

On the Kirkwood Superposition Approximation

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The Kirkwood superposition approximation is investigated by making use of the expansion theorem for the potential of average force and its correction term is found in a form of the expansion in powers of particle number density ρ . The lowest order term of this correction is calculated for a special configuration of three particles in a fluid consisting of hard spheres and the Kirkwood approximation is shown to overestimate the distribution function of triplets in this case. The Kirkwood integral equation with correction is solved by expanding the radial distribution function in powers of ρ . The solution thus obtained is shown to be exact up to the order of ρ^2 , while the Kirkwood approximation itself does not yield the correct term of the order of ρ^2 . It is shown that the distribution function of triplets can, in principle, be expanded in terms of radial distribution functions and a few terms of this expansion are calculated explicitly, of which the first term just corresponds to the Kirkwood superposition approximation.

§ 1. Introduction

The integral equation method for evaluating distribution functions in fluid systems has provided a useful means, for instance, to determine the radial distribution function itself, to calculate the equation of state, to study the condensation phenomena, etc. Since both the Kirkwood¹⁾²⁾³⁾ and the Yvon-Born-Green⁴⁾⁵⁾⁶⁾ integral equations involve the distribution functions in sets of three particles in addition to the radial distribution functions, it is necessary to introduce the assumption of a suitable closure, that is, a relation between these distribution functions, in order to solve the integral equations. The closure which has been widely used is the Kirkwood superposition approximation in which the potential of average force in a set of three particles is assumed to be the sum of the three pair potentials of average force. This is equivalent to assuming that the triplet distribution function is a product of the three radial distribution functions.

So far, there have been several methods for estimating the extent of validity of the superposition approximation. The most rigorous method is to compare the exact expansions of the radial distribution function and the pressure in powers of the number density ρ with the corresponding one obtained with the aid of the superposition approximation. This has been done for the case of the third and fourth virial coefficients in a system of hard spheres, by Hart, Wallis and Pode⁷⁾, Rushbrooke and Scoins⁸⁾, Nijboer and van Hove⁹⁾, and Hiroike¹⁰⁾. They found that

the superposition approximation yields an exact third virial coefficient, independently of the potential used. (Meeron¹¹) has shown that this comparison is less significant than it seems since the third virial coefficient is given correctly by any closure whatsoever.) However, the fourth virial coefficient is no longer exact and, in the case of hard spheres, the discrepancy amounts to about 21.5%. Also, as was shown by Nijboer and van Hove⁹, the fourth virial coefficient for hard spheres shows a wide discrepancy when calculated via the virial theorem, using the superposition approximation, and via the compressibility integral. Thus the superposition approximation seems to be inaccurate even at low densities and gives inconsistent results when substituted in different equations.

On the other hand, the direct comparison^{12,13} of the equation of state and radial distribution function obtained via the superposition approximation with the experimental data shows a surprisingly good agreement up to quite high densities. Likewise, a comparison with the results calculated by the Monte Carlo method^{14,15,16} shows good agreement up to moderately high densities. Thus the superposition approximation seems to be more accurate than could be expected from the comparison with the fourth virial coefficient.

Confronting with these facts, several authors have attempted to justify^{11,17} or to improve¹⁸ the superposition approximation or to use^{19,20} it so that it will lead to less inaccuracies than those associated with the previous equations. However, it seems that the statistical mechanical foundation of the superposition approximation has not yet been fully discussed.

The purpose of this paper is to investigate the Kirkwood superposition approximation by means of the general theorem in statistical mechanics. In § 2 using the expansion theorem for the potential of average force, the meaning of the superposition approximation is clarified and its correction term is given in a form of expansion in powers of ρ . In § 3 the lowest order term of this correction is retained and calculated in the case of hard spheres for a special configuration of three particles. The Kirkwood approximation is shown to overestimate the triplet distribution function in this case. Then § 4 is devoted to generalize the method to the case in which the coupling parameter is included and the Kirkwood integral equation with correction is solved by expanding the radial distribution function in powers of ρ . It is confirmed that the exact terms up to ρ^2 are obtained by this procedure. In § 5 a possible representation of triplet distribution function in terms of radial distribution function is discussed and a few terms of this representation are calculated explicitly, of which the first term corresponds to the Kirkwood superposition approximation.

§ 2. Expansion of potential of average force

When a subset of m particles (denoted by M) at fixed configuration is chosen out of all the particles in a fluid system, there is a potential of average force act-

ing on this set, averaged over the possible configurations of the remaining particles. If the distribution function normalized to unity of this subset is denoted by $g_m(\mathbf{M})$, the potential of average force $W_m(\mathbf{M})$ is defined by

$$g_m(\mathbf{M}) = \exp[-\beta W_m(\mathbf{M})] \tag{1}$$

where $\beta \equiv 1/kT$ with k Boltzmann's constant, T the absolute temperature. In this section we shall briefly explain the expansion theorem proved by Meeron²¹⁾ and find the correction term to the superposition approximation in a form of expansion in powers of ρ .

For a one-component system with no long-range order, $W_m(\mathbf{M})$ is expanded in powers of ρ as follows²¹⁾,

$$W_m(\mathbf{M}) = U(\mathbf{M}) - \frac{1}{\beta} \sum_{N \geq 1} \frac{\rho^N}{N!} \int Q(\mathbf{M}, \mathbf{N}) d(\mathbf{N}). \tag{2}$$

Here $U(\mathbf{M})$ is the direct interaction potential in the set \mathbf{M} and $d(\mathbf{N})$ denotes the integration over the coordinates of n particles of the set \mathbf{N} . The $Q(\mathbf{M}, \mathbf{N})$ is a sum of products of f functions,

$$f(ij) \equiv \exp[-\beta U(ij)] - 1, \tag{3}$$

where $U(ij)$ is the two-body potential function.* This sum is characterized by**

- (a) every particle of the set \mathbf{N} is connected to at least two particles of the set \mathbf{M} , either directly or by two or more independent paths, i.e., paths which involve mutually exclusive sets of intermediate particles,
- (b) all particles of the set \mathbf{N} are also connected among themselves independently of the set \mathbf{M} , i.e., without involving particles of \mathbf{M} ,
- (c) particles of \mathbf{M} are not directly connected among themselves.

Some of bond diagrams contributing to $W_3(123)$ are given in Fig. 1 and Fig. 2.

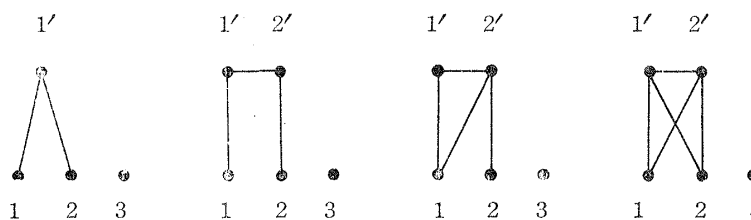


Fig. 1. Diagrams contributing to $W_3(123)$. The points 1' and 2' mean the particles of the set \mathbf{N} . These diagrams are characterized by the absence of f -bond connected to the particle 3.

It may be easily seen that the summation of all the diagrams of the type shown in Fig. 1 together with $U(12)$ yields $W_2(12)$. The similar diagrams in which the particle 1 or 2 is not connected together with $U(23)$ or $U(31)$ will lead to $W_2(23)$ or $W_2(31)$. Therefore, if we write²²⁾

* The potential energy of the system is assumed to be a sum of pair potentials.
 ** A detailed account may be found in references 11) and 21).

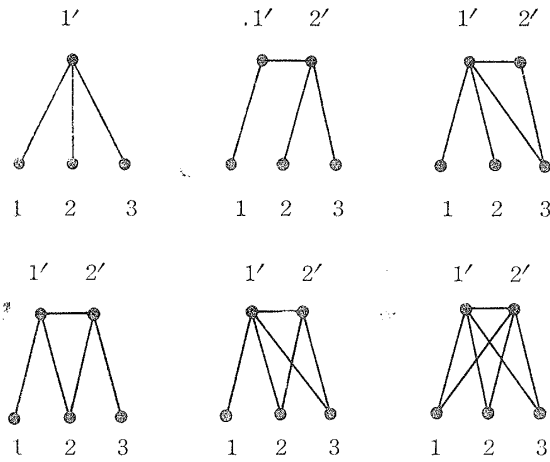


Fig. 2. Diagrams contributing to $W_3(123)$ in which every particle of the set $M (=1, 2, 3)$ is connected by f -bond.

$$W_3(123) = W_2(12) + W_2(23) + W_2(31) + w_3(123), \tag{4}$$

$w_3(123)$ may be characterized by the bond diagrams shown in Fig. 2, i.e.,

$$w_3(123) = -\frac{1}{\beta} \sum_{N \geq 1} \frac{\rho^N}{N!} \int q(3, N) d(N), \tag{5}$$

where $q(3, N)$ is a sum of products of f functions subject to, (d) every particle of the set $(1, 2, 3)$

is connected to at least one particle of the set N , besides the conditions (a), (b) and (c) mentioned above.

Substituting eq. (4) in eq. (1), we have

$$g_3(123) = g(12)g(23)g(31) \exp[-\beta w_3(123)], \tag{6}$$

where $g(12)$ is the radial distribution function, i.e., $g(12) = \exp[-\beta W_2(12)]$. From this equation it follows that the Kirkwood superposition approximation, $g_3(123) = g(12)g(23)g(31)$, corresponds to assuming that $w_3(123) = 0$, i.e., to neglecting the contributions of the bond diagrams shown in Fig. 2. In other words, $w_3(123)$ represents the correction term to the Kirkwood approximation.

If we retain the lowest order term in eq. (5), eq. (6) becomes

$$g_3(123) = g(12)g(23)g(31) \exp[\rho \int f(11')f(21')f(31') d\tau_1']. \tag{7}$$

§ 3. Validity of superposition approximation

In the previous section, we have derived the correction term to the superposition approximation in a form of power series of ρ . In this section we shall calculate the lowest order term of this correction for a special configuration of three particles in a fluid of hard spheres.

For hard spheres of diameter a , f function is given by

$$f(ij) = -1, \text{ for } |\mathbf{x}_i - \mathbf{x}_j| < a, \\ = 0, \text{ for } |\mathbf{x}_i - \mathbf{x}_j| > a.$$

Therefore, as is seen from eq. (7), the calculation of the correction term reduces to calculating the volume of the region belonging to all three spheres of radius a , of which centers are separated by $|\mathbf{x}_1 - \mathbf{x}_2|$, $|\mathbf{x}_2 - \mathbf{x}_3|$ and $|\mathbf{x}_3 - \mathbf{x}_1|$, respectively. For simplicity, however, we shall consider a special configuration in which these centers form a regular triangle. If we measure the distance in units of a , we obtain from

eq. (7)

$$g_3(123)/g(12)g(23)g(31) \equiv F(r) = \exp[-\rho a^3 Q(r)] \tag{8}$$

for this special configuration. Here $Q(r)$ implies the volume of the region common to three unit spheres of which centers are separated from each other by r , with $r = |x_1 - x_2|/a = |x_2 - x_3|/a = |x_3 - x_1|/a$.

The calculation of $Q(r)$ is rather lengthy, though it is elementary. Here we quote only the result (the derivation is given in the Appendix):

$$Q(r) = \pi + \frac{r^2}{6} \sqrt{3-r^2} + \frac{\pi}{8} r(r^2-12) - \frac{1}{4} r(r^2-12) \sin^{-1}\left(\frac{r}{\sqrt{3}\sqrt{4-r^2}}\right) + \sin^{-1}\left[\frac{6-2r-r^2}{\sqrt{3}(2-r)\sqrt{4-r^2}}\right] - \sin^{-1}\left[\frac{6+2r-r^2}{\sqrt{3}(2+r)\sqrt{4-r^2}}\right], \quad 1 \leq r \leq \sqrt{3},$$

$$= 0, \quad r \geq \sqrt{3}. \tag{9}$$

Using this equation, we calculated the function $F(r)$ for $\rho a^3 = 0.5$ and 1.0, respectively, and the result is shown graphically in Fig. 3.

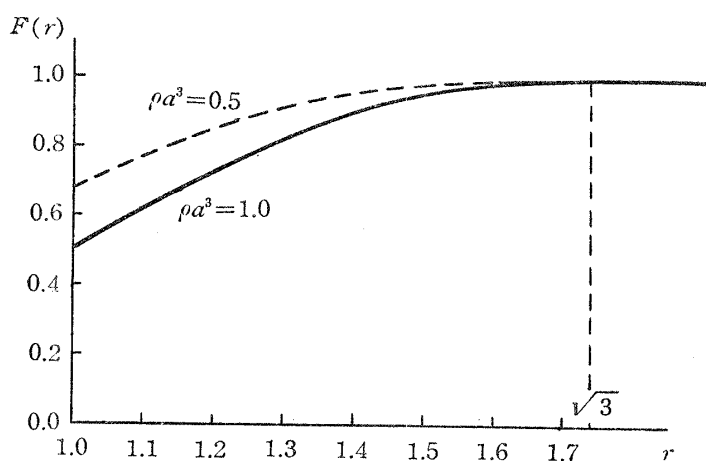


Fig. 3. The function $F(r)$ for $\rho a^3 = 0.5$ and 1.0.

From this figure, it may follow that the superposition approximation is exact for $r \geq \sqrt{3}$ to the extent of the approximation employed here, but the error produced by the superposition approximation increases as r decreases and amounts to about 50% for $r = 1$, i. e., for the configuration of closest packing. Furthermore, $F(r) \leq 1$ for $1 \leq r \leq \sqrt{3}$, that is, the superposition approximation overestimates the

triplet distribution function in this case. However, the numerical value obtained here should not be taken too literally, for we have considered only the lowest order term of correction, neglecting the higher order terms. These terms seem to have the compensating effects, i. e., the tendency to increase $F(r)$, since the contribution of the products of even number of f functions is opposite in sign to that of the lowest order term. Furthermore, if the potential has the attractive part, the f function becomes positive in the region of attraction, thus $F(r)$ may increase even in the lowest order approximation.

Although the quantitative feature of the superposition approximation is somewhat obscure as mentioned above, the qualitative feature may be that it overestimates the triplet distribution function for the repulsive potential and underestimates for the attractive potential. To clarify the situation it may be necessary to solve the Yvon-Born-Green integral equation with correction and compare the results with those

obtained without correction. We shall not enter into this problem here, but in the next section we shall solve the Kirkwood integral equation with correction expanding the radial distribution function in powers of ρ .

§ 4. Superposition approximation in Kirkwood's integral equation

In the Kirkwood integral equation for the distribution function, there appears the coupling parameter $\hat{\xi}$ which is defined so that we have the fully coupled real fluid of N particles for $\hat{\xi}=1$, but if $\hat{\xi}=0$, particle 1 does not interact with any of remaining $N-1$ particles. The generalization of the results obtained in § 2 to this case is straightforward if we regard the particle 1 as an impurity and apply the expansion theorem for the two-component system²¹⁾. Then it is readily verified that

$$g_3(123, \hat{\xi}) = g(12, \hat{\xi})g(23)g(31, \hat{\xi}) \exp[-\beta w_3(123, \hat{\xi})], \quad (10)$$

where $w_3(123, \hat{\xi})$ is given, as previously done, by eq. (5), but the f function connected to the particle 1 is replaced by

$$f(1i, \hat{\xi}) = \exp[-\beta \hat{\xi} U(1i)] - 1. \quad (11)$$

If we retain only the lowest order term as in § 3, we have

$$-\beta w_3(123, \hat{\xi}) = \rho \int f(11', \hat{\xi}) f(21') f(31') d\tau_1'. \quad (12)$$

We shall solve the Kirkwood integral equation in the following, using eqs. (10) and (12).

Let us consider the exact Kirkwood integral equation, without superposition,

$$\begin{aligned} \partial g(12, \hat{\xi}) / \partial \hat{\xi} = & -U(12)g(12, \hat{\xi}) + \beta \rho \int U(13)g(13, \hat{\xi}) d\tau_3 \\ & - \beta \rho \int U(13)g_3(123, \hat{\xi}) d\tau_3. \end{aligned} \quad (13)$$

In solving this equation, we expand $g(12, \hat{\xi})$ as follows,

$$g(12, \hat{\xi}) = e^{-\beta \hat{\xi} U(12)} [1 + \rho g_1(12, \hat{\xi}) + \rho^2 g_2(12, \hat{\xi}) + \dots]. \quad (14)$$

Substituting eqs. (10), (12) and (14) in eq. (13) and comparing the coefficient of ρ , we have

$$\partial g_1(12, \hat{\xi}) / \partial \hat{\xi} = -\beta \int U(13) e^{-\beta \hat{\xi} U(13)} f(23) d\tau_3.$$

If we integrate this equation by $\hat{\xi}$, we have

$$g_1(12, \hat{\xi}) = \int f(13, \hat{\xi}) f(23) d\tau_3. \quad (15)$$

Using eq. (15) and comparing the coefficients of ρ^2 , we obtain

$$\begin{aligned} \frac{\partial}{\partial \hat{\xi}} g_2(12, \hat{\xi}) = & \frac{\partial}{\partial \hat{\xi}} \int \{ f(13, \hat{\xi}) f(14, \hat{\xi}) f(23) f(24) / 2 + f(13, \hat{\xi}) f(14, \hat{\xi}) f(23) f(34) \\ & + f(13, \hat{\xi}) [1 + f(23)] f(24) f(34) + f(13, \hat{\xi}) f(14, \hat{\xi}) f(23) f(24) f(34) / 2 \} d\tau_3 d\tau_4. \end{aligned} \quad (16)$$

The integration over $\tilde{\xi}$ from 0 to 1 leads to the exact form for the $g_2(12)$, if one notes that $g_2(12, 0) = f(12, 0) = 0$. Therefore, if one calculates the fourth virial coefficient by using $g(12)$ obtained here, it may be given correctly both via the virial theorem and via the compressibility integral. On the other hand, if the Kirkwood approximation, i. e., $g_3(123, \tilde{\xi}) = g(12, \tilde{\xi})g(23)g(31, \tilde{\xi})$ is substituted in eq. (13), the $g_2(12)$ is no longer exact though the $g_1(12)$ is given correctly, and hence the fourth virial coefficient is not given correctly though the third one is correct.

From the above argument, it may follow that the fourth virial coefficient is given exactly if the term of the order of ρ is taken into account in $w_3(123)$. Similarly, if the terms up to ρ^2 are retained in $w_3(123)$, the fifth virial coefficient may be calculated exactly, and so forth. Thus the superposition approximation can be improved up to any virial coefficients as one desires.

§ 5. Possible representation of g_3 in terms of g

Since the integral equations for the distribution functions involve g_3 in addition to g , if it is possible to represent g_3 in terms of g , we shall have an exact equation to determine the radial distribution function and hence all thermodynamic functions will be calculated. In this section we shall discuss this representation, using the results obtained in the previous sections (we consider the case $\tilde{\xi} = 1$.)

It is easily seen that g_3 can, in principle, be expressed in terms of g ; for since both g_3 and g involve the potential function U , the elimination of U from g_3 and g may give the functional relation of g_3 with g , independently of the potential used. Based on this consideration, we shall calculate a few terms of the expansion of g_3 in terms of g .

Let us first consider the expansion of $W_2(12)$ in powers of ρ :

$$\begin{aligned}
 W_2(12) = & U(12) - \frac{\rho}{\beta} \int f(11')f(21') d\tau_1' - \frac{\rho^2}{\beta} \int [f(11')f(22') \\
 & + f(11')f(21')f(22') + f(11')f(21')f(12') \\
 & + \frac{1}{2}f(11')f(12')f(21')f(22')] f(1'2') d\tau_1' d\tau_2' + 0(\rho^3). \tag{17}
 \end{aligned}$$

We can regard this equation conversely as a defining equation of $U(12)$ in terms of $W_2(12)$ and f functions, and solve $U(12)$ as a function of W_2 and g functions by an iteration procedure:

$$\begin{aligned}
 U(12) = & W_2(12) + \frac{\rho}{\beta} \int h(11')h(21') d\tau_1' - \frac{\rho^2}{\beta} \int [h(11')h(22') \\
 & - \frac{1}{2}h(11')h(12')h(21')h(22')] h(1'2') d\tau_1' d\tau_2' + 0(\rho^3), \tag{18}
 \end{aligned}$$

where

$$h(12) = g(12) - 1. \tag{19}$$

From eqs. (3) and (18), we have

$$f(12) = h(12) - \rho g(12) \int h(11') h(21') d\tau_1' + O(\rho^2). \tag{20}$$

Then the substitution of eq. (20) in eq. (5) leads to

$$-\beta w_3(123) = \rho \int h(11') h(21') h(31') d\tau_1' + \frac{\rho^2}{2} \sum'_{ij} \left[\int h(i1') h(j2') \right] h(1'2') d\tau_1' d\tau_2' + O(\rho^3), \tag{21}$$

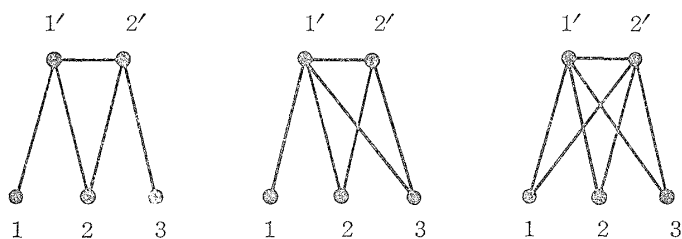


Fig. 4. Diagrammatic representation of h -bond contributing to w_3 .

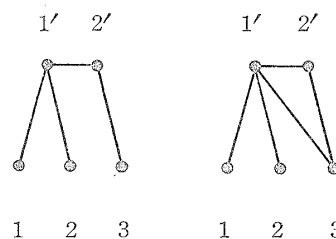


Fig. 5.

where \sum' implies a summation over the bond diagrams of the type shown in Fig. 4. However, the bond diagrams given in Fig. 5, which appear when $w_3(123)$ is expressed in terms of f functions, do not appear in the new series.

It may seem that eq. (21) is a power series of ρ , but, in reality it is not the case, since h function depends on ρ . If eq. (21) is substituted in eq. (6), it is evident that $g_3(123)$ is expressed in terms of g . Obviously, the procedure developed here can be carried out to any desired higher order terms, although the procedure rapidly becomes very tedious. Furthermore, the procedure may be readily extended to the case in which the coupling parameter is not equal to 1.

In this section we have considered the problem in the classical theory of statistical mechanics. However, we believe that the conjecture developed here may be generalized to the case of quantum theory. For instance, the functional relation between the reduced density matrices may be obtained by eliminating the potential function from these quantities, as we have done in this section. We hope that such a procedure may prove useful in connection with the quantum mechanical treatments of the many-body problem.

In conclusion the author wishes to express his sincere thanks to Prof. H. Ichimura, Dr. K. Hiroike and Dr. T. Morita for their valuable discussions. A part of this work was supported by the Scientific Research Expenditure of the Ministry of Education.

Appendix. Calculation of $\mathcal{Q}(r)$

If we set the volume of the region cut from the sphere with radius 1 by two plains ① and ② illustrated in Fig. 6 to be $V(p, l, \theta)$, it is easy to see that

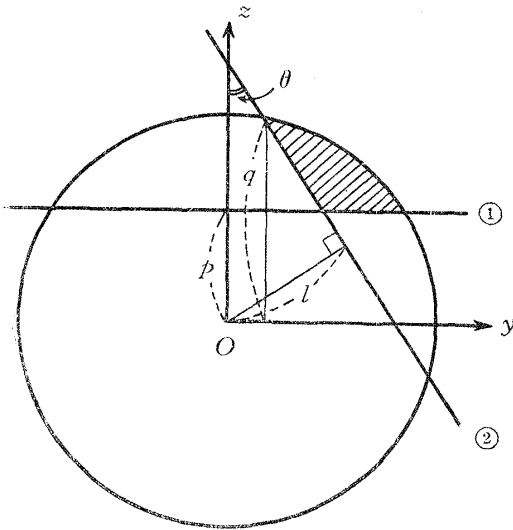
$$\Omega(r) = 3V(r/2, r/2, \pi/6), \tag{A1}$$

and therefore the calculation of $\Omega(r)$ reduces to that of $V(p, l, \theta)$.

Let the equation of plain ② in Fig. 6 be

$$y/y_0 + z/z_0 = 1.$$

Furthermore, if q denotes the distance illustrated in Fig. 6, $V(p, l, \theta)$ is given by



$$V(p, l, \theta) = \int_p^q \left[\frac{\pi}{2} (1-z^2) - (1-z^2) \sin^{-1} \left(\frac{y_0(1-z/z_0)}{\sqrt{1-z^2}} \right) - y_0(1-z/z_0) \times \sqrt{1-z^2-y_0^2(1-z/z_0)^2} \right] dz, \tag{A2}$$

for $z_0 > 1$. The integration of the second term on the right-hand side can be carried out in terms of elementary functions by the use of partial integration, and after a very lengthy calculation, we obtain

$$V(p, l, \theta) = \pi/6 \cdot p(p^2-3) \cdot \{1+2/\pi \cdot \sin^{-1}[(p \sin \theta - l)/\sqrt{1-p^2 \cos \theta}]\}$$

Fig. 6. ① and ② indicate two plains of which perpendicular distances from the origin 0 are p and l , respectively. The volume of shaded region is $V(p, l, \theta)$.

$$+ \pi/6 \cdot l(l^2-3) \cdot \{1+2/\pi \cdot \sin^{-1}[(l \sin \theta - p)/\sqrt{1-l^2 \cos \theta}]\} + 2tpl \cos \theta/3 - t \sin \theta \cos \theta/3 + t^3 \sin \theta \cos \theta/3 + (\pi - J)/3, \tag{A3}$$

where

$$t^2 = [\cos^2 \theta + 2pl \sin \theta - p^2 - l^2] / \cos^2 \theta \tag{A4}$$

and

$$J = \sin^{-1} \left[\frac{(1+l \sin \theta)(1+p) - (l+\sin \theta)^2}{(1+p)\sqrt{1-l^2 \cos \theta}} \right] - \sin^{-1} \left[\frac{(1-l \sin \theta)(1-p) - (l-\sin \theta)^2}{(1-p)\sqrt{1-l^2 \cos \theta}} \right]. \tag{A5}$$

From eqs. (A1), (A3), (A4) and (A5), we immediately have eq. (9) in § 3. It should be noted here that the J given by eq. (A5) can be written in a more symmetrical form:

$$J = \sin^{-1} [2t \cos \theta (pl - \sin \theta) / (1-p^2)(1-l^2)]. \tag{A6}$$

However, this form for J is not convenient, for the value of $\sin^{-1}x$ is not uniquely determined in eq. (A6). On the other hand, in eq. (A5) this value is unique and takes the principal value, i. e., $-\pi/2 \leq \sin^{-1}x \leq \pi/2$.

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