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Solution of an inverse problem: Maximum-overlap Jastrow function of the Lennard-Jones Bose fluid

G. L. Masserini
Dipartimento di Fisica, Università di Milano, I-20133 Milano, Italy

L. Reatto
Dipartimento di Fisica, Università di Parma, I-43100 Parma, Italy
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We determine for the Lennard-Jones Bose fluid the Jastrow function that has the maximum overlap with the exact ground state as determined by the Green's-function Monte Carlo method. We solve the resulting inverse problem with an iterative predictor-corrector method using integral equations and Monte Carlo methods, respectively. The maximum-overlap Jastrow function gives also a good value for the energy.

A common approach to dense quantum fluids like the helium liquids is the variational one based on minimization of the expectation value of the Hamiltonian of the system. Recently, one of us[1] has proposed the use of a different variational principle, one based on the maximization of the overlap integral $\langle\psi_{\rm mod}|\psi_0\rangle$ between the exact ground state ψ_0 and a model function $\psi_{\rm mod}$. The attractive aspect of this method is that one does not have to assume a definite form for the Hamiltonian, but one can get direct information on the ground state from knowledge of correlation functions of the system. Such knowledge at the level of the radial distribution function (RDF) can be obtained from scattering experiments and, in addition, from exact simulation of many-body systems, for example, by the method[2] of Green's function Monte Carlo (GFMC), one can get also the higher distribution functions. Here we present the first application of the maximum-overlap criterion to the case of a Bose fluid.

From variational computation of the energy it is known that the Jastrow function

$$\psi_J(\{\vec{r}_i\}|u)=\prod_{i<j}\exp\{-\tfrac{1}{2}u(r_{ij})\} \tag{1}$$

already represents[2] a rather good approximation to the exact ground state. The maximum of the overlap integral $\langle\psi_J|\psi_0\rangle$ obtains for that pseudopotential $u(r)$, which we call $\bar{u}(r)$, for which the RDF $g_J(r|\bar{u})$ equals[3] the exact RDF $g_0(r)$

$$g_J(r|\bar{u})=g_0(r)\ . \tag{2}$$

This implies a similar equality for the structure factor. In order to solve the functional equation (2) for $\bar{u}$, one could use one of the integral equations, like the hypernetted chain equation (HNC), that relates $g_J(r|u)$ to $u(r)$. However, it is known that in a dense system $g_J(r|u)$ is rather insensitive to the detailed shape of $u(r)$ so that these equations, when solved with respect to $u(r)$, are unstable with respect to the input $g_J(r)$. In this way any inaccuracy of the integral equation would bias the resulting $u(r)$ without control. We do not believe that any of these equations are accurate enough at all distances to be directly useful to the present inverse problem. For this reason we have introduced a new method, a predictor-corrector method.

Our method uses a blend of the integral equation approach and of Monte Carlo (MC) computation. Suppose that for a given pseudopotential $u^{(0)}(r)$ we know $g_J(r|u^{(0)})$. Then we use a suitable integral equation (the predictor) in order to estimate the variation $\delta(r)=\bar{u}(r)-u^{(0)}(r)$ needed in order that $g_J(u^{(0)}+\delta)=g_0$. However, this estimate of δ, which we call $\delta^{(1)}$, is only approximate due to the inaccuracy of the integral equation. Thus we perform an "exact" computation (the corrector) of $g_J(r|u^{(1)})$, where $u^{(1)}=u^{(0)}+\delta^{(1)}$, using simulation method. In general, $g_J(u^{(1)})-g_0$ is nonzero due to the approximate nature of the predictor, but we can use this difference with the predictor to obtain a new estimate $\delta^{(2)}=u-u^{(1)}$. The procedure is repeated until $\Delta g_n=g_J(r|u^{(n)})-g_0(r)$ is less than a given standard of error, and the final estimate of $\bar{u}$ is $\bar{u}=u^{(0)}+\sum_1^n\delta^{(i)}$. We notice that our approach has some general similarity of spirit with a method used by Campbell and Pinski[4] to minimize the energy.

As predictor, we have used two different algorithms. The first[1] is based on the random-phase approximation (RPA) that states that the change $\delta c(r)$ of the Ornstein-Zernike direct correlation function $c(r)$ equals $-\delta(r)$, the variation of the pseudopotential. When used as predictor, RPA gives

$$\delta^{(i)}_{\rm RPA}(r)=\rho^{-1}(2\pi)^{-3}\int d^3k\, e^{i\vec{k}\cdot\vec{r}}\,[\,S_J^{-1}(k|u^{(i-1)})-S_0^{-1}(k)\,]\ , \tag{3}$$

where ρ is the number density, $S_0(k)$ and $S_J(u^{(i-1)})$ are, respectively, the exact structure factor and the one corresponding to $g_J(u^{(i-1)})$. As a second predictor we have used one based on the modified HNC (MHNC) equation[1]

$$\delta^{(i)}_{\rm MHNC}(r)=\delta^{(i)}_{\rm RPA}(r)+\ln[g_J(r|u^{(i-1)})/g_0(r)]+g_J(r|u^{(i-1)})-g_0(r)\ . \tag{4}$$

The MHNC approximation is known to be accurate at short distance, in the region of the core, where $g(r)$ is rapidly vanishing.

Our computation proceeds as follows. We start from an initial guess for $u^{(0)}$ using the result of a variational computation of the energy. Then we perform a series of the predictor-corrector cycle using either the RPA and the MHNC predictor, depending where $\Delta g(r)$ is largest until the differences $\Delta S_i(k)=S_J(k|u^{(i)})-S_0(k)$ and $\Delta g_i(r)$ are below a set level of accuracy for all k and r. The level of accuracy is determined by the statistical noise of the simulation computation. For standard MC and GFMC runs, it is

generally estimated to be of the order of 1%, where $S(k)$ and $g(r)$ are of order unity.

We have studied the Lennard-Jones (LJ) Bose fluid and as input we use $g_0(r)$ obtained from a GFMC computation that is supposedly exact. Our predictors require the structure factor so we need to extend g_0 and g_J at distance $r > L/2$, where L is the side of the simulation cube. For this we use a method found satisfactory with GFMC computations:[5] g_0 and g_J are fitted in the tail region $r \lesssim L/2$ by $\mathrm{Re}[Ar^{-1}\exp(zr)]$ with respect to the complex numbers A and z, and this function is used to represent the RDF for $r > L/2$. The use in our MC of the same number of particles (64) used in the GFMC computation[4] minimizes the errors introduced by the extension.

The first computation has been performed at the equilibrium density $\rho\sigma^3 = 0.3648$. For $u^{(0)}$, we have used a pseudopotential determined in a variational computation[6] of the energy [it corresponds to the parameters $A = 0.3$, $\lambda = 1.5$, $\Lambda = 1$; $C = 0.3$, $d = 1.8$, $D = 0.6$ in Eq. (11) of Ref. 5]. At the sixth iteration, the computation was considered converged since the deviations Δg_6 and ΔS_6 are well below the estimated statistical noise of S_0 and g_0: the maximum of $|\Delta S_6(k)|$ for $k > 2\pi/L$ is below 0.003 and the average of $|\Delta g_6(r)|$ is about 0.003 in the region, where $g_0(r) > 0.5$ and is $\sim 5\%$ in the region closer to the core. In Fig. 1, we show $\Delta S_i(k)$ for some of the iterations together with this difference in the case of the McMillan's pseudopotential $u(r) = (b\sigma/r)^5$ that is widely used as an approximate wave function. Some of the $u^{(i)}(r)$ are plotted in Fig. 2. The most noticeable feature is that $\bar{u}(r)$ has a definite structure at intermediate distances, at least a minimum followed by a maximum. In order to check the convergence of our method, we have repeated[7] the computation starting from a different $u^{(0)}$: the result is the same within the statistical noise [$\delta u(r) \approx 0.03$].

A second computation has been performed at the density $\rho = 1.2\rho_{\mathrm{eq}}(\rho\sigma^3 = 0.4378)$ that is close to solidification of the system. Again we started with a $u^{(0)}$ determined with a variational computation of the energy (the parameters[6] are $A = 0.3$, $\lambda = 1.41$, $\Lambda = 0.94$; $C = 0.3$, $d = 1.694$, $D = 0.565$). After eight iterations, the convergence is as good as at ρ_{eq}, and the initial and the final pseudopotentials are shown in Fig. 3. Also in this case, $\bar{u}(r)$ has a structure and it is even more pronounced than at ρ_{eq}.

The GFMC computation is for a small number of particles so that it produces a ground state without the contribution of the zero-point motion of phonons with $k < \pi/L$. As a consequence, $S_0(k=0)$ is not zero as for a bulk system, and the $\bar{u}(r)$ that we compute does not contain the long-range tail r^{-2} due to those phonons. In order to obtain $\bar{u}$ for a bulk system, we can estimate the missing phonon contribution by

$$\delta_{\mathrm{ph}}(r) = \rho^{-1}(2\pi)^{-3}\int d^3k\, e^{i\vec{k}\cdot\vec{r}}\,[\bar{S}_0^{-1}(k) - S_0^{-1}(k)]\ , \quad (5)$$

where $\bar{S}_0(r)$ is equal to $S_0(k)$ modified at small k so that it

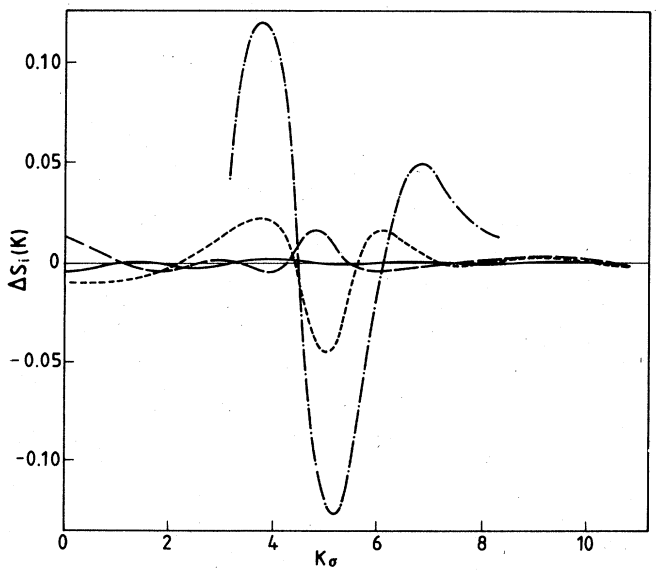

FIG. 1. $\Delta S_i(k) = S_J(k|u^{(i)}) - S_0(k)$ for the LJ Bose fluid at ρ_{eq} for $i=0$ (---), $i=2$ (— — —), $i=6$ (——), and for McMillan's u (— · — ·).

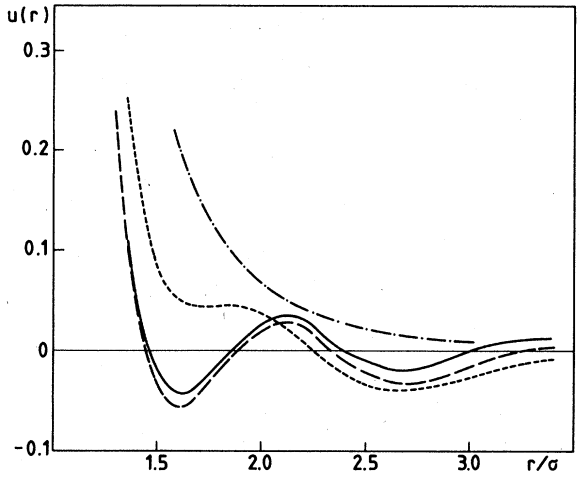

FIG. 2. Pseudopotentials $u^{(i)}$ as in Fig. 1.

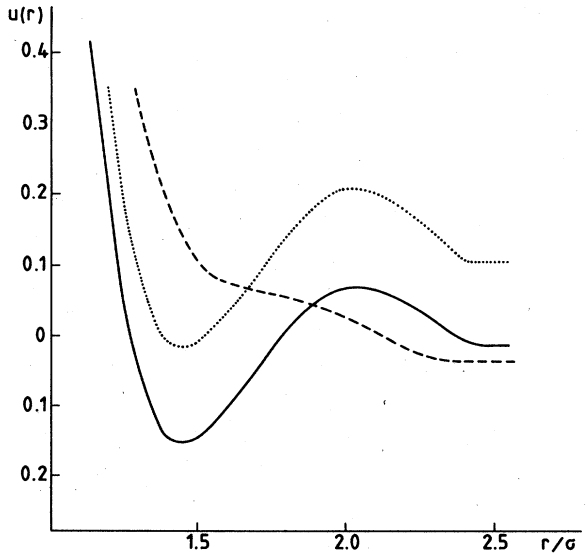

FIG. 3. Pseudopotentials for the LJ Bose fluid at $\rho = 1.2\rho_{\mathrm{eq}}$ for $i=0$ (— — —), $i=8$ (——), and $u^{(8)} + \delta_{\mathrm{ph}}$ (· · · ·).

joins the Feynman's form $2mc/\hbar k$, where c is the sound velocity[5] of this LJ system. Since $S_J(k|\bar{u}) = S_0(k)$, this same correction δ_{ph} applies also to $\bar{u}(r)$ and this is shown in Fig. 4 for ρ_{eq}, and in Fig. 3 for $\rho = 1.2\rho_{eq}$.

The maximum-overlap principle is quite distinct from the energy one so, in general, it should not also give a good value for the energy. However, since both criteria have the exact ground state as absolute extrema if the variational subspace includes the ground state, a good value of the energy for the maximum overlap $\bar{u}$ indicates that the exact ground state is rather close to the subspace of Jastrow functions. We find that this is indeed the case. In fact at ρ_{eq}, we find that $\bar{u}$ gives an expectation value of the energy per particle $E_0 = -5.80$ K. This is quite similar to the value found with the McMillan's pseudopotential ($E_0 = -5.68$ K) and only slightly higher than the best Jastrow result[6] ($E_0 = -6.10$ K). At $\rho = 1.2x\rho_{eq}$, $\bar{u}$ gives $E_0 = -4.61$ K, to be compared with the McMillan's value ($E_0 = -4.5$ K) and with the best Jastrow value[6] $E_0 = -4.91$ K.

The maximum-overlap pseudopotential $\bar{u}(r)$ of the LJ Bose fluid has a structure at an intermediate distance of order of the position of the first minimum and second maximum of $g(r)$. At higher density, the structure of $\bar{u}(r)$ becomes stronger and it is displaced to smaller distance. Comparison of Fig. 2 with Fig. 3 shows that this displacement roughly corresponds to $(\rho_{eq}/\rho)^{1/3} = (1.2)^{-1/3} = 0.94$ and this points to a collective effect as the origin of the structure. Also, energy computations have given pseudopotentials with a rather similar structure. The result[8] of a functional minimization of the energy in HNC approximation is shown in Fig. 4 and our initial $u^{(0)}$, shown in Fig. 2, similarly comes from an energy computation by Monte Carlo.[6] Notice that only in the HNC computation is the correct tail r^{-2} of $u(r)$ taken into account. We conclude that both energy and maximum-overlap computations give a Jastrow function with some structure that is density dependent, and this feature appears to be more pronounced when the maximum-overlap criterion is used. Such structure has been interpreted[6] as a result of zero-point motion of roton excitations.

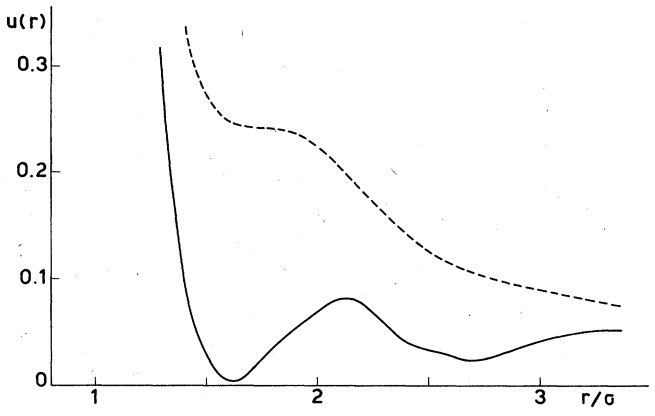

FIG. 4. Pseudopotentials at ρ_{eq}: $u^{(6)} + \delta_{ph}$ (——) and from Ref. 8 (– – –).

In summary we have shown that the maximum-overlap criterion can be used to obtain information on the ground state of a Bose fluid. The maximum-overlap pseudopotential has a structure at intermediate distance and it gives a very good value for the energy. This is another indication of the goodness of the Jastrow function. Our predictor-corrector method is rather efficient in solving an inverse problem, the determination of a distribution function starting from structural data. Using the well-known formal similarity between Jastrow theory and a fluid of classical particles, our method can be used to determine the pair interaction of a classical fluid starting from structural information. Many extensions of the present computation for quantum systems can be envisaged,[1] for instance, the inclusion of triplet terms in the wave function and a study of Slater-Jastrow functions for Fermi particles.

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