Generation and characterization of mid-infrared femtosecond pulses

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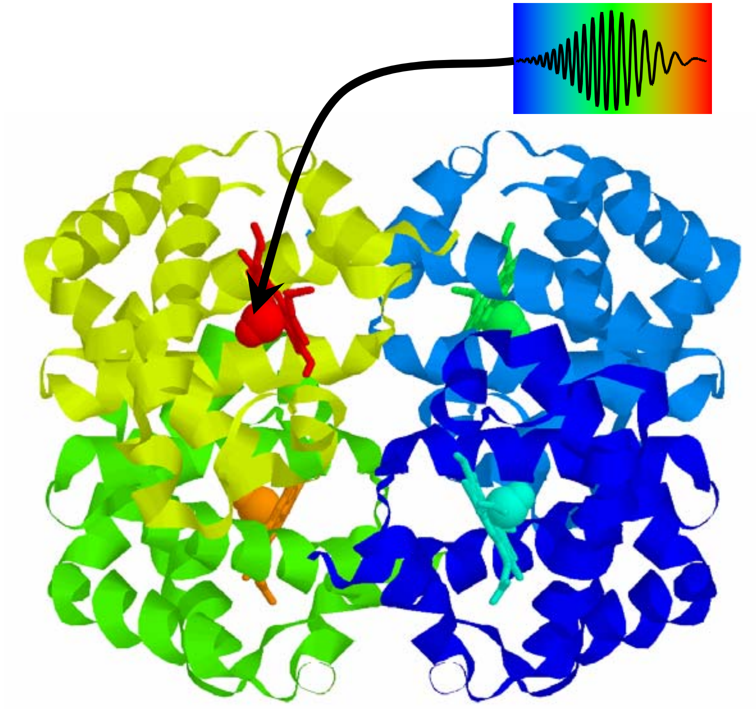
Motivations

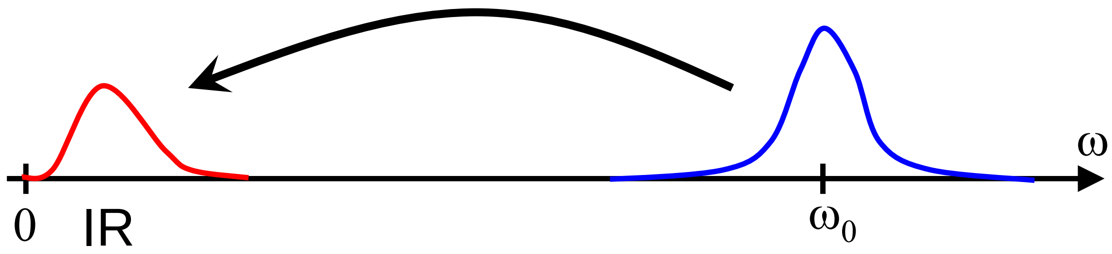


Time-resolved infrared spectroscopy
Multidimensional spectroscopy
Vibrational coherent control

1. Generation of infrared femtosecond pulses

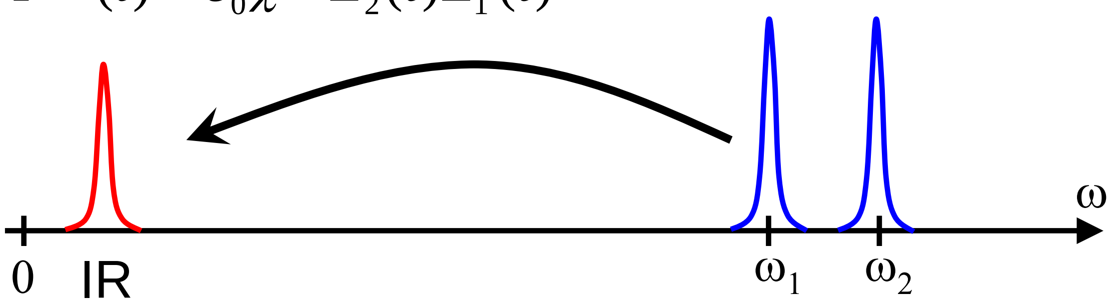
a) Optical rectification (inverse electro-optic effect)

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E(t) E^*(t)$$

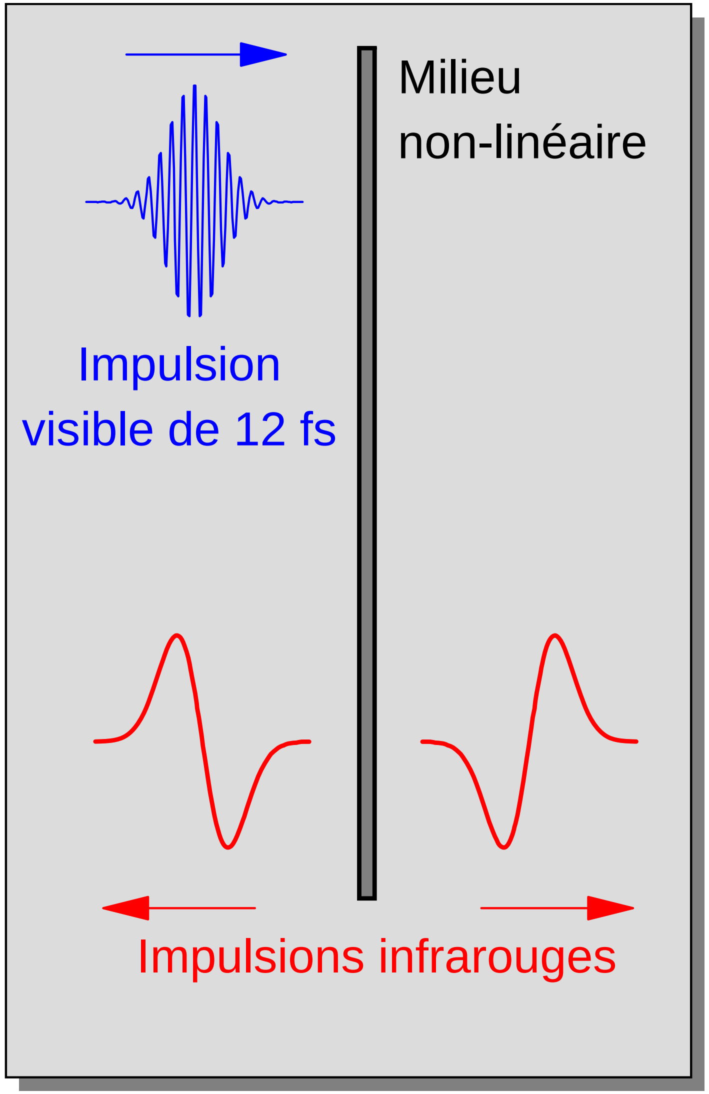


b) Difference frequency mixing

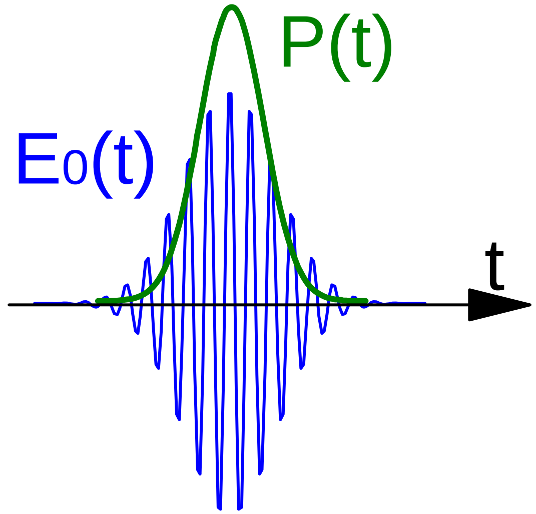
$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E_2(t) E_1^*(t)$$



Infrared generation using optical rectification



- Optical rectification : $P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left| E_0(t) \right|^2$

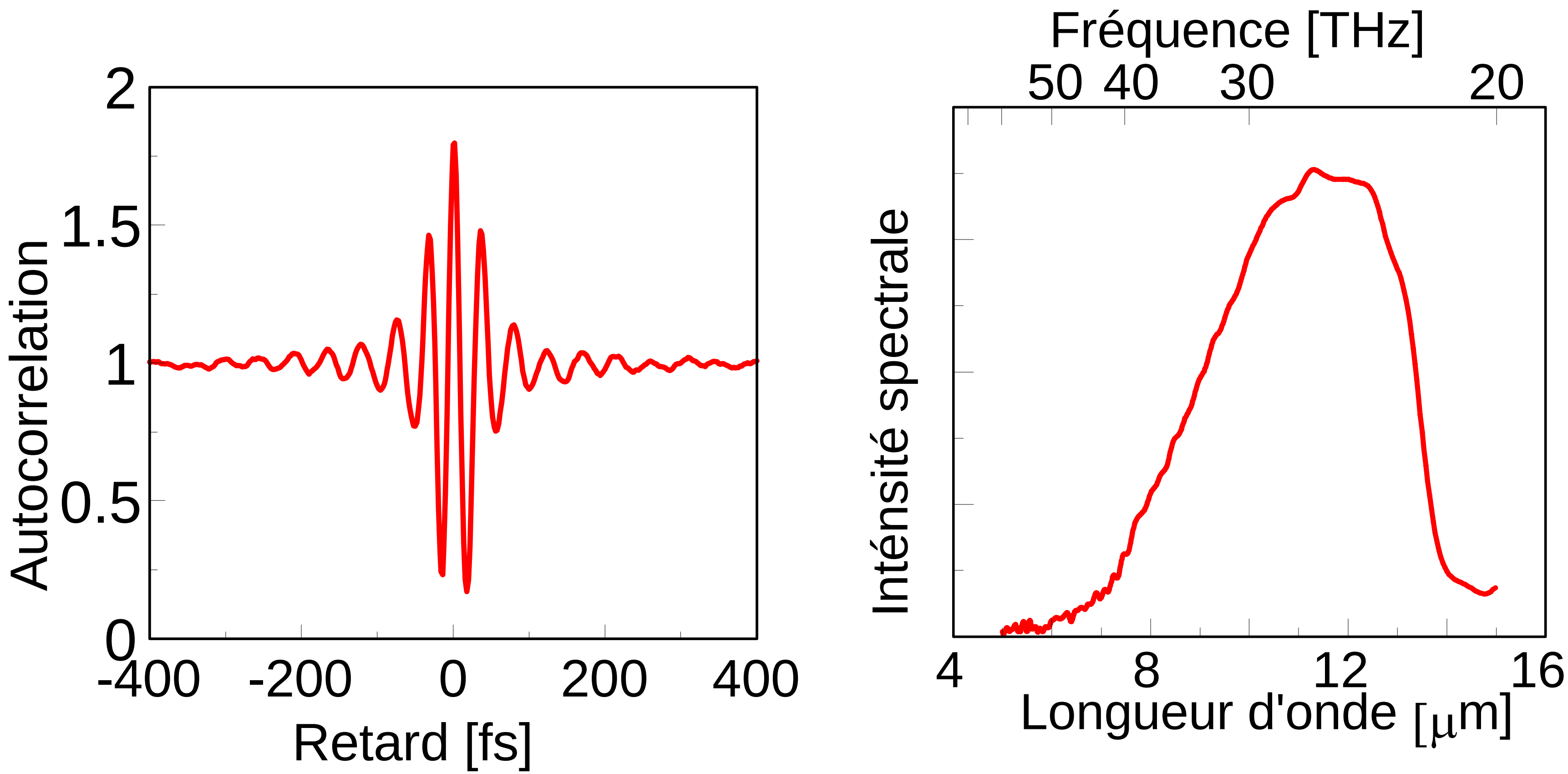


- Propagation : $\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P^{(2)}(t)}{\partial t^2}$

$$E(t) = -\frac{nL}{2\varepsilon_0 c}\frac{\partial P^{(2)}}{\partial t}$$

A. Bonvalet et al., Appl. Phys. Lett., **67**, 2907 (1995)

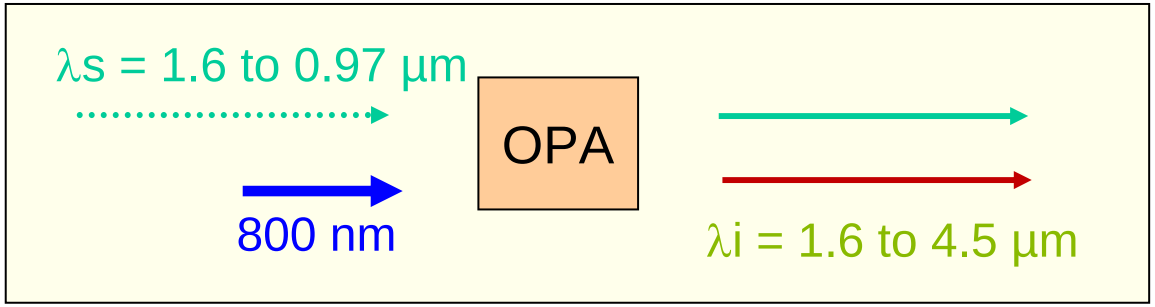
Infrared generation using optical rectification



A. Bonvalet et al., Appl. Phys. Lett., **67**, 2907 (1995)

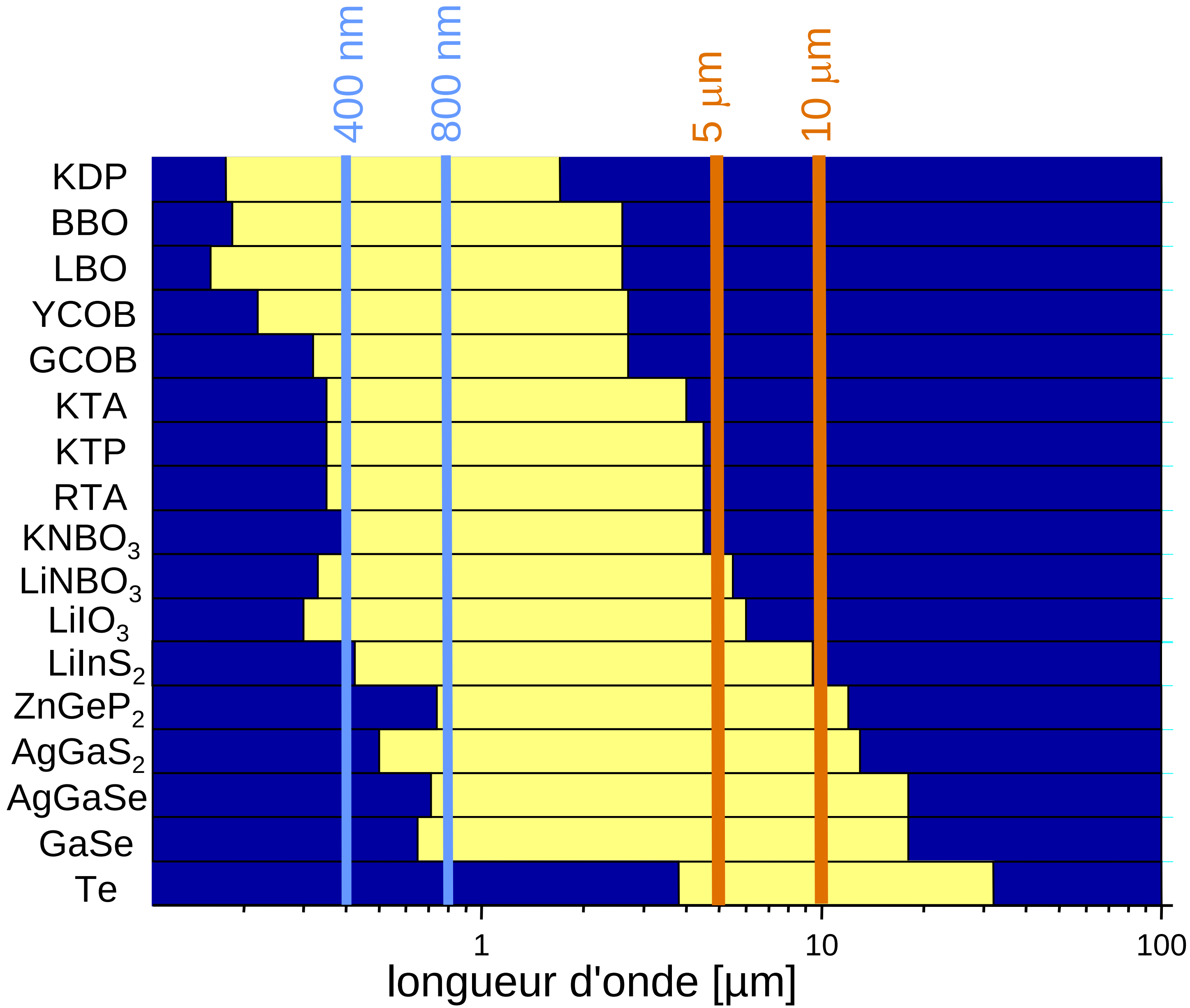
Frequency mixing : one or two stages ?

1) One stage



Simple but less efficient above 4.5 µm due to two-photon absorption

Transparency of nonlinear materials



Ternary chalcogenides $LiBC_2$ (B = In, Ga; C = S, Se, Te) for mid-IR nonlinear optics

L. Isaenko [a], A. Yelisseyev [a], S. Lobanov [a], P. Krinitsin [a], V. Petrov [b,*], J.-J. Zondy [c]

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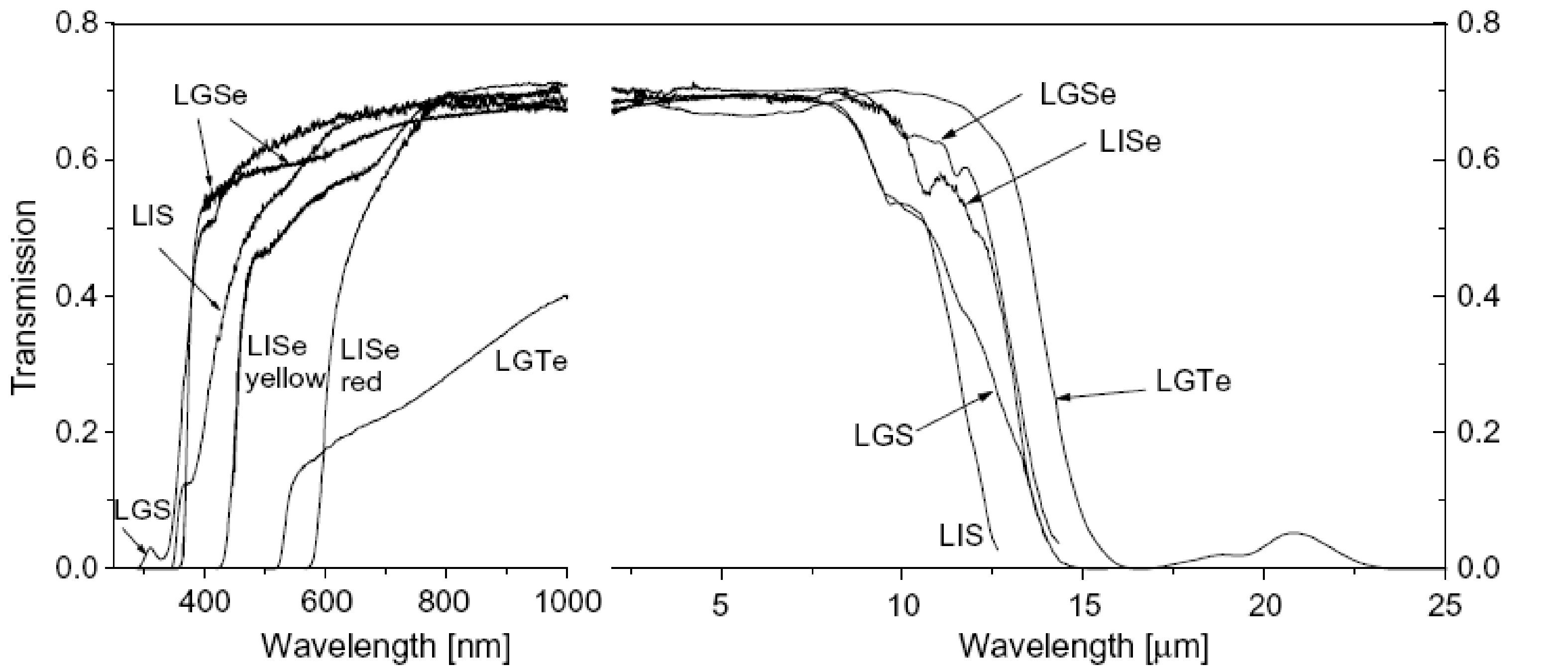
[b] Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str., D-12489 Berlin, Germany
[c] Institut National de Métrologie, Conservatoire National des Arts et Métiers, 292 rue Saint-Martin, F-75003 Paris, France

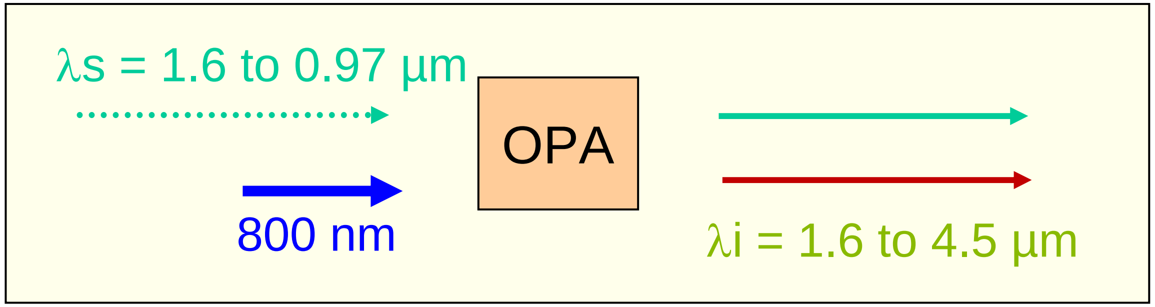
Available online 2 June 2006



J. Non-crystalline solids **352**, 2439 (2006)

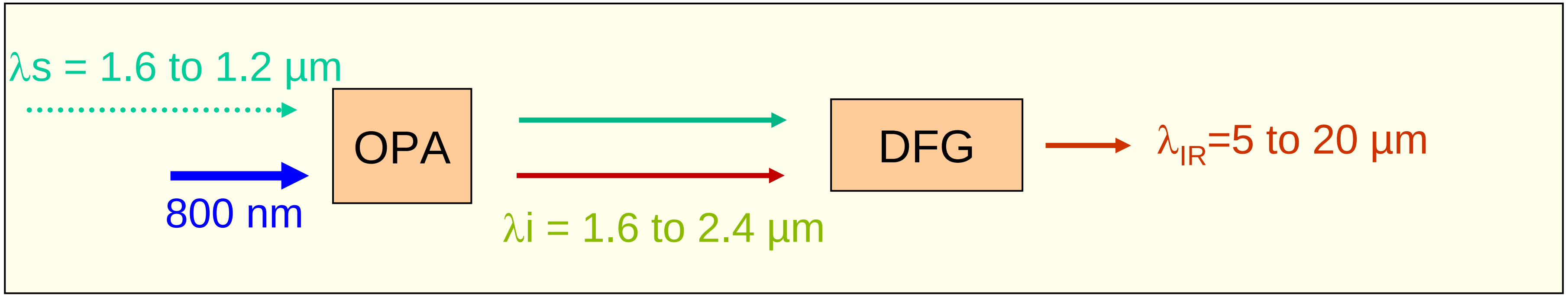
Frequency mixing : one or two stages ?

1) One stage

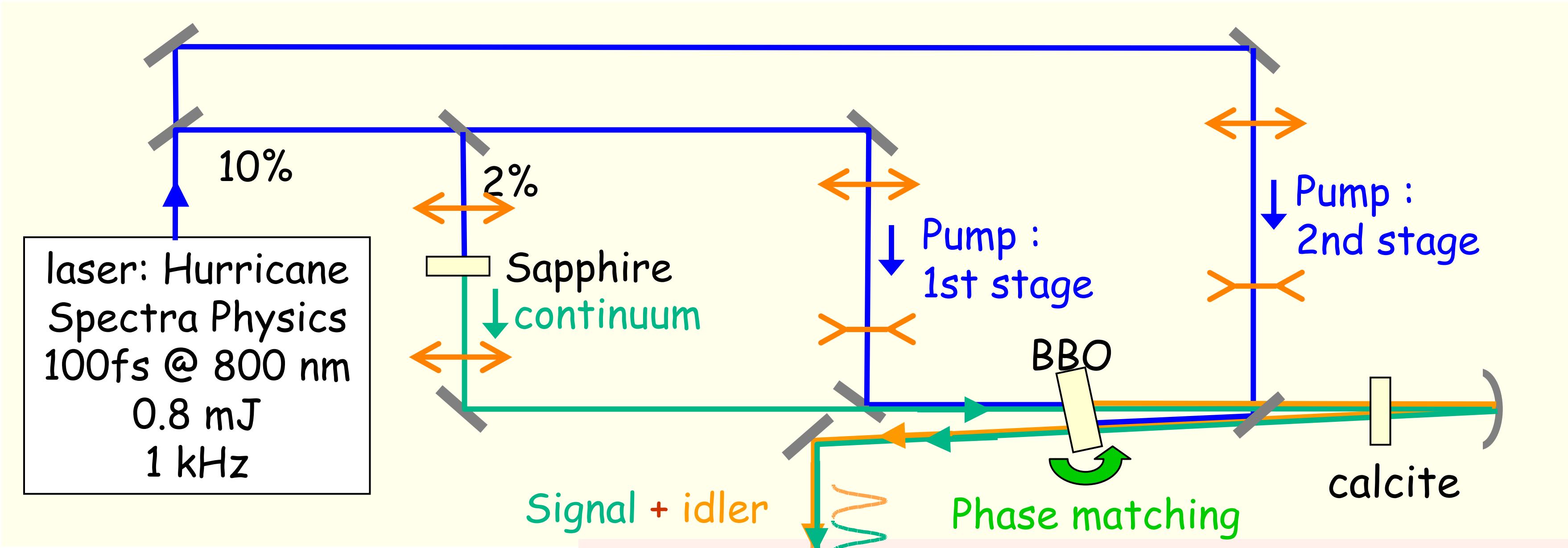


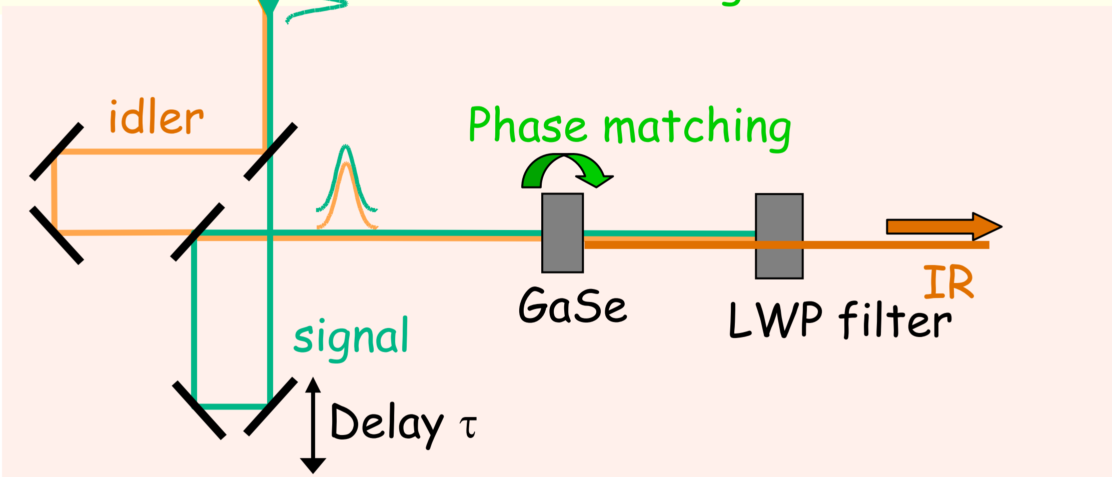
Simple but less efficient above 4.5 µm due to two-photon absorption

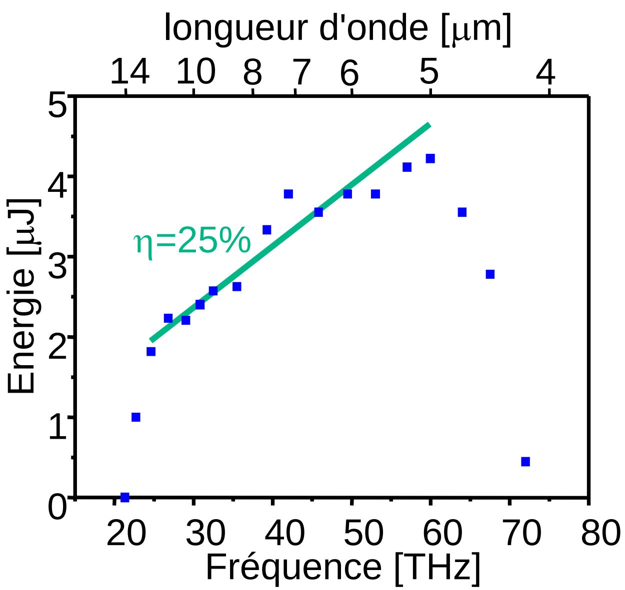
2) Two stages



OPA + DFG

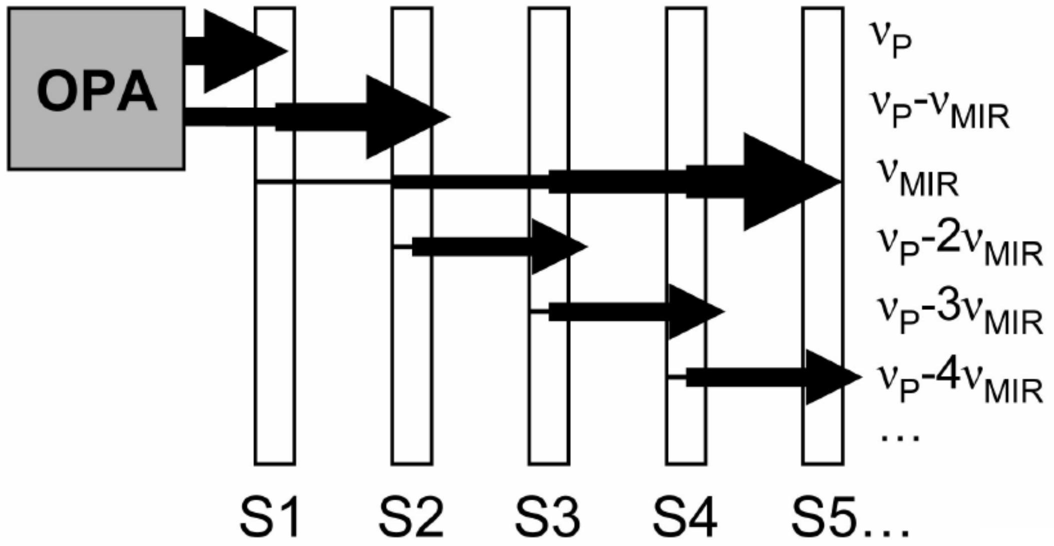


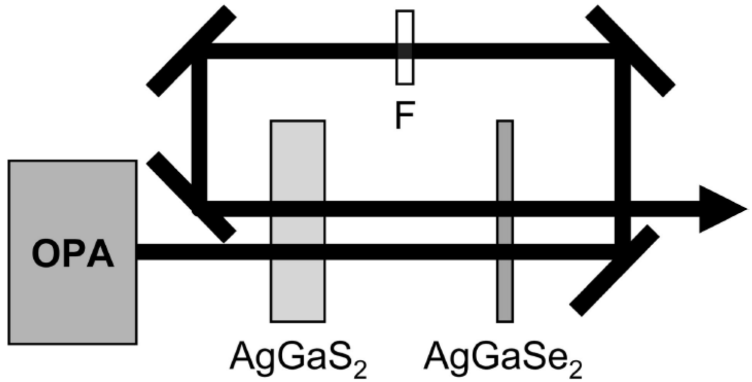




Kaindl *et al.*, JOSA B **17**, 2086 (2000)
Ventalon *et al.*, JOSA B **23**, 332 (2006)

IR generation through parametric cascade downconversion



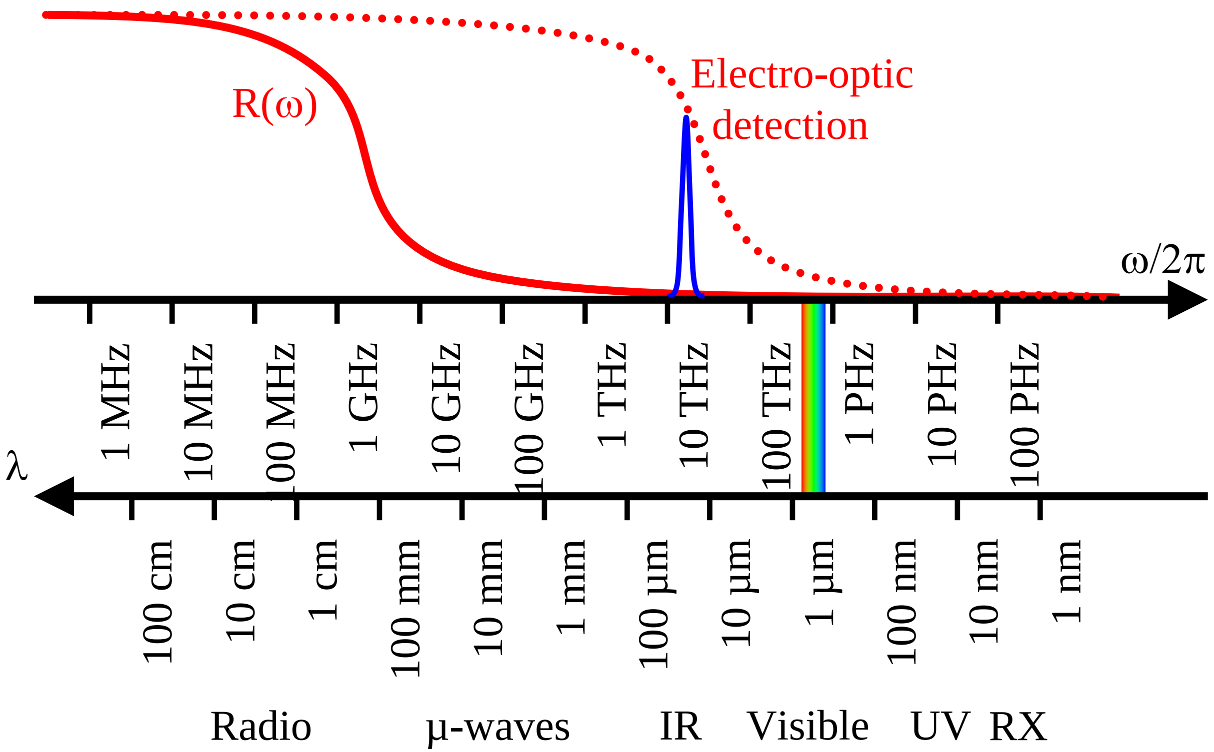


J.M. Fraser, C. Ventalon
Parametric cascade downconverter for intense ultrafast mid-IR generation beyond the Manley-Rowe limit
Appl. Opt. **45**, 4109 (2006).

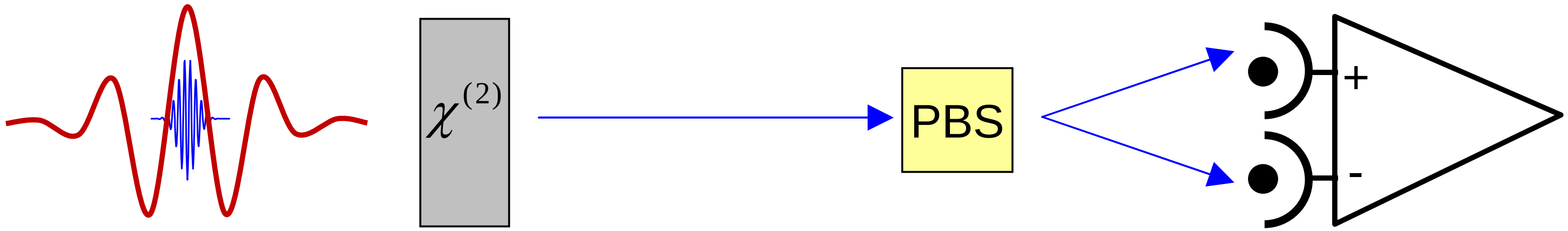
2. Characterization

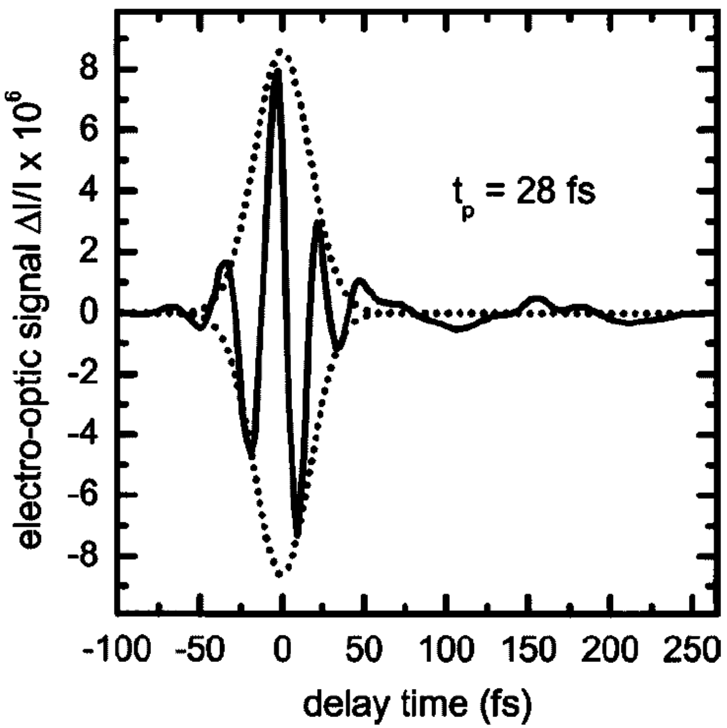
Detection method linear with the electric field

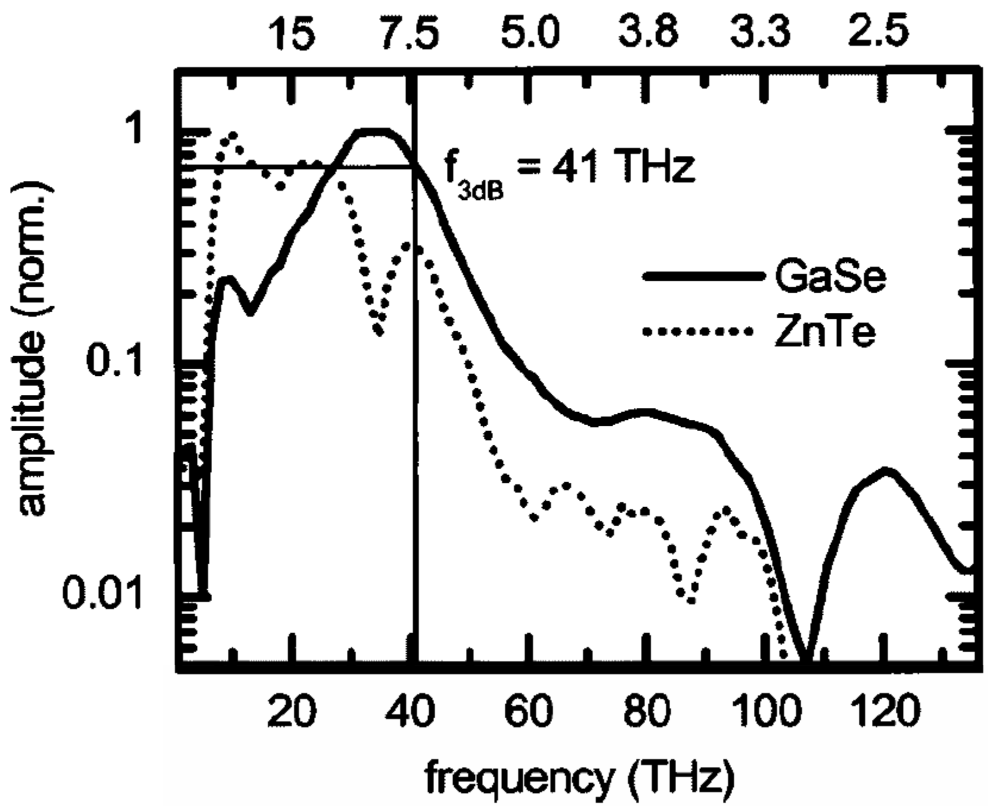
$$S(t) = R(t) \otimes E(t) \longrightarrow S(\omega) = R(\omega)\, E(\omega)$$



Electro-optic detection





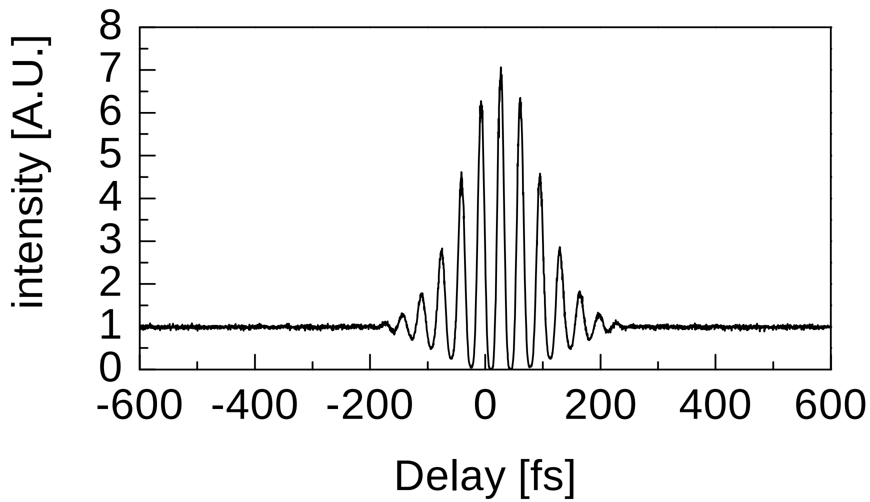


C. Kubler, R. Huber, S. Tubel, A. Leitenstorfer
Ultrabroadband detection of multi-terahertz field transients with GaSe electro-optic sensors: Approaching the near infrared
Appl. Phys. Lett. 85, 3360-3362 (2004)

Measurements based on quadratic detectors

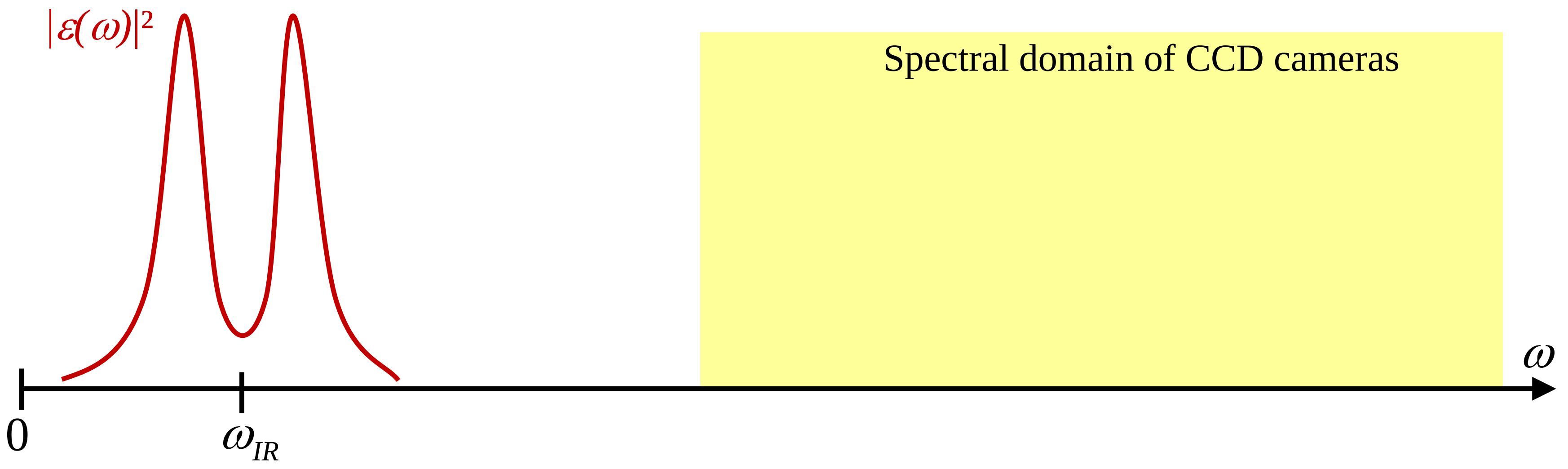
Quadratic and stationary detection results in a signal depending on $|E(\omega)|^2$ only :

Information on the spectral phase can be retrieved only using nonlinear optics !

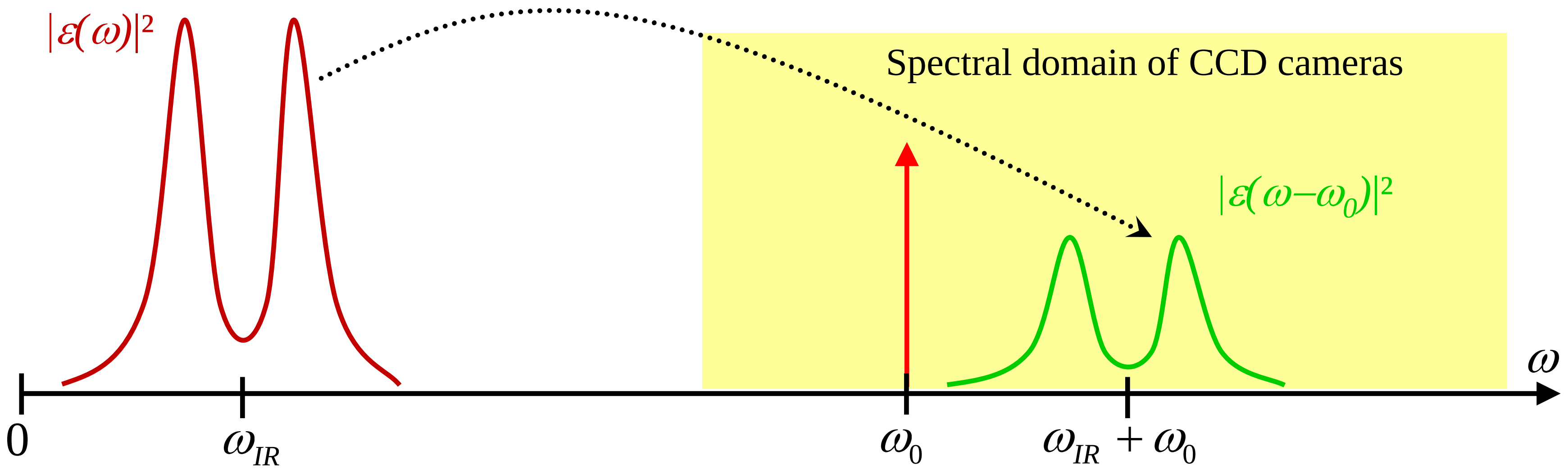


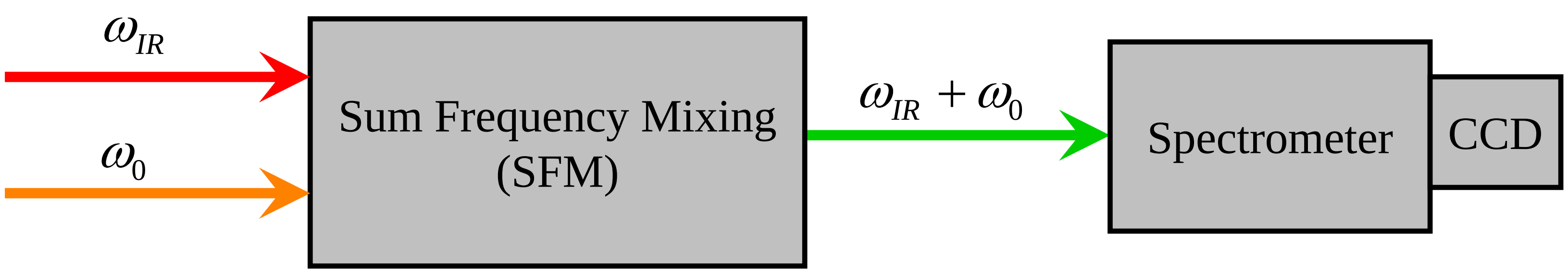
- **Second-order nonlinear autocorrelation**
- **FROG :** B. A. Richman et al., Opt. Lett. **22**, 721 (1997) ; T. Witte et al., Opt. Lett. **27**,131 (2002)
- **Pump-probe :** S. Yeremenko et al., Opt. Lett. **27**, 1171 (2002)
- **Time-domain HOT SPIDER :** C. Ventalon et al., Opt. Lett. **28**, 1826 (2003)
- **Chirped-Pulse Up-conversion** : K. J. Kubarych et al., Opt. Lett. **30**, 1228 (2005)

The problem of IR detection

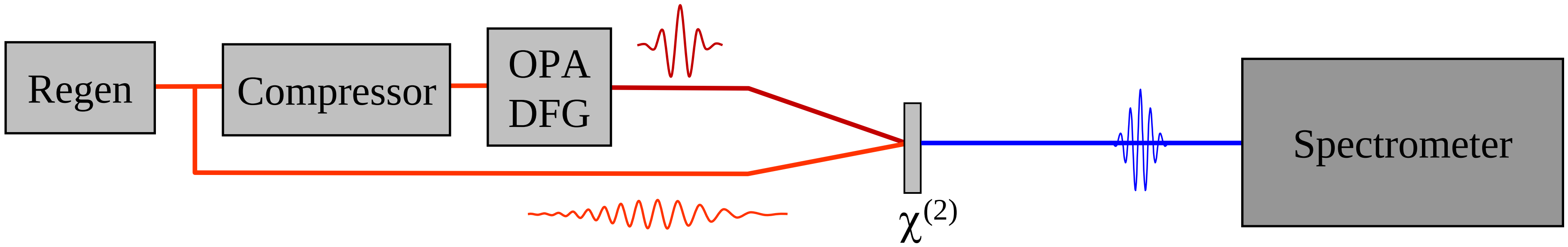


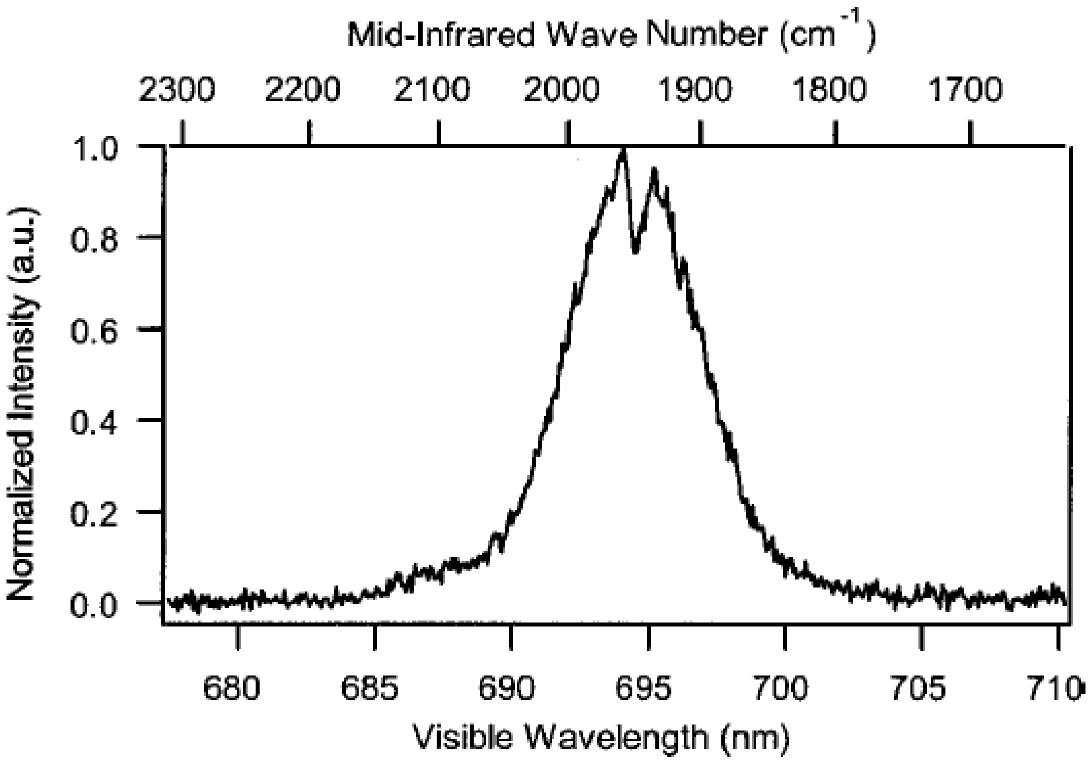
Translation from IR to visible





Translation from IR to visible



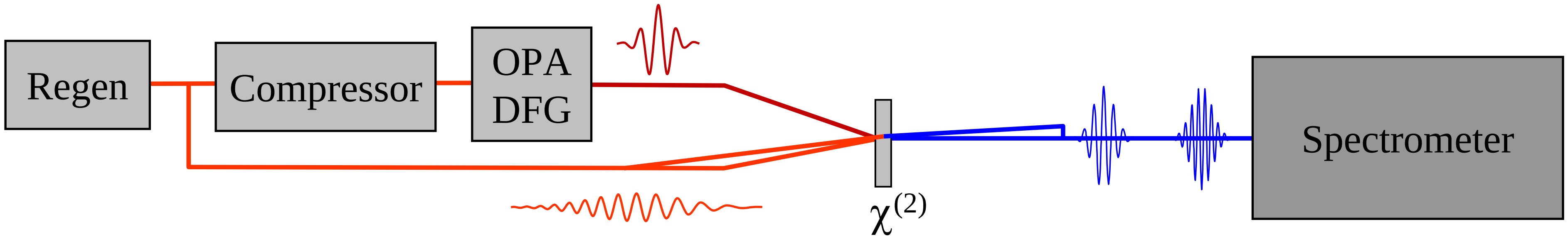


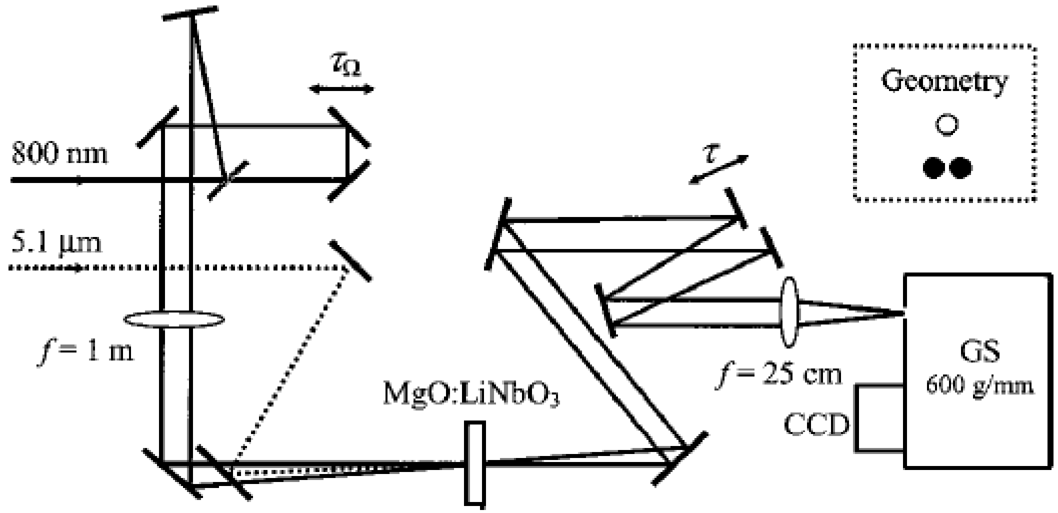
K.J. Kubarych, M. Joffre, A. Moore, N. Belabas, D. M. Jonas

Mid-infrared electric field characterization using a visible charge-coupled-device-based spectrometer

Opt. Lett. **30**, 1228-1230 (2005)

IR ZAP SPIDER





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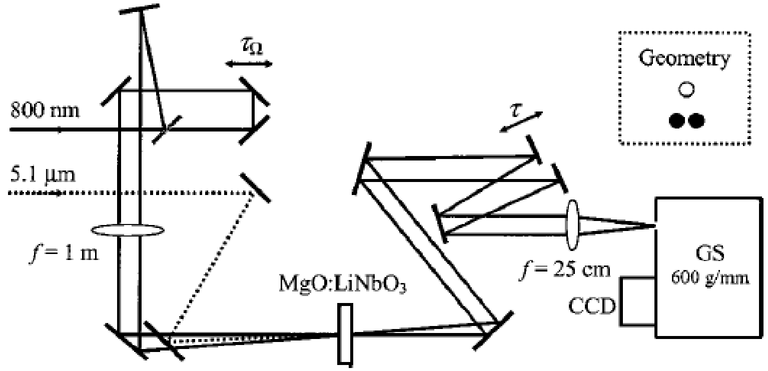
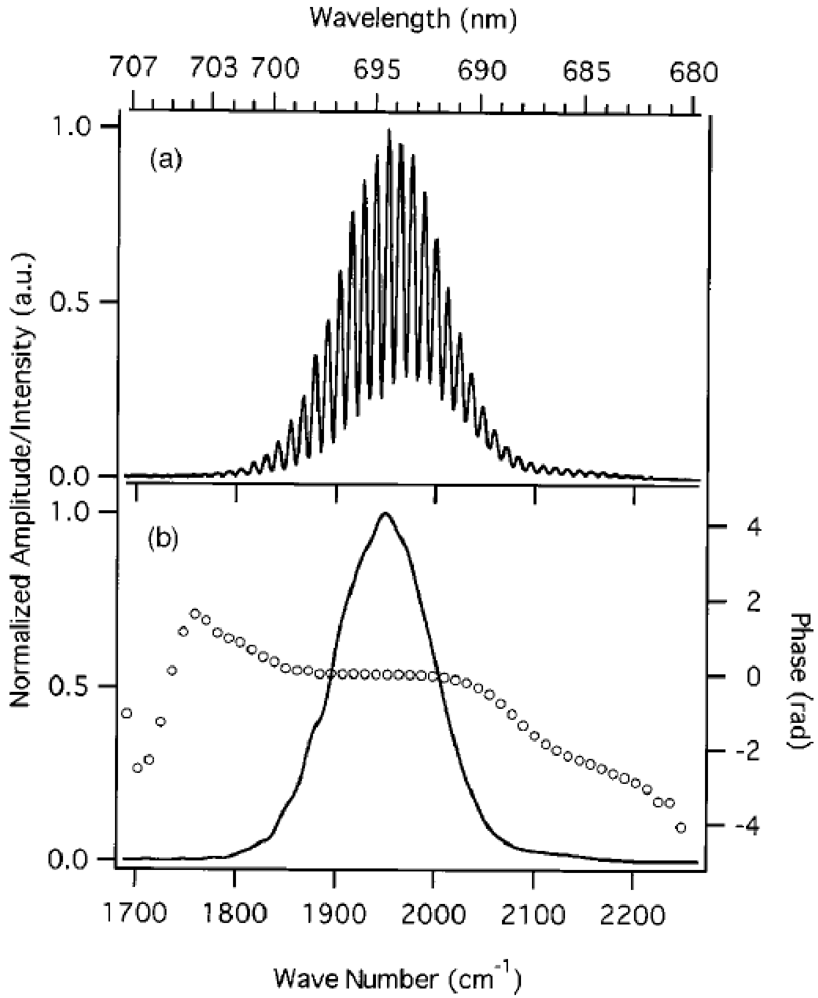




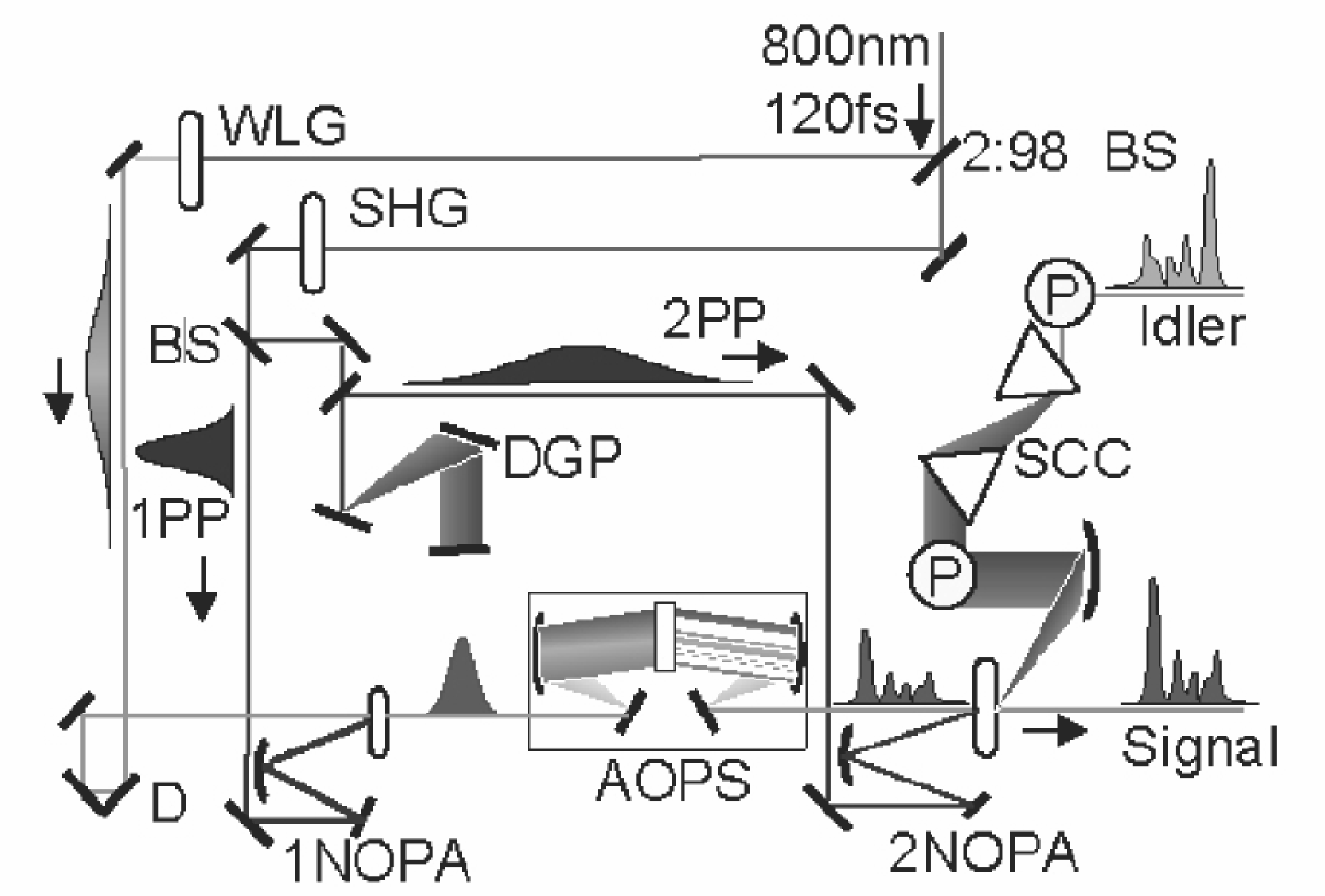
K.J. Kubarych, M. Joffre, A. Moore, N. Belabas, D. M. Jonas

Mid-infrared electric field characterization using a visible charge-coupled-device-based spectrometer

Opt. Lett. **30**, 1228-1230 (2005)

3. Mid-infrared pulse shaping

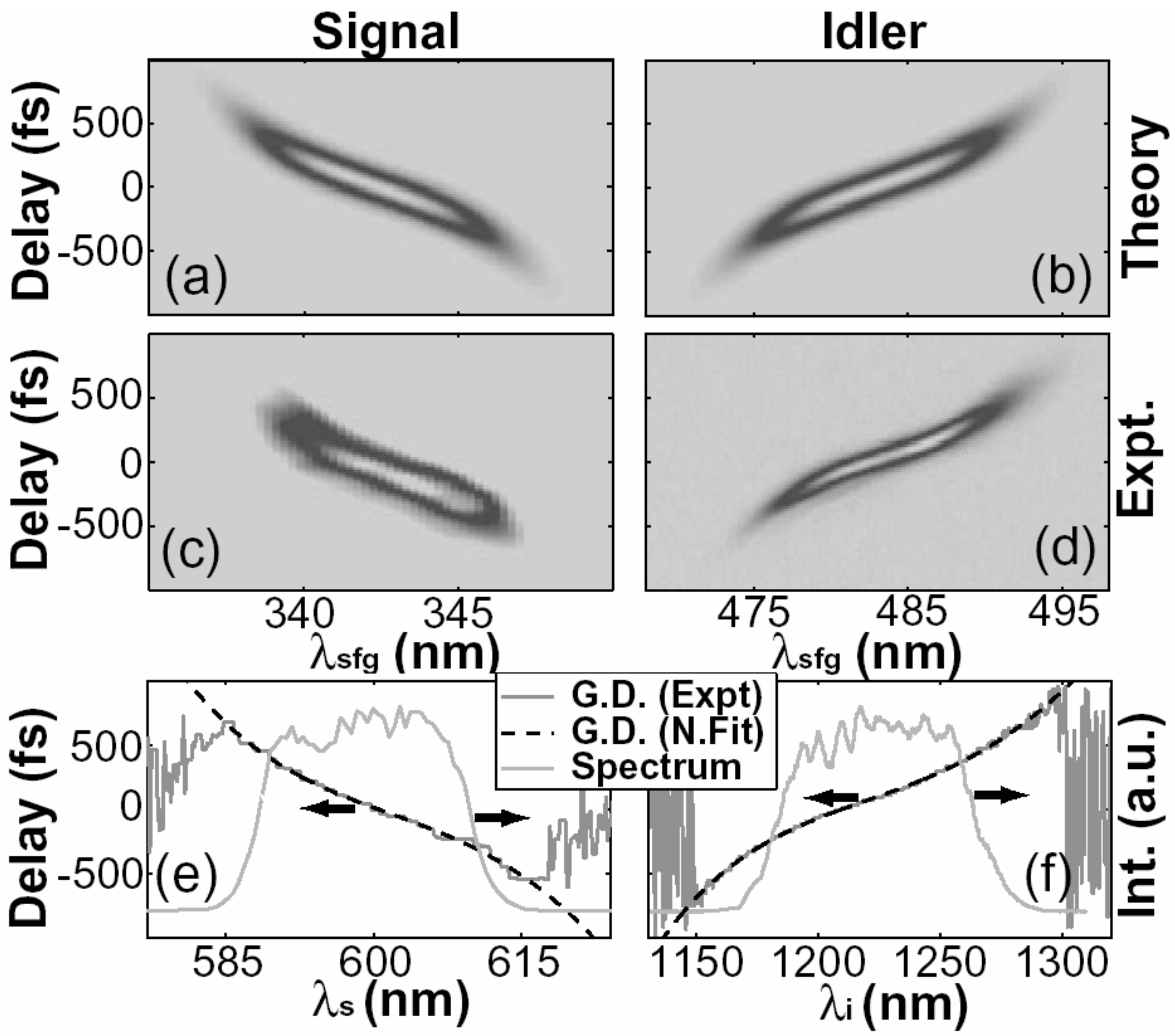
Indirect mid-IR pulse shaping



$$E_{IR}(\omega) = \int E_P(\omega_P) E_S^*(\omega_S) d\omega_P d\omega_S \delta(\omega_P - \omega_S - \omega_{IR}) \approx E_S^*(\omega_P - \omega_S)$$

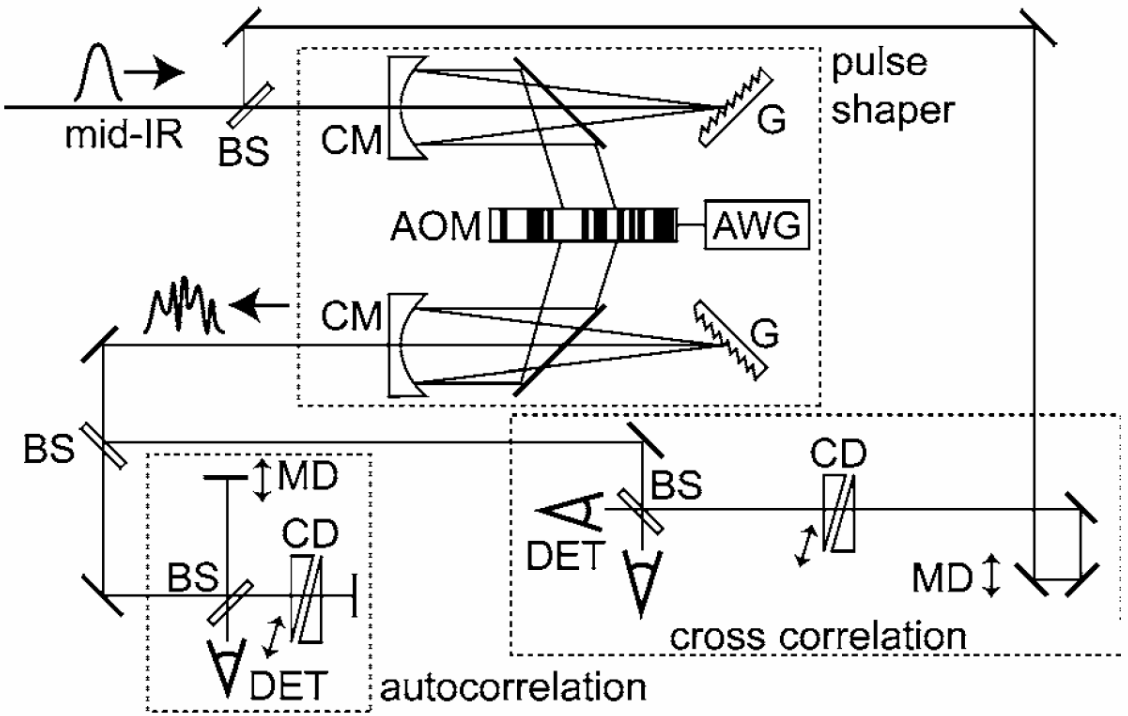
H.-S. Tan, E. Schreiber, W.S. Warren, Opt. Lett. **27**, 439 (2002)

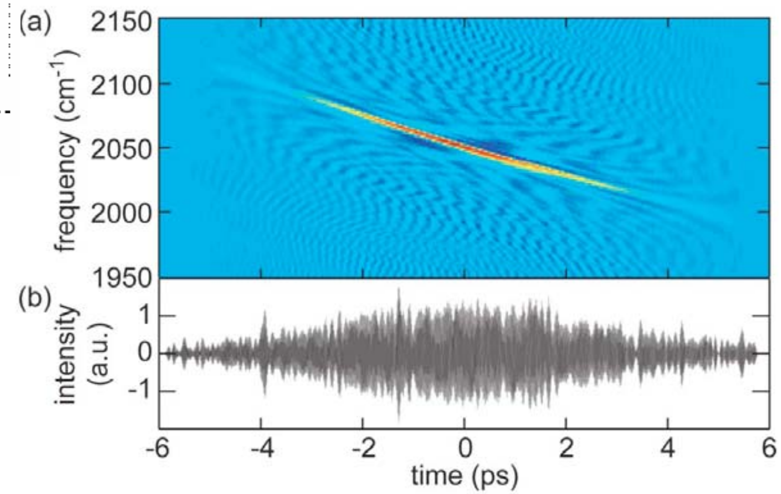
Indirect mid-IR pulse shaping



H.-S. Tan, E. Schreiber, W.S. Warren, Opt. Lett. **27**, 439 (2002)

Direct mid-IR pulse shaping





S.-H. Shim, D. B. Strasfeld, E. C. Fulmer, M. T. Zanni
Femtosecond pulse shaping directly in the mid-IR using acousto-optic modulation
Opt. Lett. **31**, 838-840 (2006)

Conclusion

- The reliability of mid-infrared femtosecond sources (5-20 µm) based on OPA is nowadays comparable to that of visible femtosecond sources.
- Efficiency remains limited due to small energy of infrared photons and to the use of two amplification stages. There is room for improvement through the use of new nonlinear materials or of primary laser sources directly in the near infrared (1-2µm).