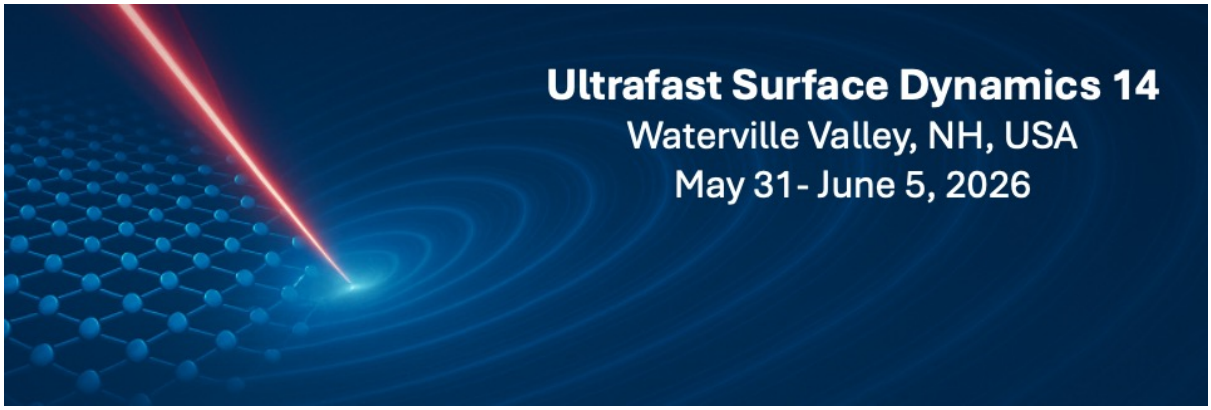

USD14 Poster Abstracts

Plasmon-driven exciton formation in a non-equilibrium Fermi liquid

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Collective modes in Fermi liquids are usually regarded as dissipation channels that relax electronic excitations through Landau damping. Whether such modes can instead mediate the formation of correlated electronic states under non-equilibrium conditions remains an open question. Here we show that, under optical photo-doping, a bulk plasmon can drive correlated inter-band transfer within a transient electronic continuum. Using time- and angle-resolved photoemission spectroscopy (Tr-ARPES) on $EuCd_2As_2$ supported by electronic structure calculations, we observe that at high excitation density, plasmons transfer energy from a weakly dispersing bulk band into unoccupied surface states. This bulk-to-surface redistribution stabilizes a long-lived, energy-localized spectral feature consistent with a Mahan exciton. Our results uncover a non-equilibrium regime of Fermi-liquid physics in which collective modes do not merely dissipate energy, but also stabilize correlated bound states.

References: R. Acharya et al. arXiv:2603.xxxxx

Momentum Microscopy with multifunctional modular solutions

SPECS Surface Nano Analysis GmbH

Saumya Mukherjee, Stefan Böttcher, Nils Weber, Matthias Escher, Marten Pratt

Complementary to conventional electron microscopes operating at high energy, one of the key developments in the field of imaging appeared in the form of low energy new generation electron spectrometers. A solution was developed by combining hemispherical analyzers and PEEM lens system to provide higher energy- and lateral- resolution in both momentum and real space imaging. Such a system was coined as momentum microscope (MM) which operates at relatively lower kinetic energy and allows aperture based selective area imaging and band dispersion analysis. Together with advancement in deflector and aperture technology it provides scanning and zooming ability, like the standard electron microscopes, but with added functionality to perform such functions not only in real space but also in momentum space. This provides an alternative opportunity to the scientific community to perform multifunctional operations within the same instrument such PEEM-like imaging and ARPES-like band dispersion. In addition to low operational kinetic energy of electrons, these systems benefit from easy-to-handle sample preparation and can host a wide variety of materials. The fundamental operation is based on photoemission spectroscopy and due to growing demands of multifunctionality our microscopes are also adapted to host spin-resolved imaging and time-resolved spectroscopy studies. In this talk, I will introduce our product portfolio on MM-systems and provide a clear overview of the possibility of reaching multi-functionality using a modular approach. Our recent system design, together with NanoESCA-MARIS as the primary work horse we are attempting to establish MM as a complementary tool in imaging, band structure study, spin- and time-resolved analysis. We will demonstrate the advancement in the performance achieved and thereby, a detailed overview of these new developments for MM technology at SPECS will be presented.

Dynamics of Interlayer Excitons in WS_2/WSe_2 Heterostructures

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Heterostructures of two-dimensional materials exhibiting electron and hole localization have emerged as exciting platforms for studying complex quantum phenomena. Transition metal dichalcogenides (TMDs) are archetypal systems in which interlayer interactions can drastically alter the direct vs. indirect character of excitons in both real space and momentum space. TMD heterostructures in particular often have complex energy landscapes supporting many different exciton states [1]. However, these excitons can be difficult to fully characterize with optical measurements due to the existence of momentum indirect "dark states" which cannot be optically excited or probed.

Alternatively, photoemission spectroscopy methods can access occupied states across the entire Brillouin zone, offering a direct account of the complete energy landscape without dark states. At Stony Brook, we use a unique combination of cavity-enhanced high-harmonic generation and time-of-flight momentum microscopy to achieve tr-ARPES of excitons in TMDs with high sensitivity [2].

We will present preliminary tr-ARPES data on interlayer exciton dynamics in WS_2/WSe_2. Optical measurements have indicated that the lowest-energy exciton in WS_2/WSe_2 is momentum-indirect, and that intervalley phonon scattering between direct and indirect excitons controls the exciton dynamics [3]. In contrast, our data reveal the energy alignment in WS_2/WSe_2 to be nearly degenerate between direct and indirect exciton valleys. Our data also show formation dynamics of excitons, indicating that formation of the momentum-direct interlayer exciton is delayed compared to the indirect interlayer exciton by roughly 1 ps.

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Hybrid Frenkel–Wannier excitons facilitate ultrafast energy transfer at a 2D–organic interface

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Two-dimensional transition metal dichalcogenides (TMDs) and organic semiconductors (OSCs) have emerged as promising material platforms for optoelectronic devices. Combining the two is predicted to yield emergent properties while retaining their individual advantages. In both materials, the optoelectronic response is governed by excitons, i.e., correlated Coulomb-bound electron-hole pairs. Consequently, excitons play a central role in determining the optoelectronic properties of the host materials, rendering a detailed understanding essential for efficiently harvesting and controlling exciton-mediated energy conversion pathways.

In OSCs, the optoelectronic response is typically dominated by tightly bound Frenkel-type excitons arising from localized molecular orbitals, whereas TMDs host delocalized Wannier-type excitons. However, much less is known about the characteristics of excitons at hybrid interfaces between these materials, which determine the possible energy- and charge-transfer pathways.

Here, we use femtosecond photoemission momentum microscopy to gain unprecedented insights into the exciton landscape and dynamics at a 2D-organic interface [1]. Building on our previous work [2], in which we demonstrated that time-resolved momentum microscopy provides direct access to the entangled exciton wavefunction, we now identify a hybrid exciton at a PTCDA/WSe_2 interface. We show that this hybrid exciton, formed predominantly via resonant Förster energy transfer, comprises both Frenkel- and Wannier-type contributions: intralayer electron–hole transitions within the organic semiconductor layer and interlayer transitions across the interface give rise to an exciton wavefunction with mixed character.

References: [1] Bennecke et al., Nat. Phys. 21, 1973-1980 (2025) [2] Bennecke et al., Nat. Commun. 15, 1804 (2024)

Subcycle dynamics of the interplay between intraband acceleration and interband absorption in Bi_2Te_3 with 2D momentum resolution

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Spin-momentum locking suppresses electron scattering in the topologically protected surface states (TSS) of topological insulators as evidenced by the observation of ballistic electron acceleration over 1 ps with subcycle ARPES [1]. The resulting long coherence time facilitated the exploration of unexpected phenomena in the strong-field regime of light-matter interaction, such as a novel mechanism of THz HHG [2] and the build-up of Floquet-Bloch states in less than two cycles of light [3].

Here, we report on the interplay of intraband and interband excitation in the perturbative regime of light-matter interaction. The experiments were conducted with a subcycle ARPES setup that combines CEP-stable MIR pump pulses (25 THz, 75 kV/cm at the Bi_2Te_3 surface) with a 15-fs two-photon-photoemission probe at a repetition rate of 200 kHz. In contrast to previous subcycle experiments, which were limited to 1D cuts through the Brillouin zone [1,3], the deflector lens of our hemispherical Scienta DA30 analyzer allows for imaging the TSS in 2D momentum space.

We observe that intraband acceleration is strongly influenced by the hexagonal warping of the TSS. For the accelerating field along the $\overline{\Gamma K}$-direction, the pump photons ($\hbar\omega_{\mathrm{MIR}} =$ 100 meV) also induce resonant interband absorption, predominantly in the perpendicular $\overline{\Gamma M}$-direction. The distinct phase relation between the two mechanisms with respect to the driving field leads to a reduced amplitude and phase shift of the measured electron displacement. In contrast to similar experiments performed for graphene in the strong-field regime [4], Landau-Zener-Majorana transitions do not contribute to the observed dynamics.

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Probing Ultrafast Structural Dynamics in Multilayer Graphene Using ULEEM

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Multilayers of graphene (MLG) exhibit electronic and structural properties that vary strongly with layer number and stacking, making it an ideal model platform for studying ultrafast phenomena in two-dimensional materials [1]. Here, we employ a newly developed ultrafast low-energy electron microscope (ULEEM) [2] to investigate the behavior of MLG after ultrashort optical excitation. The instrument features nm-spatial combined and sub-ps temporal resolutions, combined with ultimate surface sensitivity. We observe transient excitations in both real and reciprocal space and report a strong energy dependence of the ultrafast image contrast. Finally, we discuss the broader potential of ULEEM studies as a new tool for ultrafast surface science.

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Ultrafast Carrier Relaxation and Phonon Bottleneck Effect in Layered PEA2PbI4 Perovskite Films

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Two-dimensional Ruddlesden–Popper perovskites such as PEA_2PbI_4 have attracted significant attention due to their strong light–matter interactions and potential for hot-carrier optoelectronics. In this work, we investigate ultrafast carrier relaxation dynamics in ∼300–400 nm thick PEA_2PbI_4 thin films using femtosecond pump–probe spectroscopy. Static photoluminescence (PL) measurements reveal a pronounced emission peak centered at approximately 520 nm. The samples were excited at 400 nm and probed at 800 nm to monitor the transient carrier response.

The measured dynamics exhibit a fast decay that is well described by a single-exponential function, yielding a characteristic relaxation time of $\tau = 2.08 \pm 0.02$ ps under the present excitation conditions. These results indicate rapid photoexcited-carrier relaxation in PEA_2PbI_4 and establish a baseline for understanding energy-dissipation pathways in layered perovskites.

Ongoing measurements employ broadband white-light probing to obtain spectrally resolved transient absorption and extract the time-dependent carrier temperature. In addition, fluence-dependent studies are being performed to investigate the potential role of a hot-phonon bottleneck in governing the cooling dynamics of photoexcited carriers in this material system.

Efficient light upconversion via resonant exciton-exciton annihilation of dark excitons in few-layer transition metal dichalcogenides

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Materials capable of light upconversion—transforming low-energy photons into higher-energy ones—are pivotal in advancing optoelectronics, energy solutions, nanophotonics, and photocatalysis. However, the discovery in various materials pays little attention on few-layer transition metal dichalcogenides, primarily due to their indirect bandgaps and weaker light-mater interactions. Here, we report a pronounced upconversion photoluminescence in few-layer transition metal dichalcogenides, especially focusing on WSe_2. This joint theory-experiment study attributes the upconversion photoluminescence to a resonant exciton-exciton annihilation involving a pair of dark excitons with opposite momenta, followed by the spontaneous emission of upconverted bright excitons, which can have a high upconversion efficiency. Additionally, the upconversion photoluminescence is generic in MoS_2, $MoSe_2$, WS_2, and WSe_2, showing a high tuneability from green to ultraviolet light (2.34–3.1 eV). The findings pave the way for further exploration of light upconversion regarding fundamental properties and device applications in two-dimensional semiconductors.

Implementation of a time-resolved momentum microscope with tunable mid-infrared pump and deep-ultraviolet probe for ultrafast photoemission mapping

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Time and angle-resolved photoemission spectroscopy (trARPES) is a critical tool for investigating the non-equilibrium states in quantum materials. However, the traditional trARPES suffers from macroscopic spatial averaging, obscuring the intrinsic dynamics of multi-domain and spatially confined systems. To overcome this, we present the design, integration and commissioning of a novel time-resolved momentum microscope facility that enables simultaneous space- and time-resolved photoemission mapping. The momentum microscope features a field aperture to isolate photoelectrons from defined microscopic regions, achieving a spatial resolution down to a few micrometers. This capability enables the extraction of electronic band structures from targeted micro-domains, individual exfoliated flakes and other spatially confined samples. The system is integrated with a mid-infrared pump and a 6 eV probe ultrafast laser system. By combining an optical parametric amplifier and differential frequency generation, and a custom-designed optical transfer line, the excitation wavelength can be tuned between 3 and 12 μm (corresponding to photon energies of approximately 100-400 meV) with selectable polarization. This architecture enables us to manipulate non-equilibrium states via Floquet engineering, mode-selective, or in-gap low-energy excitation.

Temporospatial modulation of nematicity in FeSe

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Broken symmetry often manifests not only through uniform order parameters but also through emergent spatial textures that encode the microscopic origin of a quantum phase. Electronic nematicity in iron-based superconductors exemplifies this behavior: an electronically driven breaking of fourfold (C_4) rotational symmetry couples to the lattice and produces orthorhombic distortion and twin domains [1]. Although the nematic order in FeSe has been heavily studied via spatially averaged techniques, recent photoemission electron microscopy (PEEM) has uncovered previously hidden ∼100 nm mesoscale modulations that evade global probes [2].

In this work, we directly visualize and track the dynamics of this electronic texture using time- and energy-resolved dark-field PEEM with a time-of-art momentum microscope system. By rotating the linear polarization of a 4.8 eV probe pulse, we are able to image d_{xz} versus d_{yz} orbital character through linear dichroism contrast. The dichroic signal varies with energy and is enhanced near the Fermi surface, consistent with the prevailing view that nematicity is governed by low-energy electronic states. Having established the spatial modulation of nematicity in FeSe, we further probe its nonequilibrium behaviors. Following ultrafast optical excitation, the texture undergoes ultrafast melting and recovery, accompanied by coherent oscillations.

References: [1] J.-H. Chu et al. Science, 337, 710–712 (2012) [2] T. Shimojima et al. Science, 373,1122–1125, (2021)

Coherent Ferron Emission and Propagation

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Ultrafast excitation of ordered quantum materials can reveal collective modes that govern energy transport and phase dynamics. In ferroelectrics, the transverse optical (TO) phonon mode enables polarization waves whose quanta, ferrons, carry electric dipole moment and represent the amplitude (Higgs) mode of the ferroelectric order parameter. Here we report the ultrafast generation, coherent emission, and spatiotemporal propagation of polarization waves in the van der Waals ferroelectric $NbOI_2$. Upon femtosecond optical excitation, we observe intense, narrow-band terahertz (THz) radiation at the ferroelectric TO frequency, directly evidencing coherent ferron emission. The polarization wave modulates the ferroelectric order parameter and propagates uniaxially along the polar axis with hypersonic velocities (10^5 m/s). The dynamics exhibit long coherence times of 220 $\pm$ 80 ps at 4 K and 20 $\pm$ 7 ps at 295 K, exceeding previously reported room-temperature coherence in canalized hyperbolic phonon polaritons in related van der Waals systems. Comparative measurements on WO_2Br_2 confirm the presence of polarization waves in structurally related ferroelectrics, while their absence in non-ferroelectric $TaOBr_2$. These results establish ferrons as long-lived, providing a platform for ultrafast dipole transport, narrow-band THz emission, and coherent control of ferroelectric order.

References: [1] J. Choe, T. Handa et al., arXiv:2505.22559 (2025) [2] P. Tang et al., Phys. Rev. B 106, 8 L081105 (2022)

Probing Nonlinear Optics via Ultrafast Atomic Force Microscopy Noise Measurements

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The nonlinear optical response of materials is central to emerging optoelectronic and quantum technologies, particularly in low-dimensional systems such as transition metal dichalcogenides (TMDs) [1, 2]. A key challenge is to spatially resolve variations in the second-order electric susceptibility, $\chi^{(2)}$, at the nanoscale and correlate them with structural defects. Ultrafast atomic force microscopy (AFM) enables characterization of the $\chi^{(2)}$ by probing the second-order polarization induced by ultrafast laser pulses, providing nanometer-scale spatial resolution without relying on far-field photon emission [3]. In this work, we demonstrate improved characterization of $\chi^{(2)}$ using noise-based measurements with ultrafast AFM. Instead of performing time consuming interferometric autocorrelations at each spatial point, we measure nonlinear polarization fluctuations. Because the nonlinear polarization fluctuations scale with $\chi^{(2)}$ and optical intensity, the noise amplitude provides a rapid proxy for the local nonlinear response. This approach reduces acquisition time by more than an order of magnitude per pixel, enabling substantially improved spatial resolution. We apply this technique to organic thin films, monolayer TMDs, and reference substrates, achieving $\sim$10 nm spatial resolution and demonstrating clear contrast between regions with different nonlinear responses. Noise-based ultrafast AFM thus provides a promising route toward correlating nanoscale defect structure with local nonlinear optical properties.

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Structure and Dynamics of Na^+ and Cs^+ adsorbed on Cu(111) and solvated by D_2O molecules

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Alkali cations in liquid water represent widely studied model systems of solvation science in which the water-water interaction competes with the local water-cation coupling. On electrode surfaces additional interactions become relevant. In our investigation using time-resolved two-photon photoemission and low-temperature scanning tunneling microscopy, we compare Na^+ and Cs^+ on Cu(111) coadsorbed with individual D_2O molecules. $D_2O/Na^+/Cu(111)$ forms 10 nm wide, flat-lying aggregates. We observe a linear increase of the electronic lifetime of the 3s electron transfer resonance at the Cu(111)-Na^+ interface as a function of the number n of adsorbed D_2O molecules per ion combined with a maximal energy transfer to solvent modes ΔE of 0.6 eV at $n = 7$ [1]. On $D_2O/Cs^+/Cu(111)$ we find exclusively one or three D_2O attached to Cs^+. This leads to a stepwise increase in the electron lifetime with n. ΔE is half that of $Na^+/Cu(111)$, which we associate with differences in ion core diameter and modified interactions with Cu(111) [2]. Comparing both systems we identify pronounced differences rooted in the microscopic connection between structure and dynamics. Funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy EXC 2033-390677874-RESOLV, MO 960/27-1, and by Project ID 278162697-SFB 1242 is gratefully acknowledged.

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Collective lattice excitations in a relaxor ferroelectric probed by time-resolved coherent x-ray scattering

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Relaxor ferroelectrics are a well-known but poorly understood class of functional materials in which the electromagnetic response functions are renormalized by the presence of nanoscale heterogeneity in the ground-state polarization texture. Despite decades of research, however, the low-energy excitations associated with this phase have yet to be fully characterized, and the origin of the observed dynamical behavior remains ambiguous. In this work, we use time-resolved coherent x-ray scattering to investigate the low-energy collective modes of a relaxor ferroelectric $Pb(Mg_{1/3}Nb_{2/3})_{1-x}Ti_x\ O_3$) thin film, in which we identify two novel excitations: (1) a $350m/s$ surface acoustic wave associated directly with the collective motion of ferroelectric domain boundaries, and (2) a fully localized, non-dispersive collective lattice excitation at a frequency characteristic of an inter-domain interaction scale. Upon tuning the system away from the relaxor ferroelectric ground state via epitaxial strain, we find that the spectral weight of these excitations is greatly reduced, directly implicating them in the relaxor-like behavior in this material. These findings not only shed new light on the low-energy collective mode spectrum associated with relaxor ferroelectrics, but also represent the first demonstration of "vibrational x-ray photon correlation spectroscopy" in which the dynamics of a material's mesoscale domain structure may be directly measured in reciprocal space using coherent x-rays.

Giant dynamical magnetoelectric coupling in a van der Waals multiferroic

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The recent discovery of multiferroicity in few-layer helical van der Waals magnets has raised the prospect of realizing unprecedented magnetoelectric couplings in two dimensions. Nevertheless, the exact nature and strength of these couplings have remained elusive to date. Here, we perform a precision measurement of the dynamical magnetoelectric coupling in the van der Waals multiferroic NiI_2. We evaluate this interaction in resonance with a collective electromagnon mode, capturing the impact of its oscillations on the material's dipolar and magnetic orders. We find that NiI_2 possesses a giant natural optical activity at terahertz frequencies, greatly outclassing other known multiferroics. First-principles calculations reveal that this dynamical magnetoelectric coupling originates from direct interactions between the electronic spin and orbital degrees of freedom, resulting in substantial enhancements over lattice-mediated effects. Our findings highlight the potential for intertwined electronic states to enable unique functionalities in the two-dimensional limit and pave the way for the development of nanoscale magnetoelectric devices operating at terahertz speeds.

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From moiré ferroelectricity to nanoscale defect array in hexagonal Boron Nitride

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Defects in wide bandgap semiconductors have emerged as an important building block for quantum technologies, offering robust, optically addressable electronic spin states and single-photon emissions. However, the nanoscale deterministic spatial control of these defects is still a roadblock to a scalable integration with photonic circuits and Qubits based on entangled defect arrays. Existing techniques for patterning nanoscale defect arrays (e.g., ion implantation, localized strain, laser writing) are limited to micron-scale precision and non-deterministic yield [1]. A compelling alternative would be to spatially engineer the property of the material itself. In this talk, we present our latest results exploring the spatial distribution of defects in twisted hexagonal Boron Nitride (hBN), a system that exhibits moiré ferroelectricity [2]. Using photoemission electron microscopy (PEEM), we map the spatial distribution of occupied states at different energies including valence bands, defect states and ferroelectric domain walls. Surprisingly, we find that for small moiré period ($\lesssim$1μm), defects are preferentially located at the ferroelectric domain wall triple intersections, forming defect arrays with pitch as small as ∼270nm. At larger moiré periods, the defects form a one-dimensional structure along the domain walls. Photoluminescence shows that: most of the light emission comes from the photoemission-bright defects and that defects exhibit spectral signatures of known C-based single quantum emitters. This discovery sets moiré ferroelectricity in twisted hBN as a new and powerful way to pattern and shape quantum emitters arrays at the nanoscale.

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Non-Equilibrium Energy Flow among Electrons and Phonons in Ultrathin Pb Films on Si(111)

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The pathways of flow of energy subsequent to an impulsive optical excitation through various degrees of freedom of the electronic and lattice systems were studied for ultrathin lead films on Si substrates. The response of the electron system is determined using time-resolved photoelectron spectroscopy while the lattice excitation is determined by means of the Debye-Waller effect in ultrafast reflection high-energy electron diffraction. Epitaxial 5 ML thick Pb films on Si(111) were excited by fs-IR laser pulses. We observe thermalization of the electronic excitation in less than 200 fs and cooling in 1.4 ps. The electronic heat decays even faster in only 0.5 ps while the lattice temperature rises – depending on excitation density – slowly in 3.5 to 8 ps, leaving a gap of 3-7 ps, where the energy is hidden. We propose that the hidden energy is transiently stored in high-frequency phonon modes for which diffraction is insensitive and which are excited in 0.5 ps. Within a three-temperature model we use three heat baths, namely electrons, high-frequency and low-frequency phonon modes to simulate the observations. The excitation of low-frequency acoustic phonons, i.e., thermalization of the lattice is facilitated through anharmonic phonon-phonon coupling. Quantitative access to the temperature of the high frequency phonon subsystem is provided through the slow drop of temperature of the electron system on longer time scales longer than 3 ps which supports the proposed model. Equilibration among the phonon system, i.e., among high and low frequency phonons, takes up to 80 ps, cooling of the Pb film towards the substrate occurs finally in 1 ns.

Ultrafast table-top three-dimensional photoemission orbital tomography

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Photoemission orbital tomography (POT) enables orbital-resolved imaging of molecular systems with sub-ångström spatial resolution. When combined with photon-energy-dependent measurements, 3D-POT can reconstruct the complete three-dimensional structure of molecular orbitals. However, typical implementations require synchrotron facilities and extensive measurement times, preventing applications in time-resolved studies of ultrafast dynamics.

Here, we demonstrate the first table-top 3D-POT experiment using a femtosecond high-harmonic generation light source [1]. We combine time-of-flight momentum microscopy with a pulse-preserving EUV monochromator covering an energy range from 13 to 71 eV, enabling complete 3D data acquisition without photoelectron energy or angular scanning. Critically, we developed an advanced reconstruction algorithm that recovers both amplitude and phase information from sparse data, reducing sampling requirements by an order of magnitude [2].

We benchmark this approach on PTCDA/Ag(110), achieving full 3D reconstruction of both HOMO and LUMO orbitals using only seven photon energies. The reconstructed orbitals show excellent agreement with DFT calculations, with sub-ångström spatial resolution capturing the complete out-of-plane orbital structure. With intrinsic femtosecond time resolution, our approach opens new avenues for studying orbital hybridization in organic/inorganic heterostructures [3], exciton dynamics in organic semiconductors, and light-induced orbital evolution in photochemical reactions.

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Momentum-resolved electron–phonon coupling revealed by high harmonic spectroscopy

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Understanding how lattice motion reshapes electronic structure on femtosecond timescales is essential for controlling emergent phases in quantum materials [1,2]. Here we employ high harmonic spectroscopy (HHS) in a reflection geometry driven by an ultrafast mid-infrared (MIR) pulse to probe coherent phonons excited by a near-infrared pump in the type-II Weyl semimetal WTe_2. The harmonic emission originates strictly from the near-surface region, where we observe pronounced photoinduced modulations of the harmonic yield. By rotating the MIR driver polarization, we reveal a field-dependent anisotropy in the phonon-modulated harmonic response, directly linking vibrationally induced band structure changes to the emitted harmonics. HHS is shown to provide momentum-resolved access to the anisotropy of electron–phonon coupling for multiple phonon modes simultaneously, establishing it as a powerful probe of coherent lattice dynamics.

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Nanoscale symmetry breaking directs energy flow in black phosphorus

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Understanding the spatial directionality of energy flow at surfaces is essential for controlling nonequilibrium carrier and phonon dynamics in two-dimensional materials. Because of their all-surface nature, two-dimensional materials are exceptionally prone to extrinsic modifications, such as edge effects and localized strain, which modify electronic wave functions and band energetics. The resulting morphology-modified carrier and phonon dynamics can compete with or even overwhelm intrinsic behaviors. I investigate this interplay of intrinsic and extrinsic broken symmetries in a model system, anisotropic black phosphorus (BP), from the electronic and vibrational perspectives, using complementary electron microscopies to achieve spatial resolutions of single to tens of nanometers, beyond the reach of optical microscopies. I combine polarization-dependent photoemission electron microscopy (PEEM) and ultrafast transmission electron microscopy (UEM) to investigate the role of edge modification on the electronic structure and coherent acoustic phonon transport in BP by directly interrogating the above-bandgap optical transitions and photoexcitation-induced lattice dynamics, respectively. Using PEEM, I show that spatially confined wavefunctions at BP flake edges result in a rotated transition dipole moment, with implications for edge-selective excitation and the design of nanostructured 2D materials. With UEM, I show that the bonding anisotropy-driven mixing of longitudinal and transverse acoustic phonon modes results in a strongly reduced group velocity of in-plane coherent acoustic phonons. These results suggest that intentional design of edge structures and intrinsic bonding anisotropy offer a novel route towards controlling in-plane acoustic phonon and heat propagation.

Spontaneous breaking of mirror symmetry in a cuprate beyond critical doping

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Identifying ordered phases and their associated symmetries in high-temperature superconductors is essential for uncovering the microscopic origin of superconductivity, as critical fluctuations near phase transitions may directly influence pairing. In cuprates, the search for such ordering has largely focused on symmetry breaking within the pseudogap regime. By contrast, the overdoped Fermi-liquid–like region beyond the critical doping—where the pseudogap vanishes—has generally been regarded as a conventional, quantum-disordered metallic state. Here, we reveal broken mirror symmetry in the Fermi-liquid–like phase of $(Bi,Pb)_2Sr_2CaCu_2O_{8+\delta}$ beyond critical doping by tracking the temperature evolution of rotational-anisotropy second-harmonic generation at two doping levels. The nonlinear optical response exhibits order-parameter–like behavior with an onset temperature that coincides with the strange-metal–to–Fermi-liquid crossover, challenging the prevailing view of a trivial overdoped state. I will also describe the extension of our second-harmonic-generation platform to incorporate a tunable THz pump generated using multiple organic crystals. This capability enables selective excitation of low-energy collective modes while monitoring the nonlinear optical response, providing a pathway to identify the low-energy excitations coupled to the observed symmetry breaking.

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Theoretical and computational framework for nonlinear phononics

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Experiments in nonlinear phononics have shown that strong terahertz excitation can control a material's structural [1], magnetic[2], electric[3], and superconducting[4] properties by driving specific lattice modes. However, the lack of a fully ab initio computational framework limits the ability to screen candidate materials and complicates the interpretation of pump–probe measurements.

In this work, we present a theoretical and computational framework for modeling nonlinear phononics from first principles. The method treats the coupled dynamics of electrons, thermal phonons, and coherent phonon modes simultaneously. Electrons and thermal phonons evolve according to the Boltzmann transport equation, while the coherent amplitudes of all phonon modes follow nonlinear equations of motion. The framework includes both the damping of coherent modes due to electrons and thermal phonons, and the reciprocal impact of coherent phonons through their decay into electronic and phonon scattering channels. All electron–phonon and three-phonon interactions are computed strictly from first principles, without fitted parameters. Applications to a model system illustrate how this approach captures the key mechanisms governing terahertz-driven nonlinear phonon dynamics.

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Stimulated Moiré Phonon Scattering and Coherent Moiré Exciton-Phonon Transduction

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We demonstrate the first phonon-exciton transduction in a near-0 degree moiré $MoSe_2$ /WS_2 heterostructure via stimulated Raman scattering. An ultrafast below-bandgap pump drives out-of-plane lattice vibrations—arising from coupled interlayer breathing and shear phonons—that coherently modulate the moiré exciton resonances by altering the interlayer spacing. A later THz wavepacket, generated by pump-induced graphene acoustic phonons, excites the same modes through an incoherent driving mechanism.

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Carrier-coupled ultrafast structural dynamics and interlayer energytransfer of substrate-supported two-dimensional transition metaldichalcogenide heterostructures

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Understanding energy flow across layered two-dimensional heterostructures is crucial for controlling carrier and phonon transport as well as thermal management in next-generation optoelectronic devices. By using reflection ultrafast electron diffraction, we directly examine the photoinduced out-of-plane structural dynamics of supported MoS_2/WS_2 heterostructures and their monolayers separately. Experimental evidence reveals surprising early-time impacts of interlayer charge transfer on the out-of-plane atomic motions of van der Waals (vdW) heterojunctions compared to those of the individual monolayers. The enhanced optical absorption observed for heterostructures is also consistent with the notable interlayer electronic coupling. Following carrier–phonon coupling and carrier annihilation, we observe thermalized atomic motions that can be well described by the Debye model and a thermal dissipation picture. A higher thermal boundary conductance (TBC) across MoS_2/WS_2 heterostructures is obtained compared to those at the monolayer–substrate interfaces; however, the similar TBC values suggest comparable phonon scatterings across the vdW contacts. These results further shed light on the optical, phonon, and interfacial thermal properties of vertically-stacked vdW heterostructures.

The support by the Welch Foundation (E-2263) and the National Science Foundation (CHE-2154363) is acknowledged.

Imaging GHz Magnon Dynamics with Ultrafast Electron Microscopy

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Microwave-driven spin waves or magnons in magnetic thin films are promising medium for low-energy data transfer, where information is encoded in the phase and amplitude of coherent magnons. Operations in the GHz range also facilitate integration of magnonic devices with modern electronics systems. However, the propagation of the coherent magnons is often affected by magnetic defects (e.g. domain walls) and structural defects (grain boundaries) in magnetic films. Therefore, direct visualization and imaging of magnon propagation is key to understanding wave-defect interactions and optimizing next-generation magnonic device designs.

Here, we present our study of ultrafast imaging of coherent magnons [1] using a Lorentz transmission electron microscope (LTEM) equipped with a radiofrequency (RF) electron pulser [2-6]. The electron pulser acts as an ultrafast chopper that generates electron pulses from a continuous electron beam with a tunable repetition rate. To excite coherent magnons, we apply an RF signal at the same frequency to the sample via a RF compatible holder. This excitation signal is synchronized to the pulsed electron beam, enabling ultrafast real-space imaging. In this presentation, we show our study results obtained from soft magnetic thin films. We visualize how coherent magnons generate, propagate and scatter from defects. We also perform micromagnetic simulations to further interpret the observed phenomena [7].

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Upconversion photoluminescence from dark excitons via resonant exciton-exciton annihilation in few-layer transition metal dichalcogenides

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Two-dimensional semiconductors have become a promising platform for both studying the fundamental excitonic physics and realizing optoelectronic nanodevices. Benefit from the reduced dimensionality, the inherently strong Coulomb interactions give rise to pronounced excitonic effects, exhibiting strong light-matter interaction and exceptional optical properties. Despite the majority of exciton states are optically inactive (dark exciton), the dark exciton states are highly involved in the exciton dynamics and considerably prolong the lifetime of excitons, which further enhances the effects of inter-excitonic many-body interactions. Among the inter-excitonic many-body interactions, the exciton-exciton annihilation (EEA) is widely considered as the underlying mechanism that fundamentally limits the performance of two-dimensional semiconductors. In our joint theory-experiment work [1], we discover the upconversion photoluminescence (UPL) in few-layer transition metal dichalcogenides (TMDs) which is recognized as optically inactive materials due to the indirect band-gap. Based on the Wannier tight-binding (WTB)-based Bethe-Salpeter equation (BSE) [2], incorporating the first-principles calculated quasiparticle band structures and numerically carried out using the WannierBSE package [3], together with the microscopic many-body theory for excitons [4], we attribute the upconversion photoluminescence to a resonant exciton-exciton annihilation involving a pair of dark excitons with opposite momenta, followed by the spontaneous emission of upconverted high-lying bright excitons. Moreover, our simulations suggests that the interlayer coupling in few-layer TMDs plays crucial roles favoring the resonant upconversion process. Our findings reposition the roles of the EEA and open up a new route for both studying fundamental dark exciton physics and potential device applications.

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Probing broken time-reversal symmetry in 2D materials with tailored-light photocurrent generation

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Photocurrent generation underpins a wide range of electronic applications, such as current sources, switches, and photovoltaics. Due to the nonlinear process behind it, photocurrents can also be used to probe material properties in and out of equilibrium. In our recent work [1], we present a method to investigate fundamental symmetry breaking in a coupled light-matter system. Using a biharmonic field, we individually break or maintain time-reversal symmetry (TRS) and mirror symmetry. In inversion-symmetric graphene, two-color photocurrents originate from injection currents associated with asymmetric carrier populations. For preserved TRS in the combined system, equal excitation probabilities at conjugate momenta enforce symmetry-protected current suppression. From the interplay between field and material symmetries, we systematically derive photocurrent selection rules in the driven system. We benchmark our experimental results against time-dependent density functional theory simulations. Extending the approach to systems with broken TRS, we simulate two-color photocurrents in the magnetic 2D metal CrI_3 and in a Floquet topological insulator. The calculations reveal photocurrent generation driven by intrinsic TRS breaking, even for TRS-maintaining light fields. This enables direct, background-free detection of symmetry breaking in a material. Distinct phase dependencies further demonstrate the sensitivity of this method to underlying symmetry properties. Our work establishes photocurrent selection rules from ultrafast biharmonic fields as a non-invasive probe of symmetry-broken phases of matter, without using magnetic fields or circularly polarized light. Looking forward, it opens a pathway to directly probing material properties such as magnetic order, Floquet-engineered phases, and nonequilibrium symmetry breaking on femtosecond to attosecond timescales.

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Leveraging Ultrafast Microscopy to Probe Charge Carrier Dynamics at Semiconductor Surfaces

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Understanding how surfaces and interfaces influence charge carrier dynamics is critical for the performance of modern optoelectronic and energy materials. While bulk carrier transport has been extensively investigated, the microscopic processes governing carrier behavior near surfaces and heterointerfaces remain less well understood, particularly on ultrafast timescales. This challenge is especially pronounced in emerging material systems such as halide perovskites and two-dimensional semiconductors, where strong surface sensitivity, reduced dimensionality, and interfacial interactions can substantially reshape charge transport and recombination pathways.

Here we investigate how surface structure and interfacial coupling influence nonequilibrium charge carrier dynamics in semiconductor materials. Using a combination of ultrafast optical and electron-based techniques, we resolve carrier motion with both temporal and spatial sensitivity, allowing us to probe how surface termination, crystallographic orientation, and interfacial interactions modify carrier diffusion, recombination, and extraction pathways.

Ultrafast microscopic techniques, such as ultrafast electron microscopy (U-SEM) and transient photoluminescence microscopy (TPLM), provide a powerful route to directly visualize carrier dynamics across both space and time. By combining femtosecond temporal resolution with nanoscale spatial sensitivity, these approaches enable observation of carrier diffusion, trapping, and relaxation pathways that are often obscured in spatially averaged measurements. These measurements allow correlations between local structural or chemical variations and the resulting electronic response, revealing how microscopic heterogeneity governs the evolution of photoexcited carriers.

Establishing these relationships provides a framework for rational surface and interface engineering in semiconductor systems. By identifying how specific structural and chemical features influence carrier transport pathways, these insights can guide the design of materials and heterostructures that promote efficient charge extraction and suppress nonradiative losses, ultimately enabling improved performance in next-generation optoelectronic and energy-conversion technologies.

Nonlinear Lattice Dynamics Across a Ferroelectric Phase Transition with Two Dimensional THz Coherent Spectroscopy

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Thiourea ($SC(NH_2)_2$) stands as a classic example of molecular ferroelectricity, exhibiting a complex phase diagram driven by temperature-dependent lattice distortions. Upon cooling, the material leaves the paralectric phase and passes through a sequence of three distinct incommensurate phases characterized by molecular canting, before settling into a ferroelectric ground state [1]. A signature of this structural change is an infrared-active phonon mode that exhibits softening during cooling, followed by anomalous hardening subsequent to the ferroelectric phase transition. While linear spectroscopy can track these frequency shifts, it lacks the capacity to resolve the anharmonic interactions and microscopic couplings that fundamentally drive these transitions.

In this work, we employ two-dimensional terahertz (2D THz) coherent spectroscopy to directly probe the nonlinear lattice dynamics of thiourea across the temperature range where the structural phase transitions occur. While 2D THz methods have been widely applied to magnon and free-carrier dynamics [2,3], their application to structural phase transitions and phonon nonlinearities have been limited [4,5]. By transmitting a pair of intense time-delayed THz pulses through the sample, we generate 2D frequency-frequency correlation spectra that isolate the nonlinear vibrational response from the linear background. This multi-dimensional approach allows us to map the evolution of the soft mode's anharmonicity and its homogeneous dephasing. We present temperature-dependent spectra that elucidate how the vibrational nonlinearities and inter-mode couplings influence the lattice instability. Our findings provide a microscopic view of the energetic landscape governing the incommensurate-to-ferroelectric transformation, demonstrating the utility of 2D THz spectroscopy in characterizing the order parameters of soft molecular quantum materials.

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Control of Finite-Momentum Excitons in Transition Metal Dichalcogenide Monolayers by Spin-Orbit-Coupled Twisted Light

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In transition metal dichalcogenide monolayers (TMD-MLs), reduced dielectric screening enhances electron-hole Coulomb interactions, leading to pronounced excitonic effects and strong light-matter coupling. Here we theoretically study finite-momentum excitons driven by twisted light carrying spin angular momentum (SAM) and orbital angular momentum (OAM). To determine the band structure of finite-momentum excitons, we solve the Bethe-Salpeter equation (BSE), which requires the evaluation of electron-hole Coulomb matrix elements. To reduce this computational cost, we implement the WannierBSE package [1-3] to construct and solve the BSE within a density functional theory based Wannier tight-binding framework, fully incorporating direct and exchange Coulomb interactions. This approach quantitatively reproduces experimentally observed fine structures of bright, dark, and gray excitons and provides a transparent and physically intuitive formalism. Using momentum-dependent exciton dipole moments, we analyze exciton-twisted light interactions via time-dependent perturbation theory. We show that for bright excitons, the interplay between the valley phase with winding number two and optical OAM generates OAM-dependent exciton wave packets [4,5]. For gray excitons, we demonstrate that coupling between their out-of-plane dipole moments and the longitudinal field of twisted light, arising from optical spin-orbit interaction, enhances with optical OAM [6]. Moreover, we show that superposing twisted light fields with opposite OAM and SAM forms vector vortex beams capable of imprinting angular momentum information onto gray excitons even in the incoherent limit [6]. These results establish a route toward angular-momentum-based optical control of charge-neutral excitons in TMD-MLs.

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Steady-state nonequilibrium electron distribution in metals driven by CW laser excitation

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Ultrashort laser pulses excite electrons in metals to a state far from equilibrium, which subsequently thermalize back to equilibrium on a femto- to picosecond timescale. Thus, effects arising from the nonequilibrium electron distribution desirable for applications are considered to be inherently limited to ultrashort timescales. However, both experimental and theoretical studies indicate that an electronic nonequilibrium could persist for much longer under certain circumstances [1, 2]. Here, we demonstrate that a nonequilibrium electron distribution can be maintained as a steady state upon excitation with a continuous wave (CW) laser. Thus, the nonequilibrium properties become available at arbitrary timescales, which effectively corresponds to a novel state of matter. With a kinetic model based on microscopic Boltzmann collision integrals we find that the steady-state is determined by the competition between the continuous excitation, the thermalization due to electron-electron scattering, and different energy dissipation pathways. We show that the steady-state nonequilibrium can be controlled by the parameters of the exciting CW laser and that it affects observables like the material's optical response. Therefore, this novel state can be exploited to tailor the optical properties of metals. We also identify signatures of the steady-state nonequilibrium that can be probed experimentally with ultrashort laser pulses.

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Trefoil Floquet Engineering of Electronic Polarization

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Light-induced ferroelectric polarization has been demonstrated using resonant phonon excitation in the THz [1] and mid-infrared [2] regimes, where lattice motion plays a central role. In contrast, Floquet engineering offers a non-resonant route to manipulate electronic band structure and potentially induce purely electronic polarization without involving ionic displacement. However, direct observation of symmetry-breaking electronic polarization in centrosymmetric materials driven purely by near-infrared light has remained elusive. Here, we report transient electronic polarization induced by an engineered three-fold symmetric light field far off resonance in the centrosymmetric cubic perovskite $SrTiO_3$ at room temperature. The induced polarization, persisting for approximately 90 femtoseconds, is attributed to a Berry-phase mechanism arising from the Floquet–Bloch band structure. The non-monotonic fluence dependence reveals direct waveform control of electronic polarization. Time-dependent density functional theory simulations reproduce both the orientation and fluence dependence of the induced polarization. Our results establish a general protocol of symmetry-selective Floquet engineering applicable to a broad class of large-gap centrosymmetric materials.

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Disentangling exciton resonances in 2D materials and their heterostructures by interferometric time-resolved XUV-ARPES

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Ultrafast and coherent electron dynamics in 2D materials and their heterostructures are governed by the complex energy landscape of various excitons. These quasi-particles are often separated by only tens of meV, making them difficult to distinguish in conventional time-resolved experiments due to the spectral linewidths of ultrashort pulses. This challenge can be circumvented by interferometric multi-dimensional photoemission experiments, which have, for instance, been successfully employed to resolve (coherent) excitation pathways in molecular hybrid structures [1]. However, these experimental schemes rely primarily on optical pulses, which limit the accessible momentum space. This prevents the investigation of coherent phenomena in 2D or molecular materials, where their key spectroscopic features are often located at large electron momenta. To address these limitations, we present a modified version of interferometric time-and angle-resolved photoemission that employs two active, phase-stabilized IR pump pulses and a fs-XUV probe pulse for photoionization. We first show the capability of our approach by examining the A-exciton transition energies in a WSe_2 bulk crystal. Employing a temporal Fourier analysis of the ARPES spectrum allows us to identify distinct optical transition energies within our pump spectrum, which we can link to congruent excitation paths within the WSe_2 band structure. Similarly, we show how our methodology can disentangle intra-layer excitations and interlayer charge processes at a FePc/WSe_2 hybrid interface. In this way, our methodology provides a versatile tool to obtain a detailed view of competing excitations in low-dimensional hybrid structures.

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Observing phonon-driven Froquet states using quantum beat spectroscopy

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Floquet states, quantum states dressed by time-periodic Hamiltonians, provide a powerful framework for exploring nonequilibrium electronic configurations and ultrafast control of quantum matter. While light-driven Floquet states have been identified through energy-domain replicas in time-resolved photoemission, their experimental characterization is often limited by short lifetimes (∼100 fs) arising from weak nonlinear light-matter coupling [1].

Here, we report a direct time-domain observation of phonon-driven Floquet states at the graphene/Ir(111) surface, enabled by the intrinsically strong electron-phonon interaction of graphene. Using time-resolved two-photon photoemission spectroscopy (tr-2PPE), we observe long-lived oscillations in photoelectron intensity persisting for more than 6 ps. These oscillations are attributed to coherent graphene ZA-mode phonons [2] and manifest as pronounced quantum beats [3] in high-order image-potential state (IPS) populations.

The observed beating frequencies coincide with the coherent phonon oscillation frequency, providing direct evidence for phonon-dressed Floquet subbands emerging at the Brillouin-zone center. Importantly, phase-resolved population dynamics of the IPSs reveal a measurable delay between the lattice oscillation and electronic population, resolving the temporal ordering of Floquet physics: Floquet subband formation occurs promptly with the coherent phonon, followed by subsequent electronic population. [4]

These results establish image-potential states as a minimal and highly sensitive platform for probing driven quantum matter at surfaces, and demonstrate that coherent phonons can sustain Floquet states on timescales and with coupling strengths distinct from conventional optical driving. This work opens new avenues for exploring charge dynamics, energy flow, and nonequilibrium phases at strongly driven interfaces.

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Terahertz time-domain micro-spectroscopy of a dual-gated bilayer graphene device

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Achieving sub-diffraction spatial resolution in terahertz (THz) time-domain spectroscopy traditionally requires near-field confinement or on-chip integration, both of which involve extensive electromagnetic modeling and labor-intensive device fabrication. Here, we interface a dual-gated bilayer graphene device with the near field of a spintronic THz emitter and locally trigger THz emission using an ultrashort laser pulse. The confined THz source launches an in-plane graphene plasmonic wave that subsequently radiates into the far field, enabling comprehensive mapping of the device's THz electrodynamics. This approach establishes a pathway toward high-throughput, microscale THz characterization of two-dimensional devices.

Towards fully tunable time-resolved Momentum Microscopy at arbitrary (0-60 MHz) repetition rate

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Time- and angle-resolved photoemission spectroscopy (tr-ARPES) at 10s of MHz repetition rate driven by cavity-enhanced high-harmonic generation (CE-HHG) enables experiments that are difficult or impossible with conventional methods [1]. However, achieving complimentary tunable pump pulses at these high repetition rates has been an outstanding challenge. Moreover, certain experiments may require a reduced pump repetition rate to ensure complete sample recovery, suppress cumulative heating, or access material dynamics on longer time scales.

Conventional tunable pump sources at lower repetition rate based on optical parametric amplifiers (OPAs) rely on high pulse energy. Efficient nonlinear conversion in the low- pulse-energy regime for tens of MHz systems requires a different approach. Achieving both wavelength tunability and flexible repetition rate control while maintaining synchronization with the CE-HHG probe is thus nontrivial.

Here we present a tunable pump laser system with arbitrary (0-60 MHz) repetition rate, derived from the same master oscillator that seeds the CE-HHG probe source. The system is based on a high-power erbium fiber frequency comb incorporating chirped-pulse amplification. Tunable pump pulses spanning 1.3–2.0 eV are generated using fan-out periodically poled lithium niobate. A polarization modulation scheme implemented with an electro-optic modulator allows 0-60 MHz repetition rate control [2]. This source extends the flexibility of tr-ARPES by enabling resonant excitation across a broad range of materials while maintaining full synchronization and adjustable repetition rate.

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Two-Color Pump-Probe STM of Coherent Phonon Dynamics in Ultrathin ZnO/Ag(111)

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We use photon-assisted ultrafast STM (ph-USTM) to study optically resonant tunneling and coherent phonon (CP) dynamics in ultrathin ZnO/Ag(111) at the nanoscale. In a previous work [1], it was suggested that resonant optical excitation of the interface state to the ZnO conduction band is essential for CP spectroscopy via ph-USTM. Employing a new two-color pump-probe STM using different laser photon energies, we now systematically study the importance of on- and off-resonant optical excitation for ph-USTM-based CP spectroscopy. We show that the photocurrents and image contrast are determined by the optical resonance condition, and that the underlying optically resonant tunneling is crucial for detecting CPs via ph-USTM. Additionally, we discuss the local mode-selective coupling to the optical transition involved in the resonant photon-assisted tunneling process used as a probe. Furthermore, by measuring the dependence of the CP amplitude on the pump polarization, we show that the CPs are excited via the highly localized surface plasmon inside the tunneling gap.

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Exciton localization and dynamics in $MoSe_2/WS_2$ Heterostructures

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Moiré heterobilayers of transition metal dichalcogenides (TMDs) offer platforms for tuning of the electrical band structure [1], exciton localization [2], and potentially exciton hybridization [3] through precise control of the relative twist angle. Here we study the exciton landscape of $MoSe_2/WS_2$ moiré heterostructures using first-principles GW-BSE calculations, as well as complementary time- and angle-resolved photoemission spectroscopy (trARPES). We find that large twist angle bilayers have a type-1 band alignment resulting in $MoSe_2$ intralayer excitons while small twist angle bilayers simultaneously have regions of type-1 band alignment and type-2 band alignment yielding both long-lived intra- and interlayer excitons observable in trARPES. We find no exciton hybridization but instead explain the $MoSe_2$ absorption peaks using moiré-induced lattice reconstructions. In addition, we comment on the many-body effects arising in the trARPES data observed in these samples.

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What is the signature of a trion in photoemission?

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Recent advances in time- and angle-resolved photoemission spectroscopy (tr-ARPES) allow for the probing of multiparticle excited-states in reciprocal space. While neutral two-particle excitations (excitons) have been observed in tr-ARPES, signatures of trions—three-quasiparticle bound states—have only been probed via optical spectroscopy. In this presentation, we develop a general theory for the ARPES signature of trions in the model system of a monolayer transition metal dichalcogenide (TMD). We simulate the ARPES signals of both positively and negatively charged trions and show that the interaction of the residual holes, or electron and hole, lead to large energy shifts, on the order of the exciton binding energy, compared to the exciton signal. For positive trions, the additional momentum degree of freedom of the residual particles leads to distinctive asymmetric lineshapes. For negative trions, the photoemission process causes the tr-ARPES spectrum to reproduce inverted images of the exciton band structure for multiple exciton states, encompassing both spin-allowed and spin-forbidden states, providing a direct momentum-resolved probe of both trion and exciton physics.

Ultrafast electron dynamics in strongly correlated perovskites $LaFeO_3(001)$ and $BiFeO_3(001)$

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Perovskite oxides are characterized by their highly tunable structural and electronic properties. A key factor influencing their electronic behaviour is electron-electron correlation, which can lead to metal-insulator transitions based on the strength of on-site Coulomb repulsion (U). We are interested in the electron dynamics of strongly correlated perovskites, particularly we study the charge-transfer insulators $LaFeO_3$ and $BiFeO_3$, bridging from antiferromagnetic to ferroelectric properties. Up to now, time-resolved measurements with above-band gap excitation of highly correlated systems are quite rarely studied. Two examples beyond perovskites are measurements on NiO(100) [1] and $FePS_3$ (Mott insulator) [2], revealing complex relaxation pathways, which are coupled to the antiferromagnetic spin system.

We use time-resolved two-photon photoemission to study the electron dynamics in epitaxially grown $LaFeO_3(001)$ and $BiFeO_3(001)$ layers on $SrRuO_3/DyScO_3$. We excite electrons below and above band gap, which opens between strongly hybridized oxygen 2p/Fe 3d and Fe 3d minority states for both perovskites. Pumping above band gap, we find in both systems a broad state at $\sim$1.3 eV above E_F with a short lifetime of 30 fs. Towards the conduction band minimum, we observe a biexponential decay, with significantly varying lifetimes of 10 to 30 fs and 1 ps [3]. The slower decay channel shows an energy dependent increase in lifetime towards the Fermi level. For $BiFeO_3$ the fast decay channel shows a low amplitude, with ultrafast dynamics (¡10 fs) attributed to coupling with bulk states, whereas $LaFeO_3$ exhibits a higher probability for the fast decay channel.

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Detailed energy transport dynamics observed across solid-molecule junctions

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Interfaces play a crucial role in energy transport at the nanoscale. However, direct experimental observations of interfacial thermal conductance across molecular junctions have remained challenging. In this talk, we report dynamic energy transport processes across a molecular junction observed at the atomic level by employing reflection ultrafast electron diffraction. A clear temporal sequence of energy transfer is revealed at early times following photoexcitation of Au(111) surfaces chemically bonded with self-assembled monolayers (SAMs) of alkanethiols: from the gold surface layer (SL) to the head groups (HGs) of a SAM and then to the methylene lattice. Remarkably, the structural dynamics of the gold SL differ significantly from those of clean gold. Furthermore, the methylene lattice dynamics exhibit chain-length independence but with a length-dependent retention time to reach full thermalization. We find an ultrahigh thermal conductivity for the methylene lattice and a high interfacial thermal conductance at early times under the condition of impulsive heating. Moreover, a transient ordering at the Au–HG is observed at longer times. From the rich dynamics information, we anticipate that this time- and spatially-resolved experimental approach will enable future quantitative assessment of interfacial heat transport across solid–molecule junctions.

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Selective control of nonequilibrium multiferroic dynamics with tunable strain

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Quantum materials are characterized by intertwined orders, and a fundamental goal in condensed matter physics is to achieve their selective control. While significant progress has been made in thermal equilibrium, realizing such control out of equilibrium remains highly challenging. Here we employed *in situ* tensile strain to individually modulate the photoinduced lattice dynamics associated with ferroelectric and magnetic orders in multiferroic $BiFeO_3$. By applying MeV ultrafast electron diffraction to freestanding $BiFeO_3$ membranes under tunable strain, we showed that tensile strain markedly suppresses the ultrafast photoinduced reduction of the ferroelectric displacement. By contrast, the photoinduced dynamics of the antiferrodistortive rotation of the oxygen octahedra, which modulates the magnetic order, remains insensitive to strain. Not only do these findings reveal distinct microscopic pathways underlying nonequilibrium multiferroic dynamics, they also establish tunable strain as an effective route for engineering ultrafast phase control in correlated materials.

Time-resolved tunneling spectroscopy of ultrafast surface dynamics with single atom resolution

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We introduce lightwave-driven terahertz scanning tunneling microscopy (THz-STM) [1] as a platform for tracking the ultrafast evolution of local electronic structure following resonant interband excitation with single atom resolution. We apply this approach to the photoexcited GaAs(110), where we directly image femtosecond-scale carrier dynamics and track transient photocurrents in the vicinity of individual surface defects. We highlight how photoinduced charge carriers give rise to time-dependent band bending across the atomic-scale landscape of the sample surface. Critically, THz time-domain spectroscopy in the tip near-field [1] allows us to disentangle competing contributions to the tunnel current, separating the coherent sub-cycle THz-driven component from the intrinsic material response. We establish a new regime of ultrafast tunneling spectroscopy capable of tracking transient electronic structure and Fermi level shifts with unprecedented spatio-temporal precision, opening new pathways for understanding defect-mediated transport in materials.

References: [1] V. Jelic et al., Nat. Photon. 18, 898 (2024)

Spatio-Temporal Electron Propagation Dynamics in Au/Fe/MgO(001) in Non-Equilibrium: Revealing Single Scattering Events and the Ballistic Limit

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Optically excited electrons in metals travel ballistically on a femtosecond timescale until they scatter due to electron-electron or electron-phonon interactions. To gain a deeper unterstanding of the ballistic and rarely scattered electrons under non-equilibrium conditions we combine time-resolved two-photon photoelectron emission spectroscopy (*tr*-2PPE) with real-time time-dependent density functional theory (RT-TDDFT) as well as a random-walk-like transport simulation [1]. Here, we discuss the experimental details using a back-side pumped geometry on a MgO/Fe/Au epitaxial heterostructure probing the Au surface. RT-TDDFT show in conjunction with experimental data an electron injection channel at the Fe/Au interface at $E - E_\mathrm{F} = 1.7\,\mathrm{eV}$. The time delayed response of the photoelectrons at lower energies is used to investigate transport and scattering pathways. Analyzing this time delay shows an apparent acceleration with increasing film thickness. We find the electron trajectories angular dependence is key to explain the apparent thickness dependent acceleration and the random-walk-like transport simulation find ballistic carriers close to the injection energy. Funding by the DFG through Project No. 278162697 - SFB1242 is gratefully acknowledged.

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Ultrafast Charge-Driven Molecular Motion at a Hybrid Molecule–2D Material Interface

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Understanding how electronic excitations reshape interfacial energy landscapes is central to designing functional hybrid materials. In particular, non-equilibrium charge transfer at molecule-2D material interfaces can transiently modify intermolecular and molecule-substrate interactions, potentially enabling collective molecular motion that is inaccessible in equilibrium.

Here, we investigate a prototypical hybrid interface consisting of a self-assembled monolayer of copper(II)phthalocyanine (CuPc) on $TiSe_2$. Using a multimodal, time-resolved photoemission approach, we combine time- and angle-resolved photoemission (trARPES), orbital tomography, core-level spectroscopy (trXPS), and X-ray photoelectron diffraction (trXPD) to simultaneously track charge flow, orbital evolution, and atomic rearrangements with femtosecond resolution [1].

The combination of extreme-ultraviolet and soft X-ray probe pulses, provided by free-electron laser (FEL) radiation, enables momentum-resolved access to valence states together with element- and site-specific core-level sensitivity. Upon optical excitation, hot hole transfer from $TiSe_2$ into the molecular layer modifies the interfacial electrostatic potential, charging a substantial fraction of molecules within a few hundred femtoseconds. The resulting redistribution of Coulomb and van der Waals interactions drives a concerted azimuthal rotation of molecules in the densely packed monolayer, accompanied by transient out-of-plane deformation.

Our results demonstrate how FEL-based, element-specific ultrafast spectroscopy provides direct access to correlated electronic and structural dynamics at buried hybrid interfaces. This approach establishes a pathway toward controlling collective molecular motion and transient symmetry breaking in energy-driven molecular materials.

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Terahertz plasmonics at the graphene/$NiPS_3$ interface tuned by antiferromagnetic order

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The interaction between plasmons and magnons is a long-sought phenomenon with implications for fundamental physics and spin-plasmonics. In three-dimensional systems, this coupling is suppressed by the large mismatch in energy scales, but two-dimensional (2D) plasmons hybridize with electromagnetic fields to form plasmon-polaritons with acoustic-like dispersions that can overlap magnons spectrally. Despite numerous theoretical predictions, experimental observation of magnon-plasmon interaction has never been achieved.

In this work, we study a first-of-its-kind hybrid plasmon-magnon platform based on 2D materials. By deploying scattering-type scanning near-field optical microscopy (s-SNOM) with terahertz radiation, we perform spatial-temporal imaging on plasmon wavepackets at a graphene/$NiPS_3$ interface and track their dispersion across the antiferromagnetic transition of $NiPS_3$. We observe a clear renormalization of the plasmon dispersion concurrent with the onset of antiferromagnetic order, demonstrating tunability of terahertz plasmonic modes by magnetic order formation. These results establish a controllable platform for hybrid magnon–plasmon interactions in 2D materials and open avenues for coherent spin–plasmon devices and tunable terahertz spintronic components.

Visualizing Rabi-Oscillations in Graphene using strong-field time-resolved ARPES

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The interaction of intense few-cycle laser pulses in the petahertz regime with solids enables access to non-perturbative light–matter coupling on sub-femtosecond timescales. In graphene, the gapless Dirac dispersion and strong optical coupling provide ideal conditions to drive coherent interband dynamics in the strong-field regime [1]. Under sufficiently high peak electric fields, the electronic population undergoes Rabi oscillations between valence and conduction band states, resulting in characteristic quasi-periodic modulations of the transient band occupation within the Dirac cone [2].

Here, we discuss first preliminary results of observing such strong-field Rabi dynamics using time- and angle-resolved photoemission spectroscopy (tr-ARPES) with few-cycle near-infrared pump pulses [3]. As a surface-sensitive probe with full energy- and momentum resolution, trARPES allows direct access to the ultrafast evolution of electronic populations at surfaces and in two-dimensional materials. The method is therefore ideally suited to resolve coherent field-driven carrier dynamics in momentum space and to disentangle them from incoherent relaxation pathways.

Based on experiments employing 7 fs pump pulses on graphene, we evaluate the experimentally accessible field-strength regime and address central challenges such as space-charge effects and ultrafast dephasing. The experimental findings are directly compared to simulations of a driven Dirac system, which reveal the expected momentum-resolved signatures of Rabi oscillations and their dependence on peak electric field strength and polarization.

These efforts aim to establish trARPES as a direct probe of coherent strong-field electron dynamics at surfaces, opening new routes toward electric-field control of ultrafast electronic processes in two-dimensional materials.

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Gate-Tunable Atomically Resolved Light Emission in a Monolayer Semiconductor

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The microscopic origin of quantum emitters in two-dimensional (2D) semiconductors remains a subject of intense debate [1, 2]. While sharp emission lines are frequently attributed to atomic point defects, verifying this hypothesis, and rationally optimizing these sources, requires correlating optical spectra with atomic structure beyond the diffraction limit. Scanning tunneling microscopy-induced luminescence (STML) offers this capability but has been hindered in 2D materials by a critical trade-off: metallic substrates quench emission and hybridize with the semiconductor, while insulating substrates impede stable electron tunneling at cryogenic temperatures.

Here, we present a device architecture that overcomes these limitations, enabling the first gate-tunable STML of monolayer $MoSe_2$ in the intrinsic limit. By using a thick hexagonal boron nitride gate dielectric with a graphene nanoribbon contact array, we ensure an electronic decoupling from the metallic substrate while maintaining stable carrier injection. This platform grants independent control over carrier density and injection energy, enabling the selective electrical generation of negatively and positively charged trions.

Importantly, we provide a direct spatial correlation between individual atomic defects and their optical signatures. Contrary to the prevailing assumption that intrinsic point defects serve as single-photon emitters, we demonstrate that charged chalcogen vacancies act as efficient non-radiative recombination centers that quench luminescence, whereas isovalent defects are optically inactive. Our work opens new opportunities for functionalizing atomically confined emitters and investigating nanoscale optical properties in 2D semiconductors, including moiré superlattices.

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Signatures of THz-Frozen Orbital Order in $KCuF_3$

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The effect of strong terahertz (THz) fields on the fluctuations in materials has garnered attention due to their ability to stabilize fluctuating order [1] or create non-equilibrium states [2]. However, the THz response of systems with dynamical orbital fluctuations coupled to quantum magnetism remains unexplored. We report signatures of THz-stabilized orbital order and spin chain excitations in $KCuF_3$, a material in which strong fluctuations are present in both the orbital and spin degrees of freedom.

$KCuF_3$ exhibits dynamical fluctuations between two orbital ordering configurations that freeze below $T_R \sim 50$ K, as well as quantum fluctuations of spin-$\frac{1}{2}$ Heisenberg antiferromagnetic chains that order at $T_N \sim 40$ K. In our measurements, we find linear THz excitation of Raman active modes both above and below T_R, contradicting the selection rule expected for an inversion-symmetric fluctuating regime. This hints at the possibility of THz-induced freezing of dynamical orbital fluctuations. Furthermore, we detect a displacement signal driven by THz, which persists above T_N. 2D THz spectroscopy reveals that the displacement originates from THz rectification with spectral weight below 0.4 THz, pointing to a possible signature of the low-energy excitations of the quantum spin chain.

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