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## Supercooled liquids and glass transition

Understanding how viscos slow down determines the route followed by a supercooled liquid in becoming a glass well below the freezing point still represents one of the major challenges in condensed matter physics. In fact, in spite of countless and huge efforts on both the theoretical and experimental sides, many significant questions on the very nature of the glassy state remain unanswered yet.



General consensus exists about the kinetic nature of the experimentally observed glass transition. On cooling, molecular motions become slower and slower until they appear arrested over any experimental accessible time scale. There exists, however, a strong belief that this experimental phenomenon is only the signature of a more fundamental thermodynamic transition occurring at a well-specified

temperature lower than the experimental glass transition temperature, as it was historically hypothesized by Kauzmann. He pointed out that the entropy of a supercooled liquid decreases, while cooling, more rapidly than the entropy of the corresponding solid until, at a system dependent temperature, the two entropies equate. Extrapolating this behavior further, down to absolute zero, would represent a violation of the third law of thermodynamics. Kauzmann advanced the hypothesis that a second order phase transition, occurring at a specific temperature, higher than that of the vanishing entropy difference, could be the way the Nature adopts for avoiding such a thermodynamic paradox.

However this argument revealed erroneous, because it implies the occurrence of an isothermal process which can produce the solid phase moving from the metastable liquid. This is thermodynamically impossible because such a process would imply the coexistence of liquid and solid at a temperature lower than their coexistence temperature.

On these bases, it appears that most of the ideas which have led to the hypothesis of a thermodynamic transition reveal to be ill

founded. In particular, we have shown how distinguishing between supercooled liquids and equilibrated glasses can reduce to a mere semantic question. The apparent differences between them are only related with a cross-over temperature at which the experimental time scale becomes shorter than the configurational relaxation time of the system. Once the main motivation for assuming an underlying thermodynamic transition beyond the experimentally observed glass transition has been ruled out, many questions remain unsolved as the consequence. As an example, the assumption of a glass metastable phase would imply that a glass would be ergodic while the perspective which describes the system in terms of a kinetic arrest, over the observation time, would imply

that it is intrinsically not ergodic. A further significant example concerns with the question if it is thermodynamically reasonable to hypothesize for water the existence of a liquid-liquid phase transition in the deeply supercooled region, with a liquid-liquid coexistence line which can possibly be prolonged well below the supposed homogeneous nucleation temperature, reaching the coexistence line between two distinct amorphous phases which have been experimentally observed.

This research is aimed to explore if there are surviving enough motivations suggesting that the glass obtained from a supercooled molecular liquid can be really described as a metastable phase, other than the metastable liquid. In particular, we are trying to explore the possibility of finding an answer to many of the open questions within the framework of a simple out of equilibrium thermodynamic approach.

The research is carried on both with theoretical and experimental approaches (fast imaging techniques, Calorimetry, Raman and Brillouin Scattering).

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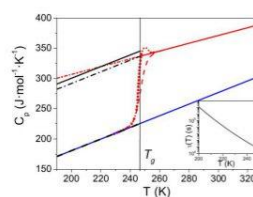


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