# Ultrashort Pulse Laser Filamentation & Nonlinear Effects in Optical Materials

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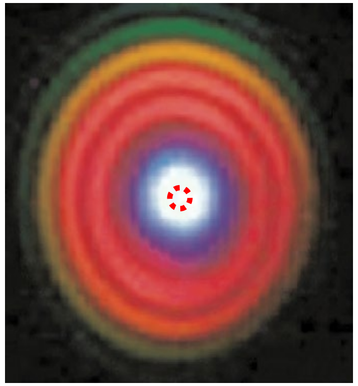
# Characterizing USPL Filamentation in Optical Materials

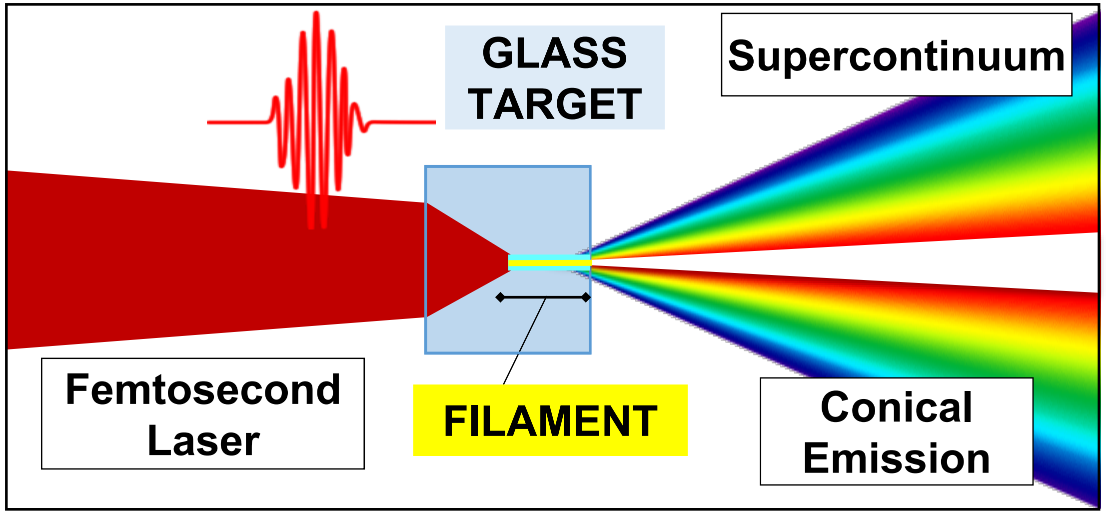


**BLUF:**

We measure ultrashort pulse laser (USPL) filamentation in transparent optical materials to characterize extreme nonlinear optical effects and begin to survey the broad parameter space of laser controls associated with the capability.



- **BACKGROUND & EXPERIMENTAL CONCEPT:**
  - Exploiting nonlinear optical effects of high-intensity USPL propagation
- **RESULTS:**
  - Calibrated measurements of filament emission and nonlinear effects
  - Experimental parameter dependencies and minimum thresholds
- **CONCLUSIONS AND OUTLOOK**



# Motivation & Context

- **There has been significant, rapid development of USPL technologies in recent years -** transitioning from experimental systems to reliable tools for industry and defense.
- **ARL is developing techniques and uncovering fundamental physics to understand and exploit the unique capabilities of USPL to aid the Service member.**
- **Propagation of USPL through nonlinear media produces wide array of interesting optical effects.** Much work has been done on filamentation of 800-nm lasers in air [ref 1-3], but high intensity propagation is still poorly understood, especially in transparent solids or at longer IR wavelengths [ref 4-6]. **ARL has the capabilities to study these effects.**
- **This ongoing project is a coordinated effort with internal and external participants -** joint experimental efforts with ARL/WMRD, C5ISR, and joint CA with the U. of AZ for numerical modeling efforts.

**What do we mean by "ultrashort"?**

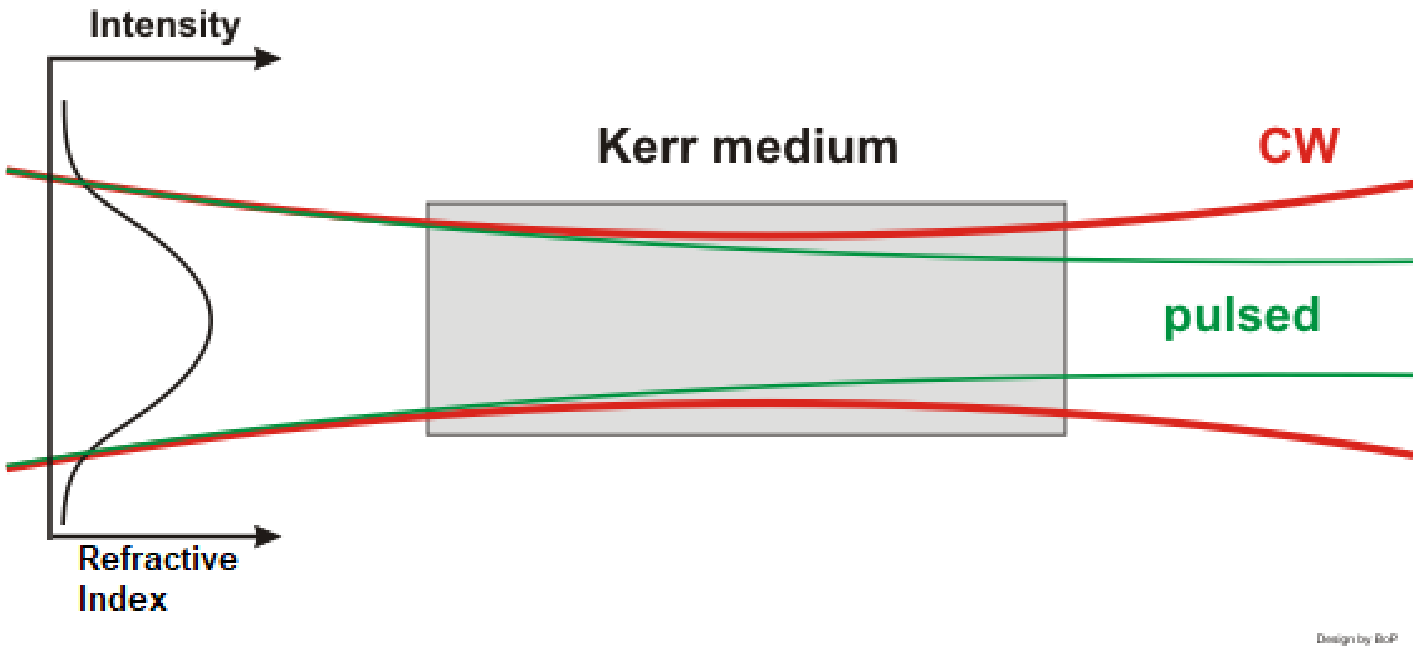
- USPLs produce extremely short EM bursts.
  - 10-100 femtoseconds
  - 1 fs = millionth of billionth of 1 second
  - Timescale of atomic motions
- Condense moderate energy into such short time generates IMMENSE peak intensities.
  - Hundreds of terawatts per $cm^2$!
- At these intensities, light interacts with matter in entirely novel ways.


References (by DOI)

1) 10.1134/S1054660X11190054
2) 10.1063/1.4896722
3) 10.1088/0953-4075/48/9/094005
4) 10.3952/physics.v57i3.3541
5) 10.1103/PhysRevE.72.037601
6) 10.1364/JOSAB.36.000SG1

# Filamentation, Conical Emission, and Supercontinuum Generation

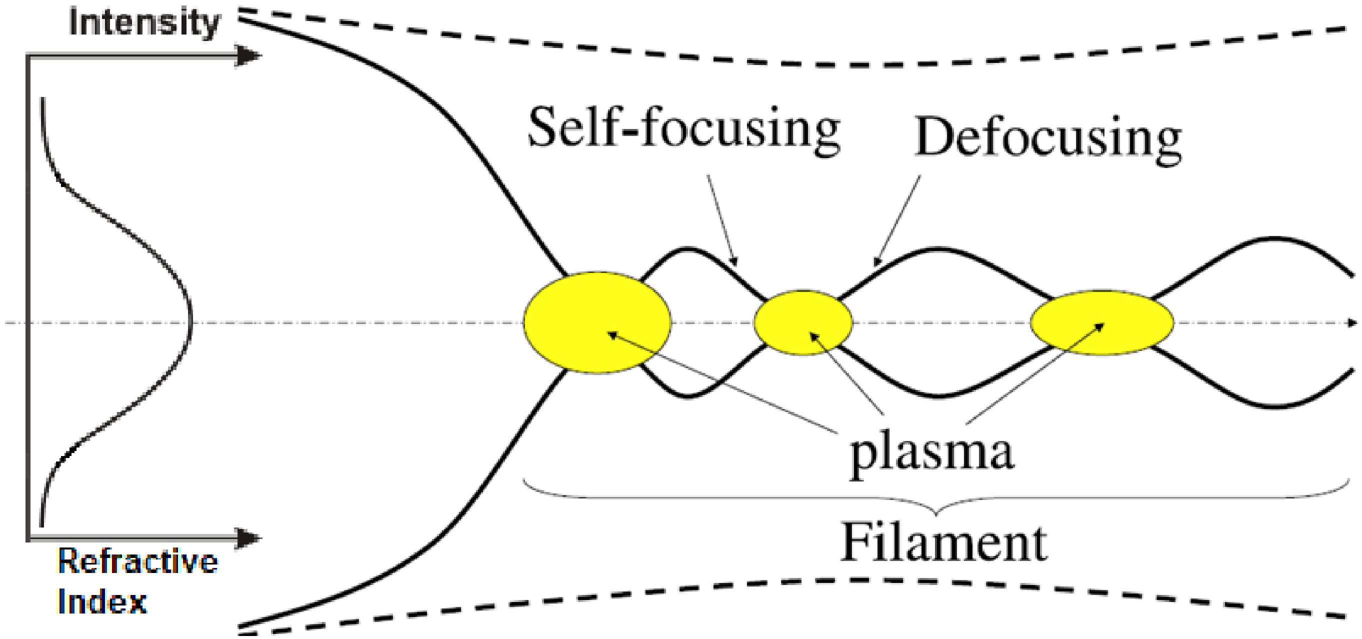
## Nonlinear Index of Refraction → Self-Focusing




- Adapted from: BoP (talk) (Uploads), CC BY-SA 3.0, https://en.wikipedia.org/w/index.php?curid=628504

# Filamentation, Conical Emission, and Supercontinuum Generation

## Self-Focusing → Filamentation



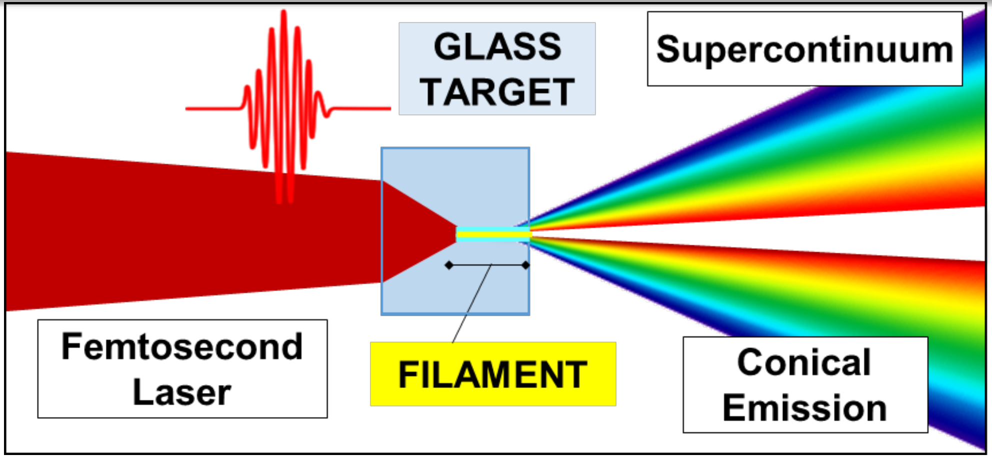

$$P_{Cr} = \frac{3.77\,\lambda_o^2}{8\pi n_0 n_2} = \sqrt{\frac{4\ln 2}{\pi}}\,\frac{E_{pulse}}{\tau_{FWHM}}$$

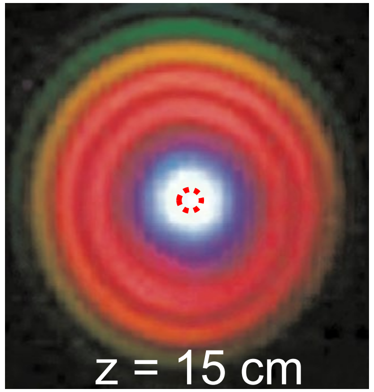
$$z_C \approx 0.367 z_{Lin}\left[\left[\sqrt{P_{in}/P_{Cr}} - 0.852\right]^2 - 0.0219\right]^{-1/2}$$

- Adapted from: Couairon, Mysyrowicz, “Femtosecond Filamentation in Transparent Media,” Physics Reports 441 (2007)

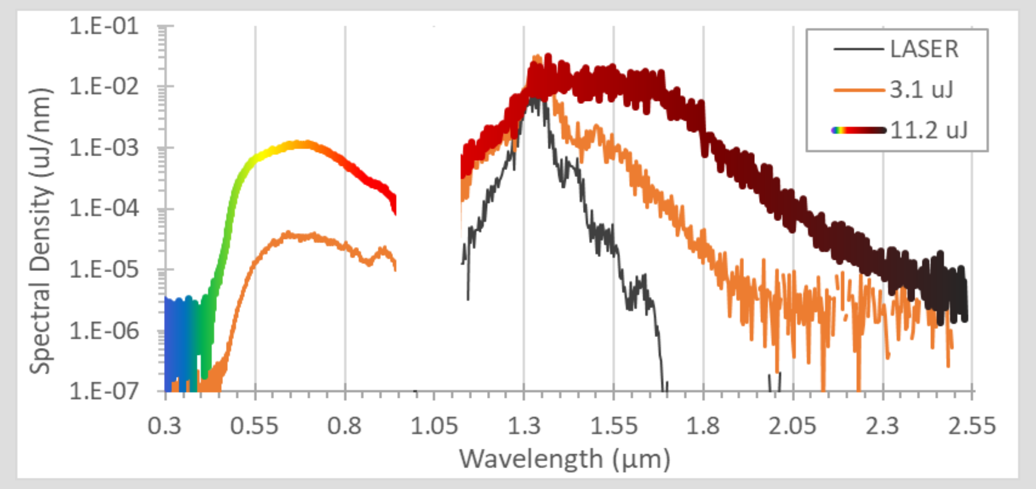
# Filamentation, Conical Emission, and Supercontinuum Generation

## Filamentation → Supercontinuum Generation + Conical Emission



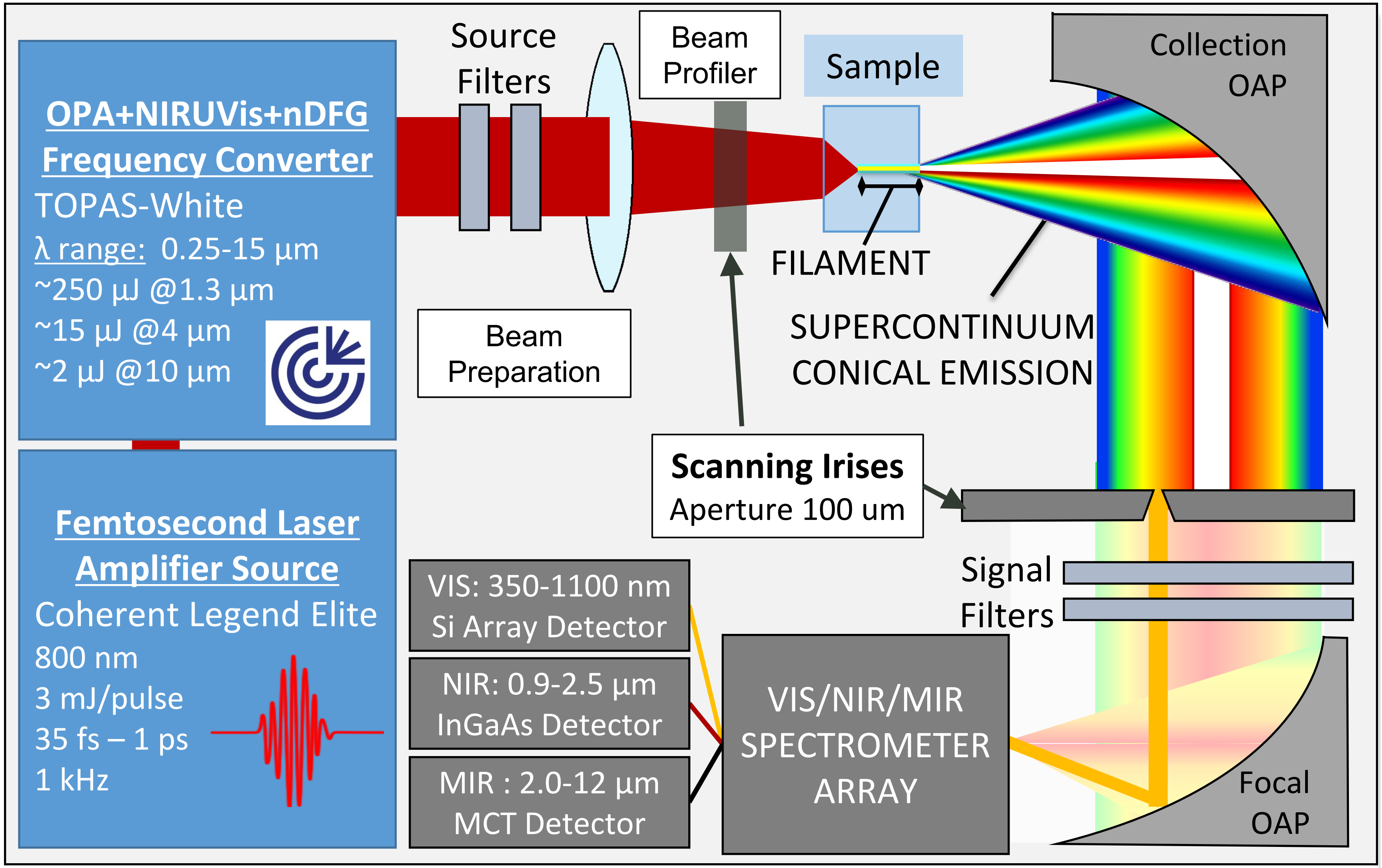

**Long-Distance, High-Intensity Propagation Enables Strong, Nonlinear, Optical Interactions**
Self-Phase Modulation, 4-Wave Mixing, Stimulated Raman, Harmonic Generation

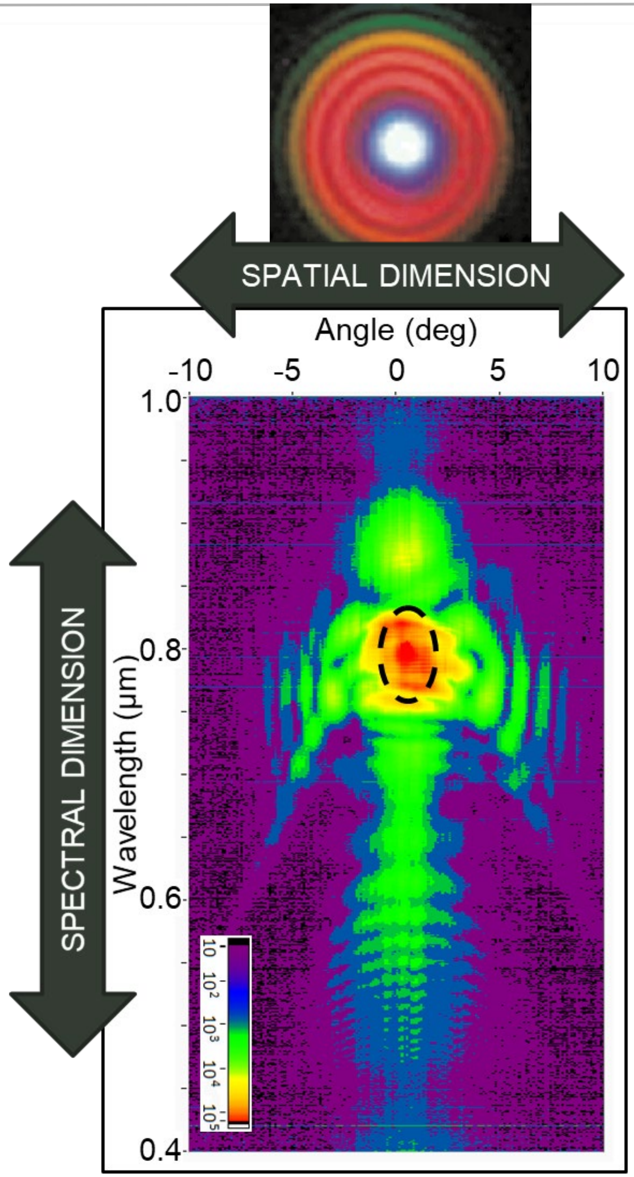



**Supercontinuum in Sapphire Glass With 1.3 um OPA Source**

- Spectrum continuously spans UV – beyond NIR

# Experimental Diagram



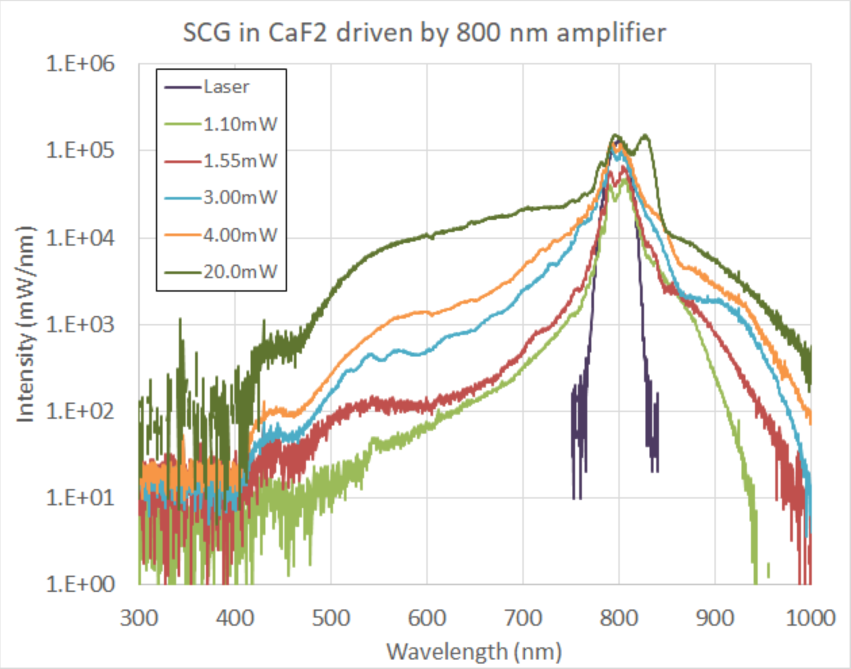
# Optical Materials of Interest

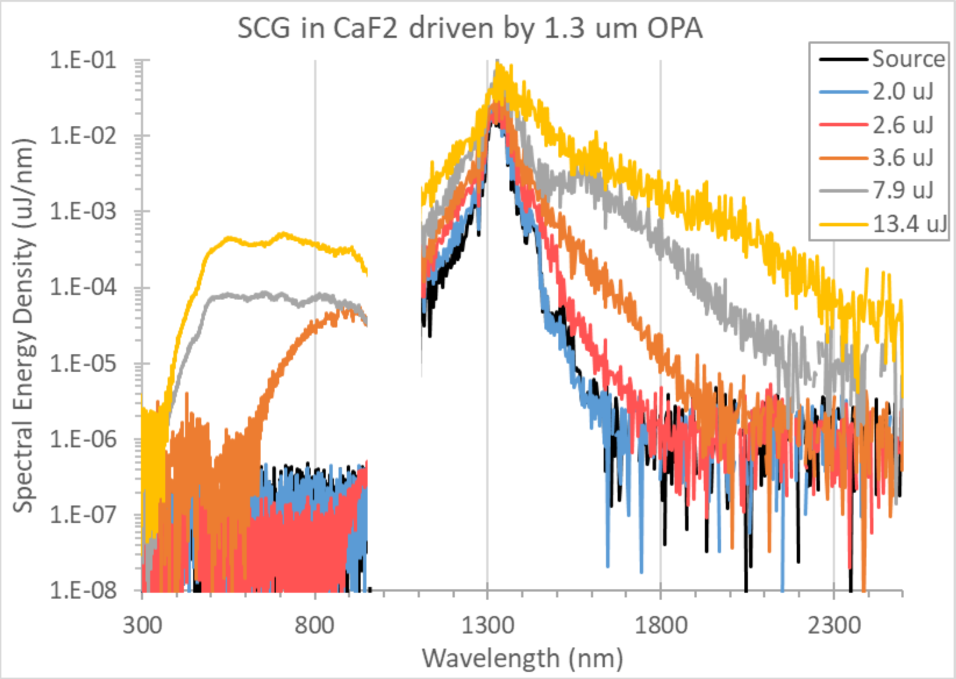
| | Sapphire | $CaF_2$ | $MgF_2$ | ZnSe |
|---|---|---|---|---|
| Transmission Range (um) | 0.15 - 5 | 0.18 - 8 | 0.2 - 6 | 0.55 - 20 |
| n Index of Refraction | 1.75 | 1.42 | 1.37 | 2.51 |
| $n_2$ Nonlinear Index (800 nm) (x10^-20 m2/W) | 3.1 | 1.9 | 0.92 | 166 |
| CRITICAL POWER (800 nm) (MW) | 1.75 | 3.5 | 7.6 | 0.0218 |
| n2 Nonlinear Index (1350 nm) (x10^-20 m2/W) | 2.8 | 1.9 | 0.92 | 58.4 |
| CRITICAL POWER (1350 nm) (MW) | 5.6 | 10.1 | 21.7 | 0.165 |

**Upcoming Materials of Interest:**

- Silicon & germanium
- Sample quality effects: crystalline vs. polycrystalline, “research grade” vs. production grade
- More chalcogenides and other specialty materials for IR

# Supercontinuum Generation in $CaF_2$

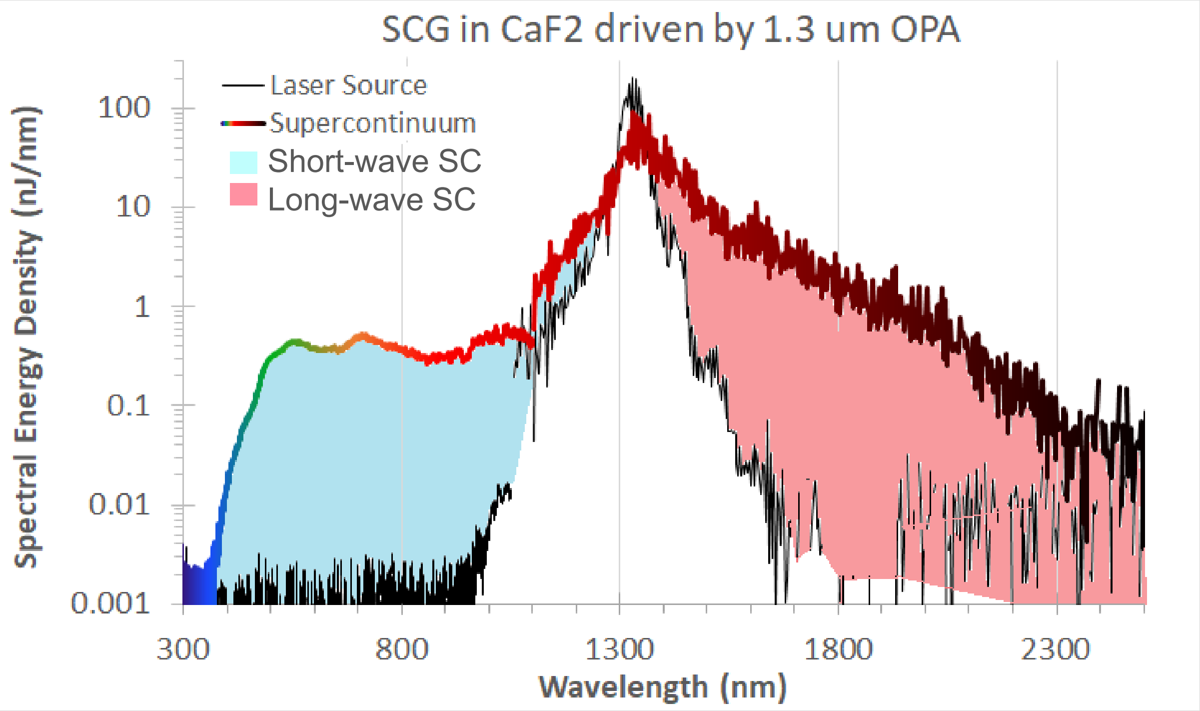


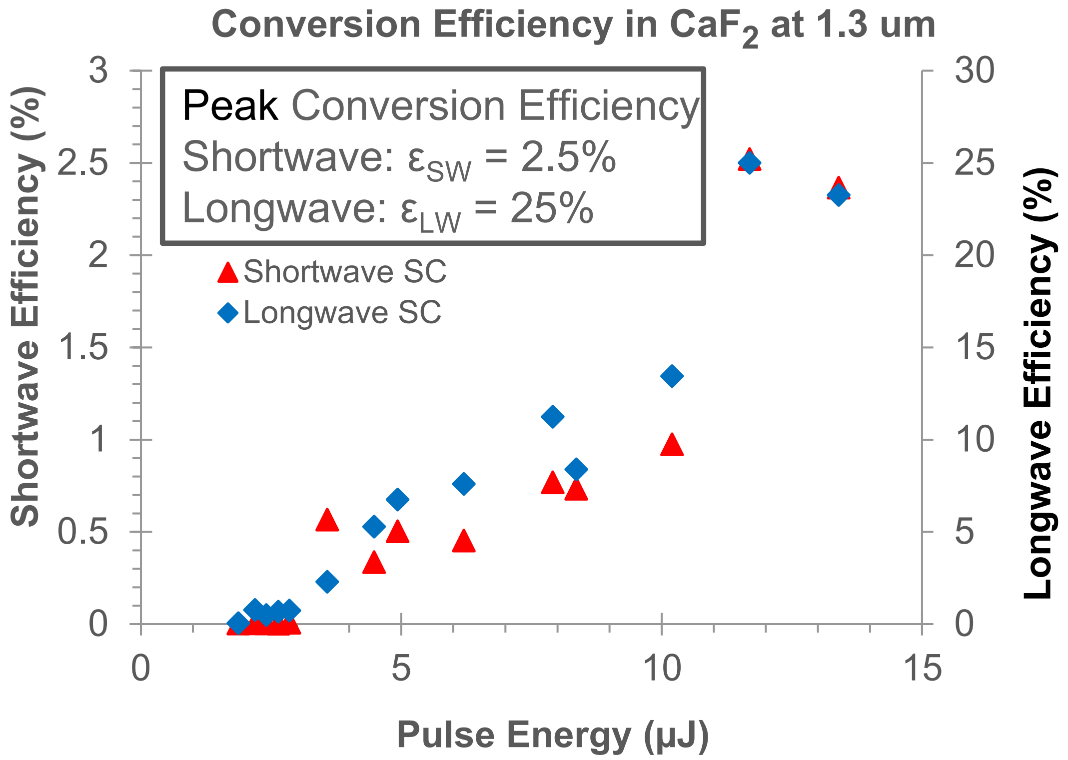




**Optical-grade $CaF_2$ sample, thickness: 4 mm**

- SC is broad and flat in shortwave, exponentially decreasing in longwave.
- Shortwave edge (λ~400 nm in $CaF_2$) target dependent. No longwave edge observed.
- Conversion efficiency: ratio of energy shifted to out-of-band wavelengths.
  - 1.3-µm source – max measured longwave conversion up to 25%.

# Supercontinuum Generation in $CaF_2$

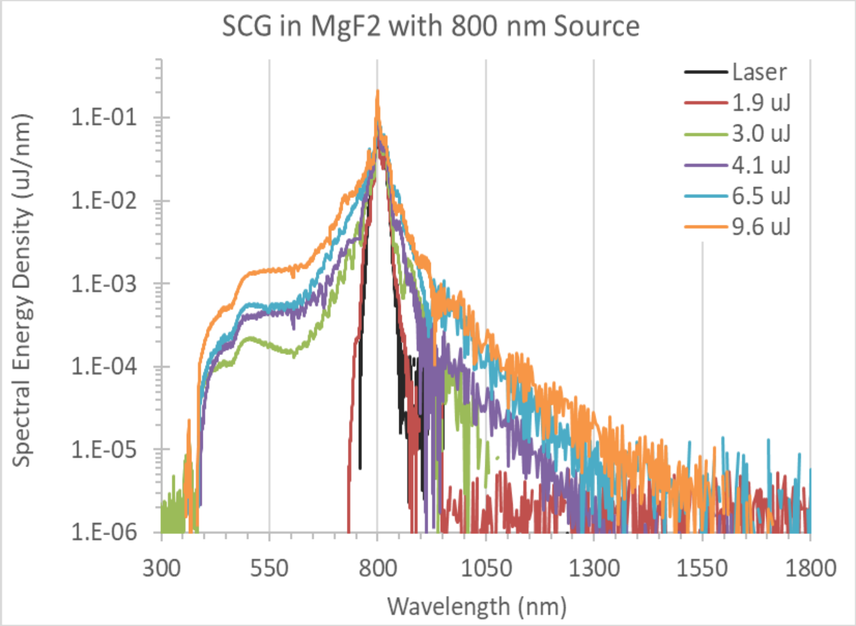


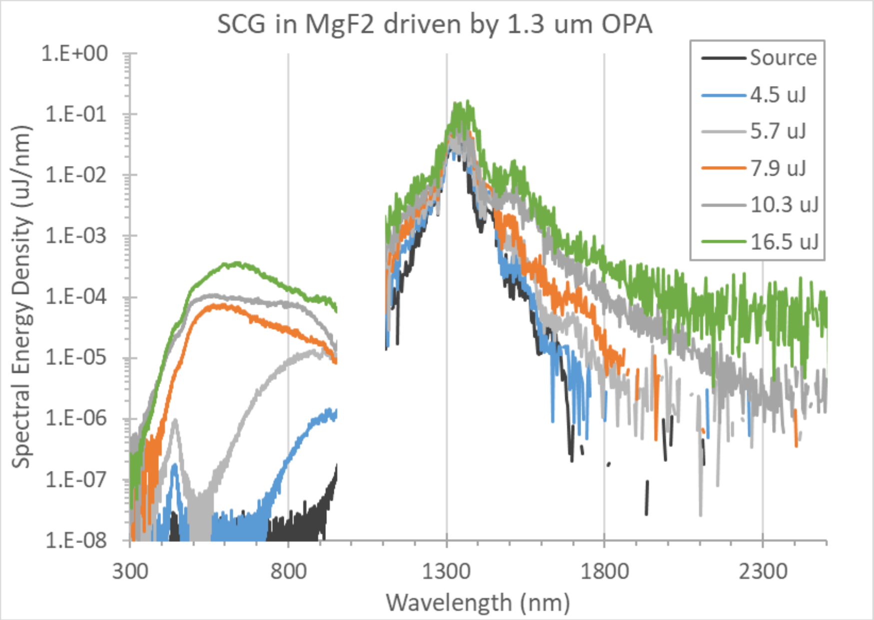



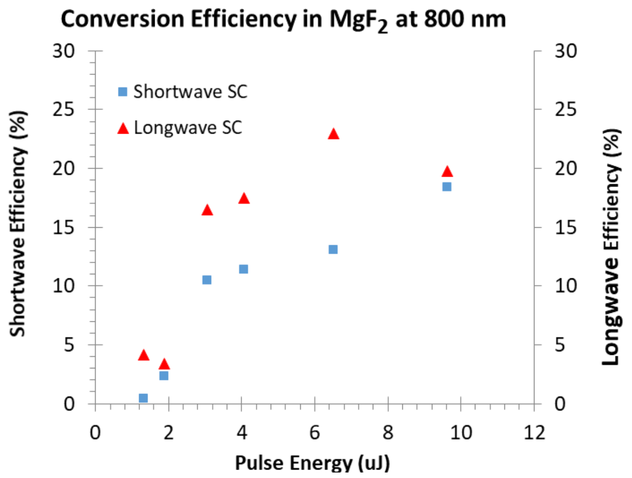

Track integrated energy transferred to supercontinuum – determine minimum threshold pulse energy for significant SCG.

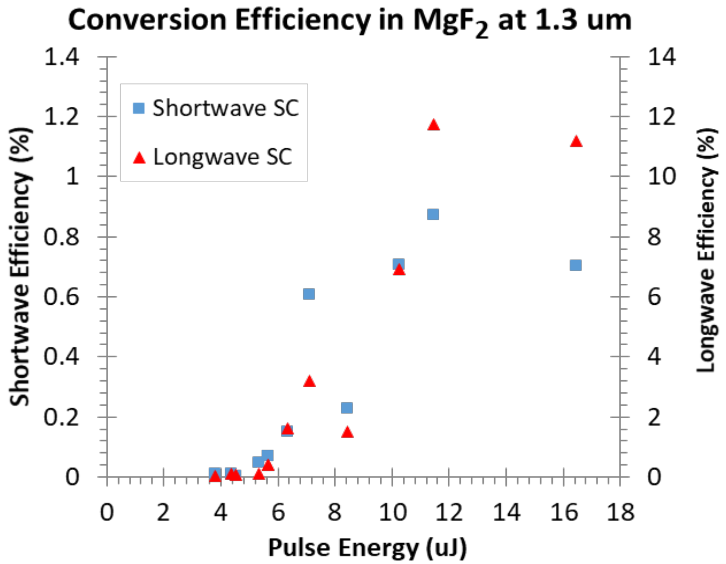
- For $CaF_2$, at 1.3 um $E_{Thresh}$ = 2.07 μJ.
  at 0.8 um $E_{Thresh}$ = 0.66 μJ.
- Long wavelength conversion ~10x stronger than short wavelength SC.
- At higher input energy, SCG begins to saturate. Sample damage limits maximum energies.

# Supercontinuum Generation in $MgF_2$



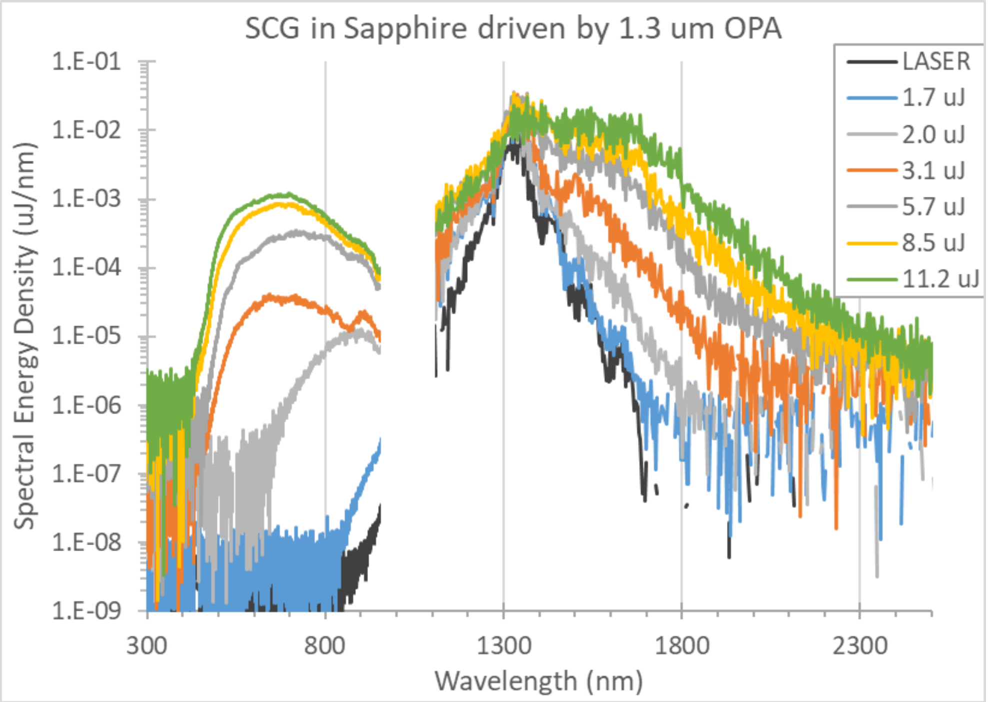



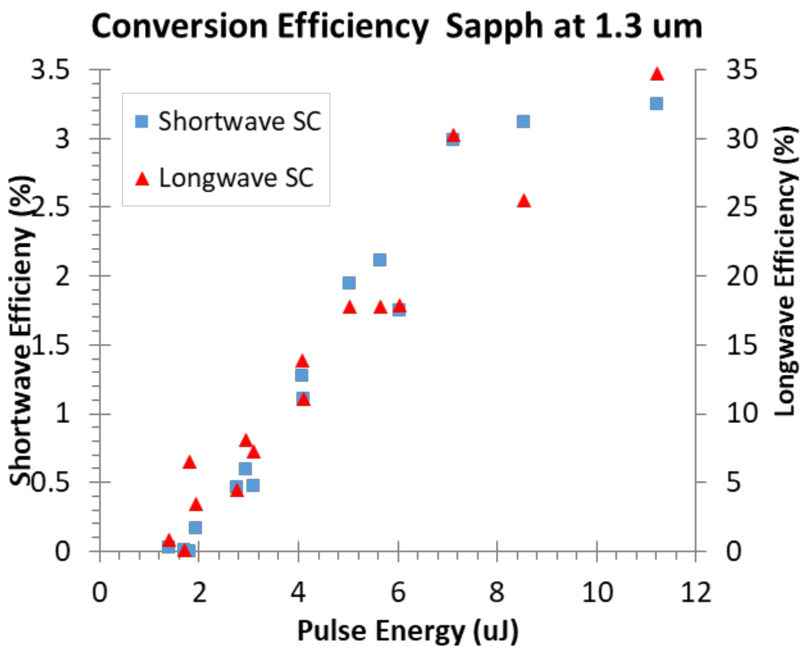




**<u>Optical-grade $MgF_2$ sample, thickness: 4 mm</u>**

- Higher threshold pulse energy:
  at 0.8 um $E_{Thresh}$ = 1.9 μJ; at 1.3 um $E_{Thresh}$ = 4.4 μJ.
- Lower conversion efficiency:
  Shortwave: $\varepsilon_{SW}$ = 0.77%.
  Longwave: $\varepsilon_{LW}$ = 12%.

# Supercontinuum Generation in Sapphire

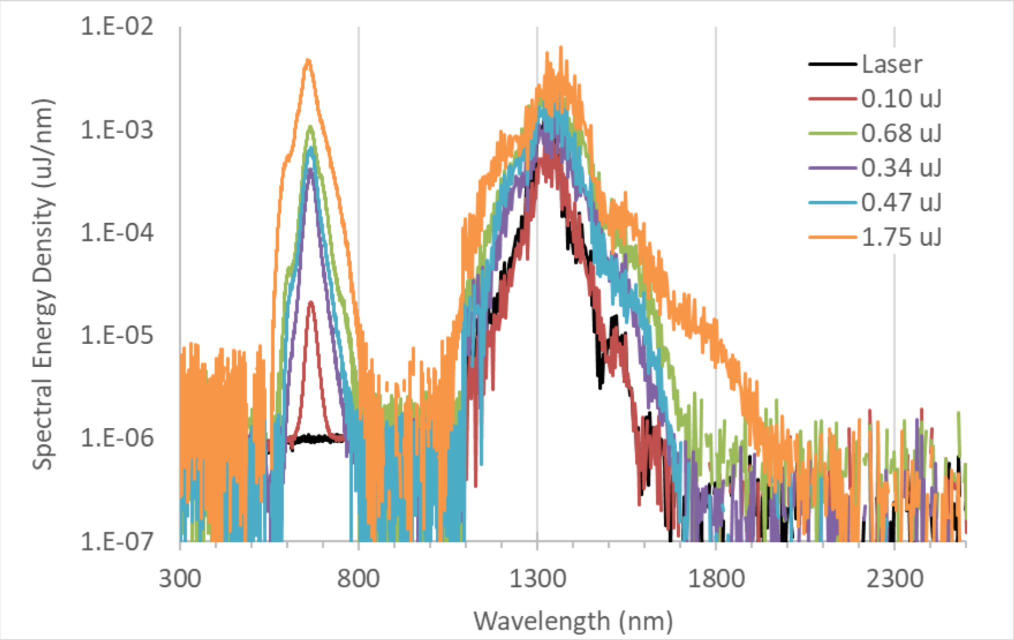


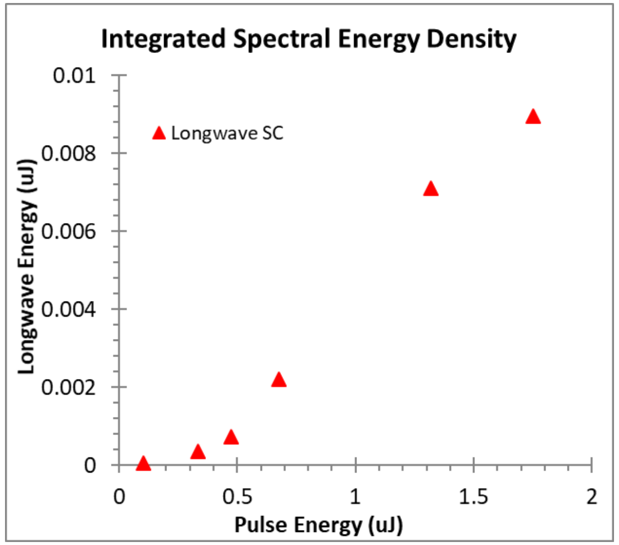



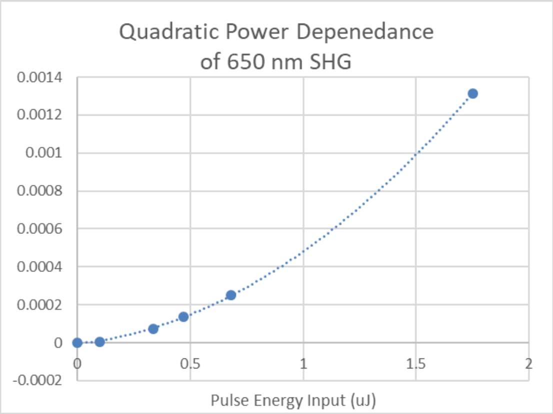

- **<u>Optical-grade sapphire (random-cut), thickness: 4 mm</u>**
  - $E_{Thresh}(0.8\ \mu m) = 0.30\ \mu J$.
  - $E_{Thresh}(1.3\ \mu m) = 1.5\ \mu J$.
- **Sapphire shows the highest SCG efficiency.**
- Max SC conversion efficiency: shortwave: $\varepsilon_{sw} = 3.3\%$.
  longwave: $\varepsilon_{LW} = 35\%$.

- SC spectral energy density comparable to primary source at highest powers.
- Growth of SC intensity more consistent: no discontinuous jumps when multiple filaments form.

# Supercontinuum Generation(+SHG) in ZnSe




**<u>Polycrystalline ZnSe, thickness: 4 mm</u>**

ZnSe undergoes SCG <u>and</u> SHG/TPA with 1.3 um source.

- Particularly low critical power due to high $n_2$. *However,* SCG threshold $E_{thresh}$= 0.075 uJ.

  Much higher multiple of $P_{crit}$ (~8.54x) than other samples.

<u>Competing Optical  Processes</u>

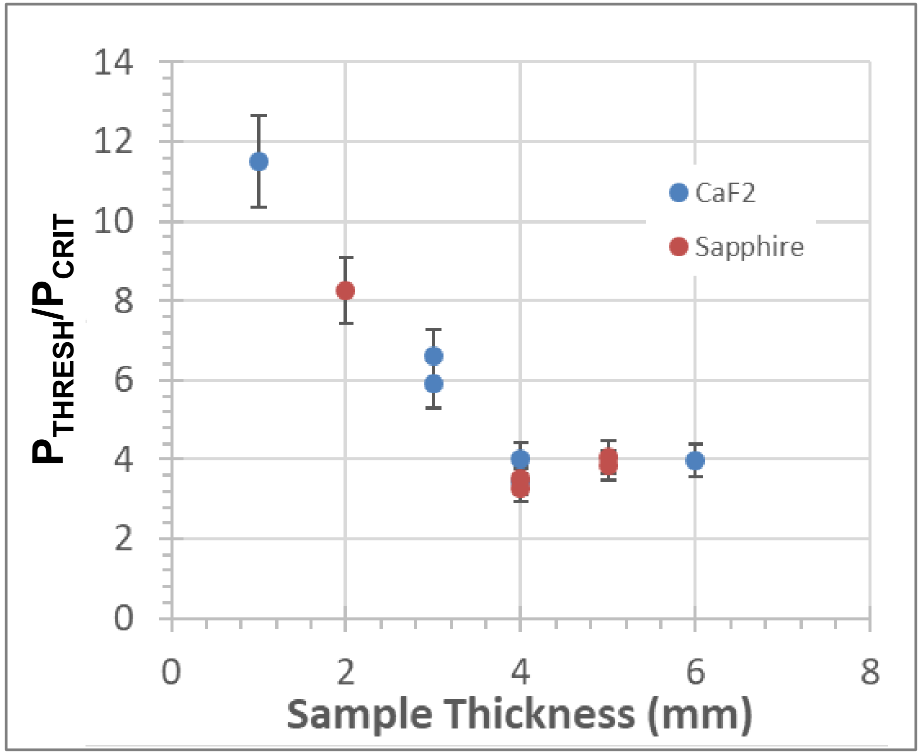
- Nonlinear absorption edge halts shortwave SC.
- $X_2$ polarizability – efficient second harmonic generation.
  - SHG overwhelms SCG.
- Expect third harmonic gen. at 433 nm, re-absorbed during propagation.

# Threshold Pulse Energy

## Conserved Trends in Threshold Pulse Energy

- Threshold energy scales with predicted $P_{CRIT}$ with ~4-5x scalar for finite length and experimental geometry.
- ZnSe has much higher scalar to threshold energy.
  - Attributed to TPA/SHG interference.

| Material | $E_{Thresh}$ (μJ) | | $P_{THRESH}/P_{CRIT}$ | |
|---|---|---|---|---|
| | λ=0.8 μm τ=35 fs | λ=1.3 μm τ≈50 fs | λ=0.8 μm τ=35 fs | λ=1.3 μm τ≈50 fs |
| **$MgF_2$** | 1.92 | 4.4 | 5.0x | 4.1x |
| **$CaF_2$** | 0.66 | 2.1 | 4.4x | 4.1x |
| **Sapph** | 0.30 | 1.5 | 4.0x | 4.6x |
| **ZnSe** | N/A | 0.075 | N/A | 8.5x |



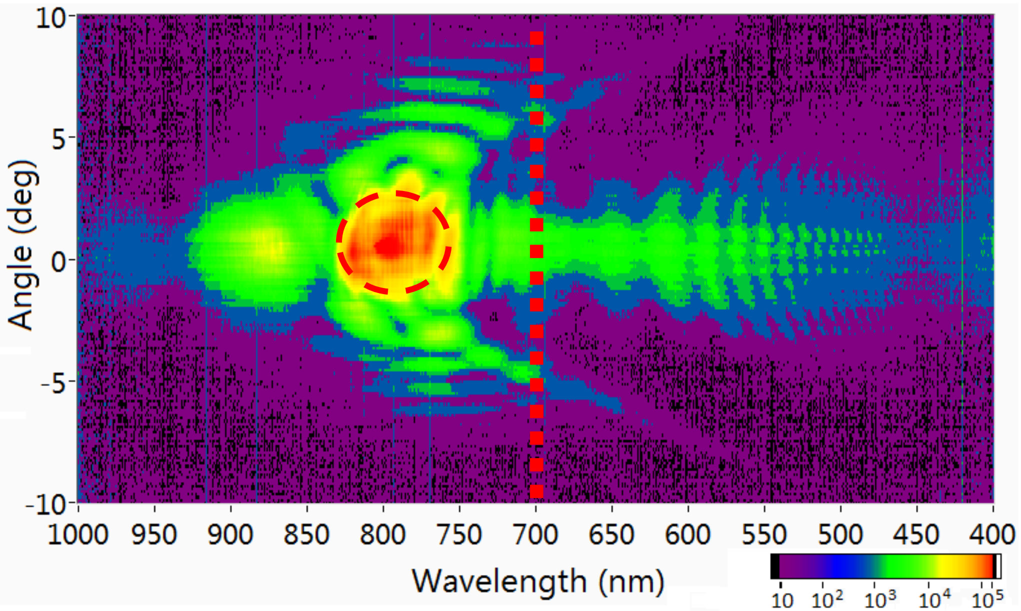

## Sample Thickness Dependence:

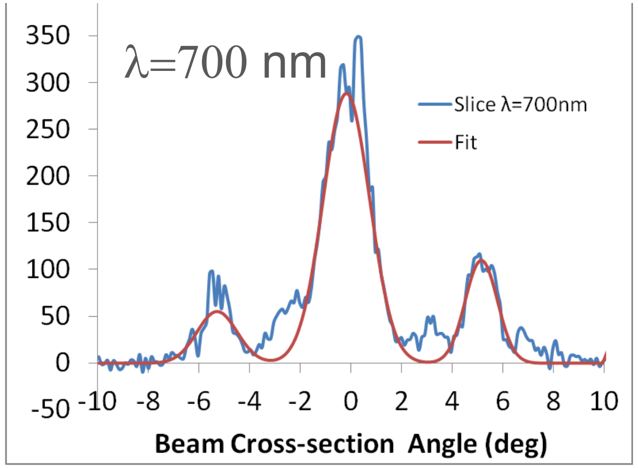
- $E_{THRESH}$ inversely related to target thickness (shorter interaction requires higher peak power).
- Thinner samples generate wider angle of conical emission.
  - Attributed to relaxed phase-matching conditions in rapid collapse of focus to filament.

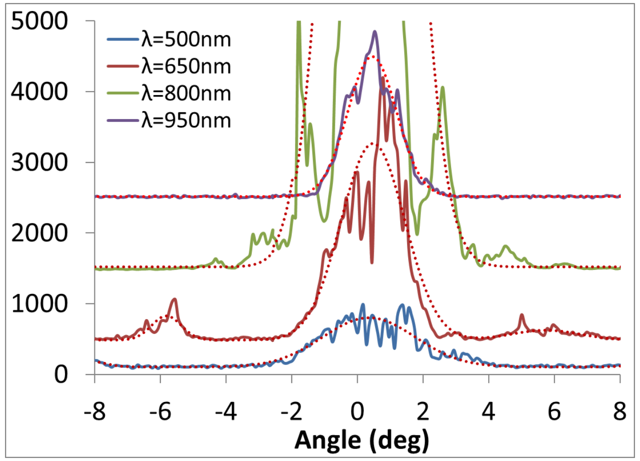
# Angle-Resolved Spectra of Filaments

**Supercontinuum emitted in broad angular cone with prominent spatial structure:**

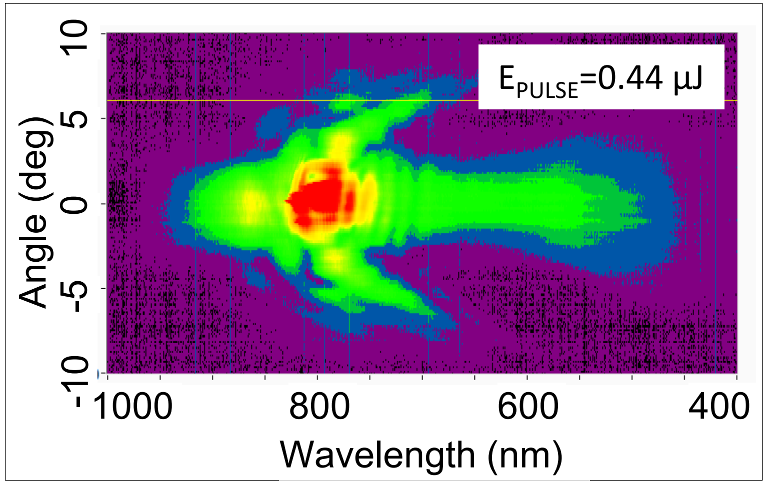
- $CaF_2$ target, λ=800 nm, $E_P$=1.4 uJ, divergence ~6.3°
- Original laser marked in dashed outline
- Angular structure due to phase-matching nonlinear processes in filament and interference of multiple filaments
- (Below): Integrating 10-nm stripe at select wavelengths to fit beam profile and extract angular width of conical emission

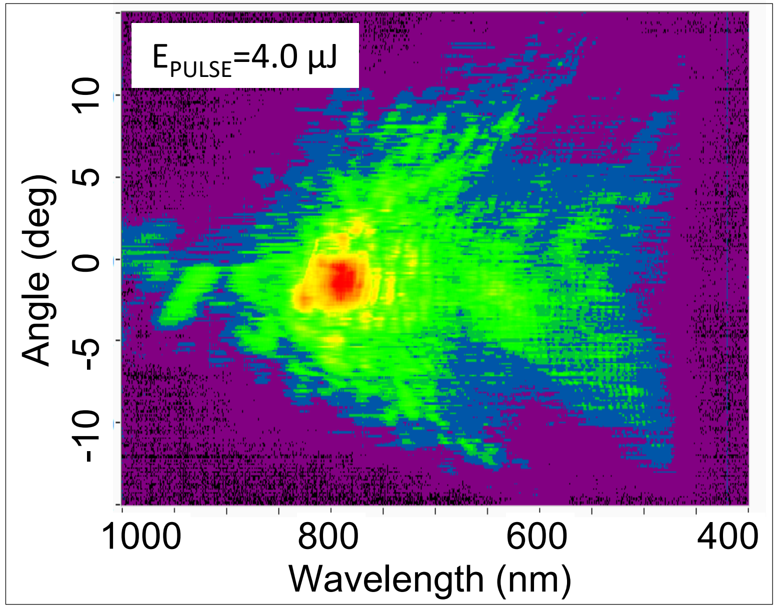


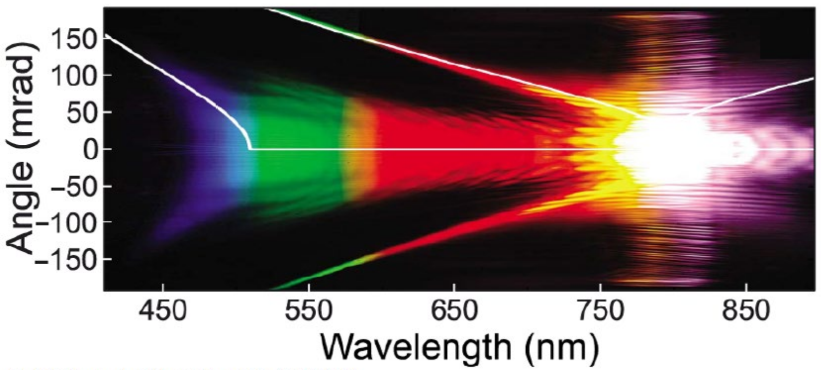



# Angle-Resolved Spectra of Filaments




Dubietis et al., Lith. J. Phys. 57, 113-157 (2017).

## Single vs. Multiple Filamentation

- Angle-resolved SCG spectra from filamentation of 800-nm beam in 6-mm sapphire at 0.44 µJ and 4.0 µJ.
- Spatially-resolved structure reveals drastic difference between single and multiple filamentation domain.
- More intense supercontinuum generation along with broader conical emission, as well as strong fringing resulting from interference between multiple filaments.

## Compare to Simulations and Similar Measure in Literature:

- Angle-resolved SCG in sapphire from 800 nm source; white curves indicate best fits to “X-wave” propagation model.

# Conclusions

- **Observed and characterized USPL filamentation in selection of mission-relevant optical materials.**
- **Broad survey of measurements to determine key laser parameters:**
  - Determine threshold pulse energy for observable filament effects.
  - Chart how effects scale with laser power, wavelength, and sample geometry.

## ONGOING RESEARCH

- **Longer wavelength laser source:**
  - OPA with attachments can span UV to FIR, with sufficient energy to see filamentation effects.
- **Expand library of materials and control space of parameters studied.**
- **Incorporate simulations to inform experiments and use experimental results to check and enhance models of ultrashort pulse laser filamentation.**
  - Ongoing collaborative agreement with professors J. Moloney and M. Kolesik (University of Arizona) to simulate propagation at high intensities in transparent materials.

**This work is a coordinated effort with multiple teams across CCDC-ARL and other organizations:** would like to acknowledge and thank T. Ensley, W. Shensky, Andrew Mott, Paul Pellegrino, S. Roberson, T. Valenzuela, L. Vanderhoeff, A. Schweinsberg, and many others for assistance, guidance, and support.

# Thank You for Your Attention