

Picosecond time-resolved cyclotron resonance in semiconductors

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A promising method for studying intraband carrier dynamics in semiconductors is monitoring the evolution of far-infrared (FIR) absorption induced by photoexcited carriers. By monitoring the photoinduced FIR absorption as a function of magnetic field, we performed time-resolved cyclotron resonance of photocreated electrons in InSb with picosecond resolution. © 1999 American Institute of Physics. [S0003-6951(99)04234-5]

Understanding the nonequilibrium carrier dynamics in semiconductors is of great technological and scientific importance. One means of studying such dynamics is by using intense ultrashort pulses of light with photon energies greater than the band gap to create nonequilibrium electron-hole pairs. The nonequilibrium carriers with excess energies relax toward the band edge through various scattering processes before eventually recombining to cause luminescence.¹ It immediately follows that interband photoluminescence spectroscopy is unable to provide direct information on intraband carrier relaxation dynamics; it is only sensitive to radiative (or “bright”) electron-hole pairs with center-of-mass momentum $\mathbf{K}=\mathbf{k}_{\text{photon}}\approx 0$, with the obvious exception of phonon-assisted processes where $\mathbf{K}=\mathbf{k}_{\text{photon}}+\mathbf{k}_{\text{phonon}}$.^{2,3} On the other hand, intraband far-infrared (FIR) spectroscopy obeys its own set of selection rules, independent of whether or not the states involved are interband active, thus providing a rare opportunity to directly probe these “dark” states. A wide variety of vertical ($\Delta\mathbf{K}\approx 0$) intraband transitions are allowed, e.g., inter-Landau level transitions (cyclotron resonance). Since cyclotron resonance (CR) is a powerful FIR spectroscopic method for investigating the dynamical conductivity of carriers in solids,⁴⁻⁶ performing *time-resolved* CR (TRCR) should provide very useful information about the relaxation dynamics of semiconductors in the presence of a magnetic field.

In this letter, we report the use of picosecond TRCR of photocreated nonequilibrium carriers in InSb using two-color [near-infrared (NIR) and FIR] laser spectroscopy. The NIR pulses are used to create a nonequilibrium distribution of carriers in a semiconductor sample. The subsequent dynamics are studied by monitoring the CR by using FIR absorption as a function of time delay between the NIR and FIR pulses. InSb was chosen as the test sample for the picosecond TRCR technique for the following reasons: (1) InSb has very small carrier effective masses which allows us to perform electron TRCR measurements in the magnetic field and FIR wavelength ranges available to us, and (2) the Landau level structure of InSb has been well studied.⁶ The TRCR experiment allows us to directly monitor the time evolution

of the effective mass, carrier density, and scattering time of the photocreated nonequilibrium carriers in the presence of a magnetic field.

The experimental setup for the two-color TRCR spectroscopy is illustrated schematically in Fig. 1. We use a femtosecond mode-locked NIR Ti:sapphire laser synchronized (to within a few ps)⁷ with the Stanford Picosecond free electron laser (FEL) as a source of synchronized NIR and FIR laser pulses. The NIR ($\lambda\sim 800$ nm) laser source is a Spectra Physics Tsunami Ti:sapphire laser seeding a Positive Light Spitfire regenerative amplifier, which generates ~ 200 fs pulses with pulse energies as high as 1 mJ. The FIR source is the Stanford picosecond FEL, which produces pulses with wavelengths extending through the midinfrared (3–15 μm) and FIR (15–60 μm). These pulses have durations ranging from 600 fs to 2 ps, and energies as high as ~ 1 μJ . The NIR beam is directed through a delay stage to allow fine adjustment of the time delay between the NIR and FIR pulses. The NIR beam is then spatially overlapped and made collinear with the FIR beam using a Si Brewster plate before both beams are focused onto the sample using a parabolic mirror. Any FIR photoinduced absorption in the Si Brewster plate was made negligible by enlarging the incident NIR spot size incident on the Si plate. The InSb sample studied is undoped, with a residual electron density of $\sim 5\times 10^{14}$ cm^{-3} and a 77 K electron mobility of $\sim 2\times 10^5$ $\text{cm}^2/\text{V s}$. The sample is wedged $\sim 3^\circ$ (to avoid multiple-reflection interference ef-

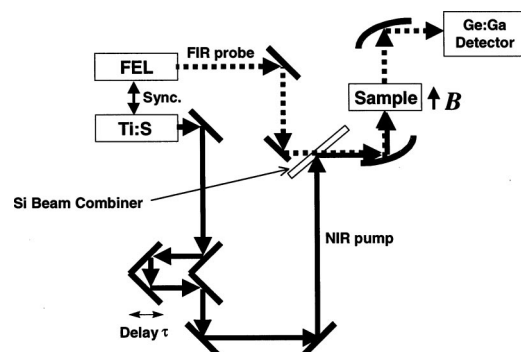


FIG. 1. Schematic diagram of the experimental setup for picosecond time-resolved photoinduced FIR absorption and cyclotron resonance.

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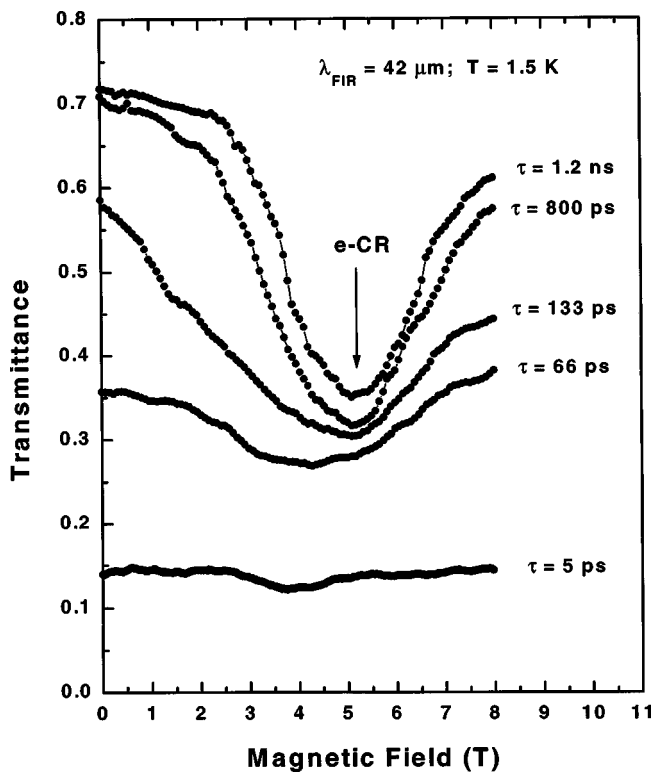
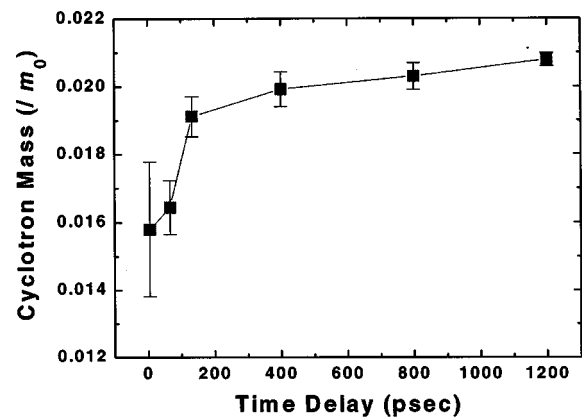


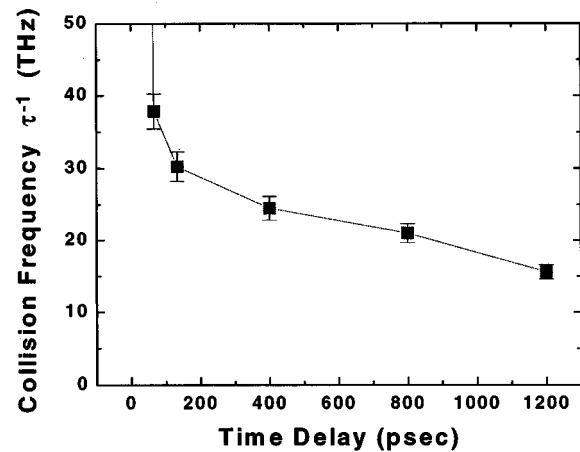
FIG. 2. Picosecond time-resolved CR of photoexcited nonequilibrium electrons in InSb. The magnetic field is scanned at fixed time delays.

fects) and polished to $\sim 150 \mu\text{m}$. The sample is placed in an Oxford Instruments Spectromag 4000 superconducting magnet with sapphire cold windows and polypropylene room temperature windows. The magnet produces a uniform magnetic field of up to 9 T parallel to the optical path. The transmitted FIR pulse is then recollimated and directed to a Ge:Ga photoconductive detector. The FIR output of the FEL consists of 10 Hz macropulses (up to 5 ms in duration). These macropulses each contain many ~ 1 ps duration micropulses separated by 84.6 ns. The Ti:sapphire laser is synchronized with the (10 Hz) macropulse output of the FEL, allowing only one NIR pulse for every FIR macropulse ($\sim 60\,000$ micropulses). This arrangement allows us to compare the intensity of transmitted FIR probe pulse before and after the NIR pump pulse. Micropulse to micropulse energy fluctuations are factored out using a FIR reference detector before the sample (not shown in Fig. 1). The experiments are performed by either keeping the magnetic field fixed and monitoring the photoinduced absorption (PIA) of the FIR pulse as a function of time delay, or the time delay remains fixed and the PIA is monitored as a function of magnetic field. Using a combination of the optical delay stage and electronic delays in the synchronization, we are able to selectively attain delays from 0 to 84.6 ns with a resolution of a few picoseconds (limited by timing jitter).

Figure 2 shows TRCR data obtained at a variety of fixed time delays. Figure 3 shows the effective mass of the electron [Fig. 3(a)] and the scattering time [Fig. 3(b)] as a function of time delay extracted from the peak position and the width of the cyclotron resonance peak (Fig. 2), using a simple Lorentzian fit to the peak. This is the first observation of picosecond TRCR (only microsecond time resolution has been achieved to date⁴), and it displays two interesting fea-



(a)



(b)

FIG. 3. (a) Electron mass and (b) collision time extracted from the cyclotron resonance peak position and width as a function of time delay. The lines are guides to the eye.

tures. First, there is no well-defined resonance at the smallest measured time scale. In this time regime (a few picoseconds after the creation of a high density of nonequilibrium free carriers), the carrier scattering rate is high enough to broaden the CR peak until it is completely obscured (i.e., the mean time between collisions τ_c is smaller than $1/\omega_c$). The observed TRCR linewidths are much larger than the transform limited spectral width of the picosecond FIR pulse, which, at 3.6 meV, corresponds to a CR linewidth of only ~ 0.6 T in InSb. Second, the CR peak position moves toward higher magnetic fields as a function of time delay. This is indicative of an electron effective mass that *increases* as a function of time, which is opposite to the behavior expected due to the strong nonparabolicity of the InSb conduction band. The effective masses of carriers in the InSb conduction band increase with increasing energy. With 800 nm (laser) excitation, the initially excited carriers are well above the band edge. Therefore, one would expect that as the carriers relaxed toward the band edge, their effective mass should decrease, thereby shifting the CR peak toward lower magnetic fields. The behavior displayed in the data is clearly opposed to these assertions, and requires further explanation.

We believe that the electron effective mass behavior may be explained by the carrier-carrier interaction which leads to band gap renormalization. Although Kohn's

theorem⁸ states that the carrier interactions should have no effect on the resonance peak in equilibrium CR, we have created a highly nonequilibrium state for which this may not apply. More specifically, Kohn's theorem breaks down when two species of carriers with different masses (such as electrons and holes) are present.⁹ It is well known¹⁰ that, when carrier densities are high, a significant decrease of the gap energy from its equilibrium value occurs as a result of correlation and exchange forces between carriers. This decrease in gap energy yields decreased electron effective masses, due to the increased $\mathbf{k}\cdot\mathbf{p}$ interaction between the conduction and light hole valence band. Therefore, the electron effective mass *increases* with time as the carrier density decreases and the gap energy approaches its equilibrium value. Using the dependence of the band gap with carrier density in conjunction with a simple two-band model,^{6,11} we can calculate the expected change in E_g and the resultant change in electron Landau level energies. The initial carrier density is estimated to be $10^{19}/\text{cm}^3$ (using the incident NIR intensity and the optical absorption depth). This density corresponds to a 25 meV (11% change) decrease in the gap energy, resulting in a 17% decrease in the band edge effective mass, which qualitatively agrees with the data. Taking into account other many-body effects such as dynamic screening and phase-space filling may provide quantitative agreement with the data.

In summary, we performed picosecond two-color (NIR and FIR) time-resolved cyclotron resonance spectroscopy on InSb. We observed a shift toward higher magnetic field in the time evolution of the CR peak, implying an increase in effective mass with decreasing carrier energy. Although this behavior is the opposite of what is anticipated due to the well-studied nonparabolicity of the InSb conduction band, it may be explained by band gap renormalization at high carrier concentrations. Also, we observed a dramatic broadening of the CR line at the smallest measured time scales. This behavior may be explained by a dramatically reduced mean time between collisions, resulting in an unobservably broad CR line. The data represent the time evolution of the electron

CR spectrum over time scales much shorter than the interband electron-hole recombination time (the time scale available through PL studies). The results demonstrate that this new type of spectroscopy is a promising tool for the study of intraband carrier dynamics.

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