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Complex Non-equilibrium Dynamics in Plasmas

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We will illustrate the universality of strongly coupled plasmas by discussing two new forms of these plasmas, which have only recently become possible to create and observe in the laboratory. They exhibit a wealth of intriguing complex behavior, which can be studied experimentally, in many cases for the first time. Plasmas, gases of charged particles, are universal in the sense that certain properties of complex behavior only depend on ratios of characteristic parameters of the plasma, not on the parameters themselves. Therefore, it is of fundamental and far-reaching consequence, to be able to create and observe a strongly coupled plasma since its behavior is paradigmatic for an entire class of plasmas.

Introduction

We will illustrate the universality of strongly coupled plasmas by discussing ultracold plasmas[1] and laser-generated nanoplasmas.[2] They exist at temperatures that are about 10 orders of magnitudes apart and at densities that are more than 12 orders of magnitude different, yet they share essential features of complex non-equilibrium dynamics.[3,4]

Before we describe these plasmas in detail we briefly recall the prerequisites of complex behavior in the context of charged particle dynamics, i.e. a special form of non-linear systems.

Complex behavior in dynamical systems based on interacting particles

Massive charged particles, such as electrons or ions, interact through the Coulomb force, which is a two-body force, attractive for charges of opposite sign and repulsive for charges of the same sign. The result is a seemingly simple dynamical system, governed by Newtonian dynamics and classical equations of motion. These equations of motion are, in their form, identical to those that describe the dynamics of stars and planets due to gravitation. In this context it has been known for a very long time that the three-body problem (e.g. sun, moon and earth) is not analytically solvable and can even give rise to chaotic behavior.[5]

However, regular motion and purely chaotic behavior are both quite simple. Complex behavior emerges through an interplay of regular and chaotic dynamics, one can say complex systems live at the 'edge of chaos'.[6] Complex behavior for three particles does, loosely speaking, not occur in the following situations.

(a) If the motion of all three particles is mutually unbound, they will, if at all, approach each other and escape again to infinity after a single collision: this will not generate complex dynamics.
(b) If two particles are tightly bound to each other, relative to the third one, the three-body system effectively decouples into a two + one body system, which also shows simple behavior.
(c) As aforementioned, under certain circumstances three (or more) particles show completely chaotic behavior in the sense that the dependence on initial conditions does not play any role after a very short time of motion. This, so called, ergodic behavior is also simple: it is in its unpredictability reliably predictable. A good analogy is the most irrational of all irrational numbers, the golden mean. It is most irrational since its decimal representation has period one, i.e. one cannot say which digit follows if the previous one is known. On the other hand, in the representation of a chain fraction, the golden mean is the simplest number, given by $1/(1 + 1/(1 + 1/(1 + \ldots$

Hence, the parameters of the system must be such that (a)–(c) are avoided, i.e. that the three (or more) particles are confined to a certain volume and are forced to meet each other again and again, creating a history of collisions and eventually very complex behavior, without losing memory of the past, which happens in a billiard game. Here, a human analogy comes immediately to mind: complex relationships only develop through repeated encounters between persons who thus have memories of previous encounters.

Plasmas

Plasmas are a prototype of complex systems in physics regarding property (b) as described in the previous section. Innocently defined as a neutral gas of charged particles, they exhibit a wealth of phenomena, which require the cooperative if not collective motion of individual particles. The collective behavior known best is collective motion at specific frequencies, where the fundamental one, the plasma-frequency Ω, depends only on the density ρ, mass m and charge eZ of the particles,

$$\Omega = \sqrt{4\pi e^2 Z^2 \rho / m} \tag{1}$$

Other, so-called plasmon modes, refer, for example, to surface excitations and depend critically on the geometrical shape of the plasma.

Plasmas are abundant in the universe and therefore also called the fourth state of matter. The interior of Jupiter and other giant stars consists of a plasma so dense that quantum mechanical effects play a role. More familiar plasmas are the corona of the sun, and man-made, magnetically confined plasmas for fusion reactions. An overview in term of temperature (kinetic energy of the plasma) and density (potential energy) is given in Figure 1.

From the view point of complexity, most of the plasmas in Figure 1, however, are not so interesting since they fulfill property (c). Being similar to ideal gases, they lack – as an ensemble of particles – the relevance of memory regarding their collision history. In this limit of ideal gases, their Coulomb coupling parameter Γ is very small. It gives the ratio between potential and kinetic energy in the plasma, expressed through density (or inverse mean distance a between particles) and temperature T,

$$\Gamma = e^2 / (akT) \tag{2}$$

while k is the Boltzmann constant. On the other hand, strongly coupled plasmas are characterized by dominant potential energy, $\Gamma > 1$ and exhibit self-organization such as crystallization of the gas for $\Gamma > 174$,[7] since the charged particles tend to minimize their energy, which requires maximizing their mutual distances, as in a crystal, if potential energy dominates. One can see in Figure 1 that the few plasmas that are strongly coupled, such as the core of Jupiter, are experimentally difficult to access.

Most attempts to realize strongly coupled plasmas in the laboratory have focused on dense matter. With the advent of ultracold atomic physics, the possibility to cool atoms in a trap to temperatures where the gas condenses into a Bose-Einstein condensate – a new path to strongly coupled plasmas – has opened up: ultracold plasmas.[1] These plasmas are extremely dilute, with densities of less than 10^{10} particles per cubic centimeter. Created from laser cooled atoms through photo-ionization they are, however, so cold (in the micro-Kelvin regime for ions) that despite their diluteness with large inter-particle separations, potential energy wins over kinetic energy, allowing for $\Gamma > 1$, see the yellow bar in Figure 1. A significant advantage in terms of experimental controllability is the time and spatial scale of these plasmas: the fundamental plasma frequency (equation (1)) reveals that they live on a nanosecond/microsecond timescale for electrons/ions, respectively. Spatially, the plasma clouds have a size of 1/10 mm, predefined by the size of the traps. These scales are convenient to experiment with.

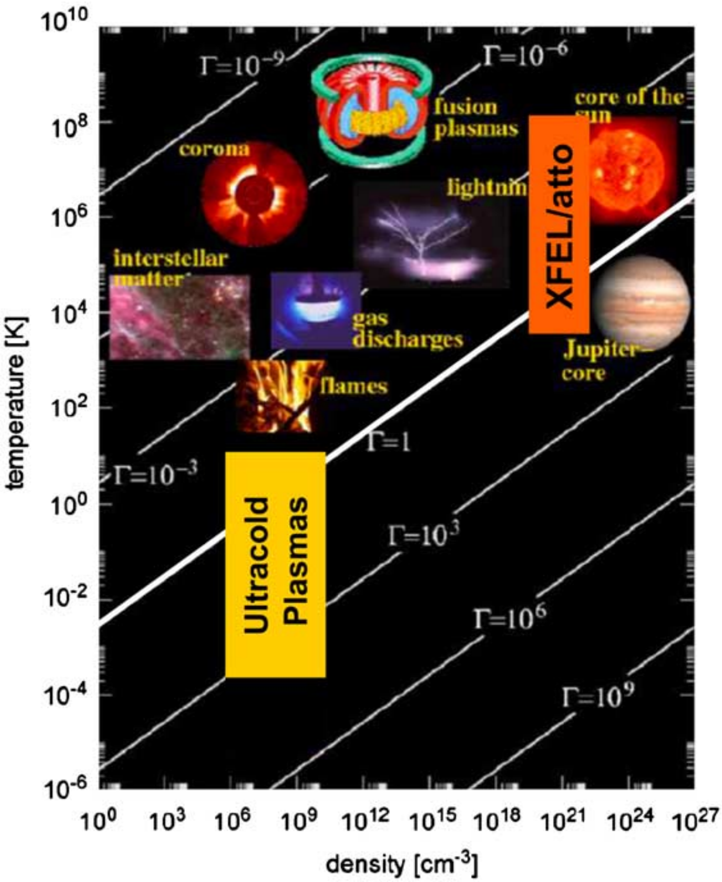

Figure 1. Natural and man-made plasmas in the density-temperature plane (after Ref. 1). Lines of equal Coulomb coupling parameter Γ (see text) are given in white, the thick white line corresponds to $\Gamma = 1$, the dividing line between ideal plasmas ($\Gamma << 1$) and strongly coupled plasmas ($\Gamma >> 1$). Parameter ranges of the plasmas discussed here, are indicated by the yellow and orange bars for ultracold and laser pulse generated plasmas, respectively

Much more difficult to observe in the experiment due to the almost solid state density is the second kind of plasma we are interested in: laser generated nanoplasmas. They live on a scale of 1 femtosecond (10^{-15} s) and have extensions in the nanometer to sub-nanometer range, see the orange bar in Figure 1.

Yet, recent advances in technology (attosecond (10^{-18} s) laser pulses) will make it easier in the future also to observe these strongly coupled plasmas. While living in very different ranges of space and time, laser-generated plasmas share one property with ultracold plasmas that is an important prerequisite for complex behavior: specific geometric shapes due to confinement, either in a trap or due to the form of the cluster, from which the nanoplasma forms.

This property of finite extension is important for complex behavior, as follows from conditions (a) and (c) above.

Ultracold plasmas

Plasma creation

The creation of an ultracold plasma is sketched in Figure 2. First, a gas of atoms is cooled in a magneto-optical trap. Next, the gas is excited to high Rydberg states or gently ionized just above threshold. As a consequence, electrons leave the gas and quickly a positive net background charge of ions forms (the ions are so cold that they stand still on this time scale) and, eventually, the ionized electrons cannot leave the gas anymore but form a plasma with very low kinetic energy. At the same time, the ions also form a plasma whose Coulomb coupling parameter is typically much higher due to the lower temperature of the ion plasma. Formally, ionic Coulomb coupling parameters up to 10,000 are possible. This is too good to be true, and indeed, it does not happen. In fact, the laws of

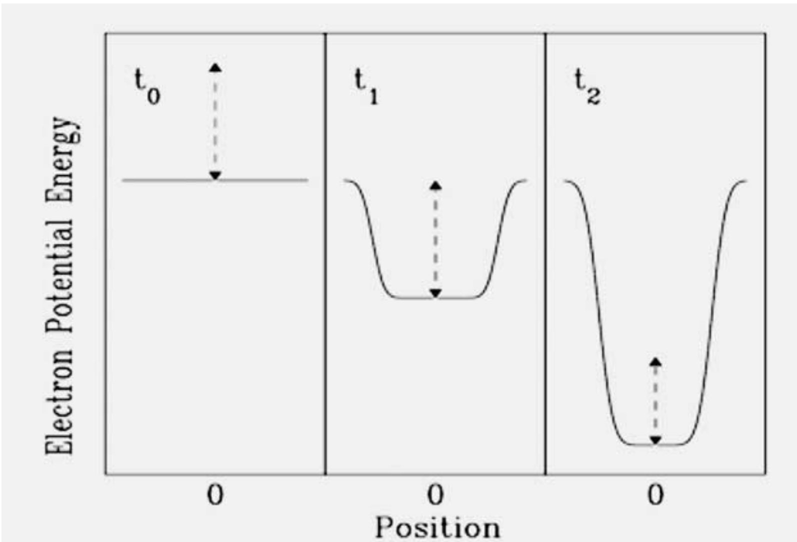

Figure 2. Sketch of plasma formation by photo ionization of a geometrically confined gas (after Ref. 8). The left panel shows the neutral gas in the beginning at time t_0. Electrons ionized acquire a kinetic energy E which is the difference between the photon energy ω and the atomic binding energy I of the electron, $E = \omega - I$, indicated by the vertical dashed lines. The center panel captures the onset of plasma formation: due to multiple ionization, a positive background potential of ions has formed which attracts the electrons. It is deep enough that the kinetic energy of a further electron ionized (indicated by an arrow) is not sufficient to escape from the cluster. Right panel: photo ionization of electrons into the plasma

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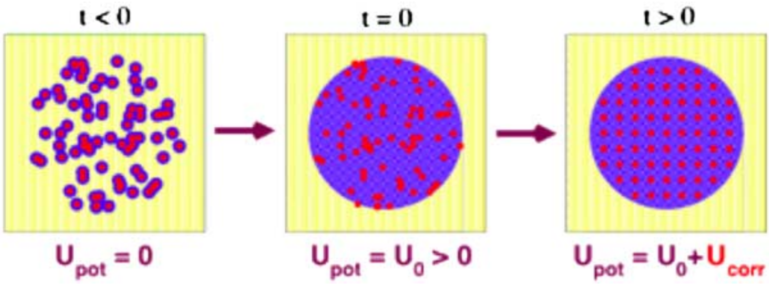

Figure 3. A gas with randomly placed atoms (left panel, $t < 0$) is ionized at time $t = 0$. As a consequence, there is potential energy $E_{pot} = U_0$ between the ions. Subsequently, correlations develop by which the potential energy is lowered, the generated positive correlation energy U_{corr} appears as kinetic energy (higher temperature) of the ions ($t > 0$)

thermodynamics teach us to expect, due to equi-partitioning of energy in the various degrees of freedom, $<E_{pot}> \approx <E_{kin}>$ and therefore $\Gamma = <E_{pot}>/<E_{kin}> \approx 1$. This is indeed the case but now the question arises, from where do the initially very cold ions $<E_{kin}> \approx 0$ acquire so much energy? The reason is disorder-induced heating sketched in Figure 3. The positions of the neutral atoms in the gas are random with a mean distance a given by the density, $a \propto \rho^{-1/3}$. After ionization the potential energy of the now ions is much higher than in an ordered, crystal like, state. Hence, through collisions, potential energy is converted into kinetic energy leading to $\Gamma \approx 1$. The process is quite violent and violates all laws of equilibrium thermodynamics rendering it very interesting but beyond the scope of our present considerations. Due to its large size ($\approx 100\ \mu$m) and slow dynamics ($\approx 1\ \mu$s) ionic ultracold plasmas can be relatively easily monitored, as described in Ref. 1.

The pertinent question, however, is: can one realize a strongly coupled ultracold plasma despite the disappointing heating effect? In principle, one could try to drive both components, the electronic and the ionic one, to the strong coupling regime. However, since they evolve on a slower and experimentally therefore easier accessible time scale, we concentrate on the ions. Our proposal, which we have underlined by calculations, consists of additional laser cooling for the ions via an optical transition.

Cooling to the strong coupling regime

Additional cooling of the ions in the plasma can be achieved for strontium, where the ion still has one optically active electron, while the first electron has been fed into the plasma. One question remains, which only the calculation or the experiment can answer: is there enough time after the creation of the ions to cool them down? The cooling competes with the slow, but steady expansion of the plasma, which is no longer held in the trap. Cooling can be modeled quite reliably with a stochastic process,[9] and the first promising observation is that the expansion of the ions under cooling slows down, not only quantitatively, but also qualitatively. While the mean width σ of the initially Gaussian-shaped cloud of the plasma in the trap (which remains Gaussian through a self-similar expansion process) grows quadratically without couling $\sigma \propto t^2$, this changes to a long-time square-root dependence under cooling, $\sigma_{\text{cool}} \propto (t/\tau)^{1/2}$. Very important for the experimental feasibility is the time scale $\tau = \beta m \sigma^2 /(kT_e)$. It can be controlled by the cooling rate β, the mass of the ion species m and the initial width σ and electronic temperature T_e of the plasma. Increasing any but the last parameter increases τ and therefore favors crystallization. For better cooling with larger β this is obvious, a larger mass also makes the system expand more slowly, and so does a larger initial size (at the same density) since it implies reduced repulsion of the ions, which slows down the Coulomb explosion. Only increasing the initial electron temperature T_e drives the ions more easily apart and should therefore be avoided.

Self-organized crystallization during expansion

What cooling achieves is to increase the Coulomb coupling parameter Γ, which should lead to a structure in the disordered gas of ions. Indeed, for sufficiently large τ the ions organize themselves on concentric shells with no ions in between, i.e. they start to form an ion crystal during the expansion of the ion gas – without any external force or geometric boundary, see Figure 4.

This crystallization is still a prediction, which awaits experimental verification but faces two difficulties: first, sufficient cooling, second, a detection scheme, which can prove the crystallization. This is indeed somewhat more tricky.

However, another signature of complex behavior, the temporal evolution of the Coulomb coupling parameter, has already been measured and we will come back to it later.

Plasmas generated by short laser pulses

Plasma creation

This type of plasma lives from the efficient deposition of energy to material in a very short time. Electrons absorb an enormous amount of energy from a very intense laser pulse, which lasts typically from a couple to some 100 fs, and delivers 10^{14}–10^{18} W/cm^2 power with a pulse profile in time that is close to Gaussian. If the pulse is ramped up fast enough, electrons are ionized almost simultaneously, but certainly on the timescale of the respective plasma of a material of density 10^{22} cm^{-3}. This timescale is set by the electronic plasma frequency, which is now of the order of a femtosecond. The process of creating a

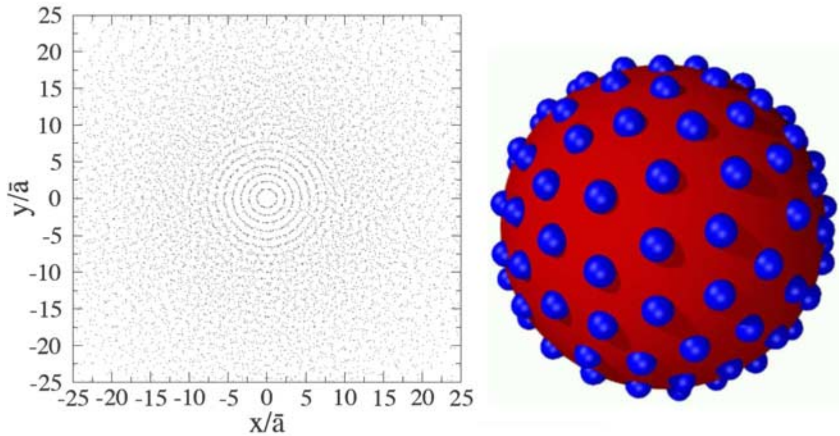

Figure 4. Self-organization of a plasma of 80000 Beryllium ions after expansion time $t = 216/\Omega$ into a crystal upon cooling with a rate of $\beta = 0.2\,\Omega$. The initial electronic Coulomb coupling parameter was $\Gamma_e = 0.08$ (after Ref. 10). Shown on the left is a cut through the center of the plasma, with the length scaled by the local mean distance a of ions. Distributions for several orientations have been superimposed for clarity of the structure. On the right the fourth shell of ions is shown (fourth circle from the center in the left figure)

plasma is very similar to the ultracold plasma (see Figure 2), only much faster: while the first ionized electrons can leave, a positive background charge forms, which further electrons cannot escape from since they do not have sufficient kinetic energy. When this happens depends mostly on the laser frequency ω, which determines – through the energy of a single photon transferred to an electron bound by energy I – how much kinetic energy E is left to escape from the cluster with a (time-dependent) background potential $V(t)$. Plasma formation sets in when $E + V(t) = 0$.

We restrict ourselves here to clusters, i.e. droplets of atoms, which can exist from extremely small sizes (a few atoms) to almost macroscopic sizes (micrometer diameter). The relevant property in terms of complex behavior is that they provide a geometric shape and well-defined confinement for the plasma (see Figure 5), and it is important for particles to undergo repeated encounters and thereby not to lose memory of them (see conditions (b) and (c)). Clearly, this confinement is only of transient character since, after multiple ionization, a cluster with repulsive ions will eventually undergo Coulomb explosion. On the plasma timescale, the cluster holds together more than long enough for the complex plasma dynamics to develop. Much more problematic is the detection of the plasma and of its evolution in time, since all this happens on a femtosecond scale.

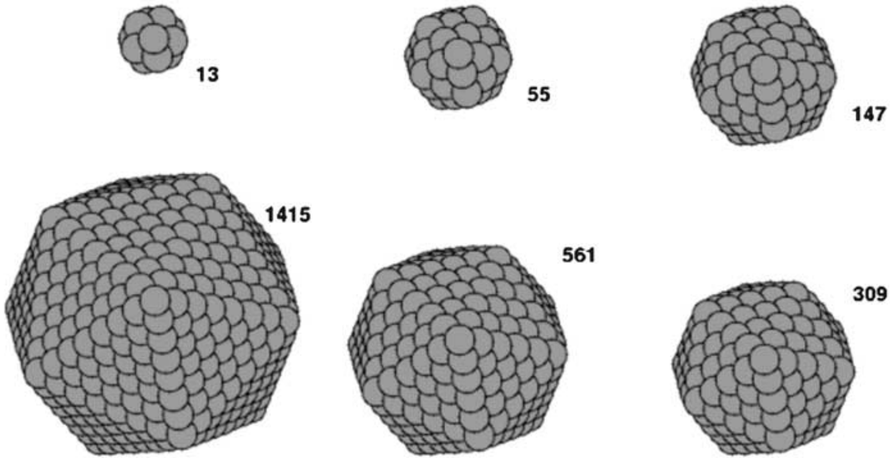

Figure 5. Closed shell configurations of rare gas clusters form so called isocaeders and are particularly stable. They contain the number of atoms as indicated

Detection of ultrafast plasma dynamics

We will discuss the scenario of a possible detection of the ultrafast plasma dynamics in the context of laser pulses from unconventional light sources. One such source is a free electron laser (FEL) at soft X-ray and soon hard X-ray frequencies. It is expected to deliver light pulses with frequencies of $\omega = 12\,\text{eV}$–$12{,}000\,\text{eV}$ over less than 50 fs and with unprecedented intensities of more than $10^{20}\,\text{W/cm}^2$. Light pulses with such fantastic properties have their price: they are generated by first accelerating electrons to almost the speed of light over 2 km in a tunnel, then sending the electrons through a series of magnets to force them into bent trajectories. Thereby, they radiate and organize themselves in bunches, leading overall to a dramatic coherent amplification of the emitted light over a certain, narrow frequency range. This principle is called SASE (Self-Amplified Stimulated Emission) and machines working with it are built at DESY in Hamburg and at SLAC in Stanford.

The second new development is attosecond pulses ($1\,\text{as} = 10^{-18}\,\text{s}$) whose duration comes close to the period of about 100 as an electron needs, in the ground state of hydrogen, to travel around the proton and whose photon energy is between 20–150 eV. These pulses are generated from certain fractions of intense longer laser pulses at longer wavelengths and, consequently, their intensity is much lower than the light from FELs. More details can be found in Ref. 11.

Not only is the detection itself difficult, but the monitoring of the plasma must be synchronized with its creation; in other words, one needs to know when the clock started. These set-ups are called pump-probe experiments: the first ‘pump’ pulse creates the dynamics one is interested in while the second ‘probe’ pulse, probes the dynamics at a well-defined time delay Δt.

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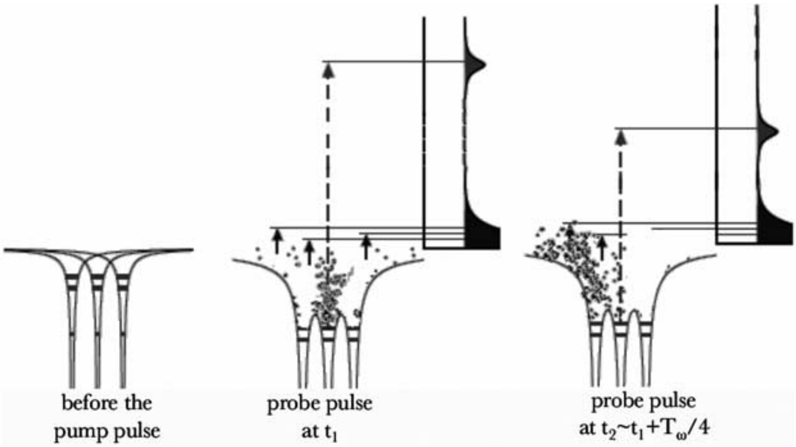

Figure 6. Sketch of a double-pulse pump-probe measurement of electronic plasma dynamics, for details, see text

A possible scenario is sketched in Figure 6. Before the first pulse hits (left panel of Figure 6), all electrons are bound to their mother atoms (the electronic levels are indicated by dark/red horizontal bars in the individual atomic potentials). The pump pulse removes the upper electrons in the atoms, and the potential barriers between the ions are bent down. Some of the electrons remain in the cluster and form a plasma (small dots in the middle panel of electrons Figure 6). A fraction of the distribution of plasma electrons has kinetic energy high enough to be able to escape from the cluster – they are measured as a continuous distribution (dark shaded) in energy. If the probe pulse is applied at a time t_1 and the plasma electrons (which are assumed here to move collectively in a kind of a wave) are in the center of the cluster, then electrons ionized by the probe pulse from the dark/red (core) levels in the atoms are taken to the continuum (dashed arrow) and they are measured at a certain energy (light shaded peak). If the probe pulse arrives when the plasma cloud has passed the center of the cluster, then the electrons it ionizes appear at lower energy in the continuum (light shaded peak in the right panel of Figure 6). The reason for the lower energy is that at time t_1 the ionized electrons have to escape from smaller positive background charge since the ions are screened through the plasma electrons. This is not the case at time t_2, and therefore the ionized electrons lose more energy on the way to the detector, which becomes visible there.

Hence, it is possible to observe indirectly, by ionization of core electrons at different probe times, the oscillation of the plasma electrons as an oscillation of the kinetic energy of the ionized core electrons, as shown in Figure 7. The oscillation of the plasma electrons has a period of below 1 fs and can only be observed by laser pulses, which are significantly shorter, in our simulation the pump and probe pulses last for 0.25 fs or 250 as.

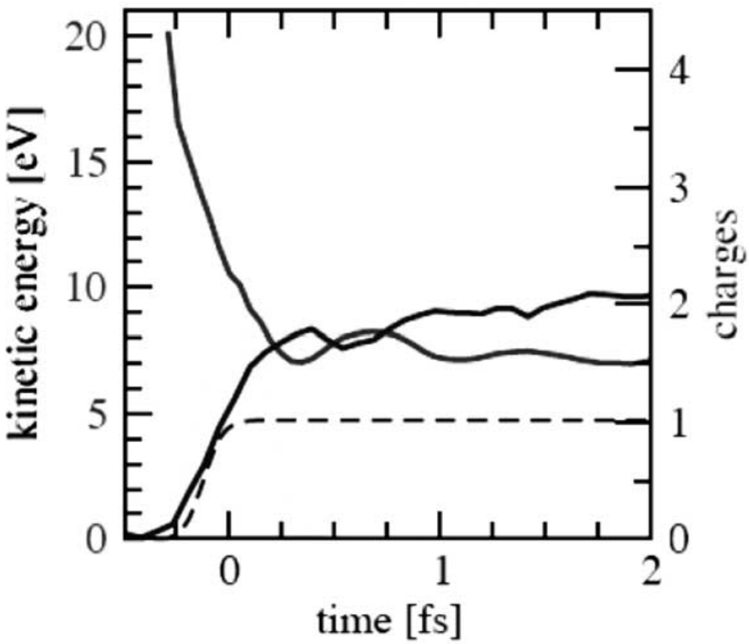

Figure 7. Oscillation of the charges (black/blue, right axis) probed as in Figure 6 with a pulse of variable time delay against the pump pulse (shown shaded, centered at time $t = 0$), which triggers the plasma dynamics in a cluster of 55 argon atoms (after Ref. 12). The probed charges indirectly map out the oscillation of the plasma electrons as is evident by the average kinetic energy of the plasma electrons (grey, left axis), oscillating out of phase but with the same frequency. Also shown is the average charge state of the ions (dashed) which stays constant after the laser pulse is over. The two laser pulses have a photon energy of $\omega = 20\,\text{eV}$, peak intensity of $5 \times 10^{14}\,\text{W/cm}^2$ and a duration of 250 attoseconds

Non-equilibrium plasma dynamics

The oscillation of the plasma electrons as described in the previous section is a quite complex collective response of a strongly coupled plasma to a sudden perturbation: the system is driven out of equilibrium by the initial ionization and creation of the plasma within the 250 as of the pump pulse. This is too fast for the system to adopt smoothly to the new situation. Rather, it ‘overreacts’, which manifests itself in the oscillation of the plasma. This oscillation damps out as the systems reaches equilibrium, but also due to loss processes.

Why is this oscillation a signature of complex behavior? It is organized, in the sense that the electrons move with a joint period, yet it is accidental since the electrons are not required to have a specific initial condition for their motion, which is strongly influenced by collisions.

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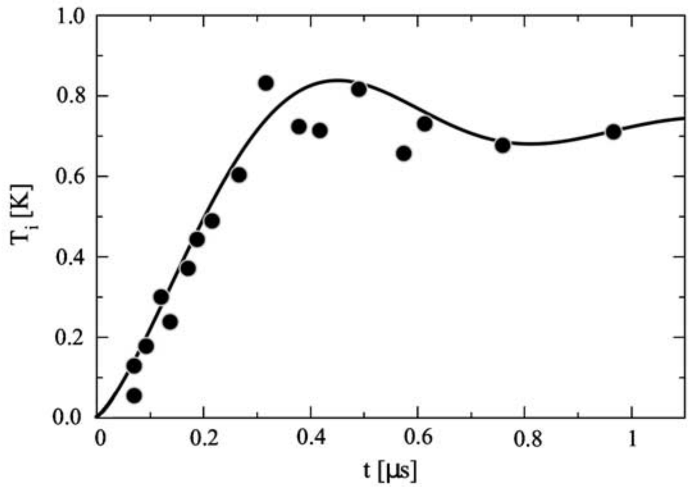

Figure 8. Calculated[3] (line) and measured[13] (points) ion temperature of an expanding plasma of 1million strontium ions with initial density $\rho = 2 \times 10^9/\mathrm{cm}^3$ and initial electron temperature of $T_e = 38\,\mathrm{K}$

Even more fascinating, at least for a physicist, is the fact that this behavior is universal: it occurs in the same way for the ultracold plasma we have discussed previously (Figure 7), but on a completely different, in comparison, extremely slow time scale of microseconds, see Figure 8. Why is the ultracold plasma driven out of equilibrium by an excitation as slow as nanoseconds? Simply because its response time scale due to the diluteness of the ions is microseconds, and on this time scale ionization and onset of plasma formation over a nanosecond is quite abrupt. For the temperature, which is a measure for the kinetic energy of the (in this case ionic) plasma, we observe exactly the same damped oscillatory behavior in time as in the case of the cluster.

Hence, making use of the universality of the strong coupling phenomenon with complex collective behavior, depending only on the Coulomb coupling parameter being larger than unity, one can relatively easily see this behavior in the ultracold plasma with its slow time scale compared with the laser-generated plasma where the dynamics happen in complete analogy but extremely fast. However, even this fast motion will be accessible experimentally in the near future.

Summary

We have discussed universal complex behavior in two forms of microscopic plasmas, i.e. gases of charged particles. This complex behavior manifests itself in an organized collective motion of the plasma particles. The motion can happen on extremely different time scales; in our case, the time scales are ten orders of

magnitude apart. Complexity is present as, for both these dynamics, elements of order and chaos are necessary in order to make it happen. This is a signature of complex systems, which live on the boundary between regular and chaotic motion – colloquially speaking, near the 'edge' of chaos.

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