

MICROSCOPIC

LAWNS

OF NATURE

MICROSCOPIC LAWS OF NATURE

PREDICTING THE PROPERTIES OF COMPLEX SYSTEMS

MAURITS W. HAVERKORT

While it is possible to describe and predict the properties of a simple molecule or crystal using the microscopic laws of quantum mechanics, it is much more difficult to predict the behaviour of complex systems. This is because, at each complexity level, new phenomena can emerge that are not intuitively obtained from the underlying microscopic laws. They arise from competing interactions between the constituents that form the material. The resulting interplay between weak and strong interactions can lead to complex emergent properties. Developing and improving new methods to model such systems both at microscopic and macroscopic scales is at the heart of research carried out at Heidelberg University's Institute for Theoretical Physics.

T

The microscopic laws of nature that underlie the physical phenomena we see in our daily lives are known in great detail. The Maxwell equations, combined with the Schrödinger equation, provide a detailed description of the properties of small systems, like a single Hydrogen atom. When introduced in 1927, Schrödinger's theory of quantum mechanics was quickly and widely accepted as its predictions agree well with experiments. For example, the equations explain the discrete absorption and emission spectrum of atoms. When atoms in a diluted gas are heated, they emit light of a distinct colour. Each element emits light at a set of element-specific wavelengths. The wavelength and thus the energy of the photons emitted is given by the difference between the discrete allowed energies the electrons can have when bound to the nucleus. Schrödinger's equation not only explains why the light frequencies emitted and absorbed by atoms are discrete. It can also be used to quantitatively predict the energies of the photons that are emitted.

Interactions between fundamental particles

Schrödinger's equation is not the most precise microscopic equation to describe the interaction between fundamental particles. Inclusion of relativistic effects into the Schrödinger equation leads to the Dirac equation. The Dirac equation introduces the spin of the electron as well as the existence of anti-particles into the description of quantum mechanics. Besides this important qualitative change, the Dirac equation leads to quantitative changes. However, for the description of matter at ambient pressure and temperature, the changes induced by the relativistic effects captured in the Dirac equation are of the order of a few percent. For an even more precise agreement between theoretical predictions and experimental observations of small systems one needs to include the interaction between photons and matter as described in the theory of Quantum Electro Dynamics. It predicts the magnetic moment of an electron, related to its angular momentum, with at least 14 digits of accuracy. If one wants to calculate the binding energy of the electron in hydrogen-like atoms with even higher precision one needs to include the internal structure and extent of the nucleus. Nucleons are built up from quarks and gluons and their interactions are given by the equations of Quantum Chromo Dynamics. The model that brings all



PROF. DR MAURITS W. HAVERKORT joined Heidelberg University's Institute for Theoretical Physics in 2016 and has served as the institute's director since 2022. He studied physics at the University of Groningen (Netherlands) and in 2005 earned his doctorate at the University of Cologne, where he subsequently worked as a postdoc. In 2008 he accepted a research position at the Max Planck Institute for Solid State Research. Following a stint as visiting researcher at the University of British Columbia in Vancouver (Canada), he transferred to the Max Planck Institute for Chemical Physics of Solids in Dresden. Maurits Haverkort's research focuses on understanding and predicting the properties of quantum materials with competing local and itinerant interactions. The numerical methods he developed are used to predict, understand and optimise a number of different material properties in various fields of research, among them solar cells, nuclear waste and battery research.

Contact: m.w.haverkort@thphys.uni-heidelberg.de

particles and interactions together is known as the Standard Model of particle physics, which was developed in the 1970s and has since been tested to a great extent.

Testing the Standard Model

One can test the Standard Model by looking at systems with higher energy, as done in particle colliders, or by looking at systems with very high precision. Our group contributes to high-precision tests by providing accurate calculations of quantum few body systems. We perform high-precision theoretical calculations of multi-electron ions, in collaboration with Dr Zoltán Harman of the Max Planck Institute for Nuclear Physics, in the framework of IsoQuant, a DFG-funded Collaborative Research Centre on quantum systems in extreme conditions based at Heidelberg University. Another research collaboration where our group contributes to high-precision physics is the Research Unit ECHO lead by Prof. Dr Christian Enns and Junior Professor Dr Loredana Gastaldo, also from Heidelberg University. ECHO aims to determine the neutrino mass, one of the few undetermined parameters in the Standard Model. An isotope of the element Holmium can decay to an excited state of Dysprosium and a neutrino. The total energy released is shared between the neutrino and the Dysprosium ion. Comparing the experimentally observed decay probability as a function of the neutrino energy to the theoretically predicted value allows one to determine the neutrino mass. In our group we calculate the decay rate as a function of the released neutrino energy. We solve the Dirac equations for the ground-state of a Holmium nucleus with 67 electrons and for the dynamics of this system after the nucleus decays to Dysprosium.

High-precision measurements are exciting tests of the Standard Model. At the same time, for the prediction of most of the physical phenomena we see in our daily lives, the Schrödinger equation, with some first-order corrections given by the Dirac equation, is accurate enough as an underlying microscopic theory. This notion led Dirac in 1929 to state: "The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known". That said, it is astounding that these few and relatively simple equations should explain the wealth of physical phenomena we see around us. On the one hand, to be able to predict the properties of a molecule or crystal, starting from the Schrödinger equation, sounds plausible. On the other hand, using the Schrödinger equation to predict the behaviour of complex systems such as human decision-making sounds rather implausible. As stated by Nobel Prize laureate P. W. Anderson in his paper 'More is Different': "The ability to reduce everything to simple fundamental laws does not imply the ability to start from those laws and reconstruct the universe." At each complexity level, new phenomena can emerge that are not

intuitively obtained from the underlying microscopic laws and are best treated by new, overarching theories.

Predicting the properties of complex systems

One challenge in predicting the properties of complex systems, starting from the microscopic laws of quantum mechanics, originates from the complexity of these equations. In classical physics, the state of a material is described by the positions and velocities of all particles involved. The number of variables needed to describe a system scales linearly with the number of particles. With modern computers one can model really large classical systems, such as classical molecular dynamics simulations of bio-molecules or electro-hydrodynamics calculations of star formation. In quantum mechanics, the state of a material that describes all its properties is given by a wave-function. The wave-function for a single particle can be written as the sum over a set of basis-functions each multiplied by a complex number. One can generate a wave-function for n particles by multiplying n different one-particle wave-functions. This however is not a general n -particle state. A general n -particle state is given by a sum over all possible different products of n one-particle states, each of them again multiplied by a complex number. The number of terms in this sum scales (worse than) exponentially with the number of particles. A quantum superposition, i.e. the fact that an n -particle quantum state is not given by the product of the wave-functions of the individual particles but by a sum over different products, is essential for the functioning of a quantum computer. At the same time, it hinders the possibility to calculate and predict the behaviour of a quantum system on a regular computer.

The challenge in predicting the properties of complex systems comes from the fact that some complex systems do not seem to behave in a way one would expect judging from the behaviour of their constituents. The electrons in

many molecules and metal crystals will start to oscillate in an oscillating electromagnetic field. In complex molecules like photosystem II, which consists of several thousand carbon and hydrogen atoms as well as a centre containing four manganese ions, many more effects happen. The former follows the simple behaviour one would expect from the underlying microscopic laws, the latter is not straightforward to predict.

This raises the question of how to model complex materials such that we can predict their properties. Modelling macroscopic quantum systems requires new theories that are based on the microscopic theory but are not necessarily equivalent. For molecules and solids, one normally uses a theory whereby the atomic nuclei are treated classically and form a symmetry broken state, i.e. a static state where some symmetry properties of the underlying macroscopic laws are not present in macroscopic systems at the lowest reachable temperatures. The electrons are treated quantum mechanically. The equations for approximately 10^{23} electrons moving on a background of a broken symmetry potential generated by the atomic nuclei is still vastly too complicated to be solvable. Furthermore, not all symmetry broken phases are structurally driven. Magnetically ordered phases are symmetry broken phases in which the lattice, i.e. the atomic positions, only plays a secondary role. There are many more electronically driven phase transitions present in nature. There is thus a need for further effective theories.

Mean-field theories

Some of the most successful and widely applied effective theories are known as mean-field theories. There is a whole class of mean-field theories, with the Hartree-Fock approximation and Density Functional Theory within the Kohn-Sham orbital formulation as prominent examples. Within these theories, the interactions between all

“It is astounding that these few and relatively simple equations should explain the wealth of physical phenomena we see around us.”

particles are replaced by a single potential that is equivalent for all particles. The many-body problem one needs to solve reduces to the problem of how a single particle moves through such a potential. This determines which single particle quantum states are occupied. The occupied quantum states determine the potential. The problem one needs to solve thus depends on the solution one finds. The final solution is a self-consistent solution, whereby the solution of the equations defines the potential used in these equations. Mean-field theories are in general solvable, even for very large systems. Mean-field theories do allow for symmetry broken phases and can, for example, predict magnetic phase transitions. In some sense the approximate macroscopic theory describes the reality better than the intuitive solution one would expect from the underlying microscopic theory.

Mean-field theories do not always work. All states described by mean-field theories can be written as a single product of one-particle states. There is thus no (non-trivial) quantum entanglement possible on a mean-field level. Mean-field theories tend to work when the resulting physics of the complex system follows the intuitive behaviour of its constituents. In turn, many of the more complex phenomena we see in the real world are not well described by mean-field theories.

Mean-field theories tend to fail when there are competing interactions between the constituents that form the material. The equations that determine the physical properties of a material can be split into different parts. The Schrödinger

equation contains a term related to the kinetic energy of the particles as well as a term describing the interaction of the particles. Each part by itself is often simple to solve. The equations remain solvable when one of the interactions is much stronger than the other. When the different terms in the Schrödinger equation are neither too strong nor too weak as compared to each other, complex behaviour emerges. The kinetic energy and Coulomb interactions between electrons and atomic nuclei are fundamentally given by the underlying microscopic theory. The Coulomb interaction strength depends on the distance between the particles. At low densities and large average distances, the Coulomb interaction is weak compared to the kinetic energy. At the densities found in atoms, the Coulomb interaction can be the leading term. As a result, one finds bound states between the atomic nuclei and electrons. The resulting effective interaction between the electrons bound to an atom can be weak or strong compared to the kinetic energy. The resulting interplay between weak and strong interacting systems leads, in the domain where these interactions compete, to the very rich phenomena we see in the physical world around us.

Determining the effective strength of an interaction is not always obvious. Electrons are Fermions. As such, by the Pauli principle, no two electrons can be in the same quantum state. For many materials such as the Alkali metals, aluminium or the semiconductors we use in our electronics, the interaction between the electrons is in principle strong compared to the kinetic energy. At the same time, the scattering between most electrons is blocked while

“When the different terms in the Schrödinger equation are neither too strong nor too weak as compared to each other, complex behaviour emerges.”

MIKROSKOPISCHE NATURGESETZE

DIE EIGENSCHAFTEN VON KOMPLEXEN SYSTEMEN VORHERSAGEN

MAURITS W. HAVERKORT

Die Teilchen, aus denen unser Universum besteht, folgen den erstmals von Erwin Schrödinger formulierten Gesetzen der Quantenphysik, die heute als Standardmodell der Physik gelten. Präzise Tests der mikroskopischen Wechselwirkungen zwischen Teilchen zeigen eine erstaunliche Übereinstimmung zwischen den theoretischen Berechnungen und der Realität im Experiment. Jedoch ist es mitunter schwierig, auf der Grundlage dieser mikroskopischen Naturgesetze die Eigenschaften makroskopischer Systeme vorherzusagen, die manchmal den physikalischen Gesetzen scheinbar zuwiderlaufende Verhaltensweisen zeigen. Diese emergenten Eigenschaften der Materie treten vor allem dann auf, wenn verschiedene das System beschreibende Wechselwirkungen im direkten Vergleich weder zu stark noch zu schwach sind.

Dank hochpräziser Berechnungen isolierter Quantensysteme mit extremen elektrischen Feldern können wir die mikroskopischen Interaktionen zwischen Teilchen des Standardmodells mit hoher Genauigkeit untersuchen und sogar Forschungsansätze über das Standardmodell hinaus erarbeiten. Um die Eigenschaften komplexer Quantensysteme vorherzusagen zu können, müssen neue theoretische Methoden und Modelle entwickelt werden.

In Heidelberg entwickeln wir numerische Verfahren und führen präzise Berechnungen für isolierte Quantensysteme durch, so dass wir die Grenzen, innerhalb derer zutreffende Vorhersagen möglich sind, ausloten und weiter verschieben können. Des Weiteren entwickeln wir effektive Methoden zur Vorhersage der Eigenschaften komplexer Materialien. Unsere Methoden wurden in einem in Heidelberg entwickelten Softwarepaket namens QUANTY gebündelt, das mittlerweile von Wissenschaftler:innen weltweit genutzt wird; zu den Anwendungsfeldern zählen etwa die Solarzellen- und Batterieforschung oder die Suche nach Lösungen für eine sichere Lagerung radioaktiver Abfälle. ●

„Die Konkurrenz zwischen unterschiedlichen effektiven Wechselwirkungen im Material führt zu komplexen emergenten Eigenschaften.“

PROF. DR. MAURITS W. HAVERKORT hat seit 2016 eine Professur am Institut für Theoretische Physik der Universität Heidelberg inne und ist seit 2022 dessen Direktor. Nach seinem Physikstudium an der Universität Groningen (Niederlande) wurde er 2005 an der Universität Köln promoviert, wo er anschließend als Postdoktorand arbeitete. Ab 2008 forschte er am Max-Planck-Institut für Festkörperforschung in Stuttgart, bevor er 2013, nach einem Aufenthalt als Gastwissenschaftler an der University of British Columbia in Vancouver (Kanada), an das Max-Planck-Institut für Chemische Physik fester Stoffe in Dresden wechselte. Maurits Haverkorts Forschung konzentriert sich auf das Verständnis und die Vorhersage von Eigenschaften von Quantenmaterialien mit konkurrierenden lokalen und itineranten Wechselwirkungen. Die von ihm entwickelten numerischen Methoden werden zur Vorhersage, zum Verständnis und zur Optimierung einer Vielzahl unterschiedlicher Materialeigenschaften in verschiedenen Forschungsbereichen eingesetzt, darunter Solarzellen, Atommüll und Batterieforschung.

Kontakt: m.w.haverkort@thphys.uni-heidelberg.de

the states they would like to scatter to are occupied. As a result, the effect of the strong interaction is still weak compared to the kinetic energy scale. One finds that these materials are often well described by mean-field theories. This leads to a great understanding of their properties such that we can use these materials to build devices.

Developing new methods

The question of how to predict material properties when their behaviour is not well described by mean-field theories is at the heart of the research performed in our group. For small systems, we study how well the microscopic laws can be solved. This can be compared to several high precision measurements done in Heidelberg and eventually lead to the improvement of the microscopic theories. For macroscopic systems we use a multitude of different methods. At the moment, there is not a single best approach. One promising approach is to represent the state of a material not by its wave-function, which for modestly large systems would require one to store more complex numbers than atoms in the universe, but by how the system reacts to external influences. The functions that describe how a system reacts to external influences are known as the response functions. With the use of diagrammatic techniques known from Quantum Field Theory one can describe the response of a large system by equations that only include the response functions of its constituents. This results in a set of equations one needs to solve self-consistently. The dynamical mean-field theory is one such approximation, but many more theories along this line are possible.

We develop new methods and implement them into computer codes. As there is no simple single solution, we developed an open-source script language, QUANTY, where calculations in quantum mechanics can be done using different methods and effective theories. In order to determine how well these theories can predict reality we compare results for calculations on specific materials to different experiments. The type of materials and research topics where our implementations are used ranges wildly between fields and materials. For example, we have investigated fundamental interactions in Nickel or Cobalt containing compounds that can be used in batteries. We investigate lanthanide compounds in order to obtain a fundamental understanding of their magnetism and the competition between local and delocalised states. Furthermore, in a recent collaboration with the Karlsruhe Institute of Technology, we investigated how the chemistry and physics of Actinides can be understood. These findings are important for the safe storage of nuclear waste or understanding how such elements bind to different ligands, which could be useful for future applications in nuclear medicine.

For all the latter examples of research we do in our group, it is the competition between different effective interactions

“It is the competition between different effective interactions in the material that leads to complex emergent properties.”

in the material that leads to complex emergent properties. The behaviour and properties of these materials cannot simply be extrapolated from the microscopic laws of physics. The microscopic laws of physics are not violated, but they cannot be solved to predict the properties of interest for these materials either. In order to predict material properties, we use a set of effective theories. We get many good results, but none of these effective theories are perfect. The main part of the research in our group is therefore focused on the improvement and development of new effective methods to predict material properties. ●