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Foreword

Advances in the microscopic theory of inhomogeneous fluids: A tribute to Stefan Sokołowski on his 75th Birthday

This special issue is a collection of papers written by friends and colleagues of Prof. Stefan Sokołowski in memory of 75 years from his birth. We remember and miss him. This foreward contains a brief description of scientific contributions by Stefan principally focused in the construction of microscopic theory of inhomogeneous fluids. It refers mostly to our happy times while working together along some of the topics mentioned below. We apologize that several issues of scientific interest have not been discussed in detail. Nevertheless, we hope that this text reflects motto of Stefan's personality and besides will attract the attention of younger readers to a set of problems unsolved so far.

The scientific work of Stefan Sokołowski covers a broad and coherent range of problems in the statistical mechanics of fluids, adsorption and soft matter. Although his earliest training was in chemistry, the core of his mature research belongs to liquid state theory and the microscopic description of inhomogeneous systems. Across five decades, he repeatedly returned to a number of fundamental questions: how the presence of a surface modifies the structure of a fluid; how directional interactions and association reshape interfacial behaviour; how confinement and quenched disorder change phase transitions; and how classical theoretical tools may be combined with computer simulations to obtain a predictive description of realistic complex systems. In this sense, his bibliography is not a collection of unrelated topics, but rather a sustained research programme built around fluid–solid interfaces, structure and phase behaviour of fluids, adsorption and confinement effects.

A notable feature of the works by Stefan is their methodological diversity. Sokołowski moved with ease between virial expansions, Born–Green–Yvon constructions, integral equation theory, density functional theory (DFT), lattice models, Monte Carlo simulations, molecular dynamics, and later coarse-grained and dissipative particle approaches. Yet the underlying aim remained stable: to develop microscopic theories capable of describing adsorption, phase behaviour, and structure in systems where nonuniformity is essential. This broad but internally consistent perspective explains why his work ranges from classical adsorption on heterogeneous surfaces to associating fluids, bulk and surface phase transitions, fluids in slit pores and disorder porous matrices, electric double layers, polymer-grafted interfaces, Janus particles and related systems.

Adsorption and the statistical mechanics of interfaces

Sokołowski's earliest publications were devoted to adsorption on heterogeneous solid surfaces and to the statistical mechanics of adsorbed layers [1, 2]. These studies emerged from the Lublin school of adsorption theory associated with W. Rudzinski and M. Jaroniec and addressed a classical but difficult problem: how to connect measurable adsorption behaviour with microscopic models of surface heterogeneity. Already at this stage one sees a characteristic feature of Sokołowski's later work — a preference for microscopic formulations rather than purely phenomenological adsorption equations.

This direction naturally developed into a more formal statistical-mechanical treatment of nonuniform fluids. In particular, his work with J. Stecki on virial expansions and interfacial structure examined the validity of reduced-dimensional descriptions for adsorption and the relation between density profiles and microscopic interactions [3, 4]. These papers are important not simply as early contributions, but because they define a line of thought that continued in later decades: adsorption should be understood as a problem in the statistical mechanics of nonuniform fluids.

An important advance came with the use of integral equation methods for fluids near walls. In a couple of papers from 1980 and 1983, Sokołowski studied the inhomogeneous pair distribution function for a hard-sphere fluid in contact with a hard wall, thus going beyond a sole focus on the singlet density profile [5, 6]. This shift from adsorption isotherms to correlation functions is scientifically significant: it placed interfacial adsorption into the broader context of microscopic liquid state theory. The influence of classical approaches to simple liquids is evident here, but the application to spatially nonuniform systems required both formal care and considerable numerical effort.

The interfacial line of research was developed further during the period of collaboration with W.A. Steele. Using local DFT, Sokołowski and Steele studied adsorption and ordering of fluids on crystalline surfaces [7, 8]. These papers belong to an important stage in the broader development of DFT for nonuniform fluids after the seminal review of Evans [9]. They also illustrate Sokołowski's typical style: the adoption of a powerful general method, but always in direct contact with specific interfacial problems. Work with J. Fischer later extended these interests to nonlocal functionals, density profiles of mixtures in pores, and the combination of DFT ideas with the method of integral equations [10–12].

A parallel line of work with A. Patrykiejew focused on adsorbed films on crystalline substrates, especially noble-gas and related submonolayer systems, where Monte Carlo simulations provided a route to microscopic phase behaviour at surfaces [13, 14]. Although these papers partly belong to surface phase transitions rather than fluid adsorption in the narrow sense, they are fully consistent with the interfacial research line: they explore how substrate structure and confinement control ordering, demixing, freezing and commensurability in low-dimensional adsorbed systems.

Associating fluids and directional interactions

Among Sokołowski's most important and sustained contributions is the theory of associating fluids. The general theoretical background for this field was provided by Wertheim's thermodynamic perturbation theory for site–site bonding [15, 16]. Sokołowski's role was not to simply extend the bulk theory, but to adapt and develop it for inhomogeneous, adsorbed, and confined systems, where directional bonding and spatial non-uniformity act simultaneously.

A key set of papers from the mid-1990s addressed the density profiles of chemically reacting or dimerizing fluids near hard and crystalline surfaces [17–26]. Taken together, these works form a coherent body of research on the influence of association on adsorption structure. They examine how bond formation changes near-surface density distributions, the role of specifically adsorbing sites, and the interplay between directional interactions and confinement.

Especially noteworthy is the methodological development represented by the nonuniform Wertheim Ornstein–Zernike equation [27]. Here, the physics of association is explicitly coupled to the correlation structure of an inhomogeneous fluid. This is more than a technical extension. When the associating fluid theory is used in the bulk, one may often separate the bonding thermodynamics from structural correlations; near surfaces such a separation is no longer justified. Sokołowski's work therefore contributed to a consistent microscopic theory of associating interfacial fluids.

At the same time, he developed integral equation approaches for associating Lennard–Jones systems [28]. These studies helped to clarify how association alters the structure and thermodynamics even before one turns to confinement. The next step towards confined associating fluids then followed naturally. In slit-like and cylindrical pores, associating Lennard–Jones fluids were studied by A. Huerta, B. Millan Malo, O. Pizio and Sokołowski, revealing changes in phase behaviour and structure caused by pore geometry [29–31]. Here, confinement does not merely shift coexistence lines, but it also leads to the network formation and may stabilize or suppress the bonded structures.

This work on associating fluids is also connected with lattice models and network-forming systems. In the paper by Patrykiejew, Pizio and Sokołowski on a two-dimensional network-forming lattice fluid [32], directional bonding was represented in a simplified approximation that nevertheless captured the unusual phase behaviour. Such models were useful not because they reproduced all microscopic details, but because they clarified generic mechanisms of network formation, ordering and fluid–fluid coexistence.

Another important aspect of this research was its broad methodology. In addition to integral equations and DFT, Monte Carlo simulations were repeatedly used to validate the theory and to provide microscopic insight [24, 33, 34]. This interplay of theory and simulation is one of the reasons why Sokołowski's papers on associating fluids remain valuable: they not only propose formalisms, but also test them on physically well-defined model systems.

Fluids in disordered porous media and replica Ornstein–Zernike theory

A second cornerstone of Sokołowski's scientific output is the theory of fluids in disordered porous media. In these systems the porous matrix is quenched while the fluid remains annealed; hence the problem differs qualitatively from ordinary equilibrium mixtures. The conceptual background was established by the replica treatment of quenched–annealed systems and the replica Ornstein–Zernike (ROZ) formalism developed by Given and Stell [35, 36]. Sokołowski was one of the researchers who applied this approach to adsorption and liquid state problems in detail.

A sequence of papers from 1996–1999 forms a remarkably consistent line of work on the subject [37–46]. These studies addressed adsorption, structure, capillary condensation and the role of confinement in random matrices. Their significance lies not only in the number of systems analysed, but in showing that quenched disorder can be treated with the same microscopic rigor as conventional fluid structure, provided the correct formalism is used.

The 1997 paper on associative ROZ equations is especially characteristic [39]. Here the problems of quenched disorder and association are combined in a single framework. This is scientifically demanding, because both features — specific bonding and matrix disorder — independently complicate the description of correlations. The paper shows one of Sokołowski's main strengths: his readiness to tackle problems where several nontrivial effects come together.

This research line was later widened in several important directions. One was the treatment of polydisperse templated porous materials, where the matrix itself carries a distribution of sizes rather than a single characteristic length scale [47]. Another was the extension to mixtures and especially to two-component fluids in disordered media, where matrix effects compete with demixing and interfacial coexistence [48, 49].

The 1998 studies of hard-sphere and Lennard–Jones fluids in disordered slit pores linked the formal ROZ theory with physically observable adsorption phenomena [41, 45]. In these works, matrix disorder alters the local structure and capillary phase behaviour in ways that cannot be understood by simply replacing the pore with an effective free volume. The matrix acts through correlations as well as steric exclusion, and this is exactly what the replica formalism captures.

The combined use of theory and simulation was again essential. In the paper by Duda, Henderson, Pizio and Sokołowski [42], simulations were used alongside the ROZ approach to test the predictions of the fluid structure. Such comparisons strengthened the credibility of the method of integral equations and clarified its limits. They also showed that Sokołowski's investigation of fluids confined in porous media was not merely formal, but was firmly connected to physically meaningful questions of adsorption, structure and phase transitions.

This line of work later connected to collaborations with J. Ilnytskyi and others at ICMP. The paper on the nematic–isotropic transition in a lattice model with quenched impurities [50] may be viewed as a conceptual extension of porous media thinking into orientationally ordered systems. Even when the model changes, the central idea remains familiar: quenched disorder modifies the effective conditions for phase transition and orientation ordering.

The papers on ROZ and fluids in disordered porous media occupy one of the central places in Sokołowski's bibliography. They also connect naturally with several related areas, including adsorption, confinement, associating fluids, ionic systems and later — porous structures decorated with polymers.

Electrolytes and electric double layers

Another major direction of Sokołowski's work concerns ionic fluids and electric double layers. This research was developed intensively during and after the collaboration with D. Henderson and co-workers. Primitive and solvent primitive models of electrolytes near interfaces provided the main physical context. These models are classical in liquid state theory, yet their nonuniform versions remain highly nontrivial because of long-range Coulomb correlations and surface-induced charge inhomogeneity.

The papers by Boda, Henderson, Fawcett and Sokołowski [51] and by Reszko-Zygmund, Sokołowski, Henderson and Boda [52] belong to this line. They analyse electric double layers using DFT and related methods and pay attention to contact values, density profiles and interfacial thermodynamics. These studies are scientifically important because they connect the theory of nonuniform fluids with electrochemically relevant observables such as differential capacitance and charge layering.

Later work extended this direction to slit-like of nanoscopic dimensions and to the solvent primitive model [53–57]. In these papers, Sokołowski returned to a question that repeatedly emerged in his research: how does confinement modify an already nontrivial fluid? For ionic systems, the answer involves the interplay of pore geometry, excluded volume, electrostatic screening and adsorption.

Surface phase transitions, films and confinement

Although adsorption and confinement are present in almost all of Sokołowski's research, there is also a more specific body of work devoted to surface phase transitions, films and low-dimensional ordering. The long collaboration with A. Patrykiejew produced a significant series of studies on the films adsorbed on crystalline substrates, particularly rare-gas and related model systems [14, 58–65]. These papers investigated demixing, freezing, domain-wall formation, commensurate–incommensurate behaviour and symmetry effects in two-dimensional or quasi-two-dimensional systems.

A comprehensive review of this field by Patrykiejew, Sokołowski and Binder [14] remains an important reference. It demonstrates that Sokołowski's contribution to interfacial physics was not limited to adsorption profiles or mean-field descriptions, but extended to subtle questions of ordering and finite-size behaviour on structured substrates. In methodological terms, these studies relied heavily on Monte Carlo simulation, but they were informed by the same microscopic and thermodynamic thinking that marked his work on fluids.

Polymer-modified surfaces and tethered chains

From the 2000s onward, Sokołowski increasingly turned to surfaces modified by grafted chains and to fluids interacting with polymer brushes. This was a natural extension of his long-standing interest in interfacial nonuniformity: a grafted layer creates a surface whose structure is itself soft, responsive and coupled to adsorption. Such systems are relevant both for fundamental statistical mechanics and for applications in chromatography, lubrication and materials design.

This field was developed in close collaboration with M. Borówko and T. Staszewski. Theoretical studies addressed adsorption from oligomer–monomer and binary solutions, structural changes of tethered layers induced by adsorption, and the behaviour of chemically bonded chain layers with active groups [66–73]. These studies illustrate a mature phase of Sokołowski's work, where DFT, simulation and a strong intuition for interface design were combined to analyse increasingly complex interfacial architectures.

A closely related direction involved the studies of the pores with their walls decorated by patterned polymer brushes using the mesoscopic off-lattice approach of the dissipative particle dynamics. These studies, performed in close collaboration with J. Ilnytskyi and T. Patsahan, were focused on the pattern-induced phase separation in a binary mixture of immiscible fluids and related structural transitions [74–76]. In these papers one can note how Sokołowski's earlier interests in confinement and adsorption migrate naturally into modern soft-matter settings.

Wetting, prewetting and water-like models

The problem of wetting and prewetting is present implicitly in many Sokołowski's studies of adsorption, but in the later period it became explicit and more sharply focused. In these papers the central question is no longer only how much the fluid adsorbs, but which exactly interfacial state is thermodynamically stable: partial wetting, complete wetting, or thin/thick film coexistence associated with prewetting. This focus is particularly natural within Sokołowski's research programme because DFT is one of the most powerful microscopic tools for analysing surface phase behaviour.

A sequence of works devoted to water and model water-like fluids, carried out in collaboration with L. Pusztai, W. Rżysko, V. Trejos, O. Pizio, illustrates this development [77, 78]. Water adsorption and phase behaviour in nanoscopic slit pores were analysed within DFT [79, 80]. Further studies considered the adsorption of water in slit-like pores with grafted chains and the associated solvation-force and brush-interdigitation effects [81]. A particularly important extension involved the wetting transitions and contact angles for water-like models on graphite-like and chemically modified substrates [82–84]. These papers are valuable not only because they revisit the classical wetting problem for a difficult fluid, but because they connect it to the broader Sokołowski programme on adsorption, confinement and structured surfaces.

Janus and hairy particles, patchy colloids and anisotropic systems

The later bibliography of Sokołowski includes another natural extension of the associating-fluid research: anisotropic colloids, Janus and hairy particles and patchy systems. Here, directional interactions are embodied not by abstract association sites alone but by explicitly anisotropic particles. The resulting self-assembly and interfacial behaviour link classical liquid state concepts to modern colloid science.

Studies in this area include Janus particles at walls modified with tethered chains, Janus dimers at liquid–liquid interfaces, bilayers of Janus-like particles, two-patch colloids in two dimensions, and related integral equation descriptions performed in collaboration with M. Borówko, W. Rżysko, T. Staszewski, A. Patrykiejew, Yu. Kalyuzhnyi [85–89]. Related studies include various types of hairy particles focusing on their structure near walls [90–92] and specific features of macromolecular gel structure depending on the decoration pattern of hairy particles, related to their catalytic applications [93].

These works mark an important extension of Sokołowski's research towards anisotropic colloids, Janus particles, and patchy systems, where directional interactions, confinement, and interfaces jointly determine the self-assembly and phase behaviour. Their significance also lies in combining simulation and theory to show that concepts developed earlier for associating fluids can be successfully carried over to colloidal systems of clear current interest.

Other directions

Several additional directions complement the main topics. The work on granular media with Herrmann, Gallas and Pöschel [94, 95] is perhaps the clearest example of Sokołowski's readiness to move beyond the conventional boundaries of equilibrium liquid theory. Although this project differs from his main adsorption and soft-matter lines, it still reflects his computational expertise and his ability to adapt methods from one field to another.

A further, perhaps less central, but scientifically important branch of his work concerns soils and agrophysical systems, often in collaboration with Z. Sokołowska and co-workers [96–100]. These papers again show a preference for connecting theory with physically complex real materials.

Considered as a whole, Sokołowski's scientific legacy lies not only in particular models or equations, but in a way of developing liquid state theory. He repeatedly chose the problems where interfaces, confinement or disorder made the physics richer and the mathematics harder, and he addressed them with a combination of microscopic theory and computational methods. The resulting body of work has a lasting value precisely because it is both methodologically rigorous and physically well focused.

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