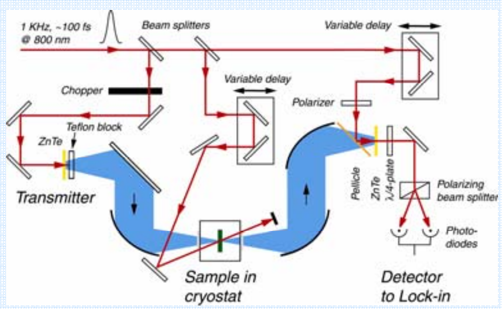
Ultrafast Dynamics in Complex Functional Nanomaterials

Introduction

Ultrafast optical spectroscopy has attained prominence in chemistry, biology, and condensed matter physics in recent years due to its ability to resolve dynamics in conventional metals and semiconductors at the fundamental timescales of electron and lattice motion. However, the application of ultrafast spectroscopy to materials of more contemporary interest is still in its infancy, especially in the mid-and-far-infrared (IR) spectral range. Therefore, the ability to probe the dynamics of these novel systems on an ultrafast time scale stands to reveal a great deal of information that will increase understanding of their basic physics and aid in their optimization for a variety of applications.

At CINT, we have several ultrafast laser systems that provide sub-100 femtosecond (fs) pulses from the visible to the far-infrared (see below). This enables experiments that can dynamically resolve elementary excitations in a wide range of nanomaterials (some examples are given in the panels on the right), a capability that will be critical in supporting users as well as in supporting original research by CINT scientists. Future research directions will include ultrafast measurements of isolated nanoparticles, resulting in a greater understanding of the influence of inhomogeneous broadening and the local environment on the dynamic response of a single nanoparticle.

Associated Capabilities



Laser systems at the CINT Core Facility:

- Spectra-Physics Spitfire Pro ultrafast amplifier, producing 35 fs, 1 mJ pulses at 800 nm and 1 kHz repetition rate
- Coherent RegA ultrafast amplifier system (including 9450 and 9850 optical parametric amplifiers), producing sub-100 fs pulses tunable from 400 nm-2.4 μm at 100 kHz
- KML Cascade cavity-dumped oscillator producing sub-20 fs, 35 nJ pulses at 800 nm and 2 MHz

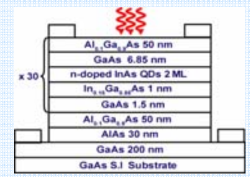
The flexibility and unique capabilities afforded by these laser systems will enable several unique experiments including:

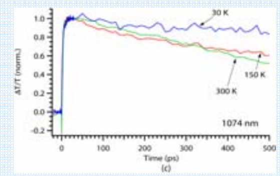
- Optical-pump THz-probe spectroscopy over broad bandwidths (~0.1-15 THz)
- High sensitivity (~10^{-6}) optical-pump optical-probe spectroscopy
- Widely tunable ultrafast spectroscopy featuring independently tunable pump and probe wavelengths between 400 nm-2.4 μm

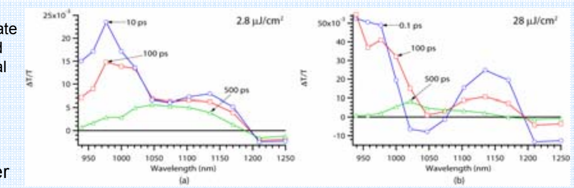
Science Examples

Ultrafast Carrier Dynamics in an InAs/GaAs Quantum Dots-in-a-Well Detector

- DWELL: Dots-in-a-well design offers several advantages over conventional quantum dot (QD) and quantum well (QW) detectors
  - Control of the operating wavelength by altering the thickness and composition of the QW
  - Recently incorporated into 640x512 focal plane arrays for thermal imaging applications
- Differential transmission spectroscopy (DTS) with near-IR probe wavelengths is used to time-resolve carrier relaxation into and out of energy levels within DWELL as a function of temperature and photoexcited carrier density
  - Density dependence: low fluence measurements indicate the possibility of a phonon bottleneck at the QD excited state, while higher fluence measurements reveal optical gain or induced absorption
  - Temperature dependent measurements of the signal near the DTS peak agrees well with typical DWELL operating temperatures
- Experiments provide insight into physics of carrier relaxation from three to two to zero dimensions



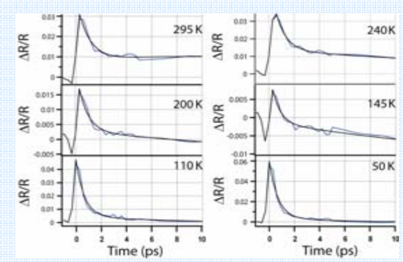


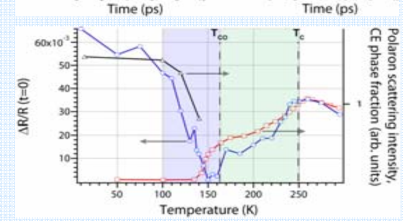


R. P. Prasankumar et al, *CLEO 2004, LEOS* 2005, submitted to *CLEO* 2007 (in collaboration with CINT user S. Krishna's group at UNM).

Nanoscale phase inhomogeneities in the manganite $Nd_{0.5}Sr_{0.5}MnO_3$ probed through the polaronic response

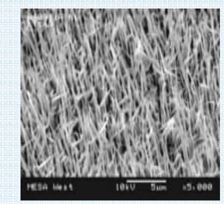
- Colossal magnetoresistance (CMR) manganites are of great fundamental and practical interest due to their unique properties
- Nanoscale phase inhomogeneities (e.g., correlated and uncorrelated polarons) strongly influence manganite physics
  - Uncorrelated and correlated Jahn-Teller polarons exist in the paramagnetic (PM) and ferromagnetic (FM) phases of various manganite systems
  - Correlated polarons are thought to govern the CMR effect
- Ultrafast optical-pump, mid-infrared-probe measurements used to investigate quasiparticle dynamics in $Nd_{0.5}Sr_{0.5}MnO_3$(NSMO)
  - NSMO: Ferromagnetic below T_C=250 K, charge-and-orbital ordered (COO) below T_{CO}=160 K
- Fast sub-picosecond relaxation corresponds to excitation and redressing of uncorrelated lattice polarons for $T>T_{CO}$
  - Close correspondence in temperature dependence of $\Delta R/R$ (t=0) and number of uncorrelated polarons as determined by x-ray scattering
  - Peak reflectivity change is a direct measure of CE phase fraction below T_{CO}
- First demonstration that ultrafast spectroscopy is sensitive to the presence of nanoscale phase inhomogeneities in complex oxides



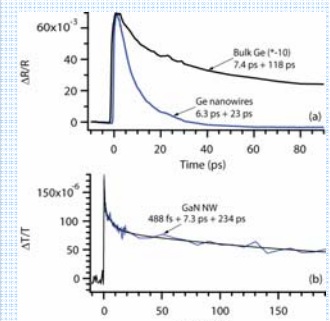


R. P. Prasankumar et al, to be submitted to *Phys. Rev. Lett*

Ultrafast Dynamics in Semiconductor Nanowires



- Semiconductor nanowires (NW) are a model system for exploring the effects of two-dimensional carrier confinement
- Single crystal Ge and GaN NWs grown by MOCVD through the vapor-liquid-solid mechanism
- First optical pump-probe measurements on semiconductor nanowires
  - Faster carrier relaxation in nanowires than in bulk material
  - Carrier trapping and recombination at surface states increases relaxation rates in nanowires
- Experiments demonstrate potential of ultrafast spectroscopy for probing carrier dynamics in semiconductor nanowires
  - Enables optimization of properties for potential applications in thermoelectrics and photonics



R. P. Prasankumar et al, submitted to *CLEO* 2007

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