Phase-matching in cascaded third-order frequency mixing

<u>Charles G. Durfee</u>

Department of Physics
Colorado School of Mines
Golden, CO 80401
cdurfee@mines.edu

Lino Misoguti,
Sterling Backus,
Margaret M. Murnane and
Henry C. Kapteyn

Department of Physics/JILA
University of Colorado
Boulder, CO 80303

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Outline

- Applications of ultrafast shortwavelength pulses
- Background: alternatives for frequency conversion
- Techniques for hollow-core waveguide phase matching
- Phase-matching of cascaded mixing
  - analytic phase-matching conditions
  - simulations
  - experimental results
- Scaling to greater conversion and higher orders

Applications of ultrafast short-wavelength pulses

- Chemical reactions
  - > time resolution
  - > intermediate states
  - > coherent control
- Materials
  - > micromachining/nanofabrication
  - > lithography
  - > synchrotron applications
- Physics
  - > atomic stabilization
  - > high density plasma interactions
  - > x-ray nonlinear optics
- Biophysics
  - > cross-linking DNA/proteins
  - > water-window holography

Frequency conversion with nonlinear crystals

Limitations of nonlinear crystals:

• Transparency	~ 150-200 nm
• Phase-matching range	~ 190 nm
• Phase-matching bandwidth/ group velocity walkoff	~ 150 fs @266nm
• Periodically-poled materials	~ 350 nm
• Crystal damage for high-power pulses	

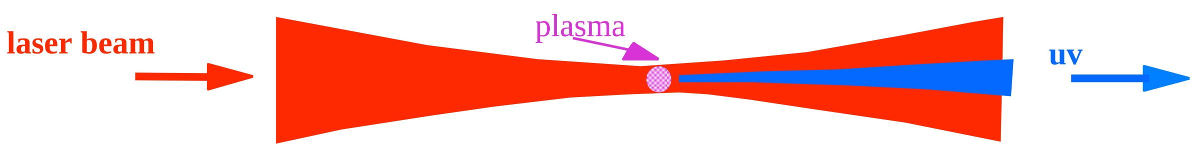
Frequency conversion in gases

<u>Use of gases circumvents some of these problems:</u>

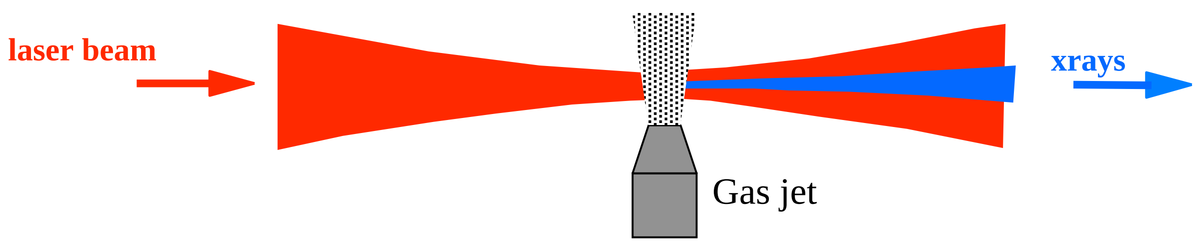
- High transparency: He: >50 nm, Ar: >105nm
- Low dispersion preserves pulse width
- No damage limitations

Focused beam geometry: Guoy phase shift

- direct third-harmonic
  (1 μJ, 0.1%, 12 fs)



- High-order harmonic generation



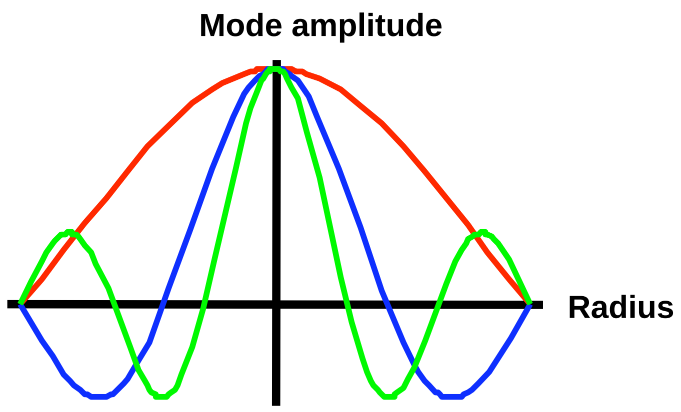
Hollow waveguides for high-intensity guiding

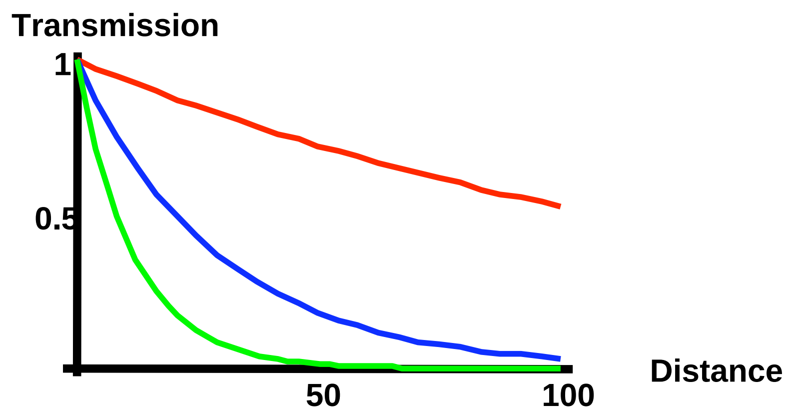
<u>Low conversion efficiency in gases:</u>

- Improve *intensity* x *density* x *length*
- Phase-matching

<u>Capillary waveguides:</u>

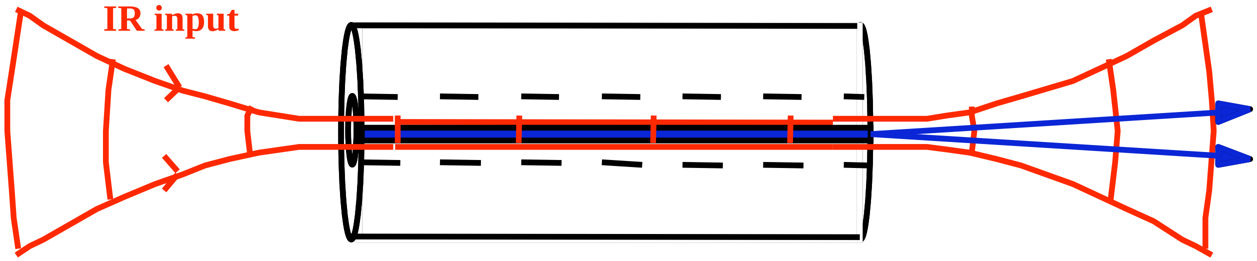
- guiding is lossy but allows high-intensity interactions





150 μm core dia, λ = 800 nm

Hollow waveguide propagation phase



The waveguide modes have flat wavefronts → z-independent contribution to phase.

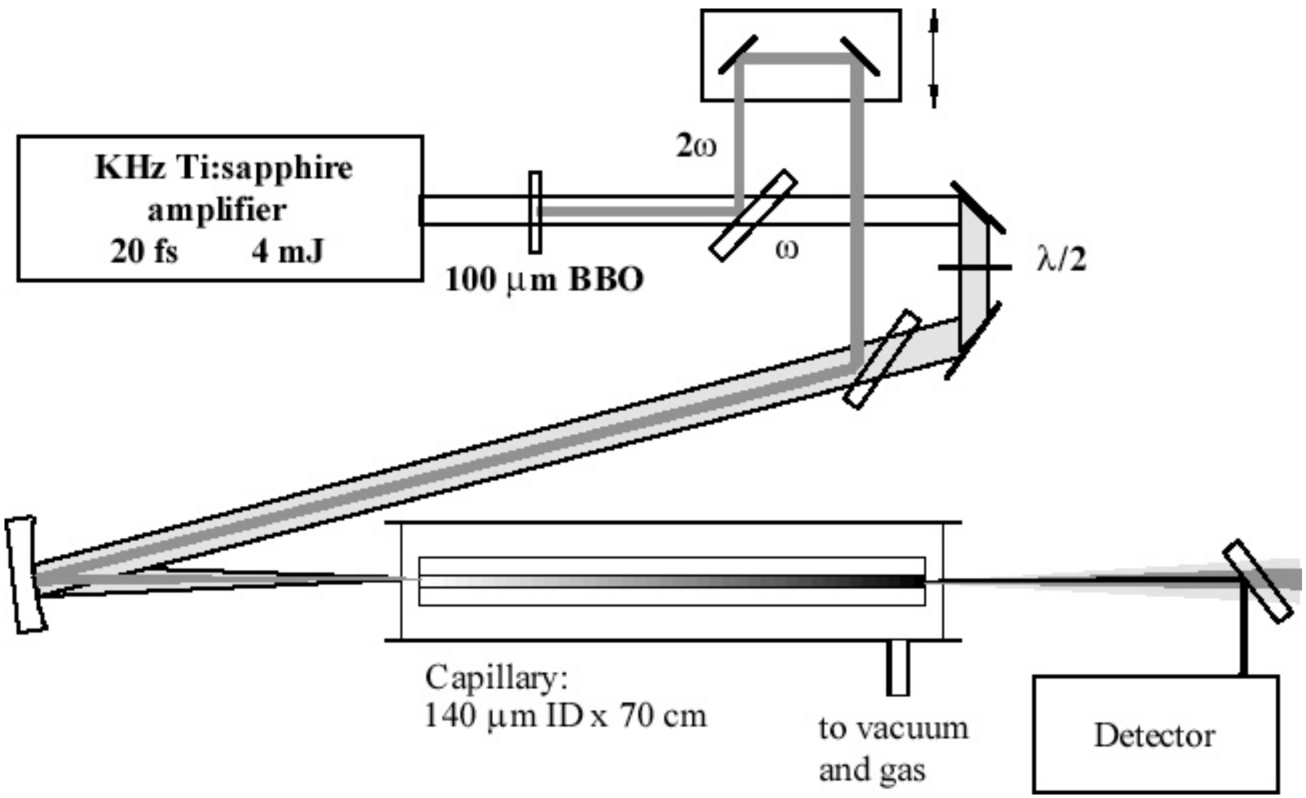
$$k = \frac{2\pi}{\lambda}\left[1 + P\delta(\lambda) - \frac{1}{2}\left(\frac{u_{nm}\lambda}{2\pi a}\right)^2\right]$$

vacuum gas waveguide

Waveguide phase from countering diffraction: strong effect on long wavelength

Phase match ($\Delta k = 0$) by varying gas pressure to balance gas and waveguide dispersion.

Experimental setup



Capillary lengths vary or can be segmented

Techniques for waveguide phase matching

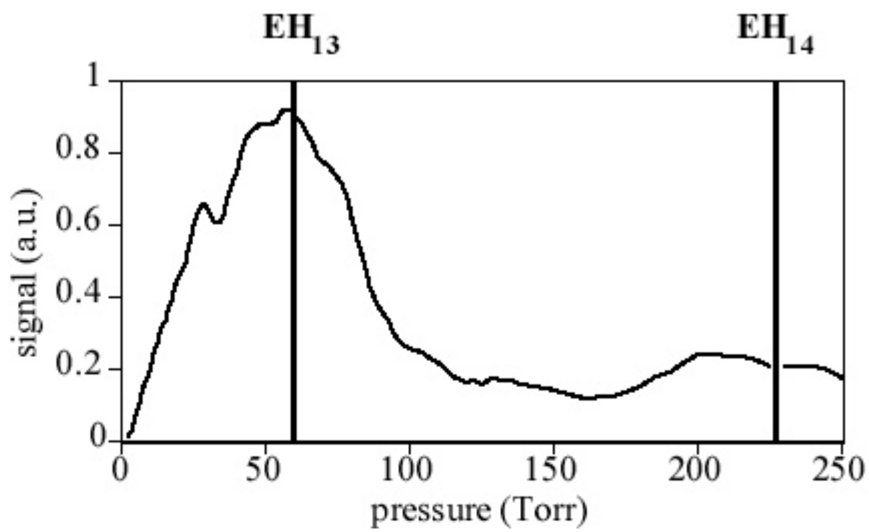
Modal phase matching for direct harmonic generation

(Durfee et al, Opt. Lett. **22** p. 1565 (1997))

$$\Delta k = \frac{2\pi P}{\lambda_3}\left[\delta(\lambda_3) - \delta(\lambda_1)\right] - \frac{1}{4\pi a^2}\left(u_{13}^2\lambda_3 - 3u_{11}^2\lambda_1\right)$$

$$\Delta k_{material} > 0 \qquad \Delta k_{mode} > 0$$

Experimental data: Argon

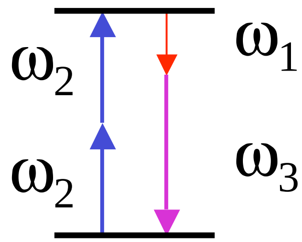


Efficiency is low (~0.2%) because of poor mode overlap

Techniques for waveguide phase matching

Frequency mixing allows all beams to be in lowest mode

$$\omega_3 = 2\omega_2 - \omega_1$$



ω_1= idler

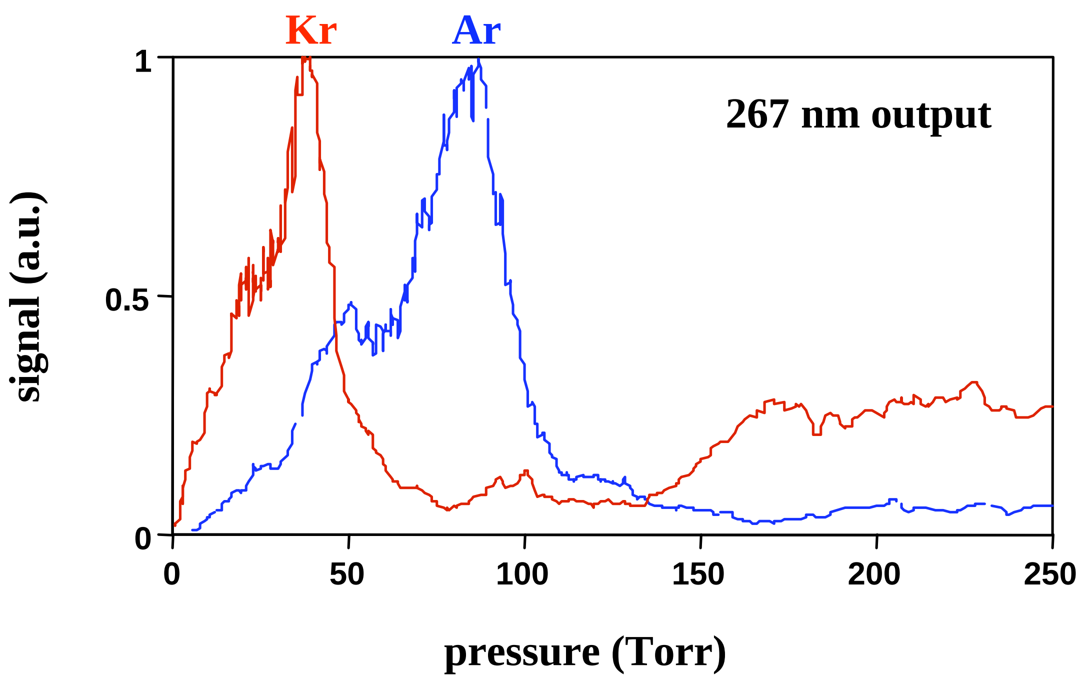
ω_2= pump

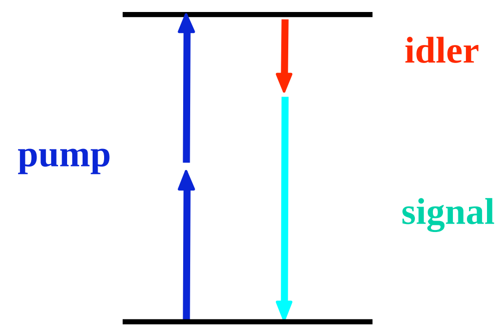
ω_3= signal

$$\Delta k = 2\pi P\left[\frac{\delta(\lambda_3)}{\lambda_3} - \frac{2\delta(\lambda_2)}{\lambda_2} + \frac{\delta(\lambda_1)}{\lambda_1}\right] - \frac{u_{11}^2}{4\pi a^2}\left(\lambda_3 - \cancel{2\lambda_2} + \cancel{\lambda_1}\right)$$

Phase-matching: Tune pressure to balance gas and modal terms

Waveguide phase matching: UV results





$\omega_{signal} = 2 \times \omega_{pump} - \omega_{idler}$

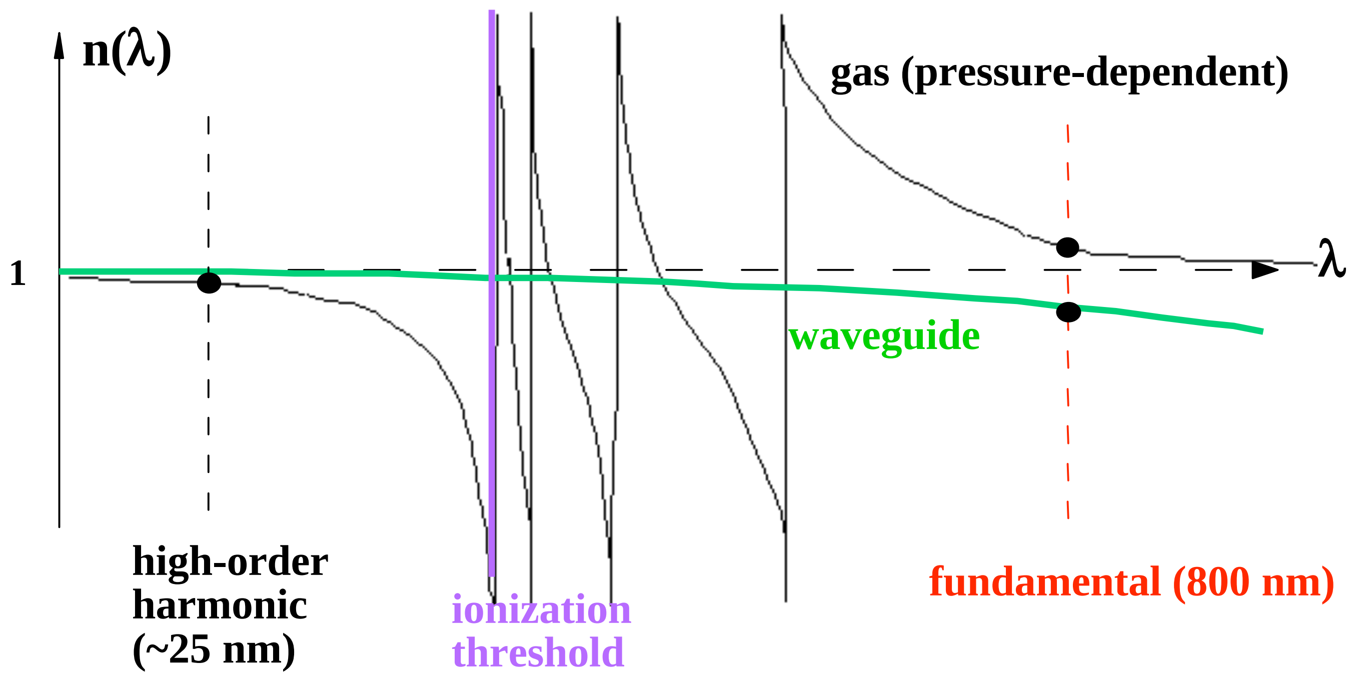
~ 40% conversion from blue pump
near-Gaussian output mode
< 7 fs walkoff

Tunable throughout the deep-UV
by using OPA output as idler

Durfee et al, Opt. Lett. **22** p. 1565 (1997)

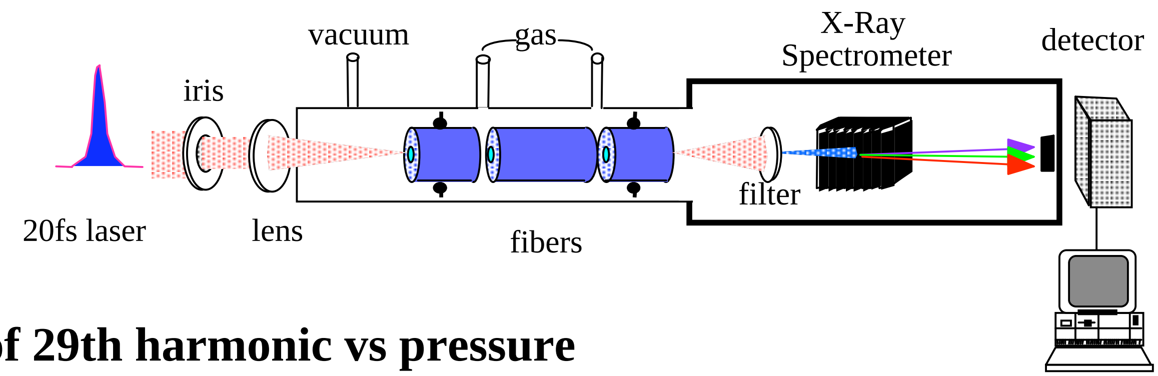
Which gas? *High nonlinearity=high dispersion=lower pressure*.
Doesn't really matter.

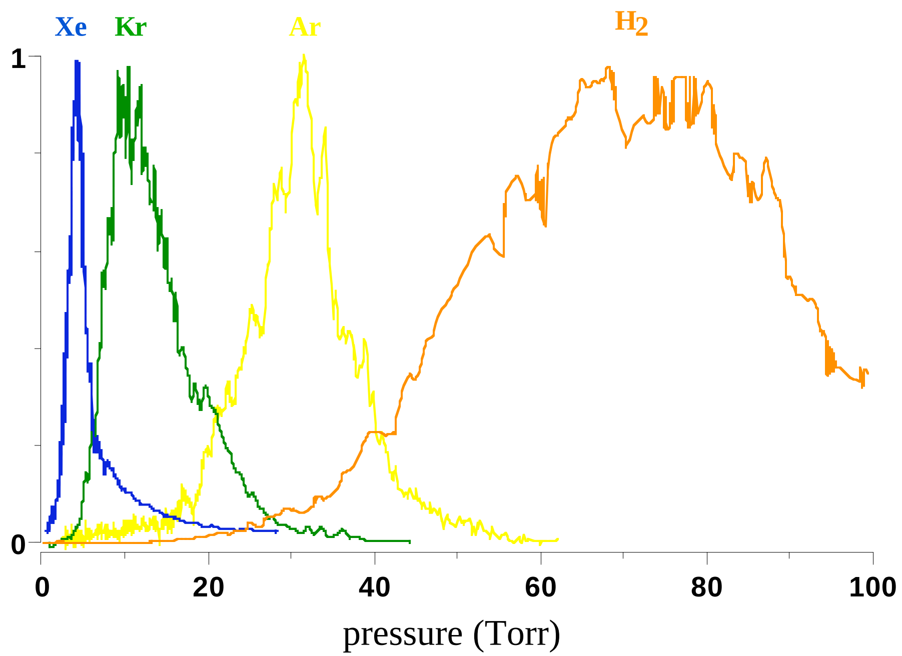
Waveguide phase matching: high harmonics



- Gas, waveguide dispersion balance.
- n < 1 @ 25 nm -> Simple HHG can be phase-matched.
- Absorption of signal is now an issue

Phase-matched HHG results





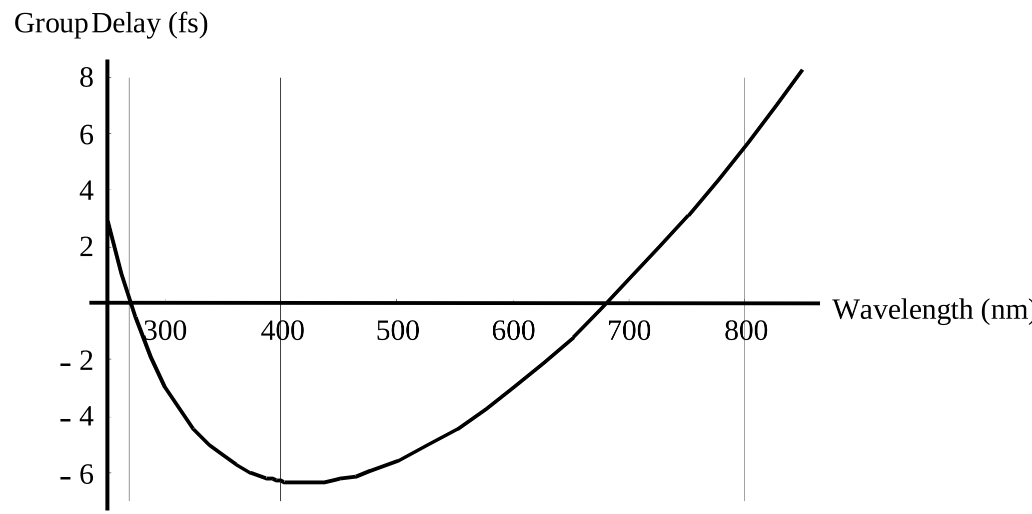
- Output enhanced by 100-1000x
- Yield limited by absorption
- optimum pressure varies with gas
- no defocusing, little absorption of fundamental

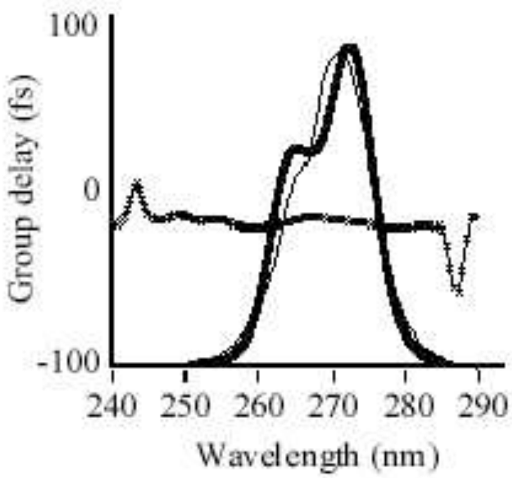
Rundquist *et al*, Science **280**, 1412 (1998)
Durfee *et al*, PRL **83**, 2187 (1999)

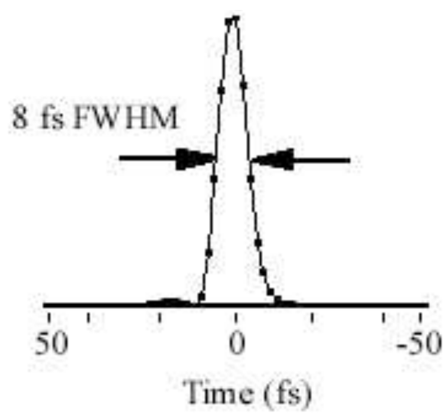
Short UV pulse generation/compression

High pulse intensity: SPM, XPM

Low degree of walkoff allows short pulse generation





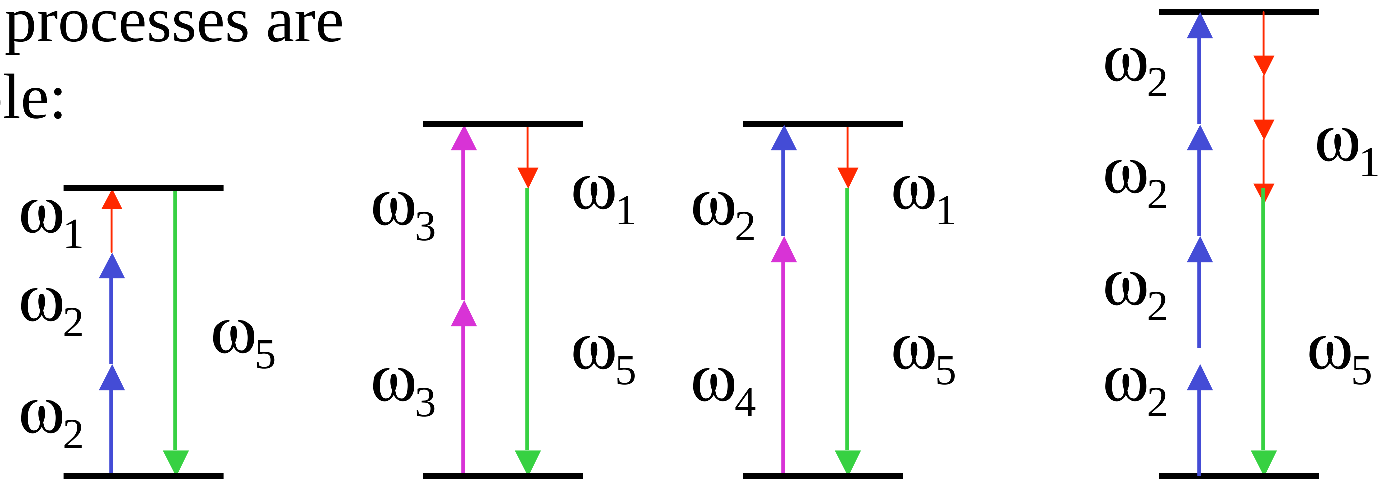


Post-compression with gratings yields ~ 8 fs, μJ pulses

Cascaded processes

High conversion to third harmonic drives cascaded processes.

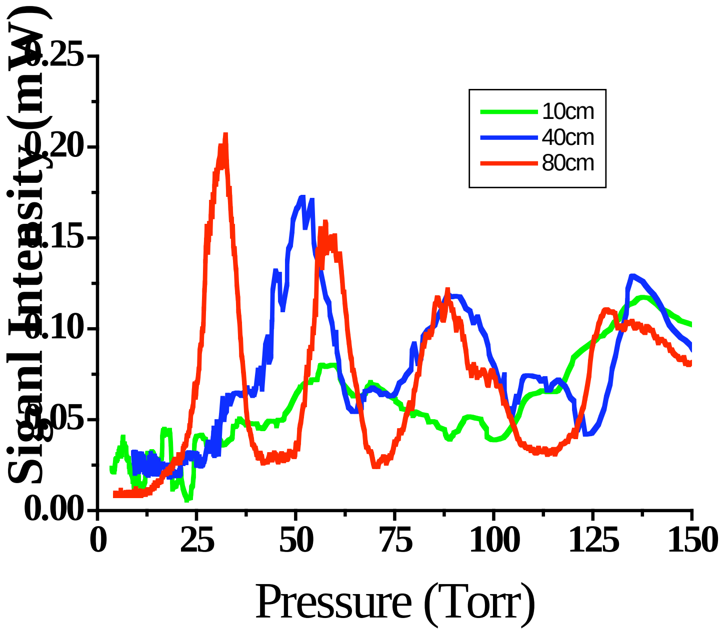
Many processes are possible:



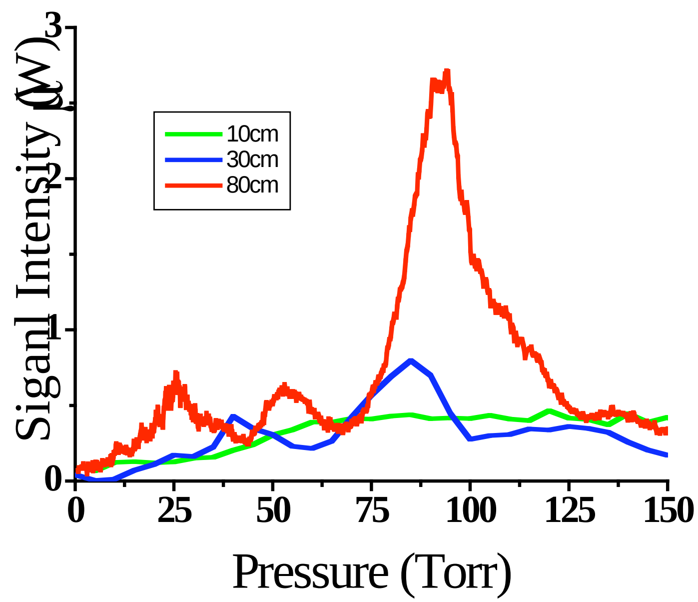
Which paths are most important?

- phase-matching
- effective order of nonlinearity

Cascaded generation: experimental results



ω_4 = 200nm (~1%)



ω_5 = 160nm (~0.01%)

Cascaded conversion is relatively efficient, shows distinct signature of phase matching.

Approaches to calculations

Two approaches to calculations:

1. Analytic - small-signal analysis
- phase-matching conditions
- estimate relative efficiency of different processes

2. Numerical modeling
- saturation
- walkoff
- phase-modulation effects
- losses
- higher-order modes

Small-signal analysis: phase-matching

To derive phase-matching conditions:
use non-depleted growth of third harmonic as source for cascading

Example:

$$\frac{dE_4}{dz} = ik_4\chi^{(3)}E_2E_3(z)E_1^* \exp[i\Delta k_4 z]$$

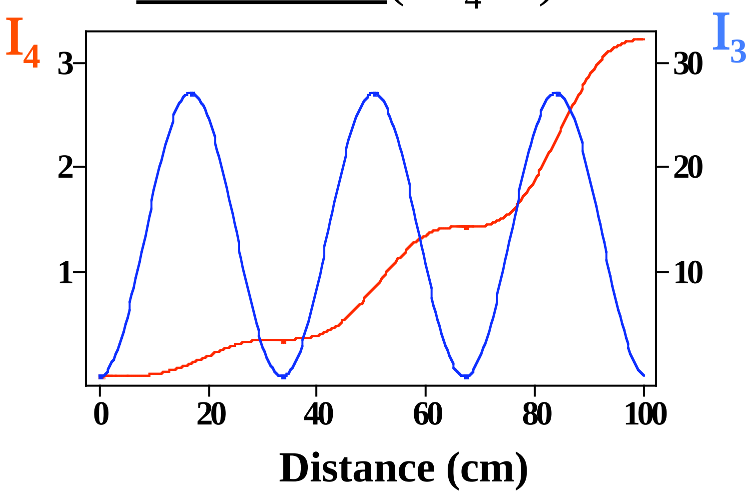
Where

$$E_3(z) = \frac{-2k_3\chi^{(3)}E_2^2E_1^* \exp[i\Delta k_3 z/2]\sin[\Delta k_3 z/2]}{\Delta k_3}$$

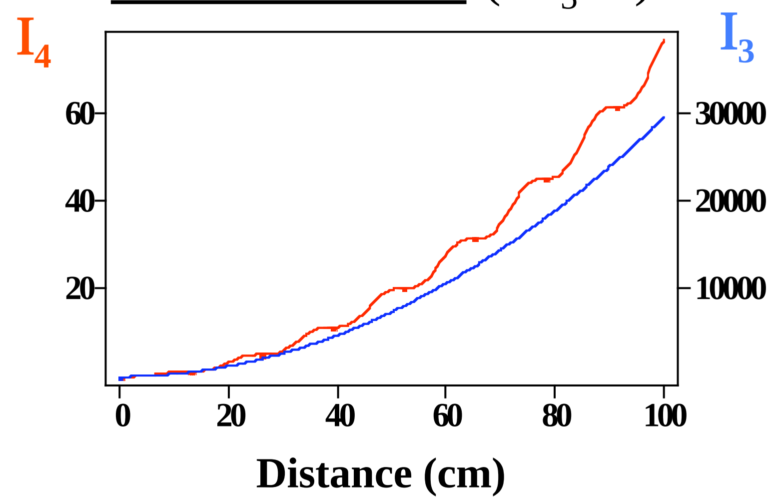
E_4 grows under several different phase matching conditions...

Cascaded phase-matching mechanisms

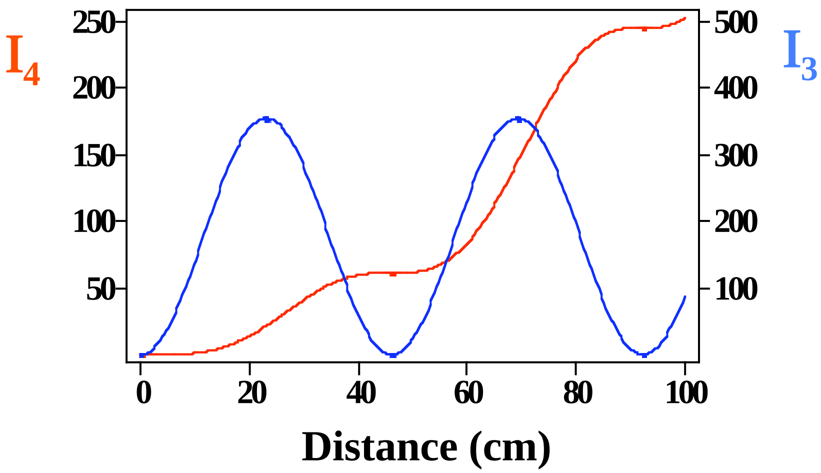
Standard (Δk_4=0)



Gain-assisted (Δk_3=0)



Cascaded QPM ($\Delta k_3 + \Delta k_4 = 0$)



Gain-assisted PM:
Increase in $E_3(z)$ gives buildup in E_4 in spite of phase mismatch

Cascaded QPM:
P_{NL} is modulated by $E_3(z)$ @ $1/\Delta k_4$

Cascaded vs. direct processes

For sufficiently long interaction length, cascaded low-order processes can dominate direct high-order.

Effective nonlinearity for cascading:

$$\chi_{eff}^{(5)} \approx \frac{2\pi\, k_3 \left(\chi^{(3)}\right)^2}{\Delta k_3}$$

Residual phase mismatch (Δk_3) is *small* in gases:

Example: for $\omega_4=\omega_2+\omega_3-\omega_1$ and $\Delta k_4+\Delta k_3=0$

$I_{direct}/I_{cascaded} \sim 10^{-5}$

Nonperturbative effects:

- χ does not drop off as steeply: plateau in high orders >7

Propagation code calculates saturated conversion

Input fields:
- energy, pulse duration, chirp, relative delay

Pressure loop

Propagation step loop: split-step + Runge-Kutta

Time domain:
- spm, xpm
- nonlinear mixing

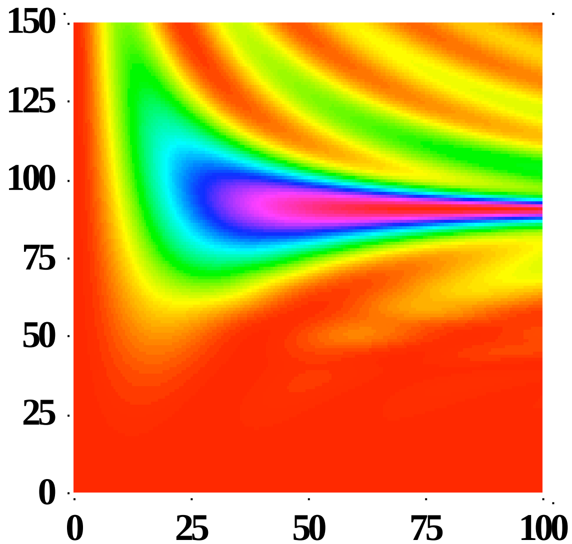
Frequency domain:
- dispersion, losses

Output processing:
- energy calculation
- post-compression

Major assumptions:
- *discrete-mode propagation*
- *five harmonic fields*
- *no bending losses*
- *no ionization*

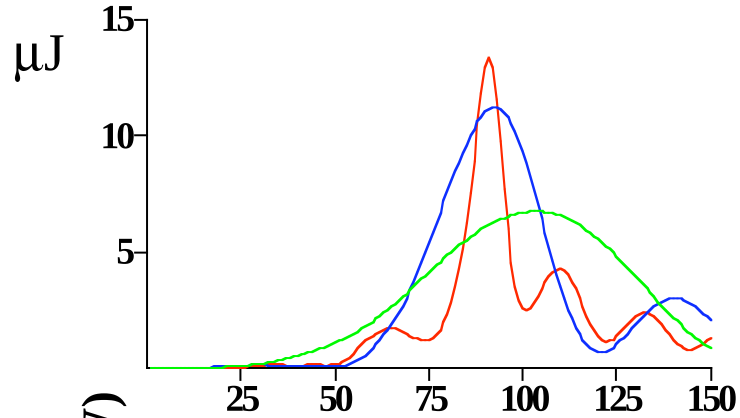
Code results: third harmonic

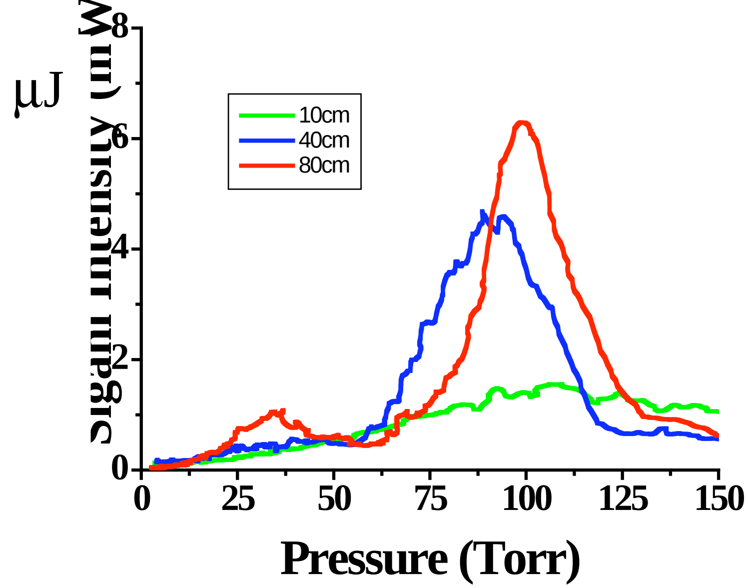
P (torr)



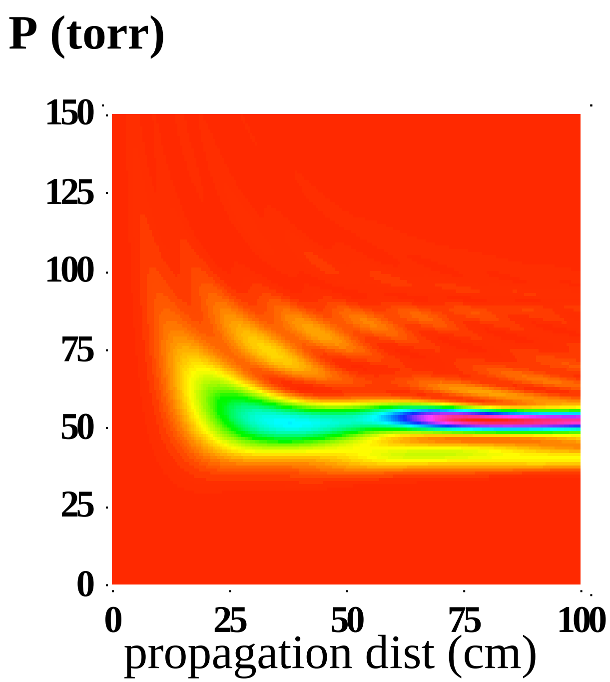
propagation dist (cm)

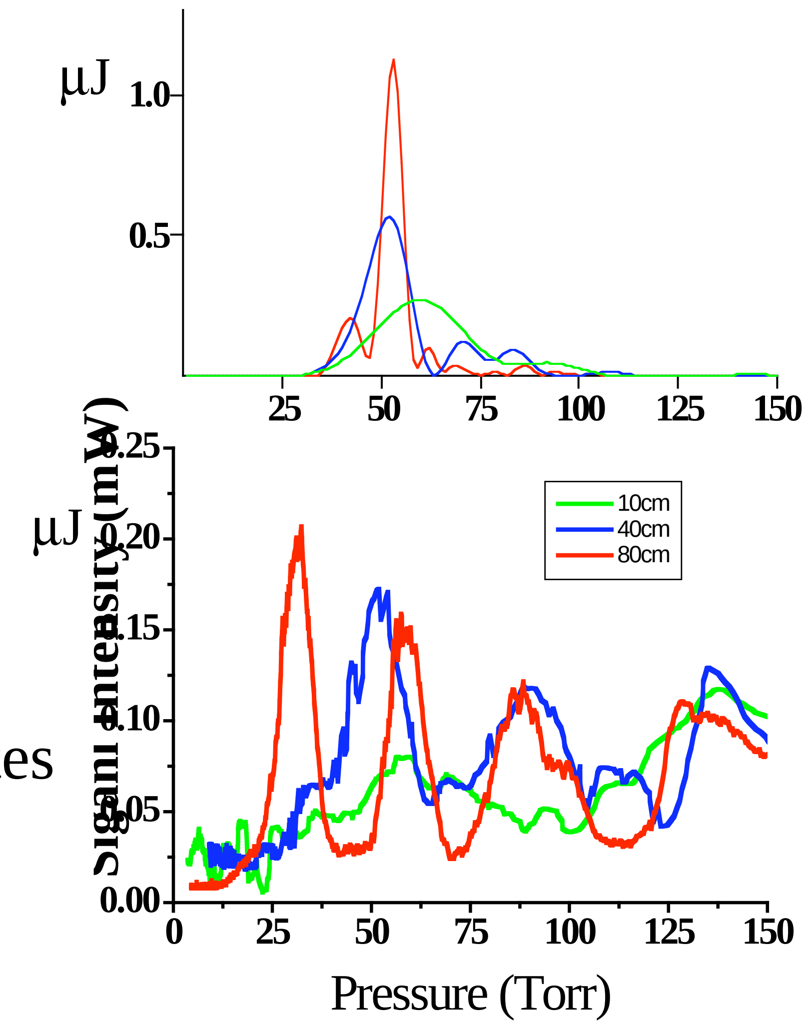
Low pressure:
peak broad, shifted high





Code results: Fourth harmonic





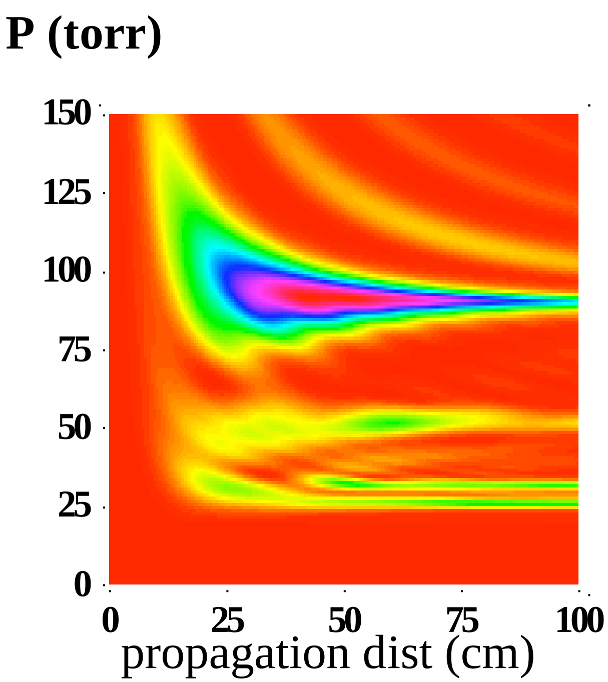
Code:

- cascaded QPM peak dominates

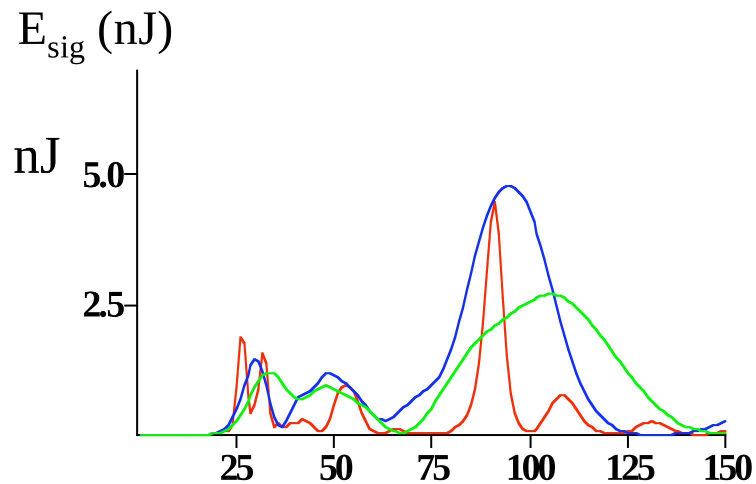
Measurement:

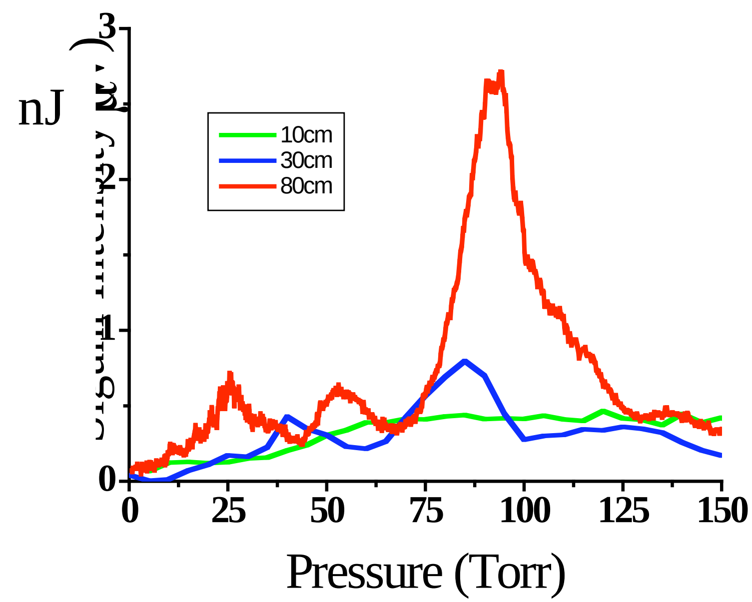
- all three important

Code results: fifth harmonic



Gain-assisted peak is strongest

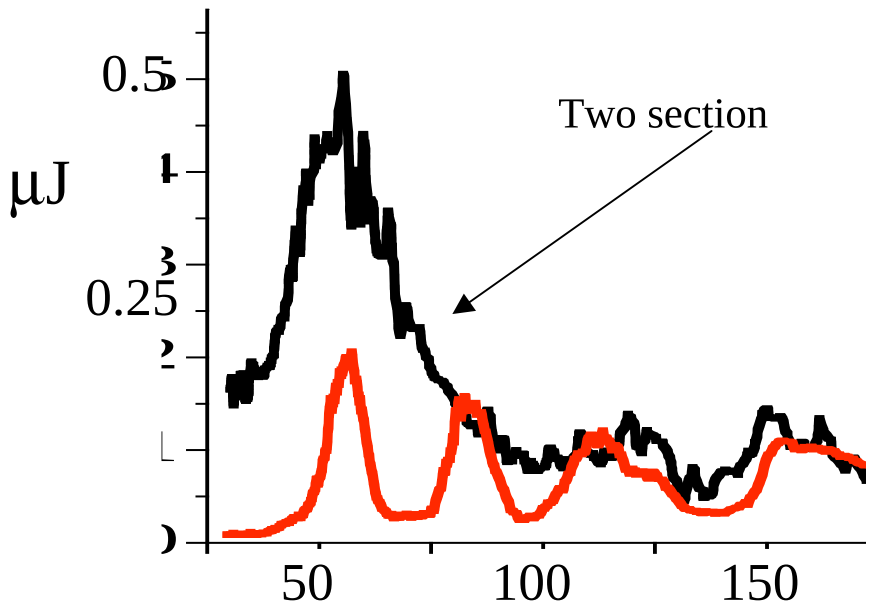




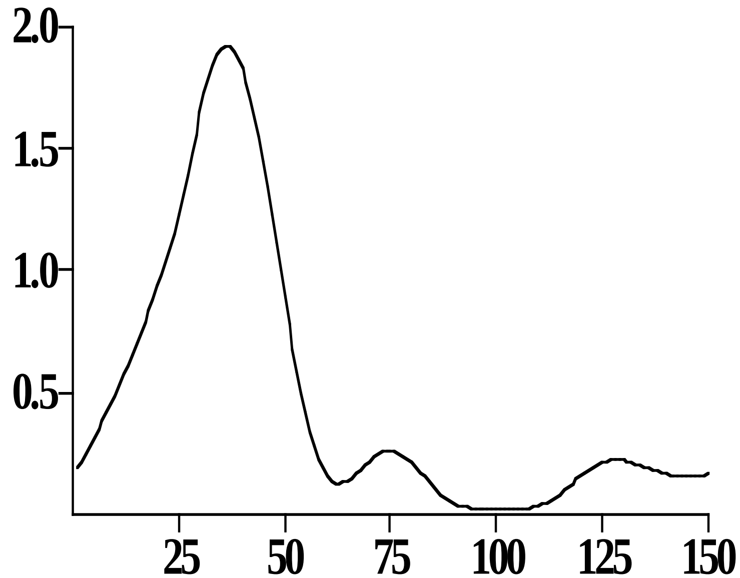
Two-stage generation

Improve cascading efficiency by using two stages:

1. Generate w3: $\Delta k_3=0$
2. Generate w4: $\Delta k_4=0$ (not CQPM)



Expt: 20cm+20cm

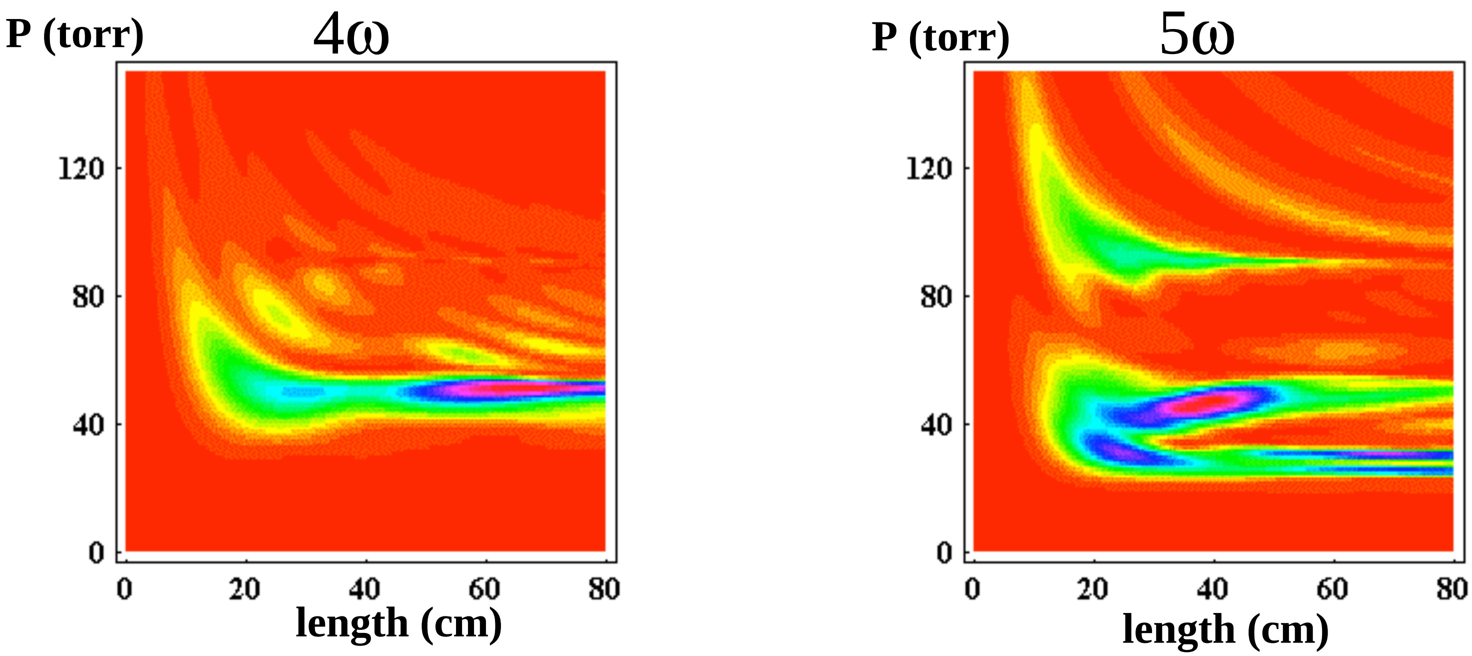


Calculation

Scaling to high conversion

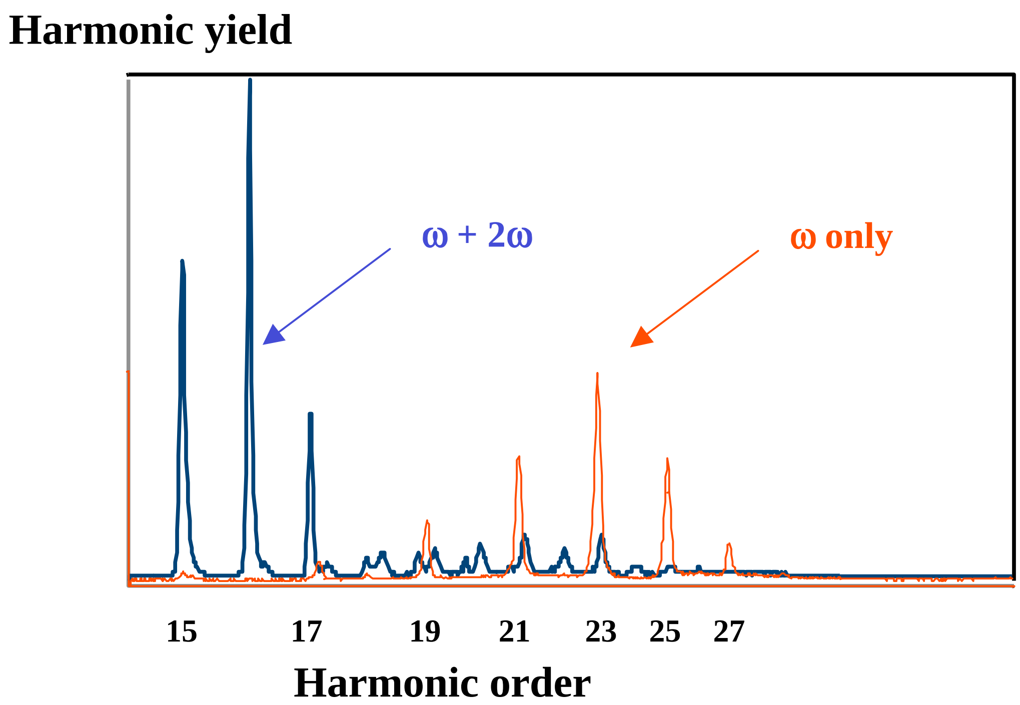
Department of Physics Colorado School of Mines

High intensity blue (400nm) drives conversion to 4ω and 5ω
- should approach ~20%, 2% conversion



At 150μJ (400nm), 10μJ (800nm), optimum 5ω shifts to peak 4w output (gain-assisted).

High-order cascading



3 cm capillary
150μJ @ω
90 μJ @2ω

Mixing ω and 2ω gives significant enhancement of HHG *inside absorption window*

Conclusions and future work

Summary:

- efficient upconversion to deep-UV, VUV, and XUV
- three types of phase-matching:
  - standard
  - cascaded quasi-phase-matching
  - gain-assisted phase-matching

Future work:

- improve efficiency of cascading
- extend to higher orders
- scale to higher pulse energies
- develop high power fs sources at 400nm and UV wavelengths