

Thermodynamic Properties of an Electron Plasma*

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The thermodynamic properties of an electron plasma are investigated on the basis of the Bogoliubov–Born–Green–Kirkwood–Yvon hierarchy equation. The correlation energy has been calculated up to the ϵ^2 term (ϵ is the standard plasma parameter) in terms of an exact solution of the binary correlation function taking into account the short-range collision effect. The contribution of ternary correlations to the correlation energy has been determined in lowest order, which turns out to be of the second order in ϵ . The total sum of the corrections due to the short-range collision effect and the ternary correlation effect is found to be in agreement with the result obtained by Abe, Bowers, and Salpeter within the regime of Gibbs' statistical thermodynamics. Although the appearance of the binary correlation function obtained in the present studies differs from those derived by Bowers–Salpeter and by O'Neil–Rostoker, it is shown that all of these expressions are essentially equivalent. According to the present analysis, however, it is proven that these expressions do not describe correctly the way in which correlation effects diminish at large distances and therefore the statement made by DeWitt saying that $g(x) \sim \frac{1}{2}(\ln 3)\epsilon^2 \exp(-k_D r)$ as $r \rightarrow \infty$ is not correct.

I. INTRODUCTION

During the past years, kinetic properties of plasmas in the nonequilibrium state have been extensively examined by many authors on the basis of the Liouville equation or the Bogoliubov–Born–Green–Kirkwood–Yvon hierarchy equation,^{1,2} while thermodynamic properties of plasmas have been investigated by applying Mayer's cluster expansion method within the framework of Gibbs' statistical thermodynamics.^{3,4} Recently, however, several authors have attempted to investigate thermodynamic properties of plasmas by solving the BBGKY hierarchy equation for the equilibrium case.^{5–7} It is worth investigating the way in which various physical effects contribute to thermodynamic properties of plasmas through the BBGKY hierarchy equation.

Guernsey⁶ has solved the BBGKY hierarchy equation for the equilibrium state by a method of double series expansion and has presented a "dynamic" derivation of the equation of state for an electron plasma, which is correct through second order in the plasma parameter $\epsilon = (4\pi n \lambda_D^3)^{-1}$. n is the number density of electron and λ_D is the Debye distance. On the other hand, O'Neil and Rostoker⁷ have determined a binary correlation function for an equilibrium electron plasma by solving analytically the BBGKY hierarchy equation. In obtaining the solution, they have introduced an arbitrary distance r_0 to divide the whole space into the inner region $r < r_0$ where the short-range collisions play a dominant role and the outer region $r > r_0$ where the effects of short-range collisions can be disregarded. Different expansions in the plasma parameter ϵ have

been applied for the equations obtained in each region. The solutions valid in each region have been matched approximately at the distance $r = r_0$. The same authors have also obtained the correlation energy which is correct up to the ϵ^2 term, by arbitrarily defining r_0 as the mean distance between particles $n^{-1/3}$.

The present paper removes the arbitrariness inherent in the above-mentioned interpolation method for solving the BBGKY hierarchy equation for the equilibrium case. We present the fundamental equations in Sec. II. In Sec. III, ignoring the ternary correlation effect, we examine the influence of the short-range collisions on the binary correlation function in some details. Following the calculation of Lamb and Burdick,⁵ we can calculate corrections to the Debye–Hückel limiting value of the correlation energy up to the second order in ϵ as follows:

$$E^{(C)}/\kappa T = -\frac{1}{2}\epsilon^2 \ln \epsilon - \frac{1}{2}(\gamma - \frac{1}{4})\epsilon^2, \quad (1)$$

where $\gamma = 0.5772$ is the Euler constant, κ is the Boltzmann constant, and T is the electron temperature. In Sec. IV, we analyze the effect of the ternary correlation on the binary correlation function in some detail. The contribution of the ternary correlation to the correlation energy is calculated as

$$E^{(T)}/\kappa T = -\frac{1}{4}(\ln 3 - \frac{5}{6})\epsilon^2. \quad (2)$$

Adding up Eqs. (1) and (2) to the Debye–Hückel limiting value of the correlation energy $-\epsilon/2$, we obtain

$$E/\kappa T = -\frac{1}{2}\epsilon - \frac{1}{4}\epsilon^2 \ln \epsilon - \frac{1}{2}(\gamma - \frac{2}{3} + \frac{1}{2} \ln 3)\epsilon^2 \quad (3)$$

which is the result obtained by Abe,³ and by Bowers and Salpeter.⁴ Thus, the present analysis demonstrates explicitly that the approach based on the method of solving the BBGKY hierarchy equation can provide such detailed information as contributions of the dynamical effect of short-range collisions and of ternary correlations to the thermodynamic properties of an electron plasma. In the last section, we discuss the present result for the binary correlation function by comparing it with that obtained by Bowers and Salpeter.

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¹ R. Balescu, *Phys. Fluids* **3**, 52 (1960).

² A. Lenard, *Ann. Phys. (N. Y.)* **3**, 390 (1963).

³ R. Abe, *Progr. Theoret. Phys. (Kyoto)* **22**, 213 (1959).

⁴ D. L. Bowers and E. E. Salpeter, *Phys. Rev.* **119**, 1180 (1960).

⁵ G. L. Lamb, Jr., and B. Burdick, *Phys. Fluids* **7**, 1087 (1963).

⁶ R. L. Gurnsey, *Phys. Fluids* **7**, 792 (1963).

⁷ T. O'Neil and N. Rostoker, *Phys. Fluids* **8**, 1109 (1965).

II. FUNDAMENTAL EQUATIONS

We consider a system composed of N electrons imbedded in a neutralizing uniform background of ions in a large volume V . Assuming the system is spatially uniform, we have the following expressions for the BBGKY hierarchy equation:

$$\frac{\partial}{\partial t} F(v_1) = \frac{n}{m} \int d^3 \frac{\partial \phi(r_{12})}{\partial \mathbf{r}_1} \cdot \frac{\partial}{\partial \mathbf{v}_1} G(1, 2) \tag{4a}$$

$$\begin{aligned} \left\{ \frac{\partial}{\partial t} + \mathbf{v}_1 \cdot \frac{\partial}{\partial \mathbf{r}_1} + \mathbf{v}_2 \cdot \frac{\partial}{\partial \mathbf{r}_2} \right\} G(1, 2) &= \frac{1}{m} \frac{\partial \phi(r_{12})}{\partial \mathbf{r}_1} \cdot \left(\frac{\partial}{\partial \mathbf{v}_1} - \frac{\partial}{\partial \mathbf{v}_2} \right) [F(v_1)F(v_2)] + \frac{1}{m} \frac{\partial \phi(r_{12})}{\partial \mathbf{r}_1} \cdot \left(\frac{\partial}{\partial \mathbf{v}_1} - \frac{\partial}{\partial \mathbf{v}_2} \right) G(1, 2) \\ &+ \frac{n}{m} \left\{ \frac{\partial F(v_1)}{\partial \mathbf{v}_1} \cdot \int \frac{\partial \phi(r_{13})}{\partial \mathbf{r}_1} G(2, 3) d^3 + \frac{\partial F(v_2)}{\partial \mathbf{v}_2} \cdot \int \frac{\partial \phi(r_{23})}{\partial \mathbf{r}_2} G(1, 3) d^3 \right\} \\ &+ \frac{n}{m} \int \left(\frac{\partial \phi(r_{13})}{\partial \mathbf{r}_1} \cdot \frac{\partial}{\partial \mathbf{v}_1} + \frac{\partial \phi(r_{23})}{\partial \mathbf{r}_2} \cdot \frac{\partial}{\partial \mathbf{v}_2} \right) H(1, 2, 3) d^3, \end{aligned} \tag{4b}$$

and

$$\begin{aligned} \left\{ \frac{\partial}{\partial t} + \sum_{i=1}^3 \mathbf{v}_i \cdot \frac{\partial}{\partial \mathbf{r}_i} \right\} H(1, 2, 3) &= \frac{1}{m} \left\{ G(2, 3) \frac{\partial F(v_1)}{\partial \mathbf{v}_1} \cdot \left(\frac{\partial \phi(r_{12})}{\partial \mathbf{r}_1} + \frac{\partial \phi(r_{13})}{\partial \mathbf{r}_1} \right) + (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2) \right\} \\ &+ \frac{1}{m} \left\{ F(v_1) \left(\frac{\partial \phi(r_{21})}{\partial \mathbf{r}_2} \cdot \frac{\partial}{\partial \mathbf{v}_2} + \frac{\partial \phi(r_{31})}{\partial \mathbf{r}_3} \cdot \frac{\partial}{\partial \mathbf{v}_3} \right) G(2, 3) + (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2) \right\} \\ &+ \frac{1}{m} \left\{ \frac{\partial \phi(r_{12})}{\partial \mathbf{r}_1} \cdot \left(\frac{\partial}{\partial \mathbf{v}_1} - \frac{\partial}{\partial \mathbf{v}_2} \right) H(1, 2, 3) + (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2) \right\} \\ &+ \frac{n}{m} \left\{ \frac{\partial F(v_1)}{\partial \mathbf{v}_1} \cdot \frac{\partial}{\partial \mathbf{r}_1} \int \phi(r_{14}) H(2, 3, 4) d^4 + (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2) \right\} \\ &+ \frac{n}{m} \left\{ \int \left(\frac{\partial \phi(r_{14})}{\partial \mathbf{r}_1} \cdot \frac{\partial}{\partial \mathbf{v}_1} + \frac{\partial \phi(r_{24})}{\partial \mathbf{r}_2} \cdot \frac{\partial}{\partial \mathbf{v}_2} \right) G(1, 2) G(3, 4) d^4 \right. \\ &\left. + (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2) \right\}, \end{aligned} \tag{4c}$$

where $F(v)$ is the one-particle distribution function. The binary correlation function $G(1, 2)$ and the ternary correlation function $H(1, 2, 3)$ are introduced through the following decomposition of the two-particle distribution function $F(1, 2)$ and the three-particle distribution function $F(1, 2, 3)$:

$$F(1, 2) = F(v_1)F(v_2) + G(1, 2), \tag{5a}$$

$$\begin{aligned} F(1, 2, 3) &= F(v_1)F(v_2)F(v_3) + \{F(v_1)G(2, 3) \\ &+ (1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1) + (1 \rightarrow 3, 2 \rightarrow 1, 3 \rightarrow 2)\} \\ &+ H(1, 2, 3). \end{aligned} \tag{5b}$$

In Eqs. (4c) and (5b), the terms denoted as $(i \rightarrow j, j \rightarrow k, k \rightarrow 1)$ represent the term obtained by applying the assigned permutation to the first term in the curly brackets of these equations. The integral over the specified number i denotes the integral over the velocities and the spatial coordinates of the i th particle. The

interaction potential $\phi(r_{ij})$ is the Coulomb potential,

$$\phi(r_{ij}) = e^2/|\mathbf{r}_i - \mathbf{r}_j|. \tag{6}$$

We have normalized the distribution functions as

$$1 = \frac{1}{V} \int F(v_1) d^1 \tag{7a}$$

$$F(v_1) = \frac{1}{V} \int F(1, 2) d^2 \tag{7b}$$

and have applied the limiting procedure of $N \rightarrow \infty, V \rightarrow \infty$, with $n = N/V$ kept finite.

In the thermal equilibrium state, the correlation functions G and H can be expressed as

$$G(1, 2) = F(v_1)F(v_2)g(r_{12}) \tag{8a}$$

and

$$H(1, 2, 3) = F(v_1)F(v_2)F(v_3)h(r_{12}, r_{23}, r_{31}). \tag{8b}$$

Then it is straightforward to reduce Eqs. (4b) and (4c) to,

$$\frac{d^2}{dr^2} g + \left(\frac{2}{r} - \frac{b}{r^2} \right) \frac{d}{dr} g - k_D^2 g = \frac{k_D^2}{4\pi} \frac{\partial}{\partial \mathbf{r}_1} \cdot \int \frac{\hat{r}_{13}}{r_{13}^2} h(r_{12}, r_{23}, r_{31}) d\mathbf{r}_3 \quad (9a)$$

and

$$\begin{aligned} \frac{\partial}{\partial r_{12}} h(r_{12}, r_{23}, r_{31}) - \frac{k_D^2}{4\pi} \int \frac{\hat{r}_{12} \cdot \hat{r}_{14}}{r_{14}^2} h(r_{23}, r_{34}, r_{42}) d\mathbf{r}_4 - (b/r_{12}^2) h(r_{12}, r_{23}, r_{31}) \\ = (b/r_{12}^2) [g(r_{12}) + g(r_{23})] + (k_D^2/4\pi) \int \frac{\hat{r}_{12} \cdot \hat{r}_{14}}{r_{14}^2} g(r_{13}) g(r_{24}) d\mathbf{r}_4, \end{aligned} \quad (9b)$$

where k_D is the Debye wave number $(4\pi e^2 n / \kappa T)^{1/2}$ and b is the Landau distance defined as

$$b = (e^2 / \kappa T). \quad (10)$$

The vector \hat{r}_{ij} is a unit vector defined as

$$\hat{r}_{ij} = (\mathbf{r}_i - \mathbf{r}_j) / |\mathbf{r}_i - \mathbf{r}_j|. \quad (11)$$

Equations (9a) and (9b) are the set of fundamental equations of the present studies. Solving Eqs. (9a) and (9b) for $g(r)$, we can calculate the correlation energy E as

$$\frac{E}{\kappa T} = \frac{1}{2} \frac{n}{\kappa T} \int d\mathbf{r} \frac{e^2}{r} g(r) = \frac{1}{2} k_D^2 \int_0^\infty dr r g(r) \quad (12)$$

and thus we can determine thermodynamic properties of the electron plasma.

In Eq. (9a), the term $-(b/r^2) dg/dr$ is due to the second term on the right-hand side of Eq. (4b). This term describes the effect of the short-range collisions. The term $-k_D^2 g$ of Eq. (9a) is due to the third term on the right-hand side of Eq. (4b). This term represents the screening effect of the long-range Coulomb potential. The right-hand side of Eq. (9a) expresses the modification of the binary correlation function resulting from ternary correlations between particles.

Neglecting contributions of short-range collisions and ternary correlations, we obtain the well-known expression for the binary correlation function as

$$g(r) = -(b/r) \exp(-k_D r). \quad (13)$$

Substituting Eq. (13) into Eq. (12), we obtain the Debye-Hückel limiting value of the correlation energy

$$E^{(D-H)} / \kappa T = -\frac{1}{2} \epsilon. \quad (14)$$

In the following sections, we investigate the influence of short range collisions and ternary correlations on the binary correlation function. We then determine the corresponding corrections to the Debye-Hückel limiting value of the correlation energy.

III. EFFECT OF SHORT-RANGE COLLISIONS ON THE BINARY CORRELATION FUNCTION

It is evident that the binary correlation function obtained as Eq. (13) loses its validity at small distances. Lamb and Burdick⁴ succeeded in eliminating this defect by taking into account the influence of short-range collisions. Disregarding the influence of ternary correlations, Eq. (9a) reduces to the following equation:

$$(d^2/dr^2)g + [(2/r) - (b/r^2)](d/dr)g - k_D^2 g = 0. \quad (15)$$

Introducing the transformations

$$g(r) = (b/r) \exp(-b/2r) U(r), \quad (16a)$$

$$x = k_D r, \quad (16b)$$

we can eliminate the first derivative term from Eq. (15), and obtain the equation for $U(x)$,

$$(d^2/dx^2)U(x) - [1 + (\epsilon^2/4x^4)]U(x) = 0. \quad (17)$$

With the aid of the transformation,⁸

$$x = (\epsilon/2)^{1/2} \exp(Z), \quad U(x) = x^{1/2} Y(Z), \quad (18)$$

Eq. (17) is reduced to the associated Mathieu equation for $Y(Z)$

$$(d^2/dZ^2)Y(Z) - [\lambda + 2h^2 \cosh(2Z)]Y(Z) = 0 \quad (19a)$$

with

$$\lambda = \frac{1}{4}, \quad h^2 = \epsilon/2. \quad (19b)$$

The solutions of Eq. (19a) are expressed as

$$Y(Z) = \begin{cases} \sum_{m=-\infty}^{\infty} (a_{2m}/a_0)^{\nu} I_m(h e^{-Z}) I_{m+\nu}(h e^Z) & (20a) \\ \sum_{m=-\infty}^{\infty} (a_{2m}/a_0)^{\nu} I_m(h e^Z) I_{m+\nu}(h e^{-Z}) & (20b) \\ \sum_{m=-\infty}^{\infty} (-1)^m (a_{2m}/a_0)^{\nu} I_m(h e^{-Z}) K_{m+\nu}(h e^Z) & (20c) \\ \sum_{m=-\infty}^{\infty} (-1)^m (a_{2m}/a_0)^{\nu} I_m(h e^Z) K_{m+\nu}(h e^{-Z}), & (20d) \end{cases}$$

⁸ The region of $x > (\epsilon/2)^{1/2}$ corresponds to the region of $0 < Z < +\infty$, while the region of $x < (\epsilon/2)^{1/2}$ corresponds to the region of $-\infty < Z < 0$.

where the coefficients a_{2m} are determined by the recurrence formula

$$a_{2m}^\nu [(2m+\nu)^2 - \lambda] - h^2 (a_{2m+2}^\nu + a_{2m-2}^\nu) = 0 \quad (21)$$

ν is related to λ via

$$\lambda = \nu^2 + [2(\nu^2 - 1)]^{-1} h^4 + [(5\nu^2 + 7)/32(\nu^2 - 1)^3(\nu^2 - 4)] h^8 + \dots \quad (22)$$

In the present problem, ν is determined from Eq. (22) for the given values of λ and h^2 as defined by Eq. (19b).

Inspection of Eq. (17) determines the following asymptotic behavior of the function $U(x)$,

$$U(x) \sim e^{\pm x} \text{ as } x \rightarrow \infty, \quad (23a)$$

and

$$U(x) \sim (2/\epsilon)^{1/2} x \exp(\pm \epsilon/2x) \text{ as } x \rightarrow 0. \quad (23b)$$

Therefore, two independent solutions of Eq. (19a) are chosen from the solutions Eqs. (20a)~(20d) to be in accord with the above asymptotic behavior of the function $U(x)$. For the outer region $x > (\epsilon/2)^{1/2}$, these solutions are given by

$$U_{<}^{(o)}(x) = (2\pi)^{1/2} x^{1/2} \sum_{m=-\infty}^{\infty} (a_{2m}/a_0)^\nu I_m(\epsilon/2x) I_{m+\nu}(x) \quad (24a)$$

$$U_{>}^{(o)}(x) = (2/\pi)^{1/2} x^{1/2} \sum_{m=-\infty}^{\infty} (-1)^m (a_{2m}/a_0)^\nu I_m(\epsilon/2x) K_{m+\nu}(x) \quad (24b)$$

and for the inner region $x < (\epsilon/2)^{1/2}$ by

$$U_{<}^{(i)}(x) = (2/\pi)^{1/2} x^{1/2} \sum_{m=-\infty}^{\infty} (-1)^m (a_{2m}/a_0)^\nu I_m(x) K_{m+\nu}(\epsilon/2x), \quad (25a)$$

$$U_{>}^{(i)}(x) = (2\pi)^{1/2} x^{1/2} \sum_{m=-\infty}^{\infty} (a_{2m}/a_0)^\nu I_m(x) I_{m+\nu}(\epsilon/2x). \quad (25b)$$

The superscripts (o) and (i) stand for the outer and the inner region, respectively. The above set of solutions is used to calculate the effect of ternary correlations on the binary correlation function. We notice that the solution $U(x)$ should behave as

$$U(x) \sim \exp(-x) \text{ for } x \rightarrow \infty \quad (26a)$$

and

$$U(x) \sim -(x/\epsilon) \exp(\epsilon/2x) \text{ for } x \rightarrow 0 \quad (26b)$$

so that the boundary conditions for the binary correlation function $g(x)$ [i.e., $g(\infty) = 0$ and $g(0) = -1$] are satisfied. Therefore, the solutions of Eq. (17) which satisfy the required boundary conditions are obtained as

$$U^{(o)}(x) = B \sum_{m=-\infty}^{\infty} (-1)^m a_{2m}^\nu x^{1/2} I_m(\epsilon/2x) K_{m+\nu}(x), \quad (27a)$$

for $x > (\epsilon/2)^{1/2}$,

and

$$U^{(i)}(x) = \sum_{m=-\infty}^{\infty} a_{2m}^\nu x^{1/2} I_m(x) I_{m+\nu}(\epsilon/2x) + C \sum_{m=-\infty}^{\infty} (-1)^m a_{2m}^\nu x^{1/2} I_m(x) K_{m+\nu}(\epsilon/2x), \quad (27b)$$

for $x < (\epsilon/2)^{1/2}$.

In the limit of $x \rightarrow 0$, the function $U^{(i)}(x)$ behaves asymptotically as

$$U^{(i)}(x) \sim a_0^\nu (\pi\epsilon)^{-1/2} x \exp[\epsilon/(2x)]. \quad (28)$$

By comparing Eq. (28) with Eq. (26b), we can determine the coefficient a_0^ν as

$$a_0^\nu = -(\pi/\epsilon)^{1/2} \quad (29)$$

Since ν is determined from Eq. (22) as

$$\nu = \frac{1}{2} + \frac{1}{8}\epsilon^2 + O(\epsilon^4), \quad (30)$$

we have

$$(a_2/a_0)^\nu = \frac{1}{12}\epsilon, \quad (31a)$$

$$(a_4/a_0)^\nu = \frac{1}{480}\epsilon^2. \quad (31b)$$

The constants B and C are determined from the following boundary conditions at $x = (\epsilon/2)^{1/2}$,

$$U^{(o)}[(\epsilon/2)^{1/2}] = U^{(i)}[(\epsilon/2)^{1/2}], \quad (32a)$$

$$dU^{(o)}/dx |_{x=(\epsilon/2)^{1/2}} = (dU^{(i)}/dx) |_{x=(\epsilon/2)^{1/2}}. \quad (32b)$$

Keeping terms of the order ϵ^2 , we can determine the constants B and C as,

$$B = (2/\pi)^{1/2} \epsilon^{1/2} (1 + \frac{1}{2}\epsilon) \quad (33a)$$

and

$$C = -\pi^{-1}\epsilon. \quad (33b)$$

In order to calculate the correlation energy up to the order ϵ^2 , it is sufficient to take $\nu = \frac{1}{2}$ and $m = 0$. Thus Eqs. (27a) and (27b) are reduced to

$$U^{(o)}(x) = -(2/\pi)^{1/2} (1 + \frac{1}{2}\epsilon) x^{1/2} I_0(\epsilon/2x) K_{1/2}(x) \quad (34a)$$

and

$$U^{(i)}(x) = -(\pi/\epsilon)^{1/2} x^{1/2} \{ I_0(x) I_{1/2}(\epsilon/2x) - (\epsilon/\pi) I_0(x) K_{1/2}(\epsilon/2x) \}. \quad (34b)$$

These expressions can be simplified further by retaining leading terms to obtain the correlation energy correctly up to second order in ϵ . Finally, the binary correlation function becomes

$$g^{(o)}(x) = -\epsilon(1 + \frac{1}{2}\epsilon)x^{-1} \exp[-x - (\epsilon/2x)] \quad x > (\epsilon/2)^{1/2} \quad (35a)$$

and

$$g^{(i)}(x) = -1 + (1 + \epsilon) \exp(-\epsilon/x) \quad x < (\epsilon/2)^{1/2}. \quad (35b)$$

Substituting Eqs. (35a) and (35b) into Eq. (12), we obtain

$$\begin{aligned} \frac{E^{(B)}}{\kappa T} &= \frac{1}{2} \int_0^{(\epsilon/2)^{1/2}} x g^{(i)}(x) dx + \frac{1}{2} \int_{(\epsilon/2)^{1/2}}^{\infty} x g^{(o)}(x) dx \\ &\approx -\frac{1}{2}\epsilon - \frac{1}{4}\epsilon^2 \ln \epsilon - \frac{1}{2}(8 - \frac{1}{4})\epsilon^2. \end{aligned} \quad (36)$$

Comparing this result with Eq. (14), we conclude that

by taking into account short-range collisions we obtain the correction to the Debye-Hückel limiting law to order $\epsilon^2 \ln \epsilon$. There arise terms of order ϵ^2 as well. Since, however, the coefficients of the ϵ^2 term in Eqs. (36) and (3) do not agree, we conclude that ternary correlations must be included to calculate this term properly.

IV. CONTRIBUTION OF TERNARY CORRELATIONS ON THE BINARY CORRELATION FUNCTION

In order to investigate the effect of ternary correlations, let us write the binary correlation function as

$$g(r) = g^{(B)}(r) + g^{(T)}(r), \quad (37)$$

where the function $g^{(T)}(r)$ is presumed to be of higher order than the function $g^{(B)}(r)$. We may then decompose the set of Eqs. (9a) and (9b) into the following set of equations:

$$\frac{d^2}{dr^2} g^{(B)} + \left(\frac{2}{r} - \frac{b}{r^2}\right) \frac{d}{dr} g^{(B)} - k_D^2 g^{(B)} = 0 \quad (38a)$$

$$\frac{d^2}{dr^2} g^{(T)} + \left(\frac{2}{r} - \frac{b}{r^2}\right) \frac{d}{dr} g^{(T)} - k_D^2 g^{(T)} = \frac{k_D^2}{4\pi} \frac{\partial}{\partial \mathbf{r}_1} \cdot \int \frac{\hat{r}_{13}}{r_{13}^2} h^{(B)}(r_{12}, r_{23}, r_{31}) d\mathbf{r}_3, \quad (38b)$$

where $h^{(B)}(r_{12}, r_{23}, r_{31})$ is determined from the following equation:

$$\begin{aligned} \frac{\partial}{\partial r_{12}} h^{(B)}(r_{12}, r_{23}, r_{31}) - \frac{k_D^2}{4\pi} \int \frac{\hat{r}_{12} \cdot \hat{r}_{14}}{r_{14}^2} h^{(B)}(r_{23}, r_{34}, r_{42}) d\mathbf{r}_4 \\ = \frac{b}{r_{12}^2} [g^{(B)}(r_{12}) + g^{(B)}(r_{23})] + \frac{k_D^2}{4\pi} \int \frac{\hat{r}_{12} \cdot \hat{r}_{14}}{r_{14}^2} g^{(B)}(r_{13}) g^{(B)}(r_{24}) d\mathbf{r}_4. \end{aligned} \quad (38c)$$

Solutions of Eq. (38a) have been discussed in detail in the preceding section. Here, we wish to solve Eqs. (38b) and (38c) and thereby calculate the contribution of ternary correlations to the correlation energy.

Eliminating the first derivative term of Eq. (38b) by a transformation defined by Eqs. (16a) and (16b), we can reduce Eq. (38b) to

$$(d^2/dx^2) U_T(x) - [1 + (\epsilon^2/4x^4)] U_T(x) = \lambda(x) \quad (39a)$$

$$\lambda(x_{12}) = \frac{1}{4\pi} \exp\left(\frac{\epsilon}{2x_{12}}\right) \left[x_{12} \frac{d}{dx_{12}} + 2 \right] \int \frac{\hat{x}_{12} \cdot \hat{x}_{13}}{x_{12}^2} h(x_{12}, x_{13}, x_{23}) d\mathbf{x}_3. \quad (39b)$$

The boundary conditions for the function $U_T(x)$ are $U_T(x) \rightarrow 0$ for $x \rightarrow \infty$ and $x \rightarrow 0$. We define the Greens function $K(x, x')$ as the solution of

$$(d^2/dx^2) K(x, x') - [1 + (\epsilon^2/4x^4)] K(x, x') = \delta(x - x') \quad (40)$$

with the boundary conditions

$$K(x, x') \rightarrow 0 \quad \text{for } x \rightarrow \infty \quad \text{and } x \rightarrow 0, \quad (41a)$$

and

$$(dK/dx) |_{x=x'+\delta} - (dK/dx) |_{x=x'-\delta} = 1 \quad (\delta > 0). \quad (41b)$$

The solution of Eq. (39a) is then given by

$$U_T(x) = \int_0^{\infty} K(x, x') \lambda(x') dx'. \quad (42)$$

Since we have obtained the two independent solutions of the homogeneous equation, Eq. (17), as Eqs. (24a), (24b) and Eqs. (25a), (25b), we can construct the Greens function $K(x, x')$ as follows:

$$K^{(\omega)}(x, x') = \begin{cases} K_{<}^{(\omega)}(x, x') = -\frac{1}{2}U_{<}^{(\omega)}(x)U_{>}^{(\omega)}(x'), & (\epsilon/2)^{1/2} < x < x', \\ K_{>}^{(\omega)}(x, x') = -\frac{1}{2}U_{>}^{(\omega)}(x)U_{<}^{(\omega)}(x'), & (\epsilon/2)^{1/2} < x' < x, \end{cases} \quad (43a)$$

and

$$K^{(i)}(x, x') = \begin{cases} K_{<}^{(i)}(x, x') = -\frac{1}{2}U_{<}^{(i)}(x)U_{>}^{(i)}(x'), & x < x' < (\epsilon/2)^{1/2}, \\ K_{>}^{(i)}(x, x') = -\frac{1}{2}U_{>}^{(i)}(x)U_{<}^{(i)}(x'), & x' < x < (\epsilon/2)^{1/2}. \end{cases} \quad (44a)$$

The constant factor $-\frac{1}{2}$ has been determined from the boundary condition given by Eq. (41b). Substituting Eqs. (42a), (43b), (44a), and (44b) into Eq. (42), we obtain

$$U_T^{(\omega)}(x) = -\frac{1}{2} \left[U_{>}^{(\omega)}(x) \int_{(\epsilon/2)^{1/2}}^x U_{<}^{(\omega)}(x') \lambda(x') dx' + U_{<}^{(\omega)}(x) \int_x^{\infty} U_{>}^{(\omega)}(x') \lambda(x') dx' \right], \quad [x > (\epsilon/2)^{1/2}], \quad (45a)$$

and

$$U_T^{(i)}(x) = -\frac{1}{2} \left[U_{>}^{(i)}(x) \int_0^x U_{<}^{(i)}(x') \lambda(x') dx' + U_{<}^{(i)}(x) \int_x^{(\epsilon/2)^{1/2}} U_{>}^{(i)}(x') \lambda(x') dx' \right] \quad [x < (\epsilon/2)^{1/2}]. \quad (45b)$$

Let us proceed to calculate the function $\lambda(x)$. Since it has been confirmed that the contribution of $g^{(T)}(x)$ to the correlation energy will be in the second order of ϵ , it is sufficient to determine the function $h^{(B)}(r_{12}, r_{23}, r_{31})$ correctly up to the second order of ϵ . Examining the structure of Eq. (38c), we find that the function $g^{(B)}(x)$ can be approximated by its lowest-order expression as

$$g^{(B)}(x) \sim g^{(0)}(x) = -(\epsilon/x) \exp(-x), \quad (46)$$

Then, as shown by O'Neil and Rostoker, the ternary correlation function $h^{(B)}(x_{12}, x_{23}, x_{31})$ is obtained as a solution of Eq. (38c) as follows:

$$h^{(B)}(x_{12}, x_{23}, x_{31}) = g^{(0)}(x_{12})g^{(0)}(x_{31}) + g^{(0)}(x_{31})g^{(0)}(x_{23}) + g^{(0)}(x_{23})g^{(0)}(x_{12}) + n \int g^{(0)}(x_{14})g^{(0)}(x_{24})g^{(0)}(x_{34}) dx_4. \quad (47)$$

This expression of the ternary correlation function is taken to be valid over the region $\infty > x > (\epsilon/2)^{1/2}$ in the lowest-order approximation, while O'Neil and Rostoker have assumed that this expression is valid only in the region $\infty > x > (4\pi\epsilon)^{1/3}$. Having determined the ternary correlation function $h^{(B)}(x_{12}, x_{23}, x_{31})$, we can proceed to calculate the function $\lambda(x)$. Details are discussed in the Appendix. Since the function $\lambda(x)$ itself is a quantity of second order in ϵ , we may evaluate the function $U_T(x)$ given by Eqs. (45a) and (45b) in the asymptotic limit of $\epsilon \rightarrow 0$. Therefore, Eq. (A17) can be approximated by

$$\begin{aligned} \lambda^{(0)}(x) &= -\frac{1}{2}(\pi)\epsilon^2 \{ 2(1+x)(1/x^3) \exp(-x) \\ &\quad - (5x^2+4x+2)(1/x^3) \exp(-2x) + \frac{1}{2}[\ln 3 \exp(-x) \\ &\quad + \text{Ei}(-x) \exp(-x) + \text{Ei}(-3x) \exp(+x)] \}. \end{aligned} \quad (48)$$

To the lowest order in ϵ , we have simply

$$U_{>}^{(\omega)}(x) = \exp(-x), \quad (49a)$$

and

$$U_{<}^{(\omega)}(x) = 2 \sinh(x). \quad (49b)$$

Thus, taking the asymptotic limit of $\epsilon \rightarrow 0$, we can evaluate Eqs. (45a) and (45b), and obtain

$$\begin{aligned} U_T^{(\omega)}(x) &\simeq -e^{-x} \int_0^x \sinh(x') \lambda(x') dx' \\ &\quad - \sinh(x) \int_x^{\infty} e^{-x'} \lambda(x') dx' \\ &= -\frac{1}{8}\epsilon^2 \{ -\frac{4}{3}(2x-3)(1/x)e^{-x} - \frac{4}{3}(x+3)(1/x)e^{-2x} \\ &\quad - (3+x) \text{Ei}(-3x)e^{+x} + (3-x)[\text{Ei}(-x) + \ln 3]e^{-x} \} \end{aligned} \quad (50a)$$

and

$$U_T^{(i)}(x) \simeq 0. \quad (50b)$$

Substituting Eqs. (50a) and (50b) into Eq. (12) with the transformations Eqs. (16a) and (16b), we can calculate the correlation energy resulting from the ternary correlation effect as

$$\begin{aligned} \frac{E^{(T)}}{\kappa T} &= \frac{1}{2} \int_0^{\infty} x g^{(T)}(x) dx \simeq \frac{1}{2} \int_0^{\infty} U_T(x) dx \\ &\simeq -\frac{1}{4}(\ln 3 - \frac{5}{6})\epsilon^2. \end{aligned} \quad (51)$$

V. CONCLUDING DISCUSSION

In the preceding sections, we have discussed a systematic approach to solve the BBGKY hierarchy equation in the thermal equilibrium case. Summing up Eqs. (36) and (51), we obtain the correlation energy of the electron plasma correct up to the second order in ϵ .

According to the analysis in Sec. III, we have shown explicitly that the dynamic effect of short-range collisions becomes appreciable at distances of the order of

$$x_0 = (\epsilon/2)^{1/2} \lambda_D = (4\pi)^{-1/3} (\epsilon/8)^{1/6} n^{-1/3}, \quad (52)$$

which is far smaller than the mean distance between particles $n^{-1/3}$. Since the Coulomb interaction is rather weak, this is physically a reasonable result.

In Sec. IV, we have examined the contribution of ternary correlation on the binary correlation function without taking into account the effect of short-range collisions. It has been confirmed that this approximation is consistent in calculating the thermodynamic properties of the electron plasma up to the second order in ϵ . In view of Eq. (51), we can conclude that the binary correlation function due to the ternary correlation effect $g^{(T)}(x)$ is given by

$$g^{(T)}(x) = (\epsilon^2/8) x^{-1} \left\{ \frac{4}{3} (2x-3) x^{-1} e^{-x} + \frac{4}{3} (x+3) x^{-1} e^{-2x} \right. \\ \left. + (3+x) \text{Ei}(-3x) e^x - (3-x) [\text{Ei}(-x) + \ln 3] e^{-x} \right\} \quad 0 < x < \infty \quad (53)$$

while the binary correlation function $g^{(B)}(x)$ is obtained as

$$g^{(B)}(x) = \begin{cases} -\epsilon(1+\frac{1}{2}\epsilon)x^{-1} \exp[-x - (\epsilon/2x)], & \infty > x > (\epsilon/2)^{1/2} \\ -1 + (1+\epsilon) \exp(-\epsilon/x), & (\epsilon/2)^{1/2} > x > 0. \end{cases} \quad (54a) \quad (54b)$$

The point $x_0 = (\epsilon/2)^{1/2}$ is determined from structure of the differential equation for the function $g^{(B)}(x)$, and thus the arbitrariness involved in the interpolation method of O'Neil and Rostoker has been eliminated completely in the present approach.

It would be worthwhile to discuss the relationship between the various expressions for the binary correlation function, since our expressions as given by Eqs. (53), (54a), and (54b) appear to be different from the Bowers-Salpeter expression and the O'Neil-Rostoker expression. In Ref. 4, the binary correlation function which gives rise to the correlation energy correctly up to the order of ϵ^2 is given as,⁹

$$g^{B-S}(x) = \{ \exp[-(\epsilon/x)e^{-x}] - 1 \} + w_1(x) \quad (55a)$$

with the abbreviation

$$w_1(x) = -(\epsilon^2/8) x^{-1} \left\{ \frac{4}{3} (e^{-x} - e^{-2x}) + (3-x) \right. \\ \left. \times [\text{Ei}(-x) + \ln 3] e^{-x} - (3+x) \text{Ei}(-3x) e^x \right\}. \quad (55b)$$

First of all, the O'Neil-Rostoker expression of the binary correlation function is essentially equivalent to the Bowers-Salpeter expression because the first two terms of the function $\phi_I(x)$, which is given in Sec. 5 of Ref. 7, are the first two terms of the expansion

$$\exp[-(\epsilon/x)e^{-x}] - 1 \approx -(\epsilon/x)e^{-x} + \frac{1}{2}(\epsilon/x)^2 e^{-2x} + \dots \quad (56)$$

while the remaining terms of $\phi_I(x)$ are nothing but the function $w_1(x)$ of Bowers and Salpeter.¹⁰ Turning to the present expression of the binary correlation function, we can show that our expression is also equivalent to the Bowers-Salpeter expression as follows: the function $g^{(T)}(x)$ can be rewritten as

$$g^{(T)}(x) = w_1(x) + (\epsilon^2/2x^2) \{ (x-1)e^{-x} + e^{-2x} \}, \quad (57)$$

hence the present expression of the binary correlation function can be expressed as,

$$g^{L-I}(x) = g^*(x) + w_1(x) \quad (58a)$$

with the abbreviation

$$g^*(x) = g^{(B)}(x) + (\epsilon^2/2x^2) \{ (x-1)e^{-x} + e^{-2x} \}. \quad (58b)$$

With the expression for $g^{(B)}(x)$ as given by Eq. (54a), the function $g^*(x)$ reduces to

$$g^*(x) \approx \epsilon(1+\frac{1}{2}\epsilon)(1/x)e^{-x} [1 - (\epsilon/2x)] \\ + (\epsilon^2/2x^2) \{ (x-1)e^{-x} + e^{-2x} \} \\ = -(\epsilon/x)e^{-x} + \frac{1}{2}(\epsilon/x)^2 e^{-2x} + \dots \quad (59)$$

which agrees with the expansion of Eq. (56) for large values of x . In the region for which $x < (\epsilon/2)^{1/2}$, we can

⁹ Recently DeWitt (Ref. 11) has obtained the same expression as $g^{B-S}(x)$ by a diagrammatic method which is slightly different from that used by Bowers and Salpeter.

¹⁰ The factor $\frac{1}{2}$ in front of $\exp(-2x)$ of $\phi_I(x)$ should be replaced by $\frac{1}{3}$. This error does not affect the correlation energy calculated by O'Neil and Rostoker, since they have used the Fourier transformation of $\phi_I(x)$ in order to calculate the correlation energy.

approximate the second term of Eq. (58b) by

$$(\epsilon^2/2x^2) \{ (x-1)e^{-x} + e^{-2x} \} \approx -\frac{1}{2}\epsilon^2. \quad (60a)$$

On the other hand, Eq. (54b) can be written,

$$\begin{aligned} g^{(B)}(x) &= -1 + (1+\epsilon) \exp(-\epsilon/x) \\ &\approx -1 + \exp\{ -(\epsilon/x)(1-x) \} \\ &\approx -1 + \exp\{ -(\epsilon/x)e^{-x} \}. \end{aligned} \quad (60b)$$

The contribution of Eq. (60a) to the correlation energy is of higher order than ϵ^2 . Thus, the present expressions given by Eqs. (53), (54a), and (54b) are essentially equivalent to the Bowers-Salpeter expression.

Finally, let us discuss a controversial point discussed in Ref. 11. DeWitt has pointed out that when x is larger than $8/(\epsilon \ln 3)$ the correction term $z_1(x)$ dominates over the lowest-order Debye-Hückel term. Within the framework of the present approach, this means the contribution of ternary correlation on the binary correlation function dominates over the lowest order binary correlation effect at very large distances. We should remember, however, that the decomposition of Eqs. (9a) and (9b) into the set of Eqs. (38a), (38b), and (38c) is carried out by assuming the function $g^{(T)}(x)$ to be of higher order than the function $g^{(B)}(x)$ in Eq. (37). Hence, the fact that $g^{(T)}(x)$ becomes larger than $g^{(B)}(x)$ at large distances indicates simply that the above decomposition of the equations loses its validity when x becomes very large. Returning to Eq. (4b), we can see explicitly that the third term, which is essential to screen the long-range interaction, dominates over the last term in the limit of $r_{12} \rightarrow \infty$. Therefore, at very large distances, the exact binary correlation function should behave as

$$g(x) \sim -\epsilon x^{-1} \exp(-x). \quad (61)$$

If we could solve Eqs. (9a) and (9b) exactly, the asymptotic form of the binary correlation function would be

$$g(x) \sim \epsilon x^{-1} \exp\{-x - \Psi(x)\}, \quad (62a)$$

where the function $\Psi(x)$ behaves asymptotically as

$$\Psi(x) \sim \frac{1}{8}\epsilon \ln 3x \exp\{-\frac{1}{8}\epsilon \ln 3x\}. \quad (62b)$$

As long as x remains smaller than $8/(\epsilon \ln 3)$, Eqs. (62a) and (62b) can be expanded as

$$g(x) \sim -(\epsilon/x)e^{-x} + \frac{1}{8}\epsilon^2 \ln 3e^{-x} \quad (63)$$

which agrees with the asymptotic form of $g^{B-S}(x)$. When x becomes larger than $8/(\epsilon \ln 3)$, Eq. (62a) is reduced to Eq. (57) since $\Psi(x)$ vanishes in the limit of $x \rightarrow \infty$. DeWitt has shown that the higher-order term $w_1(x)$ dominates over the lowest-order term $(\epsilon/x) \exp(-x)$ at large distances. He relates this to a similar phenomenon that occurs in the study of the dynamic behavior of a plasma, where the correlation damping can become larger than the Landau damping in the long wavelength region. (This effect has been discussed first by one of the present authors¹² on the basis of the BBGKY equations.) Our analysis shows explicitly, however that this is not the case. Furthermore, according to the above analysis, we can conclude that the partial summations of the cluster diagrams carried out by Bowers, Salpeter, and DeWitt are inadequate to determine the correct behavior of the binary correlation function at large distances.

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APPENDIX: CALCULATION OF THE FUNCTION $\lambda(x)$

Here, we describe some details of calculation of the function $\lambda(x)$. We have

$$\lambda(x) = (4\pi)^{-1} \exp[\epsilon/(2x)] \left(x \frac{d}{dx} + 2 \right) \int \frac{\hat{x} \cdot \hat{x}_{13}}{x^2} h(x, x_{13}, x_{23}) d\mathbf{x}_3, \quad (A1)$$

where

$$h(x, x_{13}, x_{23}) = g^{(0)}(x) g^{(0)}(x_{13}) + g^{(0)}(x) g^{(0)}(x_{23}) + g^{(0)}(x_{13}) g^{(0)}(x_{23}) + (4\pi\epsilon)^{-1} \int g^{(0)}(x_{14}) g^{(0)}(x_{24}) g^{(0)}(x_{34}) d\mathbf{x}_4. \quad (A2)$$

In Eqs. (A1) and (A2), the relative coordinate x_{12} is denoted as x for the simplicity of notations. First, let us discuss the following integral:

$$\int \frac{\hat{x} \cdot \hat{x}_{13}}{x_{13}^2} g^{(0)}(x_{23}) d\mathbf{x}_3 = -4\pi(d/dx) \left[x^{-1} \int_0^x x_{23}^2 g^{(0)}(x_{23}) dx_{23} + \int_x^\infty x_{23} g^{(0)}(x_{23}) dx_{23} \right] = \frac{4\pi}{x^2} \int_0^x x_{23}^2 g^{(0)}(x_{23}) dx_{23}. \quad (A3)$$

¹¹ H. E. DeWitt, Phys. Rev. **140**, A466 (1965).

¹² Y. H. Ichikawa, Progr. Theoret. Phys. (Kyoto) **24**, 1083 (1960).

The integral given by Eq. (A3) can be decomposed into two terms as follows:

$$\int_0^x x_{23}^2 g^{(0)}(x_{23}) dx_{23} = \int_0^{(\epsilon/2)^{1/2}} x_{23}^2 g^{(0)}(x_{23}) dx_{23} + \int_{(\epsilon/2)^{1/2}}^x x_{23}^2 g^{(0)}(x_{23}) dx_{23}. \quad (\text{A4})$$

If we evaluate the first term of Eq. (A4) by substituting Eq. (35b) for $g^{(0)}(x)$, we can confirm that the contribution of this term is of higher order than ϵ^2 . The second term of Eq. (A4) can be calculated by using the approximate expression given by Eq. (46) as follows:

$$\int_{(\epsilon/2)^{1/2}}^x x_{23}^2 g^{(0)}(x_{23}) dx_{23} = \epsilon \{ (1+x) \exp(-x) - [1 + (\epsilon/2)^{1/2}] \exp[-(\epsilon/2)^{1/2}] \}. \quad (\text{A5})$$

Therefore, we have the following approximate result:

$$\int \frac{\hat{x} \cdot \hat{x}_{13}}{x_{13}^2} g^{(0)}(x_{23}) d\mathbf{x}_3 = (4\pi\epsilon/x^2) \{ (1+x) \exp(-x) - [1 + (\epsilon/2)^{1/2}] \exp[-(\epsilon/2)^{1/2}] \}. \quad (\text{A6})$$

Now, excluding the domain of $x_{ij} < (\epsilon/2)^{1/2}$ from the following calculation, we may proceed to discuss the integral

$$I(x) = \int \frac{\hat{x} \cdot \hat{x}_{13}}{x_{13}^2} \left\{ g^{(0)}(x_{13}) g^{(0)}(x_{23}) + (4\pi\epsilon)^{-1} \int g^{(0)}(x_{14}) g^{(0)}(x_{24}) g^{(0)}(x_{34}) d\mathbf{x}_4 \right\} d\mathbf{x}_3. \quad (\text{A7})$$

First, we notice the following relation:

$$\begin{aligned} \int \frac{\hat{x}_{13}}{x_{13}^2} g^{(0)}(x_{34}) d\mathbf{x}_3 &= \hat{x}_{14} \frac{\partial}{\partial x_{14}} \int x_{13}^{-1} g^{(0)}(x_{34}) d\mathbf{x}_3 \\ &= \hat{x}_{14} (4\pi\epsilon/x_{14}^2) \{ (1+x_{14}) \exp(-x_{14}) - [1 + (\epsilon/2)^{1/2}] \exp[-(\epsilon/2)^{1/2}] \}. \end{aligned} \quad (\text{A8})$$

Therefore, the integral $I(x)$ is reduced to

$$\begin{aligned} I(x) &= \int \frac{\hat{x} \cdot \hat{x}_{14}}{x_{14}^2} g^{(0)}(x_{13}) g^{(0)}(x_{23}) [1 + \{ (1+x_{14}) \exp(-x_{14}) - [1 + (\epsilon/2)^{1/2}] \exp[-(\epsilon/2)^{1/2}] \}] \\ &= \{ 1 - [1 + (\epsilon/2)^{1/2}] \exp[-(\epsilon/2)^{1/2}] \} \int \frac{\hat{x} \cdot \hat{x}_{14}}{x_{14}^2} g^{(0)}(x_{14}) g^{(0)}(x_{24}) d\mathbf{x}_4 + \int \frac{\hat{x} \cdot \hat{x}_{14}}{x_{14}^2} (1+x_{14}) \exp(-x_{14}) g^{(0)}(x_{14}) g^{(0)}(x_{24}) d\mathbf{x}_4 \\ &\simeq \epsilon^2 \int (\hat{x} \cdot \hat{x}_{14}) (x_{14}^2 x_{24})^{-1} (1+x_{14}^{-1}) \exp(-2x_{14}-x_{24}) d\mathbf{x}_4 \quad x_{14} > (\epsilon/2)^{1/2}, \quad x_{24} > (\epsilon/2)^{1/2}, \end{aligned} \quad (\text{A9})$$

where the first term of the second line has been disregarded since it is smaller than the second term by a factor ϵ^2 . Integration of Eq. (A9) can be carried out by introducing prolate spherical coordinates,

$$x_{14} = (\xi + \eta) x_{12} / 2 \quad (\text{A10a})$$

$$x_{24} = (\xi - \eta) x_{12} / 2. \quad (\text{A10b})$$

The domain of variables ξ and η is given by

$$1 < \xi < +\infty, \quad -1 < \eta < 1. \quad (\text{A10c})$$

The allowed domain of the integral of Eq. (A9) then becomes

$$\xi - a > \eta > -\xi + a \quad (\text{A11})$$

with the abbreviation $a = (2\epsilon)^{1/2}/x$. Equation (A9) then reduces to

$$I(x) = 2\pi \int d\xi \int d\eta \frac{1+\xi\eta}{(\xi+\eta)^2} \left(1 + \frac{2}{x} \frac{1}{\xi+\eta} \right). \quad (\text{A12})$$

Changing the variable η into the new variable ζ by a transformation

$$\zeta = \xi + \eta, \tag{A13}$$

we can transform Eq. (A12) as follows:

$$I(x) = 2\pi \left[\int_1^{1+a} d\xi \int_a^{2\xi-a} d\zeta + \int_{1+2}^{\infty} d\xi \int_{\xi-1}^{\xi+1} d\zeta \right] \left(\frac{1-\xi^2}{\zeta} + \xi \right) \zeta^{-1} \left(1 + \frac{2}{x\zeta} \right) \exp(-x\xi - \frac{1}{2}x\zeta), \tag{A14}$$

where the domain of integration corresponds to the case $x > (2\epsilon)^{1/2}$. Since the contribution arising from the region of $(2\epsilon)^{1/2} > x > (\epsilon/2)^{1/2}$ to the function $U_T^{(0)}(x)$ is found to be higher order than ϵ^2 , this region can be disregarded in the present analysis. Performing the integration in Eq. (A14) results in,

$$I(x) = \pi\epsilon^2 \{ (1/x^2)(1+x) [\ln 3 + \text{Ei}(-x)] \exp(-x) + (1/x^2)(1-x) \text{Ei}(-3x) \exp(x) \}, \quad x > (2\epsilon)^{1/2}. \tag{A15}$$

Combining the contributions of Eqs. (A6) and (A15), we find

$$\int \frac{\hat{x} \cdot \hat{x}_{13}}{x_{13}^2} h(x, x_{13}, x_{23}) d\mathbf{x}_3 = \pi\epsilon^2 [(4/x^3) \{ (1+x) \exp(-2x) - [1 + (\epsilon/2)^{1/2}] \times \exp[-x - (\epsilon/2)^{1/2}] \} + (1/x^2)(1+x) [\ln 3 + \text{Ei}(-x)] \exp(-x) + (1/x^2)(1-x) \text{Ei}(-3x) \exp(x)], \tag{A16}$$

Substituting Eq. (A16) into Eq. (A1), we finally obtain the function $\lambda(x)$ as

$$\lambda(x) = -\frac{1}{4}\epsilon^2 \exp[\epsilon/(2x)] \{ 4[1 + (\epsilon/2)^{1/2}](1/x^3)(1+x) \exp[-x - (\epsilon/2)^{1/2}] - (2/x^3)(2+4x+3x^2) \exp(-2x) + [\ln 3 + \text{Ei}(-x)] \exp(-x) + \text{Ei}(-3x) \exp(x) \}. \tag{A17}$$