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Topical: Hunting for the elusive glassy state of complex plasmas

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Hunting for the elusive glassy state of complex plasmas

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Motivation: There are numerous unresolved issues concerning supercooled liquids and glasses, even of fundamental theoretical nature. Recent experimental and computational advances have generated rich information about these metastable and non-equilibrium states. However, there is still no first principle theoretical framework capable of explaining all observations, although the possible candidates are many. Experiments have demonstrated that soft matter systems have very similar metastable and glassy characteristics to molecular systems (dynamic heterogeneity, growth of microscopic relaxation time scales, dramatic viscosity growth, ageing behavior, etc). Hence, considering also the Newtonian nature of dynamics and the long-range pair interactions, complex plasma glasses qualify as unique model systems for the study of the glass transition. In what follows, we shall briefly review earlier work in metastable complex plasmas, discuss the theoretical input required for successful realization of complex plasma glasses and describe a simple experimental protocol tailor-made for the **com**plex **pla**sma fa**c**ili**t**y (COMPACT) on the International Space Station (ISS).

Metastable liquids and glasses: Quenching of liquids by means of rapid cooling or compression in a manner that prevents crystallization leads to a substantial slowdown in dynamics as well as a remarkable increase in their viscosity without a significant change in structure [Debenedetti and Stillinger 2001]. Beyond a point, a dramatic dynamic arrest intervenes so that it is no longer possible to equilibrate the system within reasonable experimental times; the metastable liquid thus transforms into a non-equilibrium glass [Debenedetti 1996]. Glasses are as mechanically rigid as crystals, but are simultaneously isotropic and lack long-range order similar to liquids [Ediger 2000, Dyre 2006]. Metastable liquids and glasses have a rich phenomenology, whose microscopic origin is far from being completely understood in spite of intensive research in the last two decades, as evidenced by the large number of theoretical approaches available [Donth 2001]. Among the key questions is whether the liquid glass transition is linked to an underlying (avoided) thermodynamic phase transition or whether the vitrification process is predominantly dynamical [Berthier and Biroli 2011, Turci et al. 2017]. Given their accessible spatiotemporal scales compared to molecular systems, soft matter systems have played a crucial role in the study of slow dynamics [Pusey and van Megen 1987, Hunter and Weeks 2012]. Hence, the addition of complex plasma glasses to the short list of colloidal glasses would be invaluable, especially given their softer interactions and virtually undamped atomistic motion [Morfill and Ivlev 2009].

Complex plasmas: The discovery of plasma crystals signaled the birth of the complex plasma field [Thomas et al. 1994]. The experimental confirmation that complex plasmas can be modelled as point particles whose interaction is described by a screened Coulomb (Yukawa) pair potential [Konopka et al. 1997] enabled theoretical as well as computational advances with established statistical mechanics methods in parallel to the experimental developments. A large number of investigations were dedicated to the phase behavior, thermodynamic and structural properties, dynamic responses, collective modes and transport properties of complex plasmas [Fortov et al. 2005, Donko et al 2008, Bonitz et al 2010, Khrapak 2016]. These investigations focused on 2D systems relevant for ground-based experiments and 3D systems relevant for microgravity, extended bulk systems and finite size systems, externally forced and equilibrium systems, crystals and liquids. The experimental effort was spear-headed by the microgravity experiments performed on the International Space Station (PKE-Nefedov, PK-3 Plus and PK-4) [Nefedov et al. 2003, Thomas et al. 2008, Pustylnik et al. 2016]. Despite the remarkable progress, the glassy state of complex plasmas has remained elusive experimentally and few observations of metastable complex plasmas have been reported (all in 2D). This is partly due to the well-known difficulties in the experimental realization of metastable states, but also due to the incomplete theoretical understanding of stable / metastable Yukawa mixtures.

Binary complex plasmas: Since experiments nearly exclusively employed monodisperse dust particles, most theoretical studies focused on one-component Yukawa systems with much less attention paid to polydisperse systems. Nevertheless, nowadays, binary Yukawa mixtures are considered to be a frontier topic in complex plasma research [Block and Meltzer 2019]. As aforementioned, 2D metastable complex plasmas have been already engineered in ground-based experiments [Su et al. 2012, Du et al. 2019]. This was achieved by: **(a)** introducing dust size deviation to suppress crystallization (either continuous polydispersity or binary individually monodisperse systems), **(b)** rapidly dropping the discharge power to promptly quench the system. It should be pointed out that size disparity translates to deviations in sheath levitation heights, thus these are quasi-2D systems. In the 2D case, the glass transition line has been estimated for one-component systems based on mode coupling theory with structural input from a scaled-hypernetted chain approach [Yazdi et al. 2015] (an approximation later revealed to be inaccurate [Lucco Castello and Tolias 2021a]) and dynamics have been studied with Langevin Dynamics simulations [Lin et al. 2018]. In the 3D case, the glass transition line has been estimated for one-component systems based on mode coupling theory with structural input from the standard hypernetted chain approach [Yazdi et al. 2015] as well as from the more accurate variational modified and the isomorph-based empirically modified hypernetted chain approaches [Lucco Castello and Tolias 2021b]. For completeness, we mention a number of pioneering binary complex plasma experiments performed under microgravity conditions that were dedicated to lane formation [Du et al. 2012], phase separation [Killer et al. 2016] and interfacial phenomena [Yang et al. 2017].

Theoretical gaps: Suppression of crystallization in glass-forming liquids has been attributed to geometric frustration characterized by the competition between a short-range tendency for the extension of long-lived locally preferred structures and global constraints that prevent periodic tiling of Euclidean space with such a motif. The most straightforward way to promote this competition is by introducing size disparity in the constituents of the physical system, as known from binary alloy glasses, colloidal glasses and computer simulations of model glasses. In order to reduce the complexity, it would be preferable to avoid polydisperse dust particles and focus on binary complex plasma mixtures. One-component Yukawa systems are characterized by two dimensionless thermodynamic parameters; the coupling parameter Γ that is the ratio of the unscreened potential energy at the mean interparticle distance over the thermal energy and the screening parameter κ that is the ratio of the mean interparticle distance over the screening length. Binary Yukawa mixtures are characterized (apart from Γ, κ now defined for the denser species) also by the external parameter σ_R that is the dust size ratio (which translates to a charge ratio within the validity of the orbit motion limited approach) and the thermodynamic parameter χ that denotes the concentration ratio. This leads to four control parameters of the state point. In order to increase the chances of the experimental realization of complex plasma glasses, the following characteristics need to be available:

- an accurate description of the structural and the thermodynamic properties of both stable and metastable Yukawa liquid mixtures that can be acquired by employing an advanced integral equation theory approach,

- an accurate estimate of the liquid glass transition line that can be acquired by utilizing mode coupling theory (conventional or generalized), as extended for multi-component systems,
- an accurate estimate of the spinodal line that can be acquired on the basis of advanced integral equation theories, *i.e.* beyond the random phase approximation,
- an accurate estimate of the crystallization line that can be acquired by utilizing two-phase Molecular Dynamics simulations for a restricted number of state points and by expanding the results with the aid of isomorph theory [Pedersen 2016],
- an empirical determination of the state points that are more robust against crystallization with the aid of specially designed Monte Carlo simulations.

Currently, none of the above input is available for binary Yukawa mixtures. In particular, even simpler input such as an equation of state has not yet been accurately determined. Considering the three thermodynamic parameters and one external parameter of the binary Yukawa system, these tasks are far from trivial but feasible.

Experimental protocol: The above theoretical input will lead to a selection of a specific (χ, σ_R) combination that is most likely to be robust against crystallization and de-mixing. Then, the (Γ, κ_1) pre-quenching state will be selected to be close to the crystallization line and the targeted $(\Gamma, \kappa_2 < \kappa_1)$ post-quenching state will be selected to lie beyond the glass transition line. The quenching will be realized by rapidly dropping the discharge power. At low pressures, the dust charge can be considered to be independent of the plasma density, while the dust kinetic temperature still remains collisionally locked to the neutral temperature. Hence, the coupling parameter should remain nearly constant during the quenching, while the screening parameter is decreased due to the increase in the plasma screening length. Since the quenching occurs on the much faster plasma time-scales, it can be considered to be nearly instantaneous for dust and it corresponds to a horizontal transition in the Γ-κ phase diagram that is completely analogous to rapid cooling or compression in the n-T diagram, see the sketch of figure 1.

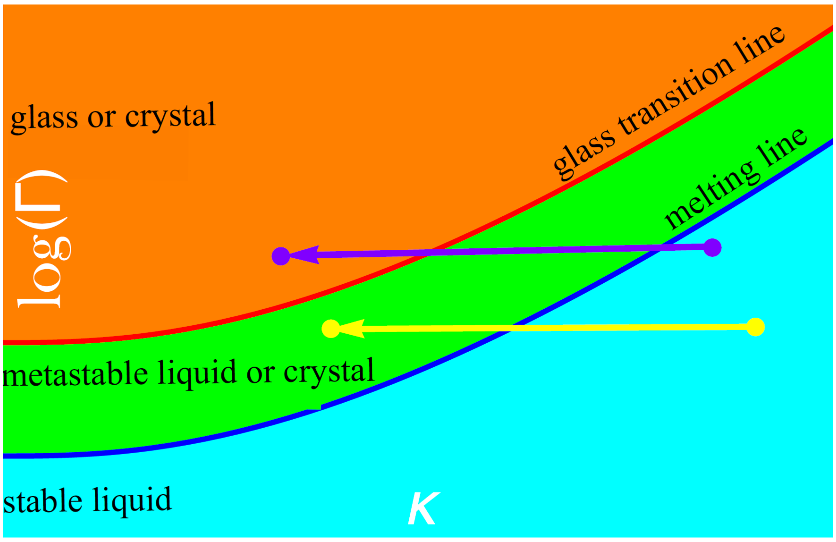

Figure: *Sketch of instantaneous phase diagram* $log[\Gamma], \kappa$ *jump of the binary complex plasma during rapid decrease of discharge power (given* χ, σ_R*). The liquid-solid phase transition curve will be computed by two-phase MD simulations and the liquid-glass transition curve by mode coupling theory with input from integral equation theory approaches. The "purple transition" will lead to glassy state observations and the "yellow transition" to metastable liquid state observations. The screening length controlled quenching in the* $\Gamma - \kappa$ *phase diagram is analogous to supercooling in* $n - T$ *phase diagram.*

Experimental device: There are two very important requirements for a successful experimental realization. *First,* the pre-quenching state should be very close to thermodynamic equilibrium.

Second, the sudden discharge power drop should not lead to localized flows that compromise homogeneity and equilibrium. Both requirements are satisfied in COMPACT, the next generation complex plasma laboratory for the ISS, which constitutes the most promising set-up for the realization of the glassy state of plasmas. COMPACT is a capacitively coupled rf plasma discharge that features a multi-segmented electrode system with a high degree of user control in plasma generation; it is this flexible plasma manipulation system that leads to the satisfaction of both requirements. In addition, COMPACT's flexible broad performance over a wide range of plasma conditions provides freedom concerning the quenching depth. Moreover, its advanced stereoscopic particle diagnostics imply that numerous physical quantities can be accurately extracted in the metastable and the glassy state. Finally, its ability to produce an extended 3D homogeneous plasma ensures a large observation volume, a diminished role of the system size and negligible effect of the boundary. Naturally, extended-in-time microgravity conditions are essential for the realization of the experiments due to the long observation times.

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