



UNIVERSITY OF LEEDS

This is a repository copy of *Sub-cycle high-order nonlinearities in a terahertz quantum cascade laser*.

White Rose Research Online URL for this paper:

<https://eprints.whiterose.ac.uk/193800/>

Version: Accepted Version

---

**Proceedings Paper:**

Riepl, J, Raab, J, Abajyan, P et al. (11 more authors) (2022) Sub-cycle high-order nonlinearities in a terahertz quantum cascade laser. In: 2022 47th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz). 2022 47th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz), 28 Aug - 02 Sep 2022, Delft, Netherlands. IEEE , pp. 1-2. ISBN 978-1-7281-9428-8

<https://doi.org/10.1109/irmmw-thz50927.2022.9895597>

---

© 2022 IEEE. Personal use of this material is permitted. Permission from IEEE must be obtained for all other uses, in any current or future media, including reprinting/republishing this material for advertising or promotional purposes, creating new collective works, for resale or redistribution to servers or lists, or reuse of any copyrighted component of this work in other works.

**Reuse**

Items deposited in White Rose Research Online are protected by copyright, with all rights reserved unless indicated otherwise. They may be downloaded and/or printed for private study, or other acts as permitted by national copyright laws. The publisher or other rights holders may allow further reproduction and re-use of the full text version. This is indicated by the licence information on the White Rose Research Online record for the item.

**Takedown**

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing [eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk) including the URL of the record and the reason for the withdrawal request.



[eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk)  
<https://eprints.whiterose.ac.uk/>

# Sub-cycle high-order nonlinearities in a terahertz quantum cascade laser

J. Riepl<sup>1</sup>, J. Raab<sup>1</sup>, P. Abajyan,<sup>2</sup> H. Nong<sup>2</sup>, J. R. Freeman,<sup>3</sup> L. H. Li,<sup>3</sup> E. H. Linfield,<sup>3</sup> A. G. Davies,<sup>3</sup> A. Wacker<sup>4</sup>, T. Albes<sup>5</sup>, C. Jirauschek<sup>5</sup>, C. Lange<sup>6</sup>, S. S. Dhillon<sup>2</sup> and R. Huber<sup>1</sup>

<sup>1</sup>Department of Physics, University of Regensburg, 93040 Regensburg, Germany

<sup>2</sup>Laboratoire de Physique de l'Ecole Normale Supérieure, CNRS, Sorbonne Université, Université de Paris, 75005 Paris, France

<sup>3</sup>School of Electronic and Electrical Engineering, University of Leeds, Woodhouse Lane, Leeds LS2 9JT, UK

<sup>4</sup>Mathematical Physics and NanoLund, Lund University, Box 118, SE-221 00 Lund, Sweden

<sup>5</sup>Department of Electrical and Computer Engineering, Technical University of Munich, 80333 Munich, Germany

<sup>6</sup>Department of Physics, TU Dortmund University, 44227, Dortmund, Germany

**Abstract**—Field-resolved two-dimensional terahertz spectroscopy reveals the sub-cycle gain dynamics of a free-running semiconductor laser and disentangles resonantly enhanced coherent and incoherent nonlinearities up to eight-wave mixing in a regime of negative absorption.

Utilizing ultrafast electron dynamics in quantum cascade lasers (QCLs) holds enormous potential for intense, compact mode-locked terahertz (THz) sources, frequency mixers, squeezed THz light, and comb-based metrology systems [1-4]. Yet the important sub-cycle dynamics have been notoriously difficult to access in operational THz QCLs.

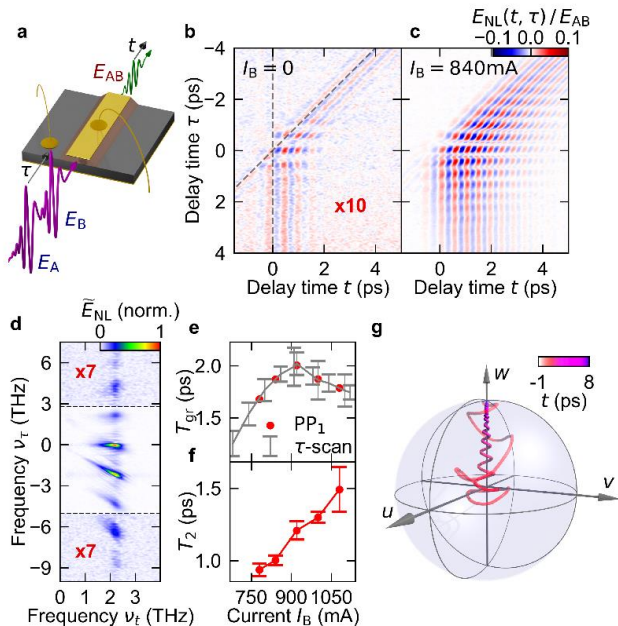
Here, we present the first ultrafast two-dimensional spectroscopy of a free running THz QCL [5]. Two identical, phase-stable, single-cycle THz waveforms  $E_A(t)$  and  $E_B(t, \tau)$  are mutually delayed by a variable time  $\tau$  and focused onto the QCL (Fig.

1a). The total transmitted field  $E_{AB}(t, \tau)$  is recorded with absolute phase and amplitude. Synchronous mechanical chopping of the two incident THz beams allows us to isolate the nonlinear response  $E_{NL}(t, \tau)$ . Figure 1b shows the resulting nonlinear response  $E_{NL}(t, \tau)$  for the unbiased laser. In sharp contrast, switching on the QCL (bias current of  $I_B = 840$  mA) leads to a qualitatively different response (Fig. 1c). First,  $E_{NL}$  increases by more than one order of magnitude, up to almost 10% of  $E_{AB}$ . Second, the coherent modulation following the second pulse is more long-lived and blue-shifted to the laser resonance of  $\nu_L = 2.2$  THz. Third, the modulation of  $E_{NL}$  along  $t$ , observed only near  $|\tau| = 0$  for the unbiased structure, now persists for much larger delays, signifying the presence of coherent nonlinear processes. To expand  $E_{NL}$  systematically into its constituent nonlinear processes [6], we perform a 2D Fourier transform of the time-domain data and apply a Liouville path analysis.

Remarkably we record coherent nonlinearities up to eight-wave mixing (Fig. 1d), which qualifies the QCL as an exceptionally efficient nonlinear optical medium and enables a direct extraction of the gain recovery time  $T_{gr}$  (Fig. 1e) and the dephasing time  $T_2$  (Fig. 1f) as a function of the bias current. The nonlinearities not only reveal extremely short gain recovery times, but also reflect the nonlinear polarization dynamics of the QCL laser transition for the first time. A density-matrix approach reproducing all nonlinearities and their ultrafast evolution, allows us to map the coherently induced trajectory of the Bloch vector (Fig. 1g). The observed nonlinearities benefit from resonant enhancement in a regime of negative absorption and bear potential for various future applications, ranging from efficient intracavity frequency conversion and mode proliferation to passive mode locking. Moreover, highly efficient and tunable multi-wave mixing opens up exciting perspectives for intracavity frequency conversion and multiplexing in a single compact electrically pumped device.

## REFERENCES

- [1] D. Burghoff et al. "Terahertz laser frequency combs" *Nat. Photon.* **8**, 462 (2014)
- [2] M. Rösch et al. "Octave-spanning semiconductor laser" *Nat. Photon.* **9**, 42 (2015).
- [3] S.S. Dhillon et al. "The 2017 terahertz science and technology roadmap" *J. Phys. D: Appl. Phys.* **50**, 043001 (2017).
- [4] F. Wang et al. "Ultrafast Response of Harmonic Modelocked THz Lasers" *Light Sci. Appl.*, **9**, 51 (2020).
- [5] J. Riepl et al. "Field-resolved high-order sub-cycle nonlinearities in a terahertz semiconductor laser" *Light Sci. Appl.* **10**, 246 (2021).
- [6] J. Raab et al. "Ultrafast terahertz saturable absorbers using tailored intersubband polaritons" *Nat. Commun.* **11**, 4290 (2020).



**Fig. 1. Field-resolved 2D THz spectroscopy of a QCL.** **a**, Experimental scheme. **b**, Nonlinear response  $E_{NL}(t, \tau)$  of the unbiased QCL. The electric field is multiplied by a factor of 10 and color-coded as in **(c)**. **c**,  $E_{NL}(t, \tau)$ , as in **(b)**, but for the biased QCL ( $I_B = 840$  mA). **d**, 2D amplitude spectrum of the nonlinear response in **(c)**. For  $\nu_\tau < -5$  THz and  $\nu_t > 2.8$  THz the spectrum is multiplied by a factor of 7. **e**, Gain recovery time  $T_{gr}$  of the QCL as a function of the bias current, extracted from the  $PP_1$  signal (red dots) and the full nonlinear time domain signal (grey). **f**, Decay time of the coherent population  $T_2$  as a function of the bias current. **g**, Temporal evolution of the QCL's density matrix, shown in a reduced Bloch sphere with radius 0.2 for a delay time of  $\tau = 0$  ps and a bias current of  $I_B = 780$  mA.