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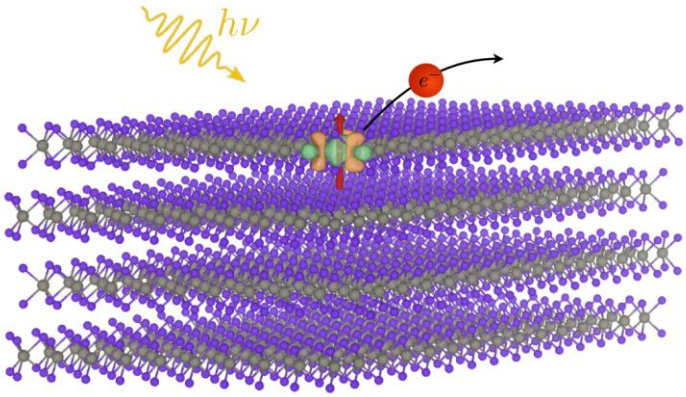
PRESS RELEASE

A paradigm shift in calculating the spectral properties of semiconductors

A new approach allows calculating the band structures of semiconductors in a simple and low-cost computational manner. The research has been published in Physical Review Research.



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Accurate and predictive first-principles calculations of spectral properties of materials, such as the band gap of semiconductors, are key to understanding, discovering and designing materials for countless applications including electronics, energy harvesting, and photonics.
In spite of the enormous theoretical progress and the exponential growth of accessible computing power over the last fifty years, such simulations are still challenging in many ways. Established methods, based on Feynman diagrams, are limited by their complexity and computational cost, and often neglect a consistent treatment of spin-dependent interactions. This is particularly relevant if considering spin-orbit coupling, a relativistic effect that is strong in presence of heavy chemical elements and often play a major role in cutting-edge scientific or technological applications.
In a new article just published in the journal Physical Review Research, Antimo Marrazzo from Scuola Internazionale Superiore di Studi Avanzati in Trieste, Italy, and Nicola Colonna, from the Paul Scherrer Institute in Villigen, Switzerland, advocate for a paradigm shift by introducing a new functional approach that allows calculating band structures of semiconductors in a simple way and at low computational cost, even in presence of spin-orbit coupling or complex magnetic

configurations. This development will make computational screenings of materials databases much more efficient and accurate, and enable simulating complex materials under more realistic conditions, such as in presence of defects or at finite temperature.

Physicists, chemists and materials scientists have made enormous progresses in the last decades in their capacity to simulate, explain and predict the properties of materials using computational tools that rely on the fundamental laws of quantum mechanics. But there are still blind spots, and one of them has to do with calculating the spectral properties of some semiconductors.
The staple method for materials simulations, density functional theory (DFT) can effectively predict the properties of many materials at their ground state, which is the stable state at the lowest possible energy level. But it does not give access to spectral properties, such as how the electrons of a material interact with light (i.e., photons). There are other theories that can solve the problem, like Green's functions and many-body perturbation theory, often based on the so-called Feynman diagrams. But they are very complex and often do not treat consistently spin, that is a fundamental property of particles and atoms, especially relevant for magnetic behavior.

"Dealing with spin degrees of freedom is difficult on a theoretical level, and computationally very expensive" say Marrazzo and Colonna. The limitations of current methods become particularly relevant for materials, such as semiconductors, that have a strong spin-orbit coupling, a relativistic effect that is particular evident in heavy chemical elements that often play a major role in cutting-edge scientific or technological applications.

In a new article just published in Physical Review Research, Marrazzo and Colonna advocate for a paradigm shift by introducing a new approach that allows calculating band structures of semiconductors in a simple way and at low computational cost, even in presence of spin-orbit coupling or complex magnetic configurations. The band structure is a fundamental quantity that is key to describe many properties of semiconductors, ranging from light absorption to electronic transport, and is the main workhorse of semiconductors physics.

Their work deals with Koopmans-compliant functionals, where the key is idea is to move from a functional theory of the total electronic density (i.e., DFT) to a functional theory of individual orbitals densities, such that the energy that comes out of the theory should increase or decrease linearly if we add or remove electrons from the system. So far, the theory has not been capable to deal with materials with strong spin-orbit coupling or with spins that are not all aligned

along the same directions, and the two authors expanded it to these relevant cases. “While DFT and Koopmans are based respectively on the total density of electrons and on the orbital density of each electron, here we added also the density of spins, or the density of spin magnetization” they say. The result is a calculation that depends on significantly fewer variables than state-of-the-art Green’s functions, and thus more manageable.

“When we started the project, our aim was to be able to simulate band structures (and spectral properties in general) in materials where relativistic effects are strong, so to get there we set sail for developing a dedicated non-collinear orbital-dependent theory and implementing the resulting method in Quantum ESPRESSO.” says Antimo Marrazzo, first author of the work. “But in the process, we actually ended up discovering and understanding a more fundamental and general aspect of the electronic structure: including spin in the effective interaction between electrons in a material is essential to describe accurately its spectral properties even in systems where spin-orbit coupling is weak or there is no magnetism at all, such as silicon”.

“Our approach”, continues Marrazzo, “crucially include spin-dependent interactions and screening effects that are either missing or rather complex to include in competing approaches, while they emerge very naturally in our non-collinear orbital-dependent functional framework”.

The new computational method was validated on some well-studied materials such as gallium arsenide, a semiconductor with many known applications, and other systems characterized more recently such as CsPbBrI, a member of the perovskite family of materials that are excellent candidates for solar cells applications; tungsten diselenite (WSe_2), that is interesting for his strong spin-orbit coupling and also because it can be exfoliated into two-dimensional monolayers, and CrI_3 that is both ferromagnetic and has strong spin-orbit coupling.

The results of the calculations proved in very good agreements with other well-established, but more costly and unwieldy theories, such as many-body perturbation theory, and with experiments.

The method has been developed and made publicly available in Quantum ESPRESSO, an integrated suite of open-source computer codes for electronic-structure calculations and materials modeling at the nanoscale. Quantum ESPRESSO is an open initiative, in collaboration with many groups world-wide, coordinated by the Quantum ESPRESSO Foundation that includes the Scuola Internazionale Superiore di Studi Avanzati (SISSA), the Abdus Salam

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International Centre for Theoretical Physics (ICTP), the CINECA Italian National Supercomputing Center, the Ecole Polytechnique Fédérale de Lausanne (EPFL), the Oden Institute for Computational Engineering and Sciences, The University of Texas at Austin, in partnership with the Italian National Research Council (CNR).

“Trieste, and SISSA in particular, has a very strong tradition and world-wide leading role in electronic structure simulations” says Marrazzo, “especially in ab initio methods, that require no experimental inputs but are instead based on the fundamental equations of quantum mechanics, electromagnetism and special relativity, thus allowing predicting complex properties of materials with computers.”

“Interactions with the core Quantum ESPRESSO developers and other colleagues here in Trieste, both at SISSA and at the Quantum ESPRESSO foundation have been instrumental for this work” Marrazzo says, “as well as the support by the PNRR and by local, national and European institutions, such as SISSA, the university of Trieste and the Italian ICSC–Centro Nazionale di Ricerca in High Performance Computing (HPC), Big Data and Quantum Computing”.
The work was funded by the European Union– NextGenerationEU, through the ICSC HPC National Center and the Italian PRIN Project “Simultaneous electrical control of spin and valley polarization in van der Waals magnetic materials” and also by Regione Friuli Venezia Giulia through the Microgrant 2023 program of the University of Trieste.

Colonna and Marrazzo say that this development will allow efficient and accurate computational screenings of materials databases and enable simulating complex materials under more realistic conditions, such as in presence of defects or at finite temperature. For the future, the duo aims to applying the method to other quantum materials and keeping exploring magnetic systems.
“We will teach our method to other scientists with dedicated workshops and summer schools” they say, “hopefully empowering them with a powerful new asset to understand, discover and design materials”.

USEFUL LINKS
Full paper

IMAGE
Crediti: Antimo Marrazzo&Nicola Colonna

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