Fundamental theory of statistical particle dynamics

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We present a fundamental theory for the kinetics of systems of classical particles. The theory represents a unification of kinetic theory, Brownian motion, and field theory. It is self-consistent and is the dynamic generalization of the functional theory of fluids in equilibrium. This gives one a powerful tool for investigating the existence of ergodic-nonergodic transitions near the liquid-glass transition.

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I. INTRODUCTION

We present here a theory[1] for the dynamics of classical particles which solves the chronic problem of self-consistency. This theory unites the desirable elements of kinetic theory [1], Brownian motion [2], and modern field theory [3].

Kinetic theory is one of our oldest [4] theoretical disciplines. Despite its many successes it has never been constructed in a fully self-consistent form [5]. Thus one of the most famous approximations in all of science, the *stosszahlansatz* of Boltzmann [6] and the treatment of the collision integral in the Boltzmann equation [7] have not been investigated systematically. The theory introduced here provides the tools to remedy this situation.

The stosszahlansatz, also referred to as the assumption of molecular chaos, is representative of decoupling approximations appearing in many [8] problems and characterized as uncontrolled by the approximation police. Of particular current interest is the validity of mode-coupling theory (MCT) [9] used in theories of the liquid-glass transition [10]. We should be able to answer the question: is the liquid-glass transition accompanied by an ergodic-nonergodic (ENE) transition [11]? The construction [12] of mode-coupling models using traditional kinetic theory is *ad hoc* and short of convincing. It has been completely ineffective in exploring corrections to conventional mode-coupling theory. The field has moved away from kinetic theory treatments and turned instead to field theoretical models [13] where one has the promise of perturbative control. Thus we recently introduced the random diffusion model which can support an ergodic-nonergodic transition [11] at one loop order. However going to two-loop order one finds that the system cannot sustain the ENE solution. The problem with such models, compared to microscopic models organized in terms of a pair potential, is that the short-distance structure is not treated naturally. In the case of colloidal systems, there has been some convergence on the Dean-Kawasaki (DK) model as the simplest field theoretic model that describes the kinetics of the colloidal systems operating under Smoluchowski dynamics. It has been difficult, for technical reasons, to establish whether the Dean-Kawasaki model supports an ENE transition even at one loop order.

It is demonstrated here how mode-coupling theory [14] naturally occurs in the application of our theory to colloidal systems governed by Smoluchowski dynamics. The approach, which allows for compatible approximations for higher-order correlation functions [15], is applicable to a large set of dynamical systems [16], reversible [17] and dissipative, including Newtonian [18], Fokker-Planck [19], and Smoluchowski [20] dynamics. The theory is organized in terms of a coupling to time and space dependent external fields. This allows for great flexibility in using functional methods in developing various types of perturbation theory.

A key point is that the equilibrium equal-time fluid structure [21] has been understood from a self-consistent field theoretical point of view for a long time. The work presented here is the natural extension to the dynamic regime of the beautiful diagrammatic/functional development for the static properties. The theory in the static case proceeded first through the introduction of graphical methods by Mayer [22] and others [23] in the 1940s and then greatly profited from graphical resummation techniques [24] which were subsequently supplemented by functional methods [25] as discussed below. Of particular interest here is the functional formulation of Percus [21,26] which cleanly connects with the widely applied self-consistent approximations named Percus-Yevick [27] and hypernetted chain [28] which fit prominently into the tool kit of anyone studying the statics of fluids. The theory, which has been applied to a large variety of systems, is in the form of nonlinear integral equations connecting the radial (pair) distribution function, the direct correlation function and the Ornstein-Zernike relation [29].

Why has it taken 40 years to extend the static theory to the dynamic regime?

Part of the answer is connected with the difficulties in developing an efficient field theoretical [30] description for classical field dynamics. It was not until the work of Martin, Siggia, and Rose [31], introducing field doubling via conjugate response fields, and its generalization by Janssen [32], De Dominicis [33], Graham [34], and others [35] that we had a self-consistent treatment for field-theoretic models such as the time-dependent Ginzburg Landau models [36] and all of the models representing dynamic critical phenomena univer-

[1]The title of this paper suggests that the theory is more widely applicable than to Smoluchowski dynamics. This is true. The basic development goes through for Newtonian dynamics, Fokker-Planck dynamics and trapped systems. However, presenting the theory in its most general form makes the development more complicated. Instead, I have chosen to present the theory in its simplest application, the case of Smoluchowski dynamics. The case of Newtonian dynamics will be available soon (S. Das and G. Mazenko).

sality classes [37]. In organizing these dynamical theories it is important to carefully incorporate causality. This is related to the issue of the proper treatment [38] of the Jacobian of the transformation from a Langevin description in terms of noise to the path-integral description in terms of physical fields. There are some apparent ambiguities in determining the Jacobian. This Jacobian was identified at least qualitatively in the early work of Onsager and Machlop. The treatment of the Jacobian has led to a fascinating set of extensions of the theory to include the topics such as ghost fermions [39], supersymmetry [40], Onsager's Reciprocity relations [41], and Jarzynski and fluctuation theorems [42]. We intend to return to these topics in future work since they can be explored in the case of particle models of the type studied here. Kinetic theory is complicated [43] compared to conventional field theories because the collective variables, particle density and phase-space density, are distributions (sums of δ functions) not smooth fields. This leads to nonlinear constraints like

$$\rho(x_1)\rho(x_2) = \rho(x_1)\delta(x_1 - x_2) + \text{two particle terms}. \quad (1)$$

One important aspect of the kinetic theory problem is that static development in terms of the density is strongly non-Gaussian [44]. By this we mean that the density fluctuations of an ideal gas are not Gaussian and one has an set of irreducible vertex functions which cannot be treated as small. The connected vertex functions for an ideal gas are not small. We return to this important point below.

II. SMOLUCHOWSKI DYNAMICS

Let us begin by defining the dynamical system of interest. Consider a system of N particles with configurations specified by the coordinates R_i which satisfy the equations of motion

$$\dot{R}_i = DF_i + \eta_i, \quad (2)$$

where the particles experience force

$$F_i = -\frac{\partial}{\partial R_i}U(R), \quad (3)$$

with total potential

$$U(R) = \frac{1}{2}\sum_{i\neq j} V(R_i - R_j), \quad (4)$$

where we choose $V(0)=V'(0)=V''(0)=0$ and we have suppressed vector labels to unclutter the equations. There is a noise source η_i for each coordinate which is taken to be Gaussian with variance

$$\langle \eta_i(t)\eta_j(t')\rangle = 2k_BTD\delta(t-t')\delta_{ij}, \quad (5)$$

where D is a diffusion coefficient. It is conventional to develop kinetic theory in terms of the phase-space density $f(1)=\Sigma_{i=1}^N \delta(x_1-R_i(t_1))\delta(p_1-P_i(t_1))$ and its cumulants. For the case of Smoluchowski dynamics this would suggest using the particle density $\rho(1)=\Sigma_{i=1}^N\delta(x_1-R_i(t_1))$ as the Martin-Siggia-Rose (MSR) field. The key to our development is that we break with tradition and treat the particle coordinates as our MSR fields with accompanying conjugate response fields.

In the approach developed here we keep track of degrees of freedom by coupling to them with external source fields. In principle we can keep track of all the degrees of freedom in the system. In practice, however, we are interested in following a small set of collective variables we label Φ. For this system, the density ρ is essential since it governs the static equilibrium behavior and, from the point of view developed here, is always included in $\Phi=(\rho,\ldots)$. For reasons that will be developed below we must also include in Φ a field B which is constructed and interpreted below. The set of collective variables treated $[\Phi=(\rho,B,\ldots)]$, is flexible and controlled by pairing each observable with a conjugate external field $(H=H_\rho,H_B,\ldots)$. The set Φ must include the fields ρ and B since we need both to control and manipulate the interactions in the system. One can include other observables in the problem, like the potential energy density, but they play a more passive role in the development.

We take advantage of the fact that, while the density is strongly non-Gaussian, the positions R_i may be more profitably thought of as Gaussian variables. Therefore, in developing our theory we do not work in the Langevin description, but instead work in the MSR representation. (As discussed in Appendix A, one has at least three approaches to choose from: Langevin, Fokker-Planck and Martin-Siggia-Rose.) The generator of cumulants in the MSR representation is given by $W_N[H]$ which is related to the N-particle partition function by

$$e^{W_N[H]} = Z_N[H] = \int \prod_{i=1}^N [\mathcal{D}(R_i)\mathcal{D}(\hat{R}_i)d^dR_i^{(0)}]P_0(R_i^{(0)})e^{-A_R}e^{H\cdot\Phi}, \quad (6)$$

where we have a probability distribution $P_0(R_i^{(0)})$ governing the system at the initial time t_0. The shorthand notation $H\cdot\Phi$ means $\int d^dx_1\int_{t_0}^\infty dt_1[H_\rho(x_1,t_1)\rho(x_1,t_1)+H_B(x_1,t_1)B(x_1,t_1)+\cdots]$. In most of our discussion here we assume the system is in equilibrium initially and the initial distribution for a set of N particles is canonical,

$$P_0[R_0] = e^{-\beta U(R_0)}/Z_0, \quad (7)$$

where U is the potential energy defined by Eq. (4) and β is the inverse temperature. The MSR action for the problem is given by

$$A_R = \int_{t_0}^\infty dt_1 \sum_{i=1}^N \{\hat{R}_i(t_1)k_BTD\hat{R}_i(t_1) + i\hat{R}_i(t_1)\cdot[\dot{R}_i(t_1) - DF_i(t_1)]\} + A_J, \quad (8)$$

where the contribution to the action A_J is from the notorious Jacobian [38]. (The steps leading from the Langevin description to the MSR field theory description are discussed in Appendix A.) The Jacobian plays a crucial role in this kinetic problem and is defined by

$$J = \det \frac{\delta \eta_i(t')}{\delta R_j(t)} = \det \frac{\delta}{\delta R_j(t)} \left(\frac{\partial R_i(t')}{\partial t'} - D F_i(t') \right)$$
$$= \det\ \delta_{ij} \left[\frac{\partial}{\partial t'} \delta(t-t') - D \frac{\partial F_i(t')}{\partial R_i(t')} \delta(t-t') \right]. \quad (9)$$

Exponentiating to write the Jacobian as a contribution to the action gives [45]

$$A_J = -\ln J = -\int_{t_0}^{\infty} dt\, \theta(0) \left(\sum_{i=1}^{N} D \frac{\partial F_i(t)}{\partial R_i(t)} \right), \quad (10)$$

where $\theta(0)=1/2$. Together, Eqs. (6), (8), and (10) define the problem of interest.

Now we want to make a separation of the degrees of freedom into two groups; one group consists of some collective variables to be chosen, and the second group consists of all the rest of the degrees of freedom. The first step in this separation is to rewrite Eq. (10) in terms of the particle density, $\rho(1)=\Sigma_{i=1}^{N}\delta[x_1-R_i(t_1)]$. We find

$$A_J = \theta(0) \int_{t_0}^{\infty} dt \int d^d x d^d y D \nabla_x^2 \rho(x,t) V(x-y) \rho(y,t) + \text{const}$$
$$= \int_{t_0}^{\infty} dt \int d^d x d^d y B_J(x,t) V(x-y) \rho(y,t) + \text{const}, \quad (11)$$

where we have defined the quantity

$$B_J(x,t) = \theta(0) D \nabla_x^2 \rho(x,t) \quad (12)$$

and the constant can be absorbed into the normalization of the partition function.

Next, notice that the dynamic part of the interaction contribution to the action can be rewritten in the form

$$D \int_{t_0}^{\infty} dt \sum_{i=1}^{N} i\hat{R}_i(t) F_i(t) = \int_{t_0}^{\infty} dt \int d^d x d^d y B_0(x,t) V(x-y) \rho(y,t), \quad (13)$$

where

$$B_0(x,t) = D \sum_{i=1}^{N} i\hat{R}_i(t) \cdot \nabla_{R_i} \delta[x - R_i(t)]. \quad (14)$$

We can then combine this contribution to the action with the contribution from the Jacobian to obtain the dynamic part of the interaction in the form

$$A_I = \int d1 d2 B(1) V(12) \rho(2), \quad (15)$$

where $B(1)=B_0(1)+B_J(1)$ is the field discussed above, $\int d1 = \int_{t_0}^{\infty} dt_1 d^d x_1$, and

$$V(12) = V(x_1 - x_2)\delta(t_1 - t_2). \quad (16)$$

Writing things out explicitly, the conjugate field is given by

$$B(1) = D \sum_{i=1}^{N} \{ [\hat{R}_i i \nabla_1 + \theta(0) \nabla_1^2] \} \delta[x_1 - R_i(t_1)]. \quad (17)$$

We can then write the partition function given by Eq. (6) in the form

$$Z_N = \int \prod_{i=1}^{N} [\mathcal{D}(R_i) \mathcal{D}(\hat{R}_i) d^d R_i^{(0)}] P_0(R_i^{(0)}) e^{-A_0 - A_I + H \cdot \Phi}, \quad (18)$$

where A_0 is the quadratic part of the action excluding the quadratic contribution to the initial probability distribution

$$A_0 = \int_{t_0}^{\infty} dt_1 \sum_{i=1}^{N} [\hat{R}_i k_B T D \hat{R}_i + i\hat{R}_i \cdot \dot{R}_i]. \quad (19)$$

Notice that we have constructed things such that the coordinates are constrained to have the values $R_i^{(0)}$ at $t=t_0$. We then average over these values. Here we are explicitly treating the case where the system is in equilibrium at $t=t_0$, but more general situations are clearly compatible with the development. The interaction part of the action (including the initial probability distribution) is given in the compact form

$$A_I = \frac{1}{2} \sum_{\alpha,\nu} \int d1 d2 \Phi_\alpha(1) \sigma_{\alpha\nu}(12) \Phi_\nu(2), \quad (20)$$

where the Greek labels range over ρ and B and we introduce the interaction matrix

$$\sigma_{\alpha,\nu}(12) = [-\beta V(12)][\hat{\rho}_\alpha \hat{\rho}_\nu \delta(t_1 - t_0) - \beta^{-1}(\hat{\rho}_\alpha \hat{B}_\nu + \hat{B}_\alpha \hat{\rho}_\nu)], \quad (21)$$

where we have introduced the useful notation

$$\hat{\rho}_\alpha = \delta_{\alpha,\rho} \quad (22)$$

and

$$\hat{B}_\alpha = \delta_{\alpha,B}. \quad (23)$$

The canonical partition function can be written in the convenient form

$$Z_N = Tr^{(N)} e^{-A_I + H \cdot \Phi}, \quad (24)$$

where we have introduced the average

$$Tr^{(N)} \mathcal{O} = \int \prod_{i=1}^{N} [\mathcal{D}(R_i) \mathcal{D}(\hat{R}_i) d^d R_i^{(0)}] P_0(R_i^{(0)}) e^{-A_0} \mathcal{O}(R). \quad (25)$$

Notice that the single-particle contribution to the action A_0 [Eq. (19)] is included in the weight in Tr. Thus the class of problems of interest are defined in terms of a path-integral formulation.

Note that the case where there is a strong external potential acting on the system of particles is easily treated within the development. Suppose that the total force acting in Eq. (2) on particle i is of the form

$$F_i^T = F_i + F_i^E \tag{26}$$

and the external force is generated by a potential

$$F_i^E = -\nabla_{R_i}\int d^dx U_E(x,t)\rho(x,t) = -\nabla_{R_i} U_E[R_i(t),t]. \tag{27}$$

An important practical example is the case of optical tweezers where this external potential or trap can be taken to be of the form

$$U_E(x,t) = \frac{1}{2}\kappa(t)[x - R_0(t)]^2, \tag{28}$$

where $\kappa(t)$ is a controllable amplitude for the potential and $R_0(t)$ is the position of the trap. If one follows the development of the previous section, one finds that the external force generates a term in the action

$$A_{U_E} = \int d1 B(1) U_E(1), \tag{29}$$

which can be included in the one-body term in the action. In this case, the initial conditions can be influenced in several ways. If the trap is turned on for times $t > t_0$ there is no change in initial conditions, while one could prepare the system in a static trap where one would need to add a term to the initial potential energy,

$$U \to U + \int d^dx U_E(x,t_0)\rho(x), \tag{30}$$

and $U_E(x,t) = U_E(x,t_0)$ for $t < t_0$. There are many other possibilities. The net result of introducing this external potential U_E that couples to the density is to physically produce the dynamic coupling to the field $B(1)$,

$$H_B(1) = U_E(1). \tag{31}$$

If one is interested in fluctuations in equilibrium in the presence of a time-independent inhomogeneous potential $u(x_1)$, then one makes the replacements $H_\rho(1) = u(x_1)\delta(t_1 - t_0)$ and $H_B(1) = u(x_1)$. H_ρ adjusts the initial condition and H_B has the effect of changing the equation of motion to include the force due to $u(x)$.

We have succeeded in writing our nonequilibrium problem as a path integral characterized by a field-dependent partition function written in the very compact symmetrical form in the grand canonical ensemble,

$$Z_T[H] = \sum_{N=0}^{\infty} \frac{\rho_0^N}{N!} Tr^{(N)} e^{\int d1 H(1)\cdot\Phi(1)} e^{1/2 \int d1 d2 \Phi(1)\cdot\sigma\cdot\Phi(2)}, \tag{32}$$

which emphasizes the role of the collective fields ρ and B. We have yet to show that this can be expressed in a form which produces a self-consistent form of perturbation theory.

III. SELF-CONSISTENT DEVELOPMENT FOR THE GENERATING FUNCTIONAL

We now want to rewrite the partition function in a form that allows us to formally carry out the average in Eq. (32). We can use the functional identity [46]

$$e^{-A_I + H\cdot\Phi} = e^{\hat{A}_T} e^{H\cdot\Phi}, \tag{33}$$

where we define the operators

$$\hat{A}_T = \frac{1}{2}\int d1 d2 \sum_{\alpha\beta} \sigma_{\alpha\beta}(12)\hat{H}_\alpha(1)\hat{H}_\beta(2) \tag{34}$$

and

$$\hat{H}_\alpha(1) = \frac{\delta}{\delta H_\alpha(1)}. \tag{35}$$

The interaction matrix $\sigma_{\alpha\beta}$ is given by Eq. (21). Then, using Eq. (33) in Eq. (24) gives

$$Z_N = e^{\hat{A}_T} Tr^{(N)} e^{H\cdot\Phi}. \tag{36}$$

Next, we restrict the set of fields Φ_i to those that are one-particle additive,

$$\Phi_i = \sum_{\alpha=1}^{N} \phi_i^\alpha, \tag{37}$$

which is true for the particle density ρ and conjugate field B, and notice that the sum over the degrees of freedom in Eq. (36) factorizes into a product of sums over the degrees of freedom of each particle. Together, these observations lead to the result

$$Z_N^{(0)} = Tr^{(N)} e^{H\cdot\Phi} = (Z_1)^N, \tag{38}$$

where the noninteracting partition function for a single particle is

$$Z_1 = Tr^{(1)} e^{H\cdot\phi^{(1)}}. \tag{39}$$

Working in the grand canonical ensemble, the grand partition function for the interacting problem is given by

$$Z_T = \sum_{N=0}^{\infty} \rho_0^N \frac{Z_N}{N!} = \sum_{N=0}^{\infty} \frac{\rho_0^N}{N!} e^{\hat{A}_T} Z_N^{(0)} = e^{\hat{A}_T} \sum_{N=0}^{\infty} \frac{\rho_0^N}{N!} Z_1^N = e^{\hat{A}_T} e^{W_0}, \tag{40}$$

where ρ_0 is the fugacity or bare density and

$$W_0 = \rho_0 Tr^{(1)} e^{H\cdot\phi^{(1)}} = \tilde{T}r e^{H\cdot\phi}, \tag{41}$$

where in the last line we have dropped the particle label on the trace and the field ϕ. The cumulants of the fields Φ_i are generated by taking functional derivatives of the generating functional

$$W[H] = \ln Z_T \tag{42}$$

with respect to H_i. The one-point average in a field is given by

$$G_i = \frac{\delta}{\delta H_i} W[H],$$

where we have used a compact notation where i labels space, time and fields ρ or B. Substituting for $W[H]$ using Eq. (40) we find

$$G_i = \frac{1}{Z^T} e^{\hat{A}_T} e^{W_0} \frac{\delta}{\delta H_i} W_0 = \frac{1}{Z^T} \tilde{T}r \phi_i e^{\hat{A}_T} e^{\phi \cdot H} e^{W_0}$$

$$= \frac{1}{Z^T} \tilde{T}r \phi_i e^{\hat{A}_T} e^{\phi \cdot H} e^{-\hat{A}_T} e^{\hat{A}_T} e^{W_0}. \quad (43)$$

It is not difficult to prove the functional identity,

$$e^{\hat{A}_T} e^{H \cdot \phi} e^{-\hat{A}_T} = e^{H \cdot \phi} e^{\left[E + \Sigma_i F_i \hat{H}_i\right]}, \quad (44)$$

where

$$E = \frac{1}{2} \sum_{ij} \sigma_{ij} \phi_i \phi_j \quad (45)$$

is a self-interaction contribution and

$$F_i = \sum_j \sigma_{ij} \phi_j \quad (46)$$

will play an important role as we go along. Using Eq. (44) back in Eq. (43), we obtain

$$G_i = \frac{1}{Z^T} \tilde{T}r \phi_i e^{H \cdot \phi} e^{[E + F \cdot \hat{H}]} e^{W[H]} = \tilde{T}r \phi_i e^{H \cdot \phi + E + \Delta W[H]}, \quad (47)$$

where

$$\Delta W[H] = W[H + F] - W[H]. \quad (48)$$

An interesting check on the theory is to show that the self-interaction E given by Eq. (45) vanishes. This follows if the potential is constructed to be zero at the origin $[V(0)=0]$. Our most important result is given by

$$G_i = \tilde{T}r \phi_i e^{H \cdot \phi + \Delta W[H]}. \quad (49)$$

Another result useful in this description follows from taking the derivative of Eq. (40) with respect to ρ_0, following steps similar to those leading to Eq. (49), and integrating with respect to ρ_0 which leads to the result

$$W[H, \rho_0] = \int_0^{\rho_0} dx Tr e^{H \cdot \phi + W[H+F,x] - W[H,x]}. \quad (50)$$

It takes a little calculus to show that the derivative of Eq. (50) with respect to H leads to Eq. (49). Equation (50) can be rewritten in terms of the more fundamental identity

$$\frac{\partial}{\partial \rho_0} Z_T[H, \rho_0] = \tilde{T}r e^{H \cdot \phi} Z_T[H + F, \rho_0]. \quad (51)$$

What about response functions? The response of the density to an external potential U_E which couples to the density is given by

$$\chi_{\rho\rho}(12) = \frac{\delta}{\delta U_E(2)} \langle \rho(1) \rangle = \frac{\delta}{\delta H_B(2)} \langle \rho(1) \rangle = G_{\rho B}(12)$$

$$= \frac{\delta}{\delta H_\rho(1)} \frac{\delta}{\delta H_B(2)} W[H]. \quad (52)$$

We must now work to show why Eq. (49) is very desirable. There are several ways one can use Eq. (49) to build an approximate theory. In comparison with the static theory one would guess that density expansions would be the most successful. This may be so, but working with expansions in the pair interaction is conceptionally simpler and more direct. It seems clear that in developing density expansions one will be able to make contact with the hypernetted chain and Percus-Yevick approximations. There appears much one can do about coupling constant renormalization. We return to discuss density expansions elsewhere.

The dependence of the theory on the pair potential is controlled by the quantity $\Delta W[H] = W[H+F] - W[H]$. We can expose the dependence on the potential by constructing the functional Taylor-series expansion

$$\Delta W[H] = \sum_i F_i \frac{\delta}{\delta H_i} W[H] + \sum_{ij} \frac{1}{2} F_i F_j \frac{\delta^2}{\delta H_i \delta H_j} W[H] + \cdots \quad (53)$$

and we can conveniently introduce the set of cumulants,

$$G_{ij\ldots k} = \frac{\delta}{\delta H_i} \frac{\delta}{\delta H_j} \cdots \frac{\delta}{\delta H_k} W[H] \quad (54)$$

to obtain

$$\Delta W[H] = \sum_i F_i G_i + \sum_{ij} \frac{1}{2} F_i F_j G_{ij} + \sum_{ijk} \frac{1}{3!} F_i F_j F_k G_{ijk} + \cdots \quad (55)$$

with F_i given by Eq. (46). Clearly, in this form we can take ΔW to be a functional of G_i. One can then use functional differentiation to express higher-order cumulants in terms of the one- and two-point correlation functions G_i and G_{ij}. One has, for example, the manipulation expressing the three-point cumulant in terms of lower order objects,

$$G_{ijk} = \frac{\delta}{\delta H_k} G_{ij} = \sum_{mnp} - G_{im} G_{jn} G_{kp} \Gamma_{mnp}, \quad (56)$$

where the irreducible three-point vertex is given as a functional derivative of the two-point irreducible vertex

$$\Gamma_{ijk} = \frac{\delta}{\delta G_k} \Gamma_{ij} \quad (57)$$

and Γ_{ij} is precisely the matrix inverse of the two-point cumulant

$$\sum_k \Gamma_{ik} G_{kj} = \delta_{ij}. \quad (58)$$

The beauty of the modern field theoretical development is that an approximation for the two-point vertex as a functional of the G_i and G_{ij} generates self-consistent approximations for all higher-order correlation functions. The method is set up to carry out various types of renormalization like replacing the bare interactions with effective interactions. This will be exploited elsewhere. We expect the situation here to be similar to quantum many-body theory. Self-consistency and conservation laws can be brought together to suggest ways of generating approximations as done by Kadanoff and Baym with their Φ-derivable [47] approximations. A key

constraint is equilibrium is the fluctuation-dissipation theorem.

Some of the structure of the theory can be appreciated via the establishment of a dynamic generalization of the static Ornstein-Zernike relation [29]. Starting with the functional equation for the two-point cumulant, one can use the chain-rule for functional differentiation to obtain

$$G_{ij} = \frac{\delta}{\delta H_j} G_i$$
$$= \tilde{Tr}\phi_i\phi_j e^{H\cdot\phi+\Delta W} + \sum_k \tilde{Tr}\phi_i e^{H\cdot\phi}\left(\frac{\delta}{\delta G_k}e^{\Delta W}\right)\frac{\delta}{\delta H_j}G_k$$
$$= \mathcal{G}_{ij} + \sum_k c_{ik}G_{kj}, \tag{59}$$

where

$$\mathcal{G}_{ij} = \tilde{Tr}\phi_i\phi_j e^{H\cdot\phi+\Delta W} \tag{60}$$

is a single-particle quantity and we have the memory function [48], self-energy, or dynamic direct correlation function given by

$$c_{ij} = \tilde{Tr}\phi_i e^{H\cdot\phi+\Delta W}\frac{\delta}{\delta G_j}\Delta W. \tag{61}$$

Since ΔW can be treated as a functional of G_i we see at this stage that we have available a self-consistent theory. If we define the matrix inverse

$$\sum_k \gamma_{ik}\mathcal{G}_{kj} = \delta_{ij} \tag{62}$$

then the two-point vertex is given without approximation as

$$\Gamma_{ij} = \gamma_{ij} + K_{ij}, \tag{63}$$

where

$$K_{ij} = -\sum_k \gamma_{ik}c_{kj}. \tag{64}$$

IV. NONINTERACTING SMOLUCHOWSKI SYSTEM

The first step in applying this theory is to work out the noninteracting cumulants for the fields $\Phi=(\rho,B)$. This calculation for the fundamental objects R, $\hat{R}$ is carried out in Appendix B. The cumulants for the collective fields are worked out in detail in Appendix C.

The final results are

$$G^{(0)}_{B\ldots B\rho\ldots\rho}(1,\ldots,\ell,\ell+1,\ldots,n)$$
$$= \rho_0(2\pi)^d\delta\left(\sum_{i=1}^n q_i\right)b(1)\cdots b(\ell)e^{N_n}, \tag{65}$$

where

$$N_n = \frac{1}{2}\bar{D}\sum_{i=1}^n\sum_{j=1}^n q_i\cdot q_j|t_i - t_j| \tag{66}$$

and

$$b(j) = \bar{D}\sum_{i\neq j=1}^n q_i\cdot q_j\theta(t_i - t_j) \tag{67}$$

and where $\bar{D}=k_BTD$. Explicitly, the two-point cumulants are

$$G^{(0)}_{\rho\rho}(q,q';t,t') = \rho_0(2\pi)^d\delta(q+q')e^{-\bar{D}q^2|t-t'|}, \tag{68}$$

$$G^{(0)}_{\rho B}(q,q';t,t') = -\rho_0(2\pi)^d\delta(q+q')Dq^2\theta(t-t')e^{-\bar{D}q^2(t-t')}, \tag{69}$$

$$G^{(0)}_{B\rho}(q,q';t,t') = -\rho_0(2\pi)^d\delta(q+q')Dq^2\theta(t'-t)e^{-\bar{D}q^2(t'-t)} \tag{70}$$

and

$$G^{(0)}_{BB}(q,q';t,t') = 0. \tag{71}$$

Notice that $G_{\rho B}$ is retarded and proportional to q^2, while $G_{B,\ldots,B}=0$. The results for density cumulants agree with the results from recent work [49] that shows statistical dynamics of the density of noninteracting Brownian particles can be described by a cubic field theory where the density is the fundamental field.

V. PERTURBATION THEORY FOR THE TWO-POINT CUMULANT

The perturbation theory can be organized in terms of the irreducible vertex functions. It is clear from the generalized Ornstein-Zernike equation [Eq. (59)] that the matrix inverse of the two-point cumulant is given by Eq. (63) with the matrix γ defined by Eq. (62) and the self-energy K_{ij} by Eq. (64). To get started, one constructs the noninteracting two-point cumulant using Eq. (65) and one finds the matrix inverses to be given by

$$\gamma^{(0)}_{B\rho}(12) = -\frac{1}{\rho_0 Dk_1^2}\left(\frac{\partial}{\partial t_1} + \bar{D}k_1^2\right)\delta(t_1 - t_2), \tag{72}$$

$$\gamma^{(0)}_{\rho B}(12) = -\frac{1}{\rho_0 Dk_1^2}\left(-\frac{\partial}{\partial t_1} + \bar{D}k_1^2\right)\delta(t_1 - t_2), \tag{73}$$

$$\gamma^{(0)}_{BB}(12) = -\frac{2D}{\rho_0 Dk_1^2}\delta(t_1 - t_2), \tag{74}$$

$$\gamma^{(0)}_{\rho\rho} = 0. \tag{75}$$

Working to first order in zero external field, $\Delta W=\Sigma_u F_u G_u$ and one has contributions to $\mathcal{G}_{ij}$ given by

$$\mathcal{G}_{ij} = \tilde{Tr}\phi_i\phi_j(1 + \Delta W + \cdots)$$
$$= \tilde{Tr}\phi_i\phi_j\left(1 + \sum_u F_u G_u\right)$$
$$= G^{(0)}_{ij} + \sum_{k,u} G^{(0)}_{ijk}\sigma_{ku}G_u.$$

It takes some manipulation to carry out the various contributions, as will be discussed in detail elsewhere, but ultimately

$$\mathcal{G}_{ij} = \frac{\bar{\rho}}{\rho_0} G_{ij}^{(0)},$$

where $\bar{\rho}=\langle\rho\rangle$ is the physical average density, corrected at first order to be $\bar{\rho}=\rho_0/[1+\rho_0\beta V(q=0)]$. This follows from the perturbation theory analysis of G_i.

Turning to the dynamic direct correlation function, we have at first order

$$\frac{\delta}{\delta G_j}\Delta W = \frac{\delta}{\delta G_j}\sum_u F_u G_u = F_j = \sum_k \sigma_{jk}\phi_k. \tag{76}$$

Putting this result directly into the defining equation for the dynamic direct correlation function, Eq. (61), gives

$$c_{ij}^{(1)} = \tilde{T}r\phi_i \sum_k \sigma_{jk}\phi_k = \sum_k G_{ik}^{(0)}\sigma_{kj}. \tag{77}$$

The contribution to the two-point irreducible vertex is given by the very simple result

$$K_{ij}^{(1)} = -\sum_{k,\ell} \gamma_{i\ell}^{(0)} G_{\ell k}^{(0)} \sigma_{kj} = -\sigma_{ij}. \tag{78}$$

Let us look at the first-order theory for the two-point correlation function. It satisfies, where it is understood that ρ_0 is replaced by $\bar{\rho}$, the matrix kinetic equation

$$G_{ij} = G_{ij}^{(0)} + \sum_{k\ell} G_{ik}^{(0)}\sigma_{k\ell}G_{\ell j}. \tag{79}$$

This is a matrix equation which holds for times $t_i, t_j \geq t_0$ and the two-point cumulant is written more explicitly as

$$G_{ij} \to (2\pi)^d \delta(q_i + q_j) G_{\alpha_i\alpha_j}(q_i, t_i, t_j), \tag{80}$$

where α_i takes on the values ρ and B and the translational invariance of the system is reflected in the multiplying δ function.

Traditionally, there have been two recipes or protocols [50] for evaluating the two-point cumulant. Both reduce the problem to effectively a one-time problem. The kinetic theory protocol (KTP) is to treat the problem as an initial value problem with the system in equilibrium at time t_0 and we determine the single-time correlation function $G_{\rho\rho}(q, t_1 - t_0)$. This quantity is available in the current approach by setting $t_2=t_0$ in Eq. (59) and using Laplace transforms. Traditionally, one organizes kinetic theory via the time correlation function

$$C_{AB}(q,t) = \langle B_{-q} e^{i\hat{L}t} A_q \rangle, \tag{81}$$

where $\hat{L}$ is the Liouville operator [51] in the case of Newtonian dynamics.

In the second protocol, called the field theory protocol (FTP), one takes $t_0 \to -\infty$ and builds up the equilibrium structure from the noise. One of the technical advantages of this approach is that one can maintain time-translational invariance over the time line and there is only one time in the problem, $t_1 - t_2$. It is natural to work in terms of time Fourier transforms in this case. This allows one to understand the causal structure in terms of properties on the complex plane. Our theory here is similar to quantum many body theory where one builds up the equilibrium correlation using thermal Green's functions [52]. A difference is that in the quantum case in equilibrium one must satisfy the KMS boundary [53] conditions. Both protocols are included in the development here. It offers the opportunity of developing approximations that are internally self-consistent and one would prefer both procedures to produce the same results.

One can work out the full solution to the set of matrix equations given by Eq. (79) with the simple result

$$G_{\rho\rho}(q, t_1, t_2) = S(q)\tilde{F}(q, |t_1 - t_2|), \tag{82}$$

where

$$\tilde{F}(q,t) = e^{-\tilde{D}(q)q^2 t}. \tag{83}$$

The static structure factor [54] is given by

$$S(q) = \frac{\bar{\rho}}{[1 + \bar{\rho}\beta V(q)]} \tag{84}$$

and the physical wave number dependent diffusion coefficient is given by [55]

$$\tilde{D}(q) = D\beta\bar{\rho}S^{-1}(q). \tag{85}$$

Notice that we can, at this level of approximation, introduce the notion of an effective potential. Comparing Eq. (84) with the static Ornstein-Zernike [29] relation we can identify the effective interaction

$$V_{EFF}(q) = -\beta^{-1} c_D(q), \tag{86}$$

where $c_D(q)$ is the physical direct correlation function which is assumed to be known by other means. We can, for example, assume that $c_D(q)$ is given in the Percus-Yevick approximation for hard spheres [56]. With this effective interaction one can work out the results of perturbation theory in $V_{EFF}(q)$.

One can also use the two protocols discussed above to analyze Eq. (79). In the KTP one sets $t_2=t_0$ and notices that only retarded quantities remain in the kinetic equation which can be solved directly by taking the Laplace transform. In the FTP where one takes $t_0 \to -\infty$, ones sees, after taking the limit, all reference to the equilibrium static structure is gone and one has time translational invariance. After Fourier transforming over time, the equations are reduced to a set of algebraic equations which are simply inverted to give the same solution in the frequency regime. Inverting the Fourier transform leads back to results found from the complete two-time solution.

To demonstrate the versatility of the method, consider a system initially ($t=t_0$) in equilibrium at temperature T_I, but with noise driving the system at T for $t > t_0$. At first order in perturbation theory, the system still satisfies Eq. (79) but with the $t=t_0$ contribution in σ at temperature T. The solution of this problem is only slightly more complicated than the equilibrium case. One finally has the solution

$$G_{\rho\rho}(q,t_1,t_2) = S(q)\tilde{F}(q,|t_1-t_2|) + [S_0(q)-S(q)]\tilde{F}(q,t_1-t_0)\tilde{F}(q,t_2-t_0), \tag{87}$$

where $S_0(q)$ is the static structure factor at temperature T_I and $S(q)$ is the static structure factor at temperature T. Now time-translational invariance is broken but is restored as the system decays to equilibrium at temperature T.

It was claimed earlier that this method could provide self-consistent approximations for higher-order cumulants. Within the first-order theory one can generate expressions for the triplet correlation functions. We easily find for the first-order theory that

$$G_{ijk} = -\sum_{vup} G_{iv}G_{ju}G_{kp}\gamma^{(0)}_{vup}, \tag{88}$$

where these are the first order Gs on the right hand side and the zeroth-order three-point vertex is given by

$$\gamma^{(0)}_{vup} = -\sum_{vup} \gamma^{(0)}_{iv}\gamma^{(0)}_{ju}\gamma^{(0)}_{kp}G^{(0)}_{vup}, \tag{89}$$

where all zeroth-order quantities can be evaluated using Eqs. (65) and (72)–(75).

At second order in the effective potential we have two contributions to the two-point vertex. The first piece comes from the self-energy contribution to the dynamic Ornstein-Zernike equation, Eq. (61), where, keeping the second-order terms, we have

$$\frac{\delta}{\delta G_k}\Delta W^{(2)} = \frac{\delta}{\delta G_k}\sum_{ij} F_iF_jG_{ij} = -\sum_{ij} F_iF_j\sum_{\ell,p} G_{i\ell}\Gamma_{\ell kp}G_{pj} \tag{90}$$

and, in the simplest second-order approximation, we replace the three-point vertex with the zeroth-order result. After some simple manipulations that will be described in detail elsewhere, the second-order contributions to the two-point vertex coming from the self-energy can be written in the mode-coupling form

$$\Gamma^{MC}_{ij} = -\frac{1}{2}\sum_{kp\ell n} \gamma^{(0)}_{ik\ell}\delta G_{kp}\delta G_{\ell n}\gamma^{(0)}_{jpn}, \tag{91}$$

where the new element is that the high wave number convergence of the integrals comes not from the three point vertices but from the subtraction in [12(a)]

$$\delta G_{kp} = G_{kp} - G^{(0)}_{kp}. \tag{92}$$

In some field theoretical treatments of the DK model one obtains memory function kernels which do not vanish in the noninteracting limit. To complete the second-order model we must also work out the second-order terms coming from $\mathcal{G}$. This term is linear in the full two-point correlation function and if there is an ENE transition at this order, it could show the stretching phenomena found phenomenologically in Goetze's F_{12} model [57]. The complete second-order model will be treated in a separate publication.

VI. CONCLUSIONS

We have presented here a reformulation of kinetic theory which is self-consistent. It allows one to study problems which are difficult to treat using other methods.

(1) We outlined a clean derivation of the mode-coupling model at second order in perturbation theory. We will analyze whether this model supports ENE transitions elsewhere.

(2) The method presented here allows for a systematic method for analyzing corrections to this second-order result including higher-order correlation functions.

(3) The method allows one to treat nonequilibrium problems like temperature quenches as demonstrated above.

(4) Completely unexplored is the fact that the perturbation theory has been developed in the presence of space and time-dependent external fields. Thus this method could be useful in problems of optical pinning and highly inhomogeneous situations.

(5) It seems likely that this method will be useful in treating meta- and unstable systems. A first approach within perturbation theory is to formulate a dynamical van der Waals theory. Similarly it seems likely that this approach will be useful in developing a dynamic theory of melting once a few ideas from density-functional theory are integrated into the development.

(6) Our focus here, because of its simplicity, has been on Smoluchowski dynamics, but as will be discussed elsewhere, the method developed here can be applied both to Newtonian dynamics and Fokker-Planck dynamics as well as a broader class of models. In the case of Newtonian dynamics, this approach offers an alternative to the conventional development in terms of the Liouville operator. It may be important that in this approach it is not necessary to use basis states in perturbation theory labeled by a continuous momentum index.

(7) It is clear that we can study using these methods the mapping of Fokker-Planck dynamics onto Smoluchowski dynamics in the large mass limit. This is of course an elaborated version of the calculation leading to Einstein's relation [58] between the friction coefficient and the diffusion coefficient. Of particular interest to us is whether in integrating out of the momentum degrees of freedom one generates density dependent diffusion coefficients. The effects of such density dependence have been shown to be physically important within the random diffusion model [13].

(8) All of this development is compatible with the bizarre developments initiated with the introduction of ghost fermions [39] into the treatment of stochastic dynamics. We expect the subsequent developments like supersymmetry [40] to manifest themselves as in field theories where one can make a connection with Onsager's reciprocal relations [41] and the fluctuation theorems [42] found in the strongly nonequilibrium regime.

(9) Independent of the relevance of point 8, the connection to the fluctuation theorems and Jarzynski equalities should be explored carefully.

(10) These methods allow one to study one-point quantities in more complicated situations. In the calculation outlined above for homogeneous systems we did not mention the self-consistent determination of the equation of state be-

cause the results are rather dull. This will not be the case for inhomogeneous systems.

(11) It seems clear that a more quantitative method can be established if one determines the best way of expanding ΔW in a density expansion. This would make close contact to the work of Percus and the standard approximations of Percus-Yevick and hypernetted chain.

(12) This method allows us to investigate the claim by Das and Mazenko that momenta are associated with a mechanism which cuts off any possible ENE transition in conventional fluids.

(13) Even in the case of Smoluchowski dynamics, the connection between the theory at the microscopic level and fluctuating nonlinear hydrodynamics has not been established. Can the noninteracting field theory of Ref. [49] be extended to the interacting regime and connected up to the microscopic theory treated here?

(14) The existence of fluctuation-dissipation theorems is very important in equilibrium. This will be explored in depth elsewhere.

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APPENDIX A: LANGEVIN, FOKKER-PLANCK, AND MSR DYNAMICS

1. Langevin model

The Langevin model we study is given by

$$\frac{\partial R_i(t)}{\partial t} = DF_i(t) + \eta_i(t), \tag{A1}$$

which is valid for $t > t_0$ and has the initial condition

$$R_i(t_0) = R_i^{(0)} \tag{A2}$$

governed by the probability distribution $P_0[R_i^{(0)}]$. $F_i(t)$ is the force acting on $R_i(t)$ and is a local function of $R_i(t)$. We assume that the noise $\eta_i(t)$ is Gaussian white noise with variance

$$\langle \eta_i(t)\, \eta_j(t') \rangle = 2k_B T D \delta_{ij} \delta(t - t'), \tag{A3}$$

where D is a diffusion coefficient. The associated probability distribution is given by

$$P[\eta] = N_0 \exp\left[- \int_{t_0}^{\infty} dt \frac{\sum_i \eta_i^2(t)}{4\bar{D}} \right], \tag{A4}$$

where N_0 is a normalization factor. The partition function in an external field is given by

$$Z[h] = \int \mathcal{D}\eta P[\eta] \int \mathcal{D}R^{(0)} P_0[R^{(0)}] S(h \cdot R), \tag{A5}$$

where the time-dependent external field couples to the system via

$$S(h \cdot R) = \exp\left[\int_{t_0}^{\infty} dt \sum_{i=1}^{N} R_i(t) h_i(t) \right] \tag{A6}$$

and where, via the Langevin equation and its initial condition, the field R_i is a functional of the noise and the initial condition $R_i^{(0)}$. Our goal is to determine the generator of cumulants, $W[h] = \ln Z[h]$.

2. Fokker-Planck dynamics

It is convenient to analyze this set of dynamical models using the Fokker-Planck description. We follow here the development of Kim and Mazenko [59]. If we define the field

$$g_\phi(t) = \prod_{i=1}^{N} \delta[\phi_i - R_i(t)], \tag{A7}$$

we may then consider the time correlation functions

$$G_{\phi,\phi'}(t) = \langle g_\phi(t) g_{\phi'} \rangle, \tag{A8}$$

where the average is over the noise and the initial condition.

Taking the time derivative of $G_{\phi,\phi'}(t)$ using the chain rule for differentiation and the Langevin equation, Eq. (A1), one is left with

$$\frac{\partial}{\partial t} G_{\phi,\phi'}(t) = - \sum_i \frac{\delta}{\delta \phi_i} \left[F_i(\phi) G_{\phi,\phi'}(t) - \sum_i \langle \eta_i(t) g_\phi(t) g_{\phi'} \rangle \right]. \tag{A9}$$

It is then not difficult to show, remembering that η is Gaussian, that

$$\langle \eta_i(t) g_\phi(t) g_{\phi'} \rangle = - \bar{D} \frac{\delta}{\delta \phi_i} G_{\phi,\phi'}(t), \tag{A10}$$

where we have used $\bar{D} = k_B T D$, the result

$$\frac{\delta R_i(t)}{\delta \eta_i(t)} = \frac{1}{2} \delta_{ij}, \tag{A11}$$

which follows from Eq. (A1) and the assumption that the initial field configuration is independent of the noise for $t \geq 0$,

$$\frac{\delta R_i(t_0)}{\delta \eta_i(t)} = 0. \tag{A12}$$

Using Eq. (A10) back into Eq. (A9) we can write

$$\frac{\partial}{\partial t} G_{\phi,\phi'}(t) = D_\phi G_{\phi,\phi'}(t), \tag{A13}$$

where the Fokker-Planck operator is defined by

$$D_\phi = D\sum_i \frac{\delta}{\delta\phi_i}\left[-F_i(\phi) + k_B T\frac{\delta}{\delta\phi_i}\right]. \tag{A14}$$

The formal solution to Eq. (A13) is

$$G_{\phi,\phi'}(t-t_0) = e^{D_\phi(t-t_0)}G_{\phi,\phi'}(0) = e^{D_\phi(t-t_0)}[\delta(\phi-\phi')P_0(\phi')]. \tag{A15}$$

Integrating over all ϕ' gives the equilibrium probability distribution

$$P(\phi,t) = \int \mathcal{D}(\phi')e^{D_\phi(t-t_0)}[\delta(\phi-\phi')P_0(\phi')] = e^{D_\phi(t-t_0)}P_0(\phi). \tag{A16}$$

It is easy to see that the equilibrium solution is given by

$$P_\phi(0) \equiv W_\phi = \frac{e^{-\beta\mathcal{H}_\phi}}{Z}, \tag{A17}$$

where

$$F_i = -\frac{\partial\mathcal{H}}{\partial\phi_i}. \tag{A18}$$

Equation (A15) then takes the form

$$G_{\phi,\phi'}(t) = e^{D_\phi t}[\delta(\phi-\phi')W_\phi]. \tag{A19}$$

Any two-time correlation can then be written in the form

$$\begin{aligned} C_{AB}(t) &= \langle A[R(t)]B[R(0)]\rangle \\ &= \int \mathcal{D}\phi\mathcal{D}\phi' A(\phi')B(\phi)G_{\phi,\phi'}(t) \\ &= \int \mathcal{D}\phi B(\phi)e^{D_\phi t}A(\phi)W_\phi, \end{aligned} \tag{A20}$$

where in the final step we assume the system is in equilibrium.

In our development it is useful to introduce the adjoint Fokker-Planck operator

$$\tilde{D}_\phi = D\sum_i \left[F_i - k_B T\frac{\delta}{\delta\phi_i}\right]\frac{\delta}{\delta\phi_i}. \tag{A21}$$

If the equilibrium average is defined as

$$\langle A\rangle = \int \mathcal{D}(\phi)W_\phi A(\phi), \tag{A22}$$

one can show that

$$C_{AB}(t) = \int \mathcal{D}(\phi)B(\phi)e^{D_\phi t}(A(\phi)W_\phi) = \langle B(\phi)e^{\tilde{D}_\phi t}A(\phi)\rangle. \tag{A23}$$

3. Multiple time correlations

Consider the multiple time correlation function

$$\begin{aligned} &G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_n}(t_0,t_1,t_2,\ldots,t_n) \\ &\quad = \langle g_{\phi_n}(t_n)\ldots g_{\phi_2}(t_2)g_{\phi_1}(t_1)g_{\phi_0}(t_0)\rangle, \end{aligned} \tag{A24}$$

where $g_\phi(t)$ is defined by Eq. (A7) and we assume $t_n \geq t_{n-1} \geq \cdots \geq t_2 \geq t_1 \geq t_0$. Using the same approach as used for treating the two-point quantity we have

$$\begin{aligned} &\frac{\partial}{\partial t_n}G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_n}(t_0,t_1,t_2,\ldots,t_n) \\ &\quad = -\sum_i \frac{\delta}{\delta\phi_i}\Big[DF_i G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_n}(t_0,t_1,t_2,\ldots,t_n) \\ &\qquad - \sum_i k_B T\langle\eta_i(t_n)g_{\phi_{n-1}}(t_{n-1})\cdots g_{\phi_2}(t_2)g_{\phi_1}(t_1)g_{\phi_0}(t_0)\rangle\Big]. \end{aligned} \tag{A25}$$

Because of causality, $g_{\phi_i}(t_i)$ for $t_i < t_n$ is independent of the noise at t_n,

$$\begin{aligned} &\langle\eta_i(t_n)g_{\phi_n}(t_n)\cdots g_{\phi_2}(t_2)g_{\phi_1}(t_1)g_{\phi_0}(t_0)\rangle \\ &\quad = \bar{D}\left\langle\frac{\delta g_{\phi_n}(t_n)}{\delta\eta_{\alpha_n}(t_n)}g_{\phi_{n-1}}(t_{n-1})\cdots g_{\phi_2}(t_2)g_{\phi_1}(t_1)g_{\phi_0}(t_0)\right\rangle. \end{aligned} \tag{A26}$$

The treatment of $\frac{\delta g_{\phi_n}(t_n)}{\delta\eta_n(t_n)}$ is the same as the two-time case and we obtain the result

$$\begin{aligned} &\frac{\partial}{\partial t_n}G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_n}(t_0,t_1,t_2,\ldots,t_n) \\ &\quad = D_{\phi_n}G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_n}(t_1,t_2,\ldots,t_n), \end{aligned} \tag{A27}$$

where D_ϕ is the Fokker-Planck operator given by Eq. (A14). This has the formal solution

$$\begin{aligned} &G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1},\phi_n}(t_0,t_1,t_2,\ldots t_{n-1},t_n) \\ &\quad = e^{D_{\phi_n}(t_n-t_{n-1})}G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1},\phi_n}(t_0,t_1,t_2,\ldots t_{n-1},t_{n-1}). \end{aligned} \tag{A28}$$

However,

$$\begin{aligned} &G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1},\phi_n}(t_0,t_1,t_2,\ldots,t_{n-1},t_{n-1}) \\ &\quad = \langle g_{\phi_n}(t_{n-1})g_{\phi_{n-1}}(t_{n-1})\cdots g_{\phi_2}(t_2)g_{\phi_1}(t_1)g_{\phi_0}(t_0)\rangle \\ &\quad = \delta(\phi_n-\phi_{n-1})G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1}}(t_0,t_1,t_2,\ldots t_{n-1}). \end{aligned} \tag{A29}$$

Clearly we can work this out recursively:

$$G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1},\phi_n}(t_0,t_1,t_2,\ldots,t_{n-1},t_n) = e^{D_{\phi_n}(t_n-t_{n-1})}\delta(\phi_n-\phi_{n-1})G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1}}(t_0,t_1,t_2,\ldots,t_{n-1})$$
$$= e^{D_{\phi_n}(t_n-t_{n-1})}\delta(\phi_n-\phi_{n-1})e^{D_{\phi_{n-1}}(t_{n-1}-t_{n-2})}\delta(\phi_{n-1}-\phi_{n-2})G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-2}}(t_0,t_1,t_2,\ldots,t_{n-2})$$
$$= e^{D_{\phi_n}(t_n-t_{n-1})}e^{D_{\phi_{n-1}}(t_{n-1}-t_{n-2})}\cdots e^{D_{\phi_2}(t_2-t_1)}e^{D_{\phi_1}(t_1-t_0)}$$
$$\times\delta(\phi_n-\phi_{n-1})\delta(\phi_{n-1}-\phi_{n-2})\cdots\delta(\phi_1-\phi_0)W_{\phi_0}. \tag{A30}$$

If we introduce the notation

$$U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1}) = e^{D_{\phi_n}(t_n-t_{n-1})}\delta(\phi_n-\phi_{n-1}), \tag{A31}$$

we can write

$$G_{\phi_0,\phi_1,\phi_2,\ldots,\phi_{n-1},\phi_n}(t_0,t_1,t_2,\ldots,t_{n-1},t_n)$$
$$= U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1})U_{\phi_{n-1};\phi_{n-2}}(t_{n-1}-t_{n-2})\cdots$$
$$\times U_{\phi_2;\phi_1}(t_2-t_1)U_{\phi_1;\phi_0}(t_1-t_0)W_{\phi_0}. \tag{A32}$$

This is the result we need in developing the path-integral approach.

4. Path integral form

How is the partition function in a field related to these multiple-time correlations? In $Z[h]$, given by Eq. (A5), we make the special choice for the external field

$$h_i(t) = \sum_{s=0}^{\ell}[i\lambda_i^s + h_i^s]\delta(t-t_s), \tag{A33}$$

which amounts to dividing up the time interval into a grid. Next, multiply by

$$\prod_{s=0}^{\ell} e^{-\lambda_i^s\phi_i^s}$$

and integrate over λ_i^s. Then, we have

$$\int \mathcal{D}\lambda\prod_{s=0}^{\ell} e^{-i\lambda_i^s\phi_i^s}Z\left[\sum_{s=0}^{\ell}[i\lambda_i^s+h_i^s]\delta(t-t_s)R_i(t_s)\right]$$
$$= \int \mathcal{D}\eta P[\eta]\int \mathcal{D}R^{(0)}P_0[R^{(0)}]$$
$$\times\int \mathcal{D}\lambda\prod_{s=0}^{\ell} e^{-i\lambda_i^s\phi_i^s}e^{i\lambda_i^sR_i(t_s)}e^{\Sigma_{s,i}h_i^sR_i(t_s)}$$
$$= \int \mathcal{D}\eta P[\eta]\int \mathcal{D}R^{(0)}P_0[R^{(0)}]\prod_{s=0}^{\ell} g_{\phi_s}(t_s)e^{\Sigma_{s,i}h_i^sR_i(t_s)}, \tag{A34}$$

where again the g_ϕ are δ-functions which allow us to make the replacement

$$e^{\Sigma_{s,i}h_i^sR_i(t_s)} = e^{\Sigma_{s,i}h_i^s\phi_i^s} = S[h\cdot\phi], \tag{A35}$$

which comes out from the average over noise and initial conditions. We have

$$\int \mathcal{D}\lambda\prod_{s=0}^{\ell} e^{-i\lambda_i^s\phi_i^s}Z\left[\sum_{s=0}^{\ell}[i\lambda_i^s+h_i^s]\delta(t-t_s)R_i(t_s)\right]$$
$$= S[h\cdot\phi]G_{\phi_0,\phi_1,\ldots,\phi_n}(t_0,t_1,\ldots,t_n). \tag{A36}$$

We finally obtain the result for the partition function we want by doing the functional integral over ϕ

$$Z[h] = \int \mathcal{D}\phi S[h\cdot\phi]G_{\phi_0,\phi_1,\ldots,\phi_n}(t_0,t_1,\ldots,t_n). \tag{A37}$$

The key here is to notice that we have an explicit expression for the multitime correlation function $G_{\phi_0,\phi_1,\ldots\phi_n}(t_0,t_1,\ldots,t_n)$. Inserting this result into Eq. (A37), we obtain

$$Z[h] = \int \mathcal{D}\phi S[h\cdot\phi]U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1})$$
$$\times U_{\phi_{n-1};\phi_{n-2}}(t_{n-1}-t_{n-2})\cdots$$
$$\times U_{\phi_2;\phi_1}(t_2-t_1)U_{\phi_1;\phi_0}(t_1-t_0)W_{\phi_0}. \tag{A38}$$

There are no constraints on the choice of time slices. Let us take the slices to be uniformly divided, $t_{s+1}=t_s+\Delta$, and we work in the limit of small Δ and large n. This defines the continuum limit for the theory. As a check on the development, notice that the normalization as $h\to 0$ is preserved,

$$Z[0] = \int \mathcal{D}\phi U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1})U_{\phi_{n-1};\phi_{n-2}}(t_{n-1}-t_{n-2})\cdots$$
$$\times U_{\phi_2;\phi_1}(t_2-t_1)U_{\phi_1;\phi_0}(t_1-t_0)W_{\phi_0} = 1. \tag{A39}$$

The result given by Eq. (A38) is naturally interpreted as a path-integral. Let us focus on the intermediate time quantities

$$U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1}) = e^{D_{\phi_n}(t_n-t_{n-1})}\delta(\phi_n-\phi_{n-1}). \tag{A40}$$

Using the integral representation for the δ-function we can diagonalize the Fokker-Planck operator,

$$U_{\phi_n;\phi_{n-1}}(t_n-t_{n-1}) = e^{D_{\phi_n}(t_n-t_{n-1})}\int d\hat{\phi}_n e^{i\hat{\phi}_n(\phi_n-\phi_{n-1})}. \tag{A41}$$

Then, we can evaluate

$$D_{\phi_n} e^{i\hat{\phi}_n \phi_n} = \frac{\delta}{\delta \phi_n}\left[-F_n(\phi) + \bar{D}\frac{\delta}{\delta \phi_n}\right] e^{i\hat{\phi}_n \phi_n} = A_n^F e^{i\hat{\phi}_n \phi_n}, \tag{A42}$$

where

$$A_n^F = -i\hat{\phi}_n F_n(\phi) - \bar{D}\hat{\phi}_n^2 - \frac{\delta}{\delta \phi_n} F_n(\phi) \tag{A43}$$

and

$$U_{\phi_n;\phi_{n-1}}(t_n - t_{n-1}) = \int d\hat{\phi}_n e^{A_n^F \Delta} e^{i\hat{\phi}_n(\phi_n - \phi_{n-1})} = \int d\hat{\phi}_n e^{A_n \Delta}, \tag{A44}$$

where

$$A_n = -\bar{D}\hat{\phi}_n^2 + i\hat{\phi}_n\left[(\phi_n - \phi_{n-1})/\Delta - F_n(\phi) - \frac{\delta F_n}{\delta \phi_n}\right]. \tag{A45}$$

Putting Eq. (A45) back into Eq. (A38), one has

$$Z[h] = \int \mathcal{D}\phi \mathcal{D}\hat{\phi} \mathcal{D}\phi_0 P_0(\phi_0) e^{\int_{t_0}^{\infty} dt A_R}, \tag{A46}$$

where A_R is the standard MSR action in the presence of an external field,

$$A_R = -\bar{D}\hat{\phi}^2(t) + i\hat{\phi}(t)\{\dot{\phi}(t) - F[\phi(t)]\} + h(t)\phi(t) - \frac{\delta F_\phi(t)}{\delta \phi(t)}. \tag{A47}$$

APPENDIX B: GAUSSIAN SINGLE-PARTICLE PROBLEM

The noninteracting correlations for a system driven by Smoluchowski dynamics is governed by the MSR action

$$A_0 = \int_{t_0}^{\infty} dt[\hat{R}(t)\bar{D}\hat{R}(t) + i\hat{R}(t)\dot{R}(t) - h(t)R(t) - \hat{h}(t)\hat{R}(t)], \tag{B1}$$

where $h(t)$ and $\hat{h}(t)$ are the external source fields and $\bar{D}=k_B T D$. We then have the identities that hold in the range $t_0 < t < \infty$,

$$\int \mathcal{D}(R)\mathcal{D}(\hat{R}) d^d R_0 P_0[R_0] \frac{\delta}{\delta R(t)} e^{-A_0} = 0 \tag{B2}$$

and

$$\int \mathcal{D}(R)\mathcal{D}(\hat{R}) d^d R_0 P_0[R_0] \frac{\delta}{\delta \hat{R}(t)} e^{-A_0} = 0. \tag{B3}$$

Let us begin with the initial condition

$$P_0[R_0] = \delta(R_0 - X_0). \tag{B4}$$

Evaluating the derivatives of A_0, we obtain

$$2\bar{D}\hat{G}(t) + i\frac{\partial G(t)}{\partial t} = \hat{h}(t). \tag{B5}$$

and

$$-i\frac{\partial \hat{G}(t)}{\partial t} = h(t), \tag{B6}$$

where

$$G(t) = \langle R(t)\rangle_{X_0} \tag{B7}$$

and

$$\hat{G}(t) = \langle \hat{R}(t)\rangle_{X_0} \tag{B8}$$

where the averages over $R(t)$ and $\hat{R}(t)$ are in the range $t_0 < t$. We must now solve these equations to obtain the generating functional.

Using the initial condition $\hat{R}(t_0)=0$, we find that

$$\hat{G}(t) = -i\int_t^{\infty} d\bar{t} h(\bar{t}) = \int_{t_0}^{\infty} d\tau g(\tau, t) h(\tau) \tag{B9}$$

and

$$\begin{aligned} G(t) &= X_0 + \int_{t_0}^{t} d\bar{t}[2i\bar{D}\hat{G}(\bar{t}) - i\hat{h}(\bar{t})] \\ &= X_0 - i\int_{t_0}^{t} d\bar{t}\hat{h}(\bar{t}) + 2\bar{D}\int_{t_0}^{t} d\bar{t}\int_{\bar{t}}^{\infty} d\bar{t}' h(\bar{t}') \\ &= X_0 + \int_{t_0}^{\infty} d\tau g(t,\tau)\hat{h}(\tau) + \int_{t_0}^{\infty} d\tau C(t,\tau) h(\tau), \end{aligned} \tag{B10}$$

where

$$g(t,t') = -i\theta(t - t') \tag{B11}$$

and

$$C(t,t') = 2\bar{D}\int_{t_0}^{t} d\bar{t}\int_{\bar{t}}^{\infty} d\bar{t}' \delta(t' - \bar{t}'). \tag{B12}$$

The generating functional satisfies

$$\hat{G}(t) = \frac{\delta \ln Z_0(h,\hat{h},X_0)}{\delta \hat{h}(t)} \tag{B13}$$

and

$$G(t) = \frac{\delta \ln Z_0(h,\hat{h},X_0)}{\delta h(t)}. \tag{B14}$$

The generating functional solution to this set of equations [Eqs. (B9), (B10), (B13), and (B14)] is given by

$$\ln Z_0(h,\hat{h},X_0) = \frac{1}{2}\int dt \int dt' h(t)C(t,t')h(t') + \int dt \int dt' h(t)g(t,t')\hat{h}(t') + \int dt h(t)X_0. \tag{B15}$$

The full generator requires averaging over the initial conditions,

$$Z[h,\hat{h}] = \int d^dR_0 P_0[R_0] e^{(1/2)h\cdot C\cdot h + h\cdot g\cdot\hat{h} + h\cdot i g\cdot R_0} = e^{(1/2)h\cdot C\cdot h + h\cdot g\cdot\hat{h}} \int d^dR_0 P_0[R_0] e^{h\cdot i g\cdot R_0}. \tag{B16}$$

All of the equilibrium cumulants can be constructed from Eq. (B12) as

$$\begin{aligned} C(t,t') &= 2\bar{D}\int_{t_0}^{t} d\bar{t}\int_{\bar{t}}^{\infty} d\bar{t}'\,\delta(t'-\bar{t}') \\ &= 2\bar{D}\int_{t_0}^{t} d\bar{t}\,\theta(t'-\bar{t}) \\ &= 2\bar{D}\,\theta(t-t')\int_{t_0}^{t'} d\bar{t} + 2\bar{D}\,\theta(t'-t)\int_{t_0}^{t} d\bar{t} \\ &= 2\bar{D}\,\theta(t-t')(t'-t_0) + 2\bar{D}\,\theta(t'-t)(t-t_0). \end{aligned} \tag{B17}$$

APPENDIX C: COLLECTIVE ϕ CORRELATIONS

We need to evaluate ϕ-correlations in the noninteracting case. We start with

$$W_0[H] = \tilde{T}re^{H\cdot\phi}, \tag{C1}$$

which generates all $\phi=(\rho,B)$ correlations. We proceed by reintroducing the microscopic sources h and $\hat{h}$ and treating

$$Z_0[H,h,\hat{h}] = \tilde{T}re^{H\cdot\phi}e^{h\cdot R+\hat{h}\cdot\hat{R}}, \tag{C2}$$

where

$$h\cdot R = \int_{t_0}^{\infty} dt h(t)R(t). \tag{C3}$$

Next, we express the ϕ in terms of $R(t)$ and $\hat{R}(t)$ as

$$\phi_\rho(1) = e^{-ik_1R(t_1)} \tag{C4}$$

and

$$\phi_B(1) = -D[(k_1\cdot\hat{R}(1) + \theta(0)k_1^2)\phi_\rho(1)]. \tag{C5}$$

Let us introduce the operators

$$\hat{\phi}_\rho(1) = e^{-ik_1(\delta/\delta h(t_1))} \tag{C6}$$

and

$$\hat{\phi}_B(1) = \hat{b}(1)\hat{\phi}_\rho(1), \tag{C7}$$

where

$$\hat{b}(1) = -D\left[k_1\frac{\delta}{\delta\hat{h}(t_1)} + k_1^2\theta(0)\right] \tag{C8}$$

so we may write

$$e^{H\cdot\phi}e^{h\cdot R+\hat{h}\cdot\hat{R}} = e^{H\cdot\hat{\phi}}e^{h\cdot R+\hat{h}\cdot\hat{R}} \tag{C9}$$

and

$$Z_0[H,h,\hat{h}] = e^{H\cdot\hat{\phi}}Z_0[h,\hat{h}], \tag{C10}$$

where $Z_0[h,\hat{h}]$ was determined in Appendix B.

Taking functional derivatives, we can determine all of the noninteracting cumulants of the complete set of densities ϕ. We have

$$\begin{aligned} G_{B,\ldots,B\rho,\ldots,\rho}(1,\ldots,\ell,\ell+1,\ldots,n) &= \frac{\delta}{\delta H_B(1)}\cdots\frac{\delta}{\delta H_B(\ell)}\frac{\delta}{\delta H_\rho(\ell+1)}\cdots\frac{\delta}{\delta H_\rho(n)} \\ &\quad\times e^{H\cdot\hat{\phi}}e^{1/2h\cdot C\cdot h}e^{h\cdot g\cdot\hat{h}}\int d^dR_0P_0[R_0]e^{h\cdot i g\cdot R_0}\big|_{H=h=\hat{h}=0} \\ &= \hat{\phi}_B(1)\cdots\hat{\phi}_B(\ell)\hat{\phi}_\rho(\ell+1)\cdots\hat{\phi}_\rho(n) \\ &\quad\times e^{(1/2)h\cdot C\cdot h}e^{h\cdot g\cdot\hat{h}}\int d^dR_0P_0[R_0]e^{h\cdot i g\cdot R_0}\big|_{H=h=\hat{h}=0}. \end{aligned} \tag{C11}$$

All n-point cumulants have n factors of $\hat{\phi}_\rho$ and ℓ factors of $\hat{b}$ corresponding to the number of B insertions,

$$\begin{aligned} G_{B,\ldots,B\rho,\ldots,\rho}(1\ldots,\ell,\ell+1,\ldots,n) &= \hat{b}(1)\cdots\hat{b}(\ell)\hat{\phi}_\rho(1)\cdots\hat{\phi}_\rho(n)e^{1/2h\cdot C\cdot h}e^{h\cdot g\cdot\hat{h}} \\ &\quad\times\int d^dR_0P_0[R_0]e^{h\cdot i g\cdot R_0}\big|_{h=\hat{h}=0}. \end{aligned} \tag{C12}$$

Because the $\hat{\phi}_\rho(j)$ are translation operators, it is not difficult to show that

$$\hat{\phi}_\rho(1)\ldots\hat{\phi}_\rho(n)F[h(j)] = F[h(j)+L_n(j)], \tag{C13}$$

where

$$L_n(j) = -i\sum_{s=1}^{n} k_s\delta(t_j-t_s). \tag{C14}$$

Thus, we have

$$G_{B,\ldots,B\rho,\ldots,\rho}(1,\ldots,\ell,\ell+1,\ldots,n)$$
$$= \hat{b}(1)\cdots\hat{b}(\ell)e^{(1/2)(h+L_n)\cdot C\cdot(h+L_n)}e^{(h+L_n)\cdot g\cdot\hat{h}}$$
$$\times\int d^dR_0P_0[R_0]e^{(h+L_n)\cdot ig\cdot R_0}|_{h=\hat{h}=0}$$
$$= \hat{b}(1)\cdots\hat{b}(\ell)e^{(1/2)L_n\cdot C\cdot L_n}e^{L_n\cdot g\cdot\hat{h}}\int d^dR_0P_0[R_0]e^{L_n\cdot ig\cdot R_0}|_{\hat{h}=0}$$
$$= \hat{b}(1)\cdots\hat{b}(\ell)e^{N_n}e^{L_n\cdot g\cdot\hat{h}}\int d^dR_0P_0[R_0]e^{L_n\cdot ig\cdot R_0}|_{\hat{h}=0} \quad \text{(C15)}$$

where we have defined

$$N_n = \frac{1}{2}L_n\cdot C\cdot L_n. \quad \text{(C16)}$$

Using the definition of $\hat{b}$, we similarly find that

$$\hat{b}(j)e^{L_n\cdot g\cdot\hat{h}} = b_n(j)e^{L_n\cdot g\cdot\hat{h}}, \quad \text{(C17)}$$

where

$$b_n(j) = -D[k_jL_n(\bar{t})g(\bar{t}-t_j) - \theta(0)k_j^2]$$
$$= Dk_j\sum_{s=1}^{n}k_s\theta(t_s-t_j) - D\theta(0)k_j^2$$
$$= Dk_j\sum_{s\neq j=1}^{n}k_s\theta(t_s-t_j). \quad \text{(C18)}$$

In the average over initial conditions, we need

$$L_n\cdot ig = -i\sum_{s=1}^{n}k_s \quad \text{(C19)}$$

and

$$\int d^dR_0P_0[R_0]e^{L_n\cdot ig\cdot R_0} = \int d^dR_0P_0[R_0]\exp\left[-i\left(\sum_{s=1}^{n}k_s\right)R_0\right]$$
$$= (2\pi)^d\delta\left(\sum_{s=1}^{n}k_s\right), \quad \text{(C20)}$$

which enforces translational invariance in space. Finally, putting this all together we have

$$G_{B,\ldots,B\rho,\ldots,\rho}(1,\ldots,\ell,\ell+1,\ldots,n)$$
$$= \rho_0b(1)\cdots b(\ell)e^{N_n}(2\pi)^d\delta\left(\sum_{s=1}^{n}k_s\right). \quad \text{(C21)}$$

The argument of the exponential contribution can be put into a more symmetric form. Starting with N_n given by Eq. (C16) and inserting L_n from Eq. (C14), we have

$$N_n = -\frac{1}{2}\sum_{i=1}^{n}\sum_{j=1}^{n}k_ik_jC(t_i,t_j). \quad \text{(C22)}$$

The zeroth-order correlation function for the Brownian coordinates is given by Eq. (B17) as

$$C(t,t') = 2\bar{D}[\theta(t-t')(t'-t_0) + \theta(t'-t)(t-t_0)], \quad \text{(C23)}$$

which, at equal times, reduces to

$$C(t,t) = 2\bar{D}(t-t_0). \quad \text{(C24)}$$

In the zeroth-order density correlation functions we find the quantity

$$N_n = -\frac{1}{2}\sum_{i,j}^{n}k_ik_jC(t_i,t_j) \quad \text{(C25)}$$

with the constraint that $\Sigma_ik_i=0$. This quantity should be time translationally invariant.

To see this, let us first define

$$D_{ij} \equiv C_{ii} + C_{jj} - 2C_{ij}$$
$$= 2\bar{D}\{t_i + t_j - 2[\theta(t_i-t_j)t_j + \theta(t_j-t_i)t_i]\} \quad \text{(C26)}$$

$$= 2\bar{D}\{t_i[\theta(t_i-t_j) - \theta(t_j-t_i)] + t_j[\theta(t_j-t_i) - \theta(t_i-t_j)]\}$$
$$= 2\bar{D}(t_i-t_j)\mathrm{sgn}(t_i-t_j) = 2\bar{D}|t_i-t_j|. \quad \text{(C27)}$$

Notice that this result holds if $t\to t-t_0$ in Eq. (C26).

We then have for the argument of the exponential for the $\rho-B$ correlation functions

$$N_n = -\frac{1}{2}\sum_{i,j}^{n}k_ik_jC(t_i,t_j)$$
$$= -\frac{1}{2}\left(\sum_{i=1}^{n}k_i^2C_{ii} + \sum_{i\neq j}k_ik_jC_{ij}\right)$$
$$= -\frac{1}{2}\left(\sum_{i=1}^{n}k_i^2C_{ii} + \sum_{i\neq j}k_ik_j\frac{1}{2}[C_{ii}+C_{jj}-D_{ij}]\right)$$
$$= -\frac{1}{2}\left(\sum_{i=1}^{n}k_i^2C_{ii} - \sum_{i=1}^{n}k_i^2C_{ii} - \frac{1}{2}\sum_{i\neq j}k_ik_jD_{ij}\right)$$
$$= \bar{D}\frac{1}{2}\sum_{i\neq j}k_ik_j|t_i-t_j|. \quad \text{(C28)}$$

For the special case of $n=2$, we have

$$N_2 = -\bar{D}k_1^2|t_1-t_2|. \quad \text{(C29)}$$

[1] By kinetic theory we mean the statistical dynamics of N classical particles. See G. Mazenko, *Nonequilibrium Statistical Mechanics* (Wiley, New York, 2006) for a discussion of kinetic theory including the modern correlation function approach.

[2] Brownian motion is interpreted here as a dynamics that is noise driven. For an individual Brownian particle located at position R acted upon by noise η, one has $\dot{R}=\eta$. This is clearly the continuum version of the random-walk problem.

[3] Field theory methods enter via the techniques for organizing self-consistent perturbation theory in terms of the roles of external fields, cumulants, and irreducible vertex functions. The fundamental reference is J. Zinn-Justin, *Quantum Field Theory and Critical Phenomena*, 4th ed. (Clarendon, Oxford, 2002). We henceforth refer to this reference as JZJ.

[4] Kinetic theory's early history is briefly reviewed in G. Mazenko, *Equilibrium Statistical Mechanics* (Wiley, New York, 2000), p. 101.

[5] By self-consistency we mean that the correlation and response functions of interest satisfy kinetic equations where the collision kernels, self-energies, or memory functions can be expressed in terms of the full physical correlation and response functions. In the areas of critical dynamics and the liquid-glass transition, this is essential in generating symmetry breaking solutions. This can generate a multiplicity of solutions not available in bare perturbation theory.

[6] Boltzmann's Stosszahlansatz is a prime example of a decoupling approximation. In the collision integral of the Boltzmann equation he assumed that the two-particle distribution function factorizes into a product of two one-particle distributions, $f_2 \to f_1 f_1$.

[7] L. Boltzmann, Math-Naturwiss, KI **66**, 275 (1872); English translation in S. G. Brush, *Kinetic Theory* (Pergamon, New York, 1966), Vol. 2.

[8] Besides Boltzmann's establishment of the H-theorem, there are many examples of decoupling factorization approximations contributing to advancing our understanding of a phenomena. Examples include, long-time tails in fluids. M. H. Ernst, E. H. Hauge, and J. M. J. van Leeuwen, Phys. Rev. Lett. **25**, 1254 (1970); K. Kawasaki, Phys. Lett. A **32**, 379 (1970); Prog. Theor. Phys. **45**, 1691 (1971); Y. Pomeau and P. Resibois, Phys. Rep. **19**, 63 (1975); and dynamic critical phenomena: M. Fixman, J. Chem. Phys. **36**, 310 (1962); L. P. Kadanoff and J. Swift, Phys. Rev. **166**, 89 (1968); K. Kawasaki, Ann. Phys. (N.Y.) **61**, 1 (1970).

[9] The role of mode-coupling theory has been characterized by E. Zaccarelli, G. Foffi, F. Sciortino, P. Tartaglia, and K. A. Dawson, Europhys. Lett. **55**, 157 (2001). In the weak supercooling regime, detailed predictions for the space and time dependence of the long-time decay of density correlations have been formulated using the ideal mode-coupling theory MCT, one of the first approaches to identify the existence of the crossover temperature. The agreement of MCT predictions with experimental findings and molecular dynamics simulations both for atomic and molecular models supports the view that MCT is indeed able to describe the slow dynamics in weak supercooled states.

[10] W. Goetze, in *Liquids, Freezing, and Glass Transition*, edited by J. P. Hansen, D. Levesque, and J. Zinn-Justin (North-Holland, Amsterdam, 1991); and S. Das, Rev. Mod. Phys. **76**, 785 (2004).

[11] The important idea of an ergodic-nonergodic (ENE) transition exists independent of whether it conforms to the details of mode-coupling theory. The defining property of an ENE transition is that as a function of a control parameter there is crossover from an ergodic phase $[\lim_{t\to\infty} G_{\rho\rho}(q,t)=0]$ to a region with nonergodic kinetics $[\lim_{t\to\infty} G_{\rho\rho}(q,t)=A^2(q)>0]$.

[12] Kinetic theory led to the early development of MCT. See for example G. F. Mazenko, Phys. Rev. A **7**, 209 (1973); L. Sjogren and A. Sjolander, J. Phys. C **12**, 4369 (1979); E. Leutheusser, Phys. Rev. A **29**, 2765 (1984); U. Bengtzelius, W. Goetze, and A. Sjolander, J. Phys. C **17**, 5915 (1984); More recent efforts to make microscopic contact (derive) mode-coupling theory are reflected in Ref. [9] where they comment: "Despite its remarkable practical success, the presence of apparently uncontrolled approximations in the derivation of the MCT equations makes it difficult to gain insights into possible improvements of the theory. The aim of this paper is to present a new derivation of the ideal MCT equations, starting from the microscopic equations for the evolution of the density (Newtons equations) and writing them as a linear generalized Langevin equation. A formally exact expression for the memory kernel is derived and, on making the approximation that the noise in the Langevin equation is Gaussian, the standard MCT equations are obtained. Note that the proposition of Gaussian noise implies that the density fluctuations are also Gaussian."; Similarly in J. Wu and J. Cao, Phys. Rev. E **67**, 061116 (2003) the authors make contact with MCT using a decoupling approximation which treats the density as a gaussian variable. Their characterization of the status of the theory is: "Although successful, the standard mode-coupling approximation has not been obtained in a systematic and straightforward fashion. A simple understanding of mode-coupling effects and their validity for describing low-temperature dynamics is still lacking. In this paper, we explore an alternative route to obtaining ideal mode-coupling equations via the direct Gaussian factorization of the multiple-point correlation function in the memory kernel."

[13] Field theory models for the glass transition are treated in S. P. Das and G. F. Mazenko, Phys. Rev. A **34**, 2265 (1986); Phys. Rev. E **79**, 021504 (2009); G. F. Mazenko, *ibid.* **78**, 031123 (2008); D. S. Dean, J. Phys. A **29**, L613 (1996); K. Kawasaki and S. Miyazima, Z. Phys. B: Condens. Matter **103**, 423 (1997); K. Miyazaki and D. R. Reichman, J. Phys. A **38**, L343 (2005); G. Biroli and J.-P. Bouchaud, Europhys. Lett. **67**, 21 (2004); A. Andreanov, G. Biroli, and A. Lefevre, J. Stat. Mech.: Theory Exp. (2006), P07008; B. Kim and K. Kawasaki, J. Stat. Mech.: Theory Exp. (2008), P02004; A. Crisanti, Nucl. Phys. B **796**, 425 (2008) .

[14] In the case of Smoluchowski dynamics it has been shown by B. Cichocki and W. Hess, Physica A **141**, 475 (1987) that the memory function can be rewritten in terms of an irreducible memory function. This argument was generalized by K. Kawasaki, *ibid.* **215**, 61 (1995). For our purposes here the point is that there is not a unique memory function form at lowest nontrivial order in perturbation theory. Thus for a variety of models one can rearrange perturbation theory to have model which supports an ENE transition. One must go to higher order to check the self-consistency of an ENE transition.

[15] The theory we develop here has great potential for treating higher-order correlation functions. The reason why this is in-

teresting is because there has been a significant amount of work associated with the concept of dynamic heterogeneity, e.g., S. C. Glotzer, J. Non-Cryst. Solids **274**, 342 (2000). Out of this research has come the idea that the order parameter for the structural glass transition problem is a time and space displaced product of particle densities, $\rho(\mathbf{r},t)\rho(\mathbf{r}+\mathbf{r}_0,t+\tau)$. The hypothesis is that the associated order parameter correlation function $C_4(\mathbf{r},t)=\langle\delta[\rho(0,0)\rho(0+\mathbf{r}_0,\tau)]\delta[\rho(\mathbf{r},t)\rho(\mathbf{r}+\mathbf{r}_0,t+\tau)]\rangle$ scales with a length ℓ as one goes near the liquid-glass transition such as $C_4(\mathbf{r},t)=F(r/\ell)/r^{(d-2+\eta)}$, where η is a critical index, the length ℓ blows up at the *transition* $\ell\approx\epsilon^{-\nu}$, and $\epsilon=T-T_c$ There is both experimental and numerical support for this hypothesis [See L. Berthier, Phys. Rev. E **69**, 020201 (2004).]; Our interest here is the theoretical work of Biroli and Bouchaud in Ref. [13(f)]. Biroli and Bouchaud sketched a field theoretical calculation of $C_4(\mathbf{r},t)$ compatible with mode-coupling theory. This calculation leads to a diverging length scale ℓ at the ideal glass transition. They find an upper critical dimension of six in their calculation. This work is very provocative since at the level of the dynamic structure factor it is well known that MCT does not contain a large length as one approaches the ergodic-nonergodic transition. Biroli and Bouchaud suggest that one must dig deeper into the theory, look at the four-point quantity C_4, to find this diverging length. However this calculation paints a picture rather than gives the results of a rigorous calculation. One of the goals for the theory developed here is to connect the collective behaviors found at the two-point level to the behavior of the three- and four-point density correlation functions.

[16] Reversible terms in generalized Langevin equations are constructed in a Poisson bracket structure, see S. Ma and G. Mazenko, Phys. Rev. B **11**, 4077 (1975).

[17] Purely dissipative Langevin systems have long been used to describe the order parameter dynamics in magnetic and superfluid systems. There is a balance between the force due to the gradient of the effective free energy and the persistent noise in a thermalized system.

[18] The path-integral description of classical Newtonian dynamics is developed in a series of sophisticated papers by Gozzi and collaborators. E. Gozzi, Phys. Lett. B **201**, 525 (1988); E. Gozzi, M. Reuter, and W. D. Thacker, Phys. Rev. D **40**, 3363 (1989); E. Deotto, E. Gozzi, and D. Mauro, J. Math. Phys. **44**, 5937 (2003); E. Gozzi and M. Reuter, Chaos, Solitons Fractals **4**, 1117 (1994). Their interest is in the symmetry structure of the coupled spaces when one allows the classical system to be coupled to a system of ghost fermions. See below.

[19] Fokker-Planck dynamics in modern terms has two complementary meanings. For systems of particles governed by phase-space coordinates and with a damping component in their dynamics the equation satisfied by the associated probability distribution is called a Fokker-Planck equation. As pointed out by Lax and Zwanzig [20], the associated equation of motion for the phase-space variables is referred to as a Langevin equation. Subsequently the transition generally from a stochastic equation of motion to the time evolution of the associated probability description is called going from a generalized Langevin equation description to generalized Fokker-Planck description. See Ref. [16].

[20] Smoluchowski dynamics has become identified with the over damped kinetics in colloidal systems where the momenta become equilibrated much faster than the positions and one has a dynamics which is subsequently organized in terms of the positions. This is the point of view taken by A. Einstein, Ann. Phys. **322**, 549 (1905) in his seminal treatment of Brownian motion; M. V. Smoluchowski, Phys. Z. **17**, 557 (1916) , still working in coordinate space, generalized Einstein's work to include external forces acting on the Brownian particles. These results were valid for long times P. Langevin, Compt. Rend. **146**, 530 (1908) shortly thereafter developed the Langevin equation description for the momenta of the Brownian particle; Working in terms of probability distributions in velocity space, A. D. Fokker, Ann. Phys. **43**, 812 (1914); and M. Planck, Sitzungsber. K. Preuss. Akad. Wiss. **23**, 324 (1917) , established the simplest form of the Fokker-Planck equation; These works were tied together by a treatment of position and momentum by O. Klein, Ark. Mat., Astron. Fys. **16**, 5 (1922); This work was repeated by H. Kramers, Physica **7**, 284 (1940); This field has been famously reviewed by G. E. Uhlenbeck and L. S. Ornstein, Phys. Rev. **36**, 823 (1930); S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943); M. Lax, *ibid.* **32**, 25 (1960); J. Phys. Chem. Solids **14**, 248 (1960); Rev. Mod. Phys. **38**, 541 (1966); The modern, linear response, correlation function treatment of Brownian motion is due to J. L. Lebowitz and E. Rubin, Phys. Rev. **131**, 2381 (1963); The associated Fokker-Planck description for more than one Brownian particle was developed by J. Deutch and I. Oppenheim, J. Chem. Phys. **54**, 3547 (1971); The extension of the multiple particle Fokker-Planck description into the over damped Smoluchowski regime was carried out by T. J. Murphy and J. L. Aguirre, *ibid.* **57**, 2098 (1972) R. Zwanzig, Adv. Chem. Phys. **15**, 325 (1969) using results reviewed by M. Lax, showed the equivalence of the Fokker-Planck and Langevin descriptions; Important papers treating the many-particle Smoluchowski dynamics system include B. J. Ackerson, J. Chem. Phys. **64**, 242 (1976); W. Dieterich and I. Peschel, Physica A **95**, 208 (1979).

[21] A modern introduction to the equilibrium theory of fluids is given by J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids*, 3rd ed. (Academic Press, New York, 2006). The static theory of liquids includes thermodynamics as well as structure. The static structure is characterized by the pair distribution, $\rho^2 g(r)=\langle\frac{1}{2}\Sigma_{i\neq j=1}^{N}\delta(r-R_i)\delta(R_j)\rangle$. An interesting contemporaneous treatment of this material is given by The Equilibrium Theory of Classical Fluids by H. L. Frisch and J. L. Lebowitz, Benjamin, 1964. Of particular interest are the two reviews, The Pair Distribution in Classical Statistical Mechanics by J. K. Percus and Cluster Expansions for Classical Systems in Equilibrium by G. Stell.

[22] Graphical methods were introduced into the treatment of clustering in equilibrium systems in a series of papers by Mayer. J. E. Mayer, J. Chem. Phys. **5**, 67 (1937); J. E. Mayer and P. G. Ackermann, *ibid.* **5**, 74 (1937); J. E. Mayer and S. F. Harrison, *ibid.* **6**, 87 (1938); J. E. Mayer and E. Montroll, *ibid.* **9**, 2 (1941); and E. Montroll and J. E. Mayer, *ibid.* **9**, 626 (1941). This work showed how a primitive density expansion could be used in dense systems.

[23] J. G. Kirkwood, J. Chem. Phys. **3**, 300 (1935); J. Yvon, Actualités Sci. Ind. **203** (1935); H. D. Ursell, Proc. Cambridge Philos. Soc. **23**, 685 (1927); and M. Born and H. S. Green, *A General Kinetic Theory of Liquids* (Cambridge University Press, Cambridge, England, 1949).

[24] Graphical Summation techniques were introduced by R. J. Riddell and G. E. Uhlenbeck, J. Chem. Phys. **21**, 2056 (1953); G. W. Ford and G. E. Uhlenbeck, PNAS **42**, 122 (1956); **42**, 529 (1956); **43**, 163 (1957); G. W. Ford, R. Z. Norman, and G. E. Uhlenbeck, Proc. Natl. Acad. Sci. U.S.A. **42**, 203 (1956); K. Hiroike, J. Phys. Soc. Jpn. **12**, 864 (1957); T. Morita and K. Hiroike, Prog. Theor. Phys. **23**, 385 (1960); **24**, 679 (1960); **23**, 1003 (1960); J. M. J. van Leeuwen, J. Groenvald, and J. de Boer, Physica **25**, 792 (1959); E. Meeron, J. Chem. Phys. **27**, 1238 (1957); J. Yvon, Nuovo Cimento **9**, 144 (1958); C. De Dominicis, J. Math. Phys. **3**, 983 (1962); **4**, 255 (1963).

[25] Functional methods were introduced by N. N. Bogolyubov, J. Phys. (USSR) **10**, 265 (1946); *Studies in Statistical Mechanics* (North Holland, Amsterdam, 1962), Vol. 2.

[26] J. K. Percus, in *The Equilibrium Theory of Classical Fluids*, edited by H. L. Frisch and J. L. Lebowitz (Benjamin, New York, 1964).

[27] J. K. Percus and G. S. Yevick, Phys. Rev. **110**, 1 (1958).

[28] See the discussion of J. S. Rowlinson, Rep. Prog. Phys. **28**, 169 (1965).

[29] The Ornstein-Zernike relation connects the radial distribution function to the direct correlation function. See L. S. Ornstein and F. Zernike in Ref. [21].

[30] Early treatments of classical field dynamics include L. Onsager and S. Machlup, Phys. Rev. **91**, 1505 (1953); R. H. Kraichnan, J. Fluid Mech. **5**, 497 (1959); J. Math. Phys. **2**, 124 (1961); **3**, 205 (1962); Phys. Fluids **7**, 1723 (1964); H. W. Wyld, Jr., Ann. Phys. (N.Y.) **14**, 143 (1961)

[31] P. C. Martin, E. D. Siggia, and H. A. Rose, Phys. Rev. A **8**, 423 (1973).

[32] H. Janssen, Z. Phys. **B23**, 377 (1976); R. Baush, H. K. Janssen, and H. Wagner, *ibid.* **B24**, 113 (1976).

[33] C. De Dominicis and L. Peliti, Phys. Rev. Lett. **38**, 505 (1977); Phys. Rev. B **18**, 353 (1978); C. De Dominicis, E. Brézin, and J. Zinn-Justin, *ibid.* **12**, 4945 (1975).

[34] R. Graham, *Springer Tracts in Modern Physics* (Springer, Berlin, 1973) Vol. 66.

[35] U. Deker and F. Haake, Phys. Rev. A **11**, 2043 (1975); R. Phythian, J. Phys. A **8**, 1423 (1975); **9**, 269 (1976); U. Deker, Phys. Rev. A **19**, 846 (1979); F. Langouche, D. Roekaents, and E. Tirapegui, Physica A **95**, 252 (1979); R. V. Jensen, J. Stat. Phys. **25**, 183 (1981).

[36] TDGL models were first studied using these new techniques by C. De Dominicis, Lett. Nuovo Cimento **12**, 567 (1975).

[37] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977)

[38] In JZJ [3] see pages 69 and 413.

[39] Ghost fermions are introduced into the analysis of stochastic dynamics with the realization that the Jacobian associated with the change of primary field from the noise to an order parameter field occurs in the numerator of the generating functional central to the development of the theory. One also notices that the trace over a Gaussian distribution of fermions is equal to the determinant of a matrix, $\det M = Tre^{-\bar{\psi}M\psi}$. See page 14 in JZJ [3]. These are the ghost fermions and eliminating the determinant in favor of the fermions gives a coupled fermion-nonfermion system, G. Parisi and N. Sourlas, Phys. Rev. Lett. **43**, 744 (1979); Nucl. Phys. B **206**, 321 (1982); G. Parisi and Wu-yong-shi, Sci. Sin. **24**, 484 (1981); M. V. Feigel'man and A. M. Tsvelik, Sov. Phys. JETP **56**, 823 (1982); Phys. Lett. A **95**, 469 (1983); E. Gozzi, Phys. Rev. D **28**, 1922 (1983); J. Zinn-Justin, Nucl. Phys. B **275**, 135 (1986); G. Munoz and W. S. Burgett, J. Stat. Phys. **56**, 59 (1989).

[40] Once one has the ghost fields as described in Ref. [39] in the generating functional for the field, one can search for symmetries of the action reflected in the whole space of the enlarged system. For a large class of stochastic systems one finds a supersymmetry which mixes fermionic and conventional field degrees of freedom. The key references are given in Ref. [39]. One can go further in the development as indicated by the title "Ward Takahashi Identities and Fluctuation-Dissipation Theorem in a superspace formulation of the Langevin Equation" by S. Chaturvedi, A. K. Kapoor, and V. Srinivason, Z. Phys. B **57**, 249 (1984); J. Phys. A **17**, 2037 (1984). By rewriting the total action in terms of a superfield one obtains a highly compact and symmetric action. This is discussed in some detail in JZJ [3]. A strong cautionary note: Using supersymmetry arguments, it is possible demonstrate that one has dimensional reduction in the random field Ising model (RFIM). Thus the RFIM in four dimensions maps onto the pure Ising model in two dimensions. Unfortunately the four dimensional RFIM does not behave like the two-dimensional pure Ising model under conditions of equilibrium as measured by numerical studies. The explanation put forth by JZJ is that the physical solution of the field equations is a supersymmetry broken solution. For a discussion see G. Grinstein, J. Appl. Phys. **55**, 2371 (1984).

[41] See Onsager's Principle of Microscopic Reversibility and Supersymmetry, E. Gozzi, Phys. Rev. D **30**, 1218 (1984); L. Onsager, Phys. Rev. **37**, 405 (1931); **38**, 2265 (1931); N. B. Casimir, Rev. Mod. Phys. **17**, 343 (1945).

[42] There are a collection of fluctuation theorems which appear to connect strongly nonequilibrium systems and which should have a counterpart in the particle systems discussed here. Outing V. Y. Chernyak, M. Chertkov, and C. Jarzynski, J. Stat. Mech.: Theory Exp. (2006), P08001; The *fluctuation theorem* refers to a set of exact relations describing the statistical mechanics of systems away from equilibrium, generically expressed by the formula, $P(+\Sigma)/P(-\Sigma)=e^{\Sigma}$ where $P(\Sigma)$ is the distribution of observed values of a quantity representing dissipation or entropy production. Such theorems have been worked out for a variety of nonequilibrium systems: D. J. Evans, E. G. D. Cohen, and G. P. Morris, Phys. Rev. Lett. **71**, 2401 (1993); D. J. Evans and D. J. Searles, Phys. Rev. E **50**, 1645 (1994); G. Gallavotti and E. G. D. Cohen, Phys. Rev. Lett. **74**, 2694 (1995); J. Kurchan, J. Phys. A **31**, 3719 (1998); J. L. Lebowitz and H. Spohn, J. Stat. Phys. **95**, 333 (1999); G. N. Bochkov and Yu. E. Kuzovlev, Zh. Eksp. Teor. Fiz. **72**, 238 (1977) [Sov. Phys. JETP **45**, 125 (1977)]; C. Jarzynski, Phys. Rev. Lett. **78**, 2690 (1997); Phys. Rev. E **56**, 5018 (1997); G. E. Crooks, J. Stat. Phys. **90**, 1481 (1998); Phys. Rev. E **60**, 2721 (1999). These theorems should have counterparts in the theories of particles developed here. There is the rather amazing claim by K. Mallick, M. Moshe, and H. Orland, e-print arXiv:0711.2059, that these theorems are a consequence of an underlying supersymmetry. ″This supersymmetry in turn allows one to generate the fluctuation-dissipation theorem to far from equilibrium situations.

[43] H. Rose, J. Stat. Phys. **20**, 415 (1979).

[44] Suppose we have an ideal Newtonian fluid in a field, $u(\mathbf{x})$, that couples to the density. Next we determine the generating functional with the result $W[u]=\rho_0\int d^dx e^{u(x)}$. If the density is a Gaussian variable, then $W[u]=\int d^dx_1 d^dx_2 u(x_1)G(x_1-x_2)u(x_2)$.

[45] JZJ [3] gives an argument on page 63 for choosing $\theta(0)=1/2$.

[46] This is just the functional generalization of the standard matrix identity $\exp[\frac{1}{2}\Sigma_{ij}K_{ij}\frac{\delta^2}{\delta h_i \delta h_j}]e^{\Sigma_i h_i\phi_i}=e^{\Sigma_i h_i\phi_i}\exp[\frac{1}{2}\Sigma_{ij}K_{ij}\phi_i\phi_j]$.

[47] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, 1962); G. Baym and L. P. Kadanoff, Phys. Rev. **124**, 287 (1961); G. Baym, *ibid.* **127**, 1391 (1962).

[48] In different contexts the kinetic kernels have different names. In the field theory protocol the kernels are typically called self-energies, in the kinetic theory protocol, where the analysis is in terms of retarded quantities, the kernel is called a memory function, and in the general case it can be called a dynamic direct correlation function.

[49] R. Velenich, C. Chamon, L. Cugliandolo, and D. Kreimer, J. Phys. A **41**, 235002 (2008).

[50] W. Kohn and J. Luttinger, Phys. Rev. **108**, 590 (1957) see the introduction and Appendix D.

[51] The classical operator formalism for dynamics was introduced by J. von Neumann, Proc. Natl. Acad. Sci. U.S.A. **18**, 70 (1932); H. Koopman and J. von Neumann, *ibid.* **18**, 255 (1932).

[52] Quantum many-body theory is close to the development here. In particular the functional formulation of P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1342 (1959); as popularized by L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, New York, 1962) is very close to our classical development.

[53] Quantum Thermal Green's functions must satisfy Kubo-Martin-Schwinger Boundary Conditions, see references in Ref. [52].

[54] See the discussion of this result for the statics in J. K. Percus, in *The Equilibrium Theory of Classical Fluids*, edited by H. L. Frisch and J. L. Lebowitz (Benjamin, New York, 1964), p. II-72.

[55] The decay time in the problem is inversely proportional to the static structure factor. This clearly, away from $q=0$, leads to a slowing down near the first structure factor peak.

[56] There is an exact solution to the Percus-Yevick approximation for hard spheres. See the discussion in J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids*, 3rd ed., (Academic Press, New York, 2006), Chap. 4.

[57] The F_{12} model and stretching are discussed in detail by W. Goetze in *Liquids, Freezing, and Glass Transition*, edited by J. P. Hansen, D. Levesque, and J. Zinn-Justin (North-Holland, Amsterdam, 1991).

[58] R. M. Mazo, *Brownian Motion* (Clarendon, Oxford, 2002).

[59] B. S. Kim and G. F. Mazenko, J. Stat. Phys. **64**, 631 (1991).