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Book Review: *Liquids, Freezing and Glass Transition*

Liquids, Freezing and Glass Transition. Les Houches, Session LI (3–28 July 1989), J. P. Hansen, D. Levesque, and J. Zinn-Justin, eds., North-Holland, Amsterdam, 1991.

These two volumes (of nearly 1000 pages) comprise summaries of ten lecture courses and seven seminars presented at the 51st session of the Les Houches Summer School, which took place 3–28 July 1989, on the subject of the statistical mechanics of liquid structure and dynamics. The purpose of the Summer School was twofold: (1) to provide state-of-the-art overviews of recent extensions of theory, simulation, and experimental techniques developed over the past two decades for simple liquids to the analysis of increasingly complex systems and phenomena; (2) to provide introductions to newly-developed "first-principles" microscopic theories of freezing and relaxation in supercooled liquids up to the glass transition. The scope and quality of the lectures and seminars is clear from the Contents:

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Seminar 5. Transport and relaxation phenomena near the glass transition—Assessment of theory, by C. A. Angell
Seminar 6. Exact results for the liquid–solid interface, by L. Blum and D. A. Huckaby
Seminar 7. Wetting and spreading: Some experiments, by L. Leger

In Course 1, The Structure of Simple Liquids, Ian McDonald presents a "gentle introduction" to the statics of the liquid state. The role of correlation functions (CF) and especially the structure factor $S(k)$ as a static response function are highlighted by a derivation of the relationship between a variation in the singlet density at a point in the fluid to a small change in an applied external field using the techniques of *functional derivatives*. Appropriate generalizations of $S(k)$ for two-component ionic fluids are then presented along with properties for the case of the Debye–Hückel model, the latter appearing as a simple approximation of the Ornstein–Zernike equation. The Ornstein–Zernike idea of partitioning the total correlation between atoms in a fluid into a direct, local atom–atom correlation plus a nonlocal one is also shown to lead to fruitful results (in the sense of suggesting tractable approximation schemes) for the theory of polar fluids, for the familiar RISM model of molecular liquids, and even for polymers.

McDonald's article introduces several themes which recur throughout the courses and seminars. The central role of correlation functions (CF) [or structure factors $S_{\alpha\beta}(k)$] for the interpretation of experiment results in their behavior and computation becoming major goals of theory and simulation. Functional analysis is a natural tool for describing inhomogeneous fluids, and it proves to be a powerful one for developing both exact and approximate theories. The steep growth of mathematical difficulty with increasing fluid complexity is demonstrated in the case of binary ionic liquids. The ability of simulation to reveal unanticipated patterns in advance of experiment (such as *charge ordering* in ionic fluids) underscores a strong complementarity and interdependence of the three approaches to liquid structure and dynamics: experiment, theory, and simulation.

The conceptual unity underlying recent developments is evident in the individual lectures and chapters. This unity owes much, I suspect, to the emergence of a common language (correlation functions, structure factors,...) so that results and insights in one speciality are quickly communicated and become accessible to the whole community. A few examples perhaps suffice to illustrate the phenomena.

Course 3, Crystallization of Liquids: A Density Functional Approach, by D. Oxtoby, reviews recent progress in the equilibrium and dynamic aspects of the crystallization of a simple liquid. He emphasizes that four successful techniques generally employed to study other phase transitions

(lattice models, renormalization group, bifurcation theory, and computer simulation) have severe limitations for studying crystallization. The density functional methods were introduced early on by Kirkwood and Onsager to study the equilibrium aspects of first-order phase transitions, and Oxtoby shows how refinements and extensions of the free energy model and density ansatz have brought the theory to the semiquantitative stage for hard spheres and Lennard-Jones liquids. The lectures conclude by indicating that an appropriate dynamic generalization of density functional methods is needed to properly address nucleation and growth.

In Course 5, Aspects of Structural Glass Transitions, by W. Gotze, we find an exhaustive, four-part (over 200-page) discussion of the recent mode-coupling theories (MCT) of structural glass transitions. This is a welcome summary of a very large literature which has sprung up over the past 10 years or so. Part 1 summarizes the phenomenology of glass formation, and discusses seven features which Gotze believes are endemic to glass transitions. While most of these are classical and well known, a few rely on some recent neutron scattering experiments (also discussed separately by F. Mezei in Course 8) which are largely unfamiliar to the "glass-transition" community. Part 2 is written as a "mathematical essay," describing aspects of the ergodic to nonergodic transition which is found to take place in a number of extremely simple nonlinear equations for the density–density correlation function. Part 3 illustrates how such nonlinear equations can arise from a first-principles treatment using MCT, and Part 4 addresses the agreement between the theory and experiment.

At first sight, the ad hoc character of the MCT equations and the intuitive idea that hydrodynamic effects should not have much to do with T_g is discouraging. Still, the unexpected simplicity of the equations, the insensitivity of many results to the exact memory functional model (i.e., F1, F2, F13,...) used, and the relative success of MCT to describe cage effects and backflow in normal liquids are all compelling arguments to ask whether some connections exist with other, more traditional interpretations of the glass transition. In any event, it is certainly true that the MCT approach has resulted in renewed experimental, theoretical, and simulation activity in glass formation.

All in all, this is an excellent collection of articles describing many recent developments in the field of liquid structure and dynamics and can be highly recommended.

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