

## Featuring soft-matter for information storage applications

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*Among the direct consequences of the verification of a crystal-to-glass transition in 2D interacting particles is that heat pulses can switch patterns, suggesting the design of bit-writing protocols for monodisperse systems.*

Not very often the time comes for a theoretical physics project to be able to conduct a proof of concept, i.e. a demonstration in principle with the aim of verifying that a concept or theory has practical potential. That's usually a privilege of the experimental realm. But right away after we understood the out-of-equilibrium properties of 2D cluster-forming systems, in a joint effort involving a fairly large collaboration with researchers from Strasbourg, Innsbruck, Stellenbosch and Stockholm [1], it became clear that the steep change in the order parameters, allowing for temperature-controlled switch applications, was one of these happy opportunities.

This is actually a long story, so in order to make it short let's go quickly through the basics: in two dimensions, interacting systems in which particles are all of the same type (monodisperse) have a very contrasting ordering behavior when compared to systems with different types of particles (polydisperse). Essentially, monodisperse systems are hard to keep in a disordered state, while polydisperse cannot even order in the general case. That's a killing property for memory applications based on phase-change, i.e. materials with the ability to switch between amorphous and ordered configurations, instrumental for rewritable non-volatile-electronic or optical storage. Not surprisingly, devices for phase-change memory, which is a promising candidate for both the next generation of high-density ultra-fast memories and the emergence of cognitive computing hardware, are typically made of bulky alloys. The search, however, continues for 2D and quasi-2D materials, since they could improve density packaging, low power, etc.

Then it turned out that, while studying the apparently distant context of cold atomic systems with quantum supersolid properties, the concept of cluster-forming ability (the property of spontaneously forming small bunches of particles) arose as crucial feature for the bosonic potential interactions. As a result, a thorough set of computer simulations was performed to characterize, among other things, the evolution of the non-equilibrium dynamics of these cluster configurations after quenches. The study of cluster-forming potentials, also called ultra-soft in some communities, was an already established field of research, remarkably connecting a large number of soft-matter topics of diverse nature, from cold atoms and vortexes in type-1.5 superconductors to colloids and polymers. Consequently, the finding of stable amorphous states

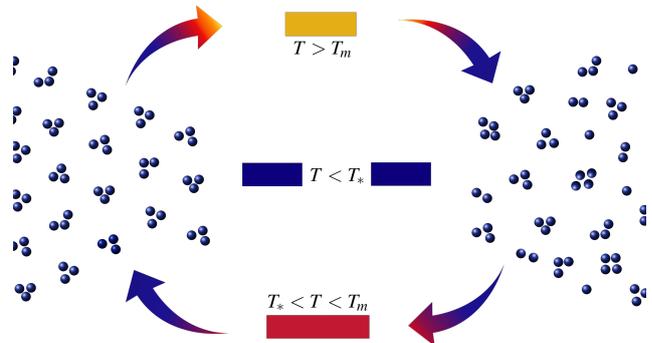


Figure 1: Schematic representation of the phase-change switching for a particle system interacting via cluster-forming potentials. The system is kept at a temperature  $T < T_*$  (blue reservoir), a switch from the crystalline to the amorphous phase is obtained by heating the system to a temperature corresponding to the disordered phase  $T > T_m$  (yellow reservoir), followed by a sudden cooling to the initial temperature. In turn, the ordered configuration can be reestablished by a moderate heating to a temperature  $T_* < T < T_m$  (red reservoir).

that do not relax towards equilibrium after quenches in a vortex-matter model with ultrasoft interactions [1], was immediately generalizable to all cluster forming potentials.

In a nutshell, the trick is that clusters form quickly, in the very early stage of the evolution after quench, and then a slower cluster dynamics sets on, in which the whole system rearranges and, most importantly, individual particles hop between neighboring clusters. But the latter is an activated process, meaning that there will be a well-defined freezing temperature  $T_*$  below which individual particles have not enough energy to overcome the hopping barrier, forcing each cluster to keep a constant size. In turn, this random distribution of sizes act as a self-assembled effective polydispersity for the clusters ensemble, making the clustered system to depart from the normal monodisperse behavior, unable to go beyond the amorphous state and eventually arresting the dynamics<sup>b</sup>.

In Fig. 1, a simple schematic representation of a bit writing protocol is illustrated, on the basis of this now possible stabilization of both, amorphous and ordered states depending on the temperature ramps. The system operates at a temperature below  $T_*$ . To go from crystalline to amorphous state, the system must be temporarily heated above the melting temperature of the crystal  $T > T_m$  so that particles become fluid and

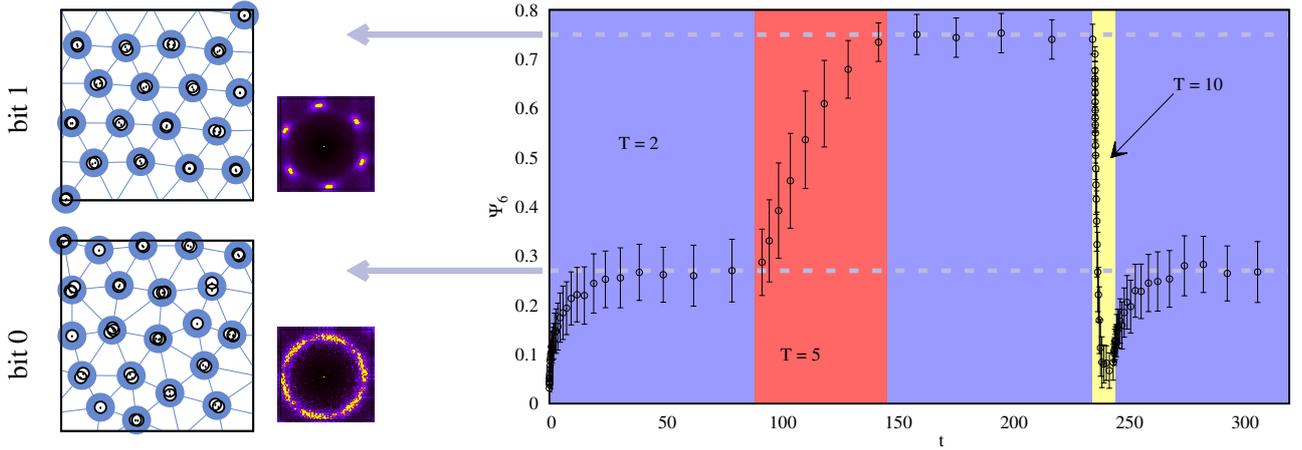


Figure 2: Simulation of the bit-writing operation in a system interacting via an ultrasoft potential with  $T_m = 8$  and  $T_* = 4$ . The order parameter  $\Psi_6$  is plotted as a function of time, and the temperature of the reservoirs along the time line is encoded with blue ( $T = 2$ ), red ( $T = 5$ ) and yellow ( $T = 10$ ) shadows. The system is quenched at  $t = 0$  from a fully disordered state to the operating temperature  $T = 2$ , and is quickly trapped in the amorphous state (bit 0), corresponding to an order parameter of about  $\Psi_6 \approx 0.27$ . A first pulse with  $T = 5$  orders the system in a crystalline configuration (bit 1), corresponding to a parameter of  $\Psi_6 \approx 0.75$ . A second pulse with  $T = 10$  fully disorder the arrangement and recovers the amorphous state. Left panels are typical configurations and structure factors corresponding to the two stable values of  $\Psi_6$  at the origin of the arrows. Dashed lines in the graphics and the connecting shadow in the configuration panels are guides for the eye. Units of time and temperature are normalized to the units of friction coefficients and potential energy respectively (see Ref. [2]).

the subsequent quench develops the self-induced polydispersity explained above. On the other hand, going from amorphous to crystalline is achieved by heating to subcritical temperature, but fairly above  $T_*$ , so that the hopping dynamics is activated again and the system can evolve to the ordered configuration; which is of course kept when getting back to the operational temperature [2]. The range of real materials that can perform such a protocol is very wide in nature and, for most of them, the state of the art in experimental setups, controlling interactions, polydispersity and dimensionality, makes this idea very likely to be tested.

As a generic proof of concept, we performed computer simulations of such bit-writing process for an ultrasoft potential [2] with well-known melting and freezing temperatures. The aim was to demonstrate that the crystalline and amorphous structures can be easily tuned by heat pulses, so introducing a new type of phase-change behavior relevant for information storage applications. The results are depicted in Fig. 2, where the right plot shows time evolution of the six-fold orientational order parameter  $\Psi_6$ , that is 1 for a perfect triangular structure and vanishes for fully disordered configurations. As can be observed, the two stable structures at the operational temperature, shown in the left panels with representative snapshots, can be switched by means of the application of targeted heat pulses. This verifies the feasibility of the hypothesized bit-writing protocol.

As briefly mentioned before, typical bit-reading principle for this kind of applications come in two variants associated to the crystalline-amorphous change: changes in the electronic properties (e.g. resistance),

and changes in optical properties. While the former is highly dependent on the specific nature of the system at hand, the latter can be easily characterized by the diffraction pattern insight obtained with the calculation of structure factors. In the small left-panels of Fig. 2 a quite sharp contrast is observed for the peak patterns of the two bits.

The discovered self-assembled polydispersity is thus an underlying mechanism with the potential to start a new branch of information storage devices. Whether the “disc” will be prototyped or not is way beyond our envision capability. Our numerical proof of concept was, however, both a hopeful call to spotlight cluster-forming systems for practical use, and a quite enjoyable stop in the way to understand the statistical physics of soft-matter.

## Notes

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- Actually, the dynamics become glassy-like, with a two-step relaxation behavior (see Ref. [1])

## References

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- [2] R. Díaz-Méndez, G. Pupillo, F. Mezzacapo, M. Wallin, J. Lidmar and E. Babaev, Phase-change switching in 2D via soft interactions, *Soft Matter*, **15** (2019) 355-358