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Dynamical Heterogeneities in Glasses, Colloids and Granular Media

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Preface

Many materials of industrial importance are amorphous, from window glasses and plastic bottles to emulsions, foams, and dense granular assemblies. Understanding the formation of these different types of disordered solids via the so-called glass and jamming transitions is a challenge that resisted a large research effort in condensed-matter physics over the last decades, and that is of interest to several fields from statistical mechanics and soft matter to material sciences and biophysics. From a fundamental point of view, the question is to know whether the sudden, but continuous freezing is due to a genuine underlying phase transition, or if it is a mere crossover with little universality in the driving mechanisms.

In recent years, evidence has mounted that the dynamical slowing down of supercooled liquids, colloids and granular media might indeed be related to the existence of genuine phase transitions, but of very peculiar nature. Contrasting with usual phase transitions, the dynamics of these materials dramatically slows down with nearly no changes in their conventional structural properties. One of the most interesting consequences of these ideas is the existence of dynamical heterogeneities, which have been discovered to be, in the space-time domain, the analog of critical fluctuations in standard phase transitions. Dynamic heterogeneity is now recognized to be a key feature of glassy dynamics, after a major research effort from a large community of researchers originating from a broad spectrum across condensed-matter physics, from liquid-state theorists to experimentalists working on foams or grains. Yet, there is no textbook specifically dedicated to this large body of work and research community, and we felt that this gap needed to be filled.

The present book provides a broad and up-to-date overview of the current understanding of dynamic heterogeneity in glasses, colloids and granular media. It is multidisciplinary in nature. It contains chapters dedicated to theory, numerical simulations and experiments. Its content spans from the physics of molecular glass formers to soft glassy materials, foams and grains and covers both equilibrium aspects related to the glass transition, and non-equilibrium features such as aging phenomena. The book contains formal chapters about recent theoretical developments and very phenomenological ones concerned with more practical and experimental aspects.

Such a broad scope would have been difficult to cover by a single author. We have therefore mustered different scientists, who have all made important contributions to the field. The book in fact originated from a workshop organized by three of us and held in the Lorentz Center in Leiden in September, 2008. The workshop had an unusual format: relatively long, in-depth oral contributions were systematically followed by a summary of the presented work and a critical discussion by a "discussant". Although standard in economics for example, this format is not very common in physics and should be encouraged. It has indeed fostered a very animated and insightful meeting.

We have tried to have the book reflect the special atmosphere of the conference by insisting that most chapters should be jointly written by researchers working on closely related topics, but usually with different points of view, backgrounds, or interests, and sometimes from competing groups! Out of this original construction, where some consensus had to be reached within each chapter, has emerged a series of contributions that has no equivalent in the published literature. The resulting book has, we believe, a much broader, equilibrated and lively perspective than conventional review articles.

A particular effort was made in each chapter towards explaining the topic in very pedagogical terms, such that a broad range of readers—graduate students and more experienced researchers, experimentalists and theoreticians—can benefit from the book. The book is hopefully well suited to people who want to learn and discover what dynamic heterogeneities are and why they are important and useful. It is also meant to be a valuable piece for researchers who already know the field and want to understand or delve into a more detailed aspect they know less about, or bring forth some experiments or simulations they are unaware of. Each chapter contains a large number of references, making the book useful also for a literature search. Finally, we made sure that each chapter can be read independently from the rest of the book, while making many cross-references between chapters to guide the readers.

The book opens with yet another unusual item: the parallel interviews of four condensed-matter luminaires: Jorge Kurchan, Jim Langer, Tom Witten and Peter Wolynes. We have asked them to answer, in an informal way, eleven questions about the glass transition and dynamics heterogeneities, the last one being: *If you met an omniscient God and were allowed one single question on glasses, what would it be?* The result is both delightful and insightful, and we hope the reader will enjoy these lively snippets of science as much as we did.

We hope that this book will fulfill a useful function in bringing together, within a single cover, most of the recent developments on dynamic heterogeneity studies. We would like to thank all the authors for their efforts and care in preparing their chapters, all the referees who accepted to review the chapters and helped the authors to produce a text of high scientific quality and rigor. We also thank the speakers, discussants and participants of the Lorentz Center workshop in Leiden in September 2008 who contributed to a successful meeting and eventually gave birth to this book.

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Contents

1
Scientific interview

Jorge KURCHAN, James S. LANGER,
Thomas A. WITTEN and Peter G. WOLYNES

Abstract

Four leading scientists in the field of the glass transition answer to a series of questions formulated by the Editors. No specific format has been imposed to their answers: the scientists were free to answer or not to any given question, using how much space they felt appropriate. After a first round of answers, each author was given the opportunity to read the answers by his colleagues and to adjust his own answers accordingly.

The questions were:

- Q1) In your view, what are the most important aspects of the experimental data on the glass transition that any consistent theory explain? Is dynamical heterogeneity one of these core aspects?
- Q2) Why should we expect anything universal in the behavior of glass-forming liquids? Is the glass-transition problem well defined? Or is any glassy liquid glassy in its own way?
- Q3) In spin-glasses, the existence of a true spin-glass phase transition has been well established by simulations and experiments. Do you believe that a similar result will ever be demonstrated for molecular glasses?
- Q4) Why are there so many different theories of glasses? What kind of decisive experiments do you suggest to perform to rule out at least some of them?
- Q5) Can you briefly explain, and justify, why you believe your pet theory fares better than others? What, deep inside, are you worried about, that could jeopardize your theoretical construction?
- Q6) In the hypothesis that RFOT forms a correct skeleton of the theory of glasses, what is missing in the theoretical construction that would convince the community?
- Q7) Exactly solvable mean-field glass models exhibit an extraordinary complexity requiring impressive mathematical tools to solve them. Are you worried that for

both the spin and structural glass problems it proves so hard to establish the validity of mean-field concepts in finite dimensions? Why are finite-dimensional versions of mean-field glasses always behaving in a very non-mean-field manner?

- Q8) In your view, do the recent ideas and experimental developments concerning jamming in granular media and colloids contribute to our understanding of molecular glasses, or are they essentially complementary?
- Q9) If a young physicist asked you whether he or she should work on the glass problem in the next few years, would you encourage him or her and if so, which aspect of the glass problem would you recommend him or her to tackle? If no, what problem in condensed-matter theory should he or she tackle instead? If yes, what particular aspect of the glass problem seems the most exciting at present?
- Q10) In twenty years from now, what concepts, ideas or results obtained on the glass transition in the last twenty years will be remembered?
- Q11) If you met an omniscient God and were allowed one single question on glasses, what would it be?

1.1 Jorge Kurchan answers

Note: only references to papers that are difficult to identify are given here. For a general presentation, see the review article (Cavagna, 2009).

Q1) In your view, what are the most important aspects of the experimental data on the glass transition that any consistent theory explain? Is dynamical heterogeneity one of these core aspects?

The mere existence of heterogeneities is not in itself an additional fact, but a mathematical necessity: there have to be heterogeneities if there is a time scale that grows faster than Arrhenius in a system of soft particles. The only way a system can construct zero modes or higher barriers is through cooperativity. Of course, if one has access to the detailed space-time form of the heterogeneities, then this is extra information.

A model of fragile glasses should capture the relation between growing time scales and vanishing entropy. I would add that fluctuation–dissipation relations should reproduce the Lennard-Jones results of Barrat and Berthier Barrat and Berthier (2002), a useful discriminant since the precise form of these relations is model dependent. All these aspects are subject to the uncertainties associated with short times.

If we make a multidimensional scatter plot of the parameters describing existing glass formers, we shall find that there are correlations. Those between fragility, specific heat and stretching exponents have been discussed by Wolynes and coworkers, and there must be others that I do not know. To the extent that they are significant, correlations must be explained. If they are imperfect, this has to be explained too.

Having said this, I think that all in all it will be the internal logic and the tractability of the theory—*the fact that the objects it invokes and relates have a clear definition*—that will make it satisfactory.

Q2) Why should we expect anything universal in the behavior of glass-forming liquids? Is the glass-transition problem well defined? Or is any glassy liquid glassy in its own way?

Here one should separate two levels. The word "glassy" is used (a) in the general sense of slow (or *slowing*) dynamics. (b) for "fragile" glasses with their rather specific features of entropy and time scales.

The general sense (a) includes systems such as the two- and three-dimensional spin-glasses (including those having Potts or p-spin interactions), "dirty" ferromagnets, crystal ripening and defect annealing, quasicrystal annealing, strong and fragile glasses, and also kinetically constrained models, frustration-limited domains, "Backgammon", "car-parking", Tower of Hanoi, and a myriad other models. The class of fragile glasses (b) is much more specific: it includes a group of systems having slowing down of dynamics in a characteristic (non-power-law) super-Arrhenius way, with what seems to be a concomitant decrease in entropy. Polymers and some molecular glasses are of this kind.

In the general class (a) there might be many ways of being glassy. One might still expect some universal features that are just a consequence of the dynamics being slow, or entropy production small. There are relations between time scale increase and dynamics correlations, bounds for the violations of the fluctuation–dissipation relation, generic features of response of the system to driving, etc., that apply to every slow system. We would like to know much more, as this might be a front where out-of-equilibrium thermodynamics might develop—it being less ambitious than the generic non-equilibrium problem.

Class (b) is much more restrictive, to the point that some consider it empty. Super-Arrhenius behavior—the logarithm of the time scale growing faster than linearly in the inverse temperature—implies growing dynamic *and static* length scales (see Q1), and this leads to the search of some form of hidden order, with perhaps complete ordering eventually avoided. Here, the expectation is that things are much more universal in the sense of applying to fewer systems but for those, essentially in the same way. Nobody will be happy with a mechanism, an order parameter or a length scale, that is defined differently for polymers and a monoatomic glasses.

Q3) In spin-glasses, the existence of a true spin-glass phase transition has been well established by simulations and experiments. Do you believe that a similar result will ever be demonstrated for molecular glasses?

The question whether there is a spin-glass transition in a magnetic field is still very much unresolved, and is the object of an impassioned and contradictory literature. The spin-glass example is interesting, as it illustrates a new development that we are witnessing: simulation time scales are for the first time overlapping experimental ones. What in fact we discover is that there is quite a good continuity between experiments and simulations, *and that there are issues that neither will resolve.*

To illustrate how simulations may have a paradoxical effect, consider the example of the JANUS aging simulations The JANUS (2008). They measure two-time correlation