

Jacob Nissim Israelachvili  
(1944 – 2018)

Jacob Israelachvili made fundamental contributions to our understanding of intermolecular and intersurface forces. Such forces are central to a broad area of science, spanning surface and colloid chemistry, condensed matter physics, materials engineering and the properties of 'soft' and biological matter. Israelachvili's pioneering work opened a new window that enabled a clear vision of a previously opaque domain, that of direct measurement of molecular and interfacial forces down to the angstrom level. This was many years before the advent of scanning probe microscopy, and Israelachvili's experimental methods and findings led to a transformation of the field.

Israelachvili, who died in Santa Barbara, California, on 20 September following a year-long struggle with cancer, was born in Tel Aviv, Israel. After attending boarding school in the UK and serving as a medic during his national service in Israel, Israelachvili studied Physics at Cambridge University, where he continued to a PhD with David Tabor at the Cavendish Laboratory. There he researched forces between interacting surfaces, an area that, while not exclusive to his interests, remained a central theme of his research career.

Following postdoctoral periods at the Cavendish and as an EMBO Fellow at the Biophysics and Karolinska Institutes at Stockholm University, Israelachvili moved in 1974 as a faculty member to the Department of Applied Mathematics at the Australian National University in Canberra. There, building on his PhD work, he developed an instrumental strategy for measuring directly forces between atomically-smooth surfaces across liquids, with or without attached surface layers, at controlled surface separations from the micron down to the angström level, which he termed a Surface Force Apparatus (SFA, also known as a surface force balance, or SFB). This nano-scale length domain is the one over which the properties of novel nanomaterials and of soft matter, including colloidal dispersions, are determined. In 1986 Israelachvili moved with his family from Canberra to Santa Barbara to take a position at UCSB as a professor in the departments of Chemical Engineering and Materials.

Intermolecular and surface forces are among the most fundamental issues in chemistry and physics, and indeed in biology, and have been investigated and exploited from antiquity. Thus, the steric stabilization of colloidal carbon black dispersions by gum arabic to form long-lasting inks, and the use of lubricants to slide heavy loads, both of which were documented in ancient Egypt, are two examples of such forces and how they were modified for practical use. In the modern era such luminaries as Newton, Maxwell, Boltzmann and van der Waals, and, in the past century, Hamaker, Langmuir, Landau, Overbeek and de Gennes, and many others, were major contributors to the theory of such forces. Classical investigations tended to focus on the forces that act between simple molecules in a gas, or between pure solids. However, the enormous area of intermolecular and surface interactions in and across liquids received relatively little attention. A major reason for this was the dearth of direct measurements concerning the nature of interparticle forces in such systems. While the theoretical treatments provided a framework for understanding such ubiquitous effects as van der Waals forces and double-layer electrostatic interactions between molecules and surfaces in liquids, and the effect of steric forces between polymer-coated surfaces, direct experimental confirmation of these forces at the molecular level was not available. Israelachvili was the first to enable and to carry out such measurements.

Israelachvili's prolific studies using the SFA revealed the nature of molecular forces and interfacial forces with a directness and resolution that had not previously been possible. Such studies also established the paradigms incorporated in subsequent techniques for studying molecular interactions, such as atomic force microscopy. At the same time, the SFA/SFB approach, now used by many groups world-wide, remains the gold standard for measuring forces between surfaces, or between the molecules which may coat them, across liquid media. The introduction of this method, and its exploitation for remarkable new discoveries, as well as for illuminating the origins of known interactions, is Israelachvili's chief monument. In the following I describe just a few of his main achievements.

Among the striking new molecular interactions discovered by Israelachvili and his co-workers were the oscillatory forces (known also as solvation or structural forces) that act between surfaces across simple liquids as they approach to contact, arising from the one-by-one expulsion of confined molecular layers. His experiments also revealed and clarified the crucial role of hydration layers in modifying interactions in watery environments. The resulting hydration repulsion overturns the long-standing scenario of surface interactions at nanometer separations in aqueous media. This scenario, which was based on a balance between attractive dispersive (van der Waals) forces and counterion osmotic pressure, and which predicted van-der-Waals dominance as the surfaces approach to contact, long held sway. As Israelachvili showed, however, hydration repulsion may overcome van der Waals attraction between charged surfaces in salt solution, so that they do not come into adhesive contact as predicted by the classic theory. Moreover, as seen subsequently by my own group and others, as well as in Israelachvili's laboratory, the related effect of hydration lubrication may account for the low friction between sliding tissues in animals. This in turn relates closely to such diseases as osteoarthritis, a debilitating condition associated with lubrication breakdown in mammalian joints.

In a different series of studies, Israelachvili and his associates first measured, and shed new light on the nature of the forces between hydrophobic surfaces, showing ultimately the short-ranged nature of such forces. Israelachvili was also directly involved with the development of new theories, such as the theory of self-assembly of amphiphilic molecules in solution, where both repulsive hydration and attractive hydrophobic and van der Waals interactions play a role – the paper on this is a citation classic that has become the 'standard model' in the field.

By coating the molecularly-smooth test substrates in the SFA with well-defined molecular layers, the forces between specific molecular species could, for the first time, be directly measured. Exploiting this, Israelachvili and his group first measured the biospecific ligand-receptor interaction potentials between biological molecules and surfaces directly, including the highly asymmetric time-dependent processes of binding (capture) and unbinding. These findings led to a profusion of work by other groups repeating and adding to this body of knowledge using similar or newer techniques which confirmed the fundamentality of Israelachvili's contributions. Non-specific molecular forces could also be readily measured. In some of the earliest direct studies on steric interactions between polymer-bearing surfaces, used widely for stabilization of colloids, Israelachvili, in collaboration with Matthew Tirrell

and myself, determined how forces between surface-attached chains varied with the transition between poor and theta solvency conditions for the polymer.

Following his pioneering studies of interfacial and molecular forces, Israelachvili extended his measurements also to time-dependent properties of confined liquids and molecular layers. Such studies revealed how the viscosity of organic liquids or oils could diverge as they were progressively confined between two surfaces. These related naturally to measurements of friction at well-defined surface separations and at controlled surface compositions and conditions. The SFA/SFB results in this area led the field of nanotribology, and enormously expanded our understanding of the nature of frictional dissipation between sliding surfaces in contact. In particular, Israelachvili and his team demonstrated the fundamental relation between friction and adhesion *hysteresis* in a wide range of conditions, as well as the conditions under which stick-slip friction would occur.

Israelachvili's impact on the field was propagated by the large number of students and post-docs who worked with him, many of whom achieved their own distinction as academic faculty all over the world. His book, *Intermolecular and Surface Forces*, now in its 3<sup>rd</sup> edition, was a landmark synthesis of classic and modern ideas, and has been hugely influential, as have been his some-500 research publications. He received many national and international awards for his work, including the Tribology Gold Medal, the top award in the field.

Israelachvili was a much-loved mentor and supervisor, as I witnessed at the meeting celebrating his 60<sup>th</sup> birthday, where, with a singular but characteristic lack of self-consciousness, he broke out in song following the dinner (as he also did at a formal Oxford College high-table dinner at another meeting that I attended). Israelachvili was a scientist of the utmost rigour and integrity, and demanded similar standards of his co-workers; at the same time he showed them great consideration. He was capable of remarkable acts of kindness, as when he took into his home and, with his wife, cared for an extended period for an overseas postdoc in his group who had developed terminal cancer.

I first met Jacob Israelachvili over 60 years ago, as we attended the same school, and our paths crossed many times since. His kindness, generosity of spirit and sense of humour will be very much missed, not only by his family, and by his many friends, students and colleagues, but by our community as whole.

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