MA 24: Focus Session: Nonlinear Spectroscopy of Collective Excitations in Quantum Magnets (joint session TT/MA)

In recent years, significant progress has been made in understanding strongly correlated quantum magnets, with a particular focus on fractionalized states of matter such as quantum spin liquids. These achievements in understanding have been made possible by remarkable developments in both materials science and experimental techniques. In particular, improvements in both traditional experimental tools (e.g., inelastic neutron scattering, Raman scattering, resonant X-ray scattering, etc.) and the introduction of innovative techniques such as 2D coherent THz spectroscopy and sophisticated noise experiments have advanced studies of quantum matter to qualitatively new levels of insight. This focus session will discuss these recent advancements in nonlinear spectroscopy techniques along with theoretical inroads in describing the nonlinear spectroscopic signatures of complex quantum magnets.

Organizers: Simon Trebst (Universität zu Köln), Johannes Knolle (TU München)

Time: Wednesday 9:30–12:45 Location: H36

Topical Talk MA 24.1 Wed 9:30 H36

Detecting Anyons Using Nonlinear Pump-Probe Spectroscopy — •Max McGinley[1,2], Michele Fava[2], and Sid Parameswaran[2] — [1]Cambridge University, UK — [2]Oxford University, UK

Topologically ordered two-dimensional systems can host excitations that possess statistics that interpolate between bosonic and fermionic—so called anyons. In this talk, I will explain how the presence of such anyonic excitations can be inferred from nonlinear spectroscopic quantities. In particular, we consider pump-probe spectroscopy, where a sample is irradiated by two light pulses with an adjustable time delay between them. The relevant response coefficient exhibits a universal form that originates from the statistical phase acquired when anyons created by the first pulse braid around those created by the second. This behaviour is shown to be qualitatively unchanged by non-universal physics including non-statistical interactions and small finite temperatures. In magnetic systems, the signal of interest can be measured using currently available terahertz-domain probes, highlighting the potential usefulness of nonlinear spectroscopic techniques in the search for quantum spin liquids. I will discuss future prospects for inferring properties of collective excitations using analogous techniques.

Topical Talk MA 24.2 Wed 10:00 H36

Two-Dimensional Nonlinear Dynamic Response of Frustrated Magnets — •Wolfram Brenig — Institute for Theoretical Physics, Technical University Braunschweig, D-38106 Braunschweig, Germany

Two-dimensional nonlinear (2DNL) coherent optical spectroscopy is of great interest in order to deconvolute excitation continua in correlated magnets, potentially allowing to analyze individual quasiparticles, including those of fractionalized magnets. We discuss the relevant response functions for the coupling of spin systems to electric fields and analyze the 2DNL dynamical susceptibilities for two scenarios of frustrated magnetism, namely for a quantum spin-liquid (QSL) as well as for a case of incommensurate spiral long-range order (ICO). For the former, we consider the Kitaev magnet, which hosts a quantum spin-liquid, featuring fractionalization in terms of mobile Majorana fermions and static flux-visons. We show that the 2DNL response does not only probe characteristic features of both fractional excitations, but also allows to extract single quasiparticle lifetimes from its multi-particle continua. These properties will be discussed over a wide range of temperatures. For the case of 2DNL response from a magnet with ICO, we chose the J_1-J_3 spin-model on the square lattice. Here, some features of the 2DNL spectra are found to be remarkably similar to those of the QSL case. Going beyond a bare quasiparticle approach, we will also comment on the impact of final-state interactions.

Work done in collaboration with Olesia Krupnitska and profiting from interactions with Roser Valentí, Natalia Perkins, Marius Möller, Anna Keselman, and David Kaib.

Topical Talk MA 24.3 Wed 10:30 H36

Imaging Magnetization Dynamics and Collective Spin Excitations in Compensated Magnets on Ultrafast Timescales — •Benjamin Stadtmüller — Experimentalphysik II, Institute of Physics, Augsburg University, 86159 Augsburg, Germany

Fundamental to the advancement of spintronics and quantum technologies is the ability to encode, manipulate and store information in the spin angular momentum of electrons on ever faster timescales. In this contribution, we therefore discuss the ultrafast magnetic response of compensated magnets, which are interesting candidates for applications due to their robustness against external fields and their fast manipulation speed. We start with the ultrafast magnetization dynamics of conventional antiferromagnets (AFMs), for which the possibility of optical excitation of collective magnon modes on ps timescales has already been demonstrated. For the case of NiO, we show that these timescales can be further reduced by exploiting the strong nonequilibrium excitation with fs laser pulses. These conditions lead to a significant loss of magnetic order and to the excitation of collective magnon modes. We then turn to the ultrafast optical response of the recently discovered class of altermagnets with their d-wave-like spin split band structure. By combining theoretical calculations with ultrafast magneto-optical experiments, we demonstrate the generation of a macroscopic spin polarization in the otherwise fully compensated altermagnet RuO_2, which can additionally be controlled by the excitation geometry [1]. [1] M. Weber and S. Wust et al. arXiv: 2408.05187

15 min. break

Topical Talk MA 24.4 Wed 11:15 H36

Revealing Dynamics of Hidden Sectors with Nonlinear Spectroscopy — •Yoshito Watanabe[1], Simon Trebst[1], and Ciarán Hickey[2,3] — [1]Institute for Theoretical Physics, University of Cologne, Cologne, Germany — [2]School of Physics, University College Dublin, Belfield, Dublin 4, Ireland — [3]Centre for Quantum Engineering, Science, and Technology, University College Dublin, Dublin 4, Ireland

Nonlinear spectroscopy, especially in its two-dimensional coherent spectroscopy (2DCS) form, is an emerging and promising tool for studying the dynamics of quantum materials. Unlike traditional linear probes, 2DCS employs a multi-pulse approach that reveals intricate dynamics, including the ability to resolve fractional excitation continua as sharp spinon-echo signals and to study interactions between excitations, phenomena often obscured in traditional measurements.

In this work, we focus on the potential of 2DCS to detect and characterize quadrupolar excitations in quantum magnets. Using exact diagonalization and establishing an effective Hamiltonian that reflects the dynamics of hidden sectors and higher-order excitations, we identify distinct spectroscopic features, including new signatures associated with quadrupolar excitations. These results provide a guide for the experimental detection and characterization of hidden dynamics in quantum materials.

Topical Talk MA 24.5 Wed 11:45 H36

Theory of Nonlinear Spectroscopy of Quantum Magnets — Anubhav Srivastava[1,2], •Stefan Birnkammer[1], GiBaik Sim[3], Michael Knap[1], and Johannes Knolle[1,4] — [1]Technical University of Munich, Garching, Germany — [2]Indian Institute of Science, Bengaluru, India — [3]Hanyang University, Seoul, Korea — [4]Imperial College London, London, United Kingdom

Two-dimensional coherent spectroscopy (2DCS) is an established method for probing molecules and has been proposed in the THz regime as a new tool for probing exotic excitations of quantum magnets but the precise nature of coupling between pump field and spin degrees of freedom has remained unclear. Here, we develop a general response theory of 2DCS and show how magneto-electric as well as polarization couplings contribute to 2DCS in addition to the standardly assumed magnetization. We propose experimental protocols to distill individual coupling contributions, for example from exchange-striction or spin current mechanism. We provide example calculations for the paradigmatic twisted Kitaev chain material $CoNb_2O_6$ and highlight the crucial role of contributions from cross-coupling between polarization and magnetic nonlinear susceptibilities. Our work paves the way for systematic studies of light-matter couplings in quantum magnets and for establishing 2DCS as a versatile tool for probing fractional excitations of exotic magnetic quantum phases.

MA 24.6 Wed 12:15 H36

Quantitative Prediction of the Dynamics of In-Gap States in Correlated Materials as Seen in Pump-Probe PES, XAS and RIXS Experiments: A NiO Case Study — •Sina Shokri and Maurits W. Haverkort — Universität Heidelberg, Institut für Theoretische Physik, Philosophenweg 19, Heidelberg 69120 Germany

Attosecond pump-probe experiments allow one to study and steer quantum materials on their fundamental time-scales. For atoms and small molecules one can theoretically predict the electronic and vibrational dynamics induced by ultra-fast light pulses [1,2]. In solids a theoretical understanding is much harder. The coupling to many continuous degrees of freedom can result into rapid loss of coherence. Quantitative predictions how coherently driven excitations decohere is highly non-trivial. Correlated Mott- Hubbard or charge transfer insulators can show a variety of long lived excitonic excitations within the optical gap. With attosecond pump-probe spectroscopy it is possible to investigate the propagation and decay of such excitations, as recently shown by two-photon photo-emission spectroscopy of NiO. These experiments show photo-induced, long-lived in-gap states with coherent oscillations [3]. In this talk we will show, using non-linear response theory, how to quantitatively predict the dynamics of in-gap states in correlated materials after an optical excitation. We will furthermore show how this dynamics can be probed with different pump-probe experiments including photo-emission spectroscopy, x-ray absorption spectroscopy and resonant inelastic x-ray scattering.

[1] PRL 128, 153001 (2022).
[2] PRA 108, 032816 (2023).
[3] Nat. Commun. 11, 4095 (2020).

MA 24.7 Wed 12:30 H36

Higher-Order Susceptibilities in Extended Kitaev Models Computed Via Krylov-Space Based Methods — •David Kaib, Marius Möller, and Roser Valenti — Institut für theoretische Physik, Goethe-Universität Frankfurt

Recently, it was proposed that techniques measuring higher-order dynamical response, such as two-dimensional coherent spectroscopy (2DCS), could provide more distinguishable signatures in analyzing the excitations of different systems. This is particularly true when linear response reveals only a featureless continuum, which could arise from various different types of excitations, or, for example, static disorder. The numerical evaluation of nonlinear response functions can, however, be computationally very demanding. Here, we propose an efficient Lanczos-based method that computes higher-order susceptibilities directly in the frequency domain. As an application case, we consider extended Kitaev models, that are relevant to α-$RuCl_3$ and related materials. We compare the nonlinear response from our method to the one obtained within linear spin-wave theory, showcasing that nonlinear response measurements can distinguish whether an observed excitation continuum is of conventional two-magnon type or has a different origin.