Mid-infrared Ultrafast and Nonlinear Spectroscopy of Semiconductors

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This paper describes our recent experiments using 140 fs pulses of intense, coherent, and tunable mid-infrared (MIR) radiation produced by an optical parametric amplifier. These experiments explored MIR ultrafast and nonlinear optical phenomena in bulk semiconductors, InMnAs/GaSb ferromagnetic heterostructures, and single-wall carbon nanotubes.

1. Introduction

The advent of long-wavelength coherent sources, such as free-electron lasers, optical parametric amplifiers, and Terahertz antennas, has created a new class of opportunities to study small-energy phenomena in solids in the time domain and/or high-intensity regimes. Long-wavelength light can couple with a wide variety of electronic and vibrational excitations in solids. This paper reviews our recent progress in MIR ultrafast optics in bulk and quantum-confined semiconductors.

2. Experimental methods

2-1. Optical Parametric Amplifier System

Our MIR source was an optical parametric amplifier (OPA) pumped by a Ti:Sapphire-based chirped pulse amplifier (CPA) (Model CPA-2010, Clark-MXR, Inc.). The OPA produced tunable, intense radiation from 522 nm to 20 μm using different mixing crystals. The CPA produced pulses of near-infrared (NIR) radiation with pulse energy ~ 1 mJ and pulse duration ~ 140 fs at a repetition rate of 50 Hz – 1 kHz.

2-2. MIR-pump/NIR-probe Spectroscopy

Here we measured near-band-edge transmission spectra in the presence of intense MIR radiation. The broadband NIR probe was produced in a sapphire plate. The probe and pump were focused onto the sample using an off-axis paraboloid. After passing through the sample, the spectrum of the probe was dispersed using a grating monochromator and detected using a Si CCD camera. Spectra were obtained with the driving field at various time delays with respect to the probe, and were subsequently normalized to the probe spectrum without the sample, to obtain the absolute transmission.

2-3. Two-Color Time-Resolved Magneto-Optical Kerr Effect (MOKE) Spectroscopy

Here we used a small fraction (~ 10^{-5}) of the CPA beam as a probe and the intense OPA beam as the pump. The two beams were made collinear by a non-polarizing beam splitter and focused onto the sample inside a magnet. The reflected NIR probe entered a Wollaston prism which spatially separated the *s*- and *p*-components of the probe, which we focused on a balanced bridge. The signal, which was proportional to the induced MOKE angle change, was fed into a lock-in amplifier or a boxcar integrator.

3. Experimental Results

3-1. Dynamic Franz-Keldysh Effect in GaAs

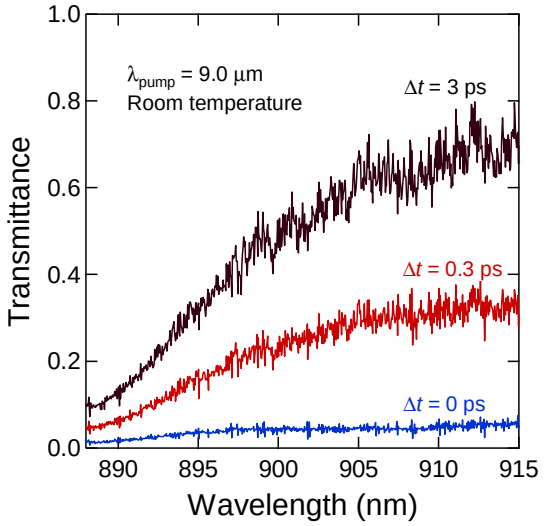

Fig. 1. Near-band-edge transmission spectra for GaAs under MIR excitation at three different time delays.

Intense AC fields can coherently and strongly modify the band structure of semiconductors. Intense long-wavelength laser fields are ideal for this purpose since they can minimize interband absorption and sample damage while maximizing the ponderomotive potential (quiver energy).[1)] Shown in Fig. 1 are near-band-edge transmission spectra for GaAs taken in the presence of a 9.0 μm MIR driving field with ~ 2×10^{10} W/cm^2 peak

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intensity. We observe a dramatic decrease in transmission below the band edge (885 nm). This decreased transmission, due to induced absorption, occurs only during the presence of the intense MIR pulse, demonstrating the virtual nature of the effect, i.e., no MIR excitation of carriers across the band gap and/or lattice-heating effects are involved.

3-2. Ultrafast Optical Manipulation of Ferromagnetic Order in InMnAs/GaSb

There is currently much interest in ultrafast spin dynamics in ferromagnets.[2] Ferromagnetic metals have been studied extensively using ultrafast magneto-optical spectroscopies. III-V ferromagnetic semiconductors can add new dimensions to this problem because of the *carrier-induced* nature of ferromagnetism in these systems. Here we studied a ferromagnetic InMnAs/GaSb heterostructure. We created transient carriers in the magnetic InMnAs layer using MIR pulses and probed the induced magnetization changes through the MOKE angle of NIR probe pulses. Our data showed that magnetic properties can be significantly modified by intense MIR pulses. Figure 2 shows two traces representing the pump-induced MOKE signal change versus time delay taken under the excitation of the MIR pump with two opposite senses of circular polarization, i.e., σ^+ and σ^-. Opposite polarizations result in *opposite signs* of the photo-induced MOKE change.

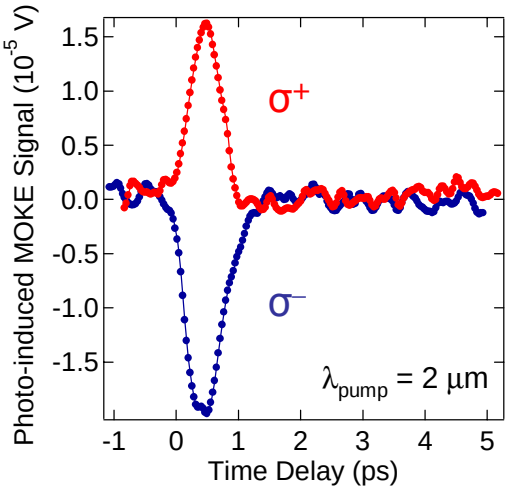

Fig. 2. Photo-induced MOKE signal at a temperature of 16 K versus time delay under pumping with circularly polarized MIR radiation.

3-3. Pump-Probe Spectroscopy of Carbon Nanotubes

Single-wall carbon nanotubes (SWCNTs) represent natural, ideal one-dimensional (1D) systems that can be used to explore a variety of novel 1D physics. 1D quantum confinement significantly increases the importance of Coulomb interactions between carriers, resulting in a collective state known as a Luttinger liquid as well as extremely large exciton binding energies. However, their optical properties have been poorly explored, due to the lack of methods for preparing pure samples consisting of SWCNTs with a single chirality. We performed pump-probe spectroscopy measurements on SWCNTs in a wide spectral range (visible to mid-infrared). In the samples we studied, nanotubes were suspended in micelles (and wrapped in polyvinyl pyrrolidone in some cases) to minimize tube-tube interactions.[3] Our data showed fast relaxation of photo-induced carriers, whose dynamics were strongly wavelength-dependent, as shown in Fig. 3.

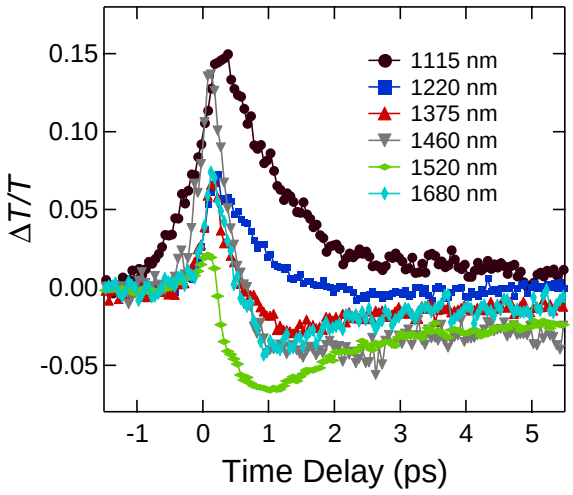

Fig. 3. Degenerate pump-probe traces for single-wall carbon nanotubes taken at six different wavelengths.

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