Thermo-optic detection of terahertz radiation from a quantum cascade laser

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We investigate the asynchronous detection of terahertz radiation from a quantum cascade laser using an electro-optic sampling apparatus. The signal does not vary substantially upon rotating the detection crystal, while a characteristic angle dependence is exhibited for synchronized time-domain pulses from a photoconductive emitter. Upon increasing the electrical modulation frequency of the cascade laser the unsynchronized signal decreases in good agreement with a thermal detector. Rather than being electro-optic in origin, we therefore ascribe the unsynchronized signal to a thermo-optic modulation of the refractive index. A simple model is in good agreement with the effect's frequency dependence. © *2010 American Institute of Physics*. [doi:10.1063/1.3528456]

In terahertz time-domain spectroscopy[1] a sub-100fs near-infrared pulse from an unltrafast laser is converted into a single-cycle pulse of terahertz radiation, either via optical rectification[2] or photoconductive emission.[3] A time-delayed fraction of the original near-infrared pulse is used to perform a gated detection of the terahertz pulse, often by electro-optic sampling.[4] Here, the electric field of the terahertz pulse induces a birefringence in a crystal via the linear electro-optic (or Pockels) effect.[5,6] This birefringence can be measured using crossed-polarizers on either side of the electro-optic crystal and a single photodiode[4] or alternatively with a quarter waveplate, a Wollaston prism, and a pair of balanced photodiodes.[7] The latter method is generally preferred, as it is less sensitive to laser noise fluctuations.

Recently, it has been reported that it is possible to detect terahertz and mid-infrared radiation that is not synchronized with the probe beam in standard electro-optic sampling setups. Gaal *et al.*[8] reported a single-shot technique that stroboscopically measures mid-infrared radiation from a CO_2 laser using 12 fs pulses from a Ti:sapphire laser and a ZnTe electro-optic crystal with the balanced photodiode detection scheme. The detection of terahertz radiation from quantum cascade lasers (QCLs) has also been reported: at 2 THz using a CdTe crystal[9] and at 3 THz using a periodically poled GaP crystal.[10] In both cases a cross-polarized detection scheme without synchronization was used. The detection of continuous-wave (CW) terahertz radiation using a CW laser diode and a ZnTe electro-optic crystal has also been reported.[11] Apart from being of fundamental interest, an investigation of unsynchronized detection schemes may lead to an enhanced responsivity. In addition, since electro-optic sampling can be used to determine the gain of terahertz quantum cascade lasers operating above threshold[12] it is important to assess whether the asynchronous signal can also contribute.

In this letter we report the thermo-optic detection of terahertz radiation from QCLs. We investigated the unsynchronized signal from a 3.2 THz QCL using a balanced photodiode detection scheme with a ZnTe crystal and compared it to the gated detection of terahertz pulses from a photoconductive emitter. The dependence of the unsynchronized signal upon the angle of the ZnTe crystal and the QCL's modulation frequency are indicative of a slow thermo-optic effect, rather than an ultrafast electro-optic effect. A simple model is presented that accounts for the frequency dependence of the unsynchronized differential photodiode signal.

A terahertz time-domain spectrometer was used to detect terahertz radiation either from a QCL or from a photoconductive emitter. The 1.56 μm light from a 100 fs, 80 MHz Er-fiber laser was frequency doubled to 780 nm, where the beam's average power was 108 mW. About 75% of the beam was used to generate single-cycle pulses of terahertz radiation using a photoconductive switch (PCS) biased by a square wave.[13] These pulses were collimated and focused by off-axis parabolic mirrors before their gated detection via electro-optic sampling using the remaining 25 mW of the near-infrared beam. The electro-optic detection scheme used a ⟨110⟩ ZnTe crystal mounted in a rotation stage, a $\lambda/4$ plate, a Wollaston prism, and a pair of balanced photodiodes (inset of Fig. 1). The photodiodes were always balanced (by rotating the $\lambda/4$ plate) to give zero signal in the absence of terahertz radiation. For the intensity detection measurements the

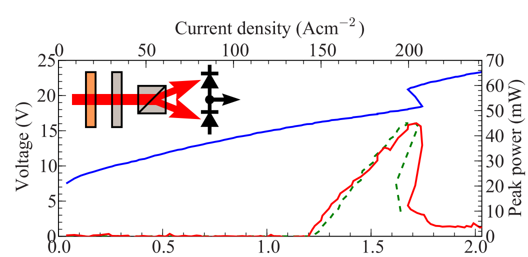

FIG. 1. (Color online) The current-voltage characteristic of the QCL (upper solid line, left-hand y-axis) when operated at 5% duty-cycle at 10 K. The balanced photodiode signal ΔI (dashed line) closely follows the intensity signal from a pyroelectric detector (lower solid line) in the range of laser action (current range of 1.2–1.7 A). Both detection schemes were calibrated by use of an absolute power meter, allowing the peak power to be plotted (right-hand y-axis). (Inset) Schematic of detection setup. From left to right the components are the ZnTe crystal, a quarter waveplate, a Wollaston prism, and a pair of balanced photodiodes.

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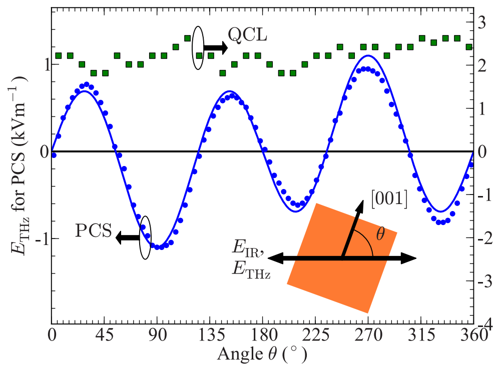

FIG. 2. (Color online) Crystal-angle θ dependence of the photodiode signal for the PCS and QCL. The peak of the terahertz pulse from the PCS (E_{THz}, points) depends on θ in a way that is reproduced by Eq. (1) (solid line). Using a 3.2 THz QCL as the emission source ($f=15$ Hz) produces a photodiode signal with responsivity R (squares, right axis) that does not depend on θ. (Inset) Geometry of experiment.

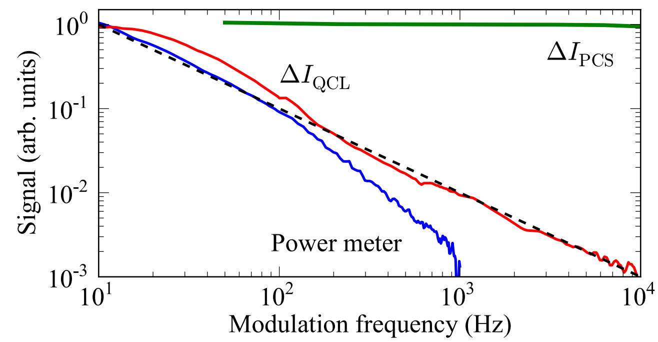

FIG. 3. (Color online) Dependence of the photodiode signal upon modulation frequency using the peak amplitude of the pulse from a photoconductive switch (ΔI_{PCS}) or a QCL (ΔI_{QCL}) as the source. Also shown are the power measured by a thermal detector and a line proportional to $1/f$ (dashed line). The electro-optic signal ΔI_{PCS} from a photoconductive switch is frequency independent, while $\Delta I_{QCL} \propto 1/f$ for the QCL and the power meter.

emitter pump beam was blocked, and the photoconductive switch was replaced by a single plasmon quantum cascade laser mounted in a cryostat at a heat-sink temperature of 10 K. The QCL was modulated electrically with a micropulse/macropulse scheme at a macropulse frequency f (50% duty-cycle) and a constant micropulse duration of 550 ns (10% duty-cycle). Two different QCL active region designs were utilized, emitting either at 3.2 THz (Ref. 14) or at 1.8 THz.[15] The 3.2 THz QCL has been previously reported to have a peak power of 45 mW and a narrow far-field emission pattern.[14] The terahertz radiation was detected at the focal spot of an off-axis parabolic mirror by either a pyrometer, a terahertz absolute power meter, or the electro-optic scheme using a lock-in amplifier referenced at f. The QCL and photoconductive emitter were oriented such that they produced polarized terahertz radiation in the same plane.

The differential signal recorded for the 3.2 THz QCL is reported in Fig. 1 as a function of the current in the QCL using either the pyrometer or the electro-optic detection scheme. The differential photodiode signal ΔI (dashed line in Fig. 1) is in good agreement with that measured by the pyrometer (solid line), indicating that $\Delta I \propto E_{THz}^2$ rather than the expected $\Delta I \propto E_{THz}$ for an electro-optic signal [see Eq. (1) and Refs. 5 and 6]. No measurable signal was observed from the 1.8 THz single plasmon QCL.

In order to elucidate the origin of the observed differential photodiode signal ΔI we rotated the detection crystal about the direction of the 780 nm probe beam using either the photoconductive switch or the 3.2 THz QCL (operated at maximum power) as the source. In our balanced photodiode detection scheme the differential photodiode signal from electro-optic sampling is given by[6]

$$\frac{\Delta I}{I_{tot}} = \frac{2\pi n^3 r_{41} L}{\lambda} \psi(\theta) E_{THz}, \tag{1}$$

where $n=2.7$ is the refractive index of ZnTe at $\lambda=780$ nm, $L=200$ μm is the thickness of the crystal, and $r_{41}=3.97\times10^{-12}$ mV^{-1}. The function $\psi(\theta)=(\cos\theta \sin 2\theta + 2\sin\theta\cos 2\theta)/2$ describes the angular dependence of the electro-optic signal. The geometry is shown in the inset of Fig. 2, where θ is defined as the angle between the [001] axis of the electro-optic crystal and the electric field directions of the near-infrared probe beam (E_{IR}) and terahertz beam (E_{THz}).[5]

In Fig. 2 the peak electric field strength E_{THz} for the photoconductive switch (dots) is plotted for different crystal angles θ. The solid line indicates a fit using $\psi(\theta)$, which reproduces well the angular dependence of E_{THz}. In contrast, ΔI_{QCL} exhibits no substantial angular dependence, as indicated by the responsivity $R=\Delta I_{QCL}/IP_{av}$ (squares) remaining constant at a value of $R=2.2$ mrad W^{-1} (where P_{av} is the average incident power from the QCL). The observation that ΔI_{QCL} is angle independent suggests that it is not produced by the Pockels effect.

A possible origin for ΔI_{QCL} is the quadratic electro-optic (or Kerr) effect, which can be unambiguously observed in isotropic centrosymmetric media without a second-order polarization $\chi^{(2)}$. In noncentrosymmetric crystals possessing a $\chi^{(2)}$ (such as ZnTe, GaP, and CdTe) the Kerr effect can become apparent at high fields. To assess this possibility we calculate E_{THz} for the QCL and PCS. From Eq. (1) the maximum amplitude of E_{THz} for the photoconductive switch is 1 kV m^{-1}. E_{THz} for the QCL can be deduced from the intensity at the focus $I_{QCL}\simeq 4.1\times10^4$ W m^{-2}, as obtained from the peak power (Fig. 1) and the spot-size at the ZnTe ($\sigma=590$ μm). Using the relation $I_{QCL}=n\epsilon_0 c E_{THz}^2/2$ the QCL is found to have a peak electric field strength of roughly 3.4 kV m^{-1} in the ZnTe crystal. While this is higher than the electric field of the PCS, it is several orders of magnitude lower than the value of $E_{THz}\simeq 10$ MV m^{-1} typically necessary to observe the Kerr effect at terahertz frequencies.[16] Therefore the Kerr effect is unlikely to contribute to the observed ΔI_{QCL}.

Further insight can be gained by measuring ΔI_{QCL} as a function of the macropulse frequency f at a constant micropulse duty-cycle (10%). In Fig. 3 the amplitude of $\Delta I_{QCL}(f)$ is plotted in the range of 10 Hz$<f<$10 kHz for the 3.2 THz QCL. Toward higher modulation frequencies ΔI_{QCL} decreases in accordance with $\Delta I_{QCL}\propto 1/f$ (dotted line). This behavior is typical of thermal detectors such as bolometers, pyrometers, and other power meters. For comparison the frequency dependence of the power measured by an absolute power meter is also reported in Fig. 3. In stark contrast, for a photoconductive switch the value of ΔI_{PCS} at the peak of the terahertz pulse is frequency independent, as expected for the time-gated detection of terahertz radiation via electro-optic sampling.

The frequency dependence of the unsynchronized signal ΔI_{QCL} points toward a thermal origin, as the signal is proportional to the total energy contained in a macropulse. The thermo-optic detection of intense terahertz radiation from a free-electron laser (0.6 MW peak power) has been recently reported using an absorptive medium (glass) via the change in the interference pattern under illumination with a CW 650 nm laser.[17] In the results reported here the frequency of the QCL lies close to the TO-phonon frequency of the detection crystal (5.3 THz for ZnTe and 4.2 THz for CdTe, Ref. 18). Consequently the absorption coefficient α of terahertz radiation in the detection crystal is not negligible.[18] In the following we therefore estimate how the absorbed energy alters the refractive index of the detection crystal.

For ZnTe, $\alpha=32\ \text{cm}^{-1}$ at 3.2 THz (Ref. 18), implying that $1-e^{-\alpha L}=47\%$ of the incident QCL beam is absorbed in our $L=200\ \mu$m thick crystal. From the measured optical power of the QCL we estimate that the energy deposited during one 16.6 ms long macropulse ($f=30$ Hz, 10% micropulse duty-cycle) is $Q=35\ \mu$J. The heat capacity C and density ρ of ZnTe and the spot-size σ allow us to estimate the local temperature change in the crystal induced by each macropulse as $\Delta T=Q/(C\pi\sigma^2 L\rho)\simeq 0.1$ K. For the 1.8 THz QCL the temperature change is much smaller ($\Delta T\simeq 1$ mK) as a consequence of the reduced $\alpha=13\ \text{cm}^{-1}$ at 1.8 THz and the lower optical power of the QCL ($\sim$1 mW). The average power of the photoconductive emitter is too low ($<1\ \mu$W) in the frequency range with high absorption to produce a significant thermo-optic signal.

The temperature dependence of the refractive index can be related to the thermal expansion coefficient and the temperature dependence of the excitonic bandgap[19] and is described by the thermo-optic coefficient dn/dT. At a wavelength λ the thermo-optic coefficient can be described by the equation

$$2n\frac{dn}{dT}=GF+HF^2, \tag{2}$$

where the function $F=\lambda^2/(\lambda^2-\lambda_g^2)$ for a material with refractive index n and isentropic bandgap at a wavelength λ_g.[19] For ZnTe the parameters G and H are less well known, but using the same values as for CdTe ($G=-9.2\times10^{-5}\ \text{K}^{-1}$, $H=6.0\times10^{-4}\ \text{K}^{-1}$, and $\lambda_g=0.51\ \mu$m) (Ref. 19) and with a probe wavelength of 0.78 μm (giving $F=1.8$) results in $\Delta n_{\text{th}}=\Delta T dn/dT\simeq 3\times10^{-5}$. This refractive index change is giant in comparison with that induced in electro-optic sampling of a PCS ($\Delta n\simeq 6\times10^{-8}$). However, in order to produce a signal ΔI in the balanced photodiode scheme the intensity of the horizontally and vertically polarized beams must differ, which necessitates a birefringence to be induced in the crystal. If Δn_{th} differs for the two principal axes of the crystal (which have refractive indices n_1 and n_2) then the thermally induced birefringence Δn_{th}^* would be

$$\Delta n_{\text{th}}^*=\Delta n_2-\Delta n_1=\Delta T\left(\frac{dn_2}{dT}-\frac{dn_1}{dT}\right),$$

$$=\Delta T\frac{GF+HF^2}{2}\left(\frac{1}{n_2}-\frac{1}{n_1}\right), \tag{3}$$

making use of Eq. (2) evaluated for n_1 and n_2 and assuming that G and H are isotropic. ZnTe has been reported previously to exhibit a birefringence induced by strain.[20] The thermally induced birefringence change is $\Delta n_{\text{th}}^*\propto\Delta T\propto Q\propto 1/f$, implying that the observed signal ΔI_{QCL}, which is proportional to the phase change,[6] obeys $\Delta I_{\text{QCL}}\propto\Delta n_{\text{th}}^*\propto 1/f$. This is in accordance with the experimental frequency dependence reported in Fig. 3.

Further experimental studies are necessary to identify the origin of the thermo-optic signal reported here and to establish whether Eq. (3) predicts the correct magnitude of the signal. Alternative mechanisms include piezoelectricity and pyroelectricity, which may be present as ZnTe has a noncentrosymmetric crystal structure. The possible influence of an asymmetric terahertz beam from the QCL could be examined by raster-scanning the position of the focused terahertz beam with respect to a fixed gate beam.

In summary, we have demonstrated that the intensity of terahertz radiation from quantum cascade lasers can be sampled asynchronously using a pulsed laser beam at 780 nm via a thermo-optic effect. The laser's emission characteristic can therefore be recorded using the same optics and electronics as needed to measure the gain of the laser via terahertz time-domain spectroscopy. Further investigation may lead to an enhanced detection efficiency either with materials with higher thermo-optic coefficients or with suitable reflective coatings.

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