Elettra
Sincrotrone
Trieste

Wave-mixing experiments with multi-colour seeded FEL pulses

Filippo Bencivenga

Outline

1) The TIMER project: collective atomic dynamics in the "mesoscopic" range by EUV/soft x-ray transient grating

2) Transient grating and wave-mixing

3) "mini-TIMER"@DiProI: FEL-stimulated **transient grating** (preliminary results, experiment done a few weeks ago!)

4) **Two-colours** FEL-pump/FEL-probe experiment on Ti

5) Outlook: **two-colour** (better three...) + **transient grating** setup
→ advanced four-wave-mixing applications at FERMI

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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?

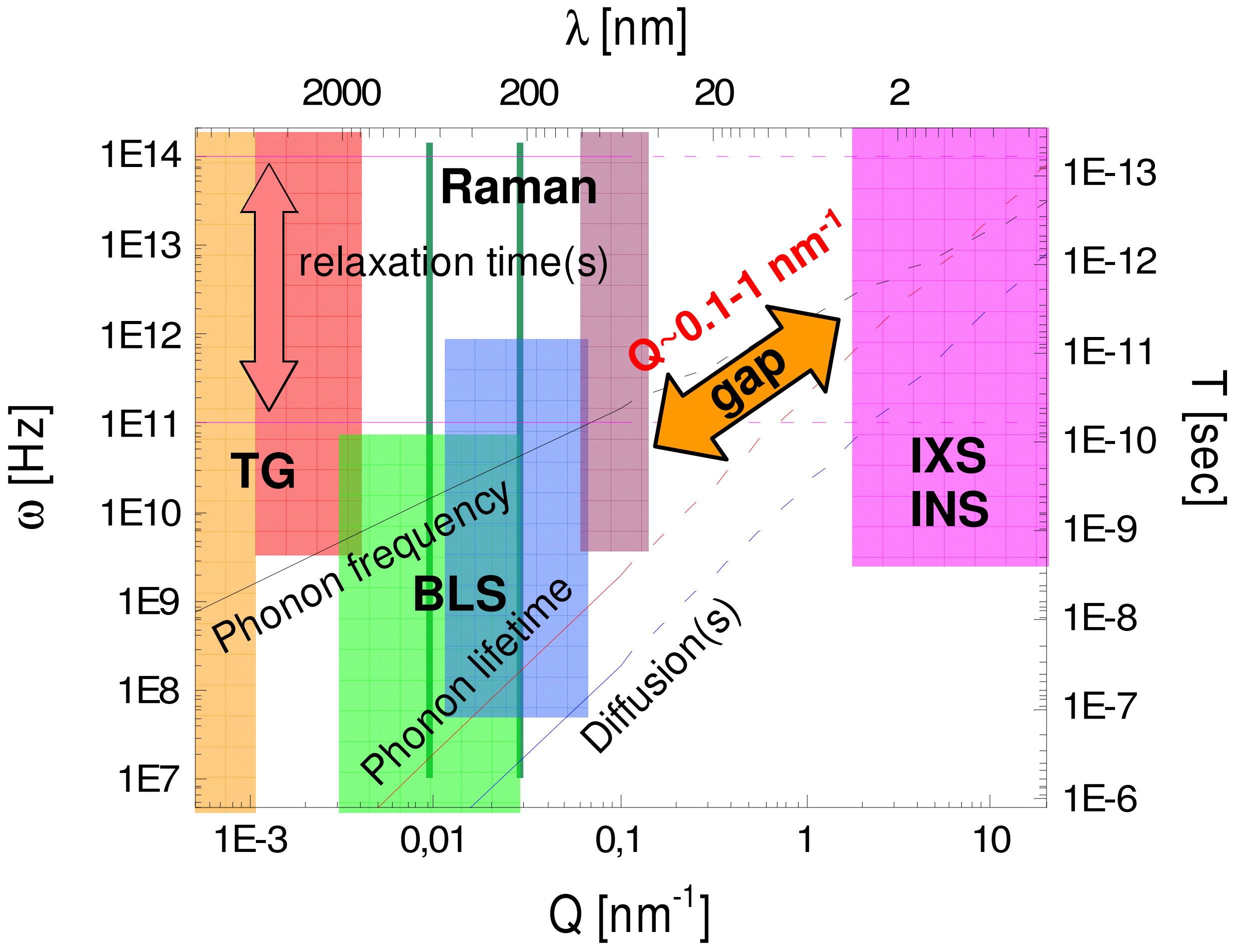
Sonoluminescence
What causes the emission of short bursts of light from imploding bubbles in a liquid when excited by sound?

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



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 …)

Methods

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

TIMER's goal is to fill the gap

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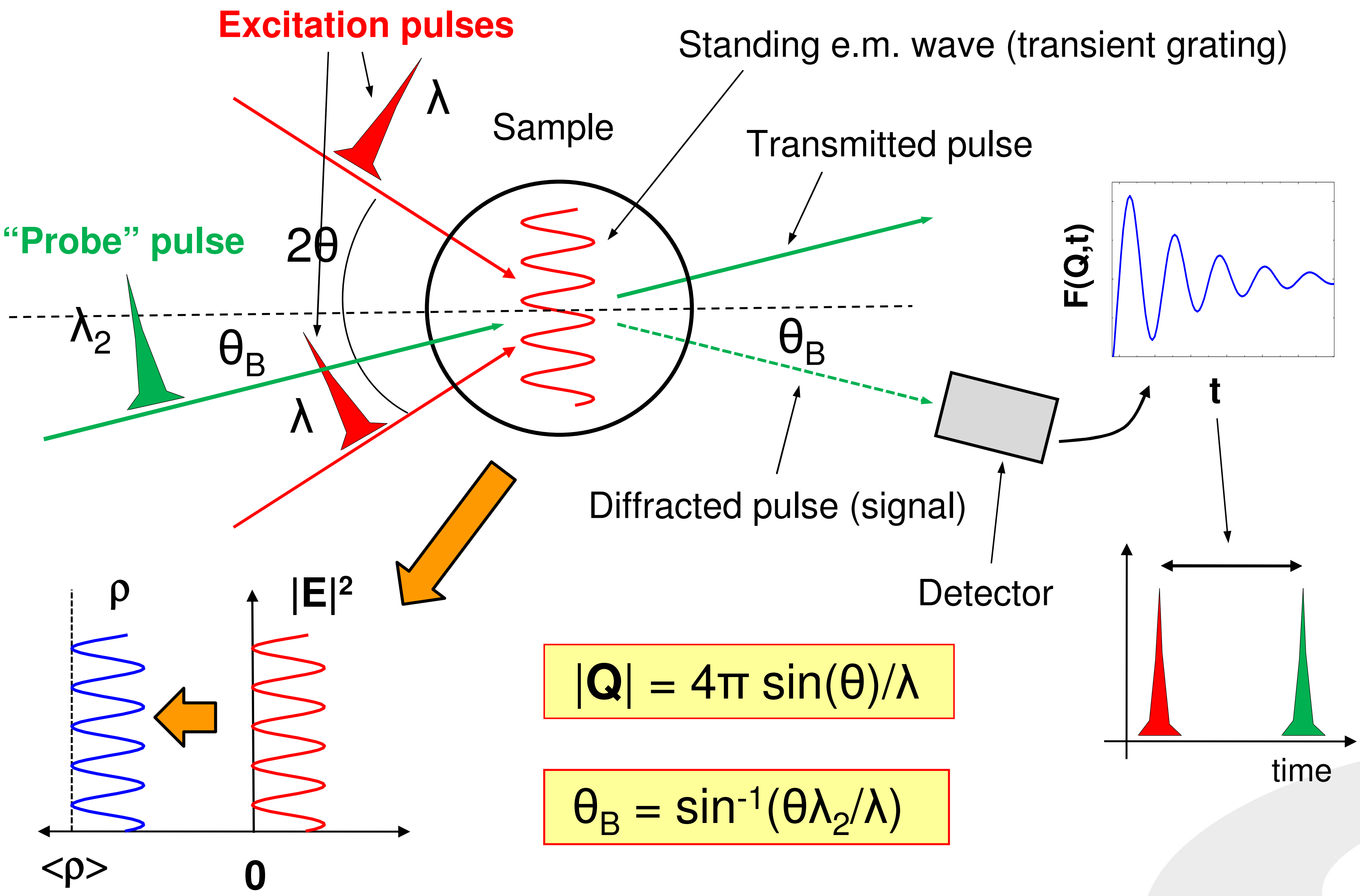
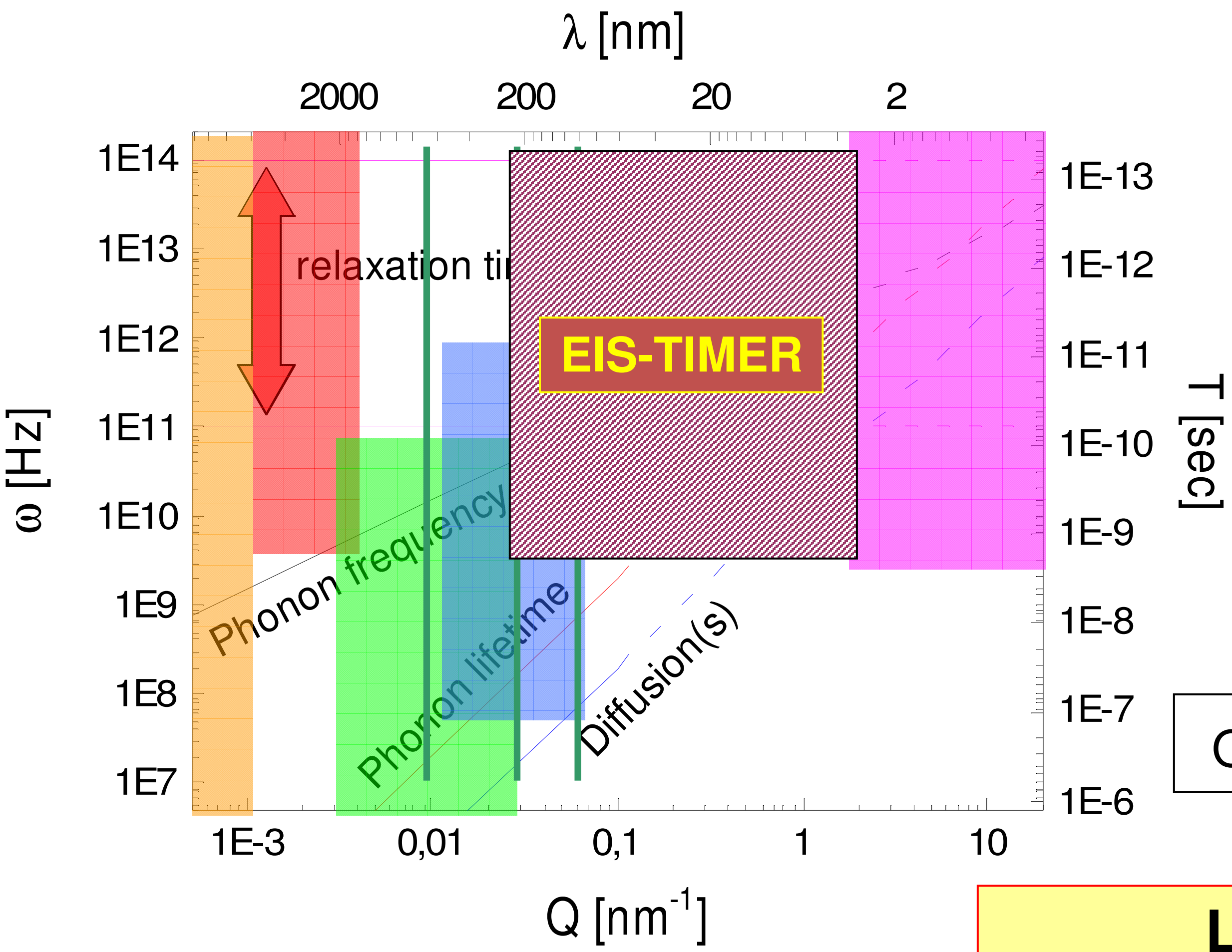
Transient grating method



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

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

EUV/x-ray transient grating



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

EIS-TIMER beamline[1]:
$\theta = 9.2^{o} - 52.7^{o}$
$\lambda = 60 - 10$ nm (~ 3 nm)
$\lambda_{pr} = \lambda / 3$

$Q = 0.03 - 1\ nm^{-1}$ (~ $3\ nm^{-1}$)

L* ~ 2π / Q*

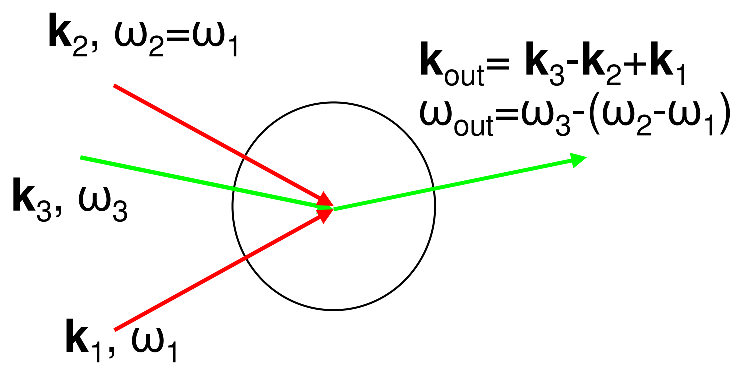
- **disordered systems (L* ~ 10 nm)**
- **nanostructures (L* ~ 1-100 nm)**

1) F. Bencivenga and C. Masciovecchio, NIMA (2008)

Non-linear (wave-mixing) signal

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

Driving forces 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. For TG: $\omega_{out} = \omega_1 - \omega_1 + \omega_3 = \omega_3$

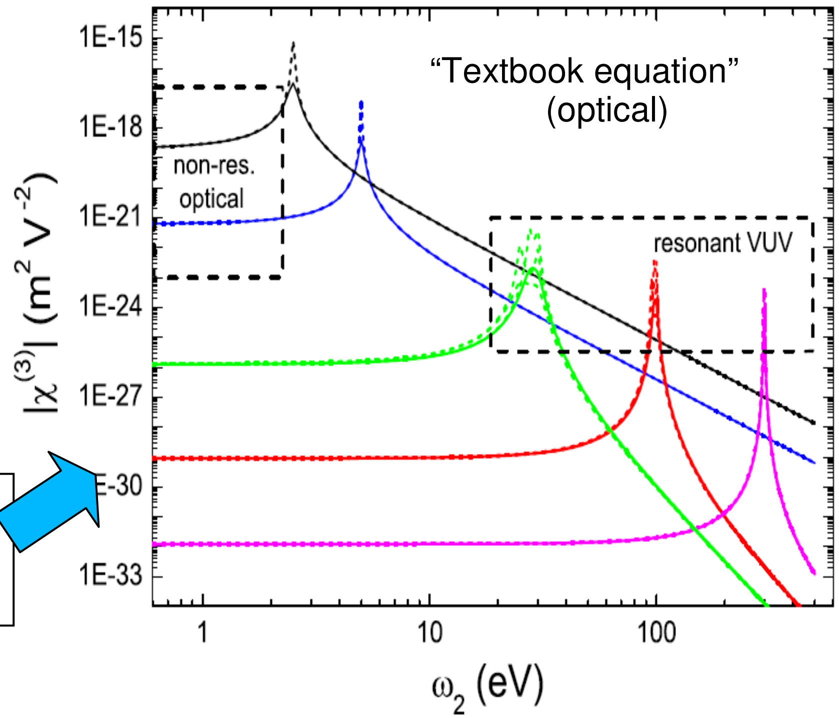


$\chi^{(2n)}=0$ (inv. sym.)
→ only available exp. evidence of x-ray induced wave-mixing[1]

$\chi^{(n)} \sim E_a^{-n+1}$ ($E_a \sim$ 10-100 V/nm)
$E_i < 1$ V/nm (e.g., damage)

$\chi^{(3)} E_j^2 / \chi << 10^{-4}$
→ $I_{fwm}/I_{lin} << 10^{-8}$

$\chi^{(n)}$ decreases on increasing ω_i [2]



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

2) B. D. Patterson, SLAC-TN (2011); F. Bencivenga et a., NJP (20013)

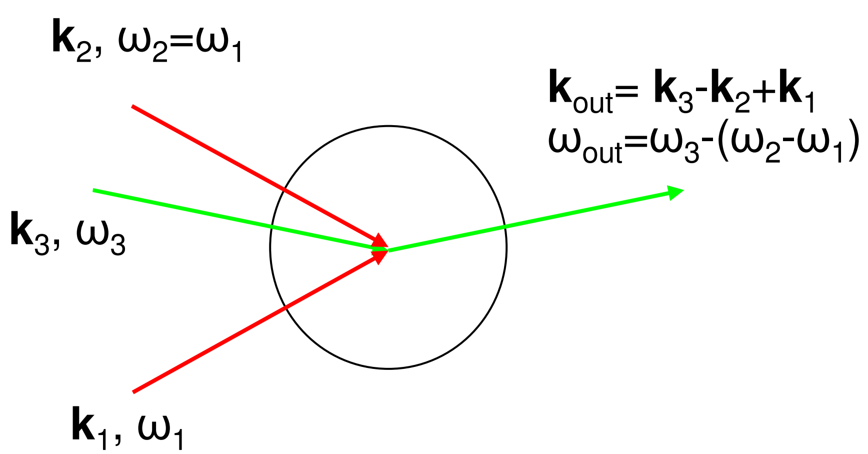
Non-linear (wave-mixing) signal

Phase matching → E_{out} fields radiated by the N elementary scatterers in different sample locations (within δk^{-1}: coherence length of the non-linear process) **add in amplitude** ($I_{out} \sim |\Sigma E_{out}|^2 \sim N^2$) not in intensity ($\Sigma |E_{out}^2| \sim N$) **along k_{out}**$= \Sigma_{p(i,j,k,...)} \pm k_i$ → $\delta k = k_{out} - \Sigma_{p(i,j,k,...)} \pm k_i$; $\delta k \neq 0$ because, e.g., **finite bandwidth** (coherence) and divergence

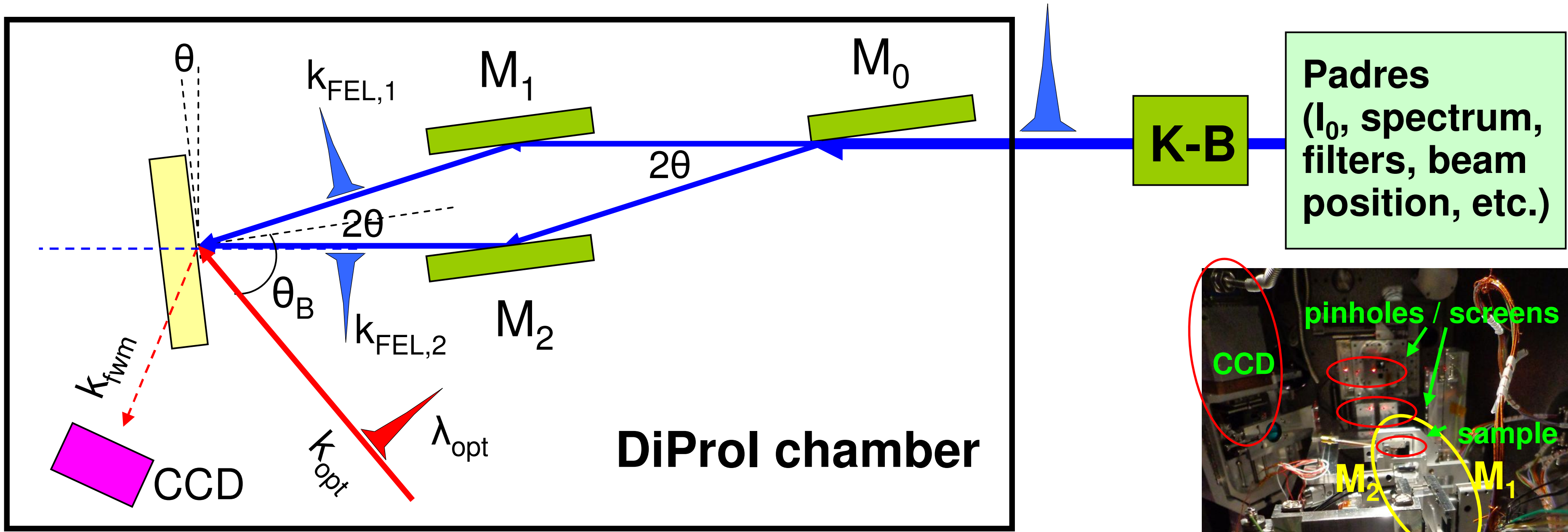
Directionality: Non-linear "phase matched" signal under $\Delta\Omega \sim 10^{-6}$ srad, linear signal much more isotropic ($\Delta\Omega \sim 4\pi$ srad) → non-linear/linear gain along $k_{out} \sim 10^7$

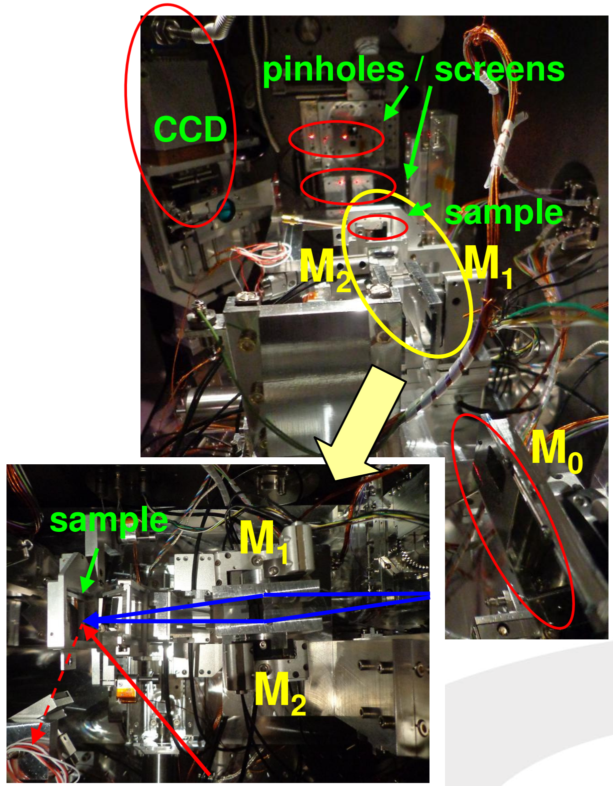
Coherent addition: the non-linear signal may become dominating, even a macroscopic beam (e.g., harmonic generation)

FERMI: EUV pulses with narrow (almost Fourier-limited) bandwidth → increase in δk^{-1} → increase in N → **N^2 increase** of I_{fwm} **along k_{fwm}**

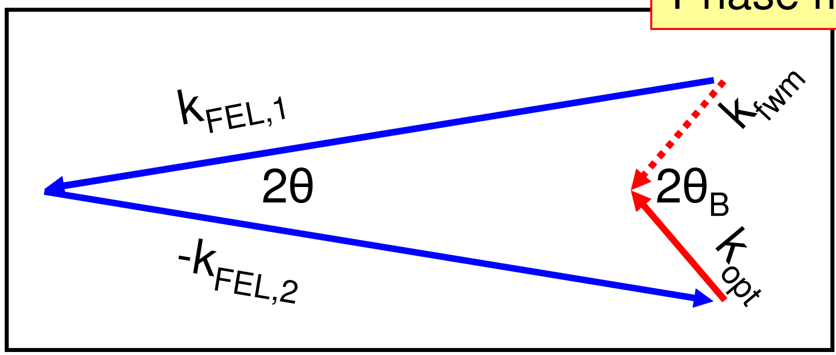


“mini-TIMER” (@DiProI)



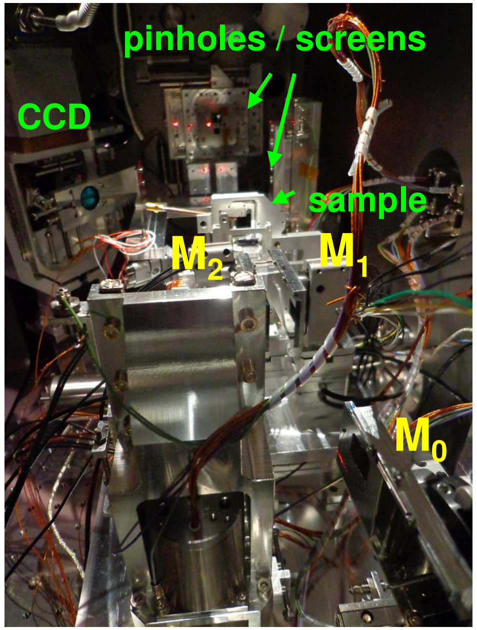


Phase matching

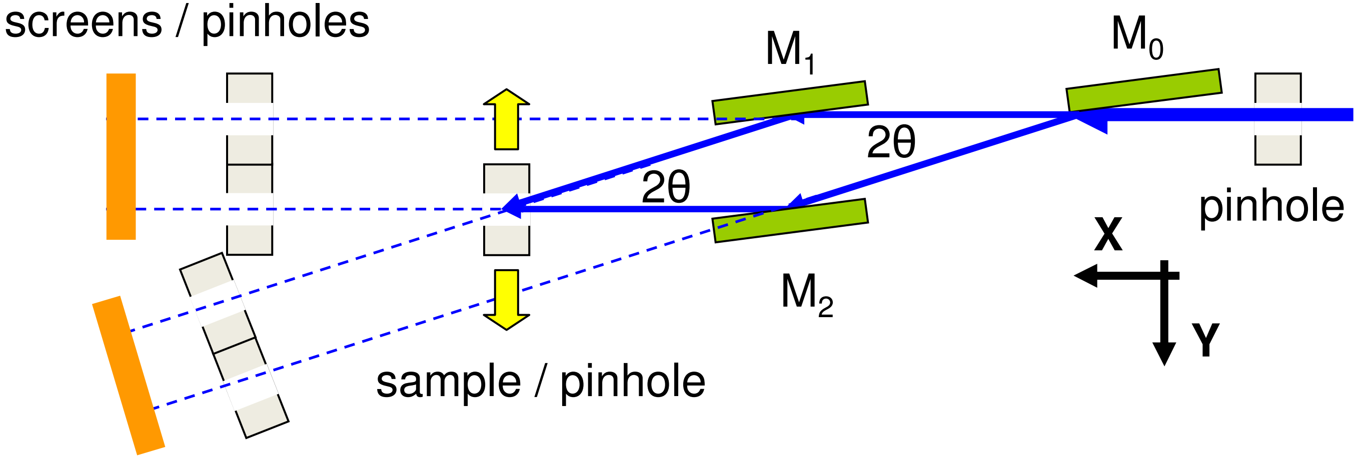


$2θ = 6^o$
$θ_B = 48.2^o$
$λ_{opt}$ =392.8 nm
$λ_{FEL}$ =27.6 nm

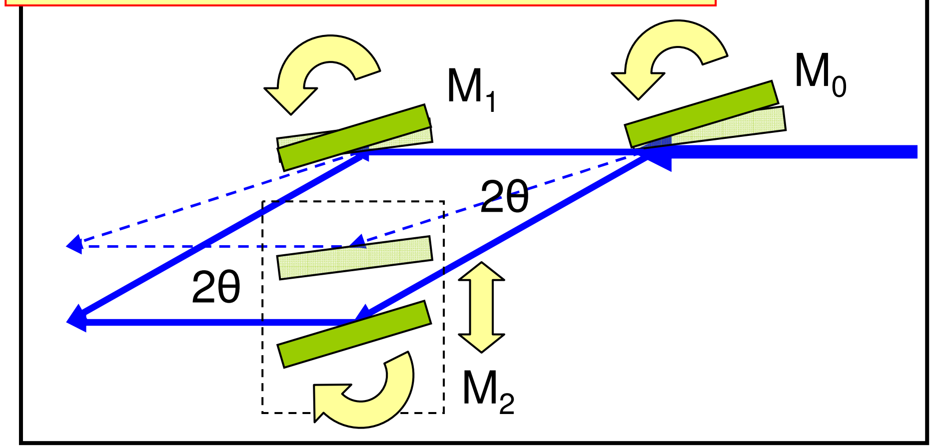
“mini-TIMER” (@DiProI)



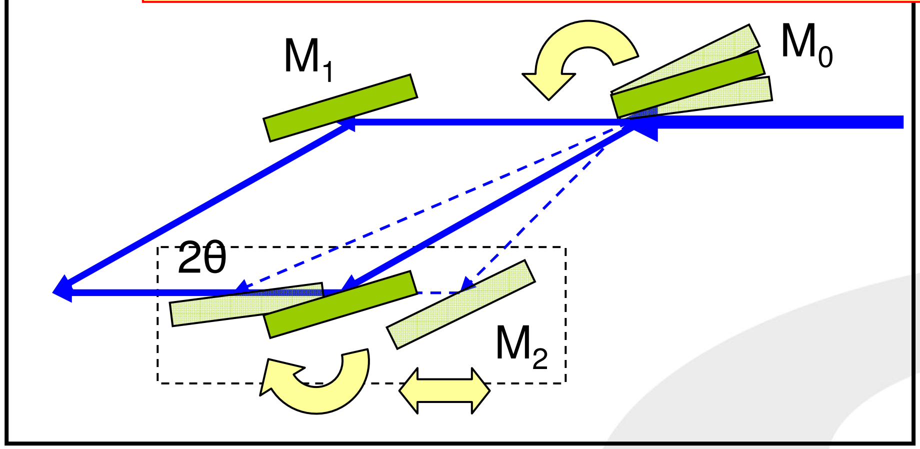
Alignement (reference pinholes + screens) → $\delta\theta \approx 0.2^o$
Degrees of freedom: $M_{1,2,3}$ pitch-roll-Z-Y; $M_{1,2}$ X;
sample/sample pinhole X,Y,Z,pitch,roll



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



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



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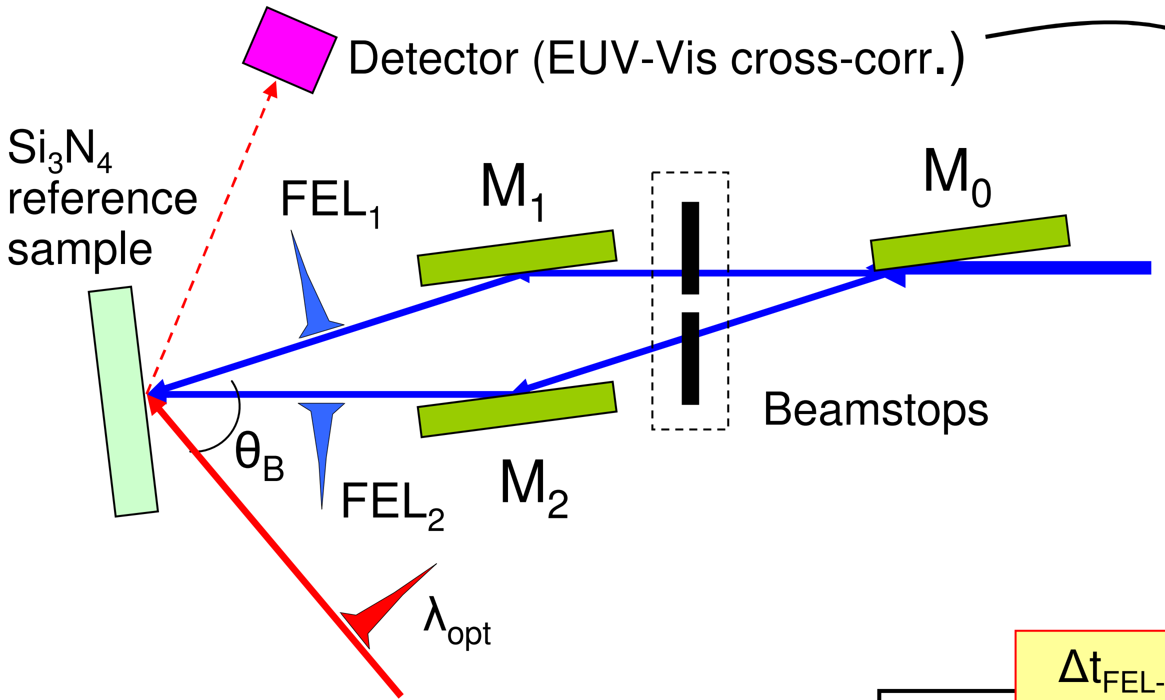
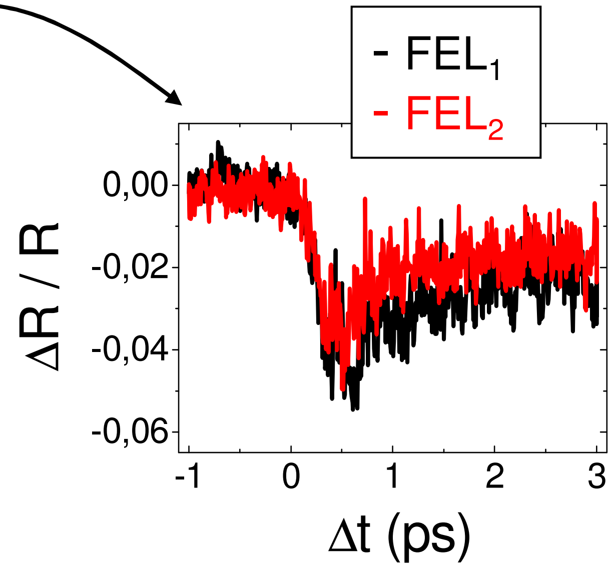
“mini-TIMER” (@DiProI)

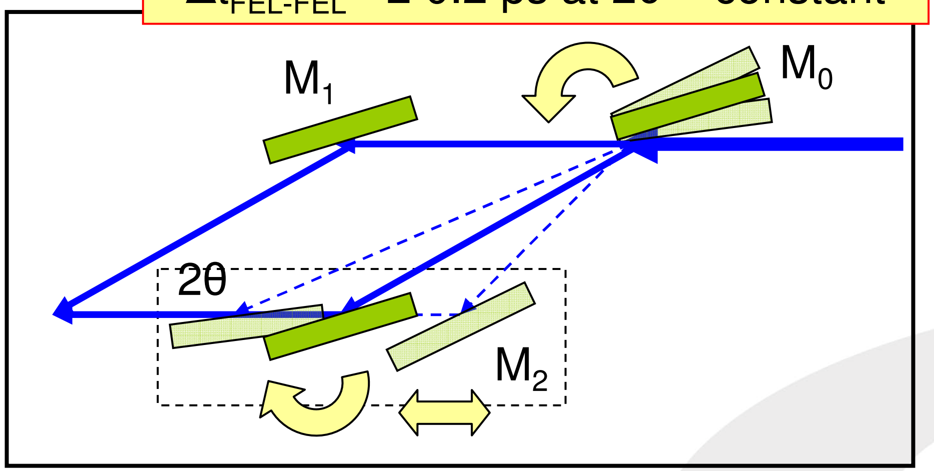




If TR signals are equal, then all pulses are in time-space coincidence and similar FEL fluence in the interaction region

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 ($2\theta > 0$)

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



F. Capotondi (unpublished)

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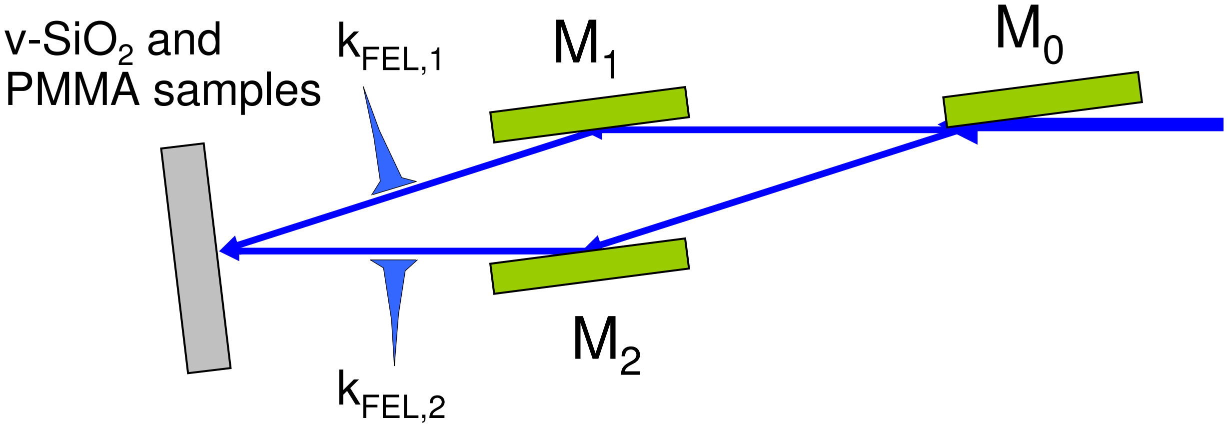
“mini-TIMER” (@DiProI)

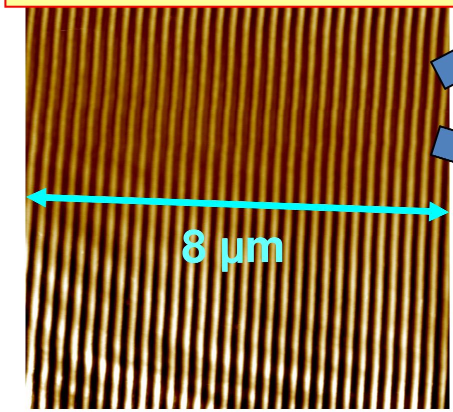


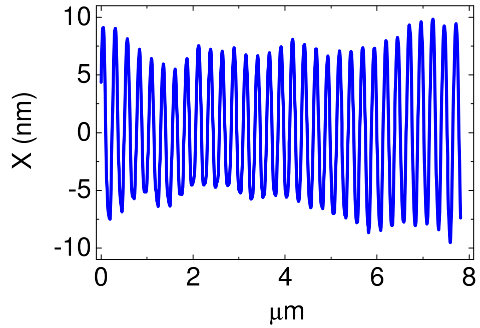
Inprints on PMMA (and SiO_2) → $2\theta = 3.16^o$

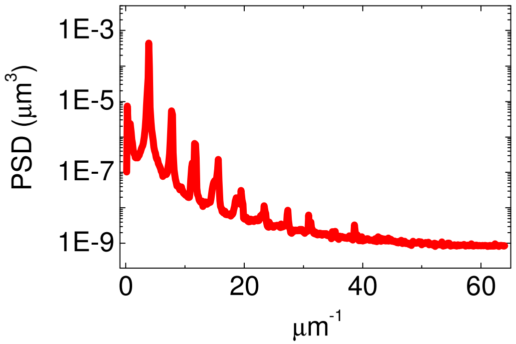
Grating visibility after multi-shot explosure → FEL_1-FEL_2 optical path difference < λ_{FEL} (< 27.6 nm)

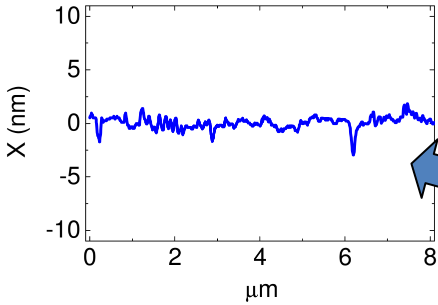
Quantitative analysis (also single/multi-shot on PMMA) is running

Permanent gratings on SiO_2 (@ FEL flux > 50 mJ/cm^2)

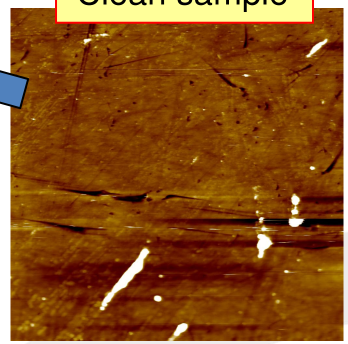




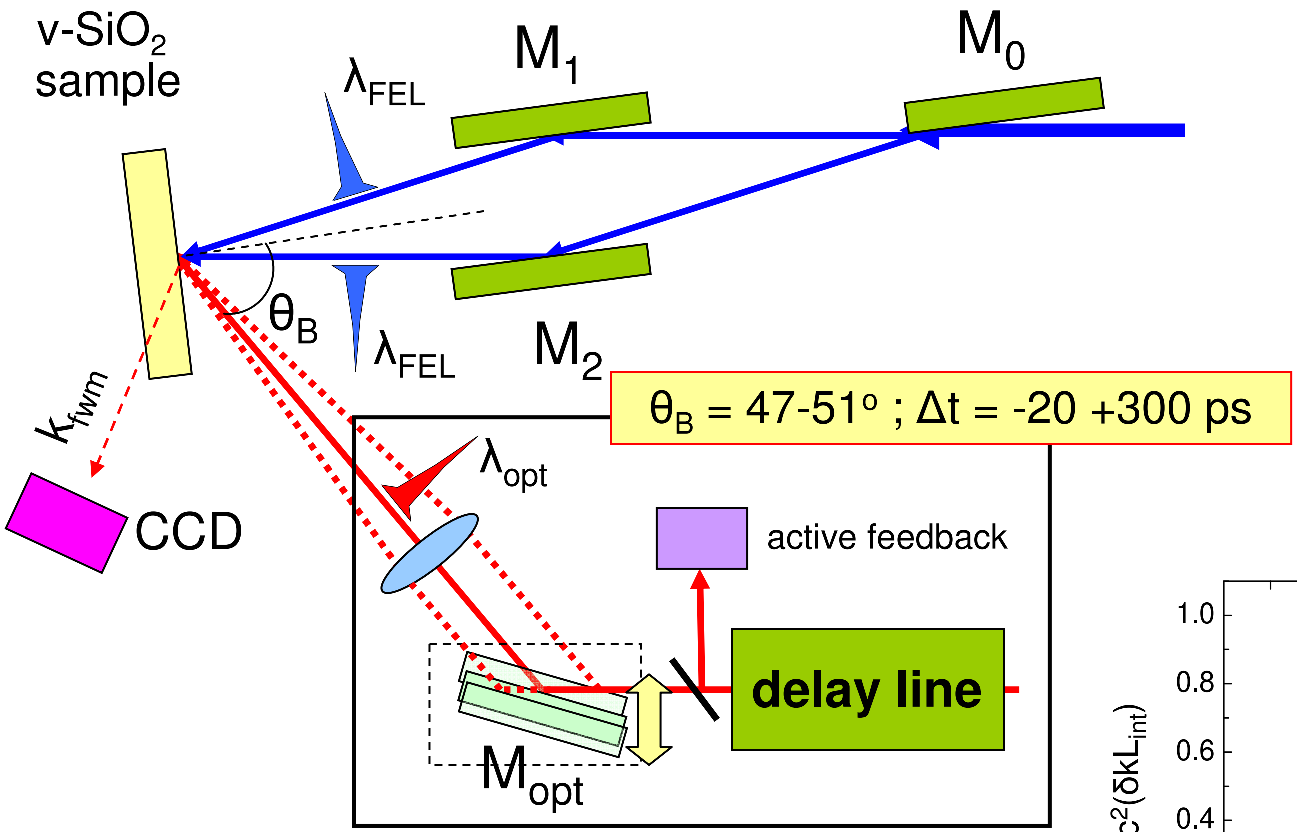




Clean sample



“mini-TIMER” (@DiProI)



Phase matching

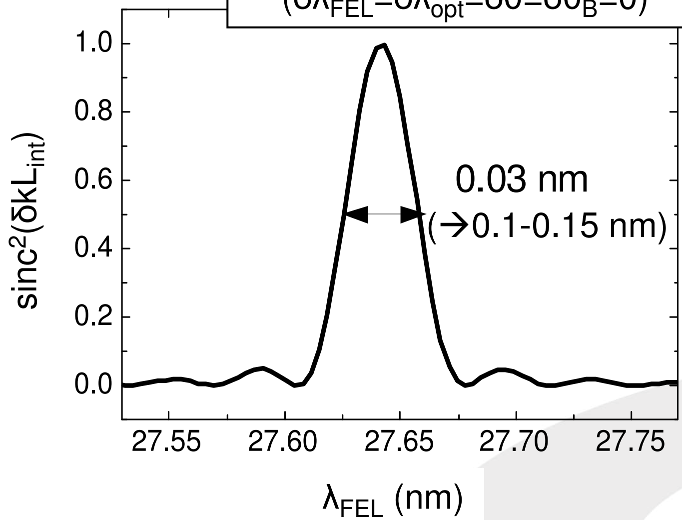
$2\theta = 6.16^o$

$\theta_B = 49.9^o$

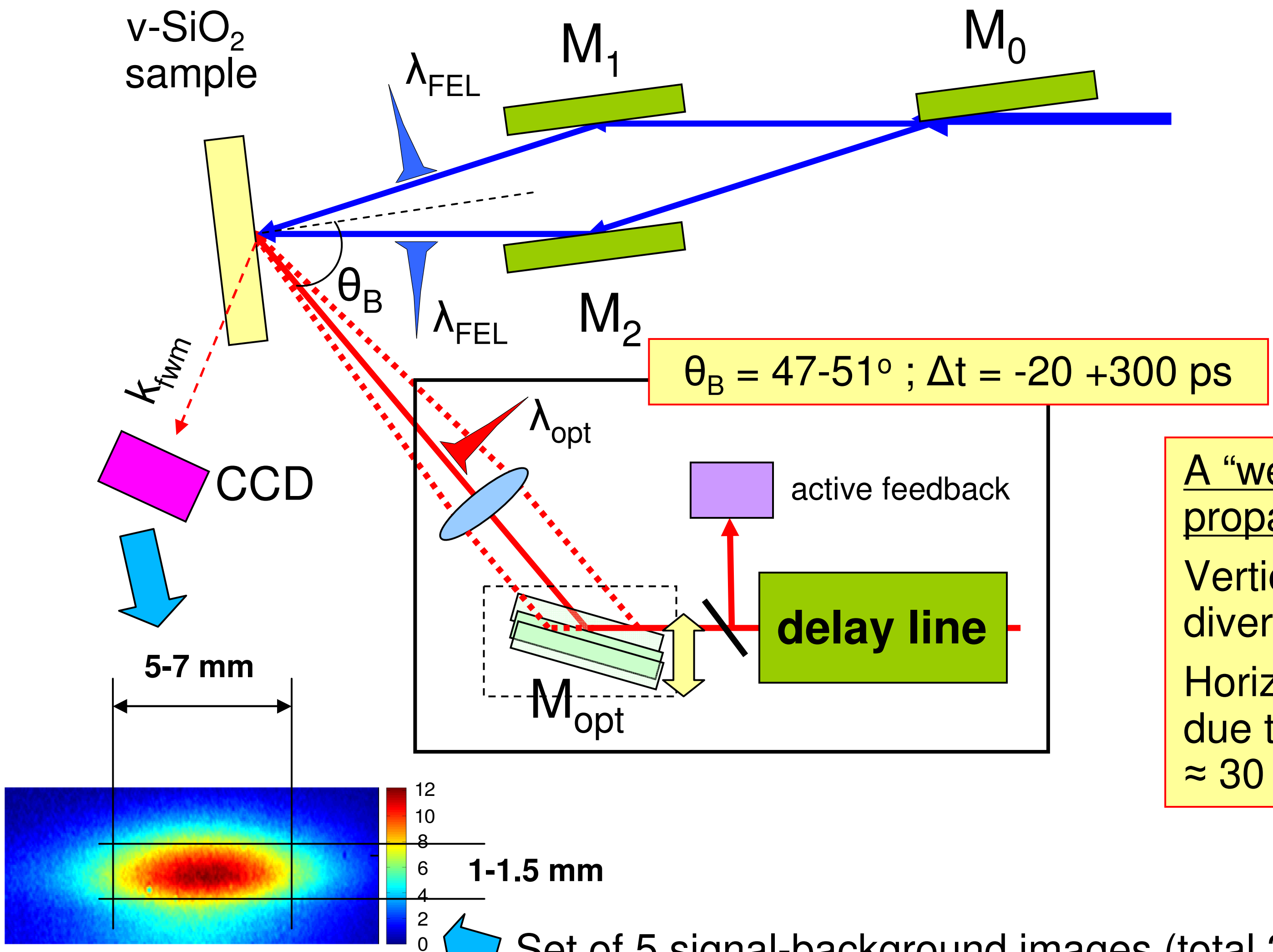
$\lambda_{opt} = 392.8$ nm

$\lambda_{FEL} = 27.6$ nm

Ideal phase matching
($\delta\lambda_{FEL}=\delta\lambda_{opt}=\delta\theta=\delta\theta_B=0$)



FEL-stimulated EUV-FWM signal



Phase matching

$2\theta = 6.16°$
$\theta_B = 49.9°$
λ_{opt} =392.8 nm
λ_{FEL} = 27.6 nm

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} ≈ 30 nm < λ_{opt})

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

F. Bencivenga (unpublished)

FEL-stimulated EUV-FWM processes

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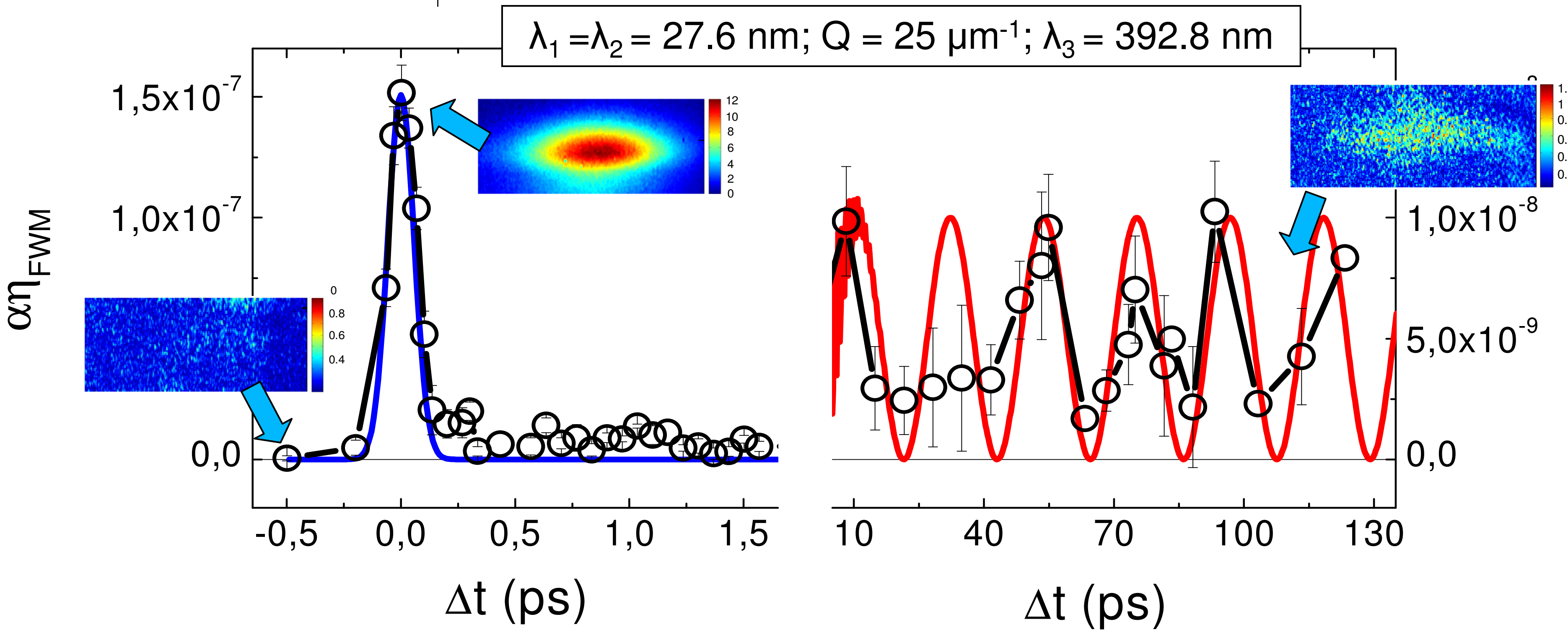
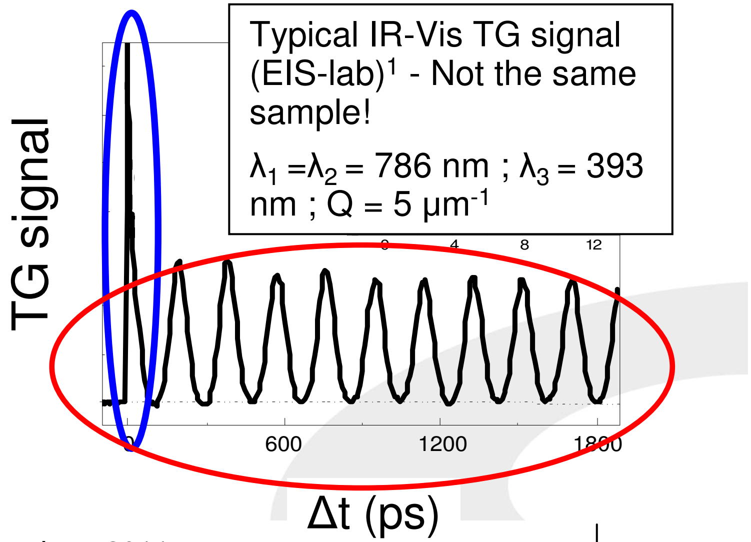


Δt ≈ 0: sharp TG peak ($R_{cc}(\Delta t)$; ≈130-140 fs FWHM, resolution limited) → **electronic response** (coherent spike)

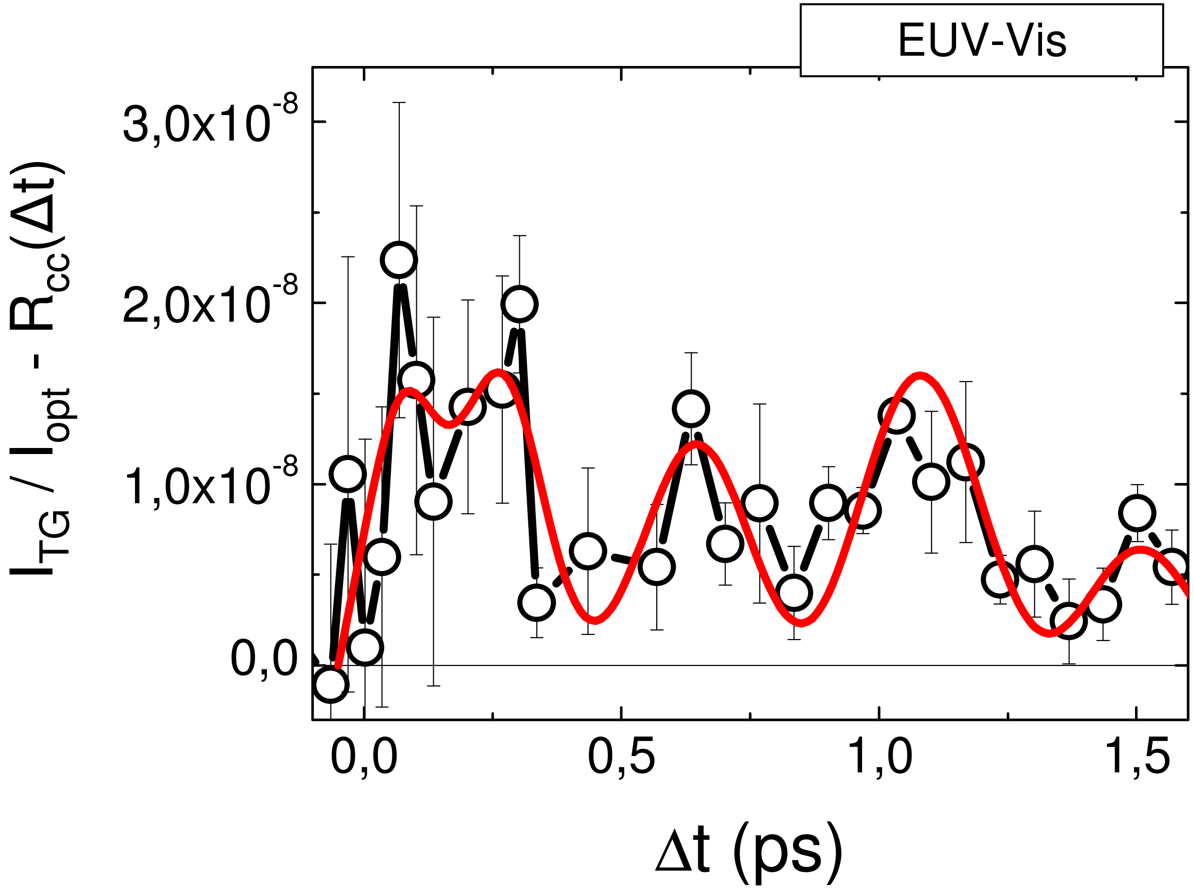
TG signal extends up to Δt ≈ 100 ps → **Longitudinal acoustic mode** at (almost) the expected frequency ($\omega_{LA} = c_s Q \approx 0.145$ THz) and lifetime > 1 ns

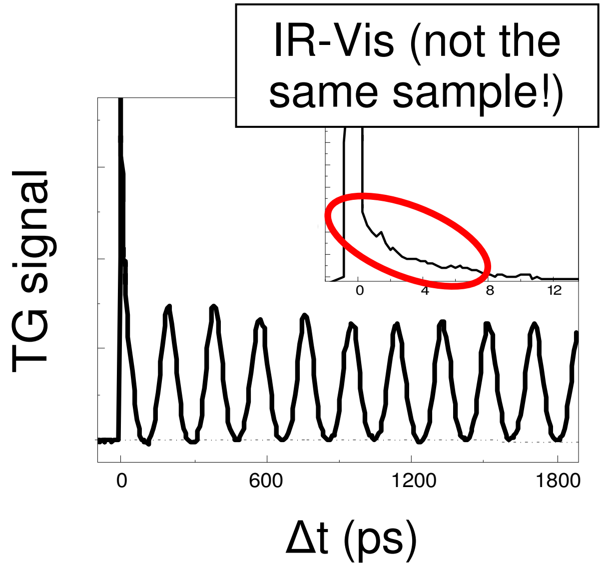
TG efficiency (I_{TG} / I_{opt}) at Δt=0 ≈ 10^{-7} (lower but still comparable to the IR-VIS) and $I_{TG}(\Delta t>0)/I_{TG}(\Delta t=0) \approx 10^{-2}$ (much larger than in the IR-VIS, typical ≈ 10^{-5})

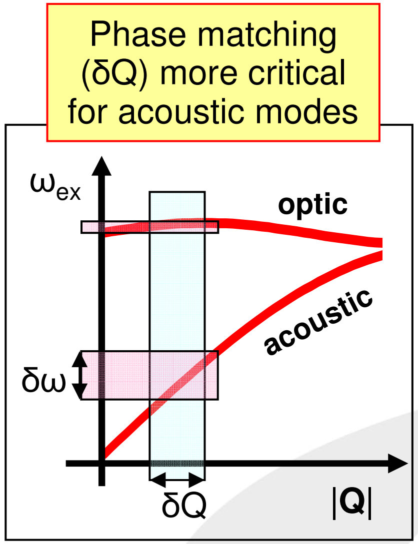


1) R. Cucini et al., Opt. Lett. 2011

FEL-stimulated EUV-FWM processes







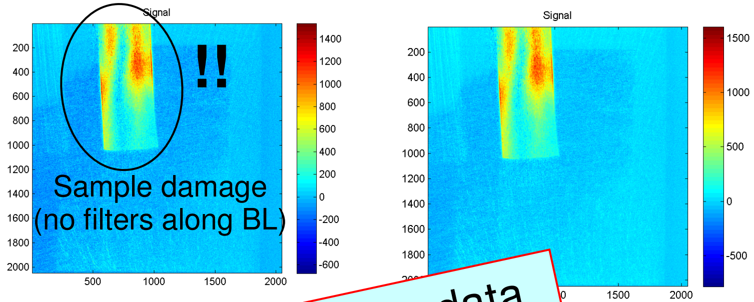
Δt = 0-1.5 ps: **two oscillations** ("optic modes") at $\omega_1 \approx 7.2$ THz (F_1 hyper-Raman mode → tetrahedral rotations) and $\omega_1 \approx 26$ THz (ν_{2b} Raman mode → tetrahedral bendings).

F. Bencivenga (unpublished)

FEL-stimulated EUV-FWM processes

A lucky event: permanent gratings for heterodyne-detection of FWM signals?

A = bkg (no FEL)-bkg*   B = signal-bkg*   (bkg* → no FEL signal taken in another (nearby) portion of the sample)

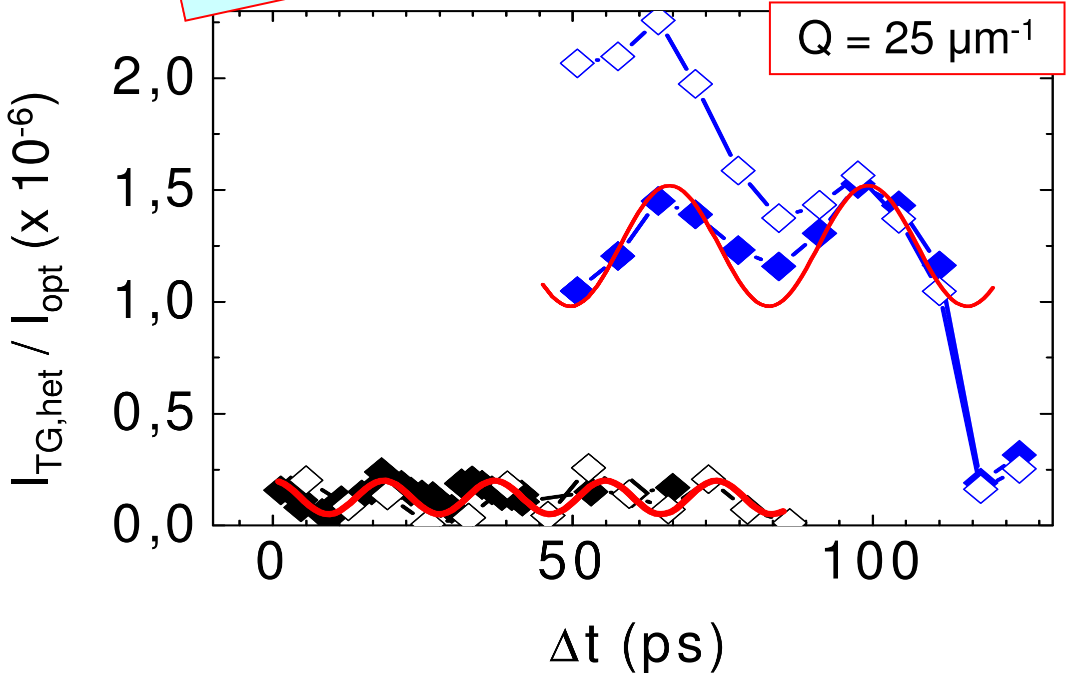


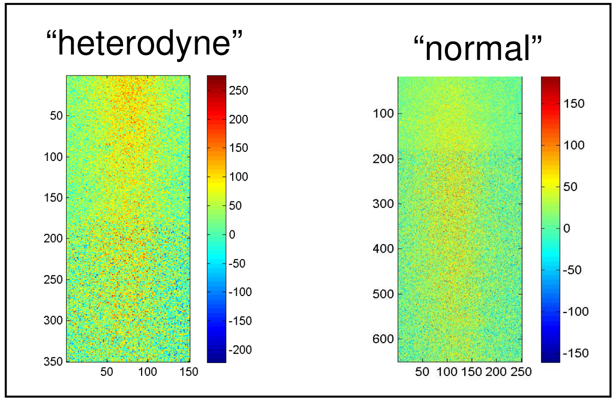
$A = |E_L|^2$ → Δt-independent signal ("local field")

$B = |E_{fwm}(\Delta t)+E_L|^2$

$B - A = |E_{fwm}(\Delta t)|^2+|E_L||E_{fwm}(\Delta t)|\sin(\Delta\varphi)$

Preliminary data

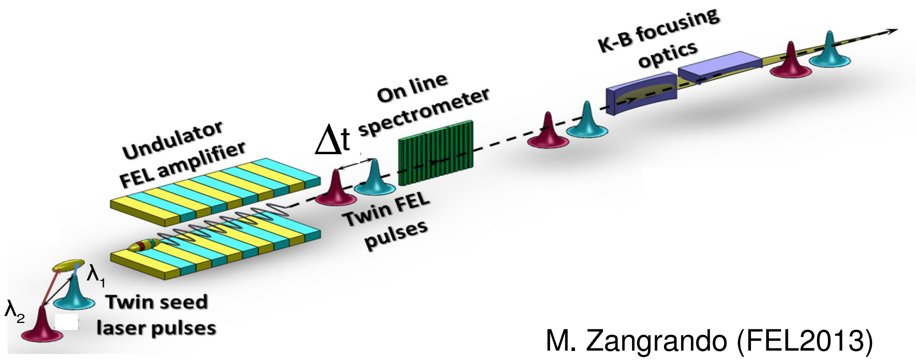




Permanent gratings might be used for "phase-locked" local fields → enhancement of the FWM signal (extraction of both amplitude and phase of E_{fwm}?)

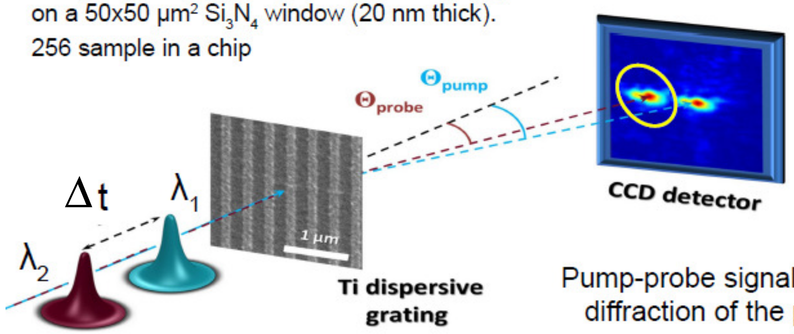
Two-colour FEL-pump/FEL-probe

“twin-seed” two-colour[1]

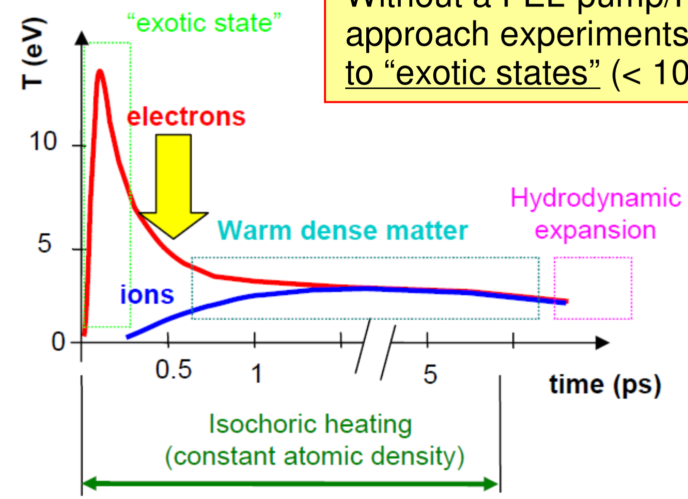


M. Zangrando (FEL2013)

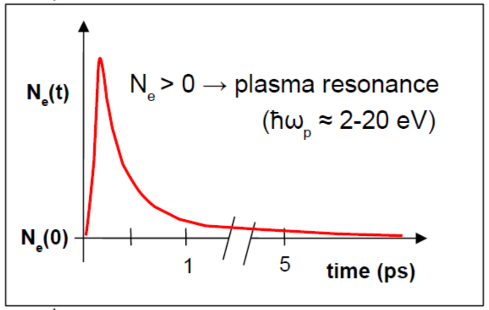
Sample: Ti strips (165 nm wide, 80 nm thick) on a 50x50 μm² Si_3N_4 window (20 nm thick). 256 sample in a chip



Pump-probe signal: Bragg diffraction of the probe

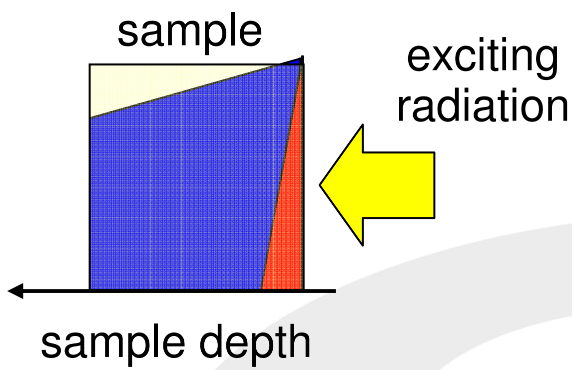


Without a FEL-pump/FEL-probe approach experiments are limited to “exotic states” (< 100 fs)



$\omega < \omega_p$: high reflectivity and limited penetration depth (large **excitation gradient**)

$\omega > \omega_p$: **homogeneous excitation**



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

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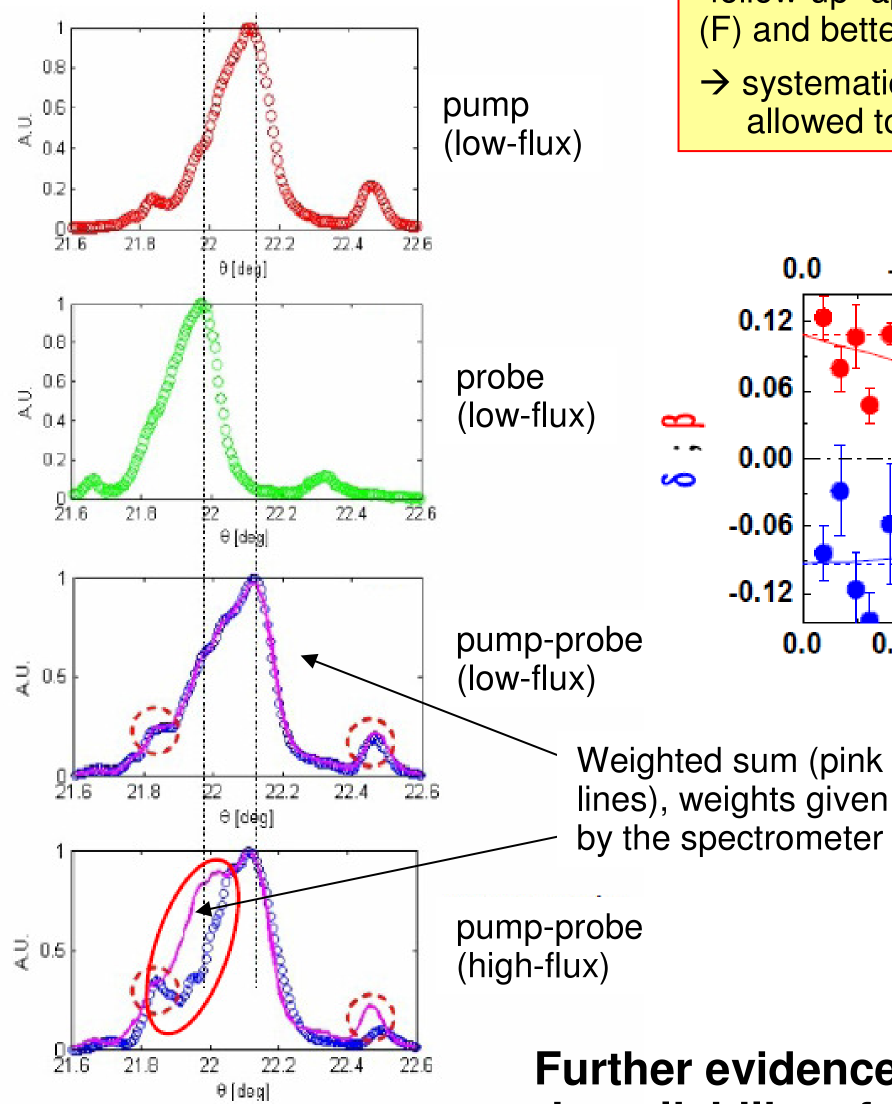
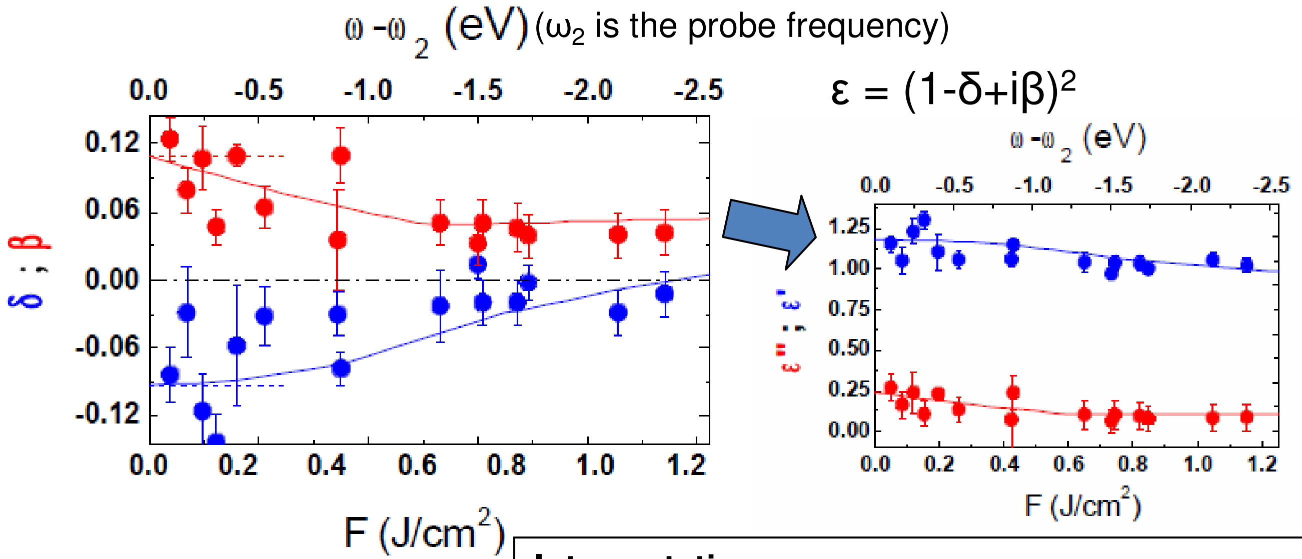
Two-colour FEL-pump/FEL-probe

"follow-up" application (warm-dense Ti)[1]: complete data vs FEL fluence (F) and better samples, still improvable...

→ systematic data analysis, based on Huygens-Fresnel calculations allowed to determine the optical constants (**δ** and **β**)





Interpretation:

XUV absorption at the Al L-edge[2] (no time-resolution: t < 20 fs, i.e. in the "exotic" non-equilibrium state)

→ **β** tends to 0 on increasing F (EUV-transparency), due to massive ionization (depletion of inner shell absorbers) that leads to an ω-shift of the edge.

Further evidence of the reliability of the "twin-seed" mode

The (equilibrium?) state reached after 0.5 ps is featured by FEL induced transparency, as in the non-equilibrium "exotic" state reached in the sub-20 fs timescale

 1) F. Bencivenga et al., Faraday Discus. (2014); 2) B. Nagler at al., Nat. Phys. (2009)

Transient grating + two-colour

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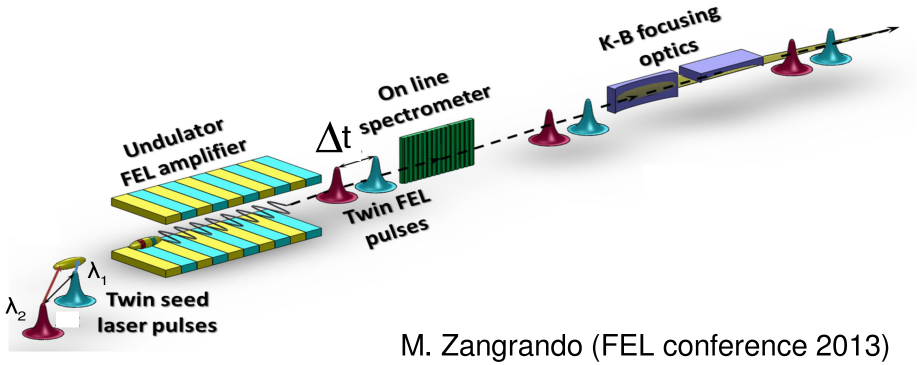
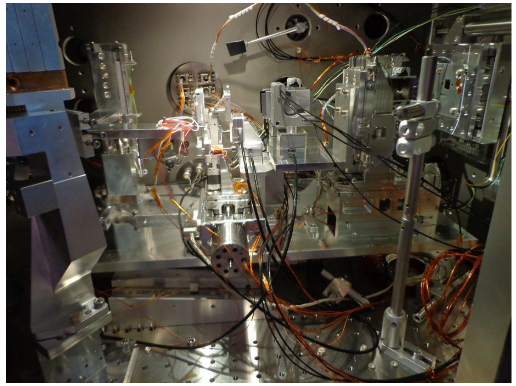
"twin-seed" two-colour

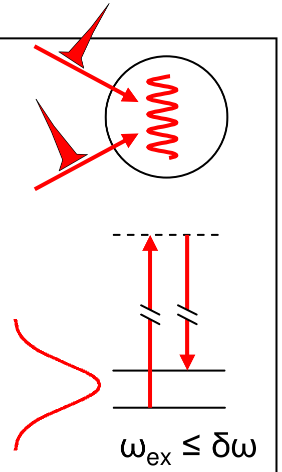


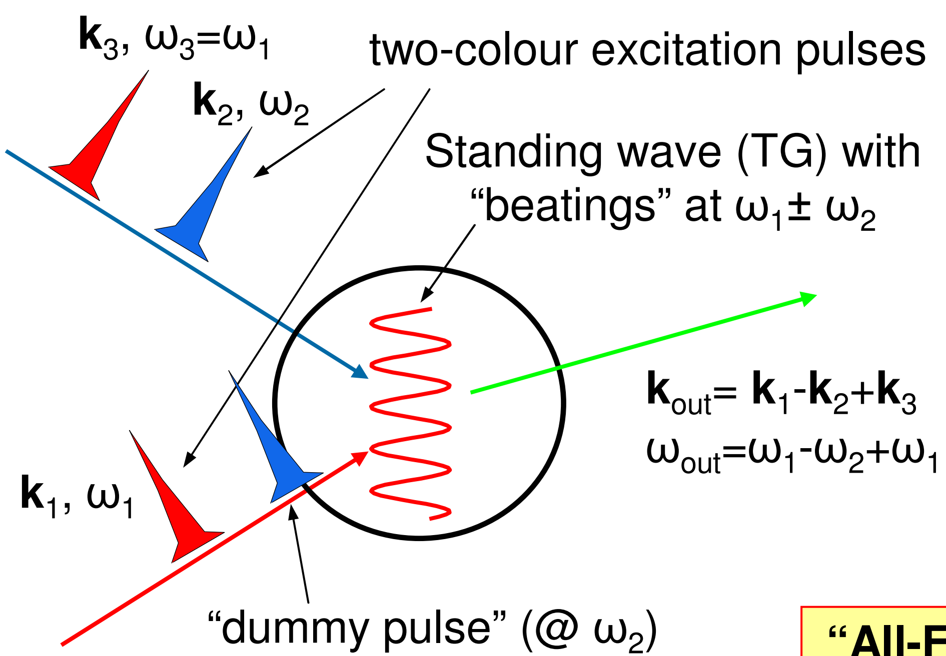
M. Zangrando (FEL conference 2013)

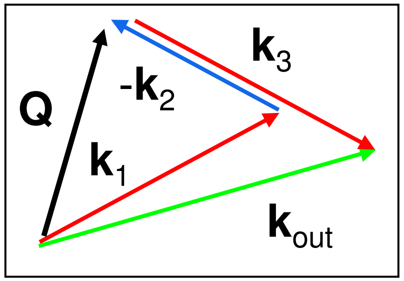
+

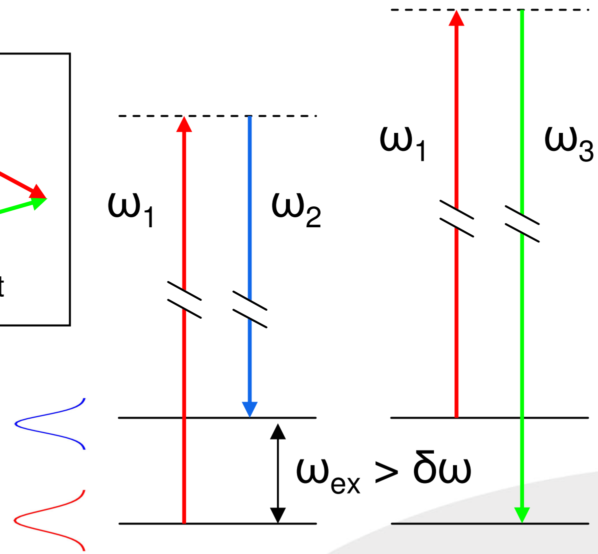
"mini-TIMER"











"All-FEL" coherent (anti-stokes) Raman scattering

F. Bencivenga et al., Faraday Discus. (2014)

Transient grating + two-colour

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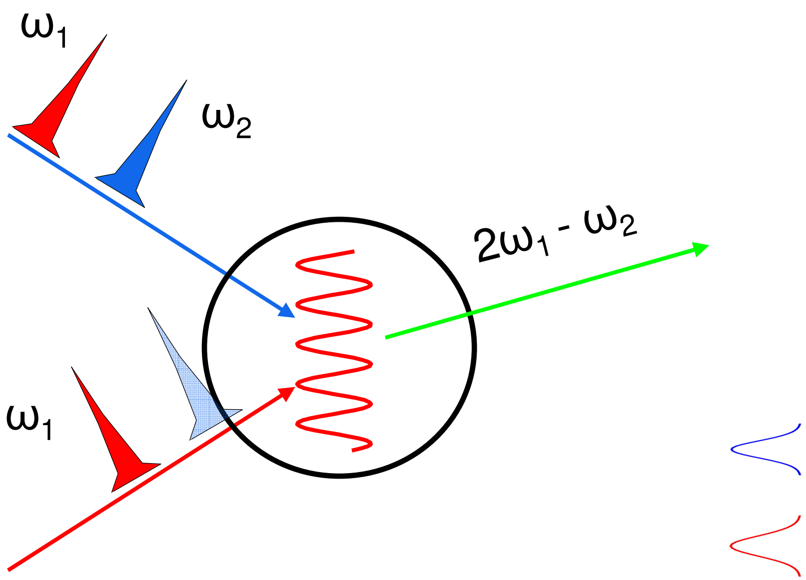
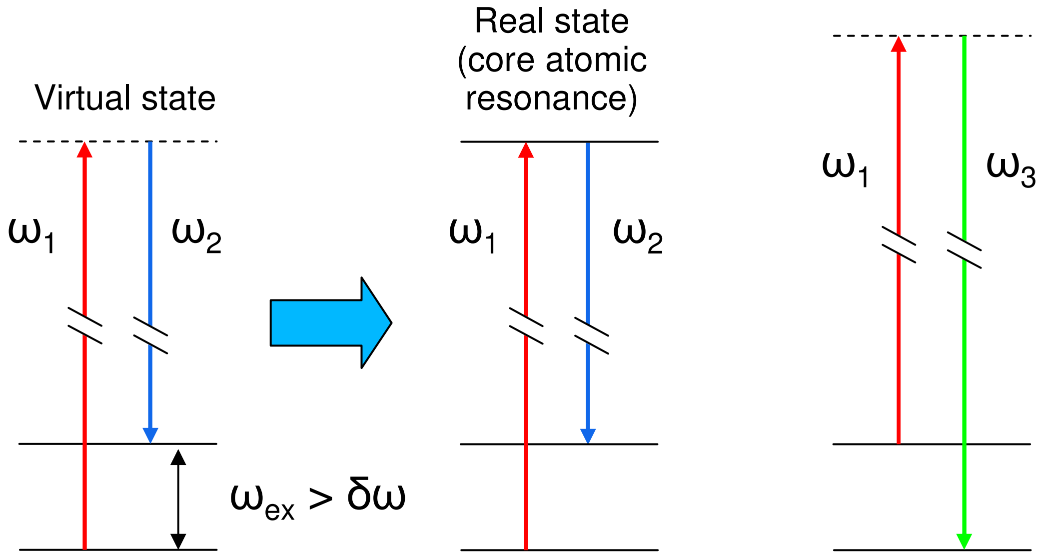
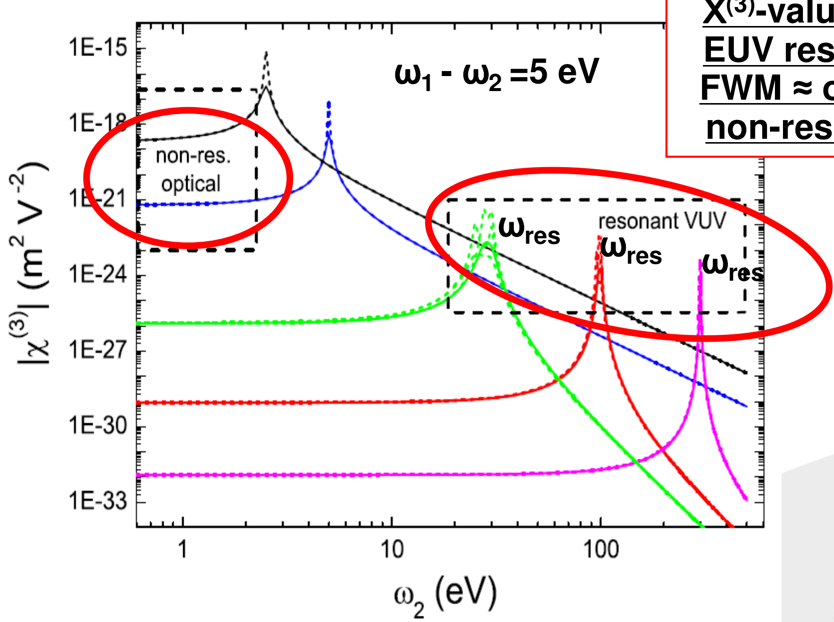




Optical input fields ($\omega_i \sim$ eV)
→ $\omega_{ex} < 0.1$'s eV (vibrations)

EUV/x-ray fields ($\omega_i > 100$'s eV)
→ **$\omega_{ex} \sim$ 1-10 eV's** (excitons)

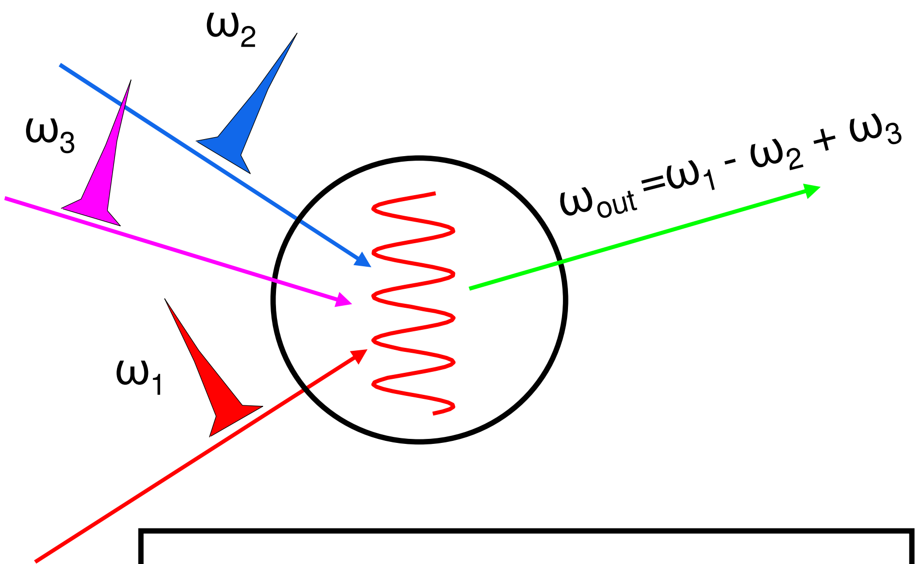
Atomic seletivity through core resonances ($\omega_1=\omega_{res}$)

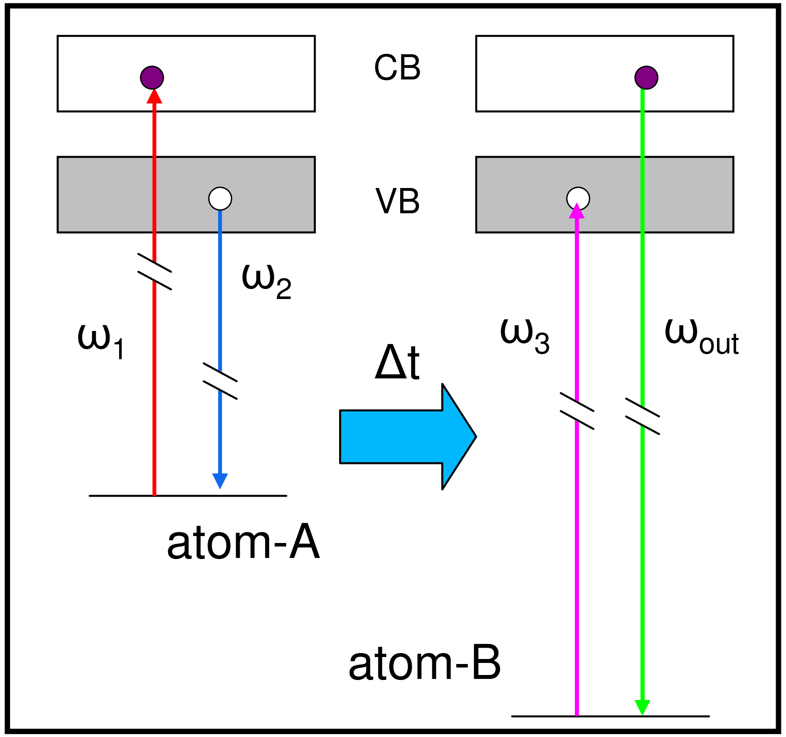


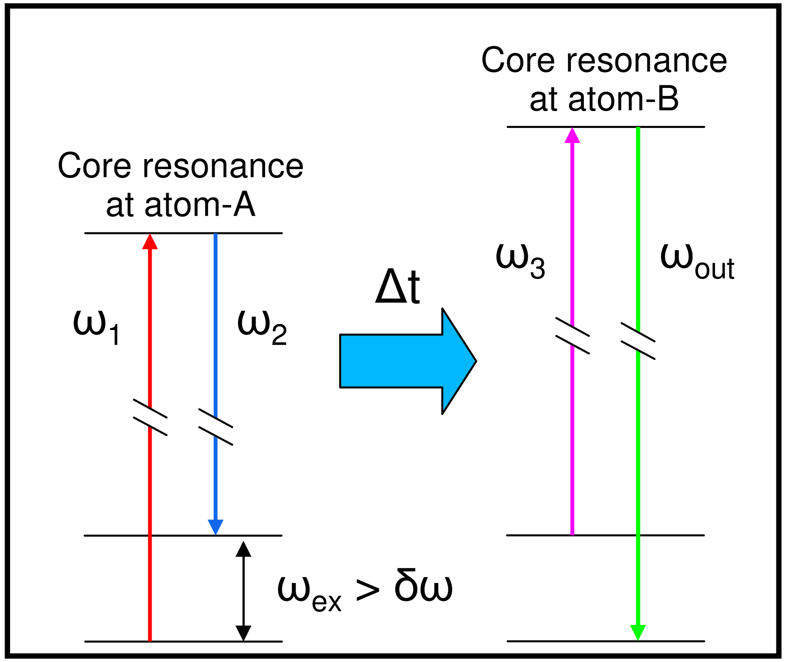
$X^{(3)}$-values for EUV resonant FWM ≈ optical non-resonant

F. Bencivenga et al., Faraday Discus. (2014)

Transient grating + three-colour



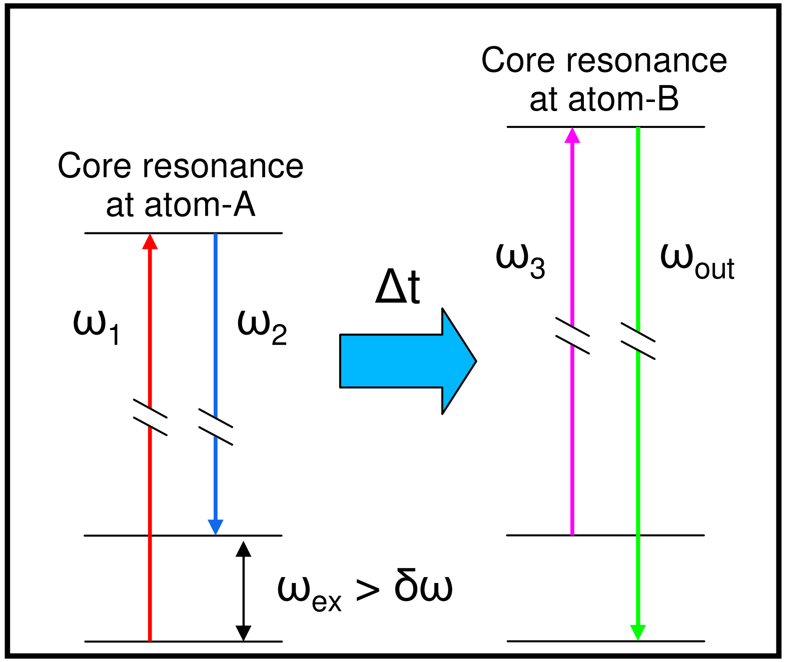


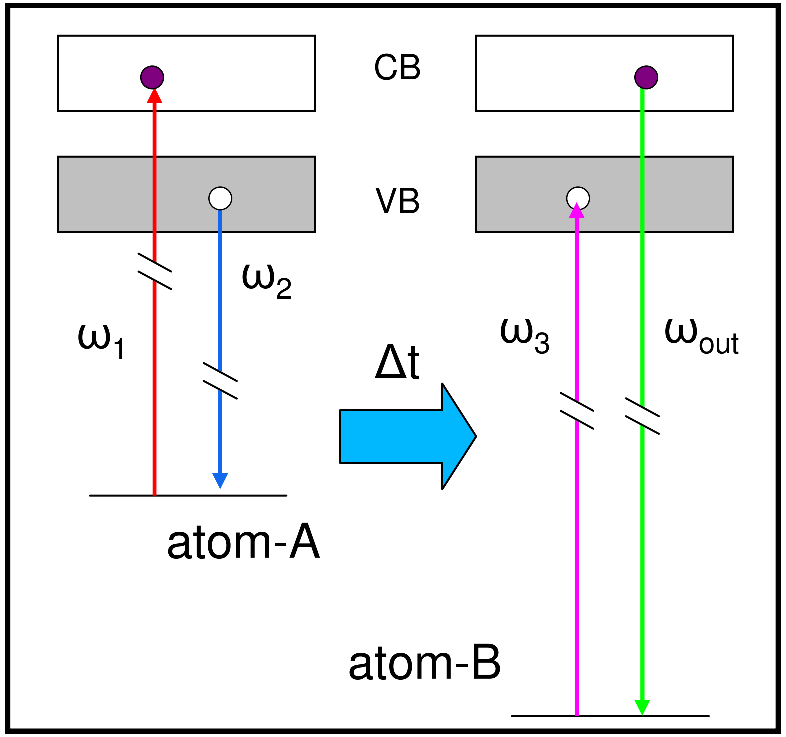


S. Tanaka and S. Mukamel, PRL (2002)

Transient grating + three-colour

FWM measures the **coherence between two atoms**: 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/energy transfer processes, non-local nature of valence band excitations, etc.





If λ_i's compare to the molecular size, then **dipole approximation does not apply** → possible to probe the entire manifold of electronic transitions without dipole selection rules

S. Tanaka and S. Mukamel, PRL (2002)

Transient grating + two-colour

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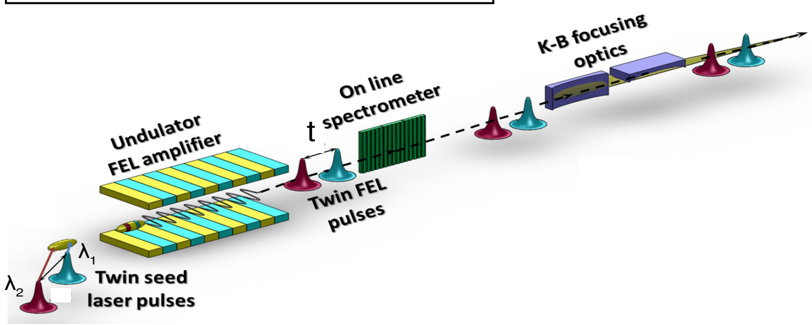
"twin-seed" two-colour



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
SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA
(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 →

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

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

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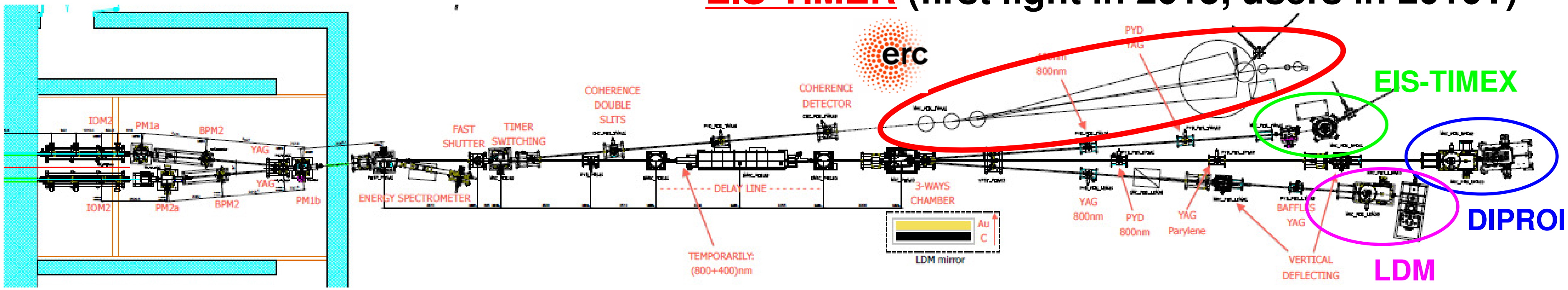
[2] *SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*
[3] *Argonne National Laboratory, Argonne, Illinois 60439, USA*
(Received 24 March 2014; published 10 July 2014)

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.

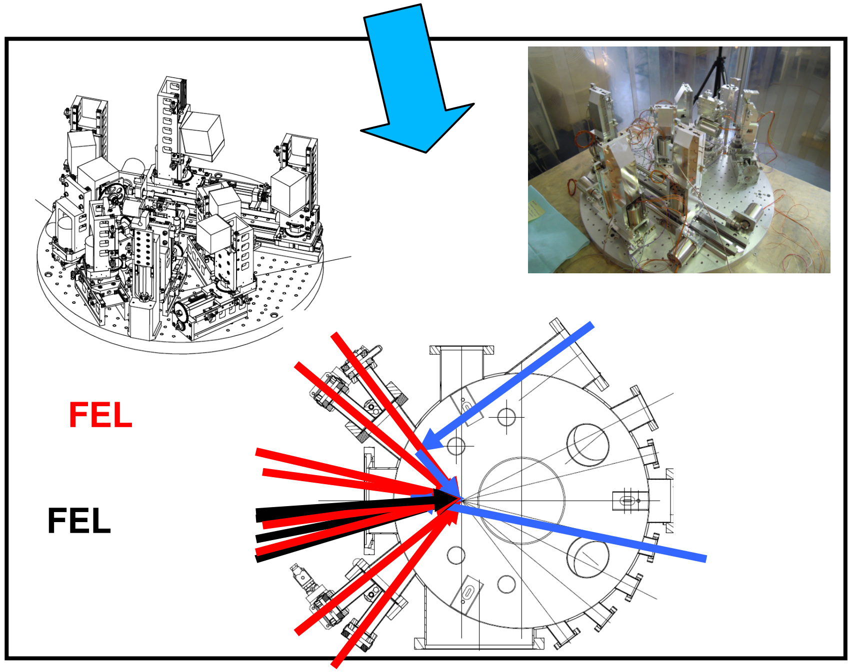
On the experimental side a lot can be done with the present state of the art, however, the full exploitation of FWM needs more flexible multi-colour (and coherent) FEL's

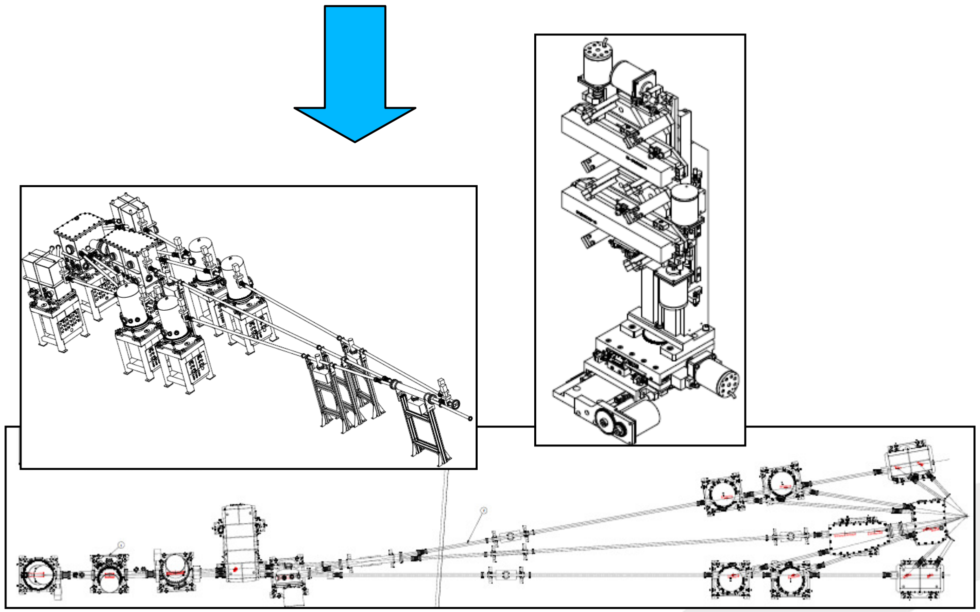
Outlook (EIS-TIMER beamline)

EIS-TIMER (first light in 2015, users in 2016?)



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





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Conclusions

- Experimental-end station (EIS-TIMER) for EUV/soft x-ray non-linear, wave-mixing experiments will be available at FERMI in 2015 (original goal is to study vibrational modes in the 0.1-1 nm^{-1} Q-range in disordered systems and nanostructures)

- First experimental evidence 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 electron / nuclear TG signal in the EUV range is larger than in the optical one
  → Observed three oscillating features, ascribable to vibrational modes (phonons)
  → Possible to use permanent gratings for heterodyning
  → My feeling is that the key role is played by coherence / phase-matching

- The possibility to exploit a multi-colour seeded FEL source and an experimental setup for transient grating experiments ("mini-TIMER"@DiProI or EIS-TIMER) would allow to develop at FERMI advanced four-wave-mixing methods, as coherent Raman scattering, in the next future

Elettra Sincrotrone Trieste

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