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Picosecond time-resolved cyclotron resonance of non-equilibrium carriers in semiconductors

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Abstract

We have investigated the intraband dynamics of non-equilibrium carriers in semiconductors, using a picosecond far-infrared (FIR) free-electron laser synchronized with a femtosecond near-infrared (NIR) Ti : sapphire laser. An intense NIR pulse creates carriers, which interact with a delayed FIR pulse. We detect time-resolved cyclotron resonance by monitoring the intensity of the transmitted and reflected FIR pulse as functions of magnetic field and time delay between the NIR and FIR pulses. Results reveal a variety of intraband dynamical phenomena over time scales much shorter than the interband lifetime.

Keywords: Cyclotron resonance; Carrier relaxation; Terahertz spectroscopy

Ultrashort pulses of far-infrared (FIR) radiation allow the exploration of low-energy dynamical phenomena in semiconductors that are *inaccessible* to usual interband optical spectroscopy. This is because *intraband* FIR spectroscopy is independent of whether the states involved are *interband*-active, thus providing a rare opportunity to directly access dark states. Probing and/or strongly driving these dark states should provide valuable new insight into the complicated dynamics of interacting non-equilibrium carriers. However, because of the rarity of ultrashort FIR sources, research in this direction has not been actively pursued.

The Stanford free-electron laser (FEL) [1] produces intense tunable picosecond pulses of FIR radiation. Utilizing this unconventional laser, we have been investigating dynamical phenomena in various intraband transitions [2,3]. The present work is an extension of our earlier work [3], and here we describe results on the picosecond time-resolved cyclotron resonance (TRCR) of photogenerated carriers in an InSb sample with a higher mobility than the previously studied sample. Furthermore, we monitored both FIR transmission and reflection simultaneously.

Our experimental apparatus utilizes a unique combination of two synchronized short-pulse lasers – a femtosecond near-infrared (NIR) Ti : sapphire laser and the picosecond FIR FEL. Intense NIR pulses create non-equilibrium carriers in the sample, which then interact with FIR pulses. More details about the setup can be found in Refs. [2,3]. The ~ 150 μm thick InSb sample was undoped and had an electron density of 8.0×10^{13} cm^{-3} and a mobility of 8.3×10^{5} cm^{2} V^{-1} s^{-1} at 78 K. It was wedged 3° to minimize interference effects. The

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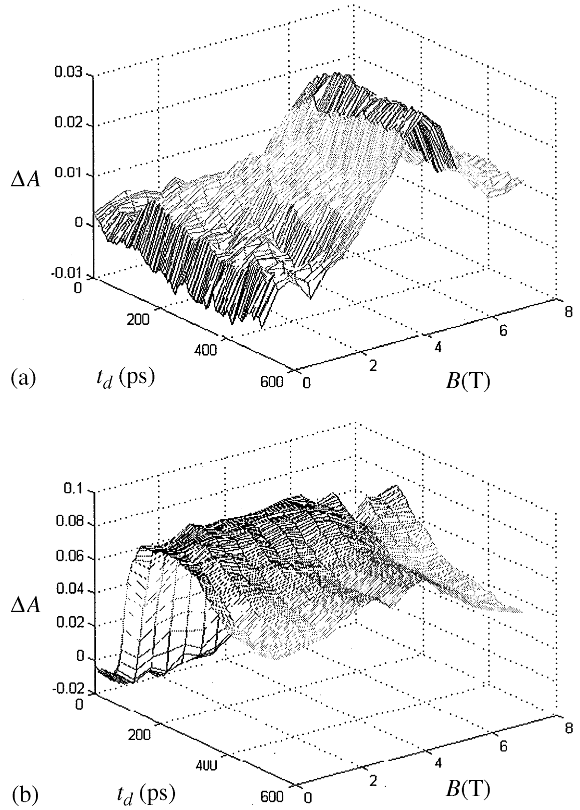

Fig. 1. Picosecond TRCR spectra of photo-created electrons in InSb. The photo-induced absorption change is plotted as a function of time delay and magnetic field for (a) low ($\sim$ 15 μJ/cm^2) and (b) high ($\sim$4 mJ/cm^2) excitations. The photo-induced absorption, ΔA, is defined as $(1 - R - T)/(1 - R) - (1 - R_0 - T_0)/(1 - R_0)$, where R and T are the reflectance and transmittance, respectively, and $R_0 = 18\%$ and $T_0 = 9\%$ are the values before the arrival of the NIR pump pulse. In both cases, the wavelength of the FIR radiation was 42 μm and the sample temperature was 1.5 K.

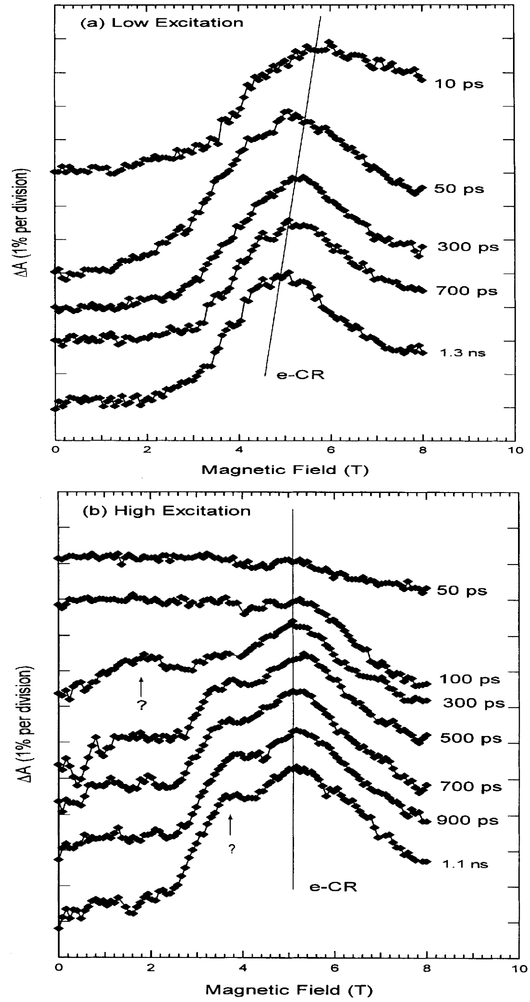

Fig. 2. Magnetic field scans at fixed time delays for (a) low ($\sim$15 μJ/cm^2) and (b) high ($\sim$4 mJ/cm^2) NIR excitations.

sample was tilted 45° with respect to the magnetic field (B) which was parallel to the p polarized FIR beam.

Fig. 1 shows a typical example of TRCR data. Here, absorption of the FIR beam is plotted as a function of time delay (t_d) and B. We can concentrate on particular aspects of the data by looking at either slices along t_d or B. Typical B-scan data for low and high excitations are shown in Fig. 2. These traces demonstrate directly the evolution of the cyclotron resonance (CR) spectrum over time scales much shorter than the interband lifetime. All of the effects seen here are therefore the result of intraband processes. The low-excitation data in Fig. 2(a) shows a smooth evolution of the electron CR spectrum from 10 ps to 1.3 ns. At 10 ps after excitation, the CR line is broadened to higher B, i.e., to heavier effective mass. This results from the strong non-parabolicity of the InSb conduction band (see, e.g., Ref. [4]). To elaborate, the initially excited carriers, having a Boltzmann-like distribution with a high carrier temperature, populate some states with high Landau indices, where the cyclotron masses are higher than that of the lowest electron Landau level. As time progresses, the electrons relax towards the lowest Landau level, resulting in a more or less symmetric line shape at 1.3 ns. The high-excitation case [Fig. 2(b)] differs significantly from the low-excitation case. First, note the dramatically broadened CR at a time delay of 50 ps, similar to our earlier observation [3]. In addition, however, owing to the higher mobility of the current sample, a new feature around 3.5 T with a mass of $\sim 0.014m_0$, is clearly resolved. This feature could be attributed to the light-hole CR or an electron–hole combined plasma resonance, but more measurements, especially with circularly polarized FIR radiation, are necessary to elucidate its origin.

Fig. 3 shows the reflectivity versus t_d at fixed B. The data exhibits qualitatively different behavior under different excitation conditions. First, all the high-excitation traces [Fig. 3(b)] show significantly enhanced reflectivity immediately after excitation. This is because the plasma frequency (ω_p) initially *exceeds* the photon energy of the FIR probe (ω_{FIR}) (29.5 meV). The estimated initial density in the high-excitation case is $\sim 10^{19}\ \mathrm{cm}^{-3}$, which corresponds to $\omega_p \sim 200$ meV. The low-excitation data at 0 T, on the other hand, shows a reflectivity *drop*. This completely opposite behavior is due to the fact that, when $\omega_p < \omega_{FIR}$, an increased density leads to a decreased refractive index, or equivalently, a decreased reflectivity. This behavior very quickly disappears as we increase B from 0 to 1.5 T. A small peak appears, which grows in intensity with B, reaches a maximum at ~ 3 T, stays roughly constant up to ~ 5 T, and finally goes away at higher B. We interpret this intriguing behavior as the interplay of the cyclotron frequency and ω_p. That is to say, as we increase B, the plasma edge moves up in frequency (see, e.g., Ref. [5]), eventually exceeding ω_{FIR}. More detailed simulations are in progress and will be discussed elsewhere [6].

In summary, we performed picosecond two-color (NIR and FIR) TRCR spectroscopy on InSb. We simultaneously monitored the dynamics of FIR transmission and reflection while we varied the magnetic field and the time delay between the NIR and FIR pulses. This has resulted in considerable clarification of our previous results [3]. Also, use of a high-mobility sample led to the observation of a new resonance feature in B-scans, which appeared only when the NIR pump fluence was sufficiently high. Finally, we found that the reflectivity dynamics are drastically affected by B. All these results demonstrate the power of this new FIR technique for investigating the dynamics of non-equilibrium carriers in semiconductors at very low energy scales.

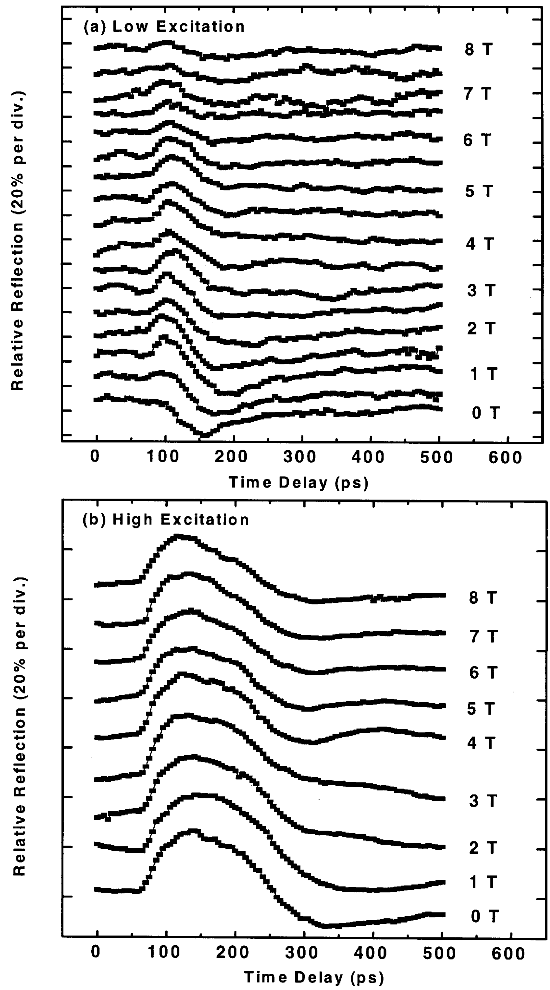

Fig. 3. Time-delay scans at fixed magnetic fields for (a) low ($\sim 15\ \mu\mathrm{J/cm^2}$) and (b) high ($\sim 4\ \mathrm{mJ/cm^2}$) NIR excitations.

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