

PREFACE

Fifty years of liquid state physics

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Preface

Fifty years of liquid state physics

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1. Historical overview

The first liquid state theories were aimed at the investigation of fundamental properties of simple liquids and liquid mixtures, and an excellent presentation of their key elements can be found in the book by Hansen and McDonald [1]. Nowadays, despite the growing importance of simulation methods in most areas of physics, chemistry, biology and engineering, theoretical approaches continue playing a significant role in providing a deep understanding on the physics of fluid (and more generally disordered) systems, and in many cases represent a complement or even a judicious alternative to computer simulation [2, 3]. In this Special Issue, we have tried to review some of the state-of-the-art approaches to problems of current interest in liquid state physics, focusing on those theoretical contributions whose origin can be traced back to George Stell's seminal works spanning the last fifty years. In the lines that follow, we have, however, aimed at providing the reader with a somewhat broader and general overview, and therefore we have included some areas of research that were not so close to George Stell's original scientific interests.

Perhaps one of the key issues in liquid state theory has been the study of phase equilibria and criticality. From a chemical engineering standpoint, thermodynamic perturbation theories (TPT) played a significant role in the seventies and early eighties, at least for fluids that could more or less be treated as composed of spherical particles [4]. When it comes to providing an accurate description of both the structure and the thermodynamics of liquids, integral equation theories show a remarkable performance. A fairly comprehensive review of their application to phase equilibrium problems can be found in the works of Caccamo [5] and Bomont [6]. Integral equation theory (IET) is in many respects very much indebted to the fundamental contributions of George Stell on cluster expansions, which provided a deep insight into the structure of closure approximations for the Ornstein–Zernike relation (OZ).

However, when describing critical behavior, both TPT and standard IET approaches fail badly to account for the non-classical behavior of critical exponents. It was the pioneering work of Høye and Stell [7] that paved the way for the introduction of the idea of enforcing thermodynamic self-consistency on integral equation approximations to improve their performance close to the critical point. In practice, the self-consistent Ornstein–Zernike approximation (SCOZA) took advantage of the analytic form of the mean spherical approximation (MSA) solution to transform the self-consistency condition into a set of partial differential equations. The lattice gas solution by Dickman and Stell in 1996 [8] proved that the equation could not only yield extremely accurate vapor-liquid phase diagrams but also non-classical subcritical indices quite close to those predicted by the renormalization group (RG) approach.

The RG theory [9] provides a framework to account for the universality of critical phenomena, but fails to predict the non-universal features. The hierarchical reference theory (HRT), later developed by Parola and Reatto [10], and the collective variables (CV) theory due to Yukhnovskii and collaborators [11] are in principle able to describe both the non-universal and the universal properties of phase transitions. Under certain conditions the SCOZA and the HRT can be expressed in a unified form, as shown by Høye and Reiner [12], and the CV theory was shown to be equivalent to the Hubbard–Stratonovich field theory [13]. These approaches have been applied to systems with various potentials, including electrostatic interactions.

In addition to bulk thermodynamic and structural properties, a great deal of effort has been invested in the problem of fluid confinement near solid surfaces, both for simple

regular geometries and in porous media. Near an external wall and in the interface region the fluid is inhomogeneous, and this fact has a strong impact on thermodynamic and mechanical properties. In particular, capillary condensation [14], wetting [15], adsorption and critical adsorption [16] have been the subject of intense theoretical and simulation studies. The physics of interfaces, and the effects of confinement in a single pore are described in the book by Rowlinson and Widom [17], and in the review by Evans [18]. In this connection, inhomogeneous integral equation theories have dealt with fluids confined in regular geometries, such as slit pores, spherical cavities, etc. These approaches experienced substantial development in the 80s and 90s for simple fluids [19].

In the particular case of random porous media, the translational degrees of freedom associated with the porous matrix are quenched, and those associated with the confined fluid are annealed. A powerful tool in studies of such systems is the replica Ornstein–Zernike (ROZ) equation developed by Given and Stell [20, 21], and further analyzed by Rosinberg *et al* [22] among others. This approach has been exploited in recent years for a variety of systems, such as templated materials [23], sponge-like porous media [24, 25], etc.

Alternatively, inhomogeneities at the microscopic length scale can be successfully described by the classical density-functional theory (DFT) [26]. The history and recent developments in DFT are summarized in the recent special issue of Journal of Physics (Condensed Matter) [27]. DFT has been very successful in the description of the near-surface structure, but the results for the structure of the interface and for surface critical phenomena strongly deviate from exact results or simulations. Correct results have been obtained in the capillary wave theory of interfaces [28], and in the RG theory for systems with boundaries [16], respectively. The presence of a near-critical fluid induces an effective long-range interaction between the confining surfaces because of the restrictions on the critical fluctuations imposed by the confinement. This phenomenon which strongly recalls the quantum Casimir force, was first predicted by Fisher and de Gennes [29], and its existence has been recently confirmed by direct measurements [30]. In particular, the thermodynamic Casimir potential is observed between surfaces of colloidal particles.

The above observation of effective interactions between colloidal particles leads the way to complex liquids or soft-matter systems which have been the main subject of numerous recent studies. Complexity in this context means strongly non-spherical shapes and/or internal molecular degrees of freedom (e.g. polarizability or flexibility), large size asymmetry between the components, long-range interactions or interactions that are strongly anisotropic, or exhibit some competing tendencies. In fact water can also be deemed a complex fluid, as is evidenced by the large amount of work devoted to its anomalous behavior. In this case, complexity is induced by the presence of the hydrogen bond network which endows water and related substances (e.g. alcohols) with unique properties. The book by Barrat and Hansen [31] contains an excellent account of recent advances in the theory of simple and complex liquids.

For molecular fluids, polymers and mixtures, TPTs have been superseded in the last two decades by the Statistical Association Fluid Theory [2] (SAFT) in its different formulations, all of them deeply rooted in Wertheim's associative thermodynamic perturbation theory [32].

Over the last four decades a monumental effort has been dedicated to expand applicability of the IET into the realm of molecular and complex fluids (such as polymers). In this connection, it is worth recalling the three main classes of integral equation approaches developed to tackle this issue. Firstly, we have those theories rooted in the expansion of molecular correlation functions in the basis of orthogonal functions (e.g. rotational invariants) [33], and secondly we have those approaches based on the decomposition into site–site functions—reference interaction site model (RISM) [33]—, which, although less accurate, are computationally less demanding and algorithmically less involved. These are easily extendable to relatively complex fluids and even polymers [34]. As a third alternative, and of particular relevance in solvation problems [3] and even for confined fluids [35] is the three dimensional Ornstein–Zernike approximation which, when combined with the RISM equation, has shown to be able to tackle complex solvation problems approaching chemical accuracy [3]. More recently, the aforementioned RISM approximation has also been extended to describe molecular fluids in the presence of inhomogeneities [36].

Also in the realm of complex fluids, the asymmetric shape of stiff particles leads to formation of thermotropic liquid crystals (LC), and strongly anisotropic interactions between amphiphilic molecules (surfactants, lipids) and solvent molecules lead to formation of lyotropic LCs. Liquid crystals, microemulsions, and block co-polymers have been successfully investigated within Landau-type, DFT or field theories and by generic lattice models [37–40]. One such model for microemulsions and lyotropic LCs was developed by Stell and collaborators [41]. In the case of lyotropic liquid crystals and block co-polymers, however, only qualitative agreement with experiment and simulations was obtained by the approximate theories. More recently star polymers, dendrimers, and colloid particles with internal structure, in particular Janus, patchy, soft, and active particles, and a variety of mixtures with polymers or LCs, have been intensively studied. Simulation and experimental techniques are the dominant methods in the study of these complex systems.

In the case of ionic systems (molten salts, electrolytes, room-temperature ionic liquids (RTIL)) a still popular model is the primitive model (PM), where hard spheres are uniformly charged in a structureless solvent that is characterized only by the dielectric constant. In 1976 Stell predicted the phase separation into ion-poor and ion-rich phases with a corresponding critical point [42]. The nature of criticality in the RPM was a subject of a long debate [43, 44]. Stell argued that despite the long-range of the Coulomb potential the critical point in the RPM should belong to the Ising universality class [43]. His arguments were confirmed later by field-theoretic methods [45, 46], by the HRT and CV theories [47, 48] and by simulations [49]. In contrast to simple fluids, however, on a simple cubic lattice a tricritical point associated with a transition to the charge-ordered ionic crystal is present instead of the critical point of the gas–liquid transition [50].

Certainly, phase behavior in ionic systems is of fundamental interest, but in recent years the most intensive studies have focused on the effect of nano-confinement and its relation with electric properties such as capacitance, mainly due to its relevance for electrochemistry. In the study of effects of confinement on ionic systems the most popular approach is still based on the Poisson–Boltzmann theory and on its various modifications [51].

In the case of globular proteins and charged colloid particles, the sizes and the charges of the particles and counter-ions are different. Moreover, the Coulomb potential often competes with the specific short-range interactions. As a result, an inhomogeneous distribution of charges and/or masses is observed, especially in the case of large size and charge disparities [52, 53]. Recently size- and charge asymmetric models have been introduced and studied, with a special focus on the effects of confinement. In the case of very large size asymmetry only the larger particles with solvent-induced interactions are taken into account. In addition to the DLVO theory, model systems with effective short-range attraction and long-range repulsion (SALR) between the particles [54] have recently been under consideration. Such effective interactions are present in the case of complex solvents, containing in particular some depletion agents inducing strong effective short-range attraction. The latter may even lead to self-assembly into clusters, layers, networks or bubbles.

Polar liquids form another class of important complex systems that have been intensively studied. Stell with coworkers made a very significant contribution to this field, of which some part is reviewed in [55, 56].

Multiple phase transitions, polydispersity, solvation, association, self-assembly, chemical reactions, protein folding and crystallization, glass transition, gelation, percolation etc are just a few examples of the rich behavior that can be found in what are commonly known as complex fluids. The work of Stell and his collaborators has been very influential in many if not all of these areas [41, 57–66].

2. Articles in this issue

This special issue of Journal of Physics (Condensed Matter) contains articles by collaborators and former students of George Stell. His very broad interests are reflected in the variety of subjects on which his work has had a lasting impact. In the majority of the articles, different theoretical approaches are being introduced, modified or further developed, or well-established methods are applied to various systems [67–80]. Particular attention is paid to

systems with long-range interactions. The growing role of computer simulations in today's liquid state research is well illustrated in works [81–87].

Seven articles are directly or indirectly devoted to electrostatic interactions in liquids, one of George Stell's favorite topics. Thus we have contributions dedicated to the structure of RTILs by Yethiraj [86], to the evaluation of the correlation energy of the electron gas using the path integral formulation of quantum statistical mechanics, by Høye and Lomba [67], and to the effects of confinement on ionic systems [69, 71]. Specifically, Holovko *et al* [69] have studied the vapor-liquid transition of an ionic fluid in porous random media using the CV and SPT theories and the replica method. In [71] Lian *et al* resort to DFT in order to study electric-double-layer capacitors in ionophobic and ionophilic nanopores. The dielectric constant of saline solutions using various water models is under consideration in [72] using a diagrammatically proper ISM theory. Fructose aqueous solutions of concentrations ranging from 1.0 to 4.0 mol L⁻¹ are investigated by means of molecular dynamics simulations by Sonda *et al* in [84]. Finally, a two-dimensional fluid of dipolar hard disks in the presence of an electric field is studied by Caillol, Levesque and Weis [83] by means of Monte Carlo simulations. The dielectric properties of the system in the liquid and in the living polymer phase are investigated.

Complex systems, namely molecular fluids, asymmetric mixtures, self-assembling and associating systems are studied in [73–77, 81]. Munao *et al* extend the molecular closure formalism developed earlier for PRISM to a more general RISM theory and to molecular models with continuous interactions [73]. Structured molecules are investigated also in the work by Lee and Pellicane [74] using closures that implement a certain degree of thermodynamic self-consistency. In order to model a polyatomic molecule, they propose the 'composite-sphere' model, where the spherical symmetries of the individual pair potentials are preserved so that the isotropic OZ equation for multi-component mixtures can be used as a theoretical framework. Another thermodynamic self-consistent integral equation theory is applied to asymmetric hard-core Yukawa mixtures by Pellicane and Caccamo in the framework of the modified hypernetted chain approximation [75]. Though based in the same ideas as the SCOZA, these self-consistent thermodynamic approaches do not aim at a precise description at the critical point, but in any case they provide a far better description of the fluid behavior when compared to other microscopic theories. Finally, a self-consistent theory is also developed by Ciach and Gozdz [76] for systems with competing interactions leading to self-assembly. Their approach reproduces the anomalous features of the equation of state of a system with aggregated particles when the four-point direct correlation function is taken into account. Self-assembly and association, are the subjects of [81] and [77]. Static and dynamic properties of a two dimensional system with competing interactions are studied by Schwanzer *et al* [81] by MC simulations over a broad temperature range and for different densities. It is worth noticing that the SALR model of reversible self-assembly is in fact a 'softer' version of the shielded attractive shell (SAS) model of dimerization due to Cummings and Stell [63], generalized by Rescic *et al* in [77] to the case of polymerization. The combination of the resummed thermodynamic perturbation theory for associating potentials and the concept of thermodynamic self consistency has led to good agreement with simulations.

With distant roots in its work on probability functions in two-phase random media, Torquato presents in [78] a thorough investigation of disordered hyperuniform heterogeneous materials, a topic of deep impact for its implication in materials design and photonics.

Ben-Amotz [79], Raineri [80] and Shao and Hall [85] consider interfacial solvation, hydration, and adsorption of proteins on nanoparticles, respectively. Ben-Amotz focuses on questions regarding the role of direct (solute–solvent) and indirect (solvent–solvent) interactions in driving adsorption to macroscopic interfaces, and other macromolecular, colloidal, and mesoscopically heterogeneous domains. In [80] the Cumulants-based coupling parameter formalism for calculating free energy changes of the solute particle from the initial to final state beyond the linear response approximation is further developed.

Recalling his previous works on kinetic equations in collaboration with Stell in the nineties, Polewczak presents in [88] an analysis of various hard sphere kinetic models for reactive mixtures. The active granular fluid model of an embryo is studied by Gao *et al* [87]. In their model, cells are treated as circular particles connected by a predefined force network.

Depending on the external stress, particles tend to form chain-like structures or compact clusters with a remarkable similarity to *in vivo* observations.

Finally, phase transitions are the core issue of three contributions. Wilding proposes two simulation methods that—when compared to standard approaches—offer improved sampling and hence provide estimates of the gas–liquid coexistence parameters and surface tension for larger system sizes and lower temperatures [82]. Navascués *et al* [68] consider isostructural solid–solid transitions that can be induced in the presence of two-scale short range repulsive interactions, a work in which the Hemmer–Stell potential [57] is recalled. Here, the full phase diagram in the crystal region is obtained by means of a perturbation theory for classical solids and MC simulations. From a much more fundamental standpoint, Lebowitz and Scaramazza’s work [70] focuses on the presence of Lee–Yang zeros of the partition function in the negative half plane with an explicit calculation for a monomer–dimer system in 1D. They discuss the connection between the position of the zeros and the sign of the virial coefficients.

In summary, this issue covers a wide range of topics that have experienced enormous progress over the last fifty years, illustrating the key role of George Stell’s pioneering work and his long lasting influence on liquid state theory up to now and for years to come.

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