Electric field distributions in strongly coupled plasmas

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(Received 26 October 1984)

Corrections to the adjustable-parameter exponential (APEX) model recently proposed by Iglesias, Lebowitz, and MacGowan for electric field distributions in a strongly coupled plasma are defined and discussed. The results for a neutral point are compared with those from Monte Carlo calculations for two values of the plasma parameter, and good agreement is obtained.

I. INTRODUCTION

The probability density for electric field values at a neutral atom or charged ion in a plasma is an important theoretical tool for the description of many spectroscopic experiments.[1] For small values of the plasma parameter (average Coulomb energy/average kinetic energy) there are several accurate methods to calculate such electric microfield distributions for both classical[2] and quantum[3] plasmas. The diagnosis of inertial confinement plasmas currently produced in many laboratories requires accurate microfield distributions for strongly coupled plasmas where the above theories are inapplicable. Recently, Iglesias, Lebowitz, and MacGowan[4] have proposed a phenomenological, but highly successful method to calculate quite simply the microfield distribution function in strongly coupled plasmas. This method is known as APEX (adjustable-parameter exponential approximation) and is essentially an effective independent-particle model. Comparisons of APEX calculations with Monte Carlo and molecular-dynamics results for the electric fields at the highly charged points relevant for laser-produced plasmas are in excellent agreement even for very large values of the plasma parameter. The results of APEX have not been tested as critically for neutral points where, for reasons described below, it may be expected to be less accurate. It is of some interest therefore to understand better why APEX works so well and to provide a means for calculating corrections to it when required. One interesting discussion along these lines has been given by Alastuey *et al.*,[5] but their results do not strictly reproduce APEX and are furthermore restricted to charged points. The objective here is to imbed the key ideas of APEX in the standard Baranger-Mozer cluster representation for microfield distributions.[2] In this way some contact between APEX and standard small plasma-parameter theories is established, while also providing APEX as the leading term in a series from which corrections can be calculated.

In the next section the Baranger-Mozer formulation is briefly reviewed and the structure of the resulting cluster series is discussed qualitatively. Next, the basic assumptions of APEX are given and motivated. Finally, a formal relationship of APEX to the Baranger-Mozer series is established and the first two terms of a renormalized series are given explicitly. To test APEX most critically, the correction terms are calculated at a neutral point for two values of the plasma parameter and compared with results from Monte Carlo calculations. For simplicity, a one-component classical plasma is assumed throughout.

II. BARANGER-MOZER FORMULATION

The probability density $Q(\boldsymbol{\epsilon})$ for the electric field at a neutral or charged point is most easily described in terms of its generating function

$$Q(\boldsymbol{\epsilon})\equiv\int\frac{d\boldsymbol{\lambda}}{(2\pi)^3}e^{-i\boldsymbol{\lambda}\cdot\boldsymbol{\epsilon}}F(\boldsymbol{\lambda})\;,\qquad F(\boldsymbol{\lambda})\equiv\langle e^{i\boldsymbol{\lambda}\cdot\mathbf{E}}\rangle\;, \tag{2.1}$$

where $\mathbf{E}$ is the field at a test charge arising from N positive charges in a uniform neutralizing background and the angular brackets $\langle\;\rangle$ denote an equilibrium ensemble average. In the limit of a formally infinite system the average becomes translationally invariant, and the location of the test charge may be taken as the origin without loss of generality. The electric field then has the form

$$\mathbf{E}=\sum_{i=1}^{N}\mathbf{E}(i)\;, \tag{2.2}$$

where $\mathbf{E}(i)$ is the Coulomb field due to the ith charge.

The Baranger-Mozer formulation results from two transformations of Eq. (2.1). The first is motivated by the fact that the required average is the product of single-particle functions, $\exp[i\boldsymbol{\lambda}\cdot\mathbf{E}(i)]$, which have a value close to one over most of the volume of the system. This suggests a first transformation to the set of single-particle functions

$$\phi(i)\equiv e^{i\boldsymbol{\lambda}\cdot\mathbf{E}(i)}-1 \tag{2.3}$$

which have the more desirable property of being zero over most of the volume. [The spirit of this transformation to the functions $\phi(i)$ is similar to the use of Mayer's f functions for thermodynamic properties of gases.] Substitution of (2.3) in (2.1) then leads directly to the series

$$F(\lambda)=\sum_{p=1}^{\infty}\left[\frac{1}{p!}\int d\mathbf{r}_1\cdots d\mathbf{r}_p f_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})\prod_{i=1}^{p}\phi(i)\right], \tag{2.4}$$

where the $f_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})$ are the usual equilibrium correlation functions representing the probability density for p charges at $\mathbf{r}_1,\ldots,\mathbf{r}_p$, and the test charge at the origin. The range of integration for each term in the series (2.4) is now limited by the functions $\phi(i)$. However, this restriction is not uniform with respect to λ, and particularly for large values of λ the functions $\phi(i)$ can differ from zero over a correspondingly large volume. Consequently, a second transformation is desirable,

$$F(\lambda)\equiv\exp G(\phi)\ , \tag{2.5}$$

where by a standard theorem of equilibrium statistical mechanics,[6] $G(\phi)$ is determined from (2.4) as

$$G(\phi)=\sum_{p=1}^{\infty}\frac{1}{p!}\int d\mathbf{r}_1\cdots d\mathbf{r}_p h_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})\prod_{i=1}^{p}\phi(i)\ . \tag{2.6}$$

Here $h_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})$ are the Ursell cluster functions associated with the set of correlation functions $f_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})$. For example,

$$\begin{aligned}h_1(\mathbf{r}_1\mid\mathbf{0})&=f_1(\mathbf{r}_1\mid\mathbf{0})\ ,\\ h_2(\mathbf{r}_1,\mathbf{r}_2\mid 0)&=f_2(\mathbf{r}_1,\mathbf{r}_2\mid 0)-f_1(\mathbf{r}_1\mid\mathbf{0})f_1(\mathbf{r}_2\mid\mathbf{0})\ .\end{aligned} \tag{2.7}$$

The significant difference between the series (2.6) and that of (2.4) is that the cluster functions h_p vanish when any members of the p particles are sufficiently far apart, whereas the correlation functions do not have this property. Consequently, the range of the integrals in (2.6) is controlled by *both* the functions $\phi(i)$ and the Ursell functions $h_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})$. The latter restricts the integration to volumes characterized by the correlation length which depends on the thermodynamic-state condition but is independent of λ. Qualitatively, therefore, the Baranger-Mozer formalism provides a series representation whose terms are controlled by the range of $\phi(i)$ for small λ and by the range of the Ursell functions for large λ. For weakly coupled plasmas, the Ursell functions of order $p+1$ are typically of the order of the plasma parameter to the power p, and the series (2.6) may be truncated at first or second order to good approximation.[7]

In spite of its success for weakly coupled plasmas, there are some notable limitations of this formalism to be expected for applications to the strongly coupled case. For practical purposes it is too difficult to calculate correlation functions of higher order than h_2. Consequently, the Baranger-Mozer representation must be sufficiently rapidly convergent for truncation at the first two terms even for strongly coupled plasmas. A related problem is the occurrence of the "bare" single-particle fields $\mathbf{E}(i)$ in the functions $\phi(i)$. All other interactions (e.g., interparticle potentials) have been removed in favor of the correlation functions that incorporate many-body effects. For a strongly coupled system it is expected that a proper formulation would eliminate the original fields $\mathbf{E}(i)$ in favor of some more representative quantity involving the correlation effects of surrounding particles. The formal procedure for carrying out such a "renormalization" has been developed[8] and presumably would be required here for strongly coupled plasmas.

III. APEX INDEPENDENT-PARTICLE MODEL

If all interactions between the plasma particles, except those with the test charge, are neglected, then the Ursell functions h_p vanish for $p\neq 1$. The Baranger-Mozer series (2.6) then reduces to only the leading term,

$$\begin{aligned}G^{(0)}(\phi)&=\int d\mathbf{r}_1 h_1^{(0)}(\mathbf{r}_1\mid\mathbf{0})\phi(1)\\ &=\int d\mathbf{r}_1 f_1^{(0)}(\mathbf{r}_1\mid\mathbf{0})\phi(1)\ .\end{aligned} \tag{3.1}$$

The superscript (0) denotes the corresponding quantity without interactions among plasma particles. The APEX model of Iglesias *et al.*[4] retains the independent-particle form of (3.1),

$$G(\phi)\rightarrow\int d\mathbf{r}_1 f_1^*(\mathbf{r}_1\mid\mathbf{0})\phi^*(1)\ , \tag{3.2}$$

with the assumption that the important effects of correlations can be accounted for by an effective pair distribution function $f_1^*(\mathbf{r}_1\mid\mathbf{0})$ and a screened field $\mathbf{E}^*(i)$ replacing $\mathbf{E}(i)$ in $\phi(i)$,

$$\phi^*(i)\equiv(e^{i\boldsymbol{\lambda}\cdot\mathbf{E}^*(i)}-1)\ . \tag{3.3}$$

Two constraints are imposed to determine f_1^* and $\mathbf{E}^*(i)$. The first is a requirement that the "quasiparticle" field due to the effective charge density at $\mathbf{r}_1$ is equal to the corresponding exact field

$$f_1^*(\mathbf{r}_1\mid\mathbf{0})\mathbf{E}^*(\mathbf{r}_1)=f_1(\mathbf{r}_1\mid\mathbf{0})\mathbf{E}(\mathbf{r}_1)\ . \tag{3.4}$$

The second requirement is that the APEX microfield distribution yield the exact second moment

$$\int d\epsilon\, Q_{\mathrm{APEX}}(\epsilon)\epsilon^2=\langle\mathbf{E}^2\rangle\ , \tag{3.5}$$

where the right side of (3.5) is known as a simple function of the plasma parameter.[4] More specifically the effective single-particle field $\mathbf{E}^*(i)$ is arbitrarily (but physically) chosen to have the Debye form

$$\begin{aligned}\mathbf{E}^*(i)&=\mathbf{E}(i)(1+\alpha r)e^{-\alpha r}\\ &\equiv\mathbf{E}(i)/R(r)\end{aligned} \tag{3.6}$$

and the parameter α is adjusted to fit the second-moment equation (3.5). These conditions then define the APEX model as

$$[G(\phi)]_{\mathrm{APEX}}\equiv\int d\mathbf{r}_1 f_1(\mathbf{r}_1\mid\mathbf{0})R(r_1)\phi^*(1)\ . \tag{3.7}$$

In practice $f_1(\mathbf{r}_1\mid\mathbf{0})$ is calculated from the hypernetted-chain integral equation.[9]

The APEX model is plausible since it incorporates the key effects for large and small field values ϵ (small and large λ values, respectively). The large fields are predominantly due to configurations with a single-plasma particle near the test charge. In this case correlations are less important and the independent-particle model is increasingly accurate. Conversely, small fields are due to the additive effects of many particles at large distances; the fields due to distant particles are asymptotically Gaussian distributed and characterized by the second moment $\langle \mathbf{E}^2 \rangle$ which is exactly included in APEX by condition (3.5). The main uncertainty, therefore, is how well APEX represents configurations of particles intermediate between the strong-field nearest neighbor and large-distance weak fields. However, this uncertain region diminishes for increasing test charge since the effect of the test charge is to exclude plasma particles from the region around it. The excluded region increases for large test charge so that only the nearest-neighbor and weak-field configurations are ultimately relevant. Conversely, APEX is expected to be least reliable for the neutral-point distribution. The condition (3.5) is ill-defined for neutral points since $\langle \mathbf{E}^2 \rangle$ diverges in this case; however, the parameter α in (3.6) is still well defined from (3.5) in the limit of vanishing test charge.[5]

IV. RENORMALIZED CLUSTER SERIES

The comments at the end of Sec. II and the success of APEX indicate that the Baranger-Mozer series could be improved if the single-particle field in the functions $\phi(i)$ is replaced by a screened field representing the effects of correlations in strongly coupled plasmas. In this spirit, a new functional series is obtained in terms of the renormalized functions ϕ^* of (3.3) by the definition

$$G^*(\phi^*) \equiv G(\phi) \tag{4.1}$$

which is readily obtained from the Baranger-Mozer series (2.6) and the functional relationship of $\phi(i)$ to $\phi^*(i)$,

$$\phi = -1 + (1+\phi^*)^R . \tag{4.2}$$

This last form is an identity that follows from the definitions (2.3), (3.3), and (3.6). Elimination of ϕ on the right side of (4.1) using (4.2) gives the desired renormalized cluster series,

$$G^*(\lambda \mid \phi^*) = \sum_{p=1}^{\infty} \left[\frac{1}{p!} \int d\mathbf{r}_1 \cdots d\mathbf{r}_p \, h_p^*(\mathbf{r}_1, \ldots, \mathbf{r}_p \mid \mathbf{0}) \times \prod_{i=1}^{p} \phi^*(i) \right] . \tag{4.3}$$

The function $h_p^*(\mathbf{r}_1, \ldots, \mathbf{r}_p \mid \mathbf{0})$ is recognized as the pth-order functional derivative of $G(\phi)$,

$$h_p^*(\mathbf{r}_1, \ldots, \mathbf{r}_p \mid \mathbf{0}) = \left. \frac{\delta^p G(\phi)}{\delta\phi^*(1) \cdots \delta\phi^*(p)} \right|_{\phi=0} . \tag{4.4}$$

More explicitly the first two terms in (4.3) are found to be

$$\begin{aligned} G^*(\lambda \mid \phi^*) = & \int d\mathbf{r}_1 \, h_1(\mathbf{r}_1 \mid \mathbf{0}) R(r_1) \phi^*(1) \\ & + \tfrac{1}{2} \int d\mathbf{r}_1 d\mathbf{r}_2 \{ h_2(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{0}) R(r_1) R(r_2) \\ & - \delta(\mathbf{r}_1 - \mathbf{r}_2) h_1(\mathbf{r}_1 \mid \mathbf{0}) R(r_1)[1 - R(r_1)] \} \\ & \times \phi^*(1) \phi^*(2) + \cdots . \end{aligned} \tag{4.5}$$

The first term of (4.5) is seen to be precisely APEX. The factor $R(r_1)$ occurs automatically here from the renormalization and eliminates the somewhat *ad hoc* assumption of APEX (3.4). It is also interesting to reinterpret the second-moment constraint of APEX (3.5), which is used to determine the parameter α in the screened field. In the present context it might appear more reasonable to choose α to improve convergence of the series (4.3). A similar procedure is used in thermodynamic perturbation theory where the corresponding parameters of the leading term are chosen to make the next order terms vanish.[10] This is not quite possible here without making α a function of λ. However, it is possible to choose α independent of λ such that all of the corrections to APEX vanish to order λ^2. That is, for $p \geq 2$,

$$\left[\frac{\partial^2}{\partial \lambda^2} \int d\mathbf{r}_1 \cdots d\mathbf{r}_p \, h_p^*(\mathbf{r}_1, \ldots, \mathbf{r}_p \mid \mathbf{0}) \phi^*(1) \cdots \phi^*(p) \right]_{\lambda=0} = 0 . \tag{4.6}$$

It is straightforward to verify that condition (4.6) is exactly the same as the second-moment condition (3.5). The latter may be understood, therefore, as a maximization of the independent-particle contribution relative to the renormalized Baranger-Mozer series.

V. NEUTRAL-POINT CALCULATIONS

The discussion at the end of Sec. III suggests that the corrections to APEX in Eq. (4.5) should be most significant for the microfield distribution at a neutral point. In this case the order of all correlation functions decreases by 1, since the test charge is not present, and (4.5) simplifies to

$$\begin{aligned} G^*(\phi^*) = & \, n \int d\mathbf{r}_1 R(r_1) \phi^*(1) \\ & + \tfrac{1}{2} \int d\mathbf{r}_1 d\mathbf{r}_2 \{ n^2 h(\mathbf{r}_1 - \mathbf{r}_2) R(r_1) R(r_2) \\ & - n \delta(\mathbf{r}_1 - \mathbf{r}_2) R(r_1)[1 - R(r_1)] \} \\ & \times \phi^*(1) \phi^*(2) + \cdots , \end{aligned} \tag{5.1}$$

where $h(r) \equiv g(r) - 1$, $g(r)$ is the radial distribution function,[9] and n is the average density. The angular parts of the integrals of the second term can be performed with a spherical harmonic expansion, just as in the usual Baranger-Mozer calculations, with the result that the first two terms of (5.1) are

$$\begin{aligned} & G^*(\phi^*) \to [G(\phi^*)]_{\mathrm{APEX}} + \Delta G^*(\phi^*) , \\ & \Delta G^*(\phi^*) = 2\pi n \int_0^\infty dr \, r^2 R(r)[R(r) - 1] \\ & \qquad \times \{ [j_0(2\lambda E^*) - 1] - 2[j_0(\lambda E^*) - 1] \} \\ & \qquad + \tfrac{1}{2} n \int_0^\infty dk [1 - S(k)] J(k) . \end{aligned} \tag{5.2}$$

In the last term $S(k)$ is the static structure factor for the one-component plasma[9] (OCP) and $J(k)$ is defined by

$$J(k) \equiv 8 \sum_{l=0}^{\infty} (-1)^l (2l+1)[k J_l(k)]^2 , \tag{5.3}$$

$$J_l(k) \equiv \int_0^\infty dr \, r^2 R(r) j_l(kr)[j_l(\lambda E^*) - \delta_{l,0}] . \tag{5.4}$$

TABLE I. Values of $\Delta G/G_{\mathrm{APEX}}$ for several values of λ at both Γ values.

	$\Delta G/G_{\mathrm{APEX}}$	
λ	$\Gamma=3.9$	$\Gamma=10$
0.02	−0.011	−0.011
0.10	−0.039	−0.044
0.20	−0.061	−0.074
0.50	−0.094	−0.12
1.0	−0.11	−0.15
2.0	−0.10	−0.15
5.0	−0.062	−0.090

Also, j_l is the spherical Bessel function of order l. In practice it appears sufficient to terminate the sum in Eq. (5.3) at $l=3$ and to evaluate $S(k)$ from the hypernetted-chain (HNC) approximation.[9] Finally, the parameter α is found from Eq. (4.6) to be[5]

$$\alpha=-\frac{2\beta U_{\mathrm{exc}}(\Gamma)}{a\Gamma}, \tag{5.5}$$

where $a=(4\pi n/3)^{-1/3}$, $\Gamma=\beta e^2/a$, β^{-1} is the temperature and $U_{\mathrm{exc}}(\Gamma)$ is the excess internal energy of the OCP. The latter may be determined either from the HNC equation or from a numerical fit due to DeWitt.[11]

Equations (5.1)—(5.5) have been evaluated for $\Gamma=3.9$ and 10, and Table I displays values of $\Delta G/G_{\mathrm{APEX}}$ for several values of λ at both Γ values. For the $\Gamma=3.9$ case, the relative correction to G_{APEX} obtains a maximum value of about 10% near $\lambda=1$. Similarly, for $\Gamma=10$ the correction at $\lambda=1$ is about 15%. Since the correction is opposite in sign to G_{APEX}, the microfield predicted from the corrected APEX theory (APXC) should have a higher peak value and a smaller width. This expectation is borne out in Figs. 1 and 2 which show the microfields for the two cases (ϵ is measured in units of $\epsilon_0 \equiv e^2/a$). In both figures, the microfield predicted by the first two terms of the Baranger-Mozer series is also shown. It is seen that the Baranger-Mozer curves differ substantially from the APEX and APXC results.

Both figures also show the results calculated from Monte Carlo simulations on a system consisting of 50 particles of charge 1 and 50 particles of charge 10^{-4}. The dark circles indicate the distribution of fields felt at the "uncharged" points. This distribution seems to be in good agreement with that predicted by APXC, indicating that APEX does not completely account for perturber-perturber correlations, and, therefore, overestimates the probability of large fields.

VI. DISCUSSION

The microfields predicted by APEX for the case of highly charged radiators, relevant to laser-fusion plasmas, are very accurate[4] and its corrections are quite small. The case of a neutral radiator considered here is of much less practical interest, but it does help clarify the region of validity of APEX, and it gives some insight into why it is so successful.

In particular, the plausibility arguments, given at the end of Sec. III, for APEX as a model interpolating between the proper small and large λ limits appear to be borne out in Table I. The most significant corrections occur in the region near $\lambda=1$, suggesting a failure to describe properly electric fields due to configurations of particles intermediate between the nearest neighbor, and

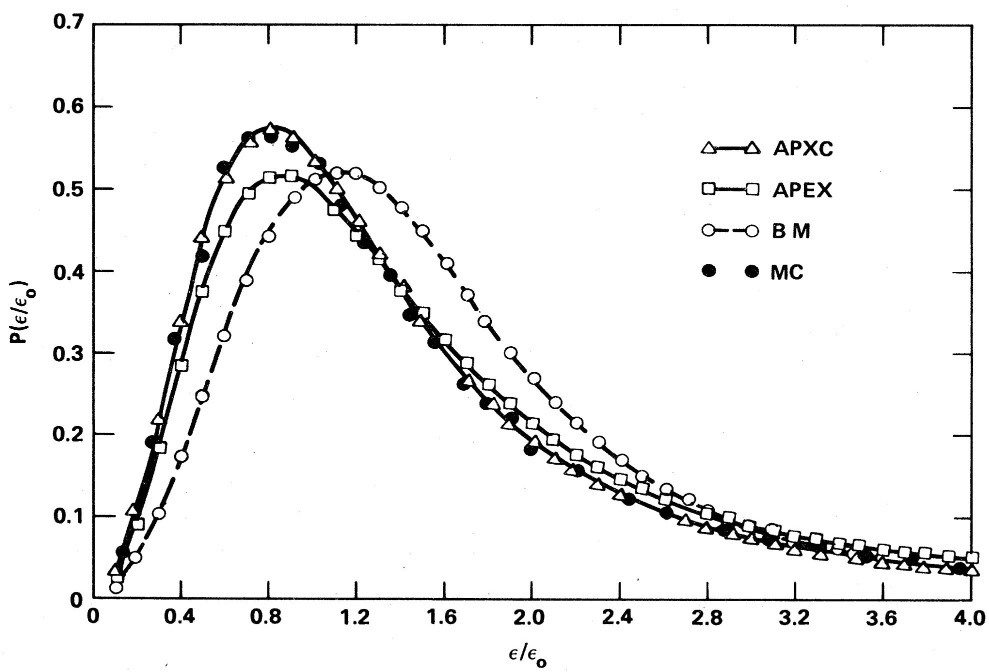

FIG. 1. $P(\epsilon)\equiv 4\pi\epsilon^2 Q(\epsilon)$ as a function of ϵ, for $\Gamma=3.9$.

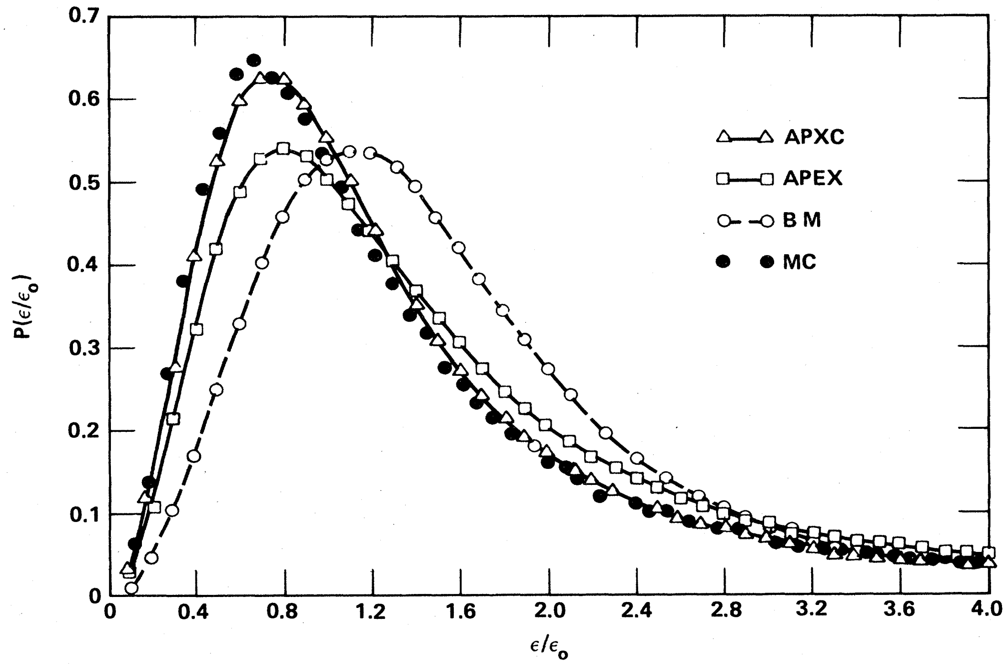

FIG. 2. Same as Fig. 1, for $\Gamma=10$.

large distance limits. However, the usual Baranger-Mozer series truncated at the first two terms also satisfies condition (3.5) and interpolates between the large λ and small λ limits, but it does so much less successfully. Apparently, the APEX field E^* is helping to reduce the size of the region of intermediate configurations. The second-moment condition controls errors only in the large λ limit, but the screened field of APEX reduces the small λ contributions, too, since ϕ^* goes to zero for the relevant configurations more rapidly than ϕ. Indeed, if the Baranger-Mozer calculations are parametrized with a screened field, the results are substantially improved.[4]

In spite of its important role in the theory, the precise functional form of E^* remains arbitrary, except that it satisfies (3.5). A natural alternative to (3.6) would be the mean force field,[9] defined in terms of the equilibrium distribution functions [see Eq. (A5)]. However, the results obtained using this choice are not as accurate[5] as those obtained from the parametrized Debye field (3.6).

In summary, APEX is least accurate when perturber-perturber correlations dominate the correlations of individual perturbers with the central point. For a neutral point, the corrections to APEX involving correlations between pairs of quasiparticles can be evaluated and the agreement with Monte Carlo calculations for $\Gamma<10$ is quite good. However, it can be seen in Fig. 2 that the Monte Carlo data seems to give a slightly higher probability to small fields than does APXC, indicating that even more terms in the renormalized Baranger-Mozer series may be required if Γ is increased any further. Since the microfield for a neutral point is not asymptotically Gaussian for large Γ, it seems probable that for very large Γ both the standard and renormalized Baranger-Mozer series will have to be summed to all orders to give accurate results.

ACKNOWLEDGMENTS

This work was initiated at a workshop on Dense Plasmas sponsored by Centre European de Calcul Atomique et Moleculaire and the authors are indebted to Carl Mozer for his hospitality and financial support during this period. The Monte Carlo data was provided by Hugh DeWitt, and helpful conversations with him and with Roy Pollock are gratefully acknowledged. The work of Boercker and Iglesias was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48.

APPENDIX: RELATIONSHIP TO OTHER RESULTS

It is straightforward to relate the Baranger-Mozer formalism and its renormalization to the formalism used by Iglesias *et al.* from the identity

$$\begin{aligned} G[\phi(\lambda)] &= G[\phi(0)] + \int_0^{\lambda} dl \frac{\partial G[\phi(l)]}{\partial l} \\ &= \int_0^{\lambda} dl \int d\mathbf{r} \frac{\partial \phi(l)}{\partial l} \frac{\delta G[\phi(l)]}{\delta \phi(l;\mathbf{r})} \\ &\equiv \int_0^{\lambda} dl \int d\mathbf{r}\, i\hat{l}\cdot\mathbf{E}(\mathbf{r}) f(\mathbf{r}\,|\,\phi(l)) \end{aligned} \tag{A1}$$

with the definition

$$f(\mathbf{r}\mid\phi(l))\equiv e^{il\cdot\mathbf{E}(r)}\frac{\delta G[\phi(l)]}{\delta\phi(l;r)} . \qquad \text{(A2)}$$

(It may be noted that this one-particle functional is the same as that used by Percus to derive integral equations for the radial distribution function of dense fluids.[6,9,12]) Equation (A1) is the formalism on which the discussion of Ref. 4 is based and Eq. (A2) provides its precise connection with the Baranger-Mozer formalism. More explicitly this is

$$f(\mathbf{r}\mid\phi)=e^{il\cdot\mathbf{E}(r)}\left[h_1(\mathbf{r}\mid 0)+\sum_{p=2}^{\infty}\frac{1}{(p-1)!}\int d\mathbf{r}_2\cdots d\mathbf{r}_p h_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid \mathbf{0})\phi(2)\cdots\phi(p)\right] , \qquad \text{(A3)}$$

where the Ursell functions $h_p(\mathbf{r}_1,\ldots,\mathbf{r}_p\mid\mathbf{0})$ are the same as those in the Baranger-Mozer expansion.

The approximations discussed in Ref. 5 are based on an expansion of $\ln[f(\mathbf{r}\mid\phi)/f(\mathbf{r}\mid 0)$ to order l^2, leading to

$$\begin{aligned}f(\mathbf{r}\mid\phi)&=f(r\mid 0)\exp[i\mathbf{l}\cdot\mathbf{E}_{\mathrm{MFF}}(\mathbf{r})+l^2\Delta(\mathbf{r})]\\&=f(r\mid 0)e^{il\cdot\mathbf{E}_{\mathrm{MFF}}(r)}[1+l^2\Delta(\mathbf{r})] .\end{aligned} \qquad \text{(A4)}$$

Here $\mathbf{E}_{\mathrm{MFF}}(r)$ is the mean force field defined by

$$\mathbf{E}_{\mathrm{MFF}}(\mathbf{r})=\mathbf{E}(\mathbf{r})+\int d\mathbf{r}'\,h_2(\mathbf{r},\mathbf{r}'\mid\mathbf{0})\mathbf{E}(\mathbf{r}') . \qquad \text{(A5)}$$

The expression for $\Delta(r)$ is also readily obtained from (A3) and depends on h_p for $p=1,2,3$. The result obtained by neglecting $\Delta(r)$ is an independent-particle model of the same form as that of APEX except with $\mathbf{E}^*(\mathbf{r})=\mathbf{E}_{\mathrm{MFF}}$ rather than the Debye form (3.6). It may be verified also that the second-moment condition (3.5) is satisfied. Nevertheless, this approximation is not as accurate as APEX, even with the corrections due to $\Delta(r)$.[5] Furthermore the λ expansion is possible only for the charged-point case.

The formalism (A1) can be renormalized using the identity (4.1) to give

$$G[\phi(\lambda)]=\int_0^{\lambda}dl\int d\mathbf{r}\,\hat{l}\cdot\mathbf{E}^*(r)f^*[r\mid\phi^*(l)] \qquad \text{(A6)}$$

with

$$\begin{aligned}f^*(\mathbf{r}\mid\phi^*(l))&\equiv e^{il\cdot\mathbf{E}^*(r)}\frac{\delta G^*(\phi^*)}{\delta\phi^*(r)}\\&=e^{il\cdot\mathbf{E}^*(r)}\left[h_1^*(\mathbf{r}\mid\mathbf{0})+\sum_{p=2}^{\infty}\frac{1}{(p-1)!}\int d\mathbf{r}_2\cdots d\mathbf{r}_p\,h_p^*(\mathbf{r}_1,\ldots,\mathbf{r}_p)\phi^*(2)\cdots\phi^*(p)\right] .\end{aligned} \qquad \text{(A7)}$$

Truncation of this expansion to first order in ϕ^* and substitution in (A6) leads to the results of Sec. IV.

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