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Approximation scheme for strongly coupled plasmas: Dynamical theory

K. I. Golden
Department of Electrical Engineering, Northeastern University, Boston, Massachusetts 02115

G. Kalman
Department of Physics and Center for Energy Research, Boston College, Chestnut Hill, Massachusetts 02167
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The authors present a self-consistent approximation scheme for the calculation of the dynamical polarizability $\alpha(\vec{k},\omega)$ at long wavelengths in strongly coupled one-component plasmas. Development of the scheme is carried out in two stages. The first stage follows the earlier Golden-Kalman-Silevitch (GKS) velocity-average approximation approach, but goes much further in its application of the nonlinear fluctuation-dissipation theorem to dynamical calculations. The result is the simple expression for $\alpha(\vec{k},\omega)$, $\alpha_{GKS}(\vec{k},\omega) = \alpha_{RPA}(\vec{k},\omega)[1 + \upsilon(\vec{k},\omega)]$, where the dynamical screening function $\upsilon(\vec{k},\omega)$ is expressed in terms of quadratic polarizabilities, and RPA stands for random-phase approximation. Its zero-frequency limit $\upsilon(\vec{k},0)$ has already been established and analyzed in the earlier GKS work. At high frequency, $\alpha_{GKS}(\vec{k},\omega\rightarrow\infty)$ exactly satisfies the $1/\omega^4$ moment sum rule. In the second stage, the above dynamical expression is made self-consistent at long wavelengths by postulating that a decomposition of the quadratic polarizabilities in terms of linear ones, which prevails in the $k\rightarrow 0$ limit for weak coupling, can be relied upon as a paradigm for arbitrary coupling. The result is a relatively simple quadratic integral equation for α. Its evaluation in the weak-coupling limit and its comparison with known exact results in that limit reveal that almost all important correlational and long-time effects are reproduced by our theory with very good numerical accuracy over the entire frequency range; the only significant defect of the approximation seems to be the absence of the "dominant" $\gamma\ln\gamma^{-1}$ (γ is the plasma parameter) contribution to Im $\alpha(\vec{k},\omega)$.

I. INTRODUCTION

The behavior of strongly coupled one-component plasmas (ocp's) has been the subject of a great number of recent investigations,[1] both as a result of Monte-Carlo[2] and molecular-dynamics[3] computer studies and as a result as well of theoretical model buildings.[1] The coupling strength of such systems is conveniently described through the plasma parameter $\gamma=\kappa^3/4\pi n$ (κ^{-1} being the Debye length, $\kappa^2=4\pi\beta e^2 n$, n the density, and β the inverse temperature) which is greater than unity for strong coupling. The physical characteristics of the system manifest themselves either as static properties (thermodynamic quantities, pair-correlation function, etc.) or as dynamic properties (dynamical-structure factor, dynamical response functions, collective modes, etc.). Among the theoretical methods pertaining to the former, the hypernetted chain (HNC) approximation[4] has been taken over from the theory of dense fluids, while others have been formulated with a view to their application to strongly coupled plasmas. The method proposed by Totsuji and Ichimaru (TI)[5] is based on the decomposition of the three-particle correlation function by mimicking its weakly coupled plasma structure. Singwi, Tosi, Land, and Sjolander (STLS)[6] and later Vashishta and Singwi[7] worked out a method for the calculation of the dielectric response function by approximating the nonequilibrium pair-correlation function. Some time ago, the present authors and Silevitch (GKS)[8] proposed a somewhat similar scheme, where, however, the approximation was carried out in a much more satisfactory way by evoking the velocity-average approximation (VAA) as its principal assumption.

On the dynamical level, the dynamical continuation of the STLS and the dynamical extension of the TI schemes turned out to be wholly unsatisfactory: actually they are structurally equivalent to each other[9] and to the mean-field theories of Nelkin and Ranganathan[10] and of Lebowitz, Percus, and Sykes.[11] As such, they are unable to reproduce any collisional or long-time effects in the dynamical form factor and in the behavior of collective modes. Independent methods, geared principally to the study of the dynamical form factor in dense neutral fluids but adapted or adaptable to the dense ocp situation, have been put forward in great variety. Generally speaking, there are attempts to improve the mean-field approach,[10-12] assumptions for the Mori memory function,[11,13] and schemes that inject approximations into the equation of motion for the dynamical form factor.[14,15] All these approaches share the common feature that they start with an already given form of the static pair-correlation function which is assumed to be determined either by computer or other experimental data or by an independent theoretical approach. Another line of approach originates from quantum many-body formalism and calculates the dynamical response function. Mukhopadhyay, Kalia, Singwi, and Gupta[16] and Ichimaru[17] should be mentioned in this connection.

The GKS theory has, probably for the first time, provided an unambiguous unified scheme from which both static and nontrivial dynamical approximations can be generated. There are two principal building blocks to the construction of the scheme. The first is the VAA which, as explained in greater detail in Sec. II, consists of replacing the "irreducible" part of the nonequilibrium two-particle distribution function by a properly chosen velocity average. The formal advantage of this step is that it allows one to replace the nonequilibrium two-particle distribution function by a nonequilibrium two-point density-correlation function, which, in turn, can be traded for an equilibrium three-point function. The second building block of the scheme is the application of the nonlinear (quadratic) fluctuation-dissipation theorem (NLFDT),[18] which introduces quadratic response functions as the basic objects whose approximation is required. There seems to be a much more natural and direct way, especially in the dynamical case, to generate approximate structures for these quadratic response functions than for any other quantity that might be a candidate for occupying a central place in the theory.

The GKS scheme has already yielded excellent formal results in the static case, where it goes further than any other approach in satisfying compressibility sum rules for the linear and quadratic polarizability response functions.[19] The VAA has also been shown to be rigorously compatible with the first two Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) equations for an ocp[20]—even though there are situations, e.g., the two-component plasma,[21] when the equivalence ceases to prevail and certain exact symmetry requirements are violated.

That the GKS theory is capable of handling dynamical effects beyond the level of the mean-field description was evident from its original formulation.[8] However, no detailed development of the dynamical theory that could lead to actual calculations has so far been given. The exposition of such a development is the primary purpose of the present paper. Our ultimate goal is the establishment of a formalism based on self-consistency for the calculation of linear and quadratic response functions, which can lead to the determination of these response functions, at least in certain parameter domains, for arbitrary values of γ. The fundamental relation satisfying this objective is set forth in Sec. V, in the form of a relatively simple quadratic integral equation for $\alpha(\vec{k}, \omega)$, the linear polarizability. The parameter domain to which it is restricted consists of the regions of small k, but arbitrary ω values; thus it is readily amenable to the study of the dispersion of collective modes. We contend that the relationship there derived is the first approximation scheme arrived at from first principles and exhibiting correct and nontrivial static and dynamical features.

Section II of the paper displays the derivation via the VAA of the basic dynamical formula in terms of equilibrium three-point functions. In Sec. III the dynamical NLFDT[18] is introduced and is exploited to provide a basic link between linear and quadratic polarizabilities. The relationship plays a pivotal role in the theory, since further progress or ramifications depend upon the particular approximation adopted for the elimination of the quadratic polarizabilities in favor of linear ones. The method espoused in this paper consists of (i) studying only the small-k behavior of the quadratic polarizability and (ii) postulating that a decomposition of the quadratic polarizability in terms of linear polarizabilities, which prevails in this limit for the weakly coupled ocp, can be relied upon as a paradigm for arbitrary γ. The manipulation of these assumptions leads to the already discussed relation in Sec. V.

The reliability of the approximation scheme is investigated in two independent ways. In Sec. IV we demonstrate that for arbitrary $\vec{k}$ and γ values, $\alpha(\vec{k}, \omega)$ as expressed in terms of the quadratic polarizabilities, exactly satisfies the $1/\omega^4$ moment sum rule.[22] In Sec. VI we compare our results in the small-γ limit with the exact weak-coupling results[23] valid to order γ. We find that all the important correlational and long-time effects, with one exception, are reproduced by our theory with very good numerical accuracy. In particular, the real part of the correlational contribution of $\alpha(\vec{k}, \omega)$ is found to be equal to the exact result within 14% at $\omega \to 0$ and within 0.5% at $\omega = \omega_0$ [$\omega_0 = (4\pi n e^2/m)^{1/2}$ is the plasma frequency]. At high frequencies exact agreement up to the coefficient of the $1/\omega^4$ term is guaranteed by virtue of the satisfaction of the $1/\omega^4$ sum rule; the agreement as to the coefficients of the higher powers of $1/\omega$ is less satisfactory. In the imaginary part of the correlational contribution of $\alpha(\vec{k}, \omega)$, responsible for the collisional damping of the collective modes, the part proportional to γ agrees with the exact result with good accuracy; the so-called dominant $\gamma \ln\gamma^{-1}$ term, however, is not reproduced by the theory. This is the deficiency referred to above; the study of its origin and suggestions for possible remedies will be dealt with in a later work.

II. VELOCITY-AVERAGE APPROXIMATION

External and internal polarizabilities are the principal transport coefficients which provide information about dynamical processes in the system. They are also the central objects in the approximation scheme discussed in this paper.

External polarizabilities, closely related to the more popular density-response functions, are defined through the relations[24]

$$\mathcal{E}^{(1)}(\vec{k}, \omega) = -\hat{\alpha}(\vec{k}, \omega)\hat{E}(\vec{k}, \omega), \tag{1}$$

$$\mathcal{E}^{(2)}(\vec{k}, \omega) = -\sum_{\vec{q}\,\mu} {}_2\hat{\alpha}(\vec{q}, \mu; \vec{k}-\vec{q}, \omega-\mu) \times \hat{E}(\vec{q}, \mu)\hat{E}(\vec{k}-\vec{q}, \omega-\mu) \tag{2}$$

connecting the plasma field $\mathcal{E}$ to the weak external perturbation $\hat{E}$. Internal polarizabilities (usually referred to as polarizabilities) connect $\mathcal{E}$ to the total electric field $E = \hat{E} + \mathcal{E}$. They are defined through the relations

$$\mathcal{E}^{(1)}(\vec{k}, \omega) = -\alpha(\vec{k}, \omega)E^{(1)}(\vec{k}, \omega), \tag{3}$$

$$\mathcal{E}^{(2)}(\vec{k}, \omega) = -\sum_{\vec{q}\,\mu} \frac{{}_2\alpha(\vec{q}, \mu; \vec{k}-\vec{q}, \omega-\mu)}{\epsilon(\vec{k}, \omega)} \times E^{(1)}(\vec{q}, \mu)E^{(1)}(\vec{k}-\vec{q}, \omega-\mu), \tag{4}$$

where $\epsilon(\vec{k}, \omega) = 1 + \alpha(\vec{k}, \omega)$ is the wave-vector- and frequency-dependent dielectric response function. We note that

$$\hat{\alpha}(\vec{k}, \omega) = \alpha(\vec{k}, \omega)/\epsilon(\vec{k}, \omega), \tag{5}$$

$${}_2\hat{\alpha}(\vec{q}, \mu; \vec{k}-\vec{q}, \omega-\mu) = \frac{{}_2\alpha(\vec{q}, \mu; \vec{k}-\vec{q}, \omega-\mu)}{\epsilon(\vec{q}, \mu)\epsilon(\vec{k}-\vec{q}, \omega-\mu)\epsilon(\vec{k}, \omega)}. \tag{6}$$

The calculation of the first-order response functions proceeds from the first BBGKY kinetic equation

$$\left(\frac{\partial}{\partial t} + \vec{v}\cdot\frac{\partial}{\partial \vec{x}} - \frac{e}{m}\hat{\vec{E}}(\vec{x}, t)\cdot\frac{\partial}{\partial \vec{v}}\right) F(\vec{x}, \vec{v}; t) = -\frac{1}{m}\frac{\partial}{\partial \vec{v}}\cdot \int d\vec{v}' \int d\vec{x}' \vec{K}(|\vec{x}-\vec{x}'|)G(\vec{x}, \vec{v}; \vec{x}', \vec{v}'; t) \tag{7}$$

for the one-particle distribution function F (normalized to N, the total number of electrons), where G is the two-particle distribution function [normalized to $N(N-1)$] and $\vec{K}(|\vec{x}-\vec{x}'|) = \vec{\nabla}(e^2/|\vec{x}-\vec{x}'|)$ is the interaction force between the field electron (at $\vec{x}$) and a typical source electron (at $\vec{x}'$).

Following the procedure outlined in Sec. I, we now present a method for converting the right-hand side of (7) into expressions involving nonequilibrium two-point and, subsequently, equilibrium three-point correlation functions. We then calculate the average-density response.

Our basic assumption is that G is well described by its velocity average, in the restricted sense, where only one of the velocity arguments is averaged out:

$$G(\vec{x}, \vec{v}; \vec{x}', \vec{v}'; t) = \tfrac{1}{2}\Big[f(\vec{x}, \vec{v}; t) \int d\vec{v}'' G(\vec{x}, \vec{v}''; \vec{x}', \vec{v}'; t) + f(\vec{x}', \vec{v}'; t) \int d\vec{v}''\, G(\vec{x}, \vec{v}; \vec{x}', \vec{v}''; t)\Big], \tag{8}$$

$$f(\vec{x}, \vec{v}; t) \equiv F(\vec{x}, \vec{v}; t)/\, n(\vec{x}, t), \quad \int d\vec{v} f(\vec{x}, \vec{v}; t) = 1.$$

While this structure is exact in equilibrium, it is certainly an approximation for the perturbed system. The resulting double velocity-space integral term

$$f(\vec{x}, \vec{v}; t) \int d\vec{v}' \int d\vec{v}''\, G(\vec{x}, \vec{v}'; \vec{x}', \vec{v}''; t)$$

which replaces

$$\int d\vec{v}'\, G(\vec{x}, \vec{v}; \vec{x}', \vec{v}'; t)$$

in (7) can then, in turn, be expressed in terms of the nonequilibrium microscopic density-density correlation function $\langle n(\vec{x})n(\vec{x}')\rangle(t)$ since

$$\int d\vec{v} \int d\vec{v}'\, G(\vec{x}, \vec{v}; \vec{x}', \vec{v}'; t) = \langle n(\vec{x})n(\vec{x}')\rangle(t) - \delta(\vec{x}-\vec{x}')n(\vec{x}, t). \tag{9}$$

$$n(\vec{x}) = \sum_{j=1}^{N} \delta(\vec{x}-\vec{x}_j).$$

The $n(\vec{x})$ and $n(\vec{x}')$ are equal-time operators and the notation $\langle\ \rangle(t)$ refers to the time evolution carried by the phase-space distribution function [see Eq. (A1)].

Upon combining (7) to (9) and taking the Fourier transform of the result, one obtains the VAA kinetic equation

$$-i(\omega - \vec{k}\cdot\vec{v})F(\vec{k}, \vec{v}; \omega) - \frac{e}{m}\sum_{\vec{q}\,\mu} \hat{\vec{E}}(q, \mu)\cdot\frac{\partial}{\partial \vec{v}}F(\vec{k}-\vec{q}, \vec{v}; \omega-\mu) = \frac{i}{m}\frac{1}{V}\sum_{\vec{q}} \vec{q}\phi(q)\cdot\sum_{\vec{p}\,\nu} \langle n_{\vec{k}-\vec{p}-\vec{q}}\, n_{\vec{q}}\rangle(\omega-\nu)\frac{\partial}{\partial \vec{v}}f(\vec{p}, \vec{v}; \nu), \tag{10}$$

$\phi(q)=4\pi e^2/q^2\,.$

Equation (10) is valid to all orders in $\hat{E}$. The introduction of $\hat{E}$ will perturb $F^0(v)$, the equilibrium Maxwell distribution, by

$$\overline{F}=\sum_{n\geq 1}\overline{F}^{(n)}\,.$$

Application of the subsequent perturbation expansions to (10) results in a hierarchy of coupled VAA kinetic equations. Only the first of these,

$$i(\omega-\vec{k}\cdot\vec{v})\overline{F}^{(1)}(\vec{k},\vec{v};\omega)+\frac{e}{m}\hat{\vec{E}}(\vec{k},\omega)\cdot\frac{\partial F^0(v)}{\partial\vec{v}}=-\frac{i}{m}\frac{1}{N}\sum_{\vec{q}}\vec{q}\phi(q)\cdot\frac{\partial F^0(v)}{\partial\vec{v}}\langle n_{\vec{k}-\vec{q}}n_{\vec{q}}\rangle^{(1)}(\omega)\,, \tag{11}$$

will be of interest in this paper; the second equation of the series has been used in the static development of the theory.[19] The subsequent conversion of the right-hand-side nonequilibrium two-point function into equilibrium three-point functions is made possible by means of straightforward theoretical statistical-mechanical perturbation calculations (Appendix A). Thus from Eq. (A10),

$$\langle n_{\vec{k}-\vec{q}}n_{\vec{q}}\rangle^{(1)}(\omega)=\frac{i\beta e}{Vk}\hat{E}(\vec{k},\omega)\left(i\omega\int_0^\infty dt\,\exp(i\omega t)\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}+\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(0)\rangle^{(0)}\right). \tag{12}$$

From (11) and (12) one then obtains the average density

$$\begin{aligned}n^{(1)}(\vec{k},\omega)&=\int d\vec{v}\,\overline{F}^{(1)}(\vec{k},\vec{v};\omega)\\&=\frac{ik}{4\pi e}\alpha_0(\vec{k},\omega)\hat{E}(\vec{k},\omega)\left[1-\frac{\kappa^2}{k^2}\frac{1}{N^2}\sum_{\vec{q}}\frac{\vec{k}\cdot\vec{q}}{q^2}\left(i\omega\int_0^\infty dt\,\exp(i\omega t)\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}\right.\right.\\&\qquad\left.\left.+\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(0)\rangle^{(0)}\right)\right],\end{aligned} \tag{13}$$

where

$$\alpha_0(\vec{k},\omega)=\frac{\phi(k)}{m}\int d\vec{v}\,\frac{\vec{k}\cdot\partial F^0(v)/\partial\vec{v}}{\omega-\vec{k}\cdot\vec{v}}$$

is the RPA value of the linear polarizability.

III. POLARIZABILITY FORMULATIONS

Equation (13) and the well-known density-response relation

$$n^{(1)}(\vec{k},\omega)=(ik/4\pi e)\hat{\alpha}(\vec{k},\omega)\hat{E}(\vec{k},\omega)$$

immediately lead to the VAA polarizability expression

$$\hat{\alpha}(\vec{k},\omega)=\alpha_0(\vec{k},\omega)\left[1-\frac{\kappa^2}{k^2}\frac{1}{N^2}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left(i\omega\int_0^\infty dt\,\exp(i\omega t)\langle n_{\vec{k}-\vec{p}}(0)n_{\vec{p}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}+\langle n_{\vec{k}-\vec{p}}(0)n_{\vec{p}}(0)n_{-\vec{k}}(0)\rangle^{(0)}\right)\right]\,. \tag{14}$$

Equation (14) can be represented in the form

$$\hat{\alpha}(\vec{k},\omega)=\hat{\alpha}_0(\vec{k},\omega)[1+\hat{v}(\vec{k},\omega)]\,, \tag{15}$$

or, equivalently, expressed as a relation for the internal polarizability,

$$\alpha(\vec{k},\omega)=\alpha_0(\vec{k},\omega)[1+v(\vec{k},\omega)]\,, \tag{16}$$

$$\hat{v}(\vec{k},\omega)=v(\vec{k},\omega)/\epsilon(\vec{k},\omega)\,, \tag{17}$$

where the screening functions $\hat{v}(\vec{k},\omega)$ or $v(\vec{k},\omega)$ are responsible for the finite coupling correction.

We now further transform $\hat{v}(\vec{k},\omega)$ into a more transparent expression through a series of algebraic manipulations. First decompose the equilibrium three-point function into its "proper" and "improper" components:

$$\begin{aligned}\langle n_{\vec{k}-\vec{p}}(0)n_{\vec{p}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}&\\=N(\delta_{\vec{p}}+\delta_{\vec{k}-\vec{p}})\langle n_{\vec{k}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}&\\+\langle n_{\vec{k}-\vec{p}}(0)n_{\vec{p}}(0)n_{-\vec{k}}(-t)\rangle^{(0)}\big|_{\vec{p},\vec{k}-\vec{p},\vec{k}\neq 0}\,,&\end{aligned} \tag{18}$$

and define the quadratic dynamical and static structure factors

$$S(\vec{p},\mu;\vec{k}-\vec{p},\nu)=\frac{1}{N}\int_{-\infty}^{\infty}\frac{d\chi}{2\pi}\langle n_{\vec{p}}(\mu)n_{\vec{k}-\vec{p}}(\nu)n^{*}_{\vec{k}}(\chi)\rangle^{(0)}\Big|_{\vec{p},\vec{k}-\vec{p},\vec{k}\neq 0}, \tag{19a}$$

$$S(\vec{p},\vec{k}-\vec{p})=\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{+\infty}\frac{d\nu}{2\pi}S(\vec{p},\mu;\vec{k}-\vec{p},\nu). \tag{19b}$$

$\hat{v}(\vec{k},\omega)$ can now be written as

$$\hat{v}(\vec{k},\omega)=-\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left(\frac{i\omega}{2\pi}\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\,\delta_{+}(\omega-\mu-\nu)\times S(\vec{p},\mu;\vec{k}-\vec{p},\nu)+S(\vec{p},\vec{k}-\vec{p})\right). \tag{20}$$

Further progress can be made and, ultimately, self-consistency can be achieved first by expressing $\hat{v}$ in terms of quadratic polarizability response functions. This is to be accomplished with the aid of the dynamical NLFDT[18] which relates the quadratic structure factor to the latter:

$$S(\vec{p},\mu;\vec{k}-\vec{p},\nu)=-2\,\mathrm{Im}\left(\frac{\hat{\alpha}(\vec{p},\mu;\vec{k}-\vec{p},\nu)}{\mu\nu}-\frac{\hat{\alpha}(\vec{p},-\mu;-\vec{k},\mu+\nu)}{\mu(\mu+\nu)}-\frac{\hat{\alpha}(-\vec{k},\mu+\nu;\vec{k}-\vec{p},-\nu)}{\nu(\mu+\nu)}\right), \tag{21}$$

where the $\hat{\alpha}$'s are quadratic external polarizabilities conveniently normalized to

$${}_2\alpha_0(\vec{p},0;\vec{k}-\vec{p},0)=2\pi i\beta^2 ne^3/(kp|\vec{k}-\vec{p}|),$$

the static ($\mu=\nu=0$) value of ${}_2\alpha(\vec{p},\mu;\vec{k}-\vec{p},\nu)$ in the RPA limit[18]: for example,

$$\hat{\alpha}(\vec{p},\mu;\vec{k}-\vec{p},\nu)\equiv i\,{}_2\alpha(\vec{p},\mu;\vec{k}-\vec{p},\nu)/{}_2\alpha_0(\vec{p},0;\vec{k}-\vec{p},0). \tag{22}$$

Since $S(\vec{p},\mu;\vec{k}-\vec{p},\nu)$ is expected to be nonsingular, the $\mu=0$, $\nu=0$, and $\mu=-\nu$ singularities in (21) are spurious, and the nonlinear fluctuation-dissipation theorem remains unchanged if one stipulates that each frequency denominator in (21) is a (double) principal-value denominator. With this understanding, the injection of Eq. (21) into Eq. (20) leads to expressions which are then amenable to Kramers-Kronig analysis. They are

$$\begin{aligned}\hat{v}'(\vec{k},\omega)=-\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\Bigg(&\hat{\alpha}''(\vec{p},0;\vec{k}-\vec{p},0)+2\omega\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\hat{\alpha}''(\vec{p},\mu;\vec{k}-\vec{p},\nu)P\frac{1}{\mu}P\frac{1}{\nu}P\frac{1}{\omega-\mu-\nu}\\&-2\omega\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\hat{\alpha}''(\vec{p},-\mu;-\vec{k},\mu+\nu)P\frac{1}{\mu}P\frac{1}{\mu+\nu}P\frac{1}{\omega-\mu-\nu}\\&-2\omega\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\hat{\alpha}''(-\vec{k},\mu+\nu;\vec{k}-\vec{p},-\nu)P\frac{1}{\nu}P\frac{1}{\mu+\nu}P\frac{1}{\omega-\mu-\nu}\Bigg),\end{aligned} \tag{23}$$

$$\begin{aligned}\hat{v}''(\vec{k},\omega)=\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\Bigg(&\omega\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\hat{\alpha}''(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)P\frac{1}{\mu}P\frac{1}{\omega-\mu}\\&-\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\hat{\alpha}''(\vec{p},-\mu;-\vec{k},\omega)P\frac{1}{\mu}-\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\hat{\alpha}''(-\vec{k},\omega;\vec{k}-\vec{p},\mu-\omega)P\frac{1}{\omega-\mu}\Bigg),\end{aligned} \tag{24}$$

where $\hat{v}=\hat{v}'+i\hat{v}''$, $\hat{\alpha}=\hat{\alpha}'+i\hat{\alpha}''$, etc. The first right-hand-side term of (23) comes from the static NLFDT,[18]

$$S(\vec{p},\vec{k}-\vec{p})=\hat{\alpha}''(\vec{p},0;\vec{k}-\vec{p},0). \tag{25}$$

In view of the fact that $\hat{\alpha}$ is a plus function of its frequency arguments, the expression (24) for $\hat{v}''$ further simplifies to

$$\hat{v}''(\vec{k},\omega)=\frac{\kappa^2}{k^2}\frac{1}{2N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left(\frac{\omega}{\pi}PP\int_{-\infty}^{\infty}\frac{d\mu}{\mu(\omega-\mu)}\hat{\alpha}''(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)+\hat{\alpha}'(\vec{p},0;-\vec{k},\omega)+\hat{\alpha}'(-\vec{k},\omega;\vec{k}-\vec{p},0)\right). \tag{26}$$

In order to convert $\hat{v}'$ into a form similar to (26), we manipulate the second right-hand-side term in (23),

$$PPP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(\vec{p},\mu;\vec{k}-\vec{p},\nu)}{\mu\nu(\omega-\mu-\nu)}$$

$$=\frac{1}{\omega}\left(PP\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\hat{a}''(\vec{p},\mu;k-\vec{p},\nu)+PP\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu}\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\hat{a}''(\vec{p},\mu;\vec{k}-\vec{p},\nu)\right.$$

$$\left.+PP\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\int_{-\infty}^{\infty}\frac{d\nu}{\omega-\mu-\nu}\hat{a}''(\vec{p},\mu;\vec{k}-\vec{p},\nu)+PP\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu}\int_{-\infty}^{+\infty}\frac{d\nu}{\omega-\mu-\nu}\hat{a}''(\vec{p},\mu;\vec{k}-\vec{p},\nu)\right)$$

$$=-\frac{\pi^2}{\omega}[\hat{a}''(\vec{p},0;\vec{k}-\vec{p},0)-\hat{a}''(\vec{p},\omega;\vec{k}-\vec{p},0)]$$

$$-\frac{\pi}{\omega}\left[P\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{a}'(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)+P\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu}\hat{a}'(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)\right]. \tag{27}$$

For the third right-hand-side term one can similarly show that

$$PPP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(\vec{p},-\mu;-\vec{k},\mu+\nu)}{\mu(\mu+\nu)(\omega-\mu-\nu)}=\frac{\pi^2}{\omega}[\hat{a}''(\vec{p},0;-\vec{k},0)-\hat{a}''(\vec{p},0;-\vec{k},\omega)]. \tag{28}$$

Finally, the fourth right-hand-side term,

$$PPP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(-\vec{k},\mu+\nu;\vec{k}-\vec{p},-\nu)}{\nu(\mu+\nu)(\omega-\mu-\nu)}$$

$$=\frac{1}{\omega}\left(PP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(-\vec{k},\mu+\nu;\vec{k}-\vec{p},-\nu)}{\nu(\mu+\nu)}+PP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(-\vec{k},\mu+\nu;\vec{k}-\vec{p},-\nu)}{\nu(\omega-\mu-\nu)}\right)$$

$$=-\frac{1}{\omega}\left(PP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(-\vec{k},\mu-\nu;\vec{k}-\vec{p},\nu)}{\nu(\mu-\nu)}+PP\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\frac{\hat{a}''(-\vec{k},\mu-\nu;\vec{k}-\vec{p},\nu)}{\nu[\omega-(\mu-\nu)]}\right), \tag{29}$$

can be transformed with the application of the Poincaré-Bertrand theorem[25] into

$$-\frac{1}{\omega}\left(PP\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\int_{-\infty}^{\infty}\frac{d\mu}{\mu-\nu}\hat{a}''(-\vec{k},\mu-\nu;\vec{k}-\vec{p},\nu)+\pi^2\hat{a}''(-\vec{k},0;\vec{k}-\vec{p},0)\right)$$

$$-\frac{1}{\omega}\left(PP\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\int_{-\infty}^{\infty}\frac{d\mu}{\omega-(\mu-\nu)}\hat{a}''(-\vec{k},\mu-\nu;\vec{k}-\vec{p},\nu)-\pi^2\hat{a}''(-\vec{k},\omega;\vec{k}-\vec{p},0)\right)=0. \tag{29a}$$

Combining Eqs. (27)–(29) according to (23) and taking account of symmetry properties like $\hat{a}(\vec{p},\omega;\vec{k}-\vec{p},0)=\hat{a}(-\vec{k},\omega;\vec{k}-\vec{p},0)$ (see Appendix B), one obtains

$$\hat{v}'(\vec{k},\omega)=\frac{\kappa^2}{k^2}\frac{1}{2N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left(\frac{\omega}{\pi}PP\int_{-\infty}^{\infty}\frac{d\mu}{\mu(\omega-\mu)}\hat{a}'(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)-\hat{a}''(\vec{p},0;-\vec{k},\omega)-\hat{a}''(-\vec{k},\omega;\vec{k}-\vec{p},0)\right). \tag{30}$$

Equations (30) and (26) then combine, in turn, to give the desired expression:

$$\hat{v}(\vec{k},\omega)=\frac{i\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{a}(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)+\hat{a}(\vec{p},\omega-\mu;\vec{k}-\vec{p},\mu)]. \tag{31}$$

Similarly,

$$v(\vec{k},\omega)=\frac{i\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\left(\frac{a(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)}{\epsilon(\vec{p},\mu)\epsilon(\vec{k}-\vec{p},\omega-\mu)}+\frac{a(\vec{p},\omega-\mu;\vec{k}-\vec{p},\mu)}{\epsilon(\vec{p},\omega-\mu)\epsilon(\vec{k}-\vec{p},\mu)}\right), \tag{32}$$

where

$$a(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)\equiv i_2\alpha(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)/_2\alpha_0(\vec{p},0;\vec{k}-\vec{p},0). \tag{33}$$

Equations (31) and (32), together with (15) and (16), constitute the central relations of our approximation scheme. They determine the linear polarizabilities in terms of the quadratic ones. As such, they are evidently not self-consistent; however, they open up avenues to further approximation methods that lead to self-consistency. The seminal question, of course, is how closure can be accomplished. A relatively simple way is to postulate a decomposition of $a(\vec{p}, \mu; \vec{q}, \nu)$ in terms of linear α's. A more ambitious approach is to relegate the closure to a higher level, i.e., to express a (or ${}_2\alpha$) in terms of the cubic polarizability ${}_3\alpha$ with the aid of the first BBGKY kinetic equation perturbed to higher order in $\hat{E}$. Closure is then sought by expressing ${}_3\alpha$ in terms of ${}_2\alpha$ and α. This latter method has been pursued to some extent, rather satisfactorily, in the static ($\omega = 0$) situation,[19] where it has been shown to give excellent results from the point of view of satisfying compressibility sum-rule requirements. In the present paper, however, we will develop the former, simpler decomposition scheme. This will be applicable to the parameter domain $(k/\kappa)\omega_0/\omega \ll 1$ and, therefore, it has no static counterpart. In Sec. IV we will analyze the RPA quadratic polarizability and will show that it has a simple decomposition in terms of linear polarizabilities. This relationship will then be postulated to serve as the basis of the self-consistency scheme for arbitrary values of γ.

We close this section with an important remark about the structure of Eq. (16). Its $\omega = 0$ limit[8,19,20]

$$\alpha(\vec{k}, 0) = \alpha_0(\vec{k}, 0)[1 + v'(\vec{k}, 0)], \tag{34}$$

$$v'(\vec{k}, 0) = -\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\frac{a''(\vec{p}, 0; \vec{k}-\vec{p}, 0)}{\epsilon(\vec{p}, 0)\epsilon(\vec{k}-\vec{p}, 0)}, \tag{35}$$

is *identical to the second* BBGKY *static equation*

$$S(k) - 1 = -\frac{\kappa^2}{k^2 + \kappa^2}\left(1 + \frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}S(\vec{p}, \vec{k}-\vec{p})\right)$$

relating the linear and quadratic structure factors $S(k)$ and $S(\vec{p}, \vec{k}-\vec{p})$. This is readily seen by application of the static NLFDT Equation (25) and $S(k) = \hat{\alpha}(\vec{k}, 0)/\alpha_0(\vec{k}, 0)$ to (34) and (35).

IV. HIGH-FREQUENCY BEHAVIOR

In this section we demonstrate that the approximation scheme satisfies the known ω^4 high-frequency sum rule.[22] We return to expression (20), in terms of the quadratic structure factor,

$$\hat{v}'(\vec{k}, \omega) = -\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left(S(\vec{p}, \vec{k}-\vec{p}) - \omega P\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\frac{S(\vec{p}, \mu; \vec{k}-\vec{p}, \nu)}{\omega - \mu - \nu}\right). \tag{36}$$

For ω large, the principal-part denominator can be expanded and one obtains

$$\begin{aligned}\hat{v}'(\vec{k}, \omega\to\infty) &= \frac{\kappa^2}{k^2}\frac{1}{N\omega^2}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}(\mu+\nu)^2 S(\vec{p}, \mu; \vec{k}-\vec{p}, \nu)\\ &= -\frac{\kappa^2}{k^2}\frac{1}{N\omega^2}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\left[\left(\frac{\partial}{\partial t'} + \frac{\partial}{\partial t''}\right)^2 S(\vec{p}, t'; \vec{k}-\vec{p}, t'')\right]_{t'=t''=0}\\ &= \frac{\kappa^2}{k^2}\frac{1}{N^2\omega^2}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}[\vec{p}\cdot\langle\vec{J}_{\vec{p}}n_{\vec{k}-\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}\cdot\vec{k} + (\vec{k}-\vec{p})\cdot\langle\vec{J}_{\vec{k}-\vec{p}}n_{\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}\cdot\vec{k}]\Big|_{\vec{p}, \vec{k}-\vec{p}, \vec{k}\neq 0},\end{aligned} \tag{37}$$

$$\vec{J}_{-\vec{k}} = \sum_{j=1}^{N}\vec{v}_j\exp(i\vec{k}\cdot\vec{x}_j).$$

The calculation in Appendix C provides

$$\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}[p\cdot\langle\vec{J}_{\vec{p}}n_{\vec{k}-\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}\cdot\vec{k} + (\vec{k}-\vec{p})\cdot\langle\vec{J}_{\vec{k}-\vec{p}}n_{\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}\cdot\vec{k}]\Big|_{\vec{p}, \vec{k}-\vec{p}, \vec{k}\neq 0} = k^2(1/\beta m)\sum_{\vec{p}}\chi^2[S(|\vec{k}-\vec{p}|) - S(p)], \tag{38}$$

where $\chi \equiv \vec{k}\cdot\vec{p}/kp$. The resulting statement for $\hat{v}'$,

$$\hat{v}'(\vec{k}, \omega\to\infty) = \frac{\omega_0^2}{\omega^2}\frac{1}{N}\sum_{\vec{p}}\chi^2[S(|\vec{k}-\vec{p}|) - S(p)], \tag{39}$$

is tantamount to the $1/\omega^4$ sum rule.[22,26] Recalling that $\hat{v}(\vec{k}, \omega) = v(\vec{k}, \omega)/\epsilon(\vec{k}, \omega)$, note that, to order $1/\omega^4$, the right-hand side of (39) is also the correct expression for $v(\vec{k}, \omega)$. In the known long-wavelength limit,

$$\hat{v}'(\vec{k}\to 0,\omega\to\infty)=-\frac{4\gamma}{15\pi}\frac{\omega_0^2}{\omega^2}\frac{k^2}{\kappa^2}\left|\int_0^\infty dx\,[S(x)-1]\right|, \tag{40}$$

$x\equiv p/\kappa$.

Finally, in the $\gamma\to 0$ limit where $S(p)=S_0(p)=p^2/(p^2+\kappa^2)$, one recovers from (39) the long-wavelength Debye-Hückel sum-rule coefficient

$$\hat{v}_0'(\vec{k}\to 0,\omega\to\infty)=-\tfrac{2}{15}\gamma(k^2/\kappa^2)\omega_0^2/\omega^2. \tag{41}$$

V. LONG-WAVELENGTH FORMULATION

The main physical interest lies in the long-wavelength dynamical behavior of the system, especially the long-wavelength damping and dispersion of plasma oscillations. We therefore now turn to the derivation of the long-wavelength ($k\to 0$) formula for $v(\vec{k},\omega)$. Recalling the discussion of Sec. III about the structure of a, we shall suppose that it can be described by an RPA-like structure. The latter is

$$a_0(\vec{p},\mu;\vec{q},\nu)=\frac{i}{\beta mn}\int d\vec{v}\,F^0(v)\frac{1}{(\omega-\vec{k}\cdot\vec{v})^2}\times\left(\vec{k}\cdot\vec{p}\frac{\vec{q}\cdot\vec{v}}{\nu-\vec{q}\cdot\vec{v}}+\vec{k}\cdot\vec{q}\frac{\vec{p}\cdot\vec{v}}{\mu-\vec{p}\cdot\vec{v}}\right), \tag{42}$$

where $\vec{q}=\vec{k}-\vec{p}$, $\nu=\omega-\mu$. The subsequent development in $|\vec{k}\cdot\vec{v}/\omega|$ to order k^2 then results in the following relationship between a_0 and α_0:

$$\begin{aligned}a_0(\vec{p},\mu;\vec{q},\nu)=&(\omega_0^2/\omega^2)[A^{(1)}(\vec{p},\mu)+A^{(1)}(\vec{q},\nu)]\\&+2(\omega_0^3/\omega^3)[A^{(2)}(\vec{p},\mu)+A^{(2)}(\vec{q},\nu)]\\&+3(\omega_0^4/\omega^4)[A^{(3)}(\vec{p},\mu)+A^{(3)}(\vec{q},\nu)],\end{aligned} \tag{43}$$

$$A^{(1)}(\vec{p},\mu)=-\frac{ip^2\vec{k}\cdot\vec{q}}{\kappa^4}\alpha_0(\vec{p},\mu), \tag{44}$$

$$A^{(2)}(\vec{p},\mu)=-\frac{i\mu}{\omega_0}\frac{\vec{k}\cdot\vec{p}\vec{k}\cdot\vec{q}}{\kappa^4}\alpha_0(\vec{p},\mu), \tag{45}$$

$$A^{(3)}(\vec{p},\mu)=-\frac{ik^2p^2\vec{k}\cdot\vec{q}}{\kappa^6}\left(1-\chi^2+\frac{\mu^2}{\omega_0^2}\frac{\kappa^2}{p^2}\chi^2\right)\alpha_0(\vec{p},\mu)-\frac{i\vec{k}\cdot\vec{q}(\vec{k}\cdot\vec{p})^2}{\kappa^4p^2}. \tag{46}$$

Now write v in the form

$$v_0(\vec{k},\omega)=(\omega_0/\omega)^2v^{(1)}+2(\omega_0/\omega)^3v^{(2)}+3(\omega_0/\omega)^4v^{(3)}, \tag{47}$$

$$v^{(j)}=i\frac{\kappa^2}{k^2}\frac{1}{N}\sum_{\vec{p}}\frac{\vec{k}\cdot\vec{p}}{p^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\left(\frac{A^{(j)}(\vec{p},\mu)+A^{(j)}(\vec{k}-\vec{p},\omega-\mu)}{\epsilon_0(\vec{p},\mu)\epsilon_0(\vec{k}-\vec{p},\omega-\mu)}+\frac{A^{(j)}(\vec{p},\omega-\mu)+A^{(j)}(\vec{k}-\vec{p},\mu)}{\epsilon_0(\vec{p},\omega-\mu)\epsilon_0(\vec{k}-\vec{p},\mu)}\right)\quad(j=1,2,3). \tag{48}$$

For $v^{(1)}$ we have from (48),

$$\begin{aligned}v^{(1)}\simeq\frac{1}{N}\int_{-\infty}^{\infty}d\mu\delta_-(\mu)\sum_{\vec{p}}\Bigg(&\frac{k^2\chi^2}{\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)+\hat{\alpha}_0(\vec{p},\omega-\mu)-2\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)]\\&-\frac{2kp\chi^3}{\kappa^2}[\hat{\alpha}_0(\vec{k}-\vec{p},\mu)+\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu)]\\&-\frac{kp(\chi-2\chi^3)}{\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu)+\hat{\alpha}_0(\vec{k}-\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)]\\&-\frac{p^2\chi^2}{\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)-\hat{\alpha}_0(\vec{k}-\vec{p},\mu)+\hat{\alpha}_0(\vec{p},\omega-\mu)-\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu)]\Bigg).\end{aligned} \tag{49}$$

By virtue of the plus-function character of $\hat{\alpha}(\omega)$, Eq. (49) splits into

$$v^{(1)}=v^{(1)}_{\rm dyn}+v^{(1)}_{\rm stat}, \tag{50}$$

where

$$\begin{aligned}v^{(1)}_{\rm dyn}=-\frac{2}{N}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\sum_{\vec{p}}\Bigg(&\frac{k^2\chi^2}{\kappa^2}\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)\\&+\frac{kp(\chi-2\chi^3)}{2\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu)+\hat{\alpha}_0(\vec{k}-\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)]\Bigg)\end{aligned} \tag{51}$$

can be reduced to

$$v^{(1)}_{\rm dyn} \simeq -\frac{k^2}{\kappa^2}\frac{1}{N}\sum_{\vec{p}}(1-4\chi^2+4\chi^4) \times \int_{-\infty}^{\infty} d\mu\, \delta_-(\mu)\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu), \tag{52}$$

and where

$$v^{(1)}_{\rm stat} = \frac{1}{N}\sum_{\vec{p}}\left(\frac{k^2\chi^2}{\kappa^2}\hat{\alpha}_0(\vec{p},0) - 2\frac{kp\chi^3}{\kappa^2}\hat{\alpha}_0(\vec{k}-\vec{p},0) - \frac{p^2\chi^2}{\kappa^2}[\hat{\alpha}_0(\vec{p},0)-\hat{\alpha}_0(\vec{k}-\vec{p},0)]\right) \simeq -\tfrac{2}{15}\gamma k^2/\kappa^2. \tag{53}$$

The reduction of $v^{(2)}$ is carried out along similar lines:

$$\begin{aligned} v^{(2)} \simeq \frac{2}{N}\int_{\infty}^{\infty} d\mu\, \delta_-(\mu) \sum_{\vec{p}} & \frac{k^2\chi^2}{\kappa^2}\left(\frac{\mu}{\omega_0}\hat{\alpha}_0(\vec{p},\mu) + \frac{\omega-\mu}{\omega_0}\hat{\alpha}_0(\vec{p},\omega-\mu) - \frac{\omega}{\omega_0}\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)\right) \\ & - \frac{kp\chi^3}{\kappa^3}\left(\frac{\mu}{\omega_0}\hat{\alpha}_0(\vec{k}-\vec{p},\mu) + \frac{\omega-\mu}{\omega_0}\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu)\right) \\ & + \frac{\omega}{\omega_0}\frac{kp\chi^3}{\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu) + \hat{\alpha}_0(\vec{k}-\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu)] \\ = v^{(2)}_{\rm dyn} + v^{(2)}_{\rm stat}\;, & \end{aligned} \tag{54}$$

where

$$\begin{aligned} v^{(2)}_{\rm dyn} &= -\frac{2}{N}\frac{\omega}{\omega_0}\int_{-\infty}^{\infty} d\mu\, \delta_-(\mu) \sum_{\vec{p}}\left(\frac{k^2\chi^2}{\kappa^2}\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu) - \frac{kp\chi^3}{2\kappa^2}[\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{k}-\vec{p},\omega-\mu) + \hat{\alpha}_0(\vec{p},\omega-\mu)\hat{\alpha}_0(\vec{k}-\vec{p},\mu)]\right) \\ &= -\frac{k^2}{\kappa^2}\frac{\omega}{\omega_0}\frac{1}{N}\sum_{\vec{p}}(-\chi^2+2\chi^4)\int_{-\infty}^{\infty} d\mu\, \delta_-(\mu)\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu), \end{aligned} \tag{55}$$

while

$$v^{(2)}_{\rm stat} = 0 \tag{56}$$

by virtue of the fact that both

$$\int d\mu\, \delta_-(\mu)(\omega-\mu)\hat{\alpha}_0(\omega-\mu) = 0$$

and

$$\int d\mu\, \delta_-(\mu)\mu\,\hat{\alpha}_0(\mu) = 0\,.$$

Finally, one can readily demonstrate that

$$v^{(3)} = 0 \tag{57}$$

to order k^2. We note that one can infer (53) together with (56) and (57) from the general sum-rule result of Sec. IV, Eq. (39).

We now combine Eqs. (47), (52), (55)–(57), and (53); as to the last, however, we replace its contribution by that corresponding to the exact sum-rule result (39) valid for $v(\vec{k},\omega)$ as well. The resulting long-wavelength formula,

$$v(\vec{k},\omega) \simeq \frac{\omega_0^2}{\omega^2}\frac{1}{N}\sum_{\vec{p}}\chi^2[S(|\vec{k}-\vec{p}|)-S(p)] - \frac{\omega_0^2}{\omega^2}\frac{k^2}{\kappa^2}\frac{1}{N}\sum_{\vec{p}}(1-6\chi^2+8\chi^4) \times \int_{-\infty}^{\infty} d\mu\, \delta_-(\mu)\hat{\alpha}(\vec{p},\mu)\hat{\alpha}(\vec{p},\omega-\mu), \tag{58}$$

we now propose to be valid for *arbitrary coupling*. This, together with our exact relation (16), comprises the proposed self-consistent approximation scheme.

VI. WEAK-COUPLING LIMIT

The fundamental expression (16), together with the small-k approximation for $v(\vec{k},\omega)$, Eq. (58), is based on the VAA and the assumption of the validity of the RPA-like structure of $a(\vec{p},\mu;\vec{q},\nu)$ in terms of linear α's. In order to assess the physical contents of the VAA in the present context, we will evaluate the weak-coupling limit of $v(\vec{k},\omega)$. Since the exact $\gamma \ll 1$ expression for $\alpha(\vec{k},\omega)$ is known[23] to order γ, this calculation will allow us to compare, to this order, $v(\vec{k},\omega)$

with its exact counterpart.

The only approximation required for the evaluation of $v(\vec{k}, \omega)$ is the "static approximation," which amounts to replacing

$$\hat{\alpha}(\vec{p}, \mu) = \alpha(\vec{p}, \mu)/\epsilon(\vec{p}, \mu)$$

and

$$\hat{\alpha}(\vec{p}, \omega-\mu) = \alpha(\vec{p}, \omega-\mu)/\epsilon(\vec{p}, \omega-\mu)$$

by $\alpha(\vec{p}, \mu)/\epsilon(\vec{p}, 0)$ and $\alpha(\vec{p}, \omega-\mu)/\epsilon(\vec{p}, 0)$, respectively. This approximation, which is probably quite good, except for $\omega \simeq 2\omega_0$ has been used[15] rather extensively; it is also instrumental in a calculation leading to the evaluation[23] of the exact $v(\vec{k}, \omega)$ and, thus it indeed provides the appropriate basis for the desired comparison.

Using now both the well-known RPA expression for α and the static approximation, Eq. (58) can be rewritten as

$$v(\vec{k}, \omega) = -\gamma \frac{\omega_0^2}{\omega^2} \frac{k^2}{\kappa^2} [\mathcal{V}_{\text{stat}} + \mathcal{V}_{\text{dyn}}(\omega)],$$

$$\begin{aligned}\mathcal{V}_{\text{dyn}}(\omega) &= \frac{6}{5\pi} \int_0^\infty dx\, x^2 \int_{-\infty}^\infty d\mu\, \delta_-(\mu) \hat{\alpha}_0(\vec{p}, \mu) \hat{\alpha}_0(\vec{p}, \omega-\mu) \\ &\simeq \frac{6}{5\pi} \int_0^\infty dx \frac{x^6}{(1+x^2)^2} H(\vec{p}, \omega), \quad x = p/\kappa, \end{aligned} \tag{59}$$

$$H(\vec{p}, \omega) = \int_{-\infty}^\infty d\mu\, \delta_-(\mu) \alpha_0(\vec{p}, \mu) \alpha_0(\vec{p}, \omega-\mu).$$

It already has been pointed out that the value of $\mathcal{V}_{\text{stat}}$ is in agreement with the sum-rule requirement, i.e., $\mathcal{V}_{\text{stat}} = \frac{2}{15}$. Exploiting now the fact that $\alpha(\vec{p}, \mu) \equiv \alpha^*(\vec{p}, \mu) + 2i\alpha''(\vec{p}, \mu)$ is a plus function and therefore $\alpha^*(\vec{p}, \mu)$ as well as $\alpha(\vec{p}, \omega-\mu)$ are minus functions of μ, and that $\alpha''(\vec{p}, 0) = 0$, $H(\vec{p}, \omega)$ becomes

$$H(\vec{p}, \omega) = \frac{1}{\pi} \int_{-\infty}^\infty \frac{d\mu}{\mu} \alpha_0''(\vec{p}, \mu) \alpha_0(\vec{p}, \omega-\mu). \tag{60}$$

The RPA expression for $\alpha(p, \nu)$ in terms of the plasma-dispersion function (o is a positive infinitesimal quantity)

$$Z(w) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^\infty dz \frac{\exp(-z^2/2)}{z - w - io} \tag{61}$$

is

$$\alpha_0(\vec{p}, \nu) = (\kappa^2/p^2)[1 + wZ(w)], \tag{62}$$

where $w = \nu/ap$ and $a = w_0/\kappa$. Introducing (62) into (60), one obtains

$$\begin{aligned} H(\vec{p}, \omega) &= \frac{\kappa^4}{\pi p^4} \left[\int_{-\infty}^\infty dw\, Z''(w) + \frac{\omega}{ap} \int_{-\infty}^\infty dw\, Z''(w) Z\left(\frac{\omega}{ap} + w\right) \right. \\ &\quad \left. + \int_{-\infty}^\infty dw\, w Z''(w) Z\left(\frac{\omega}{ap} + w\right)\right], \end{aligned} \tag{63}$$

where $Z = Z' + iZ''$. The evaluation of the integrals can be done by using a method originally suggested by Coste.[23] The details are given in Appendix D. The result for $H(\vec{p}, \omega)$ is

$$H(\vec{p}, \omega) = \frac{\kappa^4}{2p^4} \left[1 + \frac{1}{\sqrt{2}} \frac{\omega}{ap} Z\left(\frac{\omega}{\sqrt{2}\, ap}\right)\right]. \tag{64}$$

Eq. (64) substituted into (59) now leads to

$$\begin{aligned}\mathcal{V}_{\text{dyn}}(\omega) &= \frac{3}{5\pi} \int_0^\infty dx \frac{x^2}{(1+x^2)^2} [1 + uZ(u)] \\ &= \frac{3}{5\pi} \left[\frac{\pi}{4} + \frac{1}{2\sqrt{2}} \left(\frac{\omega}{\omega_0}\right)^3 \int_{-\infty}^\infty \frac{du}{u^3} \frac{Z(u)}{(1+\omega^2/2u^2\omega_0^2)^2}\right], \end{aligned} \tag{65}$$

where $u = \omega\sqrt{2}\, ap$. The integral can be evaluated with the aid of a formula given by Baus[15]; details are again relegated to Appendix D. One is led to the final result

$$\begin{aligned}\mathcal{V}_{\text{dyn}}(\omega) &= \tfrac{3}{20}\{1 - 2\eta^2[1 - \sqrt{\pi}\, \eta e^{\eta^2}(1 - \text{erf}\,\eta)]\} \\ &\quad - (\tfrac{3}{10} i/\sqrt{\pi})\eta[\eta^2 e^{\eta^2} E_1(\eta^2) - 1], \end{aligned} \tag{66}$$

$$\eta = \omega/2\omega_0, \quad E_1(\eta) = \int_\eta^\infty \frac{dt}{t} \exp(-t).$$

The comparison with the exact result can now be carried out on two levels. On the structural level, the exact equivalent of (59) is[23]

$$\begin{aligned}\mathcal{V}_{\text{dyn}}(\omega) &= \frac{46}{15\pi} \int_0^\infty dx\, x^2 \int_{-\infty}^\infty d\mu\, \delta_-(\mu) \hat{\alpha}_0(\vec{p}, \hat{\mu}) \hat{\alpha}_0(\vec{p}, \omega-\mu) \\ &\quad - \frac{16}{15\pi} \frac{\omega_0^2}{\omega^2} \int_0^\infty dx \left(\frac{x^2}{1+x^2} - x^4 \int_{-\infty}^\infty d\mu\, \delta_-(\mu)[\hat{\alpha}(\vec{p}, \mu)\alpha(\vec{p}, \omega-\mu) + \alpha(\vec{p}, \mu)\hat{\alpha}(\vec{p}, \omega-\mu)]\right) \\ &\simeq \frac{46}{15\pi} \int_{-\infty}^\infty dx \frac{x^6}{(1+x^2)^2} H(\vec{p}, \omega) - \frac{16}{15\pi} \frac{\omega_0^2}{\omega^2} \int_0^\infty dx \frac{x^2}{1+x^2} (1 - 2x^4 H(\vec{p}, \omega)). \end{aligned} \tag{67}$$

TABLE I. Values of $\mathcal{V}(\omega)$ at $\omega=0$, $\omega=\omega_0$, and $\omega \gg \omega_0$.

ω	VAA	Exact
$\omega=0$	0.283	$0.250 + i(\omega_0/\omega)0.301\ln\gamma^{-1}$
$\omega=\omega_0$	$0.249 + i0.056$	$0.248 + i(0.301\ln\gamma^{-1} + 0.064)$
$\omega\to\infty$	$0.133 + i(\omega_0/\omega)0.338$	$0.133 + i(\omega_0/\omega)[0.301\ln(2\omega_0\gamma^{-1}/\omega) + 0.778]$

In comparing (59) and (67), we see that the difference lies (i) in the different numerical coefficient in front of the first integral and (ii) in the appearance of the second integral. To some extent, the two effects are compensatory. However, the imaginary part of the second integral is divergent as $x\to\infty$. The usual device of cutting the integral off at $p_{\max}=\gamma^{-1}\kappa$ leads one to the familiar $\gamma\ln\gamma^{-1}$ contribution[15,23,28] to the imaginary part of $\epsilon(\vec{k},\omega)$. This feature is absent in the present approximation.

On the numerical level, we exhibit in Table I the values[23] of $\mathcal{V}(\omega)=\frac{2}{15}+\mathcal{V}_{\rm dyn}(\omega)$ at $\omega=0$, $\omega=\omega_0$, and $\omega\gg\omega_0$. Apart from the absence of the $\ln\gamma^{-1}$ term (which does not affect the real part), the agreement is very good. We note in particular that the change in the dispersion of the plasma oscillation due to finite γ effects is virtually identical in the VAA to what is predicted by the exact theory; both indicate that for small but finite γ, $d\omega/dk < d\omega/dk|_{\gamma=0}$.

VII. CONCLUSIONS

The purpose of the present paper has been to formulate a self-consistent approximation scheme especially suitable for the description of the dispersion of collective modes at long wavelengths in strongly coupled ocp's. This has been accomplished in two stages.

In the first stage, the application of the dynamical NLFDT to the first BBGKY kinetic equation suitably prepared in the VAA led to the important new formulations (15), (16), (31), and (32) of the linear polarizabilities in terms of quadratic ones; these equations are valid at arbitrary values of k, ω, and γ. In the $\omega=0$ limit the approximation scheme goes far beyond satisfying the compressibility sum rules for the linear and quadratic polarizabilities; in fact, one can reproduce the exact second BBGKY static equation from (16), attesting to the accuracy of the VAA at the level of the quadratic polarizability. For arbitrary values of k and γ and at high frequency ($\omega\to\infty$), we demonstrated [cf. Eq. (39)] that $\hat{\alpha}(\vec{k},\omega)$ [and, of course, $\alpha(\vec{k},\omega)$] satisfies the $1/\omega^4$ sum rule.

In the second stage, we made Eqs. (16) and (32) self-consistent in the long-wavelength limit by postulating the decomposition of the dynamical quadratic polarizabilities in terms of linear ones, in analogy with the relation which prevails in this limit for $\gamma\ll 1$. The result is a relatively simple quadratic integral equation for α [obtained from (16) and (58)]. We evaluated (58) in the small-γ limit and compared the result given by Eqs. (59) and (66) with the known exact $\gamma\ll 1$ result (67). Apart from the absence of the $\gamma\ln\gamma^{-1}$ term (which affects the damping but not the dispersion of the collective modes), we see from Table I that over the entire frequency range all other important correlational and long-time effects are reproduced by our theory with very good numerical accuracy.

The decomposition of ${}_2\alpha$ in terms of linear α's has an obvious resemblance to various well-established approximations for the static three-particle correlation function in terms of a superposition of pair-correlation functions. However, while the latter has a long tradition of physical interpretation, we are still in the dark as to the precise physical contents of this dynamical superposition scheme. Much more work will be necessary to develop the correspondence between this novel language and the physical model it represents.

The absence of the $\gamma\ln\gamma^{-1}$ dominant damping term in the $\gamma\to 0$ limit points to the likelihood of the incorrect treatment of the cumulative effect of distant collisions. This defect—which, incidentally, is more significant for small- than for high-γ values—is, however, more likely to be related to the original VAA hypothesis than to the dynamical superposition approximation.

We have made no attempt to actually solve the integral equation (58) combined with (16) for arbitrary γ values. Two obvious avenues for approaching the problem can be envisioned: iteration, and reduction to an algebraic equation by modeling $\alpha(\vec{p},\mu)$ and parametrizing it through adjustable constants. Further developments along these lines will be discussed in subsequent publications.

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APPENDIX A

In this Appendix, we show how the nonequilibrium two-point function can be expressed in terms of equilibrium three-point functions. Its calculation by ensemble averaging in the Eulerian picture,

$$\langle n_{\vec{k}-\vec{q}} n_{\vec{q}}\rangle(t) = \int d\Gamma\, \Omega(\Gamma,t) n_{\vec{k}-\vec{q}} n_{\vec{q}}, \tag{A1}$$

$$d\Gamma = d\vec{x}_1 d\vec{v}_1 \cdots d\vec{x}_N d\vec{v}_N ,$$

calls for solving the Liouville equation

$$\left(\frac{\partial}{\partial t} + i\mathcal{L}(\Gamma,t)\right)\Omega(\Gamma,t) = 0, \tag{A2}$$

$$\mathcal{L}(\Gamma,t) \equiv -i[H(\Gamma,t),\ldots], \tag{A3}$$

for the time evolution of the nonequilibrium distribution function $\Omega(\Gamma,t)$.

The unperturbed state of the system at $t=0$ is characterized by the macrocanonical distribution function

$$\Omega^{(0)}(\Gamma) = Q\exp[-\beta H^{(0)}(\Gamma)], \tag{A4}$$

where

$$Q^{-1} = \int d\Gamma \exp[-\beta H^{(0)}(\Gamma)],$$

and

$$H^{(0)}(\Gamma) = \sum_{i=1}^{N} \frac{p_i^2}{2m} + \frac{1}{2}\sum_{i\neq j} \frac{e^2}{|\vec{x}_i - \vec{x}_j|}$$

is the unperturbed Hamiltonian.

The introduction of a sufficiently weak external potential $\hat{\phi}$ into the system then perturbs $H^{(0)}$ and $\mathcal{L}^{(0)}$ by amounts

$$H^{(1)}(\Gamma,t) = -\sum_{i=1}^{N} e\hat{\phi}(\vec{x}_i,t)$$

$$= -\frac{1}{V}\sum_{\vec{k}} e\hat{\phi}(\vec{k},t) n_{-\vec{k}}, \tag{A5}$$

$$\mathcal{L}^{(1)}(\Gamma,t) = -i[H^{(1)}(\Gamma,t),\ldots]$$

$$= \frac{ie}{V}\sum_{\vec{k}} \hat{\phi}(\vec{k},t)[n_{-\vec{k}},\ldots]. \tag{A6}$$

The subsequent perturbation of (A2) results in the formal solution

$$\Omega(\Gamma,t) = \Omega^{(0)}(\Gamma) + \Omega^{(1)}(\Gamma,t) + \cdots, \tag{A7}$$

with

$$\Omega^{(1)}(\Gamma,t) = -i\int_0^\infty d\tau \exp(-i\tau\mathcal{L}^{(0)})\mathcal{L}^{(1)}(t-\tau)\Omega^{(0)}.$$

The explicit form of $\mathcal{L}^{(1)}$ exhibited in (A6) leads to $\Omega^{(1)}(\Gamma,t)$ and $\langle n_{\vec{k}-\vec{q}} n_{\vec{q}}\rangle^{(1)}(t)$ as follows:

$$\mathcal{L}^{(1)}(t-\tau)\Omega^{(0)} = \frac{ie}{V}\sum_{\vec{k}''} \hat{\phi}(\vec{k}'',t-\tau)(n_{-\vec{k}''},\Omega^{(0)}) = \frac{i\beta e\Omega^{(0)}}{V}\sum_{\vec{k}''}\hat{\phi}(\vec{k}'',t-\tau)[H^{(0)}, n_{-\vec{k}''}]$$

$$= -\frac{i\beta e\Omega^{(0)}}{V}\sum_{\vec{k}''} \hat{\vec{E}}(\vec{k}'',t-\tau)\cdot\vec{J}_{-\vec{k}''}; \tag{A8}$$

$$\Omega^{(1)}(\Gamma,t) = -\frac{\beta e\Omega^{(0)}}{V}\sum_{\vec{k}''}\int_0^\infty d\tau\, \hat{\vec{E}}(\vec{k}'',t-\tau)\cdot\exp(-i\tau\mathcal{L}^{(0)})\vec{J}_{-\vec{k}''}; \tag{A9}$$

$$\langle n_{\vec{k}-\vec{q}} n_{\vec{q}}\rangle^{(1)}(t) = -\frac{\beta e}{V}\sum_{\vec{k}''}\frac{1}{k''}\int_0^\infty d\tau\, \hat{\vec{E}}(\vec{k}'',t-\tau)\int d\Gamma\, \Omega^{(0)} n_{\vec{k}-\vec{q}} n_{\vec{q}}\, e^{-i\tau\mathcal{L}^{(0)}}\vec{k}''\cdot\vec{J}_{-\vec{k}''},$$

where $\vec{J}_{\vec{k}} = \sum_i \vec{v}_i \exp(-i\vec{k}\cdot\vec{x}_i)$. In the Eulerian picture the microscopic density and current operators have no explicit time dependence. The time dependence, however, can be generated by shifting the representation to the Lagrangian picture. Then, choosing $t=0$ as an arbitrary reference time and letting the time evolution operator act on $\vec{J}_{-\vec{k}''}$, one arrives at the result

$$\langle n_{\vec{k}-\vec{q}} n_{\vec{q}}\rangle^{(1)}(t) = -\frac{\beta e}{V}\sum_{\vec{k}''}\frac{1}{k''}\int_0^\infty d\tau\, \hat{E}(\vec{k}'',t-\tau)\langle n_{\vec{k}-\vec{q}}(0) n_{\vec{q}}(0)\vec{k}''\cdot\vec{J}_{-\vec{k}''}(-\tau)\rangle^{(0)}$$

$$= \frac{i\beta e}{Vk}\left(\hat{E}(\vec{k},t)\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(0)\rangle^{(0)} - \frac{\partial}{\partial t}\int_0^\infty d\tau\, \hat{E}(\vec{k},t-\tau)\langle n_{\vec{k}-\vec{q}}(0)n_{\vec{q}}(0)n_{-\vec{k}}(-\tau)\rangle^{(0)}\right). \tag{A10}$$

APPENDIX B

In this Appendix, we derive the useful triangle symmetry relations

$$\hat{a}(\vec{p},0;\vec{k}-\vec{p},\omega) = \hat{a}(\vec{p},0;-\vec{k},\omega), \tag{B1}$$

$$\hat{a}(\vec{p},\omega;\vec{k}-\vec{p},0) = \hat{a}(-\vec{k},\omega;\vec{k}-\vec{p},0), \tag{B2}$$

with the aid of the dynamical NLFDT equation (21). The salient point of the derivation is that we assert that $S(\vec{p},\mu;\vec{q},\nu)$ is bounded at $\mu=0$ and $\nu=0$.

A zero-frequency singularity would be indeed unusual.

Consider now Eq. (21) rewritten in the form

$$i\omega S(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)=(b^*-b)/\mu+(\tilde{b}^*-\tilde{b})/(\omega-\mu), \tag{B3}$$

where

$$b(\mu,\omega-\mu)=\hat{a}(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)-\hat{a}(\vec{p},-\mu;-\vec{k},\omega), \tag{B4}$$

$$\tilde{b}(\mu,\omega-\mu)=\hat{a}(\vec{p},\mu;\vec{k}-\vec{p},\omega-\mu)-\hat{a}(-\vec{k},\omega;\vec{k}-\vec{p},\mu-\omega). \tag{B5}$$

The assumed boundedness of $S(\vec{p},0;\vec{k}-\vec{p},\omega)$ requires that $b(0,\omega)=b^*(0,\omega)$. Thus there are two possibilities:

$$\text{(i)}\quad b(0,\omega)=0=b^*(0,\omega), \tag{B6}$$

implying condition (B1) above, or

$$\text{(ii)}\quad \mathrm{Im}\,b(0,\omega)=0, \tag{B7}$$

implying the weaker condition

$$\hat{a}''(\vec{p},0;\vec{k}-\vec{p},\omega)=\hat{a}''(p,0;-\vec{k},\omega). \tag{B8}$$

However, by virtue of the Kramers-Kronig relations that $\hat{a}(\vec{p},0;\vec{k}-\vec{p},\omega)$ and $\hat{a}(\vec{p},0;-\vec{k},\omega)$ satisfy, (B8) leads to the equality of the real parts as well, which then renders (B7) tantamount to (B6). Note that at the RPA level, one can rigorously demonstrate from Eq. (42) that

$$\hat{a}_0(\vec{p},0,\vec{k}-\vec{p},\omega)=\hat{a}_0(\vec{p},0;-\vec{k},\omega).$$

Similar arguments for the $\mu=\omega$ case result in symmetry relation (B2).

APPENDIX C

The following calculation shows how the current-density-current equilibrium correlation function reduces to the pair-correlation function.

$$\begin{aligned}
\langle \vec{J}_{\vec{p}}n_{\vec{k}-\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}\Big|_{\vec{p},\vec{k}-\vec{p},\vec{k}\neq 0} &= \sum_{i=1}^{N}\sum_{j=1}^{N}\sum_{l=1}^{N}\int d\Gamma\,\Omega^{(0)}\vec{v}_i\exp(-i\vec{p}\cdot\vec{x}_i)\exp[-i(\vec{k}-\vec{p})\cdot\vec{x}_j]\vec{v}_l\exp(i\vec{k}\cdot\vec{x}_l)\\
&=\sum_i\sum_j\sum_l\int d\Gamma\exp(-i\vec{p}\cdot\vec{x}_i)\exp[-i(\vec{k}-\vec{p})\cdot\vec{x}_j]\exp(i\vec{k}\cdot\vec{x}_l)\vec{v}_i\Omega^{(0)}\frac{\partial H^{(0)}}{\partial\vec{p}_l}\\
&=-\frac{1}{\beta}\sum_i\sum_j\sum_l\int d\Gamma\exp(-i\vec{p}\cdot\vec{x}_i)\exp[-i(\vec{k}-\vec{p})\cdot\vec{x}_j]\exp(i\vec{k}\cdot\vec{x}_l)\vec{v}_i\frac{\partial\Omega^{(0)}}{\partial\vec{p}_l}\\
&=\overleftrightarrow{1}\frac{1}{\beta m}\sum_i\sum_j\int d\Gamma\,\Omega^{(0)}\exp i(\vec{k}-\vec{p})\cdot(\vec{x}_i-\vec{x}_j)\\
&=\overleftrightarrow{1}\frac{1}{\beta m}\langle n_{k-p}(0)n_{p-k}(0)\rangle^{(0)}=\overleftrightarrow{1}(N/\beta m)[S(|\vec{k}-\vec{p}|)+N\delta_{\vec{k}-\vec{p}}],
\end{aligned}\tag{C1}$$

in which $\overleftrightarrow{1}$ is the unit tensor, and where we have made use of Hamilton's equation $\vec{v}_i=\partial H^{(0)}/\partial\vec{p}_i$ ($i=1,2,\ldots,N$) and the form (A4) of the macrocanonical distribution $\Omega^{(0)}$. Similarly,

$$\langle \vec{J}_{\vec{k}-\vec{p}}n_{\vec{p}}\vec{J}_{-\vec{k}}\rangle^{(0)}|_{\vec{p},\vec{k}-\vec{p},\vec{k}\neq 0}=\overleftrightarrow{1}(N/\beta m)[S(p)+N\delta_{\vec{p}}]. \tag{C2}$$

Note that in applying (C1) and (C2) to Eq. (38), the terms $\overleftrightarrow{1}(N^2/\beta m)\delta_{\vec{k}-\vec{p}}$ and $1(N^2/\beta m)\delta_{\vec{p}}$ should not be included since they are exactly compensated by the $-\overleftrightarrow{1}(N^2/\beta m)_{\vec{k}-\vec{p}}$ and $-\overleftrightarrow{1}(N^2/\beta m)\delta_{\vec{p}}$ contributions from the static positive background.

APPENDIX D

In this Appendix we evaluate first the μ and then the $\vec{p}$ integrals that appear in (59) and subsequent formulas that lead to (66). Analysis of the μ integral leads to integrals of the type

$$\begin{aligned}
I_1&=\int_{-\infty}^{\infty}dw\,Z''(w),\\
I_2(y)&=\int_{-\infty}^{\infty}dw\,Z''(w)Z(w+y),\\
I_3(y)&=\int_{-\infty}^{\infty}dw\,wZ''(w)Z(w+y),
\end{aligned}\tag{D1}$$

where $Z(w)$ is the plasma-dispersion function and Z'' is its imaginary part. The evaluation of I_1 is immediate, yielding

$$I_1 = \pi . \tag{D2}$$

Following Coste,[23] I_2 and I_3 can be evaluated by using the integral representation of Z and interchanging the order of integration.

$$I_2(y) = \int_{-\infty}^{\infty} dw\, \sqrt{\pi/2}\, e^{-w^2/2} \int_{-\infty}^{\infty} dt \frac{1}{\sqrt{2\pi}} \frac{e^{-t^2/2}}{t-w-y-io} \tag{D3}$$

where o is a positive infinitesimal quantity. With

$$w = \tfrac{1}{2}(z-s), \quad t = \tfrac{1}{2}(z+s), \tag{D4}$$

this becomes

$$I_2(y) = \int_{-\infty}^{\infty} dz\, e^{-z^2/4} \int_{-\infty}^{\infty} ds \frac{e^{-s^2/4}}{s-y-io} = (\pi/\sqrt{2})Z(y/\sqrt{2}). \tag{D5}$$

Similarly,

$$\begin{aligned} I_3(y) &= \int_{-\infty}^{\infty} dw\, w\, \sqrt{\pi}/2\, e^{-w^2/2} \int_{-\infty}^{\infty} dt \frac{1}{\sqrt{2\pi}} \frac{e^{-t^2/2}}{t-w-y-io} \\ &= \tfrac{1}{8}\int_{-\infty}^{\infty} dz\, z\, e^{-z^2/4} \int_{-\infty}^{\infty} ds \frac{e^{-s^2/4}}{s-y-io} - \tfrac{1}{8}\int_{-\infty}^{\infty} dz\, e^{-z^2/4}\int_{-\infty}^{\infty} ds\, e^{-s^2/4} - \tfrac{1}{8} y \int_{-\infty}^{\infty} dz\, e^{-z^2/4} \int_{-\infty}^{\infty} ds \frac{e^{-s^2/4}}{s-y-io} \\ &= -\tfrac{1}{2}\pi\left(1+\frac{y}{\sqrt{2}}Z\left(\frac{y}{\sqrt{2}}\right)\right). \end{aligned} \tag{D6}$$

$H(\vec{p},\omega)$ is now given by

$$H(\vec{p},\omega) = \frac{\kappa^4}{\pi p^4}\left[I_1 + \frac{\omega}{ap} I_2\left(\frac{\omega}{ap}\right) + I_3\left(\frac{\omega}{ap}\right)\right], \tag{D7}$$

which, by using (D2), (D5), and (D6) becomes

$$H(\vec{p},\omega) = \frac{\omega^4}{2p^4}\left[1 + \frac{1}{\sqrt{2}}\frac{\omega}{ap} Z\left(\frac{\omega}{\sqrt{2}\,ap}\right)\right], \tag{D8}$$

the result quoted in (64).

The p integration of the above leads to the integral

$$J(a) = \int_0^{\infty} du \frac{u}{(u^2+a^2)^2} Z(u) = \tfrac{1}{2}\int_0^{\infty} du \frac{1}{u^2+a^2}\frac{d}{du}Z(u). \tag{D9}$$

By referring to the differential equation

$$\frac{d}{du}Z(u) + uZ(u) + 1 = 0, \tag{D10}$$

and the alternative integral representation

$$Z'(u) = -e^{-u^2/2}\int_0^u dt\, e^{t^2/2} \tag{D11}$$

that the real part of $Z(u)$ satisfies, we transform (D9) into

$$J'(a) = \frac{\pi^{3/2}}{2^{5/2}} - \frac{1}{2a}\left(\frac{\pi}{2} + \frac{a^3}{\sqrt{2}}\int_0^{\infty}\frac{du}{u}\frac{e^{-u^2}}{u^2+\frac{1}{2}a^2}\int_0^u dt\, e^{t^2}\right). \tag{D12}$$

Baus[15] quotes the integration formula given by J. W. Turner:

$$\int_0^{\infty}\frac{du}{u}\frac{e^{-u^2}}{u^2+\eta^2}\int_0^u dt\, e^{t^2} = \frac{\pi^{3/2}}{4\eta^2}[1 - e^{\eta^2}(1-\operatorname{erf}\eta)]. \tag{D13}$$

Substitution of (D13) into (D12) gives the desired result for the real part of the integral:

$$J'(a) = -\frac{\pi}{4a} + \frac{\pi^{3/2}}{2^{5/2}} e^{a^2/2}\left[1 - \operatorname{erf}\left(\frac{a}{\sqrt{2}}\right)\right]. \tag{D14}$$

The imaginary part of $J(a)$,

$$J''(a) = \int_0^{\infty} du \frac{u}{(u^2+a^2)^2} Z''(u) = \left(\frac{\pi}{2}\right)^{1/2}\int_0^{\infty} du \frac{u}{(u^2+a^2)^2} e^{-u^2/2}, \tag{D15}$$

is integrable in terms of the modified exponential integral $E_1(\eta)$

$$E_1(\eta) = \int_{\eta}^{\infty}\frac{dt}{t} e^{-t} \tag{D16}$$

with the result

$$J''(a) = -\frac{\pi^{1/2}}{2^{3/2}}\frac{1}{a^2}\left[\frac{a^2}{2} e^{a^2/2} E_1\left(\frac{a^2}{2}\right) - 1\right]. \tag{D17}$$

APPENDIX E

In order to facilitate the comparison between our approximate results for $\gamma \to 0$ and the exact results derived from Coste's formalism,[23] we sketch in this Appendix the main points leading to the exact results exhibited in Table I. A detailed derivation will be given elsewhere. We also point out that, some erroneous statements notwithstanding,[28] this result is in perfect agreement with the requirements of the Kramers-Kronig relations.

Coste[23] obtained an exact expression to order γ for $\Delta\alpha(\vec{k},\omega)=\alpha(\vec{k},\omega)-\alpha_0(\vec{k},\omega)$ from the formal expansion of the first two equations of the BBGKY hierarchy. This result, further expanded for $k\to 0$ and valid to order k^2, is given in Eqs. (3)–(9) of his Paper II. With a little algebra this result can be rewritten in the notation of (16) and (59) of this paper with

$$\Delta\alpha(\vec{k},\omega)=\gamma\frac{k^2}{\kappa^2}\frac{\omega_0^4}{\omega^4}[\mathcal{V}_{\rm stat}+\mathcal{V}_{\rm dyn}(\omega)],$$

$$\mathcal{V}_{\rm dyn}(\omega)=\mathcal{V}_D(\omega)+\mathcal{V}_E(\omega),$$

$$\mathcal{V}_D(\omega)=\frac{46}{15\pi}\int_0^\infty dx\,x^2 \times\int_{-\infty}^{+\infty}d\mu\,\delta_-(\mu)\hat{\alpha}_0(\vec{p},\mu)\hat{\alpha}_0(\vec{p},\omega-\mu),$$

$$\mathcal{V}_E(\omega)=\frac{16}{15\pi}\frac{\omega_0^2}{\omega^2}\int dx\,x^2\left(x^2\int_{-\infty}^{+\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_0(\vec{p},\omega)\alpha_0(\vec{p},\omega-\mu)+\alpha_0(\vec{p},\omega)\hat{\alpha}_0(\vec{p},\omega-\mu)]-\frac{1}{1+x^2}\right). \quad \text{(E1)}$$

Approximating[15] $\hat{\alpha}(\vec{p},\omega)$ by $\alpha(\vec{p},\omega)/\epsilon(\vec{p},0)$, the above expression leads to

$$\mathcal{V}_D(\omega)=\frac{46}{15\pi}\int_{-\infty}^{+\infty}dx\frac{x^2}{(1+x^2)^2}H(\vec{p},\omega),$$

$$\mathcal{V}_E(\omega)=-\frac{16}{15\pi}\int_{-\infty}^{+\infty}dx\frac{x^2}{1+x^2}[1-2x^4H(\vec{p},\omega)], \quad \text{(E2)}$$

$$H(\vec{p},\omega)=\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{d\mu}{\mu}\alpha_0''(\vec{p},\omega)\alpha_0(\vec{p},\omega-\mu).$$

The integral in the expression for $\mathcal{V}_D(\omega)$ has been evaluated in Appendix D and in (61)–(66), with the result

$$\mathcal{V}_D(\omega)=\tfrac{23}{60}\{1-2\eta^2[1-\sqrt{\pi}\,\eta e^{\eta^2}(1-\mathrm{erf}\eta)]\} -i\tfrac{23}{30}\sqrt{\pi}\,\eta[\eta^2e^{\eta^2}E_1(\eta^2)-1],$$

$$\eta=\omega/2\omega_0, \quad \text{(E3)}$$

$$E_1(\eta)=\int_\eta^\infty\frac{dt}{t}e^{-t}.$$

Similar evaluation of $\mathcal{V}_E(\omega)$ leads to

$$\mathcal{V}_E(\omega)=-\tfrac{2}{15}\sqrt{\pi}\frac{1}{\eta}[1-e^{\eta^2}(1-\mathrm{erf}\eta)] +i\tfrac{2}{15}\sqrt{\pi}\frac{1}{\eta}[E_1(\gamma^2\eta^2)-e^{\eta^2}E_1(\eta^2)]. \quad \text{(E4)}$$

The appearance of the γ-dependent imaginary part is the result of the cutoff of a logarithmically divergent integral at $p_{\rm max}=\kappa/\gamma$. When evaluated at $\eta=\frac{1}{2}$, (E3) and (E4) lead to the results quoted in the second column of Table I. The imaginary part of $\mathcal{V}_E(\omega)$ contains a $\ln\gamma^{-1}$ which is obviously dominant as $\gamma\to 0$. When the Kramers-Kronig formula is applied and the Hilbert transform of this term is taken, it leads to a *negative* contribution in $\mathrm{Re}\Delta\alpha(\vec{k},\omega=\omega_0)$. From this fact it has erroneously been argued[28] that $\mathrm{Re}\Delta\alpha(\vec{k},\omega=\omega_0)$ must be *negative* as $\gamma\to 0$. This line of argument tacitly presupposes that the Kramers-Kronig relations carry the dominant term in the imaginary part of $\Delta\alpha(\vec{k},\omega)$ into a dominant term in the real part. However, it is well known that there is no such dominant term in $\mathrm{Re}\Delta\alpha$; indeed, all its contributions are of order γ as $\gamma\to 0$. The resolution of this apparent paradox lies in observing that the cutoff applied in the calculation on the integral in $\mathrm{Im}\Delta\alpha$ is ω dependent, which cannot be ignored; otherwise the applicability of the Kramers-Kronig formulas is violated. The proper procedure is to (i) use the integral representation of the divergent quantity, which then permits the exact evaluation of the Kramers-Kronig integral through the interchange of the order of the two integrations, and (ii) account for the plus-function character of the singular denominator $1/\omega^4$ originating from the conversion of $\mathcal{V}(\omega)$ into $\Delta\alpha(\omega)$. Done this way, the counterpart of the term containing $\gamma\ln\gamma^{-1}$ contribution in $\mathrm{Im}\Delta\alpha$ is of order γ in $\mathrm{Re}\Delta\alpha$, as it should be. Even though it is still negative, it now has to compete with other order-γ terms: one is the counterpart of the nondominant contribution, proportional to γ, in $\mathrm{Im}\Delta\alpha$; the other is the counterpart of a δ-function type singular contribution, related to the $1/\omega^4$ denominator. Both of these contributions are *positive*, neutralizing and overwhelming the negative piece due to the "dominant" term: the result is the net *positive* $\Delta\alpha(\vec{k},\omega=\omega_0)$ as indicated on Table I. This procedure is now exhibited in some detail below.

First we consider the Kramers-Kronig relations satisfied by $\mathcal{V}_D(\omega)$ and $\mathcal{V}_E(\omega)$.

$$\mathrm{Re}\mathcal{V}_{D,E}(\omega)=\frac{1}{\pi}\int_{-\infty}^{+\infty}d\mu\frac{\mathrm{Im}\mathcal{V}_{D,E}(\mu)}{\mu-\omega}. \quad \text{(E5)}$$

Using the integral representations of $\mathcal{V}_D(\omega)$ and $\mathcal{V}_E(\omega)$ as given by (E2) and interchanging the order of the x and μ integrations, we realize that (E5) is satisfied provided $H(\vec{p}, \omega)$ is a plus function. That this is indeed so follows from (D8), where the explicit expression for the $H(\vec{p}, \omega)$ is given: since $Z(\vec{p}, \omega)$ is a plus function, so is $H(\vec{p}, \omega)$ and thus (E5) is automatically satisfied. It remains to observe that the x integral in $\mathrm{Im}\mathcal{V}_E(\omega)$ is logarithmically divergent, while it is convergent in the expression for $\mathrm{Re}\mathcal{V}_E(\omega)$: the convergence factor is supplied by the μ integration.

As for the analytic properties of $\Delta\alpha(\vec{p}, \omega) \propto [\mathcal{V}_{\mathrm{stat}} + \mathcal{V}_{\mathrm{dyn}}(\omega)]/\omega^4$, one has to realize that the correct interpretation of the $1/\omega^4$ factor is

$$\lim_{o\to 0} 1/(\omega + io)^4 = 1/\omega^4 + \tfrac{1}{6} i\pi\delta'''(\omega).$$

Thus

$$\mathrm{Re}\Delta\alpha(\vec{k}, \omega) = \gamma \frac{k^2}{\kappa^2}\omega_0^4 \left(\frac{\mathcal{V}_{\mathrm{stat}} + \mathrm{Re}\mathcal{V}_{\mathrm{dyn}}(\omega)}{\omega^4} - \frac{\pi}{6}\delta'''(\omega)\,\mathrm{Im}\mathcal{V}_{\mathrm{dyn}}(\omega)\right), \quad \text{(E6)}$$

$$\mathrm{Im}\Delta\alpha(\vec{k}, \omega) = \gamma \frac{k^2}{\kappa^2}\omega_0^4 \left(\frac{\mathrm{Im}\mathcal{V}_{\mathrm{dyn}}(\omega)}{\omega^4} + \frac{\pi}{6}\delta'''(\omega) \times [\mathcal{V}_{\mathrm{stat}} + \mathrm{Re}\mathcal{V}_{\mathrm{dyn}}(\omega)]\right). \quad \text{(E7)}$$

Substituting (E7) in the Kramers-Kronig integral, one has

$$\int_{-\infty}^{+\infty} \frac{d\mu}{\pi} \frac{\mathrm{Im}\Delta\alpha(\vec{k},\mu)}{\mu-\omega} = \gamma\frac{k^2}{\kappa^2}\omega_0^4\left[\int_{-\infty}^{+\infty}\frac{d\mu}{\pi}\frac{\mathrm{Im}\mathcal{V}_{\mathrm{dyn}}(\mu)}{(\mu-\omega)\mu^4} - \frac{1}{6}\frac{d^3}{d\mu^3}\left(\frac{\mathrm{Re}\mathcal{V}_{\mathrm{dyn}}(\mu)}{\mu-\omega}\right)\Bigg|_{\mu=0^+} + \frac{\mathcal{V}_{\mathrm{stat}}}{\omega^4}\right] = \gamma\frac{k^2}{\kappa^2}\frac{\omega_0^4}{\omega^4}[\mathcal{V}_{\mathrm{stat}} + \mathrm{Re}\mathcal{V}_{\mathrm{dyn}}(\omega)]. \quad \text{(E8)}$$

The difference between this expression and Eq. (E6) for $\mathrm{Re}\Delta\alpha(\vec{k}\omega)$ is due to the singularity of $\mathrm{Im}\Delta\alpha(\vec{k}, \omega)$ at $\omega = 0$.

Finally it is instructive to display the three contributions to $\mathrm{Re}\Delta\alpha(\vec{k}, \omega=\omega_0)$ separately:

$$\mathcal{V}_{\mathrm{stat}} = 0.133, \quad \mathrm{Re}\mathcal{V}_D(\omega_0) = 0.296, \quad \mathrm{Re}\mathcal{V}_E(\omega_0) = -0.181. \quad \text{(E9)}$$

$\mathcal{V}_E(\omega_0)$ is indeed negative, but certainly not dominant. It is evident that replacing the full $\mathrm{Re}\mathcal{V}(\omega_0)$ by $\mathrm{Re}\mathcal{V}_E(\omega_0)$ yields, even qualitatively, completely incorrect results.

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