Nonlinear dynamics of light-matter interaction in quantum cascade lasers

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Outline

• Optical nonlinearities in QCLs
• Saturation nonlinearity and its many faces
• Frequency and phase locking of laser modes
• Frequency combs
• Harmonic frequency combs
• Generation of mode-locked pulses
  – Passive mode locking
  – Active mode locking
• Conclusions
Optical nonlinearities relevant for QCLs

• Nonlinear saturation of the ISB transition
  – Controls everything related to laser operation, regimes, mode locking, frequency combs (lectures by J. Faist and A. Hugi)

• Optical nonlinearity of a bulk semiconductor crystal or free carriers (both second and third order)
  – QCL as a heterodyne receiver or transceiver (e.g. M. Wanke)

• Structural nonlinearity of a multiple quantum well system: e.g. a confinement potential which breaks the inversion symmetry
  – Frequency self-mixing (SFG, DFG) – lecture by M. Belkin
  – Frequency shifting and generation of THz subbands by injection of a NIR radiation into a THz QCL (Dhillon, Sirtori et al.)
Saturation nonlinearity and its many faces

– Limits growth of the EM field and polarization
– Determines steady-state output power
– Determines laser response to fast modulation
– Couples different EM modes, leading to phase coupling and mode locking

\[
N_2 - N_1 \sim \frac{(N_2 - N_1)_{E=0}}{1 + (1/I_s) \sum E_\mu E_\nu}
\]

\(|E|^2\)
Quantum cascade laser

injector

injector (n-doped)

active region

injector (n-doped)

active region

Period

520 meV

Picture from C. Gmachl’s LEOS lecture
Simple model that we use in simulations

Short gain relaxation time $T_1 \sim 1$ ps

Cavity roundtrip time $T_{RT} \sim 50$ ps

Photon lifetime $T_p \sim 10$ ps

Dephasing time $T_2 \sim 0.1$ ps

$T_2 < T_1 << T_{p,RT}$

A Class-A laser!

All other solid-state and diode lasers are Class B: $T_2 << T_{RT,p} << T_1$

However, dynamical times can be of order $T_{1,2}$

$$\omega_c^2 \sim \frac{4\pi d^2 \omega \Delta N}{\hbar}; \quad \Omega_R = \frac{dE}{\hbar}$$
Single-mode laser, identical two-level “atoms”, exact resonance with the gain transition

\[ \frac{\partial E}{\partial t} + \frac{c}{\mu} \frac{\partial E}{\partial z} + \alpha E = \Gamma \frac{2\pi i \omega_0 N d}{\mu^2} \sigma \]

\[ \frac{\partial \sigma}{\partial t} + \frac{\sigma}{T_2} = -\frac{idE}{2\hbar} \Delta \]

\[ \frac{\partial \Delta}{\partial t} = \frac{\Delta_p - \Delta}{T_1} - \frac{2 \text{Im}(dE \sigma^*)}{\hbar} \]

Inversion supported by pumping \( \Delta_p \)

Effective index \( \mu \approx 3.3 \); overlap \( \Gamma \approx 0.5 \)

\[ \frac{\Delta_p}{T_1} \propto j / e \quad \text{Injection current} \]

Note a dimensionless parameter:

\[ \left( \frac{dE}{\hbar} \right)^2 T_1 T_2 = \frac{E^2}{E_s^2} \]

\[ \Delta = \frac{N_2 - N_1}{N} \]

\[ N = N_2 + N_1 \]

\[ P = 2 N d \sigma \]

Polarization amplitude

Very short \( T_2 \)

Very short \( T_1 \)

\[ \Delta = \frac{\Delta_p}{1 + \left( \frac{dE}{\hbar} \right)^2 \frac{T_1 T_2}{T_1 T_2}} = \frac{\Delta_p}{1 + \left( \frac{E}{E_s} \right)^2} \]

\[ E^2 = E_s^2 \left( \frac{\Delta_p}{\Delta} - 1 \right) \]

\[ E_s^2 = \frac{\hbar^2}{d^2 T_1 T_2} \]
Simple numerical estimates

\[ \sigma = -\frac{idE}{2\hbar}T_2\Delta \]

\[ \Delta = \frac{\Delta_p}{1 + \left(\frac{dE}{\hbar}\right)^2 T_1T_2} = \frac{\Delta_p}{1 + \left(\frac{E}{E_s}\right)^2} \]

\[ \frac{\partial E}{\partial t} + c \frac{\partial E}{\mu \partial z} + \alpha E = \Gamma \frac{2\pi i \omega_0 N d}{\mu^2 E^2} \sigma \]

\[ d = e \times 2 \text{ nm}, \quad N\Delta_p \sim \frac{10^{10} \text{ cm}^{-2}}{50 \text{ nm}} \sim 2 \times 10^{17} \text{ cm}^{-3}; \quad \mu \sim 3.3; \quad \Gamma \sim 0.5 \]

\[ E_s \sim 10 \text{ kV/cm}; \quad I_s \sim 300 \text{ kW/cm}^2; \quad \text{corresponds to intracavity power} \sim 100 \text{ mW} \]

\[ |\chi^{(3)}| = \left| \frac{\chi^{(1)}}{E_s^2} \right| \sim 10^{-14} \text{ m}^2 / \text{V}^2 \]

In transparent crystals

\[ \chi^{(3)} \sim 10^{-20} - 10^{-23} \text{ m}^2 / \text{V}^2 \]

Saturation nonlinearity: very strong (resonant; large dipole moment)
ultrafast (1 ps), broadband (10^{13} \text{ s}^{-1})
Multimode operation, two levels

\[
\frac{d\sigma}{dt} + \frac{\sigma}{T_2} = -\frac{id}{2\hbar} \Delta \sum_\lambda a_\lambda^* E_\lambda (\mathbf{r})
\]

\[
\frac{d\Delta}{dt} = \frac{\Delta_p - \Delta}{T_1} - \frac{id}{\hbar} \sum_\lambda E_\lambda (\mathbf{r})(a_\lambda^* \sigma - a_\lambda \sigma^*)
\]

\[
\frac{da_\lambda}{dt} + (\alpha_\lambda + i(\omega_\lambda - \omega_c))a_\lambda = 4\pi i\omega_0 Nd \frac{1}{V_c} \int \sigma E_\lambda (\mathbf{r}) dV
\]

Field \( E(\mathbf{r},t) = \sum_\lambda (1/2)a_\lambda(t) \exp(-i\omega_0 t + i\varphi_\lambda) E_\lambda (\mathbf{r}) + \text{c.c.} \)

Polarization \( P = Nd \sigma e^{-i\omega_0 t} + \text{c.c.} \)

Population inversion \( \Delta = \frac{N_2 - N_1}{N} \)

Due to a difference in scales, 3D cavity modes can be split into longitudinal and transverse.
Each transverse mode creates a “comb” of longitudinal modes (not phase-locked) and these “combs” overlap.

Note close grouping of modes with different transverse (and longitudinal) indices.

EM modes in QCLs: interaction through saturation nonlinearity

Longitudinal modes

\[ |E(z)|^2 \]

\[ \Delta N = N_2 - N_1 \]

Transverse Modes

- \( \text{TM}_{00} \)
- \( \text{TM}_{01} \)
- \( \text{TM}_{02} \)

Cavity cross-section

Fabry-Perot cavity

Multimode operation
Saturation nonlinearity is a fast and efficient mechanism of mode competition and coupling.

Saturation is inhomogeneous in space and in frequency (hole burning). It is also phase-dependent.

\[ N_2 - N_1 \sim \frac{(N_2 - N_1)_{E=0}}{1 + \left(\frac{1}{E_s^2}\right)\sum E_\mu E_\nu} \]
The first observation of phase-locked frequency combs was in broad-area QCLs with multiple lateral modes.
Frequency and phase locking of transverse modes

**Observed signatures:**

- Anomalous near-field and far-field beam pattern which cannot be explained by adding mode intensities
- Beam steering by current
- Locking to commensurate frequencies or synchronization of lateral modes to a single comb
- These effects appear and disappear as a bifurcation, with a slight change in injection current

Yu et al. PRL 2009; Wojcik et al. OE review 2010, PRL 2011, JMO review 2011
Frequency and phase locking of three combs

![Graphs and diagrams showing voltage, peak power, intensity, and wavelength measurements.](image)

Waveguide width 19 μm

Wojcik et al. PRL 2011
Multimode operation, two levels

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\frac{d\Delta}{dt} = \frac{\Delta_p - \Delta}{T_1} - \frac{id}{\hbar} \sum \lambda E_{\lambda}(\mathbf{r})(a_{\lambda}^* \sigma - a_{\lambda} \sigma^*)
\]

\[
\frac{da_{\lambda}}{dt} + (\alpha_{\lambda} + i(\omega_{\lambda} - \omega_c))a_{\lambda} = 4\pi i\omega_0Nd\frac{1}{V_c} \int \sigma E_{\lambda}(\mathbf{r})dV
\]

Field \( E(\mathbf{r},t) = \sum \lambda (1/2)a_{\lambda}(t)\exp(-i\omega_0 t + i\varphi_{\lambda})E_{\lambda}(\mathbf{r}) + \text{c.c.} \)

Polarization \( P = Nd\sigma e^{-i\omega_0 t} + \text{c.c.} \)

Population inversion \( \Delta = \frac{N_2 - N_1}{N} \)
• Adiabatic elimination of inversion and polarization ($T_{1,2} \rightarrow 0$)
• $X^{(3)}$ approximation

Coupled equations for modal amplitudes:

$$\frac{da_j}{dt} + \left( \alpha_j + i(\omega_{ej} - \omega_0) \right) a_j = \frac{g_j}{2} \left( \sum_{k} a_k \int \varepsilon E_j E_k \, dV - \frac{2}{E_s^2} \sum_{k,l,m} G_{jklm} a_k a_l^* a_m \right)$$

Cavity dispersion/loss  Modal gain  Nonlinear mixing

Nonlinear overlap

$$G_{jklm} = \int_{AR} \varepsilon E_j E_k E_l E_m \, dV$$

Saturation intensity

$$E_s^2 = 2\hbar^2 / (d^2 T_1 T_2)$$

Fast gain relaxation $T_1 \sim 1$ ps (Type A laser) damps relaxation oscillations and leads to stable frequency pulling and phase locking. No saturable absorber or external modulation needed! (compare with diode lasers)
• Adiabatic elimination of inversion and polarization ($T_{1,2} \rightarrow 0$)
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Coupled equations for modal amplitudes:

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\frac{da_j}{dt} + (\alpha_j + i(\omega_{cj} - \omega_0))a_j = \frac{g_j}{2} \left( \sum_k a_k \int \varepsilon E_j E_k dV - \frac{2}{E_s^2} \sum_{k,l,m} G_{jklm} a_k a^*_l a_m \right)
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- Modal gain
- Nonlinear mixing

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- **Cavity dispersion/loss**
- **Modal gain**
- **Nonlinear mixing**

**Nonlinear overlap**

\[
G_{jklm} = \int_{AR} \varepsilon E_j E_k E_l E_m \, dV
\]

**Saturation intensity**

\[
E_s^2 = \frac{2\hbar^2}{d^2 T_1 T_2}
\]

In the limit of $T_{1,2} \rightarrow 0$ and neglecting cavity dispersion the coupled-oscillator equations can be derived from the Hamilton’s principle and coincide with the “maximum emission principle”: the laser has “urge” to maximize energy extraction (Statz and Tang 1967; Schwarz and Gordon 1969).
A system of weakly coupled oscillators:

Could be the most popular model of various phenomena in physics, biology, geosciences, chemical reactions, psychology, economics, ...

http://web.mac.com/chuck_krousgrill/iWeb/ME563_fall06
First scientific study of synchronization?

In 1665 Christiaan Huygens observed an “odd kind of sympathy” between pendulum clocks hung together.


Theory: Appleton & van der Pol, 1920s

http://www.scholarpedia.org/article/Synchronization

Original drawing by Huygens
Synchrony of fireflies

Chirping of crickets

http://www.youtube.com/watch?v=sROKYelaWbo
Spontaneously synchronized clapping in a concert hall

"... This perhaps explains why in the smaller and culturally more homogeneous eastern European communities, synchronized clapping is a daily event, whereas it happens only sporadically in western European and North American audiences."

Overlapping spectral combs

Nonlinear interaction leads to frequency pulling

Three combs can lock into equidistant triplets or even to a single comb (synchronization)
9 Modes - Dynamics of 3 Close Triplets

30 initial conditions for each gain

Now interaction between different triplets is important
Dynamics is more complex and less stable
Two regions of synchronization separated by multi-stability or chaotic dynamics
For applications you want a single lateral mode and more predictable behavior.

Phase-locked frequency combs from high-performance, single lateral mode QCLs

Jerome Faist’s group (ETH Zurich), 2012

Combs vs. No Combs

Multimode generation of uncoupled modes

- Modes are non-equidistant
- Total field $E(t)$ is not periodic
- Phase differences are random and change with time

From M. Piccardo slides
- Adiabatic elimination of inversion and polarization ($T_{1,2} \rightarrow 0$)
- $X^{(3)}$ approximation

Coupled equations for modal amplitudes:

$$\frac{da_j}{dt} + (\alpha_j + i(\omega_{ej} - \omega_0))a_j = \frac{g_j}{2} \left( \sum_k a_k \int \varepsilon E_j E_k dV - \frac{2}{E_s^2} \sum_{k,l,m} G_{jklm} a_k a_l^* a_m \right)$$

Nonlinear mixing

$$2\omega_2 \leftrightarrow \omega_1 + \omega_3$$

$k = l, m = j$: phase-insensitive

Otherwise: phase sensitive

Frequency pulling to equidistant comb

Phase locking
Frequency combs

- Equidistant modes: the field is strictly periodic in time
- Stable phase differences over a large number of round-trips (could be $10^8$ or more)

\[ f_n = f_{ceo} + n \cdot f_{rep} \]
How does the field of a comb look in time domain?

- Generation of sidebands corresponds to a signal being modulated in time.
- Is the signal amplitude-modulated, frequency modulated, or both?

\[
\frac{d\sigma}{dt} + \frac{\sigma}{T_2} = -\frac{i d}{2\hbar} \sum_{\lambda} a_{\lambda} E_{\lambda}(\mathbf{r})
\]

\[
\frac{d\Delta}{dt} = \frac{\Delta_p - \Delta}{T_1} - \frac{id}{\hbar} \sum_{\lambda} E_{\lambda}(\mathbf{r})(a_{\lambda}^* \sigma - a_{\lambda} \sigma^*)
\]

\[
\frac{da_{\lambda}}{dt} + \left( \alpha_{\lambda} + i(\omega_{\lambda} - \omega_c) \right) a_{\lambda} = 4\pi i \omega_0 Nd \frac{1}{V_c} \int \sigma E_{\lambda}(\mathbf{r}) dV
\]

- In the limit of \( T_{1,2} \to 0 \), polarization and inversion instantaneously follow the field.

- In this limit any amplitude modulation should be suppressed and the signal should be close to FM. Indeed, an ultrafast gain medium quenches any amplitude fluctuations.

- However, experiments reveal a significant amplitude modulation: between 40% and 90% (Singleton et al., Optica 2018, Burghoff et al., Opt.Expr. 2015 and Nat. Phot. 2014)
Strong amplitude modulation
Singleton et al. 2018
• Relaxation times are finite
• In a strong enough field, populations will oscillate

Cooperative frequency: controls superradiant regimes

\[ \omega_c^2 = \frac{2\pi \omega_0 d^2 N}{\hbar} \]

Rabi frequency: \( \Omega = \frac{dE}{\hbar} \)
Coherent oscillations of populations and polarization

Oscillations of populations create parametric gain for sidemodes, cause phase coupling of modes or/and proliferation of equidistant modes with fixed phases

The multimode generation is always an interplay of two mechanisms: population grating (spatial hole burning) and population oscillations (coherent parametric process)

Gain for sidemodes in the presence of a strong central mode has maxima separated by 100s of GHz or THz frequencies

These plots suggest that the spectra may have widely separated maxima. Turns out, a regime is possible where intermediate modes don’t even lase!


D. Kazakov et al., Nat. Phot. 2017
Harmonic frequency comb
The Harvard experiment

- Observed in many QCLs of different designs and wavelengths
- Stable and reproducible
- Sensitive to any optical feedback
- Shows hysteresis
Delayed optical feedback destroys the harmonic state

![Spectral evolution of a QCL obtained collecting its spectral output by different schemes. (a) Purely diverging QCL output. (b) QCL output collimated with an off-axis parabolic mirror (OAP). (c) QCL output collimated using a ZnSe lens.](image)

M. Piccardo et al. Opt. Expr. 2018
Space-time domain simulations

Resonant tunneling:

\[ J = \frac{e\Omega^2\gamma}{\hbar(\Delta^2 + \gamma^2)} \left( n_g e^{-|\Delta|/k_B T} - n_u \right) \]

\[ \partial_t n_g = \frac{n_u}{T_{ug}} + \frac{n_l}{T_{lg}} - J \]

\[ \partial_t n_u = J - \frac{n_u}{T_{ul}} - \frac{n_u}{T_{ug}} - i\frac{dE}{\hbar} (\rho_{ul} - \rho_{ul}^*) \]

\[ \partial_t n_l = \frac{n_u}{T_{ul}} - \frac{n_l}{T_{lg}} + i\frac{dE}{\hbar} (\rho_{ul} - \rho_{ul}^*) \]

\[ \partial_t \rho_{ul} = -(i\omega + 1/T_2)\rho_{ul} - i\frac{dE}{\hbar} (n_u - n_l) \]

\[ \partial_z^2 E - \frac{n^2}{c^2} \partial_t^2 E = \frac{\Gamma d}{\epsilon_0 c^2 L_p} \partial_t^2 (\rho_{ul} + \rho_{ul}^*) \]
Space-time domain simulations
Analytic theory for one strong central mode and two strong subbands

Stable lasing on three modes separated by 420 GHz exists in a certain range of pumping parameters
Harmonic frequency comb

Corresponds to beatnote frequency of 100s GHz

Modes are exactly equidistant and phase locked

D. Kazakov et al., Nat. Phot. 2017
M. Piccardo et al. Optica 2018; Opt. Expr. 2018
P. Chevalier, APL 2018
Multiheterodyne, self-detection measurement

Normalized to the optical carrier frequency (66.7 THz), the spacing uniformity of the harmonic comb is verified with an accuracy of $4.9 \times 10^{-12}$.

D. Kazakov et al., Nat. Phot. 2017
Tuning of the sidebands separation by weak optical injection

Tuning between 0.3 and 1.3 THz!
We get a THz beatnote “for free”. Can we use it for THz applications?!

M. Piccardo et al., Optica 2018
A laser radio transmitter

Coherent oscillations of populations give rise to the lateral current oscillating at the beatnote frequency and its harmonics.
A laser radio transmitter

M. Piccardo et al., Science Advances, under review

This is for a “dense” frequency comb with a beatnote at 5.5 GHz. THz is work in progress
Is it possible to lock modes with “right” phases, so that the output is isolated pulses?

\[ E(t, z) = \sum_{m} E_m(z)e^{i(\omega_m t + \phi_m)} + \text{c.c.} \]
Mode locking

Resonantly or parametrically pumps energy into the laser pulse by periodic (self-) modulation of some parameter

Active mode-locking

Passive mode-locking

Fast saturable absorber as an amplitude modulator

Fast gain medium is a frequency modulator. It suppresses amplitude modulation

Picture from Rick Trebino’s lecture
Conditions for stable passive mode locking:

**Gain recovery time** \( T_1 \gg T_{RT} = 2nL_c/c \)

Gain should stay saturated below losses except the peak of the pulse when absorption is saturated.

Cross section for absorption > cross section for amplification.

Picture from Pietro Malara’s slide.
Passive mode locking works well in diode lasers

Rafailov Nature Phot. 2007
To achieve stable mode locking:

\[ \text{gain recovery time} \ > \ \text{roundtrip time} = 2nL_c/c \]

In QCLs this condition is not fulfilled.

Gain should stay saturated below losses except the peak of the pulse when absorption is saturated.

Picture from Pietro Malara’s slide.
For mode-locking, make sure that $\omega_M$ is close to mode spacing. This means that:

$$\omega_M = \frac{2\pi}{\text{cavity round-trip time}} = \frac{2\pi}{(2L/c)} = \frac{\pi c}{L}$$

Modes are locked into equidistant combs until dispersion moves them too far from resonance. For active mode locking fast gain recovery is not a problem; it even makes pulses more stable. Limitations on the power and duration?

Pictures from Rick Trebino slides
Active mode locking in a multi-section cavity

\[ P = P_0 + A \sin(2\pi ft) \]

Gain is modulated in a short section at the round-trip frequency \( f = 1/T_{RT} \)

2-Photon Autocorrelation shows 3-ps pulses

C. Wang et al. OE 2009

Mode locking existed only close to laser threshold
Insufficient gain modulation? Spatial hole burning? (Gkortzas et al., OE 2010)

Theory shows that AML should be possible in lasers with short gain recovery
time, both monolithic and external cavity

CW threshold including mirror losses

Modulated section
Active mode locking of THz QCLs

Effectively, modulation of only a small part of the cavity?

Freeman et al. APL 2012
Advantages of the AML in external cavity QCLs

The whole laser chip is modulated!
Deep gain modulation; robust AML operation over the whole dynamic range

Modulation frequencies are reduced from GHz to MHz.
Easier to handle; low-cost instrumentation

Easy to incorporate additional elements inside the cavity

Revin et al., Nature Comm. 7, 11440 (2016)
The periodic emission pulses (~3 ns long and limited by the speed of the detector) are observed if the QCL is driven near the fundamental frequency (~80.7 MHz, ~12.4 ns) of the round trip time.

Revin et al., Nature Comm. 7, 11440 (2016)
Performance of a ring cavity QCL

Maximum out-coupled averaged power is 2.9 mW (at least 10mW peak power).

Emission pulses exist through the whole dynamic range of the laser (up to laser roll-over).

Revin et al., Nature Comm. 7, 11440 (2016)
Collapse of the beatnote width from MHz to < 1 Hz

Single peak emission: sharp single peak RF spectra – high stability between relative phases of the modes – the modes are locked.

Multiple peaks emission: complicated broad RF spectra – much reduced phase stability. Broad emission: ~ 20 MHz wide pedestal – much reduced phase stability.

Revin et al., Nature Comm. 7, 11440 (2016)
Optical nonlinearity of a bulk semiconductor

Anharmonic oscillations of localized electrons

\[ \ddot{x}_k + \omega_k^2 x_k + \beta x_k^2 + \ldots \approx \frac{e}{m} E_0 e^{-i\omega t} \]

For electron displacement \( a \) and binding energy \( U_b \sim 5-10 \text{ eV} \),

\[ P = \frac{1}{V} \sum e x_k = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \]

\[ \frac{P^{(2)}}{P^{(1)}} \approx \frac{\chi^{(2)} E^2}{\chi^{(1)} E} \sim \frac{eaE}{U_b} \sim \frac{E}{E_{at}} \]

Scales as work done by the field during one oscillation period divided by binding energy

\( \chi^{(2)} \sim 10^{-6} \text{ esu} \sim 10^{-10} \text{ m/V} \) in narrow-gap semiconductors

\( \chi^{(2)} \sim 10^{-7} \text{ esu} \sim 10^{-11} \text{ m/V} \) in standard nonlinear crystals

Typical \( \chi^{(3)} \sim 10^{-12} - 10^{-15} \text{ esu} (10^{-20} - 10^{-23} \text{ m}^2/\text{V}^2) \)
Resonant nonlinearities by design

Coupled quantum well structures can be designed to have huge resonant optical nonlinearity (known for 30 years)

\[ |\chi^{(2)}| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)} \]

- \( \Delta_{ij} \) – detunings
- \( \gamma_{ij} \) – linewidths
- \( d_{ij} \) – dipole moments

\[ |\chi^{(2)}| \sim 10^4 - 10^6 \text{ pm/V} \]

Compare with 1-100 pm/V for bulk crystals
A way to get around resonant absorption

Resonant optical nonlinearity is accompanied by resonant absorption

\[ |\chi^{(2)}| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)} \]

Solution: resonant nonlinear medium with gain

This leads to nonlinear semiconductor lasers

Belyanin et al. PRA 2001
Quantum-cascade lasers with resonant optical nonlinearities

- Maximizing the product of dipoles $d_{23}d_{34}d_{24}$
- Quantum interference between cascades I and II

$\chi^{(2)} \sim 10^5 \text{ pm/V at } \sim 7-9 \mu\text{m laser wavelength}$

This is NOT sequential photon absorption/reemission

PRL 2003, APL 2004

Milliwatt power in SHG:
O. Malis et al. EL 2004
Collaboration with F. Capasso and C. Gmachl
Room-temperature THz injection laser based on difference frequency generation in mid-IR QCLs

- Powerful mid-IR QCL emitting at two modes
- Strong nonlinearity for frequency mixing process
- Low-loss, phase-matched waveguide for all three modes

Subsequent development by Belkin, Razeghi et al.
Similar ideas developed for interband diode lasers
Conclusions

• Saturation nonlinearity of the gain transition limits laser power, gives rise to multimode generation, and couples laser modes to enable frequency and phase locking
• Saturable gain suppresses amplitude modulation and leads to a nearly FM output. However, amplitude modulation exists
• Frequency combs
• Harmonic frequency combs
• Coherent intracavity current at beatnote frequency and its harmonics
  – Laser radio
  – Generation of THz and sub-THz radiation
• Generation of mode-locked pulses
  – Passive mode locking still seems to be impossible
  – Active mode locking has been achieved in both monolithic and external-cavity lasers