Optical detection and manipulation of spontaneous gyrotropic electronic order

Qiong Ma

Boston College

Typically, geometrical chirality is predefined by the chiral lattice structure of a material, which is fixed on formation of the crystal. By contrast, in materials with gyrotropic order, electrons spontaneously organize themselves to exhibit macroscopic chirality in an originally achiral lattice. Although such order—which has been proposed as the quantum analogue of cholesteric liquid crystals—has attracted considerable interest, no clear observation or manipulation of gyrotropic order has been achieved so far. Here we report the realization of optical chiral induction and the observation of a gyrotropically ordered phase in the transition-metal dichalcogenide semimetal 1T-$TiSe_2$. We show that shining mid-infrared circularly polarized light on 1T-$TiSe_2$ while cooling it below the critical temperature leads to the preferential formation of one chiral domain. The chirality of this state is confirmed by the measurement of an out-of-plane circular photogalvanic current, the direction of which depends on the optical induction.

Giant c-axis nonlinear anomalous Hall effect in T_d-$MoTe_2$ and WTe_2

Adam W. Tsen

University of Waterloo

While the anomalous Hall effect can manifest even without an external magnetic field, time reversal symmetry is nonetheless still broken by the internal magnetization of the sample. Recently, it has been shown that certain materials without an inversion center allow for a nonlinear type of anomalous Hall effect (NLAHE) even when retaining time reversal symmetry. First, I will discuss our observation of an extremely large c-axis NLAHE in the non-centrosymmetric T_d phase of $MoTe_2$ (as well as WTe_2) without intrinsic magnetic order, which is driven by asymmetric scattering. Application of higher bias yields a record Hall angle and anomalous Hall conductivity. Then, I will discuss some possible consequences of the NLAHE in these materials and their experimental signatures.

Nonlinear Optical Response of Topological Semimetals and its Application for High Performance Photodetection

Dong Sun

Peking University

The experimental manifestation of topological effects in bulk materials under ambient conditions, especially those with practical applications, has attracted enormous research interest. Weyl semimetals and chiral Fermions supply ideal material platforms for such endeavors. The Berry curvature in these materials becomes singular at the node points, creating an effective magnetic monopole in the k-space. Such singular topology can have profound impact on the nonlinear optical response of these materials and it is directly applicable to photodetection. In this talk, we started with our experimental work on the revealing of the singular topology in a type-II Weyl semimetal $TaIrTe_4$ in the photo responses, which are shown to be directly related to the divergence of Berry curvature. As a result of the divergence of Berry curvature at the Weyl nodes, $TaIrTe_4$ exhibits unusually large photo responsivity of 130.2 mA/W with 4-μm excitation in an unbiased field effect transistor at room temperature arising from the third-order nonlinear optical response. We further elucidate such effect is applicable to high performance photodetectors based on topological semimetals, especially for long wavelength (low photon energy) range. Some recent progresses along this direction from our group will be discussed.

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Nonlinear terahertz emission spectroscopy of topological chiral multifold semimetals

Liang Wu

University of Pennsylvania

The absence of mirror symmetry, or chirality, is behind striking natural phenomena found in systems as diverse as DNA and crystalline solids. A remarkable example occurs when chiral semimetals with topologically protected band degeneracies are illuminated with circularly polarized light. Under the right conditions, the part of the generated photocurrent that switches sign upon reversal of the light's polarization, known as the circular photogalvanic effect (CPGE), is predicted to depend only on fundamental constants. The conditions to observe quantization are non-universal, and depend on material parameters and the incident frequency. In my talk, I will discuss nonlinear terahertz emission spectroscopy with tunable photon energy from 0.2 eV - 1.1 eV in the chiral topological semimetals CoSi [1,2] and RhSi[3]. Particularly, we identify a large longitudinal photocurrent peaked at 0.4 eV reaching ~ 550 $\mu A/V2$ in CoSi, which is much larger than the photocurrent in any chiral crystal reported in the literature. Using first-principles calculations we establish that the peak originates from topological band crossings, reaching 3.3±0.3 in units of the quantization constant. Our calculations indicate that the quantized CPGE is within reach in CoSi upon doping and increase of the hot-carrier lifetime. If time permits, I will also briefly introduce our recent work on nonlinear optical studies on 2D antiferromagnets [4,5].

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Geometry and topology in nonlinear optical responses of quantum materials

Naoto Nagaosa[1,2]

[1]*RIKEN Center for Emergent Matter Science (CEMS)*
[2]*Department of Applied Physics, University of Tokyo*

Studies on optical responses of solids have the long history, and have been considered to be well established. However, a new development has been on-going recently, which explores the geometric and topological nature of the electronic states in solids and its crucial role in optical processes including those in nonequilibrium states.

In this talk, I will discuss the geometry and topology in the nonlinear optical responses, which includes (i) shift and injection currents in noncentrosymmetric quantum materials related to Berry phase [1],[2],[3], and (ii) Riemannian geometry in nonlinear optical responses [4], and (iii) topological spin textures in nonequilibrium state [5].

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Nonlinear electric and optical responses in van der Waals nanomaterials

Toshiya Ideue

Department of Applied Physics, University of Tokyo

Nonlinear electric and optical responses such as bulk rectification effect and bulk photovoltaic effect are unique functionalities of noncentrosymmetric materials and also a powerful probe of band geometry or anomalous scattering process [1].
In this talk, I will review our recent works on the nonlinear electric and optical responses in van der Waals nanostructures. Large bulk rectification effect has been observed in layered polar crystals or trigonal superconductors, which reflect the spin-splitting band structure or superconducting fluctuation/vortex dynamics [2,3]. We also found the giant enhancement of bulk photovoltaic effect in symmetry engineered van der Waals nanostructures including WS_2 nanotubes and WSe_2/Black phosphorus heterointerfaces [4,5]. Observed photovoltaic response can be well interpreted by a quantum geometric shift current.
These results indicate that van der Waals nanomaterials are ideal platform for realizing large nonlinear electric/optical responses and also for exploring new emergent nonlinear responses in solids.

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Nonlinear photocurrent generation in halide and organic polar semiconductors

Masao Nakamura

RIKEN Center for Emergent Matter Science (CEMS)

Photoexcitation of materials lacking the spatial inversion symmetry gives rise to an electric current without external bias, known as the bulk photovoltaic effect. A primary origin of the bulk photovoltaic effect is recently revealed to be the shift current that arises from the geometric phase of electron wave functions. In contrast to the conventional drift-diffusion current, shift current is expected to be non-dissipative due to the topological origin.

We have experimentally elucidated unique features of shift current in a representative ferroelectric semiconductor SbSI. We have demonstrated that shift current is quite robust to the localization of carriers associated with the temperature variation and impurity/defect scattering by employing an optimum electrode material, which is a hallmark of the dissipation-less nature [1, 2]. We have also revealed by time-resolved terahertz emission spectroscopy that the shift current shows an ultrafast response for pulse excitation [3].In addition to inorganic compounds like SbSI, we have explored the shift current in organic ferroelectrics. An organic charge-transfer complex TTF-CA is revealed to exhibit a giant shift current [4]. I will also talk about the recent progress in the molecular beam epitaxy of high-quality halide thin films toward the device application of shift current photovoltaics [5, 6].

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Novel in-gap bulk current rectification mechanisms

Inti Sodemann

Max Plank Institute

We develop a unified formalism for current rectification processes of electronic Bloch bands in response to oscillatory electric fields that accounts microscopically for relaxation processes by including the coupling to a physical heat bath. This allows us to describe the impact of inelastic energy relaxation processes on intra-band phenomena such as the non-linear Hall effect and on inter-band phenomena such as shift and injection current effects. Moreover, we describe the conditions under which these effects survive even when the frequency of the impinging radiation is inside the optical absorption band-gap and discuss when these effects are allowed to produce a rectified current and perform work on an external circuit without violating the fundamental laws of thermodynamics. These considerations will allow us to uncover novel facet of the non-linear Hall effect as an efficient amplifier of circularly polarized light.