# "Physics at the Interface" in Auckland

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## INTRODUCTION

From networks to nanowires, from jellium to Janus particles, from microfluidics to mass-spec: the range of research undertaken by the University of Auckland's *Physics at the Interface* group is broad, interdisciplinary, and often exotic. This breadth is unsurprising when the nature of the team is considered. Representing eight academics within a full-service Department of Physics, the group has roots in condensed matter physics, but we have diverse interests and collaborations which reflect up-to-the-minute global trends. The range of our work also reflects the nature of the local research funding environment. Three of New Zealand's Centres of Research Excellence are strongly represented:

- Te Pūnaha Matatini, literally 'the meeting place of many faces' is hosted in the Department – see Complex Systems and Networks, below.
- The MacDiarmid Institute for Advanced Materials and Nanotechnology has four investigators within the group.
- The Dodd-Walls Centre for Photonic and Quantum Technologies is represented by two investigators.

"At the Interface" aptly describes not only our research subject matter, but also our external collaborations, and the nature of our funding and administrative structures. In this article, the group's research and capability are summarized. Contributions from students and emerging researchers feature prominently. In addition to those pursuing an academic path, recent graduate researchers from the group have been employed by (for example) data science, medical technology, and nanotech startup companies.

#### MANY BODIES

Building on conventional condensed matter physics, researchers in the group are addressing a range of interesting materials problems. The universality of the many-body methods used to study materials is also being exploited in research into social networks and biological systems, for example.

#### **Electronic Structure Theory of Materials**

The advance of computationally feasible electronic structure physics has opened up new ways to understand the peculiar properties of compounds and materials that may even be inaccessible by experiments. This is especially true when going from bulk materials to surfaces and nanostructures. A prominent example is gallium nanoclusters, which have a melting temperature higher than the bulk material (Fig.1(a)). Only recently has it been possible to ascribe a conclusive physical explanation using density functional theory (DFT) and molecular dynamic (MD) simulations [1]. Those methods revealed quasi-2D structures forming in the liquid cluster phase, which lead to an entropically favoured solid cluster phase. The emerging quasi-2D gallium layers are interesting not only for promoting the higher-than-bulk melting temperature, but also because their pseudo-hexagonal structure bears resemblance to bilayer graphene.

Besides the lower stability of other phases, certain metal clusters are sometimes directly stabilized for another reason. In 1983 Knight et al. [2] found an abundance of certain sodium clusters that fulfil the shell closing criterion for jellium-like valence electrons. Such an electronic structure can even mimic electronic shell closing in atoms, which makes it possible to use those clusters as bonding partners. Such metal clusters are often referred to as superatoms, as an atomic model is helpful in understanding their interactions. One way of studying this is to look at substitution patterns in clusters, and the change of the electronic properties involved [3]. A major step towards understanding and exploiting the concept of superatoms is to understand how local electronic structure contributes to delocalized valence electrons. This is particularly challenging for transition metal clusters and the contribution of d-electrons. The superatomic concept could allow application of chemical intuition to determine new materials or to understand intercluster compounds [4] (Fig.1(b)).

In the transition from atoms and molecules to bulk materials, it is not only clusters that have exceptional behaviour. The inherent benefits of nanomaterials over bulk, for example, are often due to larger surface area and the emergence of steps and kinks. Often those features pronounce surface reactions. An effective description can often be achieved by modelling a small surface area. Interesting topics include adsorption of molecules as in heterogeneous catalysis. Naturally, these subjects are not limited to theoretical research. Our ongoing collaborations are addressing materials of experimental interest within the MacDiarmid Institute, including perovskites, bismuth and ZnO / SnO<sub>2</sub> / GdN [5] / BN [6] surfaces.



**Fig.1:** (a) Illustrative caloric curves of  $Ga_n^+$  clusters (n = 32-36) in comparison to bulk a-Ga explain greater-than-bulk melting temperatures. Reprinted with permission from Steenbergen and Gaston, Nano Lett. 16, 21 (2015). Copyright (2015) American Chemical Society. (b) Example of a cluster ( $Co_6Se_8(PPh_3)_6$ ) and a fullerene ( $C_{60}$ ) forming an inter-cluster compound ([ $Co_6Se_8(PPh_3)_6$ ]] that resembles the lattice structure of Cdl<sub>2</sub>. Reproduced from Ref. [4] with permission from the PCCP Owner Societies.

#### Skyrmions

Another research thrust concerns skyrmions, which are topological defects much like the magnetic domain wall and the superconducting vortex. If skyrmions can be made energetically stable they should occur within many systems showing broken symmetry and a characteristic set of excitations [7]. We are using theoretical techniques to study how skyrmion phases occur, and the skyrmionic texture, in a variety of materials. Textures include not only the Bloch-like skyrmion occurring in B20 chiral magnets, but also the Neel-type skyrmion. In bulk materials, the skyrmion lattice (SkL) phase generally occurs within a narrow region of the magnetic field-temperature phase diagram. The nature of the phase transition, the possibility of precursor phases, and the critical behavior on entering the skyrmion phase all require more detailed study.

How does dimensionality affect the skyrmion? When bulk, crystalline samples are thinned down the skyrmion phase becomes more stable, extending over a greater range of temperatures and applied magnetic fields. However, the observation of skyrmions in thin magnetic films has proved highly controversial. Any manipulation of skyrmions in devices will rely on magnetic ultrathin films and we are investigating the confinement of the SkL along with phases and spin textures realized in films [8].

We are also interested in how skyrmions evolve on different length, time and energy scales. It is expected that the skyrmion phase should promote a range of emergent physics. There is also recent evidence that the extent of the SkL phase is dependent on sample cooling rates. Investigations of non-equilibrium effects could allow us to stabilize these objects. Also, little is known about the effects of disorder on the skyrmion phase. Materials such as the B20 chiral magnet MnGe are thought to host disordered skyrmion phases formed from randomly oriented helices, but this disordered phase remains poorly understood [9].

Our methodology for studying skyrmions starts with exploiting the similarity between the SkL and another topological defect state, the vortex lattice in type-II superconductors. We employ multiscale theoretical and computational methods which involve spin models to identify the physics at play, followed by calculations of static properties and excitations such as magnons and phonons, which can be tested via neutron scattering. Lattice spin models and Monte Carlo simulations are used to establish phase diagrams. Spin dynamics simulations probe skyrmion dynamics, interactions, confinement effects, interactions with defects, and the magnetoelectric coupling in multiferroic films (Fig.2) [10].



**Fig. 2:** (a) Magnetic skyrmions and induced electric 'Skyrmions' in a static magnetic field. Closer top-views of a magnetic Bloch Skyrmion in (b) and an electric footprint 'Skyrmion' in (c). The color scale represents the magnitude of the z component. Reprinted figure with permission from Z. Wang and M.J. Grimson, Phys. Rev. B 94, 014311 (2016). Copyright (2016) by the American Physical Society.



## Aluminium



**Fig. 3:** MD results demonstrating contrast between the melting dynamics of different metals. Nickel (solid = black, liquid = blue) preferentially completely surface melts, while aluminium (solid = darker, liquid = lighter) only partially surface melts due to the anisotropic nature of its surface energies.

## **Theoretical and Computational Materials Physics**

We have tackled further problems relating to materials at the nano- and micro-scale via a combination of atomistic simulation and phenomenological models. For example, we study the melting of metal nanowires using molecular dynamics and the phenomenological theory of surface melting [11] (Fig.3). Studies of single nanowires are being extended to large nanowire networks, which can be used to create neuromorphic chips to simulate neural networks in the brain or for other electronic applications. We are also interested in the properties of nanostructured surfaces. Surfaces patterned with polymers can be designed to capture and collect water vapour as droplets, are being studied using MD and continuum models. We are interested in the equilibrium and dynamic properties of droplets on such surfaces, which could be used to collect drinkable water from water vapour, for example. Similarly we have developed a theoretical approach to study the effective catalytic properties of patterned surfaces (where activity may vary spatially) and tested this using kinetic Monte Carlo methods [12].

We are also studying Janus spheres, which are microor nanospheres that have hemispheres with differing physical or chemical properties. We have used molecular dynamics simulations to calculate the forces and torques on these particles as a function of their orientation to the fluid flow, and compared the results with theoretical models [13]. These calculated forces are now being tested experimentally, and could be used to find new ways to assemble and manipulate collections of colloidal particles.

## **Complex Systems and Networks**

The Department of Physics hosts one of the ten national Centres of Research Excellence: Te Pūnaha Matatini, the Centre for Complex Systems and Networks. Te Pūnaha Matatini is a trans-disciplinary research collaboration that brings together physical scientists with economists, ecologists, anthropologists, and even historians to study complex economic, social, and environmental systems.

We have recently conducted work on symmetry breaking and phase transitions in pedestrian flows [14]. Modelling the movement of crowds is important for urban design and event management. Our work focuses on the emergence of a preference for passing on the left or the right amongst pedestrians who regularly encounter each other, in an urban environment or otherwise. We have developed an Ising-like model to describe the corresponding breaking of symmetry and incorporated this into a standard 'social physics' simulation for crowd movement. The simulations show that the emergence of a preference can lead to significant improvements in pedestrian flow, but that this emergence may not occur if there is a very high variance in walking speeds.

Other recent work has used methods from statistical mechanics and network science to study political speeches in the New Zealand parliament [15], how undergraduate student outcomes in physics are related to gender and the corresponding portfolio of courses students take [16], and the network structure of New Zealand legislation. We are also very active in science communication and utilize a variety of media, including television, radio, and social media including twitter. Our work is funded both by government agencies as well as from private sector sources.

#### **Encoding Biological Information**

Biological systems regulate not only the construction of complex molecules, but also their highly heterogeneous molecular and phase composition, by processing information that is stored in the form of DNA sequences, dynamically uncoupled from the statistical mechanical processes that are computationally controlled. Understanding the origin and evolution of matter with these extraordinary information-processing capabilities, including self-constructive computation, is a major task of theoretical biology, but also raises the question of whether the spontaneous processes of evolutionary selforganisation that generate and maintain biological systems can be recapitulated in other environments where there is externally imposed selection for technological performance.

The MICREAgents project [17], funded by the European Union's Future and Emerging Technologies programme, succeeded in producing autonomous 100  $\mu$ m scale, electronically-controlled microreactor 'lablets' that selfassembled in aqueous 'solution' to perform specific electrochemical tasks, but fell short of achieving systems with an interaction 'ecology' complex enough to provide for the emergence of an electronic to chemical translation code [18]. In the meantime, our research in this general area focuses on details of the system of genetic coding employed universally by molecular biological systems. By applying methods of structural and sequence bioinformatics we are investigating the physico-informatic boundary conditions that were required to initiate the stepwise process whereby self-constructive computation emerged from predominant molecular disorder as the surface of the planet cooled to suitable temperatures approximately 4 Gyr in the past.

Having articulated the system of autocatalysis that underpins genetic coding, especially the production of the 'translatase' aaRS coding enzymes (the aminoacyl-tRNA synthetases), we are now seeking accurately to define the branching order of their ancient phylogeny through which the genetic code evolved from its initial binary specificity to the extremely versatile 20-amino acid alphabet that is found in the genetic language of every living cell. The theme of autocatalysis in functional protein production is also being explored in relation to the potential involvement of programmed genetic recoding by the aberrant form of the prion protein, which is the ætiological agent responsible for transmissible spongiform encephalopathies such as scrapie in sheep and Creutzfeldt-Jakob disease in humans.

## **DEVICES AND FABRICATION**

Here we profile four experimental teams within the *Physics at the Interface* group which bridge the gap between fundamental and applied research. Each group carries out interdisciplinary research using advanced experimental capability. Emerging themes include the use of photonics and microfluidic devices, development of sensors, applications in food and agriculture, and the physics of colloids, droplets, and biomaterials.

#### **The Photon Factory**

In the Photon Factory, physicists, chemists, biologists and engineers work together on problems ranging from fundamental molecular understanding through to microfluidic diagnostics. The fundamental studies have typically used femtosecond transient absorption spectroscopy (fsTRA) alongside computational chemistry. These techniques have allowed understanding of significant molecular absorption changes in aryl diphosphenes, depending on the molecular geometry. Using fsTRA to probe the introduction of boron into porphyrins enables investigation of singlet oxygen generation for use in photodynamic therapy. Using nsTRA (nanosecond transient absorption spectroscopy) to complement the fsTRA, we have elucidated electron transfer mechanisms and kinetics in artificial solar harvesting dyes. Recently, surprising emergent behaviour has been observed for interacting superconductors and magnets - offering new insight into major, long-term problems in materials physics.

Our transition between early stage and applied research is demonstrated by laser machining studies using short (ns) and ultrashort (fs) pulsed lasers. These lasers have been used to pattern reduced graphene oxide onto graphene oxide, revealing the mechanism of the reduction process [19]. Likewise, titanium dioxide surfaces have been patterned to template the formation of anodized titanium dioxide nanotubes with laser induced periodic surface structures (LIPSS), resulting in enhanced visible light absorption for solar photocatalysis that can be applied to different transition metal oxides. Femtosecond lasers are not yet in widespread use in industry due to slow cutting speeds, but they show superior precision and accuracy for various materials due to their 'cold-cutting' mechanism. The Photon Factory has studied cutting efficiency improvements in dielectrics, ceramics and biological hard tissue by tailoring the pulse wavelength, and spatial and temporal properties.

Our diverse knowledge of photonics and its applications also allows us to tackle high impact problems, and two start-ups are solving large problems in the dairy industry. *Engender* uses laser-based inertial microfluidics to sort bull sperm by sex, therefore allowing the choosing of offspring sex. *Orbis Diagnostics* focuses on real-time pointof-cow testing using centrifugal microfluidics to quantify biomarkers and exogenous analytes [20] (Fig.4). These start-ups aim to transfer research from the laboratory to the field in order to solve some of the major problems seen in the dairy industry.



**Fig. 4:** Simplified view of a centrifugal microfluidic setup for milk analysis in realtime, consisting of a strobe (A), camera (B), photodiode (C), LED (D) with the disk containing the microfluidic circuit (E) positioned at the centre with the rotor (F) and motor (G).

## **Biophotonics**

The biophotonics team responds to specific needs from the food and health sectors, and our research tackles most aspects of the sensing and imaging techniques being used in this area. Intrinsically, this work involves the physics of light-matter interactions, so part of our research is very fundamental, exemplified by work on dispersion measurements using optical coherence tomography (OCT) [21]. Examples of technological challenges include developing light sources for cost effective OCT sources with extended penetration depth [22]. We have used these sources together with our technique to measure dispersion of the eye [23]. More recently, we have improved the accuracy of the technique and developed a comprehensive understanding of dispersion measurement in OCT ([24], Fig.5). These results are part of a full research programme on early signs of blinding diseases while our work on polarization sensitive OCT has led to the study of early stages of osteoarthritis [25]. All our work is done in close collaboration with local and overseas biomedical researchers, and hospital-based physicians. We also work on quantitative bacterial detection using spectroscopic fluorescence with representatives from NZ industry [26].



Fig. 5: High resolution image of a rat eye that has been used to analyse the chromatic dispersion of the cornea and the aqueous humour.

## **Dynamic Microfluidics and Nanofluidics**

The Dynamic Microfluidics Laboratory features two high-speed cameras, alongside supporting capability suitable for studying the dynamics of liquids on microscopic scales. Principally, we are interested in drop impacts and capillary update. Both are experiments of high importance in applications which involve millimetric water drops. Drop impacts are carried out on interesting surfaces, such as superhydrophobics, soils, leaves, and regular microscale pillar arrays. In the latter case [27], the spreading symmetry of a drop is affected by the array (Fig.6). The Laplace pressure of a small drop enhances capillary uptake, even allowing passive uptake into a nonwetting capillary [28]. Our nanofluidics capability is highlighted by the study of submicron pores and pipettes. For tunable resistive pulse sensing, a particle-by-particle electrochemical sensing method [29], we have made particular contributions to measurement protocols. An important aspect of this work is understanding the interplay between ion and particle transport mechanisms and the sensing signal within a conical pore geometry. We have recently extended this work to similar apertures in glass pipettes. Capability has been developed akin to scanning ion conductance microscopy, but with the potential to mechanically probe nanoscale colloids.



**Fig. 6:** (a) Schematic for a typical drop impact experiment on to a regular square array of PDMS (polydimethylsiloxane) micropillars. (b) Asymmetries in the drop spread, viewed from a camera below the array. Reproduced from Ref. [27] with permission from The Royal Society of Chemistry.

#### **Ion Innovations Laboratory**

Major research areas in the Ion Innovations Laboratory include biophysics and photovoltaic devices, investigated through the lens of mass spectrometry. Understanding the biophysics of protein folding is of critical concern for disease recognition, drug design, and materials engineering. The current theory describes protein folding as being directed along a funnel-like energy landscape, which reduces the number of sampled conformations and allows proteins to fold in  $\mu$ s-s. Conventional experimental techniques have examined protein folding at timescales  $\geq$  ms. A new modified ionization source, theta nano-electrospray ionization (nESI), is being investigated as a microreactor for short-time mixing. In theta nESI, two flowing solutions are separated in a glass capillary by a septum wall, and allowed to interact at the exit orifice, before being detected by mass spectrometry [30]. The physical properties of the interfacing liquids determine the mixing dynamics. By selecting appropriate conditions, reaction times of  $\mu$ s to ms can be selected in realtime, and theta nESI can be used to interrogate changes in protein structure. The laboratory is also extending this technique to investigate the mixing dynamics of immis-



Fig. 7: Fabrication and analysis of photovoltaic devices and their degradation products.

cible solutions, as applied to electrospray deposition for solar cell device fabrication.

Organic and perovskite solar cells offer a more cost effective solution than conventional silicon based devices; however, they are lower in efficiency and less stable. Thin layer electrospray deposition by theta nESI is being investigated as a tool to mix and apply solutions and blends of materials. The laboratory is uniquely equipped to manufacture devices using new techniques, as well as to probe the device structure (Fig.7). In addition to traditional investigative techniques (e.g. SEM, TEM, AFM, UV-Vis), mass spectrometry is used to interrogate the interfacial interactions between different layers in solar cell devices, providing information on their stability and degradation products and pathways [31]. This generates a feedback loop for chemical selection, and informs the design of next generation devices.

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