TT 62: Ultrafast Dynamics of Light-Driven Systems

Time: Friday 9:30–11:45 Location: HSZ 03

TT 62.1 Fri 9:30 HSZ 03

Ultrafast dynamics of spin-density wave order in $BaFe_2As_2$ under high pressures — Ivan Fotev[1,2], Stephan Winnerl[1], Saicharan Aswartham[3], Sabine Wurmehl[3,4], Bernd Büchner[3,4], Harald Schneider[1], Manfred Helm[1,2], and •Alexej Pashkin[1] — [1]Helmholtz-Zentrum Dresden-Rossendorf, Germany — [2]Technische Universität Dresden, Germany — [3]IFW Dresden, Germany — [4]Würzburg-Dresden Cluster of Excellence ct.qmat, Germany

We utilize pump-probe spectroscopy to measure the quasiparticle relaxation dynamics of $BaFe_2As_2$ in a diamond anvil cell at pressures up to 4.4 GPa and temperatures down to 8 K. Tracing the amplitude of the relaxation process results in an electronic phase diagram that illustrates the variation of the spin-density wave (SDW) order across the whole range of the applied pressures and temperatures. We observe a slowing down of the SDW relaxation dynamics in the vicinity of the phase transition boundary. However, its character depends on the trajectory in the phase diagram: the slowing down occurs gradually for the pressure-induced transition at low temperatures and abruptly for the thermally-driven transition. Our results suggest that the pressure-induced quantum phase transition in $BaFe_2As_2$ is related to the gradual worsening of the Fermi-surface nesting conditions.

TT 62.2 Fri 9:45 HSZ 03

Probing the superconducting transition in a cuprate superconductor by multi-dimensional terahertz spectroscopy — •Albert Liu[1], Danica Pavicevic[1], Michael Fechner[1], Pedro Lozano[2], Genda Gu[2], and Andrea Cavalleri[1,3] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — [2]Brookhaven National Laboratory, New York, USA — [3]Oxford University, Oxford, UK

We develop two-dimensional terahertz spectroscopy in both a noncollinear and reflection geometry. We demonstrate the technique by distilling the terahertz optical response of a superconducting plasma into its constituent nonlinearities, up to fifth-order. Subsequent measurements at temperatures approaching and crossing the phase transition reveal a phase-disordering transition in lieu of a BCS transition. An outlook for probing light-induced superconducting states will be provided.

TT 62.3 Fri 10:00 HSZ 03

Tracing the dynamics of the superconducting order via transient THG — •Minjae Kim[1,2,3], Gennady Logvenov[2], Bernhard Keimer[2], Dirk Manske[2], Lara Benfatto[4], and Stefan Kaiser[1,2,3] — [1]Institute of Solid State and Materials Physics, Technical University Dresden, 01062 Dresden, Germany — [2]Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany — [3]4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70569 Stuttgart, Germany — [4]Department of Physics and ISC-CNR, Sapienza University of Rome, 00185 Rome, Italy

Nonlinear THz third harmonic generation (THG) was shown to directly probe internal degrees of freedoms of the superconducting condensate and its exposure to external collective modes in the framework of driven Higgs modes [1-3]. Here we extend this idea to light-driven nonequilibrium states in superconducting La2-xSrxCuO4 establishing a transient Higgs spectroscopy. We perform an optical pump-THz-THG drive experiment and using 2D-spectroscopy we disentangle the driven TH response into the excited quasiparticles and condensates response. As such the light induced changes of the THG signals probe the ultrafast pair breaking dynamics and transient pairing amplitude of the condensate.

[1] Nat. Comm. 11, 1 (2020)
[2] Nat. Comm. 11, 287 (2020)
[3] arXiv:2109.09971

TT 62.4 Fri 10:15 HSZ 03

Influence of impurities on the Higgs mode in noncentrosymmetric superconductors — •Simon Klein and Dirk Manske — Max-Planck Institut für Festkörperforschung, Stuttgart, Deutschland

Recent interest for collective amplitude (Higgs) excitations focused on third harmonic generation (THG) experiments in single- and multiband superconductors, both for singlet *s*- and *d*-wave gap structure. A resonance in the THG intensity appears, when matching the driving frequency to the energy of the corresponding investigated mode, leading to a phase jump at the resonance frequency. We extend these studies to superconductors without an inversion symmetry, which can be effectively described by a two-band model with an order parameter, consisting of spin singlet (even parity) and spin triplet (odd parity) components. We calculate the THG response, showing that it contains contributions from three distinguishable sources, namely the Higgs mode, the Leggett mode and quasiparticles. In the clean limit the quasiparticle contributions dominate the collective modes for all singlet-triplet ratios of the gap structure. By including already a small amount of impurities in the system, we find a significant enhancement of the Higgs mode contributions to the THG signal. Furthermore, we notice a significant change in the phase jump, which helps to differentiate between clean and dirty superconductors.

15 min. break

TT 62.5 Fri 10:45 HSZ 03

Non-equilibrium phenomena in a two-band superconductor MgB_2 driven by narrow-band THz pulses — •Sergei Sobolev[1], Amon Lanz[1], Tao Dong[2], Alexej Pashkin[3], Amrit Pokharel[1], Zi Zhao Gan[2], Li Yu Shi[2], Nan Lin Wang[2], Stephan Winnerl[3], Manfred Helm[3], and Jure Demsar[1] — [1]Uni-Mainz, Mainz — [2]Peking Uni., Beijing — [3]HZDR, Dresden

Excitation of a superconductor (SC) with a low energy electromagnetic pulse leads to a non-equilibrium state, which may profoundly differ from a quasi-thermal one, driven by optical excitation. Such a non-equilibrium may be especially pronounced in a multi-band SC. Here, we report on systematic time-resolved studies of the dynamics of the SC order following resonant excitation with intense narrow-band THz pulses in thin films of MgB_2, the prototype two-band SC with two distinct superconducting gaps. We demonstrate that the temperature and excitation density dependent dynamics qualitatively follows the behavior predicted by the phenomenological Rothwarf-Taylor model for dynamics in a single gap BCS superconductor implying strong coupling between the two condensates on the ps timescale. Tracking the dependence of the amplitude of the THz driven gap suppression, however, displays a pronounced minimum near $0.6T_c$, that cannot be accounted by the phenomenological model. Comparison of the results to those obtained by excitation with NIR pulses suggests that resonant THz excitation results in long-lived (100 ps timescale) non-thermal quasiparticle distribution, which gives rise to Eliashberg-type enhancement of superconductivity, competing with pair-breaking.

TT 62.6 Fri 11:00 HSZ 03

Semi-classical analysis of high harmonic spectra in Dirac materials — •Wolfgang Hogger[1], Vanessa Junk[1], Alexander Riedel[1], Cosimo Gorini[2], Juan-Diego Urbina[1], and Klaus Richter[1] — [1]Institute for theoretical physics, University of Regensburg, Germany — [2]Université Paris-Saclay, CEA, CNRS, SPEC, 91191, Gif-sur-Yvette, France

The study of high-order harmonic generation (HHG) in solids by virtue of intense laser pulses provides a fascinating platform to study ultrafast electron dynamics as well as material properties. We theoretically investigate HHG on the basis of massive Dirac Fermions, serving as a prototypical model for topologically non-trivial matter. The high harmonic spectra resulting from a numerical solution of the equations of motion for the density-matrix are supplemented and compared to a semiclassical saddle-point analysis known as the Lewenstein or simpleman model[1,2]. We demonstrate that HHG can be interpreted as a result of interfering classical trajectories generated at different halfcycles of the laser pulse. A transparent and compact analytical formula for the high harmonic spectrum in the limit of long pulses is provided and discussed regarding the range of its validity as well as corrections arising from short waveforms.

[1] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A 49, 2117 (1994)
[2] G. Vampa, C. R. Mcdonald, G. Orlando, D. D. Klug, P. B. Corkum, and T. Brabec, Phys. Rev. Lett. 113, 073901 (2014)

TT 62.7 Fri 11:15 HSZ 03

Driven lattice fluctuations in quantum paraelectric $SrTiO_3$
— •Michael Fechner[1], Michael Först[1], Ankit Disa[1], Michele Buzzi[1], Alex von Hoegen[1], Gal Orenstein[2], Viktor Krapivin[2], Quỳnh Le Nguyen[2], Roman Mankowsky[3], Mathias Sander[3], Henrik Lemke[3], Yunpei Deng[3], Mariano Trigo[2], and Andrea Cavalleri[1] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — [2]Stanford Linear Accelerator Center, Menlo Park, United States of America — [3]Paul Scherrer Institute, Villigen, Switzerland

Enhanced fluctuations are the precursors of quantum phase transition. In the case of ferro- and antiferrodistortive structural transitions, these fluctuations arise at specific points in reciprocal space. Here we use time-resolved x-ray diffuse scattering to show how a resonantly driven zone center optical phonon in cubic $SrTiO_3$ selectively modulates lattice fluctuations at the Brillouin zone boundary. On short time scales, enhanced and oscillating lattice fluctuations are found at the R-point, which reduce to below the equilibrium value on the picosecond time scale. We attribute the fast dynamics to a finite-momentum fourth-order phonon-phonon interaction between the driven infrared-active Ti-O stretch mode and the antiferrodistortive zone-boundary soft mode. The long-term reduction indicates nonlinear coupling of the antiferrodistortive distortions to strain. We discuss the implications of these findings for the recently observed light-induced ferroelectric state of $SrTiO_3$[1].
[1] T.F. Nova et al., Science 364, 1075 (2019)

TT 62.8 Fri 11:30 HSZ 03

Decay of photoinduced orbital excitations in the spin-Peierls system TiOCl: a study from first principle
— •Paul Fadler[1], Philipp Hansmann[1], Kai Phillip Schmidt[1], Angela Montanaro[2], Daniele Fausti[1,2], and Martin Eckstein[3] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg — [2]University of Trieste — [3]Universität Hamburg

In spin-Peierls materials, the magnetic interactions caused by the electronic correlations drive a phase transition in which the unit cell is distorted. TiOCl is a quasi one-dimensional transition metal oxide with well-defined orbital (d-d) excitations that transitions to a spin-Peierls state below 67K, by doubling the unit cell along the 1-D chain. Recently, optical transmissivity measurements after a double pump excitation into the orbital excitations revealed a dependence of the decay time on the excitation sequence, which may indicate different lifetimes of various orbital excitations. To unveil the microscopic origins of this observation, we study a material-realistic multi-orbital Hubbard Hamiltonian derived from ab initio density-functional theory. The localized strong-coupling native of TiOCl then allows for perturbative elimination of charge-excitations, leading to a spin-orbital model of Kugel-Khomskii type. We compute its spin dispersion in the absence of orbital excitations. Based on this, and the computed intra and inter-orbital matrix elements, we then estimate the decay time of orbital excitations via (multi) spinon emission, and compare to the experimental findings.