Prize Talk PRV I Tue 13:15 HSZ 01

Seeing is believing: Nonlinear optics on ferroic materials — •Manfred Fiebig — Dept. of Materials, ETH Zurich, Switzerland — Laureate of the Stern-Gerlach-Medal 2023

For millennia, ferromagnetism was the only form of ferroic order known to humankind. Now, however, a large variety of magnetic, electrical and mechanical types of ferroic phenomena is discussed. All of these all have one property in common: The ferroic ordering breaks the symmetry of the host material. Nonlinear optical processes are very sensitive to these symmetry changes. Even its simplest representative, doubling of the frequency of the light or "second harmonic generation" (SHG), therefore couples to the ferroic order parameter and accesses important features of the ferroic state that are often inaccessible to non-optical techniques. Novel ferroic states like ferrotoroidicity as a spontaneous order of magnetic whirls can thus be probed. Ultrafast processes can be resolved — how fast can a magnetic state be switched? In particular, the coexistence of different types of ferroic order in a material can be imaged by SHG. Thus, SHG became an invaluable tool for resolving the magnetoelectric coupling of domains in multiferroics as materials uniting magnetic and ferroelectric order. In my talk I will give an overview of the most important milestones in the classification of (multi-)ferroic materials by nonlinear optics. I will discuss basic questions such as the search for yet unknown types of ferroic order and correlations. I will also address highly application-relevant issues such as the use of SHG for tracking the emergence of ferroic order in thin films in-situ, during the growth process. A not-too-serious concept for "magnetoelectric teleportation" will conclude the lecture.

Prize Talk PRV II Wed 13:15 HSZ 01

Towards chemical and optical band structure engineering in molecular-based heterostructures — •Benjamin Stadtmueller — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany — Institute of Physics, JGU Mainz, 55128 Mainz, Germany — Laureate of the Gaede-Prize 2023

Optical excitations with femtosecond light pulses offer the intriguing opportunity to control material properties on ever-shorter timescales down to the duration of the optical excitation itself. One of the most promising ways to reduce the timescale of the ultrafast material response is to exploit optical-induced spin and charge transfer processes that can directly act on the material's band structure and its population. In this presentation, I will introduce molecular-based heterostructures as a highly intriguing platform to chemically and optically tailor charge and spin carrier functionalities on the nanoscale. Using time-, spin- and momentum-resolved photoemission with fs-XUV radiation, I will first demonstrate that optical excitation of charge transfer states in molecular materials can instantaneously alter the local energy level alignment within the molecular film on ultrafast timescales [1,2]. This approach can be transferred to heterostructures between molecular and 2D semiconductors where it allows us to transiently uncover the otherwise hidden spin polarization of the prototypical layered semiconductor WSe2. These findings will open new avenues for optical controlling and functionalizing spin phenomena in molecular-based heterostructures on ultrafast timescales. [1] Nat. Commun. 10, 1470 (2019), [2] J. Electron. Spectros. Relat. Phenomena 252, 147110 (2021).

Prize Talk PRV III Thu 13:15 HSZ 01

High-lying excitons and excitonic quantum interference in 2D semiconductors — •Kai-Qiang Lin — Department of Physics, University of Regensburg, Regensburg, Germany — College of Chemistry and Chemical Engineering, Xiamen University, Xiamen, China — Laureate of the Walter-Schottky-Prize 2023

Two dimensional semiconductors such as transition-metal dichalcogenide (TMDC) monolayers show a wealth of exciton physics. We present the existence of a novel excitonic species, the high-lying exciton (HX), in TMDC monolayers with almost twice the energy of the band-edge A-exciton but with a linewidth as narrow as that of band-edge excitons. The HX is populated through momentum-selective optical excitation in the K-valleys, and is identified experimentally in up-converted photoluminescence and theoretically in ab initio GW-BSE calculations. These calculations show that the HX is comprised of electrons of negative effective mass. The coincidence of such high-lying excitonic species at around twice the energy of band-edge excitons gives rise to a well-defined excitonic three-level system, which enables quantum-interference phenomenon revealed in optical second-harmonic generation. We show that the temporal dynamics in such a three-level system can be probed through time-resolved sum-frequency generation and four-wave mixing. The HXs can also be tuned over a wide range by twisting and Stark effect in bilayer WSe2, which gives control over the excitonic quantum interference and the corresponding optical nonlinearities. Finally, we show how an electrical gate can be used to tune excitonic quantum interference in a monolayer TMDC transistor device by forming trions.