

## Electric microfield distributions in strongly coupled plasmas

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A new method is developed for calculating electric microfield distributions in a plasma. The method uses a coupling-parameter integration technique to express the Fourier transform of the microfield distribution in terms of a special pair-distribution function. An approximation of this function yielding the exact second moment of the microfield distribution gives results which agree well with computer simulations for strongly coupled one-component plasmas.

### I. INTRODUCTION

The spectral line shapes of atoms or ions radiating in a plasma contain a wealth of information and therefore provide valuable diagnostic tools.<sup>1</sup> These shapes are determined<sup>2</sup> by the interactions of the radiator with all the components of the plasma. In connection with this problem, Holtmark<sup>3</sup> and later Margenau<sup>4</sup> developed a statistical theory of collisional broadening. In their theory the radiator is immersed in a statistically fluctuating field produced by the configuration of the plasma during the time of emission; this is assumed short compared to times in which the configuration changes significantly. Thus the problem is reduced to determining the probability distribution of the perturbing electric fields.

Various approximate theories<sup>3-12</sup> have been proposed to evaluate the electric microfield distribution  $W(\vec{\epsilon})$ . However, none of these theories provides reliable numerical results for strongly coupled plasmas. In the present paper we propose a new scheme for calculating microfield distributions. The method is based on a formalism previously introduced by one of us<sup>13</sup> which expresses the Fourier transform of  $W(\vec{\epsilon})$  in terms of a special pair-distribution function. Here, we approximate the special function by a form containing a free parameter which is then fixed to give the exact second moment of  $W(\vec{\epsilon})$ . The numerical results obtained from this scheme agree well with computer simulations for strongly coupled one-component plasmas.

Our model system consists of  $N$  particles each of charge  $Ze$  moving in a uniform neutralizing background contained in a volume  $\Omega$ . In addition, for treating the problem of the electric field distribution at an ion, a zeroth particle of charge  $Z_0e$  is included. Here,  $Z_0$  and  $Z$  are positive integers and  $e$  the magnitude of the elementary charge. The potential energy of the total system is a sum of pairwise additive Coulomb interactions

$$V = \sum_{\substack{i=0 \\ i < j}}^N \sum_{j=0}^N v_{ij} + V_B, \quad (1.1)$$

where

$$v_{ij} = Z_i Z_j e^2 / r_{ij},$$

$$r_{ij} = |\vec{r}_i - \vec{r}_j|, \quad (1.2)$$

$$Z_i = \begin{cases} Z_0, & i = 0 \\ Z, & i \neq 0 \end{cases}$$

$\vec{r}_j$  is the position of the  $j$ th particle, and  $V_B$  is the contribution to the potential energy due to the background. The electric field acting on the zeroth particle is given by the superposition of single-particle Coulomb fields plus a contribution  $\vec{E}_B$  from the background,

$$\vec{E} = \sum_{j=1}^N \vec{\epsilon}(\vec{r}_{0j}) + \vec{E}_B, \quad (1.3)$$

$$\vec{\epsilon}(\vec{r}_{0j}) = \frac{Ze}{r_{0j}^2} \hat{r}_{0j},$$

where  $\hat{r}_{0j}$  is a unit vector in the direction  $\vec{r}_0 - \vec{r}_j$ . This system models what is usually referred to as the high-frequency component of the field in a real plasma.<sup>8</sup>

We now define the electric microfield distribution  $W(\vec{\epsilon})$  as the probability density of finding an electric field  $\vec{\epsilon}$  equal to  $\vec{E}$  at  $\vec{r}_0$ . Assuming that the total system is described by classical equilibrium statistical mechanics, we have, in the limit of a macroscopic (formally infinite) system,

$$\begin{aligned}
W(\vec{\epsilon}) &= \langle \delta(\vec{\epsilon} - \vec{E}) \rangle \\
&= \lim_{\substack{N, \Omega \rightarrow \infty \\ N/\Omega = \rho}} \int_{\Omega} \cdots \int_{\Omega} d\vec{r}_0 d\vec{r}_1 \cdots d\vec{r}_N \frac{e^{-\beta V}}{Q(N, \Omega, T)} \\
&\quad \times \delta(\vec{\epsilon} - \vec{E}), \tag{1.4}
\end{aligned}$$

where  $Q(N, \Omega, T)$  is the configurational partition function and  $\beta = (k_B T)^{-1}$ .

We assume here that this thermodynamic limit exists and that all the correlation functions are translation invariant and isotropic in the limit. For proofs (in some cases) and discussion we refer the interested reader to Refs. 14 and 15.

The remainder of the paper is organized as follows. In Sec. II, we derive exact expressions for the second and fourth moments of the microfield distribution. In Sec. III we review the formalism of Ref. 13 and introduce simplifying approximations. We present numerical results in Sec. IV followed by a brief conclusion in Sec. V.

## II. EXACT MOMENT RELATIONS

Knowledge of moment sum rules is often useful in developing approximation schemes for fluids and plasmas. Here, we derive exact expressions for the second and fourth moments of the microfield distribution. Although the result for the second moment is "known"<sup>16</sup> it has not been previously incorporated into the calculation of microfield distributions (at least to our knowledge).

The second moment may be written in the form

$$\langle \vec{E} \cdot \vec{E} \rangle = \langle \vec{\nabla}_0 V \cdot \vec{\nabla}_0 V \rangle / (Z_0 e)^2, \tag{2.1}$$

where  $\vec{\nabla}_0$  is the gradient with respect to  $\vec{r}_0$  and the average is over the canonical ensemble defined in Eq. (1.4). Noting that

$$e^{-\beta V} \vec{\nabla}_0 V = -\beta^{-1} \vec{\nabla}_0 e^{-\beta V}, \tag{2.2}$$

substituting Eq. (2.2) into (2.1), integrating by parts, and setting the surface terms equal to zero yields

$$\langle \vec{E} \cdot \vec{E} \rangle = (Z_0^2 e^2 \beta)^{-1} \langle \nabla_0^2 V \rangle. \tag{2.3}$$

We now take advantage of Poisson's equation and write

$$\langle (\vec{F} \cdot \vec{F})^2 \rangle = \frac{3}{\Gamma^2} \frac{Z}{Z_0} \left[ 5 + 12 \int_0^\infty dx \frac{g^0(x)}{x^4} + 18 \int_0^\infty \frac{dx}{x} \int_0^\infty \frac{dx'}{x'} \int_{-1}^1 d\mu g_3^0(x, x', \mu) P_2(\mu) \right]. \tag{2.9}$$

Here,  $\rho g^0(x)$  is the density of plasma particles at a distance  $x$  from the zeroth particle,  $g_3^0(x, x', \mu)$  is the triplet distribution function involving the zeroth particle and any two plasma particles located at distances  $x$  and  $x'$  from the zeroth particle with  $\cos(\vec{x}, \vec{x}') = \mu$ , and  $P_2(\mu)$  is the Legendre polynomial of order 2. For  $Z_0 = Z$ ,  $g^0$  and  $g_3^0$  are just the ordinary pair- and triplet-distribution functions for a one-component plasma.

$$\nabla_0^2 V = 4\pi Z_0 Z e^2 \left[ \rho - \sum_{j=1}^N \delta(\vec{r}_j - \vec{r}_0) \right], \tag{2.4}$$

where  $-Ze\rho = \vec{\nabla}_0 \cdot \vec{E}_B / 4\pi$  is the background charge density, which remains unchanged in the thermodynamic limit even though  $E_B = 0$  in that limit. We then obtain from Eq. (2.3)

$$\langle \vec{E} \cdot \vec{E} \rangle = \frac{4\pi\rho}{\beta} \frac{Z}{Z_0} \tag{2.5}$$

since the  $\delta$  function only contributes when the positions of the two particles coincide and this has zero probability. We remark that only in going from Eq. (2.3) to (2.5) do we use special properties of the Coulomb interaction. For systems with different pairwise interactions, e.g., screened Coulomb systems, the evaluation of the second moment of the force acting on a particle can be done directly from Eq. (2.3) if the pair-distribution function is known.

It is convenient to introduce the dimensionless quantities

$$\vec{F} = \frac{\vec{E}}{Ze/a^2}, \quad \vec{x} = \frac{\vec{r}}{a}, \tag{2.6}$$

where  $a$  is the interparticle spacing,

$$\frac{4}{3}\pi\rho a^3 = 1.$$

Equation (2.5) then becomes

$$\langle \vec{F} \cdot \vec{F} \rangle = \frac{3}{\Gamma} (Z/Z_0), \tag{2.7}$$

where  $\Gamma = \beta Z^2 e^2 / a$  is the plasma-coupling constant. Equation (2.7) is a simple and exact result which may be incorporated into the calculation of the microfield distribution.

For an isotropic system the only nonvanishing fourth moments are of the form  $\langle E_l^2 E_m^2 \rangle$  where  $E_l$  is the component of  $\vec{E}$  in the  $l$  direction. Also from isotropy we have the relations

$$\langle (\vec{E} \cdot \vec{E})^2 \rangle = 3\langle E_l^4 \rangle + 6\langle E_m^2 E_n^2 \rangle = 5\langle E_l^4 \rangle \tag{2.8}$$

since, as is easily seen using spherical coordinates,

$$\langle E_l^4 \rangle = 3\langle E_m^2 E_n^2 \rangle,$$

for any  $l, m$ , and  $n$  such that  $m \neq n$ . Therefore, all these moments can be obtained from  $\langle (\vec{E} \cdot \vec{E})^2 \rangle$  which, using the procedure outlined in the Appendix, is given by

## III. THEORY AND APPROXIMATIONS

It was first noted by Morita<sup>17</sup> that the virial expansion for the Fourier transform of the microfield distribution  $T(\vec{k})$  is formally similar to that of the excess chemical potential. Recently, this similarity was used to express  $T(\vec{k})$  in terms of a special pair-distribution function<sup>13</sup> involving the zeroth particle and one of the plasma particles.

Only the main results are quoted here.

The Fourier transform of  $W(\vec{\epsilon})$  is defined by

$$W(\vec{\epsilon}) = \int \frac{d\vec{k}}{(2\pi)^3} e^{-i\vec{k}\cdot\vec{\epsilon}} T(\vec{k}), \quad (3.1)$$

or equivalently,

$$T(\vec{k}) = \langle e^{i\vec{k}\cdot\vec{\epsilon}} \rangle. \quad (3.2)$$

Since the system is assumed isotropic we may write,<sup>6</sup> setting  $\epsilon = |\vec{\epsilon}|$ ,

$$P(\epsilon) = 4\pi\epsilon^2 W(\vec{\epsilon}) \\ = \frac{2\epsilon}{\pi} \int_0^\infty dk k \sin(k\epsilon) T(k). \quad (3.3)$$

We now use a coupling parameter integration technique which parallels a well-known procedure<sup>18</sup> that expresses the excess chemical potential in terms of the pair-distribution function. This gives

$$T(k) = \exp \left[ i\rho\hat{k} \cdot \int_0^k d\lambda \int d\vec{r} \vec{\epsilon}(\vec{r}) [g^\lambda(\vec{r}) - 1] \right], \quad (3.4)$$

where  $\hat{k}$  is a unit vector in the direction of  $\vec{k}$  and  $\rho g^\lambda(\vec{r})$  is the "density" of plasma particles at position  $\vec{r}_0 + \vec{r}$  in a system whose potential energy is modified to include an extra coupling between the zeroth particle and the plasma particles. That is,

$$V(\lambda) = V - \frac{i\lambda}{\beta} \hat{k} \cdot \vec{E}, \quad (3.5)$$

$$g^\lambda(\vec{r}_0) = \lim_{\substack{N, \Omega \rightarrow \infty \\ N/\Omega = \rho}} \Omega^2 \int_\Omega \cdots \int_\Omega d\vec{r}_2 \cdots d\vec{r}_N \frac{e^{-\beta V(\lambda)}}{Q^\lambda(N, \Omega, T)},$$

with

$$Q^\lambda(N, \Omega, T) = \int_\Omega \cdots \int_\Omega d\vec{r}_0 \cdots d\vec{r}_N e^{-\beta V(\lambda)}.$$

The central problem now is the evaluation of  $g^\lambda(\vec{r})$ . Unfortunately for  $\lambda \neq 0$  the potential is imaginary so that some of our intuitive ideas about correlation functions, and how to approximate them, may not be valid. Nevertheless, we proceed and try approximation schemes of the type commonly employed in fluid theory.

Many of these schemes are based on "thermodynamic perturbation theory."<sup>19</sup> The system with potential  $V(\lambda=0) = V$  is chosen as reference system and its structure is assumed known to a good approximation. The perturbation potential is then given by a sum of pairwise interactions between the zeroth particle and the plasma particles

$$V(\lambda) - V = -\frac{i\lambda}{\beta} \hat{k} \cdot \sum_{j=1}^N \vec{\epsilon}(\vec{r}_{0j}). \quad (3.6)$$

The exponential approximation,<sup>20</sup> which gives good results in many cases, is particularly easy to try here. It makes the ansatz,

$$g^\lambda(\vec{r}) \simeq g^0(\vec{r}) \exp[\mathcal{C}(\vec{r}; \lambda)] \quad (3.7)$$

with the "renormalized potential"  $\mathcal{C}(\vec{r}; \lambda)$  given as a sum

of "generalized chains."<sup>20</sup> Due to the form of the perturbation potential in Eq. (3.6) this sum contains only two terms which give

$$\mathcal{C}(\vec{r}; \lambda) = i\lambda \hat{k} \cdot \vec{\epsilon}^*(\vec{r}), \quad (3.8)$$

$$\vec{\epsilon}^*(\vec{r}_{01}) = \vec{\epsilon}(\vec{r}_{01}) + \rho \int d\vec{r}_2 \vec{\epsilon}(\vec{r}_{02}) [g(r_{21}) - 1].$$

Here  $g(r)$  is the radial distribution function for the bulk plasma so that  $\vec{\epsilon}^*(\vec{r})$  can be interpreted as an effective single-particle field.

The substitution of Eqs. (3.7) and (3.8) into Eq. (3.4) yields

$$T(k) \simeq \exp \left[ 4\pi\rho \int_0^\infty dr r^2 g^0(r) \frac{\epsilon(r)}{\epsilon^*(r)} [j_0(k\epsilon^*(r)) - 1] \right], \quad (3.9)$$

$$\epsilon^*(r) = \frac{Ze}{r^2} \left[ 1 + 4\pi\rho \int_0^r dr' r'^2 [g(r') - 1] \right], \quad (3.10)$$

where we have written

$$\vec{\epsilon}^*(\vec{r}) = \epsilon^*(r) \hat{r}, \quad (3.11)$$

$$\vec{\epsilon}(\vec{r}) = \epsilon(r) \hat{r},$$

$j_0$  is the spherical Bessel function of order zero, and the integrations over the coupling parameter  $\lambda$  and the angles have been done.

It is now possible to evaluate  $T(k)$  from Eqs. (3.9) and (3.10) in terms of the radial distribution functions of the reference system. Such a calculation leads to a microfield distribution which does not satisfy the second-moment sum rule, Eq. (2.7). This last point may be demonstrated by first noting that  $T(k)$  is the moment generating function in the sense that the coefficients of its Taylor expansion in  $k$  are simply related to the moments

$$T(k) = 1 - \frac{k^2}{6} \langle \epsilon^2 \rangle + \frac{k^4}{120} \langle \epsilon^4 \rangle + \cdots \quad (3.12)$$

Thus the second moment obtained from Eqs. (3.9) and (3.10) is

$$\langle \epsilon^2 \rangle = \rho \int d\vec{r} g^0(r) \vec{\epsilon}(\vec{r}) \cdot \vec{\epsilon}(\vec{r}) \\ + \rho^2 \int d\vec{r} d\vec{r}' g^0(r) g(|\vec{r} - \vec{r}'|) \vec{\epsilon}(\vec{r}) \cdot \vec{\epsilon}(\vec{r}'). \quad (3.13)$$

This corresponds to replacing the triplet correlation function  $g_3^0(\vec{r}_0, \vec{r}_1, \vec{r}_2)$  by  $g^0(r_{01})g(r_{12})$  and so (3.13) is not exact. In the next section we compare microfield distributions obtained from Eqs. (3.9) and (3.10) with computer simulations and the agreement is poor. This fact, in conjunction with other results discussed in Sec. IV, suggests that the second-moment condition provides an important constraint on approximation schemes for evaluating the microfield distribution.

In order to satisfy the second-moment sum rule, we make the *ad hoc* assumption that  $\epsilon^*(r)$  can be approximated by a modified Debye screened field,

$$\epsilon^*(r) \simeq \epsilon_a(r) \equiv \frac{Ze}{r^2} (1 + \alpha r) \exp(-\alpha r), \quad (3.14)$$

where  $\alpha$  is an inverse screening length to be determined from Eq. (2.7). Substitution of the ansatz (3.14) into Eq. (3.10) leads to

$$T(L) \simeq \exp \left[ 3 \int_0^\infty dx \frac{g^0(x)}{F_\alpha(x)} [j_0(LF_\alpha(x)) - 1] \right] \quad (3.15)$$

with  $\alpha$  given by

$$\int_0^\infty dx g^0(x) F_\alpha(x) = \frac{Z}{\Gamma Z_0} \quad (3.16)$$

Equations (3.15) and (3.16) are expressed in terms of the dimensionless quantities introduced in Eq. (2.6) and

$$L = \frac{Ze}{a^2} k, \quad F_\alpha = \frac{\epsilon_\alpha}{Ze/a^2} \quad (3.17)$$

#### IV. NUMERICAL RESULTS

The second-moment sum rule provides a constraint on approximate calculations of  $P(F)$ . However, the success of these calculations depends, in part, on the relative importance of the constraint. In Fig. 1 we plot the position of the peak of  $P(F)$  vs  $\Gamma$  obtained from molecular dynamics<sup>10</sup> (MD). This is compared to the position of the peak given by a Gaussian approximation to the microfield distribution with the exact variance. Clearly, as  $\Gamma$  increases the two curves rapidly approach each other. Therefore, it seems reasonable to require that approximations to  $P(F)$  take into account the second-moment sum rule. Unfortunately this alone is not sufficient, e.g., the Gaussian approximation decays much too fast at large fields. We also mention later an example of an *a priori* more reasonable approximation with exact second moment which nevertheless agrees poorly with MD simulations.

In Figs. 2–6 we present the results of various approximations for  $P(F)$  at several values of  $\Gamma$  for  $Z_0 = Z$ . The results are compared to MD or Monte Carlo<sup>21</sup> (MC) except at the value  $\Gamma = 0.213$  where the comparison is to Hooper's approximation.<sup>11</sup> The latter is generally believed<sup>2,11,12</sup> to give accurate results for  $\Gamma \lesssim 1$ . It is clear

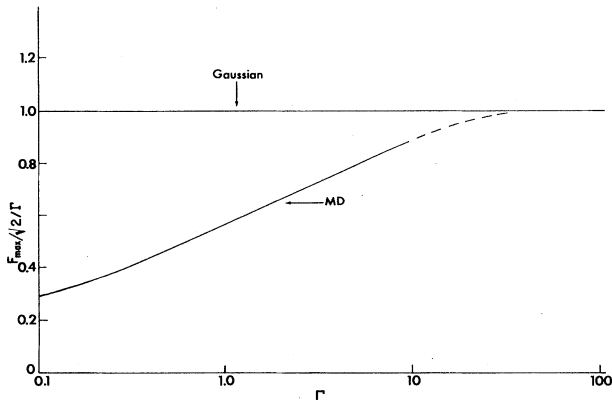


FIG. 1. Comparison of the peak position of  $P(F)$ , denoted  $F_{\max}$ , from MD and a Gaussian approximation to  $W(\vec{F})$  with exact variance.

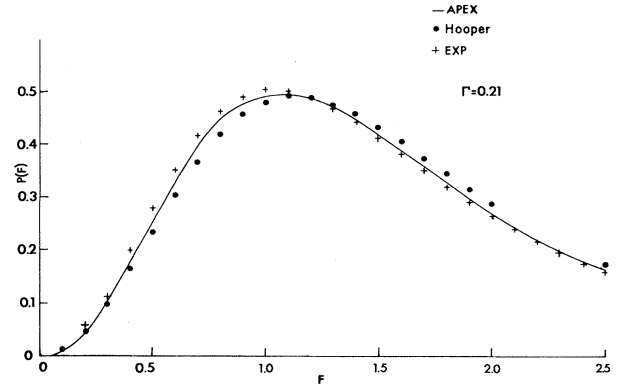


FIG. 2. Comparison of  $P(F)$  curves for  $Z_0 = Z$  at  $\Gamma = 0.21$  in units defined by Eq. (2.6).

from the figures that the  $P(F)$  obtained from Eqs. (3.15) and (3.16), hereafter called the “adjustable parameter exponential approximation” (APEX), are in good agreement with MD and MC. We do not include results for weakly coupled plasmas ( $\Gamma \ll 1$ ) since in this limit  $g^0(r)$  is given accurately by the Debye-Hückel theory and  $\alpha$  approaches the inverse Debye length so that APEX reduces to a simple approximation [see Eq. (3.1) of Ref. 13] which is in good agreement<sup>13</sup> with the Hooper results. In our calculations the radial distribution function  $g^0(r)$  is evaluated in the hypernetted chain (HNC) approximation<sup>22</sup> for  $\Gamma \leq 10$ . In addition, we used fits to the Monte Carlo  $g^0(r)$  data<sup>23</sup> for  $\Gamma = 5$  and 10 but the resulting  $P(F)$  were not significantly different from those using the HNC  $g^0(r)$ . Only the Monte Carlo  $g^0(r)$  was used for  $\Gamma = 100$ .

In Figs. 2 and 3 we have included the  $P(F)$  results from the exponential approximation (EXP) defined by Eqs. (3.7)–(3.10) and find poor agreement with MD. Although EXP is valid at  $\Gamma \ll 1$  (where it reduces to a Debye-Hückel theory) it does not satisfy the second-moment rule (see Sec. III). For example, at  $\Gamma = 1.03$  it underestimates the second moment  $\langle F^2 \rangle$  by 10%.

We also tried a parametrized Baranger and Mozer scheme (PBM) which neglects all but the first term in the virial expansion of  $T(k)$  and replaces the bare Coulomb

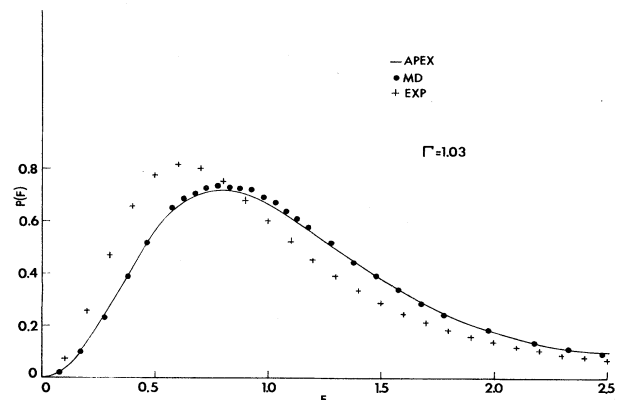
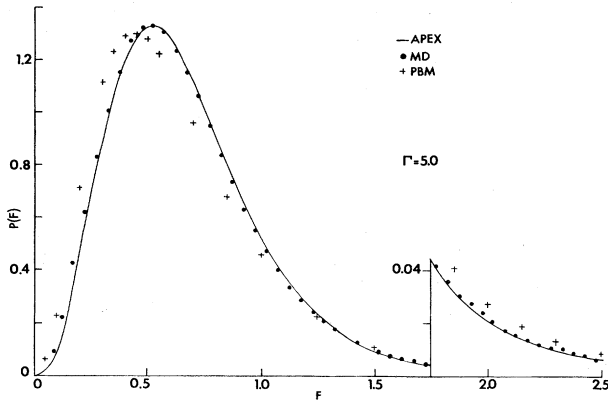


FIG. 3. Same as in Fig. 2 at  $\Gamma = 1.03$ .

FIG. 4. Same as in Fig. 2 at  $\Gamma = 5.0$ .

field by  $\vec{\epsilon}_\alpha$ . In this approximation

$$T(L) \simeq \exp \left[ 3 \int_0^\infty dx x^2 g^0(x) [j_0(LF_\alpha(x)) - 1] \right], \quad (4.1)$$

and  $\alpha$  is chosen to satisfy the second-moment rule

$$\int_0^\infty dx x^2 g^0(x) F_\alpha^2(x) = \frac{Z}{\Gamma Z_0}. \quad (4.2)$$

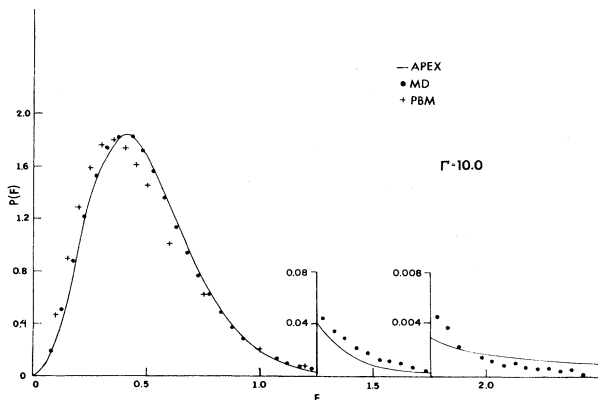
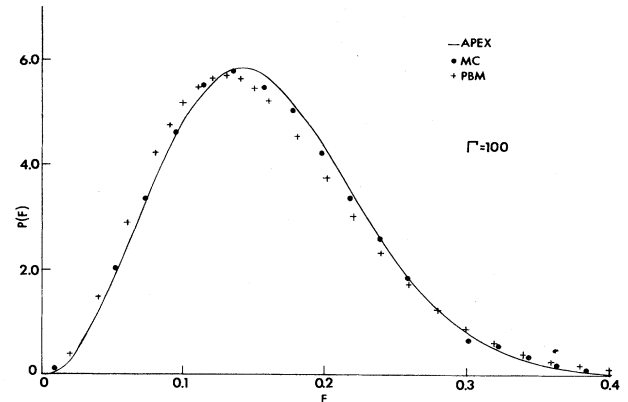
The resulting  $P(F)$  are plotted in Figs. 4–6 but the agreement with MD is not as good as APEX.

In an effort to further understand the relative success of APEX we compare Eqs. (3.17) and (4.1) revealing that APEX and PBM are quite similar. Both approximations treat the plasma as independent quasiparticles, each producing an effective field  $\vec{\epsilon}_\alpha$  at the zeroth particle. The difference between the approximations is in the distribution of quasiparticles about  $\vec{r}_0$ , which we denote by  $G(r)$ ,

$$G(r) = \begin{cases} g^0(r) \epsilon(r) \epsilon_\alpha^{-1}(r) \\ g^0(r) \end{cases} \quad (4.3)$$

for APEX and PBM, respectively.

A plausibility argument for selecting the APEX expression for  $G$  may be made as follows: Assume that an approximation of the plasma by independent quasiparticles with effective field  $\vec{\epsilon}_\alpha$  is useful for obtaining  $P(F)$ . It would then be reasonable to choose their density  $G(r)$  in

FIG. 5. Same as in Fig. 2 at  $\Gamma = 10.0$ .FIG. 6. Same as in Fig. 2 at  $\Gamma = 100$ .

such a way that the average field produced by quasiparticles at  $\vec{r}$  be the same as in the real plasma for all  $\vec{r}$ . This requires that  $\rho G(r) \vec{\epsilon}_\alpha(\vec{r}) = \rho g^0(r) \vec{\epsilon}(\vec{r})$  which is just the APEX result.

## V. CONCLUSION

We have developed a method for calculating electric microfield distributions which provides good agreement with computer simulations for strongly coupled one-component plasmas. The method is also reliable in the weakly coupled or Debye-Hückel regime. However, in the intermediate region  $0.1 \lesssim \Gamma \lesssim 1.0$ , there is some discrepancy between our results and those of Hooper.

Although the numerical results have been limited to one-component plasmas ( $Z_0 = Z$ ), it is possible to extend the calculation to cases where  $Z_0 \neq Z$ . Monte Carlo simulations are impractical when there is only one particle of a different charge but HNC calculations of  $g^0(r)$  are expected to be sufficiently accurate. Furthermore, the method can easily be extended to other systems where the second moment of  $W(\vec{\epsilon})$  is not known exactly but can be obtained from knowledge of  $g^0(r)$  which is in any case a necessary ingredient for the calculations.

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## APPENDIX

We outline the derivation of Eq. (2.9). It is easily shown with the help of Eq. (2.2) that

$$\langle \vec{\nabla}_0 V \cdot \vec{A} \rangle = \beta^{-1} \langle \vec{\nabla}_0 \cdot \vec{A} \rangle \quad (A1)$$

whenever  $|\vec{A}|$  decays sufficiently rapidly for its surface integral, coming from integrating by parts, to vanish. It then follows that

$$\begin{aligned}
(Z_0 e)^2 \beta \langle (\vec{E} \cdot \vec{E})^2 \rangle &= \langle \vec{\nabla}_0 \cdot [(\vec{E} \cdot \vec{E}) \vec{\nabla}_0 V] \rangle \\
&= \langle \vec{\nabla}_0 V \cdot \vec{\nabla}_0 (\vec{E} \cdot \vec{E}) \rangle + \langle (\vec{E} \cdot \vec{E}) \nabla_0^2 V \rangle \\
&= \beta^{-1} \langle \nabla_0^2 (\vec{E} \cdot \vec{E}) \rangle + (Z_0 e)^2 \beta \langle (\vec{E} \cdot \vec{E})^2 \rangle,
\end{aligned}
\tag{A2}$$

where the last line follows from Eq. (A1) and Poisson's equation, Eq. (2.4). The first term on the right-hand side of Eq. (A2) may be rewritten as

$$\langle \nabla_0^2 (\vec{E} \cdot \vec{E}) \rangle = 2 \sum_{l=1}^3 \sum_{m=1}^3 \langle E_{ml}^2 \rangle + 2 \langle \vec{E} \cdot \nabla_0^2 \vec{E} \rangle, \tag{A3}$$

where  $E_{ml}$  denotes the derivative of the component of  $\vec{E}$  in

$$\begin{aligned}
\sum_{l=1}^3 \sum_{m=1}^3 \langle E_{ml}^2 \rangle &= \frac{4\pi\rho Ze}{3} \left[ 4\pi\rho Ze + 2 \left\langle \sum_{j=1}^N \vec{\nabla}_0 \cdot \vec{e}(\vec{r}_{0j}) \right\rangle \right] + \sum_{l=1}^3 \sum_{m=1}^3 \left\langle \sum_{i=1}^N \sum_{j=1}^N \epsilon_{mi}(\vec{r}_{0i}) \epsilon_{ml}(\vec{r}_{0j}) \right\rangle \\
&= (\beta Z_0 e \langle \vec{E} \cdot \vec{E} \rangle)^2 + 6\rho(Ze)^2 \int d\vec{r} g^0(r)/r^6 + 6(Ze\rho)^2 \int d\vec{r} d\vec{r}' g_3^0(r, r', \mu) P_2(\mu)/(rr')^3,
\end{aligned}
\tag{A4}$$

where  $g^0(r)$ ,  $g_3^0(r, r', \mu)$ , and  $P_2(\mu)$  are defined at the end of Sec. II. Finally, combining Eqs. (A2)–(A4), performing the trivial angular integrations, and expressing the results in dimensionless variables yields Eq. (2.9).

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<sup>6</sup>A. A. Broyles, *Phys. Rev.* **100**, 1181 (1955).

<sup>7</sup>G. Ecker, *Z. Phys.* **148**, 593 (1957).

<sup>8</sup>M. Baranger and B. Mozer, *Phys. Rev.* **115**, 521 (1959).

<sup>9</sup>B. Held and C. Deutsch, *Phys. Rev. A* **24**, 540 (1981).

<sup>10</sup>J. C. Weisheit and E. L. Pollock, in *Spectral Line Shapes*, edited by B. Wende (deGruyter, Berlin, 1981).

<sup>11</sup>C. F. Hooper, Jr., *Phys. Rev.* **149**, 177 (1966).

<sup>12</sup>C. A. Iglesias and C. F. Hooper, Jr., *Phys. Rev. A* **25**, 1049 (1982).

<sup>13</sup>C. A. Iglesias, *Phys. Rev. A* **27**, 2705 (1983).

the  $m$  direction with respect to the  $l$  component of  $\vec{r}_0$ . By using the vector identity

$$\nabla_0^2 \vec{E} = \vec{\nabla}_0 (\vec{\nabla}_0 \cdot \vec{E}) - \vec{\nabla}_0 \times (\vec{\nabla}_0 \times \vec{E})$$

together with

$$\vec{\nabla}_0 \times \vec{E} = 0$$

and Poisson's equation, it is clear that the last term in Eq. (A3) vanishes.

Further progress is made by doing the required differentiations explicitly being careful to include the background contributions [see the remark immediately following Eq. (2.4)]:

<sup>14</sup>*Rigorous Atomic and Molecular Physics*, edited by G. Velo and A. S. Wightman, (Plenum, New York, 1981); J. Fröhlich and T. Spencer, p. 327; D. C. Brydges and P. Federbush, p. 371; M. Aizenman, p. 441; J. L. Lebowitz, p. 467.

<sup>15</sup>B. Jancovici, *Phys. Rev. Lett.* **46**, 386 (1981); L. Blum, Ch. Gruber, J. L. Lebowitz, and Ph. A. Martin, *ibid.* **48**, 1769 (1982).

<sup>16</sup>B. Jancovici (private communication).

<sup>17</sup>T. Morita, *Prog. Theor. Phys.* **23**, 1211 (1960).

<sup>18</sup>D. A. McQuarrie, *Statistical Mechanics* (Harper and Row, New York, 1976).

<sup>19</sup>R. W. Zwanzig, *J. Chem. Phys.* **22**, 1420 (1954).

<sup>20</sup>H. C. Andersen and D. Chandler, *J. Chem. Phys.* **57**, 1918 (1972).

<sup>21</sup>H. DeWitt (private communication).

<sup>22</sup>K. C. Ng, *J. Chem. Phys.* **61**, 2680 (1974).

<sup>23</sup>C. Deutsch, Y. Furutani, and M. M. Gombert, *Phys. Rep.* **69**, 87 (1981), and references therein.