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Reply to "Comment on 'Quantum virial expansion approach to thermodynamics of ^{4}He adsorbates in carbon nanotube materials: Interacting Bose gas in one dimension' "

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The calculation of the second virial coefficient in one dimension (1D) is performed within a scattering shifts approach. It is shown that when one carefully considers the (anti)symmetrization requirements for gases of spinless bosons and fermions in 1D, the second virial coefficients turn out to be identical. These results are used to recalculate the second virial coefficient for ^{4}He atoms adsorbed in grooves of the carbon nanotube bundles.

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In a recent article,[1] I pointed out that ^{4}He atoms adsorbed in the grooves of carbon nanotube bundles can be treated as interacting Bose gas in one dimension. I calculated the second quantum virial coefficient (B_2) for such ^{4}He gas. In a comment[2] to this article, Bruch found certain inadequacies in the treatment I presented. There are two basic points in his comment:

(i) The "exchange" part of virial coefficient must be vanishingly small in 1D. This fact can easily lead to conclusion that the formula for B_2 I presented in Eq. (18) of Ref. 1 has an odd appearance due to the fact that the ideal gas (quantum statistic) contribution explicitly figures in it.

(ii) The "direct" part of virial coefficient must be larger from the classical value for B_2. On the basis of this fact, he questions the values obtained for B_2 in Ref. 1, since they do not fulfill this inequality as he explicitly shows.

The answer to point (i) is easy. Namely, the quantum virial coefficient can be written in two ways. The first one (used in Ref. 1) is to write the virial coefficient as a sum of ideal gas term and the remaining part. The remaining part can be written as an integral over the relative wave vector, in which the phase shifts of the two-body problem figure explicitly. This representation of B_2 is very convenient from the calculational point of view. The other way (used by Bruch in Ref. 2) is to write the B_2 as a sum of the so-called "direct" and "exchange" terms. This "other way" is merely a specific choice of the rearrangement of terms appearing in the expression for B_2. The equivalence of the two approaches has been demonstrated in Ref. 3. In particular, the ideal gas term appears as the part of the "exchange" virial coefficient. Thus, there is nothing *a priori* strange about the appearance of this term in Eq. (18) of Ref. 1.

Point (ii) is certainly more serious. I have calculated the classical second virial coefficient and I find that at a temperature of 10 K the classical value of the second virial coefficient is 0.83 Å, in approximate agreement with the value that Bruch finds. This confirms the problems with the B_2 coefficient obtained in Ref. 1, and the appropriateness of point (ii) raised by Bruch. The reason for inadequacy of Eq. (18) of Ref. 1 is subtle and is not due to a "lost" minus sign. It is related to transition from a sum over discrete states in Eq. (17) to an integral in Eq. (18) of Ref. 1. As Bruch points out, a correct formula has been given in Ref. 4. This reference deals with subtleties of the virial coefficient evaluation in 3D, 2D, and 1D. A very specific behavior of the virial coefficient in 1D, and the reason for the "slippage" that led to Eq. (18) of Ref. 1 can be, however, more transparently illustrated as follows.

In 1D, one has to (anti)symmetrize the part of the two-body wave function in relative "radial" coordinate. This is not the case in 2D and 3D treatments where the (anti)symmetrization can be performed by choosing (anti)symmetric combinations of spherical functions in anglular variables. When the symmetrization requirement is applied to two spinless particles in 1D, one obtains that the allowed relative wave vectors for bosons are

$$k_i^0 = (i+1/2)\pi/L;\; k_i = [(i+M+1)\pi - \eta(k_i)]/(L-a), \quad i = 0,1,2,\ldots, \tag{1}$$

while for fermions they are

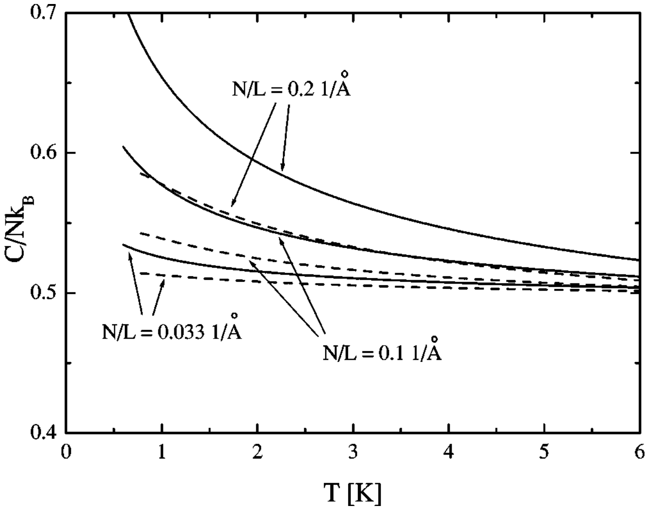

FIG. 1. Specific heat of ^{4}He gas adsorbed in the grooves of SWCNT bundles for three different linear densities (0.033 Å^{-1}, 0.1 Å^{-1}, and 0.2 Å^{-1}). Full line: Calculation with the SAPT1 potential. Dashed line: Calculation with the scaled SAPT1 potential.

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$$k_i^0 = (i+1)\pi/L;\ k_i = [(i+M+1)\pi - \eta(k_i)]/(L-a),\quad i = 0,1,2,\ldots. \tag{2}$$

In the above equations, k_i^0 represents the relative wave vector for two *noninteracting* particles and k_i is a relative wave vector for two particles *interacting via a hard-core potential*. $2L$ is the length of the 1D quantization box $([-L,L])$, $\eta(k_i)$ is the phase shift of the two-body problem [see Eqs. (13) and (14) in Ref. 1], a is the hard core length of the two-body interaction potential $(0 < a \ll L)$, and M is the number of bound states for the particle of reduced mass [μ, see Eq. (13) in Ref. 1]. There is an important difference in the ground state energy of two noninteracting bosons and fermions contained in any finite length quantization box in 1D that arises due to the wave function (anti)symmetrization requirement. The ground state energy is lower for two noninteracting bosons, which can be seen from the difference between the lowest allowed relative wave vectors k_i^0 for noninteracting bosons and fermions, respectively. When one inserts Eqs. (1) and (2) in Eq. (17) of Ref. 1 and performs the transition from summation over i to integration over k_i (k), the following result[5] is obtained:

$$B_2 = \frac{\lambda}{2\sqrt{2}} - \lambda\sqrt{2}\sum_b \exp(-\beta\epsilon_b) - \frac{\lambda\sqrt{2}}{\pi}\int_0^\infty \frac{d\eta(k)}{dk} \times \exp\left(-\beta\frac{\hbar^2k^2}{2\mu}\right)dk, \tag{3}$$

where the same notation as in Ref. 1 is used. The appearance of additional contribution of $\lambda/\sqrt{2}$ to B_2 for bosons, not present in the results of Ref. 1 is due to the mismatch (level shift) between the interacting and noninteracting energy spectra [factor of $\pi/2L$ in Eq. (1)]. This effect is not present for fermions in 1D. Equation (3) should be used instead of Eq. (18) of Ref. 1. Intriguingly (and satisfactory) enough, the same expression for B_2 is obtained both for fermions and bosons interacting via a hard-core potential in 1D, due to the subtle difference in allowed noninteracting relative wave vectors (k_i^0) in Eqs. (1) and (2). This is in accord with the opening sentence of Bruch's comment.[2] Equation (3) gives values of 2.13 Å and 2.3 Å at a temperature of 10 K, for B_2 calculated with SAPT1 and scaled SAPT1 potentials, respectively.[1] Both values are now larger from their classical counterparts. This holds for all temperatures, which is the expected behavior.

In light of this correction, Eq. (20) of Ref. 1 should be modified to

$$n\beta^2\frac{d^2B_2}{d\beta^2} = n\lambda\sqrt{2}\left[-\frac{1}{16} + \frac{S_0+I_0}{4} + \beta(S_1+I_2) - \beta^2(S_2+I_2)\right]. \tag{4}$$

This modification gives rise to different behavior of specific heat of ^{4}He atoms in grooves, namely it rises *above* the noninteracting value as the temperature is decreased. The new prediction for specific heat is displayed in Fig. 1, which should be used instead Fig. 5 of Ref. 1.

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[5] The transition from integral over $\eta(k)$ to integral over $d\eta(k)/dk$ is performed using one partial integration as in 2D and 3D treatments (Ref. 3).