Variational Study of Fermionic Helium Dimer and Trimer in Two Dimensions [#]

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In variational calculation, we have obtained the binding energy of helium 3 dimer in two dimensions. The existence of one bound state, with the binding energy –0.014 mK, has been definitively found. Also, the existence of a binding state of helium 3 trimer having spin –1/2 with the energy below –0.0057 mK is indicated. This re-opens the question of the existence of the gas phase of many helium 3 atoms on the surface of superfluid helium 4.

Key words: helium dimer, helium trimer, binding energy of helium 3 dimer.

INTRODUCTION

About thirty years ago, it was demonstrated[1] that, in dilute bulk ^{3}He–^{4}He solution, atoms of ^{3}He prefer to float on the surface of the ^{4}He rather than to be dissolved in the bulk. All atoms in the solution are pulled down by gravity. A ^{3}He atom is less massive than a ^{4}He atom and therefore its zero point motion energy is greater than that of ^{4}He (approximately by a factor 1.3). Due to this motion, it tends to have no ^{4}He nearby. This tendency leads it to sit on the surface of the ^{4}He, where it has an empty space above. Thus, a ^{3}He atom at low temperatures (below 0.1 K), on the surface of bulk liquid ^{4}He behaves as a spin –1/2 Fermi particle in two dimensions.

[#] Dedicated to Academician Krunoslav Ljolje on the occasion of his 70th birthday.

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In our recent papers,[2–4] we have considered binding of helium diatomic molecules in confined and unconfined geometries. It has been shown that in infinite space helium fermionic dimer exists only in two dimensions. In confined geometry, two helium atoms were studied in 2 and 3 dimensions. Motion of atoms has been confined by spherically external holding potentials.[2] Using a similar procedure, diatomic helium molecules have been studied in the external holding potential that depends on one coordinate as well.[4] All the considered systems might be thought as models for the interactions between helium atoms in a specific real physical environment. For example, in solid matrices, where helium dimers form the condensation seed for helium clusters, in nanotubes, with a diameter between 10 and 100 Å, and in »condensation« on a solid or liquid substrate.

We are not convinced that the atoms of ^{3}He form a gas on the surface. This doubt is based on the fact that there is one bound state of two ^{3}He atoms in 2 D space with the binding energy of about –0.02 mK.[2] This result was achieved after numerical solving of the Schrödinger equation. Of course, a variational calculation is desired as well. No successful variational calculation showing the binding of helium 3 dimer in 2 D has been done so far.

The first goal of this paper is to derive a trial radial wave function and perform variational calculation in finding the binding energy of fermionic helium 3 dimer in 2 D and the mean value of the internuclear distance. The second goal is to examine the possibility of the existence of helium 3 trimer with spin –1/2.

DERIVATION OF THE TRIAL WAVE FUNCTION

Very good trial wave functions describing the ground state of helium 4 dimer and the molecule consisting of one atom of helium 4 and one atom of helium 3 were obtained and used in Ref. 2. They describe a short-range correlation between two atoms, like in the Jastrow wave function for liquid helium state. Long-range correlations are described by decreasing the exponential function. Comparing our results with the numerical solution of the Schröedinger eq., we found that the best form was a product of the functions, which describe short and long range correlations divided by the square root of the distance:

$$\psi = \frac{1}{\sqrt{r}} \exp\left[-\left(\frac{a}{r}\right)^{\gamma} - sr\right] , \tag{1}$$

where a, γ and s are variational parameters.

Our experience showed that this function, although very good for helium 4 dimer and ^{4}He–^{3}He molecule, was not good enough to give the bound state of the fermionic helium dimer. This dimer is very large (the largest molecule we know) and the behaviour of the wave function in-between short and long range is very important. Using Gnuplot graphics and data from the numerical solution of the Schrödinger equation, we were able to construct the following trial wave function.

$$\psi(r) = \frac{1}{\sqrt{r}} f(r) = \frac{1}{\sqrt{r}} \sum_{i=1}^{6} f_i(r) \ , \tag{2}$$

where

$$f_1(r) = a_1 \exp\left[-\left(\frac{a_2}{r}\right)^{a_3}\right] \ , \quad r \in [r_1, r_2]$$

$$f_2(r) = b_1 (\ln(r - b_2))^{b_3} \ , \quad r \in [r_2, r_3]$$

$$f_3(r) = c_1 \exp\left[-\left(\frac{r_4 - r}{c_2}\right)^{c_3}\right] \ , \quad r \in [r_3, r_4]$$

$$f_4(r) = d_1 \exp\left[-\left(\frac{r - r_4}{d_2}\right)^{d_3}\right] \ , \quad r \in [r_4, r_5]$$

$$f_5(r) = e_1 \exp\left[-\left(\frac{r - r_4}{e_2}\right)^{e_3}\right] \ , \quad r \in [r_5, r_6]$$

$$f_6 = g_1 \exp(-sr) \ , \quad r \in [r_6, \infty]$$

and $r_1 = 1$ Å, $r_2 = 2.97$ Å, $r_3 = 34.57$ Å, $r_4 = 165.1$ Å, $r_5 = 228.5$ Å, $r_6 = 2000$ Å. It has 17 parameters $a_1, a_2, a_3, b_1, b_2, b_3, c_1, c_2, c_3, d_1, d_2, d_3, e_1, e_2, e_3, g_1, s$ and 8 of them are independent. Namely, using the continuity of the wave function and the first derivative in points r_2, to r_6, one finds the following nine equations among the parameters; there would be ten, but our wave function has its maximum at point $r = r_4$ and the equation which demands continuity of the first derivative disappears (with the constraint that $d_2 > 1$):

$$a_1 \exp\left[-\left(\frac{a_2}{r_2}\right)^{a_3}\right] = b_1 \left(\ln(r_2 - b_2)\right)^{b_3} \quad , \tag{3}$$

$$\left(\frac{a_2}{r_2}\right)^{b_3} \frac{a_2}{r_2} = \frac{b_3}{r_2 - b_2} \frac{1}{\ln(r_2 - b_2)} \quad , \tag{4}$$

$$b_1 \left(\ln(r_3 - b_2)\right)^{b_3} = c_1 \exp\left[-\left(\frac{r_4 - r_3}{c_2}\right)^{c_3}\right] \quad , \tag{5}$$

$$\frac{b_3}{r_2 - b_2} \frac{1}{\ln(r_3 - b_2)} = \frac{c_3}{c_2}\left(\frac{r_4 - r_3}{c_2}\right)^{c_3 - 1} \quad , \tag{6}$$

$$c_1 = d_1 \quad , \tag{7}$$

$$d_1 \exp\left[-\left(\frac{r_5 - r_4}{d_2}\right)^{d_3}\right] = e_1 \exp\left[-\left(\frac{r_5 - r_4}{e_2}\right)^{e_3}\right] \quad , \tag{8}$$

$$\frac{d_3}{d_2}\left(\frac{r_5 - r_4}{d_2}\right)^{d_3 - 1} = \frac{e_3}{e_2}\left(\frac{r_5 - r_4}{e_2}\right)^{e_3 - 1} \quad , \tag{9}$$

$$e_1 \exp\left[-\left(\frac{r_6 - r_4}{e_2}\right)^{e_3}\right] = g_1 \exp(-s r_6) \quad , \tag{10}$$

$$\frac{e_3}{e_2}\left(\frac{r_6 - r_4}{e_2}\right)^{e_3 - 1} = s \quad . \tag{11}$$

We chose coefficients a_2, a_3, b_2, c_1, c_3, d_2, d_3 and s as variational parameters, and for the others, using relations (3)–(11), we obtained the following expressions:

$$d_1 = c_1 \quad , \tag{12}$$

$$b_3 = \left(\frac{a_2}{r_2}\right)^{a_3} \frac{a_3}{r_2} (r_2 - b_2)\ln(r_2 - b_2) \; , \tag{13}$$

$$c_2 = \left[(r_3 - b_3)\ln(r_3 - b_2)\frac{c_3}{b_3}(r_4 - r_3)^{c_3-1} \right]^{1/c_3} \; , \tag{14}$$

$$b_1 = c_1 \frac{\exp\left[-\left(\dfrac{r_4 - r_3}{c_2}\right)^{c_3}\right]}{(\ln(r_3 - b_2))^{b_3}} \; , \tag{15}$$

$$a_1 = b_1 \frac{(\ln(r_2 - b_2))^{b_3}}{\exp\left[-\left(\dfrac{a^2}{r_2}\right)^{a_3}\right]} \; , \tag{16}$$

$$e_3 = \frac{\ln[s(r_6 - r_4)]\left(\dfrac{d_2}{r_5 - r_4}\right)^{d_3}\dfrac{1}{d_3}}{\ln\left(\dfrac{r_6 - r_4}{r_5 - r_4}\right)}, \tag{17}$$

$$e_2 = \left(\frac{e_3}{s}\right)^{1/e_3}(r_6 - r_4)^{1-1/e_3} \; , \tag{18}$$

$$e_1 = d_1 \exp\left[\left(\frac{r_5 - r_4}{e_2}\right)^{e_3} - \left(\frac{r_5 - r_4}{d_2}\right)^{d_3}\right] , \tag{19}$$

$$g_1 = e_1 e^{sr_6} \exp\left[-\left(\frac{r_6 - r_4}{e_2}\right)^{e_3}\right] . \tag{20}$$

The coefficients are given in the order in which they are calculated.

VARIATIONAL CALCULATION OF THE DIMER

Having derived the trial wave function we performed the variational calculation

$$E \leq \frac{\int \psi^* \hat{H} \psi \, r \, dr}{\int \psi^* \psi \, r \, dr} \,, \tag{21}$$

where

$$\hat{H} = -\frac{\hbar^2}{2\mu} \Delta + \hat{V}\left(|\vec{r}_1 - \vec{r}_2|\right) \,, \tag{22}$$

$\mu = m/2$ is the reduced mass of ^{3}He, $m = 5.00649231 \times 10^{-27}$ kg and $r = |\vec{r}_1 - \vec{r}_2|$. Then, the expression for the energy can be written in the form

$$E \leq \frac{\int_0^\infty r\,dr\left(V(r)\psi^2(r) + \frac{\hbar^2}{2\mu}(\nabla\psi(r))^2\right)}{\int_0^\infty r\,dr\,\psi^2(r)} \,. \tag{23}$$

For the interatomic potential, we used *ab initio* the SAPT potential by Korona *et al.*[6] After adding the retardation effects (SAPT1 and SAPT2 versions), Janzen and Aziz[13] showed that SAPT potential recovers the known bulk and scattering data for helium more accurately than all the other existing potentials. To calculate the integrals, we used the Romberg extrapolation method[5] and by a minimization procedure obtained the binding energy of -0.014 mK. Values of variational parameters for this energy are: $a_2 = 2.873$ Å, $a_3 = 3.698$, $b_2 = 1.55$ Å, $c_3 = 5.9$, $d_2 = 573$ Å, $d_3 = 2.0$, $s = 0.0009318$ Å^{-1}. The value of parameter c_1 does not affect the binding energy, but only the normalization integral; in our calculation it has the value $c_1 = 0.03588$. We also used the boundary points r_3, r_4, r_5 and r_6 as variational parameters. Their final values are $r_3 = 19.5$ Å, $r_4 = 199$ Å, $r_5 = 282$ Å and $r_6 = 1200$ Å. Other parameters, when calculated from expressions (12)–(20) are: $a_1 = 0.01988$, $b_1 = 0.01456$, $b_3 = 0.547$, $c_2 = 217.6$ Å, $d_1 = 0.03588$, $e_1 = 0.03634$, $e_2 = 1262.9$ Å, $e_3 = 3.634$ and $g_1 = 0.05257$.

In the limit $r \to \infty$, the wave function has the asymptotic form $e^{-s_0 r}$, where s_0 is determined by relation $s_0 = \sqrt{\frac{2\mu}{\hbar^2}(-\varepsilon)}$. The value of s_0 coincides with the value of s, which confirms the correct asymptotic behaviour of the wave function Ψ.

The function $f(r)$ and its first derivative are shown in Figure 1.
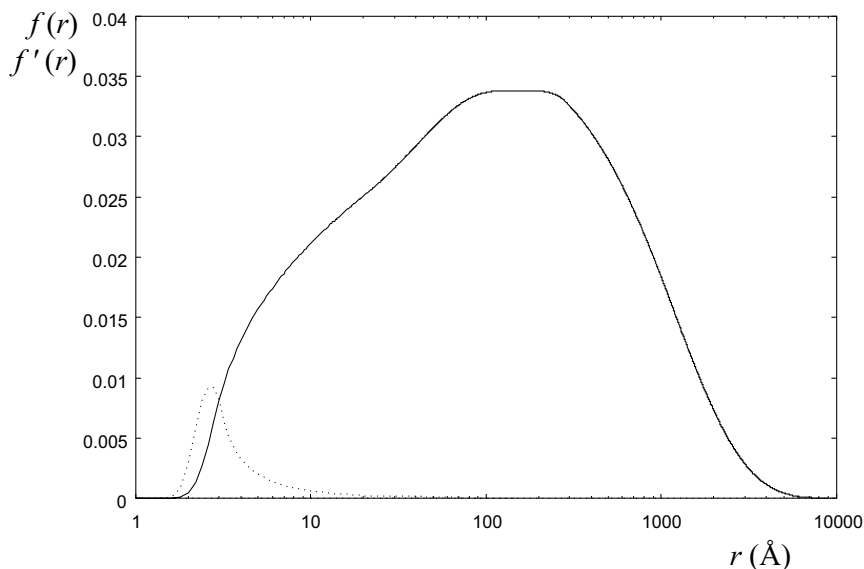

Figure 1. The figure shows the radial wave function $f(r)$ (solid line) and the first derivative $f'(r)$ (dashed line) of helium 3 in 2 D for the parameters determined by minimization of the energy.

We also calculated the mean value of internuclear distance $<r>$ and the root-mean-square (rms) deviation Δr for the $(^3\text{He})_2$.

$$<r>=\frac{\int r\psi^2(r)r\mathrm{d}r}{\int \psi^2(r)r\mathrm{d}r} \quad , \tag{24}$$

$$<r^2>=\frac{\int r^2\psi^2(r)r\mathrm{d}r}{\int \psi^2(r)r\mathrm{d}r} \quad , \tag{25}$$

and

$$\Delta(r)=\sqrt{<r^2>-<r>^2} \quad . \tag{26}$$

The obtained values of $<r> = 651$ Å and $\Delta r = 562$ Å show that $(^3\text{He})_2$ is a really huge molecule. Our results for the energy and average radius $<r>$ confirm the results of the numerical calculations from paper.[2] Since the

value of our binding energy is a bit higher than the one in Ref. 2, which is to be expected from a variational calculation, we also obtained a higher value of the average radius.

This small energy requires a high numerical precision. To verify our numerical procedure, we repeated the whole calculation, with a slightly redefined wave function and using an equivalent but different expression for the energy. Namely, the function $f_3(r)$ now reads,

$$f_3(r) = c_1 \exp\left[-\left(\frac{|r_4 - r|}{c_2}\right)^{c_3}\right] , \qquad r \in [r_3, r_4 + \delta] , \tag{27}$$

and the function $f_4(r)$ is defined for $r \in [r_4 + \delta, r_5]$ where $\delta = 1.1$ Å. From the condition that the function and the first derivative are continuous in $r_4 + \delta$, two relations for parameters d_1 and d_2 are obtained,

$$d_2 = \left(\frac{d_3}{c_3}\right)^{1/d_3} c_2^{c_3/d_3} \delta^{1 - 1/e_3} , \tag{28}$$

$$d_1 = c_1 \left[\left(\frac{\delta}{d_2}\right)^{d_3} - \left(\frac{\delta}{c_2}\right)^{c_3}\right] . \tag{29}$$

The relations for other parameters (13)–(20) are left unchanged. With the wave function defined in this way, no singularities in the second derivative of function Ψ are expected, and therefore the variational calculation can be performed using relation (23) as well as the following relation for the energy

$$E \leq \frac{\int_0^\infty r \mathrm{d}r \left(V(r)\psi^2(r) + \frac{\hbar}{2\mu}\psi(r)\Delta\psi(r)\right)}{\int_0^\infty r \mathrm{d}r \psi^2(r)} , \tag{30}$$

where the kinetic energy is expressed through the Laplace operator, Δ. The minimization of energy in both cases gave the same value of -0.014 mK, which is the same as the one obtained using the function where there is no displacement δ from the maximum in r_4. Thus, we can be certain of the applied numerical procedures.

CALCULATION OF TRIMER WITH SPIN –1/2

In 1979, Cabral and Bruch[7] considered the binding of $^3He_2$ and $^3He_3$. They performed a variational calculation, with the interatomic potentials available at the time, and concluded that both molecules are probably not bound in 2 D. Our results for the dimer led us to extend variational calculation to trimer binding. Since ^3He atoms are fermions, they form spin –1/2 trimers and spin –3/2 trimers. The results from Ref. 7 indicate that spin –1/2 trimer has a lower energy and therefore we studied only that case. The chosen form of the variational wave function, following Refs. 7 and 8 is

$$\psi = \Phi_a X_s + \Phi_s X_a \quad , \tag{31}$$

where X_s and X_a are spin doublets, symmetric and antisymmetric, respectively, under the exchange of particles 1 and 2 while ϕ_a and ϕ_s are space wave functions that are, respectively, antisymmetric and symmetric under the exchange of particles 1 and 2. Spin +1/2 projections of the doublets are

$$X_a(s_z = 1/2) = \frac{1}{\sqrt{2}}(\alpha_1 \beta_2 \alpha_3 - \beta_1 \alpha_2 \alpha_3) \quad , \tag{32}$$

$$X_s(s_z = 1/2) = \frac{1}{\sqrt{6}}(2\alpha_1 \alpha_2 \beta_3 - \alpha_1 \beta_2 \alpha_3 - \beta_1 \alpha_2 \alpha_3) \quad , \tag{33}$$

where α_i (β_i) are the usual spin up (down) eigenstates of a spin 1/2 particle and subscript i is the particle label. In the calculation for the space wave functions, we combine the following forms:

$$\phi_a = \phi_a^y \psi_0 = (y_1 - y_2)\psi_0 \text{ and, } \phi_a = \phi_a^x \psi_0 = (x_1 - x_2)\psi_0 \quad , \tag{34}$$

then

$$\phi_s = \phi_s^y \psi_0 = \frac{1}{\sqrt{3}}\left[(y_3 - y_1) + (y_3 - y_2)\right]\psi_0$$

and

$$\phi_s = \phi_s^x \psi_0 = \frac{1}{\sqrt{3}}(x_3 - x_1) + (x_3 - x_2)\psi_0 \quad , \tag{35}$$

$$\psi_0 = \frac{f(r_{12})}{\sqrt{r_{12}}} \frac{f(r_{13})}{\sqrt{r_{13}}} \frac{f(r_{23})}{\sqrt{r_{23}}} \quad , \tag{36}$$

where $f(r_{ij})$ is the new dimer wave function (2), with the δ modification. The constructed wave function is antisymmetric under the exchange of particles

1 and 2 and symmetric under cyclic exchange of particles 1,2 and 3. There-fore,[8] it is also antisymmetric under the exchange of particles 2 and 3 as well as 1 and 3.

The Hamiltonian of the system is

$$H = -\frac{\hbar^2}{2m}(\Delta_1 + \Delta_2 + \Delta_3) + V(r_{12}) + V(r_{13}) + V(r_{23}) \ . \tag{37}$$

Again, a variational ansatz was used to calculate the binding energy (21). Using the fact that the Hamiltonian is spin independent and symmet-ric under the exchange of x and y coordinates, we managed to express en-ergy by the following relations:

$$E \leq \frac{I_p + I_{ka} + I_{kb} + I_{kc}}{I_n} \ , \tag{38}$$

where

$$I_n = \frac{2}{3}C\int_0^\infty \rho_1 d\rho_1 \int_0^\infty \rho_2 d\rho_2 \int_0^{2\pi} d\theta (\rho_1^2 - \vec{\rho}_1 \vec{\rho}_2 + \rho_2^2)\psi_0^2(\rho_1,\rho_2,|\vec{\rho}_1 - \vec{\rho}_2|) \ , \tag{39}$$

$$I_p = 2C\int_0^\infty \rho_1 d\rho_1 V(\rho_1)\int_0^\infty \rho_2 d\rho_2 \int_0^{2\pi} d\theta (\rho_1^2 - \vec{\rho}_1 \vec{\rho}_2 + \rho_2^2)\psi_0^2(\rho_1,\rho_2,|\vec{\rho}_1 - \vec{\rho}_2|), \tag{40}$$

$$I_{ka} = -2C\frac{\hbar^2}{m}\int_0^\infty \rho_1 d\rho_1 \left(\frac{f''(\rho_1)}{f(\rho_1)} + \frac{1}{4\rho_1^2}\right)\int_0^\infty \rho_2 d\rho_2 \ .$$

$$\int_0^{2\pi} d\theta (\rho_1^2 - \vec{\rho}_1 \vec{\rho}_2 + \rho_2^2)\psi_0^2(\rho_1,\rho_2,|\vec{\rho}_1 - \vec{\rho}_2|) \ , \tag{41}$$

$$I_{kb} = -2C\frac{\hbar^2}{m}\int_0^\infty \rho_1 d\rho_1 \left(\frac{f'(\rho_1)}{f(\rho_1)} - \frac{1}{2\rho_1}\right)\int_0^\infty \rho_2 d\rho_2 \left(\frac{f'(\rho_2)}{f(\rho_2)} - \frac{1}{2\rho_2}\right) \ .$$

$$\int_0^{2\pi} d\theta \cos\theta (\rho_1^2 - \vec{\rho}_1 \vec{\rho}_2 + \rho_2^2)\psi_0^2(\rho_1,\rho_2,|\vec{\rho}_1 - \vec{\rho}_2|) \ , \tag{42}$$

$$I_{kc} = -3C\frac{\hbar^2}{m}\int_0^\infty \rho_1^2 d\rho_1 \left(\frac{f'(\rho_1)}{f(\rho_1)} - \frac{1}{2\rho_1}\right)\int_0^\infty \rho_2 d\rho_2 \int_0^{2\pi} d\theta \Psi_0^2(\rho_1,\rho_2,|\vec{\rho}_1 - \vec{\rho}_2|) \tag{43}$$

C is a constant, $\vec{\rho}_1 = \vec{r}_1 - \vec{r}_2$, $\vec{\rho}_2 = \vec{r}_1 - \vec{r}_3$ and θ is the angle between $\vec{\rho}_1$ and $\vec{\rho}_2$. The fact that expressions for the energy were reduced to three-dimensional integrals enabled us to perform the calculations using the same numerical methods as in the dimer case. After time consuming numerical calculations,

we found that the upper bound of the binding energy of $^3\text{He}_3$ trimer was -0.0057mK. Using the same wave function, we derived the average distance between atoms of $<\rho_1> = 4503$ Å and $\Delta\rho_1 = 3633$ Å.

DISCUSSION

To the best our knowledge, function (2) is the first trial form that in variational calculation led to the binding of helium 3 dimer. In this case, one-dimensional Romberg integration with high accuracy has been performed. The results are in good agreement with those obtained by numerical solving of the Schrödinger equation.

Having an appropriate two-body function, we were able to construct a special form of the Cabral-Bruch trimer wave function describing the state with spin 1/2. We performed a very accurate Romberg integration of three-dimensional integrals and found an upper bound to the binding energy of -0.0057 mK. This shows that helium 3 trimer with spin $-1/2$ is bound in two dimensions. (As it was showed in paper,[4] binding of diatomic helium molecules is significantly increased if they are close (about 3 Å) to the surface of liquid helium. It means that binding of trimers could be experimentally observed in the future.) This result is quite a new one. It opens the question of the phase of many helium 3 atoms on the surface of liquid helium 4. So far, it has been believed that they form a two-dimensional gas.

A qualitative estimation of our result for the trimer may be done as well. The obtained values of $<\rho_1> = 4503$ Å and $\Delta\rho_1 = 3633$ Å show that $^3\text{He}_3$ is a large molecule. Let us assume that there is a homogenous monolayer gas (or liquid) with the average distance between particles as in helium 3 trimer, then its concentration is 4.9×10^{12} m^{-2}. This concentration is several orders lower than the one of 3%, which is the upper limit for the attractive interaction between two ^3He atoms in helium 3–helium 4 film.[14] Consequently, it may be concluded that in our case the necessary condition for the binding of three helium atoms is satisfied.

Recently, new interatomic helium potentials have appeared. Van Mourik and Dunning computed a new *ab initio* potential energy curve[9] that lies between the HFD-B3-FCI1 (Ref. 12) and SAPT2 (Ref. 13) potentials, being closer to SAPT2 potential. Other authors[10,11] maintain that, according to their calculations, SAPT potential is insufficiently repulsive at short distances.

In papers,[2,3] the binding energy of helium molecules was calculated using two different potentials, HFD-B3-FCI1 and SAPT. The obtained results did not differ substaintially for these two cases. Therefore, we do not expect that the calculations with new, more precise potentials would change our results appreciably.

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SAŽETAK

Varijacijska analiza dimera i trimera helija-3 u dvije dimenzije

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Varijacijskim proračunom dobijena je energija vezanja dimera helija-3 u dvije dimenzije. Definitivno je utvrđeno postojanje jednog vezanog stanja, s energijom vezanja od – 0.014 mK. Također je određena energija vezanja odgovarajućeg trimera spina –1/2 od – 0.0057 mK. Ovaj rezultat otvara dvojbe o tome da atomi helija-3 tvore plinsku fazu na ravnoj površini suprafluidnog helija-4.