Analytical structure factor of a two-species polydisperse Percus-Yevick fluid with bimodal Schulz distributed diameters

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An analytic expression is obtained for the static structure factor $S(k)$ of a two-species polydisperse fluid of hard spheres in the Percus-Yevick approximation. The size polydispersity is included via a bimodal Schulz distribution. The derived expression is used to study the effects on $S(k)$ of the size polydispersity in a equiatomic binary mixture of hard-spheres. The main features of the effects are (1) the damping of the oscillations in $S(k)$ beyond its principal peak and larger values of $S(k)$ in the long-wavelength limit, relative to the monodisperse case; and (2) the stronger effect of the species with the larger average particle size. [S1063-651X(99)12903-9]

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I. INTRODUCTION

Colloidal dispersions, as found in nature, are normally multispecies, polydisperse, heterogeneous systems. This work is concerned with a model to study multispecies, polydisperse colloidal fluids. Given the complexity of such systems analytical models have been proposed in the form of polydisperse hard-sphere fluids [1–5], polydisperse charged hard sphere fluids [6,7], and polydisperse hard-sphere Yukawa fluids [8–11].

Griffith, Triolo, and Compere have recently derived an analytic expression for the static structure factor of a single-species polydisperse Percus-Yevick (PY) hard-sphere fluid using the Schulz distribution for the hard-sphere diameters [5]. Here we extend this work to the case of a two-species polydisperse PY hard-sphere fluid. As will be shown below, this extension is made possible by the results of our recent previous work [10,11].

Let us consider a two-species polydisperse fluid in a volume V. We represent each species as I and II, respectively. To consider the polydispersity we assume that each species is itself a multicomponent fluid with an arbitrary number of components. The number of components of species τ ($\tau=\mathrm{I}$ and II) is denoted by $n^{(\tau)}$, and the i component of species τ($\tau=\mathrm{I}$ and II) consists of $V\rho_i^{(\tau)}$ hard-sphere particles with diameter $\sigma_i^{(\tau)}$, where $\rho_i^{(\tau)}$ is the number density of component i of species τ. For such a fluid we can make use of the analytic solution of the Ornstein-Zernike (OZ) equation in the PY approximation [12,13]. In the limit of infinite number of components, the fluid consists of spheres with continuously distributed diamaters.

Below we present the analytic expression for the static structure factor $S(k)$ of the two-species polydisperse fluid of PY hard spheres with bimodal Schulz distributed diameters. Moreover, we also illustrate the effects of polydispersity on $S(k)$. In Sec. II we present a useful, compact form of $S(k)$ of a hard-sphere PY fluid which is obtained by using the method discussed in our recent previous paper [10]. The actual result for $S(k)$ is presented in Sec. III. The effects of polydispersity on $S(k)$ are shown in Sec. IV.

II. STRUCTURE FACTOR OF PY FLUID

We number the components of the fluid under study as

$$i=1,2,\ \ldots\ ,n^{(\mathrm{I})},n^{(\mathrm{I})}+1,\ \ldots\ ,n^{(\mathrm{I})}+n^{(\mathrm{II})}. \tag{2.1}$$

Henceforth we drop, until the end of this section, all the superscripts which indicate species I and II as there is no possibility of confusion.

The partial structure factor of components i and j, $S_{ij}(k)$, is given by the general expression [9]

$$S_{ij}(k)=\delta_{ij}-2\,\mathrm{Re}[\{\hat{\gamma}_s(ik)\}_{ij}], \tag{2.2}$$

where the ij element of the symmetric matrix $\hat{\gamma}_s(s)$ is defined by

$$\{\hat{\gamma}_s(s)\}_{ij}\equiv\frac{2\pi}{s}(c_ic_j)^{1/2}\rho\tilde{g}_{ij}(s). \tag{2.3}$$

In Eq. (2.3) c_i is the number concentration of the i component, ρ denotes the total number density, and

$$\tilde{g}_{ij}(s)\equiv\int_0^\infty dr\, rg_{ij}(r)\mathrm{e}^{-sr}$$

defines the Laplace transform of the partial pair distribution function $g_{ij}(r)$.

The total structure factor $S(k)$ is defined by

$$S(k)=\sum_{ij}\ (c_ic_j)^{1/2}S_{ij}(k). \tag{2.4}$$

Note that the definition of $S(k)$ used here differs from its usual definition, which also involves the scattering form factors. We follow here the definition used, for instance, in Refs. [5] and [10], and reserve the name of scattering intensity $I(k)$ to what other authors call the ''total structure factor.'' We shall not make reference to $I(k)$ in the rest of the paper. Note also that, given Eqs. (2.2) and (2.4), the calculation of $S(k)$ requires a knowledge of $\hat{\gamma}_s(s)$, to which we turn now for the particular case of the hard-sphere fluid in the PY approximation.

In the Baxter formalism, the PY solution of the OZ equation is given by the Baxter function $Q_{ij}(r)$, or its transform $\tilde{Q}_{ij}(is)$, as [13]

$$\tilde{Q}_{ij}(is)=\int_{\lambda_{ji}}^{\infty}dr\,Q_{ij}(r)e^{-sr}$$
$$=e^{s\lambda_{ij}}[\sigma_i^3\psi_1(s\sigma_i)A_j+\sigma_i^2\varphi_1(s\sigma_i)\beta_j], \quad (2.5)$$

where

$$A_j=\frac{2\pi}{\Delta}\left(1+\frac{\pi\zeta_2}{2\Delta}\sigma_j\right), \quad (2.6a)$$

$$\beta_j=\frac{\pi}{\Delta}\sigma_j, \quad (2.6b)$$

$$\psi_1(x)=[1-x/2-(1+x/2)e^{-x}]/x^3,$$
$$\varphi_1(x)=[1-x-e^{-x}]/x^2, \quad (2.6c)$$

$$\zeta_m=\sum_l\rho_l\sigma_l^m,$$

and

$$\Delta=1-\pi\zeta_3/6=1-\eta, \quad (2.6d)$$

where η denotes the packing fraction.

Using the PY solution above, the Laplace transform of the OZ equations reads [14,15],

$$\sum_l 2\pi\tilde{g}_{il}(s)[\delta_{lj}-c_l\rho\tilde{Q}_{lj}(is)]$$
$$=\left\{\left(1+\frac{s\sigma_i}{2}\right)A_j+s\beta_j\right\}\frac{e^{-s\sigma_{ij}}}{s^2}. \quad (2.7)$$

Equation (2.7) can be rewritten, by using Eq. (2.3), in the matrix form

$$\hat{\gamma}_s(s)\hat{Q}(is)=\hat{\Lambda}(s), \quad (2.8)$$

where the ij elements of the matrices $\hat{Q}(is)$ and $\hat{\Lambda}(s)$ are defined by

$$\{\hat{Q}(is)\}_{ij}\equiv\delta_{ij}-(c_ic_j)^{1/2}\rho\tilde{Q}_{ij}(is) \quad (2.9)$$

and

$$\Lambda_{ij}(s)\equiv\frac{(c_ic_j)^{1/2}\rho}{s^3}e^{-s\sigma_{ij}}\left\{\left(1+\frac{s\sigma_i}{2}\right)A_j+s\beta_j\right\}. \quad (2.10)$$

Equation (2.8) may be written as

$$\hat{\gamma}_s(s)=\hat{\Lambda}(s)\hat{R}(s), \quad (2.11)$$

where $\hat{R}(s)$ is defined by

$$\hat{Q}(is)\hat{R}(s)=1. \quad (2.12)$$

Now, with the use of Eqs. (2.6a) and (2.6b), Eq. (2.10) gives

$$\Lambda_{ij}(s)\equiv(c_ic_j)^{1/2}e^{-s\sigma_{ij}}\sum_{n=1,2}w_i^{(n)}(s)\alpha_j^{(n)}, \quad (2.13)$$

where

$$\alpha_j^{(1)}=1, \quad (2.14a)$$

$$\alpha_j^{(2)}=\sigma_j, \quad (2.14b)$$

$$w_i^{(1)}(s)=\frac{2\pi\rho}{\Delta s^3}\left(1+\frac{s\sigma_i}{2}\right), \quad (2.15a)$$

and

$$w_i^{(2)}(s)=\frac{\pi\rho}{\Delta s^3}\left\{s+\frac{\pi\zeta_2}{\Delta}\left(1+\frac{s\sigma_i}{2}\right)\right\}. \quad (2.15b)$$

Moreover, the substitution of Eq. (2.5) into Eq. (2.9), with the use of Eqs. (2.6a) and (2.6b), yields

$$\{\hat{Q}(is)\}_{ij}=\delta_{ij}-(c_ic_j)^{1/2}e^{s\lambda_{ij}}\sum_{n=1,2}Y_i^{(n)}(s)\alpha_j^{(n)}, \quad (2.16)$$

where

$$Y_i^{(1)}(s)=\frac{2\pi\rho}{\Delta}\sigma_i^3\psi_1(s\sigma_i) \quad (2.17a)$$

and

$$Y_i^{(2)}(s)=\frac{\pi\rho}{\Delta}\left\{\frac{\pi\zeta_2}{\Delta}\sigma_i^3\psi_1(s\sigma_i)+\sigma_i^2\varphi_1(s\sigma_i)\right\}. \quad (2.17b)$$

From the definition of $\hat{R}(s)$ [Eq. (2.12)], and the expression for $\{\hat{Q}(is)\}_{ij}$ [Eq. (2.16)], the ij matrix elements of the former are given by (see Refs. [9–11])

$$R_{ij}(s)=\delta_{ij}+(c_ic_j)^{1/2}e^{s\lambda_{ij}}\sum_{n=1,2}Y_i^{(n)}(s)L_j^{(n)}(s) \quad (2.18)$$

with

$$L_j^{(n)}(s)=\sum_{m=1,2}G^{(n,m)}(s)\alpha_j^{(m)}. \quad (2.19)$$

The matrix $\hat{G}(s)$, with matrix elements $G^{(n,m)}(s)$, is defined by the relation

$$\hat{G}(s)[1-\hat{F}(s)]=1, \quad (2.20)$$

with the nm matrix elements of $\hat{F}(s)$ given by

$$F^{(n,m)}(s)=\sum_i c_i\alpha_i^{(n)}Y_i^{(m)}(s). \quad (2.21)$$

We are now in a position to write down $S(k)$. First, substitute Eqs. (2.13) and (2.18) into Eq. (2.11), and also use Eq. (2.19), to obtain

$$\{\hat{\gamma}_s(s)\}_{ij}=(c_ic_j)^{1/2}e^{-s\sigma_{ij}}\sum_n\sum_m w_i^{(n)}(s)G^{(n,m)}(s)\alpha_j^{(m)}. \quad (2.22)$$

Second, substitute Eq. (2.22) into Eq. (2.2) to give

$$S_{ij}(k)=\delta_{ij}-(c_ic_j)^{1/2}2\,\mathrm{Re}\left[e^{-s\sigma_{ij}}\sum_n\sum_m w_i^{(n)}(s)G^{(n,m)}(s)\alpha_j^{(m)}\right]_{s=ik}. \tag{2.23}$$

Finally, from Eqs. (2.4) and (2.23), we obtain

$$S(k)=1-2\,\mathrm{Re}\left[\sum_n\sum_m F_w^{(n)}(s)G^{(n,m)}(s)F_\alpha^{(m)}(s)\right]_{s=ik}, \tag{2.24}$$

where

$$F_w^{(n)}(s)\equiv\sum_i c_ie^{-s\sigma_i/2}w_i^{(n)}(s) \tag{2.25a}$$

and

$$F_\alpha^{(m)}(s)\equiv\sum_i c_ie^{-s\sigma_i/2}\alpha_i^{(m)}. \tag{2.25b}$$

Equation (2.24), together with Eqs. (2.25a) and (2.25b), renders a compact and useful expression for $S(k)$ which we use in Sec. III in a specific context.

III. $S(k)$ FOR TWO SPECIES POLYDISPERSE PY FLUID

We apply the results obtained in Sec. II to study a two-species mixture of hard spheres where both the species are size polydisperse, and the distribution of hard sphere diameters is given by the bimodal Shultz distribution (see below). The application corresponds to a case that $n^{(\mathrm{I})}$ and $n^{(\mathrm{II})}$ are infinite. Write the number of hard spheres in the domain $(\sigma,\sigma+\mathrm{d}\sigma)$ by $V\rho f(\sigma)\mathrm{d}\sigma$; then the distribution of diameters $f(\sigma)$ is given by

$$f(\sigma)=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}f^{(\tau)}(\sigma), \tag{3.1}$$

with $c^{(\tau)}=\rho^{(\tau)}/\rho$. The Shultz distribution function $f^{(\tau)}(\sigma)$ is defined as

$$f^{(\tau)}(\sigma)=\left[\frac{t^{(\tau)}+1}{\sigma_0^{(\tau)}}\right]^{t^{(\tau)}+1}\frac{\sigma^{t^{(\tau)}}}{t^{(\tau)}!}\exp\left(-\left[\frac{t^{(\tau)}+1}{\sigma_0^{(\tau)}}\right]\sigma\right), \tag{3.2}$$

where $\sigma_0^{(\tau)}$ denote the average diameters, and $t^{(\tau)}$ are non-negative integers.

In order to calculate the $F_\omega^{(n)}(s)$, $F_\alpha^{(m)}(s)$, and $F^{(n,m)}(s)$ necessary for the evaluation of $S(k)$ with the above distribution of diameters, the following relations are relevant. Let $A(\sigma_i)$ be an arbitrary function of σ_i; then a quantity like $\Sigma_ic_iA(\sigma_i)$ in Sec. II can be calculated as follows:

$$\sum_i c_iA(\sigma_i)=\int_0^\infty d\sigma\, f(\sigma)A(\sigma)$$
$$=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}\int_0^\infty d\sigma f^{(\tau)}(\sigma)A(\sigma),$$

$$t_m^{(\tau)}\equiv\frac{1}{(\sigma_0^{(\tau)})^m}\int_0^\infty d\sigma\, f^{(\tau)}(\sigma)\sigma^m=\frac{(t^{(\tau)}+m)!}{t^{(\tau)}!(t^{(\tau)}+1)^m}, \tag{3.3}$$

$$f_m^{(\tau)}(a)\equiv\frac{1}{(\sigma_0^{(\tau)})^m}\int_0^\infty d\sigma\, f^{(\tau)}(\sigma)\sigma^m\mathrm{e}^{-a\sigma/\sigma_0^{(\tau)}}$$
$$=t_m^{(\tau)}\left(1+\frac{a}{t^{(\tau)}+1}\right)^{-(t^{(\tau)}+m+1)}. \tag{3.4}$$

Using the above results, from Eq. (2.25b), with Eqs. (2.14a) and (2.14b), we obtain

$$F_\alpha^{(1)}(s)=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}f_0^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right),$$
$$F_\alpha^{(2)}(s)=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}\sigma_0^{(\tau)}f_1^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right), \tag{3.5}$$

whereas Eq. (2.25a), with Eqs. (2.15a) and (2.15b), give

$$F_w^{(1)}=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}\frac{2\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}\frac{1}{(s\sigma_0^{(\tau)})^3}\times\left[f_0^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right)+\frac{s\sigma_0^{(\tau)}}{2}f_1^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right)\right] \tag{3.6a}$$

and

$$F_w^{(2)}=\sum_{\tau=I,II}\frac{c^{(\tau)}}{\sigma_0^{(\tau)}}\frac{\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}\frac{1}{(s\sigma_0^{(\tau)})^3}\left[\left(s\sigma_0^{(\tau)}+\frac{\pi\zeta_2\sigma_0^{(\tau)}}{\Delta}\right)f_0^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right)+\frac{\pi\zeta_2\sigma_0^{(\tau)}}{\Delta}\frac{s\sigma_0^{(\tau)}}{2}f_1^{(\tau)}\left(\frac{s\sigma_0^{(\tau)}}{2}\right)\right]. \tag{3.6b}$$

Moreover, using Eq. (2.21), together with Eqs. (2.14a) and (2.14b) and (2.17a) and (2.17b), we obtain

$$F^{(1,1)}=\sum_{\tau=\mathrm{I,II}}c^{(\tau)}\frac{2\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}f_a^{(\tau)}(s\sigma_0^{(\tau)}), \tag{3.7a}$$

$$F^{(2,1)}=\sum_{\tau=\mathrm{I,II}} c^{(\tau)}\sigma_0^{(\tau)}\frac{2\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}f_b^{(\tau)}(s\sigma_0^{(\tau)}), \tag{3.7b}$$

$$F^{(1,2)}=\sum_{\tau=\mathrm{I,II}} c^{(\tau)}\frac{1}{\sigma_0^{(\tau)}}\left[\left(\frac{\pi}{\Delta}\right)^2\rho\zeta_2(\sigma_0^{(\tau)})^4 f_a^{(\tau)}(s\sigma_0^{(\tau)})+\frac{\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}f_c^{(\tau)}(s\sigma_0^{(\tau)})\right], \tag{3.7c}$$

$$F^{(2,2)}=\sum_{\tau=\mathrm{I,II}} c^{(\tau)}\left[\left(\frac{\pi}{\Delta}\right)^2\rho\zeta_2(\sigma_0^{(\tau)})^4 f_b^{(\tau)}(s\sigma_0^{(\tau)})+\frac{\pi\rho(\sigma_0^{(\tau)})^3}{\Delta}f_d^{(\tau)}(s\sigma_0^{(\tau)})\right], \tag{3.7d}$$

where

$$f_a^{(\tau)}(s\sigma_0^{(\tau)})=\frac{1}{(s\sigma_0^{(\tau)})^3}\left[1-\frac{s\sigma_0^{(\tau)}}{2}-f_0^{(\tau)}(s\sigma_0^{(\tau)})-\frac{s\sigma_0^{(\tau)}}{2}f_1^{(\tau)}(s\sigma_0^{(\tau)})\right], \tag{3.8a}$$

$$f_b^{(\tau)}(s\sigma_0^{(\tau)})=\frac{1}{(s\sigma_0^{(\tau)})^3}\left[1-\frac{s\sigma_0^{(\tau)}}{2}t_2^{(\tau)}-f_1^{(\tau)}(s\sigma_0^{(\tau)})-\frac{s\sigma_0^{(\tau)}}{2}f_2^{(\tau)}(s\sigma_0^{(\tau)})\right], \tag{3.8b}$$

$$f_c^{(\tau)}(s\sigma_0^{(\tau)})=\frac{1}{(s\sigma_0^{(\tau)})^2}[1-s\sigma_0^{(\tau)}-f_0^{(\tau)}(s\sigma_0^{(\tau)})], \tag{3.8c}$$

and

$$f_d^{(\tau)}(s\sigma_0^{(\tau)})=\frac{1}{(s\sigma_0^{(\tau)})^2}[1-s\sigma_0^{(\tau)}t_2^{(\tau)}-f_1^{(\tau)}(s\sigma_0^{(\tau)})]. \tag{3.8d}$$

The use of the results above in Eq. (2.24) yields the analytic expression of $S(k)$ for a polydisperse binary mixture of hard spheres in the PY approximation promised in Sec. I.

IV. RESULTS AND DISCUSSION

We define the size-polydispersity parameter $D_\sigma^{(\tau)}$ as the square of the relative deviation of the Schulz distribution:

$$D_\sigma^{(\tau)}=t_2^{(\tau)}-1. \tag{4.1}$$

The distribution function $f^{(\tau)}(\sigma)$ is specified by two parameters, namely, $\sigma_0^{(\tau)}$ and $t^{(\tau)}$. The latter can also be written by $D_\sigma^{(\tau)}$ as

$$t^{(\tau)}=1/D_\sigma^{(\tau)}-1. \tag{4.2}$$

The analytic expression of $S(k)$ requires the specification of eight parameters, of which only six are independent. Actually we have to specify the concentration $c^{(\tau)}$, the packing fraction $\eta^{(\tau)}$, the average diameter $\sigma_0^{(\tau)}$, and the polydispersity parameter $D_\sigma^{(\tau)}$ for $\tau=$I and II, where

$$\eta^{(\tau)}=\frac{\pi}{6}\rho^{(\tau)}(\sigma_0^{(\tau)})^3 t_3^{(\tau)}. \tag{4.3}$$

These are subject to the conditions that

$$c^{(\mathrm{I})}+c^{(\mathrm{II})}=1$$

and

$$\frac{\eta^{(\mathrm{II})}}{\eta^{(\mathrm{I})}}=\frac{c^{(\mathrm{II})}}{c^{(\mathrm{I})}}\left(\frac{\sigma_0^{(\mathrm{II})}}{\sigma_0^{(\mathrm{I})}}\right)^3\frac{t_3^{(\mathrm{II})}}{t_3^{(\mathrm{I})}}. \tag{4.4}$$

In the equation above, by using Eqs. (3.3) and (4.1), $t_3^{(\tau)}$ may be written as

$$t_3^{(\tau)}=(D_\sigma^{(\tau)}+1)(2D_\sigma^{(\tau)}+1)\quad(\tau=\mathrm{I,II}).$$

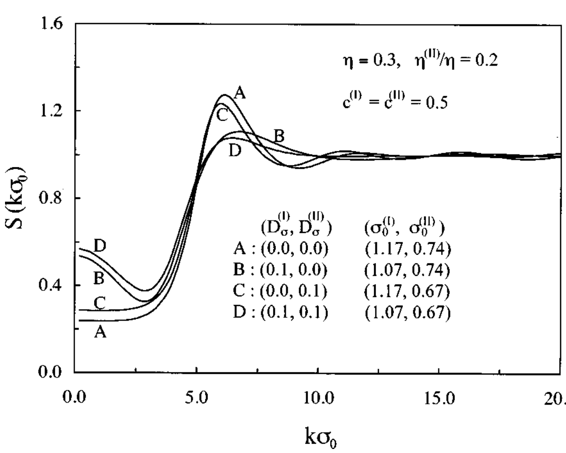

FIG. 1. Polydisperse hard-sphere PY structure factor $S(k)$ with parameters (see text) $c^{(I)}=0.5$, $\eta=0.3$, and $x=0.2$, with the following values for $(D_\sigma^{(\mathrm{I})},D_\sigma^{(\mathrm{II})})$: (0.0,0.0) for case A, (0.1,0.0) for case B, (0.0,0.1) for case C, and (0.1,0.1) for case D. Note that case A denotes the monodisperse results, which are included as a reference.

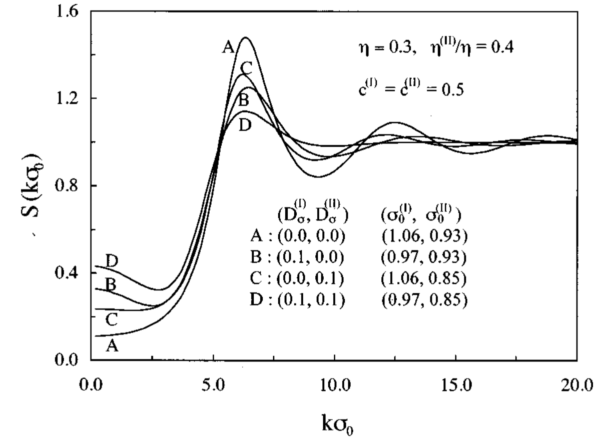

FIG. 2. Same as in Fig. 1, but with $x=0.4$.

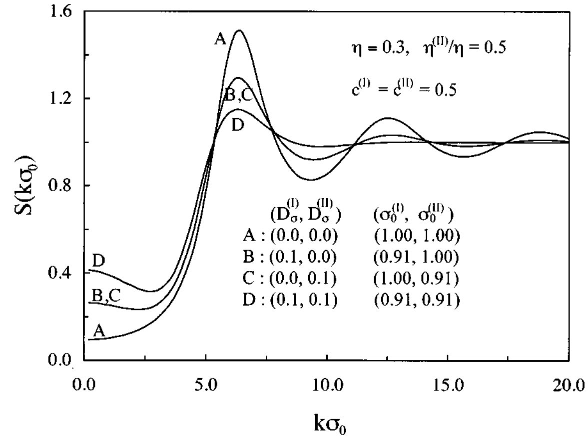

FIG. 3. Same as in Fig. 1, but with $x=0.5$.

For the calculations presented below we choose the following independent parameters: $c^{(\mathrm{I})}$, η, $x\equiv\eta^{(\mathrm{II})}/\eta$, $D_\sigma^{(\mathrm{I})}$, $D_\sigma^{(\mathrm{II})}$, and σ_0, which is chosen as the unit of length and defined by

$$\eta=(\pi/6)\rho\sigma_0^3. \tag{4.5}$$

Moreover, using Eqs. (4.3) and (4.5), we can write

$$\frac{\sigma_0^{(\tau)}}{\sigma_0}=\left(\frac{\eta^{(\tau)}}{\eta c^{(\tau)} t_3^{(\tau)}}\right)^{1/3} \quad (\tau=\mathrm{I,II}), \tag{4.6}$$

which satisfies Eq. (4.4) automatically.

In all our calculations we assume a binary mixture at equiatomic composition, $c^{(\mathrm{I})}=0.5$, and packing fraction $\eta=0.3$. Figs. 1–3 show the dependence of $S(k)$ on $D_\sigma^{(\mathrm{I})}$ and $D_\sigma^{(\mathrm{II})}$ under different packing conditions. Figure 1 shows $S(k)$ for $x=0.2$, Fig. 2 $S(k)$ for $x=0.4$, and Fig. 3 $S(k)$ for $x=0.5$. In all the figures case A represents the monodisperse case, i.e., $D_\sigma^{(\mathrm{I})}=D_\sigma^{(\mathrm{II})}=0.0$. For cases B, C, and D we used values $(D_\sigma^{(\mathrm{I})},D_\sigma^{(\mathrm{II})})=(0.1,0.0)$, $(0.0,0.1)$, and $(0.1,0.1)$, respectively.

All figures show that as the polydispersity increases, the oscillation in $S(k)$ is damped rapidly beyond the principal peak, $S(k)$ increases in the low k region, and the maximum of $S(k)$ decreases. Such a behavior of $S(k)$ is basically the same as in the single-species polydisperse PY fluid [1,2,5]. The polydispersity effect in case B is stronger than that in case C in Figs. 1 and 2, while in Fig. 3 the effects are the same in both cases B and C. When the average particle sizes in case A in each of the figures are considered, the polydispersity of the species with the larger average particle size results in the stronger effect.

Finally, the following comments are in order. First, it is interesting to know how $S(k)$ depends on the moments of the Schulz distribution function. In order to find this out, we expand Eq. (3.4) in terms of a to obtain

$$f_m^{(\tau)}(a)=\sum_{n=0}^{\infty}\frac{(-1)^n a^n}{n!}t_{n+m}^{(\tau)},$$

where $t_{n+m}^{(\tau)}$ is the nondimensional $(n+m)$th moment of the Schulz distribution defined by Eq. (3.3). This expansion shows that for the case of finite momentum transfer, $k\neq 0$, $S(k)$ depends on all the moments of the distribution. However, in the long wavelength limit, $k\to 0$ depends only on the values $m=0$, 1, 2, and 3 of $t_m^{(\tau)}$, since $f_m^{(\tau)}(a)=t_m^{(\tau)}$, a result that is consistent with the thermodynamic properties of a polydisperse hard-sphere PY fluid [16]. Second, the formalism presented above can be extended, in a straightforward fashion but not trivially, to the case of an *M*-species polydisperse fluid with appropriate Schulz distributed diameters.

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[1] A. Vrij, J. Chem. Phys. **69**, 1742 (1978); **71**, 3267 (1979).
[2] P. van Beurten and A. Vrij, J. Chem. Phys. **74**, 2744 (1981).
[3] D. Frenkel, R.J. Vos, C.G. de Kruif, and A. Vrij, J. Chem. Phys. **84**, 4625 (1986).
[4] L. Blum and G. Stell, J. Chem. Phys. **71**, 42 (1979).
[5] W.L. Griffith, R. Triolo, and A.L. Compere, Phys. Rev. A **33**, 2197 (1986).
[6] G. Senatore and L. Blum, J. Phys. Chem. **89**, 2676 (1985).
[7] D. Gazzillo, A. Giacometti, and F. Carsughi, J. Chem. Phys. **107**, 10 141 (1997).
[8] B. D'Aguanno and R. Klein, Phys. Rev. A **46**, 7652 (1992).
[9] M. Ginoza and M. Yasutomi, Mol. Phys. **93**, 399 (1998).
[10] M. Ginoza and M. Yasutomi, Phys. Rev. E **58**, 3329 (1998).
[11] M. Ginoza and M. Yasutomi, Phys. Rev. E **59**, 2060 (1999).
[12] J.L. Lebowitz, Phys. Rev. A **133**, 895 (1964).
[13] R.J. Baxter, J. Phys. Chem. **52**, 4559 (1970).
[14] L. Blum and J.S. Høye, J. Stat. Phys. **19**, 317 (1978).
[15] L. Blum, J. Stat. Phys. **22**, 661 (1980).
[16] J.J. Salacuse and G. Stell, J. Chem. Phys. **77**, 3714 (1982).