

# Application of terahertz quantum-cascade lasers to semiconductor cyclotron resonance

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Quantum-cascade lasers operating at 4.7, 3.5, and 2.3 THz have been used to achieve cyclotron resonance in InAs and InSb quantum wells from liquid-helium temperatures to room temperature. This represents one of the first spectroscopic applications of terahertz quantum-cascade lasers. Results show that these compact lasers are convenient and reliable sources with adequate power and stability for this type of far-infrared magneto-optical study of solids. Their compactness promises interesting future applications in solid-state spectroscopy. © 2004 Optical Society of America

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Recently, quantum-cascade lasers (QCLs) have been successfully operated in the terahertz (THz, or  $10^{12}\text{-s}^{-1}$ ) range.<sup>1–3</sup> This impressive technological development contributes to closing the technology gap at 0.1–10 THz, where convenient solid-state devices do not exist.<sup>4</sup> A variety of sensing and imaging applications for such THz sources was described in Ref. 5. Here we report what is to our knowledge the first application of THz QCLs to semiconductor cyclotron resonance (CR), one of a number of THz excitations in solids.

Mid-infrared QCLs have achieved sensitivities of  $10^{-4}$  to  $10^{-6}$  in chemical-sensing applications.<sup>6,7</sup> Such applications require tunability, high power, and single-mode operation. In THz solid-state spectroscopy applications, the requirement for small laser linewidths is less stringent because resonances in solids have much broader linewidths. Performing wavelength-scanned spectroscopy would require prohibitively large tunability. One can circumvent this problem, however, by tuning another experimental parameter instead, such as electric field, magnetic field (as described here), pressure, or temperature. We have found that the THz QCLs are easy to operate, reliable in terms of intensity and wavelength stability, and sufficiently

powerful for easily performing linear THz absorption measurements. Furthermore, their compactness (as compared with that of other THz devices such as Fourier-transform infrared spectrometers, free-electron lasers, CO<sub>2</sub>-laser-pumped molecular-gas lasers, and laser-based difference-frequency generators or optical parametric oscillators) permits the entire experimental setup to occupy a small volume.

CR<sup>8</sup> is a convenient tool with which to measure band parameters in semiconductors such as effective masses. We detect the resonance by applying a magnetic field and measuring the transmission of light through the sample while we vary the wavelength of the incident probe light or the magnetic field. The resonance energy is proportional to applied magnetic field  $B$  according to the formula  $\omega_c = eB/m^*$ , where  $\omega_c$  is the cyclotron frequency,  $e$  is the electronic charge, and  $m^*$  is the effective mass of the charge carriers.

In the present experiments the light sources were GaAs/AlGaAs QCLs operating at 4.7 THz (64  $\mu\text{m}$ ), 3.5 THz (86  $\mu\text{m}$ ), and 2.3 THz (127  $\mu\text{m}$ ) with a maximum cw power of  $\sim 4$  mW.<sup>2</sup> The lasers were operated at 135 Hz with a duty cycle of 25%. The light was collimated and focused onto the sample by parabolic mirrors. The sample was placed in a superconducting

magnet with cold and room-temperature  $z$ -cut quartz windows ( $f/2.4$ ), and the transmitted light was collected with a parabolic mirror and detected with a liquid-helium-cooled silicon bolometer. The entire beam path was purged with dry nitrogen. The short-term wavelength drift was  $\sim 20$  MHz over 30 s, measured by beating with a THz gas laser.<sup>9</sup> Any long-term drift was unnoticeably small during the CR measurement. The short-term intensity fluctuations were  $\sim 0.5\%$  over 1 s; long-term drift was dominated by humidity fluctuations in the beam path.

Two samples were measured: (1) 20 periods of InAs/AlSb quantum wells, with a total electron density of  $1.2 \times 10^{12} \text{ cm}^{-2}$  and a mobility of  $120,000 \text{ cm}^2/\text{Vs}$ , and (2) a single 30-nm InSb/Al<sub>0.09</sub>In<sub>0.91</sub>Sb quantum well with an electron density of  $2 \times 10^{11} \text{ cm}^{-2}$  and a mobility of  $100,000 \text{ cm}^2/\text{Vs}$ .

Figure 1 shows the transmission of 4.7-THz radiation as a function of magnetic field for InAs quantum wells from 60 to 300 K. The photon-frequency dependence of the resonance field (at 1.5 K) and the temperature dependence of the cyclotron mass (at 4.7 THz) are shown in Figs. 2(a) and 2(b), respectively. The solid line in Fig. 2(a) has a slope of 1.51 (T/THz), corresponding to an effective mass of  $0.042 m_0$ , where  $m_0$  is the free-electron mass in vacuum ( $=9.1 \times 10^{-31} \text{ kg}$ ). Landau level calculations based on an 8-band  $\mathbf{k} \cdot \mathbf{p}$  model, combined with the measured electron density, identify the observed CR at 4.7 THz as predominantly the  $(2, \uparrow) \rightarrow (3, \uparrow)$  transition, where the numbers are Landau indices and  $\uparrow$  or  $\downarrow$  specifies the spin orientation.

As shown in Fig. 2(b), the cyclotron mass increases with increasing temperature. This is the opposite of the expected behavior: As the bandgap decreases with increasing temperature, the effective mass should decrease owing to the increased coupling between the conduction and the valence bands. This behavior cannot be attributed to a change in the QCL wavelength caused by the fringe magnetic field, which is estimated to blueshift the QCL frequency by less than 1 GHz.<sup>10</sup> Two calculated curves are shown in Fig. 2(b) to highlight this unexpected behavior. These curves correspond to the  $(2, \uparrow) \rightarrow (3, \uparrow)$  and the  $(2, \downarrow) \rightarrow (3, \downarrow)$  transitions, calculated with a modified Pidgeon-Brown model<sup>11</sup> including strain, quantum confinement, and the temperature dependence of the bandgap. The CR line is fairly broad at high temperatures, and it is likely that higher-level CR transitions (e.g., 3–4) are involved, contributing to the higher masses. Although this thermal population of higher levels could certainly shift the center of gravity of the peak to higher magnetic fields (i.e., higher masses), there is no sign of a redshift of the peak even in the temperature range where the linewidth remains nearly the same (up to  $\sim 80$  K). Our calculations show that all cyclotron masses must decrease with increasing temperature, no matter which transitions are involved. Further theoretical efforts to model the observed behavior are under way.

To explore possible nonlinear phenomena in CR,<sup>12,13</sup> we performed some intensity-dependent measurements. Results are shown in Fig. 3 for the InSb

quantum well. Here the 4.7-THz QCL was used, and the sample temperature was 1.5 K. Because of the larger conduction band nonparabolicity in InSb, there are two clearly resolved resonances at this wavelength: 3.13 and 3.34 T. The two spectra shown here, taken at  $\sim 50 \mu\text{W}/\text{cm}^2$  and  $\sim 50 \text{ mW}/\text{cm}^2$ , look identical, exhibiting no sign of saturation. A study of CR in bulk InSb indicates<sup>14</sup> that saturation begins to occur at  $\sim 10^{-1} \text{ W}/\text{cm}^2$ .

In conclusion, we have used three terahertz quantum-cascade lasers operating at different frequencies to perform magneto-optical spectroscopy in semiconductors. This novel light source is more compact,

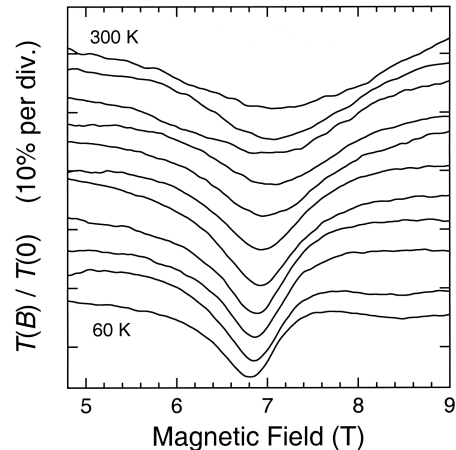


Fig. 1. Transmission as a function of magnetic field at several temperatures for InAs/AlSb quantum wells. The quantum-cascade laser wavelength is  $64 \mu\text{m}$  (4.7 THz), and the sample temperatures range from 60 to 300 K.

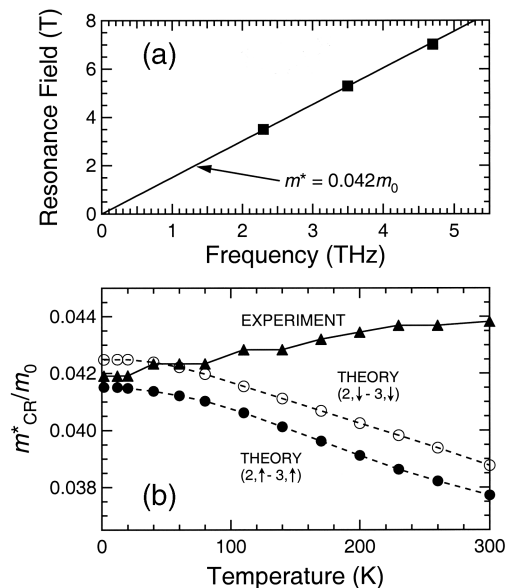


Fig. 2. (a) Resonance field as a function of photon frequency at 1.5 K for the InAs/AlSb quantum wells. The straight line has a slope of 1.51 T/THz, corresponding to an effective mass of  $0.042 m_0$ . (b) Cyclotron mass versus temperature at 4.7 THz for the InAs/AlSb quantum wells. The experimental mass (triangles) increases with increasing temperature, whereas the theoretical masses (open and filled circles) for two possible CR transitions show the opposite behavior.

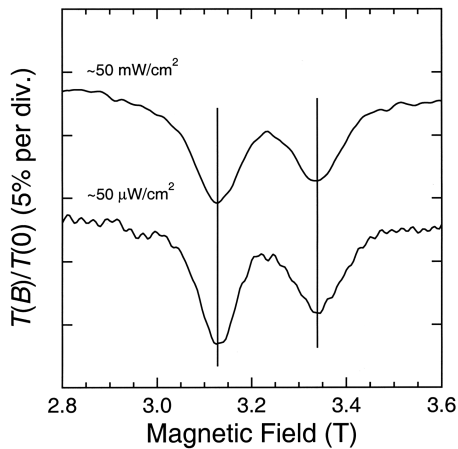


Fig. 3. Transmission as a function of magnetic field of an InSb/Al<sub>0.09</sub>In<sub>0.91</sub>Sb quantum well. The QCL's wavelength is 64  $\mu\text{m}$  (4.7 THz), and the sample temperature is 1.5 K.

more cost effective, and simpler to operate than existing far-infrared lasers. We demonstrated that these compact solid-state THz lasers have adequate power and stability for use in far-infrared magneto-optical studies of solids. We are currently developing a compact optically detected THz resonance<sup>15</sup> assembly that contains a QCL, which can be readily inserted into the bore of any superconducting magnet.

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## References

1. R. Koehler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, *Nature* **417**, 156 (2002).
2. M. Rochat, L. Ajili, H. Willenberg, J. Faist, H. Beere, G. Davies, E. Linfield, and D. Ritchie, *Appl. Phys. Lett.* **81**, 1381 (2002).
3. B. S. Williams, H. Callebaut, S. Kumar, Q. Hu, and J. L. Reno, *Appl. Phys. Lett.* **82**, 1015 (2003).
4. See, e.g., J. M. Chamberlain and R. E. Miles, eds., *New Directions in Terahertz Technology* (Kluwer Academic, Dordrecht, The Netherlands, 1997).
5. See, e.g., D. M. Mittleman, ed., *Sensing with Terahertz Radiation* (Springer-Verlag, Berlin, 2003).
6. See, e.g., F. Capasso, C. Gmachl, D. L. Sivco, and A. Y. Cho, *Phys. Today* **55**(5), 34 (2002), and references therein.
7. A. A. Kosterev and F. K. Tittel, *IEEE J. Quantum Electron.* **38**, 582 (2002).
8. J. Kono, in *Methods in Materials Research*, E. N. Kaufmann, R. Abbaschian, A. Bocarsly, C.-L. Chien, D. Dollimore, B. Doyle, A. Goldman, R. Gronsky, S. Pearton, and J. Sanchez, eds. (Wiley, New York, 2001), Unit 9b.2.
9. A. Barkan and D. M. Mittleman, Department of Electrical and Computer Engineering, Rice University, Houston, Texas 77005 (personal communication, August 21, 2003).
10. V. M. Apalkov and T. Chakraborty, *Appl. Phys. Lett.* **78**, 697 (2001).
11. C. R. Pidgeon and R. N. Brown, *Phys. Rev.* **146**, 575 (1966).
12. G. A. Rodriguez, R. M. Hart, A. J. Sievers, F. Keilmann, Z. Schlesinger, S. L. Wright, and W. I. Wang, *Appl. Phys. Lett.* **49**, 458 (1986).
13. S. K. Singh, B. D. McCombe, J. Kono, S. J. Allen, Jr., I. Lo, W. C. Mitchel, and C. E. Stutz, *Phys. Rev. B* **58**, 7286 (1998).
14. E. Gornik, T. Y. Chang, T. J. Bridges, V. T. Nguyen, J. D. McGee, and W. Müller, *Phys. Rev. Lett.* **40**, 1151 (1978).
15. J. Kono, S. T. Lee, M. S. Salib, G. S. Herold, A. Petrou, and B. D. McCombe, *Phys. Rev.* **52**, R8654 (1995).