Ultrafast phase-space dynamics of ultracold, neutral plasmas

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Abstract

The nonequilibrium phase-space dynamics of neutral, ultracold plasmas are
described. The dynamics are placed in the context of the ultrafast dynamics
of related systems that can be thought of as *photo-initiated ultrafast systems*,
which include, among others, surfaces that are melted nonthermally, some
artificial molecular machines and laser-excited clusters. The picture of energy
landscape hopping is described to unify the dynamics of these diverse systems.
The specific features of the temperature evolution of an ultracold plasma are
then discussed in detail as they compare with the short-time dynamics of the
velocity autocorrelation function. Finally, the phase-space dynamics of a one-
dimensional ultracold plasma model are visualized via molecular dynamics
calculations of phase-space trajectories.

PACS numbers: 05.20.Dd, 52.65.−y, 52.27.Gr

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Ultracold neutral plasmas (UCPs) are plasmas formed by photoionizing a dilute gas of cold,
trapped atoms. With densities near 10^9 cm^{-3} and temperatures in the μK range, these plasmas
begin their existence with a Coulomb coupling parameter of $\Gamma \sim 10^5$, where

$$\Gamma(t) = \frac{e^2}{aT_i(t)} \qquad (1)$$

measures the average ratio of potential to kinetic energy. Here, $a = (3/4\pi n)^{1/3}$ is the ion-
sphere radius and $T_i(0)$ is the initial temperature. It is now well established theoretically
[1] and experimentally [2] that UCPs will spontaneously heat until they reach a coupling
of order unity. This heating process, known as 'disorder induced heating' (DIH), can be
seen as a failing of UCPs in that their large initial coupling parameter cannot be maintained.

Consequently, since the first prediction of DIH [1], strategies have been sought to mitigate the DIH process [3]. Alternatively, the process of DIH can be embraced [4], and one can view UCPs as interesting experiments in the physics of nonequilibrium, ultrafast dynamics; that is the point of view that will be taken here.

UCPs can be thought of generally as a many-body system in which the interactions between particles have suddenly been changed. In the following section, a brief review of similar systems is given in order to place UCP dynamics in a broader context. Because of the importance the Yukawa model plays in our understanding of UCPs, the basis for that model is discussed. Details specific to UCPs are then given. In the following two sections, the connection between time correlation functions and the observed oscillations in the kinetic energy [2] is discussed. Similarities and differences between the true nonequilibrium dynamics and the velocity autocorrelation function are described. Then, the dynamics in phase space is considered in a one-dimensional UCP model that allows visualization of the DIH process. Finally, a discussion and outlook are given.

2. Survey of ultrafast systems

The focus of the present work is the ultrafast dynamics of photoexcited systems, with an emphasis on the more ubiquitous classical systems. In general, each system can be thought of as having N nuclei that interact via effective forces that arise from the neutralizing, screening electrons. Such systems can be described by the Hamiltonian

$$\mathcal{H} = \sum_{j=1}^{N} \frac{p_j^2}{2m} + U(\mathbf{r}_1, \ldots, \mathbf{r}_N), \tag{2}$$

where the potential energy term U cannot generally be written as a sum over m-body potentials, where $m < N$. In general, obtaining the form for U is difficult and requires an electronic structure calculation. The system may or may not be in contact with a thermal reservoir; but, more importantly, if the dynamics are fast enough and the coupling to the reservoir is weak enough, the ultrafast dynamics will generally be microcanonical. In the case of microcanonical evolution, the energy is a constant of the motion, that is, $\mathcal{H} = E$, and the possible configurations of the coordinates $\mathbf{r}$ are those restricted to lie on the energy surface specified by E.

Suppose that the system is photoexcited in some manner. The radiation will be absorbed predominantly by the electrons, which will be excited (or ionized) to energy levels consistent with the absorbed energy. Because the electrons determine the effective ion–ion interactions that determine the landscape defined by U, the photoexcited system will now have a new potential energy function U'. If the photoexcitation process is fast enough, the positions and momenta of the nuclei will have changed little. Now, the system can evolve on a different energy landscape E', and different configurations will be possible. One possibility is that through electron–ion collisions [5], the ions can heat. This process is, however, very slow due to the kinematically unfavorable situation arising from very disparate masses, and Pauli blocking in electron degenerate systems. Therefore, the observed dynamics that arises on very short timescales are due to the 'hop' from the energy surface E to E' alone.

Such a photoexcited energy landscape hopping process occurs in a wide variety of physical systems. As these systems are quite diverse, a short summary is given for each with the goal of motivating the importance of understanding ultrafast dynamics in general, before turning to the important specific case of ultracold plasmas.

2.1. Cluster fusion

A cluster is a small multiatom assembly of particles; anything from a few particles up to several million particles can be considered a cluster. Although the main interest in clusters is understanding the transition from atomic and molecular behavior to bulk solid behavior, it is also of interest to photoexcite clusters. One interesting application involves high-intensity, femtosecond laser-heated deuterium clusters [6]. In this system, the initial U corresponds to a potential energy that binds deuterons into D_2 molecules, and further binds the molecules into a stable cluster. As the radiation is high intensity ($>10^{16}$ W cm^{-2}), the electrons are ionized from the cluster, and the ionic interaction U' is essentially a weakly screened Coulomb system with very strong repulsive forces. These forces initiate a Coulomb explosion that accelerates the ions to keV or higher energies. When the deuterons from neighboring clusters collide, it is possible to create fusion neutrons. Suppose that one then engineers a specific form for U that enhances the forces after the laser pulse, as described by U'; such a scheme could enhance the fusion output. This, in fact, has been carried out [7], and orders of magnitude enhancements have been seen. As we will see in the following section, these experiments are similar to UCPs, except that UCPs are not nearly as dense, and Coulomb explosion does not occur.

2.2. Non-thermal melting

It is also possible to study the time development of phase transitions using photoexcitation. Interestingly, using femtosecond x-ray pulses, melting of InSb has been observed to occur on timescales much faster than the electron–phonon coupling time [8]. This is easily understood using the concept of energy landscape hopping. Initially, the semiconductor is bonded into a crystalline structure according to the landscape U. Just after photoexcitation, the bonds are weaker, and the ions forming the lattice will instantly begin to evolve on the new landscape U'. As the x-ray pulses are very short, the initial ionic momenta are unchanged and the new energy of the system is $E' = E + U' - U$. It has been pointed out that the newly formed liquid differs from the equilibrium liquid [9], a subject that forms the bulk of what follows in the present work.

2.3. Photoisomerization of molecules

In biological systems, the conformation of molecules often confers its function. The particular conformation of a molecule, such as a protein, is determined by the landscape function U, which may also include interactions with the environment. Some proteins are photoactive, such as photoactive yellow protein, and can readily become photoexcited by an electronic transition from the electronic ground state to an excited state. When this happens, the effective ionic interactions embodied in U become U', and a new set of conformations is possible [13]. Perhaps more interestingly, this photoisomerization process can be exploited for other purposes. For example, photoexcitation of engineered molecules can be used as nanoswitches [14], and can even be used in drug delivery systems in the treatment of multiple diseases, such as cancer [15].

I have only considered three examples of photoexcited, ultrafast systems here. Although there are many others that could be discussed in detail, such as the electron dynamics in semiconductors [10], I now turn to UCPs.

3. Ultracold plasmas as Yukawa systems

In what follows, I will model UCPs as a Yukawa system, an approximation that requires some justification. First, consider the electrons. The degeneracy parameter for the electrons can be

written as

$$\Theta = \frac{E_F}{T} \tag{3}$$

$$= \frac{1.7 \times 10^{-5} (n/10^{10}\ \text{cm}^{-3})^{2/3}}{T/1\ \text{meV}}. \tag{4}$$

From this expression it is clear that the electrons can typically be treated classically for most experiments. Next, consider the electron–electron Coulomb coupling parameter, which can be written as

$$\Gamma_{ee} = \frac{0.5(n/10^{10}\ \text{cm}^{-3})^{1/3}}{T/1\ \text{meV}} \tag{5}$$

and reveals that the electrons tend to be only moderately coupled. Now, since the electrons are very light compared with the ions, we can treat the electrons as instantly responding to the slow ion motions. (This assumption will be tested below.) In this case we can write a Poisson equation for the total potential of the system as

$$-\frac{1}{4\pi} \nabla^2 \phi(\mathbf{r}) = -e n_{e0}\, \mathrm{e}^{\beta e \phi(\mathbf{r})} + Ze \sum_i \delta(\mathbf{r} - \mathbf{r}_i(t)). \tag{6}$$

Although this equation is applicable to UCPs, it is difficult to solve in general due to the nonlinearity of the electron Boltzmann distribution. Since the electrons are not very strongly coupled, we can linearize the exponential to obtain the linear Poisson equation that has the well-known Yukawa or Debye–Hückel form; here, this takes the form of a superposition of Yukawa potentials from each location $\mathbf{r}_i(t)$. Note that the screening length appropriate to this problem is the classical electron Debye length. (If one is careful, there is also a constant term that arises from charge neutrality; this does not cancel here, as it usually does, since the ions are treated explicitly. The constant term has no effect on the forces, of course.) The main result is that the electrons can, to a reasonable approximation, be treated as linear screeners of the ions, although there will clearly be corrections due to moderate electron coupling.

Perhaps the most subtle assumption in the previous analysis is that the electrons statically screen the ions. Because the focus here is on ultrafast dynamics, the consequences of dynamical screening should be investigated. For simplicity, consider an ion moving in a straight line given by $\mathbf{r}(t) = \mathbf{r}_0 + \mathbf{v}t$. If this ion is dynamically screened by the electrons, again assuming weak coupling (more specifically, linear response), the total screened potential produced by this ion can be shown to be [11]

$$u_S(\mathbf{r}) = \frac{e^2}{2\pi^2} \int \mathrm{d}^3 k\, \frac{\mathrm{e}^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}(t))}}{k^2 \epsilon(k, \mathbf{k}\cdot\mathbf{v})}, \tag{7}$$

where $\epsilon(k, \omega)$ is the dielectric response of the electrons. The criteria for static screening can be stated roughly as

$$k_{De} v_{\text{typ}} \ll \omega_{pe}, \tag{8}$$

or, equivalently,

$$m_e v_{\text{typ}}^2 \ll T_e. \tag{9}$$

Here, v_{typ} is a 'typical' ion velocity. If we assume that the potential energy per particle available from the ultrafast dynamics is of order e^2/a_i, then the latter expression becomes

$$\Gamma_{ee} \ll \frac{m_i}{m_e}, \tag{10}$$

which is always satisfied in UCPs. Thus, we can expect that the Yukawa analysis, with its caveats, given above applies to current experiments.

The assumptions discussed in this section have been explored numerically, although with great difficulty because of the mass ratio involved, and results from a full two-component plasma simulation are in fact in rough agreement with a Yukawa model, although the agreement is not perfect [12]. One expects from the analysis given here that the most important weakness of the Yukawa model is the treatment of electron coupling.

4. Energy landscape hopping in ultracold plasmas

I will now focus on the specific case of ultracold plasmas, and why they are important in the context of photoexcited ultrafast dynamics. UCPs are formed by trapping and cooling a dilute atomic gas to very low temperatures (in the μK regime), and subsequently laser photoionizing some portion of the atoms into singly ionized ions. In this case, we can write the atomic (a) and plasma (p) Hamiltonians as

$$
\mathcal{H}_a = \sum_{j=1}^{N} \frac{p_j^2}{2m}
$$

$$
\mathcal{H}_p = \sum_{j=1}^{N} \frac{p_j^2}{2m} + \sum_{j<k}^{N} \frac{e^2}{r_{jk}} \, e^{-r_{jk}/\lambda},
$$

(11)

where the Yukawa interaction between ions has been assumed, and λ is the screening length of the electrons. From these Hamiltonians, it is easy to see how UCPs are unique. First, to a very good approximation, the Hamiltonians are *known* (to the degree that the Yukawa potential applies) and simple (in that only pair interactions appear). Second, the initial state is the extremely simple ideal gas state in which there are no correlations between the ions at $t = 0$, just after the laser pulse. Third, UCPs are relatively dilute, which effectively 'slows' down the timescales such that details of the evolution can be measured. Fourth, the landscape hop is enormous due to the difference in the strength of the atomic interactions (negligible) and the strong long-ranged Coulomb interaction. It is for these reasons that UCPs can play a central role in the elucidation of the physics of photo-initiated ultrafast systems.

5. Connection with the velocity autocorrelation function

It has been noted [3] that the kinetic energy oscillations appear to relax in a manner similar to that of a time correlation function. The idea is that, after photoionization, the gas behaves as a Coulomb liquid, which will generally show oscillations in its autocorrelation functions. Similar analogies have been discussed in the context of non-thermal melting [9], where the photoexcitation process melts a lattice into a liquid state and one might expect liquid-like behavior just after photoexcitation. UCP experiments measure the Doppler motion of the ions, which is a measure of the kinetic energy; as this has the form of a velocity squared, it is logical that there may be a connection between the relaxation of an UCP and the velocity autocorrelation function (VACF) of the plasma state. This connection will be explored in this section.

Let us first begin with a short review of the VACF. The normalized VACF is defined as

$$
Z(t) = \frac{\langle v(t)v(0)\rangle}{\langle v^2(0)\rangle},
$$

(12)

where $v(t)$ is the velocity of a single particle in some particular direction. Beyond its intrinsic theoretical interest in classical many-body theory [16], $Z(t)$ is important because of its relation to the diffusion coefficient, namely

$$D = \frac{T}{m} \int_0^\infty \mathrm{d}t \ Z(t). \tag{13}$$

At this point there are already two important points to call attention to. First, the VACF relates a quantity at one time to another quantity at another time. Second, the meaning of the brackets $\langle \cdots \rangle$ is a thermal average, in which it is assumed that the ergotic theorem is satisfied in that one can equivalently understand the averaging as over ensembles or over time. Explicitly, the equilibrium averaging takes the form

$$\langle \mathcal{A}(\mathbf{r}^N, \mathbf{p}^N) \rangle = \int \mathrm{d}^{3N} r \, \mathrm{d}^{3N} p \, \mathcal{A}(\mathbf{r}^N, \mathbf{p}^N) \, \frac{\mathrm{e}^{-\beta \mathcal{H}}}{\int \mathrm{d}^{3N} r \, \mathrm{d}^{3N} p \, \mathrm{e}^{-\beta \mathcal{H}}}. \tag{14}$$

As a result, the VACF is only dependent upon a single time, which is the time difference. Here, $\beta = (k_B T_i)^{-1}$ is the inverse temperature.

Because we are interested in liquid-like behavior immediately following photoexcitation, and because exact results can be written down for the short-time dynamics of equilibrium correlation functions, consider $Z(t)$ near $t = 0$; at $t = 0$ we have $Z(0) = 1$, trivially. Now, to treat the time evolution formally, I will introduce the Liouville operator

$$\mathrm{i}\hat{L} \equiv \{, \mathcal{H}\} \tag{15}$$

$$= \sum_{j=1}^N \frac{\partial \mathcal{H}}{\partial \mathbf{p}_j} \cdot \frac{\partial}{\partial \mathbf{r}_j} - \frac{\partial \mathcal{H}}{\partial \mathbf{r}_j} \cdot \frac{\partial}{\partial \mathbf{p}_j}, \tag{16}$$

which describes the time evolution of the velocity as

$$v(t) = \mathrm{e}^{\mathrm{i}\hat{L}t} v(0). \tag{17}$$

Note that I use velocities when I refer to quantities such as the VACF, which is customary, but use momenta when I refer to more general aspects of phase-space dynamics, as is customary; this should not introduce any confusion. To lowest order in a Taylor expansion, the VACF is then

$$Z(t) = \langle v^2(0) \rangle^{-1} \left\langle v^2(0) - \frac{v(0)}{m} \frac{\partial \mathcal{H}}{\partial r} t + \cdots \right\rangle, \tag{18}$$

where the term linear in t vanishes, because

$$\left\langle v(0) \frac{\partial \mathcal{H}}{\partial r} \right\rangle = \langle v(0) \rangle_p \left\langle \frac{\partial \mathcal{H}}{\partial r} \right\rangle_r \tag{19}$$

$$= 0 \tag{20}$$

as a result of the momentum and space partitioning that takes place in (14), due to the form of (2). The linear (in time) term vanishes because the system is in equilibrium.

For the actual UCP experimental situation, consider the 'temperature' quantity

$$\langle T(t) \rangle = \left\langle \frac{1}{3Nm} \sum_{i=1}^N p_i^2(t) \right\rangle. \tag{21}$$

Here, the brackets $\langle \cdots \rangle$ have a different meaning. Now, the brackets refer to an average over initial conditions, and this average may or may not have the form (14). Moreover, even if it did have that form, the *time evolution of the system is now governed by a Hamiltonian different*

from the one that appears in the averaging, or the system would not have changed. This can be written explicitly as

$$\langle T(t) \rangle = \frac{1}{3Nm} \sum_{i=1}^{N} \frac{\int d^{3N}r \, d^{3N}p \, e^{-\beta \mathcal{H}_a} \, e^{i\hat{L}_p t} \, p_i^2}{\int d^{3N}r \, d^{3N}p \, e^{-\beta \mathcal{H}_a}}, \tag{22}$$

where the averaging corresponds to the landscape U (which is zero for UCPs) and the evolution corresponds to the landscape U'. That is, the evolution operator $\hat{L}_p$ depends upon $\mathcal{H}_p$ rather than $\mathcal{H}_a$. Note that $\langle T(t) \rangle$ also depends on a single time, but this is an absolute time after photoexcitation, and not a time difference in a stationary sense. Here, the inverse temperature β now refers to the distribution of states *before* photoionization, and $T(t)$ refers to the actual temperature at time t, according to definition (21). In (14), since the temperature does not change, there is no distinction: this is an essential difference between equilibrium and non-equilibrium dynamics.

Again, we can perform a short-time expansion. Obviously, $\langle T(0) \rangle = T_i(0)$ is the initial temperature. To lowest order in time, we obtain

$$\begin{aligned} p_i^2(t) &= [e^{i\hat{L}_p t} p_i(0)]^2 \\ &= p_i^2(0) + 2p_i(0)(i\hat{L}_p)p_i(0)t + \mathcal{O}(t^2) \\ &= p_i^2(0) - 2p_i(0)\frac{\partial \mathcal{H}_p}{\partial r_i}t + \mathcal{O}(t^2), \end{aligned} \tag{23}$$

where $i\hat{L}_p p_i(0) = -\partial \mathcal{H}_p/\partial r_i$ has been used in the final step. Here, we find that we have to evaluate a quantity similar to (18), but recall that the brackets have a different meaning. Because we are assuming that the photoexcitation process is instantaneous, the $p_i(0)$ refer to the momenta before the hop, whereas the forces $-\partial \mathcal{H}_p/\partial r_i$ arise from the new, plasma state. Since the momenta before the hop are completely independent of what happens after the hop, these two quantities can be averaged separately, and we find, again, that the linear term vanishes; however, *note that this happens for completely different reasons that it did in the equilibrium case.*

This line of thought can be continued to higher orders in time, i.e., later in time [4]. At each order in time, we would find a similar situation in which the terms look similar but must be interpreted differently. In essence, for the ultrafast experiments of interest here, we must generalize each term to its nonequilibrium form. For example, at the next order, a mean force is introduced. In equilibrium, this is the mean force from the cage of neighbors that surrounds each particle in the liquid state, and gives rise to oscillations at the Einstein frequency. In the nonequilibrium UCP case, the cages have not yet formed, and the mean force now corresponds to the distributions of forces arising from configurations of the *atomic gas* [4]. In contrast, in nonthermal melting experiments, which begin in a lattice, the strong cages become too weak to continue to support a lattice structure.

6. One-dimensional model of disorder-induced heating: phase space behavior

We now consider a one-dimensional (1D) model of an ultracold plasma. The Hamiltonian of this system is

$$\mathcal{H} = \sum_{i=1}^{N} \frac{p_i^2}{2m} + \sum_{i<j}^{N} \frac{e^2}{x_{ij}} e^{-x_{ij}/\lambda}, \tag{24}$$

where $x_{ij} = x_i - x_j$ and λ is the screening length. At $t = 0$ we assume that all particles are at rest, $p_i = 0$, and that the positions x_i are distributed randomly. Molecular dynamics simulations have been used to explore the dynamics of this model.
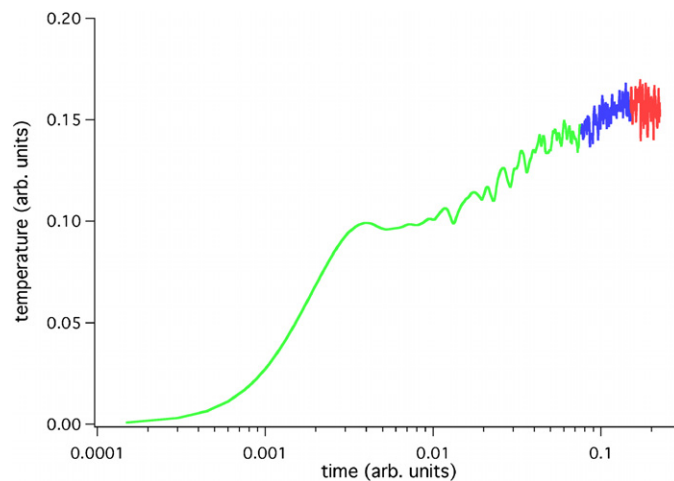
Figure 1. Temperature versus time for a model 1D UCP. Note that the usual DIH occurs, but as a two-stage process consisting of rapid heating followed by a slow rise to the final temperature.

6.1. Non-equilibrium molecular dynamics

Recognizing that experiments are the ultimate arbiter of the physics of UCPs, the most powerful tool for understanding UCP physics is molecular dynamics. Because of the interest in ultrafast processes, one utilizes non-equilibrium molecular dynamics (NEMD). The NEMD method has been well established for several decades [17], and the NEMD method used here is the same as in the previous work [1, 4]. Among the various issues that arise, an extremely small timestep is needed, far shorter than that used in equilibrium simulations by a few orders of magnitude; the actual timestep is determined on a case-by-case basis by monitoring overall convergence of the result and energy conservation. Equally important is the number of particles N needed in the simulation. Here, in contrast to previous work on nonequilibrium relaxation in dense plasmas [18], many thousands of particles are needed for convergence; again, many NEMD simulations are needed to empirically determine N, which is typically $N \sim 5000$.

6.2. DIH in one dimension

Let us begin by investigating DIH in a 1D system. NEMD simulations have been carried out for the Hamiltonian (24), and the effective temperature has been extracted; in these simulations, the average spacing and screening length have been adjusted so that their ratio is approximately the same as in real experiments (~ 0.3). An example is shown in figure 1, which displays the effective temperature versus time (on a logarithmic axis). Clearly, DIH occurs in 1D. However, there is a notable absence of strong oscillations after a first minimum, and a slow rise occurs until the final temperature is reached. The three colors shown are for the first, second and third portions of the simulation.

6.3. Single particle phase-space trajectories

Now, let us relate the global DIH process to the microscopic processes in phase space. In figure 2, four phase-space trajectories are shown from particles selected at random. The color scheme is the same as in figure 1. The most striking observation is that the underlying
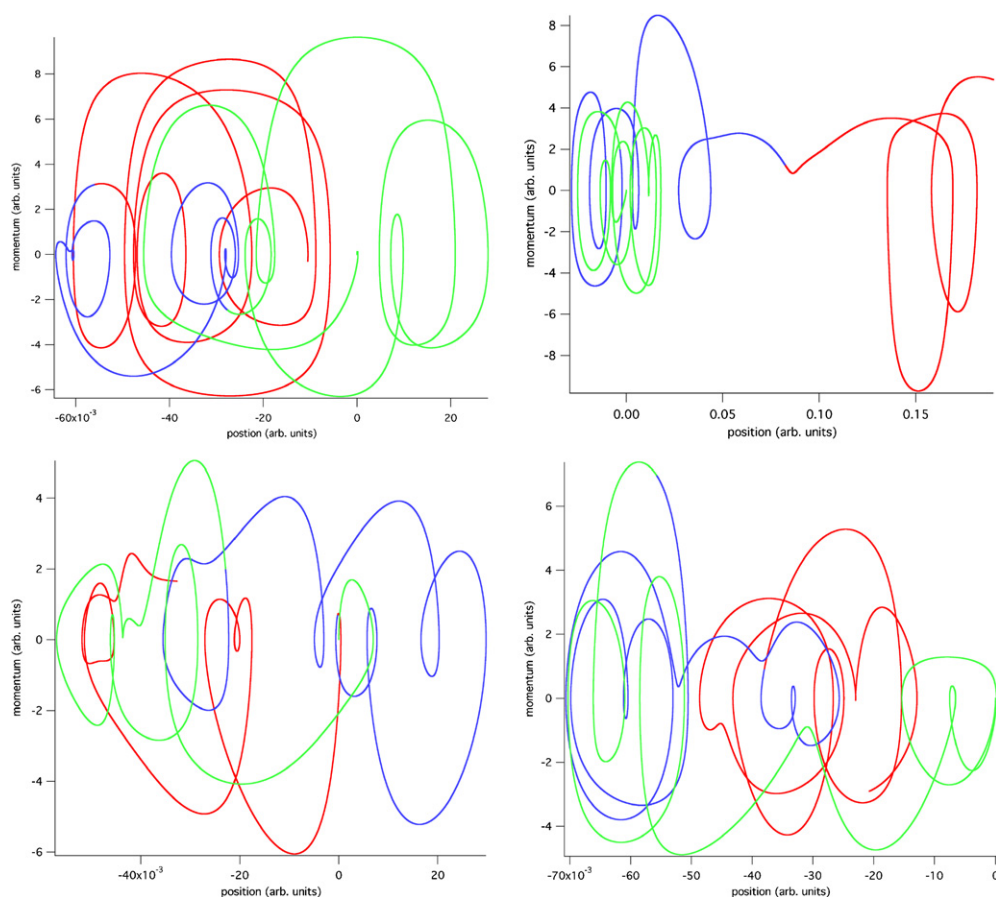
Figure 2. Phase-space trajectories for four particles chosen at random.

dynamics is much more complicated than the average behavior. In each case, there is high-frequency, harmonic-oscillator-like motion that occurs near a given location before the particle is transported to a new location where the motion continues. One property that is shared among all particles is that the final, average position is different from the initial position. This is particularly obvious in the upper right panel of figure 2. This is in contrast to the equilibrium oscillations that appear in the VACF, which arise from oscillations in a well-formed cage; for the nonequilibrium case, the cage itself is forming during the dynamics.

7. Discussion and outlook

UCPs are just one of many nonequilibrium systems that can be categorized as photo-initiated ultrafast systems. A short survey of related systems, including cluster explosion, non-thermal melting of solid surfaces and photoisomerization of molecules, has been given to place UCPs in a context broader than that of strongly coupled Coulomb systems. I have also argued that UCPs can provide a unique contribution to this nascent area of science because of their special properties.

Similar to non-thermally melted surfaces, the dynamics of an UCP can be thought of as the dynamics of a Coulomb liquid. The question arises in this context of how liquid-like the state is and, if not liquid-like, what the timescale is for quasi-equilibrium liquid-like dynamics to occur. By comparing the short-time dynamics of the temperature to that of the VACF, I have shown that the dynamics of the temperature evolution is quite different from that of the VACF, despite some interesting parallels. This analysis only highlights more clearly that the UCP dynamics cannot be reduced to a simple, known concept. A satisfying explanation of the temperature oscillations, and the timescale for their damping, is still lacking, and this will be addressed in future work.

To provide some intuition for the average dynamics usually considered, the phase-space trajectories of individual particles were also investigated using molecular dynamics. Very rapid motions were observed in the simulations that do not correspond to the observed quantities, suggesting that there are large fluctuations around the average behavior. Given the richness of this nonequilibrium physics, it is worth revisiting which quasi-thermodynamic phases [19] can be achieved with UCPs.

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