PHYSICAL REVIEW B **67**, 075403 (2003)

# Enhanced cohesion of matter on a cylindrical surface

M. K. Kostov, M. W. Cole, and G. D. Mahan
*Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802*

C. Carraro
*Department of Chemical Engineering, University of California, Berkeley, California 94720*

M. L. Glasser
*Department of Physics and Center for Quantum Device Technology, Clarkson University, Potsdam, New York 13699*

(Received 1 July 2002; published 6 February 2003)

We evaluate the cohesive energies $E_b$ of four systems in which particles move on a cylindrical surface, at fixed distance $R$ from the axis. We find quite nonuniversal dependences of $E_b$ on $R$. For the Coulomb binding problem, $E_b$ is a monotonically decreasing function of $R$. For three problems involving Lennard-Jones interactions, the behavior is nonmonotonic; $E_b$ is larger at $R=\infty$ than at $R=0$. The maximum binding corresponds to $R\sim 0.7\sigma$, where $\sigma$ is the hard core parameter. Consequences of the enhanced binding are discussed.

DOI: 10.1103/PhysRevB.67.075403 PACS number(s): 03.65.−w, 03.65.Ge, 67.90.+z

The discovery of carbon nanotubes has stimulated a rapid evolution of ideas, experiments, and understanding concerning states of matter confined to the proximity of a cylindrical surface.[1–3] Examples of such systems include electrons present within a nanotube and atoms or molecules moving just outside or within such tubes. This paper reports unexpected behavior we have found in studies of four such systems: a $+/-$ pair of charges bound by the Coulomb interaction, a pair of atoms interacting with a Lennard-Jones (LJ) interaction, an ensemble of $^4$He atoms, which condenses, and a low density fluid consisting of classical atoms. We assume that all particles move on a cylindrical surface, of radius $R$ and infinite length. This has been shown to be the case for molecules coating the inner surface of nanotubes having $R\sim 6$ to 9 Å.[4] The assumption of surface confinement simplifies the calculations without sacrificing the basic physics.

For each of these four systems, considerable attention has been directed previously to the investigation of two extreme limits of the present problem. The limit $R=\infty$, here called ‘‘flatland,’’ is that of particles moving on a plane, i.e., a two-dimensional (2D) problem. This has been extensively pursued in connection with both the 2D electron gas and monolayer films.[5–7] The opposite limit, $R$ approaching zero, is here called ‘‘lineland,’’ a 1D limit. Matter in lineland has been explored for many years as an abstract problem[8] and has recently received particular attention in connection with the possible realization of 1D phases within interstitial channels, within nanotube bundles, or grooves, on the external surface of nanotube bundles.[2,9–17] A logical question addressed in this paper is whether the properties of matter in ‘‘cylinderland’’ evolves smoothly (or even monotonically) between these limits as the value of $R$ is varied. We find that the answer is ‘‘yes’’ in just one of the four cases and that intriguing behavior arises in all four cases.

The first problem we address is the ground state binding energy $E_b(R)$ of pair of charges located on the surface of a cylinder, interacting via Coulomb potential $-e^2/r$ where $r$ is the interparticle separation. The Schrödinger equation for the hydrogen atom is exactly solvable in 2D, resulting in a ground state wave function proportional to $\exp(-2r/a_0)$, with binding energy $E_b(\infty)=2$ Hartrees and mean separation $a_0/2$ between the interacting charges.[18] Here $a_0$ is the Bohr radius. In the opposite limit of 1D, we encounter a well-known logarithmic divergence of $E_b$ as $R$ approaches zero;[19–21] the particle ‘‘falls to the center of the attractive force’’ in 1D. Recently, Kostov *et al.*[22] have evaluated $E_b$ for this system, using a variational method. Figure 1 depicts their result for a three-parameter variational solution of the problem (for details see Ref. 22). The energetics of binding is found to exhibit a *monotonic* trend as a function of the reduced curvature $C=a_0/R$. The known 1D and 2D limits of $E_b$ are accurately reproduced by the variational calculation. Note that $E_b$ is very insensitive to the value of $C$, as long as $C$ is small.[23] This weak dependence is confirmed by a perturbation theory calculation (see the inset of Fig. 1).

The second problem we consider is the ground state cohesive energy (per atom) of a fluid consisting of $^4$He atoms whose nuclei are confined to the cylindrical surface. The

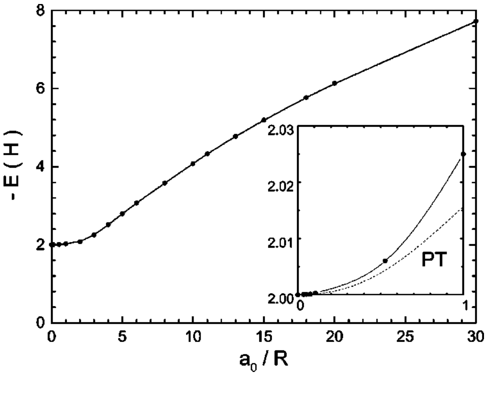

FIG. 1. Ground state energy of a $(+/-)$ Coulomb pair of charges, constrained to lie on a cylinder, as a function of curvature $a_0/R$. The points are the variational results and the solid line is a fit to them. The inset compares the variational and perturbation theory (PT) results. The energy is expressed in Hartree units.

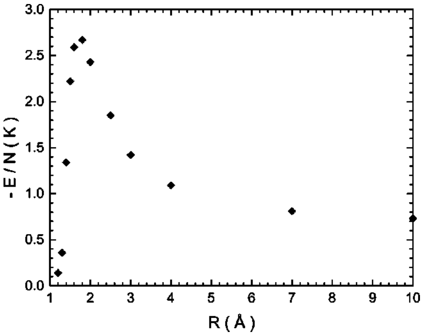

FIG. 2. Ground state cohesive energy (per atom) of a $^4$He fluid on a cylinder as a function of radius.

analogous problem has been extensively investigated in 2D,[24] for which it is relevant to superfluid films, and 1D, where it has been studied for potential application to an interacting interstitial fluid.[9–12] The problem of $^4$He has also received some attention in connection with endohedral adsorption within nanotubes.[4] In 3D, the cohesive energy of $^4$He is $E_b = 7.2$ K. In 2D, it is 0.85 K and in 1D it is $\sim 3$ mK. What $R$ dependence is expected for $E_b$ on a cylinder?

We have studied this problem variationally, using a Jastrow trial wave function, i.e., a symmetrized product of two-particle functions that prevent hard-core overlap of the atoms. The pair factor in our trial wave function is of McMillan form.[25] The interatomic potential assumed in the calculation is the modern ‘‘Aziz’’ potential.[26] The results of this liquid state calculation appear in Fig. 2. The dependence of $E_b$ on curvature is *not* monotonic. Indeed, the binding energy is a factor 3.7 higher near $R = 1.8$ Å than in flatland. The nonmonotonic dependence of $E_b$ on $C$ and the minimum value at $C = \infty$ stand in stark contrast to the monotonically *increasing* dependence of $E_b$ on $C$ and maximum value at $C = \infty$ found for the Coulomb problem.

The origin of the binding energy maximum near $R = 2$ Å is the minimum in the He-He interaction near $r_{\min} = 2.9$ Å. The He fluid has a strong binding if the geometry encourages the particles to have such a spacing. This is the case for the cylindrical geometry, as indicated by the following argument. We introduce a function called the specific area function $a(r)$, defined as the area on the cylinder's surface at distance $r$ from a specified point on the surface, per unit distance from this point. Letting this point be the origin, we have $a(r) = \int d^2 r' \, \delta(|r' - r|)$. In the flatland limit $a(r) = 2\pi r$, while in the lineland limit ($R \to 0$), $a = 2\pi R$. In the cylindrical case, $a(r)$ is proportional to an elliptic integral, which exhibits a divergence at $r = 2R$. The origin of this divergence is that a particle on one side of the cylinder has a divergent specific area at distance $2R$; the sphere of this radius is tangent to the cylinder. As a consequence, the system's energy is lowered when this distance is such that the interaction is strongly attractive. The optimal binding does not occur precisely when the minimum in the He-He potential coincides with the diameter of the cylinder, but instead at $\sim 20\%$ higher value of $R$. This difference arises from the zero-point energy of the system, which expands the nearest neighbor distance beyond $r_{\min}$. The same behavior occurs for liquid He in 2D and 3D $^4$He; the ‘‘nearest-neighbor’’ peak in the radial distribution function occurs at distance about 20% greater than $r_{\min}$.[27]

What consequences accompany the greatly enhanced binding at this value of $R$? Typically (but not always), strongly cohesive systems exhibit relatively large speeds of sound. Here, the sound propagation speed $s$ is derived from the relation appropriate to longitudinal density fluctuations propagating parallel to the axis of the cylinder $Ms^2 = \rho^2 [d^2(E/N)/d\rho^2]$. In this expression, $M$ is the atomic mass and $\rho$ is the 1D density; the derivative is evaluated at the ground state density. According to our calculations, $s = 230$ m/s at the equilibrium density $\rho = 0.3$ Å$^{-1}$ for the optimally binding radius $R = 1.8$ Å. This value may be compared to the values $s = 240$, 90, and 8.0(3.0) m/s in 3D, 2D, and 1D, respectively. We observe that the speed in cylinderland at the optimal radius is *significantly* enhanced relative to both the 1D and 2D values of $s$.

Since the cylindrical fluid is a 1D system, from the perspective of statistical mechanics, it undergoes no phase transitions at finite $T$. There is, however, a $T = 0$ transition as the system evolves from a liquid-vapor coexisting ground state to a disordered fluid at nonzero $T$, with a singular heat capacity [proportional to $\delta(T)$]. The integrated specific heat is a monotonic function of the binding energy. This might be observable, due to inhomogeneity in any real system, as a smeared out maximum in the low $T$ specific heat. The fluid's compressibility diverges as $\exp[E_b/(k_B T)]$ at low $T$, which should be observable in an adsorption isotherm. However, we have thus far no definite calculations of these quantities to compare with experiments. The problem is nontrivial because of the $T = 0$ transition, invalidating conventional low $T$ expansion methods.

The third problem we address is the curvature-dependent binding energy $E_b(R)$ of an atomic dimer on a cylinder of radius $R$. This is the two-body version of the liquid helium problem just discussed, except that here we treat the general case of atoms interacting with an arbitrary Lennard-Jones (LJ) interaction. One atom's position is fixed at cylindrical coordinates $(z, \phi) = (0,0)$ while the other moves over the surface; their separation is $r = \sqrt{z^2 + [2R \sin(\phi/2)]^2}$. By scaling distances relative to the hard-core diameter $\sigma$ and the energy relative to the well depth $\epsilon(\tilde{z} = z/\sigma, \tilde{R} = R/\sigma, \tilde{r} = r/\sigma; \mathcal{E}_b = E_b/\epsilon)$, the Schrödinger equation becomes

$$-\eta \left( \frac{\partial^2}{\partial \tilde{z}^2} + \frac{1}{\tilde{R}^2} \frac{\partial^2}{\partial \phi^2} \right) \Psi_0 + [4(\tilde{r}^{-12} - \tilde{r}^{-6}) - \mathcal{E}_b] \Psi_0 = 0. \tag{1}$$

Note that the ground state solution to this equation is determined by the boundary conditions and the value of the dimensionless *de Boer quantum parameter* $\eta = \hbar^2 / M \epsilon \sigma^2$. A large value of $\eta$ implies a large zero-point energy and the absence of any bound state, while a small value yields many bound states. Here, we focus on a specific question: what is the threshold value ($\eta = \eta_t$) separating those problems for which the dimer exists ($\eta < \eta_t$) from those for which it does

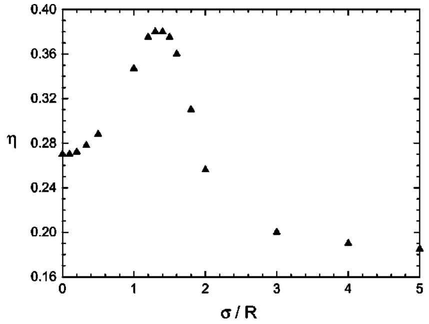

FIG. 3. Threshold value of the de Boer quantum parameter for the existence of a dimer consisting of two atoms, with an LJ interaction, as a function of inverse scaled radius.

not exist? The corresponding threshold value of $\eta_t$ is known for the limiting cases of 1D ($\eta_t = 0.1788$) Ref. 10 and 2D ($\eta_t = 0.269$).[28] We have determined the threshold for cylinderland by computing the ground state energy variationally, identifying $\eta_t$ from the point when the attractive potential energy becomes too weak for the dimer to be bound. Because the calculation is variational, the computed threshold $\eta_t$ is a lower limit to the exact value. Our $\eta_t$ results agree well with the known 1D and 2D limits cited above. For the general cylindrical case, the threshold value is shown in Fig. 3. Qualitatively, it exhibits the same phenomenon as was seen in the $^4$He liquid binding problem. That is, $\eta_t$ is particularly large when the diameter of the cylinder is such that the interatomic interaction across the cylinder is strongly attractive. There is a small difference between values of the ''optimal'' radius $R_{\text{opt}}$ for the two problems $R_{\text{opt}}/\sigma$ has the value 0.7 for the dimer problem and 0.77 for the $^4$He binding problem.

Bearing in mind the interesting behavior of the threshold $\eta_t$ in the cylinderland, we next address the specific problem of the binding energy ($E_b$) of $^4$He ($\eta = 0.18$) and $^3$He ($\eta = 0.24$) dimers on a cylinder. Employing the same variational approach as was used for $\eta_t$, we compute the ground state energy for these systems as a function of $\sigma/R$ (Fig. 4). Not surprisingly, we find significantly enhanced binding energy for the $^4$He and $^3$He dimers within a particular range of values of $R$ (near $0.65\sigma$). The enhanced binding in the cylindrical geometry is particularly dramatic for the $^3$He dimer, yielding an increase in $E_b$ of 7 orders of magnitude compared to the 2D limit for $E_b$.[29]

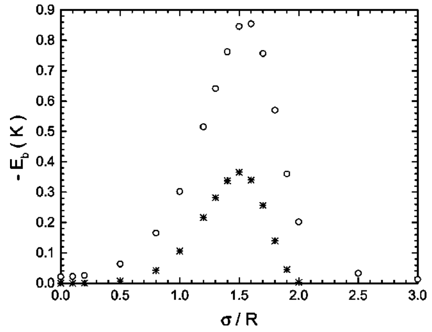

FIG. 4. Ground state energy of $^4$He (circles) and $^3$He (stars) dimers on a cylinder.

We now wish to discuss the implications of our results for spin-aligned hydrogen systems H↑ and D↑. The symbols H↑ and D↑ refer, respectively, to atomic hydrogen and deuterium pairs interacting in the $b^3\Sigma_u^+$ state. For these systems the de Boer quantum parameter $\eta$ has values larger than the 2D and 1D threshold values[28] $\eta(\text{H}\uparrow) = 0.547 \gg \eta_t(2d)$ and $\eta_t(1d)$, and $\eta(\text{D}\uparrow) = 0.274 > \eta_t(2d)$ and $\eta_t(1d)$. The very large value of $\eta$ for H↑ $[\eta(\text{H}\uparrow) > \eta_t(R_{\text{opt}})]$ implies that an H↑ dimer certainly cannot exist in 1D, 2D, or on any cylindrical surface. The situation for D↑ system is more interesting, since the D↑ dimer does not exist in 2D and 1D, but it does exist on a cylinder for extended range of radii—from $\sigma/R \approx 0.3$ up to $\sigma/R \approx 1.9$. This is a unique situation.

Recently, the ground state properties of He dimers in confined geometry were studied by Kilic *et al.*[30,31] They find a similar effect on the binding energy due to the confinement. $E_b$ is significantly enhanced when the width of the holding region is approximately equal to the range of the pair interaction. $^3$He and $^4$He dimers within a nanotube have been studied by Vranjes *et al.*[32–34] Their model is different from ours, since we constrain the atoms to move on a cylindrical surface. They also find that the binding energy depends on $R$ and exhibits nonmonotonic behavior.[35] M. Aichinger and E. Krotscheck have recently explored the $^3$He dimer problem using a fourth order real-space algorithm for solving local Schrödinger equations.[36] That technique yields an exact eigenvalue, within numerical limitations. Their results (to be published) indicate that the dimer's binding energy is even greater (by a factor about 2.6) than is reported here for the radius that binds the dimer most strongly. The value of that optimal radius is close to the value found in the present work.

Having established that the $^3$He dimer is so strongly bound for radius $R \sim 0.65\sigma$, we discuss briefly the ground state of a system of many $^3$He atoms. No condensed $^3$He liquid exists in either 2D or 1D, but perhaps one exists in cylinderland. Note that the existence of the stable dimer does not ensure the existence of a stable $N$-mer for any $N > 2$ (as was shown explicitly in the 2D case[29]). One scenario is that the ground state of the system is a gas of such $^3$He dimers (analogous to $H_2$ at room temperature). A second possibility is that the dimers coalesce to form a liquid, analogous to liquid $H_2$ between its triple point and its critical point. A third scenario is that the dimers dissolve into a many-body liquid ground state, analogous to liquid $^3$He in 3D. Neither of the first two of these possibilities has been previously found for the $^3$He many-body system. The question of which phase, among the three candidates, is the actual ground state remains open.

Finally, we address a fourth problem concerned with matter in cylinderland—the second virial coefficient of a classical gas. By analogy with well known problems in other dimensions, we write a low density (high-$T$) expansion of the 1D pressure $P = \rho^2(\partial f/\partial \rho)$, where $f$ is the free energy per particle, as $P/(\rho k_B T) \approx 1 + \rho B(T)/2\pi R + \cdots$, with

$$B(T) = \frac{1}{2}\int d^2\mathbf{r}\{1 - \exp[-\beta V(r)]\}. \tag{2}$$

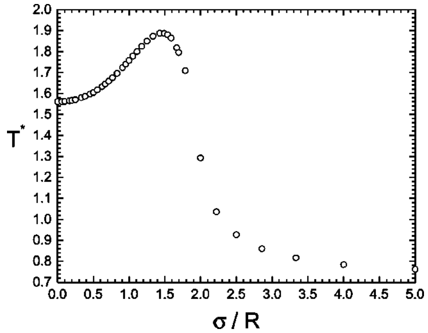

FIG. 5. Reduced Boyle temperature $T^*=k_BT/\epsilon$ for classical gases on a cylinder of radius $R$.

Here, $\beta=1/(k_BT)$ and the integration is over the cylinder's surface. We assume the usual LJ form of interaction. The results (to be reported in detail elsewhere) exhibit similar qualitative behavior to that found for the preceding two problems, both of which also involve a pair interaction with an attractive well. As in the familiar 2D and 3D contexts, $B$ is positive at high $T$ due to the repulsive interactions and $B<0$ at low $T$, where attractive effects dominate. The Boyle temperature $T_B$ is that for which $B$ vanishes, meaning that (to second order in this expansion) $P$ for the interacting system is the same as that of a noninteracting gas at the same $\rho$ and $T$. The results in Fig. 5 indicate that the Boyle temperature is the highest for a cylinder of radius $R=0.7\sigma$. In the van der Waals theory of condensation, the critical temperature is $8T_B/27$. In that mean field theory, therefore, the critical temperature is highest for a cylinder of this ''optimal'' radius. While this transition does not occur in the exact theory, one expects the virial expansion to apply at low density. Then, the data of Fig. 5 imply an onset of the effects of attraction at higher $T$ for the cylinder than for either 1D or 2D limiting cases.

In a separate work, Calbi[37] is investigating another problem in this family—the classical crystal composed of atoms on a cylindrical surface. The crystallization of the classical gas leads to an oscillatory dependence of the energy on $R$ (due to the size-dependent commensuration energy). Also found in that problem is an enhanced binding for the crystaline state. Specifically, the ground state cylindrical crystal of such an optimized radius is about 7% more strongly bound than is the 2D crystal (and a factor of 3 more strongly bound than the 1D crystal).

In summary, we have explored both classical and quantum particles confined to a cylindrical surface. In the case of a Coulomb interaction, which lacks a characteristic distance, the energetics of binding exhibits a monotonic trend as a function of curvature; there is an insensitivity of the cohesion to the value of the curvature, as long as it is small. In three problems involving interactions with a favored distance, quite distinct behavior was found: there is a particular range of values of $R$ such that the binding is enhanced; this is explicable in terms of a phase space argument relevant to interactions which have well-defined potential energy minima. We believe that this distinction between power law interacting and finite range systems is generic and should be applicable to other geometries. For example, matter confined to a spherical surface (small particles or pores) should have enhanced cohesion when the diameter is $\sim\sigma$. Other related problems merit investigation. One is the possibility of condensation of $^3$He to a liquid. The factor of $\sim 3$ enhancement of the liquid binding energy shown for cylindrical $^4$He (relative to binding in flatland) suggests that there is a range of radius over which the lighter isotopic liquid should also bind. A suggestive argument in support of that possibility is the fact that the $^3$He dimer exists over an extended range of radii $R>\sigma/2\sim 1.3$ Å, according to Fig. 3 ($\eta=0.24$ for $^3$He). For bose systems in 1D and 2D, the dimer threshold coincides with the binding threshold for the many-body bound state liquid. While we do not know the corresponding criterion for self-binding in cylinderland; it seems plausible that the cylindrical liquid $^3$He also exists.[38–40] This would be a remarkable system to investigate since it exemplifies a novel Luttinger liquid. A primary application of these results is to carbon nanotubes, whose radius can be as small as 2 Å.[41] Indeed, the so-called cylindrical shell phase of He and $H_2$ corresponds to adsorption at radial distance $\delta r\sim 3$ Å inward from the carbon atoms.[4] Thus, the pronounced effects found here for these gases at $R\sim 2$ Å (Figs. 2, 4) correspond to cylindrical phases within nanotubes of radius $\sim 5$ Å. Experimental and other theoretical study of this size tube is worth pursuing.

We are grateful to Susana Hernandez, Claire Lhuillier, Jordi Boronat, and L. W. Bruch for helpful comments. This research has been supported by Army Research Office, the Petroleum Research Fund of the American Chemical Society and the National Science Foundation (Grant No. DMR0121146). Milen Kostov is grateful to Air Products and Chemicals, Inc. (APCI) for its support. We are grateful to Eckhard Krotscheck for discussion of the $^3$He binding problem, especially concerning the orgin of the discrepancy in the $^3$He dimer energy.

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