A reformulation of simple liquids theory—Renormalization by one-, two-, and three-particle densities

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We reformulate the theory of simple liquids in a field theoretical way by taking into account the triplet potential w_{ijk} , in addition to the external potential u_i and the pair potential v_{ii} . The innovation here is the inversion method and the on-shell expansion which are the building blocks of a novel use of Legendre transformation developed in field theory. By the inversion method, we renormalize the theory in terms of one-, two-, and three-particle densities, and present a diagrammatic representation for a thermodynamical functional, which is the entropy except for a trivial constant, in terms of renormalized variables. In other words, we present an expression for the entropy in terms of only one-, two-, and three-particle densities: the *n*-particle density where $n \ge 4$ does not appear in the expression. The on-shell condition, which is a starting point of the on-shell expansion, of the thermodynamical functional thus obtained (the entropy) leads to a set of three self-consistent equations for one-, two-, and three-particle densities. Through one of the selfconsistent equations, we can systematically improve the Kirkwood's superposition approximation for the three-particle density. The on-shell conditions for other thermodynamical functionals, also obtained in this article, are found to be extentions of various well-known equations in the theory of simple liquids. The formulation presented here is complementary to the conventional re-summation techniques for renormalization of diagrams. In the present formulation, we do not have to care about the topological structure of diagrams, often characterized by the irreducibility of diagrams. Instead, by a perturbative calculation, we can automatically single out the diagrams with the topological structure predicted by the re-summation techniques. © 1998 American Institute of Physics. [S0022-2488(98)01604-1]

I. INTRODUCTION

In the theory of simple liquids, renormalization of the theory in terms of one-, and twoparticle densities is essential. (The *n*-particle density here is sometimes called the correlation function of the *n*-th order.) To perform such renormalization, re-summation techniques of diagrams had been extensively developed.¹⁻⁴ Such techniques, though elegant and attractive, are sometimes complex and difficult to understand. In such techniques, the topological structure of the diagrams for the renormalized theory is determined by using graphical terms such as irreducibility of diagrams. If one wants to obtain higher order diagrams, we have to single out, by hand, diagrams which have the topological structure thus determined, after considering all possible diagrams of that order. At a higher order, this task can be laborious.

In the present reformulation, however, we can obtain the renormalized diagrams automatically order by order through simple (but sometimes tedious) calculations. We do not have to care about the topological structure at all; the diagrams with the demanded topology are automatically singled out. In this sense, this reformulation is complementary to the conventional re-summation techniques.

The innovation here is a novel use of the Legendre transformation which has been developed in field theory and has been applied to quantum systems.⁵ In this article we present the first application to a classical system. This methodology consists of the inversion method and the one-shell expansion, which shall be reviewed briefly below. However, in short, the former is a method by which we can obtain a non-perturbative result through a sort of perturbative calculation, and the latter is a formalism in which we can systematically extract all physical information

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concerning a physical variable of interest, starting from an effective action. (The effective action is a kind of thermodynamical functional in a classical system as we see below.)

The systematic expansion of the entropy of simple liquids theory in terms of the correlation function has received considerable attention. Such an expansion for the canonical ensemble was studied in Ref. 6, which followed by the works for the grand canonical ensemble.^{7,8} Recently, these expansion schemes have been revisited^{9–12} and numerically examined^{13,14} by several researchers. In all these works, the entropy is expanded in terms of one-particle density and the *n*-th order term contains the correlation function up to the *n*-th order: the entropy is expressed by the *n*-particle densities where $n=0,1,...,\infty$.

However, as shown below, the entropy functional, which is the effective action except for a trivial term, can be described by the correlation functions of the order *up to the n-th* if we neglect higher-body potential than *n*-body. Indeed, the entropy given in Ref. 1 is described only by one-and two-particle densities since, as in conventional simple liquids theory, the triplet and higher-body potential are neglected there.

In this article we present an expression for the entropy as functional of one-, two-, and three-particle densities of the grand canonical ensemble. The expansion does not include correlation functions of the order higher than three and thus the theory is renormalized in terms of one, two-, and three-particle densities.

To renormalize the theory in terms of the three-particle density, in the present paper, we first explicitly include a triplet potential into a theory, and then, make a Legendre transformation from the triplet potential to the three particle density. The triplet potential can be set to zero after the renormalization if we like to go back to the (conventional) liquids theory without the triplet potential. Note here that, by first introducing the triplet potential and then by setting the potential at the end, we can extract new information, which can not be obtained if we neglect the potential from the start.

Extention of the liquid theory to include triplet potentials is not new and had been studied, though the above viewpoint for the triplet potential as a tool for the Legendre transformation might have not. For example, for a weak triplet potential, they included the effects of the triplet potential by modifying the pair potential with the aid of diagrams.^{15–17} Other important references can be found in Refs. 12, 18.

Furthermore, inclusion of the general *m*-body potential (m = 1, 2, 3, 4, ...) into the liquid theory is performed in Ref. 19 in a simple and clear way. There, various diagrammatic rules are neatly given and are renormalized up to the two-particle densities. In the present work, we further renormalize the theory by the three-particle densities though our case is limited to the special case where there are no more-than-three-body potentials. (See Sec. VII.)

In the present formalism, the stationary conditions for the entropy or the effective action come out as identities of the Legendre transformation. The set of conditions can be seen as a set of self-consistent equations to be solved for the variables of interest.

In this article, we obtain three kinds of the effective actions, which are closely related to the entropy. As stationary conditions for these actions, we get three sets of self-consistent equations, some of which can be seen as a generalization of well-known equations in simple liquids theory.

Especially, one of the self-consistent equations leads to successive improvement of the Kirkwood's superposition approximation⁴ for the three-particle densities. The zero-th order approximation of this theory coincides with the previously known approximation (for example, see Refs. 20, 12), which reduces to the Kirkwood's approximation for the system without the triplet potential. The first order approximation of ours can be seen as an improvement over the well-known approximations.^{21–24}

Let us review the inversion method and the on-shell expansion briefly. Consider a physical variable of interest ϕ in a system described by a Hamiltonian H. In a classical system at an equilibrium, a physical variable is a thermal expectation value of some quantity $\hat{\phi}$. (In a quantum system a physical variable is a quantum expectation value of an operator $\hat{\phi}$.) In the inversion method, we modify the Hamiltonian H to $H+J\hat{\phi}$ where J is an artificial external source which should be set to zero at the end of calculation. However, if the original Hamiltonian H already has a term proportional to $\hat{\phi}$, say $j\hat{\phi}$, we merely replace j with an artificial source J and set it back

to *j* at the end of calculation. After this modification of the Hamiltonian, we calculate ϕ by a perturbative expansion in terms of the expansion parameter, say *g*, in the presence of the artificial source *J*. We have an expansion called *the original series* of the form,

$$\phi = \phi(J) = \phi^{(0)}(J) + \phi^{(1)}(J) + \phi^{(2)}(J) + \cdots, \qquad (1.1)$$

where $\phi^{(i)}$ is the *i*-th order in g when we regard J as independent of g. Now we invert Eq. (1.1) in favor of J to get *the inversion series*,

$$J = J(\phi) = J^{(0)}(\phi) + J^{(1)}(\phi) + J^{(2)}(\phi) + \cdots, \qquad (1.2)$$

where $J^{(i)}(\phi)$ is the *i*-th order in g when we regard ϕ as independent of g. In order to obtain an explicit form of $J^{(i)}(\phi)$, we insert Eq. (1.2) into Eq. (1.1):

$$\phi = \phi^{(0)}(J^{(0)} + J^{(1)} + J^{(2)} + \dots) + \phi^{(1)}(J^{(0)} + J^{(1)} + \dots) + \phi^{(2)}(J^{(0)} + \dots) + \dots, \quad (1.3)$$

or

$$\phi = \phi^{(0)}(J^{(0)}) + \phi^{(0)}(J^{(0)})J^{(1)} + \phi^{(1)}(J^{(0)}) + \phi^{(0)}(J^{(0)})J^{(2)} + \frac{1}{2}\phi^{(0)}(J^{(0)}) \cdot (J^{(1)})^{2} + \phi^{(1)}(J^{(0)})J^{(1)} + \phi^{(2)}(J^{(0)}) + \cdots$$
(1.4)

The trick of the inversion method is to regard ϕ in the left-hand side of this equation as independent of g. Then we get a set of equations called *inversion formulae*:

$$\phi = \phi^{(0)}(J^{(0)}), \tag{1.5}$$

$$\phi^{(0)\prime}(J^{(0)})J^{(1)} + \phi^{(1)}(J^{(0)}) = 0, \qquad (1.6)$$

$$\phi^{(0)\prime}(J^{(0)})J^{(2)} + \frac{1}{2}\phi^{(0)\prime\prime}(J^{(0)}) \cdot (J^{(1)})^2 + \phi^{(1)\prime}(J^{(0)})J^{(1)} + \phi^{(2)}(J^{(0)}) = 0, \qquad (1.7)$$

$$\phi^{(0)\prime}(J^{(0)})J^{(3)} + \phi^{(0)\prime\prime}(J^{(0)})J^{(1)}J^{(2)} + \frac{1}{3!}\phi^{(0)\prime\prime\prime}(J^{(0)}) \cdot (J^{(1)})^3 + \phi^{(1)\prime}(J^{(0)})J^{(2)} + \frac{1}{2}\phi^{(1)\prime\prime}(J^{(0)}) \cdot (J^{(1)})^2 + \phi^{(2)\prime}(J^{(0)})J^{(1)} + \phi^{(3)}(J^{(0)}) = 0, \qquad (1.8)$$

and so on. In this way we can obtain $J^{(i)}(\phi)$ up to the desired order. Note here that all the derivatives of $\phi^{(i)}$ and $\phi^{(i)}$ itself in the above formulae are evaluated at $J=J^{(0)}$, which is determined by the lowest inversion formula (1.5). Finally, we set the artificially introduced source J back to the original value, say j, to go back to the original theory:

$$J^{(0)}(\phi) + J^{(1)}(\phi) + \dots = j.$$
(1.9)

If we truncate the original series (1.1) at the *i*-th order, we obtain Eq. (1.9) truncated at the *i*-th order. However, these two truncated equations are not equivalent and the latter, which can be regarded as an equation determining ϕ , is a more improved equation for ϕ than the former.

The trick of regarding ϕ as independent of g corresponds to making the Legendre transformation; ϕ and g are dealt with as mutually independent in the Legendre transformation. To further clarify this point, we introduce a generating function W which satisfies the relation

$$\phi(J) = \frac{dW(J)}{dJ}.\tag{1.10}$$

In the case of an equilibrium system W is nothing but the thermodynamical potential $[W=\Omega = -(1/\beta)\ln\Xi$: see Eq. (2.8)]. Now we introduce *the effective action* Γ by the Legendre transfor-

mation: $\Gamma = W(J,g) - J\phi(J,g)$ where $\phi(J,g) = \partial W(J,g) / \partial J$. Here, we have emphasized the *g* dependence. The variation of Γ , *if we assume that J and g are independent variables*, is expressed as

$$d\Gamma = \frac{\partial W(J,g)}{\partial J} \, dJ + \frac{\partial W(J,g)}{\partial g} \, dg - dJ \phi(J,g) - Jd \phi(J,g) = \frac{\partial W(J,g)}{\partial g} \, dg - Jd \phi(J,g).$$
(1.11)

Hence, as is well known, we see that the quantity Γ can be regarded as a function of two independent variables ϕ and $g - \Gamma = \Gamma(\phi, g)$. Equation (1.11) seems trivial but is essential in the present discussion. What is implied in Eq. (1.11) is as follows: if we solve (or invert) the relation $\phi(J,g) = \partial W(J,g)/\partial J$ in favor of *J*, assuming that two quantities ϕ and *g* are mutually independent, to obtain $J = J(\phi, g)$ and then insert this expression of *J* into all *J* appearing in $W(J,g) - J\phi$, this quantity $W(J,g) - J\phi$ is automatically written by only two independent variables ϕ and *g*. In other words, this inversion process of the Legendre transformation is carried out regarding ϕ as independent of *g*. This is the justification of the trick of the inversion method. Note that once the inversion or the Legendre transformation is performed and after the sources are set to an appropriate value, *j* for example, the resultant ϕ depends on *g*. In many quantum cases such as Quantum Electrodynamics (QED),²⁵ Superconductivity,²⁶ Hubbard model,²⁷ and Superfluidity,⁵ it is known that this trick of the inversion method reproduces the mean field approximation at the first order (in which we truncate the series at *i*=1). In this sense, the inversion method offers a way to systematically advance the mean field approximation.

Reciprocal to Eq. (1.10), there is an identity,

$$J = -\frac{d\Gamma(\phi)}{d\phi}.$$
(1.12)

This equation is equivalent to the inversion series (1.2) and, at an appropriate value of J, say j, corresponds to Eq. (1.9). In other words, Eq. (1.12) at J=j is an equation which determines ϕ . This equation corresponds to the on-shell condition in field theory. The on-shell expansion is an expansion of the right-hand side of the on-shell condition $j = -d\Gamma(\phi)/d\phi$ at the point $\phi = \phi_0 + \Delta \phi$ around the solution of the on-shell condition $\phi = \phi_0$ [i.e., $j = -d\Gamma(\phi_0)/d\phi_0$]. Through the on-shell expansion, we obtain a set of equations containing all the physical information related to ϕ such as excitations and scatterings. However, since we concentrate on equilibrium properties of liquids in this article, the higher order on-shell equations shall not be examined. Such an important issue is reserved for a future work. In this sense, a real application of the on-shell expansion to a classical system is relegated to a separate work, which shall deal with dynamical properties of simple liquids.

In the above we consider a simple case where ϕ is scalar and position independent. However, the generalization to a case where ϕ is a tensor and position dependent is straightforward [see, for example, Eq. (4.10) below].

Organization of this article is as follows. In Section II, we reformulate the simple liquids theory in a field theoretical way and successively introduce three kinds of effective actions $(\Gamma_1, \Gamma_2, \Gamma_3)$ through the first, the second, and the third Legendre transformation. We then give the on-shell condition for each effective action. It is stressed here that the effective action Γ_3 is the entropy except for a trivial term and it can be written as a functional of the correlation functions of *up to the third order*. In Section III, we present a diagrammatic expression for the thermodynamic potential with explicitly including the triplet potential w_{ijk} . The introduction of *pseudo* 2-bonds is essential for the presentation of the diagrams with the triplet potential. Sections IV to VI deal with renormalization, we obtain a diagrammatic expression for Γ_1 , Γ_2 , or Γ_3 (the entropy) and show that the on-shell condition for each Γ_i (i=1,2,3) can be considered as some extension of various well-known equations in simple liquids theory. In Section VI, we obtain a systematic way of improving the Kirkwood's superposition approximation for the three-particle densities.

II. A REFORMULATION OF THE SIMPLE LIQUIDS THEORY

Consider an N-particle classical system subject to the potential $u(\mathbf{r})$, the pair potential $v(\mathbf{r},\mathbf{r}')$, and the triplet potential $w(\mathbf{r},\mathbf{r}',\mathbf{r}'')$,²⁸

$$\mathcal{H}_{N} \equiv \mathcal{H}_{N}(\mathbf{p}_{1},\ldots,\mathbf{p}_{N},\mathbf{q}_{1},\ldots,\mathbf{q}_{N}) = \frac{1}{2m} \sum_{i=1}^{N} |\mathbf{p}_{i}|^{2} + V_{N}, \qquad (2.1)$$

$$V_{N} \equiv V_{N}(\mathbf{q}_{1},...,\mathbf{q}_{N}) = \sum_{i=1}^{N} u(\mathbf{q}_{i}) + \sum_{i< j}^{N} v(\mathbf{q}_{i},\mathbf{q}_{j}) + \sum_{i< j< k}^{N} w(\mathbf{q}_{i},\mathbf{q}_{j},\mathbf{q}_{k}).$$
(2.2)

Here the pair potential $v(\mathbf{r}_i, \mathbf{r}_j)$ and $w(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ are symmetric under the interchange of position variables $[v(\mathbf{r}_1, \mathbf{r}_2) = v(\mathbf{r}_2, \mathbf{r}_1)]$ etc.]. By introducing the *n*-particle density for an *N*-particle system (n = 1, 2, 3) by

$$\rho_N^{(1)}(\mathbf{r}) = \sum_{i=1}^N \,\delta(\mathbf{r} - \mathbf{q}_i),\tag{2.3}$$

$$\rho_N^{(2)}(\mathbf{r},\mathbf{r}') = \sum_{i \neq j}^N \,\delta(\mathbf{r} - \mathbf{q}_i)\,\delta(\mathbf{r}' - \mathbf{q}_j) = \rho_N^{(1)}(\mathbf{r})\,\rho_N^{(1)}(\mathbf{r}') - \delta(\mathbf{r} - \mathbf{r}')\,\rho_N^{(1)}(\mathbf{r}),\tag{2.4}$$

$$\rho_N^{(3)}(\mathbf{r},\mathbf{r}',\mathbf{r}'') = \sum_{i\neq j\neq k}^N \,\delta(\mathbf{r}-\mathbf{q}_i)\,\delta(\mathbf{r}'-\mathbf{q}_j)\,\delta(\mathbf{r}''-\mathbf{q}_k),\tag{2.5}$$

the grand potential $V_N - \mu N$ is cast into the form

$$V_{N} - \mu N = \int d\mathbf{r}_{1} \widetilde{u}(\mathbf{r}_{1}) \rho_{N}^{(1)}(\mathbf{r}_{1}) + \frac{1}{2} \int d\mathbf{r}_{1} \int d\mathbf{r}_{2} v(\mathbf{r}_{1}, \mathbf{r}_{2}) \rho_{N}^{(2)}(\mathbf{r}_{1}, \mathbf{r}_{2}) + \frac{1}{3!} \int d\mathbf{r}_{1} \int d\mathbf{r}_{2} \int d\mathbf{r}_{3} w(\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3}) \rho_{N}^{(3)}(\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3}), \qquad (2.6)$$

where

$$\widetilde{u}(\mathbf{r}) = -\mu + u(\mathbf{r}). \tag{2.7}$$

This expression for the potential plays a key role in our reformulation of the simple liquids theory in a field theoretical way. From this expression, we can identify $(J, \hat{\phi})$ of the inversion method with $(\tilde{u}, \rho_N^{(1)})$, $(v, \rho_N^{(2)})$, or $(w, \rho_N^{(3)})$.

The grand partition function Ξ and the thermodynamic potential,

$$\Omega = -\frac{1}{\beta} \ln \Xi, \qquad (2.8)$$

is defined by

$$e^{-\beta\Omega} = \Xi = \sum_{N=0}^{\infty} \frac{1}{N!} \frac{1}{h^{3N}} \int d\mathbf{p}^N \int d\mathbf{q}^N e^{-\beta(\mathscr{H}_N - \mu N)}, \qquad (2.9)$$

where $\int d\mathbf{p}^N$ and $\int d\mathbf{q}^N$ implies the integrations over $\mathbf{p}_1, \mathbf{p}_2, \dots, \mathbf{p}_N$ and $\mathbf{q}_1, \mathbf{q}_2, \dots, \mathbf{q}_N$, respectively.

Regarding quantities Ω and Ξ as functionals of the potentials $\tilde{u}(\mathbf{r})$, $v(\mathbf{r},\mathbf{r}')$, and $w(\mathbf{r},\mathbf{r}',\mathbf{r}')$, we have the *n*-particle density (*n*=1,2,3) by differentiating Ω with respect to \tilde{u} , v, or w:²⁹

$$\rho^{(1)}(\mathbf{r}) \equiv \frac{d\Omega}{\delta \widetilde{u}(\mathbf{r})} = \langle \rho_N^{(1)}(\mathbf{r}) \rangle, \qquad (2.10)$$

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$$\rho^{(2)}(\mathbf{r},\mathbf{r}') \equiv \frac{\delta\Omega}{\delta \frac{1}{2} v(\mathbf{r},\mathbf{r}')} = \langle \rho_N^{(2)}(\mathbf{r},\mathbf{r}') \rangle, \qquad (2.11)$$

$$\rho^{(3)}(\mathbf{r},\mathbf{r}',\mathbf{r}'') \equiv \frac{\delta\Omega}{\delta \frac{1}{3!} w(\mathbf{r},\mathbf{r}',\mathbf{r}'')} = \langle \rho_N^{(3)}(\mathbf{r},\mathbf{r}',\mathbf{r}'') \rangle.$$
(2.12)

Here, the expectation implies³⁰

$$\langle \mathcal{O}_N \rangle = \frac{1}{\Xi} \sum_{N=0}^{\infty} \frac{1}{N!} \frac{1}{h^{3N}} \int d\mathbf{p}^N \int d\mathbf{q}^N \mathcal{O}_N \ e^{-\beta(\mathcal{H}_N - \mu N)}, \tag{2.13}$$

and we have used the relations

$$\frac{\delta \widetilde{u}(\mathbf{r})}{\delta \widetilde{u}(\mathbf{r}')} = \delta(\mathbf{r} - \mathbf{r}'), \qquad (2.14)$$

$$\frac{\delta v(\mathbf{r}_{1},\mathbf{r}_{2})}{\delta v(\mathbf{r}_{1'},\mathbf{r}_{2'})} = \frac{1}{2!} \left[\delta(\mathbf{r}_{1}-\mathbf{r}_{1'}) \,\delta(\mathbf{r}_{2}-\mathbf{r}_{2'}) + \delta(\mathbf{r}_{1}-\mathbf{r}_{2'}) \,\delta(\mathbf{r}_{2}-\mathbf{r}_{1'}) \right], \tag{2.15}$$

$$\frac{\delta w(\mathbf{r}_{1},\mathbf{r}_{2},\mathbf{r}_{3})}{\delta w(\mathbf{r}_{1'},\mathbf{r}_{2'},\mathbf{r}_{3'})} = \frac{1}{3!} \left[\delta(\mathbf{r}_{1} - \mathbf{r}_{1'}) \,\delta(\mathbf{r}_{2} - \mathbf{r}_{2'}) \,\delta(\mathbf{r}_{3} - \mathbf{r}_{3'}) + (3! - 1) \,\text{terms} \right].$$
(2.16)

In order to rewrite the theory in terms of $\rho^{(1)}(\mathbf{r})$, $\rho^{(2)}(\mathbf{r},\mathbf{r}')$, and $\rho^{(3)}(\mathbf{r},\mathbf{r}',\mathbf{r}'')$ instead of $\tilde{u}(\mathbf{r})$, $v(\mathbf{r},\mathbf{r}')$, and $w(\mathbf{r},\mathbf{r}',\mathbf{r}'')$, the Legendre transformations are successively performed. The first, the second, and the third Legendre transformations are, respectively, defined by

$$\Gamma_1[\rho^{(1)}, v] \equiv \Omega[\tilde{u}, v, w] - \int d\mathbf{r}_1 \tilde{u}(\mathbf{r}_1) \rho^{(1)}(\mathbf{r}_1), \qquad (2.17)$$

$$\Gamma_{2}[\rho^{(1)},\rho^{(2)}] \equiv \Gamma_{1}[\rho^{(1)},v] - \frac{1}{2} \int d\mathbf{r}_{1} \int d\mathbf{r}_{2}v(\mathbf{r}_{1},\mathbf{r}_{2})\rho^{(2)}(\mathbf{r}_{1},\mathbf{r}_{2}), \qquad (2.18)$$

$$\Gamma_{3}[\rho^{(1)},\rho^{(2)},\rho^{(3)}] \equiv \Gamma_{2}[\rho^{(1)},\rho^{(2)},w] - \frac{1}{3!} \int d\mathbf{r}_{1} \int d\mathbf{r}_{2} \int d\mathbf{r}_{3}w(\mathbf{r}_{1},\mathbf{r}_{2},\mathbf{r}_{3})\rho^{(3)}(\mathbf{r}_{1},\mathbf{r}_{2},\mathbf{r}_{3}).$$
(2.19)

In these equations, $(\tilde{u}, \rho^{(1)})$, $(v, \rho^{(2)})$, or $(w, \rho^{(3)})$ can be regarded as (J, ϕ) of the inversion method. In the present case, the original Hamiltonian already has a term proportional to ϕ : we should set $J = -\mu + u$, v, or w (instead of J = 0) at the end. (In the conventional simple liquids theory where the triplet potential is neglected, we have to set J = w = 0 at the end.) By using the relation

$$\delta\Omega[\tilde{u}, v, w] = \int d\mathbf{r}_1 \frac{\delta\Omega}{\delta\tilde{u}(\mathbf{r}_1)} \,\delta\tilde{u}(\mathbf{r}_1) + \int d\mathbf{r}_1 \int d\mathbf{r}_2 \frac{\delta\Omega}{\delta v(\mathbf{r}_1, \mathbf{r}_2)} \,\delta v(\mathbf{r}_1, \mathbf{r}_2) \\ + \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \,\frac{\delta\Omega}{\delta w(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)} \,\delta w(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3), \qquad (2.20)$$

we can explicitly confirm that Γ_1 , Γ_2 , and Γ_3 are functionals of $(\rho^{(1)}, v, w)$, $(\rho^{(1)}, \rho^{(2)}, w)$, and $(\rho^{(1)}, \rho^{(2)}, \rho^{(3)})$, respectively, with the identities

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$$\frac{\delta\Gamma_{1}[\rho^{(1)}, v, w]}{\delta\rho_{1}^{(1)}} = -\tilde{u}_{1}, \quad \frac{\delta\Gamma_{1}[\rho^{(1)}, v, w]}{\delta\frac{1}{2}v_{12}} = \rho_{12}^{(2)}, \quad \frac{\delta\Gamma_{1}[\rho^{(1)}, v, w]}{\delta\frac{1}{3!}w_{123}} = \rho_{123}^{(3)}, \quad (2.21)$$

$$\frac{\delta\Gamma_{2}[\rho^{(1)},\rho^{(2)},w]}{\delta\rho_{1}^{(1)}} = -\tilde{u}_{1}, \quad \frac{\delta\Gamma_{2}[\rho^{(1)},\rho^{(2)},w]}{\delta\frac{1}{2}\rho_{12}^{(2)}} = -v_{12}, \quad \frac{\delta\Gamma_{2}[\rho^{(1)},\rho^{(2)},w]}{\delta\frac{1}{3!}w_{123}} = \rho_{123}^{(3)}, \quad (2.22)$$

$$\frac{\delta\Gamma_{3}[\rho^{(1)},\rho^{(2)},\rho^{(3)}]}{\delta\rho_{1}^{(1)}} = -\tilde{u}_{1}, \quad \frac{\delta\Gamma_{3}[\rho^{(1)},\rho^{(2)},\rho^{(3)}]}{\delta\frac{1}{2}\rho_{12}^{(2)}} = -v_{12}, \quad \frac{\delta\Gamma_{3}[\rho^{(1)},\rho^{(2)},\rho^{(3)}]}{\delta\frac{1}{3!}\rho_{123}^{(3)}} = -w_{123}.$$
(2.23)

Here, we have used simple and self-evident notations. These can be seen as the on-shell conditions for the first, second, and third Legendre transformations, respectively, and shall be shown to correspond to familiar equations of simple liquids theory.

The effective actions introduced above are some thermodynamic functionals and closely related to the entropy. We notice that the entropy functional is given by

$$S/k = -\beta \Gamma_3 + 3\langle N \rangle/2, \qquad (2.24)$$

where k is the Boltzmann constant, because the definition of the entropy S is given by

$$\Omega = \langle E \rangle - TS - \mu \langle N \rangle, \qquad (2.25)$$

where T is the temperature, and because there is the identity

$$\langle E - \mu N \rangle = \frac{3}{2} \langle N \rangle kT + \int d1 \widetilde{u_1} \rho_1^{(1)} + \int d1 \int d2 v_{12} \rho_{12}^{(2)} + \int d1 \int d2 \int d3 w_{123} \rho_{123}^{(3)}.$$
(2.26)

It also follows that, if we neglect the three-body potential, the entropy is expressed as

$$S/k = -\beta \Gamma_2 + 3\langle N \rangle / 2 \quad (w = 0),$$
 (2.27)

and, if we further neglect the two-body potential, it is given by

$$S/k = -\beta \Gamma_1 + 3\langle N \rangle / 2 \quad (v = w = 0).$$
 (2.28)

We stress here that the stationary condition for the entropy, which leads to the self-consistent equations, automatically comes out as identities of the Legendre transformation. For example, the identities in (2.23) are equivalent to

$$\frac{\partial S/k}{\partial \rho_1^{(1)}} = \beta \widetilde{u_1} + \frac{3}{2}, \quad \frac{\partial S/k}{\partial \rho_{12}^{(2)}} = \beta v_{12}, \quad \frac{\partial S/k}{\partial \rho_{123}^{(3)}} = \beta w_{123}.$$
(2.29)

From the above argument it may be clear that, in general, the entropy of simple liquids theory, where potentials of higher than m-body are neglected, can be written as a functional of the correlation functions of the order up to the m-th. In such a case, higher order correlation functions can be expressed by the correlation functions of up to the m-th order.

By using the results given in this section, we can establish the general statement that *n*-particle density can be given as an explicit functional of *s*-particle densities where $s \le n-1$. From the second equality in Eq. (2.21) we readily notice that $\rho^{(2)}$ is given as a functional of $\rho^{(1)}$, *v*, and *w*. From the last equality in Eq. (2.22), we notice that $\rho^{(3)}$ is given by $\rho^{(2)}$, $\rho^{(1)}$, and *w*. In the same way, if we introduce Γ_n by taking into general *m*-body potentials, we can confirm that Γ_n is a functional of $\rho^{(s)}$ (*s*=1,2,...,*n*) as a consequence of the basic property of the Legendre transfor-

mation. Since $\rho^{(n)}$ is given by the derivative of Γ_{n-1} as in Eqs. (2.21) and (2.22), we arrive at the general statement. This statement is not limited to the classical case; the parallel argument in relativistic quantum field theory is developed in Ref. 31.

III. DIAGRAMMATIC EXPRESSION FOR THE THERMODYNAMIC POTENTIAL

The thermodynamic potential can be expressed as

$$e^{-\beta\Omega} = \Xi = \sum_{N=0}^{\infty} \frac{1}{N!} \int d[N] \left[\prod_{i} z_{i}^{*}\right] \cdot \left[\prod_{i < j} (1+f_{ij})\right] \cdot \left[\prod_{i < j < k} (1+t_{ijk})\right], \quad (3.1)$$

where

$$\int d[N] \equiv \int d\mathbf{q}_1 \int d\mathbf{q}_2 \cdots \int d\mathbf{q}_N, \qquad (3.2)$$

$$z_i^* \equiv z^*(\mathbf{q}_i) = z e^{-\beta u(\mathbf{q}_i)},\tag{3.3}$$

$$f_{ij} \equiv f(\mathbf{q}_i, \mathbf{q}_j) = e^{-\beta v(\mathbf{q}_i, \mathbf{q}_j)} - 1, \qquad (3.4)$$

and

$$t_{ijk} \equiv t(\mathbf{q}_i, \mathbf{q}_j, \mathbf{q}_k) = e^{-\beta w(\mathbf{q}_i, \mathbf{q}_j, \mathbf{q}_k)} - 1.$$
(3.5)

Here, we have introduced the activity

$$z = \frac{e^{\beta\mu}}{\Lambda^3},\tag{3.6}$$

where the de Broglie thermal wavelength $\Lambda = \sqrt{2 \pi \beta \hbar^2 / m}$ originates from the momentum integration $\int dp e^{-\beta |p|^2 / 2m} / h = \Lambda^{-1}$.

Diagrammatically $-\beta\Omega$ can be expressed as a sum of connected diagrams built up with black circles, 2-bonds, 3-bonds, and pseudo 2-bonds, where every pair of circles connected by the same 3-bond should be connected by a pseudo 2-bond.³² Here, the graphical terms are defined as follows. A 2-bond connects a pair of circles but a pair can not be connected directly more than once by 2-bonds. A 3-bond connects a triplet of circles but a triplet can not be connected directly more than once by 3-bonds. A pseudo 2-bond is basically the same as the 2-bond: it connects a pair of circles but a pair can not be connected directly more than once by 3-bonds. A pseudo 2-bond is basically the same as the 2-bond: it connects a pair of circles but a pair can not be connected directly more than once by pseudo 2-bonds and pseudo 2-bonds is the following: if a pair is connected by the same 3-bond, the pair should be connected by a pseudo 2-bond; otherwise, simply by a 2-bond. The pseudo 2-bond is found to be very useful for a collective representation of diagrams which contain 3-bonds. Without use of pseudo 2-bonds, the number of diagrams is dramatically increased. We give all diagrams of $-\beta\Omega$ explicitly up to the fourth order in the activity z:

(->

$$-\beta\Omega = -\beta\Omega^{(1)} + -\beta\Omega^{(2)} + \cdots, \qquad (3.7)$$

where

$$-\beta\Omega^{(1)} = \bullet \tag{3.8}$$

$$-\beta\Omega^{(2)} = \bullet - - \bullet \tag{3.9}$$

$$-\beta\Omega^{(3)} = + + + \qquad (3.10)$$

Symbol	Eqs. (3.8)-(4.7)	Eqs. (4.12)–(5.8)	Eqs. (5.12)-(6.6)	Eqs. (6.11)–(6.14)
		Appendix A	Appendix B	
•	z_1^*	n_1	n_1	n_1
1 2	f_{12}	f_{12}	h_{12}	h_{12}
1 2	$1 + f_{12}$	$1 + f_{12}$	$1 + h_{12}$	$1 + h_{12}$
3	t ₁₂₃	t_{123}	t_{123}	γ_{123}

TABLE I. Analytical expressions corresponding to the symbols in the specified parts of the text. The white circles are the same as the black circles but no integrations are associated with them.



We note here that, as suggested above, every triplet connected by a 3-bond forms a triangle whose sides are pseudo 2-bonds. We can convince ourselves that all the diagrams at the fourth order are written out in Eq. (3.11) if we note the following three points, which, respectively, concern the fourth order diagrams including two, three, and four 3-bonds. (1) If we chose two distinct triplets from four distinct points, two (and only two) points are necessarily shared by the two triplets. (2) If we chose three distinct triplets from four distinct points, one (and only one) point is necessarily shared by the three triplets, while each of the other three points is shared by two (and only two) of the three triplets. (3) If we chose four distinct triplets from four distinct points, every point is necessarily shared by three (and only three) of the four triplets, and every pair of points should be shared by two (and only two) of the four triplets.

The algebraic expression corresponding to a diagram is obtained by the following steps (see Table I).

- 1. Assign the numbers 1,2,...,n to all the black circles in an arbitrary way. We associate the factors $z_1^*, z_2^*, ..., z_n^*$ to the black circles labeled 1,2,...,n, respectively. Here *n* is the total number of black circles in the diagram.
- 2. Associate the factor f_{ij} and $1+f_{ij}$ to the 2-bond and the pseudo 2-bond, respectively, whose ends are labeled *i* and *j* in the first step.
- 3. Associate the factor t_{ijk} to the 3-bond whose ends are labeled *i*, *j*, and *k* in the first step.
- 4. Integrate the product of all the factors z^* , f, 1+f and t over the variables $\mathbf{q}_1, \dots, \mathbf{q}_n$.
- 5. Multiply the integral by the inverse of the symmetry number S of the diagram.

The symmetry number is the number of all possible ways of labeling (one of which is done in the first step) topologically equivalent to each other. For example, if a diagram contains *j* black circles

which play equivalent roles (in the sense any interchange of the labels among those circles does not change the topology of the graph), the set of *j* circles contributes the symmetry factor *j*!. The symmetry numbers for the diagrams in Eqs. (3.8)-(3.11) are successively given by 1,2, (2,3!,3!), $(2,3!,2\cdot4,2!,2!2!,4!,...,4!)$.

Since the pseudo 2-bonds are not conventional, we present two examples of diagrams which contain the pseudo 2-bonds:



Though Eq. (3.13) is mathematically correct under the above rules 1–5 for analytical expressions, only the second expression (which consists of two diagrams) accords with our convention of distinguishing 2-bonds from pseudo ones. (For example, in the first expression, a pair which is not connected by the same 3-bond is connected by a pseudo 2-bond and thus this expression violates our convention.) Because of this, we employ the second expression in what follows.

IV. INVERSION FROM ONE-BODY POTENTIAL TO ONE-PARTICLE DENSITY

In this section we renormalize the theory in terms of the one-particle density by using the inversion method. To this end we identify ϕ in the inversion formulae with the one-particle density $n_1 \equiv \rho^{(1)}(\mathbf{r}_1)$. The diagram expression for n_1 can be easily obtained from that for $-\beta\Omega$ if we notice that the differentiation by the one-particle potential u_1 changes one of the black circles to a white circle. This is understood from

$$-\beta n_1 = -\beta \,\frac{\delta\Omega}{\delta u_1},\tag{4.1}$$

and

$$\frac{\delta z_1^*}{\delta u_{1'}} = -\beta z_1^* \delta_{11'} \,. \tag{4.2}$$

Here, a white circle should be associated with z_i^* but no integration is performed over the variable *i*. Except for this rule for a white circle, the rules for obtaining an analytical expression from a diagram are not changed (see Table I).

We give all the diagrams up to the fourth order in the activity *z*:

$$n_1 = n_1^{(0)} [\tilde{u}] + n_1^{(1)} [\tilde{u}] + \cdots, \qquad (4.3)$$

where

$$n_1^{(0)}[\widetilde{u}] = \mathbf{O} \tag{4.4}$$

$$n_1^{(1)}[\widetilde{u}] = \mathbf{0} - \mathbf{\bullet} \tag{4.5}$$



where \cdots represents derivatives of the last six diagrams written in Eq. (3.11).

In the above $n^{(0)}$ is NOT the zero-th order in z and, strictly speaking, we should identify ϕ in the inversion formulae not with n_1 but n_1/z . However, here, we have identified ϕ with n_1 since the result is the same. We have also identified J with \tilde{u} and regarded \tilde{u} as an *artificial* source, though it should be set to the original value at the end. In other words, in the following we shall use the same symbol \tilde{u} for both J and j.

From Eq. (1.5), or $n_1 = n_1^{(0)} [\tilde{u}^{(0)}]$, and from Eq. (4.4), we have

$$n_1 = e^{-\beta \tilde{u}_1^{(0)}} / \Lambda^3 \quad (-\beta \tilde{u}_1^{(0)} = \ln n_1 + 3 \ln \Lambda).$$
(4.8)

This relation implies

$$z_1^*|_{\tilde{u} \to \tilde{u}^{(0)}} = n_1. \tag{4.9}$$

This means that the replacement $\tilde{u} \rightarrow \tilde{u}^{(0)}$, as implied in the inversion formulae Eqs. (1.5)–(1.8), is equivalent to a change of interpretation of circles: in the new diagram all the white and black circles should be associated not with z_i^* but with n_i .

In the present case where $\phi = n_1$, Eq. (1.6) can be re-written as

$$\int d1' \frac{\delta n_1^{(0)}[\tilde{u}^{(0)}]}{\delta \tilde{u}_{1'}^{(0)}} \tilde{u}_{1'}^{(1)} + n_1^{(1)}[\tilde{u}^{(0)}] = 0, \qquad (4.10)$$

where, from Eq. (4.4),

$$\frac{\delta n_1^{(0)}[\tilde{u}^{(0)}]}{\delta \tilde{u}_{1'}^{(0)}} = -\beta n_1 \delta_{11'}.$$
(4.11)

Thus, from Eq. (4.5), Eq. (4.10) is diagrammatically expressed as

$$-\beta n_1 \widetilde{u}_1^{(1)} + \mathbf{o} = 0, \qquad (4.12)$$

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from which we obtain

$$\beta \tilde{u}_{1}^{(1)} = - \bullet . \tag{4.13}$$

We stress here in the above two graphs the white and black circles now correspond not to z_i^* but to n_i . This is because these diagrams originate from the inversion formula in which we replace \tilde{u}_1 by $\tilde{u}_1^{(0)}$ and, as explained above, this replacement changes the circles in the diagrams from z_1^* to n_i . By the same token, all the circles should be associated with n_i in what follows (see Table I).

The first term in Eq. (1.7) can be written as $-\beta n_1 \tilde{u}_1^{(2)}$ as above. The second term in Eq. (1.7) in this case is written as

$$\frac{1}{2} \phi^{(0)''} (J^{(1)})^2 = \frac{1}{2} \int d1' \int d2' \frac{\delta^2 n_1^{(0)} [\tilde{u}^{(0)}]}{\delta \tilde{u}_{1'}^{(0)} \delta \tilde{u}_{2'}^{(0)}} \tilde{u}_{1'}^{(1)} \tilde{u}_{2'}^{(1)}, \qquad (4.14)$$

where, from Eq. (4.4),

$$\frac{\delta^2 n_1^{(0)}[\tilde{u}^{(0)}]}{\delta \tilde{u}_{1'}^{(0)} \tilde{u}_{2'}^{(0)}} = (-\beta)^2 \delta_{11'} \delta_{12'} \mathbf{0} \quad .$$
(4.15)

From Eq. (4.13), Eq. (4.14) can be expressed as

Since $\phi^{(1)}$ in Eq. (1.7), in this case, implies [from Eq. (4.5)]

$$\frac{\delta n_1^{(1)}[\tilde{u}^{(0)}]}{\delta \tilde{u}_{1'}^{(0)}} = -\beta \quad \underbrace{\mathbf{o}}_{\mathbf{1}} \quad -\beta \delta_{\mathbf{1}\mathbf{1'}} \quad \mathbf{o} \quad \mathbf{\bullet} \quad , \tag{4.16}$$

the third term in Eq. (1.7) can be expressed as -2 -2 -2 .

Here the factor 2 in front of the second graph appears because the symmetry factor of the graph is 2. (Up to now we have omitted the index 1 for a white circle, if there is only one white circle in the diagram.) Thus, Eq. (1.7) reduces to

$$-\beta n_1 \tilde{u}_1^{(2)} + \left(\begin{array}{c} & & \\ & &$$

In this equation, we see that the three diagrams explicitly written exactly cancel the 1-reducible diagrams appearing in $n_1^{(2)}[\tilde{u}^{(0)}]$. Here, the 1-reducible diagram is a diagram in which there is at least one circle the removal of which leads to a separation of the diagram. Thus we have

$$\beta \tilde{u}_{1}^{(2)} = \bigwedge^{1} + \bigwedge^{1} .$$
 (4.18)

Concerning Eq. (1.8), in a similar manner, we have (see Appendix A)

$$\phi^{(0)'}J^{(3)} = -\beta n_1 \widetilde{u}_1^{(3)}, \qquad (4.19)$$

$$\phi^{(0)''}J^{(1)}J^{(2)} \to + + , \qquad (4.20)$$

$$\frac{1}{3!}\phi^{(0)'''}(J^{(1)})^3 \to - \checkmark , \qquad (4.21)$$

$$\phi^{(2)'}J^{(1)} \rightarrow - 2 \qquad - 2 \qquad - 3 \qquad - 3$$

By substituting these results in Eq. (1.8), we see again that all the 1-reducible diagrams exactly cancel out. As a result we have

where \cdots represents derivatives of the last six diagrams written in Eq. (3.11).

We have derived $\tilde{u_1}$ explicitly up to the third order in n_1 (black circle) and in principle we can continue such a calculation up to the desired order. Then the inversion series is given by

$$\beta \widetilde{u}_1 = \beta \widetilde{u}_1^{(0)} + \beta \widetilde{u}_1^{(1)} + \beta \widetilde{u}_1^{(2)} + \cdots, \qquad (4.26)$$

where $\tilde{u}_{1}^{(i)}$ (*i*=0,1,2,3) are given in Eqs. (4.8), (4.13), (4.18), and (4.25). As we see below in Eq. (4.30), this inversion series, obtained by the inversion method, can be regarded as the on-shell condition and as a generalization of a well-known formula in simple liquids theory.

Now we can integrate the expression (4.26) to get a generating functional $\Gamma_1[n,v,w]$ due to the relation $\beta \tilde{u_1} = -\beta \delta \Gamma_1[n,v,w]/\delta n_1$. The result is given by



We stress here that in the above diagram circles are associated with n_i and thus all the diagrams are functional not of z_i^* but of n_i . All the diagrams up to the fourth order of n_i are explicitly shown in Eq. (4.27).

Equation (4.27) can be related to the previously known results for the entropy and the thermodynamic potential. The entropy in the case where v = w = 0 can be given by $-\beta\Gamma_1 + (3/2) \times \langle N \rangle$, as stated before. Indeed, if we further set u = 0 (the ideal gas), we recover the entropy of the ideal gas, $S/k = Vn(5/2 - \ln n - 3 \ln \Lambda)$, where *n* and *V* is the density and the volume of the system, respectively. If we neglect the triplet potential, Eq. (4.27) reduces to the standard expression for the thermodynamic potential renormalized by one-particle density:^{1,2}

$$-\beta\Omega = \int d1 n_1 (1 - \ln n_1 + \ln z - \beta u_1) + \mathcal{H}^{(1)}[n, f], \qquad (4.28)$$

where $\mathscr{K}^{(1)}[n, f]$ is the whole class of the 1-irreducible (1IR) diagrams built up with *n*-circles and *f*-bonds (*f*-bonds are functionals of *v*). Here the 1IR diagram is the one in which deletion of any one of the black circles does not cause a separation of the diagram.

The on-shell condition for the first Legendre transformation [the first equation in (2.21)], or

$$\beta \, \frac{\delta \Gamma_1[n, v, w]}{\delta n_1} = -\beta \widetilde{u_1}, \tag{4.29}$$

for Γ_1 in Eq. (4.27), reduces to

$$\beta u_1 = -\ln n_1 + \ln z + \mathbf{o} + \mathbf{o}$$

which is equivalent to the inversion series (4.26), as stated above. This inversion series is a generalization of a familiar equation in the simple liquid theory:^{1,4}

$$\ln(n_1/z_1^*) = K_1^{(1)}[n, f], \qquad (4.31)$$

where $K_1^{(1)}[n,f] = \delta \mathscr{H}^{(1)}[n,f]/\delta n_1$ is a sum of all 1-irreducible diagrams consisting of one white circle labeled 1, one or more black circles, and *f*-bonds. If we again neglect the triplet potential and set *u* to zero (a homogeneous liquid), Eq. (4.30) reduces to this standard result (4.31).

V. INVERSION FROM PAIR POTENTIAL TO TWO-PARTICLE DENSITY

In this section we renormalize the theory in terms of the two-particle density by using the inversion method. To this end we try to identify ϕ in the inversion formulae with the two-particle density $n_{12} \equiv \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2)$. The diagram expression for n_{12} can be easily obtained from that for Γ_1 if we notice that the differentiation by the two-particle potential v_{12} removes one of the (pseudo) 2-bonds with making black circles of both ends white circles if we multiply the resultant diagram by a factor $1 + f_{12}$. This is understood from the relations

$$-\beta n_{12} = -\beta \frac{\delta \Omega[u, v, w]}{\delta \frac{1}{2} v_{12}} = -\beta \frac{\delta \Gamma_1[n, v, w]}{\delta \frac{1}{2} v_{12}},$$
(5.1)

and

$$\frac{\delta f_{12}}{\delta \frac{1}{2} v_{1'2'}} = -\beta (1+f_{12}) (\delta_{11'} \delta_{22'} + \delta_{12'} \delta_{21'}).$$
(5.2)

Thus, we have

$$n_{12} = (1 + f_{12})(\mathbf{o} \ \mathbf{o} + \mathbf{o} + \mathbf{o} + \mathbf{o} + \mathbf{o} + \cdots)$$
 (5.3)

We notice here that all the diagrams have two white circles, which stand for n_i . This implies that, if we consider n_i as the expansion parameter of the inversion method, $n_{12}^{(0)}$ vanishes while the inversion method assumes a non-zero $\phi^{(0)}$. To avoid this difficulty we identify ϕ with a new quantity defined by

$$h_{12} = \frac{n_{12}}{n_1 n_2} - 1. \tag{5.4}$$

It is diagrammatically given as

$$h_{12} = h_{12}^{(0)} [v] + h_{12}^{(1)} [v] + \cdots, \qquad (5.5)$$

where

$$h_{12}^{(0)}[v] = \frac{1}{1-2} \tag{5.6}$$

$$h_{12}^{(1)}[v] = (1 + f_{12})($$
 $\bigwedge_{1 = 2}^{\bullet} + \bigwedge_{1 = 2}^{\bullet})$ (5.7)

$$h_{12}^{(2)}[v] = (1+f_{12})(1+f_{$$

Here, the terms represented by \cdots are the derivatives of the last three diagrams in Eq. (4.27). From Eq. (1.5), or $h_{12} = h_{12}^{(0)} [v^{(0)}]$, and from Eq. (5.6), we have

$$h_{12} = e^{-\beta v_{12}^{(0)}} - 1 \quad [-\beta v_{12}^{(0)} = \ln(h_{12} + 1)].$$
(5.9)

This implies

$$f_{12}|_{v \to v(0)} = h_{12}. \tag{5.10}$$

Equation (1.6) in this case can be written as

$$\int d1' \int d2' \frac{\delta h_{12}^{(0)}[v^{(0)}]}{\delta v_{1'2'}^{(0)}} v_{1'2'}^{(1)} + h_{12}^{(1)}[v^{(0)}] = 0,$$
(5.11)

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which reduces to (see Appendix B)

$$-\beta(1+h_{12})v_{12}^{(1)} + (1+h_{12})(\qquad \bigwedge_{1 \qquad 2}^{\bullet} + \qquad \bigwedge_{1 \qquad 2}^{\bullet}) = 0.$$
 (5.12)

Thus, we have

$$\beta v_{12}^{(1)} = \bigwedge_{1 \ 2}^{\bullet} + \bigwedge_{1 \ 2}^{\bullet} . \tag{5.13}$$

In the above diagrams the (pseudo) 2-bonds are not *f*-bonds but (pseudo) *h*-bonds due to the replacement $v \rightarrow v^{(0)}$ in the inversion formula (1.6) [see Eq. (5.10)]. Similarly the 2-bonds and the pseudo 2-bonds appearing in the following diagrams should all be associated with h_{ij} and $1 + h_{ij}$, respectively (see Table I).

The terms in Eq. (1.7) are calculated as follows (see Appendix B):

$$\phi^{(0)'}J^{(2)} = -\beta(1+h_{12})v_{12}^{(2)}, \qquad (5.14)$$

$$\frac{1}{2}\phi^{(0)''}(J^{(1)})^2 \to (1+h_{12})(\left[1\right]^2 + \left[1\right]^2 + \left[1\right]^2 + \left[1\right]^2 \right], \qquad (5.15)$$

$$\phi^{(1)'}J^{(1)} \rightarrow -(1+h_{12})(2(\prod_{1}^{2} + \prod_{1}^{2} + \prod_{2}^{2} + \prod_{1}^{2})) + 2\prod_{1}^{2} + \prod_{2}^{2} + \prod_{2}^{2} + \prod_{1}^{2} + \prod_{2}^{2} + \prod_{2}^{2} + \prod_{1}^{2} + \prod_{2}^{2} + \prod_{2}$$

By substituting these results in Eq. (1.7), we see that all the 2-reducible diagrams except

exactly cancel out. Here, the 2-reducible diagrams are defined as follows. First, we make a 0-diagram from the diagram by connecting external points 1,2 by a (pseudo) 2-bond along with n_1,n_2 and then by integrating the diagram over the variables 1, 2. (The 0-diagram, here, is a diagram which has no external points and white circles.) Then the original diagram is 2-reducible if the resultant 0-diagram is the one in which there is at least one pair of circles the deletion of which leads to a separation of the diagram.

From Eqs. (5.14) to (5.16), we have

$$\beta v_{12}^{(2)} = - \left[\begin{array}{c} & & \\ &$$

where \cdots represents derivatives of the last three diagrams in Eq. (4.27).

We have derived v_{12} explicitly up to the second order in n_1 (black circle) and in principle we can continue such a calculation up to the desired order. Then the inversion series is given by

$$\beta v_{12} = \beta v_{12}^{(0)} + \beta v_{12}^{(1)} + \beta v_{12}^{(2)} + \cdots, \qquad (5.18)$$

where $\beta v_{12}^{(i)}$ (*i*=0,1,2) are given in Eqs. (5.9), (5.13), and (5.17). As we see below in Eq. (5.35), Eq. (5.18) can be regarded as the on-shell condition and as a generalization of some equations familiar in the theory of simple liquids.

Now we can integrate the inversion series (5.18) to get a generating functional $\Gamma_2[n,h,w]$ due to the relation $\beta v = -2\beta \delta \Gamma_2[n,h,w]/\delta \rho^{(2)}$, or

$$\beta v_{12} = -\beta \, \frac{2}{n_1 n_2} \, \frac{\delta \Gamma_2[n,h,w]}{\delta h_{12}},\tag{5.19}$$

and the result is given by

$$-\beta\Gamma_{2} = \int d1n_{1}(1 - \ln n_{1} - 3\ln \Lambda) + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] + \frac{1}{2} \int d1 \int d2n_{1}$$

We stress here that in the above diagram circles, 2-bonds, and pseudo 2-bonds are associated with n_i , h_{ij} and $1 + h_{ij}$, respectively, and thus all the diagrams are functional of n_i , h_{ij} , and w_{ijk} . All the diagrams up to the fourth order of n_i are explicitly shown in Eq. (5.20). As stated before, from this expression we can readily obtain the entropy for the system without the triplet potential, which coincides with the expression given in Ref. 1.

If we neglect the triplet potential, Eq. (5.20) reduces to the standard expression for the thermodynamic potential renormalized by one-particle density:^{1,2}

$$-\beta\Omega = \int d1n_1(1 - \ln n_1 + \ln z - \beta u_1) - \frac{\beta}{2} \int d1 \int d2\rho_{12}^{(2)} v_{12} + \frac{1}{2} \int d1 \int d2n_1n_2 \{h_{12} - [1 + h_{12}]\ln[1 + h_{12}]\} + \mathcal{N}[n,h] + \mathcal{K}^{(2)}[n,h].$$
(5.21)

 $\mathcal{N}[n,h]$ are made up of polygonal diagrams and, if we regard $[nh]_{12} \equiv n_1 h_{12}$ is the (1,2) element of the functional matrix [nh], it is given by

$$\mathcal{N}[n,h] = -\sum_{p=3}^{\infty} \frac{(-1)^p}{2p} \operatorname{Tr}[nh]^p = \frac{1}{2} \operatorname{Tr}\left[\ln(1+nh) - nh + \frac{1}{2} (nh)^2\right].$$
(5.22)

 $\mathcal{K}^{(2)}[n,h]$ is the whole class of the 2-irreducible (2IR) diagrams built up with *n*-circles and *h*-bonds. Here, a 2IR diagram is the one in which the deletion of any pair of the black circles does not cause a separation of the diagram.

In the conventional definition, a 2-irreducible diagram consists of circles and 2-bonds but not of 3-bonds and pseudo 2-bonds. However, in the following, we include such diagrams consisting of 3-bonds and pseudo 2-bonds into the definition of 2IR diagrams, and call the whole class of such 2IR diagrams $\mathscr{K}^{(2)}[n,h,w]$ instead of $\mathscr{K}^{(2)}[n,h]$. Then, Eq. (5.20) is equivalent to Eq. (5.21) where $\mathscr{K}^{(2)}[n,h]$ is replaced by $\mathscr{K}^{(2)}[n,h,w]$.

The on-shell condition for the second Legendre transformation [the first two equations in (2.22)] are equivalent to

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$$\frac{\delta\Gamma_2[n,h,w]}{\delta n_1} = -\tilde{u_1} - \int d2n_2(1+h_{12})v_{12}, \qquad (5.23)$$

$$\frac{\delta\Gamma_2[n,h,w]}{\delta\frac{1}{2}h_{12}} = -n_1 n_2 v_{12}, \qquad (5.24)$$

from which we have

$$\ln z - \ln n_{1} + \int d2n_{2} \{h_{12} - (1 + h_{12})\ln(1 + h_{12})\} + \frac{\delta \mathcal{N}[n,h]}{\delta n_{1}} + \frac{\delta \mathcal{R}^{(2)}[n,h,w]}{\delta n_{1}}$$
$$= \beta u_{1} + \beta \int d2n_{2}(1 + h_{12})v_{12}, \qquad (5.25)$$

$$-\ln(1+h_{12}) + \frac{2}{n_1 n_2} \left(\frac{\delta \mathcal{N}[n,h]}{\delta h_{12}} + \frac{\delta \mathcal{R}^{(2)}[n,h,w]}{\delta h_{12}} \right) = \beta v_{12}, \qquad (5.26)$$

where

$$\frac{\delta \mathcal{N}[n,h]}{\delta n_1} = \frac{1}{2} \left(h \cdot \frac{1}{1+[nh]} - h + h \cdot [nh] \right)_{11}, \tag{5.27}$$

$$\frac{2}{n_1 n_2} \frac{\delta \mathcal{N}[n,h]}{\delta h_{12}} = \left(h - h \; \frac{1}{1 + [nh]}\right)_{12}.$$
(5.28)

This set of equations, corresponding to Eq. (4.30) in the previous section, is to be solved selfconsistently for the two variables n and h. In the above, h and [nh] stand for the functional matrix as before. The diagrammatic expression for the sum of derivatives of $\mathcal{N}[n,h]$ or $\mathcal{K}[n,h,w]$ in Eq. (5.25) and in Eq. (5.26) can be easily obtained from the diagrams in Eq. (5.20) by removing, in all possible ways, one of the black circles and one of the (pseudo) 2-bonds (along with two black circles at the both ends), respectively.

If we introduce the direct correlation function $c(\mathbf{r}_1,\mathbf{r}_2)=c_{12}$ by the so-called Ornstein–Zernike relation,

$$h_{12} = c_{12} + \int d3 c_{13} n_3 h_{32}, \qquad (5.29)$$

or, in our simplified notation,

$$h = c + c \cdot [nh] = c + c \cdot [nc] + c \cdot [nc]^2 + \cdots,$$
 (5.30)

we have

$$c = h - c \cdot [nh] = h - h \cdot [nh] + h \cdot [nh]^2 - \dots = h \frac{1}{1 + [nh]}.$$
(5.31)

Thus we obtain

$$\frac{\delta \mathcal{N}[n,h]}{\delta n_1} = \frac{1}{2} \left(c - h + h \cdot [nh] \right)_{11}, \tag{5.32}$$

$$\frac{2}{n_1 n_2} \frac{\delta \mathcal{N}[n,h]}{\delta h_{12}} = h_{12} - c_{12}, \qquad (5.33)$$

from which we have

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$$\ln z - \ln n_{1} + \int d2n_{2} \{h_{12} - (1 + h_{12}) [\ln(1 + h_{12}) + \beta v_{12}] \} + \frac{1}{2} \left(c_{11} - h_{11} + \int d2h_{12}n_{2}h_{21} \right) + \frac{\delta \mathscr{H}^{(2)}[n, h, w]}{\delta n_{1}} = \delta u_{1},$$
(5.34)

$$-\ln(1+h_{12})+h_{12}-c_{12}+\frac{2}{n_1n_2}\frac{\delta \mathscr{K}^{(2)}[n,h,w]}{\delta h_{12}}=\beta v_{12}.$$
(5.35)

Note here that c_{12} and the derivatives of $\mathscr{K}^{(2)}[n,h,w]$ are explicitly given as functionals of n and h. This set of equations are self-consistent equations for n_i, h_{ij} , which is equivalent to the set of (5.25) and (5.26), and Eq. (5.35) can be regarded as another expression of the inversion series (5.18), as stated above.

The inversion series (5.35), obtained as the on-shell condition, is a generalization of a familiar exact relation,

$$h_{12} - c_{12} - \ln(1 + h_{12}) = \beta v_{12} - d_{12}, \qquad (5.36)$$

which was derived by van Leenwen *et al.*,⁴ if d_{12} , originally given by the *f*-bond expansion, is rewritten as an *h*-bond expansion.

The hypernetted-chain (HNC) approximation,

$$h_{12} - c_{12} - \ln(1 + h_{12}) = \beta v_{12}, \qquad (5.37)$$

is reproduced if we neglect all the 2IR diagrams in Eq. (5.35).

The Percus-Yevick (PY) approximation can be expressed as

$$\beta v_{12} + \ln(1 + h_{12}) = \ln(1 + h_{12} - c_{12}), \qquad (5.38)$$

implying the approximation

$$h_{12} - c_{12} + \frac{2}{n_1 n_2} \frac{\delta \mathscr{K}^{(2)}[n, h, w]}{\delta h_{12}} \simeq \ln(1 + h_{12} - c_{12}).$$
(5.39)

VI. INVERSION FROM TRIPLET POTENTIAL TO THREE-PARTICLE DENSITY

In this section we renormalize the theory in terms of three-particle density by using the inversion method. To this end we try to identify ϕ in the inversion formulae with the two-particle density $n_{12} \equiv \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2)$. The diagram expression for n_{123} can be easily obtained from that for Γ_2 if we notice that the differentiation by the three-particle potential w_{123} removes one of the 3-bonds with making black circles of the three ends white circles if we multiply the resultant diagram by a factor $1 + t_{123}$. This is understood from the relations

$$-\beta n_{123} = -\beta \frac{\delta \Omega[u, v, w]}{\delta \frac{1}{3!} w_{123}} = -\beta \frac{\delta \Gamma_2[n, h, w]}{\delta \frac{1}{3!} w_{123}}$$
(6.1)

and

$$\frac{\delta t_{123}}{\delta \frac{1}{3!} w_{1'2'3'}} = -\beta (1+t_{123}) [\delta_{11'} \delta_{22'} \delta_{33'} + (3!-1) \text{ terms}].$$
(6.2)

Then, we notice that all the diagrams of n_{123} have three white circles. This implies that, if we regard n_i as the expansion parameter of the inversion method, $n_{123}^{(0)}$ vanishes while the inversion

method assumes non-zero $\phi^{(0)}$. In addition, since, in all the diagrams of $\Gamma_2[n,h,w]$, every t_{ijk} is accompanied by a factor $n_i n_j n_k (1+h_{ij})(1+h_{jk})(1+h_{ki})$, it is convenient to identify ϕ not with n_{123} but with γ_{123} defined by

$$\gamma_{123} = \frac{n_{123}}{n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31})} - 1.$$
(6.3)

Then this quantity is diagrammatically given as

$$\gamma_{123} = \gamma_{123}^{(0)} + \gamma_{123}^{(1)} + \cdots, \qquad (6.4)$$

where

$$\gamma_{123}^{(0)} = \bigwedge_{1 = 2}^{3} , \qquad (6.5)$$



From Eq. (1.5), or $\gamma_{123} = \gamma_{123}^{(0)} [w^{(0)}]$, and from Eq. (6.5), we have

$$\gamma_{123} = e^{-\beta_{w_{123}^{(0)}}} - 1 \quad [-\beta_{w_{123}^{(0)}} = \ln(\gamma_{123} + 1)]. \tag{6.7}$$

This implies

$$t_{123}|_{w \to w}(0) = \gamma_{123}. \tag{6.8}$$

Equation (1.6) in this case can be written as

$$\int d1' \int d2' \int d3' \frac{\delta \gamma_{123}^{(0)}[w^{(0)}]}{\delta w_{1'2'3'}^{(0)}} w_{1'2'3'}^{(1)} + \gamma_{123}^{(1)}[w^{(0)}] = 0,$$
(6.9)

where

$$\frac{\delta \gamma_{123}^{(0)}[w^{(0)}]}{\delta w_{1'2'3'}^{(0)}} = -\frac{\beta}{3!} (1+\gamma_{123}) [\delta_{11'} \delta_{22'} \delta_{33'} + (3!-1) \text{ terms}].$$
(6.10)

Thus, we have



Here, the 3-bonds should be associated not with t_{ijk} but with γ_{ijk} [see Eq. (6.8) and Table I].

We have derived w_{123} explicitly up to the first order in n_1 (black circle) and in principle we can continue such a calculation up to the desired order. Then the inversion series is given as

$$\beta w_{123} = \beta w_{123}^{(0)} + \beta w_{123}^{(1)} + \beta w_{123}^{(2)} + \cdots, \qquad (6.12)$$

where $\beta w_{123}^{(i)}$ (*i*=0,1) are given in Eqs. (6.7) and (6.11). As we see below in Eq. (6.20), this inversion series can be obtained from the on-shell condition and can be seen as a series which systematically improve the Kirkwood's approximation for the three-particle density.

Now we can integrate the inversion series (6.12) to get a generating functional $\Gamma_3[n,h,\gamma]$ due to the relation $\beta w = -3!\beta \delta \Gamma_3[n,h,\gamma]/\delta \rho^{(3)}$, or

$$\beta w_{123} = -\beta \frac{3!}{n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31})} \frac{\delta \Gamma_3[n,h,\gamma]}{\delta \gamma_{123}}, \tag{6.13}$$

and the result is given by

$$-\beta\Gamma_{3} = \int d1n_{1}(1 - \ln n_{1} - 3\ln\Lambda) + \frac{1}{2} \int d1 \int d2n_{1}n_{2}[h_{12} - (1 + h_{12})\ln(1 + h_{12})] \\ + \frac{1}{3!} \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{123} - (1 + \gamma_{123})\ln(1 + \gamma_{123})] \\ + \int d1 \int d1 \int d2 \int d3n_{1}n_{2}n_{3}(1 + h_{12})(1 + h_{23})(1 + h_{31})[\gamma_{12} - (1 + \gamma_{123})\ln(1 + \gamma_{123})(1 + \eta_{123})(1 + \eta_{123}$$

In the above diagrams, the 3-bonds are not *t*-bonds but γ -bonds due to the replacement $w \rightarrow w^{(0)}$ in the inversion formula (1.6) [see Eq. (6.8) and Table 1]. We stress here that, in the above diagram, circles, 2-bonds, pseudo 2-bonds, and 3-bonds are associated with n_i , h_{ij} , $1 + h_{ij}$, and γ_{ijk} , respectively, and thus all the diagrams are functional of n_i , h_{ij} , and γ_{ijk} . All the diagrams up to the fourth order of n_i are explicitly shown in Eq. (6.14).

From Eq. (6.14), the renormalized entropy S/k of the system can be easily obtained as a functional of one- to three-particle densities due to the relation $S/k = -\beta\Gamma_3 + (3/2)\int dln_1$, as stated earlier. This result for the entropy should be compared with the previously known result, for example, given in Ref. 8. Up to the third order (in n_i), these two coincide with each other. However, above this order, they look different. For example, while the fourth order of $\Delta S/k$ given in Ref. 8 includes the four-particle density, our expression does not contain the four-particle density and is written in terms of only one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle density can be written in terms of one- to three-particle densities.

The result (6.14) was suggested in Ref. 2, where the diagrammatic part is collectively called $\mathscr{T}[n,h,\gamma]$. In there, $\mathscr{T}[n,h,\gamma]$ is not defined explicitly and any diagrams contained in it are not presented, unfortunately.

The on-shell conditions for the third Legendre transformation [the three equations in (2.23)] are equivalent to

$$\frac{\delta\Gamma_3}{\delta n_1} = -u_1 - \int d2n_2(1+h_{12})v_{12} - \frac{1}{2} \int d2d3n_2n_3(1+h_{12})(1+h_{23})(1+h_{31})(1+\gamma_{123})w_{123},$$
(6.15)

$$\frac{\delta\Gamma_3}{\delta h_{12}} = -\frac{1}{2} n_1 n_2 v_{12} - \frac{1}{2} n_1 n_2 \int d3 n_3 (1+h_{23})(1+h_{31})(1+\gamma_{123}) w_{123}, \qquad (6.16)$$

$$\frac{\delta\Gamma_3}{\delta\gamma_{123}} = -\frac{1}{3!} n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31}) w_{123}, \qquad (6.17)$$

from which we have

$$\ln z - \ln n_1 + \int d2n_2 [h_{12} - (1 + h_{12}) \{ \ln(1 + h_{12}) + \beta v_{12} \}] + \frac{1}{2} \int d2d3n_2n_3(1 + h_{12})(1 + h_{23})$$

$$\times (1+h_{31})[\gamma_{123} - (1+\gamma_{123})\{\ln(1+\gamma_{123}) + \beta w_{123}\}] + \frac{\delta \mathscr{Y}}{\delta n_1} = \beta u_1, \qquad (6.18)$$

$$-\ln(1+h_{12}) + \int d3n_3(1+h_{23})(1+h_{31}) [\gamma_{123} - (1+\gamma_{123})\{\ln(1+\gamma_{123}) + \beta w_{123}\}]$$

$$+\frac{2}{n_1 n_2} \frac{\delta \mathscr{Y}}{\delta h_{12}} = \beta v_{12}, \tag{6.19}$$

$$-\ln(1+\gamma_{123}) + \frac{3!}{n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31})} \frac{\delta \mathscr{F}}{\delta \gamma_{123}} = \beta w_{123}.$$
(6.20)

This set of equations, corresponding to the set of (5.25) and (5.26) in the previous section, is to be solved for the three variables n, h, and γ .

This set can also be seen as a generalization of Eqs. (6)–(8) in Ref. 12. If we approximate \mathscr{T} by the fifth term in Eq. (5) in Ref. 12, which is originally obtained in Ref. 11, this set of equations here reduce to Eqs. (6)–(8) in Ref. 12. However, we notice that our diagrammatic expression for \mathscr{T} does not have diagrams directly corresponding to the fifth term in Eq. (5) in Ref. 12. This is because this fifth term originates from the last term in Eq. (4) of Ref. 12, and this term, which should be equal to \mathscr{T} for the system without higher order potential than the third, contains correlation functions of up to the infinite order while \mathscr{T} here contains correlation functions up to the third order.

As mentioned above, Eq. (6.20) can be expressed as

$$\beta w_{123} = \beta w^{(0)} + \beta w^{(1)} + \beta w^{(2)} + \cdots, \qquad (6.21)$$

where

$$\beta w^{(0)} = -\ln(1 + \gamma_{123}), \tag{6.22}$$

$$\beta w^{(1)} = \int d4n_4 h_{41} h_{42} h_{23} + \cdots, \qquad (6.23)$$

where \cdots represents γ -dependent terms which correspond to the last seven diagrams in Eq. (6.11).

Equation (6.21) or (6.20) can be related to some of the previously known approximations. If we truncate the inversion series (6.21) at i=0, we obtain the well-known approximation $\gamma_{123}=e^{-\beta w_{123}}-1$ (for example, see Ref. 12). At $w_{123}=0$, this approximation reduces to the Kirkwood's superposition approximation,

$$n_{123} = n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31}).$$
(6.24)

If we truncate the series at i=1 and further neglect the γ -dependent terms in $\beta w^{(1)}$ (with $w_{123}=0$), we get the first order approximation given in Ref. 24,

$$n_{123} = n_1 n_2 n_3 (1+h_{12})(1+h_{23})(1+h_{31}) \exp\left[\int d4n_4 h_{41} h_{42} h_{43}\right].$$
(6.25)

Equation (6.25) with *h*-bonds in the exponential replaced by *f*-bonds had also been studied (see, for example, Refs. 21-23).

Thus, in our formulation, the zero-th order approximation reproduces the superposition approximation and the first order approximation, which is the solution of the equation [obtained from Eq. (6.11)],

$$\beta w_{123} = -\ln(1+\gamma_{123}) + \int d4n_4h_{41}h_{42}h_{43} + \int d4n_4h_{41}(1+h_{42})(1+h_{43})\gamma_{423} + \int d4n_4h_{42}(1+h_{41})(1+h_{43})\gamma_{413} + \int d4n_4h_{43}(1+h_{41})(1+h_{42})\gamma_{412} + \int d4n_4(1+h_{41})(1+h_{42})(1+h_{43})(\gamma_{412}\gamma_{423} + \gamma_{421}\gamma_{413} + \gamma_{413}\gamma_{423}) + \int d4n_4(1+h_{41})(1+h_{42})(1+h_{43})\gamma_{412}\gamma_{423}\gamma_{431}, \qquad (6.26)$$

contains more information than the first approximation in Ref. 24. Solving our first order approximation is practically an interesting problem, which is reserved for a future work. Further systematic improvement of the approximation is obtained if we calculate $\beta w^{(2)}, \beta w^{(3)}, \ldots$, by the inversion method.

VII. DISCUSSION

In this paper, we applied the inversion method to a classical system and renormalized liquid theory in terms of one-, two-, and three-particle densities in the presence of the three-body potential. Our approach here complements the previously known re-summation techniques. We showed that the entropy functional can be expressed by up to three-particle densities and gave lower order diagrams of the entropy explicitly. We also pointed out that the stationary conditions for the entropy leads to a set of self-consistent equations. In a certain case, the self-consistent equation can be a base for a systematic advancement of the Kirkwood's superposition approximation.

Some of the results presented here are mere repetition or only a slight extension of the well-known results as suggested frequently in the text. We could not avoid mixing up known and unknown results to explain our approach explicitly. Here, we appreciate and clarify to what extent these results can be considered original. In Section II, the introduction of Γ_3 and its relation to the entropy seems a rather novel viewpoint. In Sections III–V, all the results are at least implied in the previous literature (see, for example, Ref. 19). However, we pointed out that, if one introduces the pseudo 2-bonds, the diagrams of Ω and Γ_n in the presence of the 3-body potential can be written economically and we gave the diagrams of (up to) the fourth order ($\Omega^{(4)}$, etc.) explicitly; otherwise the number of diagrams is too many to be included in a paper—this may be one reason the fourth order diagrams (in the presence of the 3-body potential) have not been given explicitly in the literature. In addition, the derivation of Γ_n based on the inversion method has originality and complements the previously known ways. The results in Sec. VI are the renormalization in terms

of one-, two-, and three-particle densities. In that the entropy does not include more-than-threeparticle densities, this explicit diagrammatic result may be original. The result reduces to some previously known approximations in special cases.

The present work can be related to the work by Stell¹⁹ in the following way. In the second article in Ref. 19, he gave diagrammatic rules for some correlation functions in terms of ρ_1 , h_2 , and f_s (where $s \ge 3$) in his notation. In the special case where there are no more-than-three-body potentials, his result can be regarded as the diagrammatic rule in terms of one- and two-particle densities and the three-body potential. (Notice here that his f_3 is our t_{ijk} .) This corresponds to the description by Γ_2 , which is still a functional of the three-body potential w_{123} (or f_3), in our language: the theory is renormalized up to the two-particle densities. In the present work, though it is limited to the special case (without *m*-body potential where $m \ge 4$), we further changed the variable w_{ijk} (or f_3) of the theory to the three-particle density $\rho^{(3)}$ or γ_{ijk} . This is accomplished by use of Γ_3 , which is a functional of $\rho^{(3)}$, and then the theory is renormalized up to the three-particle density $\rho^{(3)}$ or γ_{ijk} . This is accomplished by use of Γ_3 , which is a functional of $\rho^{(3)}$, coincides with the second article in Ref. 19 (which is expressed in terms of $\rho^{(1)}$, h_{ij} , and t_{ijk}), coincides with the second term on the left-hand side of Eq. (6.20) of this article (which is expressed in terms of $\rho^{(1)}$, h_{ij} , and t_{ijk}), when renormalized up to the three-particle densities by changing the variable from t_{ijk} to γ_{ijk} .

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APPENDIX A: USEFUL FORMULAE FOR THE INVERSION FROM ONE-PARTICLE POTENTIAL TO ONE-PARTICLE DENSITY

In this appendix, we present useful formulae for the inversion process. Below $\phi^{(i)''\cdots'}$ stands for $\delta^k n_1^{(i)} [u^{(0)}] / \delta \widetilde{u}_{1'}^{(0)} \delta \widetilde{u}_{2'}^{(0)} \cdots \delta \widetilde{u}_{k'}^{(0)}$:

$$\phi^{(0)\prime} \to -\beta \delta_{11\prime} \quad \mathbf{O}$$
(A1)

$$\phi^{(0)''} \to (-\beta)^2 \delta_{11'} \delta_{12'} \, \mathop{\mathbf{o}}_{1}$$
 (A2)

$$\phi^{(0)'''} \to (-\beta)^3 \delta_{11'} \delta_{12'} \delta_{13'} \begin{array}{c} \mathbf{0} \\ 1 \end{array}$$
 (A3)

$$\phi^{(1)\prime} \to -\beta \begin{array}{c} \mathbf{O} \\ \mathbf{O} \\ \mathbf{I} \end{array} \begin{array}{c} \mathbf{O} \\ \mathbf{I} \end{array} - \beta \delta_{11\prime} \begin{array}{c} \mathbf{O} \\ \mathbf{I} \end{array} \begin{array}{c} \mathbf{O} \\ \mathbf{I} \end{array}$$
(A4)

$$\phi^{(1)\prime\prime} \to (-\beta)^2 (\delta_{11\prime} \delta_{12\prime} \ \mathbf{0}_{1} \bullet + \delta_{11\prime} \ \mathbf{0}_{2} \bullet + \delta_{12\prime} \ \mathbf{0}_{1} \bullet + \delta_{1\prime2\prime} \ \mathbf{0}_{1} \bullet \mathbf{0}_{1} + \delta_{1\prime2\prime} \ \mathbf{0}_{1} \bullet \mathbf{0}_{1}$$
(A5)

$$\phi^{(2)'} \to (\checkmark)' + (\checkmark)', (A6)$$

where

$$(\int (\delta_{11'})' = -\beta(\delta_{11'}) + (\delta_{11'}) + (\delta_{11'}) + (\delta_{11'})$$
 (A7)

$$(\bigwedge^{\mathbf{A}})' = -\beta (\bigwedge^{\mathbf{A}} + \delta_{11'} \bigwedge^{\mathbf{A}})$$
 (A8)

$$(\bigwedge^{l})' = -\beta(\delta_{11'} \bigwedge^{l} + \bigwedge^{l}) \qquad (A9)$$

$$(\beta)' = -\beta(\delta_{11'} \beta + \delta_{1'})$$
 (A10)

APPENDIX B: USEFUL FORMULAE FOR THE INVERSION FROM TWO-PARTICLE POTENTIAL TO TWO-PARTICLE DENSITY

In this appendix, we present useful formulae for the inversion process. Below $\phi^{(i)n'\cdots}$ stands for $\delta^k h_{12}^{(i)}[v^{(0)}]/\delta v_{1'}^{(0)} \delta v_{2'}^{(0)} \cdots \delta v_{k'}^{(0)}$:

$$\phi^{(0)'} \to -\frac{\beta}{2} (1+h_{12}) (\delta_{11'} \delta_{22'} + \delta_{12'} \delta_{21'}) \tag{B1}$$

$$\phi^{(0)} \rightarrow \left(-\frac{\beta}{2}\right)^2 (1+h_{12})(\delta_{11'}\delta_{22'}+\delta_{12'}\delta_{21'})(\delta_{13'}\delta_{24'}+\delta_{14'}\delta_{23'}) \tag{B2}$$

$$\phi^{(1)'} \to (1+h_{12})'(\bigwedge_{1}^{\bullet} 2 + \bigwedge_{1}^{\bullet} 2) + (1+h_{12})(\bigwedge_{1}^{\bullet} 2 + \bigwedge_{1}^{\bullet} 2)'$$
where
(B3)

where

$$(1+h_{12})' = -\frac{\beta}{2} (1+h_{1'2'}) (\delta_{11'} \delta_{22'} + \delta_{12'} \delta_{21'})$$
(B4)

$$(\bigwedge_{1} \sum_{2})' = -\frac{\beta}{2}(1+h_{1'2'})(\delta_{11'} \ \mathbf{o}_{2'-2} + \delta_{12'} \ \mathbf{o}_{1'-2} + \delta_{21'} \ \mathbf{o}_{2'-1} + \delta_{22'} \ \mathbf{o}_{1'-1})$$
(B5)

$$(\begin{array}{c} & & \\ & &$$

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- ²⁸ If we consider the system without the three-body force, $w(\mathbf{r}, \mathbf{r}', \mathbf{r}'')$ here is artificial. We can still extract new information, which otherwise could not be obtained, by setting $w(\mathbf{r}, \mathbf{r}', \mathbf{r}'') = 0$ at the end (as mentioned before).
- ²⁹The two-particle density can also be obtained by noting the following relation:

 $-(1/\beta) \left[\delta^{2} \Omega / \delta u(\mathbf{r}) \delta u(\mathbf{r}') \right] = \langle \rho_{N}^{(1)}(\mathbf{r}) \rho_{N}^{(1)}(\mathbf{r}') \rangle - \langle \rho_{N}^{(1)}(\mathbf{r}) \rangle \langle \rho_{N}^{(1)}(\mathbf{r}') \rangle = \rho^{(2)}(\mathbf{r},\mathbf{r}') + \delta(\mathbf{r}-\mathbf{r}')\rho^{(1)}(\mathbf{r}) - \rho^{(1)}(\mathbf{r})\rho^{(1)}(\mathbf{r}').$ (B7)

The second equality can be understood from $\rho^{(2)}(\mathbf{r},\mathbf{r}')$:

$$= \langle \rho_N^{(1)}(\mathbf{r}) \rho_N^{(1)}(\mathbf{r}') \rangle = \delta(\mathbf{r} - \mathbf{r}') \rho^{(1)}(\mathbf{r}), \qquad (B8)$$

which is obtained by taking the expectation of Eq. (2.4).

³⁰We tacitly assume $\rho_N^{(1)}(\mathbf{r}) = 0$ for N = 0, $\rho_N^{(2)}(\mathbf{r}, \mathbf{r}') = 0$ for $N = 0, 1, \text{ and } \rho_N^{(3)}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') = 0$ for N = 0, 1, 2.

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- ³² The right-hand side of Eq. (3.1) can be expressed by a sum of all possible diagrams built up with the four elements (black circles, 2-bonds, 3-bonds, and pseudo 2 ponds), which includes disconnected diagrams. Here, as suggested in the text, if there is a 3-bond, every pair from the triplet (connected by the 3-bond) should be connected by a pseudo 2-bond. On the other hand, due to the operation of logarithm, $-\beta\Omega$ does not include disconnected diagrams.