

[a] Reference 13.

C. The solid phase

The traditional approach to construct a solid phase wave function introduces a mean field term that localizes the particles around given lattice positions $\mathbf{l}_i$. In the Nosanow form, we use a product of Gaussian one-body factors

$$\phi(R)=\prod_i e^{-(C/2)(\mathbf{r}_i-\mathbf{l}_i)^2}. \tag{3.11}$$

IV. RESULTS

A. The liquid phase

A good trial wave function must be continuous and it is convenient to have at least the first derivative continuous as well. Usually the pseudopotentials are slightly modified near the edge of the simulation cell to avoid discontinuities. It also is possible to include long range pseudopotentials and compute their contribution using lattice sum techniques such as the Ewald method. We discuss only the cases were the correlations go to one and at least the first derivative goes to zero at or before a pair separation corresponding to half the simulation cell size. A possible way of implementing these requirements is to make a polynomial fit to the pseudopotential near the edge of the simulation cell so that it goes smoothly to zero. In Ref. 13 a third degree polynomial was used to fulfill the continuity requirements. The result obtained in this way, with the pseudopotential u_p, is displayed in Table I. An easier option to implement is given by the prescription

$$u(r)\rightarrow u(r)+u(r_B-r)-2u\left(\frac{r_B}{2}\right). \tag{4.1}$$

Here, $r_B/2$ is the cutoff length where the pseudopotentials and their first derivative are to go smoothly to zero. As we can see in Table I, for the number of particles we consider, both ways of imposing the continuity conditions give the

TABLE II. Results for a pair-product wave functions obtained through the basis set method at the equilibrium density, $\rho\,\mathrm{nm}^3 = 21.8$. The first column displays how many elements are used in the basis set. The average total energy $\langle E\rangle/N$ and the kinetic energy $\langle T\rangle/N$ per particle are given in Kelvin. The last column gives the healing distance explicitly or as $l/2$ when it is equal to half of the simulation cell. The value 0.72 nm corresponds to about half the size of the simulation cell of 64 bodies system.

BS	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)	N	Range (nm)
		HFDHE2		
7	-5.978 ± 0.011	-14.7950 ± 0.0064	64	$l/2$
7	-5.983 ± 0.019	14.805 ± 0.015	108	0.72
6	-5.9580 ± 0.0086	14.5772 ± 0.0046	64	$l/2$
5	-5.791 ± 0.012	14.1461 ± 0.0060	64	$l/2$
		HFD-B3-FCI1		
8	-6.176 ± 0.023	14.8457 ± 0.0094	108	0.72
7	-6.124 ± 0.025	14.705 ± 0.015	108	0.72
7	-6.135 ± 0.032	14.520 ± 0.017	108	$l/2$
6	-6.139 ± 0.024	14.569 ± 0.011	64	$l/2$
6	-6.097 ± 0.011	14.6675 ± 0.0059	108	0.72
5	-6.095 ± 0.034	14.302 ± 0.021	64	$l/2$
5	-6.038 ± 0.012	14.3161 ± 0.0098	108	0.72

same result. Due to its computational convenience, from now on, when dealing with pseudopotentials that need to be modified to go to zero at the edge of the simulation cell, we will use only the condition of Eq. (4.1) as a means of imposing the continuity of the wave function and its first derivative. The total energy of the system is computed using the kinetic energy estimator of the Pandharipande-Bethe form

$$\langle T\rangle_{\mathrm{PB}} = -\frac{\hbar^2}{2m}\left\langle \sum_{i=1}^{N} \frac{\nabla_i^2\Psi_T(R)}{\Psi_T(R)}\right\rangle. \tag{4.2}$$

The reported kinetic energy is obtained through an estimator of the Jackson-Feenberg form

$$\langle T\rangle_{\mathrm{JF}} = -\frac{\hbar^2}{4m}\left\langle \sum_{i=1}^{N} \frac{\nabla_i^2\Psi_T(R)}{\Psi_T(R)} - \left(\frac{\nabla\Psi_T(R)}{\Psi_T(R)}\right)^2\right\rangle. \tag{4.3}$$

As is well known the latter gives individually the lowest variance, while the total energy has a lower variance when the Pandharipande-Bethe form is used. Note that the random walk used to estimate the system properties can be the same if the only change is the two-body interacting potential. In our results this is done in those cases where we have equal values for the kinetic energy for both interacting potentials. Also, according to the results of Table I, at least for the energy, finite size effects are not noticeable at the accuracy level and for the number of particles used in our calculations.

Before we compare results obtained with pseudopotentials of different forms in trial functions of pair-product form, let us discuss some results obtained only through the basis set method.[8] From Table II we can see that at the equilibrium density, a basis set with six elements is enough to recover the energy due to two-body correlations for either one of the potentials HFDHE2 or HFD-B3-FCI1. Roughly speaking, as we change to the HFD-B3-FCI1 potential, the total energy per particle decreases by about 0.15 K. That is to say, a decrease practically equal to the deepening of the new potential. This might be seen as a further indication that the major contribution to the binding energy of the helium atoms comes from two-body interactions, and the dimer properties are reflected onto the whole system. The variational principle requires the total energy to be a minimum. However, at the accuracy level with which we perform our calculations, we can see that the kinetic energy depends on the value of the healing distance. Looking at results obtained using a basis set of seven elements for the HFD-B3-FCI1 potential we can see that by increasing the healing distance, the kinetic energy decreases. This change is probably also reflected in other properties of the system as well. If we compare our results with the experimental value of the kinetic energy, displayed in Table IV, the large healing distance gives the best result. Since we are mainly interested in comparing the two potentials using the variational principle we will not pursue this point here. As already mentioned, although this principle requires a minimum for the total energy, it says nothing about other properties. In particular, for a system of helium atoms the binding energy is the result of a delicate balance between kinetic and potential energy with large offsetting changes. Note that for a given number of basis elements, different evaluations of the total energy are in excellent agreement within statistical uncertainties.

TABLE III. Average total and kinetic energies obtained through pair-product wave functions with pseudopotentials (PP) of different forms and for the HFD-B3-FCI1 potential, at the equilibrium density. The PP used were of the McMillan and of the exponential form, first and second rows respectively, and one built using the basis set method, third row. The total number of particles used was 108. The correlations range was half of the simulation cell.

PP	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)
u_M	-5.8512 ± 0.0079	15.1081 ± 0.0072
u_e	-6.0390 ± 0.0092	14.9006 ± 0.0047
BS_6	-6.097 ± 0.011	14.6675 ± 0.0059

In Table III we compare wave functions of the Jastrow form obtained using different pseudopotentials. The results show how important it is to consider a pseudopotential that satisfies a Schrödinger-like equation at small pair separations, Eq. (3.7). The pseudopotential of the exponential form is able to recover about 75% of the additional binding energy obtained when we go from a pseudopotential of the McMillan form to one built using the basis set method.

Results obtained with explicit three-body terms are presented in Table IV. Examining the results for the HFDHE2 potential, we see that a basis set of 6 elements is large enough to give all the binding energy obtained with a wave function with optimal two-body correlation factor and explicit triplet term.[8] This is the same number of elements used in the simple pair-wise trial function, cf. Table II. For the HFD-B3-FCI1 potential this continues to be true. With this last potential, we have used two different forms of the function ξ in the triplet factors. The possibility that an exponential form gives a better result does not materialize. In fact, the result in somewhat worse. An unexpected result is that the triplet term is able to compensate for some of the defi-

TABLE IV. Average total energy $\langle E\rangle/N$ and the kinetic energy $\langle T\rangle/N$ per particle at the given healing distance, displayed in the last column, at the equilibrium density. Results obtained for variational functions (VF) of the Jastrow (J) with explicit three-body terms for the given two-body pseudopotentials and triplets functional form (T stands for a form dependent on a Gaussian and T_e on an exponential). Except for the GFMF results with the HFDHE2 potential, obtained with 64 bodies, all runs were made with 108 particles.

VF	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)	Range (nm)
	HFDHE2		
$J_{BS10}T$	-6.862 ± 0.016[a]	14.868 ± 0.052	$l/2$
$J_{BS6}T$	-6.8594 ± 0.0084	14.0330 ± 0.00862	0.72
GFMC	-7.120 ± 0.024[b]	14.47 ± 0.09[c]	
	HFD-B3-FCI1		
$J_{ue}T_e$	-6.979 ± 0.011	-14.484 ± 0.015	$l/2$
$J_{BS6}T$	-6.9983 ± 0.0049	14.2097 ± 0.0064	0.72
$J_{BS7}T$	-6.9766 ± 0.0079	14.1505 ± 0.0085	$l/2$
$J_{ue}T$	-7.0000 ± 0.0054	14.2185 ± 0.0078	$l/2$
GFMC	-7.292 ± 0.003	14.030 ± 0.021	$l/2$
	Experiment		
	-7.71[d]	14.0 ± 0.3[e]	

[a] Reference 8.

[b] Reference 2.

[c] Reference 21.

[d] Reference 22.

[e] Reference 23, average of results at temperature of 1.3 K.

ciencies of the pseudopotential of the exponential form and produces the same energy obtained with a wave function that employs the basis set method. Recall that for a pair-wise wave function, cf. Table III, the basis set method give results significantly better than those obtained with a pseudopotential of the exponential form.

We present GFMC results also in Table IV. The difference in energy between the variational and these results is 0.26 K for the HFDHE2 potential and about 0.29 K for the HFD-B3-FCI1 potential. These values agrees within their statistical errors. Our GFMC calculation gives an energy that is below the experimental value. On the other hand, the kinetic energy as now calculated and the experimental data are in excellent agreement. This suggests that three-body potential effects may account for the energy difference.

In Table V we present the variational parameters for many of the trial functions used to obtain the results we have discussed. In this same table, we also display the parameters of the variational functions used in the study of the solid phase.

B. The crystalline phase

We have investigated solid ^{4}He at the density $\rho=32.9\,\mathrm{nm}^{-3}$ using the HFD-B3-FCI1 two-body interaction potential. We have started our studies with a trial function of the Jastrow-Nosanow form

$$\Psi_{\rm JN}(R)=\Psi_J(R)\phi(R)=\prod_{i<j} f(r_{ij})\cdot\prod_{i} e^{-(C/2)(\mathbf{r}_i-\mathbf{l}_i)^2}, \tag{4.4}$$

a product of Eq. (3.2) and Eq. (3.11). In Table VI we present results obtained with this trial function. Loosely speaking, the new interacting potential (HFD-B3-FCI1) is responsible for a decrease of 0.20 K in the binding energy. Again, it is remarkable that an analytical two-body correlation factor, with the proper behavior at small distances Eq. (3.7), is able to decrease the binding energy by 0.3 K in comparison to the one with a pseudopotential of the McMillan form.

Before turning our attention to results obtained using trial functions with explicit triplet terms in the solid phase, we consider the basis set method results. It will be interesting to see how the size of the basis in this method changes with density. In Table VII we present results for crystalline ^{4}He at $\rho=32.9\,\mathrm{nm}^{-3}$ obtained with basis of sizes ranging from 6 to 10 elements. As we might expect, more than the six elements

TABLE V. Parameters for the given variational functions (VF) and the HDF-B3-FCI1 interacting potential at the equilibrium density and at a crystalline ^{4}He phase. In almost all runs the healing distance was half of the simulation cell. Only in the liquid phase, when the basis set method was used, it was equal to 0.72 nm. Note that for the $J_{BS6}T$ and $J_{BS9}TN$ trial functions the parameter values occupy more than one row. To avoid round off errors in some cases we display extra digits. The units of the parameters are the following: λ and C are given in nm^{-2}, w in nm.

VF	Variational parameters				
		Liquid phase $\rho=21.8\,\mathrm{nm}^{-3}$			
J_{ue}	$\mathcal{A}=0.0075$	$\mathcal{S}=1.40$			
J_{BS6}	$c_2=0.096$	$c_3=-0.0028$	$c_4=0.0095$	$c_5=-0.0096$	$c_6=-0.0040$
$J_{BS6}T$	$c_2=0.092$	$c_3=-0.0055$	$c_4=0.0088$	c5=−0.0057	$c_6=0.0022$
	$\lambda=-857.0$	$w=0.179$			
$J_{ue}T$	$\mathcal{A}=0.0070$	$\mathcal{S}=1.40$	$\lambda=-689.0$	$w=0.184$	
		Solid phase $\rho=32.9\,\mathrm{nm}^{-3}$			
$J_{ue}N$	$\mathcal{A}=0.0185$	$\mathcal{S}=1.16$	$C=138.0$		
$J_{ue}TN$	$\mathcal{A}=0.0165$	$\mathcal{S}=1.19$	$\lambda=-735.0$	$w=0.166$	$C=125.5$
$J_{BS9}TN$	$c_2=0.040$	$c_3=0.012$	$c_4=-0.012$	$c_5=0.0072$	
	$c_6=0.0045$	$c_7=0.0026$	$c_8=-0.0022$	$c_9=0.0012$	
	$\lambda=-918.0$	$w=0.166$	$C=109.0$		

TABLE VI. Results for solid ^{4}He at $\rho = 32.9\,\mathrm{nm}^{-3}$ using a pairwise trial functions with the given pseudopotentials and localizing factors of the Nosanow form (N). The result for both interacting potentials were obtained using a 108 bodies.

VF	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)
	HFDHE2	
$J_{uM}N$	-3.322 ± 0.019[a]	
	HFD-B3-FCI1	
$J_{uM}N$	-3.522 ± 0.019	31.051 ± 0.016
$J_{ue}N$	-3.840 ± 0.012	30.634 ± 0.012

[a]Reference 13.

used at the equilibrium density, cf. Table II, are needed. In fact, with the density increase the form of the two-body correlation factor becomes more critical and we eventually need a basis with at least nine elements to obtain an optimal two-body correlation factor.

The best variational results obtained for the ^{4}He crystal phase are displayed in Table VIII. They are obtained with variational wave functions that include a two-body correlation factor, explicit triplet terms and a Nosanow factor. It is striking that the new interacting potential used together with the basis set method produces a variational energy that is equal to the experimental value within statistical uncertainties. At these densities the triplet term is no longer able to compensate for imperfections in the two-body pseudopotential of the exponential form as happened in the lower density liquid phase, cf. Table IV.

V. CONCLUSIONS

Considering our results for both the liquid and the solid phases, we conclude that a more complete description of the He-He interaction most probably requires a three-body interaction. At the equilibrium density our best variational result disagrees with the experimental binding energy by about 2% while our solid at a density about $1.18\rho_{\mathrm{melting}}$ is in excellent agreement. Since it is likely that the variational wave function gives a worse description of the ground state at higher densities, we believe the results simply show that three-body interactions become more important as the density increases. If they give a positive contribution to the energy and are neglected, we are left with the false impression that our variational description is getting better as the density increases. The increase in the basis size in the basis set method to get the best functional form of the two-body correlation factor as the density increases is also evidence that the variational function gets relatively worse. On the other hand, if we accept that at high densities the three-body contribution to the potential energy is important, our GFMC calculations show that these interactions are important as well at the equilibrium density. As already mentioned, our GFMC calculation, performed with the HFD-B3-FCI1 potential, gives a total energy below the experimental value by about 2%.

TABLE VII. Variational and kinetic energies per particle for solid ^{4}He at $\rho = 32.9\,\mathrm{nm}^{-3}$. Resulted obtained using the basis set method, with different numbers of elements in the basis, explicit three-body terms and localizing factors of the Nosanow form. Calculations were performed using 108 particles and de HFD-B3-FCI1 potential.

VF	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)
$J_{BS6}TN$	-4.179 ± 0.016	31.805 ± 0.019
$J_{BS7}TN$	-4.209 ± 0.014	31.621 ± 0.022
$J_{BS8}TN$	-4.243 ± 0.016	31.516 ± 0.024
$J_{BS9}TN$	-4.2608 ± 0.0062	31.4406 ± 0.0097
$J_{BS10}TN$	-4.256 ± 0.010	31.412 ± 0.010

TABLE VIII. Results for crystal ^{4}He at $\rho = 32.9\,\mathrm{nm}^{-3}$. The variational functions include a factor of the Jastrow form, explicit three-body terms and one-body terms of the Nosanow form. The number of particles used in the calculations were 108.

VF	$\langle E\rangle/N$ (K)	$\langle T\rangle/N$ (K)
	HFDHE2	
$J_{uM}TN$	-3.786 ± 0.014[a]	
$J_{BS10}TN$	-4.011 ± 0.036[b]	
	HFD-B3-FCI1	
$J_{ue}TN$	-4.2345 ± 0.0082	31.201 ± 0.019
J_{BS9}TN	-4.2608 ± 0.0062	31.4406 ± 0.0097
	Experiment	
	-4.26[c]	34.0 (±0.9)[d]

[a]Reference 24.

[b]Reference 8.

[c]Linear interpolation from data of Ref. 25.

[d]Linear interpolation from data at 1.70 K of Ref. 26. The error is from the published data and is displayed only as an indication.

The importance of nonadditive contributions in systems of helium atoms has been a concern in the literature for a long time. In fact, at very high pressures, they are needed for a good description of this system.[14,15] Although many efforts[2,16,17] concentrated in considering only the long range triple-dipole Axilrod-Teller interaction, exchange effects in the electronic clouds of two atoms in the presence of a third one has also been take into account by the so called Bruch-McGee potential.[18] However, there is a great deal of uncertainty in the parameters values of these potentials and even in their functional form.[15,19] We know that corrections due to interactions of the Axilrod-Teller form are not very sensitive to the exact form of the two-body interacting potential and that they increase the energy, at the equilibrium density by about 2%.[2,16] This could bring our GFMC result in agreement with experiment at this density if we discard the exchange effects. However, due to their attractive character and similar size, these exchange effects can cancel the triple-dipole energy at short range. In other words: a good representation of the many-body components of the interaction potential for a system of helium atoms remains an outstanding problem.

A significant improvement in the description of systems of helium atoms that interact through a potential of the Hartree-Fock dispersion form is obtained by using variational wave functions with two-body correlation factors having a pseudopotential of the exponential form. Its functional form, Eq. (3.7), is simple and it is advantageous to use it not only when the HFD-B3-FCI1 potential is used, but also for any potential of the Hartree-Fock dispersion form. The variational parameter search is easily done and does not require any special attention.

Despite the excellent results obtained with the pseudopotential of the exponential form, it also has limitations. For such a functional form, fixed-phase arguments would predict a triplet term dependent on a function ξ, Eq. (3.8), of exponential rather than of Gaussian form. Our results at best say that both the exponential and the Gaussian forms are equivalent. We interpret this fact as an indication that at intermediate distances important features are still missing in our pseudopotential of the exponential form.

The HFDHE2 potential has served for years as an excellent ''effective'' pair potential. Due to the well depth of this potential, its exclusive pair-wise treatment of the helium atoms in the condensed phase has allowed the prediction of many properties in very good agreement with experiment.[2] A more recent quantum Monte Carlo investigation[19] of this system using the HFD-B(HE) interacting potential proposed by the Aziz group in 1987 (Ref. 4) also points in the direction that many-body effects might be of importance. However, as before the inclusion of three-body contributions to the total energy produces a worse equation of state.

In the past, the very good theoretical results obtained with the HFDHE2 potential have inhibited the adoption of its revised versions. Now, however, this situation seems in need of a change. It has been suggested that the HFD-B3-FCI1 interatomic potential has reached an accuracy where it can be used to calibrate the experimental apparatus used to measure virial coefficients and transport properties.[6] On the other hand, there are already path integral Monte Carlo results[20] for the two-dimensional ^{4}He system obtained with the HFD-B3-FCI1 potential. As is well known[15] this method does not depend on trial functions and so it is easier to move to a new interacting potential.

One of the reasons why variational calculations move more slowly to new interacting potentials might be because new optimizations are need. We hope that this work presenting optimum variational parameters for many useful trial functions, both at the equilibrium density and for a solid at $\rho \approx 1.18\rho_{\mathrm{melt}}$ might overcome this inertia.

The variational calculations of this paper show that three-body interactions must be considered in the He-He potential, since we do not hope to have an exact variational wave function for our system of ^{4}He atoms in the solid phase. We hope this situation will encourage new efforts in understanding the nonaddictive contributions to the potential energy for bulk helium systems.

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