

## Light–Matter Interactions and Topology: Current Status & Directions

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**From Einstein’s quanta of light to wave–particle duality and the Einstein-Podolsky-Rosen (EPR) conundrum, everything that may seem paradoxical in the physical sciences is a matter of light and insight. Therefore, our research on light–matter interactions is aimed not just at obtaining experimental measurements and findings, but at exploring innovative ways to think about them. In this report, we first outline our perspectives and motivations on this new line of research within our department. We then introduce our recent experimental studies on photoinduced magnetotransport and describe the methodologies and techniques we are developing. Note that, at the time of this report, the studies we present have not yet undergone peer review and are still subject to further refinement and validation.**

Interactions between light and matter are fundamental to perceiving and understanding the world, as everything radiates and absorbs energy. However, what if we told you that there is no light, and that the dash in the title of this report is not just a matter of style but a statement of physics? That there is no "spooky action at a distance" and nothing hiding in the dark – no dark matter or axions? That everything we see and measure is nothing but interactions becoming finite and then definite within the limits of our models?

To understand the essence of topology and light–matter interactions, one must realize that all light–matter interactions are topologically nontrivial. No lasers, photonic crystals, or exotic optical effects are required to appreciate the math. For a solid-state physicist, it might be surprising to hear that everything we perceive with our eyes inherently exhibits phenomena analogous to the quantum spin Hall effect. Why? Because electromagnetic fields are inherent to the stability of matter and carry not only energy but also momentum, such as orbital angular momentum in the presence of phase vorticity, and spin angular momentum when circularly polarized. Photons, as relativistic spin-1 particles, exhibit intrinsic spin-orbit interactions, including the Berry phase. This is why evanescent electromagnetic modes with transverse spin-momentum locking exist at all interfaces between free space and any medium, promoting robust angular momentum–direction coupling.

Through the lens of topology, even seemingly ordinary notions like “everything radiates and absorbs” gain meaning and magic beyond conventional wisdom. Topology truly shines when it comes to organizing complex phenomena in terms of their invariant properties. Analogous to how statistical mechanics tames the intangible dynamics of chaotic gas phases into meaningful quantities like temperature and pressure, topology enables physicists to identify principles of organization based on global effects that transcend specific energy scales. It provides an informational perspective that al-

lows some to study high-energy physics in solid-state crystals and others to explore a whole universe in a helium droplet. Topology reveals that physics – despite being largely organized along the QFT framework of effective field theories, and allowing us to discover symmetries, patterns, and principles of organization – leaves open what organization actually *is* and how it emerges – in models, in physics, and in life.

That said, we now turn to specific topics in the physics of magnetotransport. Here, many experimental observables, reflecting averaged thermodynamic transport quantities, are currently interpreted within theoretical frameworks assuming closed system dynamics and negligible light–matter interactions. With ‘quantumness’ engineered into the equations, charge transport phenomena are increasingly seen as responses to adiabatic electronic wave function evolution. Using experimental data and previously inaccessible observables from local photoexcitation, we present a complementary view of well-known effects, offering an open-system perspective on transport in materials with band-structure topology. We demonstrate that Berry-type fluxes naturally emerge in momentum-constrained energy–charge-coupled transport dynamics, resulting in nonequilibrium magnetization currents and a form of ‘Berryogenesis.’ Finally, we provide a brief outlook on studying and controlling these feedback mechanisms in evanescent fields, potentially leading to novel forms of Hall-type ‘activity’ in matter.

### Lighting up topological transport

Transport in topological semimetals is typically driven by electric fields, magnetic fields, and temperature gradients. When two of these fields are applied orthogonally, they generate a third field in the perpendicular direction, leading to transverse electromagnetic and thermomagnetic effects such as the Hall and Nernst effects. Additionally, collinear field gradients can induce Dirac and Weyl fermion flux, which were recently

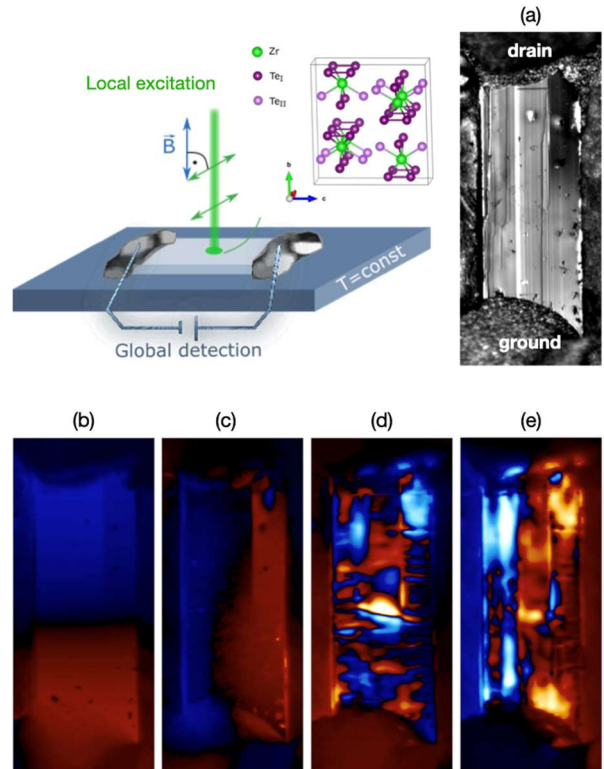
interpreted as manifestations of anomaly cancellations within their effective field theories [1]. Complementary to conventional global magnetotransport measurements, local photoexcitation can be used to drive any of the aforementioned fluxes. The electromagnetic field of a laser beam can be engineered to be energy- and momentum-selective, as well as spatiotemporally structured, making local photoexcitation a versatile probe for driving and visualizing transport. This capability has led to numerous studies on light field- and spin-orbit-driven currents in materials with nontrivial electronic band structures.

In addition to state-of-the-art methods, we developed an innovative experimental setup for determining the magnetotransport properties of millimeter-sized single crystals using cryogenic confocal laser scanning microscopy. Our high-throughput image acquisition approach allows for multidimensional parameter scans at temperatures of 2–300 K, in magnetic fields of up to 7 T, as a function of polarization, and at wavelengths covering the visible to near-infrared spectrum, using both continuous- and pulsed-wave excitations (140 fs). Our setup enables rapid image acquisition over millimeter-scale areas with micron-sized resolution, facilitating the unprecedented visualization of magnetotransport dynamics in the form of full “movies” [2].

### Phototransport maps in quantizing fields

As a model system, we investigated the photoinduced transport properties of zirconium pentatelluride ( $\text{ZrTe}_5$ ) single crystals grown using the tellurium flux method. The magnetotransport properties of  $\text{ZrTe}_5$  have been extensively studied, with reports on its chiral magnetoelectric effects, hydrodynamic transport, anomalous Hall and Nernst effects, and quasi-quantized three-dimensional (3D) Hall effects [3]. Thus,  $\text{ZrTe}_5$  is an excellent candidate for testing our approach.

Figure 1 presents our initial experimental results on the cryogenic photoinduced magnetotransport imaging of  $\text{ZrTe}_5$  single crystals, showing a characteristic dataset of two-dimensional (2D) scans. The dataset includes an optical reflection map (Figure 1a) and photovoltage maps (Figures 1b–e) obtained by raster-scanning a laser beam across the surface of the crystal. The out-of-phase maps in Figures 1b and 1c display homogeneous planar character over millimeter-length scales. By contrast, the in-phase maps in Figures 1d and 1e exhibit complex spatial patterns across the entire crystal. Moreover, the photovoltage signals show clear signatures of Shubnikov–de Haas (SdH) oscillations (Figure 2). This observation of SdH oscillations in re-



*Fig. 1: Cryogenic photoinduced magnetotransport imaging of  $\text{ZrTe}_5$  single crystal. (a) Optical reflection and (b–d) photovoltage maps: (b) out-of-phase, even; (c) out-of-phase, odd; (d) in-phase, even; and (e) in-phase, odd.*

sponse to visible-light excitation (2.2 eV) may seem counterintuitive. However, it highlights a conceptual benefit of our approach: the spatial decoupling of local excitation from signal detection. The probed voltages are related to ambient-type carriers. This makes our optoelectronic measurement schema particularly attractive for studying topological transport phenomena, where long-range global responses of charge carriers may relate to surface and/or edge states.

Moreover, this spatial decoupling allows us to relate our photoinduced data to conventional magnetotransport studies that aggregate data across an entire sample in response to globally applied fields. As an example, Figure 2a shows the out-of-phase photovoltage of many different image pixels compared to recent magnetothermoelectric transport studies [3, 4], providing compelling evidence that these photovoltages can be attributed to photothermoelectric Seebeck and Nernst effects. While photothermoelectric currents have been intensively studied, we want to highlight that these photovoltage maps show an interesting real-space characteristic; that is, a planar-type photothermoelectric response. Previous reports on photothermoelectric current maps have reported signals related to surface-

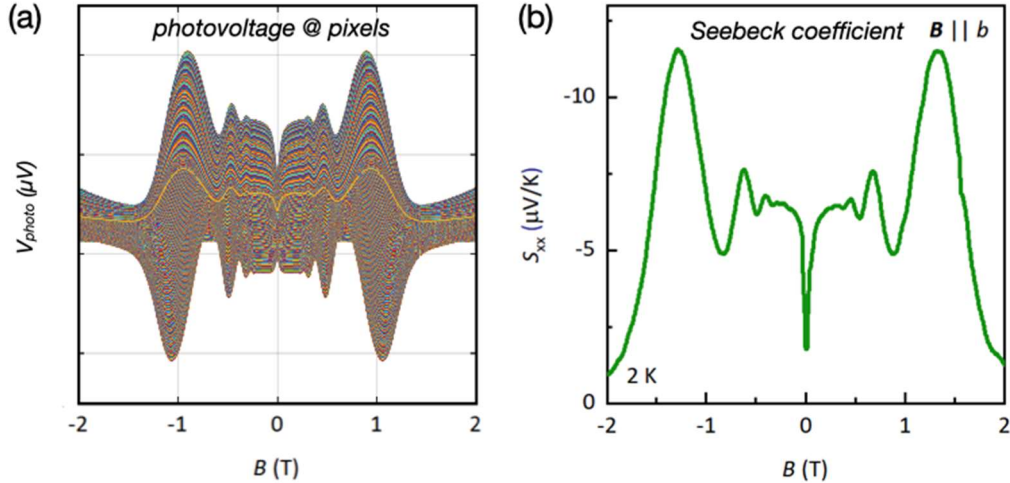


Fig. 2: (a) Shubnikov-de Haas oscillations in the out-of-phase (even) photovoltage at different image pixels and (b) comparison to Seebeck coefficient.

or interface-type inhomogeneities, such as  $p$ - $n$  junctions, electrical contacts, and lateral crystal edges, where the in-plane symmetry is intrinsically broken. We interpret this as evidence that our signals are sensitive to out-of-plane inversion symmetry breaking at the surface, which is consistent with the 3D geometric characteristics of our sample. Furthermore, we note that the in-phase photovoltage maps provide clear evidence of real-space disorder and long-range chemical potential fluctuations, similar to those observed on the surfaces of topological insulator thin films and graphene layers at low carrier densities. This suggests that recent conflicting reports on the presence of trivial Fermi pockets and multicarrier transport in pentatellurides at liquid helium temperatures may originate from real-space effects.

### Numerical modeling of observations

To quantitatively understand the correlation between long-range photovoltage patterns and microscopic potential disorder, we initially analyzed our photovoltage maps using a well-established numerical model based on the Shockley–Ramo theorem (SRT). In this model, laser photoexcitation, even when spatially distant from the current-collecting contact, creates a local electromotive force that drives ambient carriers from the excitation region to the remote contacts. The SRT model can reproduce multiple real-space features observed in the in-phase photovoltage response, such as long-range characteristics and sensitivity to surface inhomogeneities and edges. However, it fails to qualitatively reproduce planar-type responses. Overall, we find that the SRT is insufficient to fully understand the dynamics of photoinduced magnetotransport because it treats the

photocurrent response as electrostatic and is largely agnostic the microscopic mechanisms of local photocurrent generation.

To address these limitations, we are developing a new model based on the principles of thermodynamic reciprocity. This approach considers that the photoinduced transport processes in optoelectronic devices near equilibrium and in linear response are reciprocal to their time-reversed counterparts under bias, even in a magnetic field. For example, when unilluminated and voltage-biased, bolometric detectors show Joule heating; photovoltaic cells act as electroluminescence devices; and reverse-biased photothermoelectric devices act as heaters/coolers. Consequently, we consider global photovoltages to be weighted volume integrals of a local injection density term multiplied by a responsivity coefficient. While the local injection density generally depends on the local electrodynamics, including material-specific light–matter interactions, the primary relevant effect in the first approximation is absorption.

### Illuminating intrinsic potential landscapes

Considering the Onsager–Casimir reciprocity for all net transport currents, our photovoltage maps can be interpreted as reflections of internal affinity shifts in response to local energy injection. This enables us to relate our photovoltage maps to the linear response transport theory, wherein external thermodynamic driving forces induce coupled energy and charge fluxes. According to Einstein’s relation, an applied electric field is compensated for by a gradient in the chemical potential. Similarly, a temperature gradient is compensated for by the gravitational potential, where the “thermal field” is analogous to the electrostatic field.

Supporting the applicability of our model, we observed that the photothermoelectric Seebeck- and Nernst-type effects in response to laser-induced temperature gradients were out of phase with the energy-injection modulation frequency. Furthermore, the planar character of these effects suggests the existence of a thermoelectric magnetization current flow within the laser spot, which causes a net transport flux in the presence of broken inversion symmetry, such as at the crystal surface. Here, gradients in the temperature and electrochemical potential are no longer collinear and are not fully balanced, unlike those in the bulk crystal. We conclude that these unbalanced electrochemical potentials lead to measurable out-of-phase photothermoelectric voltages, demonstrating that the conventional wisdom – that the thermopower voltage is proportional to the temperature gradient multiplied by the thermopower coefficient – does not hold true in the presence of thermal gravitational potentials, anisotropic thermopower tensors, and inversion symmetry breaking at edges and surfaces.

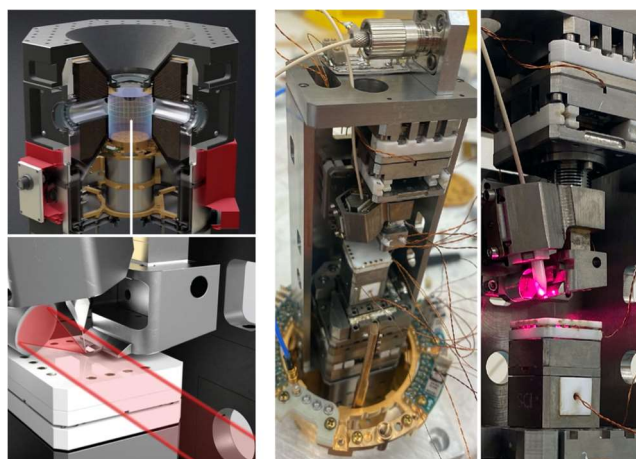
The unbalanced gravitational (thermal vector) potential causes a conjugate thermovoltage in the presence of inherent chemical potential gradients and fluctuations, such as those at interfaces and edges. Our in-phase maps clearly show this dynamic back-action effect. The signal is in phase with the energy-injection density because it is proportional to the built-in chemical potential gradients, absolute temperature (unbalanced gravitational potential), and absolute thermopower. Notably, these thermoelectric effects, which are electrodynamic rather than electrostatic, have largely been overlooked in previous magnetotransport, thermoelectric, and photocurrent studies. Further theoretical investigations based on effective field theory descriptions of transport coefficients are underway to elucidate the intricate relationships between our observed photovoltage maps and their interpretation as real-space gravitational potential maps, particularly in the quantum limit of charge and heat flow.

### Some general implications

We next want to discuss some implications of our observations for interpreting globally averaged magnetotransport measurements. In combination with considerations regarding Onsager-Casimir reciprocity and linear response, our real-space photoinduced transport maps reveal that the common assumptions of homogeneous samples and (electrostatic/gravitational) fields are generally not fulfilled in real-world crystals.

This nonhomogeneity is not merely due to sample quality or the presence of defects, but stems from the finite nature of crystals and their intrinsic thermodynamic openness to energy and momentum exchange interactions. Interpreting topological magnetotransport observables, such as the Hall voltage in a quantum Hall insulator phase, requires intrinsic finite size and disorder effects. Our photovoltage maps highlight that “disorder” serves largely as a phenomenological placeholder for the particularities of open-system energy–momentum flux dynamics that cannot be adequately described and understood within closed-system electrostatic charge transport models.

In light of our findings, the interpretation of experimental observations attributed to anomalous Nernst effects can be enriched by considering an “open-system” perspective. When there is momentum-unbalanced energy–charge flux coupling, the transverse voltage measured in response to a globally applied temperature gradient is not solely due to the Nernst effect but also the Bridgman effect. Similarly, this perspective offers a new approach for interpreting observations related to anomalous Hall effects. Although our specific experimental observations pertain to ZrTe<sub>5</sub>, the thermodynamic reciprocity principles invoked for interpretation are broadly applicable to other materials and temperature regimes. This suggests that, in many systems, purely electronic-type orbital magnetizations may be significantly influenced by magnetothermoelectric back-action effects and orbital-type thermoelectric magnetization currents.



*Fig. 3: Schematics (left) and photographs (right) of our photoinduced force scanning probe microscope, designed for operation under cryogenic conditions, high magnetic fields, and compatibility with a mid-infrared quantum cascade laser.*

### Light–matter interactions gaining momentum

Finally, we outline our research activities on photoinduced force microscopy and our plans to investigate topological electromagnetic states in relation to the hybridization of helical photons and chiral fermion states in polaritonic quantum Hall resonator devices.

To achieve our objectives, we developed a cryogenic high-magnetic-field photoinduced force microscope (Figure 3). Unlike scanning near-field optical microscopy, which retrieves local spectroscopic information from scattered light, our microscope detects electromagnetic forces from spin–angular momentum interactions. Unlike electronic spin, which can be probed using established techniques like magnetic force microscopy, tools for directly detecting electromagnetic (photon) momentum textures and interactions of polaritonic evanescent fields are still under development. Although optical forces have been extensively studied in relation to macroscopic mechanical objects, particle trapping, and enantioselective sorting, near-field imaging of photomodulated dispersion forces related to topological states and macroscopic quantum phases such as superconductivity remain largely unexplored.

As an initial experimental model, we are investigating MoO<sub>3</sub> single crystals with bi-anisotropic phonon–polariton modes. Our primary focus is on understanding photomodulated force interactions in regimes dominated by dispersion/van der Waals-type forces and pioneering methods to differentiate electromagnetic gradient forces from scattering forces. Our ultimate goal is to establish an experimental platform for characterizing and controlling cavityless polaritonic modes and resonators to demonstrate the topological ground state control of charge transport phases via spin-to-angular momentum-shaped evanescent near-field structures.

### External Cooperation Partners

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