Probing and manipulating magnetic and 2D quantum materials using ultrafast laser and high harmonic sources

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Status

The ability to probe the full dynamic response of quantum materials on the length- and time-scales (Å to attoseconds on up) fundamental to charge, spin and phonon interactions is leading to a host of new discoveries. The coupled interactions between charges, spins, orbital and lattice degrees of freedom are key to determining the state of a material, whether metallic, insulating, superconducting or magnetic. Under thermal equilibrium conditions these states can be tuned by varying the temperature, pressure, chemical doping or dimensionality. However, their inherent complexity and rich phase landscape make them challenging to understand or manipulate deterministically. Fortunately, ultrafast light sources have undergone remarkable advances in recent years, achieving what was merely a dream three decades ago, i.e., full coherent control of light fields spanning the THz to the X-ray regions. These new capabilities are providing powerful new tools for coherently manipulating and probing quantum materials using light.

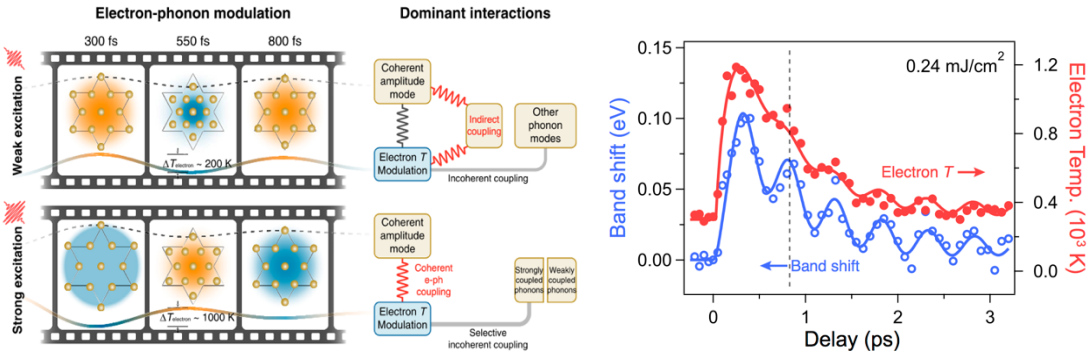

Figure 1. (We allow at most two figures that are roughly the size of this box. If the figure is reproduced or adapted from another non-IOP publication, you must seek permission for re-use from the publisher) **Coherently manipulating the charge and lattice orders and interactions using light.** After excitation by an ultrafast laser, the electron temperature in the CDW material 1*T*-$TaSe_2$ is modulated by the coherent phonon – by between 200 and 1000 K. In all past work, the electron bath cooled monotonically due to electron-phonon coupling. The dominant interactions also switch as the material enters a new 2D phase that cannot be reached via equilibrium routes.

Current and Future Challenges

Recent advances in high harmonic generation (HHG) have provided new techniques for uncovering new phases and couplings in 2D and magnetic materials, using femtosecond lasers to coherently manipulate and tune the properties and interactions in the material, and HHG as a probe to map out these new phases and couplings.[1-4] There are several beautiful recent observations of exciting new states in materials using light, but in some cases, the limited experimental data points made it challenging to validate an interpretation. The new ability of ultrafast HHG-based photoemission and magneto-optic spectroscopies to precisely probe multiple phases, electronic and magnetic order parameters, when combined with scanning the laser excitation fluence, is allowing us to map how material phases, magnetic states, and electron-phonon-spin couplings in these delicate materials.[5]

One aspect that facilitates the precise manipulation of materials using light is the large separation in timescales between the charge, spin and phonon dynamics. Much effort has been devoted to coherent control of molecular systems, and the prospects for similar control in quantum materials systems are just as exciting and appear even more achievable.

Advances in Science and Technology to Meet Challenges

High Harmonic Generation. The HHG process has made it possible to generate bright attosecond-to-femtosecond pulse trains or pulses, that can capture the fastest coupled charge, spin, phonon, and transport dynamics in materials and nano systems. Recent advances in HHG have made it possible to extend bright HHG from the extreme ultraviolet (EUV) region into the vacuum ultraviolet (VUV) and soft X-ray regions, with spectral, temporal and polarization properties that are ideal for a host of ultrafast spectroscopies.[6,7] It is now possible to generate bright HHG into the soft x-ray region of the spectrum at >kHz repetition rates required for advanced spectroscopies. It is also possible to create polarization and phase structured HHG beams (spin and orbital angular momentum) to enhance contrast, and to implement unique excitations and probes of chiral structures in magnetic materials and nanostructures.[8-10] There have also been significant advances in generating light in the VUV spectral region (~6–18 eV), a spectral region uniquely suited to probe chemical processes, as well as time- and spin-resolved angle-resolved photoemission spectroscopy (trARPES). It harnesses a new highly cascaded harmonic generation process where UV and VUV spectral lines are produced through upconversion of an infrared fiber laser such that each harmonic contributes to the cascaded formation of higher harmonics. A significant advantage of this process is that the peak intensity required for this process is in the range of 10^{12} W/cm^2 - substantially lower (x10-100) than that required for HHG, correspondingly lowering the pulse energy needed for VUV generation. This makes it possible to produce high flux, high repetition rate (100kHz to >MHz), a photon energy range from the UV to 18 eV, with variable energy (~10-40 meV) and time resolution (~50-100 fs).[6] These new HHG capabilities are ideal for a suite of ultrafast X-ray absorption spectroscopies, photoelectron spectroscopies and magneto-optic spectroscopies.

Ultrafast Electron Calorimetry. Key to using the capabilities of these new light sources to understand quantum materials is a new approach we call ultrafast electron calorimetry.[1-4] Using HHG-based trARPES, we can measure the full dynamic electronic band structure and electron temperature. Since the electrons thermalize very quickly and their heat capacity is small, heat transfer from the electrons is directly monitored. By varying the laser excitation fluence while monitoring this electron temperature at very short times, we can sensitively detect the changes in the microscopic interactions. When coupled with the ability of HHG trARPES to simultaneously capture the full dynamic band structure (which reflects the macroscopic order parameter), we can map and expand the phase diagram using light as a tuning parameter. In recent research, we used femtosecond laser pulses to coherently manipulate the electron and phonon distributions, and their couplings, in the quasi 2D charge density wave (CDW) material 1*T*-$TaSe_2$. We observed a large electron temperature modulation and a phase change of π when increasing laser fluence, suggesting a switching of the dominant coupling mechanism between the coherent phonon and electrons (Fig. 1). Moreover, it is also possible to dynamically tune the material phase all the way from the usual insulating CDW phase, through the normal undistorted metallic phase, to highly metallic CDW phase, using light.

Light-induced Spin Manipulation. For more than two decades, ultrafast lasers have been used to quench or switch the magnetic state of materials.[11,12] To date laser-induced manipulation of the magnetic state has almost exclusively been understood as a secondary process taking place on

~picosecond timescales. We use a suite of correlated high harmonic based spectroscopies: trARPES, combined with HHG-based element-specific magneto-optic probes, to detect the excitations present in the material, over a full range of laser fluences and depths. As illustrated in Fig. 2 (left), we demonstrated that a femtosecond light pulse can directly transfer spin polarization from one element to another in a half-metallic Heusler material, Co_2MnGe.[13,14] This spin transfer initiates as soon as light is incident on the material, showing that we can spatially transfer angular momentum between neighboring atomic sites on timescales <10 fs. In yet another surprising finding in Ni (Fig. 2, right), when the electron temperature is transiently driven above the Curie temperature, there is an extremely rapid change in the material response: the spin system absorbs sufficient energy within 20 fs to subsequently proceed through the phase transition, while demagnetization occur on much longer, fluence-independent, timescales of ~176 fs. These findings uncover a new timescale of ultrafast demagnetization and collectively show that previous theoretical understanding is incomplete. The addition of spin-resolved ARPES, and ultrafast soft X-ray and electron spectroscopies and imaging, can yield additional insight into ultrafast spin dynamics and transport.

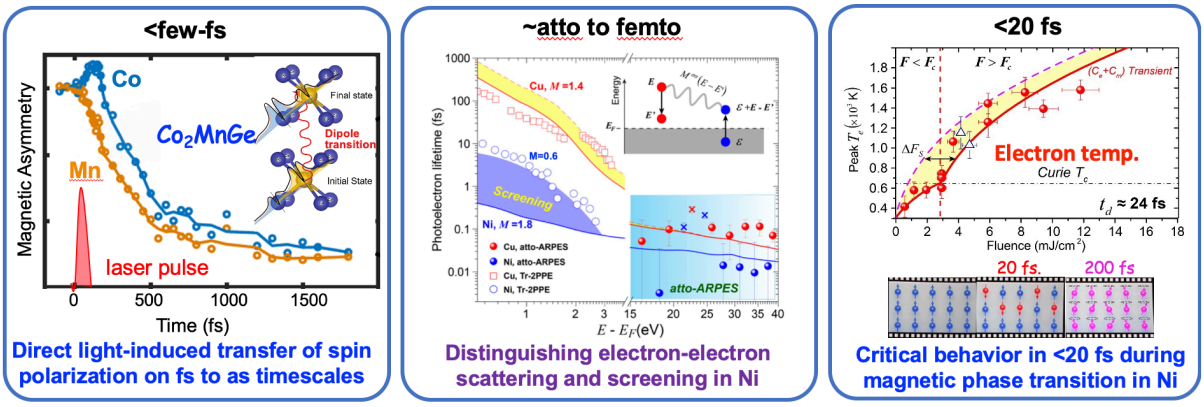

Figure 2. (We allow at most two figures that are roughly the size of this box. If the figure is reproduced or adapted from another non-IOP publication, you must seek permission for re-use from the publisher) **New timescales for spin manipulation observed experimentally.** (left) Light can transfer spin polarization between two elements in a Heusler alloy, on few-fs timescales. (centre) Distinguishing electron-electron scattering and screening in Ni. (right) Ultrafast electron calorimetry showed that the energy required to drive a true magnetic phase transition in Ni is absorbed by the spin system within ~20 fs, clearly observable as an inflection point in the measured electron temperature at the Curie temperature.

Concluding Remarks

Over the past 3 decades of studies of high harmonic generation, and 2 decades of developing new experimental methodologies using ultrashort pulse EUV light to study materials, we now have a set of tools with broad applicability, whose potential has to-date barely been tapped. As HHG sources become easier to use, they can proliferate to a larger variety of experimental venues, both to apply already-established techniques, and to develop new ones. The ability to coherently manipulate quantum materials, using HHG as a probe and expanding the use of a variety of complementary coherent techniques including Raman excitation, as well as directly driving quantum materials with mid-IR and THz radiation, are all exciting areas of future investigation. The study of high harmonic quantum light science, and the applications for time-resolved studies in material, have shown an extraordinary potential for further advances and impact on broad fields of quantum materials science.

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