Glassy dynamics: mean-field landscape pictures versus growing length scale scenarii

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This is a short review on the compatibility between (a) mean-field, mode-coupling theories of the glass transition, where potential energy landscape ideas are natural, and (b) the necessity of describing the slowing down of glassy materials in terms of a growing cooperative length, absent from mean-field descriptions. We summarize some of the outstanding questions that remain before we can say we understand why glasses do not flow.

I. THE PROBLEM

The most salient properties of fragile glasses are (a) the non exponential (“stretched”) and heterogeneous nature of the relaxation; (b) the extremely fast rise of their viscosity \( \eta \) that increases by 15 orders of magnitude as the temperature is decreased by less than a factor 2, and appears to diverge at a finite (Vogel-Fulcher) temperature; (c) the aging and memory effects of the out-of-equilibrium phase, that shows some similarities with spin-glasses. A rather remarkable aspect of the Vogel-Fulcher divergence (b) is that the extrapolated freez-temperature; (c) the aging and memory effects of the out-of-equilibrium phase, that shows some similarities with spin-glasses. A rather remarkable aspect of the Vogel-Fulcher divergence (b) is that the extrapolated freezing temperature \( T_{VF} \) is found to be very close, for a whole range of materials, to the Kauzmann temperature \( T_K \) where the extrapolated entropy of the glass becomes smaller than that of the crystal. In other words, the viscosity of glasses (a dynamical property) seems highly correlated to the number of microscopic configurations in which the glass can get stuck. The smaller this number, the larger the viscosity. The striking observation that makes the ‘problem of glasses’ interesting is that very many, totally different materials, exhibit the same properties, pointing to the existence of a somewhat universal mechanism: glassy dynamics is physics more than chemistry.

Two apparently quite different frameworks have been discussed in the (fourty years old) literature to account for this phenomenology: 1) phase space/energy landscape pictures, where the system is trapped in metastable states of varying depth. The dynamics is made of small harmonic vibrations around each metastable configurations, separated by hops between different minima of the free-energy; 2) cooperatively rearranging regions of increasing length. The dynamics becomes sluggish because larger and larger regions of the material have to move simultaneously to allow substantial motion of individual particles. Although this idea seems most reasonable, its reality has remained elusive until recently: a consistent definition of this growing length, its experimental measurement and its calculation within a theoretical model (even highly simplified) are subjects of topical activity. Interestingly, similar concepts are also relevant for the description of other “jammed” systems, such as dense granular assemblies that flow in a very jerky way.

The landscape picture can be given more flesh within mean-field theories. The Random Energy Model, for example, contains already a lot of the glass phenomenology (entropy crisis at \( T_K \), aging...). More elaborate mean field spin-glasses theories lead to dynamical equations that are identical to the “Mode Coupling” theory (MCT) of supercooled liquids. MCT is considered by many to be the only available first principle theory of the supercooled state, starting from interacting atoms and making its way up to compute the viscosity of the liquid as a function of density and temperature. This theory makes a number of quantitative predictions that can be compared to experiments, some of which in remarkable agreement with observations. This analogy between MCT and mean-field spin-glasses allows one to interpret the MCT scenario for dynamical arrest in a clear fashion: the potential energy landscape has only unstable saddle points above a certain “threshold” energy, around which the system can only pause momentarily before continuing its exploration of phase space. This corresponds to the high temperature liquid phase. At the energy threshold (corresponding to \( T_{MCT} \)), the saddles only have marginal (zero curvature) escape directions, responsible for the MCT divergence of relaxation times. For lower energies (temperature) there are only minima where the system gets trapped. The number of these minima is exponential in the size of the system as long as \( T_K < T < T_{MCT} \), but the barriers between them are (in mean-field) infinite: once trapped in a minimum, the system remains there forever.

Is this mean-field picture at all relevant for finite range interactions? Phase-space pictures cannot be directly applied when the dynamics is local: the energy surface must somehow “factorize”. There cannot be a diverging time
scale without a diverging length scale. “Hops” in phase space should correspond to definite spatial structures (vacancies? strings? fractal clusters?). The observed dynamical heterogeneities, and the corresponding viscosity/diffusion decoupling must be accounted for. Hidden behind phase space pictures, there must thus be a dynamical length scale $\xi(T)$ governing the slowing down of these materials. Contrarily to simple systems where this length scale is the characteristic size over which some order (ferromagnetic, crystalline, etc.) is established, the difficulty of glasses and spin-glasses is that no obvious local order sets in. The definition of a dynamical length scale is more subtle and requires a four point density correlation function that attempts to quantify the cooperativity of the dynamics. This four-point function plays the role of the usual two point correlation in second order phase transitions. Following this path, one can show that MCT in fact predicts the divergence of $\xi(T)$ when $T \to T_{MCT}$: correspondingly, fluctuations become dominant below six dimensions, so that MCT cannot be quantitative in three dimensions. The second, perhaps more essential, problem of MCT is that barriers between metastable states must be finite for realistic potentials, even for $T < T_{MCT}$. Therefore, the predicted divergence of the viscosity at $T_{MCT}$ is in general smeared out.

What is then the nature of the growing length scale at lower temperatures? Old free volume ideas, recently revived within the context of ‘facilitated’ models, suggest that mobility defects trigger the dynamics and become more and more dilute at temperature decreases – the length scale is then the distance between defects. A more ambitious scenario, proposed by Adam-Gibbs in the 60’s and, inspired by mean-field spin-glasses, by Kirkpatrick, Thirumalai and Wolynes in the 80’s, relates the size of collectively rearranging regions to the configurational entropy of the glass. The idea is that of entropic melting of frozen clusters: small clusters have few low energy configurations and are pinned in one of them by the external environment; large clusters can explore many configurations and free themselves from any boundary conditions. At small scales, the dynamics from state to state is fast (low barriers) but leads to nowhere – the system ends up always visiting the same state. For larger scales, the system can at last delocalize itself in phase space and kill correlations, but this takes an increasingly large time. The crossover scale $\xi$ then sets the relaxation time and behind phase space pictures, there must thus be a dynamical length scale $\xi(T)$ governing the slowing down of these materials. Contrarily to simple systems where this length scale is the characteristic size over which some order (ferromagnetic, crystalline, etc.) is established, the difficulty of glasses and spin-glasses is that no obvious local order sets in. The definition of a dynamical length scale is more subtle and requires a four point density correlation function that attempts to quantify the cooperativity of the dynamics. This four-point function plays the role of the usual two point correlation in second order phase transitions. Following this path, one can show that MCT in fact predicts the divergence of $\xi(T)$ when $T \to T_{MCT}$: correspondingly, fluctuations become dominant below six dimensions, so that MCT cannot be quantitative in three dimensions. The second, perhaps more essential, problem of MCT is that barriers between metastable states must be finite for realistic potentials, even for $T < T_{MCT}$. Therefore, the predicted divergence of the viscosity at $T_{MCT}$ is in general smeared out. 

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II. OPEN QUESTIONS

Therefore, some of the outstanding questions that remain before we can say we understand why glasses do not flow are the following:

- How relevant (if at all) are mean-field ideas/models for real glasses/spin-glasses? Is cooperativity ‘non topographic’ (non thermodynamical) as in mobility defect/facilitated models or related to an exponential degeneracy of metastable states, as in mean-field models? Can one make some (controlled) theoretical progress on a non mean-field model of glasses, or at least formulate a Ginzburg-like criterion to understand the parameter region where mean-field models are relevant to describe real glassy materials? Can a frozen, non crystalline thermodynamical glass phase exist in finite dimensions or is it always killed by ‘activated processes’, absent from MCT? How much of Parisi’s hierarchical energy landscape survives in finite dimensions?

- What is the geometry of elementary dynamical excitations in glasses and spin-glasses (strings? fractal clusters?)? Is there really, experimentally, a detectable growing dynamical length scale in glassy systems (including jamming granular materials, soft glassy materials, spin glasses)? How large can this length actually grow (probably not more than 10-20), and can this explain the apparent universality of glassy dynamics? Is this length scale impor-
tant to understand, e.g. anomalous phonon modes or fracture in these materials?

• Is quenched disorder crucial or is there a random matrix like theory of the statistics of energy landscapes, that would make MCT predictions generic? Is the idea of fragility and disorder/temperature chaos, now well established for disordered systems (spin glasses, randomly pinned objects) and possibly related to rejuvenation effects, also relevant for structural glasses?

From a wider perspective, one can ask how much glassy systems, with their profusion of quasi-equilibrium states and complex dynamics, can be used as metaphors in other contexts. Combinatorial optimization is an already well studied one. Applications in economics/finance/game theory, where equilibrium is often assumed but may never be reached, is a fascinating prospect.

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III. (VERY) FEW REFERENCES

• Some classic papers: G. Adam, J. H. Gibbs, J. Chem. Phys. 43 139 (1965). M. Goldstein, J. Chem. Phys. 51, 3728 (1969).

• The mean-field scenario for glasses: T. R. Kirkpatrick, P. G. Wolynes, Phys. Rev. B 36, 8552 (1987); T. R. Kirkpatrick, D. Thirumalai, P. G. Wolynes, Phys. Rev. A 40 (1989) 1045; M. Mézard, First steps in glass theory, in “More is different”, Ong and Bhatt Editors, Princeton University Press (2002).

• The Mode-Coupling Theory. W. Götze, L. Sjögren, Rep. Prog. Phys. 55 241 (1992)

• Length scales within mean-field models: G. Biroli, J. P. Bouchaud, Diverging length scale and upper critical dimension in the Mode-Coupling Theory of the glass transition, Europhys. Lett. 67 21 (2004); J. P. Bouchaud, G. Biroli, On the Adam-Gibbs-Kirkpatrick-Thirumalai-Wolynes scenario for the viscosity increase of glasses, cond-mat/0406317 to appear in J. Chem. Phys.

• Non topographic mobility defect models: J.P. Garrahan, D. Chandler, Proc. Natl. Acad. Sci. 100, 9710 (2003); L. Berthier, J.P. Garrahan, Phys. Rev. E 68, 041201 (2003)

• Strings and landscape: M. Vogel, B. Doliwa, A. Heuer and S.C. Glotzer, Particle rearrangements during transitions between local minima of the potential energy landscape, e-print cond-mat/0309153 and refs. therein.

• Length scales and rejuvenation in spin-glasses: D. S. Fisher, D. A. Huse, Phys. Rev B 38, 373 (1988). J.P. Bouchaud, V. Dupuis, J. Hammann, E. Vincent, Phys. Rev B 65 024439 (2001); P. Jonsson, R. Mathieu, P. Nordblad, H. Yoshino, H. Aruga Katori, A. Ito, Spin-glasses, a ghost story, e-print cond-mat/0307640.