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

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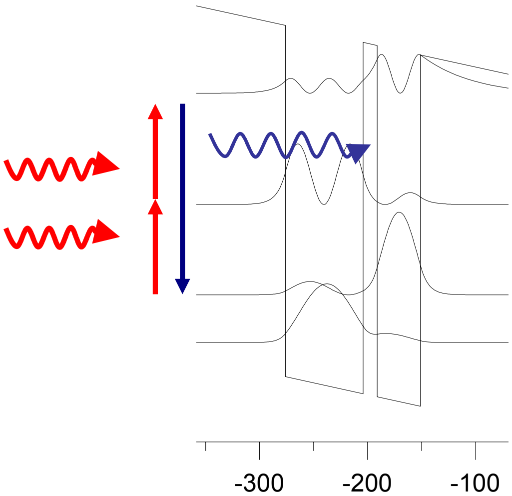
Outline

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: e.g. $\chi^{(2)}$ ~ 10^6 pm/V, Raman gain ~ 10^{-4} cm/W

Gurnick & De Temple 1983, Fejer et al. 1989; Sirtori et al. 1991, …
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)

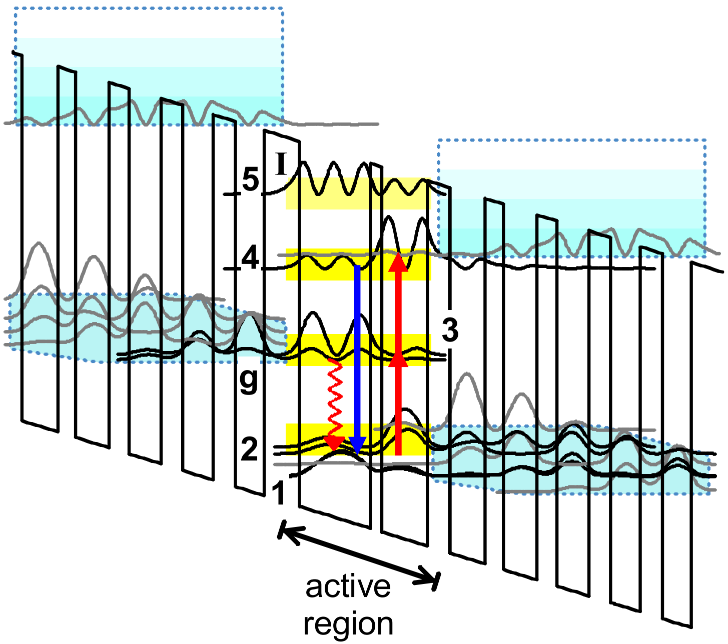
Solutions:

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



Gmachl et al. 2003

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

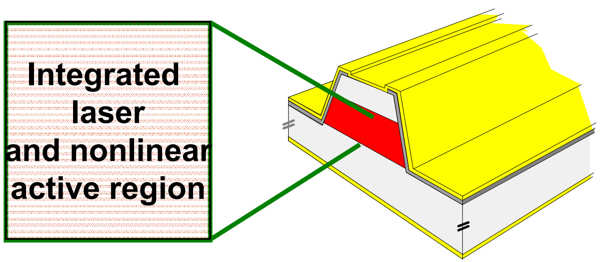
Resonant enhancement of nonlinearity: resonant absorption is compensated by laser gain

The tightest possible confinement and mode purity

No problem with external pump; an injection-pumped device

Groups led by Capasso, Gmachl, Strasser

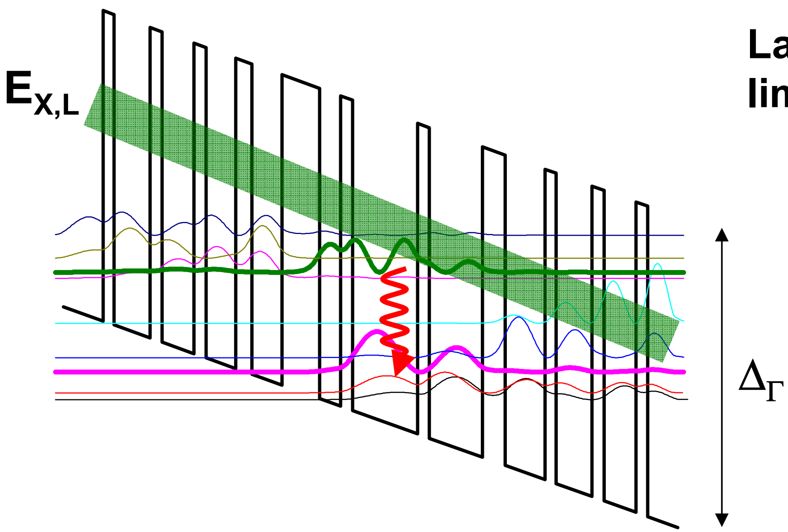
Most results are in QCLs, but there are a few in diode lasers.



Short-wavelength performance limitations of QCLs

From Tessier's talk:

	GaAs/AlAs	InGaAs/AlInAs	InGaAs/AlAsSb /InP	InAs/AlSb
Δ_Γ, eV	1	0.9	1.6	2.1
$\Delta_{\Gamma\text{-X,L}}$, eV	0.2	0.5	0.5	0.73



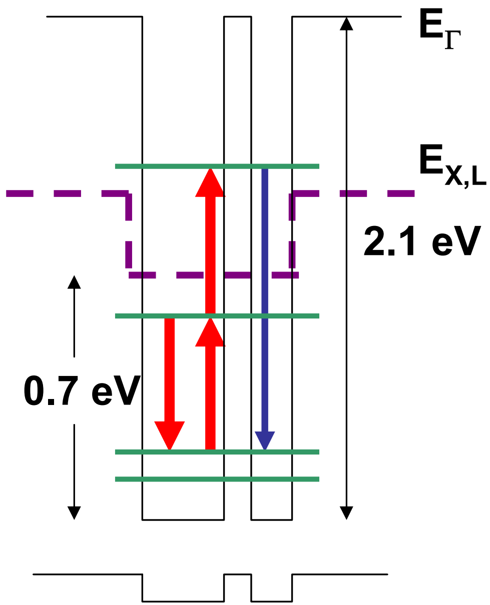
Lateral valleys may be a fundamental limitation to lasing at short wavelengths.

If so, only InAs/AlSb and may be GaN/AlN are suitable

High band offset heterostructures

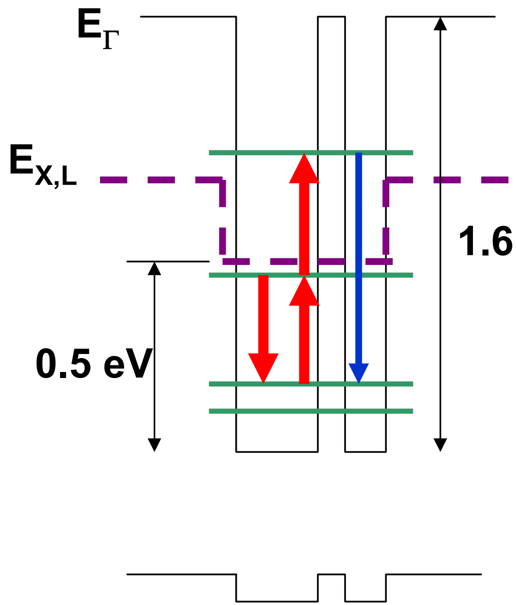
InAs/AlSb on InAs

Up to λ ~ 2.7 μm (Univ. Montpellier)



Lattice-matched GaInAs/AlAsSb/InP

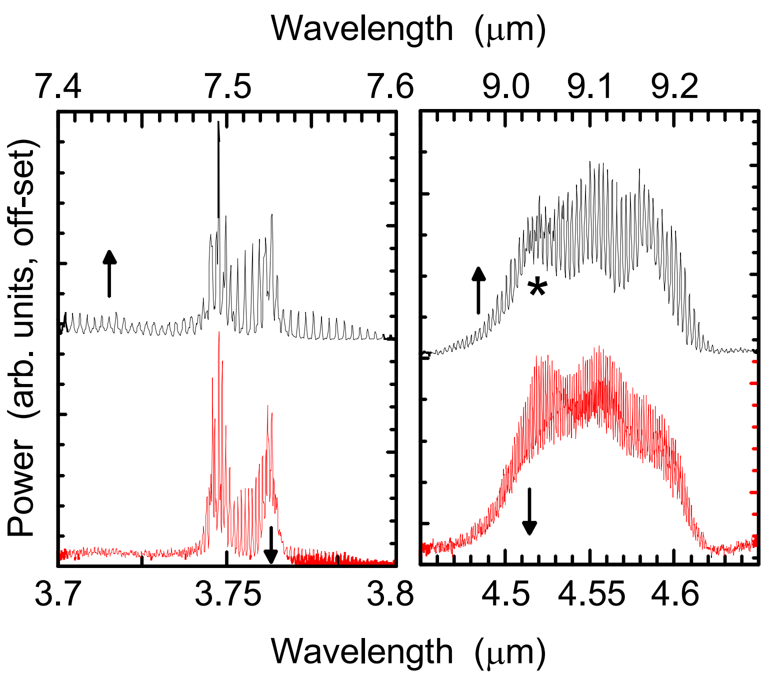
(Univ. Sheffield, Fraunhofer Inst.)



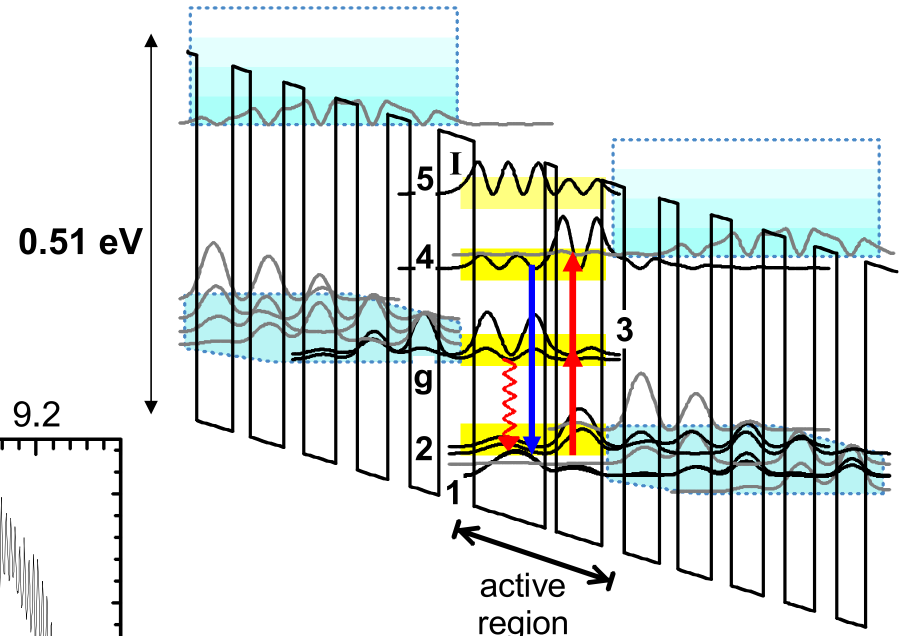
- SHG is not sensitive to the presence of lateral valleys or to the distance from the upper state to the top of the wells
- Full band offset is potentially available

- 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

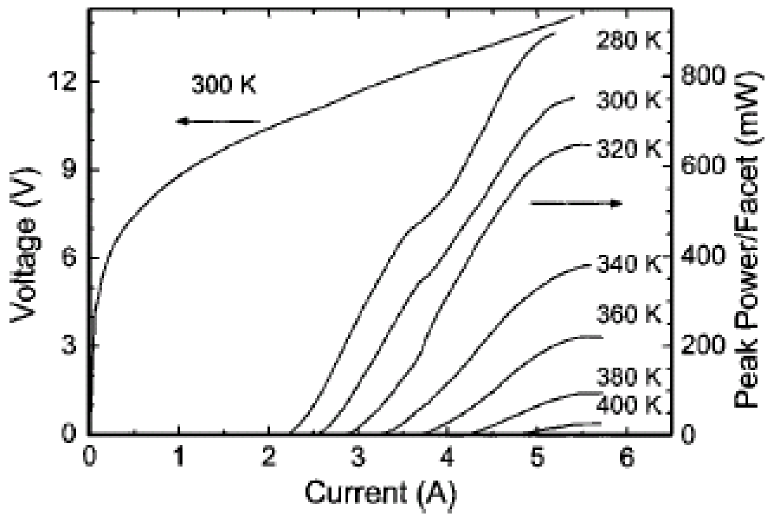


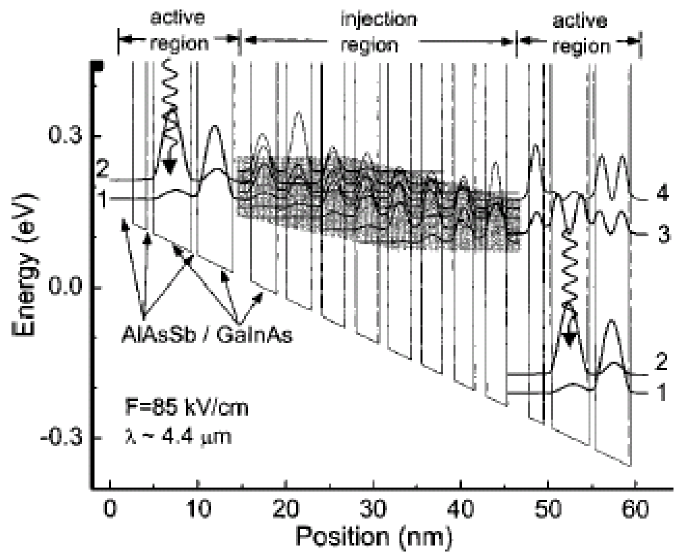
Gmachl et al. 2003



Shortest SH wavelength: 3.75 μm

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).



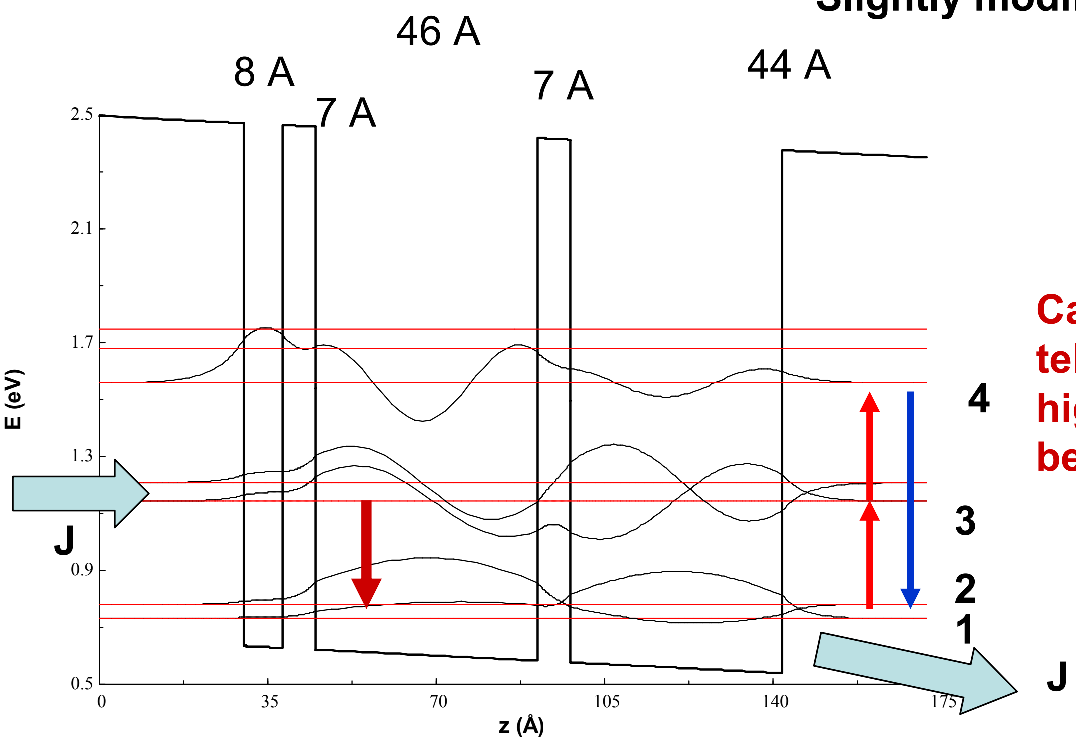


APL 86, 131107 (2005)

Lattice matched $Ga_{0.47}In_{0.53}As/AlAs_{0.56}Sb_{0.44}/InP$ QCL

Three QW active region

Slightly modified from Yang et al.



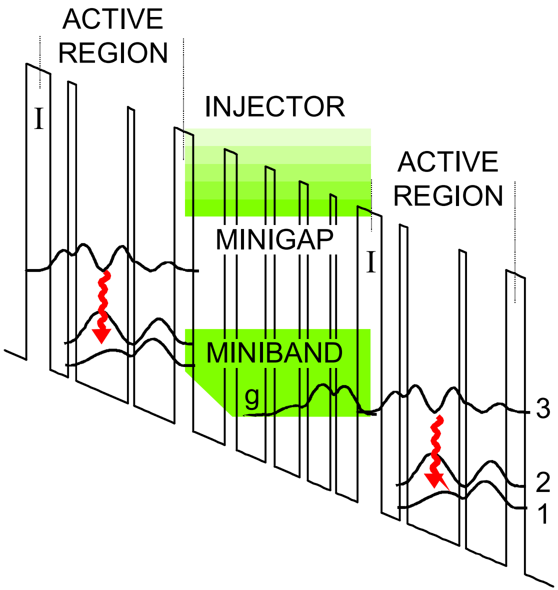
$\lambda(\omega) \sim 3.8\ \mu m$
$\lambda(2\omega) \sim 1.9\ \mu m$

Can be easily scaled to telecom wavelengths if high-power lasers at 3 μm become available!

d_{23} = 20 A
d_{24} = 0.6 A
d_{34} = 11 A

$\chi^{(2)} \sim 700$ 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~ 1 ps

Cavity roundtrip time T_c ~ 20-80 ps

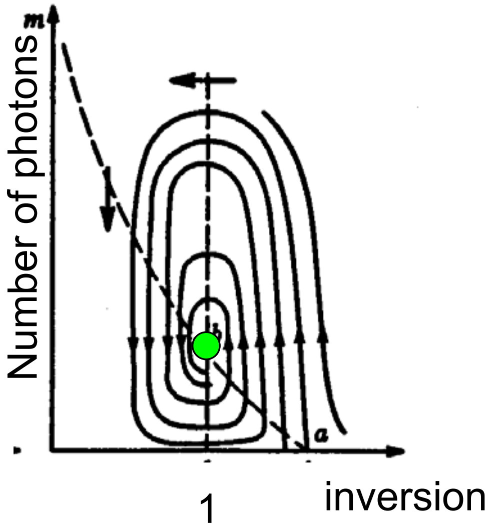
Dephasing time T_2 ~ 0.1-0.5 ps

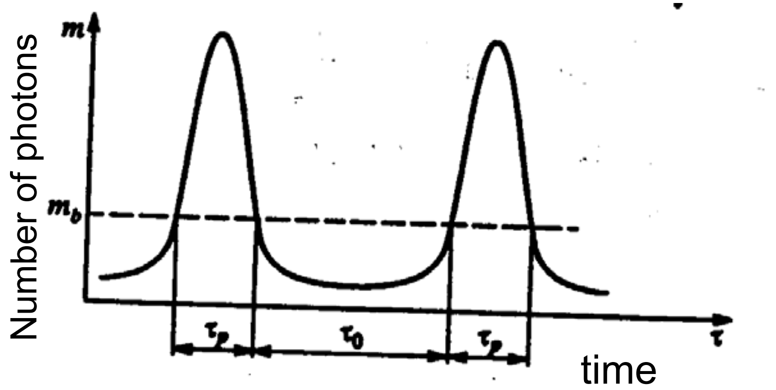
$T_2 < T_1 << T_c$

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

All other solid-state and diode lasers are Class B:
$T_2 << T_c << T_1$

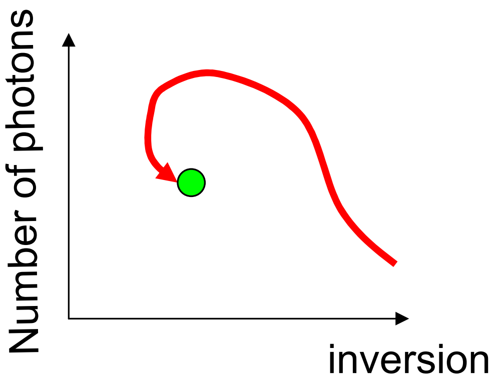
Simplest dynamics: relaxation oscillations





Solid-state lasers:
Stable focus

$$\frac{T_1}{T_c} >> 1$$



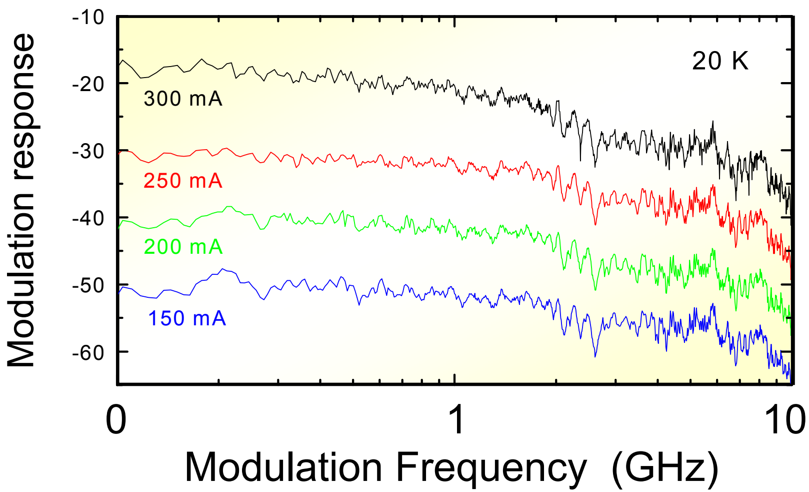
QC-lasers:
Stable node

$$\frac{T_1}{T_c} << 1$$

Overdamped oscillations

Absence of relaxation oscillations?

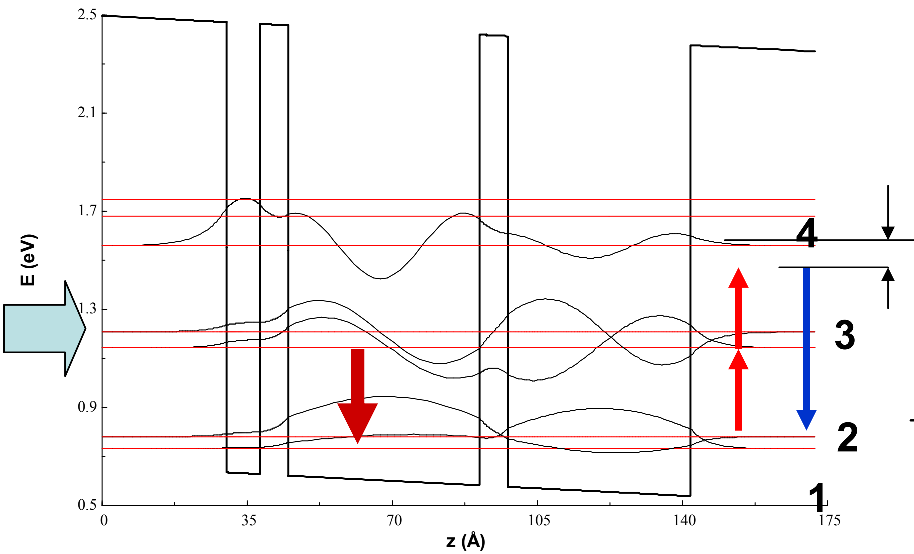
Potential for high-speed modulation up to THz rates



Capasso et al. IEEE JQE 38, 511 (2002)

Nonlinear polarization at second harmonic:

$$P(2\omega)=\frac{e^3 N_e \mathrm{E}_z^2(\omega)}{\hbar^2}\left(\frac{z_{23}z_{34}z_{24}}{\Gamma_{42}}\left(\frac{n_3-n_4}{\Gamma_{43}}+\frac{n_3-n_2}{\Gamma_{32}}\right)\right)$$



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

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

Need Δ ~ 2-3γ

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

Factors limiting $\chi^{(2)}$:
- **Product of dipole moments**
- **Broadenings γ_{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 $P_{NL} \sim \chi^{(2)} E_p^2$

$$W_s \sim \frac{32\pi^5 W_p^2 \left| \int \frac{\chi^{(2)}(z)}{\varepsilon^2(z,\omega_p)} \mathrm{H}_p^2(y,z)\mathrm{H}_s(y,z)\, dydz \right|^2 \left| L_{coh} \right|^2}{\mu_p^2 \mu_s c \lambda_s^2}$$

"p" – pump

"s" - signal

$$\left| L_{coh} \right| \sim 1 / \left| k_s - 2k_p \right|$$

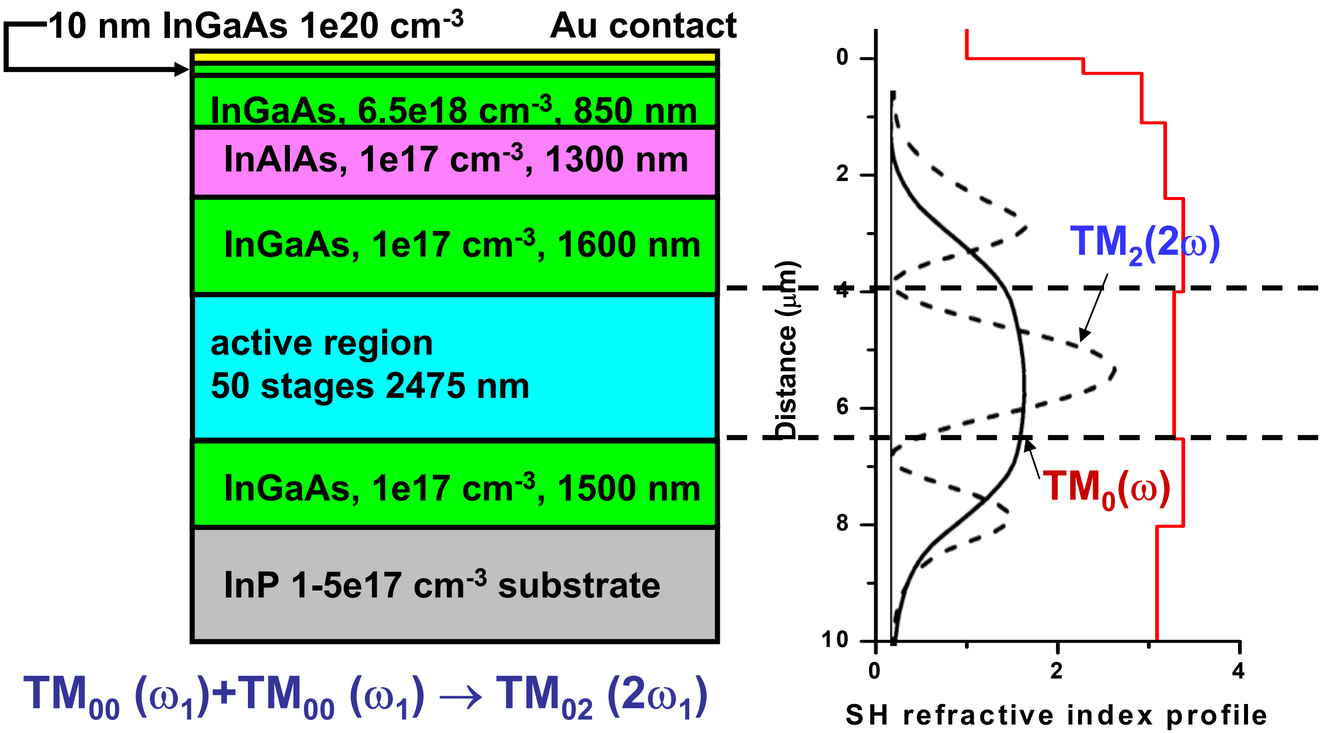
Coherence length mainly limited by phase mismatch
Without phase matching: $\Delta k_x \sim 2000$ cm^{-1}

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

- **Quasi phase matching by periodic Stark shift: APL 88, 201108 (2006)**
- **Modal phase matching: APL 84, 2751 (2004), EL 40 (2004)**
- **Off-axis or surface emission**

Modal phase matching seems to be most promising at this point

Waveguide design for phase-matched second-harmonic generation

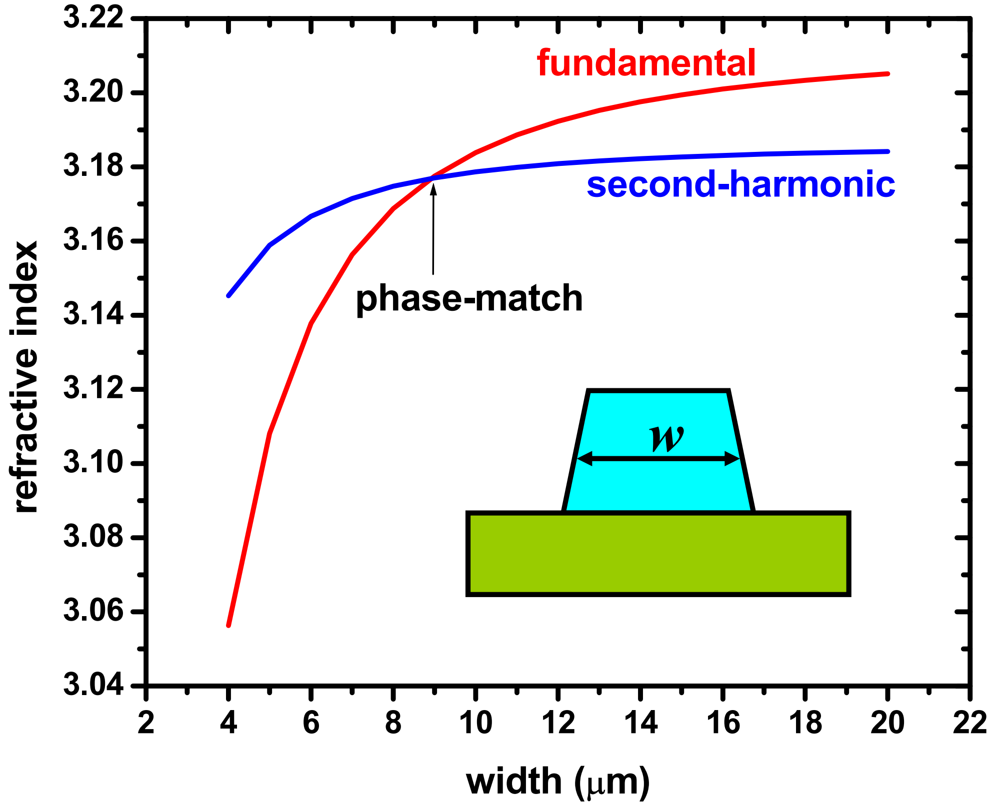


$TM_{00}(\omega_1) + TM_{00}(\omega_1) \rightarrow TM_{02}(2\omega_1)$

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

O. Malis et al., Electron. Lett. 40, 1586 (2004)

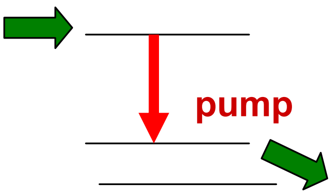
"Exact" phase-matching using ridge-width dependence of the propagation constants



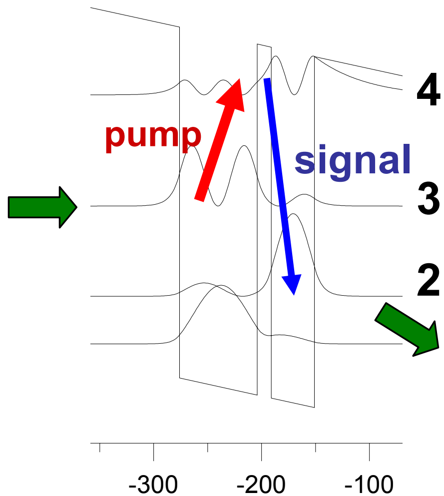
O. Malis et al., Electron. Lett. 40, 1586 (2004)

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

Laser pump stage



Raman stage

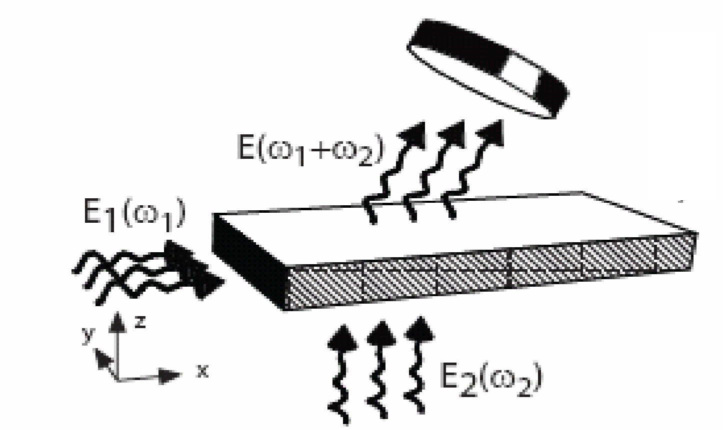


- **More powerful than SHG**
- **No phase matching is needed**
- **Has threshold; may compromise the pump laser**

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

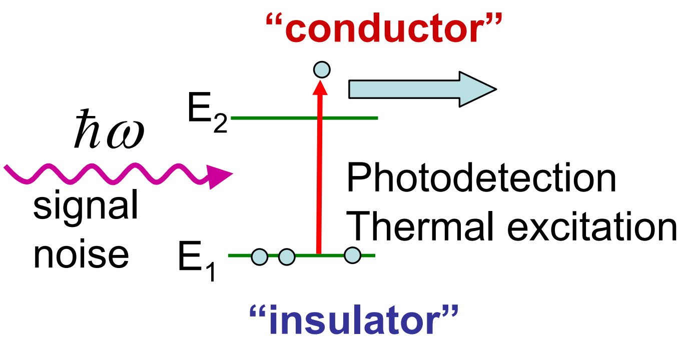
APL 87, 26113 (2005): antistokes Raman emission but no lasing

Coherent up-conversion detection of the mid/far-IR radiation using intersubband-interband electronic nonlinearity



Mid/far-IR semiconductor photodetectors

Interband, intersubband, or impurity transition

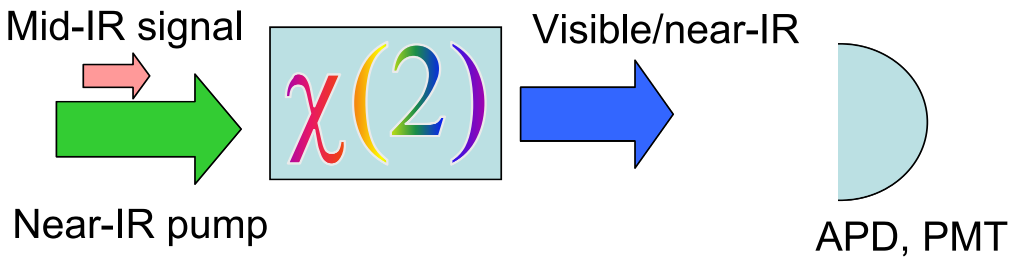


$$E_2 - E_1 \le \hbar\omega$$

- Good performance, especially when used with high-power signals (QC lasers)
- High dark current due to thermal excitations $\sim \exp\left(-\frac{E_2 - E_1}{kT_D}\right)$
- 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

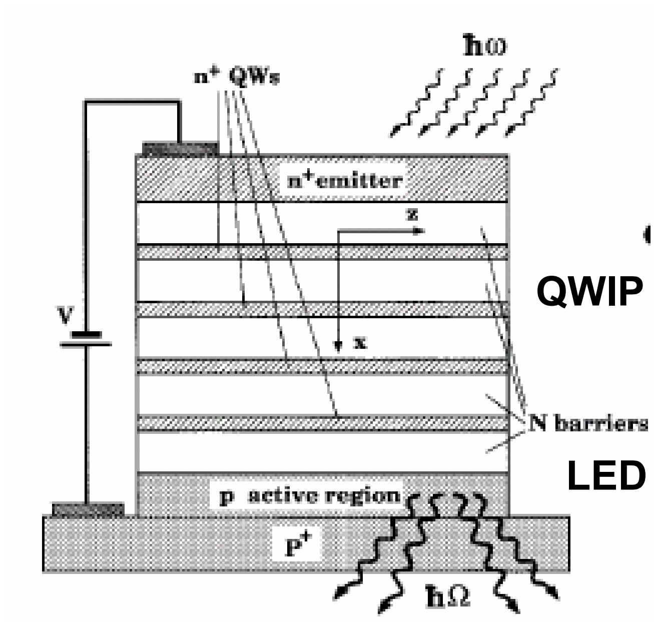
Developed since 1960s: Midwinter, Townes, Boyd, …



- 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: ~ 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 ->photocurrent-> spontaneous Near-IR emission -> photocurrent

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

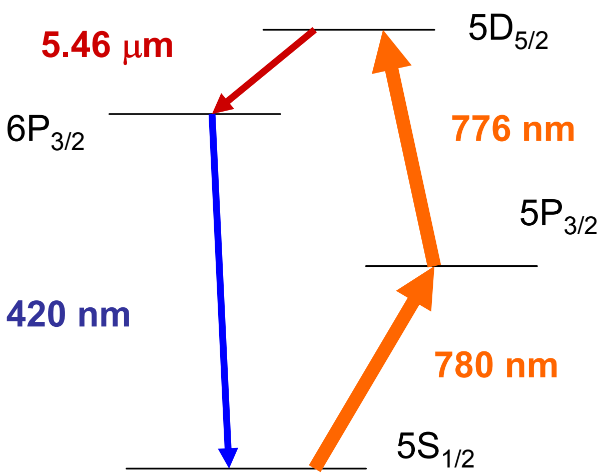
Generation of mid-infrared (Phillips, today's talk) and THz (Barbieri, next talk) sidebands on a near-IR optical carrier in a QCL

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**

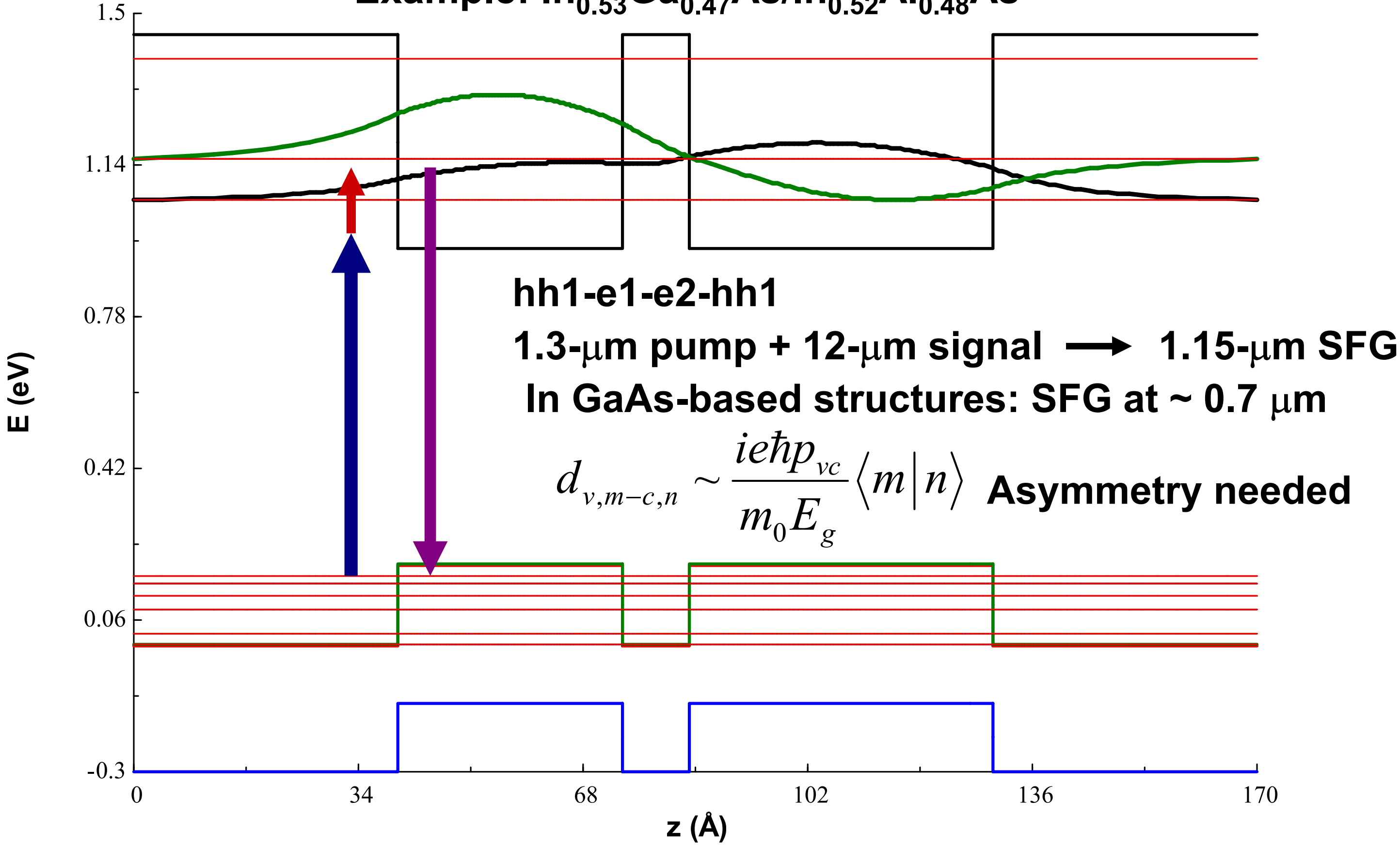
1. **Nonlinearity and spectral response**
2. **Geometry**
3. **Figures of merit**
4. **Various device designs**

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: $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$



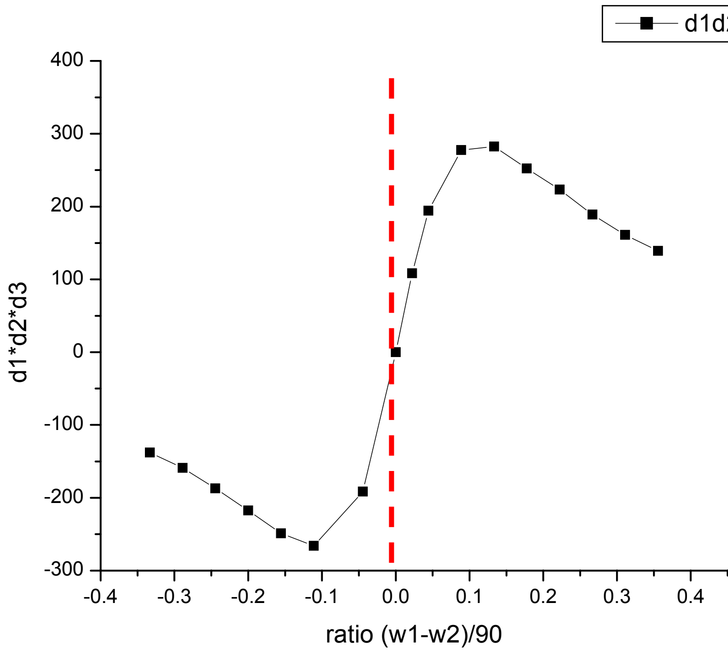
6-band VB + CB with energy-dependent mass

$d_{hh1\text{-}e1}$ = 7 A, $d_{hh1\text{-}e2}$ = 1.5 A, $d_{e1\text{-}e2}$ = 25 A

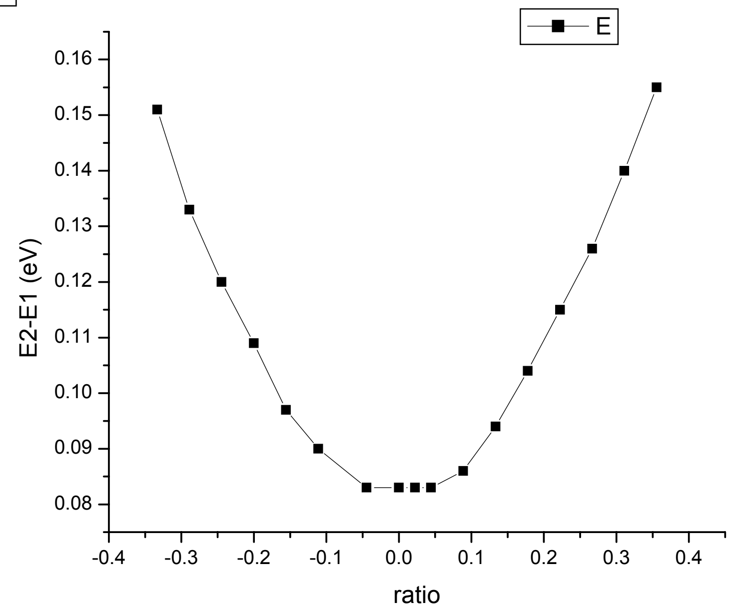
Choosing the right structure

Example: $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$

Total thickness 90A, barrier thickness=10A



Product of dipole moments vs well asymmetry ((w1-w2)/total thickness)

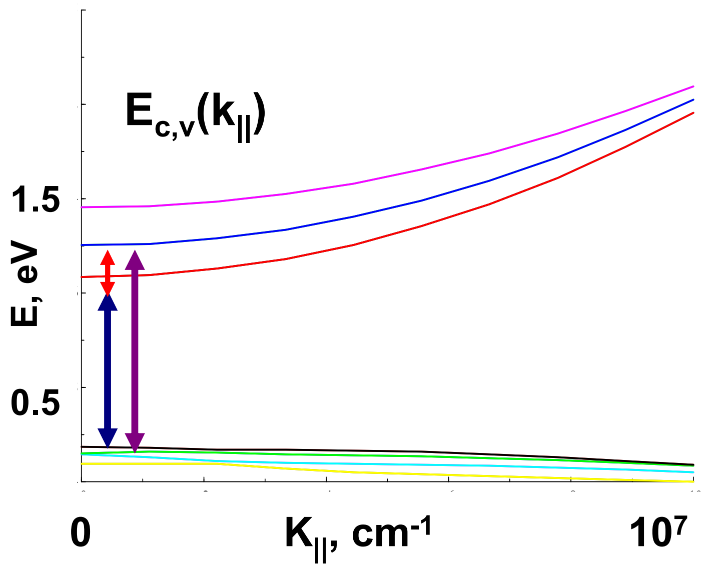


Energy difference vs well asymmetry

Calculating $\chi^{(2)}$ and spectral response

$$\chi^{(2)}\left(\omega_{up}=\omega_s+\omega_p\right)\sim$$

$$\frac{1}{2\pi L_{qw}}\int dk^2\,\frac{d_{vc1}d_{vc2}d_{c1c2}\left(f_{v1}(k)-f_{c1}(k)\right)}{\left(E_{vc2}(k)-\hbar\omega_{up}-i\hbar\gamma_{vc2}(k)\right)\left(E_{vc1}(k)-\hbar\omega_p-i\hbar\gamma_{vc1}(k)\right)}$$

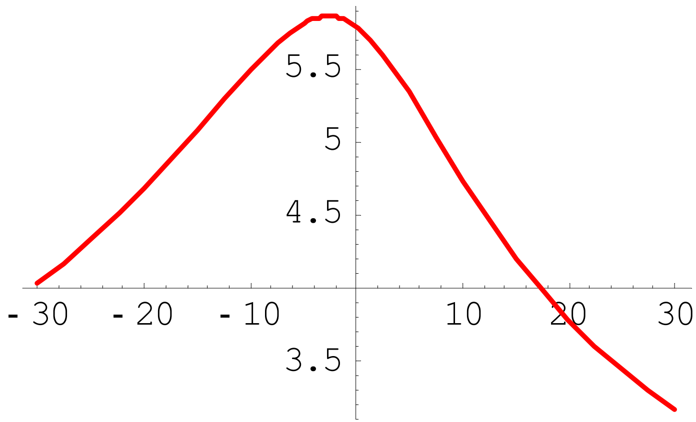


Assuming k-independent γ's and d's, parabolic bands, equal masses m_{c1} and m_{c2}, $f_v = 1$, $f_c = 0$,

$$\chi^{(2)}\left(\omega_{up}=\omega_s+\omega_p\right)\sim d_{vc1}d_{vc2}d_{c1c2}\frac{m_r}{\pi\hbar^2 L_{qw}}F(\Delta_{up},\Delta_p,\gamma_{vc1},\gamma_{vc2})$$

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

$|\chi^{(2)}|$, nm/V **Δ_p = 20 meV, γ = 10 meV**



Δ_{up}, meV

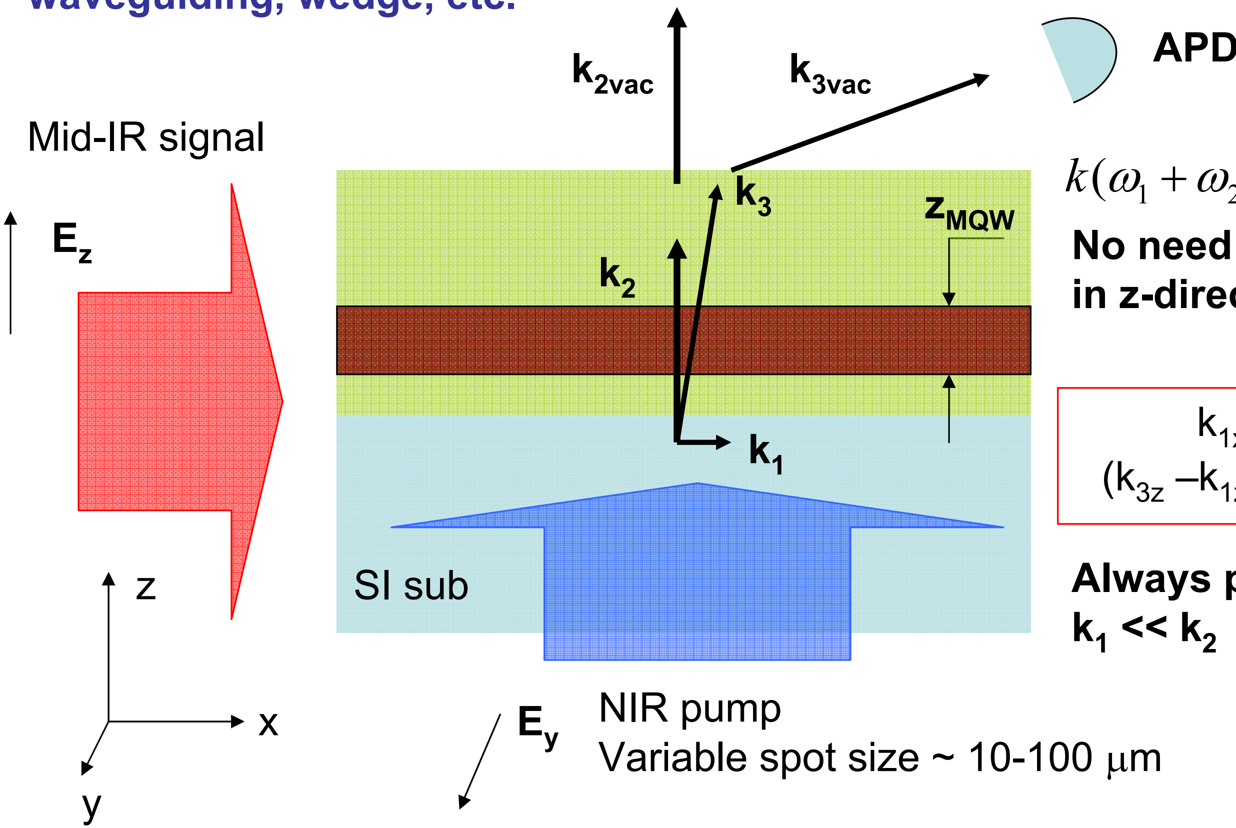
$|\chi^{(2)}|^2$ bandwidth of ~ 35 meV for a 100-meV mid-IR photon energy

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.



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

No need to phase-match in z-direction

$$k_{1x} = k_{3x}$$
$$(k_{3z} - k_{1z} - k_2)z_{MQW} << 1$$

Always possible, since $k_1 << k_2$

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

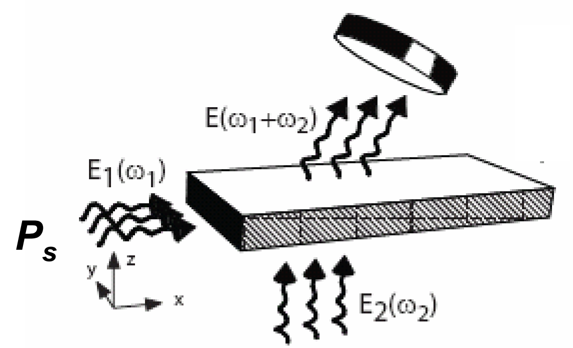
Figures of merit

Up-conversion efficiency:

$$P_{SFG} = \eta_{up} P_s$$

Responsivity: $$R = \frac{I_{NIR}}{P_s} = \eta_{up} R_{NIR}$$

R_{NIR}, D^*_{NIR}, η_{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:

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

η_{up}: in general, a 3D electrodynamics problem

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

$$\eta_{up} \sim \frac{32\pi^5 \left|\chi^{(2)}\right|^2}{\lambda_{SFG}^2 c \mu_s \mu_p \mu_{SFG}} \frac{z_{MQW}^2}{L_z L_y} I_p \sim 5.5\times 10^{-4}\,\Gamma_s I_p\,(\mathrm{W/cm^2})$$

$$\Gamma_s = \frac{z_{MQW}^2}{L_z L_y} \sim 10^{-2} \div 10^{-4}$$

Maximum z_{MQW} is limited by SFG signal absorption to ~ 50 periods

Interband absorption per well:

$$\alpha L_{qw} \approx 2\frac{e^2}{\hbar c}\frac{m_r}{m_0}\frac{E_p}{E_g}\left|\langle m|n\rangle\right|^2$$ **~ 1.5-2% for a strong allowed transition (hh1-e1)**

~ 1

Pump absorption and spontaneous recombination noise

Pump frequency is tuned 20-30 meV below band gap.

No simple formula for α_{pump} , but empirically it does not limit z_{MQW}

However, it limits pump intensity I_p!

$$\mathrm{NEP}_{NIR} > \frac{\Delta\Omega}{4\pi} P_{spon}(\omega_{SFG}) \qquad P_{spon} \sim R_{31} n_{e2} n_{hh1} \hbar\omega V_{MQW}$$

$$n_{e2} \sim n_{e1} e^{-\frac{E_{e2}-E_{e1}}{k_B T}}$$ **in equilibrium.** $$\alpha_{pump} I_p / \hbar\omega_p = n_{e1} / \tau_{e1}$$

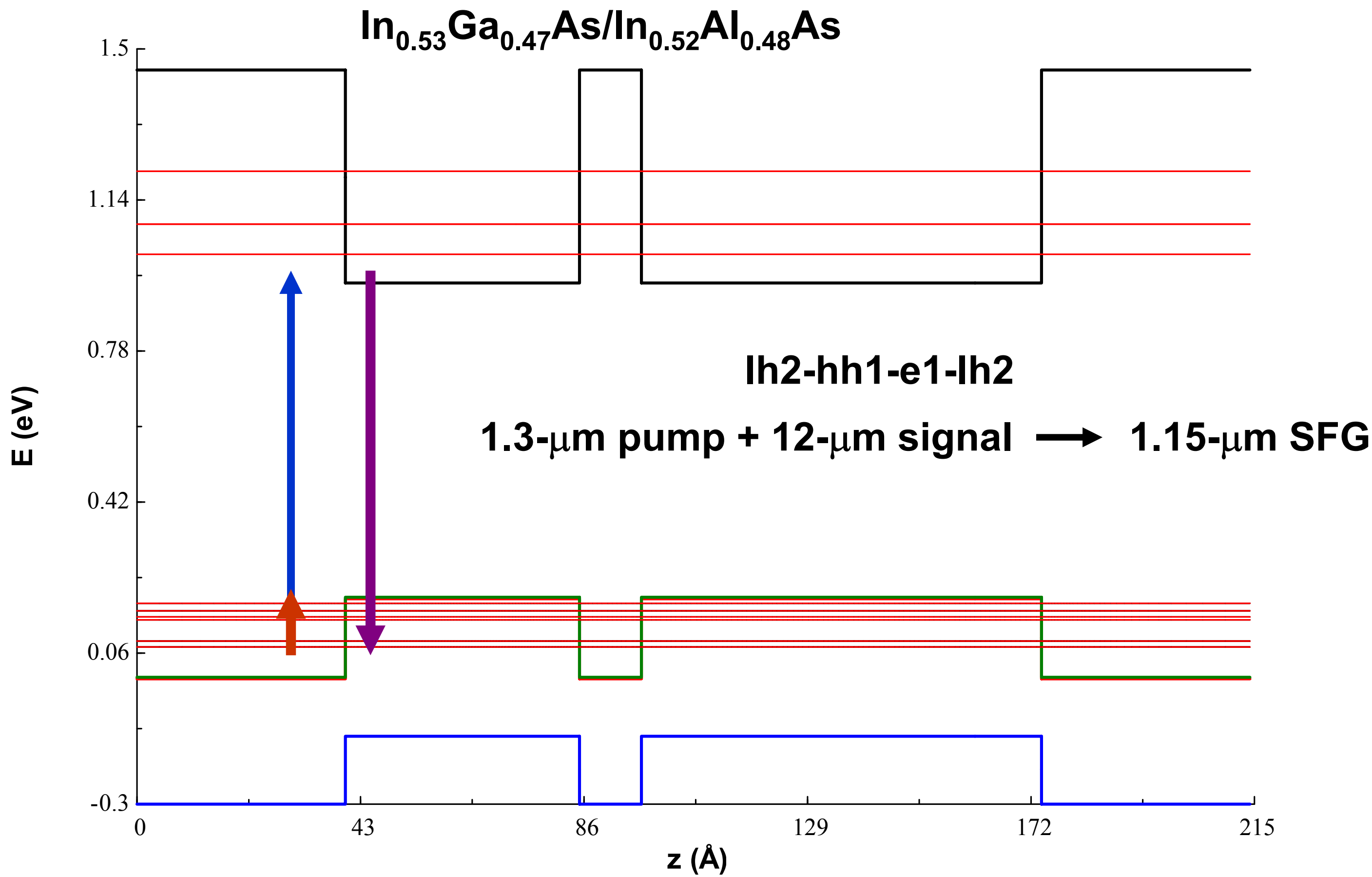
NEP_{NIR} can be extremely low: ~ 10^{-14} W, even at telecom wavelengths

(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 ~ 10^5 W/cm^2; η_{up} ~ 0.1, η_{phot} ~ 10^{-3} in the mid-infrared

Using intersubband transitions in valence band for normal incidence detection



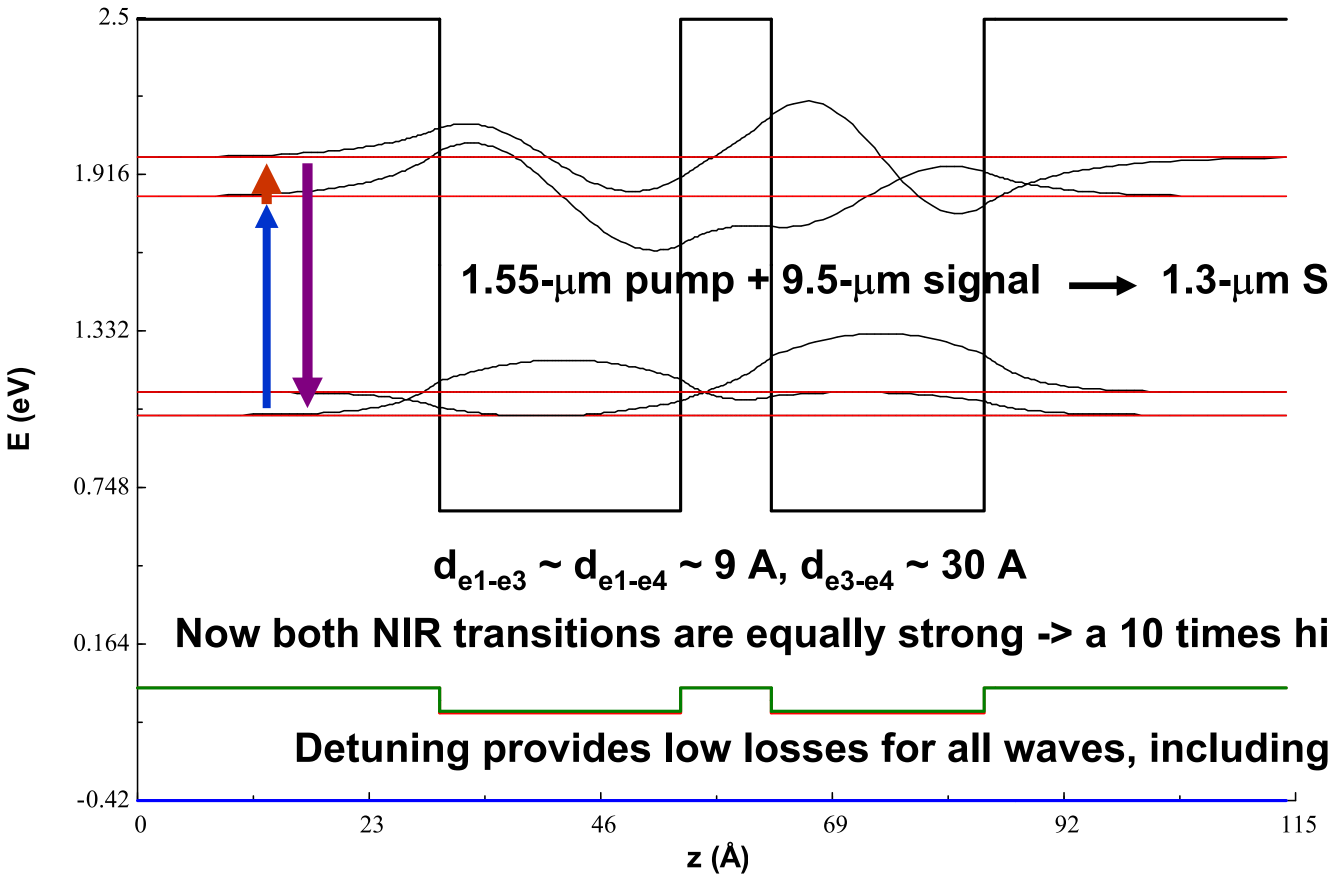
For in-plane polarized waves:

$d_{hh1\text{-}e1} \sim 7$ A, $d_{hh1\text{-}lh2} \sim 8$ A, $d_{lh2\text{-}e1} \sim 0.6$ 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



1.55-μm pump + 9.5-μm signal → 1.3-μm SFG

$d_{e1-e3} \sim d_{e1-e4} \sim 9$ A, $d_{e3-e4} \sim 30$ A

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