How to measure sub-7 fs laser pulses using second-harmonic nanocrystals

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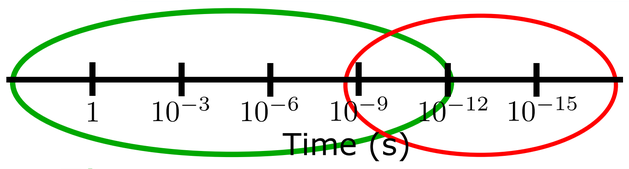
Abstract

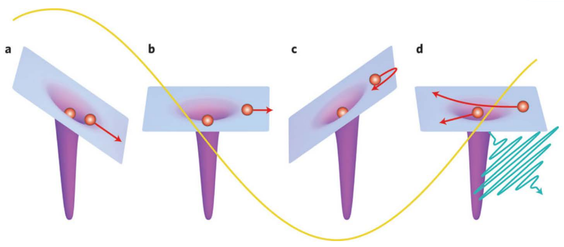
1. We demonstrate the measurement and retrieval of **sub-7 fs** laser pulses using nanocrystals (NCs).
2. The temporal measurement is based on dispersion-scan (**d-scan**) in systems which generate second harmonic (SHG).
3. Retrieval is possible by implementing a **new algorithm**, which takes **scattering** into account, and also identifies coherent and incoherent contributions to the signal.
4. Whe show that dielectric NCs are a **cheap**, and **wavelength-independent** alternative to expensive and specifically-designed bulk crystals to perform temporal measurements of **ultrafast light pulses**.

1. Shortest measurable events



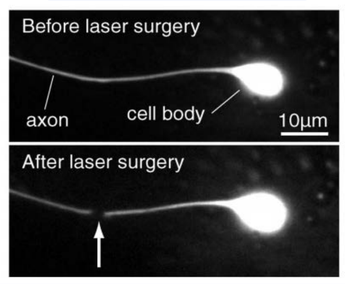
This pulse at 800 nm lasts ~6 fs
The field oscillates only twice!

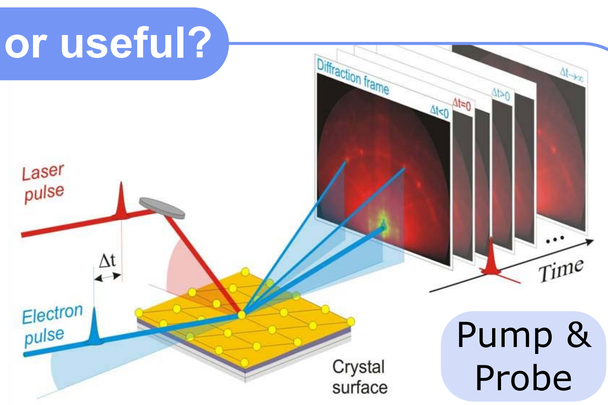
2. Are they important or useful?



High Harmonic Generation

Neuro-surgery

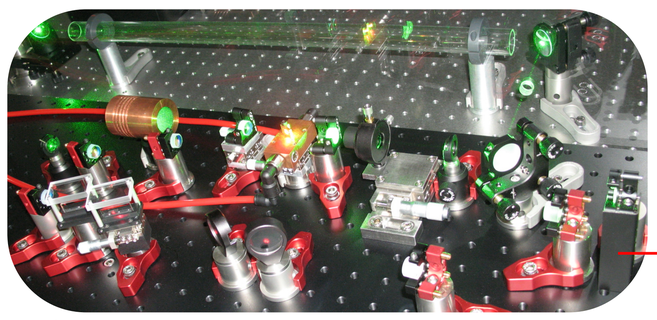




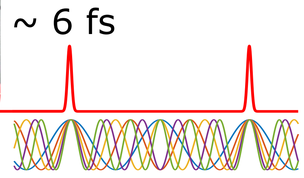
Pump & Probe

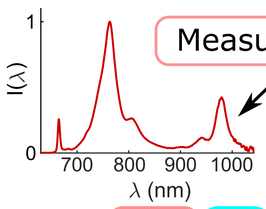
3. We generate them

Our laser: house-made broadband Ti:Sa oscillator

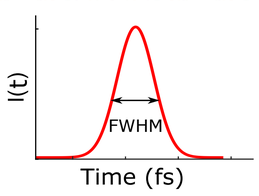


@ 800 nm
1 nJ/pulse
76 MHz rep. rate
~ 6 fs





Measured from our laser



Fourier Transform

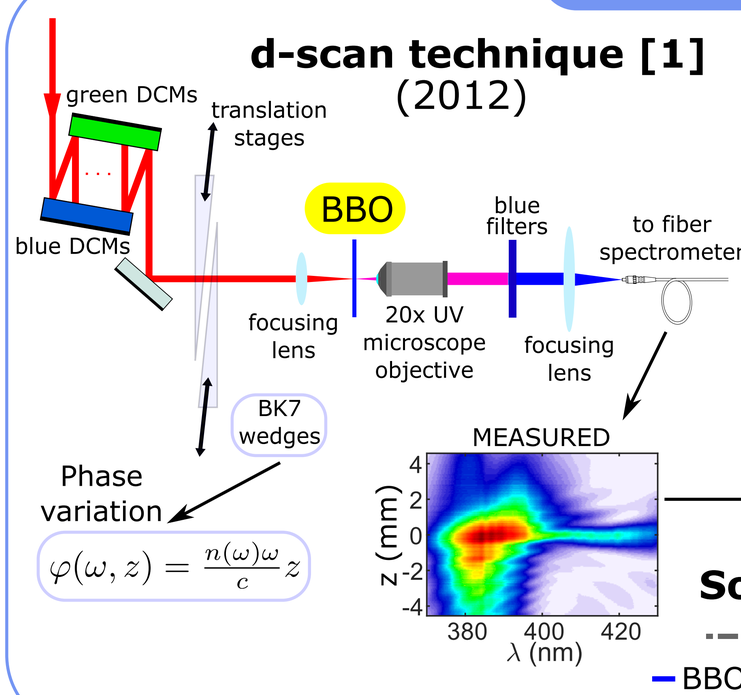
$$\tilde{E}(\omega) = \sqrt{I(\omega)}e^{i\phi(\omega)} \rightarrow E(t) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\tilde{E}(\omega)e^{i\omega t}d\omega \rightarrow I(t) = |E(t)|^2$$

If $\phi(\omega) = 0 \Rightarrow$ FWHM = Fourier Limit
(Shortest pulse achievable)

FIS2009-07870

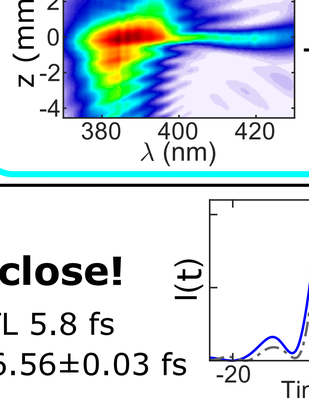
4. We measure them

d-scan technique [1] (2012)



Measured laser spectrum: $\left|\tilde{E}(\omega)\right| = \sqrt{I(\omega)}$

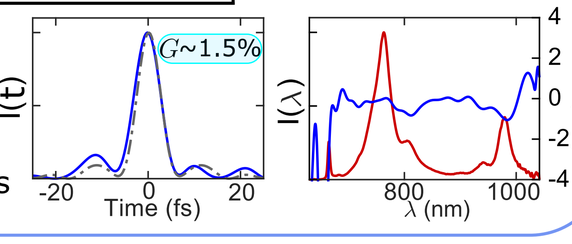
$$I_{sim}(\omega, z) = \left|\int\left[\left(\int\left|\tilde{E}(\omega)\right|e^{i\phi_j(\omega)}e^{i\varphi(\omega,z)}e^{i\omega t}\mathrm{d}\omega\right)^2\right]e^{-i\omega t}\mathrm{d}t\right|^2$$



Standard algorithm

So close!

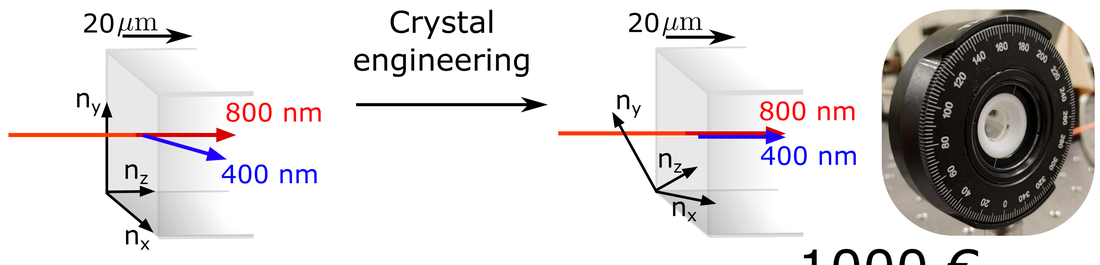
- TL 5.8 fs
- BBO 6.56±0.03 fs



5. How easy is it?

- Phase matching limits SHG
- Thin crystals are needed (~20 μm)
- Specific crystals are needed

General case → Crystal engineering → **Phase matching**



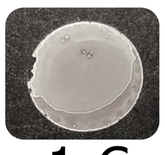
1000 €

6. Idea

To use nanocrystals instead of bulk crystals:

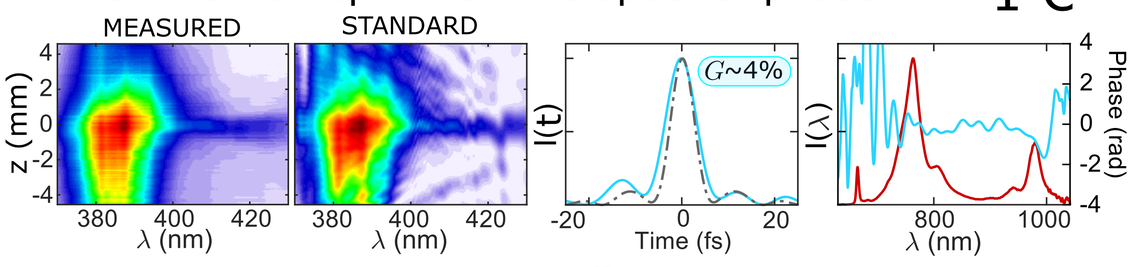
- No phase matching requirements
- The same sample doubles any spectral range [2]
- Commercially available and cheap
- Direct deposition by ourselves

$BaTiO_3$
200 nm



1 €

Does its SHG depend on the spectral phase?



Seems to work,
but still not perfect

TL 5.8 fs
$BaTiO_3$ 7.6±0.4 fs

FIS2017-87360P

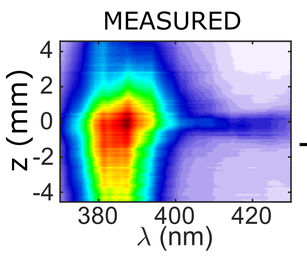
7. Problem solved! ([3], Q1)

What **is not** the problem:
- Polarization dependence
- Intensity response
- Pulse lengthening (linear dispersion)
- Kerr effect
- Particle density or size

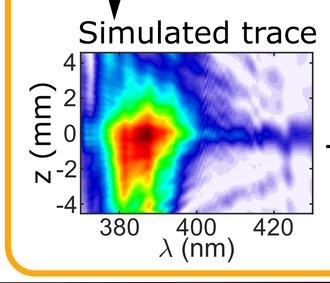
What **is** the problem: **SCATTERING** (sub-wavelength nanoparticles)

Solution $\equiv I_{sim}(\omega, z) = XI_{coherent}(\omega, z) + (1 - X)I_{scattering}(\omega, z)$

Laser spectrum $\left|\tilde{E}(\omega)\right| = \sqrt{I(\omega)}$



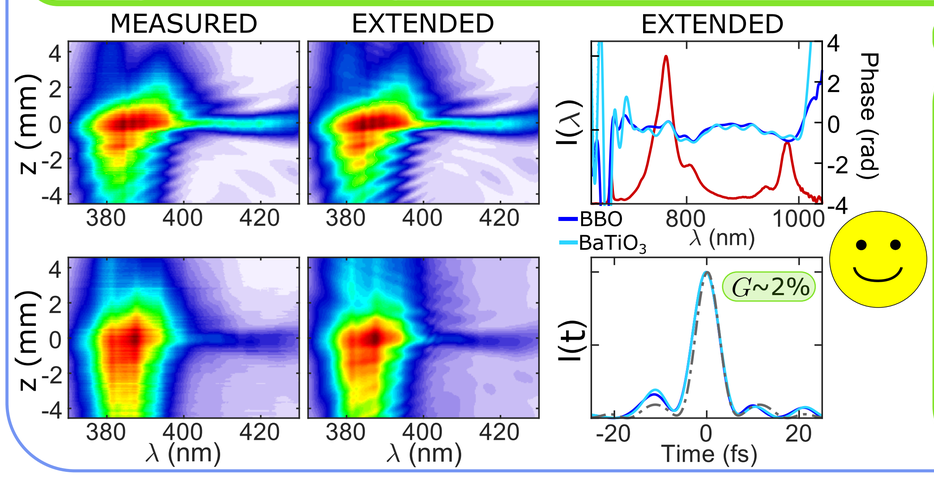
$$I_{sim}(\omega, z) = \left|\int\left[\left(\int\left|\tilde{E}(\omega)\right|\left[X_l e^{i\phi_0(\omega)} + (1 - X_l)e^{i\Phi(\omega)}\right]e^{i\varphi(\omega,z)}e^{i\omega t}\mathrm{d}\omega\right)^2\right]e^{-i\omega t}\mathrm{d}t\right|^2$$



Coherence optimization

Random (incoherent) phase $\Phi(\omega)$ → G'_{min} → X_l → G_{min} → $\phi_n(\omega)$ → G'_{min} → X_k, $\phi_n(\omega)$ → FULL RETRIEVAL
Initial (coherent) phase $\phi_0(\omega)$

Extended algorithm



Results with Extended algorithm

BBO
FWHM = 6.5±0.1 fs
X ~ 0.98
G ~ 1.5 %

$BaTiO_3$
FWHM = 6.5±0.1 fs
X ~ 0.75
G ~ 2 %

References

[1] M. Miranda, C. Arnold, T. Fordell, F. Silva, B. Alonso, R. Weigand, A. L'Huillier, and H. Crespo, "Characterization of broadband few-cycle laser pulses with the d-scan technique," Opt. Express **20**, 18732-18743 (2012).
[2] R. Weigand and E. Cabrera-Granado, "Broadband emission and tunability of the second harmonic signal generated in clusters of dielectric nanoparticles," Reunión Nacional de Óptica 2018, Castellón, Spain.
[3] Ó. Perez-Benito and R. Weigand, "Nano–dispersion–scan: measurement of sub-7-fs laser pulses using second-harmonic nanoparticles," Opt. Lett. **44**, 4921-4924 (2019).