Multimode Dynamics and Frequency Comb Generation in Quantum Cascade Lasers

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***Abstract*-In this talk I will discuss how resonant light-matter interaction in the gain medium of quantum cascade lasers gives rise to a rich nonlinear multimode dynamics and a variety of phase-locked multimode regimes, most notably optical frequency combs with separation between the comb lines changing from one to many dozen round-trip frequencies. I will review recent progress in understanding why frequency comb formation seems to be so ubiquitous in QCLs.**

Optical frequency combs with equidistant separation and locked phases between the comb lines which serve as a ruler in the frequency domain, have revolutionized spectroscopy and metrology. A standard method of producing frequency combs is based on mode-locked lasers generating ultrashort pulses, which operate mostly in the near-infrared and visible ranges. The mid-infrared (mid-IR) and terahertz (THz) spectral regions, where most chemical compounds have strong spectral fingerprints, hold enormous potential for frequency comb applications. Quantum cascade laser (QCLs) cover most of the mid-IR and parts of the THz spectral regions. Unfortunately, ultrafast gain relaxation in QCLs effectively prohibits ultrashort pulse generation through passive mode locking. Active mode locking schemes are still feasible but they are less convenient and produce pulses of limited peak power [1-5], although there are some recent advances in this direction: see [6], where a large modulation depth was achieved by careful design of the active region. Note that THz QCLs with diagonal laser transitions and longer gain recovery time have shown passive mode locking and, recently, harmonic mode locking [7,8].

So, generation of ultrashort pulses in mid-IR QCLs has proven to be an uphill battle. That is why the realization that strong resonant nonlinearity of the gain transition itself is enough to trigger the formation of frequency combs in QCLs was such a pleasant surprise [9-11]. It turned out that the phase-sensitive part of four-wave mixing (FWM) interactions mediated by the gain transition is enough to couple frequency and phases of QCL cavity modes into an equidistant comb; no additional nonlinear element is needed [10,11]. These early works showed synchronization of combs belonging to different lateral modes, which was not very practical. In [12] a much broader comb belonging to a single transverse mode in a QCL with ultra-broad gain spectrum was demonstrated, which opened the floodgate of QCL frequency comb efforts in the mid-IR and later in the THz.

Recent theoretical studies [14-16] confirmed that it is FWM mediated by the ultrafast gain transition which remains the main mechanism responsible for the comb formation. In conventional Fabry-Perot QCLs multimode instability results mainly from the nonuniform gain distribution caused by standing laser field, i.e. spatial hole burning (SHB) [17]. The frequency comb is formed when the growing side modes become frequency and phase-locked due to resonant four wave mixing. However, unlike the shorter-wavelength combs generated by ultrashort-pulse lasers, in QCLs the underlying periodic modulation is linked to a predominantly frequency-modulated (FM) optical wave [13,17]. In general, when the gain recovery time is infinitely short, an ideal FM output is mandated by the variational principle [18]. However, the gain recovery time is short but still finite. Therefore, the amplitude modulation (AM) due to population pulsations is always present to some extent and can be directly measured [19].

A new twist in the QCL frequency comb saga started a few years ago, when the harmonic state in QCL operation was discovered [20] In the harmonic state the lasing modes are separated in frequency domain by a large number of free spectral ranges (FSRs), often tens of FSRs, which makes it distinct from a conventional laser state, where laser modes are separated by one FSR. It was later verified that harmonic state is also a frequency comb, i.e. the cavity modes are strictly equidistant and phase locked [14] The separation between neighboring lasing modes in the harmonic combs reaches hundreds of gigahertz and even THz frequencies, which implies coherent (sub)picosecond intracavity modulation. Moreover, the separation can be tuned by optical seeding [21,22]. Therefore, harmonic QCL combs provide direct link between mid-IR lasing and coherent (sub)THz generation and modulation [19,23,24], which makes them promising as (sub)THz coherent sources and transceivers. At the same time, such a large frequency interval between modes makes experimental characterization of harmonic combs very difficult and requires sophisticated experimental techniques [18,19,23,24]. Some properties of the harmonic state still cannot be measured directly. Therefore, many aspects of the underlying physical mechanisms which give rise to complete suppression of neighboring cavity modes and the harmonic comb formation remain a mystery. This makes theoretical studies and in particular analytic theory insight into the harmonic comb physics particularly valuable.

It would be tempting to explain the harmonic comb formation as the multimode instability corresponding to the AM optical wave, which peaks at large frequency detunings [17,20]. However, in Fabry-Perot lasers and within the two-level model of the gain transition, the AM instability is suppressed by the FM instability and the self-starting harmonic comb seems impossible.

Yet another recent puzzle is the formation of frequency combs in ring QCLs in the unidirectional regime. Contrary to the current understanding that in the absence of SHB and inhomogeneous spectral broadening the laser should maintain single-mode operation up to extreme pumping conditions of the RNGH instability [17,20], a dramatic lowering of the

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multimode instability threshold was recently demonstrated in unidirectional ring QCLs [25].

Several recent studies are trying to solve these puzzles. For example, the phase matching requirement in dispersive waveguides was shown to suppress lasing of neighboring modes and favor harmonic combs [16]. In many QCL designs, especially in THz QCLs [8], the gain has significant contributions from at least two intersubband transitions with different central frequencies and dipole moments. This leads to the formation of sidebands with very different amplitudes and resulting AM dynamics, again favoring the harmonic combs [8]. Finally, even for a single gain transition, taking into account the second-order dispersive gain (Bloch gain) leads to strong coupling of the amplitude and phase fluctuations (the nonzero linewidth enhancement factor [26]) which favors the low-threshold comb formation even in ring QCLs [25].

To summarize, the mode locking and frequency comb formation in QCLs remains a rapidly growing research area while many puzzles still remain. It is fascinating how QCLs continue bringing new surprising features to such a well studied field as fundamental laser dynamics.

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