

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

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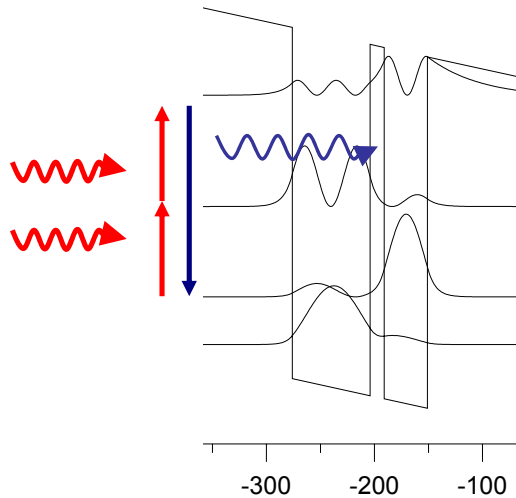
Outline

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. $\chi^{(2)} \sim 10^6$ pm/V, Raman gain $\sim 10^{-4}$ 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)

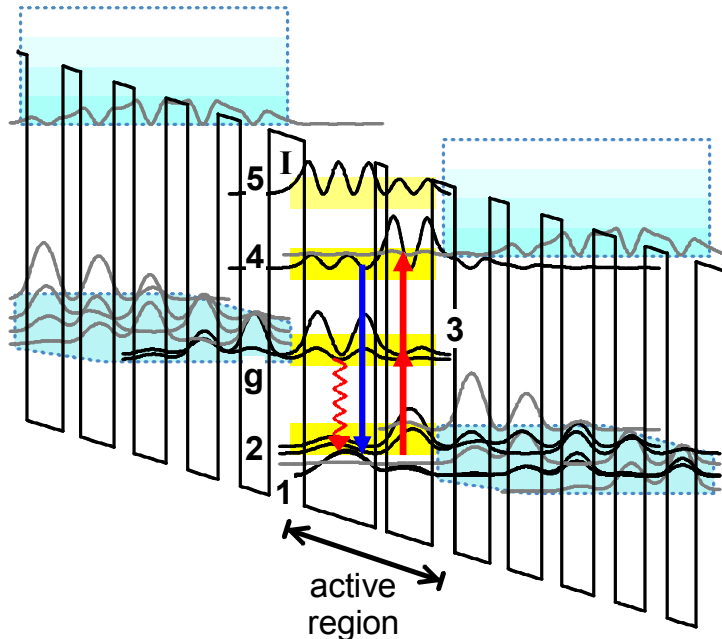
Solutions:

**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



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

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

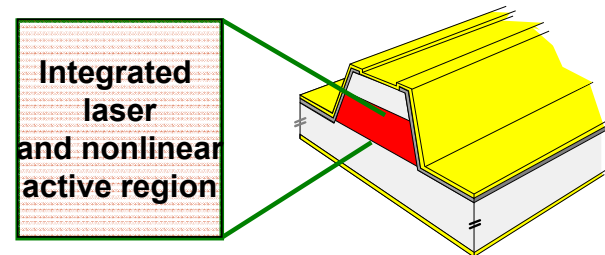
The tightest possible confinement and mode purity

No problem with external pump; an injection-pumped device

Gmachl et al. 2003

Groups led by Capasso, Gmachl, Strasser

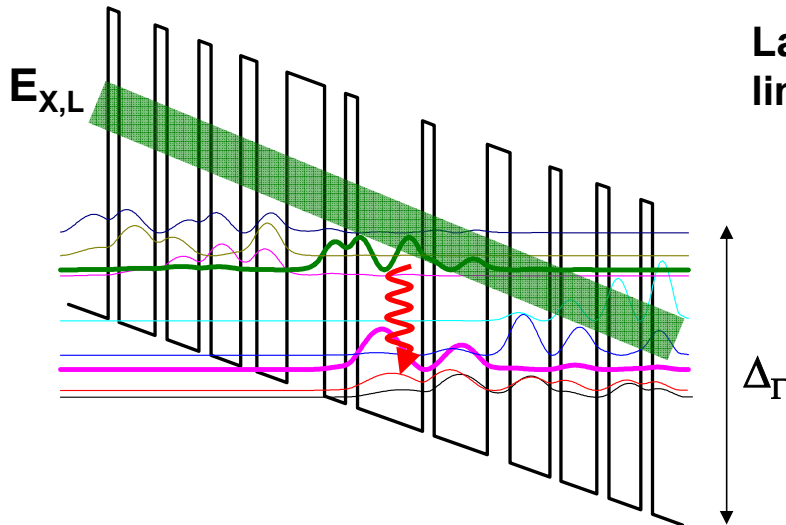
Most results are in QCLs, but there are a few in diode lasers.



Short-wavelength performance limitations of QCLs

From Tessier's talk:

	GaAs/AlAs	InGaAs/AlInAs	InGaAs/AlAsSb /InP	InAs/AlSb
Δ_{Γ} , eV	1	0.9	1.6	2.1
$\Delta_{\Gamma-X,L}$, 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

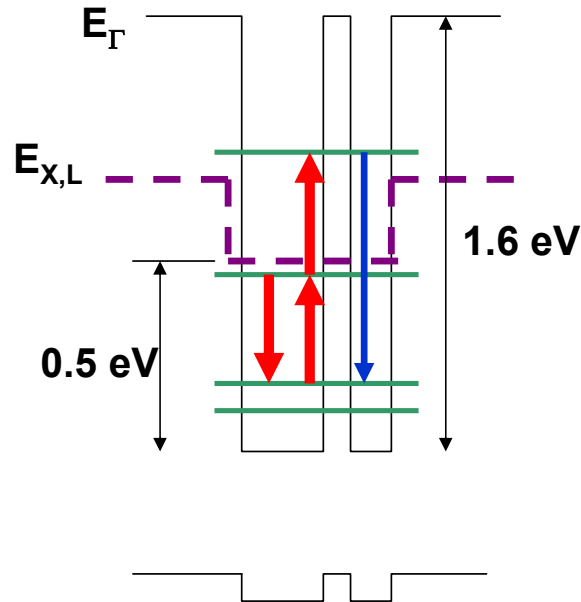
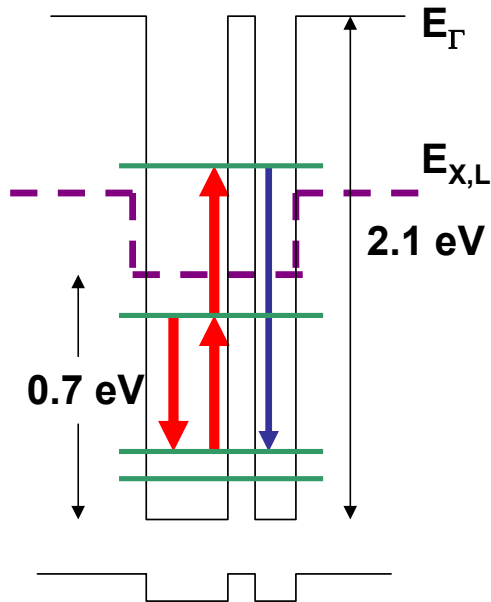
High band offset heterostructures

InAs/AlSb on InAs

Up to $\lambda \sim 2.7 \mu\text{m}$ (Univ. Montpellier)

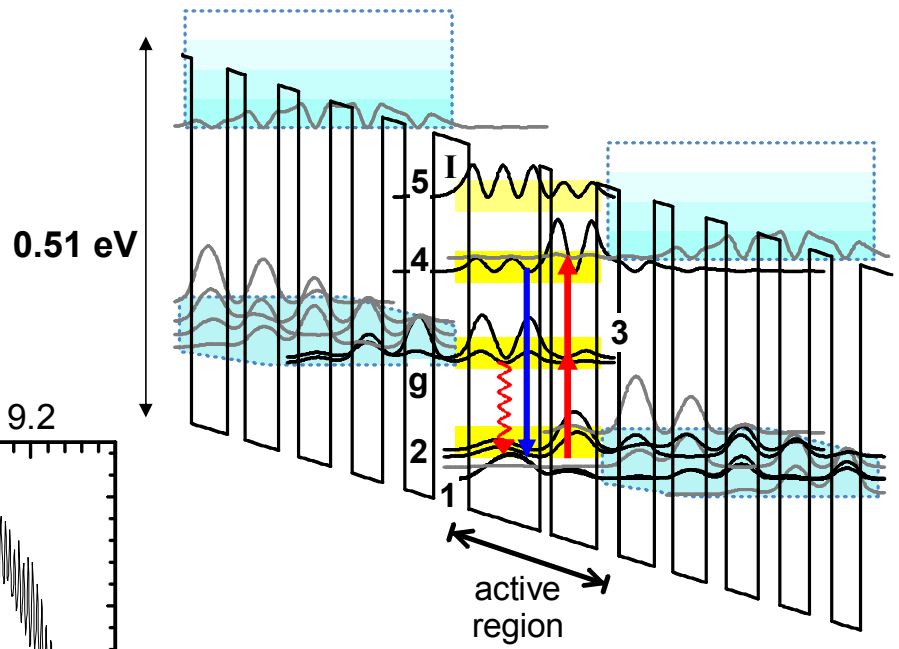
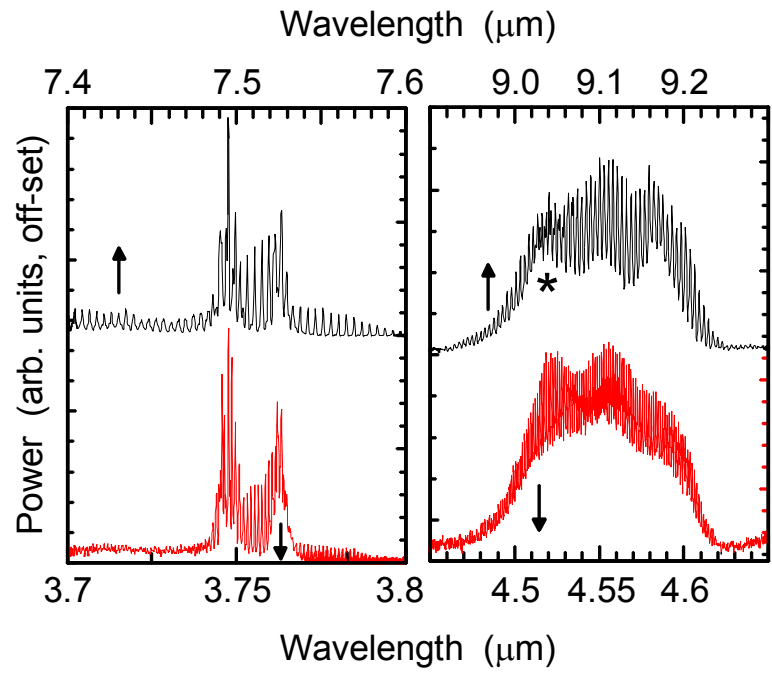
Lattice-matched GaInAs/AlAsSb/InP

(Univ. Sheffield, Fraunhofer Inst.)



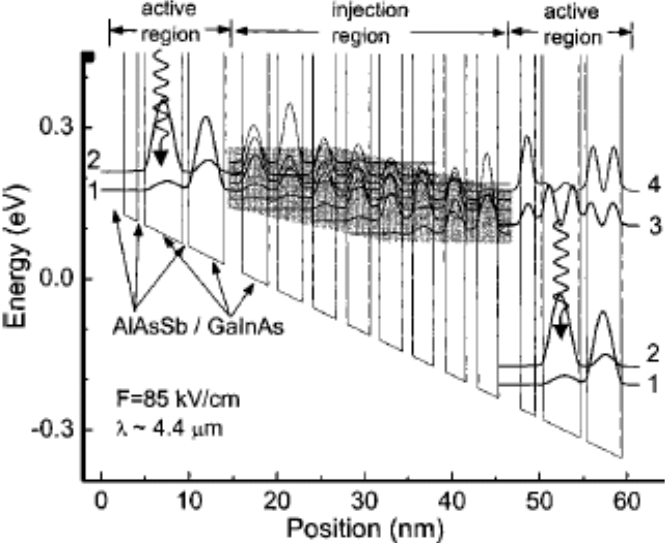
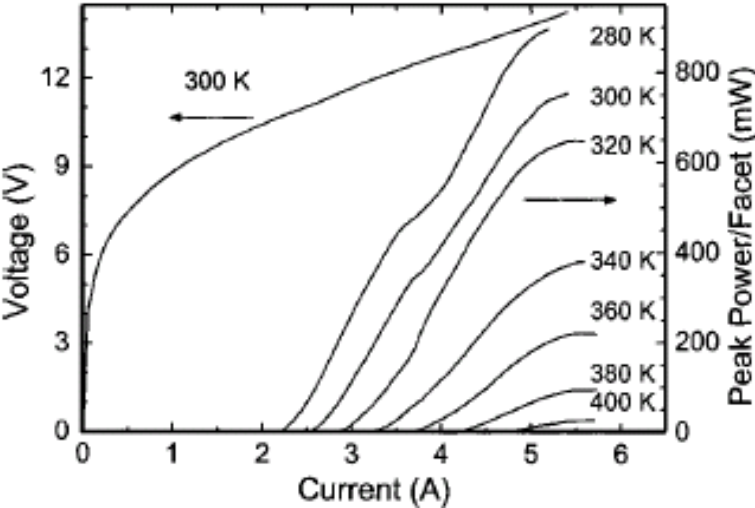
- SHG is not sensitive to the presence of lateral valleys or to the distance from the upper state to the top of the wells
- Full band offset is potentially available
- SHG needs a high-power laser and a phase-matched waveguide

All SHG lasers so far have been in lattice-matched
GaInAs/AlInAs/InP or
AlGaAs/GaAs structures



Shortest SH wavelength: 3.75 μm

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).

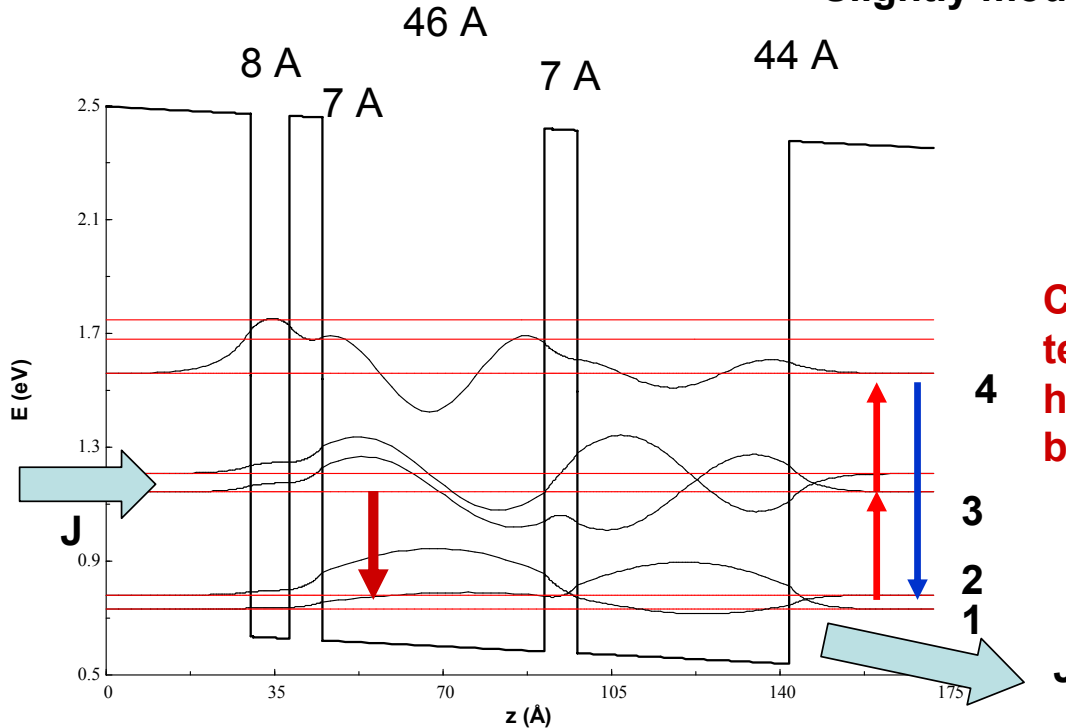


APL 86, 131107 (2005)

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.



$$\lambda(\omega) \sim 3.8 \mu\text{m}$$

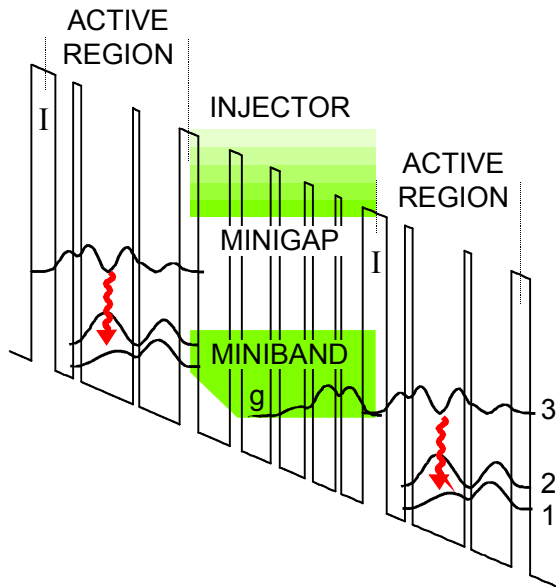
$$\lambda(2\omega) \sim 1.9 \mu\text{m}$$

Can be easily scaled to telecom wavelengths if high-power lasers at $3 \mu\text{m}$ become available!

$$\begin{aligned} d_{23} &= 20 \text{ \AA} \\ d_{24} &= 0.6 \text{ \AA} \\ d_{34} &= 11 \text{ \AA} \end{aligned}$$

$$\begin{aligned} \chi^{(2)} &\sim 700 \text{ pm/V for } N_3 = 10^{17} \text{ cm}^{-3} \text{ and } 30 \text{ meV detuning} \\ 2\gamma_{32} &= 2\gamma_{43} = \gamma_{42} = 20 \text{ meV, } E = 85 \text{ kV/cm} \end{aligned}$$

Who would care about QCLs at telecom wavelengths?



Ultrashort carrier lifetime $T_1 \sim 1$ ps

Cavity roundtrip time $T_c \sim 20-80$ ps

Dephasing time $T_2 \sim 0.1-0.5$ ps

$$T_2 < T_1 \ll T_c$$

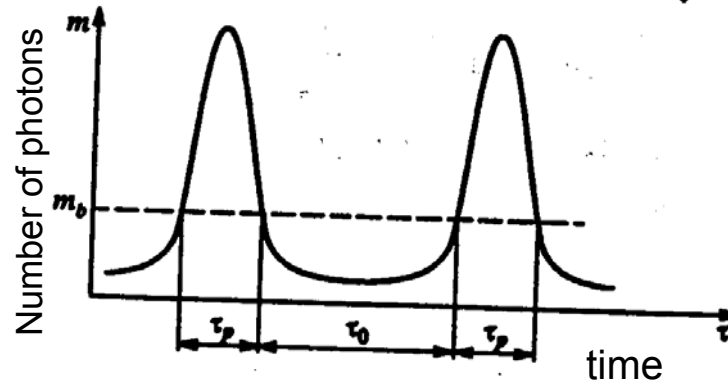
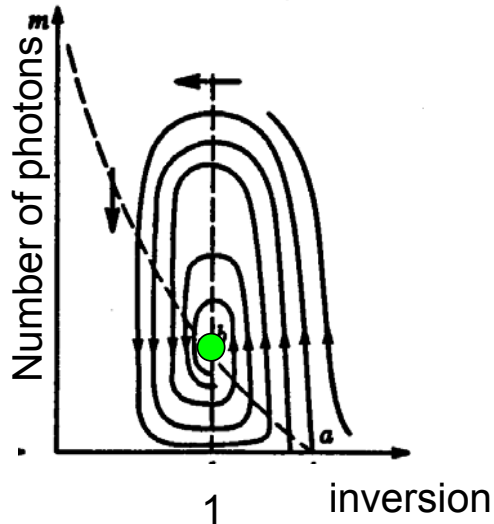
An overdamped Class A laser?

Only for small-signal modulation and low power

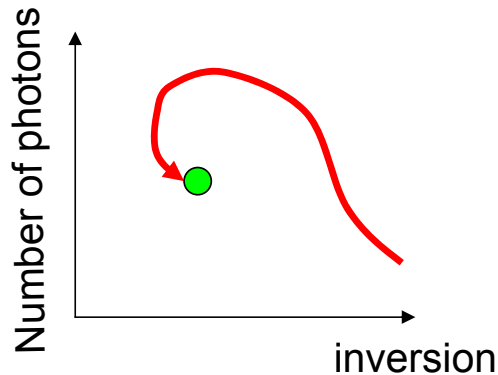
All other solid-state and diode lasers are Class B:

$$T_2 \ll T_c \ll T_1$$

Simplest dynamics: relaxation oscillations



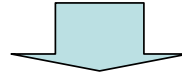
Solid-state lasers: $\frac{T_1}{T_c} \gg 1$
Stable focus



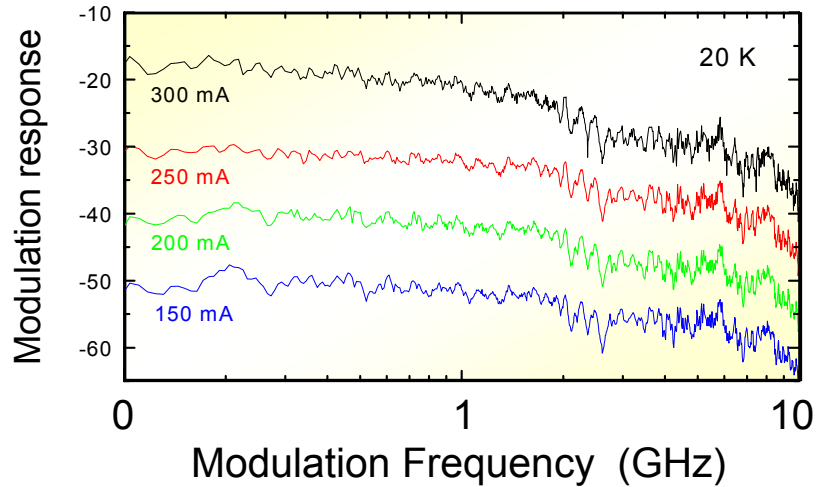
QC-lasers: $\frac{T_1}{T_c} \ll 1$
Stable node

Overdamped oscillations

Absence of relaxation oscillations?



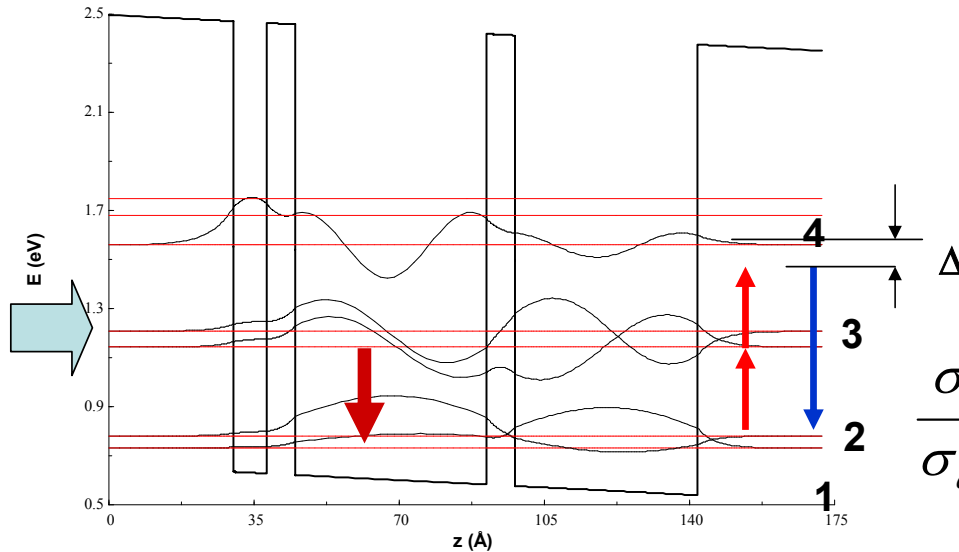
Potential for high-speed modulation up to THz rates



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

Nonlinear polarization at second harmonic:

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



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

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

Need $\Delta \sim 2-3\gamma$

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

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 $P_{NL} \sim \chi^{(2)} E_p^2$

$$W_s \sim \frac{32\pi^5 W_p^2 \left| \int \frac{\chi^{(2)}(z)}{\varepsilon^2(z, \omega_p)} H_p^2(y, z) H_s(y, z) dy dz \right|^2 |L_{coh}|^2}{\mu_p^2 \mu_s c \lambda_s^2}$$

“p” – pump
“s” - signal

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

Coherence length mainly limited by phase mismatch

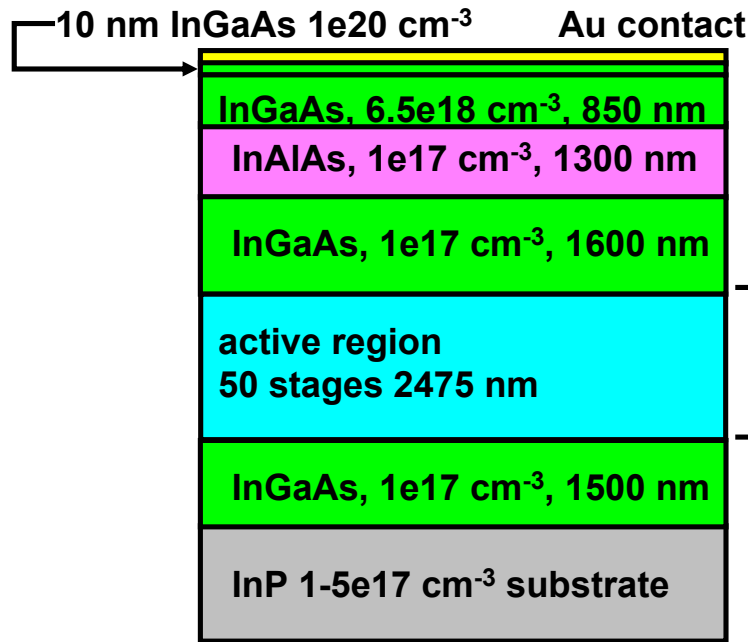
Without phase matching: $\Delta k_x \sim 2000 \text{ cm}^{-1}$

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

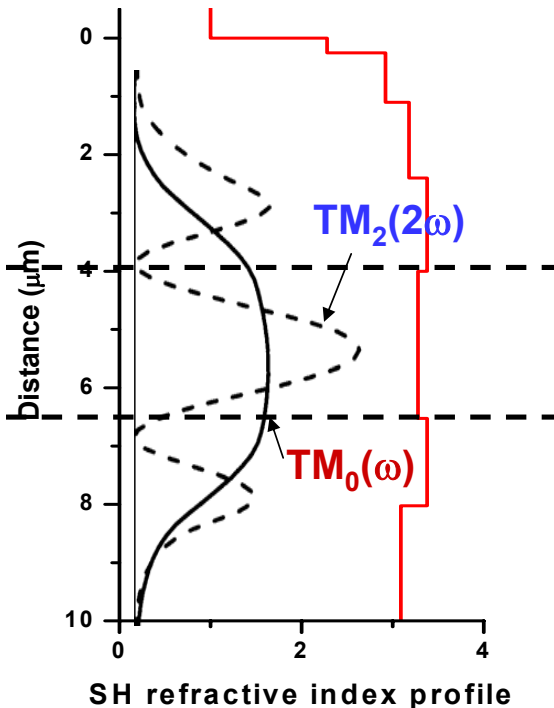
- Quasi phase matching by periodic Stark shift: APL 88, 201108 (2006)
- Modal phase matching: APL 84, 2751 (2004), EL 40 (2004)
- Off-axis or surface emission

Modal phase matching seems to be most promising at this point

Waveguide design for phase-matched second-harmonic generation



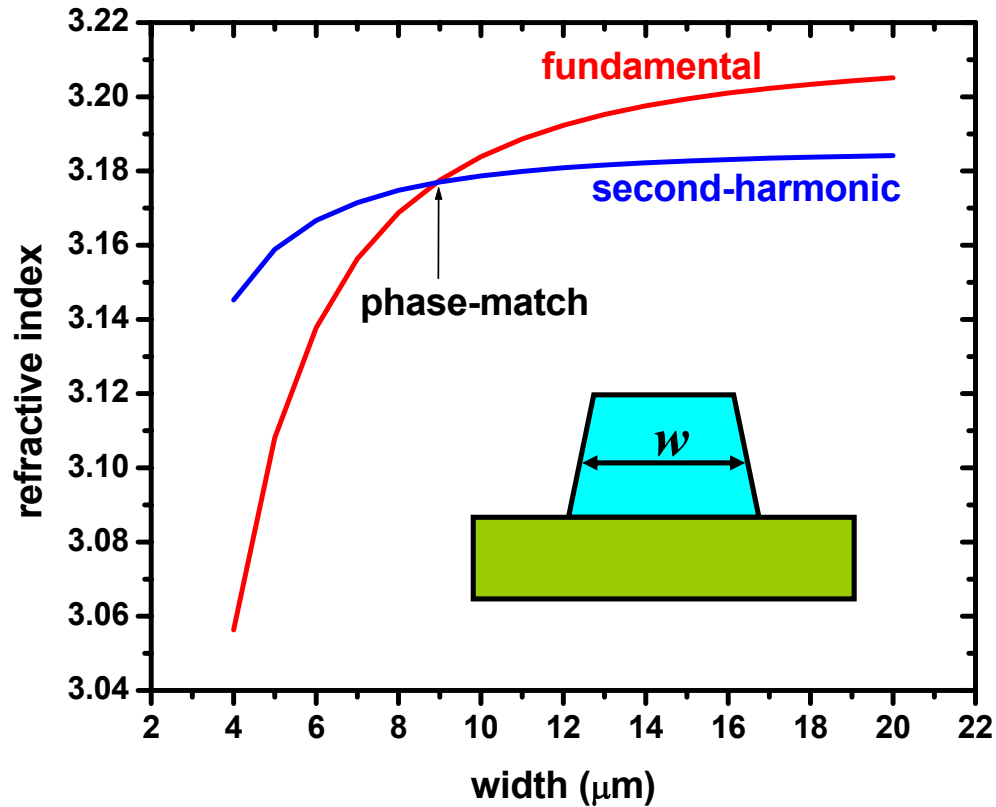
$$TM_{00}(\omega_1) + TM_{00}(\omega_1) \rightarrow TM_{02}(2\omega_1)$$



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

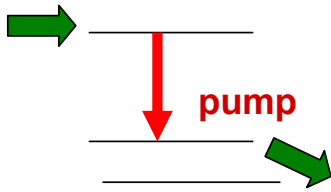
O. Malis et al., Electron. Lett. 40, 1586 (2004)

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

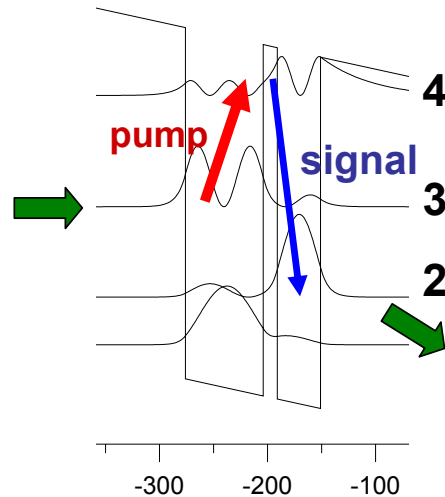


Anti-Stokes Raman injection laser for tunable frequency up-conversion

Laser pump stage



Raman stage

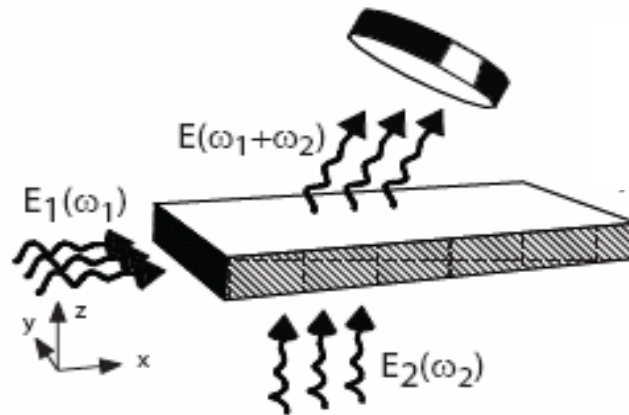


- More powerful than SHG
- No phase matching is needed
- Has threshold; may compromise the pump laser

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

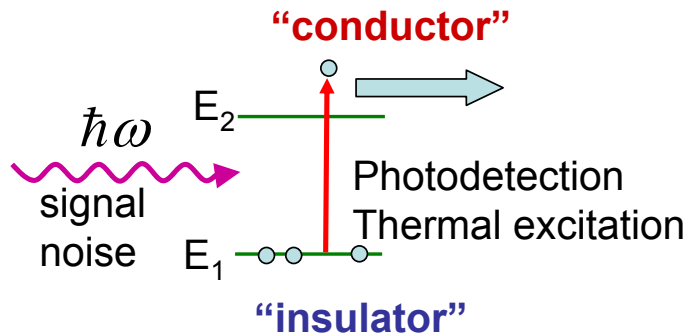
APL 87, 26113 (2005): antistokes Raman emission but no lasing

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



Mid/far-IR semiconductor photodetectors

Interband, intersubband, or impurity transition

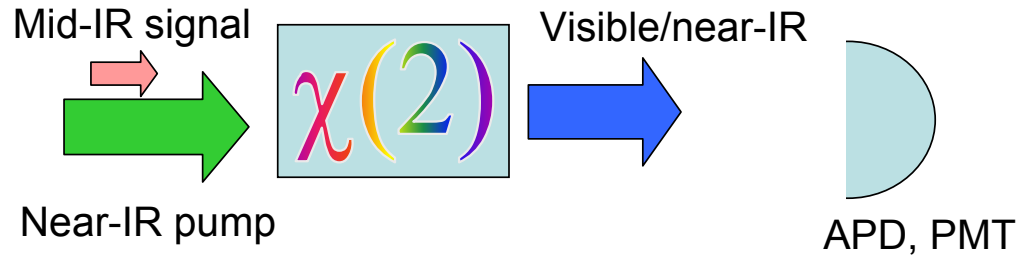


$$E_2 - E_1 \leq \hbar\omega$$

- 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

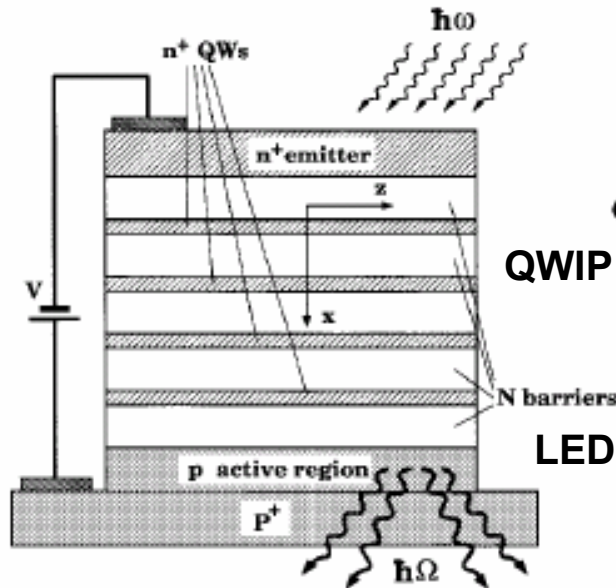
Developed since 1960s: Midwinter, Townes, Boyd, ...



- 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: $\sim 10^{-6}$;
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

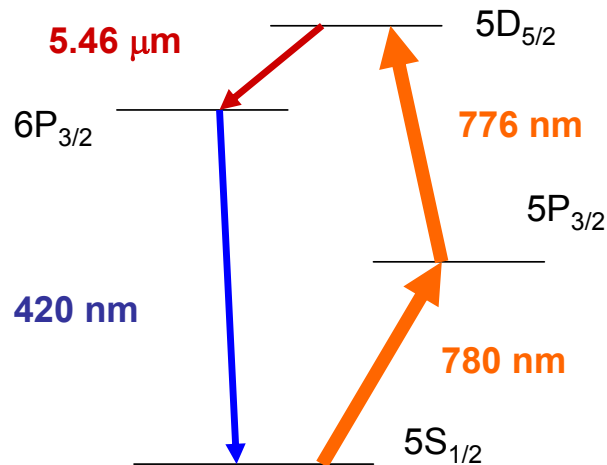
Generation of mid-infrared (Phillips, today's talk) and THz (Barbieri, next talk) sidebands on a near-IR optical carrier in a QCL

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

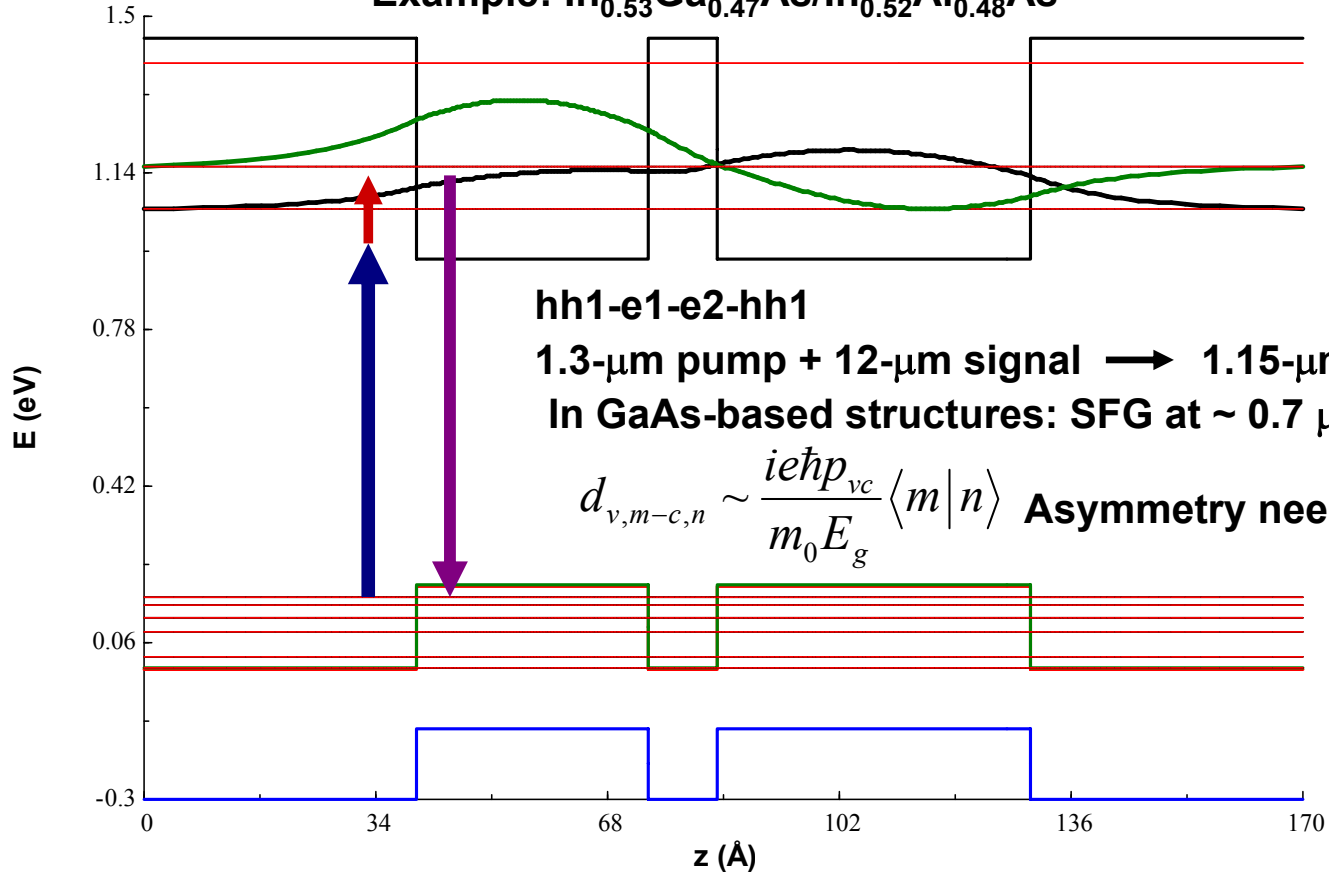
1. Nonlinearity and spectral response
2. Geometry
3. Figures of merit
4. Various device designs

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



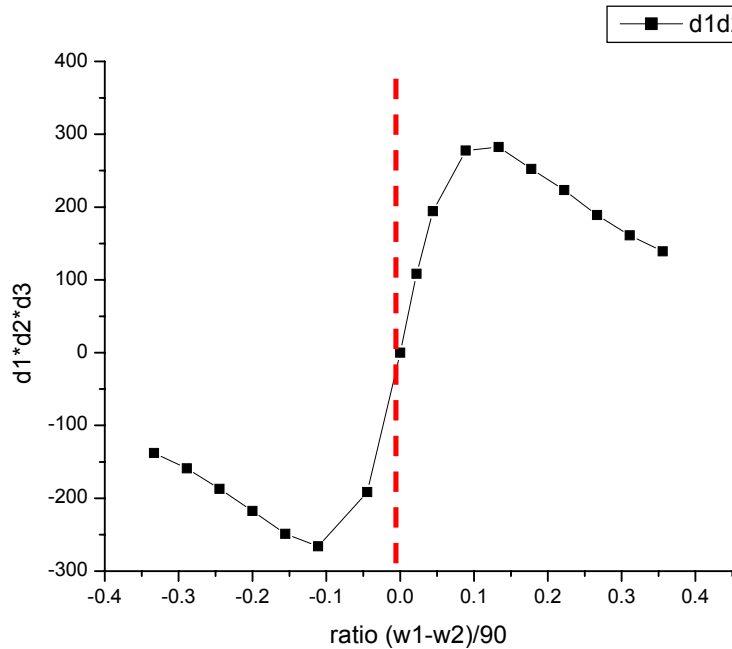
6-band VB + CB with energy-dependent mass

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

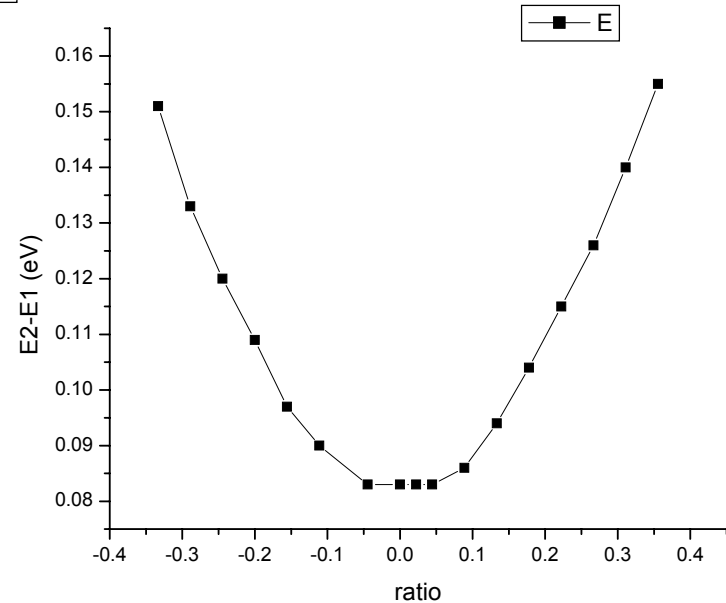
Choosing the right structure

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

Total thickness 90A, barrier thickness=10A



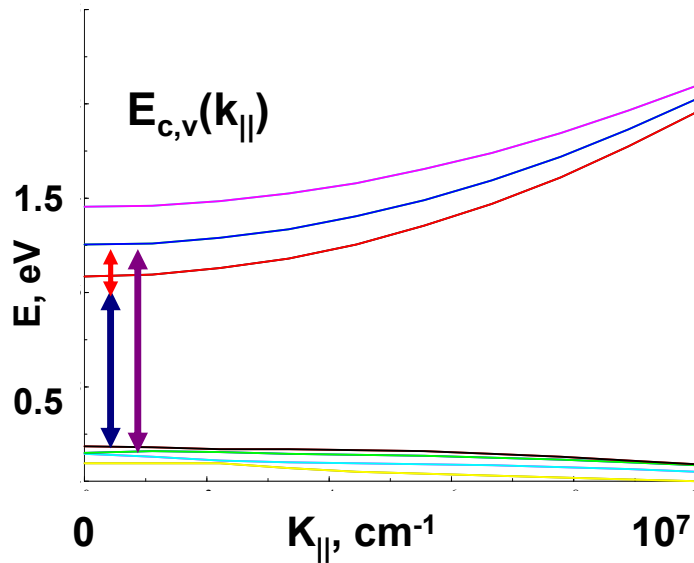
Product of dipole moments vs well asymmetry $((w1-w2)/\text{total thickness})$



Energy difference vs well asymmetry

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

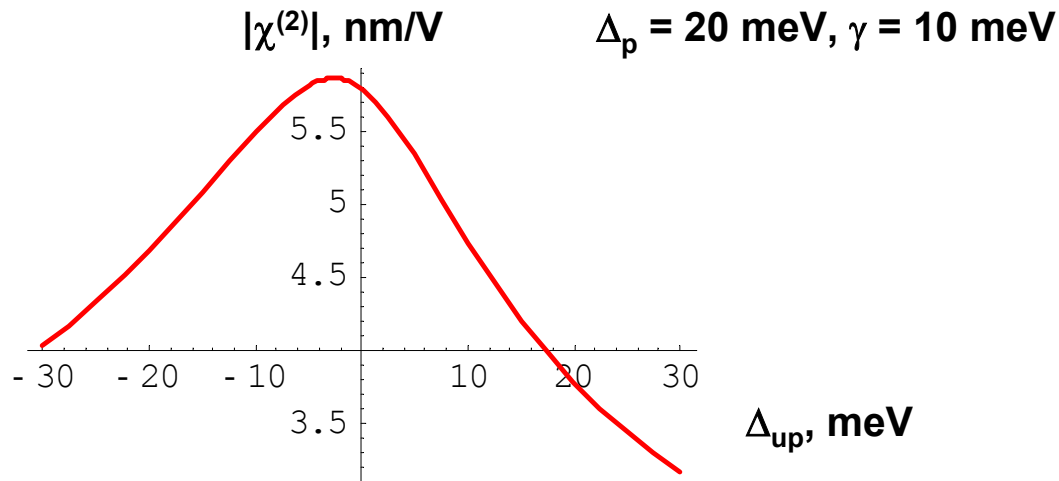
$$\chi^{(2)}(\omega_{up} = \omega_s + \omega_p) \sim \frac{1}{2\pi L_{qw}} \int dk^2 \frac{d_{vc1} d_{vc2} d_{c1c2} (f_{v1}(k) - f_{c1}(k))}{(E_{vc2}(k) - \hbar\omega_{up} - i\hbar\gamma_{vc2}(k))(E_{vc1}(k) - \hbar\omega_p - i\hbar\gamma_{vc1}(k))}$$



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

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

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



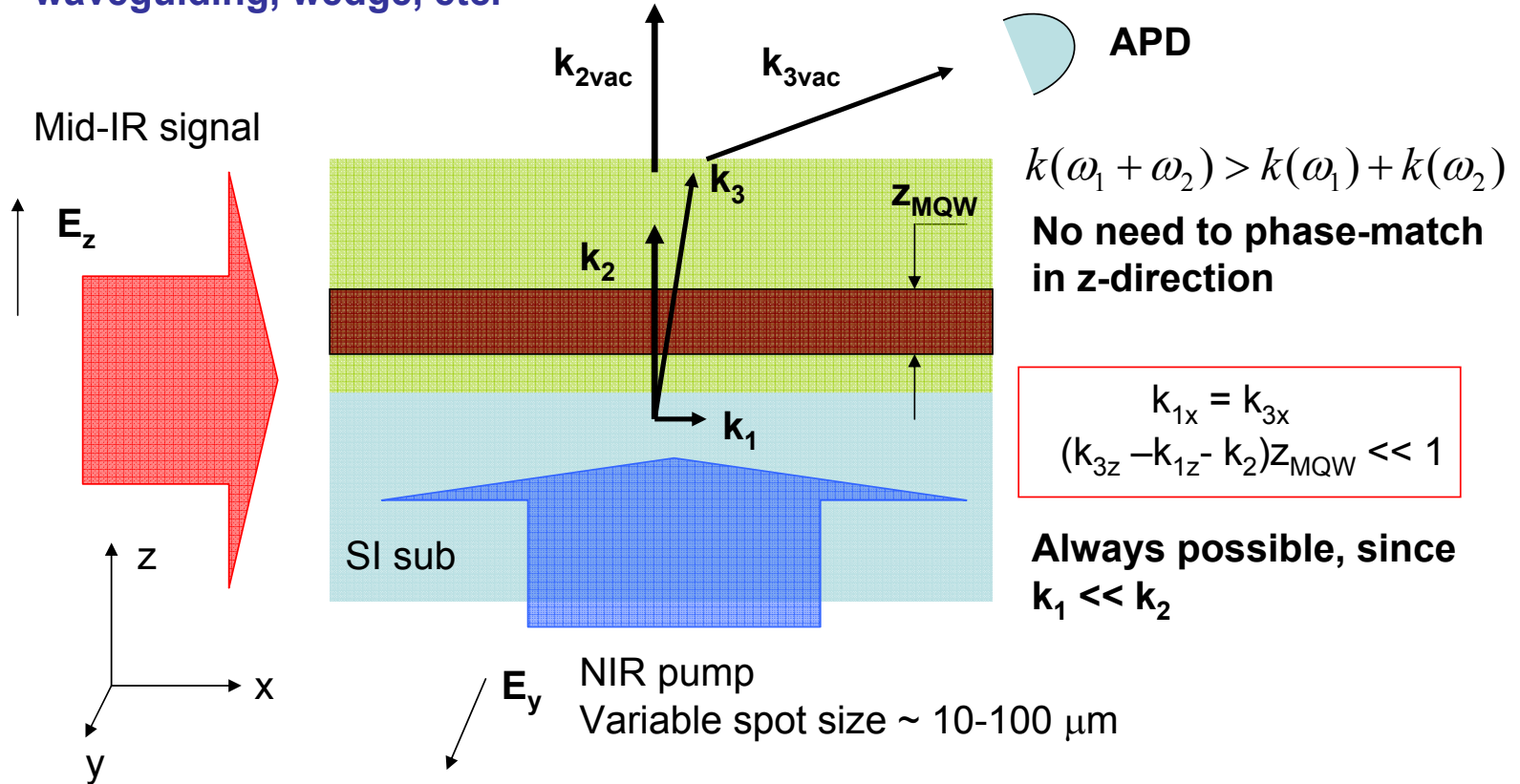
$|\chi^{(2)}|^2$ bandwidth of ~ 35 meV for a 100-meV mid-IR photon energy

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.



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

Figures of merit

Up-conversion efficiency:

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

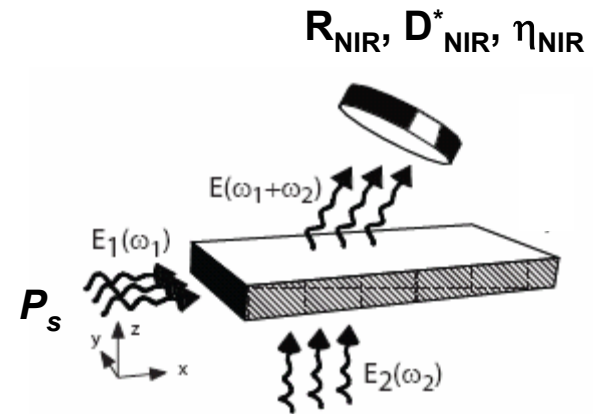
Responsivity: $R = \frac{I_{NIR}}{P_s} = \eta_{up} R_{NIR}$

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:

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



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

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

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

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

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

Interband absorption per well:

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

~ 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 !

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

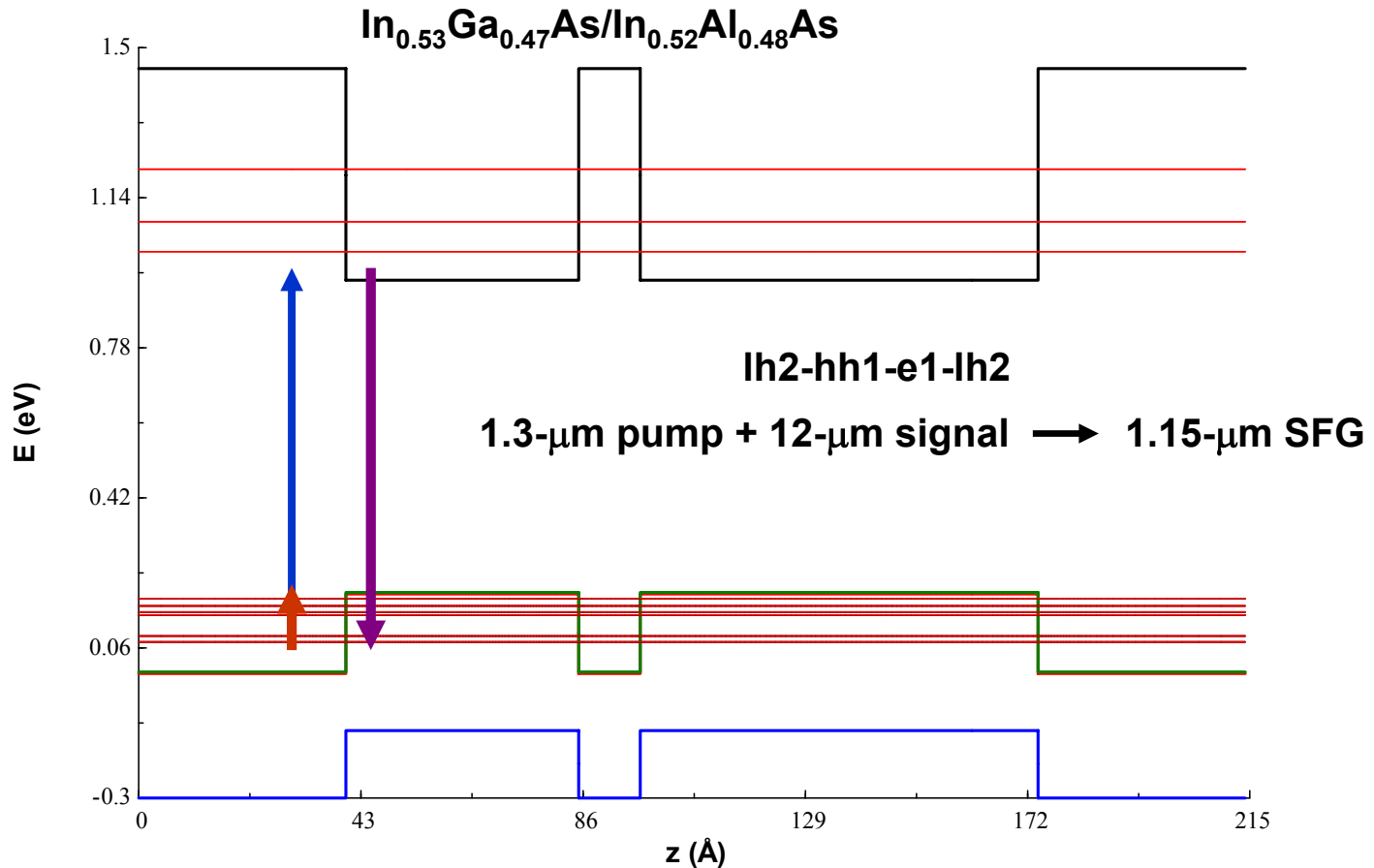
$$n_{e2} \sim n_{e1} e^{-\frac{E_{e2}-E_{e1}}{k_B T}} \quad \text{in equilibrium.} \quad \alpha_{\text{pump}} I_p / \hbar\omega_p = n_{e1} / \tau_{e1}$$

NEP_{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/cm²; $\eta_{\text{up}} \sim 0.1$, $\eta_{\text{phot}} \sim 10^{-3}$ in the mid-infrared

Using intersubband transitions in valence band for normal incidence detection



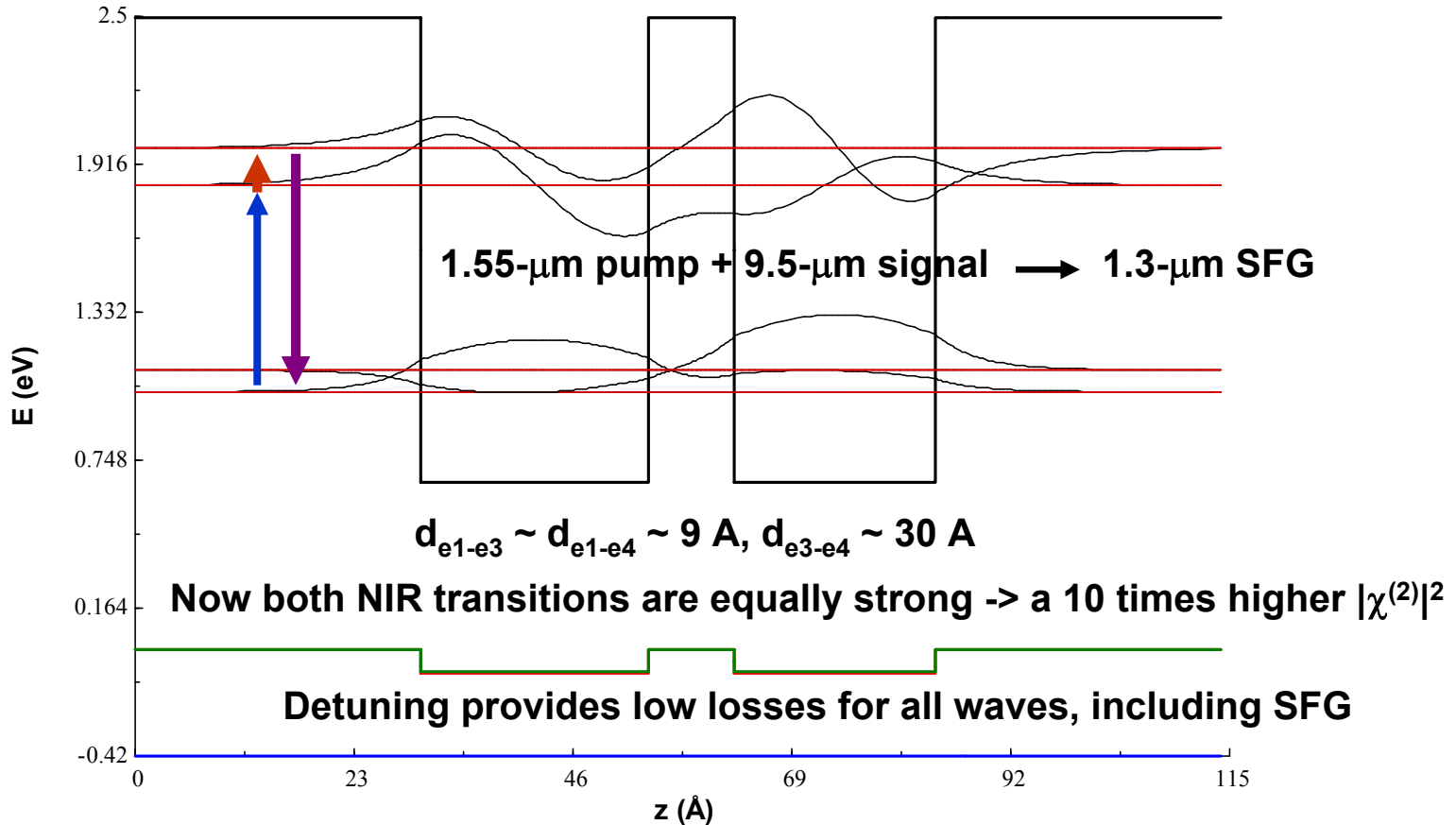
For in-plane polarized waves:

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

**Lower nonlinearity,
but simpler geometry**

SFG at telecom wavelengths in lattice matched $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}/\text{AlAs}_{0.56}\text{Sb}_{0.44}/\text{InP}$

Intersubband transitions only



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**