





## Dynamic characteristics of three-dimensional strongly coupled plasmas

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The dynamic structure factor and other dynamic characteristics of strongly coupled one-component plasmas have been studied [Yu. V. Arkhipov *et al.*, *Phys. Rev. Lett.* **119**, 045001 (2017)] using the self-consistent version of the method of moments. Within any version of the latter, the system dielectric function satisfies all involved sum rules and other exact relations automatically, and the advantage of this version is that, in addition, the dynamic characteristics (the dynamic structure factor, the dispersion, and decay parameters of the collective modes) are all expressed in terms of the static ones (the static structure factor) without any adjustment to the simulation data. The approach outlined in the aforementioned Letter is justified in detail and applied mainly to the classical Coulomb systems achieving satisfactory agreement with new numerical simulation data. It is shown how the realm of applicability of the method can be extended to partly degenerate and multicomponent systems, even to simple liquids. Some additional theoretical results are presented in the Supplemental Material.

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### I. INTRODUCTION

The description of dynamic properties of collision-dominated Coulomb systems is a challenge of contemporary statistical plasma physics. The Coulomb systems we refer to are strongly coupled plasmas and in particular warm dense matter, where the temperature and density vary in wide ranges of magnitude but in a way that the characteristic lengths such as the Debye and Wigner-Seitz radii, the thermal de Broglie wavelength, and the Landau length are all about 1 Å long. Under such conditions, thermal, Coulomb coupling, and quantum effects compete with each other and impede the construction of a crossover theory capable of including all of these effects in the description of static, kinetic, and dynamic properties of the above systems of high relevance for inertial fusion devices [1–3] and advanced laboratory studies, e.g., in ultracold plasmas [4,5], electrolytes and charged stabilized colloids [6,7], laser-cooled ions in cryogenic traps [8–10], and dusty plasmas [11–14]. Strongly coupled plasmas also appear in astrophysical contexts in white dwarfs and neutron stars [15,16].

In the present article we basically study classical fluid one-component plasmas where the coupling parameter (roughly a ratio of the Coulomb interaction energy of two particles to

their kinetic energy)

$$\Gamma = \frac{\beta e^2}{a} \in (1, 175). \quad (1)$$

The properties of the uniform partially degenerate electron gas with pronounced quantum properties are usually characterized by the degeneracy parameter defined as

$$D = \beta E_F$$

and the Brueckner parameter  $r_s = a/a_B$ . Here  $a = \sqrt[3]{3/4\pi n}$ ,  $a_B$ , and  $E_F$  are the Wigner-Seitz and Bohr radii and the (electron) Fermi energy; in addition,  $n$  is the number density of the plasma particles, the temperature  $T = (k_B\beta)^{-1}$ , and

$$r_s = \frac{\Gamma}{2D} \left( \frac{9\pi}{4} \right)^{2/3} = 1.842 \frac{\Gamma}{D}.$$

For the latest review on strongly coupled Coulomb liquids, see [17]. Despite the lack of small parameters ( $\Gamma$  or  $D$  or  $r_s$  or  $r_s^{-1}$ ) [18], static structural and kinetic characteristics of strongly coupled plasmas are relatively easy to determine numerically (see, e.g., [19,20]). On the other hand, there have recently appeared extensive numerical studies of dynamic properties of the uniform paramagnetic electron gas and warm dense matter (see [21] and references therein) and of the classical one-component plasmas [22] carried out in a wide realm of variation of the thermodynamic parameters.

The main aim of the present paper is the analytical description of the latter results along with the simulation data

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on the dynamic local-field correction [23] without using any numerical adjustment parameter. The theoretical basis is in the framework of the nonperturbative self-consistent method of moments [24,25], though the initial idea of the method of moments itself is about 40 years old [26,27]. In addition, here we will provide a detailed justification of the self-consistent approach in comparison to the well-established methods of description of dynamic properties of Coulomb systems, which are the mean-field [28] and memory-function [29,30] methods, and the quasilocalized charge approximation [31–33] along with others.

The moment approach was originally based on Nevanlinna's [34,35] noncanonical solutions of the (truncated) Hamburger moment problem consisting in the reconstruction of a non-negative distribution density using a finite number of its power moments (for more details, see Sec. I A in the Supplemental Material [36]). If we employ this approach to study the plasma dielectric function or the dynamic structure factor, the latter are actually the sum rules [47] valid independently of the small-parameter expansions. In this regard, the moment approach is nonperturbative and thus it is auspicious for the determination of dynamic properties of the above crossover systems with no small dimensionless parameters, especially if it is complemented by physically motivated simplifications or asymptotic considerations. The background of the moment approach is certainly purely mathematical with the specifics of a system reflected only in the moments; in this sense the approach is model-free. It can be applied to study dynamic properties of Coulomb and non-Coulomb liquid systems [48], irrespectively of its geometry [49,50] (see also [51–53]). Observe also that the untruncated moment approach taking into account an infinite number of moments is similar to the continued-fraction method by Hong *et al.* [54,55].

Preliminary versions of the present work were presented in [56–59]. Here, in addition, the results of [60] are partially analyzed.

The paper is organized in the following way. In Sec. II we provide mathematical and physical aspects of the moment method background. The expressions obtained there are of general character and hence they are applicable to multi-component and partially degenerate systems as well. Then, in Sec. III, our moment approach is applied to determine dynamic structure factor and dispersion and decay of the collective modes of both Coulomb and Yukawa one-component classical plasmas. Further, in Sec. IV, a comparison to the quasilocalized charge and the extended random-phase approximations is carried out. Numerical results of the moment approach are checked against the available simulation data on the dynamic characteristics of classical Coulomb systems including the dynamic local-field correction, in Secs. V and VI, respectively. A summary is given, conclusions are drawn, and perspectives are outlined in Sec. VII. Some additional details and results are presented in the Supplemental Material [36] with special attention paid to the recent data of [60].

## II. METHOD OF MOMENTS

### A. Moments

The systems we consider are presumed to be in thermal equilibrium, like unmagnetized one-component plasmas. The

keystones of our approach are the plasma (inverse) dielectric function  $\epsilon^{-1}(q, z = \omega + i\delta)$  ( $\delta \geq 0$ ), which is a genuine response function for any  $q$  [61,62], and the (non-negative and even) loss function

$$\mathcal{L}(q, \omega) = -\frac{\text{Im}\epsilon^{-1}(q, \omega)}{\omega}. \quad (2)$$

This definition is quite similar to that of the effective damping function of the hydrodynamic approach by Kadanoff, Martin, and Yip [29,30]. Here and throughout the text we use the dimensionless wave number  $q = ka$ .

The dynamic structure factor (DSF), which is the central quantity of collective and dynamic effects, is determined by the loss function via the fluctuation-dissipation theorem

$$S(q, \omega) = \frac{q^2 n}{3\pi\Gamma} B(\beta\hbar\omega)\mathcal{L}(q, \omega), \quad (3)$$

where

$$B(x) = x[1 - \exp(-x)]^{-1} \underset{x \rightarrow 0}{\simeq} 1 \quad (4)$$

is the Bose factor. Both dynamic functions  $\mathcal{L}(q, \omega)$  and  $S(q, \omega)$  behave at very low frequencies and/or in classical systems [where  $B(\beta\hbar\omega) = 1$ ] in a similar way. We will show how, on the rigorous mathematical basis complemented by simple physical considerations, the knowledge of the dynamic characteristics can be reduced to that of the static ones, precisely, the (partial) static structure factor(s).

The construction blocks of the present approach are the system sum rules which are the loss function frequency power moments

$$C_\nu(q) = \frac{1}{\pi} \int_{-\infty}^{\infty} \omega^\nu \mathcal{L}(q, \omega) d\omega, \quad \nu = 0, 2, 4. \quad (5)$$

Note that the odd-order moments vanish due to the symmetry of the loss function, and thus the general formulas of the theory of moments simplify significantly. Contrary to the multicomponent plasma situation [63], higher-order one-component plasma (OCP) sum rules converge but they are related to scarcely studied nonpairwise correlations that we neglect here (see nevertheless [64]). In classical systems, due to (4), the moments of the loss function (5) are directly proportional to those of the dynamic structure factor (see [24,25]).

The moments  $C_0(q)$ ,  $C_2$ , and  $C_4(q)$  and the characteristic frequencies

$$\omega_1(q) = \sqrt{\frac{C_2}{C_0(q)}}, \quad \omega_2(q) = \sqrt{\frac{C_4(q)}{C_2}} \quad (6)$$

are known independently; they are determined by the system composition, degeneracy, and thermodynamics. By virtue of the classical version of the fluctuation-dissipation theorem (3), the zeroth sum rule in a classical system is determined by the system static structure factor

$$C_0(q) = \frac{1}{\pi} \int_{-\infty}^{\infty} \mathcal{L}(q, \omega) d\omega = \frac{3\Gamma}{q^2} S(q). \quad (7)$$

The second moment is the  $f$ -sum rule [65]

$$C_2(q) = -\frac{1}{\pi} \int_{-\infty}^{\infty} \omega \text{Im} \epsilon^{-1}(q, \omega) d\omega = \omega_p^2, \quad (8)$$

where  $\omega_p$  is the system plasma frequency. Hence, in classical plasmas

$$\omega_1^2(q) = \frac{q^2}{3\Gamma} \frac{\omega_p^2}{S(q)}. \quad (9)$$

The loss function fourth power moment

$$C_4(q) = \frac{1}{\pi} \int_{-\infty}^{\infty} \omega^4 \mathcal{L}(q, \omega) d\omega \quad (10)$$

for OCPs has been studied by Kugler [28], Pathak and Vashishta [66], and earlier in [67–70]. It was established that in a classical Coulomb (C) or Yukawa (Y) one-component plasmas with the pairwise interaction potential [71] (see also [72])

$$\varphi(r) = \frac{e^2}{r} \exp\left(-\kappa \frac{r}{a}\right), \quad (11)$$

$$\frac{\omega_2^2(q)}{\omega_p^2} = \zeta_\alpha(q) + \frac{q^2}{\Gamma} + U_\alpha(q), \quad \alpha = C, Y,$$

$$\zeta_C(q) = 1, \quad \zeta_Y(q) = \frac{q^2}{q^2 + \kappa^2},$$

$$U_\alpha(q) = \frac{1}{3\pi} \int_0^\infty [S(p) - 1] f_\alpha(p, q) p^2 dp,$$

$$f_C(p, q) = \frac{5}{6} - \frac{p^2}{2q^2} + \frac{(p^2 - q^2)^2}{4q^3 p} \ln \left| \frac{q+p}{q-p} \right|,$$

$$f_Y(p, q; \kappa) = \frac{3q^2 - p^2 - \kappa^2}{2q^2} - \frac{2p^2}{3(p^2 + \kappa^2)} + \frac{(q^2 - p^2 - \kappa^2)^2}{8q^3 p} \ln \left( \frac{\kappa^2 + (q+p)^2}{\kappa^2 + (q-p)^2} \right). \quad (12)$$

Certainly, the Coulomb OCP (COCP) expressions are recovered when  $\kappa \rightarrow 0$ . On the other hand, the Yukawa OCP (YOCP) static structure factor  $S(q)$  depends on the screening parameter  $\kappa$ .

These results were generalized for partially degenerate systems in [27,73] within the Kubo linear-response theory and using the second-quantization technique (see [27,47,51,52] and Sec. I C in the Supplemental Material [36]).

### B. Hamburger problem and Nevanlinna formula

To find the dynamic characteristics of Coulomb systems and to relate them to the static ones, we use the solutions of the truncated Hamburger moment problem corresponding to a certain set of convergent frequency moments (5) of the distribution density  $\mathcal{L}(q, \omega)/\pi$  (for more details, see Sec. I A in the Supplemental Material [36]). It is known that the Hamburger problem is solvable, i.e., we can reconstruct the loss function by its moments whenever the set of the involved moments is positive definite [74,75]. It is obvious that the solvable truncated moment problem (except for the so-called degenerate cases [47]) may have only an infinite number of solutions. The branch of at least continuous (noncanonical) solutions of the moment problem by virtue of Nevanlinna's theorem [34,35] is parametrized by the Nevanlinna parameter functions (NPFs)  $R_\nu(z; q)$ . This one-to-one correspondence (a

bijection) is realized by the Nevanlinna formula

$$\int_{-\infty}^{\infty} \frac{\mathcal{L}(q, \omega) d\omega}{\pi(z - \omega)} = \frac{E_{\nu+1}(z; q) + R_\nu(z; q)E_\nu(z; q)}{D_{\nu+1}(z; q) + R_\nu(z; q)D_\nu(z; q)}, \quad (13)$$

$$\text{Im } z > 0, \quad \nu = 0, 1, 2, \dots;$$

the functions  $R_\nu(z = \omega + i\delta; q)$ , exactly like any response (Nevanlinna) function [74], must be analytic and possess a non-negative imaginary part in the upper half plane  $\delta > 0$ , being at least continuous on its closure  $\delta = 0$ . In addition, they must [uniformly within any angle  $\vartheta \leq \arg(z) \leq \pi - \vartheta$ ,  $0 < \vartheta < \pi$ ] satisfy the limiting condition

$$\lim_{z \rightarrow \infty} \frac{R_\nu(z; q)}{z} = 0. \quad (14)$$

This important property of the NPF guarantees the automatic satisfaction, by the density  $\mathcal{L}(q, \omega)/\pi$ , of the imposed moment conditions or sum rules. The coefficients of the one-to-one linear-fractional transformation  $\mathcal{L}(q, \omega) \longleftrightarrow R_\nu(\omega; q)$  [Eq. (13)] are real orthogonal polynomials with the weight  $\mathcal{L}(q, \omega)/\pi$  [52,74], which possess only real alternating zeros [74]. Precisely, for an even density  $\mathcal{L}(q, \omega)/\pi$ ,

$$D_0(z; q) = 1, \quad D_1(z; q) = z, \quad D_2(z; q) = z^2 - \omega_1^2(q),$$

$$D_3(z; q) = z[z^2 - \omega_2^2(q)], \dots,$$

$$E_0(z; q) = 0, \quad E_1(z; q) = C_0(q), \quad E_2(z; q) = C_0(q)z,$$

$$E_3(z; q) = C_0(q)\{z^2 - [\omega_2^2(q) - \omega_1^2(q)]\}, \dots \quad (15)$$

A contemporary proof of the Nevanlinna theorem (13) based on the Krein formula for the generalized resolvents [76] can be found in [77]. Note that if we descend on the real axis of frequencies or let  $\delta \downarrow 0$  in (13), then, due to the Sochocki-Plemelj-Dirac formula

$$\frac{1}{\omega' - \omega - i0^+} = \frac{\mathcal{P}}{\omega' - \omega} + \pi i \delta(\omega' - \omega), \quad (16)$$

( $\mathcal{P}$  standing for the Cauchy principal value), we obtain

$$\mathcal{L}(q, \omega) = -\text{Im} \frac{E_{\nu+1}(\omega; q) + R_\nu(\omega; q)E_\nu(\omega; q)}{D_{\nu+1}(\omega; q) + R_\nu(\omega; q)D_\nu(\omega; q)},$$

$$\nu = 0, 1, 2, \dots, \quad (17)$$

or, for the classical systems,

$$S(q, \omega) = -\frac{q^2 n}{3\pi\Gamma} \text{Im} \frac{E_{\nu+1}(z; q) + R_\nu(z; q)E_\nu(z; q)}{D_{\nu+1}(z; q) + R_\nu(z; q)D_\nu(z; q)},$$

$$\nu = 0, 1, 2, \dots \quad (18)$$

Earlier [52], we contrasted these results with alternative theoretical approaches like the random-phase or the extended Mermin approximations. Let us now establish an interrelation between the particular cases with  $\nu = 0, 1$  and widely employed mean-field, hydrodynamic, and memory-function models. In the simplest case of  $\nu = 0$ , even the  $f$ -sum rule is not satisfied (its similarity to the mean-field models is discussed in the Supplemental Material [36]). The memory-function approach intrinsically involves only three sum rules  $\{C_0(q), 0, C_2\}$ , but using the Gaussian memory function, one includes in the model two sum rules more. This is achieved by introducing an adjustable parameter, a characteristic relaxation time.

There are only two theoretical approaches capable of directly taking into account five sum rules  $\{C_0(q), 0, C_2, 0, C_4(q)\}$  while describing dynamic properties of strongly coupled plasmas and warm dense matter. These are the quasilocated-charge approximation (QLCA) model [31–33] and the method of moments. These two, in addition, express the dynamic characteristics in terms of the static ones, namely, in the case of one-component plasmas, the static structure factor. However, the QLCA does not account for the energy dissipation and the single-particle movement effects such as the Vlasov dispersion contribution to the fourth moment.

The advantages of the method of moments corresponding to the case  $\nu = 2$  are to be outlined further. Certainly, the numerical outcomes will be checked against available simulation data as well.

### III. DYNAMIC STRUCTURE FACTOR

#### A. Memory-function formalism

If we take into account only three sum rules  $\{C_0(q), 0, C_2\}$  ( $\nu = 1$ ), with

$$\det \begin{pmatrix} C_0(q) & 0 \\ 0 & C_2 \end{pmatrix} > 0,$$

it follows from (13) and (17) that

$$\int_{-\infty}^{\infty} \frac{\mathcal{L}_1(q, \omega) d\omega}{\pi(z - \omega)} = \frac{C_0(q)[z + R_1(z; q)]}{z^2 - \omega_1^2(q) + zR_1(z; q)} \quad (19)$$

and

$$\mathcal{L}_1(q, \omega) = \frac{\omega_p^2 \text{Im} R_1(\omega; q)}{|\omega^2 - \omega_1^2 + \omega R_1(\omega; q)|^2}, \quad \omega = \text{Re}(z + i0^+) \quad (20)$$

or

$$\epsilon_1^{-1}(q, \omega) = 1 + \frac{\omega_p^2}{\omega^2 - \omega_1^2(q) + \omega R_1(\omega; q)} \quad (21)$$

(see Sec. I B in the Supplemental Material [36] as well). Let us compare these expressions with those of the classical Gaussian memory-function model widely used recently for the description of dynamic properties of strongly coupled Yukawa plasmas [22,23] (see also [78]). Then, by employing the classical version of the fluctuation-dissipation theorem and using our notation, we get

$$\begin{aligned} \mathcal{L}_{\text{MF}}(q, \omega) \\ = \frac{\omega_p^2 \text{Re} M(q, \omega)}{[\omega^2 - \omega_1^2(q) - \omega \text{Im} M(q, \omega)]^2 + [\omega \text{Re} M(q, \omega)]^2}, \end{aligned} \quad (22)$$

where

$$M(q, \omega) = \frac{\tau_q [\omega_2^2(q) - \omega_1^2(q)]}{i\sqrt{\pi}} Z\left(\frac{\tau_q \omega}{\sqrt{\pi}}\right) \quad (23)$$

is determined by the plasma dispersion function of a real variable  $\xi = \tau_q \omega / \sqrt{\pi}$ ,

$$Z(\xi) = \exp(-\xi^2) \left( i\sqrt{\pi} - 2 \int_0^\xi \exp(s^2) ds \right). \quad (24)$$

The memory-function model (22) for the loss function is obviously equivalent to (20) with

$$R_1(\omega; q) = \frac{\tau_q [\omega_2^2(q) - \omega_1^2(q)]}{\sqrt{\pi}} Z\left(\frac{\tau_q \omega}{\sqrt{\pi}}\right). \quad (25)$$

For any correct NPF, the generic loss function satisfies the sum rules  $\{C_0(q), 0, C_2\}$  automatically and with the Gaussian memory function (23) of [79], the asymptotic form of the inverse dielectric function (21) with (25) approaches, independently of the value of the positive relaxation time  $\tau_q$ , the one corresponding to the set of five sum rules  $\{C_0(q), 0, C_2, 0, C_4(q)\}$ :

$$\begin{aligned} \epsilon_{\text{MF}}^{-1}(q, \omega) &= 1 + \frac{\omega_p^2}{\omega^2 - \omega_1^2 + \frac{\omega \tau_q (\omega_2^2 - \omega_1^2)}{\sqrt{\pi}} Z\left(\frac{\tau_q \omega}{\sqrt{\pi}}\right)} \\ &\underset{\omega \rightarrow \infty}{\simeq} 1 + \frac{\omega_p^2}{\omega^2} + \frac{\omega_2^2 \omega_p^2}{\omega^4} + O(\omega^{-6}) \end{aligned} \quad (26)$$

This means that the memory-function model (22) and (23) satisfies these sum rules and this presumably permitted to adjust the expression for the dynamic structure factor stemming from (22) to the vast amount of simulation data presented in [22,23]. Moreover, Mithen *et al.* in [22,23] showed that a very fine tuning to the dynamic simulation data can be performed by taking into account the numerical *noise* in the characteristic frequencies  $\omega_1(q)$  and  $\omega_2(q)$ . In other words, for each value of the wave number  $q$  the authors of those papers used the adjusted values of  $\tau_q$  complemented by a slight [not more than  $\pm 10\%$  of the values precalculated by (6)] variation of these frequencies to achieve an almost complete coincidence of the memory-function theoretical values of the dynamic structure factor with the simulation data. We conclude that the memory-function formalism is a special case of the solution of the problem of moments. In general, it satisfies the zero- and  $f$ -sum rules, but with a Gaussian memory function (23) it satisfies the fourth sum rule as well, independently of the value of the relaxation time  $\tau_q$ .

#### B. Five-moment problem

##### 1. Setup

Consider now the Hamburger moment problem with five convergent frequency moments  $\{C_0(q), 0, C_2, 0, C_4(q)\}$ ,  $\nu = 2$ . By virtue of the Cauchy-Bunyakovsky-Schwarz inequality, this moment sequence is positive definite (see [27,52]). Hence, this moment problem is solvable, i.e., we can reconstruct the loss function, the dynamic structure factor, and to describe the properties of the collective modes existing in the system the unshifted (diffusion) mode and the shifted (optical or acoustic-rotor) mode

$$\omega_{\text{us}}(q) = -ia(q), \quad \omega_{\pm \text{sh}}(q) = \pm W(q) - ib(q) \quad (27)$$

and other dynamic characteristics, in both one-component and multicomponent plasmas. Expressions for the loss function and the dynamic structure factor follow immediately from the

general formulas (15), (17), and (18),

$$\mathcal{L}(q, \omega) = \frac{\omega_p^2 [\omega_2^2(q) - \omega_1^2(q)] \text{Im } R_2(\omega; q)}{|\omega[\omega^2 - \omega_2^2(q)] + [\omega^2 - \omega_1^2(q)]R_2(\omega; q)|^2}, \tag{28}$$

and for the classical systems

$$S(q, \omega) = \frac{q^2 n}{3\pi\Gamma} \frac{\omega_p^2 [\omega_2^2(q) - \omega_1^2(q)] \text{Im } R_2(\omega; q)}{|\omega[\omega^2 - \omega_2^2(q)] + [\omega^2 - \omega_1^2(q)]R_2(\omega; q)|^2}. \tag{29}$$

The mode characteristics are to be found from the dispersion equation or as the poles of the inverse dielectric function. The latter can be extracted from the Nevanlinna linear-fractional transformation (13) using

$$\begin{aligned} \epsilon^{-1}(q, z = \omega + i0^+) &= 1 - C_0(q) + \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\mathcal{L}(q, \omega') d\omega'}{1 - \frac{\omega'}{z}} \\ &\underset{\omega \rightarrow \infty}{\simeq} 1 - C_0(q) + \frac{1}{\pi} \int_{-\infty}^{\infty} \left[ 1 + \frac{\omega'}{z} + \left(\frac{\omega'}{z}\right)^2 + \left(\frac{\omega'}{z}\right)^3 + \left(\frac{\omega'}{z}\right)^4 + \left(\frac{\omega'}{z}\right)^5 + \dots \right] \mathcal{L}(q, \omega') d\omega' \\ &= 1 + \frac{\omega_p^2}{\omega^2} + \frac{\omega_2^2 \omega_p^2}{\omega^4} + O(\omega^{-6}), \end{aligned}$$

and thus coincides with the expansion (26). Certainly, the dielectric function

$$\epsilon(q, z) = 1 - \frac{\omega_p^2(z + R_2(z; q))}{z(z^2 - \omega_2^2 + \omega_p^2) + R_2(z; q)(z^2 - \omega_1^2 + \omega_p^2)} \tag{32}$$

can be applied to the solution of other problems, e.g., the problem of reflection of laser radiation from shock-compressed plasmas (see [80,81] and references therein).

**2. Nevanlinna parameter reduction**

In order to employ the above expressions for the plasma dynamic characteristics and to solve the dispersion equation

$$z[z^2 - \omega_2^2(q)] + R_2(z; q)[z^2 - \omega_1^2(q)] = 0 \tag{33}$$

explicitly, one has to model the NPF  $R_2(z; q)$ . The simplest is to substitute the NPF by its static value, like it was initially suggested in [26] and followed in [82] and a number of other publications (see [47] and references therein):

$$R_2(z; q) = R_2(0; q) = ih(q), \quad h(q) > 0. \tag{34}$$

In order to relate the unknown function  $h(q)$  to the system static characteristics, precisely, to the frequencies  $\omega_1(q)$  and  $\omega_2(q)$ , consider the Fourier transform of the loss function  $\Lambda(q, t)$ , whose behavior at long times, by virtue of the Tauber or Abel theorem, will be similar to that of the Fourier transform of the dynamic structure factor, i.e., the intermediate scattering function  $K(q, t)$ . The function  $\Lambda(q, t)$  is bounded, decreasing exponentially, and becomes essentially zero for long times. Hence, taking into account the physical timescales of the problem, for  $t$  larger than the longest relaxation time of the system collective modes and in compliance with Bogolyubov's principle of weakening of correlations,

the Kramers-Kronig relations for the inverse dielectric function

$$\begin{aligned} \epsilon^{-1}(q, z) &= 1 - \int_{-\infty}^{\infty} \frac{\omega \mathcal{L}(q, \omega) d\omega}{\pi(\omega - z)} \tag{30} \\ &= 1 - C_0(q) + \frac{z}{\pi} \int_{-\infty}^{\infty} \frac{\mathcal{L}(q, \omega) d\omega}{\omega - z} \\ &= 1 + \frac{\omega_p^2 [z + R_2(z; q)]}{z[z^2 - \omega_2^2(q)] + R_2(z; q)[z^2 - \omega_1^2(q)]}, \tag{31} \\ &\text{Im } z > 0. \end{aligned}$$

Note that the asymptotic expansion of this inverse dielectric function corresponds to the number of sum rules involved independently of the form of the NPF,

both  $\Lambda(q, t)$  and  $K(q, t)$  have finite (zero) limiting values as  $t \rightarrow \infty$ . Then  $\mathcal{L}(q, \omega)$ , like the dynamic structure factor, also has a finite zero-frequency limiting value. Hence, for very low frequencies, due to the same theorems, the values of the loss function need to be weakly dependent on  $\omega$ . This is exactly what we observe on the dynamic structure factor graphs of [22–24,83]: There is no Rayleigh peak characteristic for the diffusion mode in simple liquids, so we observe broad flat extrema of both Coulomb and Yukawa dynamic structure factors near the zero frequency. A sharp Rayleigh peak observed on some graphs, e.g., in [83], might be attributed to the caging effects [22].

The loss function (and in classical systems the dynamic structure factor as well) is an even function of frequency, so from the mathematical point of view this situation corresponds to the so-called third derivative test: The first three derivatives of the loss function must vanish at  $\omega = 0$  and the value of the fourth one defines the nature of the extremum; we have a minimum if it is positive and vice versa. Therefore, as it can be easily shown, we can describe the absence of the Rayleigh peak [22] if we oblige the second derivative of  $\mathcal{L}(q, \omega)$  with respect to  $\omega$  to vanish at  $\omega = 0$ :

$$\left. \frac{d^2 \mathcal{L}(q, \omega)}{d\omega^2} \right|_{\omega=0} = 0. \tag{35}$$

The same results can be obtained if we observe that the loss function as an even function of frequency effectively depends only on the variable  $x = \omega^2$  and study the extrema of the the function  $\mathcal{L}(q, x)$  so that we can use the traditional Fermat interior extremum condition for the first derivative of the latter

function:

$$\left. \frac{d\mathcal{L}(q, x)}{dx} \right|_{x=0} = 0. \quad (36)$$

The condition (35) or (36) was justified numerically in [52].

The degenerate system dynamic structure factor (3), due to the presence of the Bose factor, does not possess the property (35), but in classical systems it permits to eventually express (within the present approach) all dynamic characteristics in terms of the static structure factor(s). The approximation (34) converts (28) into

$$\mathcal{L}(q, \omega)|_{R_2=ih} = \frac{\omega_p^2 [\omega_2^2(q) - \omega_1^2(q)] h(q)}{\omega^2 [\omega^2 - \omega_2^2(q)]^2 + h^2(q) [\omega^2 - \omega_1^2(q)]^2}. \quad (37)$$

The Maclaurin expansion of this expression,

$$\begin{aligned} \mathcal{L}(q, \omega \rightarrow 0)|_{R_2=ih} &\simeq \frac{\omega_p^2 (\omega_2^2 - \omega_1^2)}{h\omega_1^4} \\ &+ \frac{\omega_p^2 (\omega_2^2 - \omega_1^2)}{h^3 \omega_1^8} \omega^2 (\omega_2^4 - 2h^2 \omega_1^2) \\ &+ \frac{\omega_p^2 (\omega_2^2 - \omega_1^2)}{h^5 \omega_1^{12}} \omega^4 (3h^4 \omega_1^4 + 2h^2 \omega_1^4 \omega_2^2 \\ &- 4h^2 \omega_1^2 \omega_2^4 + \omega_2^8) + O(\omega^6), \end{aligned}$$

implies that the loss function possesses a broad flat extremum and no Rayleigh peak at  $\omega = 0$  if

$$h(q) = h_0(q) = \frac{\omega_2^2(q)}{\sqrt{2}\omega_1(q)}. \quad (38)$$

An alternative justification of the approximation (34) and (38) is provided in Sec. II of the Supplemental Material [36]. The sign of the fourth derivative at  $\omega = 0$  for  $h = h_0$ , i.e., the sign of the parameter

$$\theta(q) = \frac{2\omega_1(q) - \omega_2(q)}{\omega_p} \in \left( -\frac{\omega_2(q)}{\omega_p}, \frac{\omega_2(q)}{\omega_p} \right), \quad (39)$$

determines the nature of the extremum at  $\omega = 0$ : A positive  $\theta(q)$  corresponds to a minimum and vice versa. When  $\theta(q) > 0$ , the loss function and, in classical systems, the dynamic structure factor have two shifted maxima whose positions on the frequency axis, in nondegenerate and slightly degenerate systems, are determined by the following simple expression:

$$\omega_{\max}(q) = \pm \frac{\omega_2(q)}{\sqrt{3}\omega_1(q)} \sqrt{4\omega_1^2(q) - \omega_2^2(q)}. \quad (40)$$

Otherwise, this mode decays strongly and we have only a broad unshifted maximum. Observe also that the zero-frequency value

$$\mathcal{L}(q, 0)|_{R_2=ih} = \frac{\sqrt{2}\omega_p^2}{\omega_1^3(q)} \left[ 1 - \frac{\omega_1^2(q)}{\omega_2^2(q)} \right] \quad (41)$$

is a decreasing function of the parameter  $\theta(q)$ . The role of the discrimination parameter (39) was discussed in [24,25].

The important result (38) leads to the simple form for the dynamic structure factor

$$S(q, \omega) = \frac{q^2 n}{3\Gamma} \frac{\sqrt{2}\omega_1 \omega_2^2 \omega_p^2 (\omega_2^2 - \omega_1^2) B(\beta\hbar\omega)}{2\omega_1^2 \omega^2 (\omega^2 - \omega_2^2)^2 + \omega_2^4 (\omega^2 - \omega_1^2)^2}, \quad (42)$$

which is one of the main practical results of the present paper. In a classical system like a one-component plasma the Bose factor  $B(\beta\hbar\omega) = 1$ .

Further, the model (38) simplifies the dispersion equation (33) into the analytically solvable cubic equation

$$\sqrt{2}z\omega_1(q)[z^2 - \omega_2^2(q)] + \omega_2^2(q)[z^2 - \omega_1^2(q)] = 0 \quad (43)$$

whose solutions are represented by the following formulas:

$$\begin{aligned} \omega_{\text{us}}(q) &= -ia(q) = -w^2 X - wY - ih_0/3, \\ \omega_{-\text{sh}}(q) &= -W(q) - ib(q) = -X - Y - ih_0/3, \\ \omega_{\text{sh}}(q) &= W(q) - ib(q) = -wX - w^2 Y - ih_0/3. \end{aligned} \quad (44)$$

They provide direct information on the system (unshifted) diffusion and shifted (optical or acoustic-rotor) modes. Here  $w = \exp(2\pi i/3)$  and

$$\begin{aligned} X &= \sqrt[3]{h_0 V^2 / 2i + Z^3}, \quad Y = \sqrt[3]{h_0 V^2 / 2i - Z^3}, \\ Z^3 &= \sqrt{-\left(\omega_2^2/3 - h_0^2/9\right)^3 - (h_0 V^2 / 2)^2}, \\ V^2 &= -\omega_2^2/3 + \omega_1^2 + 2h_0^2/27. \end{aligned} \quad (45)$$

We have shown that in both classical and partially degenerate systems, irrespectively of the number of their components and in the present approximation determined by the number of sum rules taken into account (6) and the model (38), the physical characteristics of the plasma are contained in the characteristic frequencies  $\omega_1(q)$  and  $\omega_2(q)$ , i.e., they are described by the system static structure characteristics only. The corresponding numerical results in comparison with available simulation data are provided in Sec. V.

#### IV. COMPARISON WITH ALTERNATIVE THEORETICAL RESULTS

Observe first that the above expression for the inverse dielectric function (31) leads to the QLCA model [31–33] for the dielectric function of *purely Coulomb systems* if we neglect the Nevanlinna parameter function  $R_2(z; k)$  (responsible for the account of the energy dissipation in the system) and the zeroth-order moment and consider only the coupling contribution  $U(k)$  to the exact characteristic frequency

$$\begin{aligned} \omega_2^{\text{COCP}}(k) &= \sqrt{\frac{C_4(k)}{C_2}} \\ &= \omega_p \sqrt{1 + K(k) + U(k)} \xrightarrow{\text{QLCA}} \omega_p \sqrt{1 + U(k)}. \end{aligned}$$

Thus we understand that the quasilocalized charge approximation takes into account, and in a limited form, neglecting the kinetic energy contribution, only the second and fourth sum rules. Sophisticated QLCA results including the dispersion gap in electron bilayers can be reproduced and improved within the moment approach. However, there is no possibility to reduce the QLCA dispersion to the classical Vlasov

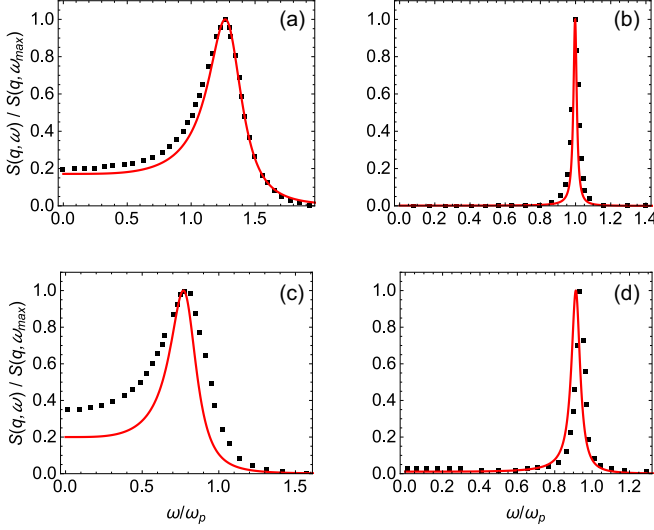


FIG. 1. Dynamic structure factor (47) (solid lines) normalized to the shifted maximum values in COCPs, compared to the molecular-dynamics results [23] (squares), at (a)  $\Gamma = 1$  and  $q = 1.02$ , (b)  $\Gamma = 10$  and  $q = 0.64$ , (c)  $\Gamma = 50$  and  $q = 2.32$ , and (d)  $\Gamma = 120$  and  $q = 1.39$ . The static structure factors were calculated using the fitting of [88].

asymptotic form contained in the kinetic contribution  $K(k)$  of the fourth moment. In Yukawa one-component plasmas, the QLCA also accounts for two contributions to the fourth moment only:

$$\left[ \frac{\omega_2^{\text{YOCP}}(k)}{\omega_p} \right]^2 = \frac{k^2 a^2}{k^2 a^2 + \kappa^2} + K(k) + U(k) \xrightarrow{\text{QLCA}} \frac{k^2 a^2}{k^2 a^2 + \kappa^2} + U(k).$$

A partial account of the fourth sum rule together with the  $f$ -sum rule has permitted one to achieve, within the QLCA, interesting versatile new results on the dispersion of the collective modes up to now [84,85]. Generally speaking, the QLCA results are imbedded in the moment formalism, but our approach takes the processes of energy dissipation into consideration and permits us to determine the DSFs and the decrements of the collective modes.

Another theoretical approach in the investigation of dynamic characteristics of the systems we are interested in here is based on the local-field corrections to the random-phase approximation [86] (see also [52]). In one-component plasmas the dynamic local-field correction is equivalent to our NPF, while in multicomponent systems the NPF stands for all partial local-field corrections which are quite difficult to model (see, nevertheless, [87] and references therein). This interrelation in one-component plasmas is discussed in Sec. VI.

## V. NUMERICAL RESULTS

### A. Dynamic structure factor

In Figs. 1–3 the numerical simulation data on the dynamic structure factor of Coulomb and Yukawa one-component plasmas [22,23,60] are compared with those calculated by

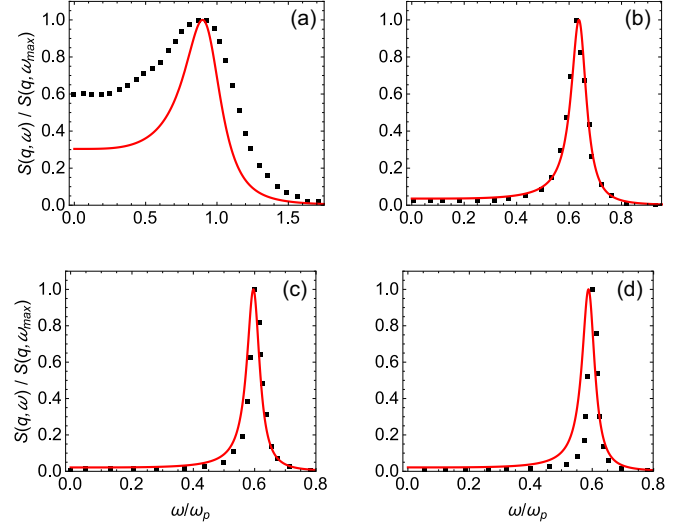


FIG. 2. Same as in Fig. 1, but in YOCPs (solid lines), compared to the MD results [22] (squares), at  $\kappa = 1$ ,  $q = 0.85$ , and (a)  $\Gamma = 1$ , (b)  $\Gamma = 10$ , (c)  $\Gamma = 50$ , and (d)  $\Gamma = 120$ . The static structure factors were calculated using the variational modified hypernetted chain procedure of [89].

Eq. (47). Some preliminary comparison with the recent data of [60] is presented in [36], Sec. III. These results complement those contained in [24]. The Fourier-transformed effective interaction potential in YOCPs is that of (11),

$$\varphi(q) = \frac{4\pi(ea)^2}{q^2 + \kappa^2} = \frac{q^2 \phi(q)}{q^2 + \kappa^2}, \quad (46)$$

where in COCPs  $\kappa = 0$  and in YOCPs  $\kappa = \sqrt{3\Gamma}$ . The simple form for the DSF stemming from (29), (34), and (38),

$$S(q, \omega) = \frac{q^2 n}{3\Gamma} \frac{\sqrt{2}\omega_1 \omega_2^2 \omega_p^2 (\omega_2^2 - \omega_1^2)}{2\omega_1^2 \omega^2 (\omega^2 - \omega_2^2)^2 + \omega_2^4 (\omega^2 - \omega_1^2)^2}, \quad (47)$$

was used and the characteristic frequencies  $\omega_1(q)$  and  $\omega_2(q)$  were calculated according to (9) and (12), respectively, with the employment of the mentioned fitted static structure factors.

In Figs. 1–3 we observe good agreement of the results of our calculations with the molecular-dynamics (MD) simulation data at least up to  $q \approx 2.3$ . We understand that for the

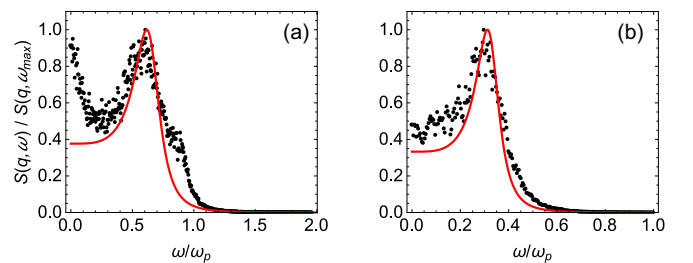


FIG. 3. Same as in Fig. 2, but compared to the MD results [60], at (a)  $\kappa = 0$ ,  $\Gamma = 150$ , and  $q = 2.71$  and (b)  $\kappa = 3$ ,  $\Gamma = 100$ , and  $q = 1.99$ . The static structure factors were calculated using (a) the fitting of [88] and (b) the variational modified hypernetted chain approximation of [89].

values of  $\Gamma \lesssim 1$  [Fig. 2(a)] the static approximation (34) is not applicable since then the NPF must be dynamic and describe the Landau decay. Notice that the quantitative agreement, especially with respect to the physically important position of the shifted maxima, has been achieved by direct calculations using no adjustment parameters. The results of [60] are also discussed in detail in [36].

### B. Dispersion properties

Analysis of the characteristics of collective modes in strongly coupled Coulomb and Yukawa classical one-component plasmas can be realized within the present self-consistent method as well by solving the dispersion equation (43). We have determined that the structure of the collective spectrum is determined by the sign of the discrimination parameter  $\theta(q)$  [Eq. (39)]: When  $\theta(q) < 0$  all modes merge and there is a unique broad line located at  $\omega = 0$ , but when  $\theta(q) > 0$  a shifted line is formed whose position  $\omega_{\max}(q)$  on the frequency axis can be calculated as in Eq. (40). In COCPs this line corresponds to the optical Langmuir-Bohm-Gross mode and in YOCPs it represents the acoustic-roton mode. When this mode decrement  $b(q)$  in (44) is small with respect to its frequency  $W(q)$  the latter real part of the solution of the dispersion equation (33) is close to  $\omega_{\max}(q)$  but, in general, they do not coincide. The results of [24,25] are complemented here by those of Sec. III in [36].

In conclusion, we stress that we thus reduce the knowledge of the dynamic characteristics to the calculation of the frequencies (9) and (12) or to that of the static structure factor  $S(q)$ . The precision of the data on  $S(q)$  affects the precision of our dynamic results significantly. One can try to adjust our results to the simulation data by fitting the values of the characteristic frequencies  $\omega_1(q)$  and  $\omega_2(q)$ , like it was done in [22,23], but this goes beyond the scope of the present work.

## VI. LOCAL-FIELD CORRECTION

We have seen that the Nevanlinna parameter function plays a significant if not crucial role in the present approach. Generally speaking, it is a nonphenomenological component of the latter. In OCPs it is nevertheless directly related to the dynamic local-field correction (DLFC) and in this section we wish to study the interrelation between the static approximation (34) and (38) and the DLFC simulation data of [23].

We have seen that the Nevanlinna formula relates the five-moment NPF  $R_2(\omega; q)$  directly to the dielectric function [see (32)]. On the other hand, by the definition of the DLFC  $G(k, \omega)$ ,

$$\epsilon(q, \omega) = 1 + \frac{\phi(q)\Pi(q, \omega)}{1 - \phi(q)\Pi(q, \omega)G(q, \omega)},$$

where  $\Pi(q, \omega)$  is the polarization function [90]. Then [82]

$$G(q, \omega) = 1 + \frac{1}{\phi(q)\Pi(q, \omega)} + \frac{\omega^2}{\omega_p^2} - \frac{\omega\omega_2^2(q) + \omega_1^2(q)R_2(\omega; q)}{\omega_p^2[\omega + R_2(\omega; q)]}, \quad (48)$$

so any NPF model implies a model for the one-component plasma DLFC and vice versa. In particular, for (34) and (38) we have

$$G_0(q, \omega) = 1 + \frac{1}{\phi(q)\Pi(q, \omega)} + \frac{\omega^2}{\omega_p^2} - \frac{\omega^2\omega_2^2 + h_0^2\omega_1^2}{\omega_p^2(\omega^2 + h_0^2)} + i\omega h_0 \frac{\omega_2^2 - \omega_1^2}{\omega_p^2(\omega^2 + h_0^2)}. \quad (49)$$

The relation between the static local-field correction and the zero-frequency moment of the loss function follows immediately from (48),

$$G(q, 0) = 1 + \frac{1}{\phi(q)\Pi(q, 0)} - \frac{\omega_1^2(q)}{\omega_p^2}, \quad (50)$$

which implies that the static local-field correction is real and is equivalent to the frequency  $\omega_1^2(q)$  only but depends on the polarization operator model. Notice also that due to (48), the dynamic local-field correction is a response function: It is analytic in the upper half plane of frequency where  $\text{Im}G(q, \omega) \geq 0$ .

The expressions (48)–(50) are statistics-free, but in a classical system we choose the polarization function taken in the random-phase approximation as in [23],

$$\begin{aligned} \Pi(q, \omega) &= \Pi_0(q, \omega) \\ &= \frac{3\Gamma}{q^2} \left[ 1 + \frac{\omega}{q\omega_p} \sqrt{\frac{3\Gamma}{2}} Z\left(\frac{\omega}{q\omega_p} \sqrt{\frac{3\Gamma}{2}}\right) \right], \end{aligned}$$

where  $Z(\xi)$  is the real argument plasma dispersion function (24). So, as it is well known [91,92],

$$G_{\text{classical}}(q, 0) = 1 + \frac{q^2}{3\Gamma} \left( 1 - \frac{1}{S(q)} \right).$$

The above model for the NPF (34) leads to reasonable agreement between the dynamic local-field correction  $G_0(q, \omega)$  determined in Eq. (49) and the simulation data of [23] (see Figs. 4 and 5). The squares there represent the simulation data of [23] (the results of the method of continued fractions are presented as well). Notice that the applicability of the Tanaka-Ichimarū extended random-phase approximation is dubious in Yukawa plasmas with no long-range interaction characteristic for the Coulomb systems. We observe that the improvement by the present model is significant at the lower coupling strengths, but at higher coupling strengths we still have no quantitative model for the dynamic local-field correction, at least within the static approximation (34) for the NPF.

On the other hand, the quantity directly computed in molecular-dynamics simulations of [23] was the intermediate scattering function  $K(q; t)$ , which is the frequency Fourier transform of the dynamic structure factor. The  $K(q; t)$  data permitted the authors of [23] to calculate both the dynamic structure factor and the dynamic local-field correction, and it was pointed out in [23] that the latter was more difficult to compute from the  $K(q; t)$  molecular-dynamics data than the dynamic structure factor. The difficulty of determination of the  $K(q; t)$  long-time asymptotic form is reflected in the accuracy of the results of [23] near the zero frequency.



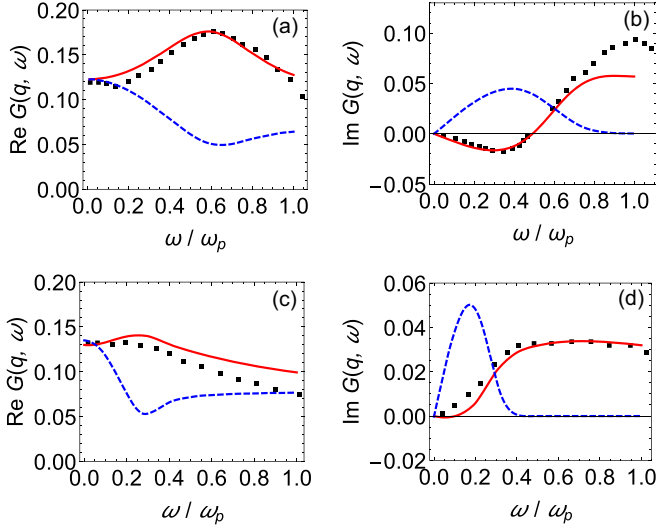


FIG. 4. Dynamic local-field correction in COCPs (solid lines), compared to the molecular dynamics results [23] (squares), at  $q = 1.02$  and (a) and (b)  $\Gamma = 10$  and (c) and (d)  $\Gamma = 50$ . Blue dashed lines correspond to the model of [55]. The static structure factors were calculated using the fitting of [88].

Three starting positions are employed here to analyze the low-frequency behavior of the dynamic local-field correction.

(i) Both the Nevanlinna parameter function and the dynamic local-field correction are analytic and holomorphic functions of the complex frequency  $z = \omega + i\delta$  in the half plane  $\delta > 0$ ; hence they admit Maclaurin expansions at  $\omega = 0$ .

(ii) Due to the Riesz-Herglotz formula for the (Nevanlinna) response functions [74], the correct zero-frequency value of the NPF is purely imaginary [47,74], say,  $ih(q)$ , with  $h(q) > 0$ .

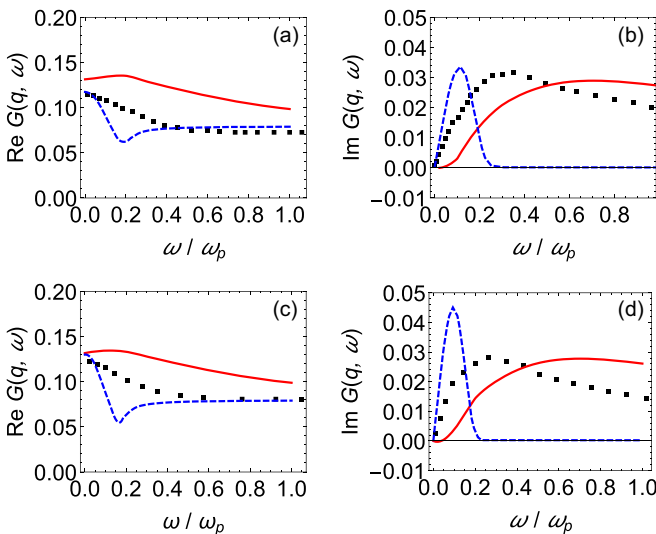


FIG. 5. Same as in Fig. 4, but for (a) and (b)  $\Gamma = 120$  and (c) and (d)  $\Gamma = 160$ .

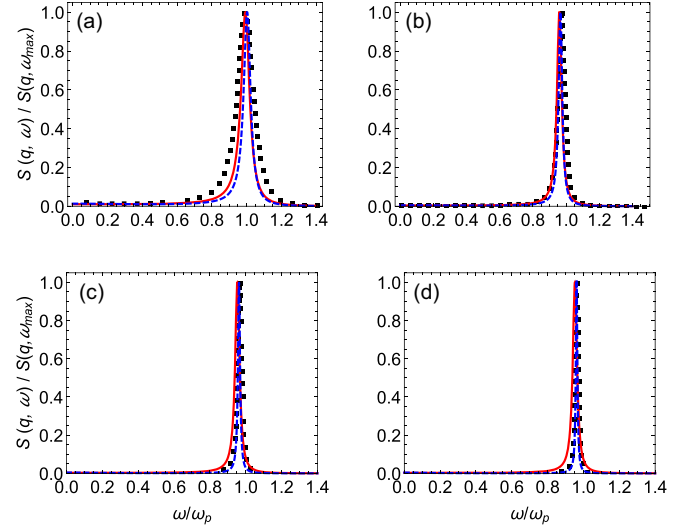


FIG. 6. Dynamic structure factor (3) normalized to the shifted maxima values in Coulomb one-component plasmas, compared to the MD results [23], at  $q = 1.02$  and (a)  $\Gamma = 10$ , (b)  $\Gamma = 50$ , (c)  $\Gamma = 120$ , and (d)  $\Gamma = 160$ . Red solid lines correspond to the dynamic structure factors with  $h_0(q)$  [Eq. (47)] and blue dashed lines display the dynamic structure factors with  $h$  [Eq. (52)]. The static structure factors were calculated using the fitting of [88].

(iii) The systems studied in [23] are classical; hence

$$\phi(q)\Pi_0(q, \omega \rightarrow 0) \simeq \frac{3\Gamma}{q^2} + 3i\sqrt{\pi} \frac{\Gamma}{q^3} \frac{a\omega}{v_T} + O(\omega^2).$$

Thus,

$$G(q, \omega \rightarrow 0) \simeq G(q, 0) - i\omega \left( \frac{\omega_1^2(q) - \omega_2^2(q)}{\omega_p^2 h(q)} + \frac{\sqrt{\pi} a q}{3\Gamma v_T} \right) + O(\omega^2)$$

or

$$G(q, \omega \rightarrow 0) \simeq G(q, 0) - i\omega p(q) + O(\omega^2).$$

Then, after some simple calculations, we obtain for the NPF from (48) the following limiting form:

$$R_2(\omega \rightarrow 0; q) \simeq i \frac{\omega_2^2(q) - \omega_1^2(q)}{\omega_p^2 \left[ \frac{\sqrt{\pi} a q}{3\Gamma v_T} - p(q) \right]} = ih(q). \quad (51)$$

These values are certainly different from  $h_0(q)$ . For each wave number, we obtained the values of the parameter  $p(q)$  from the data of [23] on the dynamic local-field correction and found that the value of  $h(q)$  in (51) is quite close to  $h_0(q)$  and gives satisfactory results for the dynamic structure factor (see Fig. 6). These calculations were carried out using the expression

$$S(q, \omega) = \frac{q^2 n_e}{3\Gamma} \frac{\omega_p^2 (\omega_2^2 - \omega_1^2) h}{\omega^2 (\omega^2 - \omega_2^2)^2 + h^2 (\omega^2 - \omega_1^2)^2} \quad (52)$$

and (47), respectively.

## VII. CONCLUSION

The nonperturbative self-consistent moment approach to the description of dynamic properties of strongly coupled Coulomb systems was discussed. The method was constructed to satisfy the first three nonvanishing sum rules of the system inverse dielectric function automatically. It is capable of reducing the knowledge of the dynamic properties to that of the static ones. It is also independent of any data input from simulations. Agreement was achieved with available numerical data on the dynamic structure factor, the collective mode characteristics, and even the dynamic local-field correction of the classical Coulomb and Yukawa plasmas. A comparison with the most important alternative theoretical approaches was carried out. The viability of the suggested approach was thus justified and confirmed. In general, it is also perfectly applicable to study dynamic characteristics of any physical system described by a response function like the inverse dielectric function; further studies in the case of the uniform electron gas or more complex systems are left for future work. The robustness of the approach with respect to the precision of the data on the static structure factors requires further examination. An investigation of the possibilities to vary the values of the sum rules thus accounting for the uncertainties in the static structure factor should be considered elsewhere.

We believe that the double-hump feature that emerges in the dynamic structure factor computed by MD [see, for example, Fig. 3(a)] at strong coupling and high wave numbers could be described within the same formalism in the nine-moment approximation, i.e., taking into account the sixth and eighth moments, which could generate two additional characteristic frequencies. The latter are related to the three- and four-point static structure factors  $S(\mathbf{k}_1, \mathbf{k}_2)$  and  $S(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ , scarcely known at the moment. This task is also beyond the scope of the present work.

## ACKNOWLEDGMENTS

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