# Wigner-Kirkwood quantum corrections for the pair distribution function in a plasma

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We study the quantum corrections for the pair distribution function $g_2(r_{12})$ in a one-component plasma. Our analysis is based on the $\hbar^2$ expansion of the Wigner-Kirkwood $N$-particle distribution function in phase space. A resulting expression for $g_2(r_{12})$ is derived exactly, at order $\hbar^4$, valid at any interparticle distance. This quantum pair distribution function is expressed in terms of the classical two-, three-, four-, and five-particle distribution functions. Analytical properties of this expression are studied, both for small and large $r_{12}$. Accurate approximate expressions, depending only on the classical pair distribution function, are proposed. [S1063-651X(98)09204-6]

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## I. INTRODUCTION

The knowledge of the static pair distribution functions $g_2(r_{12})$ plays a central role in the study of thermodynamical and transport properties of a fluid [1]. In a partially degenerated fluid, quantum expressions $g_2^q(r_{12})$ are needed. A powerful approach involves a semiclassical quantum distribution function $P(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{p}_1,\ldots,\mathbf{p}_N)$ proposed by Wigner in his original paper [2] to calculate quantum corrections for the classical thermodynamical properties. Using the so-called Wigner-Kirkwood (WK) $\hbar^2$ expansion of the quantum distribution function, the $\hbar^2$ correction for $g_2(r_{12})$ was derived by Jancovici [3] with a very simple result in the case of the one-component plasma (OCP).

The $\hbar^4$ term was already investigated by Alastuey and Martin [4] and Cornu and Martin [5] in order to analyze the absence of exponential clustering in a quantum plasma. These authors predicted the asymptotic behavior ($r_{12}\to\infty$) of the charge-charge correlation function in the OCP. As a major result, they have shown that, in contradistinction with the classical case where $g_2^c(r_{12}\to\infty)-1$ ($c$ stands for classical) decreases faster than any inverse power of $r_{12}$ (exponential clustering) [6], the $\hbar^2$ WK expansion of $g_2^q(r_{12})-1$ no longer exhibits a decay bounded by an exponential, but an algebraic one. They proved that the $\hbar^4$ term is decreasing as $r_{12}^{-10}$. As explained by these authors, this tail arises from the fluctuations of dipolar interactions which are not perfectly screened in the quantum case. The knowledge of the $\hbar^4$ term is thus shown to have a great importance in plasma statistics.

In the present work, we apply the WK formalism described in Sec. II to derive the exact $\hbar^4$ correction for the pair distribution function in the OCP (Sec. III). At order $\hbar^4$, $g_2^q(r_{12})$ is expressed in terms of the classical distribution functions $g_n^c(\mathbf{r}_1,\ldots,\mathbf{r}_n)$ (up to $n=5$). This result [Eq. (32)] has been previously presented in a Letter [7]. The analytical properties of the $\hbar^4$ term are studied in both the limits $r_{12}\to\infty$ (Sec. IV) and $r_{12}\to 0$ (Sec. V). Of course, as explained at the end of Sec. V, $r_{12}$ cannot be too small. In the other case, this semiclassical formalism would not be appropriate and the WK expansion would not converge. For large $r_{12}$, the expansion with respect to $r_{12}^{-2}$ is derived exactly at all orders [Eq. (60)]. This analysis of the behavior of the $\hbar^4$ term allows us to propose, in Sec. VI, approximate expressions for the WK pair distribution function $g_2^q(r_{12})$ at order $\hbar^4$. These very accurate approximations need only the knowledge of the classical pair distribution function $g_2^c(r_{12})$. In the last section, a few numerical results are shown.

## II. WIGNER-KIRKWOOD DISTRIBUTION FUNCTION, $\hbar^4$ CORRECTION

If we consider a set of $N$ particles enclosed in volume $V$ at temperature $T$ ($N$ and $V$ are very large), $\hbar^2$ expansion of the unnormalized $N$-particle WK distribution function in phase space [2] reads

$$P(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{p}_1,\ldots,\mathbf{p}_N)=\exp(-\beta\epsilon)+\hbar^2 f_2+\hbar^4 f_4+\cdots+\hbar^{2n}f_{2n}+\cdots, \tag{1}$$

where $\epsilon$ is the total energy,

$$\epsilon=\sum_{k=1}^{N}\frac{p_k^2}{2M_k}+U(\mathbf{r}_1,\ldots,\mathbf{r}_N), \tag{2}$$

and $\beta=1/k_BT$ ($k_B$ is Boltzmann's constant). $\mathbf{r}_k$ and $\mathbf{p}_k$ denote the position and the momentum of the $k$th particle in three-dimensional (3D) space, $M_k$ denotes its mass, and $U$ stands for the potential energy. It can be seen that $f_{2n}$ satisfies a partial differential equation [2]

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$$-\sum_{k=1}^{N}\frac{\mathbf{p}_k}{M_k}\cdot(\nabla_{\mathbf{r}_k}f_{2n})+\sum_{k=1}^{N}(\nabla_{\mathbf{r}_k}U)\cdot(\nabla_{\mathbf{p}_k}f_{2n})=\frac{1}{2^2 3!}\sum_{k_1,k_2,k_3=1}^{N}(\nabla_{\mathbf{r}_{k_1}}\nabla_{\mathbf{r}_{k_2}}\nabla_{\mathbf{r}_{k_3}}U)\cdot(\nabla_{\mathbf{p}_{k_1}}\nabla_{\mathbf{p}_{k_2}}\nabla_{\mathbf{p}_{k_3}}f_{2n})+\cdots$$
$$-\frac{(-1)^n}{2^{2n}(2n+1)!}\sum_{k_1,\ldots,k_{2n+1}=1}^{N}(\nabla_{\mathbf{r}_{k_1}}\cdots\nabla_{\mathbf{r}_{k_{2n+1}}}U)\cdot(\nabla_{\mathbf{p}_{k_1}}\cdots\nabla_{\mathbf{p}_{k_{2n+1}}}f_{2n}),\tag{3}$$

written here with the help of contracted tensor products. $\nabla_{\mathbf{r}_k}$ and $\nabla_{\mathbf{p}_k}$ denote the gradients at $\mathbf{r}_k$ and $\mathbf{p}_k$. $f_2$ was evaluated by Wigner in his original work [2]. In order to derive terms of higher orders, we conveniently make use of Wigner's notation: a point in the phase space is specified by $(r_1,\ldots,r_{3N},p_1,\ldots,p_{3N})$ where $r_1,\ldots,r_{3N}$ are the $3N$ spatial coordinates of the $N$ particles and $p_1,\ldots,p_{3N}$, the corresponding momentum coordinates. Let

$$f_{2n}=e^{-\beta\epsilon}g_{2n}.\tag{4}$$

Upon evaluating the $p$ derivatives of $e^{-\beta\epsilon}$ and $f_2$, the following equation is readily deduced:

$$-\sum_{k=1}^{3N}\frac{p_k}{M_k}\frac{\partial g_4}{\partial r_k}+\sum_{k=1}^{3N}\frac{\partial U}{\partial r_k}\frac{\partial g_4}{\partial p_k}=\frac{g_2}{3!4}\left[-\sum_{k,l,m=1}^{3N}\frac{\partial^3 U}{\partial r_k\partial r_l\partial r_m}\frac{\beta^3 p_k p_l p_m}{M_k M_l M_m}+3\sum_{k,l=1}^{3N}\frac{\partial^3 U}{\partial r_k^2\partial r_l}\frac{\beta^2 p_l}{M_k M_l}\right]$$
$$+\frac{1}{3!16}\sum_{k,l,m,n=1}^{3N}\frac{\partial^3 U}{\partial r_k\partial r_l\partial r_m}\frac{\partial^2 U}{\partial r_m\partial r_n}\frac{\beta^5 p_k p_l p_n}{M_k M_l M_m M_n}$$
$$+\frac{1}{5!16}\sum_{k,l,m,n,o=1}^{3N}\frac{\partial^5 U}{\partial r_k\partial r_l\partial r_m\partial r_n\partial r_o}\frac{\beta^5 p_k p_l p_m p_n p_o}{M_k M_l M_m M_n M_o}$$
$$-\frac{1}{3!16}\sum_{k,l,m=1}^{3N}\frac{\partial}{\partial r_k}\left(\frac{\partial^2 U}{\partial r_k\partial r_l}\frac{\partial^2 U}{\partial r_l\partial r_m}\right)\frac{\beta^4 p_m}{M_k M_l M_m}$$
$$-\frac{1}{3!32}\sum_{k,l,m,n=1}^{3N}\frac{\partial^5 U}{\partial r_k^2\partial r_l\partial r_m\partial r_n}\frac{\beta^4 p_l p_m p_n}{M_k M_l M_m M_n}+\frac{1}{128}\sum_{k,l,m=1}^{3N}\frac{\partial^5 U}{\partial r_k^2\partial r_l^2\partial r_m}\frac{\beta^3 p_m}{M_k M_l M_m},\tag{5}$$

with [2]

$$g_2=\sum_{k=1}^{3N}\frac{\beta^2}{8M_k}\frac{\partial^2 U}{\partial r_k^2}+\sum_{k=1}^{3N}\frac{\beta^3}{24M_k}\left(\frac{\partial U}{\partial r_k}\right)^2+\sum_{k,l=1}^{3N}\frac{\beta^3 p_k p_l}{24M_k M_l}\frac{\partial^2 U}{\partial r_k\partial r_l}.\tag{6}$$

Noting that $g_2$ is a sum of terms of orders $\beta^2$ and $\beta^3$, it can be checked that the right-hand member of Eq. (5) contains terms of orders $\beta^3$, $\beta^4$, $\beta^5$, and $\beta^6$. Thus $g_4$ may be written in the form

$$g_4=\beta^3 G_3+\beta^4 G_4+\beta^5 G_5+\beta^6 G_6,\tag{7}$$

where $G_3$, $G_4$, $G_5$, and $G_6$ are also solutions to four partial differential equations, easier to solve than the initial one. Equation (5) can also be solved by rearranging its right-hand member according to powers of $U$.

As a final result, $P$ at order $\hbar^4$ reads

$$P=\exp\left[-\beta\epsilon+\hbar^2\left(-\sum_{k=1}^{N}\frac{\beta^2\nabla_k^2 U}{8M_k}+\sum_{k=1}^{N}\frac{\beta^3}{24M_k}(\nabla_k U)^2+\sum_{k,l=1}^{N}\frac{\beta^3\mathbf{p}_k\mathbf{p}_l}{24M_k M_l}\cdot\nabla_k\nabla_l U\right)\right]+\hbar^4 e^{-\beta\epsilon}\left[-\frac{\beta^3}{2}\left(\sum_{k=1}^{N}\frac{\nabla_k^2}{8M_k}\right)^2 U\right.$$
$$+\frac{\beta^4}{16(3!)}\sum_{k,l=1}^{N}\frac{(\nabla_k U)\cdot\nabla_k}{M_k M_l}(\nabla_l^2 U)+\frac{\beta^4}{32(3!)}\sum_{k,l=1}^{N}\frac{(\nabla_k\nabla_l U)^2}{M_k M_l}+\frac{\beta^4}{32(3!)}\sum_{k,l,m=1}^{N}\frac{\nabla_k^2}{M_k M_l M_m}(\mathbf{p}_l\mathbf{p}_m)\cdot(\nabla_l\nabla_m U)$$
$$-\frac{\beta^5}{4(5!)}\sum_{k,l=1}^{N}\frac{(\nabla_k U)\cdot\nabla_k}{M_k M_l}(\nabla_l U)^2-\frac{\beta^5}{4(5!)}\sum_{k,l,m=1}^{N}\frac{(\mathbf{p}_k\mathbf{p}_l\nabla_m U)}{M_k M_l M_m}\cdot(\nabla_k\nabla_l\nabla_m U)-\frac{\beta^5}{2(5!)}\sum_{k=1}^{N}\frac{1}{M_k}\left(\nabla_k\sum_{l=1}^{N}\frac{\mathbf{p}_l}{M_l}\cdot\nabla_l U\right)^2$$
$$\left.-\frac{\beta^5}{16(5!)}\left(\sum_{k=1}^{N}\frac{\mathbf{p}_k}{M_k}\cdot\nabla_k\right)^4 U\right]+O(\hbar^6),\tag{8}$$

with $\boldsymbol{\nabla}_k \equiv \boldsymbol{\nabla}_{\mathbf{r}_k}$. In the last equation, the subscripts take values ranging from 1 to $N$, the number of particles. The spatial distribution function $g(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ is deduced performing the average of $P(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{p}_1,\ldots,\mathbf{p}_N)$ over the momenta:

$$g(\mathbf{r}_1,\ldots,\mathbf{r}_N)=\frac{\int\cdots\int d^3p_1\cdots d^3p_N P(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{p}_1,\ldots,\mathbf{p}_N)}{\int\cdots\int d^3p_1\cdots d^3p_N \exp(-\beta\Sigma_{k=1}^N p_k^2/2M_k)}. \tag{9}$$

It can be written in the compact form

$$g(\mathbf{r}_1,\ldots,\mathbf{r}_N)=\exp\left(\sum_{k=1}^N \frac{\lambda_k^2}{24}\nabla_k^2\right)e^{-\beta U}+\hbar^2F_2+\hbar^4F_4+O(\hbar^6), \tag{10}$$

where $\lambda_k$ is the de Broglie wavelength associated with the $k$th particle:

$$\lambda_k^2=\hbar^2\beta/M_k. \tag{11}$$

$\hbar^2F_2$ and $\hbar^4F_4$ are calculated in Appendix A with the results

$$\hbar^2F_2=-e^{-\beta U}\sum_{k=1}^N \frac{\lambda_k^2}{24}\nabla_k^2\beta U \tag{12}$$

and

$$\begin{aligned}\hbar^4F_4=&-\left(\sum_{l=1}^N \frac{\lambda_l^2}{24}\nabla_l^2\beta U\right)\sum_{k=1}^N \frac{\lambda_k^2}{24}\nabla_k^2 e^{-\beta U}-\frac{1}{60}\sum_{k,l=1}^N \frac{\lambda_k^2\lambda_l^2}{24}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)\cdot(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l e^{-\beta U})+e^{-\beta U}\left[-\frac{19}{10}\left(\sum_{k=1}^N \frac{\lambda_k^2}{24}\nabla_k^2\right)^2(\beta U)\right.\\&\left.+\frac{1}{2}\left(\sum_{k=1}^N \frac{\lambda_k^2}{24}\nabla_k^2\beta U\right)^2+\frac{1}{120}\sum_{k,l=1}^N \frac{\lambda_k^2\lambda_l^2}{24}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)^2+\frac{7}{60}\sum_{k,l=1}^N \frac{\lambda_k^2\lambda_l^2}{24}\boldsymbol{\nabla}_k(\beta U)\cdot\boldsymbol{\nabla}_k(\nabla_l^2\beta U)\right].\end{aligned} \tag{13}$$

Equation (10) agrees with a result previously derived by Alastuey and Jancovici [8], in the case of the magnetized OCP.

Terms of higher orders are investigated in Appendix B. It is proved, using a recurrence scheme, that the contribution $(1/n!)(\Sigma_k\lambda_k^2\nabla_k^2/24)^n e^{-\beta U}$ arises at all the orders in the $\hbar^2$ expansion. This explains the choice made in writing Eq. (10) with the emergence of an exponential operator.

## III. PAIR DISTRIBUTION FUNCTION IN AN OCP

For the sake of simplicity, we restrict ourselves hereafter to the case of one single-particle species. For $N$ particles of mass $M$, enclosed in a volume $V$, the pair distribution function reads as

$$g_2(r_{12})=\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N g(\mathbf{r}_1,\ldots,\mathbf{r}_N)}{\rho^2\int\cdots\int d^3r_1\cdots d^3r_N g(\mathbf{r}_1,\ldots,\mathbf{r}_N)}, \tag{14}$$

with $\rho=N/V$. It can be expanded with respect to $\hbar^2$ as Jancovici [3] did for the first order, replacing $g(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ with its expansion (10). $g_2(r_{12})$, hereafter denoted as $g_2^q(r_{12})$, becomes

$$\begin{aligned}g_2^q(r_{12})=&\left[\exp\left(\frac{\lambda^2}{12}\nabla^2\right)g_2^c(r_{12})+\hbar^2\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N F_2}{\rho^2\int\cdots\int d^3r_1\cdots d^3r_N\exp(-\beta U)}+\hbar^4\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N F_4}{\rho^2\int\cdots\int d^3r_1\cdots d^3r_N\exp(-\beta U)}+O(\hbar^6)\right]\\&\times\left[1-\hbar^2\frac{\int\cdots\int d^{3N}rF_2}{\int\cdots\int d^{3N}r\ \exp(-\beta U)}+\hbar^4\left(\frac{\int\cdots\int d^{3N}rF_2}{\int\cdots\int d^{3N}r\ \exp(-\beta U)}\right)^2-\hbar^4\frac{\int\cdots\int d^{3N}rF_4}{\int\cdots\int d^{3N}r\ \exp(-\beta U)}+O(\hbar^6)\right],\end{aligned} \tag{15}$$

where $\lambda^2$ $(=\hbar^2\beta/M)$ is the squared de Broglie wavelength. $g_2^c$ stands for the classical pair distribution function. In the classical case, $g(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ reduces to $\exp(-\beta U)$ so that

$$g_2^c(r_{12})=\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N\exp[-\beta U(\mathbf{r}_1,\ldots,\mathbf{r}_N)]}{\rho^2\int\cdots\int d^3r_1\cdots d^3r_N\exp[-\beta U(\mathbf{r}_1,\ldots,\mathbf{r}_N)]}. \tag{16}$$

$g_2^q$ can be rewritten as

$$g_2^q(r_{12})=\exp\left(\frac{\lambda^2}{12}\nabla^2\right)g_2^c(r_{12})+\hbar^2\,\frac{\frac{N(N-1)}{\rho^2}\int\cdots\int d^3r_3\cdots d^3r_N F^2-g_2^c(r_{12})\int\cdots\int d^{3N}rF_2}{\int\cdots\int d^{3N}r\,\exp(-\beta U)}$$
$$\times\left(1-\hbar^2\,\frac{\int\cdots\int d^{3N}rF_2}{\int\cdots\int d^{3N}r\,\exp(-\beta U)}\right)-\hbar^2\left(\frac{\int\cdots\int d^{3N}rF_2}{\int\cdots\int d^{3N}r\,\exp(-\beta U)}\right)\left[\exp\left(\frac{\lambda^2}{12}\nabla^2\right)-1\right]g_2^c(r_{12})$$
$$+\hbar^4\,\frac{\frac{N(N-1)}{\rho^2}\int\cdots\int d^3r_3\cdots d^3r_N F_4-g_2^c(r_{12})\int\cdots\int d^{3N}rF_4}{\int\cdots\int d^{3N}r\,\exp(-\beta U)}+O(\hbar^6). \tag{17}$$

Now we deal with a one-component plasma (OCP) made of $N$ particles of charge $Ze$ embedded in a uniform charged background of opposite sign. In such a system, the potential energy is

$$U=\frac{1}{2}\sum_{k\neq l}u(r_{kl})-\sum_k Z^2e^2\rho\int\frac{d^3r}{|\mathbf{r}_k-\mathbf{r}|}+\frac{1}{2}Z^2e^2\rho^2\int\int\frac{d^3r\,d^3r'}{|\mathbf{r}-\mathbf{r}'|}, \tag{18}$$

where

$$u(r_{kl})=\frac{Z^2e^2}{r_{kl}}, \tag{19}$$

$\mathbf{r}_{kl}=\mathbf{r}_l-\mathbf{r}_k$, and $r=|\mathbf{r}|$. $U$ satisfies Poisson's law

$$\nabla_k^2U=-4\pi Z^2e^2\sum_{l(\neq k)}\delta(\mathbf{r}_{kl})+4\pi Z^2e^2\rho \tag{20}$$

and

$$\boldsymbol{\nabla}_k\cdot\boldsymbol{\nabla}_lU=4\pi Z^2e^2\delta(\mathbf{r}_{kl})\quad\text{if }\ k\neq l. \tag{21}$$

The last equations and the relation $\delta(\mathbf{r}_{kl})e^{-\beta U}=0$ (i.e., $e^{-\beta U}$ is null if two particles are at a same position) allow us to express Eq. (17) in the form

$$g_2^q(r_{12})=\exp\left(\frac{\lambda^2}{12}\nabla^2\right)g_2^c(r_{12})-\frac{2\lambda^4}{5(24)^2}\sum_{k,l=1}^{2}\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_le^{-\beta U})\cdot(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)}{\rho^2\int\cdots\int d^{3N}re^{-\beta U}}$$
$$+\frac{\lambda^4}{5(24)^2}\sum_{k,l=1}^{N}\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_Ne^{-\beta U}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)^2}{\rho^2\int\cdots\int d^{3N}re^{-\beta U}}-\frac{\lambda^4}{5(24)^2}g_2^c(r_{12})\sum_{k,l=1}^{N}\frac{\int\cdots\int d^{3N}re^{-\beta U}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)^2}{\int\cdots\int d^{3N}r\,e^{-\beta U}}$$
$$+O(\hbar^6). \tag{22}$$

It remains to evaluate the various tensors and contracted products which appear in the last equation. After some calculations, the last summation in Eq. (22) can be written as

$$\sum_{k,l=1}^{N}\lambda^4e^{-\beta U}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)^2=\frac{N}{3}\frac{\lambda^4}{\lambda_D^4}e^{-\beta U}+\frac{4}{3}\frac{\lambda^4}{\lambda_D^4}e^{-\beta U}\sum_{k\neq l}\frac{1}{x_{kl}^6}+\frac{2}{3}\frac{\lambda^4}{\lambda_D^4}e^{-\beta U}\sum_{\substack{k,l,m\\(k\neq l\neq m\neq k)}}\frac{P_2(\hat{\mathbf{x}}_{kl}\cdot\hat{\mathbf{x}}_{km})}{x_{kl}^3x_{km}^3}, \tag{23}$$

where $x=r/a$ $[a=(\frac{4}{3}\pi\rho)^{-1/3}]$ and $\lambda_D$ $[=(4\pi Z^2e^2\beta\rho)^{-1/2}]$ is the Debye screening length. $\hat{\mathbf{x}}$ is a unit vector: $\hat{\mathbf{x}}=\mathbf{x}/x$ and $P_2$, the second-order Legendre polynomial. Therefore,

$$\lambda^4\sum_{k,l=1}^{N}\frac{\int\cdots\int d^{3N}r\,e^{-\beta U}(\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l\beta U)^2}{\int\cdots\int d^{3N}r\,e^{-\beta U}}=\frac{N}{3}\frac{\lambda^4}{\lambda_D^4}+12\frac{\lambda^4}{\lambda_D^4}\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}g_2^c(x_{34})$$
$$+18\frac{\lambda^4}{\lambda_D^4}\int\int\int\frac{d^3x_3d^3x_4d^3x_5}{(4\pi)^3x_{34}^3x_{35}^3}g_3^c(\mathbf{x}_3,\mathbf{x}_4,\mathbf{x}_5)P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35}), \tag{24}$$

where $g_3^c$ is the classical three particle distribution function [see Eq. (27)].

Consider next the second summation in the right-hand member of Eq. (22). Noting that

$$\sum_{k,l=1}^{N} \frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}(\nabla_k\nabla_l\beta U)^2}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}} = +\frac{2N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}[(\nabla_1\nabla_2\beta U)^2+(\nabla_1\nabla_1\beta U)^2]}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}$$
$$+\frac{4N(N-1)(N-2)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}(\nabla_1\nabla_3\beta U)^2}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}$$
$$+\frac{N(N-1)(N-2)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}(\nabla_3\nabla_3\beta U)^2}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}$$
$$+\frac{N(N-1)(N-2)(N-3)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}(\nabla_3\nabla_4\beta U)^2}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}, \tag{25}$$

it is possible to write

$$\lambda^4\sum_{k,l=1}^{N}\left[\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N e^{-\beta U}(\nabla_k\nabla_l\beta U)^2}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}} - g_2^c(x_{12})\,\frac{\int\cdots\int d^{3N}r\, e^{-\beta U}(\nabla_k\nabla_l\beta U)^2}{\int\cdots\int d^{3N}r\, e^{-\beta U}}\right]$$
$$=\frac{8}{3}\frac{\lambda^4}{\lambda_D^4}\frac{1}{x_{12}^6}g_2^c(x_{12})+4\frac{\lambda^4}{\lambda_D^4}\int\frac{d^3x_3}{4\pi}g_3^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3)\left[\frac{4}{x_{13}^6}+\frac{2P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}+\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}\right]$$
$$+12\frac{\lambda^4}{\lambda_D^4}\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}[g_4^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4)-g_2^c(x_{12})g_2^c(x_{34})]+12\frac{\lambda^4}{\lambda_D^4}\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}g_4^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4)$$
$$\times\left[\frac{2P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3x_{14}^3}\right]+18\frac{\lambda^4}{\lambda_D^4}\int\int\int\frac{d^3x_3d^3x_4d^3x_5}{(4\pi)^3x_{34}^3x_{35}^3}P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})[g_5^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3,\mathbf{x}_4,\mathbf{x}_5)$$
$$-g_2^c(x_{12})g_3^c(\mathbf{x}_3,\mathbf{x}_4,\mathbf{x}_5)]. \tag{26}$$

In the last equation, $g_n^c$ is the classical $n$ particle distribution function:

$$g_n^c(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_n)=\frac{N!\int\cdots\int d^3r_{n+1}\cdots d^3r_N\exp(-\beta U)}{(N-n)!\rho^n\int\cdots\int d^{3N}r\,\exp(-\beta U)}, \tag{27}$$

with $n=2$, 3, 4, or 5.

The first summation in Eq. (22) (the summation over $k$ and $l$ taking values 1 and 2) has finally to be examined:

$$\lambda^4\sum_{k,l=1}^{2}\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N(\nabla_k\nabla_l e^{-\beta U})\cdot(\nabla_k\nabla_l\beta U)}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}=\beta\lambda^4\nabla_1\nabla_1 g_2^c(r_{12})\cdot\nabla_1\nabla_1\left[4u(r_{12})-2Z^2e^2\rho\int\frac{d^3r}{|\mathbf{r}_1-r|}\right]$$
$$+2\beta\lambda^4\rho\int d^3r_3\nabla_1\nabla_1 g_3^c(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)\cdot\nabla_1\nabla_1 u(r_{13}). \tag{28}$$

Upon performing tensorial calculations, the last equation becomes

$$\lambda^4\sum_{k,l=1}^{2}\frac{N(N-1)\int\cdots\int d^3r_3\cdots d^3r_N(\nabla_k\nabla_l e^{-\beta U})\cdot(\nabla_k\nabla_l\beta U)}{\rho^2\int\cdots\int d^{3N}r\, e^{-\beta U}}=\frac{2}{3}\frac{\lambda^4}{a^2\lambda_D^2}\nabla^2 g_2^c(x_{12})+\frac{8}{3}\frac{\lambda^4}{a^2\lambda_D^2}\frac{1}{x_{12}^3}\left(\nabla^2-\frac{3}{x_{12}}\frac{d}{dx_{12}}\right)g_2^c(x_{12})$$
$$+\frac{2\lambda^4}{a^2\lambda_D^2}\int\frac{d^3x_3}{4\pi x_{13}^3}\nabla_1\nabla_1 g_3^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3)\cdot\overleftrightarrow{\mathbf{T}}_{31}, \tag{29}$$

where $\overleftrightarrow{\mathbf{T}}_{31}$ is a tensor of order 2. In the orthogonal normalized natural base associated with the vector $\mathbf{r}_{31}$, $\overleftrightarrow{\mathbf{T}}_{31}$ reads

$$\overleftrightarrow{\mathbf{T}}_{31}=\begin{pmatrix}-2&0&0\\0&1&0\\0&0&1\end{pmatrix}. \tag{30}$$

Then the contracted tensor product in the last integral of Eq. (29) can be expressed as

$$\nabla_1\nabla_1 g_3^c(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3)\cdot\overleftrightarrow{\mathbf{T}}_{31}=-2P_0(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})x_{13}\frac{\partial}{\partial x_{13}}\left(\frac{\partial}{x_{13}\partial x_{13}}\right)g_3^c(x_{12},x_{13},x_{23})-4P_1(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})\frac{\partial^2}{\partial x_{12}\partial x_{13}}g_3^c(x_{12},x_{13},x_{23})$$
$$-2P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})x_{12}\frac{\partial}{\partial x_{12}}\left(\frac{\partial}{x_{12}\partial x_{12}}\right)g_3^c(x_{12},x_{13},x_{23}), \tag{31}$$

in which $P_0$, $P_1$, and $P_2$ are Legendre polynomials of orders 0, 1, and 2.

Finally, making use of Eqs. (26) and (29) allows us to reexpress each term of the right-hand member of Eq. (22), with the result

$$g_2^q(x_{12})=\exp\left(\frac{\lambda^2}{12a^2}\nabla^2\right)g_2^c(x_{12})-\frac{\lambda^4}{a^4}\frac{\Gamma}{180}\left[\frac{\nabla^2}{4}+\frac{1}{x_{12}}\left(\frac{1}{x_{12}}\frac{d}{dx_{12}}\right)^2\right]g_2^c(x_{12}) \tag{32a}$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma}{240}\int\frac{d^3x_3}{4\pi x_{13}^3}\nabla_1\nabla_1 g_3^c(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31} \tag{32b}$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{40}\left\{\frac{1}{3x_{12}^6}g_2^c(x_{12})+\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}\right\} \tag{32c}$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{80}\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3} \tag{32d}$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{20}\left\{\int\frac{d^3x_3}{4\pi x_{13}^6}g_3^c(1,2,3)+\frac{3}{4}\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}[g_4^c(1,2,3,4)-g_2^c(x_{12})g_2^c(x_{34})]\right\} \tag{32e}$$

$$+\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{80}\left\{\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}g_4^c(1,2,3,4)\left[\frac{2P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3x_{14}^3}\right]\right.$$
$$\left.+\frac{3}{2}\int\int\int\frac{d^3x_3d^3x_4d^3x_5}{(4\pi)^3x_{34}^3x_{35}^3}P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})\,[g_5^c(1,2,3,4,5)-g_2^c(x_{12})g_3^c(3,4,5)]\right\}+O(\lambda^6/a^6), \tag{32f}$$

where $\Gamma$ is the coupling constant:

$$\Gamma=Z^2e^2\beta/a. \tag{33}$$

$g_n^c(1,2,\ldots,n)$ stands for $g_n^c(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n)$.

Expression (32) provides an exact expansion (up to $\hbar^4$) of the quantum pair distribution function in the OCP. This result is analogous with the one obtained previously by Alastuey and Martin [4] for the charge-charge correlation function in the OCP [see Eqs. (4.40) and (4.44) in their paper, hereafter denoted as I], using very compact notations. We have checked that the term (4.40b) in I corresponds exactly to the $\hbar^4$ terms of our Lines (32a) and (32b). It could also be verified that

$$\hbar^4\times\text{Eq. (4.44a-I)}=\rho^2e^2\,\frac{1}{2}\left[\frac{\lambda^4\Gamma^2}{120a^4}\frac{1}{x_{12}^6}g_2^c(x_{12})+\text{Line (32e)}\right],$$

$$\hbar^4\times\text{Eq. (4.44b-I)}=\hbar^4\times\text{Eq. (4.44a-I)}+\rho^2e^2[\text{Line (32d)}+\text{Line (32f)}+\text{remaining term in Line (32c)}]. \tag{34}$$

In accordance with Alastuey and Martin [4], we conclude that the evaluation of the $\hbar^4$ term of the Wigner-Kirkwood expansion needs the knowledge of the classical two-, three-, four-, and five-particle distribution functions.

The following sections are devoted to a study of the WK $g_2^q(r_{12})$ expressed by Eq. (32). In order to simplify the analysis of this expression in the limit $x_{12}\to\infty$ (next section) and to develop further an approximate expression (Sec. VI), it appears useful to rewrite Line (32e) in the form

$$\text{Line (32e)}=+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{20}\int\frac{d^3x_3}{4\pi x_{13}^6}[g_3^c(1,2,3)-g_2^c(x_{12})g_2^c(x_{13})]+\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{80}\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}[g_4^c(1,2,3,4)+g_2^c(x_{12})g_2^c(x_{34})$$
$$-g_2^c(x_{12})g_3^c(1,3,4)-g_2^c(x_{12})g_3^c(2,3,4)]. \tag{35}$$

The constant $\int(d^3x/4\pi x^6)g_2^c(x)$ in the first integral cancels exactly the other constant $\frac{3}{2}\int\int[d^3x_3d^3x_4/(4\pi)^2x_{34}^6][g_3^c(1,3,4)-g_2^c(x_{34})]$ in the second integral. This can be verified expanding $g_2^c$ and $g_3^c$ in Ursell functions [Eqs. (37) and (38)] and taking into account OCP sum rules [Eqs. (41) and (42)]. In a similar way, Line (32f) is rewritten as

$$\text{Line (32f)} = +\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{80}\int\int \frac{d^3x_3 d^3x_4}{(4\pi)^2}\left[\frac{2P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3x_{14}^3}\right][g_4^c(1,2,3,4)-g_2^c(x_{12})g_3^c(1,3,4)]$$
$$+\frac{\lambda^4}{a^4}\frac{9\Gamma^2}{160}\int\int\int \frac{d^3x_3d^3x_4d^3x_5}{(4\pi)^3x_{34}^3x_{35}^3}P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})[g_5^c(1,2,3,4,5)+g_2^c(x_{12})g_3^c(3,4,5)-2g_2^c(x_{12})g_4^c(1,3,4,5)]. \tag{36}$$

As previously mentioned, the constant $\propto\int\int[d^3x_3d^3x_4/(4\pi)^2x_{13}^3x_{14}^3]P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})g_3^c(1,3,4)$ (in the first integral) compensates the other one $\int\int\int[d^3x_3d^3x_4d^3x_5/(4\pi)^3x_{34}^3x_{35}^3]P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})[g_4^c(1,3,4,5)-g_3^c(3,4,5)]$ (in the second integral). This can be checked with the help of the Ursell function expansions (38) and (39) and of the sum rules (41), (42), and (43). When lines (32e) and (32f) are expressed in the forms (35) and (36), each integral does not tend to a constant as $x_{12}$ approaches infinity (see next section).

## IV. ASYMPTOTIC BEHAVIOR OF THE QUANTUM PAIR DISTRIBUTION FUNCTION

Each term of the right-hand member of Eq. (32) is considered in the limit $x_{12}\to\infty$. This study is based on properties obeyed by the classical distribution functions $g_n^c$: the exponential clustering and a number of well-known OCP sum rules [9,10].

As Alastuey and Martin [4] did, we first expand $g_n^c$ in the Ursell functions (or truncated functions) $g_{2T}^c$, $g_{3T}^c$, $g_{4T}^c$, and $g_{5T}^c$, defined by the relations

$$g_2^c(1,2)=1+g_{2T}^c(1,2), \tag{37}$$

$$g_3^c(1,2,3)=1+g_{2T}^c(1,2)+g_{2T}^c(1,3)+g_{2T}^c(2,3)+g_{3T}^c(1,2,3), \tag{38}$$

$$g_4^c(1,2,3,4)=1+g_{2T}^c(1,2)+g_{2T}^c(1,3)+g_{2T}^c(1,4)+g_{2T}^c(2,3)+g_{2T}^c(2,4)+g_{2T}^c(3,4)+g_{2T}^c(1,2)g_{2T}^c(3,4)+g_{2T}^c(1,3)g_{2T}^c(2,4)$$
$$+g_{2T}^c(1,4)g_{2T}^c(2,3)+g_{3T}^c(1,2,3)+g_{3T}^c(1,2,4)+g_{3T}^c(1,3,4)+g_{3T}^c(2,3,4)+g_{4T}^c(1,2,3,4), \tag{39}$$

$$g_5^c(1,2,3,4,5)=1+[g_{2T}^c(1,2)+(\text{permutations})]+[g_{2T}^c(1,2)g_{2T}^c(3,4)+(\text{permutations})]+[g_{3T}^c(1,2,3)+(\text{permutations})]$$
$$+[g_{2T}^c(1,2)g_{3T}^c(3,4,5)+(\text{permutations})]+[g_{4T}^c(1,2,3,4)+(\text{permutations})]+g_{5T}^c(1,2,3,4,5). \tag{40}$$

According to the exponential clustering (valid for a plasma in the classical framework) [6], $g_{nT}^c(1,\ldots,i,\ldots,j,\ldots,n)$ decreases faster than any power of any distance $x_{ij}$, as $x_{ij}$ approaches infinity (i.e., at least exponentially).

The OCP sum rules used here are [9,10]

$$\int\frac{d^3x}{4\pi}g_{2T}^c(x)=-\frac{1}{3}, \tag{41}$$

$$\int\frac{d^3x_3}{4\pi}g_{3T}^c(1,2,3)=-\frac{2}{3}g_{2T}^c(x_{12}), \tag{42}$$

$$\int\frac{d^3x_4}{4\pi}g_{4T}^c(1,2,3,4)=-g_{3T}^c(1,2,3), \tag{43}$$

$$\int\frac{d^3x}{4\pi}x^2g_{2T}^c(x)=-\frac{2}{3\Gamma}, \tag{44}$$

$$\int\frac{d^3x_3}{4\pi}x_{13}^lP_l(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})g_{3T}^c(1,2,3)=-\frac{1}{3}x_{12}^lg_{2T}^c(x_{12})\quad(\text{if } l\geqslant1), \tag{45}$$

where $P_l$ is the Legendre polynomial of order $l$. Equation (44) (the second moment of $g_{2T}^c$) is the well-known Stillinger-Lovett condition (i.e., the perfect screening condition of an infinitesimal external charge).

Hence it follows that the terms containing derivatives in the right-hand member of Eq. (32) [Line (32a)] decrease faster than any power of $x_{12}$, as $x_{12}$ tends to infinity, and

$$\frac{1}{x_{12}^6}g_2^c(x_{12})=\frac{1}{x_{12}^6}+(\text{terms decreasing faster than any } x_{12}^{-n})\quad\text{for large } x_{12}. \tag{46}$$

Next, consider in Line (32b) the contracted product expressed in Eq. (31). Making use of the expansion of $g_3$ in truncated functions [Eq. (38)] and then eliminating the terms with zero value lead to the result

$$\int \frac{d^3x_3}{4\pi x_{13}^3}\,\boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g_3^c(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}=\int \frac{d^3x_3}{4\pi x_{13}^3}\,\boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g_{3T}^c(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}, \tag{47}$$

which decreases faster than any power of $x_{12}$, for large $x_{12}$ [as $g_{3T}^c(1,2,3)$ does].

Let us consider now Line (32e) written under the form (35). One can write

$$\int \frac{d^3x_3}{4\pi x_{13}^6}[g_3^c(1,2,3)-g_2^c(x_{12})g_2^c(x_{13})]=-g_{2T}^c(x_{12})\int \frac{d^3x}{4\pi x^6}\,g_2^c(x)+\int \frac{d^3x_{13}}{4\pi x_{13}^6}[g_{2T}^c(x_{12})+g_{2T}^c(x_{23})+g_{3T}^c(1,2,3)]. \tag{48}$$

And for large $x_{12}$

$$\int \frac{d^3x_3}{4\pi x_{13}^6}[g_3^c(1,2,3)-g_2^c(x_{12})g_2^c(x_{34})]=\int \frac{d^3x_{13}}{4\pi}\left(\frac{g_{2T}^c(x_{23})}{x_{13}^6}\right)_{\text{large } x_{12}} +(\text{terms decreasing faster than any } x_{12}^{-n}) \quad \text{for large } x_{12}. \tag{49}$$

In the same way, the second integral in Eq. (35) becomes

$$\begin{aligned}\int\int \frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}&[g_4^c(1,2,3,4)+g_2^c(x_{12})g_2^c(x_{34})-g_2^c(x_{12})g_3^c(1,3,4)-g_2^c(x_{12})g_3^c(2,3,4)]=\frac{4}{3}\,g_{2T}^c(x_{12})\int \frac{d^3x}{4\pi x^6}\,g_2^c(x)\\ &+\int\int \frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}[2g_2^c(x_{14})g_2^c(x_{23})+2g_{3T}^c(1,2,3)+g_{4T}^c(1,2,3,4)]=2\int\int \frac{d^3x_3d^3x_4}{(4\pi)^2}\left(\frac{g_{2T}^c(x_{14})g_{2T}^c(x_{23})}{x_{34}^6}\right)_{\text{large } x_{12}}\\ &+(\text{terms decreasing quickly}) \quad \text{for large } x_{12}.\end{aligned} \tag{50}$$

Consider $x_{34}^{-6}$ and $x_{13}^{-6}$ in the right-hand members of Eqs. (50) and (49):

$$x_{34}^{-6}=(x_{13}^2+x_{14}^2-2\mu_4x_{13}x_{14})^{-3}, \quad x_{13}^{-6}=(x_{12}^2+x_{23}^2-2\mu_3x_{12}x_{23})^{-3}, \tag{51}$$

with $\mu_4=\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14}$ and $\mu_3=\hat{\mathbf{x}}_{21}\cdot\hat{\mathbf{x}}_{23}$. Performing angular integrations followed by expansions in terms of inverse powers of $x_{12}$ provides

$$\begin{aligned}\frac{1}{2}\int_{-1}^{+1}\frac{d\mu_3}{x_{13}^6}&=\frac{1}{x_{12}^6}\sum_{l=0}^{\infty}\frac{(2l+4)!}{4!(2l+1)!}\frac{x_{23}^{2l}}{x_{12}^{2l}} \quad \text{if } x_{12}>x_{23},\\ \frac{1}{4}\int_{-1}^{+1}d\mu_3\int_{-1}^{+1}\frac{d\mu_4}{x_{34}^6}&=\frac{1}{x_{12}^6}\sum_{m=0}^{\infty}\sum_{l=0}^{\infty}\frac{(2l+2m+4)!}{(2l+1)!(2m+1)!}\frac{x_{14}^{2l}x_{23}^{2m}}{x_{12}^{2l+2m}}=\frac{1}{x_{12}^6}+\frac{1}{x_{12}^6}\sum_{l=1}^{\infty}\frac{(2l+4)!}{4!(2l+1)!}\frac{x_{14}^{2l}+x_{23}^{2l}}{x_{12}^{2l}}\\ &+\frac{1}{x_{12}^6}\sum_{m=2}^{\infty}\sum_{l=1}^{m-1}\frac{(2m+4)!}{4!(2l+1)!(2m-2l+1)!}\frac{x_{14}^{2l}x_{23}^{2m-2l}}{x_{12}^{2m}} \quad \text{if } x_{12}>x_{23},x_{14}.\end{aligned} \tag{52}$$

Taking into account sum rules (41) and (44), term (32e) becomes

$$\text{Line (32e)}=-\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{320}\sum_{m=0}^{\infty}\sum_{l=0}^{m}\frac{(2m+8)!S_{2l+2}S_{2m-2l+2}}{(2l+3)!(2m-2l+3)!x_{12}^{2m+10}} \quad \text{for large } x_{12}, \tag{53}$$

where $S_{2n}$ is the $2n$-order momentum of $g_{2T}^c$:

$$S_{2n}=\int \frac{d^3x}{4\pi}\,x^{2n}g_{2T}^c(x)=\int_0^{\infty}dx\;x^{2n+2}g_{2T}^c(x). \tag{54}$$

We remark that [cf. Eqs. (41) and (44)]

$$S_0=-\frac{1}{3} \quad \text{and} \quad S_2=-\frac{2}{3\Gamma}. \tag{55}$$

$S_4$ is related to the compressibility [11–13]. In Eq. (53) there is no contribution of order $x_{12}^{-8}$. Thus gathering together Eqs. (46) and (53) provides

$$\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}g_2^c(x_{12})+\text{Line (32e)}=\frac{\lambda^4}{a^4}\frac{\Gamma^2}{320}\sum_{m=0}^{\infty}\sum_{l=0}^{m}\frac{(2m+8)!S_{2l+2}S_{2m-2l+2}}{(2l+3)!(2m-2l+3)!x_{12}^{2m+10}}\quad\text{for large } x_{12}. \tag{56}$$

There is no contribution of orders $x_{12}^{-6}$ and $x_{12}^{-8}$.

As proved by Alastuey and Martin [4], the quantity

$$\frac{1}{2}\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}g_2^c(x_{12})+\frac{1}{2}[\text{Line (32e)}]$$

[which corresponds to Eq. (4.44a) in I] is the only contribution to the large-$x_{12}$ behavior of $\hbar^4$ term. Thus the sum of all the other terms [corresponding to Eq. (4.44b) in I] decreases at least exponentially. This is confirmed in Appendix C where all the integrals involving Legendre polynomials in Eq. (32) are examined. From the above analysis, it follows that the terms which contribute to the large-$x_{12}$ tail come solely from the integral

$$\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}\left(\frac{g_{2T}^c(x_{14})g_{2T}^c(x_{23})}{x_{34}^6}\right)_{\text{large } x_{12}}$$

in Eq. (50), in agreement with paper I.

As shown in Appendix C, Line (32d) decreases at least exponentially as $x_{12}$ approaches infinity. Concerning Line (32c), we prove that

$$\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}=-\frac{1}{3x_{12}^6}+(\text{terms decreasing quickly})\quad\text{for large } x_{12}. \tag{57}$$

Thus we remark that $-1/3x_{12}^6$ cancels exactly the $x_{12}^{-6}$-order term in $g_2^c(x_{12})/x_{12}^6$, in the large-$x_{12}$ limit [Eq. (46)]. So Line (32c) has no algebraic contribution in this limit. It is no longer an $x_{12}^{-6}$-order term. It is worthwhile to note that

$$\text{Line (32f)}+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{40}\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}=-\frac{1}{2}\left[\text{Line (32e)}+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}g_2^c(x_{12})\right]\quad\text{for large } x \tag{58}$$

or

$$\text{Line (32f)}-\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}=-\frac{1}{2}\left[\text{Line (32e)}+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}\right]\quad\text{for large } x. \tag{59}$$

Hence Eq. (56) gives *twice* the exact long-range expansion of the $\hbar^4$ term of $g_2^q$. In a previous Letter [14], we claimed that this remarkable relation (the factor of 2) is valid only for orders $x_{12}^{-10}$ and $x_{12}^{-12}$. In fact, this relation holds at all the next orders. As a main result, we can conclude that, for a large separation,

$$\begin{aligned} g_{2T}^q(x)\equiv g_2^q(x)-1&=\text{Line (32e)}+\text{Line (32f)}+O(\lambda^6/a^6)\quad\text{for large } x\\ &=\frac{\lambda^4}{a^4}\frac{\Gamma^2}{640}\sum_{m=0}^{\infty}\sum_{l=0}^{m}\frac{(2m+8)!S_{2l+2}S_{2m-2l+2}}{(2l+3)!(2m-2l+3)!x^{2m+10}}+O\left(\frac{\lambda^6}{a^6}\right)\quad\text{for large } x\\ &=\frac{\lambda^4}{a^4}\left[\frac{7}{9x^{10}}-\frac{21\Gamma}{2x^{12}}S_4-\frac{33\Gamma}{x^{14}}S_6+\frac{2079\Gamma^2}{40x^{14}}S_4^2+O(x^{-16})\right]+O(\lambda^6/a^6), \end{aligned} \tag{60}$$

in which $x$ substitutes for $x_{12}$.

The large-$x$ expansion of $g_2^q(x)$ (at order $\hbar^4$) is then achieved through Eq. (60) *at any order*. As proved by Alastuey and Martin [4] and Cornu and Martin [5], there is no exponential clustering in a quantum plasma. The first term (in $x^{-10}$) is in agreement with their results. In the case of plasmas with Fermi or Bose statistics, recent papers by Cornu [15] confirm the presence of algebraic tails.

## V. SMALL SEPARATION BEHAVIOR OF THE QUANTUM PAIR DISTRIBUTION FUNCTION

The right-hand member of Eq. (32) is hereafter examined in the small-$x_{12}$ limit. The validity of the resulting expansion is discussed at the end of this section. For our purpose, consider the total potential energy [Eq. (18)] for the $N$ charged particles:

$$U_N(1,\ldots,N)=\frac{Z^2e^2}{r_{12}}+\sum_{k=3}^{N}\frac{Z^2e^2}{r_{1k}}+\sum_{k=3}^{N}\frac{Z^2e^2}{r_{2k}}+\frac{1}{2}\sum_{\substack{k,l=3\\(k\neq l)}}^{N}\frac{Z^2e^2}{r_{kl}}-Z^2e^2\rho\int_N\frac{d^3r}{|\mathbf{r}-\mathbf{r}_1|}-Z^2e^2\rho\int\frac{d^3r}{|\mathbf{r}-\mathbf{r}_2|}-Z^2e^2\rho\sum_{k=3}^{N}\int\frac{d^3r}{|\mathbf{r}-\mathbf{r}_k|}$$
$$+\frac{1}{2}Z^2e^2\rho^2\int\int\frac{d^3r\,d^3r'}{|\mathbf{r}-\mathbf{r}'|}. \tag{61}$$

Let us choose the coordinate origin at the mass center of the two particles 1 and 2: $\mathbf{r}_2=-\mathbf{r}_1=\mathbf{r}_{12}/2$. In order to deduce the expansion of $U_N$, $1/r_{1k}+1/r_{2k}$ and $\int d^3r/|\mathbf{r}-\mathbf{r}_1|+\int d^3r/|\mathbf{r}-\mathbf{r}_2|$ are expanded in powers of $r_{12}$ according to

$$\beta U_N(1,2,3,\ldots,N)=\frac{\Gamma}{x_{12}}+\beta U_{(N-1)'}(0,3,\ldots,N)+\frac{\Gamma}{4}x_{12}^2+\frac{\Gamma}{4}\sum_{k=3}^{N}\left[4\pi\delta(\mathbf{x}_k)x_{12}^2P_1(\mu_k)+\frac{x_{12}^2}{x_k^3}P_2(\mu_k)\right]$$
$$+O(\Gamma x_{12}^4)\quad\text{for small } x, \tag{62}$$

with $\mu_k=\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_k$. $P_1$ and $P_2$ are the Legendre polynomials of first and second orders. $U_{(N-1)'}(0,3,\ldots,N)$ is the potential energy of $N-1$ particles: one of charge $2Ze$ at the origin and the other ones of charge $Ze$ at $\mathbf{r}_3,\ldots$ and $\mathbf{r}_N$. Then, the classical pair distribution is also expanded as

$$g_2^c(x_{12})=\frac{N(N-1)\int\cdots\int d^3x_3\cdots d^3x_N\exp[-\beta U_N(1,2,\ldots,N)]}{\rho^2a^6\int\cdots\int d^3x_1\cdots d^3x_Ne^{-\beta U_N}}$$
$$=\exp\left(-\frac{\Gamma}{x_{12}}-\frac{\Gamma}{4}x_{12}^2\right)\frac{N(N-1)}{\rho^2a^6\int d^{3N}xe^{-\beta U_N}}\int\ldots\int d^3x_3\cdots d^3x_N\exp\Bigg(-\beta U_{(N-1)'}(0,3,\ldots,N)$$
$$-\frac{\Gamma}{4}\sum_{k=3}^{N}P_2(\mu_k)\frac{x_{12}^2}{x_k^3}+O(\Gamma x_{12}^4)\Bigg)\quad\text{for small } x_{12}. \tag{63}$$

It was noted by Jancovici [16] that:

$$\frac{N(N-1)\int\cdots\int d^3x_3\cdots d^3x_N\exp[-\beta U_{(N-1)'}(0,3,\ldots,N)]}{\rho^2a^6\int\cdots\int d^3x_1\cdots d^3x_Ne^{-\beta U_N}}=\exp[\beta F(0,N)-\beta F(1,N-2)]=\exp(-\mathcal{A}), \tag{64}$$

where $\beta F(M,N)$ is the excess free energy of a mixture made of $M$ particles of charge $2Ze$ and $N$ particles of charge $Ze$. Then Eq. (63) becomes

$$g_2^c(x_{12})=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}-\frac{\Gamma}{4}x_{12}^2\right)\left(1-\frac{3\Gamma}{4}\int\frac{d^3x_3}{4\pi}g_{2'}^c(x_3)P_2(\mu_3)\frac{x_{12}^2}{x_3^3}+O(\Gamma x_{12}^4)\right)\quad\text{for small } x_{12}, \tag{65}$$

in which $g_{2'}^c(x_3)$ $[=g_{2'}^c(0,3)]$ is the pair distribution function of a charge $2Ze$ and a charge $Ze$. Let $g_{3'}^c(0,3,4)$ be the three-particle distribution function of charge $2Ze$ at the origin and two charges $Ze$ at $\mathbf{r}_3$ and $\mathbf{r}_4$ (the other particles of the plasma bear charges $Ze$). The same definitions stand for $g_{4'}^c(0,3,4,5)$ and the other distribution functions (the charge $2Ze$ is located at the origin). As the integration over $\mu_3$ yields 0, the small-$x_{12}$ expansion of $g_2^c(x_{12})$ reads

$$g_2^c(x_{12})=\exp\left[-\frac{\Gamma}{x_{12}}-\mathcal{A}-\frac{\Gamma}{4}x_{12}^2\right][1+O(x_{12}^4)]\quad\text{for small } x_{12}. \tag{66}$$

In a similar way, the other distribution functions are also expanded as

$$g_3^c(1,2,3)=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)[g_{2'}^c(0,3)+O(x_{12}^2)]\quad\text{for small } x_{12},$$

$$g_4^c(1,2,3,4)=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)[g_{3'}^c(0,3,4)+O(x_{12}^2)]\quad\text{for small } x_{12},$$

$$g_5^c(1,2,3,4,5)=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)[g_{4'}^c(0,3,4,5)+O(x_{12}^2)]\quad\text{for small } x_{12}. \tag{67}$$

In the right-hand member of Eq. (32), let us consider now the terms involving $g_3^c$. The first one [Line (32b)] can be expressed with the help of tensorial calculations as

$$\int \frac{d^3x_3}{4\pi x_{13}^3}\,\boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g_3^c(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}=\int \frac{d^3x_3}{4\pi x_{13}^3}\left[\left(\frac{4\partial}{x_{12}^2\partial x_{12}}\,x_{12}^2\,\frac{\partial}{\partial x_{12}}\right)g_3^c(x_{12},x_3,\mu_3)[-2P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})]+\frac{4\partial}{x_{12}\partial x_{12}}\,g_3^c(x_{12},x_3,\mu_3)[6P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})]\right]. \tag{68}$$

It should be noted that $x_3$ and $\mu_3$ do not depend explicitly on $\mathbf{x}_1$ and that, moreover, $\mathbf{x}_1=\frac{1}{2}\mathbf{x}_{21}$. Then performing the expansions of $x_{13}$ $[=(\frac{1}{4}x_{12}^2+x_3^2+\mu_3 x_{12}x_3)^{1/2}]$ and $P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})$ $(=P_2[(x_{12}+2\mu_3x_3)/2x_{13}])$ in powers of $x_{12}$ allows us to rewrite Eq. (68) as

$$\int \frac{d^3x_3}{4\pi x_{13}^3}\,\boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g_3^c(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}=4\int \frac{d^3x_3}{4\pi x_3^3}\left[1-3\mu_3^2+\frac{3\mu_3}{2}\,(5\mu_3^2-3)\,\frac{x_{12}}{x_3}+O\left(\frac{x_{12}^2}{x_3^2}\right)\right]\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\left[\left(\frac{\Gamma^2}{x_{12}^4}-\frac{3\Gamma}{x_{12}^3}\right)g_{2'}^c(x_3)+O(x_{12}^{-2})\right]\quad \text{for small } x_{12}$$
$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)O(x_{12}^{-2}). \tag{69}$$

In a similar way, the first term in Line (32e) is expanded as

$$\int \frac{d^3x_3}{4\pi x_{13}^6}\,g_3^c(1,2,3)=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int \frac{d^3x_3}{4\pi x_3^6}\left[1-3\mu_3\,\frac{x_{12}}{x_3}+O\left(\frac{x_{12}^2}{x_3^2}\right)\right][g_{2'}^c(x_3)+O(x_{12}^2)]\quad \text{for small } x_{12}$$
$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\left[\int_0^\infty \frac{dx_3}{x_3^4}\,g_{2'}^c(x_3)+O(x_{12}^2)\right]. \tag{70}$$

The other integrals involving $g_3^c$ [Lines (32c) and (32d)] become

$$\int \frac{d^3x_3}{4\pi x_{12}^3x_{13}^3}\,P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})g_3^c(1,2,3)=\frac{1}{x_{12}^3}\,\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int \frac{d^3x_3}{4\pi x_3^3}\left(1-3\mu_3^2+\frac{3\mu_3}{2}\,(5\mu_3^2-3)\,\frac{x_{12}}{x_3}+O(x_{12}^2/x_3^2)\right)\times[g_{2'}^c(x_3)+O(x_{12}^2)]\quad \text{for small } x_{12}$$
$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)O(x_{12}^{-1}) \tag{71}$$

and

$$\int \frac{d^3x_3}{4\pi x_{13}^3x_{23}^3}\,P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})g_3^c(1,2,3)=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int \frac{d^3x_3}{4\pi x_3^6}\,[1+O(x_{12}^2)]P_2\left(\frac{(-\frac{1}{2}\mathbf{x}_{12}-\mathbf{x}_3)\cdot(\frac{1}{2}\mathbf{x}_{12}-\mathbf{x}_3)}{x_{13}x_{23}}\right)\times[g_{2'}^c(x_3)+O(x_{12}^2)]\quad \text{for small } x_{12}$$
$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\left(\int_0^\infty \frac{dx_3}{x_3^4}\,g_{2'}^c(x_3)+O(x_{12}^2)\right). \tag{72}$$

It is easy to check that the remaining integrals in the right-hand member of Eq. (32) pertaining to Lines (32e) and (32f) have an expansion of the same form; the main term is an exponential term $\exp(-\Gamma/x_{12}-\mathcal{A})$ times a constant (which depends only on $\Gamma$):

$$\int\int \frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}\,[g_4^c(1,2,3,4)-g_2^c(x_{12})g_2^c(x_{34})]=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int \frac{d^3x_3d^3x_4}{(4\pi)^2x_{34}^6}\times[g_{3'}^c(0,3,4)-g_2^c(x_{34})+O(x_{12}^2)]\quad \text{for small } x_{12}, \tag{73}$$

$$\int\int \frac{d^3x_3 d^3x_4}{(4\pi)^2} g_4^c(1,2,3,4)\left[\frac{2P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3 x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3 x_{14}^3}\right]$$

$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int\int \frac{d^3x_3 d^3x_4}{(4\pi)^2}[g_{3'}^c(0,3,4)+O(x_{12}^2)]\left[\frac{2P_2(\hat{\mathbf{x}}_3\cdot\hat{\mathbf{x}}_{34})}{x_3^3 x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_3\cdot\hat{\mathbf{x}}_4)}{x_3^6}+O(x_{12})\right] \quad \text{for small } x_{12}$$

$$=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\left\{\int\int \frac{d^3x_3 d^3x_4}{(4\pi)^2} g_{3'}^c(0,3,4)\left[\frac{2P_2(\hat{\mathbf{x}}_3\cdot\hat{\mathbf{x}}_{34})}{x_3^3 x_{34}^3}+\frac{P_2(\hat{\mathbf{x}}_3\cdot\hat{\mathbf{x}}_4)}{x_3^6}\right]+O(x_{12})\right\}, \quad (74)$$

$$\int\int \frac{d^3x_3 d^3x_4 d^3x_5}{(4\pi)^3 x_{34}^3 x_{35}^3} P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})[g_5^c(1,2,3,4,5)-g_2^c(x_{12})g_3^c(3,4,5)]=\exp\left(-\frac{\Gamma}{x_{12}}-\mathcal{A}\right)\int\int \frac{d^3x_3 d^3x_4 d^3x_5}{(4\pi)^3 x_{34}^3 x_{35}^3} P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})$$

$$\times[g_{4'}^c(0,3,4,5)-g_3^c(3,4,5)+O(x_{12}^2)] \quad \text{for small } x_{12}. \quad (75)$$

Let us examine now terms containing derivatives [Line (32a)] which are calculated through the derivatives of $g_2^c$ [Eq. (66)]. We can conclude that, for small $x$, the main contribution at $\hbar^4$ order is $\frac{1}{2}[(\lambda^2/12a^2)\nabla^2]^2 g_2^c(x)$. Finally, the WK $g_2^q(x)$ expressed by Eq. (32) is expanded as follows:

$$g_2^q(x)=g_2^c(x)\left\{\exp\left[\frac{\lambda^2}{12a^2}\left(\frac{\Gamma^2}{x^4}-\frac{\Gamma^2}{x}+O(x^0)\right)\right]+\frac{\lambda^4}{15a^4}\left[-\frac{\Gamma^3}{2x^7}+\frac{\Gamma^2}{x^6}+O(x^{-4})\right]\right\}+O\left(\frac{\lambda^6}{a^6}\right) \quad \text{for small } x. \quad (76)$$

Only

$$\exp\left(\frac{\lambda^2}{12a^2}\nabla^2\right)g_2^c(x), \quad \frac{1}{x}\left(\frac{1}{x}\frac{d}{dx}\right)^2 g_2^c(x), \quad \text{and} \quad \frac{1}{x^6}g_2^c(x)$$

are kept in Eq. (76). The other terms are smaller if $x$ is sufficiently small. The exponential term

$$\exp\left(\frac{\lambda^2}{12a^2}\frac{\Gamma^2}{x^4}\right)$$

(which goes to $\infty$ as $x$ approaches 0) is an algebraic formulation which generalizes a result due to Jancovici [3]. Nevertheless, our study does not allow us to assert that at all orders the main term, in the small-$x$ limit,comes from this exponential term. In the present work, the $x\to 0$ limit is investigated only at order $\hbar^2$ and $\hbar^4$; so we do not know the $x\to 0$ behavior of the fully resummed WK expansion.

Assuming the $\hbar^2$ expansion to be a convergent, one implies that $x$ cannot be too small. In the other case, this semiclassical formalism would not be appropriate. But only a proper study of the convergence of the WK expansion could provide the convergence criteria. Therefore, we propose only a qualitative criteria for the validity of expansion (76). It seems reasonable to impose that

$$\frac{1}{2}\left(\frac{\lambda^2}{12a^2}\nabla^2\right)^2 g_2^c(x)<\frac{\lambda^2}{12a^2}\nabla^2 g_2^c(x)<g_2^c(x), \quad (77)$$

i.e.,

$$\frac{\lambda^2}{12a^2}\frac{\Gamma^2}{x^4}<1. \quad (78)$$

It should be mentioned that the exact quantum $g_2^q(0)$ is finite, strictly positive, and cannot be derived within the WK formalism. In particular, in the zero-density limit, Davies and Storer evaluated $g_2^q(0)$ exactly [17], and Minoo, Gombert, and Deutsch [18] studied $g_2^q(x)$ and expanded it with respect to $x$ for small $x$. Vieillefosse has also worked on this topic [19].

## VI. APPROXIMATE EXPRESSIONS FOR THE QUANTUM PAIR DISTRIBUTION FUNCTION

The net evaluation of the WK pair distribution function $g_2^q$ as derived in Eq. (32) requires the whole knowledge of the classical distribution functions $g_n^c(1,2,\dots,n)$ up to $n=5$. Therefore, it appears necessary to derive an expression which approximates $g_2^q$ accurately, but which is simpler to evaluate numerically. A first approximation has been previously presented in a Letter [14].

This approximate expression has to behave like $g_2^q$ for small and large $x_{12}$ (same expansions). Thus its derivation is based on the large- and small-$x_{12}$ behaviors detailed in Secs. IV and V. For convenience, $g_2^q(x_{12})$ is split into a short and intermediate part, on the one hand, and a long part, on the other hand, according to

$$g_2^q(x_{12})=g_s^q(x_{12})+g_l^q(x_{12}), \quad (79)$$

where the subscripts $s$ and $l$ stand for short and long separations, respectively. Following our previous analysis, we first write (at order $\hbar^4$):

$$g_s^q=\text{Line (32a)}+\text{Line (32b)}+\text{Line (32c)}+\text{Line (32d)},$$

$$g_l^q=\text{Line (32e)}+\text{Line (32f)}. \quad (80)$$

### A. Preliminaries

In the following, many terms will be computed by means of Fourier techniques. We adopt the dimensionless Fourier transforms

$$\widetilde{f}(q)=3\int_0^\infty dx\ x^2 f(x) j_0(qx)$$

$$\Leftrightarrow f(x)=\frac{2}{3\pi}\int_0^\infty dq\ q^2\widetilde{f}(q) j_0(qx), \qquad (81)$$

with $j_0(u)=\sin u/u$. The Fourier transform of $g^c_{2T}(x)$ will be denoted $h(q)$. Because of the presence of Legendre polynomials in the expressions to evaluate, a number of Fourier transforms involve spherical Bessel functions of the first kind, $j_l(u)$, with $l\geqslant 0$ $[j_l(u)=\sqrt{\frac{1}{2}\pi/u}J_{l+1/2}(u)]$. Among them, the following one plays a key role:

$$\chi(q)=3q^2\int_0^\infty \frac{dx}{x}\, g^c_2(x) j_2(qx)\equiv 3q\int_0^\infty \frac{dx}{x}\frac{dg^c_2(x)}{dx}\, j_1(qx)$$

$$\equiv 3\int_0^\infty dx\ \frac{d}{dx}\left(\frac{1}{x}\frac{dg^c_2(x)}{dx}\right) j_0(qx). \qquad (82)$$

It is easy to check, writing $g^c_2(x)\equiv 1+g^c_{2T}(x)$, that

$$\lim_{q\to 0}\frac{\chi(q)}{q^2}=1. \qquad (83)$$

As the small-$q$ behavior reflects the long-range behavior of the direct functions, it follows from Eq. (83) that neglecting functions which decrease at least exponentially as $x$ tends to infinity [i.e., $g^c_2(x)=1+g^c_{2T}(x)\to 1$] is similar to simply replacing $\chi(q)$ with $q^2$ in Fourier space.

### B. Short and intermediate separation term $g^q_s$

In the following section, all terms contained in $g^q_s$ [Eq. (80)] are evaluated numerically, at least approximately. Therefore, Lines (32b), (32c), and (32d) have to be expressed in order to compute them at any distance $x$. For this purpose, we make a wide use of the superposition approximation

$$g^c_3(1,2,3)\simeq g^c_2(x_{12})g^c_2(x_{13})g^c_2(x_{23}). \qquad (84)$$

First, consider Line (32b):

$$\text{Line (32b)}=\frac{\lambda^4}{a^4}\frac{\Gamma}{240}\, I(x_{12}), \qquad (85)$$

where

$$I(x_{12})=\int \frac{d^3x_3}{4\pi x^3_{13}}\, \boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g^c_3(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}. \qquad (86)$$

When $g^c_3(1,2,3)$ is estimated by means of the superposition approximation, Eq. (31) is modified according to

$$\boldsymbol{\nabla}_1\boldsymbol{\nabla}_1 g^c_3(1,2,3)\cdot\overleftrightarrow{\mathbf{T}}_{31}$$
$$\simeq -2P_0(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})[g^c_{2T}(x_{23})+1]$$
$$\times g^c_2(x_{12})x_{13}\frac{d}{dx_{13}}\left(\frac{d}{x_{13}dx_{13}}\, g^c_2(x_{13})\right)$$
$$-4P_1(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})[g^c_{2T}(x_{23})+1]\left(\frac{d}{dx_{12}}\, g^c_2(x_{12})\right)$$
$$\times\left(\frac{d}{dx_{13}}\, g^c_2(x_{13})\right)-2P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})[g^c_{2T}(x_{23})+1]$$
$$\times g^c_2(x_{13})x_{12}\frac{d}{dx_{12}}\left(\frac{d}{x_{12}dx_{12}}\, g^c_2(x_{12})\right). \qquad (87)$$

The terms independent of $x_{23}$ in the last equation are directly integrated on $x_3$ to give a zero result. The other ones, which are factorized by $g^c_{2T}(x_{23})$, are rewritten with the help of Eq. (D2), yielding, finally,

$$I(x)\simeq -\frac{4}{9\pi}\, g^c_2(x)\int_0^\infty dq\ q^2 h(q)\chi(q) j_0(qx)$$
$$-\frac{8}{9\pi}\left(\frac{d}{dx}\, g^c_2(x)\right)\int_0^\infty dq\ q h(q)\chi(q) j_1(qx)$$
$$-\frac{4}{9\pi}\left[x\frac{d}{dx}\left(\frac{d}{x\,dx}\, g^c_2(x)\right)\right]\int_0^\infty dq\ h(q)\chi(q) j_2(qx), \qquad (88)$$

in which $h(q)$ denotes the Fourier transform of $g^c_{2T}(x)$ and $\chi(q)$ is defined by Eq. (82). In the $x\to\infty$ limit, each term in the right-hand member of Eq. (88) decreases at least exponentially. In the $x\to 0$ limit, one gets

$$g^c_2(x)\int_0^\infty dq\ q^2 h(q)\chi(q) j_0(qx)$$
$$\underset{\text{as } x\to 0}{\sim} g^c_2(x)\int_0^\infty dq\ q^2 h(q)\chi(q),$$

$$\left(\frac{d}{dx}\, g^c_2(x)\right)\int_0^\infty dq\ q h(q)\chi(q) j_1(qx)$$
$$\underset{\text{as } x\to 0}{\sim}\frac{\Gamma}{3x}\, g^c_2(x)\int_0^\infty dq\ q^2 h(q)\chi(q),$$

$$\left[x\frac{d}{dx}\left(\frac{d}{x\,dx}\, g^c_2(x)\right)\right]\int_0^\infty dq\ h(q)\chi(q) j_2(qx)$$
$$\underset{\text{as } x\to 0}{\sim}\frac{\Gamma^2}{15x^2}\, g^c_2(x)\int_0^\infty dq\ q^2 h(q)\chi(q). \qquad (89)$$

Hence the third term appears to be the leading one in this limit. It behaves like $(\Gamma^2/x^2)g^c_2(x)$ as expected [see Eq. (69)]. Moreover, it is smaller than the ones retained in the small-$x$ approximation expressed by Eq. (76). Thus we are reinforced to use Eq. (88) in order to compute approximately $I(x)$.

Let us examine now Line (32c) which can be written as

$$\text{Line (32c)} = \frac{\lambda^4}{a^4}\frac{\Gamma^2}{40} J(x_{12}), \tag{90}$$

where

$$J(x_{12}) = \frac{1}{3x_{12}^6} g_2^c(x_{12}) + \int \frac{d^3x_3}{4\pi} g_3^c(1,2,3) \frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3 x_{13}^3}. \tag{91}$$

Its numerical evaluation needs the knowledge of $g_3^c$. The latter can also be estimated with superposition approximation (84):

$$\int \frac{d^3x_3}{4\pi} g_3^c(1,2,3) \frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3 x_{13}^3} \simeq g_2^c(x_{12}) \int \frac{d^3x_3}{4\pi} g_2^c(x_{13}) g_{2T}^c(x_{23}) \frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3 x_{13}^3}. \tag{92}$$

The remaining integration involves a convolution product and a Legendre polynomial. It can be performed, for instance, by means of Fourier-Bessel techniques [Eq. (D2)]:

$$J(x_{12}) \simeq \frac{1}{3x_{12}^6} g_2^c(x_{12}) + \frac{2}{9\pi}\frac{g_2^c(x_{12})}{x_{12}^3} \times \int_0^\infty dq\, h(q)\chi(q) j_2(qx_{12}). \tag{93}$$

The last equation can be rewritten in a different form more appropriate to an accurate numerical evaluation. The basic idea is to singularize in integral (92) the only part which involves the asymptotic limit $x_{12}\to\infty$ (which is of order $x_{12}^{-6}$). In so doing, $g_2^c(x_{13})$ is first rewritten as $1+g_{2T}^c(x_{13})$. Thus Eq. (93) becomes

$$J(x_{12}) \simeq \frac{1}{3x_{12}^6} g_2^c(x_{12}) + \frac{g_2^c(x_{12})}{x_{12}^3}[A(x_{12})+B(x_{12})], \tag{94}$$

with

$$A(x_{12}) = \int \frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{13}^3} g_{2T}^c(x_{23}),$$

$$B(x_{12}) = \int \frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{13}^3} g_{2T}^c(x_{13}) g_{2T}^c(x_{23}). \tag{95}$$

Let us first consider $B(x_{12})$. In the limit $x_{12}\to\infty$, it vanishes at least exponentially as the product $g_{2T}^c(x_{13})\, g_{2T}^c(x_{23})$ does. It can be evaluated in Fourier space [with the help of Eq. (D2)] as

$$B(x) = \frac{2}{9\pi}\int_0^\infty dq[\chi(q)-q^2]h(q)j_2(qx). \tag{96}$$

In the vicinity of $x_{12}=0$, $g_2^c(x)B(x)/x^3$ behaves like $g_2^c(x)/x$. From the above considerations, we are led to conclude that $g_2^c(x)B(x)/x^3$ plays a negligible role. This conclusion has to be achieved through numerical calculations for any distance as is done in the following section. Thus $B$ can be seen as a correction to $A$ which is expressed as [Eqs. (D12) and (D14)]

$$A(x_{12}) = -\frac{1}{3} g_{2T}^c(x_{12}) + \frac{1}{x_{12}^3}\int_0^{x_{12}} dx\, x^2 g_{2T}^c(x). \tag{97}$$

Note that, if $A(x)+B(x)$ is approximated by $A(x)$,

$$J(x) \simeq \frac{g_2^c(x)}{x^3}\left[\frac{1}{3x^3}+A(x)\right], \tag{98}$$

$J(x)$ is a function which decreases at least exponentially in the limit $x\to\infty$ and behaves as $g_2^c(x)/3x^6$ does in the case of small $x$, as expected. Making use of sum rule (41), $A(x)$ in Eq. (97) can be reexpressed in an equivalent form more appropriate to numerical computations:

$$A(x_{12}) = \frac{\frac{1}{3}x_{12}^3 g_{2T}^c(x_{12}) - \int_0^{x_{12}} dx\, x^2 g_{2T}^c(x)}{3x_{12}^3\int_0^\infty dx\, x^2 g_{2T}^c(x)} = -\frac{\int_0^{x_{12}} dx\, x^3 (d/dx) g_{2T}^c(x)}{3x_{12}^3\int_0^\infty dx\, x^3(d/dx)g_{2T}^c(x)}. \tag{99}$$

Line (32d) has also to be evaluated for any $x$, at least approximately. In the superposition approximation, it becomes

$$\text{Line (32d)} \simeq \frac{\lambda^4}{a^4}\frac{\Gamma^2}{80} K(x_{12}), \tag{100}$$

in which

$$K(x_{12}) = g_2^c(x_{12}) \int \frac{d^3x_3}{4\pi} g_2^c(x_{13}) g_2^c(x_{23}) \frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3 x_{23}^3}. \tag{101}$$

The result (D10) or, more precisely, its inverted Fourier transform

$$F(x_{23})P_n(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32}) = \frac{2i^{-n}}{3\pi}\int \frac{d^3q}{4\pi} e^{-i\mathbf{q}\cdot\mathbf{x}_{32}} P_n(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{q}})\widetilde{F}_n(q), \tag{102}$$

with

$$n=2 \quad \text{and} \quad F(x_{23}) = g_2^c(x_{23})/x_{23}^3, \tag{103}$$

allows us to evaluate $K(x_{12})$. Equation (102) becomes

$$\frac{g_2^c(x_{23})}{x_{23}^3} P_n(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32}) = -\frac{2}{3\pi}\int \frac{d^3q}{4\pi} e^{-i\mathbf{q}\cdot\mathbf{x}_{32}} P_n(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{q}})\frac{\chi(q)}{q^2}, \tag{104}$$

where $\chi(q)$ is the function defined by Eq. (82). Writing $\mathbf{x}_{32} = \mathbf{x}_{31} + \mathbf{x}_{12}$ and expanding $\exp(-i\mathbf{q}\cdot\mathbf{x}_{31})$ in terms of Legendre polynomials [Eq. (D9)], the following equation is obtained:

$$K(x) = g_2^c(x)\,\frac{2}{9\pi}\int\frac{d^3q}{4\pi}\left[\frac{\chi(q)}{q^2}\right]^2 j_0(qx). \qquad (105)$$

In the $x\to 0$ limit, $K(x)$ behaves like $g_2^c(x)$. We have to check that it decreases at least exponentially, as $x$ goes to infinity. First, let us remark that Eq. (105) is the Fourier transform of $\frac{1}{3}[\chi(q)/q^2]^2$. Recall that $\chi(q)$ is the Fourier transform of the function

$$f(x) = \frac{1}{x^2}\frac{d}{dx}\left(\frac{1}{x}\frac{dg_2^c(x)}{dx}\right). \qquad (106)$$

Thus $\chi(q)/q^2$ is the Fourier transform of a function $F(x)$ expressed as

$$F(x) = \int_x^\infty \frac{dy}{y^3}\frac{d}{dy}\,g_2^c(y), \qquad (107)$$

which is decreasing at least exponentially in the large-$x$ limit. In this limit, $K(x)$ behaves like the convolution product $F(x)\circ F(x)$, which decreases as expected.

Thus the superposition approximation does not introduce any drawback in the evaluation of Line (32d) (i.e., there is no extra contribution which does not vanish at least exponentially in the large-$x$ limit).

Therefore, all terms in $g_s^q$ [Eq. (80)] can be computed approximately for any $x$, making use of the superposition approximation (84). For that purpose, only the knowledge of the classical pair distribution function $g_2^c$ is required.

### C. Large separation term $g_l^q$

It remains to derive an approximate expression for $g_l^q$ contribution [Eq. (80)] which possesses the following properties: it provides the correct asymptotic behavior [Eq. (60)] in the $x_{12}\to\infty$ limit, and it is hidden by the dominant terms in the small $x_{12}$ range [Lines (32a) and (32c)]. For that purpose, we take advantage of the fact that Line (32e) gives *twice* the exact WK $g_{2T}^q$ behavior plus an additional term $\propto x_{12}^{-6}$, in the large-$x_{12}$ limit. Hence Line (32c) is modified in order to reproduce long-range expansion (60). Consider term (32e) expressed by Eq. (35). Making use of the superposition approximation

$$g_3^c(1,2,3) \simeq g_2^c(x_{12})g_2^c(x_{13})g_2^c(x_{23}),$$

$$g_4^c(1,2,3,4) \simeq g_2^c(x_{12})g_2^c(x_{13})g_2^c(x_{14})g_2^c(x_{23})g_2^c(x_{24})g_2^c(x_{34}), \qquad (108)$$

one gets

$$g_3^c(1,2,3) - g_2^c(x_{12})g_2^c(x_{13}) \simeq g_2^c(x_{12})g_2^c(x_{13})g_{2T}^c(x_{23}) \qquad (109)$$

and

$$\begin{aligned} &g_4^c(1,2,3,4) + g_2^c(x_{12})g_2^c(x_{34}) - g_2^c(x_{12})g_3^c(1,3,4) - g_2^c(x_{12})g_3^c(2,3,4) \\ &\simeq g_2^c(x_{12})g_2^c(x_{34})[g_{2T}^c(x_{13})g_{2T}^c(x_{23}) + g_{2T}^c(x_{13})g_{2T}^c(x_{24}) + g_{2T}^c(x_{14})g_{2T}^c(x_{23}) + g_{2T}^c(x_{14})g_{2T}^c(x_{24}) \\ &\quad + g_{2T}^c(x_{13})g_{2T}^c(x_{14})g_{2T}^c(x_{23}) + g_{2T}^c(x_{13})g_{2T}^c(x_{14})g_{2T}^c(x_{24}) + g_{2T}^c(x_{13})g_{2T}^c(x_{23})g_{2T}^c(x_{24}) + g_{2T}^c(x_{14})g_{2T}^c(x_{23})g_{2T}^c(x_{24}) \\ &\quad + g_{2T}^c(x_{13})g_{2T}^c(x_{14})g_{2T}^c(x_{23})g_{2T}^c(x_{24})]. \end{aligned} \qquad (110)$$

In the square brackets of the last equation, the two products $g_{2T}^c(x_{13})g_{2T}^c(x_{24})$ and $g_{2T}^c(x_{14})g_{2T}^c(x_{23})$ are the only ones which do not decrease at least exponentially as $x_{12}$ goes to infinity. Therefore, discarding the other terms yields a first approximation called $\mathscr{e}_1$:

$$\begin{aligned} \text{Line (32e)} \simeq \mathscr{e}_1 = {}& \frac{\lambda^4}{a^4}\frac{\Gamma^2}{20}\,g_2^c(x_{12})\int\frac{d^3x_3}{4\pi}\frac{g_2^c(x_{13})}{x_{13}^6}\,g_{2T}^c(x_{23}) \\ &+ \frac{\lambda^4}{a^4}\frac{3\Gamma^2}{40}\,g_2^c(x_{12})\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}\frac{g_2^c(x_{34})}{x_{34}^6} \\ &\times g_{2T}^c(x_{14})g_{2T}^c(x_{23}) \quad \text{for large } x_{12}. \end{aligned} \qquad (111)$$

The two integrals in this equation have to be compared with the ones which appear in the right-hand members of Eqs. (49) and (50) whose asymptotic forms are very similar, but Eq. (111) can be evaluated at any $x_{12}$ because the squared dipole potentials $x_{13}^{-6}$ and $x_{34}^{-6}$ are now weighted by $g_2^c(x_{13})$ and $g_2^c(x_{34})$, respectively. In the $x_{12}\to 0$ limit, approximation $\mathscr{e}_1$ behaves like $g_2^c(x_{12})$ times a constant, and so it is hidden by the terms kept in $g_s^q$ [Eq. (80)], as required. $\mathscr{e}_1$ is then rewritten in Fourier space,

$$\mathscr{e}_1 = \frac{\lambda^4}{a^4}\frac{\Gamma^2}{180\pi}\,g_2^c(x)\int_0^\infty dq\; q^2\varphi(q)[(h(q)+1)^2-1]j_0(qx), \qquad (112)$$

in which $\varphi(q)$ is the Fourier transform of $g_2^c(x)/x^6$:

$$\varphi(q) = 3\int_0^\infty \frac{dx}{x^4}\,g_2^c(x)j_0(qx). \qquad (113)$$

Expression (112) can be expanded in the large-$x$ limit (i.e., the small-$q$ limit). For that purpose, $j_0(qx)$, which appears in the definition of $h(q)$, is expanded, providing an expansion of $h(q)$ in terms of $S_{2n}$ [Eq. (54)]:

$$\begin{aligned}\mathscr{e}_1=&\frac{\lambda^4}{a^4}\frac{\Gamma^2}{20}g_2^c(x)\Bigg\{-\frac{g_2^c(x)}{6x^6}\\&+\frac{3}{2}\sum_{m=0}^{\infty}\sum_{\ell=0}^{m}\frac{S_{2\ell+2}S_{2m-2\ell+2}}{(2\ell+3)!(2m-2\ell+3)!}\\&\times\frac{2}{3\pi}(-1)^m\int_0^\infty dq\ q^{2m+6}\varphi(q)j_0(qx)\Bigg\}\\=&\frac{\lambda^4}{a^4}\frac{\Gamma^2}{20}g_2^c(x)\Bigg\{-\frac{g_2^c(x)}{6x^6}\\&+\frac{3}{2}\sum_{m=0}^{\infty}\sum_{\ell=0}^{m}\frac{S_{2\ell+2}S_{2m-2\ell+2}}{(2\ell+3)!(2m-2\ell+3)!}\\&\times(\nabla^2)^{m+2}\left(\frac{g_2^c(x)}{x^6}\right)\Bigg\}. \qquad (114)\end{aligned}$$

Noting that

$$\begin{aligned}(\nabla^2)^{m+2}\frac{1}{x^6}&=\left[\frac{d^{2m+2}}{dx^{2m+2}}+\left(\frac{2m+4}{x}\right)\frac{d^{2m+1}}{dx^{2m+1}}\right]\frac{1}{x^6}\\&=\frac{(2m+8)!}{4!x^{2m+10}}, \qquad (115)\end{aligned}$$

it follows that expansion (53) is exactly recovered. Thus it can be concluded that the *two functions Line (32e) and* $\mathscr{e}_1$ *are equivalent for large separation.*

In Eq. (112), the term of order $x^{-6}$, which is canceled by another one coming from term (f) of Eq. (32), can be simply erased in order to get an approximate expression for $g_l^q$:

$$\begin{aligned}g_l^q(x)=&\text{Line (32e)}+\text{Line (32f)}\simeq\tfrac{1}{2}\mathscr{e}_1+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{240}\frac{[g_2^c(x)]^2}{x^6}\\=&\frac{\lambda^4}{a^4}\frac{\Gamma^2}{360\pi}g_2^c(x)\int_0^\infty dq\ q^2\varphi(q)[h(q)+1]^2j_0(qx), \qquad (116)\end{aligned}$$

which is simple and very accurate.

Equation (116) is based on several approximations: the superposition approximation and the deletion of a lot of terms in Line (32e), as explained below Eq. (110), and in Line (32f). The major interest of the approximated formula (116) relies on the fact that it reproduces *exactly* the long-range expansion (60). So we are able to study correctly the large-$x$ tail of $g_2^q$. On the other hand, in the small separation limit, approximation (116) behaves like $g_2^c(x)$ [as Line (32e) +Line (32f) does] and it is hidden by the dominant terms which are expressed by Lines (32a) and (32d). Thus the discarded terms in Lines (32e) and (32f) can be taken as negligible.

In a previous Letter [14], another approximation to $g_l^q$ was proposed. Its limiting behaviors ($x\to 0$ and $x\to\infty$) were also correctly reproduced. Nevertheless, in the $x\to\infty$ limit, only the first order of the exact expansion was recovered. Moreover, after numerical calculations, we have been led to conclude that it could be rough at intermediate separations, which is not the case of Eq. (116).

## VII. NUMERICAL RESULTS

In order to put numbers on our previous calculations, we present in this section some numerical results for the $\hbar^4$ order term of the WK pair distribution function $g_2^q(x)$. Gathering all contributions, the latter reads for small $\lambda^2/a^2$ as

$$g_2^q(x_{12})\simeq g_2^c(x_{12})+\frac{\lambda^2}{12a^2}\nabla^2g_2^c(x_{12})+\frac{1}{2}\left(\frac{\lambda^2}{12a^2}\nabla^2\right)^2g_2^c(x_{12})-\frac{\lambda^4}{a^4}\frac{\Gamma}{180}\left[\frac{\nabla^2}{4}+\frac{1}{x_{12}}\left(\frac{d}{x_{12}dx_{12}}\right)^2\right]g_2^c(x_{12}) \qquad (117a)$$

$$\begin{aligned}&-\frac{\lambda^4}{a^4}\frac{\Gamma}{540\pi}\Bigg\{g_2^c(x_{12})\int_0^\infty dq\ q^2h(q)\chi(q)j_0(qx_{12})+2\left(\frac{d}{dx}g_2^c(x_{12})\right)\int_0^\infty dq\ qh(q)\chi(q)j_1(qx_{12})\\&+\left[x\frac{d}{dx}\left(\frac{d}{x\,dx}g_2^c(x_{12})\right)\right]\int_0^\infty dq\ h(q)\chi(q)j_2(qx_{12}) \qquad (117b)\end{aligned}$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120}\frac{g_2^c(x_{12})}{x_{12}^6}\left[1-\frac{\int_0^{x_{12}}dx\ x^3\frac{d}{dx}g_{2T}^c(x)}{\int_0^\infty dx\ x^3\frac{d}{dx}g_{2T}^c(x)}\right]+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{180\pi}\frac{g_2^c(x_{12})}{x_{12}^3}\int_0^\infty dq\ h(q)[\chi(q)-q^2]j_2(qx_{12}) \qquad (117c)$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{360\pi}g_2^c(x_{12})\int_0^\infty\frac{dq}{q^2}[\chi(q)]^2j_0(qx_{12}) \qquad (117d)$$

$$+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{360\pi}g_2^c(x_{12})\int_0^\infty dq\ q^2\varphi(q)[(h(q)+1)^2-1]j_0(qx_{12})+\frac{\lambda^4}{a^4}\frac{\Gamma^2}{240}\left(\frac{g_2^c(x_{12})}{x_{12}^3}\right)^2. \qquad (117e)+(117f)$$

Here (117a), (117b), (117c), (117d), and (117e)+(117f) refer to the various terms mentioned in Sec. VI. The form retained in Eq. (117) for line (117e)+(117f) is taken intentionally in order to get more accurate numerical results. Recall that the functions $h(q)$, $\varphi(q)$, and $\chi(q)$ are the Fourier transforms of

$$g_{2T}^c(x), \quad g_2^c(x)/x^6, \quad \text{and} \quad \frac{1}{x^2}\frac{d}{dx}\left(\frac{1}{x}\frac{dg_2^c(x)}{dx}\right)$$

[see Eq. (82)], respectively.

The parameters involved in these calculations are $\Gamma$, the plasma parameter, and the quantum parameter $\lambda^2/a^2$. At order $\hbar^4$, various terms are scaled by $\lambda^4/a^4$. Consequently, all our numerical results will be given in $\lambda^4/a^4$ units.

In order to evaluate properly Eq. (117), we have to choose a ''good'' classical pair distribution function $g_2^c(x)$ for the OCP. We retain the well-known hypernetted chain (HNC) approximation, supplemented with the extracted bridge function $b(x)$ calculated by Iyetomi *et al.* [20]. Two explanations justify this choice. On the one hand, this function reproduces accurately the ''exact'' classical pair distribution function as deduced from Monte Carlo simulations. On the other hand, according to the HNC numerical scheme (see Ng [21]), $g_2^c(x)$ is computed in its standard form [with an additive term $b(x)$]

$$g_2^c(x)=\exp\left[-\frac{\Gamma}{x}+h(x)-c(x)+b(x)\right] \qquad (118)$$

simultaneously with the direct correlation function $c(x)$ computed in Fourier space by means of the Ornstein-Zernike relation

$$h(q)=\tilde{c}(q)+h(q)\tilde{c}(q). \qquad (119)$$

By imposing the well-known limits

$$\lim_{x\to\infty} c(x)=-\beta u(x)=-\frac{\Gamma}{x},$$

$$\lim_{q\to 0} \tilde{c}(q)=-\beta\tilde{u}(q)=-\frac{3\Gamma}{q^2}, \qquad (120)$$

it follows that

$$\lim_{q\to 0} h(q)=\lim_{q\to 0}\frac{\tilde{c}(q)}{1-\tilde{c}(q)}=-1,$$

$$\lim_{q\to 0} S(q)=\lim_{q\to 0}[1+h(q)]=\frac{q^2}{3\Gamma}. \qquad (121)$$

This implies that sum rules (41) and (44) for the truncated function $g_{2T}^c$ are well fulfilled with this approximation. As a consequence, the evaluation of Line (117e)+(117f) will reproduce the exact asymptotic behavior $\frac{7}{9}(1/x^{10})$.

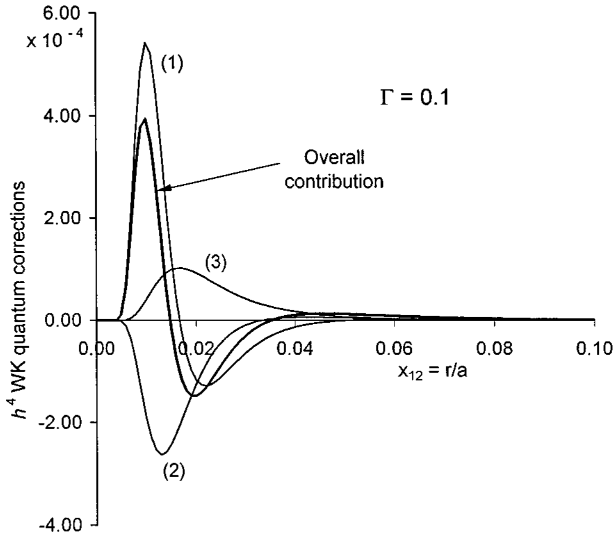


FIG. 1. The bold curve represents the WK corrections in the OCP at order $\hbar^4$ in $\lambda^4/a^4$ units as a function of the dimensionless separation $x_{12}$ computed with formula (117), in the case where $\Gamma=0.1$. $g_2^c$ is evaluated with the HNC approximation. The major contributions arise from the term proportional to the squared Laplacian [curve (1)], the term $\sim(1/x_{12})[(1/x_{12})d/dx_{12}]^2 g_2^c(x_{12})$ [curve (2)], and finally Line (117c) [curve (3)]. In this case, all the other contributions play no significant role in numerical calculations [i.e., the bold curve reduces practically to the sum of curves (1), (2), and (3)].

Numerical results were performed while computing $g_2^c(x)$ on a grid of $n=2048$ points with $dx=0.01$, in the range $0.1\leqslant\Gamma\leqslant 10$. The functions $h(q)$, $\chi(q)$, and $\varphi(q)$ as well as the other Fourier transformed functions which appear in Eq. (117) were performed by means of fast Fourier transform (FFT) techniques. Special attention was paid to the evaluation of Line (117e)+(117f), which is computed on an extended grid of 8192 points in order to extract properly its asymptotic behavior.

Figures 1–3 display some numerical results computed

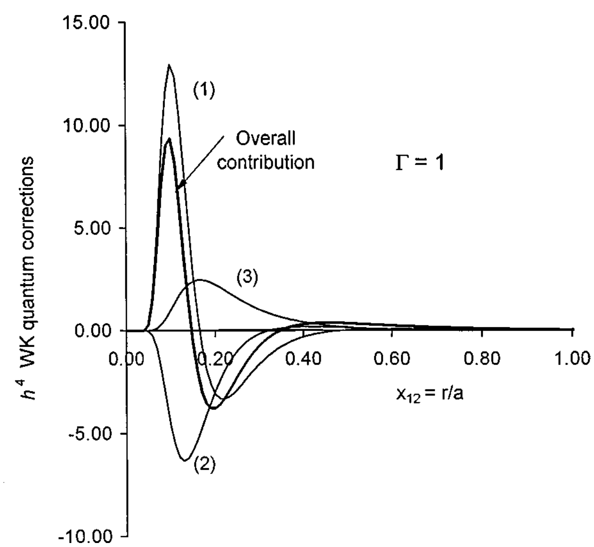


FIG. 2. WK corrections at order $\hbar^4$ in $\lambda^4/a^4$ units as a function of the dimensionless separation $x_{12}$ computed with expression (117). $\Gamma$ is kept fixed at 1. Other factors the same as in Fig. 1.

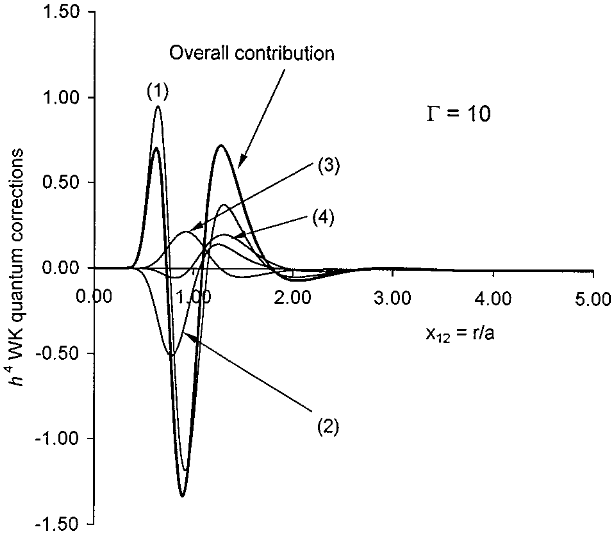


FIG. 3. Plot of various contributions to the overall $\hbar^4$ WK corrections in the OCP (bold curve which is the sum of the four other curves) in $\lambda^4/a^4$ units as a function of the separation $x_{12}$ calculated with formula (117) for $\Gamma=10$. $g_2^c$ is computed from the HNC approximation supplemented with the extracted bridge functions derived by Iyetomi *et al.* [20]. Contributions (1), (2), and (3) originate from the same terms as detailed in Fig. 1. Curve (4) corresponds to the net contribution of all the remaining terms which can no longer be neglected in the strong-coupling case. This results in an oscillating behavior of the $\hbar^4$ order.

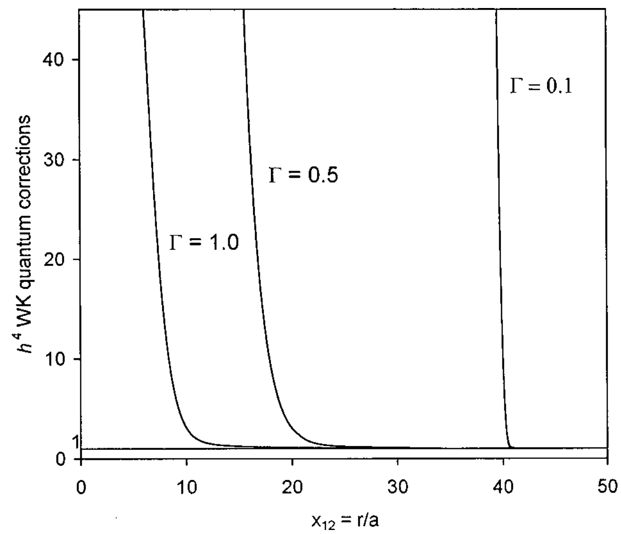


FIG. 5. Comparison between the asymptotic behavior of Line (117e)+(117f) scaled by the asymptotic limit $(7/9)x_{12}^{-10}$, for three values of $\Gamma$ in the weak-coupling range $(\Gamma\leqslant 1)$. The limit is reached at a very large distance, much larger than the distance where $g_{2T}$ can be considered to equal 0. These results are strongly dependent on $\Gamma$. The smaller $\Gamma$ is, the farther the $\hbar^4$ term behaves like $(7/9)x_{12}^{-10}$.

from Eq. (117) as detailed above, for three values of the plasma parameter $\Gamma=0.1$, 1, and 10. It is seen that in the weak-coupling range (i.e., $\Gamma\leqslant 1$), numerical contributions to the $\hbar^4$ correction for the WK pair function $g_2^q(x)$ come essentially from three terms: the squared Laplacian term, the term

$$\sim \frac{1}{x}\left(\frac{1}{x}\frac{d}{dx}\right)^2 g_2^c(x),$$

and the first term in Line (117c) [neglecting $B(x)$]. This is a consequence of the fact that at small coupling, the function $g_2^c(x)$ increases very rapidly with $x$, so that those derivative terms get significantly enhanced.

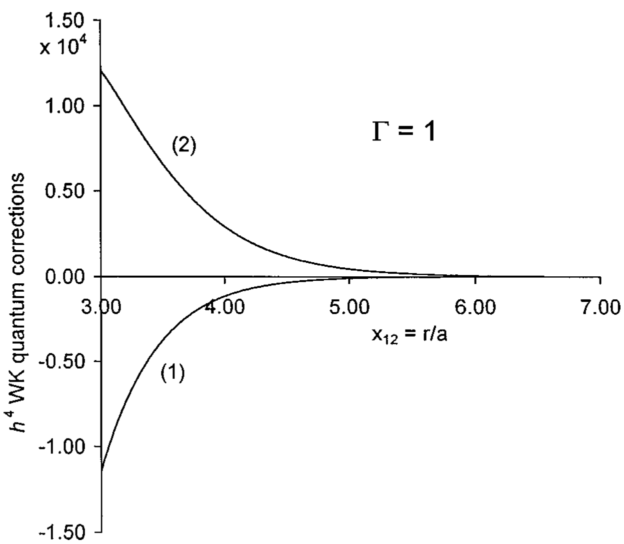


FIG. 4. Comparison between the asymptotic behaviors calculated from Line (117e)+(117f) [curve (2)] and the remaining contributions in Eq. (117) [curve (1)] for the WK corrections at order $\hbar^4$ in $\lambda^4/a^4$ units as a function of the dimensionless separation $x_{12}$, with $\Gamma=1$. This features the long-range behavior of Line (117e)+(117f) which vanishes like $(7/9)x_{12}^{-10}$, while all the other terms vanish much more quickly according to the exponential clustering.

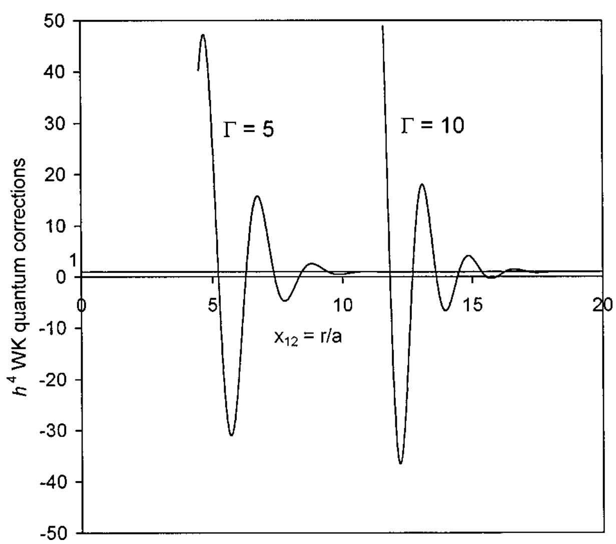


FIG. 6. Same as in Fig. 5, but it concerns two values of $\Gamma$ in the strong-coupling range $(\Gamma>1)$. As in Fig. 5, it is shown that the asymptotic limit is correctly reproduced again at a separation substantially larger than the typical values at which $g_2^c$ approaches its asymptotic limit. In contradistinction to the weak-coupling case, the term $(7/9)x_{12}^{-10}$ is now approached much more readily as $\Gamma$ is smaller. Also, it should be noticed that Line (117e)+(117f) exhibits a strong oscillating behavior at the $x_{12}\rightarrow\infty$ limit.

It should be appreciated that, at very small $\Gamma$ values (i.e., $\Gamma \leqslant 0.1$), the $\hbar^4$ correction in $\lambda^4/a^4$ units is significantly greater than 1, especially at small $x$ values. This reflects the breakdown of the WK formalism in the $x \rightarrow 0$ limit.

Results displayed in Fig. 3 show that, at large $\Gamma$ values (here $\Gamma = 10$), the contribution of the three dominant terms as described above is still important, but all the other terms [retained in Eq. (117)] contribute equally well and add to each other, resulting in an oscillating behavior.

Figures 4–6 serve to illustrate the behavior of Eq. (117) in the $x \rightarrow \infty$ limit. It should be appreciated that the rigorous asymptotic limit $\frac{7}{9}(1/x^{10})$ is correctly approached by the appropriate term retained in Eq. (117) [i.e., Line (117e) +(117f)], but only at a very large interparticle separation (much larger than the distance at which the correlation function $g_{2T}$ approaches zero), which depends on the value $\Gamma$. This is primarily a consequence of the fact that the asymptotic behavior of Line (117e)+(117f) is directly related to $S^2(q) = [1 + h(q)]^2$, in the $q \rightarrow 0$ limit. If $q^2$ is small enough, $S(q)$ is well approximated by $q^2/3\Gamma$, and if $x$ is large enough, the oscillating function $j_0(qx)$ truncates the integrant $\sim \varphi(q)(q^2/3\Gamma)^2$. Then the asymptotic limit $\frac{7}{9}(1/x^{10})$ is correctly reproduced and it arises very rapidly as $\Gamma$ increases, in the case of small $\Gamma$ values ($\Gamma \leqslant 1$) according to Fig. 5. More specifically, it should be noted that the algebraic tail appears at distances larger than the classical correlation length which controls the decay of $g_2^c(x)$. At small $\Gamma$ values, this length reduces to the Debye one $\lambda_D$ $(= a/\sqrt{3\Gamma})$ and the algebraic tail appears at distances larger and larger as $\Gamma$ decreases. In the opposite case ($\Gamma > 1$), Fig. 6 shows that the asymptotic behavior is reached much more quickly as $\Gamma$ is smaller, at least in the range of $\Gamma$ values in which computations have been performed. This is a consequence of the fact that $g_2^c$ reaches its asymptotic limit 1 at larger distances as $\Gamma$ is larger. Moreover, in this case ($\Gamma > 1$), Line (117e)+(117f) exhibits an oscillating behavior in the $x_{12} \rightarrow \infty$ limit, which is also related to the $g_2^c$ behavior.

## VIII. CONCLUSION

The $\hbar^4$ term in the Wigner-Kirkwood expansion of the pair distribution function in the one-component plasma, $g_2^q(x)$, has been derived exactly [Eq. (32)]. This term depends on the classical distribution functions for two, three, four, and five particles. Its behaviors both for large and small separations have been fully investigated with the resulting expansions (60) and (76). We have to mention that, for too small separations, the semiclassical WK formalism is not appropriate. At large distances, the polynomial behavior first proved by Alastuey and Martin [4] [term equals $(\lambda^4/a^4)\frac{7}{9}(1/x^{10})$] has been recovered and an expression for all the other terms of orders $x^{-2p}$ has been derived [Eq. (60)].

It seems unlikely to compute exactly this $\hbar^4$ correction as it would need the full knowledge of the classical distribution functions for several particles (up to 5). Then, in order to get an idea of the importance of those quantum corrections, especially of the $x^{-10}$ long-range part, an approximate but accurate expression for the $\hbar^4$ term has been derived which reproduces correctly the behavior of all the various terms in Eq. (32) in both the limits $x \rightarrow 0$ and $x \rightarrow \infty$. For that purpose, we have made much use of the superposition approximation and have suppressed some terms taken as negligible. Dealing with the long-range behavior, the approximate expansion (116) permits us to recover exactly the large distance expansion (60) at all orders. We remark that expansion (60) depends only on $g_2^c$ before any approximation. This can explain why the superposition approximation does not modify the long-distance behavior of the exact $\hbar^4$ correction. The resulting approximate expression (117) can be evaluated if the classical pair distribution function $g_2^c(x)$ is known. This was achieved numerically using the HNC approximation. The numerical results show that the asymptotic limit ($\sim x^{-10}$) is approached at very large distances, very much larger than the one at which $g_2$ reaches its asymptotic limit 1. Concerning the orders of magnitude, it appears that the long-range contribution plays, however, no significant role, in comparison with the other terms involved in Eq. (117), especially the terms which contain derivatives of $g_2^c(x)$.

We are planning to analyze the resulting effects of these quantum corrections on the thermodynamic quantities (energy, free energy, pressure, etc.) and to compare with other quantum calculations [22].

## APPENDIX A: $\hbar^4$ CORRECTION FOR $g(r_1,\ldots,r_N)$

Starting from Eq. (9) and replacing $P$ with its expansion in increasing $\hbar^2$ powers, $g$ is written in the form

$$g = \exp(-\beta U) + \hbar^2 H_2 + \hbar^4 H_4 + O(\hbar^6), \tag{A1}$$

where

$$H_2 = \int \cdots \int d^{3N}p f_2 \Bigg/ \int \cdots \int d^{3N}p \, \exp\left(-\beta \sum_{k=1}^{3N} \frac{p_k^2}{2M_k}\right) \tag{A2}$$

and

$$H_4 = \int \cdots \int d^{3N}p f_4 \Bigg/ \int \cdots \int d^{3N}p \, \exp\left(-\beta \sum \frac{p_k^2}{2M_k}\right)$$
$$= e^{-\beta U} \int \cdots \int d^{3N}p (\beta^3 G_3 + \beta^4 G_4 + \beta^5 G_5 + \beta^6 G_6) \exp\left(-\beta \sum \frac{p_k^2}{2M_k}\right) \Bigg/ \int \cdots \int d^{3N}p \, \exp\left(-\beta \sum \frac{p_k^2}{2M_k}\right). \tag{A3}$$

Replacing $f_2$ ($\equiv e^{-\beta U} g_2$) with Eq. (6) yields

$$H_2 = e^{-\beta U}\left[ -\sum_{k=1}^{3N} \frac{\beta^2}{8M_k} \frac{\partial^2 U}{\partial r_k^2} + \sum_{k=1}^{3N} \frac{\beta^3}{4(3!)M_k}\left(\frac{\partial U}{\partial r_k}\right)^2 + \sum_{k=1}^{3N} \frac{\beta^3}{4(3!)M_k^2}\frac{\partial^2 U}{\partial r_k^2}\overline{p_k^2}\right], \tag{A4}$$

where

$$\overline{f(p_k)} = \int_{-\infty}^{+\infty} dp_k f(p_k) \exp(-\beta p_k^2/2M_k) \bigg/ \int_{-\infty}^{+\infty} dp_k \times \exp(-\beta p_k^2/2M_k), \tag{A5}$$

$f(p_k)$ being any function of $p_k$. In the last sum of Eq. (6), only the terms with $k=l$ are taken into account, the rest of them becoming null after integration. The same rewriting of $H_4$ is done from expressions of $G_3$, $G_4$, $G_5$, and $G_6$. There are sums of terms containing products of two momenta:

$$\sum_{l,m=1}^{3N} \frac{p_l p_m}{M_l M_m}\frac{\partial^2 U}{\partial r_l \partial r_m} \quad \text{or} \quad \left(\sum_{l=1}^{3N}\frac{p_l}{M_l}\frac{\partial U}{\partial r_l}\right)^2.$$

Only the cases where the two momenta are the same contribute to $H_4$ (i.e., $p_k p_l$ does not contribute to $H_4$ if $p \neq l$). In the expressions of $G_5$ and $G_6$, there are products of four momenta: $p_k p_l p_m p_n$. Only the following cases are considered:

(1) $k=l$, $m=n$, and $k\neq m$,
(2) $k=m$, $l=n$, and $k\neq l$,
(3) $k=n$, $l=m$, and $k\neq l$,
(4) $k=l=m=n$,

the other terms yielding a zero result. Using the relations

$$\overline{p_k^2} = M_k/\beta \quad \text{and} \quad \overline{p_k^4} = 3M_k^2/\beta^2 \tag{A6}$$

gives

$$H_2 = e^{-\beta U}\left[ -\sum_{k=1}^{3N}\frac{\beta^2}{2(3!)M_k}\frac{\partial^2 U}{\partial r_k^2} + \sum_{k=1}^{3N}\frac{\beta^3}{4(3!)M_k}\left(\frac{\partial U}{\partial r_k}\right)^2\right] \tag{A7}$$

and

$$\begin{aligned} H_4 = e^{-\beta U}\Bigg\{ &\frac{1}{2}\left[ -\sum_{k=1}^{3N}\frac{\beta^2}{2(3!)M_k}\frac{\partial^2 U}{\partial r_k^2} + \sum_{k=1}^{3N}\frac{\beta^3}{4(3!)M_k}\left(\frac{\partial U}{\partial r_k}\right)^2\right]^2 - \frac{\beta^3}{2(5!)}\left(\sum_{k=1}^{3N}\frac{1}{M_k}\frac{\partial^2}{\partial r_k^2}\right)^2 U \\ &+ \frac{\beta^4}{3(5!)}\sum_{k,l=1}^{3N}\frac{1}{M_k M_l}\left(\frac{\partial^2 U}{\partial r_k \partial r_l}\right)^2 + \frac{\beta^4}{5!}\sum_{k,l=1}^{3N}\frac{1}{M_k M_l}\frac{\partial U}{\partial r_k}\frac{\partial^3 U}{\partial r_k \partial r_l^2} \\ &- \frac{\beta^5}{2(5!)}\sum_{k,l=1}^{3N}\frac{1}{M_k M_l}\frac{\partial^2 U}{\partial r_k \partial r_l}\frac{\partial U}{\partial r_k}\frac{\partial U}{\partial r_l}\Bigg\}. \end{aligned} \tag{A8}$$

Derivatives of $\exp(-\beta U)$ are introduced to simplify the writing of $H_2$ and $H_4$, as Jancovici [3] did. Then Eqs. (12) and (13), written by means of tensorial notation, are obtained.

## APPENDIX B: INVESTIGATION OF ORDERS HIGHER THAN $\hbar^4$

Here the purpose is to investigate the orders higher than $\hbar^4$, for the WK distribution functions.

$P(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{p}_1,\ldots,\mathbf{p}_N)$ is expanded as follows:

$$P = \exp(-\beta\epsilon)[g_0 + \hbar^2 g_2 + \hbar^4 g_4 + \cdots + \hbar^{2n} g_{2n} + \cdots]. \tag{B1}$$

$g_0$ is 1, $g_2$ is a sum of $\beta^2$- and $\beta^3$-order terms, and $g_4$ is a sum of terms of $\beta^3$, $\beta^4$, $\beta^5$, and $\beta^6$ orders. What about $g_{2n}$?

From Eq. (3), the differential equation which defines $f_{2n}$ ($=e^{-\beta\epsilon}g_{2n}$), it can be seen that the highest power of $\beta$ is $\beta^{3n}$. Let us show that this term is

$$\begin{aligned} A_{2n}\beta^{3n} &= \frac{\beta^{3n}}{n!}\left[\sum_{k=1}^{N}\frac{(\boldsymbol{\nabla}_k U)^2}{24M_k} + \sum_{k,l=1}^{N}\frac{\mathbf{p}_k\mathbf{p}_l}{24M_k M_l}\cdot\boldsymbol{\nabla}_k\boldsymbol{\nabla}_l U\right]^n \\ &= \frac{\beta^{3n}}{n!}(A_2)^n. \end{aligned} \tag{B2}$$

It is true for $n=0$, 1, and 2. Assuming Eq. (B2), $A_{2n+2}$ is a solution to the equation

$$\begin{aligned} &-\sum_{k=1}^{N}\frac{\mathbf{p}_k}{M_k}\cdot(\boldsymbol{\nabla}_{\mathbf{r}_k}A_{2n+2}) + \sum_{k=1}^{N}(\boldsymbol{\nabla}_{\mathbf{r}_k}U)\cdot(\boldsymbol{\nabla}_{\mathbf{p}_k}A_{2n+2}) \\ &= -\frac{A_{2n}}{24}\sum_{k,l,m=1}^{N}\frac{\mathbf{p}_k\mathbf{p}_l\mathbf{p}_m}{M_k M_l M_m}\cdot\boldsymbol{\nabla}_{\mathbf{r}_k}\boldsymbol{\nabla}_{\mathbf{r}_l}\boldsymbol{\nabla}_{\mathbf{r}_m}U, \end{aligned} \tag{B3}$$

which gives

$$A_{2n+2} = \frac{1}{(n+1)!}(A_2)^{n+1}. \tag{B4}$$

Considering the smallest order in $\beta$, we verify that it is

$$B_{2n}\beta^{n+1} = -\frac{\beta}{n!}\left(\sum_{k=1}^{N}\frac{\beta\nabla_k^2}{8M_k}\right)^n U. \tag{B5}$$

It is true for $g_2$ and $g_4$. If it is true until $g_{2n-2}$, the smallest power of $\beta$ in the right-hand member of Eq. (3) is $\beta^{n+1}$ and $B_{2n}$ satisfies a differential equation

$$\begin{aligned} &-\sum_{k=1}^{N}\frac{\mathbf{p}_k}{M_k}\cdot(\boldsymbol{\nabla}_{\mathbf{r}_k}B_{2n}) + \sum_{k=1}^{N}(\boldsymbol{\nabla}_{\mathbf{r}_k}U)\cdot(\boldsymbol{\nabla}_{\mathbf{p}_k}B_{2n}) \\ &= \frac{1}{8^n(n!)}\sum_{k_1,\ldots,k_n,k_{n+1}=1}^{N} \times\frac{\mathbf{p}_{k_{n+1}}}{M_1\cdots M_{k_{n+1}}}\cdot\boldsymbol{\nabla}_{\mathbf{r}_{k_{n+1}}}\nabla^2_{\mathbf{r}_{k_1}}\cdots\nabla^2_{\mathbf{r}_{k_n}}U. \end{aligned} \tag{B6}$$

$B_{2n}$ defined by Eq. (B5) is a solution to Eq. (B6).

Therefore, $P$ may be expressed as

$$\begin{aligned} P = \exp(-\beta\epsilon)[&1 + \hbar^2(B_2\beta^2 + A_2\beta^3) \\ &+ \hbar^4(B_4\beta^3 + \cdots + A_4\beta^6) \\ &+ \cdots + \hbar^{2n}(B_{2n}\beta^{n+1} + \cdots + A_{2n}\beta^{3n}) + \cdots], \end{aligned} \tag{B7}$$

where $A_{2n}$ and $B_{2n}$ are defined by Eqs. (B2) and (B5).

What is possible to write concerning $g(\mathbf{r}_1,\ldots,\mathbf{r}_N)$? The averages on the momenta modify the orders with respect to $\beta$. Each product of two momenta decreases the order in $\beta$ by 1, after the integrations. The general form of $g$ is

$$g=\exp(-\beta U)[1+\hbar^2E_2+\hbar^4E_4+\cdots+\hbar^{2n}E_{2n}+\cdots] \tag{B8}$$

Considering the expansion of $E_{2n}$ in powers of $\beta$, it is easy to check that the highest-order term is

$$C_{2n}=\frac{\beta^{3n}}{n!}\left[\sum_{k=1}^{N}\frac{(\nabla_kU)^2}{24M_k}\right]^n. \tag{B9}$$

Thus $g$ is of the form

$$g=\exp\left(\sum_{k=1}^{N}\frac{\lambda_k^2}{24}\nabla_k^2\right)e^{-\beta U}+e^{-\beta U}[\hbar^2I_2\beta^2+\hbar^4(I_4\beta^3+\cdots+J_4\beta^5)+\cdots+\hbar^{2n}(I_{2n}\beta^{n+1}+\cdots+J_{2n}\beta^{3n-1})\cdots], \tag{B10}$$

where

$$\exp\left(\sum_{k=1}^{N}\frac{\lambda_k^2}{24}\nabla_k^2\right)=\sum_{i=1}^{\infty}\frac{1}{i!}\left(\sum_{k=1}^{N}\frac{\lambda_k^2}{24}\nabla_k^2\right)^i \tag{B11}$$

is an operator applied to the function $\exp[-\beta U(\mathbf{r}_1,\ldots,\mathbf{r}_N)]$. $I_{2n}$ and $J_{2n}$ are functions of $U$, gradients $U$, and Laplacians $U$. That explains the form of Eq. (10).

## APPENDIX C: ASYMPTOTIC BEHAVIORS OF THE TERMS IN INVOLVING LEGENDRE POLYNOMIALS IN EQ. (32)

The first term comes from Line (32c):

$$\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}=\int\frac{d^3x_3}{4\pi}[g_{2T}^c(2,3)+g_{3T}^c(1,2,3)]\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13}}{x_{12}^3x_{13}^3}. \tag{C1}$$

The other terms in expansion of $g_3^c$ [Eq. (38)] yield zero integrals. $g_{3T}^c(1,2,3)$ tends to zero at least exponentially as $x_{12}$ approaches infinity. In Appendix D, it is proved that

$$A(x_{12})=\int\frac{d^3x_3}{4\pi}g_{2T}^c(x_{23})\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{13}^3}=-\frac{1}{3}g_{2T}^c(x_{12})+\frac{1}{x_{12}^3}\int_0^{x_{12}}dx\,x^2g_{2T}^c(x). \tag{C2}$$

Thus, in the large-$x_{12}$ limit, making use of sum rule (41) allows us to deduce the relation

$$\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{12}^3x_{13}^3}=-\frac{1}{3x_{12}^6}+(\text{terms decreasing quickly})\quad\text{for large }x_{12}. \tag{C3}$$

Here $-1/3x_{12}^6$ cancels exactly the large-$x_{12}$ term in $g_2^c(x_{12})/x_{12}^6$. Another integral that we have to study is [Line (32d)]

$$\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}=2\int_{x_{13}\leqslant x_{23}}\frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}[1+g_{2T}^c(x_{12})+g_{2T}^c(x_{13})+g_{2T}^c(x_{23})+g_{3T}^c(1,2,3)]. \tag{C4}$$

With the help of Appendix E [Eqs. (E6) and (E7)], the last expression is rewritten as

$$\int\frac{d^3x_3}{4\pi}g_3^c(1,2,3)\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}=\int_{x_{12}/2}^{\infty}dx_{13}\,g_{2T}^c(x_{13})\left(\frac{1}{x_{13}^4}-\frac{x_{12}}{x_{13}^5}+\frac{x_{12}^3}{8x_{13}^7}\right)+2\int_{x_{13}\leqslant x_{23}}\frac{d^3x_3}{4\pi}[g_{2T}^c(x_{23})+g_{3T}^c(1,2,3)]\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32}}{x_{13}^3x_{23}^3}. \tag{C5}$$

As $x_{23}\geqslant x_{13}\Rightarrow x_{23}\geqslant x_{12}/2$, Line (32d) is shown to decrease at least exponentially as $x_{12}$ approaches infinity. We have to consider finally term (32f) expressed by Eq. (36). The various distributions are expanded in truncated Ursell functions. In so doing, Line (32f) becomes

$$\begin{aligned}\text{Line (32f)}=&\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{40}\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2x_{13}^3x_{34}^3}P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})[g_{2T}^c(1,4)g_{2T}^c(2,3)+g_{2T}^c(1,3)g_{2T}^c(2,4)+g_{3T}^c(2,3,4)+g_{4T}^c(1,2,3,4)]\\&+\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{80}\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2x_{13}^3x_{14}^3}P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})[g_{3T}^c(2,3,4)+g_{4T}^c(1,2,3,4)]\\&+\frac{\lambda^4}{a^4}\frac{9\Gamma^2}{160}\int\int\int\frac{d^3x_3\,d^3x_4\,d^3x_5}{(4\pi)^3x_{34}^3x_{35}^3}P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})[2g_{2T}^c(1,3)g_{3T}^c(2,4,5)+4g_{2T}^c(1,4)g_{3T}^c(2,3,5)+g_{5T}^c(1,2,3,4,5)].\end{aligned} \tag{C6}$$

In this equation, the terms left out vanish, as can be checked by angular integration and involving Eq. (E6).

Appendixes D and E allow us to study the first integral in the right-hand member of Eq. (C6). Taking into account Eq. (D14), it may be seen that

$$\int\int \frac{d^3x_3\, d^3x_4}{(4\pi)^2}\left(\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3 x_{34}^3}\, g_{2T}^c(x_{23}) g_{2T}^c(x_{14})\right)_{\text{large } x_{12}} = -\frac{1}{3}\int \frac{d^3x_3}{4\pi}\left(\frac{g_{2T}^c(x_{23})}{x_{13}^6}\right)_{\text{large } x_{12}} + (\text{terms decreasing quickly}), \quad \text{(C7)}$$

since the convolution product of two fast decreasing functions is a fast decreasing function. Next, Eq. (E7) leads us to see that

$$\int \frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3 x_{34}^3}\, g_{2T}^c(x_{13})$$

is convergent and is decreasing at least exponentially as $x_{14}$ increases indefinitely. Here

$$\int\int \frac{d^3x_3\, d^3x_4}{(4\pi)^2}\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3 x_{34}^3}\, g_{2T}^c(x_{13}) g_{2T}^c(x_{24})$$

has the same property in the large-$x_{12}$ limit (it is a convolution product of two functions decreasing at least exponentially). As $x_{12}$ goes to infinity, it is deduced from Eq. (E12) that

$$\int\int \frac{d^3x_3\, d^3x_4}{(4\pi)^2}\left(\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3 x_{34}^3}\, g_{3T}^c(2,3,4)\right)_{\text{large } x_{12}} = (\text{terms decreasing faster than any } x_{12}^{-n}). \quad \text{(C8)}$$

Let us examine now the second integral in the right-hand member of Eq. (C6). In Appendix F, the following expansion valid for large $x_{12}$ is derived:

$$I = \int\int \frac{d^3x_3\, d^3x_4}{(4\pi)^2}\left(\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3 x_{14}^3}\, g_{3T}^c(2,3,4)\right)_{\text{large } x_{12}} = -\frac{2}{3x_{12}^6} S_0 - \frac{1}{18}\sum_{\ell=1}^{\infty}\frac{(\ell+1)(\ell+2)(2\ell+3)}{x_{12}^{2\ell+6}} S_{2\ell} \quad \text{for large } x_{12}$$

$$= \frac{2}{9x_{12}^6} - \frac{1}{72}\sum_{\ell=1}^{\infty}\frac{(2\ell+4)!}{(2\ell+1)!\, x_{12}^{2\ell+6}} S_{2\ell}, \quad \text{(C9)}$$

where $S_{2l}$ is defined by Eq. (54).

In the last integral of the right-hand member of Eq. (C6), the term

$$\int \frac{d^3x_4}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})}{x_{34}^3 x_{35}^3}\, g_{3T}^c(2,3,5) g_{2T}^c(x_{14})$$

is a convolution product of $g_{2T}^c(x_{12})$ and $[P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{35})/x_{13}^3 x_{35}^3] g_{3T}^c(2,3,5)$. Integrating over $\mathbf{x}_3$ and $\mathbf{x}_5$ yields, in the large-$x_{12}$ limit, a convolution product of two fast decreasing funtions [see Eq. (C8)]. Thus it decreases at least exponentially as $x_{12}$ increases indefinitely. Note also that

$$\int \frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})}{x_{34}^3 x_{35}^3}\, g_{3T}^c(2,4,5) g_{2T}^c(x_{13})$$

is a convolution product of $[P_2(\hat{\mathbf{x}}_{14}\cdot\hat{\mathbf{x}}_{15})/x_{14}^3 x_{15}^3] g_{3T}^c(2,4,5)$ with $g_{2T}^c(x_{12})$. Thus, integrating over the angles and expanding in decreasing powers of $x_{12}$, we arrive at

$$\int\int\int \frac{d^3x_3\, d^3x_4\, d^3x_5}{(4\pi)^3}\left(\frac{P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})}{x_{34}^3 x_{35}^3}\, g_{3T}^c(2,4,5) g_{2T}^c(x_{13})\right)_{\text{large } x_{12}}$$

$$= \int \frac{d^3x_3}{4\pi}\left[g_{2T}^c(x_{13})\left(-\frac{2}{3x_{23}^6} S_0 - \frac{1}{18}\sum_{\ell=1}^{\infty}\frac{(\ell+1)(\ell+2)(2\ell+3)}{x_{23}^{2\ell+6}} S_{2\ell}\right)\right]_{\text{large } x_{12}} \quad \text{for large } x_{12}. \quad \text{(C10)}$$

Integrating next $x_{23}^{-6-2\ell}$ $[=(x_{12}^2+x_{13}^2-2\mathbf{x}_{12}\cdot\mathbf{x}_{13})^{-3-\ell}]$ over the angles and expanding it in decreasing powers of $x_{12}$, the last equation becomes

$$\int\int\int \frac{d^3x_3\,d^3x_4\,d^3x_5}{(4\pi)^3}\left(\frac{P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})}{x_{34}^3x_{35}^3}\,g_{3T}^c(2,4,5)g_{2T}^c(x_{13})\right)_{\text{large }x_{12}}$$

$$=\frac{2}{9}\int\frac{d^3x_3}{4\pi}\left(\frac{g_{2T}^c(x_{13})}{x_{23}^6}\right)_{\text{large }x_{12}}-\frac{1}{72}\sum_{\ell=1}^{\infty}\sum_{m=0}^{\ell-1}\frac{(2\ell+4)!S_{2m}S_{2\ell-2m}}{(2m+1)!(2\ell-2m+1)!x_{12}^{2\ell+6}}\quad\text{for large }x_{12}. \tag{C11}$$

Note that the integral

$$\int\frac{d^3x_3}{4\pi}\left(\frac{g_{2T}^c(x_{13})}{x_{23}^6}\right)_{\text{large }x_{12}}$$

compensates the same integral in the right-hand member of Eq. (C7) [owing to the coefficients in Eq. (C6)] and that the term corresponding to $m=0$ in the last expression cancels exactly the sum over $\ell$ in Eq. (C9). Gathering together all these results [Eqs. (C7), (C9), and (C11)], term (32f) is expanded in powers of $x_{12}^{-2}$ as follows:

$$\text{Line (32f)}=\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{40}\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2}\left(\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}\,g_{2T}^c(x_{23})g_{2T}^c(x_{14})\right)_{\text{large }x_{12}}$$

$$+\frac{\lambda^4}{a^4}\frac{3\Gamma^2}{80}\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2}\left(\frac{P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})}{x_{13}^3x_{14}^3}\,g_{3T}^c(2,3,4)\right)_{\text{large }x_{12}}$$

$$+\frac{\lambda^4}{a^4}\frac{9\Gamma^2}{80}\int\int\int\frac{d^3x_3\,d^3x_4\,d^3x_5}{(4\pi)^3}\left(\frac{P_2(\hat{\mathbf{x}}_{34}\cdot\hat{\mathbf{x}}_{35})}{x_{34}^3x_{35}^3}\,g_{3T}^c(2,4,5)g_{2T}^c(x_{13})\right)_{\text{large }x_{12}}\quad\text{for large }x_{12}$$

$$=\frac{\lambda^4}{a^4}\frac{\Gamma^2}{120x_{12}^6}-\frac{\lambda^4}{a^4}\frac{\Gamma^2}{640}\sum_{m=0}^{\infty}\sum_{\ell=0}^{m}\frac{(2m+8)!S_{2\ell+2}S_{2m-2\ell+2}}{(2\ell+3)!(2m-2\ell+3)!x_{12}^{2m+10}}\quad\text{for large }x_{12}. \tag{C12}$$

At this stage, we have verified Eqs. (58) and (59), valid in the large-$x_{12}$ limit.

### APPENDIX D: EVALUATION OF $A(x_{12})$ [Eq. (C2)]

As this integral will be also used in the computations performed in Sec. VII, our purpose is to calculate it exactly for any $x_{12}$ (not only in the case where $x_{12}$ is large).

Consider $F(x)$ and $G(x)$, two functions of the distance $x$, and define another function $S_{n,F,G}(x)$ as follows:

$$S_{n,F,G}(x_{12})=\int\frac{d^3x_3}{4\pi}\,G(x_{23})F(x_{13})P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13}), \tag{D1}$$

where $n$ is an integer and $P_n$ is the Legendre polynomial of order $n$. The last integral is assumed to be convergent. The first step of this appendix is the proof of the relation

$$S_{n,F,G}(x_{12})=\frac{2}{9\pi}\int_0^\infty dq\;q^2\tilde{G}_0(q)\tilde{F}_n(q)j_n(qx_{12}) \tag{D2}$$

with

$$\tilde{H}_\ell(q)=3\int_0^\infty dx\;x^2H(x)j_\ell(qx). \tag{D3}$$

The functions $j_\ell$ are the spherical Bessel functions of first kind. Note that $\tilde{H}_0$ is the Fourier transform of $H$. Equation (D1) expresses a convolution product. Thus Eq. (D2) is verified if $n=0$ and, in the general case (for any $n$), we are allowed to rewrite $S_{n,F,G}$ with the help of Fourier transforms:

$$S_{n,F,G}(x_{12})=\frac{1}{24\pi^3}\int d^3q\;e^{-i\mathbf{q}\cdot\mathbf{x}_{12}}\tilde{G}_0(q)\times\int d^3x\;e^{i\mathbf{q}\cdot\mathbf{x}}F(x)P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}). \tag{D4}$$

In polar coordinates, let

$$\mathbf{q}=\begin{pmatrix}0\\0\\q\end{pmatrix},\quad \mathbf{x}_{12}=\begin{pmatrix}0\\x_{12}\sin\theta_2\\x_{12}\cos\theta_2\end{pmatrix},$$

$$\text{and}\quad \mathbf{x}=\begin{pmatrix}x\sin\theta\cos\varphi\\x\sin\theta\sin\varphi\\x\cos\theta\end{pmatrix}. \tag{D5}$$

Thus

$$\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}=\sin\theta_2\sin\theta\sin\varphi+\cos\theta_2\cos\theta. \tag{D6}$$

If $n\neq 0$, $P_n$ is expanded in terms of the associated Legendre functions of the first kind $P_n^m$:

$$P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}})=P_n(\cos\ \theta_2)P_n(\cos\ \theta)+2\sum_{m=1}^{n}\frac{(n-m)!}{(n+m)!}\times P_n^m(\cos\ \theta_2)P_n^m(\cos\ \theta)\cos\left(\frac{m\pi}{2}-m\varphi\right). \tag{D7}$$

Integrating over $\varphi$ cancels each term in the last expansion, except for the first one:

$$\int_0^{2\pi}d\varphi\ P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}})=2\pi P_n(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{12})P_n(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}). \tag{D8}$$

Expanding the exponential

$$e^{i\mathbf{q}\cdot\mathbf{x}}=\sum_{\ell=0}^{\infty}(2\ell+1)i^{\ell}j_{\ell}(qx)P_{\ell}(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}) \tag{D9}$$

gives

$$\begin{aligned}\int d^3x\ e^{i\mathbf{q}\cdot\mathbf{x}}F(x)P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}})&=2\pi P_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{q}})\int_0^{\infty}dx\ x^2F(x)\int_{-1}^{+1}d\mu\ e^{iqx\mu}P_n(\mu)\\&=\frac{4\pi}{3}\ i^nP_n(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{q}})\widetilde{F}_n(q).\end{aligned} \tag{D10}$$

Next, the same calculations are performed for the integration over $\theta_2$. Finally, Eq. (D2) is obtained.

Consider now the case where

$$n=2,\quad F(x)=\lim_{\varepsilon\to 0}\frac{\exp(-\varepsilon x)}{x^3}\quad\text{and}\quad G(x)=g_{2T}^c(x). \tag{D11}$$

$\widetilde{G}_0(q)$ is $h(q)$, the Fourier transform of $g_{2T}^c(x)$, and the transform $\widetilde{F}_2(q)$ is simply 1. Then Eq. (D2) becomes

$$\begin{aligned}S_{2,F,G}(x_{12})=A(x_{12})&=\frac{2}{9\pi}\int_0^{\infty}dq\ q^2h(q)j_2(qx_{12})\\&=-\frac{2}{9\pi}\int_0^{\infty}dq\ q^2h(q)j_0(qx_{12})\\&\quad-\frac{2}{3\pi x_{12}}\frac{d}{dx_{12}}\int_0^{\infty}dq\ q^2\frac{h(q)}{q^2}j_0(qx_{12})\\&=-\frac{1}{3}g_{2T}^c(x_{12})-\frac{1}{x_{12}}\frac{d}{dx_{12}}\int\frac{d^3x_3}{4\pi x_{13}}g_{2T}^c(x_{23}).\end{aligned} \tag{D12}$$

Integrating over the angles

$$\frac{1}{2}\int_{-1}^{+1}\frac{d\mu}{x_{13}}=\frac{1}{2}\int_{-1}^{+1}\frac{d\mu}{(x_{12}^2+x_{23}^2-2\mu x_{12}x_{23})^{1/2}}=\frac{1}{\sup(x_{12},x_{23})} \tag{D13}$$

yields Eq. (C2):

$$\begin{aligned}A(x_{12})&=\int\frac{d^3x_3}{4\pi}g_{2T}^c(x_{23})\frac{P_2(\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})}{x_{13}^3}\\&=-\frac{1}{3}g_{2T}^c(x_{12})+\frac{1}{x_{12}^3}\int_0^{x_{12}}dx\,x^2g_{2T}^c(x).\end{aligned} \tag{D14}$$

Note that the last integral in Eq. (D12) is convergent.

## APPENDIX E: SOME INTEGRALS INVOLVING LEGENDRE POLYNOMIALS

Consider $P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})/x_{13}^3x_{23}^3$ [cf. Eq. (C4)]. We have to prove that the integral

$$\int d^3x_3\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}=2\int_{x_{13}\leqslant x_{23}}d^3x_3\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3} \tag{E1}$$

is convergent (there is a pole in $x_{13}=0$) and to evaluate it. First, $P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})/x_{23}^3$ is expressed in terms of $x_{12}$, $x_{13}$, and $\mu$ $(=\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{13})$:

$$\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{23}^3}=(x_{12}^2+x_{13}^2-2\mu x_{12}x_{13})^{-3/2}-\frac{3}{2}(1-\mu^2)x_{12}^2(x_{12}^2+x_{13}^2-2\mu x_{12}x_{13})^{-5/2}. \tag{E2}$$

Noting that

$$x_{23}^2=x_{12}^2+x_{13}^2-2\mu x_{12}x_{13}\geqslant x_{13}^2\Leftrightarrow\mu\leqslant\frac{x_{12}}{2x_{13}}\Leftrightarrow -1\leqslant\mu\leqslant\mu_{\max} \tag{E3}$$

with $\mu_{\max}=x_{12}/2x_{13}$ if $x_{13}\geqslant x_{12}/2$,

and $\mu_{\max}=+1$ if $x_{13}\leqslant x_{12}/2$,

$P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})/x_{23}^3$ is integrated over $\mu$:

$$\begin{aligned}\int_{-1}^{\mu_{\max}}d\mu\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{23}^3}&=\frac{1}{x_{13}^3}+\frac{x_{13}-\mu_{\max}x_{12}}{x_{12}x_{13}^2(x_{12}^2+x_{13}^2-2\mu_{\max}x_{12}x_{13})^{1/2}}\\&\quad-\frac{x_{12}(1-\mu_{\max})}{2x_{13}(x_{12}^2+x_{13}^2-2\mu_{\max}x_{12}x_{13})^{3/2}}\\&\quad-\frac{(x_{12}^2+x_{13}^2-2\mu_{\max}x_{12}x_{13})^{1/2}}{x_{12}x_{13}^3},\end{aligned} \tag{E4}$$

$$\text{if}\ x_{13}\leqslant x_{12}/2,\quad\int_{-1}^{+1}d\mu\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}=0. \tag{E5}$$

As the angular integration equals zero for small $x_{13}$, integral (E1) is convergent. Therefore,

$$\int \frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}=2\int_{x_{12}/2\leq x_{13}\leq x_{23}}\frac{d^3x_3}{4\pi}\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}$$
$$=\int_{x_{12}/2}^{\infty}\frac{dx_{13}}{x_{13}}\int_{-1}^{x_{12}/2x_{13}}d\mu\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{23}^3}$$
$$=\int_{x_{12}/2}^{\infty}dx_{13}\left(\frac{1}{x_{13}^4}-\frac{x_{12}}{x_{13}^5}+\frac{x_{12}^3}{8x_{13}^7}\right)$$
$$=0\quad \text{if}\ x_{12}\neq 0. \tag{E6}$$

The following relation can also be verified:

$$2\int_{x_{13}\leq x_{23}}\frac{d^3x_3}{4\pi}g_{2T}^c(x_{13})\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32})}{x_{13}^3x_{23}^3}$$
$$=2\int_{x_{12}/2\leq x_{13}\leq x_{23}}\frac{d^3x_3}{4\pi}g_{2T}^c(x_{13})\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{32}}{x_{13}^3x_{23}^3}$$
$$=\int_{z_{12}/2}^{\infty}dx_{13}g_{2T}^c(x_{13})\left(\frac{1}{x_{13}^4}-\frac{x_{12}}{x_{13}^5}+\frac{x_{12}^3}{8x_{13}^7}\right). \tag{E7}$$

Examine

$$\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}g_{3T}^c(2,3,4)=\left[\frac{3}{2}\frac{(\mathbf{x}_{12}\cdot\mathbf{x}_{34}+\mathbf{x}_{23}\cdot\mathbf{x}_{34})^2}{|\mathbf{x}_{12}+\mathbf{x}_{23}|^5x_{34}^5}-\frac{1}{2|\mathbf{x}_{12}+\mathbf{x}_{23}|^3x_{34}^3}\right]g_{3T}^c(2,3,4) \tag{E8}$$

which appears in the first integral of the right-hand member of Eq. (C6). Let $(x_{23},\theta_3,\varphi_3)$, polar coordinates of vector $\mathbf{x}_{23}$, and $(x_{34},\theta_4,\varphi_4)$, polar coordinates of vector $\mathbf{x}_{34}$, with

$$\theta_3=(\mathbf{x}_{12},\mathbf{x}_{23}),\quad \theta_4=(\mathbf{x}_{23},\mathbf{x}_{34}),$$
$$\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{23}=\cos\ \theta_3=\mu_3,$$
$$\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{34}=\cos\ \theta_4=\mu_4. \tag{E9}$$

It follows that

$$\hat{\mathbf{x}}_{12}\cdot\hat{\mathbf{x}}_{34}=\cos\ \theta_3\cos\ \theta_4-\sin\ \theta_3\sin\ \theta_4\sin\ \varphi_4 \tag{E10}$$

and

$$d^3x_3d^3x_4=d\varphi_3d\mu_3x_{23}^2dx_{23}d\varphi_4d\mu_4x_{34}^2dx_{34}. \tag{E11}$$

Note that $g_{3T}^c(2,3,4)$ is dependent on $x_{23}$, $x_{34}$, and $\mu_4$. Carrying out angular integrations, one gets

$$\int_{-1}^{+1}d\mu_3\int_0^{2\pi}d\varphi_3\int_0^{2\pi}d\varphi_4\frac{P_2(\hat{\mathbf{x}}_{31}\cdot\hat{\mathbf{x}}_{34})}{x_{13}^3x_{34}^3}=\begin{cases}0 & \text{if}\ x_{23}<x_{12},\\ \dfrac{(4\pi)^2P_2(\mu_4)}{2x_{23}^3x_{34}^3} & \text{if}\ x_{23}>x_{12}.\end{cases} \tag{E12}$$

There is a pole in $x_{13}=0$ (i.e., $x_{12}=x_{23}$ and $\mu_3=-1$).

## APPENDIX F: EXPANSION (C9)

In this appendix, $I$ [Eq. (C9)] is expanded with respect to $x_{12}^{-1}$. From Eq. (D10), it can be shown that

$$\int\frac{d^3x_{13}}{x_{13}^3}P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})e^{i\mathbf{q}\cdot\mathbf{x}_{13}}=-\frac{4\pi}{3}P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{14}). \tag{F1}$$

Using twice the inverted form of this relation, to express $(1/x_{13}^3)P_2(\hat{\mathbf{x}}_{13}\cdot\hat{\mathbf{x}}_{14})$ and $(1/x_{14}^3)P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{14})$, $I$ is rewritten in the form

$$I=\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}\left[g_{3T}^c(2,3,4)\lim_{\varepsilon,\varepsilon'\to 0}\int\int\frac{d^3q\ d^3q'}{(6\pi^2)^2}e^{-i(\mathbf{q}+\mathbf{q}')\cdot\mathbf{x}_{12}}e^{-i\mathbf{q}\cdot\mathbf{x}_{23}-i\mathbf{q}\cdot\mathbf{x}_{24}}e^{-\varepsilon q-\varepsilon' q'}P_2(\mathbf{q}\cdot\mathbf{q}')\right]_{\text{large}\ x_{12}}. \tag{F2}$$

$e^{-i\mathbf{q}\cdot\mathbf{x}_{23}}$ and $e^{-i\mathbf{q}\cdot\mathbf{x}_{24}}$ are expanded in terms of Legendre polynomials and of spherical Bessel functions of the first kind [cf. Eq. (D9)]:

$$I=\sum_{l,l'=0}^{\infty}(-i)^{l+l'}(2l+1)(2l'+1)\int\int\frac{d^3x_3d^3x_4}{(4\pi)^2}g_{3T}^c(2,3,4)\lim_{\varepsilon,\varepsilon'\to 0}\int\int\frac{d^3q\ d^3q'}{(6\pi^2)^2}e^{-i(\mathbf{q}+\mathbf{q}')\cdot\mathbf{x}_{12}}e^{-\varepsilon q-\varepsilon' q'}P_2(\mathbf{q}\cdot\mathbf{q}')$$
$$\times j_l(gx_{23})j_{l'}(q'x_{24})P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{23})P_{l'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{24}). \tag{F3}$$

The vectors $\mathbf{q}'$ and $\mathbf{x}_{24}$ are expressed in polar coordinates as follows:

$$\mathbf{x}_{23}=\begin{pmatrix}0\\0\\x_{23}\end{pmatrix},\quad \mathbf{q}'=\begin{pmatrix}0\\q'\sin\ \theta'\\q'\cos\ \theta'\end{pmatrix},\quad \mathbf{x}_{24}=\begin{pmatrix}x_{24}\sin\ \theta_4\cos\ \varphi_4\\x_{24}\sin\ \theta_4\sin\ \varphi_4\\x_{24}\cos\ \theta_4\end{pmatrix}. \tag{F4}$$

Equation (D8) is used to write

$$\frac{1}{2\pi}\int_0^{2\pi}d\varphi_4P_{l'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{24})=P_{l'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{23})P_{l'}(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24}). \tag{F5}$$

We consider now $P_{l'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{23})$. Involving the vector $\mathbf{q}$, we process as for $P_{l'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{24})$. The vectors are expressed with new polar coordinates:

$$\mathbf{q}=\begin{pmatrix}0\\0\\q\end{pmatrix},\quad \mathbf{q}'=\begin{pmatrix}0\\q'\sin\theta\\q'\cos\theta\end{pmatrix},\quad \mathbf{x}_{23}=\begin{pmatrix}x_{23}\sin\theta_3\cos\varphi_3\\x_{23}\sin\theta_3\sin\varphi_3\\x_{23}\cos\theta_3\end{pmatrix}. \tag{F6}$$

Let

$$\mu_3=\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{23},$$
$$\mu_4=\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24}. \tag{F7}$$

Equation (F3) becomes

$$I=\sum_{l,l'=0}^{\infty}(-i)^{l+l'}(2l+1)(2l'+1)\,\frac{1}{4}\int_{-1}^{+1}d\mu_3\,P_l(\mu_3)P_{l'}(\mu_3)\int_{-1}^{+1}d\mu_4\,P_{l'}(\mu_4)\int_0^{\infty}dx_{23}x_{23}^2\int_0^{\infty}dx_{24}x_{24}^2g_{3T}^3(2,3,4)$$
$$\times\lim_{\varepsilon,\varepsilon'\to0}\int\int\frac{d^3q\;d^3q'}{(6\pi^2)^2}\,e^{-i(\mathbf{q}+\mathbf{q}')\cdot\mathbf{x}_{12}}e^{-\varepsilon q-\varepsilon'q'}P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')P_{l'}(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')j_l(qx_{23})j_{l'}(q'x_{24}). \tag{F8}$$

Because of the orthogonality of the Legendre polynomials, only the terms for which $l=l'$ contribute to the sum over $l$ and $l'$. Therefore,

$$I=\sum_{l=0}^{\infty}(-)^l(2l+1)\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2}\,P_l(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24})g_{3T}^c(2,3,4)$$
$$\times\lim_{\varepsilon,\varepsilon'\to0}\int\int\frac{d^3q\;d^3q'}{(6\pi^2)^2}\,e^{-i(\mathbf{q}+\mathbf{q}')\cdot\mathbf{x}_{12}}e^{-\varepsilon q-\varepsilon'q'}P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')j_l(qx_{23})j_l(q'x_{24}). \tag{F9}$$

Here $e^{-i(\mathbf{q}+\mathbf{q}')\cdot\mathbf{x}_{12}}$ is also expanded:

$$I=\sum_{l=0}^{\infty}(-1)^l(2l+1)\sum_{k,k'=0}^{\infty}(-i)^{k+k'}(2k+1)(2k'+1)\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2}\,P_l(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24})g_{3T}^c(2,3,4)$$
$$\times\lim_{\varepsilon,\varepsilon'\to0}\int\frac{d^3q\;d^3q'}{(6\pi^2)^2}\,e^{-\varepsilon q-\varepsilon'q'}P_k(\hat{\mathbf{q}}\cdot\hat{\mathbf{x}}_{12})P_{k'}(\hat{\mathbf{q}}'\cdot\hat{\mathbf{x}}_{12})j_k(qx_{12})j_{k'}(q'x_{12})$$
$$\times P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')j_l(qx_{23})j_l(q'x_{24}). \tag{F10}$$

As previously, it can be proved that

$$I=\sum_{l,k=0}^{\infty}(-1)^{l+k}(2l+1)(2k+1)\int\int\frac{d^3x_3\,d^3x_4}{(4\pi)^2}\,P_l(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24})g_{3T}^c(2,3,4)\lim_{\varepsilon,\varepsilon'\to0}\int\int\frac{d^3q\;d^3q'}{(6\pi^2)^2}\,e^{-\varepsilon q-\varepsilon'q'}P_2(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')$$
$$\times P_k(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{q}}')j_k(qx_{12})j_k(q'x_{12})j_l(qx_{23})j_l(q'x_{24}). \tag{F11}$$

With the help of the known relation

$$(2l+1)\mu P_l(\mu)=(l+1)P_{l+1}(\mu)+lP_{l-1}(\mu)\qquad\text{if }\;l\neq0, \tag{F12}$$

one gets

$$(2l+1)(2k+1)\int_{+1}^{+1}d\mu\;P_2(\mu)P_l(\mu)P_k(\mu)=(2l+1)(2k+1)\int_{+1}^{+1}d\mu\left(\frac{3\mu^2}{2}-\frac{1}{2}\right)P_l(\mu)P_k(\mu)$$
$$=\frac{3}{2}\int_{+1}^{+1}d\mu[(l+1)P_{l+1}(\mu)+lP_{l-1}(\mu)][(k+1)P_{k+1}(\mu)+kP_{k-1}(\mu)]$$
$$-\frac{1}{2}(2l+1)(2k+1)\int_{+1}^{+1}d\mu\;P_l(\mu)P_k(\mu)\quad\text{if }\;l,k\neq0. \tag{F13}$$

Then it is shown that

$$(2l+1)(2k+1)\int_{+1}^{+1} d\mu\ P_2(\mu)P_l(\mu)P_k(\mu)=3\,\frac{l(l-1)}{2l-1}\ \delta_{k,(l-2)}+2\,\frac{l(l+1)(2l+1)}{(2l-1)(2l+3)}\ \delta_{k,l}+3\,\frac{(l+1)(l+2)}{2l+3}\ \delta_{k,(l+2)}\,. \tag{F14}$$

Thus, in the right-hand member of Eq. (F11), the sum over $l$ and $k$ reduces to the cases where $k=l-2$, $l$, or $l+2$. Equation (F11) is then rewritten as follows:

$$I=\frac{2}{3\pi^2 x_{12}^6}\sum_{l,k=0}^{\infty}\left[\frac{l(l-1)}{2l-1}\ \delta_{k,(l-2)}+\frac{2l(l+1)(2l+1)}{3(2l-1)(2l+3)}\ \delta_{k,l}+\frac{(l+1)(l+2)}{2l+3}\ \delta_{k,(l+2)}\right]$$
$$\times\int\int\ \frac{d^3x_3\,d^3x_4}{(4\pi)^2}\ P_l(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24})g_{3T}^c(2,3,4)K_{k,l}\left(\frac{x_{23}}{x_{12}}\right)K_{k,l}\left(\frac{x_{24}}{x_{12}}\right), \tag{F15}$$

where $K_{k,l}(x/x_{12})$ is a function defined by

$$K_{k,l}\left(\frac{x}{x_{12}}\right)=\lim_{\varepsilon\to 0}\int\ \frac{d^3q}{4\pi}\ e^{-\varepsilon q}j_k(q)j_l\left(q\ \frac{x}{x_{12}}\right). \tag{F16}$$

As $x_{12}$ is large enough, $j_l(qx/x_{12})$ can be expanded in terms of increasing powers of $x/x_{12}$. This brings in the following expansion for the function $K_{k,l}$:

$$K_{k,l}\left(\frac{x}{x_{12}}\right)=\left(\frac{x}{x_{12}}\right)^l\sum_{m=0}^{\infty}(-1)^m\left(\frac{x}{2x_{12}}\right)^{2m}\ \frac{I_{(2+l+2m),k}}{m!(2l+2m+1)!!}, \tag{F17}$$

where the $I_{n,k}$'s correspond to the integrals

$$I_{n,k}=\lim_{\varepsilon\to 0}\int_0^{\infty}du\ e^{-\varepsilon u}u^n j_k(u), \tag{F18}$$

which have here to be evaluated in the only case where $n\geqslant k$, because $k$ is restricted to $l-2$, $l$, or $l+2$. If $k>0$, integrations by parts provide

$$I_{n,k}=(n+k-1)I_{n-1,k-1}=(n+k-1)(n+k-3)\cdots(n-k+1)I_{n-k,0}\quad(\text{for}\ \ n>k)$$
$$\equiv I_{n,n}=(2n-1)!!I_{0,0}\quad(\text{for}\ \ n=k). \tag{F19}$$

One gets

$$\underset{(p>0)}{I_{p,0}}=\lim_{\varepsilon\to 0}\int_0^{\infty}du\ e^{-\varepsilon u}u^{p-1}\sin u=\lim_{\varepsilon\to 0}\ \mathrm{Im}\int_0^{\infty}du\ e^{-u(\varepsilon-i)}u^{p-1}=\mathrm{Im}\left[\frac{(p-1)!}{(-i)^p}\right]=\begin{cases}0\quad(\text{if}\ \ p\ \ \text{is even})\\(-1)^{(p-1)/2}(p-1)!\quad(\text{if}\ \ p\ \ \text{is odd}),\end{cases}$$

$$I_{0,0}=\lim_{\varepsilon\to 0}\int_0^{\infty}du\ e^{-\varepsilon u}\ \frac{\sin u}{u}=\frac{\pi}{2}. \tag{F20}$$

Note that, in the present calculations, one is just concerned with even $p=n-k$ numbers. The peculiar feature that $I_{p,0}$ vanishes in that case, except for $p=0$, considerably simplifies the evaluation of expansion (F17), as it is restricted to its term $m=0$, while $k$ is also restricted to the single value $l+2$, for a given $l$ number. This allows us to express Eq. (F15) in the simple form

$$I=\frac{1}{6x_{12}^6}\sum_{l=0}^{\infty}(l+1)(l+2)(2l+3)\int\int\ \frac{d^3x_3\,d^3x_4}{(4\pi)^2}\ g_{3T}^c(2,3,4)P_l(\hat{\mathbf{x}}_{23}\cdot\hat{\mathbf{x}}_{24})\ \frac{x_{23}^l x_{24}^l}{x_{12}^{2l}}\quad\text{for large}\ \ x_{12}, \tag{F21}$$

which brings in an expansion in terms of the Legendre polynomial $P_l$. Finally, making use of OCP sum rules (42) and (45) provides Eq. (C9):

$$I=-\frac{2}{3x_{12}^6}\ S_0-\frac{1}{18}\sum_{l=1}^{\infty}\ \frac{(l+1)(l+2)(2l+3)}{x_{12}^{2l+6}}\ S_{2l}\quad\text{for large}\ \ x_{12}. \tag{F22}$$

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