

# Symposium Ultrafast Excitation Pathways of Quantum Materials (SYUE)

jointly organised by  
the Surface Science Division (O),  
the Low Temperature Physics Division (TT), and  
the Thin Films Division (DS)

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Ultrafast laser experiments provide avenues to utilizing ultrafast light-matter interaction to control macroscopic properties of quantum materials. While conventional photo-induced processes in condensed matter often rely on ultrafast laser heating, there are intriguing pathways towards emergent macroscopic properties in quantum materials that are inherently non-thermal in nature. The mechanisms behind such pathways include transient modifications of the free-energy landscape by population redistributions, the dynamical modification of coupling strengths, and coherent light-matter dressing via Floquet engineering. This symposium brings together leading researchers in the field to showcase experimental and theoretical progress and discuss new avenues.

## Overview of Invited Talks and Sessions

(Lecture hall HSZ 01)

### Invited Talks

SYUE 1.1	Wed	9:30–10:00	HSZ 01	<b>Dynamics and control in quantum materials using multi-terahertz spectroscopy</b> — ●RICHARD AVERITT
SYUE 1.2	Wed	10:00–10:30	HSZ 01	<b>Accessing the nonthermal phonon populations in 2D materials with femtosecond electron diffuse scattering</b> — ●HÉLÈNE SEILER, MARIOS ZACHARIAS, DANIELA ZAHN, PATRICK-NIGEL HILDEBRANDT, THOMAS VASILEIADIS, YOAV WILLIAM WINDSOR, YINGPENG QI, CHRISTIAN CARBOGNO, CLAUDIA DRAXL, RALPH ERNSTORFER, FABIO CARUSO
SYUE 1.3	Wed	10:30–11:00	HSZ 01	<b>Exciting potentials – Exploring the realms of ultrafast phase transitions</b> — ●LAURENZ RETTIG
SYUE 1.4	Wed	11:15–11:45	HSZ 01	<b>Sub-cycle multidimensional spectroscopy of strongly correlated materials</b> — VIKTOR VALMISPILD, EVGENY GORELOV, MARTIN ECKSTEIN, ALEXANDER LICHTENSTEIN, HIDEO AOKI, MIKHAIL KATSNELSON, MISHA IVANOV, ●OLGA SMIRNOVA
SYUE 1.5	Wed	11:45–12:15	HSZ 01	<b>Witnessing many-body entanglement in light-driven quantum materials</b> — ●MATTEO MITRANO, DENITSA BAYKUSHEVA, MONA KALTHOFF, DAMIAN HOFMANN, MARTIN CLAASSEN, DANTE KENNES, MICHAEL SENTEF
SYUE 1.6	Wed	12:15–12:45	HSZ 01	<b>Optical responses of photoexcited materials: from parametric amplification to photoinduced superconductivity</b> — ●EUGENE DEMLER

### Sessions

SYUE 1.1–1.6	Wed	9:30–12:45	HSZ 01	<b>Ultrafast Excitation Pathways of Quantum Materials</b>
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## SYUE 1: Ultrafast Excitation Pathways of Quantum Materials

Time: Wednesday 9:30–12:45

Location: HSZ 01

**Invited Talk** SYUE 1.1 Wed 9:30 HSZ 01  
**Dynamics and control in quantum materials using multi-terahertz spectroscopy** — ●RICHARD AVERITT — UC San Diego, Department of Physics

Dynamics and control of quantum materials using light has emerged as a frontier research area. There are numerous possibilities to investigate including photoinduced metastability, creating non-equilibrium states with emergent properties, driving coherent many-body dynamics, or impulsively driving an order parameter to investigate the nonlinear \*optical\* response which can encode properties not evident with linear optical probes. In this vein, terahertz to mid-infrared spectroscopy is a particularly useful probe of low energy electrostatics in quantum materials in both the static and dynamic limits. I will present examples from my research group. This includes: (i) Efficient coherent magnon generation in the Mott insulator  $\text{Sr}_2\text{IrO}_4$  using sub-gap circularly polarized mid-IR pulses. (ii) Gentle photoexcitation of the putative excitonic insulator  $\text{Ta}_2\text{NiSe}_5$  resulting in stimulated nonlinear parametric terahertz generation that serves as a reporter of the excitonic condensate and its coupling to phonons.

**Invited Talk** SYUE 1.2 Wed 10:00 HSZ 01  
**Accessing the nonthermal phonon populations in 2D materials with femtosecond electron diffuse scattering** — ●HÉLÈNE SEILER<sup>1,2</sup>, MARIOS ZACHARIAS<sup>1,3</sup>, DANIELA ZAHN<sup>1</sup>, PATRICK-NIGEL HILDEBRANDT<sup>1</sup>, THOMAS VASILEIADIS<sup>1</sup>, YOAV WILLIAM WINDSOR<sup>1,4</sup>, YINGPENG QI<sup>1</sup>, CHRISTIAN CARBOGNO<sup>1</sup>, CLAUDIA DRAXL<sup>5</sup>, RALPH ERNSTORFER<sup>1,4</sup>, and FABIO CARUSO<sup>6</sup> — <sup>1</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — <sup>2</sup>Freie Universität Berlin, Berlin, Germany — <sup>3</sup>INSA de Rennes, 35708 Rennes, France — <sup>4</sup>Technische Universität Berlin, Berlin, Germany — <sup>5</sup>Humboldt-Universität zu Berlin, Berlin, Germany — <sup>6</sup>Christian-Albrechts-Universität zu Kiel, Kiel, Germany

Femtosecond electron diffuse scattering (FEDS) is a powerful technique to access nonthermal phonon populations in photo-excited materials. We perform FEDS experiments to study the microscopic energy flow in two prototypical 2D materials, black phosphorus and  $\text{MoS}_2$ . The experiments are complemented by first-principles calculations of the coupled electron-phonon dynamics based on the time-dependent Boltzmann equations. We also introduce an efficient first-principles methodology for the calculation of the all-phonon inelastic scattering in solids, and demonstrate its broad applicability to other 2D materials. Our joint experimental-theoretical approach provides a detailed picture of nonthermal phonons in photo-excited materials.

**Invited Talk** SYUE 1.3 Wed 10:30 HSZ 01  
**Exciting potentials – Exploring the realms of ultrafast phase transitions** — ●LAURENZ RETTIG — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Phase transitions, ubiquitous in nature, are characterized by breaking of characteristic material symmetries, which can be described by an order parameter. The phase of a material is then determined by the shape and the minima of the free energy surface. While in thermal equilibrium, a system occupies the phase space close to these minima, and phase transitions occur as function of thermodynamic state variables, ultrafast laser excitation allows modifying the energy surfaces on timescales faster than the response time of the system, thereby driving ultrafast phase transitions, and triggering coherent dynamics within the transient energy surfaces. This approach not only provides information about the microscopic couplings behind a phase transition, but also allows for coherent manipulation of material properties.

In my talk, I will discuss recent experiments employing time- and angle-resolved photoemission spectroscopy (trARPES), unearthing rich dynamics of photoinduced phase transitions in prototypical charge-density wave materials, including a transition into a hidden, metastable state.

**15 min. break**

**Invited Talk** SYUE 1.4 Wed 11:15 HSZ 01  
**Sub-cycle multidimensional spectroscopy of strongly cor-**

**related materials** — VIKTOR VALMISPILD<sup>3</sup>, EVGENY GORELOV<sup>3</sup>, MARTIN ECKSTEIN<sup>6</sup>, ALEXANDER LICHTENSTEIN<sup>4,5</sup>, HIDEO AOKI<sup>7</sup>, MIKHAIL KATSNELSON<sup>8</sup>, MISHA IVANOV<sup>1,9</sup>, and ●OLGA SMIRNOVA<sup>1,2</sup> — <sup>1</sup>Max-Born-Institut Berlin — <sup>2</sup>Technische Universität Berlin — <sup>3</sup>European XFEL, Germany — <sup>4</sup>Institute of Theoretical Physics, University of Hamburg — <sup>5</sup>The Hamburg Centre for Ultrafast Imaging — <sup>6</sup>Department of Physics, University of Erlangen-Nuremberg — <sup>7</sup>National Institute of Advanced Industrial Science and Technology (AIST), Japan — <sup>8</sup>Institute for Molecules and Materials, Radboud University, — <sup>9</sup>Department of Physics, Imperial College London

Strongly correlated solids are extremely complex and fascinating quantum systems, where new states continue to emerge and where interaction with light may trigger interplay between them. In this interplay, sub-laser cycle electron response is particularly attractive as a tool for ultrafast manipulation of matter at PHz scale.

We introduce a new type of non-linear multidimensional spectroscopy, which allows us to unravel the sub-cycle dynamics of strongly correlated systems interacting with few-cycle infrared pulses and the complex interplay between different correlated states evolving on the sub-femtosecond time-scale. We use laser-driven two-dimensional Hubbard model to resolve sub-cycle transitions between Mott-insulating and metallic states, and follow pathways of charge and energy flow leading to final non-equilibrium state stabilized by correlations after the end of the laser pulse.

**Invited Talk** SYUE 1.5 Wed 11:45 HSZ 01

**Witnessing many-body entanglement in light-driven quantum materials** — ●MATTEO MITRANO<sup>1</sup>, DENITSA BAYKUSHEVA<sup>1</sup>, MONA KALTHOFF<sup>2</sup>, DAMIAN HOFMANN<sup>2</sup>, MARTIN CLAASSEN<sup>3</sup>, DANTE KENNES<sup>2,4</sup>, and MICHAEL SENTEF<sup>2</sup> — <sup>1</sup>Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA — <sup>2</sup>Max Planck Institute for the Structure and Dynamics of Matter, Center for Free-Electron Laser Science (CFEL), Luruper Chaussee 149, 22761 Hamburg, Germany — <sup>3</sup>Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, PA 19104, USA — <sup>4</sup>Institut für Theorie der Statistischen Physik, RWTH Aachen University, 52056 Aachen, Germany and JARA-Fundamentals of Future Information Technology, 52056 Aachen, Germany

Many-body entanglement in condensed matter systems can be diagnosed from equilibrium response functions through the use of entanglement witnesses and operator-specific quantum bounds. Here, we investigate the applicability of this approach for detecting entangled states in quantum systems driven out of equilibrium. We use a multipartite entanglement witness, the quantum Fisher information, to study the dynamics of a paradigmatic fermion chain undergoing a time-dependent change of the Coulomb interaction. Our results show that the quantum Fisher information is able to witness distinct signatures of multipartite entanglement both near and far from equilibrium that are robust against decoherence. We discuss implications of these findings for probing entanglement in light-driven quantum materials with time-resolved optical and x-ray scattering methods.

**Invited Talk** SYUE 1.6 Wed 12:15 HSZ 01

**Optical responses of photoexcited materials: from parametric amplification to photoinduced superconductivity** — ●EUGENE DEMLER — ETH, Zurich, Switzerland

Optical drives at terahertz and mid-infrared frequencies in quantum materials are commonly used to explore the nonlinear dynamics of interacting many-body systems. Recent experiments demonstrated several surprising optical properties of transient states induced by driving, including the appearance of photo-induced edges in the reflectivity, enhancement of reflectivity, and even light amplification. I will show that many of these unusual properties can be understood from the general perspective of reflectivity from Floquet materials, in which pump-induced oscillations of a collective mode lead to parametric generation of excitation pairs. This analysis predicts a universal phase diagram of drive induced features in reflectivity, which evidence a competition between driving and dissipation. I will argue that this mechanism explains several recent experimental observations, including photoinduced superconductivity in the pseudogap phase of high  $T_c$  cuprates.