QUASIPARTICLE, NATURAL, AND MEAN-FIELD ORBITALS IN DROPS OF LIQUID HELIUM*

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INTRODUCTION

In the last few years we have used the variational Monte Carlo (VMC) technique to find accurate ground-state wave functions for drops of liquid helium containing 8-728 ^4He atoms (Bose statistics) or 40-240 ^3He atoms (Fermi statistics). The wave functions include both two- and three-particle correlations. From comparisons¹ with GFMC results, we believe these wave functions to be among the most accurate for systems of ~ 100 strongly interacting particles.

Having determined the wave functions, we can use them to investigate the properties of the quantum-mechanical ground states of large finite systems. In previous studies we have investigated the accuracy of local-density approximations (LDA) for the pair-correlation function² and have studied the structure functions of the drops and their relationships to two-particle correlations.³ The present report is principally devoted to a recent study of single-particle orbitals in the drops.⁴ I first give a brief outline of the VMC wave functions.

THE VMC WAVE FUNCTIONS

We are interested in the ground-state wave functions for a system of N atoms interacting with pair-wise potentials; the Hamiltonian is

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

where V is the HFDHE2 He-He potential of Aziz.⁵ We use wave functions with two- and three-particle correlations:

$$\Psi_v(\vec{r}_1, \vec{r}_2, \dots, \vec{r}_N) = \Phi(\vec{r}_1, \vec{r}_2, \dots, \vec{r}_N) \prod_{i<j} f_2(r_{ij}) \prod_{i<j<k} f_3(r_{ij}, r_{jk}, r_{ik}) \quad (2)$$

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where

$$\Phi = \prod_i f_1(r_i) , \quad \text{Bose} , \quad (3)$$

$$= \det[f_i(\vec{r}_j^\uparrow)] \times \det[f_i(\vec{r}_k^\downarrow)] , \quad \text{Fermi (spin)} . \quad (4)$$

For the Fermi case the f_i ($1 \leq i \leq N/2$) are the $N/2$ lowest eigenstates of a potential well and j ($1 \leq j \leq N/2$) labels the spin-up particles while k ($N/2+1 \leq k \leq N$) labels the spin-down particles. The $\vec{r}_i^\uparrow$ includes the effects of Feynman-Cohen backflow.

The details of the wave functions may be found in Ref. 1; they contain more than 10 variational parameters. For a given set of parameters we evaluate the energy as

$$E_v = \int d^3r_1 \dots d^3r_N |\Psi_v|^2 \frac{1}{\Psi_v} H \Psi_v / \int d^3r_1 \dots d^3r_N |\Psi_v|^2 \quad (5)$$

The E_v is an upper bound to the true ground state energy. We have varied the parameters in Ψ_v and re-evaluated Eq. (5) until the minimum E_v was found. The $3N$ -dimensional integral in Eq. (5) is done by the Metropolis random walk using $|\Psi_v|^2$ as a weight function. These walks for the final sets of variational parameters have been saved in files and can be used to compute other expectation values such as the density distributions reported here.

DEFINITIONS OF SINGLE-PARTICLE ORBITALS

We have investigated three types of single-particle orbitals: mean field (MF) designated as ϕ , quasiparticle (QP) designated as χ , and natural designated as ψ . I first give a brief description of how each of these were computed.

The MF orbitals, $\phi_i(r)$, are the solutions of a potential well. The well is chosen such that the single-particle density reproduces the density of the correlated ground-state. For the Bose drops there is just one MF orbital and its wave function is just $\sqrt{\rho(r)/N}$. For Fermi systems we consider the shell closures to be the same as for a harmonic oscillator and we must numerically determine the potential well; it contains small wiggles that are necessary to reduce the oscillations in the MF density.

The QP orbitals are defined as the overlap of a correlated wave function for the system with $N-1$ particles with the N -particle ground state:

$$\chi_{n\ell}(r) \propto \int d\vec{r}_2 \dots d^3r_N \Psi_{N-1} \left[(n\ell)^{-1}; \vec{r}_2, \dots, \vec{r}_N \right] \\ \times Y_{\ell m}(\hat{r}) \Psi_N(\vec{r}, \vec{r}_2, \dots, \vec{r}_N) . \quad (6)$$

We form Ψ_{N-1} by using the same two- and three-body correlations as in the N -particle system (the dependence on N is very weak¹) and by removing one atom from Φ . In the case of the Fermi drops we can choose which $f_{n\ell}$ to remove from the Slater determinant; this choice is indicated by the $n\ell$ label!

in ψ_{N-1} . If the $f_{n\ell}$ is near the Fermi surface of the N-atom drop, then we expect that Eq. (9) will be a good (ground state or excited) wave function for the (N-1)-atom system. However, if $n\ell$ represents a several $\hbar\omega$ excitation, then, because of mixing with particle-hole excitations, Eq. (9) will not be a good wave function and we do not report QP orbitals for such cases. We consider only the 1s QP orbital for the Bose drops.

The natural orbitals, $\psi(r)$, are defined as the eigenfunctions of the one-body density matrix:⁸

$$\rho(\vec{r}, \vec{r}') = \sum_i n_i \psi_i^*(\vec{r}) \psi_i(\vec{r}') , \quad (7)$$

and the eigenvalues, n_i , are the corresponding (fractional) occupation numbers. We compute the ψ_i by making a partial-wave expansion and diagonalizing the resulting $\rho_\ell(r, r')$ matrices to get $\psi_{n\ell}(r)$ and the corresponding $n_{n\ell}$.

RESULTS

For the Bose drops we find that there is one s-wave natural orbital whose occupation is a large fraction (37% for N=70) of the drop. We designate this as the 1s orbital and (as will be explained later) refer to it as the "condensate" orbital. All of the remaining orbitals have less than 0.5 (2 ℓ +1) particles each. For the 70-atom drop the total occupation per angular momentum peaks at $\ell=4$ (excluding n_{1s}) and the sum of all $n_{n\ell}$ up to $\ell=10$ is 65, the remaining 5 atoms being in $\ell>10$ states. Thus the strong correlations in liquid helium induce a broad angular-momentum distribution.

Figure 1 shows some of the s-wave natural orbitals for the 70-atom ^4He drop. The dashed curve shows for comparison the MF orbital. The 1s orbital is peaked in the surface and in fact its contribution to the density, $n_{1s} |\psi_{1s}(r)|^2$, accounts for essentially all of the density beyond $r=4.5\sigma$. Because the contribution of a natural orbit to the density is $n_{n\ell} |\psi_{n\ell}(r)|^2$, all the natural orbitals must be confined to within the drop and the higher, less occupied, orbitals tend to peak more and more at small r .

Fig. 1. S-wave natural orbitals of the 70-atom ^4He drop.

Fig. 2. $\tilde{\chi}_{1s}(k)$ (error bars) and $\tilde{\chi}_{1s}^{LDA}(k)$ (solid curve) for the ^4He 240-atom drop.

The fact that in the Bose drops n_{1s} is large and all other $n_{ns} < 1$ can be used to show that $\chi_{1s}(r) \simeq \phi_{1s}(r)$, and that its occupation is also n_{1s} ; i.e. the QP and $1s$ natural orbitals are essentially the same.⁴ To the accuracy of our calculations of these two functions, we cannot distinguish them.

The QP functions are often of experimental interest (for example, they determine stripping-reaction form factors) and it would be useful to have a simple way to approximate them. We have found that the local density approximation (LDA):

$$\chi_{1s}^{LDA}(r) \propto \sqrt{n_0[\rho(r)]} \phi_{1s}(r), \quad (8)$$

where $n_0(\rho)$ is the condensate fraction of the infinite liquid at density ρ , provides a good approximation for the QP orbital in terms of the MF orbital. Furthermore, a simple quadratic expression may be used for $n_0(\rho)$:

$$n_0(\rho) \simeq [1 - 0.68 \rho/\rho_0]^2, \quad (9)$$

where ρ_0 is the saturation density ($0.365 \sigma^{-3}$). The excellent quality of this LDA is shown in Fig. 2 which compares χ_{1s} and χ_{1s}^{LDA} in momentum space (the tilda indicates momentum space). Figure 3 compares Eq. (9) (lower solid line) with HNC calculations⁷ of $n_0(\rho)$. The dashed and dotted curves show the ratio $\chi_{1s}[r(\rho)]/\sqrt{\rho}$ (normalized at one value of ρ) for the $N=20, 70$, and 240 drops and thus are another indication of the accuracy of Eqs. (8) and (9).

I turn now to results for the 70-atom ^3He drop. There are 9 occupied MF orbitals for this drop with the least bound being the $3s$. The natural orbitals corresponding to these MF orbitals have 50-85% occupation with the surface-peaked $3s$ orbital being the most occupied and in total account for 50 of the 70 atoms. The remaining atoms are in orbitals and angular momentum states that are unoccupied in the MF; some 4 atoms have $\ell > 10$. These MF unoccupied orbitals all have natural orbital occupations $n_{n\ell} < 0.1$.

Fig. 3. $\sqrt{n_0(\rho)}$ for liquid ^4He and $\sqrt{Z(\rho)}$ for liquid ^3He .

Figure 4 compares the 1s, 2s and 3s natural (solid curves) and MF orbitals (dashed and dotted curves). The quantities $r^2\phi^2$ and $r^2\phi_n^2$ are plotted. The MF orbitals have the standard structure (0 nodes in 1s, 1 node in 2s, etc.) and their secondary lobes are comparable to the primary peaks. However, all three of the natural orbitals have two nodes each and their secondary lobes are very small. Thus the natural orbitals are highly localized. This appears to be a general result for the fermion natural orbitals; if for a given angular momentum there are I occupied MF orbitals, then the natural orbitals all have $I-1$ nodes and are much more localized than the MF orbitals.

Fig. 4. The 1s, 2s and 3s natural and MF orbitals in the 70-atom ^3He drop.

Fig. 5. Natural (full lines) and localized MF (dashed lines) orbitals in the 70-atom ${}^3\text{He}$ drop.

Because the MF orbitals are fully occupied, any linear combination of the orbitals for a given angular momentum may be used without changing $\rho(r)$. In the spirit of the Wannier functions,⁸ we have constructed the linear combinations of the MF orbitals that maximize the peak heights. The resulting s-wave orbitals are compared with the natural orbitals in Fig. 5; they are quite similar.

The 3s QP orbital is shown (as the error bars) in Fig. 6. It is more localized and surface peaked than the MF orbital (dash-dot). Because there are more than one strongly occupied s-wave natural orbital, the QP and natural orbitals are not necessarily the same; the QP orbital is less localized

Fig. 6. 3s orbitals for 70 ${}^3\text{He}$ atoms.

than the natural orbital (dotted curve). The solid curve is an LDA for $\chi_{3s}(r)$ of the same form as was used in the Bose case:

$$\chi_{n\ell}^{\text{LDA}}(r) \propto \sqrt{Z[\rho(r)]} \phi_{n\ell}(r), \quad (10)$$

where Z is the discontinuity of the occupation number at the Fermi surface for the infinite liquid. Again we use a simple quadratic for $Z(\rho)$:

$$Z(\rho) \simeq [1 - 0.45 \rho/\rho_0]^2. \quad (11)$$

This is compared in Fig. 3, with some calculations⁹ (plus signs) of Z and estimates (circles) assuming that the experimental effective mass¹⁰ is given⁹ by $0.8/Z$. We find that the LDA is a good approximation to the QP orbital for the lowest orbital of each ℓ -value (3s, 2d, 1f, 2p and 1g in the 70-atom case).

Figure 7 shows the difference of the densities of the 70- and 69-atom ³He drops. The quantity $4\pi r^2 \rho(r)$ is plotted; its integral is unity. The error bars are the results of the Monte Carlo calculations and the solid curve is χ_{3s}^{LDA} . The 3s MF (dotted) and natural (dashed) orbitals are also shown; the QP clearly gives a good description of the difference while the MF has excessive contributions at small r . Electron scattering experiments¹¹ on lead and thallium have shown similar results; the MF 3s proton orbital predicts too large a density difference at small r .

MOMENTUM DISTRIBUTIONS IN THE DROPS

To conclude, I briefly consider the momentum distributions, $\tilde{\rho}(k)$, for these drops.⁴ Figure 8 shows the Monte Carlo calculation (error bars) of the momentum distribution of the 240-atom ⁴He drop. The dashed curve is the QP orbital and when this density (weighted by the occupation of the state) is added to the smooth background shown by the dotted curve, an excellent fit (solid curve) to $\tilde{\rho}(k)$ is obtained. Similarly good fits are obtained for all the Bose drops we studied.

Fig. 7. Density difference $\rho_{N=70}(r) - \rho_{N=69}(r)$.

Fig. 8. $\tilde{\rho}(k)$ for the 240-atom ^4He drop.

Figure 9 shows $\tilde{\rho}(k)/N$ of the 20-, 70- and 240-atom ^4He drops. We see two features:

1. The peak at small k gets narrower with increasing N . For this reason we associate the peak (and its representation as χ_{1s} or ϕ_{1s}) with the condensate; the peak will become a delta function in the infinite drop. The "condensate" fractions in the drops (30-50%) are much larger than that of the liquid (10%) because so much of these small drops is in the surface and thus at lower density.

Fig. 9. The $\tilde{\rho}(k)/N$ of $N=20$ (dotted), 70 (dashed), and 240 (dash-dot) ^4He drops, compared with that of the extended liquid from experiment¹² (dash-dash-dot), and variational calculations⁷ (solid).

Fig. 10. $\tilde{\rho}(k)/N$ for the 40- (dotted), 70- (solid) and 112- (dashed) atom ^3He drops. The dash-dot curve is for the infinite liquid (Ref. 9).

2. For large momenta the densities are nearly independent of N and are reasonably given by the density distribution of the infinite liquid at saturation density^{7,12} (solid and dash-dash-dot curves). We can thus represent $\tilde{\rho}(k)$ as the sum of the QP and infinite liquid contributions.

Figure 10 shows $\tilde{\rho}(k)/N$ for the 40-, 70- and 112-atom ^3He drops. We see features similar to those for the Bose case; the small- k behavior has a plateau that falls off at the Fermi momentum of the infinite liquid. This part of $\tilde{\rho}(k)$ is well represented by the contributions of the natural orbitals corresponding to filled MF orbitals. Because we cannot compute the QP orbitals for high excitation, we cannot compare their contribution. At larger momenta the $\rho(k)/N$ are again independent of N and approximately equal to the values for the infinite liquid.⁹

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REFERENCES

1. V. R. Pandharipande, S. C. Pieper and R. B. Wiringa, Phys. Rev. B **34**, 4571 (1986).
2. S. C. Pieper, R. B. Wiringa and V. R. Pandharipande, Phys. Rev. B **32**, 3341 (1985).
3. R. Schiavilla, D. Lewart, V. R. Pandharipande, S. C. Pieper, R. B. Wiringa and S. Fantoni, Nucl. Phys., to be published.
4. D. S. Lewart, V. R. Pandharipande and S. C. Pieper, "Single-Particle Orbitals in Liquid Helium Drops", preprint.
5. R. A. Aziz, V. P. S. Nain, J. S. Carley, W. L. Taylor and G. T. McConville, J. Chem. Phys. **70**, 4430 (1979).
6. P.-O. Löwdin, Phys. Rev. **97**, 1474 (1955).

7. E. Manousakis, V. R. Pandharipande and Q. N. Usmani, Phys. Rev. B **31**, 7022 (1985).
8. E. I. Blount, Advances in Solid State Physics, ed. F. Seitz and D. Turnbull, (Academic Press Inc. 1962) Vol. **13**, p. 305.
9. J. Carlson, R. M. Panoff, K. E. Schmidt, P. A. Whitlock and M. H. Kalos, Phys. Rev. Lett. **55**, 2367 (1985); A. Fabrocini and V. R. Pandharipande, submitted to Phys. Rev. B.
10. D. S. Greywall, Phys. Rev. B **27**, 2747 (1983).
11. J. M. Cavedon, B. Frois, D. Goutte, M. Huet, Ph. Leconte, C. Papanicolas, X.-H. Phan, S. K. Platchkov, S. Williamson, W. Boeglin and I. Sick, Phys. Rev. Lett. **49**, 978 (1982).
12. V. F. Sears, Phys. Rev. B **28**, 5109 (1983); and private communication.

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