

Identifying liquidlike and gaslike states of dusty plasmas using isomorph theory

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Equilibrium molecular dynamical simulations are performed to investigate the isomorphic properties of the supercritical liquidlike to gaslike transition in Yukawa fluids. From the calculated Pearson correlation coefficient R , the isomorph theory is always applicable in both the liquidlike and gaslike states of Yukawa systems, so that the Frenkel line is also an isomorph curve, as discussed here. Based on the isomorph theory, it is proposed that the height of the first peak of the radial distribution function $g(r_{fp})$ can be used as a convenient and sensitive criterion of the Frenkel line, which is $g(r_{fp}) = 1.2$ for two-dimensional and $g(r_{fp}) = 1.14$ for three-dimensional Yukawa fluids. This criterion of the Frenkel line from $g(r_{fp})$ well agrees with the widely accepted condition of $C_T/v_{th} = \sqrt{2}$ for the supercritical transition, as well as the equivalent of the reduced instantaneous shear modulus $G_\infty/nk_B T = 2$. The criterion of $g(r_{fp})$ provides a convenient method to identify the supercritical transition of dusty plasmas or Yukawa fluids, which may be extended to other physical systems.

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I. INTRODUCTION

When the pressure and temperature are above the so-called critical point [1], various substances become supercritical fluids, in which the traditional liquid and gas phases cannot be separated anymore by the first-order thermodynamic phase transition [2,3]. Supercritical fluids exhibit the typical features of both liquids and gases, i.e., the higher number densities and larger diffusion constants [4], leading to a variety of applications in extraction, purification, cleaning, and nuclear waste management [5]. Following the Frenkel's liquid description of the local vibrational and long-time hopping motions for individual particles [6,7], the Frenkel line is proposed to discriminate the liquidlike and gaslike states of supercritical fluids [2]. From the previous investigations [2,7], the Frenkel line is identified using different diagnostics, including the heat capacity [8], velocity autocorrelation function [9], variation transition of transport coefficients [10], and reduced instantaneous transverse sound speed [7,11]. However, it is still an open question to identify a universal boundary between the gaslike and liquidlike behaviors in supercritical fluids, since different definitions of the Frenkel line may be not always equivalent to each other, as discussed in Ref. [12].

As a theory developed within the past two decades, the isomorph theory reveals the invariance of the thermodynamic

properties, static structures, and dynamics of various simple liquids [13,14]. The isomorph theory works quite well to the R -systems [15], whose Pearson correlation coefficient R , defined using the equilibrium fluctuations of the virial ΔW and potential energy ΔU as

$$R = \frac{\langle \Delta W \Delta U \rangle}{\sqrt{\langle (\Delta W)^2 \rangle \langle (\Delta U)^2 \rangle}}, \quad (1)$$

exceeds 0.9 [13]. From the previous investigations [15–17], most van der Waals bonded, metallic, and weakly ionic or dipolar systems are all found to be R -systems over a wide range of parameters. On the phase diagram, the “isomorph curves” are defined using the condition of constant excess entropy [15], along which a number of reduced physical quantities are unchanged, called the “isomorph invariants” [13]. These reduced physical quantities are normalized using $n^{-1/d}$, $(m/k_B T)^{1/2} n^{-1/d}$, and $k_B T$ as the units of length, time, and energy, respectively [13]. Here, n is the number density, d is the dimensionality of the system, and m is the mass of each particle. It is worth noting that the systems with the inverse power law (IPL) repulsion, i.e., the so-called IPL systems, are always perfect R -systems [18] because their Pearson correlation coefficient $R = 1$, in which the isomorph theory is strictly obeyed in the entire parameter range [13].

In the previous investigations [19–21], isomorph curves are derived analytically in the IPL, Lennard-Jones (L-J), and Yukawa systems. For the L-J and Yukawa systems, it is found that the static structures, various dynamics, including diffusion procedures and nonequilibrium relaxation processes, are all quasi-invariant along these isomorph curves [11,13–15]. The origin of the excess entropy scaling of transport coefficients discovered by Rosenfeld [22] can be well understood using the isomorph theory [23]. For R -systems, the melting curve, separating the solid and liquid phases, is one typical

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isomorphic curve [13]; as a result, the melting curve of various systems with different interparticle interactions can be analytically derived using the isomorph theory [24,25]. A natural next question is whether the Frenkel line of a R -system to discriminate the liquidlike and gaslike states is also an isomorphic curve, which we have not found from our literature search, as we will study here.

As a model system, dusty plasma [26–41], also termed as complex plasma, refers to partially ionized gas containing micron-sized dust particles of solid matter [34]. Under the typical laboratory conditions, these dust particles are charged to $\sim -10^4 e$ [42,43] in the steady state within microseconds; as a result, the electric potential energy between neighboring dust particles is much greater than their kinetic energy; i.e., they are strongly coupled, exhibiting the typical properties of liquids and solids [44–49]. Due to the shielding of free electrons and ions, the interaction between dust particles in plasma can be accurately modeled using the Yukawa repulsion [50–52]. These highly charged dusts can form into either a single layer two-dimensional (2D) suspension [53–55] or a three-dimensional (3D) suspension [56,57] especially in the microgravity space station condition [58,59], i.e., 2D or 3D dusty plasmas. Each dust particle can be easily observed and tracked during experiments, so that dusty plasma provides a powerful platform to investigate collective dynamics at the individual particle level. The previous study [7] reveals the supercritical nature of dusty plasmas, where the corresponding supercritical transition is investigated using various diagnostics. From Ref. [11], it is demonstrated that the reduced transverse sound speed, i.e., the ratio of the transverse sound speed to the thermal speed C_T/v_{th} , is quasi-invariant for both 2D and 3D dusty plasma or Yukawa fluids. Previous studies [60,61] also suggest that a demarcation line between gaslike and liquidlike regimes for L-J and IPL systems might be an isomorphic curve. Thus, it is reasonable to further investigate the transition between the liquidlike and gaslike states of dusty plasma or Yukawa fluids using the isomorph theory [62], as we will do here.

In this paper, we directly combine the concepts of the isomorph theory and the supercritical transition of dusty plasma or Yukawa fluids for the first time. The rest of the paper is organized as follows. In Sec. II, we introduce our simulation method of 2D and 3D dusty plasmas. In Sec. III, after confirming that the 2D and 3D gaslike Yukawa fluids are still R -systems, we demonstrate that C_T/v_{th} for Yukawa fluids is an isomorphic invariant to a good approximation based on strict derivations for a perfect R -system and the previous Yukawa simulation results [7,11], so that the Frenkel line is regarded as an isomorphic curve for Yukawa fluids. Based on our analyzed results, finally we propose a convenient and sensitive criterion of supercritical liquidlike to gaslike transition, which is the height of the first peak of the radial distribution function (RDF) $g(r_{fp})$. In Sec. IV, we provide a brief summary of our findings reported here.

II. SIMULATION METHOD

To systematically study dynamics of 2D and 3D dusty plasmas under different conditions, we perform equilibrium molecular dynamics (MD) simulations [63,64]. In our MD

simulations, the equation of motion for the particle i is

$$m\ddot{\mathbf{r}}_i = - \sum_{j \neq i} \nabla \phi_{ij}. \quad (2)$$

Here, $\phi_{ij} \equiv Q^2 \exp(-r_{ij}/\lambda_D)/4\pi\epsilon_0 r_{ij}$ is the interparticle Yukawa repulsion [52,65], Q is the charge of each particle, r_{ij} is the distance between particles i and j , and λ_D is the Debye shielding length.

The Yukawa systems are usually characterized by the two dimensionless parameters, which are the coupling parameter $\Gamma \equiv Q^2/(4\pi\epsilon_0 a k_B T)$ and the screening parameter $\kappa \equiv a/\lambda_D$ [66]. Here, T is the kinetic temperature. To normalize the length scale, we use the Wigner-Seitz radius a , which is either $a = (n\pi)^{-1/2}$ for 2D systems [66] or $a = (4\pi n/3)^{-1/3}$ for 3D systems [63,67], where n is the number density of particles. To characterize the timescale, we use either the nominal dusty plasma frequency $\omega_{pd} = (Q^2/2\pi\epsilon_0 m a^3)^{1/2}$ for 2D systems [66] or the dusty plasma frequency $\omega_{pd} = (3Q^2/4\pi\epsilon_0 m a^3)^{1/2}$ for 3D systems [67].

We mainly focus on the fluid phase of 2D and 3D Yukawa systems in our current investigation. In our simulations, we choose the κ values from 0.5 to 5.0, and then specify the value of the relative temperature Γ_m/Γ to be 1.02, 2, 3.33, 10, 20, 100, 200, and 500, where Γ_m is the coupling parameter of the corresponding melting point from Refs. [68,69] and Ref. [70] for 2D and 3D Yukawa systems, respectively. Later, while we search for the transition point, we also specify the value of the relative temperature Γ_m/Γ from 10 to 100 with the step of 0.1, for each κ value.

Here are the details of our simulations. While performing 2D Yukawa simulations, we choose $N = 16\,384$ particles constrained in a rectangular box, with the dimensions of $243.8a \times 211.1a$. However, for 3D Yukawa simulations, we choose $N = 16\,000$ particles constrained in a cubic box, with the dimensions of $40.62a \times 40.62a \times 40.62a$. The periodic boundary conditions [71] are applied in both 2D and 3D simulations. During simulations, we specify the cutoff radius [71] of the interparticle potentials as $22.0a$ for 2D systems and $19.0a$ for 3D systems. In our simulations, we choose the canonical ensemble [18] to use the Nosé-Hoover thermostat to control the temperature of the studied system. The integration time steps are set to be $0.002\omega_{pd}^{-1}$ for both 2D and 3D systems, small enough as justified in Ref. [64]. For each simulation run of both 2D and 3D systems, we first integrate $\geq 3.0 \times 10^6$ steps to reach the steady state, and then collect the positions and velocities of all simulated particles for the next 5.0×10^6 steps, which are used for the data analysis reported here. Note that using a few test runs with larger system sizes, we confirm that our results reported here are all independent of the system size.

III. RESULTS

A. Isomorphic property of Yukawa fluids

We calculate the correlation coefficient R of virial and potential energy equilibrium fluctuations for our simulated 2D and 3D Yukawa fluids over a wide range of parameters, as presented in Fig. 1. From Fig. 1(a), when $\kappa = 0.5$, the values of R for 2D Yukawa fluids at low temperatures, i.e.,

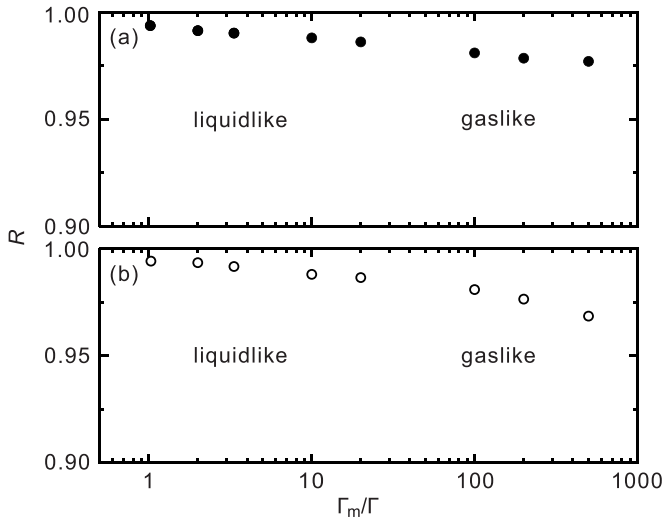


FIG. 1. Calculated correlation coefficient R for 2D [panel (a)] and 3D [panel (b)] Yukawa fluids as a function of the relative temperature Γ_m/Γ for the screening parameter $\kappa = 0.5$. From panel (a), the obtained R is always greater than 0.9 for 2D Yukawa fluids at all studied temperatures, only decreasing $\approx 1.68\%$ from 0.994 to 0.977 when the relative temperature Γ_m/Γ increases from 1.02 to 500. From panel (b) for 3D Yukawa fluids, the obtained R decreases only $\approx 2.51\%$ from 0.994 to 0.969 by increasing the relative temperature Γ_m/Γ from 1.02 to 500.

$1 < \Gamma_m/\Gamma < 10$, are very close to unity, indicating that 2D Yukawa fluids at low temperatures are typical R -systems, well agreeing with the previous studies [20,21]. When the temperature increases further, i.e., $100 < \Gamma_m/\Gamma < 500$, the R value decreases gradually to 0.977, which is still well beyond the typical criterion of R -systems of 0.9 [13]. Similarly to the results of the 2D Yukawa fluids in Fig. 1(a), our obtained R values for 3D Yukawa fluids for $\kappa = 0.5$ are also greater than 0.95 when $1 < \Gamma_m/\Gamma < 500$, as shown in Fig. 1(b), only decreasing $\approx 2.51\%$, from 0.994 to 0.969.

Besides the results presented in Fig. 1, we confirm that our calculated values of the correlation coefficient R for 2D and 3D Yukawa fluids under all other studied conditions are always greater than 0.9, with κ varying from 1.0 to 5.0 and $1 < \Gamma_m/\Gamma < 500$. Our simulation data clearly indicate that, over a wide range of parameters, even in the typical gaslike state, 2D and 3D Yukawa fluids are always typical R -systems. However, for other physical systems including the attractive interaction, such as the L-J system, when the temperature is high enough, the correlation between virial and potential energy equilibrium fluctuations is completely destroyed [15,72]. We speculate that the pure repulsion between particles, just like our studied Yukawa fluids, leads to the typical R -systems at all studied temperatures, well agreeing with the conclusion that all liquids become R -systems when the attraction interactions are ignorable drawn in Ref. [17]. As a result, it is reasonable to exploit the isomorph concept while investigating the dynamics of 2D and 3D Yukawa fluids, no matter whether they are in the liquidlike or gaslike states.

Using the strict derivation of the isomorph theory [62,73], here we first demonstrate that the reduced instantaneous transverse sound speed C_T/v_{th} in the perfect R -system is always

an isomorph invariant. In the previous investigation [11] of other R -systems, i.e., 2D and 3D Yukawa liquidlike fluids, it is found that C_T/v_{th} seems to be an isomorph invariant to a good approximation. From Refs. [74,75], C_T/v_{th} in a fluid can be expressed as

$$\frac{C_T^2}{v_{th}^2} = 1 + \frac{\pi n}{k_B T} \int_0^\infty r^d g(r) [A r \psi''(r) + B \psi'(r)] dr, \quad (3)$$

where d is the dimensionality of the studied system, $\psi(r)$ represents the pairwise potential between two particles in a perfect R -system at a distance of r , $g(r)$ is the RDF. Here, A and B are the coefficients related to the dimensionality d [11] of the studied system, which are $1/8$ and $3/8$ for 2D systems, or $2/15$ and $8/15$ for 3D systems. Note that the primes on $\psi(r)$ represent taking derivatives with respect to its argument r . As derived in detail in the Supplemental Material [76], C_T/v_{th} can be equivalently expressed in the reduced units [15] as

$$\frac{C_T^2}{v_{th}^2} = 1 + \pi \int_0^\infty \tilde{r}^d \tilde{g}(\tilde{r}) [A \tilde{r} \tilde{\psi}''(\tilde{r}) + B \tilde{\psi}'(\tilde{r})] d\tilde{r}. \quad (4)$$

Here, $\tilde{r} \equiv r n^{1/d}$ is the reduced distance and $\tilde{\psi}(\tilde{r}) \equiv \psi(\tilde{r} n^{-1/d})/k_B T$ is the reduced pairwise potential. The primes on $\tilde{\psi}(\tilde{r})$ represent taking derivatives with respect to its argument $\tilde{r}$, while $\tilde{g}(\tilde{r})$ is the reduced RDF that is an important isomorph invariant [13].

Using the expression of the total instantaneous potential energy U of a perfect R -system, we demonstrate that the terms of $\tilde{\psi}'(\tilde{r})$ and $\tilde{\psi}''(\tilde{r})$ in Eq. (4) are both invariants along isomorph curves. The total instantaneous potential energy of a perfect R -system can be expressed as [62,73]

$$U(\mathbf{r}_1, \dots, \mathbf{r}_N) = h(n, S_{ex}) \tilde{\Phi}(\tilde{\mathbf{r}}_1, \dots, \tilde{\mathbf{r}}_N) + f(n), \quad (5)$$

where $(\mathbf{r}_1, \dots, \mathbf{r}_N)$ represents the microscopic configuration of N particles. Here, $\tilde{\Phi}(\tilde{\mathbf{r}}_1, \dots, \tilde{\mathbf{r}}_N)$ is a dimensionless function of the reduced microscopic configuration $(\tilde{\mathbf{r}}_1, \dots, \tilde{\mathbf{r}}_N)$ that is invariant along isomorph curves [73], while $f(n)$ and $h(n, S_{ex})$ are functions of the number density n and excess entropy S_{ex} [62,73]. Meanwhile, the total instantaneous potential energy has a general form of $U(\mathbf{r}_1, \dots, \mathbf{r}_N) = \sum_{i < j} \psi(r_{ij})$. Taking the partial derivative of $U(\mathbf{r}_1, \dots, \mathbf{r}_N)$ with respect to $|\mathbf{r}_\mu - \mathbf{r}_\nu|$, and also combining it with Eq. (5), we confirm the invariance of both $\tilde{\psi}'(\tilde{r})$ and $\tilde{\psi}''(\tilde{r})$ along isomorph curves, as derived in detail in Ref. [76]. Thus, from Eq. (4), we achieve the conclusion that C_T/v_{th} is an isomorph invariant, i.e.,

$$C_T/v_{th} = \text{const}, \quad (6)$$

along isomorph curves for a perfect R -system. For other nonperfect R -systems with the Pearson correlation coefficient R very close to unity, such as the Yukawa fluids studied in this paper, we suggest that the isomorph invariance of C_T/v_{th} is still preserved to a good approximation, as numerically confirmed in Refs. [7,11]. Thus, it is reasonable to regard C_T/v_{th} as an isomorph invariant for Yukawa fluids and use Eq. (6) as the approximate expression of the isomorph curves in Yukawa fluids, as we do in this paper.

For both 2D and 3D Yukawa fluids, the transition curve between the liquidlike and gaslike states is an isomorph

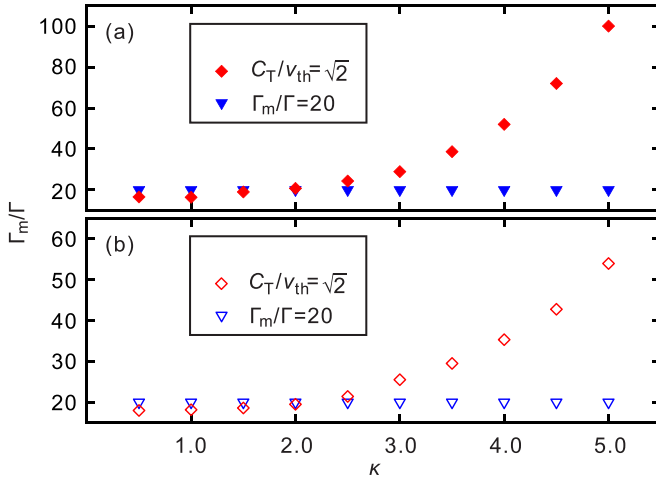


FIG. 2. Obtained data points corresponding to the condition of $C_T/v_{th} = \sqrt{2}$, as well as the data points of $\Gamma_m/\Gamma = 20$, for 2D [panel (a)] and 3D [panel (b)] Yukawa fluids. In panel (a) for 2D Yukawa fluids, the value of Γ_m/Γ under the condition of $C_T/v_{th} = \sqrt{2}$ increases monotonically with κ . A significant discrepancy between the data points of $C_T/v_{th} = \sqrt{2}$ and $\Gamma_m/\Gamma = 20$ occurs when $\kappa \geq 3.0$. In panel (b) for 3D Yukawa fluids, the value of Γ_m/Γ under the condition of $C_T/v_{th} = \sqrt{2}$ also increases monotonically with κ , deviating from $\Gamma_m/\Gamma = 20$ significantly when $\kappa \geq 3.0$.

curve, as we explain in detail next. From the isomorph theory [13], it is widely accepted that the melting curve is parallel to the nearby solid and liquid isomorph curves, so that the melting curve is definitely one isomorph curve [13]. Due to the exact same reason, we believe that the transition curve between the liquidlike and gaslike states of Yukawa fluids must be parallel to nearby liquidlike and gaslike isomorph curves. Thus, the Frenkel line, which is the transition curve to discriminate between the liquidlike and gaslike states, should be also one isomorph curve in Yukawa fluids. In fact, from Refs. [7,11], using a large amount of computer simulated data of Yukawa fluids, it is found that, for both 2D and 3D Yukawa fluids, the transition between liquidlike and gaslike states occurs when $C_T/v_{th} = \sqrt{2}$, where the number 2 comes from the equality of the kinetic contribution and the potential contribution [11]. Combining these findings with our derived Eq. (6) here, we conclude that the Frenkel line for either 2D or 3D Yukawa fluids can be approximately identified using the criterion of $C_T/v_{th} = \sqrt{2}$.

B. Frenkel line as isomorph curve

In Fig. 2, we plot our obtained data points corresponding to the Frenkel line of 2D and 3D Yukawa fluids using the criterion of $C_T/v_{th} = \sqrt{2}$ from our simulation data. To obtain the value of Γ_m/Γ corresponding to the condition of $C_T/v_{th} = \sqrt{2}$, we perform MD simulations by varying the value of Γ_m/Γ from 10 to 100 with the step of 0.1, while specifying κ from 0.5 to 5.0 with a step of 0.5 for both 2D and 3D Yukawa fluids. Using the simulation data of each run, we calculate the value of C_T/v_{th} from the corresponding RDF using Eq. (3). For each specified value of κ of either 2D or 3D Yukawa fluids, we plot the data point of C_T/v_{th} closest

to $\sqrt{2}$ among all of the achieved data, with the confirmed discrepancy of always $<1\%$. Note that we also confirm that, for each κ value, the value of $C_T/v_{th} = \sqrt{2}$ always increases monotonically with the specified Γ value, so that the achieved value of $C_T/v_{th} = \sqrt{2}$ is the only closest value to $\sqrt{2}$ for our current chosen step of 0.1 for Γ_m/Γ .

For our obtained data points corresponding to the Frenkel line as the filled and open diamond symbols shown in Figs. 2(a) and 2(b), the value of Γ_m/Γ increases monotonically with κ . Especially in the range of $3.0 \leq \kappa \leq 5.0$, the increase of Γ_m/Γ is even more drastic, from ≈ 29 to ≈ 100 and from ≈ 25 to ≈ 54 for 2D and 3D Yukawa fluids, respectively. However, from Fig. 2, within the range of $0.5 \leq \kappa \leq 2.5$, for either 2D or 3D Yukawa fluids, the value of Γ_m/Γ is always very close to 20, with only a trivial increment, i.e., $\approx 12.5\%$ for 2D and $\approx 10.0\%$ for 3D Yukawa fluids. In Ref. [7], the supercritical liquidlike to gaslike transitions for 2D and 3D Yukawa fluids are investigated using the diagnostics of the specific heat capacity, instantaneous transverse sound speed, transport coefficients, and others. The results from these diagnostics all indicate that the conditions of the Frenkel lines of both 2D and 3D Yukawa fluids are almost the same as $\Gamma_m/\Gamma = 20$ for the studied parameter range of $\kappa \leq 3.0$ [7], well agreeing with our findings in Fig. 2. However, while the variation range of κ is extended to 5.0, the previously found transition condition of $\Gamma_m/\Gamma = 20$ [7] is not appropriate any more, as further investigated next.

The discrepancy between the data points of $\Gamma_m/\Gamma = 20$ and the Frenkel line of $C_T/v_{th} = \sqrt{2}$ in Fig. 2 clearly indicates that the previous criterion of $\Gamma_m/\Gamma = 20$ [7] for supercritical transition of Yukawa fluids cannot be extended to a wider range of the κ value. As demonstrated above, the Frenkel line of Yukawa fluids is an isomorph curve, while in Ref. [21] the analytical expression of isomorph curves in the low-temperature liquidlike regime is derived as

$$\Gamma_m/\Gamma = \text{const.} \quad (7)$$

Thus, it is important to further confirm whether the expression of Eq. (7) is still valid to describe isomorph curves of Yukawa fluids at higher temperatures, even in the gaslike regime including the supercritical transition point. In fact, a similar problem to describe the melting curve of 2D Yukawa systems has also been addressed in Refs. [68,77], where the “effective coupling parameter” is used to describe the κ dependence of various thermodynamic properties. Different from the κ -independent Eq. (7), the data points corresponding to the isomorph curve of $C_T/v_{th} = \sqrt{2}$ in Fig. 2 clearly show the κ -dependent property, which we will explain in the framework of isomorph theory next.

From the isomorph theory, one can separate the two variables of n and T in the analytical expression of isomorph curves for R -systems at lower temperatures [62,73]. At lower temperatures, the isomorph curves can be expressed using $h(n)/T = \text{const}$ [62], where the variables of n and T are separated into the numerator and denominator, respectively. The function of $h(n)$ is just the melting temperature T_m [21], so that the isomorph curves are $T_m/T = \text{const}$, or equivalently Eq. (7), for Yukawa fluids. However, at higher temperatures, the function of h also depends on the excess entropy S_{ex} , a

thermodynamic quantity related to the combination of both n and T . As a result, the expression of isomorph curves is $h(n, S_{\text{ex}})/T = \text{const}$, or equivalently, $T_{\text{m}}/T = \mathcal{C}T_{\text{m}}/h(n, S_{\text{ex}})$, where $\mathcal{C}$ is a constant; i.e., the two variables of n and T cannot be separated in this expression anymore. Replacing (n, T) using the convention parameters of (κ, Γ) for Yukawa fluids at higher temperatures can be expressed as

$$\Gamma_{\text{m}}/\Gamma = \Gamma_{\text{m}}h(\kappa, S_{\text{ex}})/\mathcal{C}. \quad (8)$$

For our studied Yukawa fluids at higher temperatures, the Frenkel line is definitely an isomorph curve, which can be described using Eq. (8), reasonably leading to the κ -dependent property, especially in the range of $\kappa \geq 3.0$ as shown in Fig. 2.

In a recent paper [78] dedicated to the excess entropy S_{ex} of Yukawa fluids, it has been demonstrated that S_{ex} is a quasiuniversal function of Γ_{m}/Γ at lower temperatures. The condition of constant S_{ex} for the isomorph curves [15] clearly results in Eq. (7). When increasing the temperature and considering a wider range of κ , the excess entropy depends not only on Γ_{m}/Γ , but also explicitly on κ , so that the constant S_{ex} does not result approximately in Eq. (7) any more. The corresponding isomorph curves are then no longer well represented by Eq. (7). Note that for the IPL systems, the isomorph curves can be strictly expressed as $T_{\text{m}}/T = \text{const}$ [13,79], no matter how the temperature varies. For smaller κ values, our studied Yukawa systems are closer to the IPL system [17], so that the corresponding Frenkel lines still can be expressed using Eq. (7) as reported in Ref. [7]. However, for larger κ values, the Yukawa systems deviate further away from the IPL system [17], so that the corresponding Frenkel line should be expressed using Eq. (8) instead, reasonably leading to the discrepancy of two types of data points in Fig. 2.

We further examine the variation of the isomorph invariant $C_{\text{T}}/v_{\text{th}}$ along the condition of $\Gamma_{\text{m}}/\Gamma = 20$ for both 2D and 3D Yukawa fluids, as shown in Fig. 3. Based on our simulation data of 2D and 3D Yukawa fluids at the condition of $\Gamma_{\text{m}}/\Gamma = 20$, we calculate the corresponding data points of $C_{\text{T}}/v_{\text{th}}$ from the RDF using Eq. (3), as plotted in Figs. 3(a) and 3(b), respectively. From Fig. 3, for the condition of $\Gamma_{\text{m}}/\Gamma = 20$, when $0.5 \leq \kappa \leq 2.0$, the difference between the calculated $C_{\text{T}}/v_{\text{th}}$ values and $\sqrt{2}$ is not so significant, only varying from $\leq 4.2\%$ for 2D and $\leq 5.4\%$ for 3D Yukawa fluids, respectively. However, when $\kappa \geq 3.0$, the calculated $C_{\text{T}}/v_{\text{th}}$ values for the condition of $\Gamma_{\text{m}}/\Gamma = 20$ increase monotonically and substantially, which is $\approx 39.5\%$ for 2D and $\approx 26.0\%$ for 3D Yukawa fluids while κ increases to 5.0. The significant variations of the isomorph invariant $C_{\text{T}}/v_{\text{th}}$ for 2D and 3D Yukawa fluids in Fig. 3 further confirm that the previous criterion of $\Gamma_{\text{m}}/\Gamma = 20$ [7] for the Frenkel line cannot be extended to a wider κ range.

C. Transition between liquidlike and gaslike states from $g(r_{\text{fp}})$

Besides the isomorph invariant $C_{\text{T}}/v_{\text{th}}$, here we would like to search for a more convenient and sensitive criterion of the Frenkel line to identify the supercritical liquidlike to gaslike transition in 2D and 3D Yukawa fluids. According to the isomorph theory, the static structure of 2D and 3D Yukawa fluids is quasi-invariant along the isomorph curves [13]. To confirm the invariance of the RDFs, we examine our

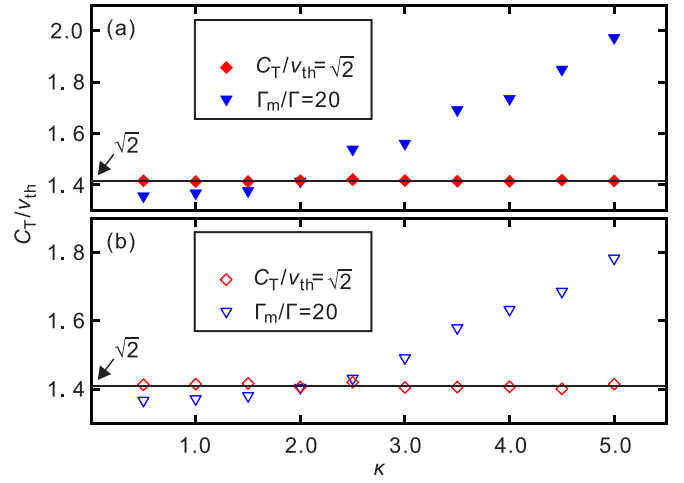


FIG. 3. Calculated $C_{\text{T}}/v_{\text{th}}$ values corresponding to the condition of $\Gamma_{\text{m}}/\Gamma = 20$, as well as the data points of $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$ from Fig. 2. In panel (a) for 2D Yukawa fluids, the value of $C_{\text{T}}/v_{\text{th}}$ under the condition of $\Gamma_{\text{m}}/\Gamma = 20$ deviates from $\sqrt{2}$ with the magnitude of $\approx -4.2\%$ when $\kappa = 0.5$, gradually increasing to $\approx 39.5\%$ when $\kappa = 5.0$. In panel (b) for 3D Yukawa fluids, as κ increases from 0.5 to 5.0, the difference between the calculated values of $C_{\text{T}}/v_{\text{th}}$ and $\sqrt{2}$ increases gradually from -5.4% to 26.1% .

calculated RDFs for 2D and 3D Yukawa fluids for different κ values along the Frenkel line.

We calculate the RDFs of 2D and 3D Yukawa fluids under the conditions corresponding to the data points in Fig. 2, as plotted in Fig. 4. Clearly, for 2D Yukawa fluids under the condition of $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$ as shown in Fig. 4(a), no matter how κ varies between 0.5 and 5.0, the height of the first peak of $g(r)$, labeled as $g(r_{\text{fp}})$, is always 1.2, only fluctuating $\approx 1.7\%$. For 3D Yukawa fluids under the condition of $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$ in Fig. 4(b), the value of $g(r_{\text{fp}})$ is also nearly a constant of 1.14, with the fluctuation of $\approx 1.7\%$ for the varying κ from 0.5 to 5.0. Note that, from Fig. 4, when $0.5 \leq \kappa \leq 5.0$, although the value of $g(r_{\text{fp}})$ is nearly unchanged under the condition of $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$, the location of the first peak decreases monotonically from $1.72a$ to $1.36a$ for 2D, and from $1.67a$ to $1.46a$ for 3D Yukawa fluids, respectively. Meanwhile, for the condition of $\Gamma_{\text{m}}/\Gamma = 20$, both the height and location of the first peak of $g(r)$ vary substantially, as shown in two insets of Fig. 4. Under the condition of $\Gamma_{\text{m}}/\Gamma = 20$ with the increasing κ from 0.5 to 5.0, $g(r_{\text{fp}})$ increases monotonically $\approx 32.2\%$ from 1.15 to 1.52 for 2D and $\approx 18\%$ from 1.11 to 1.31 for 3D Yukawa fluids, respectively. At the same time, the location of the first peak also monotonically shifts from $1.72a$ to $1.51a$ for 2D and from $1.68a$ to $1.48a$ for 3D Yukawa fluids, as shown in two insets of Fig. 4.

As the main result of this paper, we propose a sensitive criterion of the Frenkel line for Yukawa fluids using the height of the first peak of $g(r)$, which is $g(r_{\text{fp}}) = 1.2$ for 2D and $g(r_{\text{fp}}) = 1.14$ for 3D Yukawa fluids, respectively. Clearly, the criterion of $g(r_{\text{fp}}) = 1.2$ for 2D or $g(r_{\text{fp}}) = 1.14$ for 3D Yukawa fluids is equivalent to the well accepted criterion of $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$ [7] over a wide range of κ from 0.5 to 5.0, as shown in Fig. 4. Similar to the isomorph invariant $C_{\text{T}}/v_{\text{th}} = \sqrt{2}$, $g(r_{\text{fp}})$ also exhibits the excellent isomorph

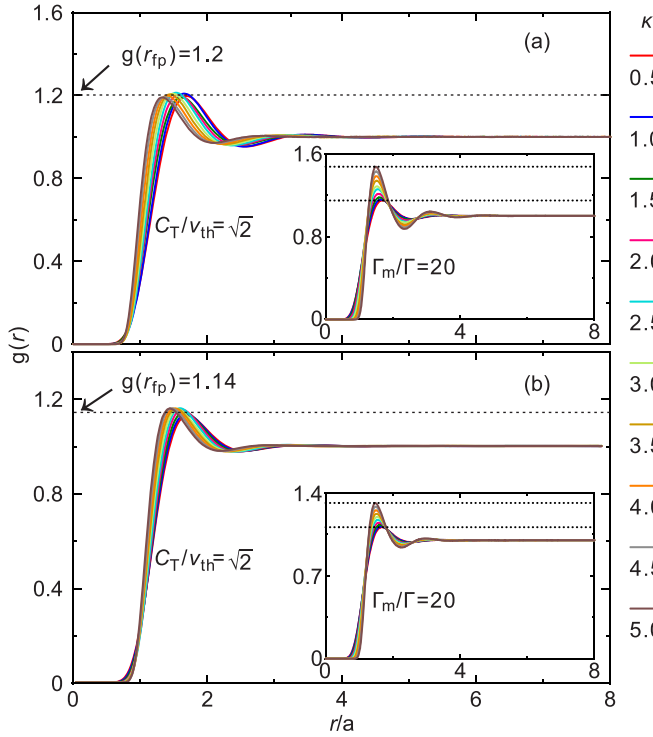


FIG. 4. Calculated RDF results corresponding to all the data points in Fig. 2. The RDFs for 2D Yukawa fluids under the condition of $C_T/v_{th} = \sqrt{2}$ in panel (a) almost collapse together, especially the height of the first peak of RDF is almost unchanged at 1.2 with the fluctuation of only $\approx 1.7\%$. The RDFs for 3D Yukawa fluids under the condition of $C_T/v_{th} = \sqrt{2}$ in panel (b) also approximately collapse into a single curve, with the nearly same height of the first peak of 1.14 with only $\approx 1.7\%$ variation. Our calculated RDFs under the condition of $\Gamma_m/\Gamma = 20$ are presented in the two insets. Clearly, for different κ values, the RDFs under $\Gamma_m/\Gamma = 20$ do not collapse together for either 2D or 3D Yukawa fluids, especially the height of the first peak of $g(r)$ changes substantially.

invariance; i.e., the criterion of $g(r_{ip}) = 1.2$ for 2D or 1.14 for 3D Yukawa fluids is able to accurately identify the isomorphous curve of $C_T/v_{th} = \sqrt{2}$, or the Frenkel line. On the other hand, compared with the instantaneous transverse sound speed C_T , the RDF $g(r)$ can be obtained much more easily, from both experiments [30,80] and simulations [81]. As a result, purely based on the height of the first peak of $g(r)$, combined with the convenient and sensitive criterion of $g(r_{ip}) = 1.2$ for 2D and 1.14 for 3D systems, the supercritical transition between the liquidlike and gaslike dusty plasmas or Yukawa fluids can be clearly identified. However, the previously suggested criterion of $\Gamma_m/\Gamma = 20$ of the Frenkel line for 2D and 3D Yukawa fluids in Ref. [7] is only accurate for smaller κ values, which cannot be generalized to a wider parameter range studied here. Note that, for an IPL system, i.e., a perfect R -system, the $g(r)$ result should be exactly unchanged along isomorphous curves [13]. As clearly plotted in Fig. 4, under the condition of $C_T/v_{th} = \sqrt{2}$, the slight shifts of both the location and height of the first peak of $g(r)$ do exist, which we attribute to the fact that Yukawa systems are not perfect R -systems [13,20].

To further confirm the proposed criterion of $g(r_{ip}) = 1.2$ for 2D and $g(r_{ip}) = 1.14$ for 3D Yukawa fluids for the

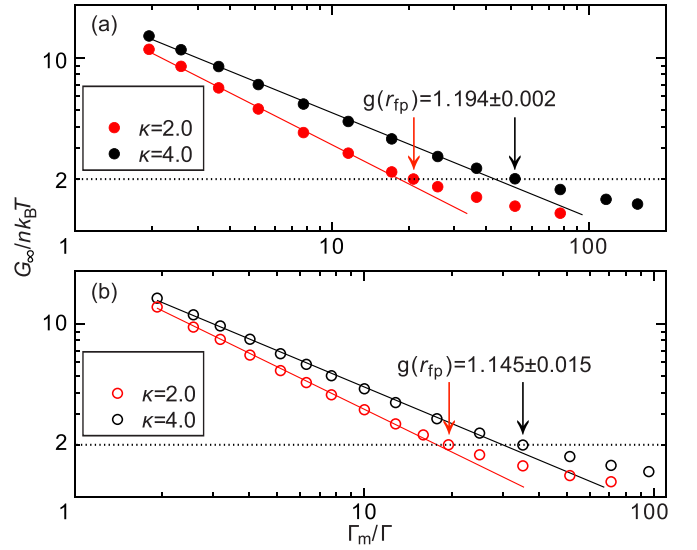


FIG. 5. Variation of the reduced instantaneous shear modulus $G_\infty/nk_B T$ with the relative temperature Γ_m/Γ of 2D [panel (a)] and 3D [panel (b)] Yukawa fluids for the two typical κ values of 2.0 and 4.0. For each panel, the solid lines are the corresponding linear least-squares fittings to all data points within the range of $G_\infty/nk_B T > 2$ in the logarithmic scale for the two axes. Clearly, for the range of $G_\infty/nk_B T > 2$, $G_\infty/nk_B T$ always perfectly decreases with Γ_m/Γ in a power law. However, in the range of $G_\infty/nk_B T < 2$, $G_\infty/nk_B T$ decreases with Γ_m/Γ differently, as indicated by the deviation of the data points from the fitting lines there. When $G_\infty/nk_B T = 2$ for all studied κ values, as two shown here, the corresponding values of $g(r_{ip})$ are very close to our proposed criterion of the supercritical transition, i.e., 1.2 for 2D and 1.14 for 3D Yukawa fluids, as marked there.

supercritical transition between liquidlike and gaslike states, we calculate the corresponding reduced instantaneous shear modulus $G_\infty/nk_B T$ results, as presented in Fig. 5. To calculate $G_\infty/nk_B T$, we follow Ref. [75] to use the formula

$$\frac{G_\infty}{nk_B T} = 1 + \frac{\pi n}{k_B T} \int_0^\infty r^d g(r) [A r \psi''(r) + B \psi'(r)] dr, \quad (9)$$

where we achieve $g(r)$ from the Ornstein-Zernike equation [79] of $g(r) = 1 + c(r) + n \int c(r') [g(|\mathbf{r} - \mathbf{r}'|) - 1] d\mathbf{r}'$ and the closure relation of $g(r) = \exp(-\psi(r)/k_B T) + g(r) - c(r) + b(r) - 1$ [79], as well as the bridge function $b(r)$ [82–84]. Note that we confirm that our achieved $g(r)$ results here well agree with the calculated $g(r)$ results from our simulation data to derive C_T above. For 2D and 3D Yukawa fluids, the $G_\infty/nk_B T$ value decreases monotonically with Γ_m/Γ for all studied κ values, as two typical values of $\kappa = 2.0$ and 4.0 presented in Fig. 5. We apply the linear least-squares fitting to all the calculated $G_\infty/nk_B T$ data points for each κ value in the logarithmic scales of the two axes, resulting in a nearly perfect power law dependence of $G_\infty/nk_B T$ on Γ_m/Γ for the range of $G_\infty/nk_B T > 2$, as the solid lines shown in Fig. 5. However, when $G_\infty/nk_B T < 2$, the decreasing trend of $G_\infty/nk_B T$ with Γ_m/Γ varies substantially, as the corresponding data points deviating from the corresponding solid line in Fig. 5.

The transition of the decreasing trend of $G_\infty/nk_B T$ with Γ_m/Γ in Fig. 5 just corresponds to the Frenkel line condition

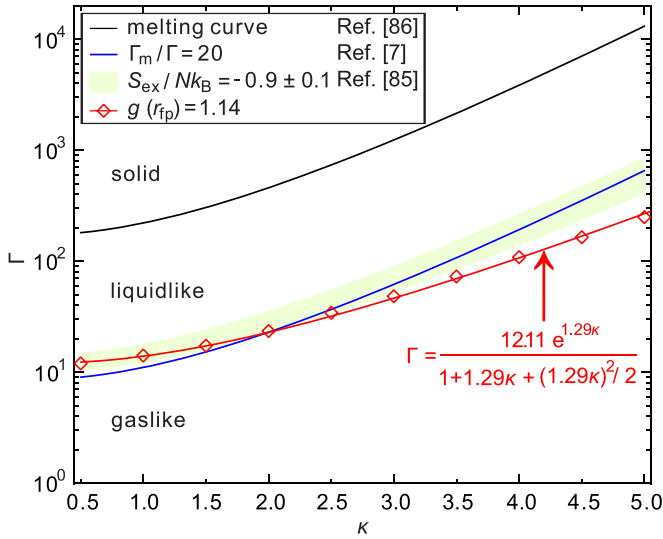


FIG. 6. Obtained phase diagram for 3D Yukawa systems. The results for the liquidlike to gaslike supercritical transition from Refs. [7,85] and our current results, as well as the melting curve from Ref. [86], are all presented here. Clearly, our current results of the supercritical transition from $g(r_{ip}) = 1.14$ are well consistent with those using the criteria of $\Gamma_m/\Gamma = 20$ [7] and $S_{ex}/Nk_B = -0.9 \pm 0.1$ [85]. We also find that our obtained transition results from $g(r_{ip}) = 1.14$ can be well fit to Eq. (10), as the curve across the data points here.

described above. Comparing Eqs. (3) and (9), the dependence of $G_\infty/nk_B T$ on Γ_m/Γ changes at $G_\infty/nk_B T = 2$, which is, in principle, equivalent to the criterion of $C_T/v_{th} = \sqrt{2}$ [11,75], just corresponding to the Frenkel line. In Fig. 5, we also calculate $g(r)$ results corresponding to the condition of $G_\infty/nk_B T = 2$, as the data points on the dashed lines. We confirm that, for 2D Yukawa fluids, the value of $g(r_{ip})$ is either 1.192 for $\kappa = 2.0$ or 1.196 for $\kappa = 4.0$ as shown in Fig. 5(a), very close to the proposed criterion of $g(r_{ip}) = 1.2$ here. In Fig. 5(b) for 3D Yukawa fluids, we obtain $g(r_{ip}) = 1.13$ for $\kappa = 2.0$ and $g(r_{ip}) = 1.16$ for $\kappa = 4.0$, well agreeing with our proposed criterion of $g(r_{ip}) = 1.14$. Note that, from our interpretation, when $G_\infty/nk_B T = 2$, the slight variation of $g(r_{ip})$ for different κ values comes from the fact that our studied Yukawa fluids are not perfect R -systems, as discussed previously. From all above, we confirm that our proposed criterion of $g(r_{ip}) = 1.2$ for 2D and $g(r_{ip}) = 1.14$ for 3D Yukawa fluids is effective and convenient to identify the supercritical transition between the liquidlike and gaslike states.

By plotting our current results with the previous results in Refs. [7,85,86], we obtain the phase diagram of 3D Yukawa systems containing the solid, liquidlike, and gaslike states, as presented in Fig. 6. To search for the conditions corresponding to our proposed $g(r_{ip}) = 1.14$ in Fig. 6, we vary the κ value with the step of 0.5 and scan the Γ value with the step of 1.0 to calculate the corresponding $g(r)$ using the Ornstein-Zernike equation and the bridge function [82–84]. For the current setting step of Γ , when the difference of the calculated $g(r_{ip})$ and 1.14 reaches its smallest value, which is always $< 1.5\%$, we just plot the corresponding values of Γ and κ , i.e., the data points in Fig. 6. We also find that these obtained data points

can be well fit using the expression

$$\Gamma = \alpha \frac{e^{\beta\kappa}}{1 + \beta\kappa + (\beta\kappa)^2/2}, \quad (10)$$

where $\alpha = 12.11$ and $\beta = 1.29$, as also plotted there. In Fig. 6, besides our results described above, the supercritical transition results of $\Gamma_m/\Gamma = 20$ [7] come from the diagnostics of heat capacity, velocity autocorrelation function, and transport coefficients, while the results of $S_{ex}/Nk_B = -0.9 \pm 0.1$ [85] from the excess entropy (which we calculate for 3D Yukawa systems using the integral of the excess energy expression [87]) are proposed from the validity of the Stokes-Einstein relation. In addition, the plotted melting curve in Fig. 6 is derived from the isomorph theory [86].

From Fig. 6, our reported results for the liquidlike to gaslike supercritical transition based on the criterion of $g(r_{ip}) = 1.14$ are well consistent with the previous investigations [7,85]. Clearly, in Fig. 6, when $0.5 \leq \kappa \leq 2.5$, the Frenkel lines identified by the three criteria of $g(r_{ip}) = 1.14$, $\Gamma_m/\Gamma = 20$ [7], and $S_{ex}/Nk_B = -0.9 \pm 0.1$ [85] almost overlap with each other. This consistency clearly indicate that our proposed criterion of the Frenkel line for Yukawa fluids is reliable. When $\kappa > 2.5$, the discrepancy between the Frenkel lines identified by the criteria of $g(r_{ip}) = 1.14$ and $\Gamma_m/\Gamma = 20$ increases gradually with the κ value. As discussed above, this discrepancy comes from the fact that our studied 3D Yukawa systems deviate further away from the perfect R -system for larger κ values.

IV. SUMMARY

In summary, we perform MD simulations of 2D and 3D Yukawa fluids to study the isomorphic properties of the supercritical liquidlike to gaslike transition in dusty plasmas. By calculating the Pearson correlation coefficient R of the equilibrium fluctuations of virial and potential energy, we confirm that isomorph theory is still applicable to the gaslike state of Yukawa systems. By demonstrating that C_T/v_{th} is an isomorph invariant to a good approximation for Yukawa fluids, we conclude that the transition line between the liquidlike and gaslike states, i.e., the Frenkel line, for Yukawa fluids is an isomorphic curve. From our analyzed results, we propose a sensitive criterion of the Frenkel line for Yukawa fluids using the height of the first peak of $g(r)$, which is $g(r_{ip}) = 1.2$ for 2D and $g(r_{ip}) = 1.14$ for 3D Yukawa fluids. This criterion is very convenient to identify the supercritical transition for dusty plasmas or Yukawa fluids, because $g(r)$ can be easily and accurately achieved in both experiments and simulations. We also confirm that this criterion of the Frenkel line for dusty plasma or Yukawa fluids using $g(r_{ip})$ well agrees with the widely accepted condition of $C_T/v_{th} = \sqrt{2}$ for the supercritical transition, as well as the equivalent condition of $G_\infty/nk_B T = 2$. We finally confirm that our results for the supercritical transition are well consistent with the previous investigations [7,85] after preparing the phase diagram of 3D Yukawa systems. Our presented results connect the isomorph theory with the supercritical dynamical transition in Yukawa fluids for the first time, which may be further extended to other physical systems in future. In future, probably other quasi-invariants may be also found in R -systems,

including Yukawa fluids, as the criteria for the location of the Frenkel line.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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