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**LETTER TO THE EDITOR**

# Analytical solution of the Ornstein–Zernike equation for a multicomponent fluid

M Yasutomi and M Ginoza

Department of Physics and Earth Sciences, College of Science, University of the Ryukyus, Nishihara-Cho, Okinawa 903-0213, Japan

E-mail: b984942@sci.u-ryukyu.ac.jp

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**Abstract.** A formal solution of the Ornstein–Zernike equation for a multicomponent fluid consisting of hard-spherical molecules is studied with the closure of the usual hard-sphere condition and the following:

$$c_{ij}(r) = \sum_{n=1} \left( \frac{K_{ij}^{(n)}}{r} + L_{ij}^{(n)} z_n \right) \mathrm{e}^{-z_n r} \qquad \sigma_{ij} < r$$

where $c_{ij}(r)$ is the direct correlation function and $\sigma_{ij}$ is the closest distance between $i$ and $j$ species of molecules. The solution is expressed in terms of the physical solutions of a system of nonlinear algebraic equations. The result is a generalization of that of Blum (1980 *J. Stat. Phys.* **22** 661).

The thermodynamical and structural properties of a fluid are described by correlation functions between molecules in the fluid. One of the most useful schemes for calculations of correlation functions is based on the Ornstein–Zernike (OZ) equation. The OZ equation is in principle solved when it is closed by a closure relation. Many workers have studied the analytical solutions of the OZ equation with the closures in the mean spherical approximation (MSA) or the generalized mean spherical approximation (GMSA). As far as the present authors are aware, the case of the most general MSA closure is solved by Blum and Høye (1978) and Blum (1980), where the closure is defined as follows:

$$g_{ij}(r) \equiv h_{ij}(r) + 1 = 0 \qquad r < \sigma_{ij} = (\sigma_i + \sigma_j)/2 \tag{1}$$

and

$$c_{ij}(r) = \sum_{n=1} \frac{K_{ij}^{(n)}}{r} \mathrm{e}^{-z_n r} \qquad \sigma_{ij} < r \tag{2}$$

where $h_{ij}(r)$ and $c_{ij}(r)$ are the total and the direct correlation functions for two spherical molecules of $i$ and $j$ species, $\sigma_i$ is the diameter of $i$ species of molecule and $K_{ij}^{(n)}$ and $z_n$ are parameters characterizing the interaction between molecules. The work is based on the Baxter formalism of the OZ equation (Baxter 1970).

For the formal solution by Blum and Høye, one of the present authors (Ginoza 1986a, b) considered the factorizable case: $K_{ij}^{(n)} = K^{(n)} d_i^{(n)} d_j^{(n)}$. This case actually gives considerable simplification for the formal solution and has been useful: the tractable expressions have been obtained for thermodynamic (Ginoza 1990, Yurdabak *et al* 1994, Herrera *et al* 1996) and structural (Ginoza and Yasutomi 1998a) quantities of a fluid with size and interaction polydispersities as well as a fluid with an arbitrary number of components. Such investigations are still progressing (Blum *et al* 1992, Ginoza and Yasutomi 1998b).

In the present Letter, a new case of the following closure relation will be considered:

$$c_{ij}(r) = \sum_{n=1} \left( \frac{K_{ij}^{(n)}}{r} + L_{ij}^{(n)} z_n \right) e^{-z_n r} \qquad \sigma_{ij} < r. \tag{3}$$

This is a generalization of the closure of equation (2). As far as the present authors are aware, no analytical solution of the OZ equation with the closure of equations (1) and (3) has been given yet. The aim of the present Letter is to present the formal solution for the new case. The work would be statistical-mechanically interesting and useful for understanding colloidal fluids as well (Sood 1991). In particular, the single term case of equation (3) corresponds to the Sogami–Ise potential (Sogami and Ise 1984); the present result prompts us to investigate the thermodynamic and structural properties of the colliodal fluids.

We consider the multicomponent fluid. In the Baxter formalism of the OZ equation, the Baxter function $Q_{ij}(r)$ plays a essential role and the OZ equation is given as follows:

$$2\pi r c_{ij}(r) = -\frac{d}{dr} Q_{ij}(r) + \sum_l \rho_l \int_{\lambda_{lj}}^{\infty} dt\ Q_{jl}(t) \frac{d}{dr} Q_{il}(r+t) \tag{4$a$}$$

$$2\pi r h_{ij}(r) = -\frac{d}{dr} Q_{ij}(r) + 2\pi \sum_l \rho_l \int_{\lambda_{jl}}^{\infty} dt\ Q_{lj}(t)(r-t) h_{il}(|r-t|) \tag{4$b$}$$

where $\rho_l$ stands for the number density of molecules of $l$ species and $\lambda_{lj} = (\sigma_l - \sigma_j)/2$. We shall obtain $Q_{ij}(r)$ by solving these equations with the closure of equations (1) and (3).

As usual (Blum and Høye 1978, Blum 1980), we write the function $Q_{ij}(r)$ as

$$Q_{ij}(r) = Q_{ij}^0(r) + Q_{ij}^1(r) \tag{5$a$}$$

where

$$Q_{ij}^0(r) = 0 \qquad r > \sigma_{ij} \text{ or } r < \lambda_{ji}. \tag{5$b$}$$

Substitution of equation (5$a$) into equation (4$a$) and the use of equations (3) and (5$b$) yields

$$2\pi \sum_{n=1} \left( K_{ij}^{(n)} + L_{ij}^{(n)} z_n r \right) e^{-z_n r} = -\frac{d}{dr} Q_{ij}^1(r) + \sum_l \rho_l \int_{\lambda_{lj}}^{\infty} dt\ Q_{jl}(t) \frac{d}{dr} Q_{il}^1(r+t)$$
$$r > \sigma_{ij}. \tag{6$a$}$$

Substitution of equation (5$a$) into (4$b$) and the use of equations (1) and (5$b$) yields

$$\frac{d}{dr} Q_{ij}^0(r) = A_j\, r + \left( \beta_j - 1/2\sigma_j A_j \right) - \frac{d}{dr} Q_{ij}^1(r) - 2\pi \sum_l \rho_l \int_{\sigma_{il}}^{\infty} dt\ g_{il}(t)\ t\ Q_{lj}^1(t+r)$$
$$\lambda_{ji} < r < \sigma_{ij} \tag{6$b$}$$

where

$$A_j = 2\pi \left( 1 - \sum_l \rho_l T_{lj}^{(0)} \right) \tag{7$a$}$$

$$\beta_j = \frac{1}{2} \sigma_j A_j + 2\pi \sum_l \rho_l T_{lj}^{(1)} \tag{7$b$}$$

with

$$T_{lj}^{(n)} = \int_{\lambda_{jl}}^{\infty} \mathrm{d}r\ r^n Q_{lj}(r). \tag{7c}$$

Now, equations (6a) and (6b) suggest the following functional form for $Q_{ij}^1(r)$:

$$Q_{ij}^1(r) = \sum_{n=1} \left( D_{ij}^{(n)} + E_{ij}^{(n)} z_n r \right) \mathrm{e}^{-z_n r}. \tag{8}$$

In fact, the direct substitution shows that equation (8) is the solution of equation (6*a*) if the following equations are satisfied:

$$2\pi K_{ij}^{(n)} = z_n \sum_l \left\{ (D_{il}^{(n)} - E_{il}^{(n)})[\delta_{lj} - \rho_l \tilde{Q}_{jl}(\mathrm{i}z_n)] - E_{il}^{(n)} z_n \rho_l \tilde{Q}_{jl}^{(1)}(\mathrm{i}z_n) \right\} \tag{9a}$$

$$2\pi L_{ij}^{(n)} = z_n \sum_l E_{il}^{(n)} \left[ \delta_{lj} - \rho_l \tilde{Q}_{jl}(\mathrm{i}z_n) \right] \tag{9b}$$

where

$$\tilde{Q}_{jl}(s) = \int_{\lambda_{lj}}^{\infty} \mathrm{d}r\ Q_{jl}(r) \mathrm{e}^{\mathrm{i}sr} \tag{10a}$$

$$\tilde{Q}_{jl}^{(1)}(s) = \int_{\lambda_{lj}}^{\infty} \mathrm{d}r\ Q_{jl}(r) r \mathrm{e}^{\mathrm{i}sr}. \tag{10b}$$

On the other hand, substituting equation (8) into equation (6*b*) and solving the resulting differential equation with the use of the boundary condition $Q_{ij}^0(\sigma_{ij}) = 0$, we obtain

$$Q_{ij}^0(r) = \frac{1}{2}(r - \sigma_{ij})(r - \lambda_{ji})A_j + (r - \sigma_{ij})\beta_j + \sum_n \left( C_{ij}^{(n)} + F_{ij}^{(n)} \right) \left( \mathrm{e}^{-z_n r} - \mathrm{e}^{-z_n \sigma_{ij}} \right)$$
$$+ \sum_n z_n F_{ij}^{(n)} \left( r\mathrm{e}^{-z_n r} - \sigma_{ij} \mathrm{e}^{-z_n \sigma_{ij}} \right) \qquad \lambda_{ji} \leqslant r \leqslant \sigma_{ij} \tag{11}$$

where

$$C_{ij}^{(n)} = -D_{ij}^{(n)} + E_{ij}^{(n)} + \sum_l \left( D_{lj}^{(n)} \gamma_{il}(z_n) + E_{lj}^{(n)} \gamma_{il}^{(1)}(z_n) \right) \tag{12a}$$

$$F_{ij}^{(n)} = -E_{ij}^{(n)} + \sum_l E_{lj}^{(n)} \gamma_{il}(z_n) \tag{12b}$$

with

$$z_n \gamma_{il}(z_n) = 2\pi \rho_l \tilde{g}_{il}(z_n) \tag{13a}$$
$$\gamma_{il}^{(1)}(z_n) = 2\pi \rho_l \tilde{g}_{il}^{(1)}(z_n) \tag{13b}$$

$$\tilde{g}_{il}(z_n) = \int_{\sigma_{il}}^{\infty} \mathrm{d}r\ \mathrm{e}^{-z_n r}\ r g_{il}(r) \tag{14a}$$

$$\tilde{g}_{il}^{(1)}(z_n) = \int_{\sigma_{il}}^{\infty} \mathrm{d}r\ \mathrm{e}^{-z_n r}\ r^2 g_{il}(r). \tag{14b}$$

Now, we have obtained the functional form of $Q_{ij}(r)$, which is equation (5*a*) with equations (5*b*), (8) and (11). As is seen from equations (12*a*) and (12*b*), $Q_{ij}(r)$ is given apparently by $A_j$, $\beta_j$ and the set $\{D_{ij}^{(n)}, E_{ij}^{(n)}, \gamma_{ij}(z_n), \gamma_{ij}^{(1)}(z_n)\}$. Below, however, we will show that $A_j$ and $\beta_j$ are given explicit expressions in terms of the set. Thus, we see that $Q_{ij}(r)$ is expressed in terms of the set. The set is determined by equations (9*a*) and (9*b*) and the additional conditions obtained from equation (4*b*) for $r > \sigma_{ij}$ which $Q_{ij}(r)$ has to satisfy. The additional equations are obtained by calculating equations (14*a*) and (14*b*) from the equation (4*b*) as follows:

$$2\pi \sum_{l} \tilde{g}_{il}(z_n) \left[\delta_{lj} - \rho_l \tilde{Q}_{lj}(\mathrm{i}z_n)\right] = \left[\left(1 + \frac{z_n\sigma_i}{2}\right) A_j + z_n\beta_j\right] \frac{\mathrm{e}^{-z_n\sigma_{ij}}}{z_n^2} - \sum_{m=1} \frac{z_m \mathrm{e}^{-(z_n+z_m)\sigma_{ij}}}{z_n+z_m} \left\{C_{ij}^{(m)} + F_{ij}^{(m)} \frac{z_m}{z_n+z_m}\left[1+(z_n+z_m)\sigma_{ij}\right]\right\} \tag{15a}$$

$$\begin{aligned}2\pi \sum_{l} \tilde{g}_{il}^{(1)}(z_n) \left[\delta_{lj} - \rho_l \tilde{Q}_{lj}(\mathrm{i}z_n)\right] &= \sum_{m=1} \frac{z_m \mathrm{e}^{-(z_n+z_m)\sigma_{ij}}}{(z_n+z_m)^2}\left[1+(z_n+z_m)\sigma_{ij}\right] \\ &\times \sum_{l} D_{lj}^{(m)}\left[\delta_{li} - \gamma_{il}(z_m)\right] - \sum_{m=1} \frac{z_m \mathrm{e}^{-(z_n+z_m)\sigma_{ij}}}{(z_n+z_m)^3} \sum_{l} E_{lj}^{(m)} \\ &\times \left\{(z_n+z_m)\left[1+(z_n+z_m)\sigma_{ij}\right]\left[\delta_{li} + \gamma_{il}^{(1)}(z_m)\right]\right. \\ &\left. - z_m\left[1+\left[1+(z_n+z_m)\sigma_{ij}\right]^2\right]\left[\delta_{li} - \gamma_{il}(z_m)\right]\right\} \\ &+ z_n \sum_{l} \gamma_{il}(z_n)\tilde{Q}_{lj}^{(1)}(\mathrm{i}z_n) + \frac{\mathrm{e}^{-z_n\sigma_{ij}}}{z_n^3} \\ &\times \left\{\left[1+(1+z_n\sigma_{ij})\left(1+\frac{z_n\sigma_i}{2}\right)\right] A_j + z_n(1+z_n\sigma_{ij})\beta_j\right\}.\end{aligned} \tag{15b}$$

The Laplace transforms $\tilde{Q}_{lj}(\mathrm{i}s)$ and $\tilde{Q}_{lj}^{(1)}(\mathrm{i}s)$ are obtained by integrating equations (10*a*) and (10*b*) with the use of the solution $Q_{lj}(r)$ (equation (5*a*)):

$$\begin{aligned}\mathrm{e}^{-s\lambda_{lj}} \tilde{Q}_{lj}(\mathrm{i}s) &= \frac{1}{2} A_j \Phi_{lj}^{(2,0)}(s,0) + B_j \Phi_{lj}^{(1,0)}(s,0) \\ &+ \sum_{n=1}\left[(C_{lj}^{(n)} + F_{lj}^{(n)})\Phi_{lj}^{(0,0)}(s,z_n) + F_{lj}^{(n)} z_n \Phi_{lj}^{(1,0)}(s,z_n)\right] \\ &+ \sum_{n=1} D_{lj}^{(n)} \frac{\mathrm{e}^{-z_n\lambda_{jl}}}{s+z_n} + \sum_{n=1} E_{lj}^{(n)} \frac{z_n \mathrm{e}^{-z_n\lambda_{jl}}}{(s+z_n)^2}\left[1+(s+z_n)\lambda_{jl}\right]\end{aligned} \tag{16a}$$

$$\begin{aligned}\mathrm{e}^{-s\lambda_{lj}} \tilde{Q}_{lj}^{(1)}(\mathrm{i}s) &= \frac{1}{2} A_j \Phi_{lj}^{(3,1)}(s,0) + B_j \Phi_{lj}^{(2,1)}(s,0) \\ &+ \sum_{n=1}\left[(C_{lj}^{(n)} + F_{lj}^{(n)})\Phi_{lj}^{(1,1)}(s,z_n) + F_{lj}^{(n)} z_n \Phi_{lj}^{(2,1)}(s,z_n)\right] \\ &+ \sum_{n=1} D_{lj}^{(n)} \frac{\mathrm{e}^{-z_n\lambda_{jl}}}{(s+z_n)^2}\left[1+(s+z_n)\lambda_{jl}\right] \\ &+ \sum_{n=1} E_{lj}^{(n)} \frac{z_n \mathrm{e}^{-z_n\lambda_{jl}}}{(s+z_n)^3}\left\{1+\left[1+(s+z_n)\lambda_{jl}\right]^2\right\}\end{aligned} \tag{16b}$$

where $B_j = \beta_j - \frac{1}{2}\sigma_j A_j$,

$$\Phi_{lj}^{(n,m)}(s,z) = \sigma_l \mathrm{e}^{-z\lambda_{jl}} \int_0^1 \mathrm{d}t \left[(\lambda_{jl} + \sigma_l t)^n \mathrm{e}^{-(s+z)\sigma_l t} - \sigma_{lj}^{n-m}(\lambda_{jl} + \sigma_l t)^m \mathrm{e}^{-z\sigma_l - s\sigma_l t}\right]. \qquad (17)$$

Equation (17) is a linear combination of integrals as

$$\phi^{(m)}(x) = \int_0^1 \mathrm{d}t\, t^m \mathrm{e}^{-xt} \qquad m = 0, 1, 2, \cdots, n. \qquad (18)$$

In particular,

$$\Phi_{lj}^{(1,0)}(s,0) = \sigma_l^2 \varphi_1(s\sigma_l)$$
$$\Phi_{lj}^{(2,0)}(s,0) = \sigma_j \sigma_l^2 \varphi_1(s\sigma_l) + 2\sigma_l^3 \psi_1(s\sigma_l)$$
$$\Phi_{lj}^{(2,1)}(s,0) = \frac{1}{2}\sigma_j \sigma_l^2 \varphi_1(s\sigma_l) + \sigma_l^3 \varphi_2(s\sigma_l)$$
$$\Phi_{lj}^{(3,1)}(s,0) = \sigma_j \sigma_l^3 \psi_1(s\sigma_l) + \sigma_l^4 \psi_2(s\sigma_l) + \frac{1}{2}\sigma_j^2 \sigma_l^2 \varphi_1(s\sigma_l) + \sigma_j \sigma_l^3 \varphi_2(s\sigma_l)$$

where

$$\varphi_1(x) = (1 - x - \mathrm{e}^{-x})/x^2$$
$$\varphi_2(x) = \left[2 - 3x/2 + x^2/2 - (2 + x/2)\mathrm{e}^{-x}\right]/x^3$$
$$\psi_1(x) = [1 - x/2 - (1 + x/2)\mathrm{e}^{-x}]/x^3$$
$$\psi_2(x) = \left[6 - 3x + x^2/2 - \left(6 + 3x + x^2/2\right)\mathrm{e}^{-x}\right]/x^4.$$

These functions are used below.

By solving equations (7*a*) and (7*b*), let us give $A_j$ and $\beta_j$ explicit expressions in terms of the set $\{D_{ij}^{(n)}, E_{ij}^{(n)}, \gamma_{ij}(z_n), \gamma_{ij}^{(1)}(z_n)\}$.

Equations (7*c*) for $n = 0$ and 1 are obtained from equations (16*a*) and (16*b*), respectively. Thus, from equations (7*a*), (16*a*), (12*a*) and (12*b*) we have

$$A_j = \frac{2\pi}{\Delta}\left[1 + \frac{1}{2}\zeta_2\beta_j + \sum_{n=1}\left(M_j^{(n)} + N_j^{(n)}\right)\right] \qquad (19)$$

where

$$M_j^{(n)} = \sum_k \rho_k C_k^M(z_n) D_{kj}^{(n)} \mathrm{e}^{-z_n\sigma_{kj}} \qquad (20a)$$

$$N_j^{(n)} = \sum_k \rho_k E_{kj}^{(n)} \mathrm{e}^{-z_n\sigma_{kj}} \left[\frac{z_n\sigma_j}{2} C_k^M(z_n) - C_k^N(z_n)\right] \qquad (20b)$$

with

$$C_k^M(z_n) = \sum_l \mathrm{e}^{z_n\lambda_{kl}} \gamma_{kl}(z_n) z_n \sigma_l^2 \varphi_1(-z_n\sigma_l) - \frac{1 + z_n\sigma_k}{z_n} \qquad (21a)$$

$$C_k^N(z_n) = \sum_l \mathrm{e}^{z_n\lambda_{kl}} \left[\gamma_{kl}(z_n) z_n^2 \sigma_l^3 \varphi_2(-z_n\sigma_l) - \gamma_{kl}^{(1)}(z_n) z_n \sigma_l^2 \varphi_1(-z_n\sigma_l)\right] + \frac{2 + z_n\sigma_k + z_n^2\sigma_k^2}{2z_n}. \qquad (21b)$$

Similarly, from equations (7*b*), (16*b*), (12*a*), (12*b*) and (19) we have

$$\beta_j = \frac{\pi}{\Delta}\sigma_j + \frac{2\pi}{\Delta}\sum_{n=1}\left[\mu_j^{(n)} + \nu_j^{(n)}\right] \qquad (22)$$

where $\Delta = 1 - \pi\zeta_3/6$, $\zeta_m = \sum_l \rho_l \sigma_l^m$,

$$\mu_j^{(n)} = \sum_k D_{kj}^{(n)} \rho_k \mathrm{e}^{-z_n\sigma_{jk}} C_k^{\mu}(z_n) \tag{23a}$$

$$\nu_j^{(n)} = \sum_k E_{kj}^{(n)} \rho_k \mathrm{e}^{-z_n\sigma_{jk}} \left[ C_k^{\nu}(z_n) + \frac{z_n\sigma_j}{2} C_k^{\mu}(z_n) \right] \tag{23b}$$

with

$$C_k^{\mu}(z_n) = \sum_l \gamma_{kl}(z_n) \mathrm{e}^{z_n\sigma_{lk}} z_n \sigma_l^3 \psi_1(z_n\sigma_l) + \frac{1}{z_n^2}\left(1 + \frac{z_n\sigma_k}{2}\right) \tag{24a}$$

$$C_k^{\nu}(z_n) = \sum_l \mathrm{e}^{z_n\sigma_{lk}} \left[ \gamma_{kl}(z_n) \frac{z_n^2\sigma_l^4}{2} \psi_2(z_n\sigma_l) + \gamma_{kl}^{(1)}(z_n) z_n \sigma_l^3 \psi_1(z_n\sigma_l) \right] + \frac{2 + z_n\sigma_k + z_n^2\sigma_k^2/4}{z_n^2}. \tag{24b}$$

In order to obtain equations (19), (20*a*, *b*), (21*a*, *b*), (22), (23*a*, *b*), and (24*a*, *b*) we used $\rho_l\gamma_{lk} = \rho_k\gamma_{kl}$ and $\rho_l\gamma_{lk}^{(1)} = \rho_k\gamma_{kl}^{(1)}$ obtained from equations (13*a*) and (13*b*).

We have discussed the formal solution of the OZ equation with the closure of equations (1) and (3). The solution $Q_{ij}(r)$ is equation (5*a*) with equations (5*b*), (8), and (11) which are expressed in terms of the set: $\{D_{ij}^{(n)}, E_{ij}^{(n)}, \gamma_{ij}(z_n), \gamma_{ij}^{(1)}(z_n)\}$. The set is the physical solutions of the algebraic equations (9*a*), (9*b*), (15*a*) and (15*b*). Since equations (15*a*) and (15*b*) are linear in respect to $D_{ij}^{(n)}$ and $E_{ij}^{(n)}$, we can solve them in terms of $\gamma_{ij}(z_n)$ and $\gamma_{ij}^{(1)}(z_n)$. Therefore, the equations to be solved are equations (9*a*) and (9*b*) for $\gamma_{ij}(z_n)$ and $\gamma_{ij}^{(1)}(z_n)$.

The use of the solution $Q_{ij}(r)$ in equation (4*a*) yields $c_{ij}(r)$ for $r < \sigma_{ij}$, and this and equation (3) give the full structure of the fluid. This route, however, would not be better than the following use of equation (16*a*):

$$\tilde{c}_{ij}(k) \equiv \int \mathrm{d}\boldsymbol{r}\, c_{ij}(r) \mathrm{e}^{\mathrm{i}\boldsymbol{kr}} = \tilde{Q}_{ij}(k) + \tilde{Q}_{ji}(k) - \sum_l \rho_l \tilde{Q}_i l(k) \tilde{Q}_{jl}(-k) \tag{25}$$

where $\tilde{Q}_{ij}(k)$ is defined by equation (10*a*).

For the interaction between $i$ and $j$ species of molecules in the fluid, the MSA closure of equation (3) suggests

$$\phi_{ij}(r) = -k_{\mathrm{B}}T \sum_{n=1} \left( \frac{K_{ij}^{(n)}}{r} + L_{ij}^{(n)} z_n \right) \mathrm{e}^{-z_n r} \qquad \sigma_{ij} < r. \tag{26}$$

The excess interaction energy per unit volume of the fluid, $\varepsilon$, is given as

$$\varepsilon = \sum_i \sum_j 2\pi\rho_i\rho_j \int_0^{\infty} \mathrm{d}r\, r^2 g_{ij}(r)\phi_{ij}(r) = \sum_i \sum_j \rho_i \sum_{n=1} z_n \left( K_{ij}^{(n)} \gamma_{ij}(z_n) + L_{ij}^{(n)} \gamma_{ij}^{(1)}(z_n) \right). \tag{27}$$

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