Quantum-Mechanical Second Virial Coefficient of a Hard-Sphere Gas at High Temperature*

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The quantum-mechanical second virial coefficient NB(T) of a gas of hard spheres is evaluated at high temperature T. The result is

$$NB(T) = \frac{2\pi a^3 N}{3} \left[1 + \frac{3}{2\sqrt{2}} \frac{\lambda}{a} + \frac{1}{\pi} \left(\frac{\lambda}{a} \right)^2 + \frac{1}{16\pi\sqrt{2}} \left(\frac{\lambda}{a} \right)^3 + \cdots \right].$$

Here N is Avogadro's number, a is the diameter of a molecule, and $\lambda = (2\pi \hbar^2 \beta/m)^{1/2}$ is the thermal wavelength, defined in terms of the molecular mass m, Planck's constant h, and $\beta = 1/kT$ where k is Boltzmann's constant. The evaluation is based upon a direct method of expanding the Bloch function, which occurs in the expression for B(T), in powers of a^{-1} . The first term in B(T) is just the result of classical statistical mechanics, the second term was obtained by Uhlenbeck and Beth, and one-half the third term was obtained by Mohling. Boyd, Larsen, and Kilpatrick calculated B(T) numerically, fitted their results to a series of the above form, and obtained coefficients which agree with our exact results to seven decimal places for the third term and nearly as many for the fourth term.

I. INTRODUCTION

A CCORDING to quantum statistical mechanics, the second virial coefficient NB(T) of a gas of hard spheres at temperature T is the sum of a direct and an exchange part, $B(T) = B_D(T) + B_E(T)$. Here N is Avogadro's number. It has been proved by Larsen, Kilpatrick, Lieb and Jordan¹ and by Lieb² that $B_E(T)$ decreases exponentially with T as T becomes infinite. Therefore, to find B(T) for T large it suffices to evaluate $B_D(T)$, which is given by

$$B_D(T) = \frac{1}{2} \int [1 - 2^{3/2} \lambda^3 G(\mathbf{x}_0, \mathbf{x}_0, \beta)] d\mathbf{x}_0.$$
 (1.1)

Here λ , the thermal wavelength, and $G(\mathbf{x},\mathbf{x}_0,\beta)$ the thermal Green's function, or Bloch function, are defined by

$$\lambda = (2\pi\hbar^2\beta/m)^{1/2}, \qquad (1.2)$$

$$\frac{\partial G}{\partial \beta} - D\Delta G = -\delta(\beta)\delta(\mathbf{x} - \mathbf{x}_0), \qquad (1.3)$$

$$G(\mathbf{x}, \mathbf{x}_0, 0-) = 0, \qquad (1.4)$$

$$G(\mathbf{x}, \mathbf{x}_0, \beta) = 0, \quad |\mathbf{x} - \mathbf{x}_c| \le a. \tag{1.5}$$

In these equations $\beta = 1/kT$, where k is the Boltzmann constant and T is the absolute temperature, \mathbf{x}_c is the center of a spherical molecule and a is its diameter

(i.e., a is the radius of the hard-core interparticle potential), m is the mass of a molecule, $D = \hbar^2/m$ and \hbar is Planck's constant divided by 2π .

We shall present a method for expressing $B_D(T)$ as a series in powers of λ/a and use it to obtain

$$NB_{D}(T) = \frac{2\pi a^{3}N}{3} \left[1 + \frac{3}{2\sqrt{2}} \frac{\lambda}{a} + \frac{1}{\pi} \left(\frac{\lambda}{a} \right)^{2} + \frac{1}{16\pi\sqrt{2}} \left(\frac{\lambda}{a} \right)^{3} + \cdots \right]. \quad (1.6)$$

Since $B(T) \sim B_D(T)$ for T large, (6) is also the expansion of B(T). The first term in (6) is the result according to classical statistical mechanics and the subsequent terms are quantum-mechanical corrections to it. The term in λ/a was obtained by Uhlenbeck and Beth.³ Mohling⁴ calculated the term in $(\lambda/a)^2$ but obtained the coefficient $1/2\pi$ instead of $1/\pi$. The fact that his result was wrong was discovered by Boyd, Larsen and Kilpatrick⁵ who evaluated B(T) numerically and fitted their results to a series of the form (6). Their third coefficient agreed to seven decimal places with $1/\pi$, which is our result. Their fourth coefficient agreed with $1/16\pi\sqrt{2}$ to nearly as many places. In fact they guessed both of these numbers, as well as the next coefficient, from their calculations.

We wish to thank Elliott Lieb for suggesting the problem of resolving the discrepancy between the numerical and analytical results.

II. EXPANSION OF $G(x,x_0,\beta)$ IN POWERS OF a^{-1}

Let us introduce a Cartesian coordinate system $\mathbf{x} = (x_1, x_2, x_3)$ with its origin on the surface of the sphere

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¹S. Larsen, J. Kilpatrick, E. Lieb, and H. Jordan, Phys. Rev. 140, A129 (1965). These authors obtained an upper bound on $B_E(T)$ of the form $\ln[B_E(T)/B_E^0(T)] \leq -2\pi (a/\lambda)^2$. Here $B_E^0(T)$ is the exchange virial coefficient for an ideal gas without a hard core.

² E. H. Lieb [J. Math. Phys. 7, 1016 (1966)] shows the exact asymptotic formula for $B_E(T)$ to be $\ln[B_E(T)/B_E^0(T)] = -\frac{1}{2}\pi^3(a/\lambda)^2 + O[(a/\lambda)^{2/3}].$

 ³ G. E. Uhlenbeck and E. Beth, Physica 3, 729 (1936).
 ⁴ F. Mohling, Phys. Fluids 6, 1097 (1963).
 ⁵ M. E. Boyd, S. Y. Larsen, and J. E. Kilpatrick (to be pub-int). lished).

 $|\mathbf{x}-\mathbf{x}_c|=a$ and with the x_1 axis passing through the center of the sphere \mathbf{x}_c and the source point \mathbf{x}_0 . Then $\mathbf{x}_c = (-a, 0, 0)$ and $\mathbf{x}_0 = (x_0, 0, 0)$, where x_0 denotes the distance of the source from the sphere. In addition the x_2 , x_3 plane is tangent to the sphere at the origin. In these coordinates the equation of the hemisphere $x_1 > -a$ is

$$x_1 = [a^2 - \rho^2]^{1/2} - a$$

= - (\rho^2/2a) - (\rho^4/8a^3) - (\rho^6/16a^5) - \cdots . (2.1)

Here $\rho^2 = x_2^2 + x_3^2$.

Since G depends upon the sphere radius a, it is helpful to indicate this explicitly by writing $G = G(\mathbf{x}, \mathbf{x}_0, \beta, a^{-1})$. When no confusion can occur, it is convenient to omit the arguments \mathbf{x}_0 and β and to write $G(\mathbf{x}, a^{-1})$. Our objective is to express G as a Taylor series in a^{-1} about

$$a^{-1}=0$$
,

$$G(\mathbf{x},a^{-1}) = G^{(0)}(\mathbf{x}) + G^{(1)}(\mathbf{x})a^{-1} + G^{(2)}(\mathbf{x})a^{-2}/2 + \cdots$$
(2.2)

Here $G^{(n)}(\mathbf{x}) = \partial^n G(\mathbf{x}, 0) / \partial (a^{-1})^n$ is the *n*th derivative of *G* with respect to a^{-1} at $a^{-1} = 0$.

To determine $G^{(0)}(\mathbf{x})$ we begin with (1.3), (1.4), and (1.5), which we rewrite on the hemisphere $x_1 > -a$ as follows, using (2.1):

$$G([a^2 - \rho^2]^{1/2} - a, x_2, x_3, a^{-1}) = 0.$$
 (2.3)

Now we set $a^{-1}=0$ in (1.3), (1.4) and (2.3) to obtain the following equations satisfied by $G^{(0)}(\mathbf{x} :$

$$\frac{\partial G^{(0)}}{\partial \beta} - D\Delta G^{(0)} = -\delta(\beta)\delta(\mathbf{x} - \mathbf{x}_0), \qquad (2.4)$$

 $G^{(0)}(\mathbf{x}) = 0$, at $\beta = 0$ - (2.5)

$$G^{(0)}(0,x_2,x_3) = 0. (2.6)$$

In (2.6) we have used the fact that for any fixed value of ρ , $\lim_{a\to\infty} [a^2 - \rho^2]^{1/2} - a = 0$.

Equations (2.4)–(2.6) show that $G^{(0)}(\mathbf{x})$ is the Green's function for the half-space $x_1 \ge 0$. It is given by

$$G^{(0)}(\mathbf{x}) = \frac{1}{8(\pi D\beta)^{3/2}} \exp\left(\frac{-\rho^2}{4D\beta}\right) \left[\exp\left(\frac{-(x_1 - x_0)^2}{4D\beta}\right) - \exp\left(\frac{-(x_1 + x_0)^2}{4D\beta}\right)\right].$$
 (2.7)

To evaluate $B_D(T)$ from (1.1) we shall need $G^{(0)}(\mathbf{x}_0)$, which is obtained by setting $\rho = 0$ and $x_1 = x_0$ in (2.7). This yields

$$G^{(0)}(\mathbf{x}_0) = 1/8 (\pi D\beta)^{3/2} [1 - \exp\{-(x_0^2/D\beta)\}] \quad (2.8)$$

To determine $G^{(n)}(\mathbf{x})$ for $n = 1, 2, \cdots$ we differentiate (1.3), (1.4) and (2.3) *n* times with respect to a^{-1} and

then set $a^{-1}=0$. Then (1.3) and (1.4) yield

$$\frac{\partial G^{(n)}}{\partial \beta} - D\Delta G^{(n)} = 0, \qquad n \ge 1 \qquad (2.9)$$

$$G^{(n)}(\mathbf{x}) = 0$$
 at $\beta = 0-$, $n \ge 1$. (2.10)

From (2.3) we obtain

$$G^{(1)}(0,x_2,x_3) = (\rho^2/2)G_{x_1}^{(0)}(0,x_2,x_3)$$

= $(x_0\rho^2/16\pi^{3/2})\exp(-(x_0^2+\rho^2)/4D\beta)$ (2.11)

$$G^{(2)}(0,x_2,x_3) = \rho^2 G_{x_1}^{(1)}(0,x_2,x_3).$$
(2.12)

In (2.11) we have used the explicit result (2.7) for $G^{(0)}$. In (2.12) we have used (2.7) to conclude that $G_{x_1x_1}^{(0)}(0,x_2,x_3)=0$. We have also used (2.1) to see that there is no term in a^{-2} in the expression for the surface of the sphere. Boundary values for higher derivatives of G can be found by further differentiation of (2.3).

We can now obtain $G^{(1)}$ by solving (2.9), (2.10), and (2.11). This is an initial-boundary value problem for the heat equation in the half-space $x_1 \ge 0$ with zero initial data and given inhomogeneous data on the plane $x_1=0$. The Green's function for this problem is $G^{(0)}$, so by Green's theorem we obtain

$$G^{(1)}(\mathbf{x}) = \frac{x_0 x_1}{128\pi^3 D^4} \int_0^\beta d\tau \int_{-\infty}^\infty dx_2' \int_{-\infty}^\infty dx_3' (\rho')^2 \\ \times [\tau(\beta - \tau)]^{-5/2} \exp\left(\frac{-(\rho')^2 - x_0^2}{4D\tau}\right) \\ \times \exp\left(\frac{-x_1^2 - (x_2 - x_2')^2 - (x_3 - x_3')^2}{4D(\beta - \tau)}\right). \quad (2.13)$$

Upon performing the x_{2}' and x_{3}' integrations in (2.13), we obtain

$$G^{(1)}(\mathbf{x}) = \frac{x_0 x_1 \exp(-\rho^2/4D\beta)}{8\pi^2 D^2 \beta^2} \int_0^\beta \left[1 + \frac{\tau \rho^2}{4D\beta(\beta - \tau)}\right] \times \frac{\exp((-x_0^2/4D\tau) - [x_1^2/4D(\beta - \tau)])}{[\tau(\beta - \tau)]^{1/2}} d\tau. \quad (2.14)$$

By setting $x = x_0$ in (2.14), we find

$$G^{(1)}(x_0) = \frac{x_0^2}{8\pi^2 D^2 \beta^2} \int_0^\beta \frac{\exp([-x_0^2 \beta/4 D\tau(\beta-\tau)])}{[\tau(\beta-\tau)]^{1/2}} d\tau.$$
(2.15)

The method we have used to obtain $G^{(1)}$ can be used to obtain all the $G^{(n)}$ successively. We shall now use it to obtain $G^{(2)}$, which satisfies (2.9), (2.10), and (2.12). In (2.12) we need the derivative of $G^{(1)}$ with respect to x_1 at $x_1=0$, which can be computed from (2.14). When (2.14) is differentiated with respect to x_1 , the right side becomes the difference of two integrals which both diverge as x_1 tends to zero. Therefore, we introduce the new variable $\mu = x_1 [4D(\beta - \tau)]^{-1/2}$ into these two integrals. Then $G_{x_1}^{(1)}$ can be written as the quotient of two expressions which both vanish at $x_1 = 0$. Application of L'Hospital's rule yields the result

$$G_{x_1}^{(1)}(0, x_2, x_3) = \frac{x_0 \exp(-\rho^2/4D\beta)}{8\pi^2 D^2 \beta^2} \int_0^\beta \frac{\exp(-x_0^2/4D\tau)}{[\tau(\beta-\tau)]^{1/2}} \times \left[1 - \frac{\rho^2}{2D\beta} \left(\frac{x_0^2}{2D\tau} + 1\right)\right] d\tau.$$
(2.16)

We now apply Green's theorem to $G^{(2)}$ and the Green's function $G^{(0)}$, taking account of (2.16) in evaluating the right side of (2.12). In this way we obtain

$$G^{(2)}(\mathbf{x}) = \frac{x_1 x_0}{64 (\pi D)^{7/2}} \int_0^\beta d\tau \int_{-\infty}^\infty dx_2' \int_{-\infty}^\infty dx_3' \frac{(\rho')^2}{\tau^2 (\beta - \tau)^{5/2}} \exp\left\{\frac{-(\rho')^2}{4D\tau} - \frac{[x_1^2 + (x_2 - x_2')^2 + (x_3 - x_3')^2]}{4D(\beta - \tau)}\right\} \\ \times \left[\int_0^\tau \frac{\exp(-x_0^2/4D\eta)}{[\eta(\tau - \eta)]^{1/2}} \left\{1 - \frac{(\rho')^2}{2D\tau} \left(\frac{x_0^2}{2D\eta} + 1\right)\right\} d\eta\right]. \quad (2.17)$$

After we set $x_1 = x_0$ and $\rho = 0$ in (2.17) we can perform the x_2' and x_3' integrations. Then (2.17) becomes

$$G^{(2)}(\mathbf{x}_{0}) = \frac{x_{0}^{2}}{4\pi^{5/2}D^{3/2}\beta^{2}} \int_{0}^{\beta} \frac{\exp(-x_{0}^{2}/4D(\beta-\tau))}{(\beta-\tau)^{1/2}} \left[\int_{0}^{\tau} \frac{\exp(-x_{0}^{2}/4D\eta)}{[\eta(\tau-\eta)]^{1/2}} \left\{ 1 - \frac{4(\beta-\tau)}{\beta} \left(\frac{x_{0}^{2}}{2D\eta} + 1 \right) \right\} d\eta \right] d\tau. \quad (2.18)$$

III. EVALUATION OF $B_D(T)$

We shall now use the preceding results in (1.1) to obtain an expansion for $B_D(T)$. First we use (1.5) in (1.1) to obtain

$$B_D(T) = \frac{2\pi a^3}{3} + \frac{1}{2} \int_{|\mathbf{x}_0 - \mathbf{x}_c| > a} \left[1 - 2^{3/2} \lambda^{3/2} G(\mathbf{x}_0, \mathbf{x}_0, \beta) \right] d\mathbf{x}_0.$$
(3.1)

Now we use the spherical symmetry of the integrand about the center of the sphere \mathbf{x}_c to replace the angular integration by multiplication by $4\pi |\mathbf{x}_0 - \mathbf{x}_c|^2 = 4\pi (x_0 + a)^2$. Then (3.1) becomes

$$B_D(T) = \frac{2\pi a^3}{3} \left\{ 1 + \frac{3}{a^3} \int_0^\infty \left[1 - 2^{3/2} \lambda^{3/2} G(x_{0,0}, 0, a^{-1}) \right] (x_0 + a)^2 dx_0 \right\} .$$
(3.2)

Next we use the expansion (2.2) for $G(\mathbf{x},a^{-1})$ in (3.2), using the results (2.8), (2.15), and (2.18) for $G^{(0)}(\mathbf{x}_0)$, $G^{(1)}(\mathbf{x}_0)$ and $G^{(2)}(\mathbf{x}_0)$. Then we multiply G by $(x_0+a)^2$ and keep terms in the braces up to and including those of order a^{-3} . In this way we obtain from (3.2)

$$B_{D}(T) = \frac{2}{3}\pi a^{3} \left\{ 1 + \frac{3}{a} \int_{0}^{\infty} \exp\left(\frac{-x_{0}^{2}}{D\beta}\right) dx_{0} + \frac{3}{a^{2}} \left\{ 2 \int_{0}^{\infty} \exp\left(\frac{-x_{0}^{2}}{D\beta}\right) x_{0} dx_{0} + \frac{1}{(\pi D\beta)^{1/2}} \int_{0}^{\infty} \int_{0}^{\beta} \frac{\exp\left[-\beta x_{0}^{2}/4D\tau(\beta-\tau)\right] x_{0}^{2}}{[\tau(\beta-\tau)]^{1/2}} d\tau dx_{0} \right\} + \frac{3}{a^{3}} \left(\int_{0}^{\infty} \exp\left(\frac{-x_{0}^{3}}{D\beta}\right) x_{0}^{2} dx_{0} + \frac{2}{(\pi D\beta)^{1/2}} \int_{0}^{\infty} \int_{0}^{\beta} \frac{\exp\left[-\beta x_{0}^{2}/4D\tau(\beta-\tau)\right] x_{0}^{3}}{[\tau(\beta-\tau)]^{1/2}} d\tau dx_{0} + \frac{1}{\pi\beta^{1/2}} \int_{0}^{\infty} dx_{0} \int_{0}^{\beta} d\tau \frac{x_{0}^{2} \exp\left[-x_{0}^{2}/4D(\beta-\tau)\right]}{(\beta-\tau)^{1/2}} \times \left[\int_{0}^{\tau} \frac{\exp\left(-x_{0}^{2}/4D\eta\right)}{[\eta(\tau-\eta)]^{1/2}} \left[1 - \frac{4(\beta-\tau)}{\beta} \left(\frac{x_{0}^{2}}{2D\eta} + 1\right) \right] d\eta \right] \right) + O(a^{-4}) \right\}.$$
(3.3)

Although some of the integrals in (3.3) look quite formidable, they can all be evaluated explicitly without much difficulty if the x_0 integration is always done first. Upon evaluating these integrals, we obtain from (3.3)

$$B_D(T) = \frac{2}{3}\pi a^3 \left[1 + \frac{3\sqrt{\pi}}{2} \left(\frac{D\beta}{a^2} \right)^{1/2} + 2 \left(\frac{D\beta}{a^2} \right) + \frac{\sqrt{\pi}}{8} \left(\frac{D\beta}{a^2} \right)^{3/2} + O(a^{-4}) \right].$$
(3.4)

From the definitions $D = h^2/m$ and the definition (1.2) of λ , it follows that $D\beta = \lambda^2/2\pi$. When we use this relation in (3.4) we obtain the result (1.6) stated in the Introduction.

In conclusion, we note that our procedure could be used to evaluate additional terms in (3.4). We also observe that we could have introduced dimensionless variables from the beginning, and then our expansion would have been in terms of a dimensionless ratio rather than in terms of a^{-1} . Finally, we should point out that our procedure yields the asymptotic expansion of $B_D(T)$ for λ/a small, rather than a convergent power series, since $B_D(T)$ has an essential singularity at $\lambda/a=0$. This singularity will be manifested by exponentially small terms similar to those occurring in $B_E(T)$ and described in footnote 2.

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Cyclotron Excitation of Electromagnetic Waves by a Gyrating Electron Beam*

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When a tenuous electron stream gyrates around the field lines of a static magnetic field in the presence of a stationary plasma, electromagnetic waves are excited which propagate perpendicular to the magnetic field. Previous theoretical studies involved the assumption that these excited waves are almost longitudinal (quasistatic approximation). Such an assumption is not made in this analysis. It is shown that the excited waves have the electron cyclotron frequency or some multiple thereof. The electric intensity vector rotates in a plane perpendicular to the magnetic field; the phase velocities are of the order of, or exceed, the velocity of light, and for certain plasma-beam systems lower harmonics of the electron cyclotron frequency cannot be excited. Expressions are given for the rate of growth of the waves as a function of the plasma-beam parameters and the harmonic number.

I. INTRODUCTION

THERE has been considerable interest recently¹ in processes which produce emission of waves from plasma, perpendicular to a magnetic field, with frequencies which are multiples of electron gyrofrequency. The relevant theory has been developed in terms of a dispersion equation based on quasistatic approximation. This approximation involves the assumption that the electrostatic effects alone control the wave propagation and is valid when the phase velocity of the wave, when compared to the velocity of light in vacuum, is sufficiently small. In a quasistatic approximation the waves are almost longidudinal, i.e., the electrical intensity \mathbf{E} is almost parallel to the wave vector \mathbf{k} .

A suggestion that waves moving perpendicular to the magnetic field are electrostatic was made by Canobbio and Croci² in their analysis of radiation observed in a Penning ion-gauge (PIG) discharge by Landauer.³ Subsequent investigations based on quasistatic approximation were made by Dory, Guest, and Harris,⁴ Crawford and Tataronis,⁵ Ikegami,⁶ and others.

This investigation is based on a different approach to the problem. It has not been assumed that the excited waves are quasistatic (i.e., no *a priori* restrictions are imposed on the orientation of **E** with respect to **k**), and it is shown that there is an emission of excited harmonic waves which are not almost longitudinal. The main characteristics of these excited waves are: (a) each is elliptically (or circularly) polarized and the electric

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