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Strong-correlation limit of four electrons in an isotropic harmonic trap

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Asymptotically exact wave functions of the four-electron harmonium atom at the strong-correlation limit are given by Slater determinants multiplied by a Jastrow factor in the form of a single exponential function with the argument bilinear in the interelectron distances. The spin orbitals that enter the determinants set the multiplicities and angular momenta of individual states whereas the Jastrow factor fully accounts for the electron correlation. Asymptotically exact expressions for the corresponding electron densities, which can be computed from such wave functions with the help of the saddle-point approximation, have the form of products of Gaussian functions and polynomials in the Cartesian coordinates.

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I. INTRODUCTION

Confinement of a small number of electrons in an isotropic harmonic potential gives rise to systems of interest to a wide audience of physicists and chemists. In the solid-state setting, it produces quantum dots that, due to their prospective use in electronic and optical devices, have been the subject of recent extensive studies [1]. In the gas phase, it is relevant to description of electromagnetic entrapment of cold plasmas that provides new exciting avenues for both theoretical and experimental research [2]. Interestingly, these two cases are directly related through the phenomenon of Wigner crystallization at the weak-confinement limit [3,4].

Equally important is the application of the three-dimensional species, called harmonium atoms, to calibration of approximate electronic structure formalisms of quantum chemistry. In the past, such calibration has been limited to the two-electron harmonium [5], which offers the distinct advantage of being exactly solvable for infinitely many (though select) magnitudes of the confinement strength [6]. Although recent investigations of the three-electron harmonium [7,8] hold the promise of further advances in this field and numerical simulations provide additional albeit limited data [9,10], the lack of a general ansatz for approximate wave functions of many-electron harmonium atoms remains a major stumbling block.

The first step in the development of such an ansatz is the construction of pertinent wave functions that are asymptotically exact at the strong-correlation limit (which corresponds to a vanishing confinement strength). Describing harmonic oscillations about potential energy minima of the respective Wigner molecules, these wave functions fully incorporate electron correlation effects and thus are highly suitable for being the starting approximation of more accurate approaches.

The aforementioned construction is readily carried out for the two- and three-electron harmonium atoms [6,8], the wave-function normalization and the calculation of the corresponding electron density involving, however, somewhat convoluted algebra in the latter case. In this paper, these developments are extended to the four-electron harmonium atom, which has the property of being the largest system of this kind that can be described in terms of a Jastrow factor [11] in the form of a single exponential function with the argument bilinear in the interelectron distances.

II. THEORY

When expressed in terms of the center-of-mass position R and the interelectron distances $\{r_{pq}\}$, the nonrelativistic Hamiltonian,

$$\hat{H} = -(1/2)\sum_{i=1}^{N} \hat{\nabla}_i^2 + (\omega^2/2)\sum_{i=1}^{N} r_i^2 + \sum_{i>j=1}^{N} r_{ij}^{-1}, \tag{1}$$

of a system of N electrons in a spherical harmonic trap with the confinement strength ω (known as an N-electron harmonium atom [5,8]) becomes

$$\hat{H} = -(1/2)\sum_{i=1}^{N} \hat{\nabla}_i^2 + (N\omega^2/2)R^2 + \hat{V}_{ee}, \tag{2}$$

where

$$\hat{V}_{ee} = \sum_{i>j=1}^{N} [(\omega^2/2N) r_{ij}^2 + r_{ij}^{-1}], \tag{3}$$

allowing for separation of the Kohn modes (described by R) [12] from relative particle motions. Here and in the following, the atomic units ($\hbar = e = m = 1$) are used. For $N < 5$, the totalities of the interelectron distances and the internal degrees of freedom are the same, making $\{r_{pq}\}$ convenient variables for the construction of wave functions with proper permutational symmetries.

Let $r_0 = (N/\omega^2)^{1/3}$ be the value of each interelectron distance for which V_{ee} attains a minimum that for N equal to 3 and 4 corresponds to particles located at the vertices of an equilateral triangle and a regular tetrahedron, respectively. At the strong-correlation limit of $\omega \to 0$, the ground-state bosonic wave functions Ψ_b pertaining to (1) describe harmonic oscillations of particles about these minima, the displacements $\{d_{ij}\}$ equaling $\forall_{i,j}\ d_{ij} = r_{ij} - r_0$. In particular,

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$$\Psi_b \equiv \Psi_b(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4) = N_b \exp(-\omega\lambda_1 R^2)\exp(-\omega\Xi) \quad (4)$$

with

$$\begin{aligned}\Xi = {} & \lambda_2\,(d_{12}+d_{13}+d_{14}+d_{23}+d_{24}+d_{34})^2 \\ & + \lambda_3\,[(d_{23}-d_{14})^2+(d_{24}-d_{13})^2+(d_{34}-d_{12})^2] \\ & + \lambda_4\,[(d_{12}+d_{34}-d_{13}-d_{24})^2+(1/3)(d_{12}+d_{13}+d_{24}+d_{34} \\ & -2d_{14}-2d_{23})^2] \end{aligned} \quad (5)$$

for $N=4$, where the terms with λ_1, λ_2, λ_3, and λ_4 describe, respectively, the Kohn modes, the A_1 breathing mode, the T_2 "squashing" modes, and the E distortion modes with the force constants of $4\omega^2$, $3\omega^2$, $(3/2)\omega^2$, and $(3/4)\omega^2$. A straightforward calculation yields

$$\lambda_1 = 2, \quad \lambda_2 = \frac{\sqrt{3}}{48}, \quad \lambda_3 = \frac{\sqrt{3/2}}{8}, \quad \text{and } \lambda_4 = \frac{\sqrt{3}}{16}. \quad (6)$$

Consequently, at the $\omega \to 0$ limit, the ground-state energy of the four-electron harmonium atom is given asymptotically by the sum of the potential energy at the minimum and the zero-point energy,

$$E = 9\,2^{-2/3}\omega^{2/3} + (1/4)(6+4\sqrt{3}+3\sqrt{6})\omega + \cdots . \quad (7)$$

One should note that, unlike fully Coulombic systems (such as the heliumlike ions that dissociate below certain critical value of the nuclear charge), the harmonium atoms remain bound for arbitrarily small confinement strengths, simply undergoing dilation in accordance with the aforementioned dependence of r_0 on ω.

The asymptotic exactness of the wave function (4) is confirmed by analysis of the local energy,

$$\begin{aligned}\Psi_b^{-1}\hat{H}\Psi_b = {} & E + 3^{1/2}2^{-5/3}\omega^{5/3}\,(d_{12}+d_{13}+d_{14}+d_{23}+d_{24}+d_{34}) \\ & - 3^{1/2}2^{-29/6}\omega^{7/3}\,(4+3\sqrt{2}) \\ & \times(d_{12}^2+d_{13}^2+d_{14}^2+d_{23}^2+d_{24}^2+d_{34}^2) \\ & + 7\,3^{-1/2}2^{-16/3}\omega^{7/3}\,(d_{12}d_{13}+d_{12}d_{14}+d_{12}d_{23} \\ & + d_{12}d_{24}+d_{13}d_{14}+d_{13}d_{23}+d_{13}d_{34}+d_{14}d_{24} \\ & + d_{14}d_{34}+d_{23}d_{24}+d_{23}d_{34}+d_{24}d_{34}) \\ & - 3^{-1}2^{-17/6}\,(5\sqrt{6}-6\sqrt{3})\,\omega^{7/3}\,(d_{14}d_{23}+d_{13}d_{24} \\ & + d_{12}d_{34}) + \cdots , \end{aligned} \quad (8)$$

which is computed by expressing $\{d_{ij}\}$ in terms of $\{\mathbf{r}_i\}$, carrying out the necessary double differentiations, reverting the variables to $\{d_{ij}\}$, and finally expanding the resulting expression around the point $\forall_{i,j}\ d_{ij}=0$. Since the displacements $\{d_{ij}\}$ have average magnitudes proportional to $\omega^{-1/2}$, the local energy differs from E by terms that decay with ω at least as fast as $\omega^{7/6}$.

Calculation of the normalization constant N_b is readily carried out in a rotating-frame coordinate system, in which the position vectors $\{\mathbf{r}_i\}$ are parametrized by the six interelectron distances $(r_{12},r_{13},r_{14},r_{23},r_{24},r_{34})$, the three components (X,Y,Z) of the center-of-mass position vector $\mathbf{R}$, and the three Euler angles $(\theta_1,\theta_2,\theta_3)$ through the relation

$$\mathbf{r}_i = \mathbf{R} + \mathbf{U}(\theta_1,\theta_2,\theta_3)(\tilde{\mathbf{r}}_i - \tilde{\mathbf{r}}_{\text{c.m.}}), \quad i=1,2,3,4, \quad (9)$$

where $\mathbf{U}(\theta_1,\theta_2,\theta_3)$ is the pertinent rotation matrix, the coordinates x_2, x_3, y_3, x_4, y_4, and z_4 that define $\{\tilde{\mathbf{r}}_i\}$,

$$\begin{gathered}\tilde{\mathbf{r}}_1 = (0,0,0), \quad \tilde{\mathbf{r}}_2 = (x_2,0,0), \\ \tilde{\mathbf{r}}_3 = (x_3,y_3,0), \quad \tilde{\mathbf{r}}_4 = (x_4,y_4,z_4), \end{gathered} \quad (10)$$

are explicit functions of $\{r_{ij}\}$, and

$$\tilde{\mathbf{r}}_{\text{c.m.}} = \frac{1}{4}(\tilde{\mathbf{r}}_1+\tilde{\mathbf{r}}_2+\tilde{\mathbf{r}}_3+\tilde{\mathbf{r}}_4). \quad (11)$$

After carrying out the necessary integrations (see the Appendix), one obtains

$$\begin{aligned}N_b &= 2^{3/2}3^{1/4}\pi^{-13/4}\lambda_1^{3/4}\lambda_2^{1/4}\lambda_3^{3/4}\lambda_4^{1/2}\omega^{13/4} \\ &= 2^{-27/8}3^{3/4}\pi^{-13/4}\omega^{13/4}. \end{aligned} \quad (12)$$

Computation of the electron density

$$\rho_b(\mathbf{r}_1) = 4\int |\Psi_b(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)|^2 d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 \quad (13)$$

that corresponds to Ψ_b is much more involved. Thanks to the spherical symmetry of Ψ_b, it can be assumed that $\mathbf{r}_1 = (0,0,r_1)$ without any loss of generality. Since [compare Eq. (9)]

$$\mathbf{R} = \mathbf{r}_1 + \mathbf{U}(\theta_1,\theta_2,\theta_3)\tilde{\mathbf{r}}_{\text{c.m.}} \quad (14)$$

and consequently

$$\begin{aligned} d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 = {} & \frac{r_{12}r_{13}r_{14}r_{23}r_{24}r_{34}}{x_2 y_3 z_4} \\ & \times \sin\theta_2\, dr_{12}dr_{13}dr_{14}dr_{23}dr_{24}dr_{34}\, d\theta_1 d\theta_2 d\theta_3, \end{aligned} \quad (15)$$

one has [compare Eq. (12)]

$$\begin{aligned}\rho_b(\mathbf{r}_1) = {} & 32\left(\frac{3\lambda_1^3\lambda_2\lambda_3^3\lambda_4^2\omega^{13}}{\pi^{13}}\right)^{1/2}\int dr_{12}\int dr_{13}\int dr_{14}\int dr_{23}\int dr_{24}\int dr_{34}\frac{r_{12}r_{13}r_{14}r_{23}r_{24}r_{34}}{x_2 y_3 z_4}\exp(-2\omega\Xi) \\ & \times\int_0^{2\pi} d\theta_1\int_0^{\pi}\sin\theta_2\, d\theta_2\int_0^{2\pi} d\theta_3\, \exp\{-2\lambda_1\omega\,[\mathbf{r}_1+\mathbf{U}(\theta_1,\theta_2,\theta_3)\tilde{\mathbf{r}}_{\text{c.m.}}]^2\}, \end{aligned} \quad (16)$$

which affords

$$\rho_b(\mathbf{r}_1)\big|_{|\mathbf{r}_1|=r_m} = 16\,2^{2/3}\pi^{-3/2}\omega^{11/6}\,(288\lambda_1^{-1}+3\lambda_2^{-1}+36\lambda_3^{-1})^{-1/2} \tag{17}$$

after integration (see the Appendix). Upon insertion of the normalization condition

$$4\pi\int_0^\infty \rho_b(\mathbf{r})r^2dr = 4, \tag{18}$$

Eq. (17) produces

$$\begin{aligned}\eta &= 192\,(96\lambda_1^{-1}+\lambda_2^{-1}+12\lambda_3^{-1})^{-1}\\ &= 2\,(4\,6^{1/2}-5\,3^{1/2}+6\,2^{1/2}-9)\\ &\approx 1.245\,97\end{aligned} \tag{19}$$

for the exponent in the known dependence of $\rho_b(\mathbf{r})$ on r [8],

$$\rho_b(\mathbf{r}) = \rho_b(\mathbf{r}_1)\big|_{|\mathbf{r}_1|=r_m}\exp[-\eta\omega\,(r-r_m)^2]. \tag{20}$$

The actual fermionic wave functions Ψ_f of the four-electron harmonium atom at the strong-correlation limit are readily constructed as products of the aforederived bosonic wave function Ψ_b that plays the role of the Jastrow factor [11] and combinations of Slater determinants ψ chosen to ensure that the resulting Ψ_f are eigenfunctions of the $\hat{\mathbf{S}}^2$, $\hat{\mathbf{S}}_z$, $\hat{\mathbf{L}}^2$, and $\hat{\mathbf{L}}_z$ operators,

$$\begin{aligned}\Psi_f &\equiv \Psi_f(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4)\\ &= (N_f/N_b)\,\psi(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4)\Psi_b(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4).\end{aligned} \tag{21}$$

Here and in the following, $\mathbf{x}$ stands for the combined spatial and spin coordinates $(\mathbf{r},s)$. Thus, for the simplest wave functions describing, respectively, the odd-parity ^5S and the even-parity ^3P states one has

$$\psi_q(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4) = \alpha(s_1)\alpha(s_2)\alpha(s_3)\alpha(s_4)\begin{vmatrix}1 & x_1 & y_1 & z_1\\ 1 & x_2 & y_2 & z_2\\ 1 & x_3 & y_3 & z_3\\ 1 & x_4 & y_4 & z_4\end{vmatrix}, \tag{22}$$

with

$$N_q = \frac{8}{\sqrt{3}}\,r_0^{-3}N_b = 2^{-19/8}3^{1/4}\pi^{-13/4}\omega^{21/4} \tag{23}$$

and

$$\psi_t(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4) = \begin{vmatrix}\alpha(s_1) & \beta(s_1) & x_1\alpha(s_1) & y_1\alpha(s_1)\\ \alpha(s_2) & \beta(s_2) & x_2\alpha(s_2) & y_2\alpha(s_2)\\ \alpha(s_3) & \beta(s_3) & x_3\alpha(s_3) & y_3\alpha(s_3)\\ \alpha(s_4) & \beta(s_4) & x_4\alpha(s_4) & y_4\alpha(s_4)\end{vmatrix}, \tag{24}$$

with

$$N_t = 2\sqrt{\frac{2}{3}}\,r_0^{-2}N_b = 2^{-77/24}3^{1/4}\pi^{-13/4}\omega^{55/12}. \tag{25}$$

On the other hand, the simplest wave function of the even-parity ^1S state requires a combination of three determinants,

$$\begin{aligned}\psi_s(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4) = &\begin{vmatrix}\alpha(s_1) & \beta(s_1) & x_1\alpha(s_1) & x_1\beta(s_1)\\ \alpha(s_2) & \beta(s_2) & x_2\alpha(s_2) & x_2\beta(s_2)\\ \alpha(s_3) & \beta(s_3) & x_3\alpha(s_3) & x_3\beta(s_3)\\ \alpha(s_4) & \beta(s_4) & x_4\alpha(s_4) & x_4\beta(s_4)\end{vmatrix}\\ &+\begin{vmatrix}\alpha(s_1) & \beta(s_1) & y_1\alpha(s_1) & y_1\beta(s_1)\\ \alpha(s_2) & \beta(s_2) & y_2\alpha(s_2) & y_2\beta(s_2)\\ \alpha(s_3) & \beta(s_3) & y_3\alpha(s_3) & y_3\beta(s_3)\\ \alpha(s_4) & \beta(s_4) & y_4\alpha(s_4) & y_4\beta(s_4)\end{vmatrix}\\ &+\begin{vmatrix}\alpha(s_1) & \beta(s_1) & z_1\alpha(s_1) & z_1\beta(s_1)\\ \alpha(s_2) & \beta(s_2) & z_2\alpha(s_2) & z_2\beta(s_2)\\ \alpha(s_3) & \beta(s_3) & z_3\alpha(s_3) & z_3\beta(s_3)\\ \alpha(s_4) & \beta(s_4) & z_4\alpha(s_4) & z_4\beta(s_4)\end{vmatrix},\end{aligned} \tag{26}$$

with

$$N_s = \frac{2\sqrt{2}}{3}\,r_0^{-2}N_b = 2^{-77/24}3^{-1/4}\pi^{-13/4}\omega^{55/12}. \tag{27}$$

Examination of the local energies pertaining to these three wave functions confirms the expected degeneracy of the fermionic energy levels with their bosonic counterpart at the $\omega \to 0$ limit. Wave functions of states with other multiplicities and angular momenta can be obtained in an analogous fashion.

Computation of electron densities corresponding to the fermionic wave functions,

$$\begin{aligned}\rho_f(\mathbf{x}_1) = 4\left(\frac{N_f}{N_b}\right)^2\int |\psi(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4)|^2\,|\Psi_b(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)|^2\\ \times d\mathbf{x}_2d\mathbf{x}_3d\mathbf{x}_4,\end{aligned} \tag{28}$$

proceeds in a straightforward manner. As in the case of the normalization constants, the sharp-peaking property of Ψ_b allows for evaluation of the necessary integrals, producing

$$\rho_q^\alpha(\mathbf{r}_1) = \frac{1}{4}\left(1+3\frac{r_1^2}{r_m^2}\right)\rho_b(\mathbf{r}_1) \quad \text{and} \quad \rho_q^\beta(\mathbf{r}_1) = 0 \tag{29}$$

for the odd-parity ^5S state,

$$\rho_t^{\alpha}(\mathbf{r}_1)=\frac{1}{4}\left(1+3\,\frac{x_1^2+y_1^2}{r_m^2}\right)\rho_b(\mathbf{r}_1)\quad\text{and}\quad\rho_t^{\beta}(\mathbf{r}_1)=\frac{1}{4}\rho_b(\mathbf{r}_1)\tag{30}$$

for the even-parity ${}^3P$ state, and

$$\rho_s^{\alpha}(\mathbf{r}_1)=\rho_s^{\beta}(\mathbf{r}_1)=\frac{1}{4}\left(1+\frac{r_1^2}{r_m^2}\right)\rho_b(\mathbf{r}_1)\tag{31}$$

for the even-parity ${}^1S$ state.

III. DISCUSSION AND CONCLUSIONS

Asymptotically exact wave functions of the four-electron harmonium atom at the strong-correlation limit are given by Slater determinants multiplied by a Jastrow factor in the form of a single exponential function with the argument bilinear in the interelectron distances. The spin orbitals that enter the determinants set the multiplicities and angular momenta of individual states (which are all degenerate with the bosonic state at the $\omega\to 0$ limit) whereas the Jastrow factor fully accounts for the electron correlation. Asymptotically exact expressions for the corresponding electron densities, which can be computed from such wave functions with the help of the saddle-point approximation, have the form of products of Gaussian functions and polynomials in the Cartesian coordinates.

Following the previous studies of analogous two-electron [5] and three-electron systems [7], in which the expressions valid for $\omega\to 0$ were employed in the construction of approximate but more general wave functions, the formulas derived in this paper can serve as the starting point in the development of accurate methods for the description of electronic structure of the harmonium atom with four electrons at all confinement strengths. One possible avenue in this direction is offered by a variational determination of the pertinent spin orbitals. The present work provides algebraic techniques necessary for computation of integrals bound to arise in such an approach.

Finally, it should be pointed out that harmonium atoms with more than four electrons will require more sophisticated treatment as in their case the number of the interelectron distances does not match that of the degrees of freedom, making the construction of the Jastrow factor with a proper symmetry much more involved, and thus significantly increasing the level of complexity of the subsequent wave-function normalization and the calculation of the electron density.

APPENDIX

In order to compute the normalization constant N_b with the help of the coordinate system defined by Eqs. (9)–(11), one begins with the observation that since

$$d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 = \frac{r_{12}r_{13}r_{14}r_{23}r_{24}r_{34}}{x_2 y_3 z_4}\times\sin\theta_2\, dr_{12}dr_{13}dr_{14}dr_{23}dr_{24}dr_{34}\, d\theta_1 d\theta_2 d\theta_3\, d\mathbf{R},\tag{A1}$$

the normalization constant satisfies the equation

$$N_b^2\int dr_{12}\int dr_{13}\int dr_{14}\int dr_{23}\int dr_{24}\int dr_{34}\int_0^{2\pi} d\theta_1\int_0^{\pi}\sin\theta_2\, d\theta_2\int_0^{2\pi} d\theta_3\int d\mathbf{R}\,\frac{r_{12}r_{13}r_{14}r_{23}r_{24}r_{34}}{x_2y_3z_4}\exp(-2\omega\lambda_1R^2)\exp(-2\omega\Xi) = 1.\tag{A2}$$

Although the integration limits for the interelectron distances in Eq. (A2) are such that $\{r_{ij}\}$ satisfy the triangle inequalities,

$$\forall_{i,j,k}\,|r_{ij}-r_{ik}|\le r_{jk}\le r_{ij}+r_{ik},\tag{A3}$$

they can be replaced by the intervals of $(-\infty,\infty)$ at the limit of $\omega\to 0$. At the same time, x_2, y_3, and z_4 can be set to r_0, $\frac{3^{1/2}}{2}r_0$, and $(\frac{2}{3})^{1/2}r_0$, respectively [these values follow from the definition (10), upon setting all interelectron distances to r_0], without any loss of asymptotic exactness. These two simplifications, which are possible due to the fact that Ψ_b peaks sharply at the interelectron distances equal to r_0, allow rewriting Eq. (A2) as

$$16N_b^2\left(\frac{\pi^7}{\lambda_1^3\omega^7}\right)^{1/2}\int_{-\infty}^{\infty}dr_{12}\int_{-\infty}^{\infty}dr_{13}\int_{-\infty}^{\infty}dr_{14}\times\int_{-\infty}^{\infty}dr_{23}\int_{-\infty}^{\infty}dr_{24}\int_{-\infty}^{\infty}dr_{34}\exp(-2\omega\Xi)=1,\tag{A4}$$

after integration over θ_1, θ_2, θ_3, and $\mathbf{R}$ (note the relationships between r_0 and ω, and between $\{r_{ij}\}$ and $\{d_{ij}\}$), which finally yields

$$N_b = 2^{3/2}3^{1/4}\pi^{-13/4}\lambda_1^{3/4}\lambda_2^{1/4}\lambda_3^{3/4}\lambda_4^{1/2}\omega^{13/4} = 2^{-27/8}3^{3/4}\pi^{-13/4}\omega^{13/4}.\tag{A5}$$

The evaluation of the integral (16) begins with angular integration, which produces

$$\rho_b(\mathbf{r}_1) = 128 \left(\frac{3\lambda_1\lambda_2\lambda_3^3\lambda_4^2\omega^{11}}{\pi^9} \right)^{1/2} r_1^{-1} \int dr_{12} \int dr_{13} \int dr_{14} \int dr_{23} \int dr_{24} \int dr_{34} \frac{r_{12}r_{13}r_{14}r_{23}r_{24}r_{34}}{x_2 y_3 z_4} \times (3r_{12}^2 + 3r_{13}^2 + 3r_{14}^2 - r_{23}^2 - r_{24}^2 - r_{34}^2)^{-1/2} \{\exp[\omega\Phi(r_1)] - \exp[\omega\Phi(-r_1)]\}. \tag{A6}$$

The quantity $\Phi(r_1)$ that enters Eq. (A6),

$$\Phi(r_1) = -2\lambda_1 r_1^2 - (\lambda_1/8)(3r_{12}^2 + 3r_{13}^2 + 3r_{14}^2 - r_{23}^2 - r_{24}^2 - r_{34}^2) - 2\Xi + \lambda_1 r_1 \sqrt{3r_{12}^2 + 3r_{13}^2 + 3r_{14}^2 - r_{23}^2 - r_{24}^2 - r_{34}^2} \tag{A7}$$

possesses a maximum at $r_{12}=r_{13}=r_{14}=r_{23}=r_{24}=r_{34}=r_0$ [note that the conditions (A3) are satisfied] and $r_1=r_m$, where $r_m=\frac{\sqrt{6}}{4}r_0$, giving rise to the expansion

$$\begin{aligned}\Phi(r_1) \approx & -\frac{1}{48}(9\lambda_1 + 96\lambda_2 + 96\lambda_3 + 128\lambda_4)(d_{12}^2 + d_{13}^2 + d_{14}^2) - \frac{1}{48}(\lambda_1 + 96\lambda_2 + 96\lambda_3 + 128\lambda_4)(d_{23}^2 + d_{24}^2 + d_{34}^2) \\ & + \frac{1}{24}(-9\lambda_1 - 96\lambda_2 + 64\lambda_4)(d_{12}d_{13} + d_{12}d_{14} + d_{13}d_{14}) \\ & + \frac{1}{24}(3\lambda_1 - 96\lambda_2 + 64\lambda_4)(d_{12}d_{23} + d_{12}d_{24} + d_{13}d_{23} + d_{13}d_{34} + d_{14}d_{24} + d_{14}d_{34}) \\ & + \frac{1}{24}(3\lambda_1 - 96\lambda_2 + 96\lambda_3 - 128\lambda_4)(d_{12}d_{34} + d_{13}d_{24} + d_{14}d_{23}) \\ & - \frac{1}{24}(\lambda_1 + 96\lambda_2 - 64\lambda_4)(d_{23}d_{24} + d_{23}d_{34} + d_{24}d_{34}) - 2\lambda_1(r_1 - r_m)^2 + \sqrt{\frac{3}{2}}\lambda_1(r_1 - r_m)(d_{12} + d_{13} + d_{14}) \\ & - \frac{1}{\sqrt{6}}\lambda_1(r_1 - r_m)(d_{23} + d_{24} + d_{34}) + \cdots. \end{aligned} \tag{A8}$$

Application of this expansion in conjunction with the saddle-point approximation allows for integration over the remaining variables, affording

$$\rho_b(\mathbf{r}_1)\big|_{|\mathbf{r}_1|=r_m} = 16\, 2^{2/3}\pi^{-3/2}\omega^{11/6}(288\lambda_1^{-1} + 3\lambda_2^{-1} + 36\lambda_3^{-1})^{-1/2}. \tag{A9}$$

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