Elettra
Sincrotrone
Trieste

Advanced pump-probe experiments at FERMI

Filippo Bencivenga

www.elettra.eu

Outline

1) The beginning of the story: the TIMER project

2) Transient grating (TG), non-linear optics and wave-mixing

3) Compact apparatus for FEL-based TG experiments

4) “mini-TIMER”@DiProI: FEL-stimulated **transient grating**
   → some are preliminary results, experiment done in July!

5) Next step: **two-colour** seeded pulses + **transient grating**
   → coherent Raman scattering

TIMER: aim of the project

Unsolved problems in physics

Wikipedia
The Free Encyclopedia

Condensed matter physics

Amorphous solids
What is the nature of the transition between a fluid or regular solid and a glassy phase? What are the physical processes giving rise to the general properties of glasses?

High-temperature superconductors
What is the responsible mechanism that causes certain materials to exhibit superconductivity at temperatures much higher than around 50 Kelvin?

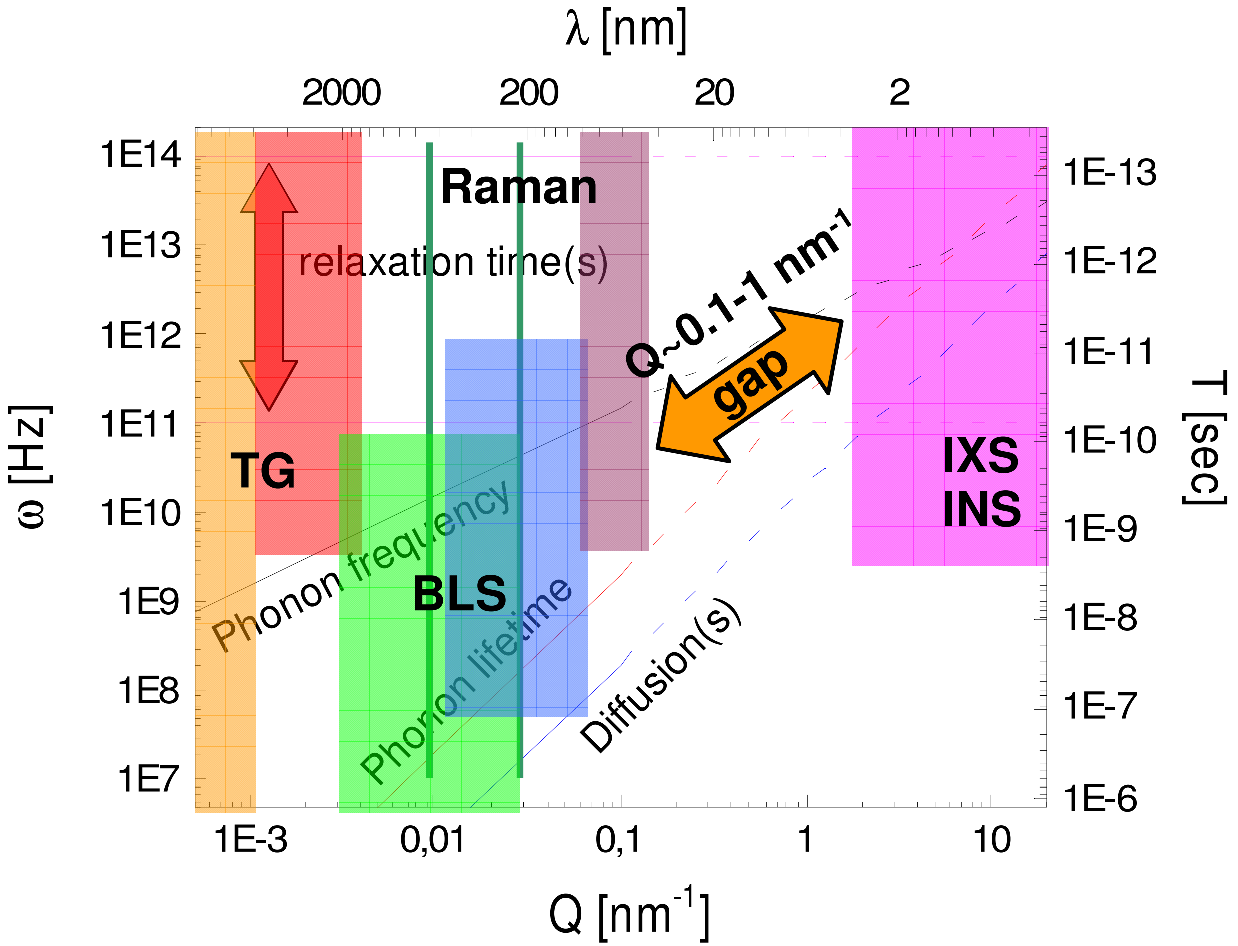
Sonoluminescence
What causes the emission of short bursts of light from imploding bubbles in a liquid when excited by sound?

Turbulence
Is it possible to make a theoretical model to describe the statistics of a turbulent flow (in particular, its internal structures)? Also, under what conditions do smooth solution to the Navier-Stokes equations exist?

Glass is a very general state of matter (a large number of systems can be transformed from liquid to glass), which shows anomalies with respect to crystals

Key role of **vibrational dynamics** in the few **THz** frequency range
→ **phonon-like modes** in the **Q=0.1-1 nm^{-1}** wavevector range

TIMER: aim of the project

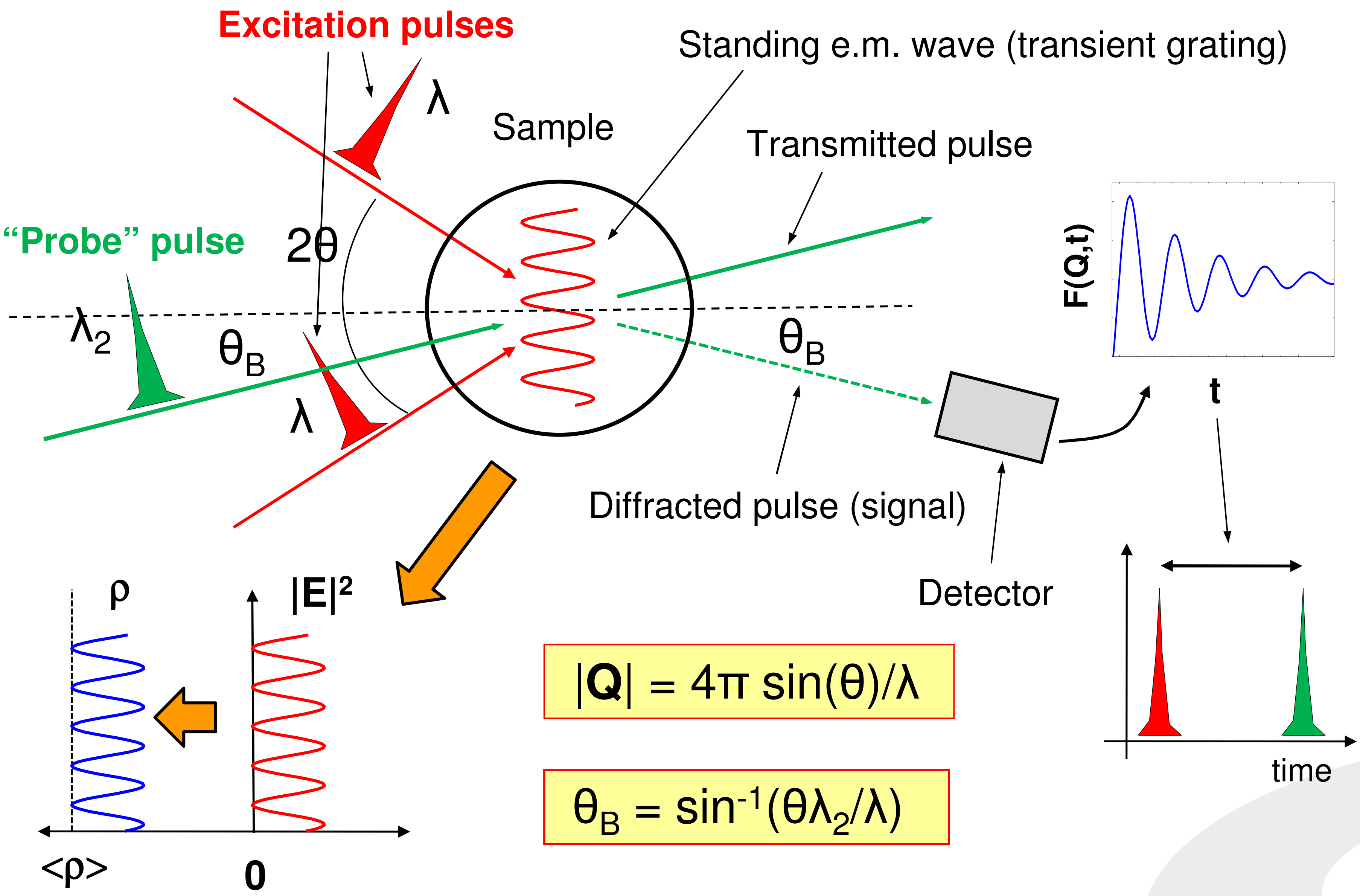


Methods
Transient grating, Raman and Brillouin light and UV scattering, IUVS (BL10.2 @Elettra), inelastic (hard) x-ray and (thermal) neutron scattering

Information
Structure and **Elasticity** (sound velocities)
Interaction potential and **Anharmonicity**
Dynamical istabilities (phonon softening)
Electron-phonon coupling
Thermodynamics (c_V, λ, Θ_D, S_D, etc …)

TIMER's goal is to fill the gap

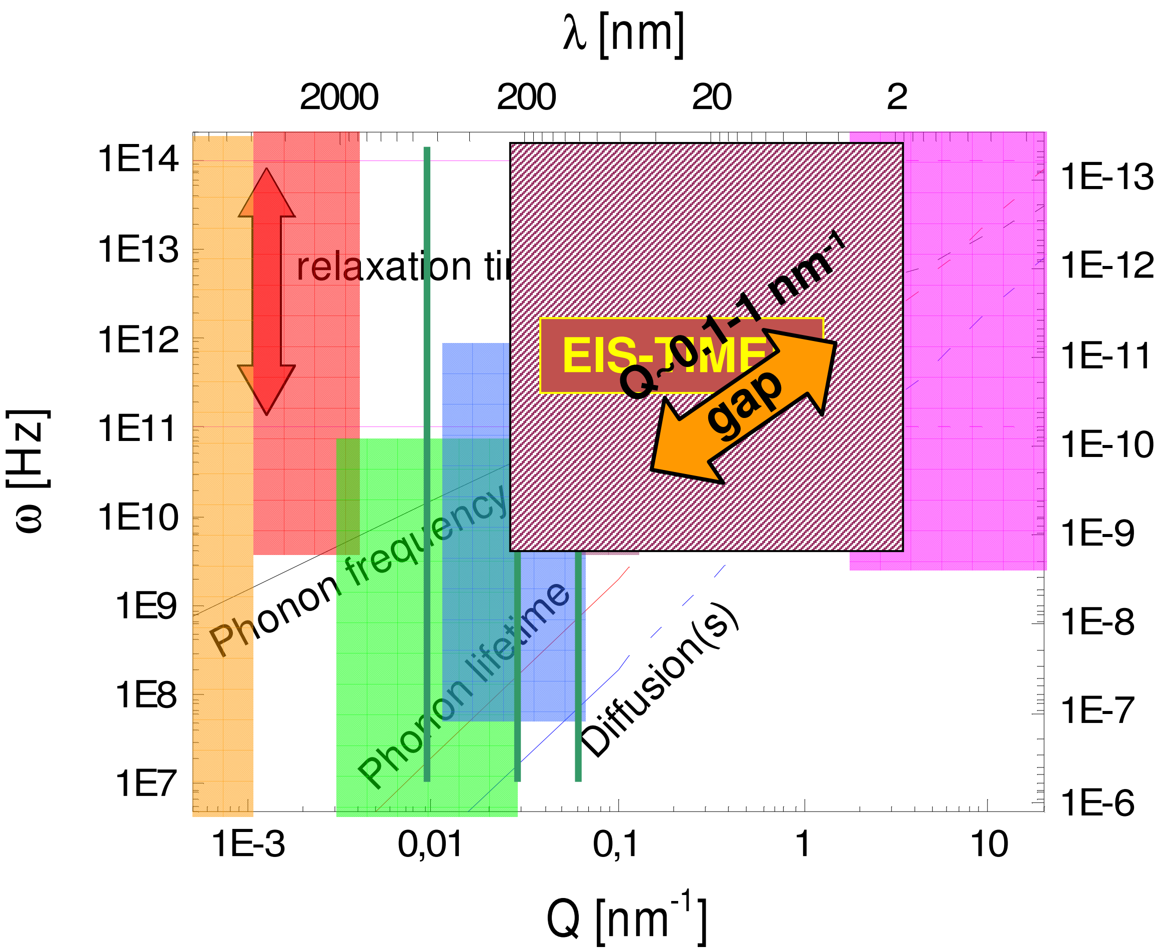
Transient grating method



$|\mathbf{Q}| = 4\pi \sin(\theta)/\lambda$

$\theta_B = \sin^{-1}(\theta\lambda_2/\lambda)$

EUV/x-ray transient grating



$|Q| = 4\pi \sin(\theta)/\lambda$

EIS-TIMER beamline:
$\theta = 9.2^o - 52.7^o$
$\lambda = 60 - 4$ nm
$\lambda_{pr} = \lambda / 3$ (3rd harm)

$Q = 0.03 - 2.5\ nm^{-1}$

$L^* \sim 2\pi / Q^*$

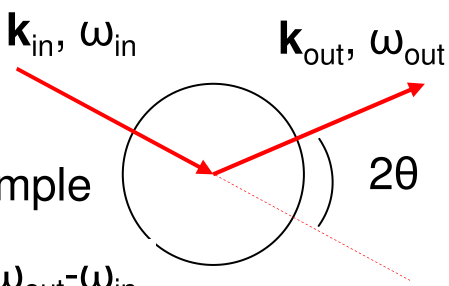
- **disordered systems ($L^* \sim 10$ nm)**
- **nanostructures ($L^* \sim 1$-100 nm)**

F. Bencivenga and C. Masciovecchio, NIMA (2009)

Linear vs non linear scattering

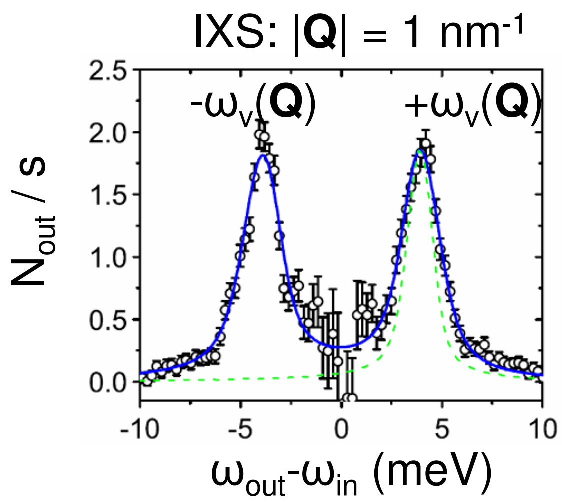
$E_{out} \sim \chi E_{in}$

Brillouin & Raman scattering



$\omega = \omega_{out} - \omega_{in}$

$\mathbf{Q} = \mathbf{k}_{out} - \mathbf{k}_{in}$



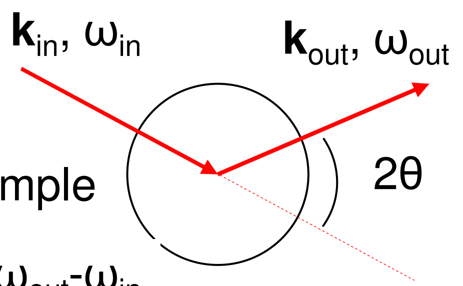
Linear vs non linear scattering

$$E_{out} \sim \Sigma_{p(i,j,k,...)} [\chi E_i + \chi^{(2)} E_i E_j + \chi^{(3)} E_i E_j E_k + ...]$$

$\chi^{(n)}$ = n-order non linear susceptibility
→ "(n+1)-wave-mixing" processes

$\chi^{(2n)}$=0 (sample with inver. sym.)
→ first evidence of x-ray induced wave-mixing[1]

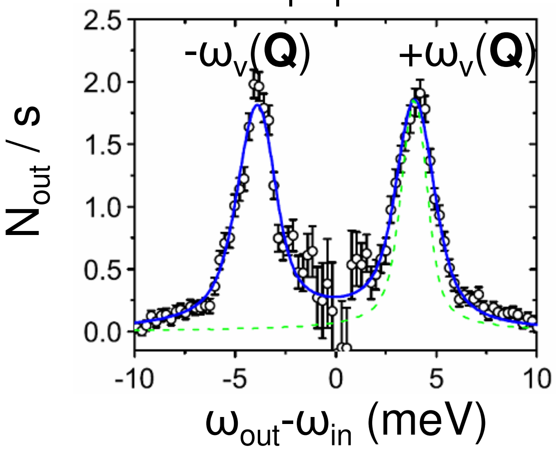
Brillouin & Raman scattering



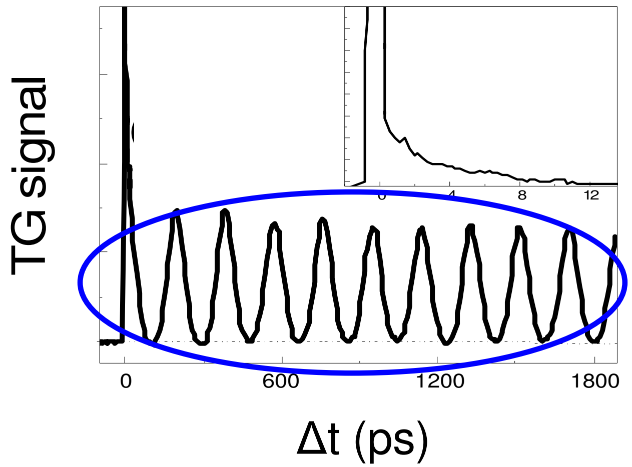
$\omega = \omega_{out} - \omega_{in}$

$\mathbf{Q} = \mathbf{k}_{out} - \mathbf{k}_{in}$

IXS: $|\mathbf{Q}|$ = 1 nm^{-1}

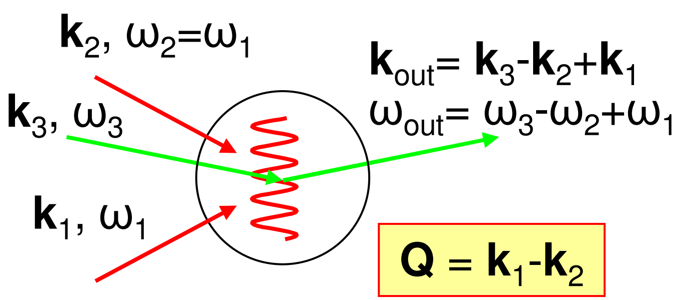


$|\mathbf{Q}|$ = 0.005 nm^{-1}



Stimulated Brillouin & Raman scattering

→ four-wave-mixing (FWM) based on TG



$\mathbf{Q} = \mathbf{k}_1 - \mathbf{k}_2$

TG signal ~ FT(IXS signal)

<u>Spontaneous vs coherent population of excited states</u>

1) T. E. Glover et al., Nature (2012)

Wave-mixing signal

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$$E_{out} \sim \Sigma_{p(i,j,k,...)} [\chi E_i + \chi^{(2)} E_i E_j + \chi^{(3)} E_i E_j E_k + ...]$$

Driving force in the wave equation: non-linear emission at $\omega_{out} = \Sigma_{p(i,j,k,...)} \pm\omega_i$, not necessarily equal to any ω_i.

$\chi^{(n)} \sim E_a^{-n+1}$ ($E_a \sim$ 10-100 V/nm)

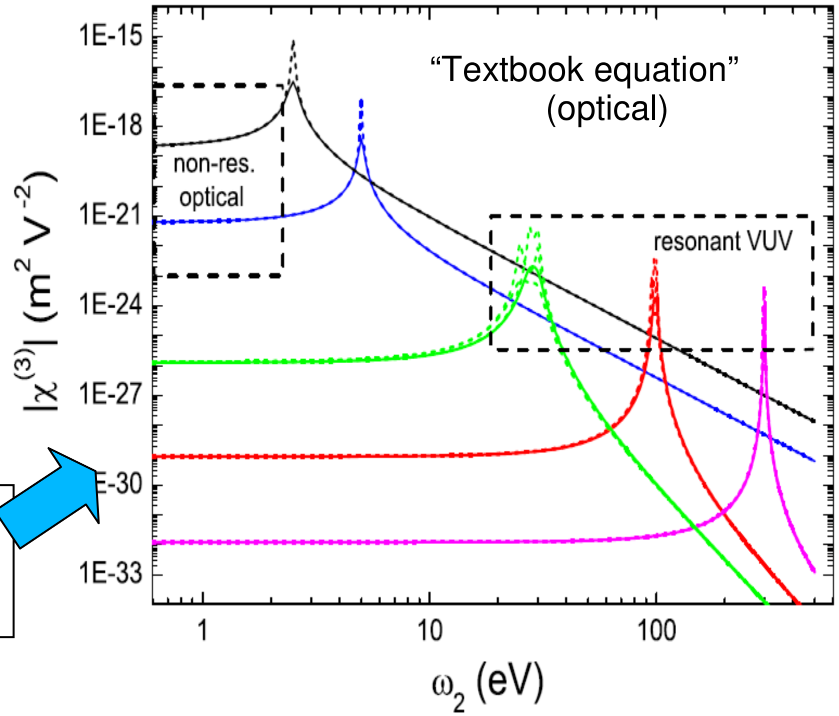
$E_i <<$ 1 V/nm (e.g., damage)

For TG (four-wave-mixing)

$\chi^{(3)} E_j^2/\chi << 10^{-4}$

$\rightarrow I_{fwm}/I_{lin} << 10^{-8}$

$\chi^{(3)}$ decreases on increasing ω_i



B. D. Patterson, SLAC-TN (2011); F. Bencivenga et a., NJP (2013)

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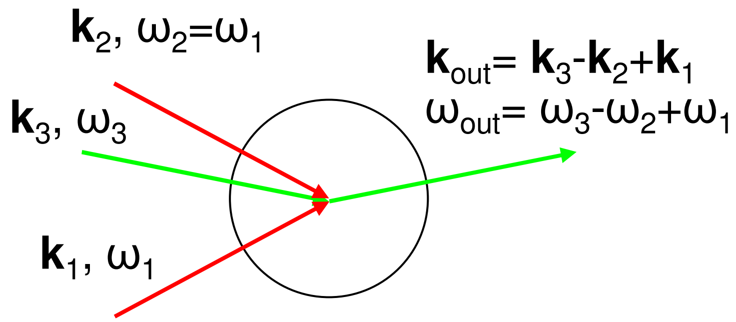
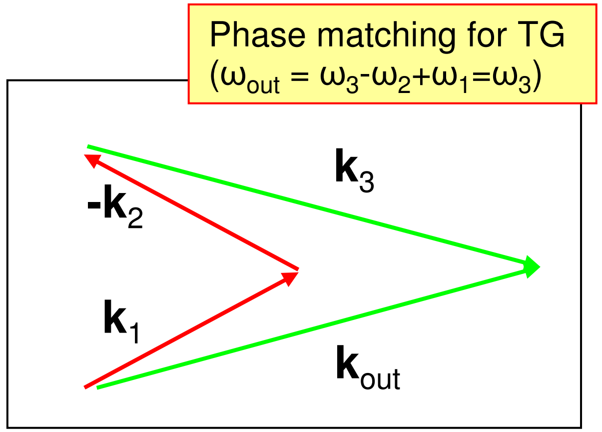
Phase matching

$E_{out} \sim \Sigma_{p(i,j,k,...)} [\chi E_i + \chi^{(2)} E_i E_j + \chi^{(3)} E_i E_j E_k + ...]$



Phase matching → non linear emission from N elementary emitters placed at different sample locations within $2\delta k^{-1}$ (coherence length of the non-linear process) **adds in phase** (intensity grows up as N^2) **along k_{out}** $= \Sigma_{p(i,j,k,...)} \pm k_i$ (for a FWM process that radiates at $\omega_{out} = \Sigma_{p(i,j,k,...)} \pm \omega$)

→ $\delta k = |k_{out} - \Sigma_{p(i,j,k,...)} \pm k_i| > 0$ because, e.g., bandwidth and divergence



Directionality + **coherent addition** may lead to a dominating non linear signal, which can even turn into a macroscopic coherent beam that propagates downstream the sample (e.g., harmonic generation[1])

FERMI: EUV pulses with (almost) Fourier-limited bandwidth → increase in δk^{-1} → increase in N (within $2\delta k^{-1}$) → substantial ($\sim N^2$) increase of I_{fwm} along k_{out}

1) Frenkel at al., Phys. Rev. (1962)

“mini-TIMER” (@DiProI)

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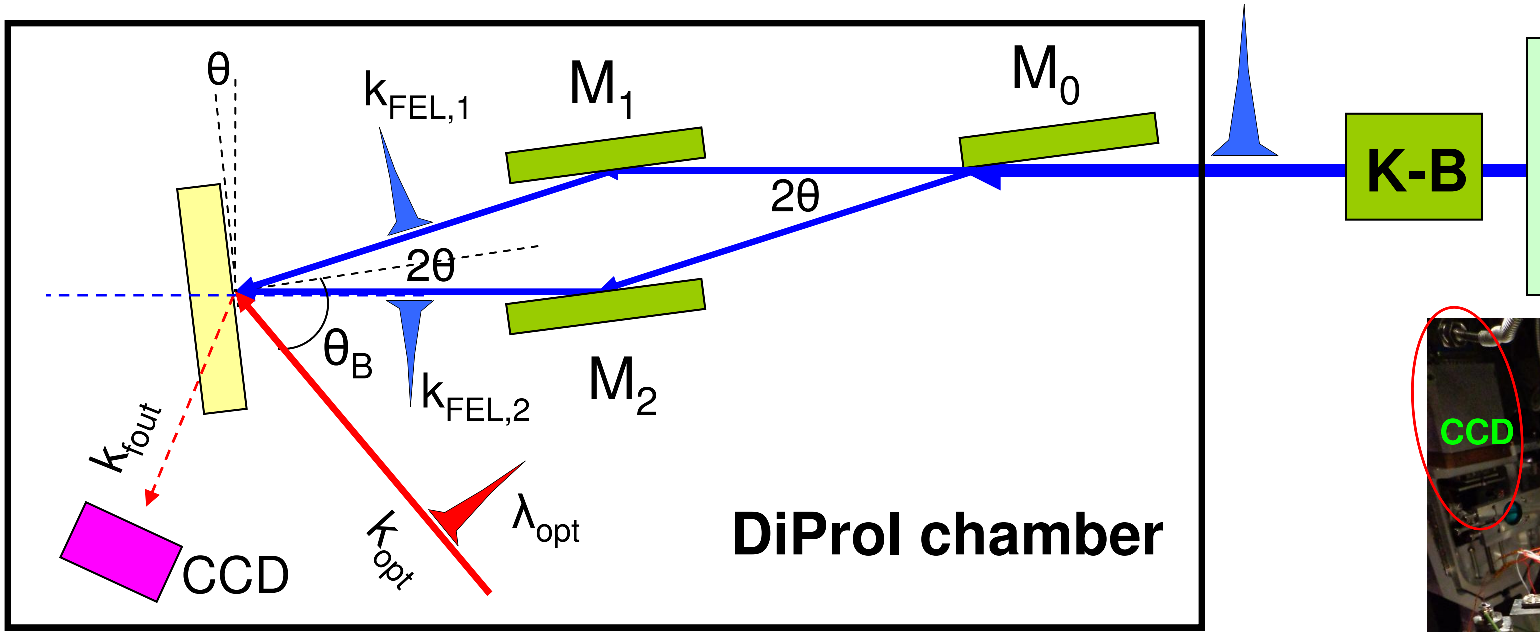
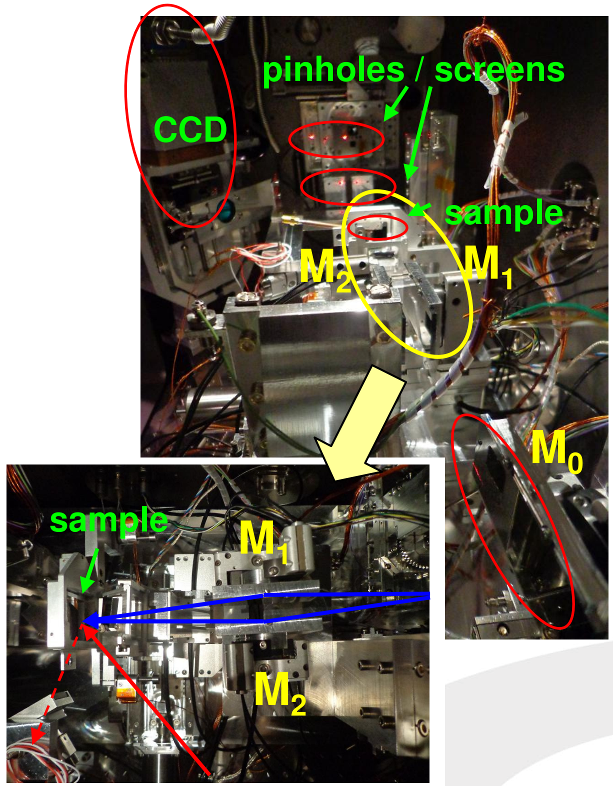
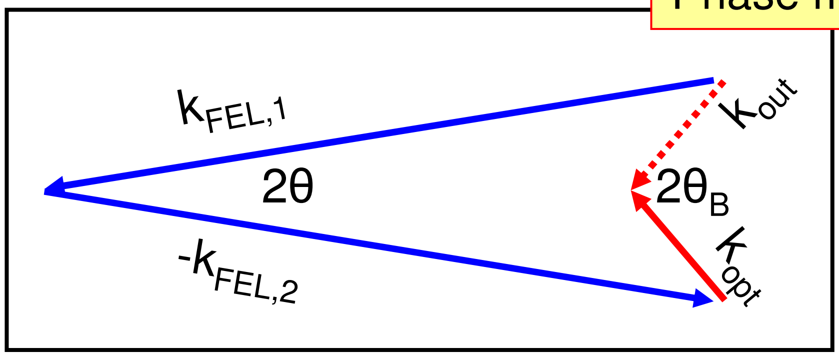


Padres (I_0, spectrum, filters, beam position, etc.)



Phase matching



$2θ = 6.16^o$
$θ_B = 49.9^o$
$λ_{opt}$ =392.8 nm
$λ_{FEL}$ =27.6 nm

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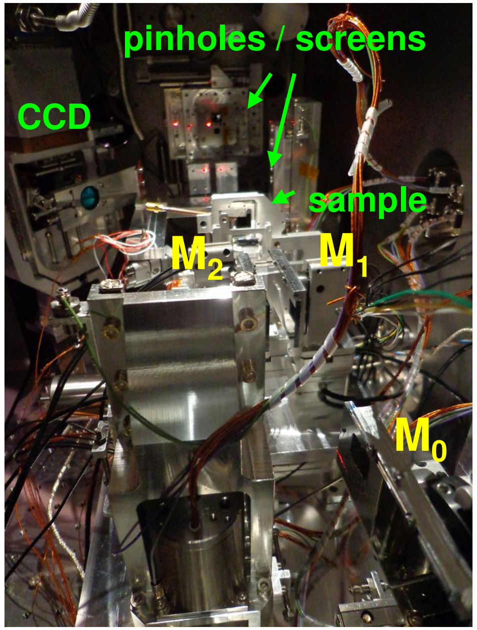
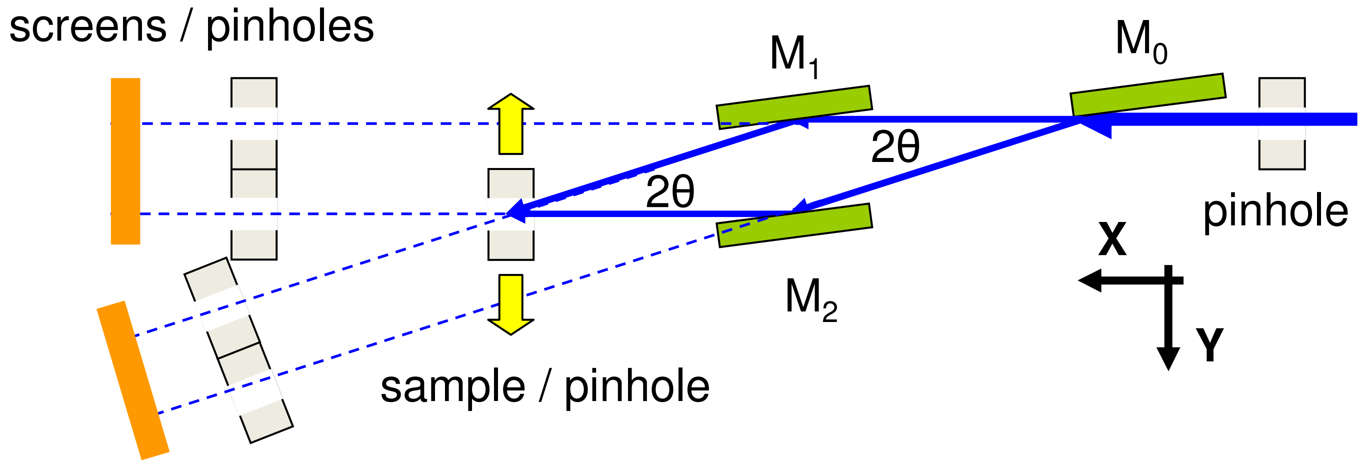
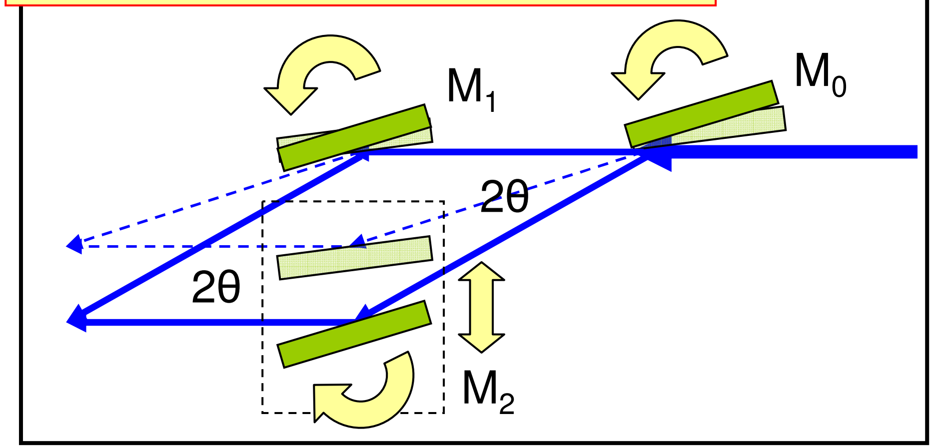
"mini-TIMER" (@DiProI)



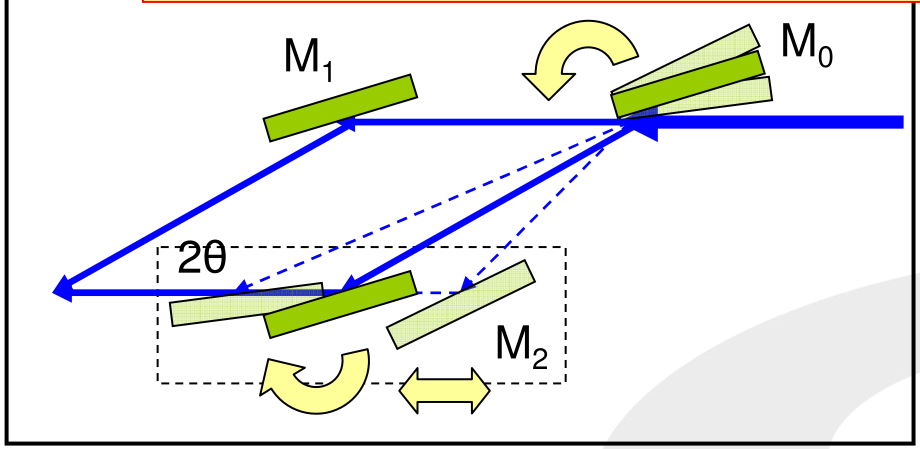
Alignement (reference pinholes + screens) → $\delta\theta \approx 0.03^o$
Degrees of freedom: $M_{1,2,3}$ pitch-roll-Z-Y; $M_{1,2}$ X;
sample/sample pinhole X,Y,Z,pitch,roll



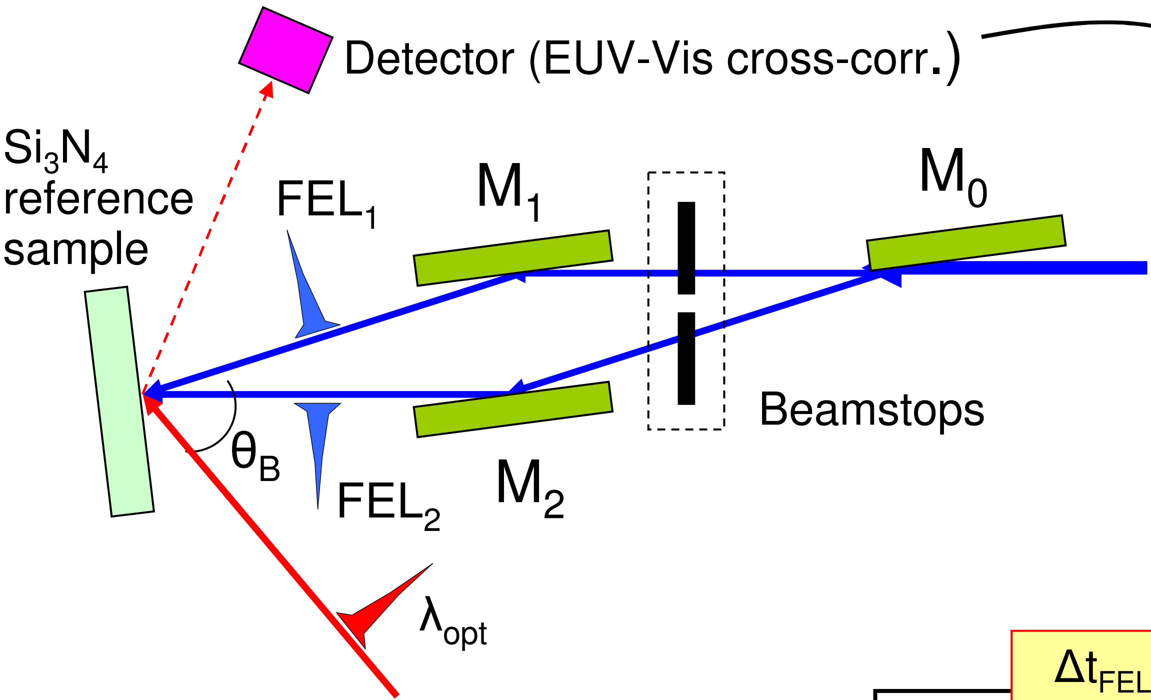
2θ = 2-9° at $\Delta t_{FEL\text{-}FEL}$ = constant

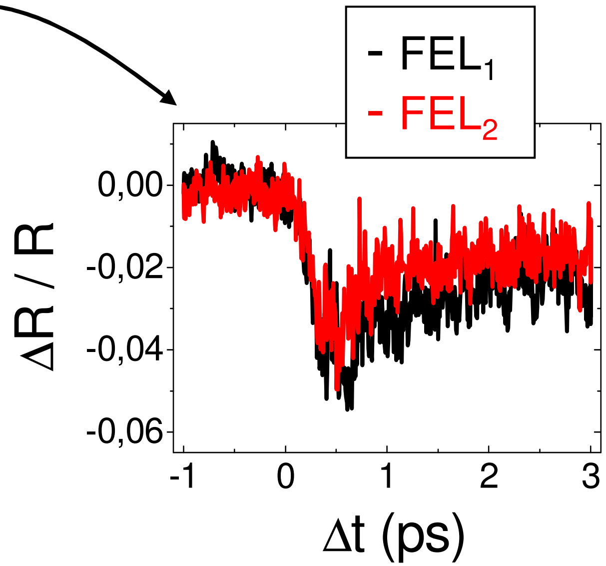


$\Delta t_{FEL\text{-}FEL}$ = ± 0.2 ps at 2θ = constant



"mini-TIMER" (@DiProI)

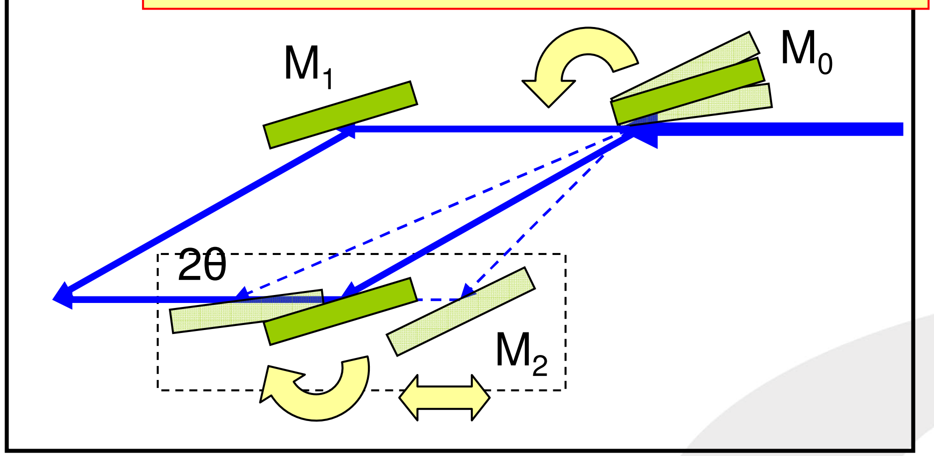




If TR signals are equal, then all pulses are in time-space coincidence and similar FEL fluence in the interaction region

Our setup can be also used as a compact split-and-delay stage for FEL-pump/FEL-probe measurements, with the advantage of spatial pump-probe separation ($2\theta > 0$)

$\Delta t_{FEL-FEL} = \pm 0.2$ ps at 2θ = constant



F. Capotondi (unpublished)

“mini-TIMER” (@DiProI)

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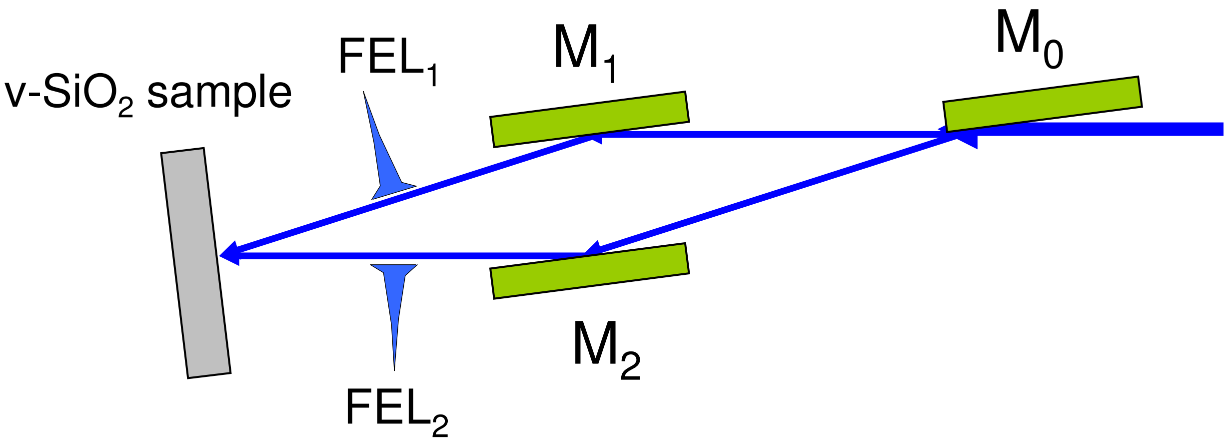
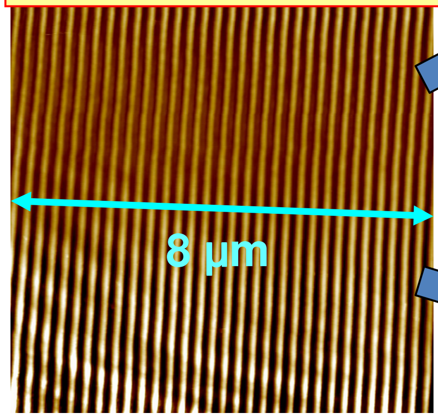


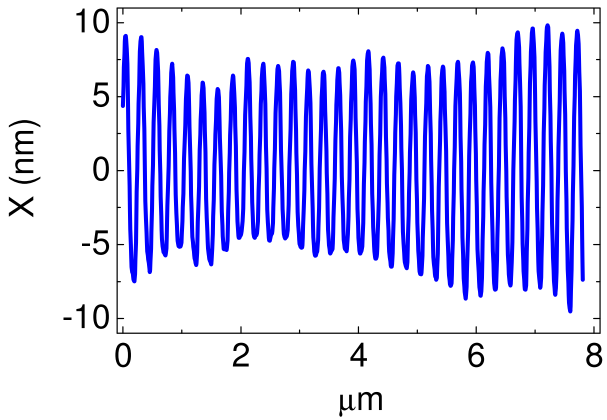
Inprints on SiO_2 (and PMMA) → 2θ = 6.16°

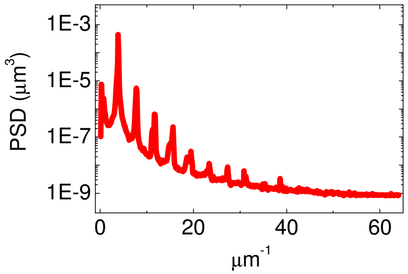
Grating visibility after multi-shot explosure → FEL_1-FEL_2 optical path difference < λ_{FEL} (< 27.6 nm)

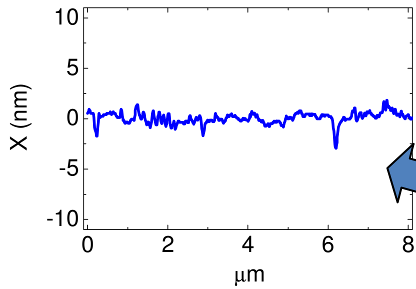
Quantitative analysis (also single/multi-shot on PMMA) is running

Permanent gratings on SiO_2 (after 1000’s shots @ FEL flux > 50 mJ/cm^2)

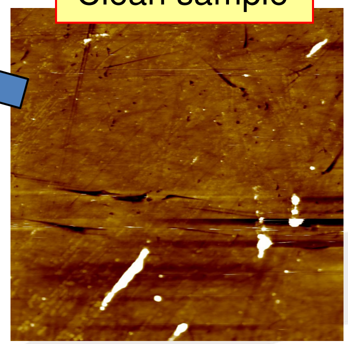




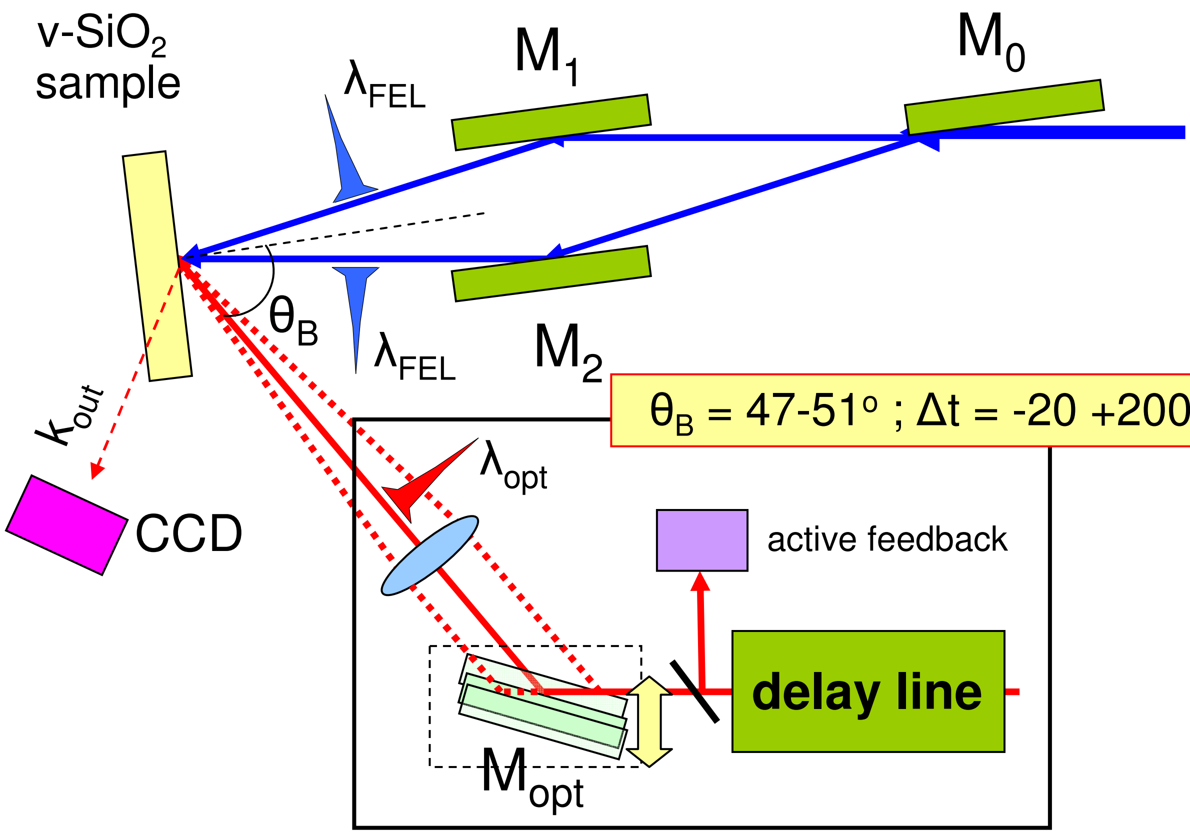




Clean sample

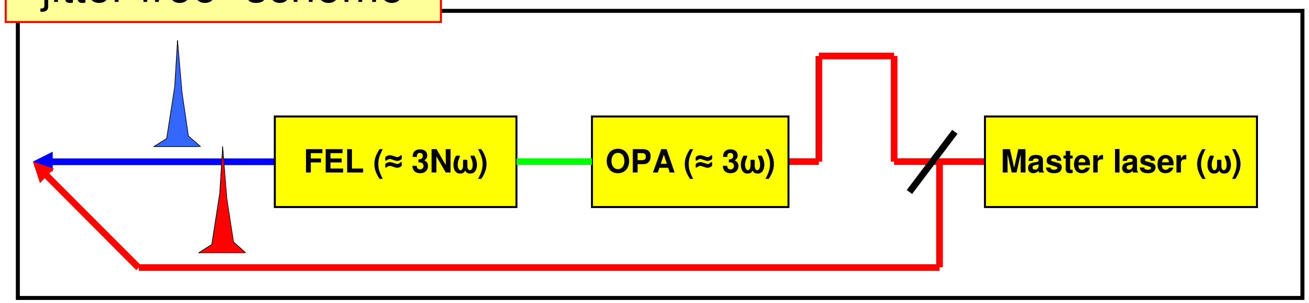


“mini-TIMER” (@DiProI)



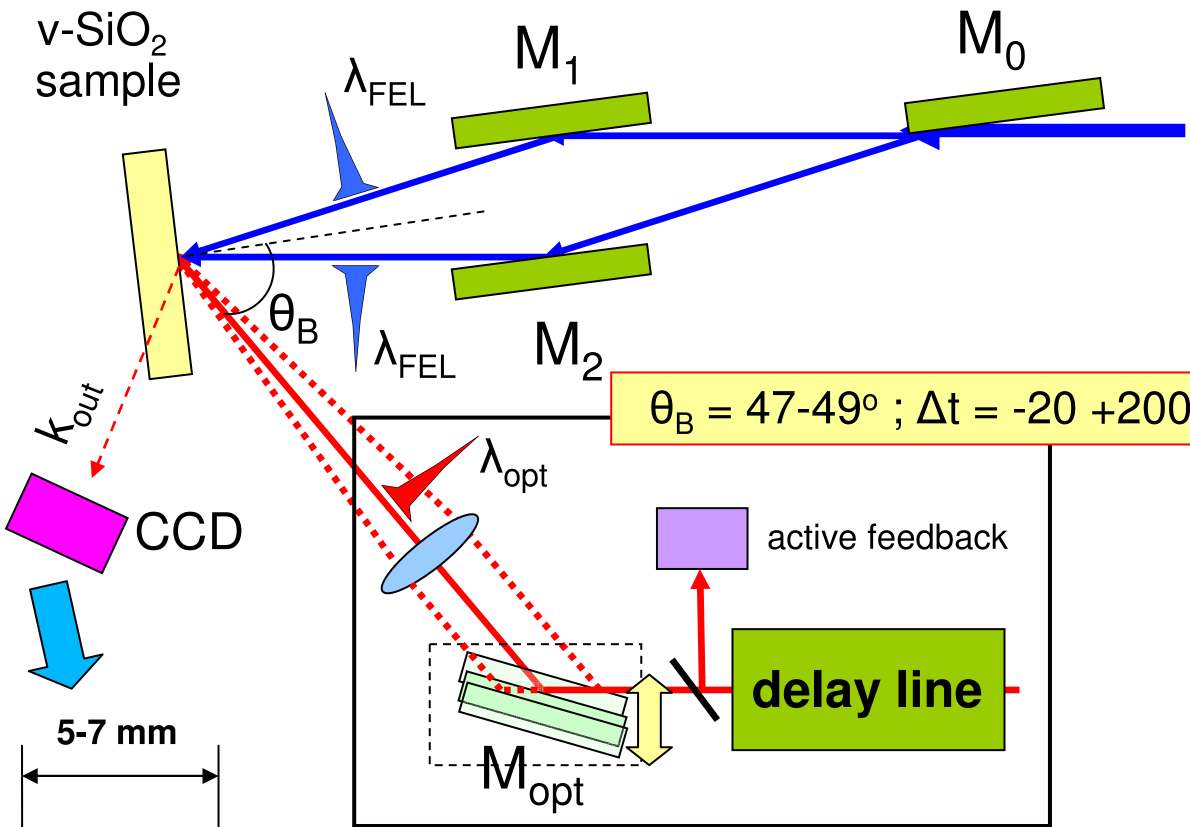
θB = 47-51° ; Δt = -20 +200 ps

“jitter free” scheme[1]

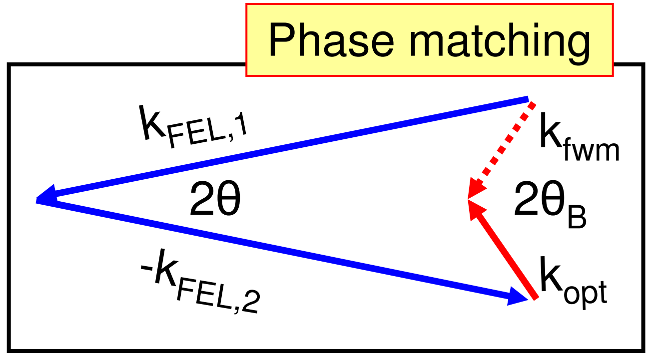


1) M. B. Danailov et al., Opt. Express (2014)

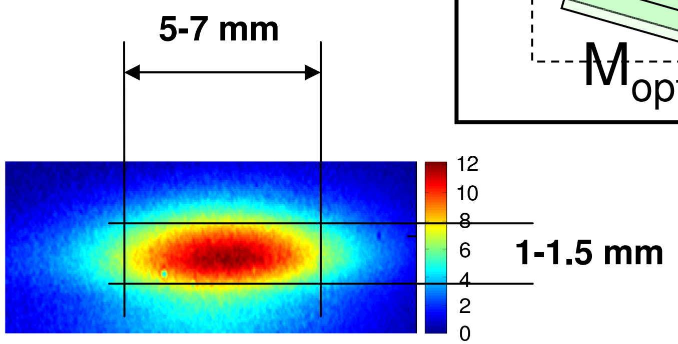
FEL-stimulated FWM signal



θ_B = 47-49° ; Δt = -20 +200 ps



2θ = 6.16°
θ_B = 49.9°
λ_{opt} =392.8 nm
λ_{FEL} =26.7 nm
$\Delta t = \Delta t_{FEL\text{-}FEL} = 0$



Set of 5 signal-background images (total 3000-3000 shots with-without FEL, integr. time ≈ 12 min / point.)

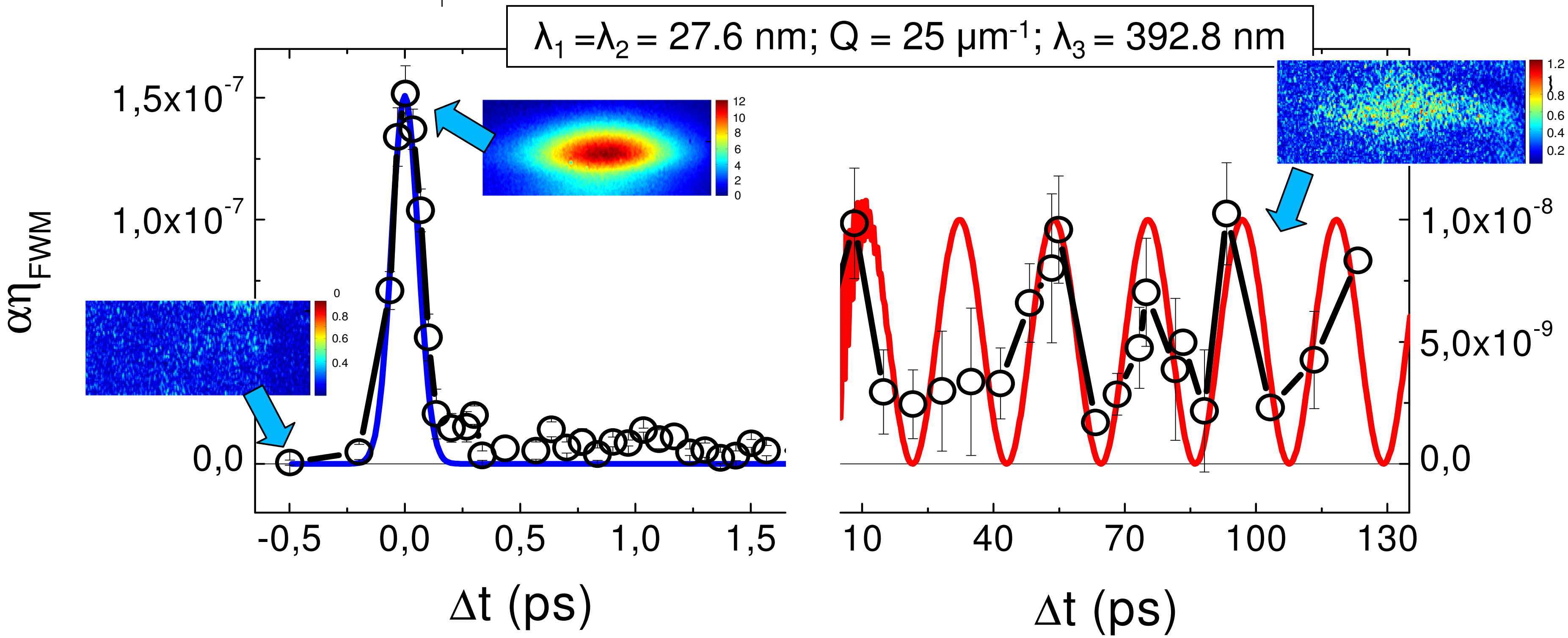
A "well defined" coherent beam propagates along k_{out} → **FWM**

Vertical dimension fits with the divergence of input beams

Horizontal dimension is larger due to thin grating effects (L_{abs} for EUV field ≈ 55 nm < λ_{opt})

F. Bencivenga (unpublished)

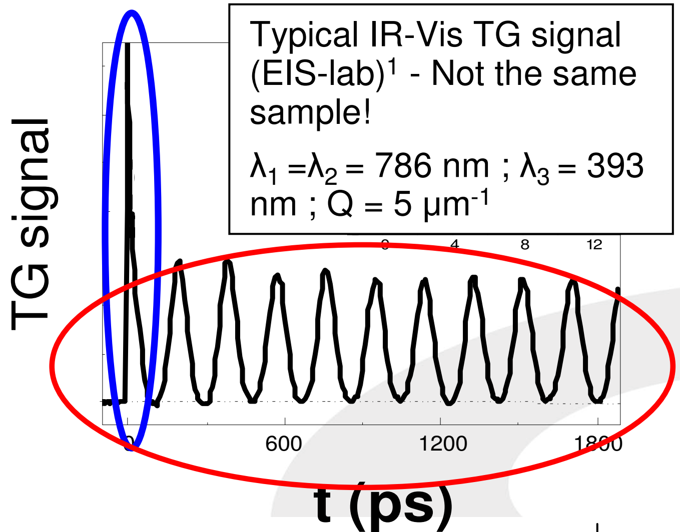
FEL-stimulated FWM signal



Δt ≈ 0: sharp TG peak ($R_{cc}(\Delta t)$; ≈130-140 fs FWHM, resolution limited) → **electronic response** (coherent spike)

TG signal extends up to Δt ≈ 100 ps → **Longitudinal acoustic mode** at (almost) the expected frequency ($\omega_{LA} = c_s Q \approx 0.145$ THz) and lifetime > 1 ns

TG efficiency (I_{TG} / I_{opt}) at Δt=0 ≈ 10^{-7} (lower but still comparable to the IR-VIS) and $I_{TG}(\Delta t>0)/I_{TG}(\Delta t=0) \approx 10^{-2}$ (much larger than in the IR-VIS, typical ≈ 10^{-5})



1) R. Cucini et al., Opt. Lett. 2011

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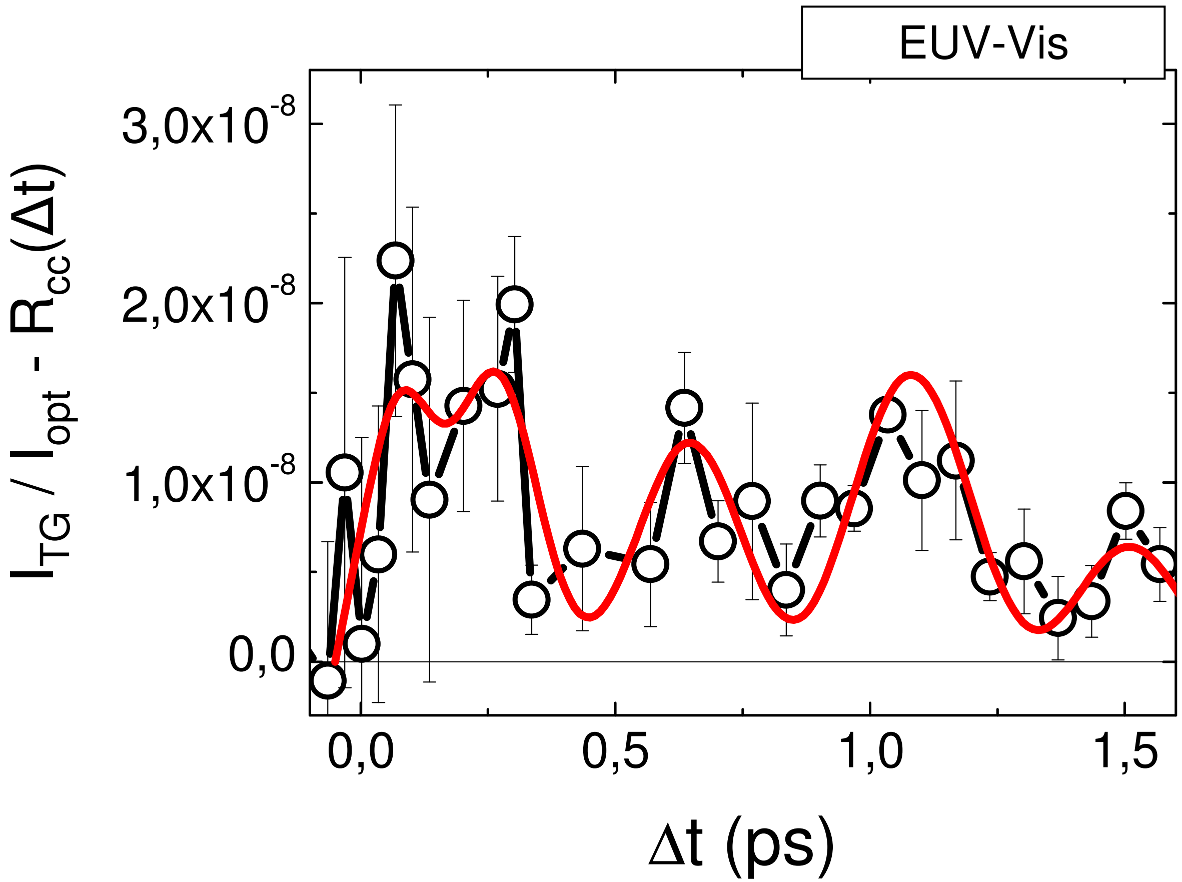
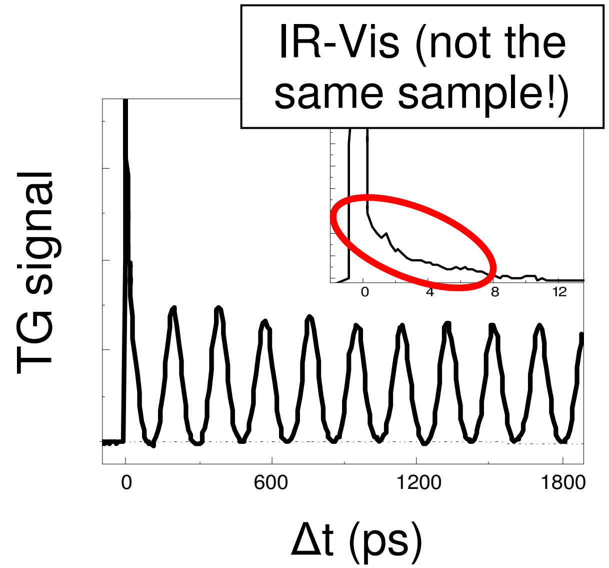
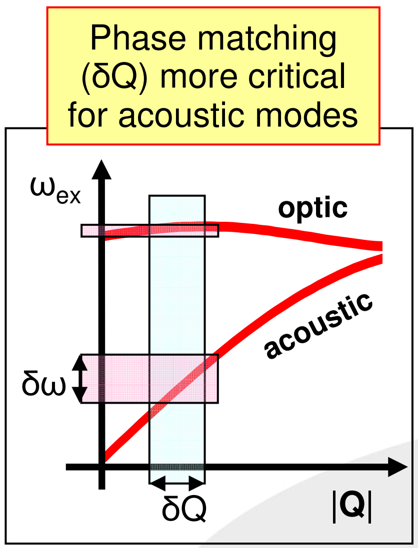
FEL-stimulated FWM processes







Δt = 0-1.5 ps: **two oscillations** ("optic modes") at $\omega_1 \approx 7.2$ THz (F_1 hyper-Raman mode → tetrahedral rotations) and $\omega_1 \approx 26$ THz (ν_{2b} Raman mode → tetrahedral bendings).

F. Bencivenga (unpublished)

Next step: TG + two-colour

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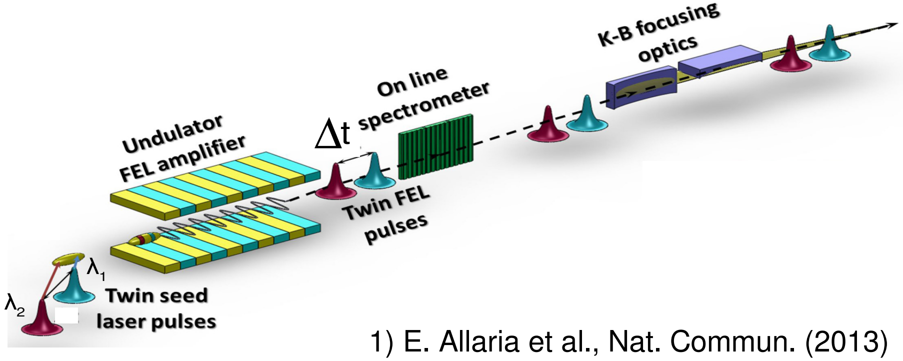
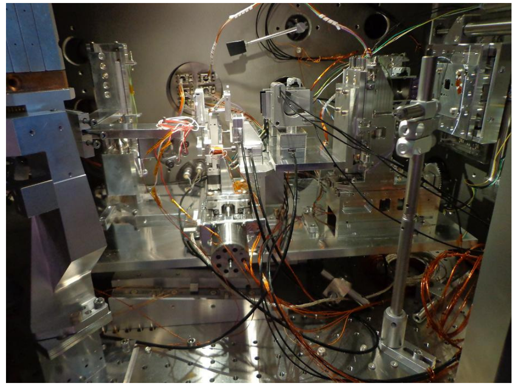
"twin-seed" two-colour[1]

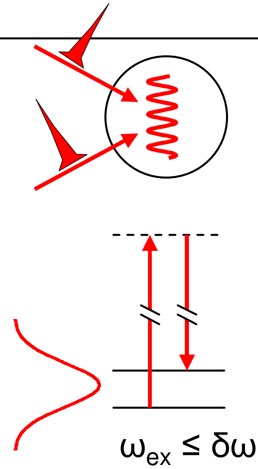


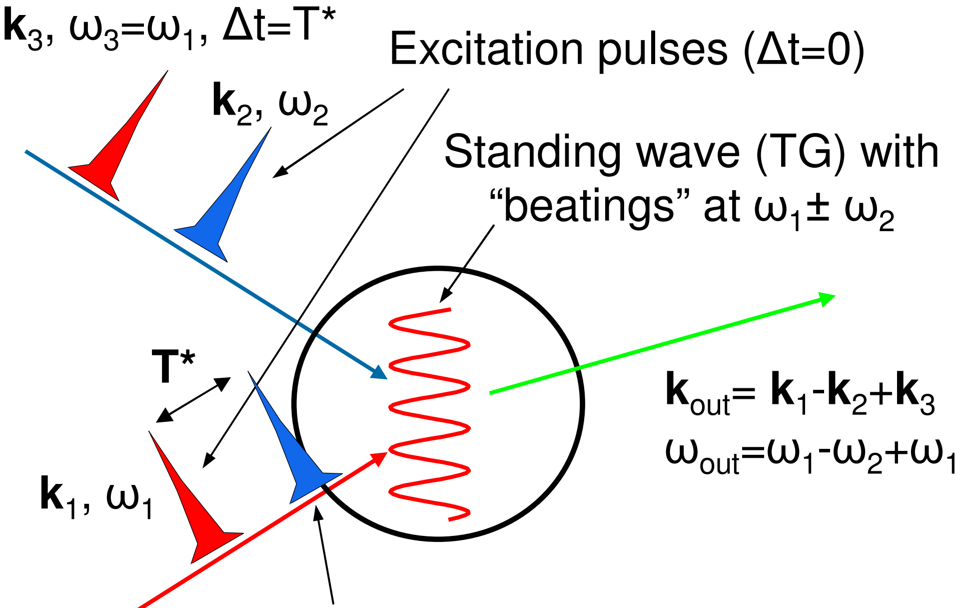
1) E. Allaria et al., Nat. Commun. (2013)

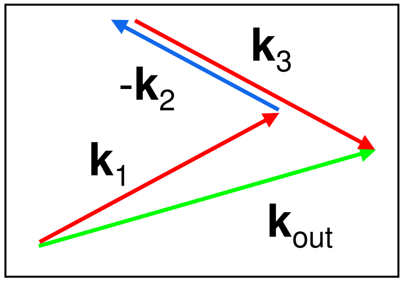
+

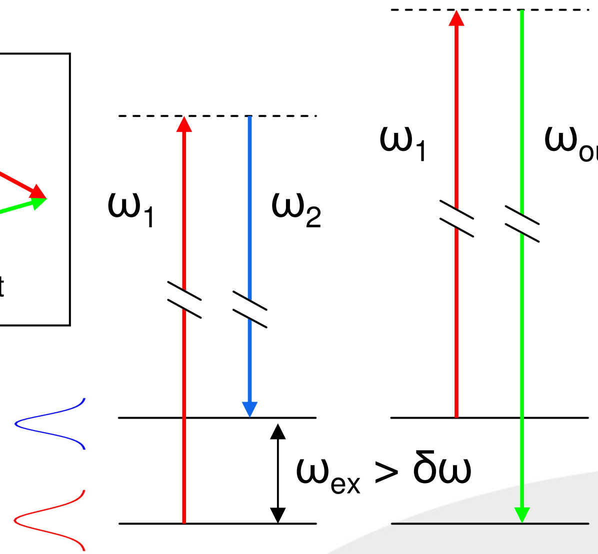
"mini-TIMER"







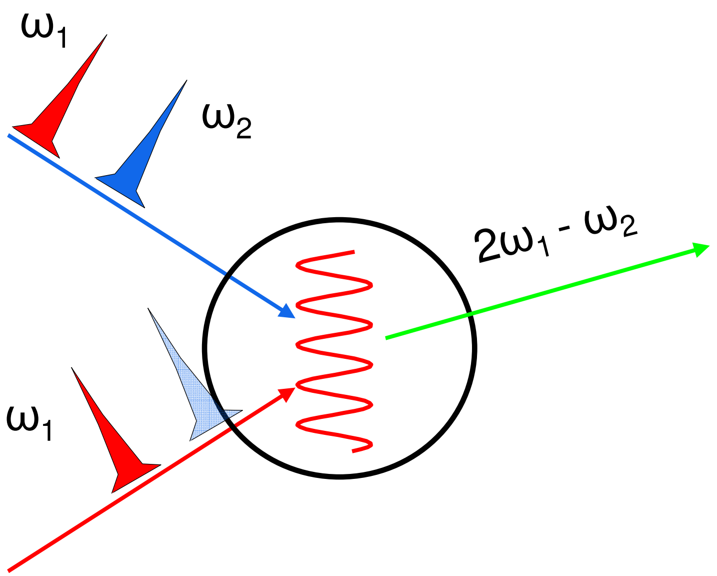


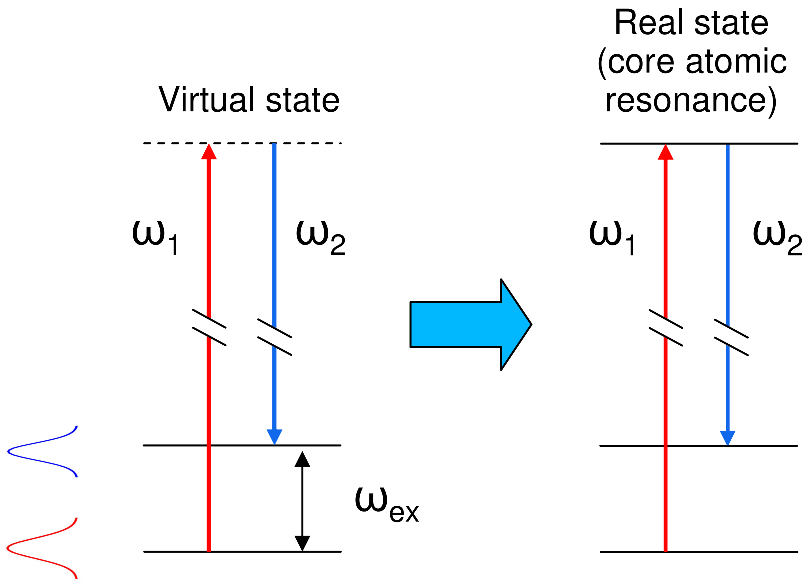


"all-FEL-based" coherent (anti-stokes) Raman scattering

F. Bencivenga et al., Faraday Discus. (2014); Exp scheduled in Sept. 2015

Next step: TG + two-colour



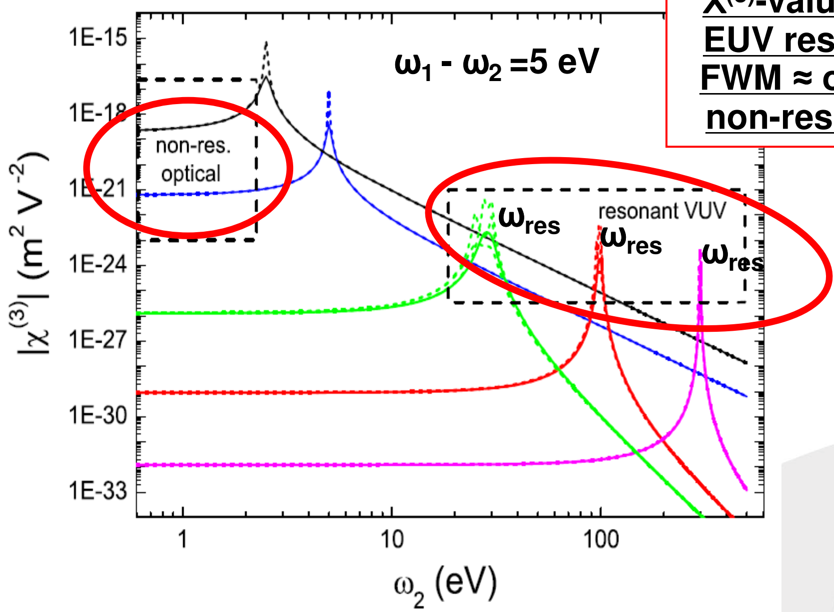


Optical input fields (ω_i ~ eV)
→ ω_{ex} < 0.1's eV (vibrations)

EUV/x-ray fields (ω_i > 100's eV)
→ **ω_{ex} ~ 1-10 eV's** (excitons)

Atomic seletivity through core resonances ($\omega_1=\omega_{res}$)

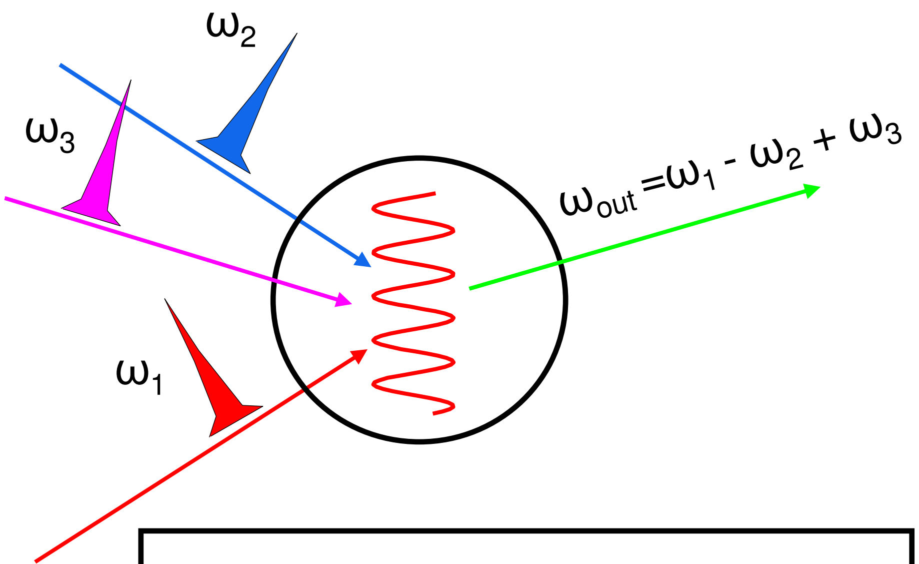
Control the atomic site where the excitation is generated

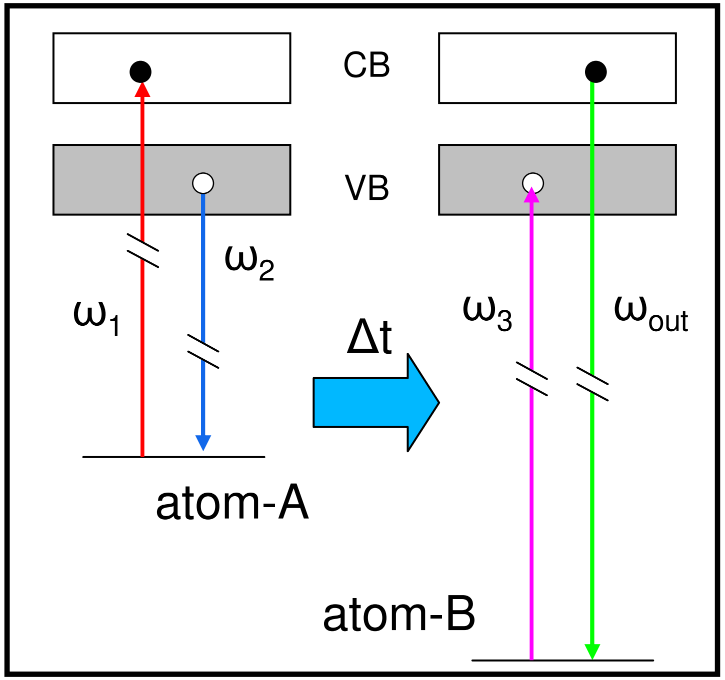


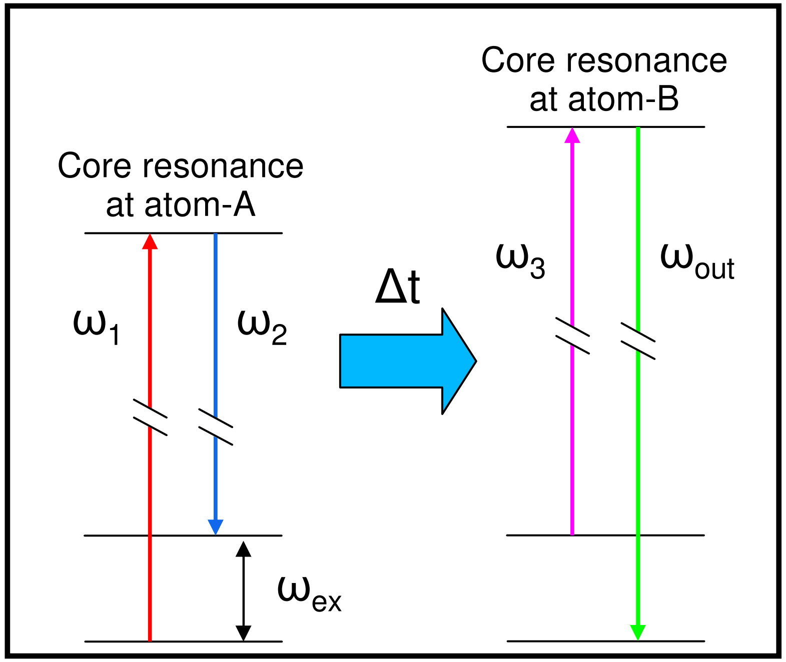
$X^{(3)}$-values for EUV resonant FWM ≈ optical non-resonant

F. Bencivenga et al., Faraday Discus. (2014); Exp scheduled in Sept. 2015

Outlook: TG + three-colour







Tuning ω_i's (to ω_{res}'s of selected elements) and Δt one can chose where a selected excitation is created, as well as where and when it is probed → delocalization of electronic states, charge and energy transfer processes, etc.

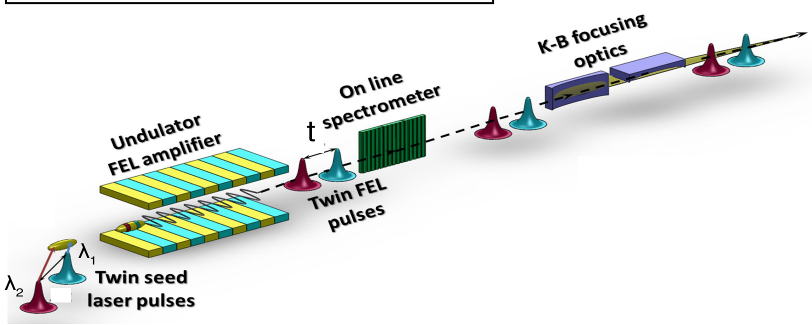
If λ_i's compare to molecular size (x-ray) then dipole selection rules do not apply → possible to probe the entire manifold of electronic transitions

S. Tanaka and S. Mukamel, PRL (2002) **and many other works from Mukamel's group**

Elettra Sincrotrone Trieste

Outlook: TG + three-colour

“twin-seed” two-colour



PRL 110, 134801 (2013) PHYSICAL REVIEW LETTERS 29 MARCH

Experimental Demonstration of Femtosecond Two-Color X-Ray Free-Electron Lasers

A. A. Lutman, R. Coffee, Y. Ding,* Z. Huang, J. Krzywinski, T. Maxwell, M. Messerschmidt, and H.-D. Nuhn
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(Received 13 December 2012; published 25 March 2013)

With an eye toward extending optical wave-mixing techniques to the x-ray regime, we present the first experimental demonstration of a two-color x-ray free-electron laser at the Linac Coherent Light Source. We combine the emittance-spoiler technique with a magnetic chicane in the undulator section to control the pulse duration and relative delay between two intense x-ray pulses and we use differently tuned canted pole undulators such that the two pulses have different wavelengths as well. Two schemes are shown to produce two-color soft x-ray pulses with a wavelength separation up to ~1.9% and a controllable relative delay up to 40 fs.

Possible to achieve a three-colour seeded FEL emission at FERMI, but the tunability in ω_i‘s is limited by the FEL gain bandwidth → ω_{ex} < 1 eV

PRL 113, 024801 (2014) PHYSICAL REVIEW LETTERS week ending 11 JULY 2

Free-Electron Laser Design for Four-Wave Mixing Experiments with Soft-X-Ray Pulses

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(Received 24 March 2014; published 10 July 2014)

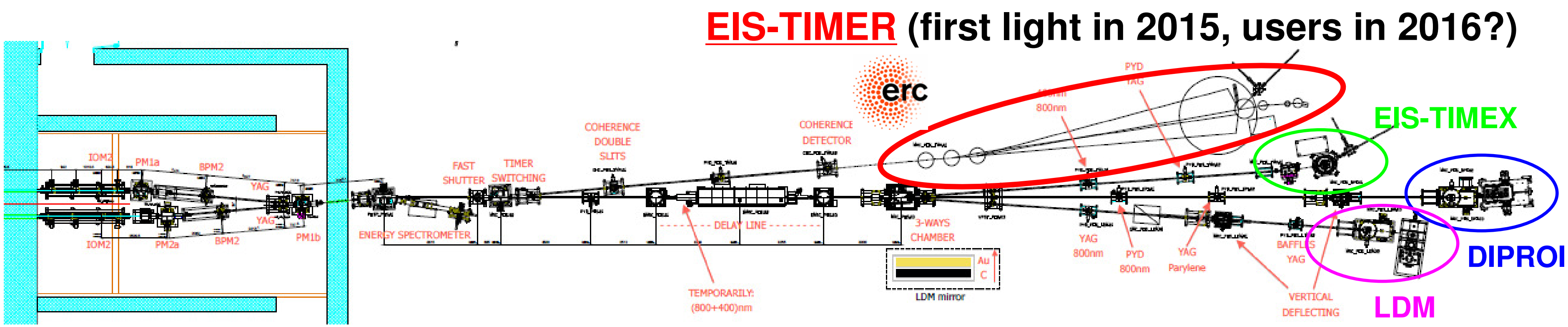
We present the design of a single-pass free-electron laser amplifier suitable for enabling four-wave mixing x-ray spectroscopic investigations. The production of longitudinally coherent, single-spike pulses of light from a single electron beam in this scenario relies on a process of selective amplification where a strong undulator taper compensates for a large energy chirp only for a short region of the electron beam. This proposed scheme offers improved flexibility of operation and allows for independent control of the color, timing, and angle of incidence of the individual pulses of light at an end user station. Detailed numerical simulations are used to illustrate the more impressive characteristics of this scheme.

New schemes to achieve larger separation (eV’s) in the photon energy are under study at FERMI

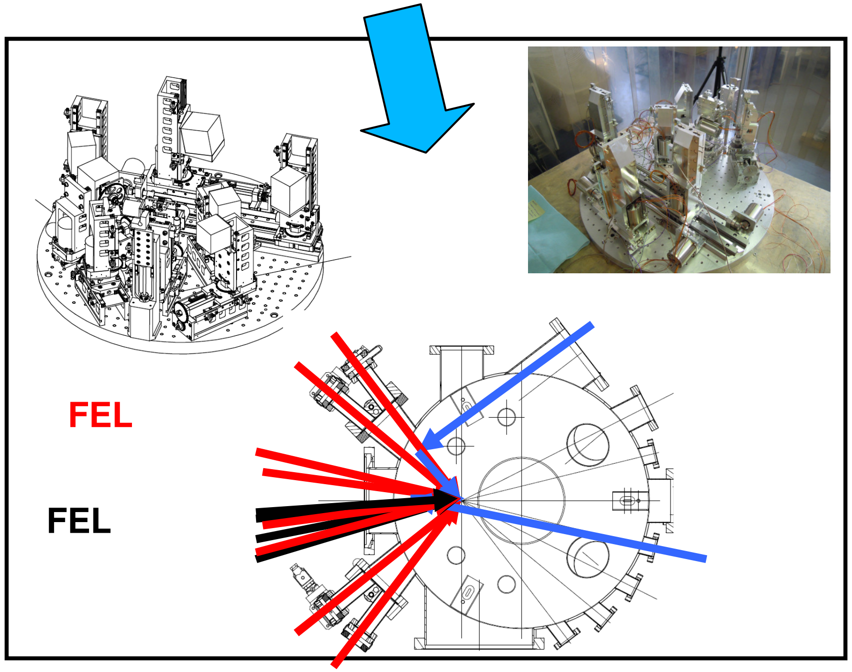
Efforts/proposal on the “machine side“ are coming up

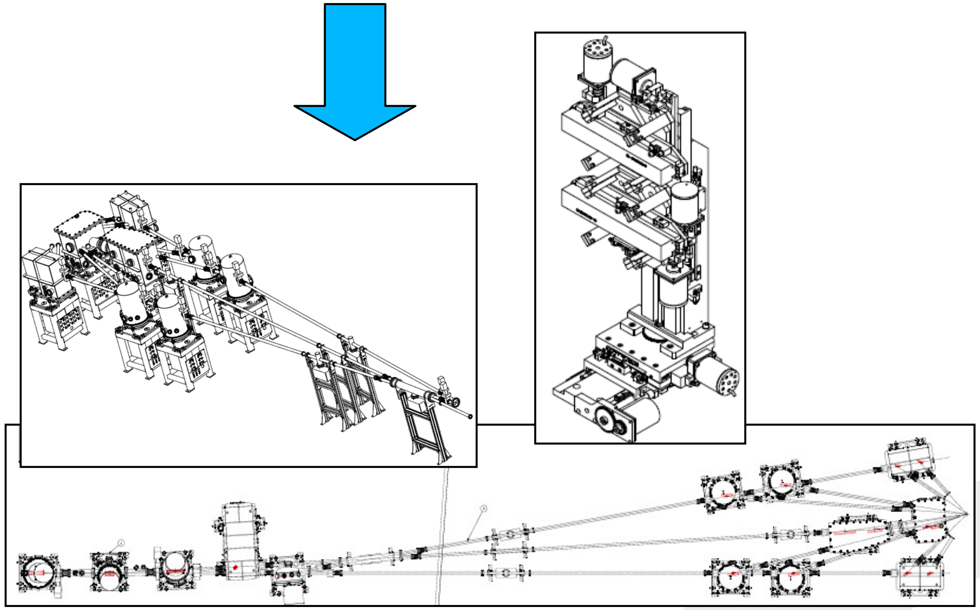
EUV/x-ray non linear coherent methods based on FEL’s might be established in a near future

Outlook: EIS-TIMER beamline



End-station ready, optics almost ready, photon transport system under construction





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Conclusions

- An experimental-end station (EIS-TIMER) dedicated to non-linear, wave-mixing experiments will be available at FERMI in 2015 (original goal is to study vibrations in the 0.1-1 nm^{-1} Q-range in disordered systems and nanostructures)

- Experimental evidences of FEL-induced four-wave-mixing processes
  → Experimental setup to carry out EUV/soft x-ray four-wave-mixing experiments (with transient gratings) at the DiProI end-station, with large room for improvements…
  → The signal is substantially larger than what expected and the electronic/nuclear signal ratio is larger than in the optical regime
  → Observed three oscillating features, ascribable to vibrational modes (phonons)

- The possibility to exploit multi-colour seeded FEL sources and experimental setups dedicated to transient grating experiments (such as “mini-TIMER” or EIS-TIMER) would allow to develop advanced FWM methods in the near future.

Elettra Sincrotrone Trieste

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