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Historical Article

B. V. Derjaguin* and J. Theo. G. Overbeek. Their Times, and Ours

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Abstract. This year is the 25th anniversary of Boris Vladimirovich Derjaguin's death. The author was priviledged to know Derjaguin and Theo. Overbeek quite well. These two Giants of Colloid Science oversaw the evolution of the subject from a qualitative backwater to center stage in the now rapidly developing enabling discipline of modern physical chemistry. This is a personal account of events in their times.

Keywords. Derjaguin, Overbeek, DLVO, colloid science, polywater, cold fusion.

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The Schools of Derjaguin (1902-1994) and Overbeek (1911-2007) dominated Colloid and Surface Science completely for over 50 years. They did so deservedly because, to quote Overbeek in his (1948) book: *The science of colloids appears to be entering upon a new stage, which is less empirical, and where the experimental study of better defined objects will be guided rather by qualitative"rules"or"working hypotheses". The theory of the stability of lyophobic colloids, as developed in this book, may serve as an example of this development*" [3].[1]

Over the following half century and more, its acolytes and disciples clung to the core foundations because for the first time there was a firm mathematical scaffolding on which to build.

DLVO theory provided the backbone of colloid science since 1948 when Theo Overbeek published his thesis on the Theory of the Stability of Lyophobic Colloids with his supervisor Verwey. Germany had taken over Philips Industries when it occupied the Netherlands. Verwey protected Overbeek who worked on his thesis. At night, Overbeek, who had three young daughters, worked for the resistence arranging for Jews to escape. Had he been caught it would have meant instant death. Not an ideal research environment.

It took he and Annie his wife 30 years before they could face going across the border to Germany. I went there with them to the border.

Independently, Derjaguin and Landau published what is essentially the DLVO theory in Russian in 1941. Their paper is distinguished by the vitriol

* A more detailed history of Derjaguin's work can be found in the introductory paper of his collected works published by this author at Derjaguin's request [1,2]

and contempt with which they put down and dismiss an earlier 1937 attempt by Sam Levine from Manchester. Landau's diatribe is worth reading, a marvel of undeserved arrogant contempt from the great man Landau! It can be found in translation [1] or Landau's collected works.

Levine's sin was to replace a non linear charging process in the theoretical development of double layer electrostatic forces by a linear one. It is ironic that in 1961 when Dzyalshinski, Lifshtz and Pitaevski developed their quantum field theory of electromagnetic interactions between colloidal particles they made the same mistake. The whole impressive edifice then collapsed to semi classical theory [4].

(The implications of this error are prodigious and unrealised still, both for physical chemistry and physics generally).

In 1952 Overbeek and Derjaguin met at a Faraday conference in Britain, and their interchanges are all recorded in the Discussions of The Faraday Society record of the Meeting. No punches were pulled Overbeek always being a gentleman, and Derjaguin definitely not.

It was about priority and while in principle the Russians might have the better of it, a manuscript in Russian in Moscow during the war was not readily accessible. The two never got on. Derjaguin liked cowboy movies, and others that, shall we say, are less cultural, at least on his visit to Canberra many years later to the author's lab.

THE POLYWATER BUSINESS

This almost certainly costed Overbeek and Derjaguin their expected Nobel prize.

Around 1967-1968 Derjaguin seized on some work of a junior worker called Nikolai Fedyakin who discovered a new form of water he called polywater. Derjaguin, anxious for a Nobel prize, published it in Nature. This was against the advice of a number of colleagues. In particular an eminent Russian Academician, an infrared spectroscopist, advised against publication, as did V. Sobelev. N. Churaev with it. (I know this from my friend Vadim Ogarev who was rumored to be nominal Head (KGB) of Derjaguin's lab. Vadim was actually a very good scientist, and his father twice Order of Lenin, invented the Soviet U235 separation technology. Everyone knew everyone on those days, much as in the USA on the Manhattan project. It was at the height of the cold war. The whole polywater thing went viral.

The author heard Boris talk about it at NIH (National Institutes of Health, Bethesda, Maryland, US) in 1969. Like a precursor of climate change the earth might be consumed when all the world turned to sticky polywater! Eminent American quantum chemists "proved" that the sceptics were wrong; polywater, like climate change, existed. Brian Pethica, pragmatic British scientist, who knew about thermodynamics, proved the contrary [5,6]. Derjaguin withdrew. The Americans had a lot of egg on their faces and Derjaguin was never forgiven. Kurt Vonnegut's 'Ice-nine' in his 'Cat's Cradle' novel was based on the polywater "discovery" [7].

Felix Franks who edited 12 large volumes on water, wrote a racy book called "Polywater" about it in 1981 while on sabbatical in the author's lab [8]. It is somewhat biased, written during the Cold War. Felix worked as British spy in Germany in the war and hated Russian communists. Pethica took him to task in a review well worth reading [9].

The Americans did something more ridiculous than polywater at the same time, when President Nixon launched his war on cancer [10]. This was a new model for science reflected in today's fashion for computer simulation. The idea was that a billion dollars would be contracted to entrepreneurs who would set up labs run by technicians (black, underpaid) who would inject mice with all the conceivable chemicals in the world to see if they cured cancer. Brilliant. Simple. An unanticipated difficulty was that the entrepreneurs underpaid their resentful technicians who injected the mice at random and, in sympathy, allowed them to escape. The main frame computer to process the data was literally rusting when Adrian Parsegian and I who were at NIH at the time went to see the program manager. Shades of the present fashion for simulation.

Some years later, Derjaguin invited the author to participate in Moscow in one of his biannual surface forces conferences. I faxed back – no e-mail then - to say that the man he really needed was Jacob Israelachvili from my lab.

Jacob had done the first direct measurements of surface forces beween molecularly smooth mica sheets, with Tabor in Cambridge, before coming to Australia [11-13]. At the time he and Richard Pashley had pioneered surface measurements between surfaces in liquids. Derjaguin faxed back that regrettably while there were was accomodation for me, every hotel room in Moscow was competely booked out. Naturally I withdrew. It did not help that Russia and Israel had no diplomatic relations. And I was informed what was going by a friend, scientific attaché in Moscow at the time. Derjaguin had a gun at his head as it were.

But Jacob Israelachvili, not one to mince words, went to war writing outrageous letters to the Royal Soci-

ety and others protesting this (Soviet) discrimination against Israel (himself).

It is again ironic that practically all direct surface forces measurements, dating back to the famous work of Israelachvili and Tabor are wrong, due to incorrect theory, incorrect use of theory, multiplicity of parameters and so on. We shall have more to say on this below (the first experiment in the West with an accuracy of 2 Å, subsequently could not be fitted to Lifshitz theory of surface forces until was realised the radii of the two crossed glass cylinders used – one or two centimeters, and measured with a schoolchild's drawing compass - had an error of 100%) [11].

COLD FUSION

Derjaguin committed another sin, with the discovery by he and his coworkers of the phenomenon of cold nuclear fusion. This controversial observation takes place when deuterium containing ionic solids are put under mechanical loading, and was published after a great deal of careful work 3 years before the competing claims of nuclear fusion [14-16], lately widely dismissed, of some Americans, by a different method.

Derjaguins discovery was derided but may not be so silly. When a hard crystalline material cracks, the crack can be 2000 Å long and a tenth of an Å wide. Electrons ripped off in the high energy grinding process are a confined instantaneously high temperature plasma. Who knows?

This was explained to me by Derjaguin when I visited him at one time in Moscow on my way to Sweden.

He instructed me that I should tell Sture Forsen, Chair of the Nobel Prize Committee in Chemistry, that he, Sture, should give Derjaguin a Nobel prize for this. I did not have the heart to tell him that in the previous year I had chaired a Committee that reviewed research in physical chemistry in Sweden. And that in a light hearted concluding paragraph I had said that"*the Committee formed the distinct impression that very shortly the entire surface of Sweden would be covered in close packed array by NMR machines. And unless they were fitted with solar collectors no good would come of it.*"This gentle hint at over emphasis on nuclear magnetic resonance research went down like a lead balloon with my friend Forsen.

MOLECULAR FORCES IN RETROSPECT

At this point we can look back at the long period of "DLVO dominance" and see where it has taken us. We will then look at the implications of the polywater business.

Newton in a letter to his friend Bishop Bentley had this to say about forces: "*That gravity should be innate, inherent and essential to matter, so that one body may act upon another at a distance through a vacuum without the mediation of anything else, by and through which their action and force may be conveyed from one to another, is to me so great an absurdity, that I believe no man who has in philosophical matters a competent faculty of thinking can ever fall into it. Gravity must be caused by an agent acting constantly according to certain laws; but whether this agent be material or immaterial, I have left to the consideration of my readers.*"

Action at a distance a-la-gravity, or via electromagnetic forces transmitted by a virtual field through space remains a mystery disguised by equations. We have no such trouble understanding "hydration" forces. (Neighboring molecules, squashed tight push against each other).

Newton tried to measure molecular (surface) forces but gave up saying "surface combinations were owing" i.e. contamination. The work of the Russian School under Derjaguin and of the Dutch led by Overbeek brought it all into sight again culminating with the simultaneous dramatic publication of the Lifshitz theory and its extension by Dzyaloshinski, Lifshitz and Pitaevski and the first direct measurements of forces between molecularly smooth (mica) surfaces by Israelachvili and Tabor [11-13,17].

The triumphs are trumpeted and now imitated by armies of people practising force measurements with AFM machines, an innovation that came from our group at the ANU in Canberra [18]. The limitations of both theory and experiment are now apparent. They have been reviewed extensively elsewhere [19]. Indeed if anyone claims agreement with DLVO theory, his measurements are wrong. The foundations of the theory, are deeply flawed even of continuum solvent theory. They include pH, pK_as, interfacial tensions, activities, interparticle interactions, zeta potentials, etc.

Since the theory with condensed media is wrong, the measurements that claim agreement must also be incorrect - except for a gallant few.

People took both the theory of Overbeek and Derjaguin outside their own claimed domain of validity.

ANOMALOUS WATER AND POLYWATER

As already remarked "polywater " burst upon the scene in1969.

Very long range water structure, if we like bulk "hydration", a new form of water, anomalous water, is invoked with monotonous regularity whenever phenomena occur that are not explained by existing theory. The classical exemplar, exhibit one, is a jellyfish. The concept has long history going back to Thomas Young who used the concept of a liquid having bulk properties right up to a molecular distance from an interface. (That is an assumption of DLVO theory as spelt out by Hamaker in his thesis and a student of de Boer).

(Jellyfish have a longer history, more than 700 million years to the Edicara era. Anomalous water is a matter of supreme existence to them)

Young's 1805 theory of interfacial tension was taken over by Laplace, dressed in fancy equations that Young went to great pains to avoid, and incorporated into Volume 6 of his Mécanique Céleste [20]. (Laplace ignored contact angles !) Poisson, in 1831 disputed the assumption and introduced the idea that a surface had to induce a change – hydration, a decay in order - in near surface liquid molecules. The debate was settled in favour of Young–Laplace by Ockham's razor. Poisson's case was not helped by a mistake in a factor or 2 in his analysis. The story is outlined superbly in two magnificent articles by the Rev. Challis of Trinity College Cambridge (Newton's College). These much neglected reports to the British Association of 1834 and 1836, on Forces and Hydrodynamics in Colloid science – for which subject he coined the term "*Mathematical Physics, for this the highest Department of Science*" — deserve to be recognised. In the 1834 paper he suggested that measurement of molecular forces could be accomplished by using the new work of Fresnel on diffraction of light, as indeed it was by Israelachvili, Winterton and Tabor 150 years later [12].

George Peacock, Professor of Mathematics at Cambridge and Young's biographer, furiously accused Laplace of plagiarism - perfidious French ! And there the matter lay until the great 1876 article of James Clerk Maxwell on Capillary Action in the 9th edition of *Encyclopeadia Britannica*, updated by Lord Rayleigh in the 11th edition. Note to Editors – J. C. Maxwell, a Scotsman, a clade of humanity famous for its impecunity, preferred publication there as they paid very well.The paper is also in his collected works .

Maxwell resolved the issue decisively in favour of Poisson. And deduced the range of the exponentially decaying hydration forces – about 3 Å. This anticipated a similar advance of Stjepan Marcelja exactly 100 years later [21].

In no sense was this "hydration water" polywater. At the same time, 1876, Hofmeister was doing his seminal work on specfic ion effects and pondering if they were due to surface (adsorption of ions) or due to effects of some very long range water structure.

So if we like polywater, anomalous water was always in the air, and for jellyfish in the sea.

Following the advances in spectroscopic chemical analysis techniques which clearly demonstrated that 'polywater' produced in fine glass capillaries was actually a silicate based solution, R. M. Pashley, a beginning PhD student of Kitchener's [22] further proposed that thin 'polywater' films produced on condensation on silica based glass plates often gave adsorption isotherms which could be accurately described by Raoult's law. That is, the vapour pressure reduction could be caused by solutes created during the adsorption process, corresponding to about a monolayer of dissolved material from the glass surface. Even Michael Faraday considered water films condensed on glass to conduct electricity due to dissolved solutes. Pashley presented this work in Stockholm in 1978, and explained Faraday's isotherms. Boris Derjaguin commented that this may indeed be the proper explanation. Pashley was also the first to measure and interpret long range hydrophobic forces.[2]

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[2] For the measurement and theory of Van der Waals-Lifshtz Forces see also [23] where the film heght was studied vs film thickmess of liquid helium on vertical crystal of cleaved calcium fluoride can reasonably claim priority see also [24].

This is the preferred story in some quarters.

The Dutch, Sparnay *et al*. tried to measure the van der Waals forces between glass spheres, Dutch industry having centuries of experience in grinding smooth lenses.

Alas, the asperities on the glass surfaces were too large, larger than 60 Å, so the experiments were doomed.

Derjaguin had the advantage of them. His step father was the great Russian physicist P. N. Lebedev, the discoverer of light radiation pressure and a friend of J. Clerk Maxwell, got Derjaguin his start in research at age 17 in a Biophyics Institute. (Deryaguin was a school mate in Moscow of George Kistokowski, who emigrated to the U.S.A and became President of M.I.T. They met up again during the Cold War). Lebedev in 1894 quoted in Ref. 25 had written this amazingly prescient paragraph that clearly inspired Derjaguin:

"... of special interest and difficulty is the process which takes place in a physical body when many molecules interact simultaneously, the oscillations of the latter being interdependent owing to their proximity. If the solution of this problem ever becomes possible we shall be able to calculate in advance the values of the intermolecuar forces due to molecular inter-radiation, deduce the laws of their temperature dependence, solve the fundamental problem of molecular physics whether all the so-called 'molecular forces' are confined to the already known mechanical interaction of light radiation, to electromagnetic forces, or whether forces of hitherto unknown origin are involved."

Lifshitz with theory in 1955, and Abrikossova and Derjaguin with experiments in 1956, confirmed Lebedev's vision on molecular forces. The work was continued also by Dzyaloshinski and Pitaevski who developed – with Lifshitz - a theory of interactions between two planar dielectric surfaces separated by a liquid. Hydration was neglected, as the liquid in contact with the two surfaces was assumed to retain its bulk properties [25-27]. The theory used measured bulk dielectric properties as a function of frequency and so avoided the impossible donkey work of pairwise summation or simulation of molecular forces. Brilliant!

THE DENOUEMENT

Derjaguin's polywater was due further to contamination from human skin. The dismissal of polywater, to this day, was very shortsighted. Jellyfish do exist, and their "anomalous" water structure is probably due to cooperative very long ranged fluctuation forces between the extremely dilute conducting polymers that permeate the carapace of the creatures. The same is true for the curious anomalous exclusion zone of nafion, a fuel cell polymer [28], and for the remarkable sustained work on colloid stability of latex spheres of Norio Ise [29]. And for the endothelial surface layer on veins and arteries in physiology [30].

These matters are made more complex by this realisation that dissolved atmospheric gas, and its self organised state in nanobubbles everywhere present is responsible for most of what we label "hydrophobic" interactions, and is truly a hidden variable.

Anomalous water is not necessary.

The Greeks told us so with their four elements: fire (energy), water, earth and air but we ignored them.

The fourth element, air, is universally ignored. Its presence and the major effects of dissolved gas are missing from classical theory and open up whole new dimensions. Refs. 30-33 allow us to see how we can look forward to bridging biology and physical chemistry. Ref. 30 and the papers on novel water technologies in an upcoming special issue of this Journal are examples computer simulation is impotent to handle this realisation.

Descartes might well have said *I breathe: therefore I am* instead of *I think: therefore I am*.

There is more. Even without the extra dimension and hidden variable provided by dissolved gas we have moved far from the simpler world of DLVO. By that statement we include all of physical, colloid and electrochemistry.

For the intuition derived from on the classical theory assumes a fundamental ansatz –that electrostatic, double layer and dispersion (quantum mechanical) forces can be dealt with separately.

They can not, and the fundamental ansatz is wrong, violating two physical principles, the Gibbs adsorption isotherm and the gauge condition on the electromagnetic field [34,35].

Once electrostatic and dispersion forces are treated consistently however [31,33], much that was mysterious and handled by fitting parameters falls into place systematically; Hofmeister, specific ion effects and hydration for example.

The situation means however that the meaning and interpretation and intuition that we are familar with

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The Russians measured the forces between conducting cylinders at large distance, the "retarded" classical regime and so can claim priority.

But credit for the first measurements of non retarded van der Waals forces goes to Isrealachvili, Winterton and Tabor in Cambridge in 1969 [12]. Winterton quit to become an Anglican priest in Yorkshire. (Rabinovich and Derjaguin almost caught up). The story is intersting and deserves retelling. The inhibition to direct measurement going back to Newton was asperities on surfaces as well as contamination. Tabor, a Reader at Cambridge worked under a Professor Bowden, a Tasmanian who was interested in friction. They transferred to Melbourne, Australia, to work on radar as part of the World War 2 effort. Their job was to work on electrical condensers that use molecularly smooth mica. So Tabor conceived the idea of using sheets of this mica glued onto glass cylinders at right angles (the same geometry as a sphere on a flat surface to do the job.) and after the war back at Cambridge set to it. Distance was measured by the interferometric method suggested by the Rev. Challis in 1834. The forces showed up as deviations of spring on which one cylinder was suspended. And so a large industry was born. The technique therefore made the journey from Australia, back to Cambridge and then back to my Department in Canberra with Isrealachvili whence his departure to San Diego 12 years later rebadged it as an American invention! Tabor also invented the term "Tribophysics" for the subject of lubrication.

Note on the discovery of long range hydrophobic interaction.

The long range hydrophobic interaction between similar surfaces was first measured and reported by Israelachvili and Pashley in 1982 [38,39] based on their experiments using the Surface Forces Apparatus (SFA), which was developed by Israelachvili. Two symmetrical, cleaved and smooth mica surfaces were coated with a hydrophobic surfactant monolayer and the forces between them was measured in various aqueous electrolyte solutions. Comparing these measured forces with the expected van der Waals attractive forces, indicated that there was an additional attractive force an order of magnitude larger than any van der Waals force out to many tens of nm., which was identified as a 'long range hydrophobic attraction'. Since then, these attractive forces have been measured at separations up to several hundred nms. The origin of these forces has generated much debate, with the likelihood that their unexpectedly long range is probably related to dissolved gas cavitation created between the hydrophobic surfaces, evidence for which was also observed in the original studies [40].

In fact there are not one but many "hydrophobic" interactions that have different mechanisms [31-33]. There may be some dispute about who measured what first when and where.

Priority may go to our colleague V. V. Yaminski then in Moscow or to Pashley or both. Yaminski, no longer with us, has the distinction of being the only person ever to have read and understood J. Willard Gibb's collected works. The works are so turgid that anyone else who claims to have read them is a liar.

A consequence is that Yaminski, given a choice between choosing to describe a phenomenon in 50 words or 200 invariably chose 10,000, so honouring his hero and obscuring his works completely.

Some hydrophobic interactions involve cavitation, an important and completely ignored driver of enzymatic interactions [41]. Some involve nanobubbles at interfaces. Some involve surfactants, and electrostatics, some polymer bridging. Nearly all involve dissolved gas [33]. The most striking are the observations that emulsions become more stable when gas is removed. Hydrophobic proteins disperse when gas is removed. Certainly hydrophobic interactions generally disappear when gas is removed. Two other explicit examples are reported in Refs. 42 and 43. More theoretical and experimental results are found in Refs. 44-51.

A more recent publication (after 20 years study) is that of Kekicheff [52]. The sustained work on water structure near hydrophobic and hydrophilic surfaces, with and without salts by novel laser optical spectroscopic techniques is now likely to move center stage as we move to incorporate the new dimension provided by dissolved gas.

Figure 1. B. Derjaguin in his laboratory.

needs reworking , for pH, pK_as, buffers, interfacial tensions, intermolecular forces, zeta potentials, Hofmeister effects, hydration.

To put matter in perspective, recall a lovely quotation [36]: *"Over a hundred years ago, in the heyday of belief in self-sufficient progress, Paul Valéry insisted emphatically on the fact that civilisations are mortal. Fifteen hundred years before, St. Augustine echoed the same thought when in a simple sermon (and not in the famous work which contains one of the few philosophies of history that the West has produced), he summed up the true functions of earthly civilisation in a single illuminating phrase: 'an architect builds a durable house with the aid of a temporary scaffolding.' Civilisations are the impressive, complicated and bewildering scaffolding, machinamenta temporalia (Sermo 362.7). The edifice that arises above it is, he maintains, the Eternal City of God".*

We can interchange the word civilisations with scientific theories. The beliefs of one era evolve into others that are very far removed. It is therefore not usually possible to value scientific contributions for at least 50 years after their appearance.

But the new theories depend on the earlier foundations.

We have moved very far from where DLVO began and developed.

Finally then, for Theo. Overbeek and Boris Derjaguin, and their followers. We honour them still. Because like the ancient Egyptians they stood steadfast to that which they once believed to be valid. And by so doing they have laid us all under an obligation. We have work to do.

POSTSCRIPT

The author was priviledged to be a friend of both Overbeek and of Derjaguin.

He was honoured by the award of the Overbeek Gold Medal of the European Colloid and Interface Society in 2014 [37]. He was one 5 lecturers at Overbeek's 85th birthday celebrations, the others being Dutch. He has the Rebinder Medal of the USSR Academy of Science.

He experienced the many sad inhibitions to research on eastern colleagues during the Cold War. His most celebrated contribution to the cold war was when the Russians launched the Sputnik in 1957. The announcement on public radio by the Australian Broadcasting Commission occasioned the immediate formation of the St. George's College Astronomical Society (University of Western Australia) whose presidency he assumed. The Secretary, one David Muschamp a philosopher was delegated to report a sighting by the Society of the Sputnik traversing the clear night sky of the city of Perth. The announcement and publication of this "first" ever satellite sighting was received by the citizenry with acclamation. Sadly, a first example of fake news, you could see the thing, a gold coated sphere 15 cm diameter, traversing the Perth evening skies. The St. Georges College Astronomical Society, overwhelmed with its successes never met again.

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