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Ultrafast and nonlinear spectroscopy of semiconductors with small energy photons

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Abstract

Ultrashort pulses of intense, coherent, and tunable far-infrared and mid-infrared radiation, achievable with free-electron lasers and optical parametric amplifiers, provide unique opportunities to investigate low energy dynamics in solids. This paper reviews our recent experiments on bulk and quantum-confined semiconductors using such radiation. These experiments include: far-infrared spectroscopy of transient plasmas, picosecond time-resolved cyclotron resonance, terahertz sideband generation, and the dynamic Franz–Keldysh effect.

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Keywords: Infrared; Free electron lasers; Semiconductors; Ultrafast and nonlinear optics

1. Introduction

The advent of long-wavelength coherent sources such as free electron lasers has created a new class of opportunities to study small-energy phenomena in solids. This paper describes some of the highlights of our recent experiments [1–7].

2. Far-infrared dynamics of transient plasmas

Here we studied the far-infrared (FIR) properties of photo-generated plasmas [1]. We simultaneously monitored the time evolution of the transmission and reflection of a FIR probe pulse after near-infrared (NIR) excitation. This allowed us to carry out a *dynamical* study of the Drude conductivity of photocreated transient plasmas. More specifically, we were able to directly determine the density and scattering lifetime of the carriers as functions of time, i.e. $n(t)$ and $\tau(t)$. In Fig. 1, we present our experimental data (a) along with fits (b) obtained using our theory.

3. Time-resolved cyclotron resonance

Here we studied the evolution of the effective mass, density, and scattering time of transient carriers in a magnetic field [2,3]. Typical data for InSb/InAlSb quantum wells are shown in Fig. 2(a). At 25 ps, the cyclotron resonance line is significantly broadened to higher B. As time progresses, the electrons relax towards the band

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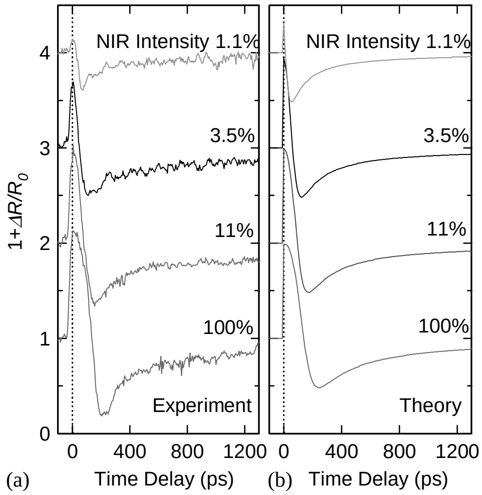

Fig. 1. Experimental (a) and calculated (b) photo-induced reflectivity of InSb as a function of time delay for selected NIR pump intensities.

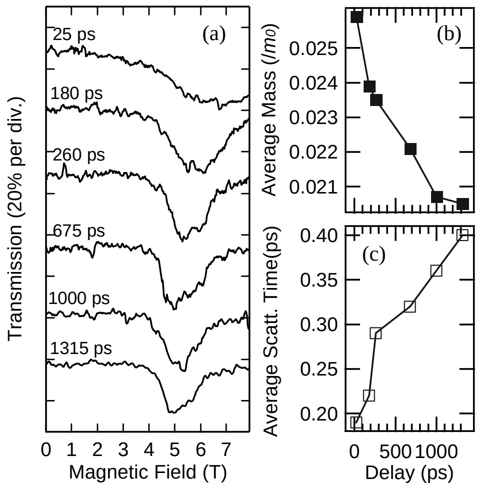

Fig. 2. Picosecond time-resolved CR in InSb quantum wells. (a) Transmission change of the FIR probe as a function of magnetic field at different delays. (b) Average cyclotron mass vs. time delay. (c) Average scattering time vs. time delay.

edge, resulting in a lighter mass at 1.3 ns. The average mass and scattering time are plotted vs. delay in Figs. 2(b) and (c), respectively. We performed a simulation of the data, which allowed us to determine the time evolution of the density and temperature of the transient Fermi gas.

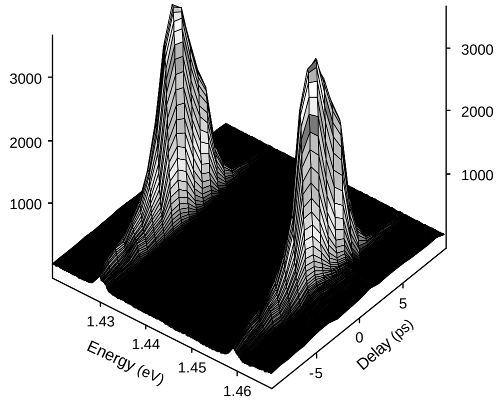

Fig. 3. Sideband intensity as a function of photon energy and relative delay between the NIR and THz pulses. This represents a convenient method for characterizing THz pulses using a Si photodetector.

4. Terahertz sideband generation

Here we demonstrate THz sideband generation, in which a weak beam passing through a THz-driven semiconductor acquires new frequencies separated by integer multiples of the THz frequency [4]. We were able to probe the evolution of the THz sidebands in the time domain, as shown in Fig. 3, where we show both the ω_{-1} and ω_{-2} sidebands. We observe that the sidebands appear only when the NIR and THz pulses temporally overlap.

5. Extreme mid-infrared nonlinear optics

Here we explored novel nonlinear phenomena using intense mid-infrared (MIR) radiation from an optical parametric amplifier [5–7]. Shown in Fig. 4 is transmission data taken using a 3.5 μm MIR driving field with $\sim 2\times10^{10}\,\mathrm{W/cm^2}$ peak intensity. We observe a dramatic *decrease* in transmission that extends past 0.2 eV below the band edge of the GaAs sample. This decreased transmission, due to induced absorption, occurs only during the presence of the intense MIR pulse. Existing models predict a laser-induced absorption with an absorption coefficient that is linear with the ponderomotive potential and with an extent that is on the order of the ponderomotive potential

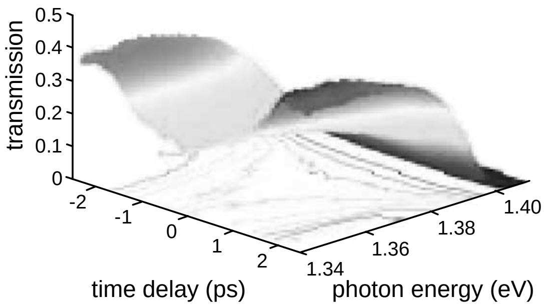

Fig. 4. Transmission of a NIR broadband probe pulse through GaAs as a function of time delay between the arrival of the intense 3.5 μm MIR driving pulse and the NIR probe pulse.

below the band edge. Both of these predictions are consistent with our data.

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