Proceedings Paper:
Riepl, J, Raab, J, Abajyan, P et al. (11 more authors) (2022) Field-resolved high-order nonlinearities in a free-running terahertz quantum cascade laser. In: Proceedings of the 2022 Conference on Lasers and Electro-Optics, CLEO 2022 - Proceedings. 2022 Conference on Lasers and Electro-Optics (CLEO), 15-20 May 2022, San Jose, CA, USA. IEEE , pp. 1-2. ISBN 9781957171050

Field-resolved high-order nonlinearities in a free-running terahertz quantum cascade laser

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***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 τ 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.

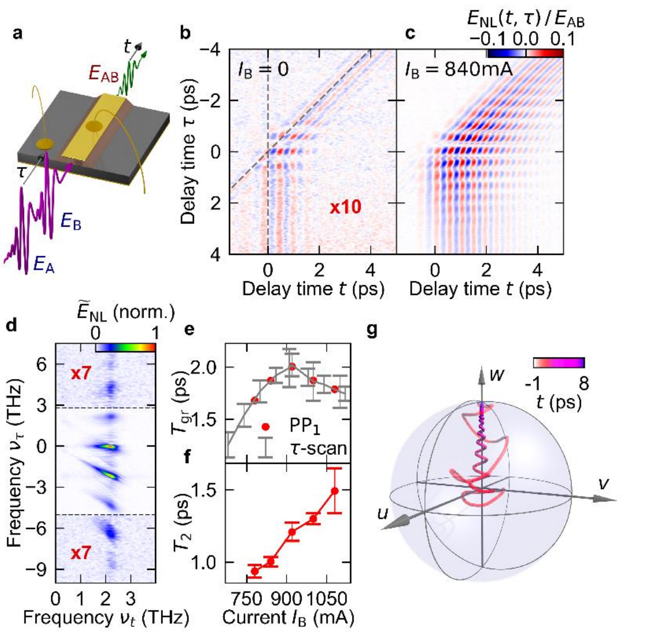

Fig. 1. Field-resolved 2D THz spectroscopy of a QCL. a, Experimental scheme. **b,** Nonlinear response $E_{NL}(t,\tau)$ of the unbiased QCL. The electric field is multiplied by a factor of 10 and color-coded as in (**c**). **c,** $E_{NL}(t,\tau)$, as in (**b**), but for the biased QCL ($I_B = 840$ mA). **d,** 2D amplitude spectrum of the nonlinear response in (**c**). For $\nu_\tau < -5$ THz and $\nu_\tau > 2.8$ 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.

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.

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