Resonant nonlinear optics in coupled quantum wells: from lasers to detectors

Alexey Belyanin
Texas A&M University

A. Wojcik, F. Xie, Y. Cho, V.R. Chaganti  
J. Kono  
Fow-Sen Choa

TAMU  
Rice  
UMBC
Outline

Introduction: electronic nonlinearity in coupled quantum wells

Frequency up-conversion with intersubband transitions

• Second harmonic generation in high band offset heterostructures
  - Integration with short-wavelength QCLs
  - SHG at wavelengths 1.5-2 μm?

• Frequency up-conversion infrared photodetectors
  - Resonant sum-frequency generation utilizing interband and intersubband transitions
  - Figures of merit
  - Normal incidence up-conversion detectors
  - Intersubband SFG detection in high band offset materials
Asymmetric coupled quantum wells: an ideal medium for nonlinear optics

Main advantage: control of the nonlinear optical response by engineering the shape (symmetry) of envelope functions.

Extreme flexibility and wavelength agility

Sharp atomic-like lines;
No cross-absorption

High optical nonlinearities: \( \chi^{(2)} \sim 10^6 \text{ pm/V}, \text{ Raman gain } \sim 10^{-4} \text{ cm/W} \)

Theory: see e.g. the review by Khurgin in Semiconductors and Semimetals V. 59;
Also: Ch. 5,6 in Intersubband Transitions in Quantum Structures, McGraw-Hill, 2006

Add advantages of a semiconductor medium: electron transport and Stark effect under applied voltage, integration with other components
A resonant nonlinear optics dilemma:
An enhancement in the nonlinearity is accompanied by such an increase in the optical absorption that it renders the nonlinearity useless.

(c.f. Murphy’s law)

Solutions:

Do nothing. This approach works nicely in gases (this fact has triggered LWI-EIT business), but not in semiconductors

Play with detuning, populations, and phase matching to make the nonlinear conversion length shorter than the absorption length

Compensate losses by gain  Integrate optical nonlinearity with a gain medium
Monolithic integration of resonant electronic nonlinearities with injection lasers

Laser field serves as an intracavity optical pump for the nonlinear process

Resonant enhancement of nonlinearity: resonant absorption is compensated by laser gain

The tightest possible confinement and mode purity

No problem with external pump; an injection-pumped device

Gmachl et al. 2003

Groups led by Capasso, Gmachl, Strasser

Most results are in QCLs, but there are a few in diode lasers.
Short-wavelength performance limitations of QCLs

From Tessier's talk:

<table>
<thead>
<tr>
<th></th>
<th>GaAs/AlAs</th>
<th>InGaAs/AlInAs</th>
<th>InGaAs/AlAsSb/InP</th>
<th>InAs/AlSb</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta \Gamma$, eV</td>
<td>1</td>
<td>0.9</td>
<td>1.6</td>
<td>2.1</td>
</tr>
<tr>
<td>$\Delta \Gamma-X_{\text{L}}$, eV</td>
<td>0.2</td>
<td>0.5</td>
<td>0.5</td>
<td>0.73</td>
</tr>
</tbody>
</table>

Lateral valleys may be a fundamental limitation to lasing at short wavelengths.

If so, only InAs/AlSb and may be GaN/AlN are suitable.
High band offset heterostructures

InAs/AlSb on InAs
Up to $\lambda \sim 2.7 \ \mu m$ (Univ. Montpellier)

Lattice-matched GaInAs/AlAsSb/InP
(Univ. Sheffield, Fraunhofer Inst.)

- SHG is not sensitive to the presence of lateral valleys or to the distance from the upper state to the top of the wells
- Full band offset is potentially available
- SHG needs a high-power laser and a phase-matched waveguide
All SHG lasers so far have been in lattice-matched GaInAs/AlInAs/InP or AlGaAs/GaAs structures

Shortest SH wavelength: 3.75 µm

Gmachl et al. 2003
However, lasers in high band offset structures become more and more refined and powerful. This is especially true for longer wavelengths around 4 µm (see Monday’s talks).

APL 86, 131107 (2005)
Lattice matched $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}/\text{AlAs}_{0.56}\text{Sb}_{0.44}/\text{InP}$ QCL

Three QW active region
Slightly modified from Yang et al.

$\chi^{(2)} \sim 700$ pm/V for $N_3 = 10^{17}$ cm$^{-3}$ and 30 meV detuning

$2\gamma_{32} = 2\gamma_{43} = \gamma_{42} = 20$ meV, $E = 85$ kV/cm
Who would care about QCLs at telecom wavelengths?

Ultrashort carrier lifetime $T_1 \approx 1 \text{ ps}$

Cavity roundtrip time $T_c \approx 20-80 \text{ ps}$

Dephasing time $T_2 \approx 0.1-0.5 \text{ ps}$

$T_2 < T_1 << T_c$

An overdamped Class A laser?
Only for small-signal modulation and low power

All other solid-state and diode lasers are Class B:

$T_2 << T_c << T_1$
Simplest dynamics: relaxation oscillations

Solid-state lasers: Stable focus \[ \frac{T_1}{T_c} \gg 1 \]

QC-lasers: Stable node \[ \frac{T_1}{T_c} \ll 1 \]

Overdamped oscillations
Absence of relaxation oscillations?

Potential for high-speed modulation up to THz rates

Capasso et al. IEEE JQE 38, 511 (2002)
Nonlinear polarization at second harmonic:

\[
P(2\omega) = \frac{e^3 N_e E_{\text{z}}^2(\omega)}{\hbar^2} \left( \frac{z_{23} z_{34} z_{24}}{\Gamma_{42}} \left( \frac{n_3 - n_4}{\Gamma_{43}} + \frac{n_3 - n_2}{\Gamma_{32}} \right) \right)
\]

\[
\Gamma_{32} = \gamma_{32} + i(\omega_{32} - \omega)
\]

\[
\Gamma_{42} = \gamma_{42} + i(\omega_{42} - 2\omega)
\]

Need \( \Delta \sim 2-3\gamma \)

\[
\frac{\sigma_g}{\sigma_{\text{abs}}} \sim \frac{d_{23}^2}{d_{34}^2} \left( 1 + \frac{\Delta^2}{\gamma^2} \right) \sim 5 - 10
\]

Factors limiting \( \chi^{(2)} \):
- Product of dipole moments
- Broadenings \( \gamma_{ij} \)
- Detuning from resonances

Factors affecting lasing:
- Resonant absorption 3-4;
- Depletion of upper laser state 3;
- Nonlinear depletion of laser mode
SHG efficiency

For second order nonlinearity \( P_{NL} \sim \chi^{(2)} E^2_p \)

\[
W_s \sim \frac{32\pi^5 W_p^2}{\int \frac{\chi^{(2)}(z)}{\varepsilon^2(z, \omega_p)} H^2_p(y, z) H_s(y, z) dy dz} \left| L_{coh} \right|^2
\]

Coherence length mainly limited by phase mismatch
Without phase matching: \( \Delta k_x \sim 2000 \text{ cm}^{-1} \)

Large \( \chi^{(2)} \), phase matching, and large nonlinear overlap are crucial for high efficiency

- Off-axis or surface emission

Modal phase matching seems to be most promising at this point
Waveguide design for phase-matched second-harmonic generation

$\text{TM}_{00} (\omega_1) + \text{TM}_{00} (\omega_1) \rightarrow \text{TM}_{02} (2\omega_1)$

2 mW power, 35 mW/W² efficiency (> 1 W/W² theoretical)

“Exact” phase-matching using ridge-width dependence of the propagation constants

Anti-Stokes Raman injection laser for tunable frequency up-conversion

Laser pump stage

Raman stage

- More powerful than SHG
- No phase matching is needed
- Has threshold; may compromise the pump laser

Gain \sim \text{Re} \left\{ \frac{\omega_z z_{24}^2 z_{34}^2}{\gamma_{24} + \left| \Omega_p \right|^2 / \gamma_{23} + i\Delta} \left[ \frac{|E_p|^2 n_{34}}{\hbar^2 \gamma_{23} (\gamma_{34} - i\Delta)} - n_{24} \right] \right\}

APL 87, 26113 (2005): antistokes Raman emission but no lasing
Coherent up-conversion detection of the mid/far-IR radiation using intersubband-interband electronic nonlinearity
Mid/far-IR semiconductor photodetectors

- Good performance, especially when used with high-power signals (QC lasers)
- High dark current due to thermal excitations
  \[ \exp \left( -\frac{E_2 - E_1}{kT_D} \right) \]
- Cooling is required for high detectivity
- Even at BLIP conditions the detectivity is limited due to exponentially growing background blackbody radiation
- No single photon counting
Frequency up-conversion to the visible/near-IR range

Developed since 1960s: Midwinter, Townes, Boyd, …

- Superior visible/near-IR detectors: avalanche photodiodes and photomultipliers
- Low dark counts and background noise
- Room-temperature operation
- Single photon counting in the mid-IR (Temporao et al., EL 2006)

Low up-conversion efficiency: $\sim 10^{-6}$; Need to use high-power pulsed lasers
Some related concepts and devices

Integrated QWIP and LED
(H.C. Liu et al., Ryzhii et al.)

This is an incoherent process:

- Mid-IR signal $\rightarrow$ photocurrent $\rightarrow$
- spontaneous Near-IR emission $\rightarrow$
- photocurrent

Vagos et al. 1993 – double photon absorption followed by Near-IR PL

Generation of mid-infrared (Phillips, today’s talk) and THz (Barbieri, next talk) sidebands on a near-IR optical carrier in a QCL

Both are coherent nonlinear mixing processes
Coherent up-conversion detection in atomic vapors
Harris; Boyd et al.; Boyd & Scully

Promise to achieve much higher up-conversion efficiency (at the expense of narrow spectral responsivity)

Four-wave mixing;
Needs two powerful lasers;
Sharp resonance
Coherent up-conversion detection in coupled QWs:

• Hoping to retain attractive properties of up-conversion in more efficient, compact devices
• Coherent phase-sensitive process; unique applications in the mid-far/IR are enabled by single photon counting

1. Nonlinearity and spectral response
2. Geometry
3. Figures of merit
4. Various device designs

Many studies of electronic nonlinearities in QWs at mixed interband/intersubband transitions: see e.g. Khurgin (1990s), Neogi et al. (1998) for SFG studies
1. Nonlinearity. (a) Interband-intersubband cascade

A stack of undoped double QWs

Example: In$_{0.53}$Ga$_{0.47}$As/In$_{0.52}$Al$_{0.48}$As

1.3-µm pump + 12-µm signal $\rightarrow$ 1.15-µm SFG

In GaAs-based structures: SFG at ~ 0.7 µm

$d_{v,m-c,n} \sim \frac{ie\hbar p_{vc}}{m_0 E_g} \langle m|n \rangle$ Asymmetry needed

6-band VB + CB with energy-dependent mass

$d_{hh1-e1} = 7$ Å, $d_{hh1-e2} = 1.5$ Å, $d_{e1-e2} = 25$ Å
Choosing the right structure

Example: In\textsubscript{0.53}Ga\textsubscript{0.47}As/In\textsubscript{0.52}Al\textsubscript{0.48}As

Total thickness 90A, barrier thickness=10A

Product of dipole moments vs well asymmetry \((w_1-w_2)/\text{total thickness}\)

Energy difference vs well asymmetry

Choosing the right structure

Example: In\textsubscript{0.53}Ga\textsubscript{0.47}As/In\textsubscript{0.52}Al\textsubscript{0.48}As

Total thickness 90A, barrier thickness=10A

Product of dipole moments vs well asymmetry \((w_1-w_2)/\text{total thickness}\)

Energy difference vs well asymmetry
Calculating $\chi^{(2)}$ and spectral response

$$\chi^{(2)}\left(\omega_{up} = \omega_s + \omega_p\right) \sim \frac{1}{2\pi L_{qw}} \int dk^2 \frac{d_{vc1} d_{vc2} d_{c1c2} (f_v(k) - f_c(k))}{\left(E_{vc2}(k) - \hbar \omega_{up} - i\hbar \gamma_{vc2}(k)\right)\left(E_{vc1}(k) - \hbar \omega_p - i\hbar \gamma_{vc1}(k)\right)}$$

Assuming $k$-independent $\gamma$'s and $d$'s, parabolic bands, equal masses $m_{c1}$ and $m_{c2}$, $f_v = 1$, $f_c = 0$,
\[
\chi^{(2)}(\omega_{up} = \omega_s + \omega_p) \sim d_{v1c1} d_{v2c2} \frac{m_r}{\pi \hbar^2 L_{qw}} F(\Delta_{up}, \Delta_p, \gamma_{v1c1}, \gamma_{v2c2})
\]

\[
\Delta_p = E_{v1-c1} - \hbar \omega_p, \quad \Delta_{up} = E_{v1-c2} - \hbar \omega_{up}
\]

|\chi^{(2)}|, nm/V

-\Delta_{up}, meV

\[|\chi^{(2)}|^2 \text{ bandwidth of } \sim 35 \text{ meV for a } 100-\text{meV mid-IR photon energy}\]

Need detuning of the pump below band gap to minimize optical pumping

May need detuning of the SFG frequency from transition hh1-c2 to minimize spontaneous recombination noise
Possible detection geometry for hh1-ee1-ee2 nonlinearity

Automatic phase-matching for various mid-IR beam directions and inputs: waveguiding, wedge, etc.

Directions of the OPA pump $k_2$ and the up-converted signal $k_3$ are separated

Mid-IR signal

Ez

z

x

y

Ez NIR pump

Variable spot size ~ 10-100 µm

$\text{SI sub}$

APD

$\vec{k}_{2\text{vac}}$

$\vec{k}_{3\text{vac}}$

$\vec{k}_2$

$\vec{k}_3$

$\vec{k}_1$

$z_{\text{MQW}}$

$k(\omega_1 + \omega_2) > k(\omega_1) + k(\omega_2)$

No need to phase-match in z-direction

$\vec{k}_{1x} = \vec{k}_{3x}$

$(\vec{k}_{3z} - \vec{k}_{1z} - \vec{k}_2)z_{\text{MQW}} \ll 1$

Always possible, since $k_1 \ll k_2$
Figures of merit

Up-conversion efficiency:

\[ P_{SFG} = \eta_{up} P_s \]

Responsivity:

\[ R = \frac{I_{NIR}}{P_{s}} = \eta_{up} R_{NIR} \]

Single photon conversion efficiency:

\[ \eta_{phot} = \eta_{up} \frac{\omega_{s}}{\omega_{SFG}} \eta_{NIR} \sim 0.01 \eta_{up} \]

Detectivity \( D^* = D^*_{NIR} \), if spontaneous emission power from the sample at SFG frequency and within acceptance angle is below NEP of the NIR detector:

\[ \text{NEP}_{NIR} > \frac{\Delta \Omega}{4\pi} P_{spon}(\omega_{SFG}) \]
\( \eta_{up} \): in general, a 3D electrodynamics problem

Neglecting pump absorption and nonlinear depletion, and for plane waves (or broad beams):

\[
\eta_{up} \approx \frac{32\pi^5 |\chi^{(2)}|^2}{\lambda_{SFG}^2 c \mu_s \mu_p \mu_{SFG} L_z L_y} \frac{z_M{QW}^2}{\Gamma_s I_p} \sim 5.5 \times 10^{-4} \Gamma_s I_p (\text{W} / \text{cm}^2)
\]

\[
\Gamma_s = \frac{z_M{QW}^2}{L_z L_y} \sim 10^{-2} \div 10^{-4}
\]

Maximum \( z_{MQW} \) is limited by SFG signal absorption to \( \sim 50 \) periods

Interband absorption per well:

\[
\alpha L_{qw} \approx 2 \frac{e^2}{\hbar c} \frac{m_r E_p}{m_0 E_g} |\langle m | n \rangle|^2 \sim 1.5-2\% \text{ for a strong allowed transition (hh1-e1)}
\]

\( \sim 1 \)
Pump absorption and spontaneous recombination noise

Pump frequency is tuned 20-30 meV below band gap.
No simple formula for $\alpha_{\text{pump}}$, but empirically it does not limit $z_{\text{MQW}}$

However, it limits pump intensity $I_p$!

$$\text{NEP}_{\text{NIR}} > \frac{\Delta \Omega}{4\pi} P_{\text{spon}}(\omega_{\text{SFG}})$$

$$P_{\text{spon}} \sim R_{31} n_{e2} n_{hh1} \hbar \omega V_{\text{MQW}}$$

$$n_{e2} \sim n_{e1} e^{-\frac{E_{e2} - E_{e1}}{k_B T}} \text{ in equilibrium.}$$

$$\alpha_{\text{pump}} I_p / \hbar \omega_p = n_{e1} / \tau_{e1}$$

$\text{NEP}_{\text{NIR}}$ can be extremely low: $\sim 10^{-14}$ W, even at telecom wavelengths
(e.g. InGaAs APD, IEEE JQE 42, 397 (2006))

Even better performance at higher photon energies in GaAs-based nonlinear structures

$I_p$ limited to $\sim 10^5$ W/cm$^2$; $\eta_{\text{up}} \sim 0.1$, $\eta_{\text{phot}} \sim 10^{-3}$ in the mid-infrared
Using intersubband transitions in valence band for normal incidence detection

\[ \text{In}_{0.53} \text{Ga}_{0.47}\text{As/In}_{0.52}\text{Al}_{0.48}\text{As} \]

\[ \text{Lh}_2\text{-hh}_1\text{-e}_1\text{-lh}_2 \]

1.3-\(\mu\m\) pump + 12-\(\mu\m\) signal \(\rightarrow\) 1.15-\(\mu\m\) SFG

For in-plane polarized waves:

\(d_{\text{hh}_1\text{-e}_1} \sim 7 \text{ A}, d_{\text{hh}_1\text{-lh}_2} \sim 8 \text{ A}, d_{\text{lh}_2\text{-e}_1} \sim 0.6 \text{ A}\)

Lower nonlinearity, but simpler geometry
SFG at telecom wavelengths in lattice matched Ga$_{0.47}$In$_{0.53}$As/AlAs$_{0.56}$Sb$_{0.44}$/InP
Intersubband transitions only

\[
\begin{align*}
E_{\text{e}_1-\text{e}_3} &\sim E_{\text{e}_1-\text{e}_4} \sim 9 \text{ Å}, \quad E_{\text{e}_3-\text{e}_4} \sim 30 \text{ Å} \\
\end{align*}
\]

Now both NIR transitions are equally strong -> a 10 times higher $|\chi^{(2)}|^2$

Detuning provides low losses for all waves, including SFG

Highly efficient SFG ($\eta \sim 1$) is possible, but both pump and signal need to propagate along the QW layers.
Conclusions

• Intracavity SHG in QC lasers: frequency up-conversion into the 1.5-3.5 μm range

• Large nonlinearity; thresholdless (low-risk); should operate CW at room T when the pump laser operates that way

• Phase-matched waveguide is crucial for phase matching, and InP- based GaInAs/AlAsSb QC lasers seem to be particularly convenient for implementing SHG.

• SFG in coupled QW structures enables mid-IR detectors with decent efficiency, high detectivity and very low dark current with a relatively low-power CW laser pump.

• Potential for integration with semiconductor pump lasers