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Direct evaluation of the physical characteristics of Yukawa fluids based on a simple approximation for the radial distribution function

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ABSTRACT

We propose a simple analytical approximation for the radial distribution function (RDF) in a single-component Yukawa fluid. The proposed RDF depends on the two input parameters – the non-ideality parameter Γ and the structure (screening) parameter κ, which determine the thermodynamic state of Yukawa systems. We demonstrate that various physical properties can be directly calculated using the proposed RDF. In particular, the internal energy and pressure, the excess entropy in the pair approximation, and the dispersion relation of longitudinal acoustic-like collective excitations are calculated. These theoretical results are compared with the results from molecular dynamics simulations and good overall agreement is observed in the investigated regime of screening and coupling parameters.

The structure of an equilibrium liquid is characterized by the presence of the so-called *short-range order*, which determines significantly various physical properties of the liquid state. The radial distribution function (RDF) $g(r)$ is a structural characteristic, which contains information about relative positions of the particles (molecules) in the system. This function characterizes pair correlations in many-particle systems [1–3] and it appears in the expressions for the basic thermodynamic properties such as the internal energy, pressure, and the pair excess entropy. Last quantity is used to approximate the actual excess entropy (with a varying degree of accuracy depending on the state point) and that stems from the difficulty of computing the higher order terms of the Nettleton-Green expansion [4,5]. In addition to the function $g(r)$, it is also necessary to know the interparticle potential $u(r)$ in order to calculate analytically these thermodynamic properties. The pairwise interaction potential of the form

$$u(r) \sim \frac{1}{r}\exp\left(-\alpha r\right), \tag{1}$$

is known as the Yukawa (screened Coulomb) potential [1–3,6]. Yukawa potential has been proven to be a suitable model to test various approximations in the theory of condensed matter. Its repulsive character mimics interaction between charged particles immersed into a neutralizing medium (like e.g. plasma). For example, in the case of the particles of the same charge Ze, where Z is the charge number and e is the electron charge, the potential becomes

$$u(r) = \frac{(Ze)^2\exp(-r/\lambda_s)}{4\pi\varepsilon_0 r}, \tag{2}$$

where λ_s is the Debye screening length, associated with the presence of neutralizing medium and ε_0 is the vacuum permittivity. The time scale of charge density fluctuations in the system is determined by the plasma frequency

$$\omega_p = \sqrt{\frac{Z^2e^2\rho}{\varepsilon_0 m}}, \tag{3}$$

where ρ is the density of particles in the system and m is the particle mass.

The equilibrium thermodynamics of Yukawa systems is specified by the two key dimensionless parameters: the non-ideality (or coupling) parameter Γ and the structural (or screening) parameter κ [1–3]. The non-ideality parameter

$$\Gamma = \frac{(Ze)^2}{4\pi\varepsilon_0 a k_B T} \tag{4}$$

represents the ratio of the pair interaction energy (excluding the screening effects) evaluated at the mean interparticle distance to the average energy of the thermal motion of the particles. In expression (4), the quantity $a = (3/4\pi\rho)^{1/3}$ is the so-called Wigner-Seitz radius. The

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structural parameter is determined as the ratio of a to the Debye screening length λ_s:

$$\varkappa = \frac{a}{\lambda_s}. \tag{5}$$

In the limit $\varkappa = 0$, the potential (2) reduces to the bare Coulomb potential. In the opposite limit $\varkappa \to \infty$, it approximates the hard-sphere interaction potential. Given the expressions (4) and (5), the formula for the potential can be rewritten in the dimensionless form as follows

$$\beta u(x) = \Gamma \frac{\exp(-\varkappa x)}{x}, \tag{6}$$

where $\beta = 1/(k_B T)$ and $x = r/a$.

Recently, it has been demonstrated [7–9] that the so-called *one-step approximation* for the RDF, of the form

$$g^{(1st)}(x) = \theta(x - x_{\text{eff}}) \tag{7}$$

can be used within the framework of the quasilocalized charge approximation (QLCA) [10,7,11–13] to obtain particularly simple analytical expressions for the dispersion of longitudinal and transverse collective excitations in Yukawa fluids. Here $\theta(x)$ is the Heaviside step-function and x_{eff} is the effective correlational hole radius, which can be determined by requiring that the internal energy or pressure are correctly evaluated from the approximation (7). Note that the approximation (7) is at first glance better appropriate for a rarefied gas of hard spheres (and real gases at sufficiently high temperatures [14]). However, it turns out to be a good approximation also for fluids with soft long-range interactions, when the cumulative contribution from long distances (where $g(x) \simeq 1$) is dominant. Nevertheless, the simplest form (7) does not account for the most prominent signature of the liquid state, which is manifested in the characteristic maximum of the RDF.

A rather accurate parameterization of the Yukawa RDF has been devised in Ref. [15]. The expression is based on combining a Coulomb RDF parameterization with an appropriate effective coupling parameter that maps Yukawa fluids into Coulomb fluids (such an effective coupling parameter has been identified in molecular dynamics (MD) simulations in Ref. [16]). In spite of considerable success of this attempt, the fundamental difference between Coulomb and Yukawa systems renders impossible a perfect match between their static structures through an effective coupling parameter [15].

In this paper we propose a much simpler approximation for RDF, which depends explicitly on Γ and $\varkappa$ and exhibits a maximum. Using this approximation we then calculate the internal energy, pressure, the pair excess entropy, and the dispersion relation of the longitudinal collective mode. The calculation is done for nine $(\Gamma,\ \varkappa)$ state points with $\Gamma = 20,\ 50,\ 100$ and $\varkappa = 1,\ 1.5,\ 2$. Theoretical results are compared with the results of MD simulations and reasonable agreement is documented.

Namely, we suggest the following *two-step approximation* for the function $g(x)$:

$$g^{(2st)}(x) = g_m \theta(x - x_1)\theta(x_2 - x) + \theta(x - x_2). \tag{8}$$

According to Eq. (8), the first maximum of $g(x)$ is modelled by the rectangular shape. Here, the distances x_1 and x_2 determine the left and right boundaries of the first maximum of the RDF and g_m is the magnitude of this maximum. The sketch of this two-step RDF is shown in Fig. 1, where approximation (8) is compared with a real RDF, typical for a liquid-like state. We require that the magnitude of the first peak of the model RDF (8) coincides with that of the real RDF. Other peaks of the RDF corresponding to the second, third, etc. coordination shells are ignored in this two-step approximation. We chose to determine the distance x_1 from the condition $g(x_1) = 0.5$ for a real RDF (see Fig. 1). Then, following Ref. [16], we can express x_1 in terms of Γ and $\varkappa$ as follows:

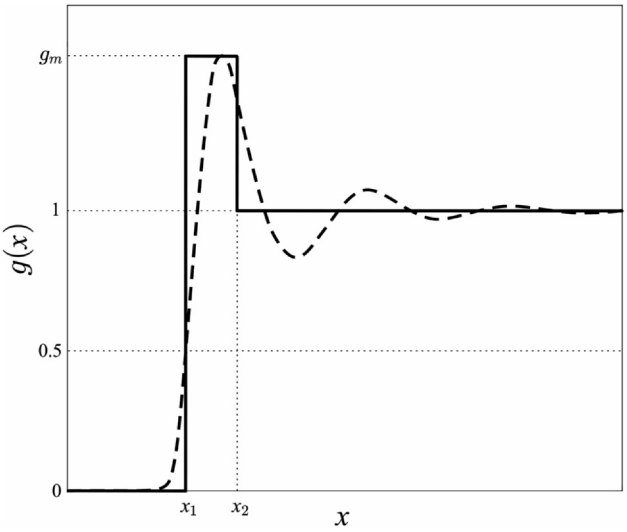

Fig. 1. Radial distribution function. The solid line corresponds to the two-step approximation (8), the dashed line corresponds to a typical RDF of a liquid-like state.

$$x_1^3 = \frac{1}{b_1} \ln \frac{\Gamma - b_2(\varkappa)}{b_3(\varkappa)}, \tag{9}$$

where

$b_1 = 1.575,$

$b_2(\varkappa) = -0.931 + 0.422\varkappa - 0.696\varkappa^2,$

$b_3(\varkappa) = 1.238 - 0.28\varkappa + 0.644\varkappa^2.$

Expression (9) allows us to determine the value of x_1 with good accuracy (the maximum deviation from the simulation results does not exceed 5.6%). To determine the outer radius of the model RDF maximum, x_2, we apply the charge neutrality condition:

$$\int_0^\infty \Big[1 - g(x)\Big] x^2 dx = \frac{1}{3}. \tag{10}$$

This condition is strictly valid for Coulomb systems, but remains a very good approximation for weakly screened Yukawa systems. After simple algebra we get

$$x_2^3 = \frac{x_1^3 g_m - 1}{g_m - 1}. \tag{11}$$

The quantity g_m can be determined from a relation suggested in Ref. [16]:

$$\Gamma = a_1(\varkappa) + a_2(\varkappa) g_m + a_3(\varkappa) g_m^2, \tag{12}$$

where the parameters $a_1(\varkappa)$, $a_2(\varkappa)$ and $a_3(\varkappa)$ satisfy the quadratic polynomial

$$a_\xi(\varkappa) = c_1^{(\xi)} + c_2^{(\xi)}\varkappa + c_3^{(\xi)}\varkappa^2,\ \ \xi = 1,\ 2,\ 3,$$

and the dimensionless coefficients $c_1^{(\xi)}$, $c_2^{(\xi)}$ and $c_3^{(\xi)}$ are constants: $c_1^{(1)} = 22.4$, $c_2^{(1)} = -7.88$ and $c_3^{(1)} = 9.68$;$c_1^{(2)} = -70.09$, $c_2^{(2)} = 20.28$ and $c_3^{(2)} = -32.48$;$c_1^{(3)} = 52.6$, $c_2^{(3)} = -12.71$ and $c_3^{(3)} = 23.73$. From Eq. (12) we obtain

$$g_m = \frac{-a_2(\varkappa) + \sqrt{a_2^2(\varkappa) - 4a_1(\varkappa)a_3(\varkappa) + 4a_3(\varkappa)\Gamma}}{2a_3(\varkappa)}. \tag{13}$$

Expression (13) allows us to determine the value of g_m with good accuracy (the maximum deviation from the simulation results does not exceed 3.3%). Then, the quantity r_2 for a particular $(\Gamma,\ \varkappa)$ state-point can be found by solving the system of Eqs. (9), (11) and (13).

The RDF $g(x)$ enters microscopic equations for various physical properties. For example, the reduced excess internal energy U_{ex} of the Yukawa fluid [17–21] is

$$U_{ex} = \frac{3\Gamma}{2}\int_0^\infty \exp\left(-\kappa x\right) g(x) x\, dx. \tag{14}$$

Using Eq. (8), we immediately obtain from Eq. (14)

$$U_{ex}^{(2st)} = \frac{3}{2}\frac{\Gamma g_m}{\kappa^2}\left(F(x_1) - \frac{g_m - 1}{g_m}F(x_2)\right), \tag{15}$$

where

$$F(x) = (\kappa x + 1)\exp(-\kappa x).$$

The reduced excess internal pressure P_{ex} of Yukawa fluids [17–21] is

$$P_{ex} = \frac{\Gamma}{2}\int_0^\infty (\kappa x + 1)\exp\left(-\kappa x\right) g(x) x dx. \tag{16}$$

Then, using Eq. (8), we find

$$P_{ex}^{(2st)} = \frac{\Gamma g_m}{2\kappa^2}\left(G(x_1) - \frac{g_m - 1}{g_m}G(x_2)\right), \tag{17}$$

where

$$G(x) = ((\kappa x)^2 + 3\kappa x + 3)\exp(-\kappa x).$$

The microscopic expression for the reduced pair excess entropy S_{ex2} in the two-particle approximation does not explicitly contain the interaction potential $u(r)$ and for an isotropic system reads [22]

$$S_{ex2} = -\frac{3}{2}\int_0^\infty \left[g(x)\ln g(x) + 1 - g(x)\right] x^2 dx, \tag{18}$$

where pair excess entropy is expressed in units Nk_B. From Eq. (8) we find

$$S_{ex2}^{(2st)} = -\frac{1}{2(g_m - 1)}[x_1^3 g_m \ln g_m - (g_m \ln g_m + 1 - g_m)]. \tag{19}$$

The interparticle interaction and structural properties determine also the collective particle dynamics of a system [23,3]. The propagation of a collective mode is characterized by the so-called dispersion relation $\omega(k)$, where ω is the frequency of these excitations, and k is the wave number. The exact expression for the dispersion law $\omega_L(k)$ of the longitudinal acoustic-like mode in liquids is not known [24]. Therefore, the dispersion $\omega_L(k)$ is calculated, as a rule, within the framework of some approximations or theoretical models [23]. For soft interactions in the plasma-related context, QLCA has been proven to adequately describe the dispersion relation $\omega_L(k)$, especially in the long-wavelength regime. For Yukawa fluids the QLCA model yields [10,7,11,12]:

$$\omega_L^{(QLCA)}(\bar{k}) = \omega_p\sqrt{\frac{\bar{k}^2}{\kappa^2 + \bar{k}^2} + D(\bar{k})}, \tag{20}$$

where $\bar{k} = ka$,

$$D(\bar{k}) = \int_0^\infty K(\bar{k}, \kappa)\left[1 - g(x)\right]\frac{\exp(-\kappa x)}{x}dx,$$

and

$$K(\bar{k}, \kappa) = 2\left(1 + \kappa x + \frac{(\kappa x)^2}{3}\right)\left(\frac{\sin(\bar{k}x)}{\bar{k}x} + \frac{3\cos(\bar{k}x)}{(\bar{k}x)^2} - \frac{3\sin(\bar{k}x)}{(kr)^3}\right) + \frac{(\kappa x)^2}{3}\left(\frac{\sin(\bar{k}x)}{\bar{k}x} - 1\right).$$

Using the approximation (8), we obtain from Eq. (20)

$$\omega_L^{(QLCA+2st)}(\bar{k}) = \omega_p\sqrt{g_m\left(E(x_1) - \frac{g_m - 1}{g_m}E(x_2)\right)}, \tag{21}$$

where

$$E(x) = \left[\left(\frac{1}{3} - \frac{2\cos(\bar{k}x)}{(\bar{k}x)^2} + \frac{2\sin(\bar{k}x)}{(\bar{k}x)^3}\right)(\kappa x + 1) - \frac{\kappa^2(\bar{k}\cos(\bar{k}x) + \kappa\sin(\bar{k}x))}{\bar{k}(\kappa^2 + \bar{k}^2)}\right]\exp(-\kappa x).$$

As follows from Eqs. (15), (17), (19) and (21), the quantities U_{ex}, P_{ex}, S_{ex2} and the dispersion relation $\omega_L^{(QLCA)}(\bar{k})$ can be directly calculated for a given $(\Gamma,\ \kappa)$ state-point. In addition, these quantities can be evaluated on the basis of microscopic expressions (14), (16), (18) and (20) if the actual RDF is known. The actual RDFs have been calculated using MD simulations [25–29]. These simulations have been performed for a Yukawa system consisting of 64 000 particles in a cubic cell with the periodic boundary conditions. The simulations have been done the *NVT* ensemble. The time step for integration of the equations of motion has been chosen as $t_{step} = 0.01/\omega_p = t_{th}/(200\sqrt{3\Gamma})$, where t_{th} is the time required for the particle to overcome the average interparticle distance $2a$, moving with the thermal velocity $v_{th} = \sqrt{k_B T/m}$.

The results of numerical calculations for the reduced excess internal energy U_{ex}, reduced excess internal pressure P_{ex} and reduced pair excess entropy S_{ex2}, obtained using the approximation (8) along with those obtained using the actual RDFs from MD simulations are presented in Table 1. The relative deviations between theoretical and simulation results are also given in this table (in percent). It is observed that the proposed model provides very accurate estimates of U_{ex} and P_{ex}. The relative deviations from "exact" MD results are usually less than 1%. This is comparable to the accuracy of other recent approximations [18,30,20,19]. More accurate integral equation theory models are

Table 1
Reduced excess internal energy U_{ex}, reduced excess internal pressure P_{ex} and reduced pair excess entropy S_{ex2} of Yukawa fluids evaluated using the actual RDF from MD simulations. The same quantities ($U_{ex}^{(2st)}$, $P_{ex}^{(2st)}$ and $S_{ex2}^{(2st)}$) are calculated from Eqs. (15), (17) and (19). The relative deviations $\delta_{U_{ex}}$, $\delta_{P_{ex}}$ and $\delta_{S_{ex2}}$ between the theoretical and simulation values are given [in percents].

κ	Γ	U_{ex}	$U_{ex}^{(2st)}$	$\delta_{U_{ex}}$	P_{ex}	$P_{ex}^{(2st)}$	$\delta_{P_{ex}}$	S_{ex2}	$S_{ex2}^{(2st)}$	$\delta_{S_{ex2}}$
1	20	21.132	21.213	0.38	24.995	25.267	1.09	−0.63	−0.856	35.87
1	50	51.562	51.681	0.23	61.805	62.363	0.9	−1.136	−1.242	9.33
1	100	101.968	101.72	0.24	122.994	123.717	0.59	−1.839	−1.604	12.78
1.5	20	7.029	7.026	0.04	9.371	9.371	0.0	−0.532	−0.738	38.7
1.5	50	16.495	16.525	0.18	22.647	22.621	0.11	−0.937	−1.038	10.8
1.5	100	31.965	31.924	0.13	44.552	44.334	0.49	−1.478	−1.418	4.06
2	20	2.948	2.902	1.56	4.311	4.274	0.86	−0.434	−0.626	44.2
2	50	6.509	6.534	0.38	9.994	10.003	0.09	−0.75	−0.944	25.9
2	100	12.154	12.215	0.5	19.184	19.151	0.17	−1.164	−1.238	6.4

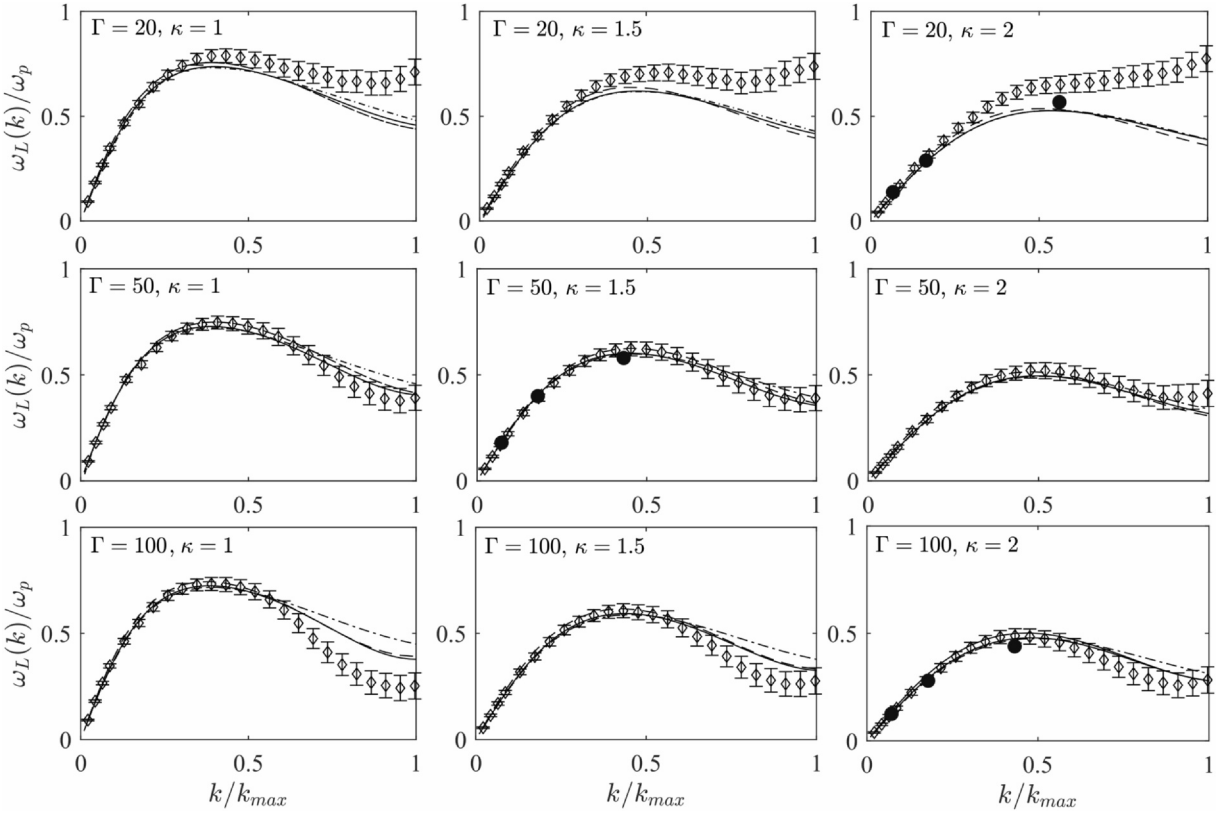

Fig. 2. Reduced frequency $\omega_L(k)/\omega_p$ of the longitudinal collective mode of Yukawa fluids versus the reduced wave vector $k/k_{\max}$ for nine (Γ, κ) state-points. Here, $k_{\max}$ is the position of the main maximum in the static structure factor $S(k)$. The symbols ⋄ represent MD results based on the analysis of the spectral density of longitudinal current fluctuations. Solid curves are the results obtained using Eq. (20) with the actual RDF taken from MD simulations. The dashed curves represent the results of Eq. (21) with the two-step approximation for RDF and the dash-dotted curves correspond to Eq. (20) with the one-step approximation (7) for RDF [7]. The symbols • represent positions of the maxima in the theoretical spectra of the dynamic structure factor from work [3].

available, albeit rather more complicated, such as for instance the variational modified hypernetted chain approach [31], and the isomorph-based empirically modified hypernetted chain approximation [32] and the empirical bridge function approach [33]. The appeal of the present approach is mostly in terms of its simplicity and physical transparency. For the reduced pair excess entropy the agreement is not so good, but increases when the coupling parameter increases. The entropy is more sensitive to the exact shape of the RDF. This is absolutely not surprising: From Eq. (18) we see that in contrast to the energy and pressure, the contribution to the reduced pair excess entropy from the region $g(x) \simeq 1$ is vanishingly small.

The dispersion relations $\omega_L(k)/\omega_p$ of the longitudinal acoustic-like mode are presented in Fig. 2 for the nine (Γ, κ) state-points investigated. Here, the theoretical results obtained using Eq. (21) are compared with the results obtained in the framework of the one-step approximation (7) and MD simulations. In MD simulations, the dispersion relations have been evaluated from the location of the maxima in the spectral density of the longitudinal current (for details see Ref. [34]).In addition, we estimated the positions of the maxima in the theoretical spectra of the dynamic structure factor from work [3], which obtained using the results of the sum-rule approach. As can be seen from Fig. 2, theoretical results reproduce properly the dispersion law for the case of low temperature states with $\Gamma = 100$ and 50. Some discrepancy between the theoretical and MD simulation results is observed only for the case of $\Gamma = 20$ for the wave number $(k_{\max}/2) < k < k_{\max}$. The main reason for disagreement here is the neglect of the kinetic contribution to the dispersion relation (which is reasonable at strong coupling, but is not so appropriate at weaker coupling). It is noteworthy that for all considered (Γ, κ) states, the theoretical model (21) reproduces very well the features of collective vibrational dynamics in the wave number range corresponding to the generalized hydrodynamics: $0 < k < (k_{\max}/2)$. In particular, the theoretical model yields the correct values of the sound velocity: $c_s = \lim_{k\to 0}\omega_L(k)/k$. For example, for the state with $\Gamma = 20$and $\kappa = 1$ we find $c_s/(\omega_p a) = 0.97$, and for the state $\Gamma = 100$ and $\kappa = 2$ we obtain $c_s/(\omega_p a) = 0.44$. Note that the dispersion relations found within the one-step approximation (7) also reproduce the MD simulation results with a very good accuracy at long wavelengths [7,8].

The main results of this work can be summarized as follows. The two-step approximation for the RDF proposed in this work yields thermodynamic quantities such as the reduced excess internal energy, the reduced excess internal pressure, the reduced pair excess entropy and also the dispersion relation $\omega_L(k)$ of the longitudinal collective mode for the Yukawa fluids. In the investigated intermediate screening and moderate-to-strong coupling regimes, energy, pressure, and the long-wavelength dispersion relations are all in excellent agreement with MD simulation results. For these quantities the exact structure of the RDF is not essential and properly designed models can be quite useful. For the reduced pair excess entropy the agreement is not so good, illustrating much stronger sensitivity to the exact shape of the RDF. In addition, it should be noted that the two-step approximation for the RDF proposed in this work for a homogeneous Yukawa fluid can also be generalized for the cases of inhomogeneous fluids, which, for example, can form at the surface of solid walls [35], as well as for the cases of two or more component Yukawa fluids [36]. In these cases, additional information is needed on the position x_1 of the left boundary of the first maximum in the two-step for the RDF approximation, determined from the condition $g(x_1) = 0.5$.

CRediT authorship contribution statement

I.I. Fairushin: Conceptualization, Data curation, Writing - original draft, Investigation. **S.A. Khrapak:** Conceptualization, Methodology, Investigation, Writing - review & editing. **A.V. Mokshin:** Conceptualization, Investigation, Writing - original draft, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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