Ultrafast Dynamics of Strongly Coupled Plasmas

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The ultrafast dynamics of a strongly coupled plasma following an energy landscape shift is studied theoretically and with simulation. To lowest order in time, the inertial dynamics on the new landscape can be characterized by the plasma microfield, which, for the randomly ordered case of an ultracold neutral plasma, is dominated by nearest neighbor interactions. Formation of the pair correlation function arises after ballistic overshoot, which leads to oscillations in the effective temperature. Warm dense matter systems are also considered in this context.

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The structural and dynamical properties of complex molecules, liquids, and solids are governed by the system's multidimensional potential energy function U [1]. The occurrence of various configurations is determined by the topology, or ruggedness, of the energy landscape as characterized by local minima in U. Recently, careful studies have been carried out in which sudden *changes* in this landscape are made by impulsive softening of the interatomic interactions with radiation. This work has resulted in a microscopic visualization of the solid-liquid transition using electron pulses [2] and x-ray pulses [3].

It is the purpose of the present work to explore the opposite case of impulsive *hardening* of the interatomic interactions. Among all possibilities, the most extreme case is that in which the initial state is an ideal gas, which has a topologically flat energy landscape. Rapid photoionization of an ideal gas produces a strongly coupled plasma that can, in principle, have a complex energy landscape [4]. A strongly coupled plasma is a plasma for which the Coulomb coupling parameter $\Gamma = Q^2/aT$ exceeds unity, where Q is the charge, $a = (3/4\pi n)^{1/3}$ is the Wigner-Seitz radius in terms of the density n, and T is the temperature in energy units. Owing to recent advances in laser-cooling technology, this scenario is made more compelling by the presence of experiments that employ a neutral, ultracold plasma (UCP) [5]. Although the long-time dynamics of UCPs is now understood [6], the recent experimental data have revealed novel short-time dynamics [5].

Here, the ultrafast dynamics of strongly coupled plasmas are studied with both theoretical models and molecular dynamics simulations that are capable of describing dynamics on arbitrarily short-time scales (the non-Markovian regime) with a nearly exact description of the classical many-body system. For the UCP case, the ultrafast ion dynamics on the new energy landscape is quantified and shown to be similar to the situation found in some condensed matter systems [3]. Pair correlations form after the inertial stage, and it is shown that this leads to oscillations in the effective temperature due to ballistic overshoot originating within the inertial stage.

Since the dynamics is expected to be sensitive to the particular initial conditions, we first choose the case of $T = 0$ and random initial positions, as in an UCP. The energy landscape shift occurs because the system is switched from a neutral system ($Q = 0$) to a charged system ($Q = |e|$). Because of the experimental accessibility to the quantity, the focus will be on the effective temperature (in energy units), defined as

$$T(t) = \frac{m}{3N}\sum_{i=1}^{N}\langle v_i^2(t)\rangle. \tag{1}$$

In what follows, (1) will simply be referred to as the temperature, an issue that will be addressed in more detail below. The ultrafast dynamics can be described by a Trotter expansion of the Liouville time-evolution operator at short times [7] to obtain the equations of motion:

$$\begin{aligned}\mathbf{v}_i(t) &= \mathbf{v}_i(0) + \frac{1}{2m}[\mathbf{F}_i(0) + \mathbf{F}_i(t)]t,\\ \mathbf{r}_i(t) &= \mathbf{r}_i(0) + \mathbf{v}_i(0)t + \frac{1}{2m}\mathbf{F}_i(0)t^2.\end{aligned} \tag{2}$$

To lowest order, for uncorrelated initial positions and velocities, it is easy to show that the temperature (1) evolves as

$$T(t) = T(0) + (t/\tau_2)^2 + (t/\tau_4)^4 + \cdots, \tag{3}$$

where now temperature has units of Q^2/a, time has units of inverse plasma periods ω_p^{-1}, and forces have units of Q^2/a^2. [Note that $T(t)$ is essentially $1/\Gamma(t)$.] Early-time heating is characterized by $\tau_2 = 3/\sqrt{\langle F^2\rangle}$, where $\langle F^2\rangle = \int_0^\infty dF F^2 P(F)$ is the second moment of the microfield $P(F)$, which represents the initial distribution of forces on the new energy landscape. Thus, to lowest order in time, the temperature rises purely from ballistic motion on the new landscape—no many-body dynamics is involved.

Equation (2) can be used to obtain higher-order predictions (i.e., later in time). Unfortunately, the results are not expressible in terms of well-known functions such as the microfield because $\mathbf{F}_i(t)$ describes the forces at the end of the interval when *all* particles have moved. An approximation can be made, however, by supposing that the local environment does not change too much over the short interval such that microfield gradients can be used to estimate the forces just after the initial, inertial dynamics. In fact, this is probably the case if the inertial dynamics is dominated by a nearest neighbor (NN). Under such conditions we can write

$$\tau_4 = \left(\frac{54N}{\sum_{i=1}^{N}\langle \mathbf{F}_i \cdot (\mathbf{F}_i \cdot \nabla)\mathbf{F}_i\rangle}\right)^{1/4}. \tag{4}$$

By taking each ion in turn and defining the total force be in the z direction, this can be written in terms of $\langle F_{iz}^2 \partial F_{iz}/\partial z\rangle$, which introduces the curvature of the energy landscape $\sim \partial^2 U/\partial z^2$. If the inertial dynamics indeed arises from the nearest neighbor, we can write this as $\langle F_{iz}^2\rangle[\partial F_{iz}/\partial z]_{F_{iz}}^{NN}$. Should this turn out to be accurate, we immediately know that the sign of the t^4 contribution is negative because the microfield gradient is [8]; essentially, this can be thought of as capturing the fact that the ions move into regimes with lower field strength.

Because of the small electron-ion mass ratio, ion dynamics can be described by a linearly screened Coulomb potential when the electrons are weakly coupled to all species, which is the case for UCPs. Consider then dynamics generated by the Yukawa pair potential $u_Y(r) = T\Gamma \exp(-\kappa r)/r$, where r is in units of a and $\kappa = a/\lambda$ is a dimensionless inverse screening length, which describes ions adiabatically and linearly screened by free electrons. Such a model serves as an accurate description for UCPs [5,6]. That the dimensionless form of (3) depends *only* upon κ reveals the quasiuniversal behavior seen in experiments [5], for which the variations in κ were very small (see below). The ion heating rate as a function of κ can be quantified by $\langle F^2\rangle = 33 - 4\kappa + 0.1\kappa^2$, as determined by placing $N = 10\,000$ particles randomly in a periodic box. Note that the heating time τ_2 is insensitive to κ. This fact makes the heating of UCPs appear to be more universal than perhaps might have been expected. This can be understood from the properties of microfields; in particular, the second moment is very sensitive to large field values, which mainly arise from the nearest neighbor. Since nearest neighbor distances for a random distribution are, on average, much less than a, the dominate forces arise from close range where nearly pure Coulomb forces are experienced. This behavior of microfields has been discussed previously in the context of dense plasma spectroscopy [8].

The ideas presented so far have been tested with molecular dynamics (MD) simulations, details of which are described elsewhere [6,8]; briefly, the equations of motion of $N = 5000$ particles have been integrated using the potential $u_Y(r)$ with periodic boundary conditions. An important property of the Yukawa model is that electrons are *not* dynamic, so that electron-ion collisional relaxation does not compete with the process under consideration [9]. Results for $\kappa = 0.5, 1.0, 2.0$ are shown in Fig. 1 for $T(t)$ versus time. In real units these are for UCP-like cases, with

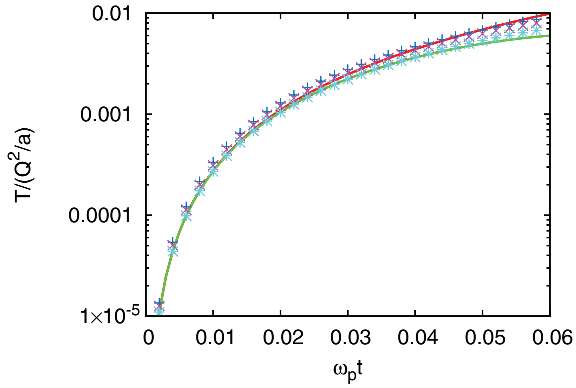

FIG. 1 (color). Simulation results for the temperature versus time in the inertial phase for three plasmas with $\kappa = 0.5$ (blue pluses), $\kappa = 1.0$ (magenta crosses), and $\kappa = 2.0$ (cyan stars). Also shown are predictions based on τ_2 alone (solid red line) and with τ_4 contributions (solid green line) for $\kappa = 2.0$.

fixed $T_e = 5$ K (Debye screening model), $Q = 1$, $\mathbf{v}_i(0) = 0$, and $n = 3.68 \times 10^6$, $n = 2.35 \times 10^8$, $n = 1.5 \times 10^{10}$ cm^{-3}. These results were compared with detailed two-component plasma simulations, and the efficacy of the Yukawa model was established; this comparison will be reported elsewhere. Note that the MD results, represented by points, are quite similar despite the differences in κ; this supports the notion that the dynamics is quasiuniversal over this large density range. The red line is based on (3) with τ_2 contributions only and the green line includes contributions through τ_4. The ultrafast dynamics is very well predicted by this microfield model, including the approximations involved in obtaining (4), which suggests that indeed nearest neighbors dominate the landscape on this time scale. Because the ultrafast dynamics is described well by a microfield model, it is clear that many-body processes *have not begun* on this time scale. This is similar to the equilibrium situation in liquid-state correlation functions [10].

Before moving to intermediate-time dynamics, it is necessary to discuss different initial conditions. In particular, there is interest [6,11,12] in precorrelated systems. In the opposite extreme of a lattice of ions, the microfield will contain a great deal of cancellation; thus, $P(F)$ will be peaked at small F, which would yield a very slow heating rate. Moreover, a term that vanished previously that scales as $\langle \mathbf{v}_i(0) \cdot \mathbf{F}_i(0)\rangle t$ no longer vanishes because the forces on each ion are no longer random; ions will initially move against the forces (on average) and the system will *cool* [12,13]. Thus, the initial heating or cooling depends on the convexity of the new local environment.

Some insight is gained by writing $T(t)$ as

$$T(t) = T(0) + \frac{2}{3N}\sum_{i=1}^{N}\int_0^t dt'\langle \mathbf{F}_i(t') \cdot \mathbf{v}_i(0)\rangle + \frac{1}{9N} \times \sum_{i=1}^{N}\int_0^t dt'\int_0^t dt''\langle \mathbf{F}_i(t') \cdot \mathbf{F}_i(t'')\rangle, \tag{5}$$

which is nothing more than (1) with $\mathbf{v}_i(t) = \mathbf{v}_i(0) + \int_0^t dt'\mathbf{a}_i(t')$. This form shows how memory enters via the time integrations. More importantly, the final term describes the force on each particle at two different times, which in turn depends on the positions of all other particles at those times, and therefore introduces a *dynamic three-particle* correlation function. A useful kinetic theory description will therefore require an accurate description of nonequilibrium, many-particle correlations. It is possible, however, to gain physical insight by writing the time derivative of $T(t)$ in terms of macroscopic variables as

$$\frac{dT(t)}{dt} = \frac{2}{3N}\int d^3r \int d^3r' \mathbf{F}(|\mathbf{r} - \mathbf{r}'|) \cdot \langle n(\mathbf{r}, t)\mathbf{j}(\mathbf{r}', t)\rangle, \quad (6)$$

where the density is $n(\mathbf{r}, t) = \sum_{i=1}^N \delta(\mathbf{r} - \mathbf{r}_i(t))$, the current is $\mathbf{j}(\mathbf{r}, t) = \sum_{i=1}^N \mathbf{v}_i(t)\delta(\mathbf{r} - \mathbf{r}_i(t))$, and it is understood that particles do not act on themselves. Positions and momenta partition (classically) in thermal equilibrium and this quantity vanishes, as it should. In general, (6) shows that the temperature can either increase or decrease depending on the average coherence between the forces and currents. In the UCP case considered above, ions mainly move *with* the force and $dT(t)/dt > 0$; for a highly correlated (latticelike) initial state, ions move mainly *against* the force and $dT(t)/dt < 0$. In general, the result (6) suggests that sign changes of $dT(t)/dt$ can occur at any time, not just initially. For example, the initially ballistic ions will eventually encounter other, repelling ions; because of inertia, the ions overshoot before reversing direction. If this can happen on about the same time scale for each ion—i.e., coherently—the temperature can reverse its trend. The temperature is unlikely to return to its initial value, because the randomness in the initial environment breaks perfect coherence. This behavior is, of course, exactly what is seen in recent UCP experiments [5].

Again, MD has been used to quantify the preceding arguments, now using $N = 10\,000$ particles. Three UCP-like cases in which the electron temperature has been fixed at $T_e = 38$ K, the initial ion temperature is taken to be $T_i(0) = 10$ mK, and the densities $n = 0.736, 1.47, 2.58 \times 10^9$ cm^{-3} have been considered. Screening lengths, $\kappa = 0.44, 0.49, 0.54$, are very similar due to the very weak n dependence of κ. These densities are e^{-1} of the peak densities in the experiments [5], which serves as a rough correction for the inhomogeneity in the experiments. The evolution of the temperatures, in mK, is shown in Fig. 2(a) and, to the accuracy of the MD model, very good agreement with the experiments is found. The results show quasiuniversal relaxation and strong temperature oscillations. Figure 2(b) shows the moment ratios $\frac{3}{5} \times \langle v^4\rangle/\langle v^2\rangle^2$ (lower set) and $\frac{9}{35}\langle v^6\rangle/\langle v^2\rangle^3$ (upper set) of the velocity distribution, which would be unity for a Maxwellian. These ratios quantify the degree to which the term "temperature" has a meaning; deviations from a time-dependent Maxwellian are evident, but both moments approach near unity quickly ($\sim \omega_p^{-1}$). During the ballistic phase ($t < 0.1\omega_p^{-1}$), strong deviations from a Maxwellian are seen.

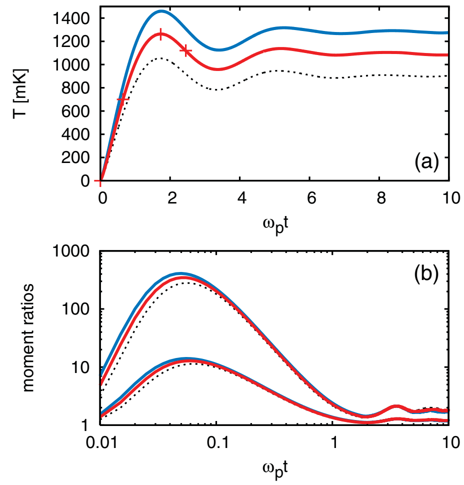

FIG. 2 (color). Temperature versus time for three UCPs is shown in panel (a). At intermediate times, strong temperature oscillations appear. Also shown, in panel (b), are the moment ratios $\frac{3}{5}\langle v^4\rangle/\langle v^2\rangle^2$ and $\frac{9}{35}\langle v^6\rangle/\langle v^2\rangle^3$ that would be unity for a pure Maxwellian. During the inertial phase there are large deviations from a Maxwellian, but quasiequilibrium distributions occur beyond one plasma period.

To corroborate the picture given above, the pair correlation function $g(r, t)$ and radial velocity distribution $v_r(r, t)$ were computed for the intermediate density case. The former quantity reveals how the pair correlations form in general, whereas the latter quantity is more closely connected to the currents in (6). Results are shown in Figs. 3(a) and 3(b) at the 4 times marked in Fig. 2(a): initial, during heating, when the temperature has stalled, and during cooling. At the earliest 2 times (red and blue lines) the pair correlation function reveals a blast wave as ions rapidly separate. This behavior is reflected in the velocity field by large, positive values. At the peak (black dotted line), when $dT(t)/dt = 0$, the pair correlation function has an equilibriumlike form and the velocity field has small values about zero. Finally, as the temperature drops, ions have reversed directions and the Coulomb hole is

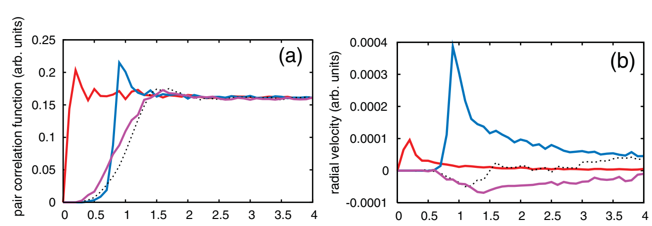

FIG. 3 (color). Formation of the pair correlation function $g(r, t)$ (a) and the velocity field $v_r(r, t)$ (b) at the 4 times marked in the previous figure. At early time, $g(r, t)$ has a blast-wave character (red and blue lines), whereas complete reversal of the current occurs after reaching the peak temperature (magenta lines).

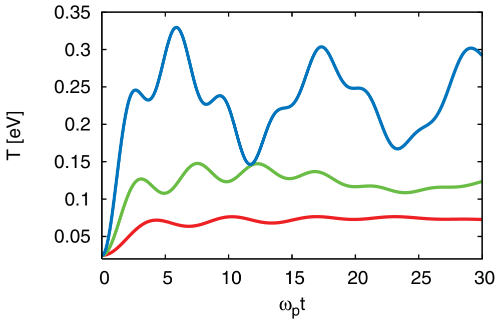

FIG. 4 (color). Temperature oscillations for warm dense aluminum at solid density and room temperature. The electrons are assumed to be heated from room temperature to $T_e(0^+) = 20$ eV (red line), $T_e(0^+) = 50$ eV (green line), and $T_e(0^+) = 200$ eV (blue line).

beginning refill (magenta line); the velocity field is now completely negative. Thus, the temperature oscillations seen in recent UCP experiments [5] directly reflect the detailed formation of the pair correlation function.

Interestingly, temperature oscillations have been seen before in MD simulations [9,14], but *no interpretation* was given for their appearance or behavior. Kinetic theory calculations do not show the oscillations, even when computed for equivalent conditions as in the MD [13]. As we now know that the oscillations are real [5], and reflect very interesting intermediate-time dynamics, further kinetic theory explorations are warranted.

It is interesting to investigate the degree to which the ideas presented so far apply to warm dense matter, since such experiments are typically underdiagnosed and incomplete knowledge of the temperature evolution has led to behavior that is difficult to explain [15]. To not cloud the underlying physics by adding complexities associated with electronic structure, the Yukawa model will be used with a finite-temperature Thomas-Fermi screening length. Consider an aluminum target initially at room temperature ($T = 300$ K) that is laser- or beam-heated instantaneously at $t = 0$. Because the energy is absorbed primarily by the electrons, it is assumed that T_e suddenly jumps to either $T_e(0^+) = 20$ eV, $T_e(0^+) = 50$ eV, or $T_e(0^+) = 200$ eV, values consistent with typical experiments. For simplicity the valence is fixed at $\langle Z \rangle = 3$ and a long equilibration crystallizes the aluminum. The temperature evolution is shown in Fig. 4 where clear evidence for disorder-induced heating (DIH) and temperature oscillations is seen. Consistent with predictions [6] for precorrelated states, the magnitude of DIH is much lower than for UCPs. Thus, dense plasmas likely experience the same DIH and temperature oscillations as UCPs do, but on a time scale that is much faster because of the higher density.

It has been shown that the ultrafast dynamics of strongly coupled plasmas is inertial on the new energy landscape, as characterized by the microfield and its gradients, similar to condensed matter systems [3]. Ions move ballistically in these fields with many-body contributions entering at about $0.1\omega_p^{-1}$. Predictions based on a short-time evolution model agree well with MD results. For random initial conditions, in which the plasma will always heat [6], the heating is quasiuniversal in the Yukawa model, with a weak κ dependence, similar to experimental observations [5]. The formation of the pair correlation function, and related temperature oscillations, has been quantified and details revealed by MD; agreement with experiment is good. A simple picture emerges in which temperature oscillations arise from ions overshooting their equilibrium positions following their ballistic motion on the new landscape. Interestingly, it has been shown that the usual definition of temperature as a mean kinetic energy has moments similar to a time-dependent Maxwellian, *after* the ballistic, inertial dynamics phase. Finally, these ideas have been applied to the formation of warm dense matter in which it has been shown that similar ultrafast dynamics occurs at 10 orders of magnitude higher density and with partially degenerate electrons, which may have implications for the interpretation of notoriously underdiagnosed experiments [15], and possibly Doppler broadening in transient inversion collisional x-ray lasers [16], which scales as $\lambda/\Delta\lambda \sim T^{-1/2}$.

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