Caging of Particles in One-Component Plasmas

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Strongly coupled Coulomb systems are characterized by localization ("caging") of particles trapped and oscillating in slowly fluctuating local potential wells. This observation constitutes the basic assumption underlying the quasilocalized charge approximation. Using molecular dynamics simulation we study the changes in the particles' surroundings (cages) in a classical three-dimensional one-component plasma. The results of our analysis show that at high coupling values, substantial changes occur only after several plasma oscillation cycles. We also analyze the oscillation frequencies of the caged particles and relate the decorrelation of the cages to the process of self-diffusion.

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Strongly coupled Coulomb systems comprise a large class of physical systems which are of importance both from a fundamental point of view and from the point of view of applicability [1]. They appear in settings as distinct as laser and particle beam compressed fusion plasmas and astrophysical plasmas (giant planetary interiors, white dwarfs, and pulsar crusts). They arise in condensed matter systems: liquid metals, electrolytes, charged polymers, semiconductor quantum wells, and quantum dots. They also occur in many realizations of mesoscopic matter: complex (dusty) plasmas, charged colloids, and ion cluster plasmas.

Centrally important for the study of such systems is the model of the classical three-dimensional one-component plasma (OCP), consisting of a single species of particles immersed in a neutralizing background. The OCP can be characterized by the ratio of the interparticle potential energy to the kinetic energy of the particles, expressed through the coupling parameter $\Gamma = q^2/(ak_BT)$, where $a = (4n\pi/3)^{-1/3}$ is the Wigner-Seitz radius, n is the density of particles, q is the charge of the particles, and T is the temperature [2,3].

A basic feature of strongly coupled plasmas in their liquid phase is the localization of charges [4–7]: there is indication that the particles spend substantial periods of time in local minima of the rough potential surface developing in such systems. At the same time, the time of localization is limited by the reformation of the potential surface due to the migration of the very particles that generate it. An approximation scheme based on the observation of localization is the quasilocalized charge approximation (QLCA) that has proven to be a very useful tool in theoretical studies of the properties of strongly coupled Coulomb systems [8–10]. In order for the QLCA to be valid one needs to assume that the period of time spent by the charges in the local potential minima extends over several oscillation cycles.

The objective of the present study is to investigate this fundamental feature of particle caging of the strongly coupled OCP in the liquid phase. This is done by analyzing the changes of the surroundings of individual particles using the correlation techniques developed by Rabani *et al.* [11,12]. The trajectories of particles are followed by molecular dynamics (MD) simulation, based on the particle-particle particle-mesh (PPPM) method [13,14], using periodic boundary conditions. The cubic simulation box has an edge length of $L = 10^{-6}$ m and the number of particles is set to $N = 1024$. The present calculations cover the $2.5 \leq \Gamma \leq 160$ range. At the start of the simulations, random initial particle configurations are set. The initial velocities of the particles are sampled from a Maxwellian distribution with a temperature corresponding to the prescribed value of Γ. The system is then thermostated for several thousand time steps, and the particle trajectories are traced in the next several thousand (2000–20 000) time steps following the thermalization period. The stability of the system is monitored by recording the system temperature (calculated from the kinetic energy of particles).

During the data collection phase information about the surrounding of each particle is stored. As discussed in [11], different definitions can be used to find the set of neighbors that make up the immediate surroundings of the particles. Here we make use of two definitions labeled (D1) and (D2): In (D1), we take the particles situated within the first coordination shell at any moment [as identified by the first minimum of the pair correlation function $g(r)$], whereas in (D2) we take the closest 14 particles at any

moment [resembling the underlying bcc (body centered cubic) structure of the 3D OCP, with the first and second closest neighbors of the lattice not being resolved in $g(r)$ in the fluid phase]. In the PP part of the simulation this is done by the efficient "chaining mesh" technique [14].

Rabani *et al.* [11,12] have defined a generalized neighbor list ℓ_i for particle i, $\ell_i = \{f(r_{i,1}), f(r_{i,2}), \ldots, f(r_{i,N})\}$. Using definition (D1), f is the Heaviside function (Θ); i.e., $f = 1$ if $r_{i,j} \leq r_c$, and $f = 0$ otherwise [11,12]. Here r_c is the cutoff radius, and the neighbors are said to be closely separated (and particle j is said to belong to the surrounding, or "cage" of particle i) if $r_{i,j} \leq r_c$. At high Γ values the first minimum of $g(r)$ occurs at $r/a = 2.42$. This value is chosen for the cutoff radius in the simulations covering the whole range of Γ. Using definition (D2) of the set of closest neighbors, the neighbor list ℓ_i always has a fixed number (14) of elements of 1, while its other elements are zero. It is noted that in the simulations using (D1) the number of closest neighbors fluctuates around 14 with a standard deviation between 1 and 2.

The similarity between the surroundings of the particles at $t = 0$ and t is measured by the "list correlation" function, derived from the scalar product of the neighbor list vectors:

$$C_\ell(t) = \frac{\langle \ell_i(t)\ell_i(0)\rangle}{\langle \ell_i(0)^2\rangle}, \tag{1}$$

where $\langle\cdot\rangle$ denotes averaging over particles and initial times. The number of particles that have left the original cage of particle i at time t can be determined as

$$n_i^{\text{out}}(t) = |\ell_i(0)^2| - \ell_i(0)\ell_i(t), \tag{2}$$

where the first term gives the number of particles around particle i at $t = 0$, while the second term gives the number of "original" particles that remained in the surrounding after time t elapsed. The cage correlation function C_{cage} can be calculated for a different number of particles leaving the cage c, as an ensemble and time average of the function $\Theta(c - n_i^{\text{out}})$, i.e.,

$$C_{\text{cage}}^c(t) = \langle \Theta[c - n_i^{\text{out}}(0,t)]\rangle. \tag{3}$$

Previous molecular dynamics studies have shown that the collective dynamics of the OCP is governed by the longitudinal plasma oscillation in the vicinity of the plasma frequency (ω_p) and by the low frequency shear mode [15,16]. The oscillation of individual particles also shows up in their phase space trajectories. The analysis of these trajectories in the $p_x - x$ phase plane reveals a dramatic qualitative difference between the quasilocalized high-Γ and the relatively low-Γ situations, as illustrated in Figs. 1(a) and 1(b). In the first case characteristic loops appear indicating the quasilocalized oscillations. The time required for a particle to proceed along one loop scatters mostly between the inverse of the Einstein frequency $\omega_E = \omega_p/\sqrt{3}$ and the inverse of the plasma frequency ω_p. While this is indeed expected on the basis of the predicted collective mode spectrum [8,9] of the 3D OCP in this coupling domain, the frequency histograms [shown in Figs. 2(a) and 2(b)] reveal additional information about the physics of the strongly coupled phase. To highlight the effect of the dynamical interaction between the particles on the frequency spectrum we have also analyzed the frequency distribution of a single caged particle in the frozen environment of the others. At high values of Γ [e.g., $\Gamma = 160$; see Fig. 2(c)] we obtain a sharp peak at the Einstein frequency; at lower values of Γ [see Fig. 2(d) for $\Gamma = 40$] the frequency distribution becomes wider. This may be due to the fact that, with decreasing Γ, there is increasing randomness and, consequently, increasing deviation from spherical symmetry in the environment sampled by the oscillating charge.

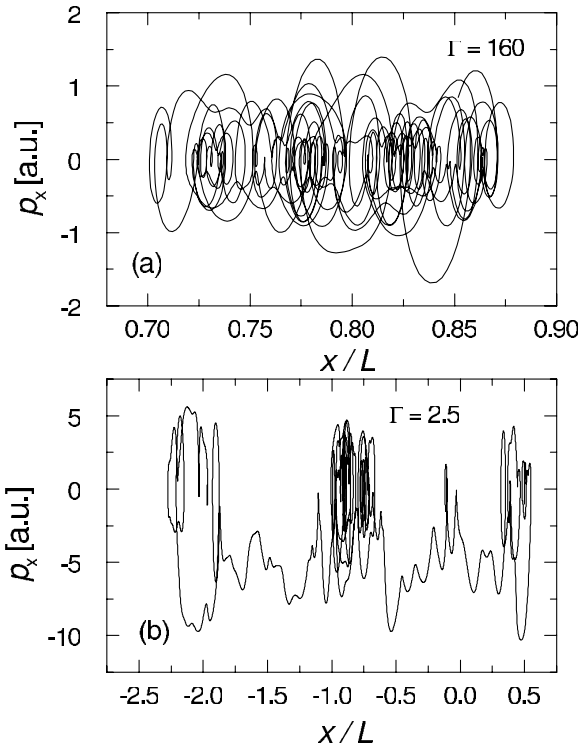

FIG. 1. $p_x - x$ phase plane trajectories of a single particle at (a) $\Gamma = 160$ and (b) $\Gamma = 2.5$.

The cage correlation functions of the 3D OCP, calculated for different values of the number of particles (c) leaving the cage [see Eq. (3)] are plotted in Fig. 3(a) for $\Gamma = 160$, as a function of time. Figure 3(b) shows the cage correlation functions obtained using $c = 7$, for several values of Γ. The data show that using definition (D2), a somewhat slower decay is obtained. We define the cages to be decorrelated when half of the particles' original neighbors leave their surroundings with 90% probability. The number of oscillation cycles T_{decorr} needed to reach this state is plotted in Fig. 3(c) as a function of Γ, for both definitions of the set of nearest neighbors. The data can be well approximated by the functional form: $T_{\text{decorr}} = A\exp(\Gamma^B)$, with approximate values $A = 0.13$ and $B = 0.35$. We can conclude that the cages of particles become decorrelated during $\approx$45 oscillation cycles at $\Gamma = 160$, while at $\Gamma = 7$ the cages decorrelate during one cycle. Near the phase

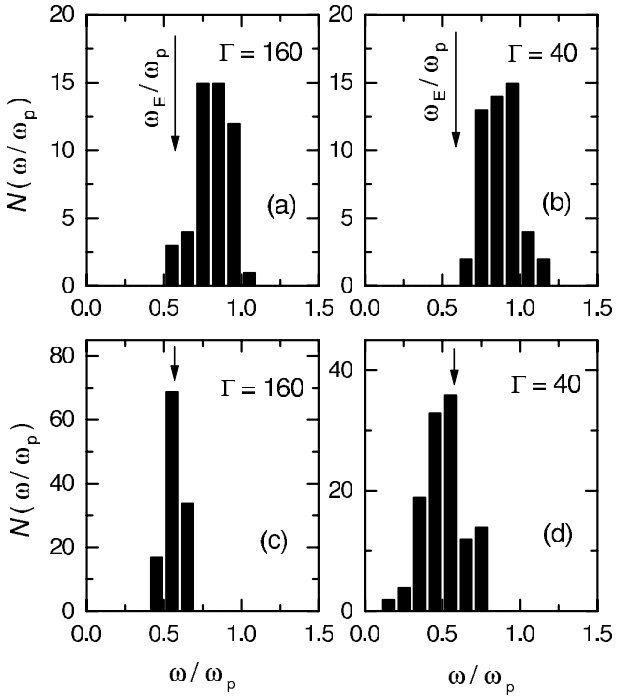

FIG. 2. Histogram spectra of closed trajectory segments of single caged particles at (a) $\Gamma = 160$ and (b) $\Gamma = 40$. (c) and (d) Same as (a) and (b), but measured in the frozen environment of the other particles.

transition point at $\Gamma \approx 172$ the localization of particles tends to be complete, and thus we can expect $T_{\text{decorr}} \to \infty$. Because of the extremely long relaxation time near this point, it is not feasible to approach this domain from the liquid side. We have, however, verified that once the system is set up in the crystalline state, e.g., at $\Gamma = 200$, T_{decorr} is indeed unmeasurably long.

Cage correlation functions can also be calculated for individual particles. Representative examples of such C_{cage} functions are shown in Fig. 4(a) for $\Gamma = 160$. The graphs (corresponding to different values of c) usually exhibit oscillations, i.e., some of the particles leaving the cage return later for some time. (These oscillations are less frequent in the case of lower Γ values.) We define T_1 as the number of oscillation cycles for the cage correlation with $c = 7$ to decay to zero (and not to return to unity again). Figure 4(b) shows a histogram of T_1 obtained from monitoring numerous individual decorrelation events. The $f(T_1)$ histogram has a quite broad distribution, the number of decorrelation cycles for $\Gamma = 160$ lies mostly between $\approx$8 and 80. The shape and the width of the histograms obtained at the lower Γ values are similar.

The escape of individual particles from the cages can be related to the process of diffusion. As the cage correlation function $C^c_{\text{cage}}(t)$ gives the probability of the escape of less than c particles from the cage, the probability of the escape of exactly one particle can be given by $P_1(t) = C^{c=2}_{\text{cage}}(t) - C^{c=1}_{\text{cage}}(t)$. Plotting P_1 as a function of time, we find an exponential decay (except at early times). The characteristic decay time t_0 relates to the probability that any

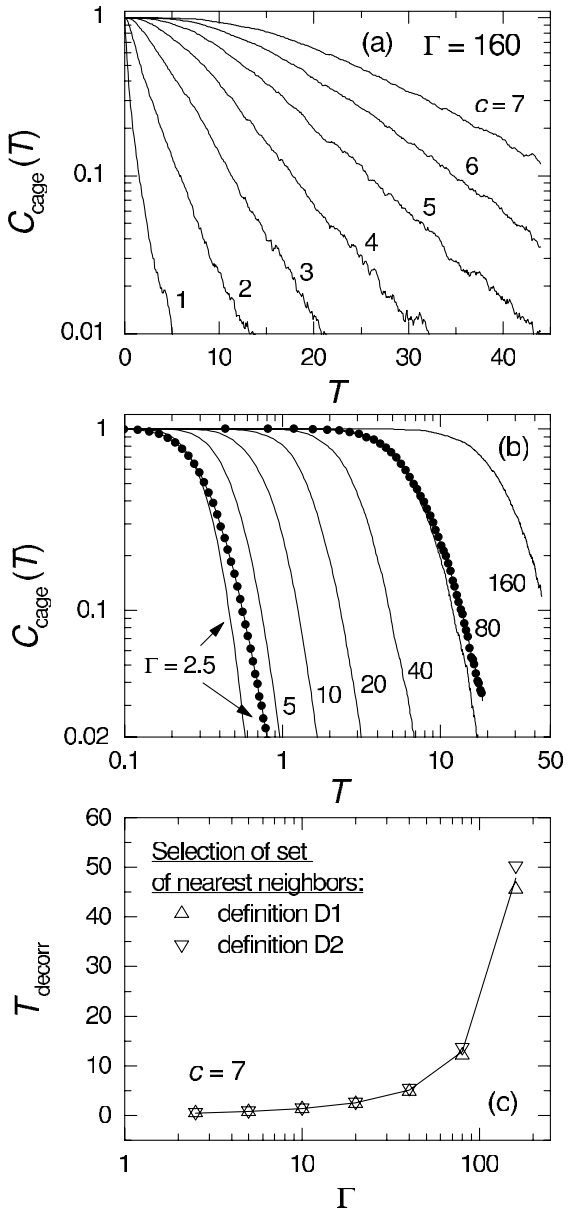

FIG. 3. (a) Cage correlation functions calculated for a different number of particles c leaving the cage at $\Gamma = 160$. (b) Cage correlation function for $c = 7$ at different values of Γ. The curves with solid lines have been obtained following definition (D1) to find the closest set of neighbors, while the dots have been obtained taking definition (D2). (c) Number of oscillation cycles during which the particle cages decorrelate.

of the 14 particles (making up the cage) leaves: a diffusion coefficient for a particle D_c can be calculated as $D_c = (\Delta x)^2/14t_0$, where Δx is the displacement of the particles associated with the escape from the cage. We have compared the D_c data choosing $\Delta x/a = 0.4$, with the independently obtained self-diffusion coefficient (calculated directly from the mean square displacement of the particles [17]): we find a good agreement in the high-Γ domain showing that escape from the cage governs the self-diffusion process in this domain. In addition, the data also agree very well with the MD Yukawa calculations

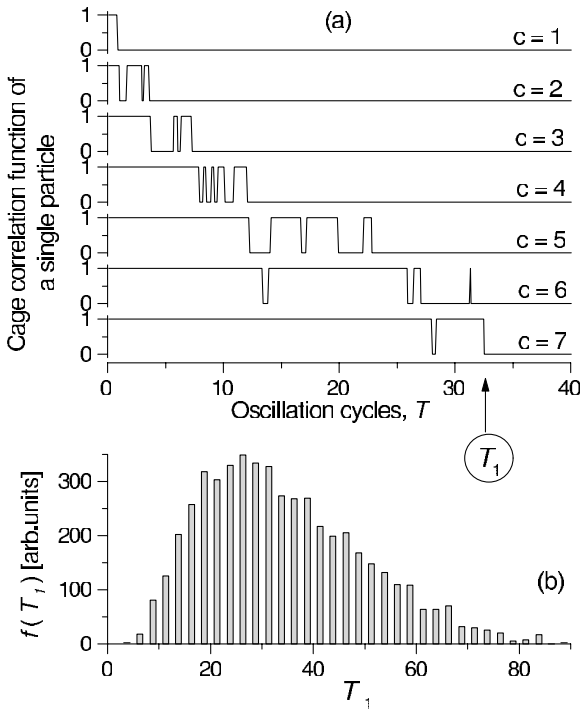

FIG. 4. (a) Examples of cage correlation functions for an individual particle, for a different number of particles leaving the cage c at $\Gamma = 160$. (b) Histogram of T_1 (number of oscillation cycles after which the $c = 7$ cage correlation function is zero) obtained from decorrelation events of individual cages.

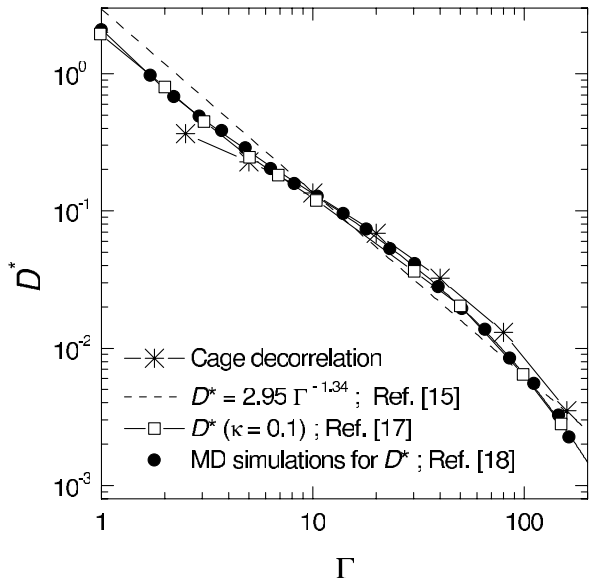

FIG. 5. Comparison of reduced self-diffusion coefficients of the OCP obtained from the decay of cage correlations, as well as from different MD simulations.

of Ohta and Hamaguchi [18] in the low κ limit. All these data reasonably emulate—although apparently with a somewhat more involved dependence on Γ—the D^* values given by the simple formula $D^* = 2.95\Gamma^{-1.34}$ of Hansen *et al.* [15]. These comparisons are shown in Fig. 5.

In conclusion, in this Letter we have been able to quantify the so far rather nebulous notion of localization in a strongly coupled OCP by analyzing through MD simulation the dependence of the particle dynamics on the coupling parameter Γ. The simulation results support the physical basis of the QLCA. They show that at high values of Γ the particle dynamics changes qualitatively: the particles are caged by their nearest neighbors and spend several oscillation cycles in local minima of the rough potential surface without experiencing substantial changes in their surroundings. The caging time is a fast growing function of Γ. The caged particles exhibit a characteristic oscillation spectrum. Escape from the cage seems to be the dominating process for self-diffusion in this coupling domain. The results have been found to be relatively insensitive to the method of selection for the set of closest neighbors.

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