PROFESSOR JULIE B STAUNTON

Paving the way for future breakthroughs

Professor Julie B Staunton shares the details of magnetic materials modelling with her disordered local moment theory, and the practical application of her research in magnetic refrigeration

What attracted you to the research field of materials' properties initially and how has this interest evolved over the years?

On university physics courses students learn about quantum mechanics and electromagnetism and how these theories can explain materials' properties. It fascinated me that such abstract ideas should, in principle, explain everything about a material; for example, what makes something a good electrical conductor or possess a permanent magnetisation or be mechanically strong.

There is then the tantalising challenge of developing this understanding until it is deep and useful enough for new materials with new functionalities to be designed on a computer. Since any material has septillions of electrons interacting with each other as well as heavier, slower atomic nuclei, this appears a completely intractable problem until one learns about a technique known as density functional theory (DFT). With the development of powerful supercomputers this method can now describe many materials in detail and is very widely used.

Can you elucidate the main theme that runs through your group's research?

The main theme is a description of the various properties of materials via a careful account of 'electronic glue', the complex fluid which is comprised of the many interacting electrons in a material. A strength of the work is that it is `first principled' and so needs no parameters other than the atomic numbers of the elements in the material. Many aspects can be tested in quantitative detail by a range of increasingly more sophisticated experimental measurements. The group's work concerns the development of the theory beyond standard DFT.

How have you adapted DFT in your disordered local moment (DLM) theory?

DFT is conventionally confined to treatments of materials at low temperatures. Our magnetic materials modelling work, although based on DFT, removes this constraint by identifying aspects of the electronic glue which form relatively slowly varying magnetic fields ('local magnetic moments'). It is the behaviour of these which determines overall magnetic properties as the temperature of a material is changed. By incorporating the effects of these local magnetic moments, whose orientations vary very slowly on the time scale of an electron travelling ('hopping') from one atomic position to another, and averaging over them, temperature-dependent effects can be described.

Do you plan to develop this approach into a tool to transform the process of discovering promising magnetic materials?

My group is currently building a computer code which is based on DFT-DLM theory for this purpose. We are producing it for magnetic materials groups to use for predictive modelling of a wide range of materials and to aid the search for new magnetic materials. Our first task is to get it to help with the optimisation of the magnetocaloric effect (MCE) which is harnessed in magnetic refrigeration. We are also incorporating those aspects of electronic glue that make the magnetisation of a magnetic material have a preferred or `easy' orientation with respect to how the atoms are arranged. Although these aspects are typically rather fine and difficult to capture accurately, they are crucial in determining which materials are likely to be useful permanent magnets.

Are there particular properties that may make magnetic refrigeration more desirable than conventional fridges?

Conventional fridges are based almost entirely on a compression/expansion refrigeration cycle. This is a technology more or less unchanged since its invention over a century ago and close to its fundamental performance limit – but well below what is thermodynamically possible. Prototype magnetic fridges demonstrated during the last decade, on the other hand, have been proven to be much more energy efficient and can span a broad temperature range around room temperature. Whereas conventional refrigeration uses hazardous liquid chemicals as refrigerants which eventually leak into the environment and are ozone layer depletive and global warming gases, magnetic refrigeration is much more environmentally friendly.

What are your plans in terms of the future trajectory of your research?

Over the short term, I want to complete development and testing of ab initio computational modelling of magnetic materials in consultation with experimental groups. This should result in the production of a robust, easy-to-use code with the results entered into a database for a wider set of researchers to use. Following on for this, I intend to examine spin-dependent and temperature-dependent electron transport for device applications, search for new permanent magnets (with reduced amounts of expensive rare earths) and investigate electronic effects near interfaces and defects and in nanostructures.

The laws of attraction

A University of Warwick research team has set out to create an *ab initio* quantum materials modelling tool that could help researchers around the world identify materials with special qualities like magnetism much more easily. The tool will have immediate applications in the field of magnetic refrigeration

DENSITY FUNCTIONAL THEORY (DFT) is a technique used to model materials and determine their structure and properties. The method has been popular in solid-state physics since the 1970s, and is widely used today in an advanced state enabled by computer technology. DFT is primarily used for determining the properties of materials by describing the electrons and their interactions with each other, as well as slower nuclei and external fields, producing diverse effects such as magnetism and superconductivity. DFT therefore allows physicists to identify materials with specific properties and put them to use in varied practical applications.

A NEW METHOD

Although DFT has become largely indispensible to physicists worldwide, it does have its limitations; the technique is typically confined to treatments of materials at low temperatures, and is therefore not suited to finding a material's temperature-dependent properties. In order to counter this shortcoming for magnetic materials, Professor Julie B Staunton has developed an extension of DFT that she calls disordered local moment (DLM). DLM allows temperature-dependent properties to be observed and described by incorporating the effects of these 'local moments' and averaging over them. A number of discoveries have already been achieved using DLM: "The technique has explained the varied magnetic structures of the heavy rare earth metals and quantified the magnetic refrigeration properties of gadolinium and the rare earth-free compound CoMnSi," Staunton highlights.

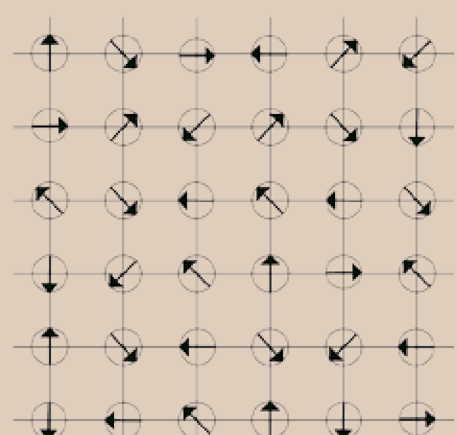
Staunton is now heading a team of research scientists at the University of Warwick with the intention of developing DFT-DLM into a computer program that magnetic materials groups will be able to use to predictively model materials. When the tool is complete, it will allow physicists everywhere to identify materials with useful electronic properties – and therefore put these materials to good use in practical applications such as magnetic refrigeration. However, the creation of the tool is made more difficult by the fact that the magnetic properties the Warwick team is interested in are very delicately poised; their modelling must track, in minute detail, the temperature dependence of the material, as well as indicating the effects of structural change.

Although several families of promising magnetic materials for refrigeration have been discovered, Staunton reveals that the search for magnetic materials, especially in the context of refrigeration, has tended to have been a heuristic one: "We hope to change this and establish our *ab initio* quantum materials modelling tool by working closely with leading experimental groups in the area".

AN INTERNATIONAL COLLABORATION

Staunton admits the goal of the Warwick team would be unfeasible without extensive collaboration. There are several key partners at both a national and international level, including electronic structure DFT groups in Munich, Halle, Budapest and the UK. Staunton has been particularly pleased with her group's active participation in the large and successful European PSI-K network, a charitable organisation dedicated to facilitating international collaboration in the field of *ab initio* materials modelling.

The Warwick team's research will primarily be a boon to other researchers, with their DLM-DFT technique paving the way for further discoveries in the field. However, unsatisfied with leaving



CARTOON OF DISORDERED LOCAL MOMENTS IN A MAGNETIC MATERIAL

INTELLIGENCE

THE CHANGING SHAPE OF MAGNETIC REFRIGERATION: AN INVESTIGATION OF ADAPTIVE MAGNETIC MATERIALS

OBJECTIVES

To describe the various properties of materials via a careful account of their 'electronic glue' using advanced computational techniques and resources available at the University of Warwick. Through a number of different projects, theoretical metallic magnetism is studied, with applications to permanent magnet design, spintronics, magnetic properties of nanostructures, magnetocaloric and magnetic shape memory materials and also electrocaloric materials.

KEY COLLABORATORS

Professor Dr Hubert Ebert; Dr Jan Minar; Dr Diemo Koedderitzsch; Dr Sven Bornemann, Ludwig-Maximilians-Universität, Munich, Germany • **Dr Arthur Ernst; Dr Sergey Ostanin; Dr Alberto Marmodoro**, Max Planck Institute of Microstructure Physics, Halle, Germany • **Dr Manuel dos Santos Dias**, Forschungszentrum Jülich, Peter Grünberg Institut, Germany • **Professor László Szunyogh**, Budapest University of Technology and Economics, Hungary • **Professor Dzidka Szotek; Dr Martin Lüders; Professor Leon Petit; Professor Walter Temmerman**, Science and Technologies Facilities Council (STFC), Daresbury, UK • **Dr Gavin Bell**, University of Warwick, UK • **Professor Vitalij K Pecharsky; Professor Karl A Gschneidner Jnr**, Ames Lab, Ames, Iowa, USA • **Dr Karl Sandeman; Dr Zsolt Gercsi**, Imperial College London, UK • **PSI-K** (Ab initio (from electronic structure) calculation of complex processes in materials) network – www.psi-k.org)

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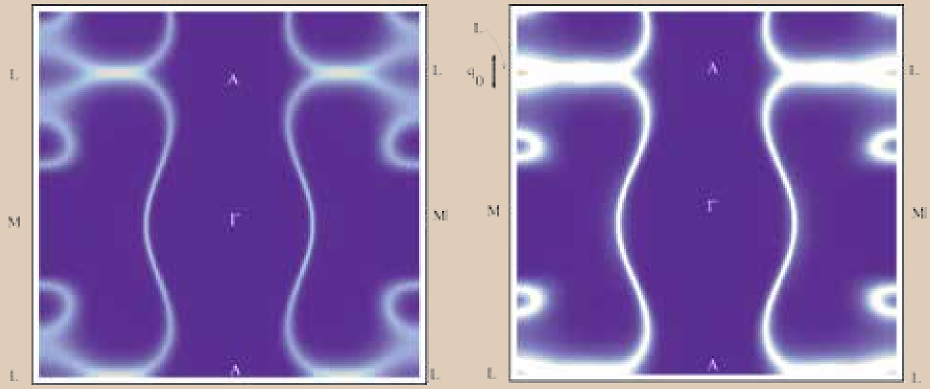
PROFESSOR JULIE B STAUNTON studied Physics at Bristol University after being introduced to the subject at Channing School by the science educationalist Joan Solomon. She carried out postgraduate research at Bristol, supervised by Professor Balazs Gyorffy, and postdoctoral work at Imperial College. Staunton became a lecturer in Warwick University's Physics Department in1984 obtaining a personal chair in 2001 and now heads the Theoretical Physics Group. She is married to Richard with one daughter, Karen.

WARWICK



Parts of the electron boundary (Fermi surface) of two rare earth magnets. The bright horizontal lines on the right signal that this metal is a helical magnet rather than a ferromagnet.

the discovery of practical applications to the physicists who follow, Staunton's group is already planning on pursuing one of the more pragmatic outcomes of their research themselves. Of particular interest to the Warwick researchers are materials that could produce a magnetocaloric effect – a novel magnetothermodynamic response that occurs when a changing magnetic field successfully alters the temperature of a material. This effect can be harnessed in order to maintain magnetic refrigeration, a developing technology that Staunton is keen to further. Hence, one of the group's main aims in this research direction is to use their modelling to suggest further candidates for materials that can act as refrigerants.

In their current state, magnetic refrigeration techniques are feasible but expensive – when the first room temperature magnetic refrigerator prototypes were tested in the 1990s, they relied on refrigerants based on rare earth metals such as gadolinium, which present a problem because they are expensive and difficult to obtain. Shortly afterwards, prototypes based around rare earth metals combined with silicon and germanium were tested, although these were still impractically expensive. Since then, much attention has been devoted to finding a magnetic refrigeration solution that does not significantly involve rare earth metals. With the Warwick team's new tool operational, these solutions could be improved and optimised.

GLOBAL WARMING VERSUS MAGNETIC COOLING

The impact of making magnetic refrigeration widely accessible could be very significant indeed. Magnetic fridges work by applying a magnetic field to the refrigerant which align its randomly orientated 'magnetic moments' and cause it to heat up. By removing this heat with a simple coolant like water, and then removing the magnetic field, the refrigerant can be made very cold, very quickly. In addition, magnetic refrigeration units have been shown to be more energy efficient than conventional fridges.

If magnetic refrigerators could be manufactured more cheaply, then they could be commercially available in the near future; which, given their higher capabilities alone, would be an exciting leap in consumer technology. However, the benefits would go far beyond increased capability – magnetic refrigerators would be less damaging to the environment in other ways as well. The harmful hydrofluorocarbons (HFCs) that most modern refrigerators use as coolant, for example, could be eliminated entirely.

As well as collaborating with a number of other research organisations to advance DLM-DFT techniques, the Warwick team is also partnering with several research consortia to further their progress in magnetocaloric refrigeration specifically. Two leading experimental groups are collaborating to support their project: one headed by Dr Karl Gschneidner and Dr Vitalij Pecharsky at Ames Laboratory in the US, and a second at Imperial College London headed by Dr Karl Sandeman. The former brings expertise in rare earths to the partnership, and has already pioneered room temperature magnetic refrigeration, while the latter specialises in improving the basic understanding of transitions between magnetic states.

THE POSSIBILITIES ARE ENDLESS

Another potential application of the tool which Professor Staunton plans to further investigate is in identifying magnetoplastic, or 'magnetic shape memory', materials – materials that respond to changing magnetic fields by altering their shape. The DLM-DFT tool will have the capability to quantify the magnetoplastic qualities of a material, and allow physicists to experiment with such materials at a nanostructural level. The practical uses of magnetoplastic materials are endless, as Staunton explains: "Such effects have a diverse set of applications, such as micropumps, sonars and magnetomechanical sensors".

At present, the group is studying an alloy of nickel, manganese and gallium that has been the subject of much experimental analysis in the past as a test case. Their findings will show whether or not their modelling approach can correctly indicate the optimal composition of the alloy at which the magnetoplastic effect peaks. As concerns refrigeration, they will not only be continuing their search for new refrigerants, but will also be attempting to nanostructure a large magnetocaloric effect by optimising the capabilities of the rare earth metals already being used in prototype refrigerators.