Dynamics of strongly correlated and strongly inhomogeneous plasmas

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Kinetic and fluid equations are derived for the dynamics of classical inhomogeneous trapped plasmas in the strong coupling regime. The starting point is an extended Singwi-Tosi-Land-Sjölander (STLS) ansatz for the dynamic correlation function, which is allowed to depend on time and both particle coordinates separately. The time evolution of the correlation function is determined from the second equation of the Bogolyubov-Born-Green-Kirkwood-Yvon hierarchy. We study the equations in the linear limit and derive a nonlocal equation for the fluid displacement field. Comparisons to first-principles molecular dynamics simulations reveal an excellent quality of our approach thereby overcoming the limitations of the broadly used STLS scheme.

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Strongly correlated plasmas (SCPs) [1] constitute an important field of current research due to their relevance for shock-compressed matter [2], planetary and stellar interiors [3], or even the exotic quark-gluon plasma [4]. In these systems, the mean interaction energy (often by orders of magnitude) exceeds the kinetic energy \( \gamma = \langle v \rangle / \langle E_{\text{kin}} \rangle \gg 1 \), giving rise to a variety of effects such as the emergence of liquidlike properties, crystallization, or anomalous transport, which are well established experimentally [1,3]. At the same time, strong correlations make a theoretical description very challenging since standard microscopic approaches from high-temperature plasmas fail. Nevertheless, in recent decades a number of concepts have been put forward that have been successful in describing selected properties of SCPs. In particular, their linear response behavior is—in part—well reproduced by the Singwi-Tosi-Land-Sjölander (STLS) scheme [5,6], the quasilocalized charge approximation (QLCA) [7], approximations for the dynamic local field corrections [8], or generalized linear response behavior is—in part—well reproduced by the Singwi-Tosi-Land-Sjölander (STLS) scheme [5,6], the quasilocalized charge approximation (QLCA) [7], approximations for the dynamic local field corrections [8], or generalized hydrodynamics [9] and kinetic theories [10] that are based on the memory function approach.

While these approaches often yield excellent results for spatially uniform plasmas, they are not applicable to systems with strong density gradients. However, in recent years an increasing number of experimental systems, including dusty plasmas [11,11], confined ions [12], ultracold neutral [13], or laser-produced and inertial confinement fusion plasmas [14], are faced with strong density inhomogeneities caused by interfaces, boundaries, and finite system sizes. Of particular interest is their collective excitation spectrum [15], which is a sensitive diagnostics of the microscopic structure and thermodynamic properties [16], just as the familiar optical line spectrum reflects the properties of the correlated electrons in atoms and molecules. In spherically confined systems, a prominent role is played by the breathing mode—the radial expansion and contraction of all particles—which is sensitive to the interparticle potential and the coupling strength and easily excited and probed experimentally in such diverse systems as dusty plasmas, trapped ions or cold atoms, and quantum dots. In this Rapid Communication, we will, therefore, use the breathing frequency as a stringent test for the accuracy of theoretical models for strongly coupled, inhomogeneous classical plasmas using new molecular dynamics (MD) data as a reference.

While there has been extensive work on particular collective modes and interactions [17], various approximation schemes\(^1\), or limiting cases\(^2\) presently no theory that is broadly applicable to the dynamic properties of strongly correlated and strongly inhomogeneous plasmas is available. It is the purpose of this Rapid Communication to fill this gap. We present a systematic microscopic theory based on the fundamental Bogolyubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy for the reduced nonequilibrium distribution functions. We decouple the hierarchy by modifying the principle of weakening of initial correlations of Bogolyubov [26], which he derived for the case \( \Gamma < 1 \) such that it is applicable at strong coupling, \( \Gamma \gg 1 \). From this we obtain a general kinetic theory, fluid equations, and equations for the collective mode spectrum. Finally, a rigorous test is performed for the breathing frequency of Yukawa clusters at finite temperature. The excellent agreement with MD simulations demonstrates that our theory overcomes the limitations of the broadly used STLS approach.

**Kinetic theory.** We consider \( N \) identical particles with mass \( m \) interacting via a pair potential \( V(\mathbf{r}) \) in a confinement potential \( V(\mathbf{r}) \) (\( d \) dimensions). Their dynamics is completely described by the BBGKY hierarchy for the reduced \( s \)-particle distribution functions \( f, f^{(2)}, f^{(3)}, \ldots \). The first two equations

\(^1\) MD simulations showed that mean-field theories [19,20] yield reasonable results for the low-order modes in Coulomb systems [21,22]. However, they cannot describe torsional modes [22,23] and become increasingly inaccurate for short-ranged Yukawa systems and modes of higher order [24]. The QLCA has been extended to inhomogeneous systems by Lee and Kalman [25].

\(^2\) The approaches above are restricted to particular excitations (scaling ansatz), weak coupling (cold and warm fluid), low-order modes (viscoelastic), or they do not account for the thermal particle motion (QLCA, harmonic approximation). More advanced methods for uniform systems exist [18] (generalized hydrodynamics, method of moments) but do not account for strong density gradients or finite system sizes.
of the hierarchy are [27]

\[ \partial_t + \hat{Q}_1 f(1,t) = -\int F_{12} \cdot \nabla_{\vec{r}_1} f^{(2)}(1,2,t) d^2 \vec{r}_2, \]  

\[ \partial_t + \hat{Q}_1 + \hat{Q}_2 + F_{12} \cdot (\nabla_{\vec{r}_1} - \nabla_{\vec{r}_2}) f^{(2)}(1,2,t) = -\sum_{i=1}^2 \int F_{i3} \cdot \nabla_{\vec{r}_i} f^{(3)}(1,2,3,t) d^3, \]

where \( \hat{Q}_i = (p_i/m) \cdot \nabla_{\vec{r}_i} + F^{ext}_{i} \cdot \nabla_{\vec{r}_i}, \) \( F_{ij} = -\nabla_{\vec{r}_i} v(r_{ij}), \) and \( r_{ij} = |r_i - r_j|, \) and the external force \( F^{ext}_{i}(r_i,t) \) includes the confinement potential \( V. \) The numbers 1, 2, 3 comprise positions and momenta, \( s = \{\vec{r}_i, \vec{p}_i\}. \)

To derive a closed kinetic equation for \( f, \) Bogolyubov introduced the concept of a time-scale hierarchy [26],

\[ \tau_{cor} \ll \tau_{rel} \ll \tau_{hyd}, \]

indicating that macroscopic processes (hydrodynamics, time scale \( \tau_{hyd} \)) are slow compared to the momentum relaxation of \( f (\tau_{rel}), \) which, in turn, is much slower than the equilibration of the higher-order distributions \( \tau_{cor}. \) This allows him to simplify the time dependence of \( f^{(2)} \) according to \( f^{(2)}(f(t); t) \rightarrow f^{(2), EQ}(f(t)), \) i.e., \( f^{(2)} \) has already relaxed to its equilibrium functional form but is still time dependent via the nonequilibrium distribution \( f(t). \) This directly leads to kinetic equations for weakly nonideal systems such as the Boltzmann equation, valid for \( \tau > \tau_{cor}. \)

However, in a strongly correlated system, the inequality (2) is violated, in particular, \( f \) and \( f^{(2)} \) relax on similar time scales, e.g., [28,29]. The solution to this problem is offered by the nature of the dynamics of an SCP, which comprises two main time scales. First, a fast time of small-scale momentum relaxation processes, \( \tau_{cor}, \) during which the particles settle close to the minima of the total potential created by external fields and all other particles. Since \( \Gamma \gg 1 \) (low \( T, \)) the subsequent dynamics in the second stage resembles that of an elastic medium, which is probed in experiments measuring the collective mode spectrum and which is, therefore, in the focus of our paper.

Although this dynamics is much slower than that of the first stage, it is very complicated since one- and two-particle quantities are strongly coupled and require a self-consistent treatment. Our modified functional hypothesis for a strongly correlated system [extended STLS (ESTLS)] therefore reads

\[ f^{(2)}(t) = f^{(2), EQ}(f(t), h(t)), \]

\[ f^{(2)}(1,2,t) = f(1,t)f(2,t)[1 + h(r_1,r_2,t)], \]

where we have introduced the pair correlation function (PCF) \( h(r_1,r_2,t). \) The only approximation made is that of momentum independence of \( h. \) This is justified for \( t \gtrsim \tau_{cor} \) when \( f^{(2)} \) has already undergone the relaxation towards the equilibrium functional form. At the same time \( f^{(2)} \) is still time dependent via the two nonequilibrium functions \( f(t) \) and \( h(t), \)

\[ \partial_t f + \frac{p}{m} \cdot \nabla f + (F^{ext} - \nabla V^{mf} + F^{cor}) \cdot \nabla p f = 0, \]

where interactions enter via the mean-field potential, \( V^{mf}(r,t) = \int v(|r - \bar{r}|) n(\bar{r},t) d\bar{r}, \) and a correlation force, \( F^{cor}(r,t) = -\int \nabla v(|r - \bar{r}|) n(\bar{r},t) h(r,\bar{r},t) d\bar{r}. \)

To proceed we derive an equation for the dynamics of \( h(r_1,r_2,t). \) This is achieved by using Eq. (4) in Eq. (1b) and integrating over \( p_1 \) and \( p_2, \) which yields a continuity equation for \( n^{(2)}(r_1,r_2,t) = n(r_1)n(r_2)[1 + h(r_1,r_2,t)], \) where \( n(r,t) \) is the particle density. It can be further simplified with the continuity equation for \( n(r,t), \) and we obtain

\[ \partial_t h + \nabla \cdot u(r_1,t) + \nabla_2 h \cdot u(r_2,t) = 0, \]

where the fluid velocity field is defined via the nonequilibrium distribution function, \( u(r,t) = \frac{1}{m} \int \hat{p} f(r,p,t) d\hat{p}. \)

Equation (7) can be expressed via the convective time derivative as \( D_t h = 0, \) which yields the time evolution of the PCF in nonequilibrium: \( h(t) \) instantaneously responds to any currents induced in the plasma and adjusts its form according to Eq. (7). The dynamics of \( h \) is entirely driven by the one-particle distribution \( f(t), \) which, in turn, is coupled to the PCF via Eqs. (5) and (6).

Equations (5)–(7) are our first main result and provide the basis for a very general, yet complicated, kinetic description of trapped SCP in nonequilibrium. A simpler description still retaining correlations follows by eliminating the momentum dependence also for \( f(t), \) i.e., by turning to a fluid approach.

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3 For weak coupling, Guernsey [30] avoided the assumption of spatial homogeneity and the time-scale separation by considering small perturbations from equilibrium. His theory was applied to the plasma conductivity and dielectric function [31].

4 A similar concept has been used for long-living correlations related to bound states in Ref. [32]
Fluid equations are obtained as usual by taking moments of Eq. (5):

\[ D_1 n = -n \nabla \cdot u, \]

\[ mn D_1 u = -\nabla \cdot \pi + m(F_{\text{ext}} - \nabla V_{\text{mf}} + F_{\text{cor}}), \]

\[ D_r \pi_{\alpha \beta} = -\pi_{\alpha \beta} \nabla \delta u_\delta - \pi_{\alpha \delta} \nabla \delta u_\beta - \pi_{\beta \delta} \nabla \delta u_\alpha, \]

where \( \pi(r,t) = m \left[ \int \nabla r \cdot u_\alpha \right] \right] \int f(r,p,t) dp \) is the pressure tensor [38,39], which contains only kinetic contributions. We close the hierarchy by neglecting the high-frequency treatment of the correlation function. This system presents an important extension of standard fluid theory to strongly correlated \( (\Gamma \gg 1) \) and strongly inhomogeneous systems via the additional force \( F_{\text{cor}} \).

We close the hierarchy by neglecting the heat flux term as inferred Eq. (9) directly from the BBGKY hierarchy and Eq. (4).

\[ \delta h \approx -\nabla_1 h_0 \cdot q(r_1,t) - \nabla_2 h_0 \cdot q(r_2,t). \]

This dependence of \( \delta h \) on the displacement \( q \) becomes important at strong coupling since there \( h_0 \) rapidly varies as a function of \( r_1 \) and \( r_2 \). Therefore, neglect of \( \delta h \) (as in the original STLS scheme [5]) is the source of dramatic errors in the description of SCP which is overcome in the present ESTLS approach, as will be shown below.\(^5\)

Linearization of the system (8) allows one to derive a closed equation for the displacement field,

\[ mn_0 \delta_2 q_\alpha = -\delta_1 n \nabla_\alpha \left[ V + V_{\text{mf}} \right] - n_0 \nabla_\alpha \delta V_{\text{mf}} + n_0 \delta F_{\text{ext}} \]

\[ - \nabla_\beta \left[ \delta \pi_{\alpha \beta} + n_0 F_{\text{cor} \alpha} q_\beta \right] + n_0 F_{\text{cor} \alpha, \text{ext}}, \]

where we introduced

\[ F_{\text{cor} \alpha, \text{ext}}(r,t) = \int \nabla_\alpha \nabla_\beta \nu(|r - \bar{r}|) n_0(\bar{r}) h_0(r,\bar{r}) \]

\[ \times \left[ q_\beta(\bar{r},t) - q_\beta(r,t) \right] d\bar{r}, \]

\[ \delta \pi_{\alpha \beta} = -\left[ \nabla p_{\alpha \beta} \cdot q \delta \pi_{\beta} + p_{\beta \alpha}^0 \left( 2 \epsilon_{\alpha \beta} + \epsilon_{\beta \delta} \delta \pi_{\alpha \delta} \right) \right], \]

and the strain tensor, \( \epsilon_{\alpha \beta} = \frac{1}{2} \left( \nabla_\alpha q_\beta + \nabla_\beta q_\alpha \right) [39]. \)

Equations (10) and (11) are our second main result. They describe the linear response of a strongly inhomogeneous SCP to external fields based on its equilibrium PCF \( h_0(r,\bar{r}) \), which is required to solve Eq. (10) self-consistently. It can be obtained, e.g., from simulations or thermodynamic strong coupling theories [40]. Equations (10) and (11) provide a generalization of existing fluid theories. In particular, when the confinement potential \( V, F_{\text{cor} \alpha} \), and the kinetic pressure are neglected, we recover the (linear) nonuniform QLCA [25]. If, further, the system is homogeneous, \( F_{\text{cor} \alpha} \) leads to the QLCA dynamic matrix while \( F_{\text{cor} \alpha}^0 \) vanishes. Finally, if \( F_{\text{cor} \alpha} \) are neglected, we recover the warm-fluid equations for a (classical) weakly coupled plasma. Naturally, the cold-fluid limit is obtained if we further discard the kinetic pressure tensor.

Discussion. Our ESTLS ansatz incorporates the dominant dynamics of pair correlations through \( D_r h = 0, \) Eq. (7). The physical meaning is that the correlations between two particles are frozen as they move along with the fluid. This is valid as long as relaxation processes are negligible, and the PCF instantly responds to the fluid displacements [see Eq. (9)], i.e., at high frequencies \( \omega \) (elastic behavior, see also Ref. [41]).

The omission of the heat flux term to the fluid equations is also known to be a high-frequency approximation [39], valid for \( \omega \gg v_0/T, \) where \( v_0 = \sqrt{k_B T/m} \) is the thermal velocity and \( l \) the length scale of interest [38]. This is well justified at strong coupling (low temperature, with the de Broglie wavelength much smaller than the interparticle spacing so a classical approach is valid).

Let us now test our theory on the important case of the breathing mode in a harmonic trap, \( V(r) = \frac{1}{2} \omega_0^2 r^2. \) Although accurate results for the breathing frequency \( (BF, \omega_0) \) were recently obtained for systems with power-law interaction [17], the case of general interactions is still open. The displacement vector is chosen radial and uniform, \( q(r,t) \sim r e^{-i \omega t} \), which was shown to be accurate (i) at infinite coupling (crystals) [42] and (ii) when the system size significantly exceeds the screening length [20]. The result for the BF within our ESTLS

FIG. 1. (Color online) Breathing frequency of three-dimensional (3D) Yukawa clusters with (a) 100 and (b) 200 particles. Comparison of MD simulations, STLS theory, the present ESTLS [Eq. (13)], and a cold-fluid approach [20].

\(^5\)Strictly speaking, the ansatz (4) requires that the strong coupling condition is not violated at any time.

\(^6\)Compared to STLS or its improvements, where \( \delta h \) is constructed from the derivative of \( h_0 \) with respect to the density [35], we have inferred Eq. (9) directly from the BBGKY hierarchy and Eq. (4).
inhomogeneous extension] and the present ESTLS. The dependence is not well understood [24]. The MD results are been studied at zero temperature [46,47] but its temperature are scaled to $\omega_0$ (dashed lines, open symbols). The frequencies are scaled to $\omega_0$—the frequency at the largest $\Gamma$ measured in the simulation.

theory reads [43]

$$\frac{\omega_0^2}{\omega^2_0} = 1 + \frac{E_{\text{kin}}}{E_{\text{pot}}} + \frac{1}{2} \frac{(v_2)_2}{E_{\text{pot}}} , \quad \text{(13)}$$

where $E_{\text{kin}} = \frac{d}{2} N k_B T$ is the average kinetic and $E_{\text{pot}} = \frac{1}{2} m \omega_0^2 (r^2)$ the potential energy. The one- and two-particle averages are $\langle r^2 \rangle_1 = \int r^2 n_0(r) \, dr$ and

$$\langle v_i \rangle_2 = \frac{1}{2} \int \int v_i(r_1) n_0(r_1) n_0(r_2) [1 + h_0(r_1, r_2)] d r_1 d r_2 ,$$

respectively ($i = 1, 2$), where $v_1(r) = v'(r) r$ and $v_2(r) = v''(r) r^2$. This generalizes the result of Ref. [17] to arbitrary interaction. Similar expressions were found for confined quantum gases [44]. Using the Virial theorem, $\langle v_1 \rangle_2 = 2(E_{\text{kin}} - E_{\text{pot}})$, we also recover the result of the harmonic approximation [44,45] if we neglect $E_{\text{kin}}$ at strong coupling.

In Fig. 1 we present MD simulation results for Yukawa interaction, $v(r) = Q^2 e^{-r/\lambda} / r$, where $Q$ is the particle charge and $\lambda$ the screening length [24], covering long (plasmas) and short (neutral systems) interaction ranges. The Yukawa BF has been studied at zero temperature [46,47] but its temperature dependence is not well understood [24]. The MD results are used to benchmark the theoretical results of STLS [using its inhomogeneous extension] and the present ESTLS.

The first observation is that ESTLS shows excellent agreement with MD over the whole range of the parameter $\Gamma = Q^2 / (a k_B T)$, where $a = (Q^2 / m \omega_0^2)^{1/3}$ is a characteristic length scale. In contrast, while STLS improves upon the cold-fluid theory of Ref. [24], see Figs. 1(a) and 1(b), the deviations from the MD result rapidly increase with $\Gamma$, for $\Gamma > 1$. The origin of this poor performance is clearly traced to the neglect of $\delta h$, i.e., the spatial derivatives of the PCF [cf. Eq. (9)], which increase with $\Gamma$. In the Coulomb limit [Fig. 1(c)], STLS slightly drops below $\omega_0/\omega_0 = \sqrt{3}$, which is the exact limit for $\Gamma \to \infty$ [47].

A detailed comparison between ESTLS and MD is shown in Fig. 2. Besides the excellent quantitative agreement, we note that ESTLS reproduces the nontrivial dependence of $d\omega_0/d\Gamma$ on the interaction range $\alpha / \lambda$ [24]. The reason for the change in slope is that in the ideal gas limit, $\Gamma \to 0$, $\omega_0$ is independent of the interaction and given by $\omega_0(0) = 2$, whereas it depends on $\alpha / \lambda$, in the infinite coupling limit, $\Gamma \to \infty$. Thus, $\omega_0(\Gamma)$ increases (decreases) if the latter lies above (below) the former.

In summary, we have presented a kinetic approach to the dynamics of strongly correlated confined plasmas. The simple ansatz, Eq. (4), and the BBGKY hierarchy directly determine the dynamics of the PCF [Eq. (7)]. The theory was applied to the breathing mode in a harmonic trap and showed excellent agreement with MD simulations. If accurate equilibrium PCFs are available, our theory provides a direct route to determining the collective modes self-consistently by solving Eq. (10). If, on the other hand, strong excitations are of interest, the nonlinear fluid equations [Eq. (8)] or the kinetic equation [Eq. (5)] are available. The presented ESTLS theory should be useful for the study of high-frequency processes in inhomogeneous SCP, in particular dusty, non-neutral, and ultracold neutral plasmas. Furthermore, the basic idea of partial weakening of initial correlations and the ansatz (4) should also be of relevance to strongly correlated confined quantum systems.

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Note that $\Gamma$, despite being similar to the coupling parameter for a homogeneous plasma, does not describe the effective coupling strength since the length scale $a$ does not take the density variation in the trapped system into account. In addition, the screening effect is not included. The parameter can be interpreted as a dimensionless inverse temperature. For a discussion of coupling in Yukawa systems, we refer to Ref. [48].