Probing Electron Dynamics in the Ultrafast Regime

Dr Oliver Busch
Dr Franziska Ziolkowski
Dr Jürgen Henk
Prof. Dr Ingrid Mertig

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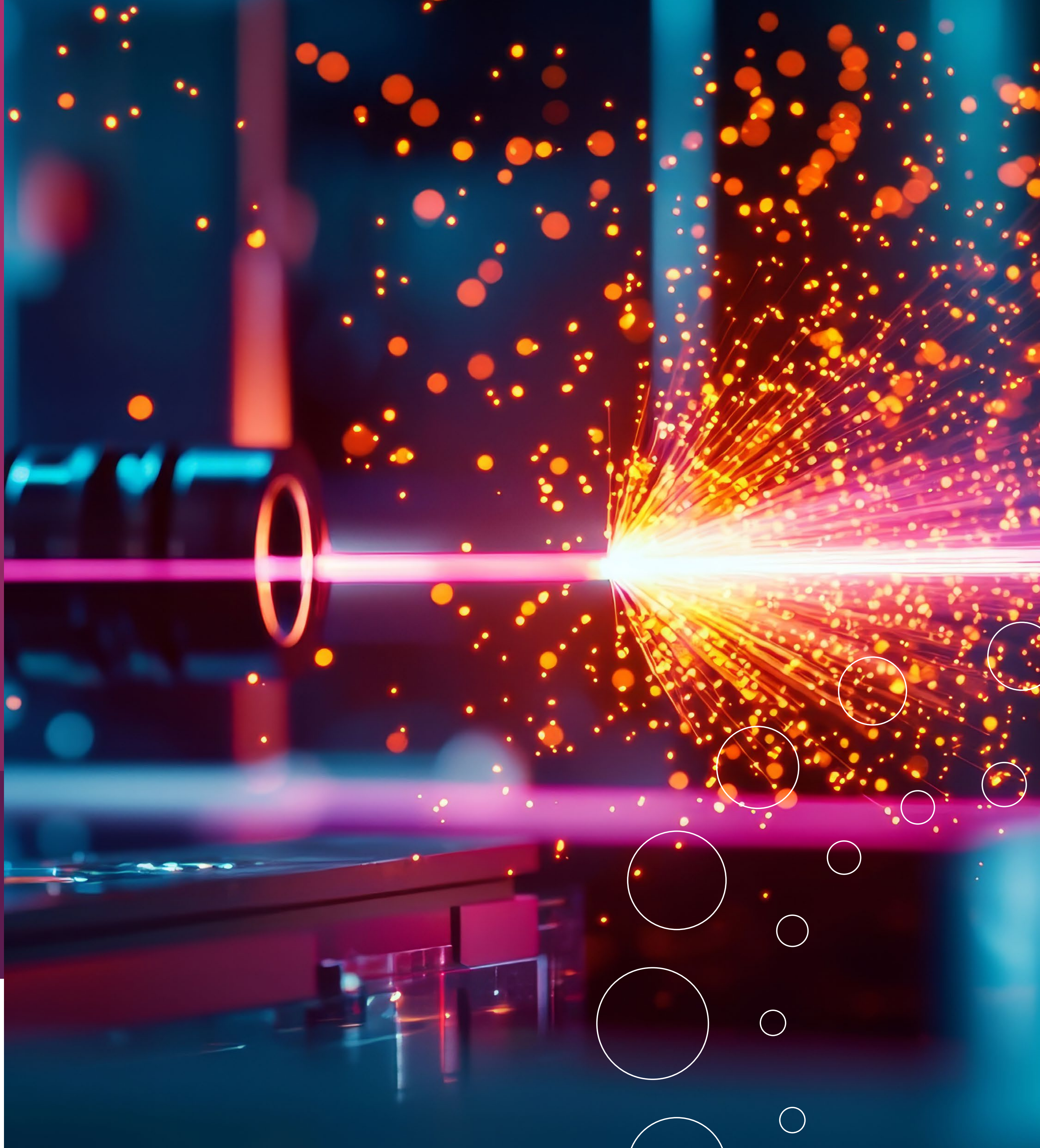


Probing Electron Dynamics in the Ultrafast Regime

In the atoms that make up the matter around us, negatively charged particles called electrons have properties such as spin and orbital angular momentum. Researchers at Martin Luther University Halle-Wittenberg have developed a theoretical framework which allows them to simulate the dynamics of the spin and orbital angular momentum of electrons in materials when probed with an ultrafast laser pulse. Using this framework, they are able to simulate different materials and improve our understanding of dynamics on an atomic scale.

Simulating Ultrafast Electron Dynamics Induced by a Femtosecond Laser Pulse

Everything around us is made up of subatomic particles, with positive protons and neutrally charged neutrons forming the nuclei of atoms, surrounded by negatively charged electrons. These electrons have a property known as spin, which describes the intrinsic rotational motion of the particle around its own axis. Understanding the dynamics of this spin helps us to further understand how these electrons move throughout different types of materials, or what happens at different material interfaces, and helps scientists to develop future experiments to examine these properties further.

Researchers from Martin Luther University Halle-Wittenberg look at ultrafast electron dynamics. They consider timescales of fractions of a second, considering femtoseconds, or one quadrillionth of a second, to picoseconds, or one trillionth of a second. By considering the dynamics when a bright, powerful light pulse of this duration from a laser is incident on a material, the team can learn more about properties like magnetism and currents generated within the material.

The research team have developed a theoretical model which can simulate these dynamics. To carry this out, they firstly represent their sample as a cluster of atoms – this could be a one-dimensional chain, a two-dimensional structure with a regular pattern, or a three-dimensional structure with defects or irregularities in the material structure. Each cluster consists of a regular arrangement of atomic nuclei and electrons and has specific boundary conditions based on its proximity to other clusters.

From this set-up, the researchers define a Hamiltonian, an operator which tells us about the energy in the system. By finding the eigenstates of this Hamiltonian, which tell us about the possible outcomes of measurements of the quantum state, the team can analyse electron dynamics. A simulated laser pulse is defined and then applied to the cluster of atoms, which causes transitions in the orbital structure of the electrons, or the structure of the electrons around the nucleus.

Another aspect considered in their simulations is thermalisation. At some time scales, bosons around the electrons, like phonons and magnons, can couple with the system and try to reach thermal equilibrium with the electrons. The team account for this through a bosonic bath, or by simulating that their atom cluster is coupled to these bosonic particles. By considering the laser pulse and this bosonic bath, they can then evolve the system during the pulse and simulate the changes to the electron and spin dynamics – and to reflect this, they call their approach 'EVOLVE'.

Simulating Electron Dynamics at Interfaces

The team use their simulation model to look at an interface between cobalt and copper. From this, they are able to consider the occupation of the orbitals or consider the energies at which electrons no longer remain in their standard orbit around the nucleus. Using these occupation profiles, they can also consider the magnetic properties of the sample. Prior to the pulse, cobalt is a ferromagnetic material containing electrons in its outermost orbitals which give it this property, whereas copper is not magnetic. As the pulse is applied, the team can simulate how the cobalt becomes demagnetised due to the motion of the electrons at the interface.

This demagnetisation occurs due to spin-transfer across the interface between the cobalt and the copper, which the team are able to simulate. The laser pulse leads to a flow of electrons from the copper to the cobalt, causing a demagnetisation of the cobalt and a slight increase above zero in the magnetisation of the copper. The research team are able to simulate this through looking at the currents, both in the flow of charge and the flow of spin angular momentum, between the atoms in their simulation.

Simulating Dynamics of Orbital Angular Momentum at Interfaces

As well as being able to simulate the dynamics of the electrons' spin angular momentum, the team also consider the dynamics arising from the orbital angular momentum of the electrons. Whilst spin is the intrinsic property of the electron itself, the motion-related orbital angular momentum tells us about the orbit of the electron around the nucleus at the centre of the atom. The team look at how ultrafast laser pulses can potentially induce orbital angular momentum, and how this is distributed across the sample.

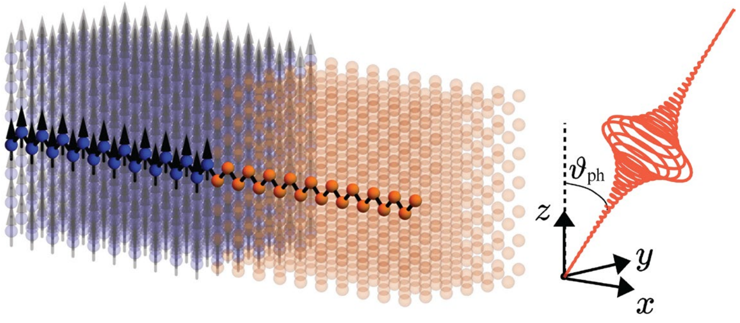
To carry this out, the researchers use the 'EVOLVE' approach as outlined above, applying it to copper and cobalt, as well as to a copper-cobalt interface. For these investigations, the team use a circularly polarized ultrafast laser pulse. Based on the geometry of the sample and the laser pulse, they can consider components of the orbital angular momentum which are induced along the longitudinal and transversal directions. The team also consider the spatio-temporal properties, or the change in the location of the induced orbital angular momentum over time, and can simulate the respective orbital currents also.

The team find that, for their simulations of copper, all the components of the laser-induced orbital angular momentum oscillate rapidly, even after the laser pulse is completed. In particular, the component in the longitudinal direction oscillates only a little, at a value which could potentially be measurable in future experiments. For the cobalt, the team observe some oscillation during the laser pulse, but this stops after the pulse. They use this to suggest how the electron dynamics described above contribute to the demagnetisation of cobalt, with little effect from the orbital angular momentum.

The research team also evaluate a copper-cobalt interface. They consider how the polarisation of the laser pulse affects the transfer of spin and orbital angular momentum across this interface, highlighting how having a p-polarised laser pulse would be needed to transfer orbital angular momentum between copper and cobalt.

Simulation of the Ultrafast Orbital Hall Effect

By investigating the orbital angular momentum in two-dimensional samples in the presence of an ultrafast laser pulse, the research team can also investigate the orbital Hall effect. The Hall effect originally shows how, if we place a flat piece of some electrically conducting material between the north and south pole of a magnet and then connect the sample to a battery so current flows through it, we produce regions of positive and negative charge –or a potential difference– in the sample. A similar effect can also be found for electron spins, where if a current flows through a sample, opposite spins accumulate on each side of the material at right angles to the direction of the current. The research team investigate the orbital counterpart of this, looking at how orbital angular momentum accumulates in the sample.



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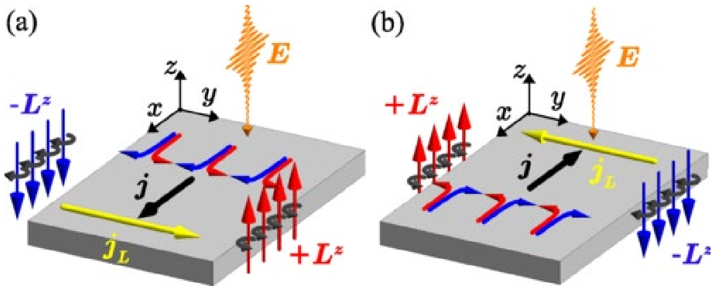
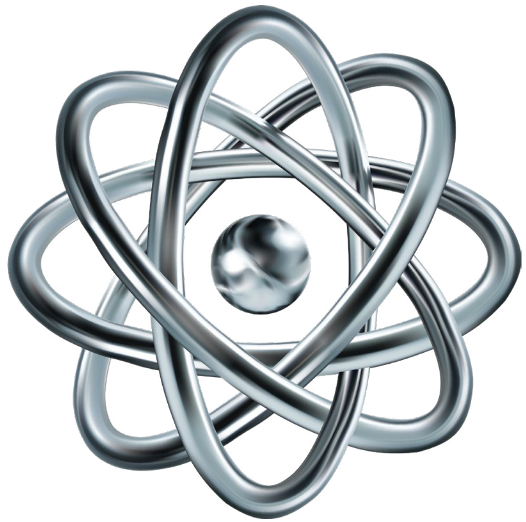
By considering the laser pulse and this bosonic bath, they can then evolve the system during the pulse and simulate the changes to the electron and spin dynamics – and to reflect this, they call their approach 'EVOLVE'.



Snapshots of the ultrafast orbital Hall effect in a two-dimensional sample (gray rectangular solid). (a) A linearly polarized femtosecond laser pulse impinges perpendicular to the surface (along the z axis) onto the sample. The laser's electric field E (orange), oscillating along the nanoribbon (±x direction), causes an oscillating longitudinal charge current j, which, at the moment depicted here, is oriented in +x direction (black arrow) and is deflected toward the ribbon's edges; confer the three representative pairs of current filaments (bent blue and red arrows). Hence, orbital angular momentum (OAM) Lz is transported across the ribbon, giving rise to a transverse OAM current jL (yellow arrow along +y direction). As a result, Lz is accumulated with opposite orientation at the edges (upward red and downward blue arrows). (b) Half a laser's period later, the reversal of E reverses the orientation of j, jL, and Lz The periodic field switching creates an ultrafast (on the femtosecond scale) orbital Hall effect.



To do this, they use a nanoribbon structure of copper, a thin two-dimensional structure which is assumed to be infinite in one direction. Their 'EVOLVE' approach is again used in this simulation, and the electric field of the laser leads to a current along the nanoribbon. By looking at the sites of the nanoribbon, the team consider if the site has orbital angular momentum, and how the orbital angular momentum changes over the duration of the laser pulse. They identify the transversal flow of an electron current to the edges of the nanoribbon, and how within half of the cycle of the laser pulse, positive orbital angular momentum is transported towards the upper edge of the nanoribbon, and then in the next half of the cycle it is transported to the lower edge. This transversal orbital angular momentum current is a signature of the orbital Hall effect and oscillates with the electric field of the laser pulse.

The researchers also study how the orbital angular momentum is distributed over the nanoribbon, to identify this accumulation at the edges of the material, which is characteristic of the orbital Hall effect. To do this, they analyse whether a site on the lattice is occupied or not and, if a site has orbital angular momentum, over time as well as looking at how these quantities are distributed in space and time. Their simulations highlight how both quantities show higher values at the edges as opposed to in the middle of the nanoribbon. From these simulations, the team reveal how the ultrafast orbital Hall effect here is laser driven. Using an oscillating field, they are able to simulate both currents of orbital angular momentum throughout the material and how the orbital angular momentum accumulates.

Overall, through their development of a theoretical model to study laser-induced electron dynamics and the creation of their 'EVOLVE' simulation framework, the research team have been able to investigate multiple electron properties in different materials with atomic and femtosecond resolution. From looking at electron dynamics to understand demagnetisation and spin currents at the copper–cobalt interface, through to the consideration of orbital angular momentum and the orbital Hall effect, the team have demonstrated how their model can help us to learn more about these dynamics and system requirements. With new experimental techniques being developed in this field, being able to effectively simulate a range of materials and the effects of different laser pulses is an invaluable tool for the solid state physics community.

Article written by Imogen Forbes, MSci

MEET THE RESEARCHERS

Quantum Theory of the Solid State Group

Martin Luther University Halle-Wittenberg, 06108 Halle (Saale), Germany

Dr Oliver Busch

Dr Oliver Busch obtained his Bachelor of Engineering in Mechanical Engineering at the Duale Hochschule Baden-Württemberg in 2014. After this, he completed his Bachelor of Science, Master of Science, and PhD in Physics at the Martin Luther University Halle-Wittenberg in 2018, 2020 and 2024, respectively. Following the completion of his doctorate, Dr Busch now works as a postdoctoral researcher at Martin Luther University Halle-Wittenberg, as part of the Quantum Theory of the Solid State group. His work focuses on electron dynamics, looking at simulations where the sample is excited with an ultrafast laser pulse using the group's computational framework, EVOLVE. By considering laser-induced transfer of the charge, spin and orbital angular momentum in materials such as copper and cobalt, Dr Busch is able to reveal more about the role of the laser's polarisation and the impact of material interfaces on these electron dynamics. Dr Busch also works on the spin and orbital Hall and Edelstein effects on ultrafast time scales.

CONTACT

oliver.busch@physik.uni-halle.de

https://www.physik.uni-halle.de/fachgruppen/qtfk/

Prof. Dr Ingrid Mertig

Prof. Dr Ingrid Mertig completed her Diploma in Theoretical Physics in 1979 before obtaining her PhD in Theoretical Physics in 1982, both at TU Dresden in Germany. Her academic education also includes a postdoctoral position at the Joint Institute for Nuclear Research in Russia. She completed her Habilitation at TU Dresden in 1995. Prof. Mertig was invited as a Guest Professor to several groups worldwide including New York, Paris, and Tokyo. She became a Professor at Martin Luther University Halle-Wittenberg in 2001 and built up the group Quantum Theory of the Solid State. She is currently a Wilhelm and Else Heraeus Senior Professor. Her research interests include spintronics, transport theory and density functional theory, and she has published widely in these areas. Her contributions to science have been recognised through numerous awards, including the Max Born prize in 2024.

Dr Jürgen Henk

PD Dr Jürgen Henk obtained his Diploma in Physics in 1986, followed by his PhD in Physics in 1991, both from the University of Kiel in Germany. Following his PhD, he worked as a postdoctoral researcher in Germany and Sweden before becoming a staff scientist, firstly at the Max Planck Institute in Halle, and at Martin Luther University Halle-Wittenberg since 2012. Since 2015, he has been the Head of "Halles Schülerlabor für Physik", a program which gives high school students an opportunity to go to the Martin Luther University to supplement their physics classes. Alongside his role as a staff scientist, Dr Henk also completed his Habilitation in 2005. Dr Henk's research focuses on the theory of electron dynamics on the ultrafast timescale, electron transport in low-dimensional systems and electron spectroscopy, and he has published extensively in these fields. In 2021, Dr Henk's contributions were also recognised as an outstanding referee for the journals of the American

Dr Franziska Ziolkowski

Dr Franziska Ziolkowski obtained her Bachelor of Science and Master of Science from Martin Luther University Halle-Wittenberg in 2015 and 2018, respectively. During her Master's degree, she was a student assistant at the Institute of Mathematics and Physics and following her Master's degree she became a research assistant at the Institute of Physics at Martin Luther University. Dr Ziolkowski also obtained a prestigious Studienstiftung des deutschen Volkes scholarship during her undergraduate and Master's degrees and received academic awards from the university. Dr Ziolkowski completed her PhD in Theoretical Physics, also at Martin Luther University Halle-Wittenberg in 2024, and she continues to work here as a postdoctoral researcher in the Quantum Theory of the Solid State group. Dr Ziolkowski's work has been widely published, and she focuses on electron dynamics, including working on the development of the EVOLVE computational framework used to simulate the effect of ultrafast laser pulses on electron dynamics in materials and at material interfaces. .

KEY COLLABORATORS

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FURTHER READING

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