The Physics of Glasses:
Relating Metallic Glasses to Molecular, Polymeric and Oxide Glasses

A proposed joint program of the Kavli Institute for Theoretical Physics
and
The International Center for Materials Research at UCSB

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Executive Summary

This proposal seeks to take advantage of a recent convergence between theoretical and computational advances toward understanding glasses within the physics and chemistry communities and practical advances in producing, characterizing and testing metallic glasses in the engineering community. The 3-month workshop to be jointly supported by KITP and ICMR is proposed for April 15, 2010 – July 15, 2010. This workshop will involve theoretical and experimental physicists, chemists and engineering scientists from a broad range of communities whose work relates to the structure, thermodynamics, dynamics and mechanics of glasses. The program will focus specifically on key questions that represent fundamental physical issues critical for glasses in general, including chalcogenides, polymeric and metallic glasses. These questions include:

- What are the universal aspects of glass formation, in diverse substances, such as metallic alloys, silica and polymeric glasses? What accounts for these universal aspects?
- How does structure connect to dynamics in glass formers, in particular to dynamical heterogeneity, fragility, jamming, aging and rejuvination?
- What underlying physics determines glass formability and suppresses crystal nucleation in multi-component systems?
- How does glass respond to the application of mechanical stresses both at and far below the glass transition? To what aspects of dynamical behavior does mechanical response under applied stress relate?

To the knowledge of the organizers, although there is avid interest in these questions in both the statistical and condensed matter physics communities and in the materials science community, there have been only a handful of opportunities for members of the physics and materials communities to interact. We have solicited feedback from both communities and have been persuaded that creating a platform for dialog between these communities at this juncture will yield rich scientific dividends. At this point over 100 researchers have expressed interest in potentially participating in this program (a list is provided at the end of this document), and 37 have expressed a desire to come for 6 weeks or more. The community of scientists who have expressed interest is international with a significant number of participants from North America (51), Western Europe (34) and Asia (16) as well as South America, Australia and the Middle East in accordance with the mission of the ICMR.

Introduction

The nature of the glass transition remains one of the grand challenges at the intersection of condensed matter and statistical physics. A great deal of controversy still surrounds theories of glass transition, and this has been a principal focus of the community. Investigations in the physics community are only beginning to expand to include other equally fundamental questions of relevance for the engineering of these systems. A number of recent developments make this an exciting time for understanding the physics of glasses in general, and metallic glasses in particular. New computational methodologies and increased computing power have made it possible to test the fundamental assumptions of well-developed theories. Furthermore new experimental techniques have been developed for characterizing the structure of glasses, materials that were once considered simply "a-morphous". But, perhaps most importantly, as a result of the great strides made by materials engineers, metallic glasses themselves have risen from obscurity to the threshold of application. Although the theoretical physics community and the metallic glasses community remain largely dissociated from each other at this time, we believe that a program such as we describe here can bridge that divide. This presents a new opportunity for the theoretical physics community to engage actively with the materials community so that the new wealth of experiment and the relevant practical concerns can serve to refocus the lines of inquiry for the field. We are confident that increased interaction between theoretical physics and materials engineering in this area will result in advances that will be fundamental, insightful and practical.

We believe that the best way to accomplish this is to organize a 3-month workshop under the auspices of the KITP and the ICMR at UCSB. This workshop would be built upon the highly successful KITP model and would bring participants into residence for no fewer than 3 weeks, with a limited number of 1-week visits made by experimental investigators. The workshop would seek to focus on a number of critical theoretical questions that are important to the glass community, and the metallic glass community in particular. We believe that by considering these questions we can refocus the attention of the theoretical effort in ways that will spur creative approaches to investigations of glassy materials.

Brief Survey of Recent Developments

While glasses have been an area of active study for a long time, there are a number of notable developments that have occurred in the last decade that broadened thinking about these materials.

One of these has been the simultaneous success and limitation of mode coupling theory (MCT). MCT provides a mean-field description of the dynamics of a glass in terms of its structure factor, which can be measured experimentally via scattering. The theory proposes a coupling between density fluctuations that results in a critical slowing down of the system as the liquid is cooled. MCT does an excellent job of relating the structure of the glass to the initial few decades of dynamical slow down as the temperature of the super-cooled liquid is lowered. It also provides insight into the scaling that is observed in this regime. However, MCT produces an incorrect prediction for the temperature of the glass transition and, being based on an uncontrolled approximation to provide closure to the dynamic equations, provides little insight into the origin of these discrepancies. Furthermore, as a mean field theory expressed in terms of Fourier space modes of the density fluctuations, the theory is divorced from the atomic scale details that control glassy behavior. This makes it difficult to use MCT as a starting point for further developments related to, for example, predicting low temperature mechanical response

or the suppression of crystal nucleation as relates to glass formability. However, there have indeed been recent attempts to both extend MCT to account for dynamical heterogeneities, and to make it useful in dealing with systems under applied external stress. The extent to which such advances will overcome the limitations mentioned above remains to be demonstrated.

Another is the advance in the energy landscape theory (ELT). ELT captured the imagination of many theoretical physicists because of its intuitive clarity and simplicity in expressing the role of structural heterogeneity in determining complex dynamics. Coupled with computational simulations this theory has led to important analysis of the relationship between the statistics of local energy minima (inherent structures), saddle points, and energy meta-basins, to slow dynamics, in a diverse range of systems, in and out of equilibrium. Different dynamical regimes, evaluation of configurational entropy and verification of the Adam-Gibbs relation, fragiliy, fragile-to-strong crossover, and aging, are some of the aspects that have been studied in this approach. Here too, however, an understanding of the atomistic underpinning of relevant quantities is still unclear, even with the aid of computer simulation. Consequently the predictive power of the ELT is limited. Some critical concepts appear to be lacking before this phenomenology can become productively wedded to a real space structure of materials.

Yet another significant development is the exploration of the concept of "fragility". This concept allows us to map out all kinds of different classes of glasses, from inorganic glasses and metallic glasses to organic and polymeric glasses, in a single chart, and to understand the common features and phenomena related to their viscous behavior. However, the microscopic origin of the variations in liquid fragility, while under active study, is not satisfactorily understood.

A number of intriguing observations have been made in computer simulations of glasses that provide additional insight into the dynamics of supercooled liquids and low temperature glasses. These include the observation of dynamical heterogeneities, correlated motions of atoms in the glass that appear to grow in length scale as the glass is reduced in temperature. The analysis of such heterogeneities, leading to an understanding of, e. g., the breakdown of the Stokes-Einstein relation, have been pursued vigorously. Length scales associated with dynamical heterogeneities have been incorporated in extending MCT, as mentioned above, but deep theoretical questions remain. In particular a school of thought emerged that sees dynamical heterogeneity as central to the phenomena of glass formation. In particular, theories of dynamic facilitation have been proposed that seek to provide a touchstone for the unification of theory regarding glasses and kinetically constrained statistical physics models.

Beyond computational approaches, there has been a recent excitement regarding experiments that employ colloidal systems as analogous systems to atomic and molecular glass formers. These experiments provide spatially and temporally resolved data of the dynamics of the glassy system, and these systems can be investigated carefully while subjected to shear. This development presents and opportunity to strengthen the connections between the glass community and the colloid and granular materials communities. It is our plan to include work on theories of "jamming" in this program. These theoretical developments seek to provide a common framework in which to think about the structurally arrested states in glasses and granular media.

Attempts to go beyond the mean-field level of description have also been attempted in work inspired by behavior in spin glass models. In the context of the random first order theory, the

"mosaic" length scale associated with nucleation events leading to structural relaxation has been analyzed.

At low temperatures interesting observations have been made regarding stress fluctuations in the glass, and the advent of local regions of rearrangement, so-called "shear transformation zones," that appear to mediate plastic flow. These observations have been used as springboards for physically based phenomenological theories of the glass transition and the glass' low temperature behavior.

Simultaneous to this theory and simulation research, which was primarily carried out in the physics community, a large amount of effort was being devoted to understanding metallic glasses in the materials engineering community. These efforts focused on the development of new alloy systems with increasing degrees of glass formability. Such investigations have taken place without very much theoretical guidance, relying instead on various empirical methods to suggest compositions that would likely fall near deep eutectics in the alloy phase diagrams. The advent of glass forming alloys with lower critical cooling rates has permitted the fabrication of material of sufficient size to permit mechanical testing. These tests have revealed beneficial mechanical properties such as high strength, significant energy return in impact and a relatively high elastic compliance. In addition they have provided further evidence of catastrophic failure modes when the material's strength was exceeded, most notably by strain localization. Understanding both the issues surrounding glass formability and those related to mechanical response of these materials will require advances in our fundamental understanding of glasses. These advances will require an understanding of the structural basis of these behaviors; analysis of structure in metallic glasses is eminently more accessible than in many favorite organic or polymeric glass forming systems, and thus provides an opportunity and a challenge to develop and test a fundamental understanding of structure - property relations in glass formers.

Program Goals

The goals of the workshop will be to address relevant and fundamental theoretical questions that seem likely to provide significant advances to the materials science of glass forming metals. These questions are:

- What are the universal aspects of glass formation, in diverse substances, such as metallic alloys, covalently bonded ceramics and polymeric glasses? What accounts for these universal aspects?

  A great deal of wisdom regarding the glass transition has been developed in allied communities of covalently bonded glasses and polymeric glasses. Clearly significant universality exists across all these systems despite the highly disparate nature of the way frustration arises in these systems. By including a broad cross-section of researchers not limited to those focused on metallic glasses we hope to elucidate the underlying reasons for this universality and, equally as important, to explore its limits.

- How does structure connect to dynamics in glass formers, in particular to dynamical heterogeneity and to fragility?

  A clear understanding does not exist of how aspects of dynamics such as fragility may be rationalized in terms of changes in structure of glass formers. Indeed, a part of the recent thrust in studying *dynamical* heterogeneity has been the lack of clear *structural*

correlations that may be associated with dynamical slow down. Though MCT is based on structural information, there is no widely accepted way of understanding diversity of glass formers in terms of their real space structural peculiarities, *via* MCT. Further, it is only recently that researchers have attempted to build dynamical heterogeneity into MCT. At the same time, serious attention to spatial heterogeneity also comes from other non-mean field approaches such as the recent nucleation theories of glassy dynamics. At the other end, there have been interesting attempts to analyze structure of metallic glass formers. Thus, a discussion of structure, structural correlations and dynamical correlations in connection to each other will be fruitful.

- What underlying physics determines glass formability and suppresses crystal nucleation in multi-component systems?

  This question is significantly different than the question of the nature of the glass transition itself. It requires us to consider the circumstances under which a material will form a glass as opposed to organizing into a structure with some degree of long-range order. As with the question of the nature of the glass transition this discussion will necessarily involve consideration of both the atomic scale structure of the glass and the kinetics that determine that structure and its evolution at a given temperature. Interest lies both in fundamental questions on the role of thermodynamic and kinetic aspects, and applied questions relating to the ability to predict glass forming ability based on composition.

- How does glass respond to the application of mechanical stresses both at and far below the glass transition? To what aspects of dynamical behavior does mechanical response under applied stress relate?

  This question provides a connection between the study of nature of glasses and aspects of their mechanical behavior that are critical for understanding their processability and applicability. These questions are tied closely to questions of structure and kinetics, but require theorists to build a connection between the fundamental aspects of the glass transition and the practical concerns of fracture and other modes of mechanical failure such as shear band formation. Consensus has yet to emerge regarding very basic considerations as to the structure of these theories, and how one ought to characterize and analyze such mechanically induced material instabilities.

Proposed Program / ICMR Involvement

We propose a 3-month long program that will last from April 15 – July 15, 2010. The program will include a strong core of theory-minded scientists from physics and materials science. These will be augmented by a cross-section of carefully chosen experimentalists who share the goal of using experiment to obtain fundamental insights into the above issues. Weekly themes will be chosen by the organizers to take best advantage of the combinations of researchers currently in residence. We anticipate organizing three formal seminars, one informal moderated discussion, and one session of 5-minute blackboard introductions each week during the program. Otherwise we anticipate allowing a large degree of flexibility so participants can spend most of their time working together on problems of mutual interest. To make sure these connections happen efficiently the organizers plan to use the web to help invited participants organize into working groups prior to their arrival. We plan to initiate a wikispace on the KITP server that participants can use to gather thoughts, references and work-in-

progress, as well as planning topical discussions during the program. The organizers and certain longer-term visitors will be tasked with fostering the use of the wiki to promote informal exchanges and to guide open and inclusive discussion in ways that help us bring about fruitful interchange of ideas. In particular we wish to see the community come to some consensus on model systems that can be studied in detail to provide deeper insight, identify promising theoretical avenues that will address questions of value to the materials engineers and provide guidance as to the best ways in which simulation can serve to bridge the gap between theory and experiment.

Involvement of the ICMR will permit us to add components to the program that are not standard in other KITP programs. One of these initiatives will be to foster student exchange between research groups. Student applications would be accepted from the research groups of researchers participating in the program. These students will be given an opportunity to participate in the program for a period of time to be determined. During the KITP program support for student travel and living expenses will be supplied by ICMR, and they will be accommodated as associates during the KITP program. The expectation will be that this support is contingent on the student subsequently spending an extended period of time visiting the group of another researcher participating in the program. ICMR has facilitated such long-term international student exchanges in the past, and are currently engaged in such a program in complex oxide systems. We strongly believe that such student exchanges, supported by ICMR, have the potential to foster longer-term intergroup collaborations.

As another component of this goal of longer-term interactions between research groups, rather than stage a conference during the workshop, we would prefer to organize a conference at the KITP 8-months to one year after the workshop ends, sometime between June-August 2011. This workshop would gather the participants and the exchange students one more time to foster continuity in the interactions that develop during the program. The additional costs of this conference, which would be higher than usual due to the need to bring all the participants back for a one-week visit, would be borne by ICMR.

Researchers Who Have Expressed Interested in Participation

C. Austin Angell, ASU, USA
Ali Argon, MIT, USA
Michael Atzmon, Michigan, USA
Biman Bagchi, IISc, INDIA
Jean-Louis Barrat, Lyon, FRANCE
Livio Battezzati, Turin, ITALY
Ludovic Berthier, Montpellier, FRANCE
Rut Besseling, Edinburgh, UK
Giulio Biroli, CEA, FRANCE
Jean Carlson, UCSB, USA
Christiane Caroli, Paris IV, FRANCE
Horacio Castillo, Ohio U, USA
Michael Cates, Edinburgh, UK
Bulbul Chakraborty, Brandeis, USA
Charusita Chakravarty, IITD, INDIA
Claudio Chamon, BU, USA
David Chandler, UC Berkeley, USA
Leticia Cugliandolo, Paris VI, FRANCE
Chandran Dasgupta, IISc, INDIA
Juan de Pablo, Wisconsin, USA
Pablo Debenedetti, Princeton, USA
Florian Dellla Torre, ETH Zurich, SWITZ
Michael Demkowicz, LANL, USA
Ken Dill, UCSF, USA
Jack Douglas, NIST, USA
Douglas Durian, U Penn, USA
Jeppe Dyre, Roskilde, DENMARK
Mark Ediger, Wisconsin, USA
Takeshi Egami, U Tenn/ORNL, USA
Michael Falk, Michigan, USA
Franz Faupel, Kiel, GERMANY
Silvio Franz, ICTP, ITALY
Daan Frenkel, Cambridge, UK
Matthias Fuchs, Konstanz, GERMANY
Juan Garrahan, Nottingham, UK
Sharon Glotzer, Michigan, USA
Bill Goddard, Caltech, USA
Lindsay Greer, Cambridge, UK
Peter Harrowell, Sidney, AUSTRALIA
Marc Heggen, Julich, GERMANY
Eduardo Jagla, Bariloche, ARGENTINA
Willlliam Johnson, Caltech, USA
Kenneth Kelton, WUSTL, USA
Nicholas Kioussis, CS Northridge, USA
Walter Kob, Montpellier II, FRANCE
Jorge Kurchan, ESPCI, FRANCE
Daniel Lacks, CWRU, USA
James Langer, UCSB, USA
Anael Lemaitre, Inst Navier, FRANCE
John Lewandowski, CWRU, USA
Andrea Liu, U Penn, USA
Evan Ma, JHU, USA
L Mahadevan, Harvard, USA
Craig Maloney, CMU, USA
Daniel Miracle, AFRL, USA
Kunimasa Miyazaki, Kochi, JAPAN
James Morris, ORNL/Tenn, USA
Zohar Nussinov, WUSTL, USA
Corey O'Hearn, Yale, USA
Akira Onuki, Kyoto, JAPAN
Dimitrios Papaconstantopoulos, George Mason, USA
John Perepezko, Wisconsin, USA
Peter Poole, St. Francis Xavier, CANADA
Wilson Poon, Edinburgh, UK
Itamar Procaccia, Weizmann, ISRAEL
U Ramamurty, IISc, INDIA
Sriram Ramaswamy, IISc, INDIA
S Ranganathan, IISc, INDIA
Madan Rao, RRI, INDIA
Mark Robbins, JHU, USA
Joerg Rottler, UBC, CANADA
Tanguy Rouxel, Uni Rennes, FRANCE
Konrad Samwer, Göttingen, GERMANY
Srikanth Sastry, JNC, INDIA
Peter Schall, Amsterdam, NETHERLANDS
Herbert Schober, Juelich, GERMANY
Christopher Schuh, MIT, USA
Kenneth Schweizer, UIUC, USA
Francesco Sciortino, La Sapienza, ITALY
Surajit Sengupta, SNBNCBS Kolkata, INDIA
Oleg Senkov, UES, USA
Scott Shell, UCSB, USA
Howard Sheng, George Mason, USA
Yoji Shibutani, Osaka, JAPAN
Peter Sollich, Kings, UK
Frans Spaepen, Harvard, USA
Matthias Sperl, DLR, GERMANY
H. Eugene Stanley, BU, USA
Hajime Tanaka, Tokyo Univ, JAPAN
Anne Tanguy, Lyon, FRANCE
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Salvatore Torquato, Princeton, USA
Thomas Truskett, UT Austin, USA
Frederic Van Wijland, Uni Paris VII, FRANCE
Damien Vandembroucq, St. Gobain, FRANCE
Thomas Voigtman, DLR, GERMANY
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Eric Weeks, Emory, USA
David Weitz, Harvard, USA
Michael Widom, CMU, USA
Gerhard Wilde, Muenster, GERMANY
Yue Wu, UNC, USA
Matthieu Wyart, Harvard, USA
Ryoichi Yamamoto, Kyoto, JAPAN