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Broadband ultrafast terahertz spectroscopy in the 25 T Split Florida-Helix

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We describe the development of a broadband (0.3–10 THz) optical pump-terahertz probe spectrometer with an unprecedented combination of temporal resolution (≤200 fs) operating in external magnetic fields as high as 25 T using the new Split Florida-Helix magnet system. Using this new instrument, we measure the transient dynamics in a gallium arsenide four-quantum well sample after photoexcitation at 800 nm. *Published by AIP Publishing.* https://doi.org/10.1063/1.5023384

I. INTRODUCTION

Ultrafast spectroscopic techniques permit the study and control of the optical and electronic properties of materials on time scales faster than traditional electronic techniques can resolve. Currently available femtosecond lasers can produce ultrafast pulses over a wide range of the spectrum including ultraviolet,[1] optical,[2] infrared,[3] and/or terahertz.[4] In these experiments, a femtosecond pump pulse from an ultrafast laser perturbs the sample, while a time-delayed probe pulse studies the transient changes to the complex dielectric constants of materials. At low pump pulse energies, these experiments study the near-equilibrium properties of materials, while as the pulse energy is increased, these can be used to study strongly non-equilibrium dynamics,[5] metastable phases,[6] and photoinduced phase transitions.[7] Using ultrafast lasers to study and manipulate the optical properties of materials, for example, can trigger phase transitions to alternate orders that cannot be easily accessed using other experimental techniques, and thus, they can help elucidate the underlying competition between different degrees of freedom in a material.[5] Investigation of non-equilibrium dynamics and metastable phases generally requires control of these material properties on a pico- or femtosecond time scale. This enables the dynamic tuning of one degree of freedom (e.g., charge, lattice, orbital, and spin) on a time scale that is faster than it is coupled to the other degrees of freedom. This enables the system to move into an otherwise thermodynamically inaccessible metastable configuration.[7]

In this paper, we describe the development of our broadband optical pump-terahertz probe spectrometer that operates in *sustained* high magnetic fields. This uses the 25 Tesla Split Florida-Helix magnet at the National High Magnetic Field Lab, which has been designed with multiple optical axes for *free-space* optical experiments. We use this new spectrometer to study light-induced changes to the terahertz dielectric constants in a 18 nm semiconductor multiple quantum well (VA0719). An excitation pulse excites the 18 nm multiple quantum well sample using the output ($h\nu_1$ = 1.55 eV) of a mode-locked titanium:sapphire laser amplifier, while the probe measures the changes to the sample transmission using a time-delayed broadband terahertz pulse derived from the same laser amplifier. We observe a reduction in the transmitted terahertz bandwidth when compared to the generated pulse that is consistent with the reststrahlen bands of gallium arsenide and aluminum gallium arsenide in the quantum well. Finally, we discuss the broader applicability of our new instrument to a wider range of material systems, including the complex and competing dynamics between electronic, orbital, lattice, and spin degrees of freedom in many correlated electron systems.

II. ULTRAFAST SPECTROSCOPY IN CORRELATED SYSTEMS AND QUANTUM-CONFINED SEMICONDUCTORS

Spin has an important role in the electronic and magnetic phases of many materials including common semiconductors,[8–11] transition metal oxides (e.g., manganites,[12,13] cuprates,[14,15] nickelates,[16] iridates,[17,18] and vanadates[19]), transition metal dichalcogenides,[20] and pnictides.[21] External control of spin in these systems either requires the use of light[22] through spin-orbit coupling in the material or external magnetic fields, which split spin states into different energy levels.[13] An improved understanding of the underlying physics responsible for these metastable phases will be aided by a new generation of ultrafast experimental tools that can measure and control the electronic, lattice, orbital, and/or spin degrees of freedom.

The utility of our instrument is its ability to study the collective and single-particle excitations within a wide range of materials of contemporary interest. Figure 1 highlights some of these excitations, which are present in a wide variety of two-dimensional materials, correlated electron materials, and

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