Measuring intensity correlations of a THz quantum cascade laser around its threshold at sub-cycle timescales

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ABSTRACT

The quantum nature of photonic systems is reflected in the photon statistics of the light they emit. Therefore, the development of quantum optics tools with single photon sensitivity and excellent temporal resolution is paramount to the development of exotic sources, and is particularly challenging in the THz range where photon energies approach $k_B T$ at $T=300$ K. Here, we report on the first room temperature measurement of field $g^1(\tau)$ and intensity correlations $g^2(\tau)$ in the THz range with sub-cycle temporal resolution (146 fs) over the bandwidth 0.3-3 THz, based on electro-optic sampling. With this system, we are able to measure the photon statistics at threshold of a THz Quantum Cascade Laser.

Keywords: Quantum Optics, Photon statistics, THz Quantum Cascade Laser, threshold, ZnTe coherence properties

1. INTRODUCTION

The Terahertz frequency range of the electromagnetic spectrum (0.1-10 THz), lying between the optics and electronics, has witnessed up to date very advanced applications in spectroscopy and matter control,\textsuperscript{1} despite technological challenges which often require cryogenics for low background noise and free-space optics for minimized absorption. Devices operating in this range generally combine technologies belonging both to optics and to high-frequency electronics, which lead to light emission by uncommon mechanisms: lasers based on intersubband transitions configured with metallic strip wave-guides, photo-conductive antennas based on charge transients just to name a few. Nevertheless, the complexity of optical systems and the variety of applications is thus hindered by technological challenges, with notable realizations in the field of afm-imaging,\textsuperscript{2} higher harmonics generation\textsuperscript{3} and the development of local oscillators.\textsuperscript{4}

In future, the Terahertz frequency range might offer a fascinating playground for quantum optics studies beyond the phenomena demonstrated at optical and microwave frequencies. In particular, the physics in the limit of ultra-strong and deep strong light matter coupling predicts light emission through non-trivial mechanisms,\textsuperscript{5,6} such as from a non-excited system in its ground state which is non-adiabatically switched between uncoupled and ultra-strongly coupled regime. Notably, the technological maturity of nanoprocessing and epitaxial growth have allowed for above $\frac{\hbar \omega_c}{\Delta \omega} = 0.5$ coupling strengths to be demonstrated first at THz frequencies\textsuperscript{7} (0.58 at 500 GHz) and the frontier has been continuously pushed further to coupling strengths close to unity\textsuperscript{8} (0.87 at 500 GHz).

The quantum nature of photonic systems and the underlying emission mechanisms of light are reflected in the temporal correlation of their emission. The pioneering work of Hanbury Brown and Twiss,\textsuperscript{9} who raised big controversy with their measurement of coherence properties from star light, has nowadays essential technological implications in the characterization of light sources (single photon emitters, lasers, thermal radiation). Crucial requirements on the detectors including very low background noise, high sensitivity but also very good temporal resolution have seriously limited the experiments that could be performed so far at THz frequencies.

In this paper, we exploit nonlinear parametric interaction between the THz field under investigation and femtosecond probing pulses for the measurement of field and intensity correlations at THz frequencies. We

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characterize our technique in comparison with state-of-the-art detector technology and highlight its advantages. We demonstrate good temporal resolution, high sensitivity and low background noise. With this technique, we measure the threshold statistics of a THz Quantum Cascade Laser.

2. CORRELATION MEASUREMENTS AT THz FREQUENCIES

Correlations are ubiquitous tools for the characterization of statistical properties of quantum fields. Two distinct quantities have been introduced for the description of light sources and their optical coherence: $g^1(\tau)$ representing the degree of first order coherence, and $g^2(\tau)$, the degree of second order coherence.

$$g^1(\tau) = \frac{\langle E_{\text{THz}}^-(t)\tilde{E}_{\text{THz}}^+(t+\tau)\rangle_t}{\sqrt{\langle E_{\text{THz}}^-(t)\tilde{E}_{\text{THz}}^+(t)\rangle_t\langle E_{\text{THz}}^-(t)\tilde{E}_{\text{THz}}^+(t+\tau)\rangle_t}}$$

$$g^2(\tau) = \frac{\langle E_{\text{THz}}^-(t)\tilde{E}_{\text{THz}}^-(t+\tau)\rangle_t\langle E_{\text{THz}}^+(t)\tilde{E}_{\text{THz}}^+(t+\tau)\rangle_t}{\langle E_{\text{THz}}^-(t)\tilde{E}_{\text{THz}}^-(t)\rangle_t\langle E_{\text{THz}}^+(t)\tilde{E}_{\text{THz}}^+(t)\rangle_t}.$$

The Hanbury Brown and Twiss experiment measures the degree of second order coherence. It is performed by splitting the radiation under investigation with a beam splitter, delaying one arm by an arbitrary time $\tau$ and measuring the coincidence of detecting photons at two distinct intensity detectors. The necessity for ultra-short detection times has been raised since its introduction, and the use of fast detection schemes (100 fs) marked the point at which Hong, Ou and Mandel measured anti-bunching of photons. Alternative ways towards very short detection times have been proposed, among which two-photon absorption lead to the first measurement of photon bunching from thermal sources (1 fs resolution).

Nowadays THz detector technology permits for detectors with relatively short detection times and good sensitivity. However, they are subject to current research and typically require cryogenic cooling for low background noise. Commercially available solutions are mostly based on inherently slow pyroelectric detectors (ms). An overview of selected THz detector technology is presented in table 1 together with established NIR technology for comparison.

Table 1. Response time of various state-of-the-art detectors compared to the detected frequency.

| Detector type                                 | Response time // in units of oscillation period, $\frac{\tau_{\text{osc}}}{T_{\text{osc}}}$ |
|----------------------------------------------|---------------------------------------------------------------------------------------------|
| Electro-optic sampling (2 THz)               | 100 fs // ≈ 0.2                                                                            |
| Graphene based hot electron bolometer (1 THz)| 50 ps // ≈ 50                                                                               |
| Pyroelectric detector (1 THz)                | 1 ms // ≈ 1000000000                                                                        |
| Two-photon absorption (1550 nm)              | 2.5 fs // ≈ 0.5                                                                             |
| Single photon APDs (1550 nm)                 | 20 ps // ≈ 4000                                                                             |

2.1 Electro-optic sampling for correlation measurements

Among the coherent detection schemes, electro-optic sampling (EOS) utilized for Terahertz Time Domain Spectroscopy, makes use of the parametric non-linear interaction between a THz wave and a femtosecond long probing beam inside a $\chi^{(2)}$ medium in a travelling wave configuration. Upon interaction, the polarization of the probing beam is altered linearly to the THz electric field magnitude (figure 1a)). The interaction time is thus limited to the width of the probing pulse, which is typically on the order of 100 fs, rendering it very suitable for correlation measurements at ultrashort timescales. We exploit EOS to measure first and second order degree of coherence at THz frequencies.

A correlation measurement requires two distinct operations: the individual measurement of the same physical quantity at two distinct time-points and the averaging subsequent over all relevant times. While in standard techniques, the first requirement is met by use of a beam splitter and two distinct detectors, in our approach,
we make use of a second, mutually delayed (by a time $\tau$) probing pulse, combined with real-time measurement (at the laser repetition rate) of the electric field (figure 1b)). The time averaging is performed digitally post-measurement. We used a frequency doubled Erbium Doped Fiber Amplifier (EDFA) with a pulse width of 146 fs. A full description of the setup comprising all technical details is given elsewhere, in Ref.14.

Figure 1. Principle of EOS and correlation measurements. a) In EOS, the polarization state of an ultra-short probe is altered locally by the presence of a THz wave, linearly to the magnitude to the electric field at the sampling point. b) The correlation measurement scheme is based on two mutually delayed probing beams which sample the THz wave at two distinct timepoints without the necessity of a beam splitter.

Electro-optic sampling presents various advantages over other detection schemes available for Hanbury Brown Twiss experiments at THz frequencies. Being a parametric coherent technique, the electro-optic effect is most efficient when the coherence condition is met: the phase index of the THz wave has to equal the group index of the probing pulse.\textsuperscript{15,16} The interplay between probe wavelength, temperature and probe width allows for wide tunability and close-to-ideal coherence properties. The use of 110-oriented ZnTe as a detection crystal with $r_{41} = 4 \text{ pm/V}$, together with tunable pulses from a Ti:Sapph laser (center wavelength 725-850 nm), allows for coherence lengths on the order of several milimeters to be achieved anywhere in the range from 0.1-3 THz, as shown in figure 2. For the computation of the coherence length we used the frequency dependent refractive index reported here.\textsuperscript{17} Unlike conventional detectors, the bandwidth of detection extends over several octaves.

Figure 2. Coherence properties on 110-oriented ZnTe crystal in the detection range 0.1-3 THz. a) Refractive index of ZnTe at both interacting wavelengths. Schematic of coherence condition. b) Coherence length of ZnTe for different probe central wavelengths within the Ti:Sapph bandwidth.

The flexibility of this technique makes it applicable to other frequencies in the neighboring lower and higher frequency regions, with a fundamental Niquist limit imposed by the length of the femtosecond pulse. Nowadays
3. PHOTON STATISTICS OF A THZ QUANTUM CASCADE LASER

The photon statistics of a laser are expected to change at its threshold, from spontaneous emission with Super-Poissonian statistics below threshold \(g^2(0) = 2\), to coherent radiation above threshold with Poissonian statistics \(g^2(0) = 1\). A Terahertz Quantum Cascade Laser (QCL) is therefore a well suited device to be investigated in our setup. In addition, it has a typical bandwidth on the order of 20-30 %, and the short temporal resolution of the correlation scheme we propose in this paper could be exploited to its full potential when performing this measurement below lasing threshold.

We investigate the current dependent photon statistics of a THz QCL with an emission centered around 2.3 THz. The temporal resolution of our measurement of 146 fs is shorter than one cycle of oscillation of the THz emission, rendering it a sub-cycle measurement of \(g^2(\tau)\). The laser is 1 mm long and cooled to 25 K inside a cryostat, and the measurements are performed at lab temperatures. For better understanding, we perform a theoretical analysis described in the following.

3.1 Maxwell Bloch formalism for emission dynamics

The theoretical model we use to describe the photon statistics and the intra-cavity photon field of the laser is based on a Maxwell-Bloch formalism combined with modal decomposition and is discussed in detail here.\(^1\)

The output describes the time-resolved evolution of all lasing modes at a given pumping, modeled as gain. It takes into account a Lorentzian gain profile, dispersion and four wave mixing. We use this model to predict the threshold statistics of the laser as well as a current-dependent analysis in the whole dynamic range. To do so, we add to the existing model the spontaneous emission, which is the limiting quantum noise and will have a major influence on the photon statistics below and during threshold. The complete description of the modal amplitudes concludes to

\[
\dot{A}_n = \begin{cases} \frac{G_n - 1 + i \left( \frac{\omega_n^2 - \omega_{nc}^2}{2\omega_n} \right)}{\text{Net gain}} A_n - \mathbf{1} \times \frac{A_n^{\text{spont}}}{\text{spontaneous emission term}}. \quad (1) \end{cases}
\]

Here \(A_n\) is the complex amplitude of each mode in the laser cavity, \(G_n\) the normalized gain, and \(A_n^{\text{spont}}\) the Langevin force which describes the quantum fluctuation. It is modeled as a memory-less Markovian random process with an amplitude corresponding to one single photon inside the laser cavity. The computation of \(g^2(\tau)\) is trivial from the steady-state solution of the complex amplitudes.

Figure 3 shows characteristic temporal traces of the laser modes below (a) and above threshold (b), together with the modes supported by the laser cavity and the expected outcome of both a sub-cycle (as in our case) and a many-cycle (as in most cases) \(g^2(\tau)\) measurement of the laser emission.

The computation predicts various expected outcomes. Below threshold (here 90% of threshold gain), the gain of the laser is smaller than the losses and only spontaneous radiation is present in the cavity. The intra-cavity power is very low (small modal amplitudes) and decays at the cavity decay rate. The many-cycle value of \(g^2(\tau = 0) = 2\), as expected for spontaneous (bunched) emission. Due to interference effects, the sub-cycle value of \(g^2(\tau = 0) = 3.5\). Above threshold (here 103% of threshold gain), the laser is operating in single-mode since the gain is bigger than the losses only at its peak value. The modes inside the laser cavity experience gain and one gets amplified until reaching steady-state. The many-cycle value of \(g^2(\tau = 0) = 1\), as expected for lasing into one single coherent mode. The sub-cycle temporal shape of \(g^2(\tau)\) represents a cosine wave with an amplitude of 0.5 and an average of 1. The periodicity of the cosine wave corresponds to twice the lasing frequency, since the instantaneous intensity of a coherent state oscillates at double the emission frequency.
3.2 Sub-cycle measurement of $g_1^1(\tau)$ and $g_2^2(\tau)$

In the following we will present the results of the measurement of $g_1^1(\tau)$ and $g_2^2(\tau)$ with the electro-optic sampling method we present in this paper.

Figure 4 shows characteristic results from the laser in the threshold region (a) and above threshold (b). We retrieve the expected lasing frequency around 2.3 THz from the measurement of $g_1^1(\tau)$, whose Fourier transform gives the spectrum of emission. In addition, we find an increased value of $g_2^2(\tau = 0) = 1.18$ for the laser in the threshold region, as expected at this operating point. Above threshold, we retrieve a characteristic shape of $g_2^2(\tau)$ as expected: a cosine wave oscillating around 1 with an amplitude of 0.5 and double the emission frequency.

Further reading concerning measurements and analysis of the sensitivity limit of the setup are extensively discussed here.\[14\]

4. OUTLOOK

We have demonstrated the successful implementation of the first setup capable of measuring the first and second order degree of coherence at THz frequencies with unprecedented sub-cycle (146 fs) resolution. The properties of this type of setup, including very detection bandwidth, high tunability and high sensitivity make it a valuable solution in the Terahertz range and might be adapted for the neighboring mid-IR and microwave regions as well.

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Figure 4. Measurement results of $g_1^1(\tau)$ and $g_2^2(\tau)$ with fast electro-optic sampling in the threshold region (a) and above threshold (b), showing an increased value of $g_1^1(\tau = 0) = 1.18$ and $g_2^2(\tau = 0) = 1$, respectively. Double-frequency oscillations are retrieved for the sub-cycle measurement of $g_2^2(\tau)$.

REFERENCES

[1] Kampfrath, T., Tanaka, K., and Nelson, K. A., “Resonant and nonresonant control over matter and light by intense terahertz transients,” Nat Photon 7, 680–690 (Sept. 2013).
[2] Eisele, M., Cocker, L., Huber, M. A., Plankl, M., Viti, L., Ercolani, D., Sorba, L., Vitiello, M., and R., H., “Ultrafast multi-terahertz nano-spectroscopy with sub-cycle temporal resolution,” Nat Photon 8, 841–845 (Nov. 2014).
[3] Hohenleutner, M., Langer, F., Schubert, O., Knorr, M., Huttner, U., Koch, S. W., Kira, M., and Huber, R., “Real-time observation of interfering crystal electrons in high-harmonic generation,” Nature 523, 572–575 (July 2015).
[4] Huebers, H.-W., Pavlov, S. G., Semenov, A. D., Khler, R., Mahler, L., Tredicucci, A., Beere, H. E., Ritchie, D. A., and Linfield, E. H., “Terahertz quantum cascade laser as local oscillator in a heterodyne receiver,” Opt. Express 13(15), 5890–5896 (2005).
[5] Ridolfo, A., Savasta, S., and Hartmann, M. J., “Nonclassical Radiation from Thermal Cavities in the Ultrastrong Coupling Regime,” Physical Review Letters 110, 163601 (Apr. 2013).
[6] Ridolfo, A., Leib, M., Savasta, S., and Hartmann, M., “Photon Blockade in the Ultrastrong Coupling Regime,” Physical Review Letters 109, 193602 (Nov. 2012).
[7] Scalari, G., Maissen, C., Turcinkova, D., Hagenmüller, D., De Liberato, S., Ciuti, C., Reichl, C., Schuh, D., Wegscheider, W., Beck, M., and Faist, J., “Ultrastrong Coupling of the Cyclotron Transition of a 2D Electron Gas to a THz Metamaterial,” Science 335, 1323–1326 (Mar. 2012).
[8] Maissen, C., Scalari, G., Valmorra, F., Beck, M., Faist, J., Cibella, S., Leoni, R., Reichl, C., Charpentier, C., and Wegscheider, W., “Ultrastrong coupling in the near field of complementary split-ring resonators,” Physical Review B 90, 205309 (Nov. 2014).
[9] BROWN, R. H. and TWISS, R. Q., “Correlation between photons in two coherent beams of light,” Nature 177, 27–29 (Jan. 1956).
[10] Hong, C. K., Ou, Z. Y., and Mandel, L., “Measurement of subpicosecond time intervals between two photons by interference,” Phys. Rev. Lett. 59, 2044–2046 (Nov 1987).
[11] Boitier, F., Godard, A., Rosencher, E., and Fabre, C., “Measuring photon bunching at ultrashort timescale by two-photon absorption in semiconductors,” Nature Physics 5, 267–270 (Jan. 2009).
[12] Cai, X., Sushkov, A. B., Suess, R. J., Jadidi, M. M., Jenkins, G. S., Nyakiti, L. O., Myers-Ward, R. L., Li, S., Yan, J., Gaskill, D. K., Murphy, T. E., Drew, H. D., and Fuhrer, M. S., “Sensitive room-temperature terahertz detection via the photothermoelectric effect in graphene,” Nat Nano 9, 814–819 (Oct. 2014).
[13] Vicarelli, L., Vitiello, M. S., Coquillat, D., Lombardo, A., Ferrari, A. C., Knap, W., Polini, M., Pellegrini, V., and Tredicucci, A., “Graphene field-effect transistors as room-temperature terahertz detectors,” *Nature Materials* **11**, 865–871 (Sept. 2012).

[14] Benea-Chelmin, I.-C., Bonzon, C., Maissen, C., Scalari, G., Beck, M., and Faist, J., “Sub-cycle measurement of intensity correlations in the Terahertz range,” *ArXiv e-prints* (Dec. 2015).

[15] Gallot, G. and Grischkowsky, D., “Electro-optic detection of terahertz radiation,” *Journal Of The Optical Society Of America B-Optical Physics* **16**, 1204–1212 (Aug. 1999).

[16] Adam, A. J., van der Valk, N. C., and Planken, P. C., “Measurement and calculation of the near field of a terahertz apertureless scanning optical microscope,” *J. Opt. Soc. Am. B* **24**(5), 1080–1090 (2007).

[17] Nahata, A., Weling, A. S., and Heinz, T. F., “A wideband coherent terahertz spectroscopy system using optical rectification and electrooptic sampling,” *Applied Physics Letters* **69**(16), 2321–2323 (1996).

[18] Villares, G. and Faist, J., “Quantum cascade laser combs: effects of modulation and dispersion,” *Optics Express* **23**(2), 1651 (2015).