Non-linear optics at short wavelengths

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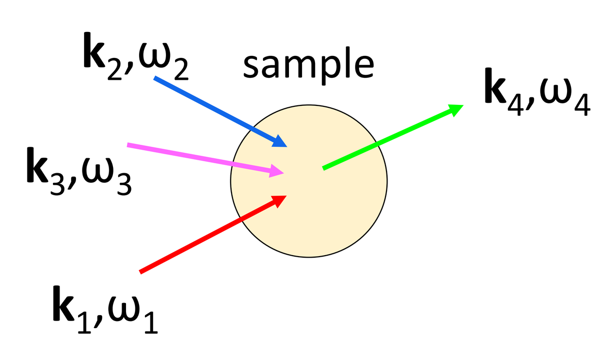
Four-wave-mixing: three light fields (E_i) with frequency ω_i and wavevector $\mathbf{k}_i$ (i=1,2,3) interact with the sample giving rise to a fourth field $E_4(\omega_4,\mathbf{k}_4)$, through 3rd order non-linear process (third harmonic generation, coherent Raman scattering, etc.)

Linear light-matter interactions (diffraction, absorption, emission, inelastic scattering, etc.)

$\chi^{(2n)}=0$ (in samples with inversion symmetry) → first evidence of x-ray wave-mixing[1]

$$E_4 \sim \Sigma_{p(i,j,k,\dots)}\, \chi E_i + \chi^{(2)} E_i E_j + \chi^{(3)} E_i E_j E_k + \dots$$

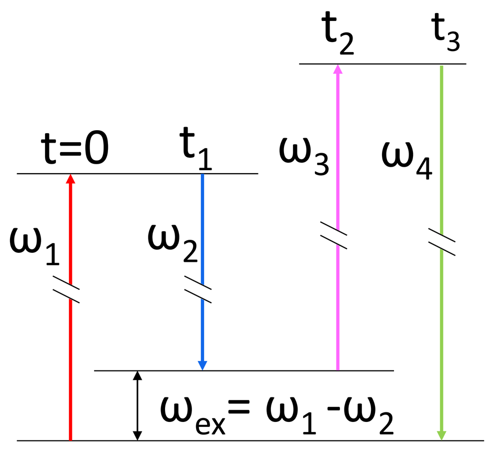
Four-wave-mixing (FWM)

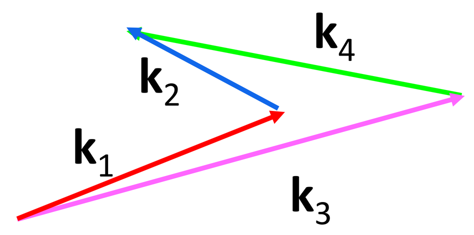


"phase matching"

$\mathbf{k}_4 = \mathbf{k}_3 - \mathbf{k}_2 + \mathbf{k}_1$

$\omega_4 = \omega_3 - \omega_2 + \omega_1$





1) Glover et al., Nature 2012; Shwartz et al., PRL 2014

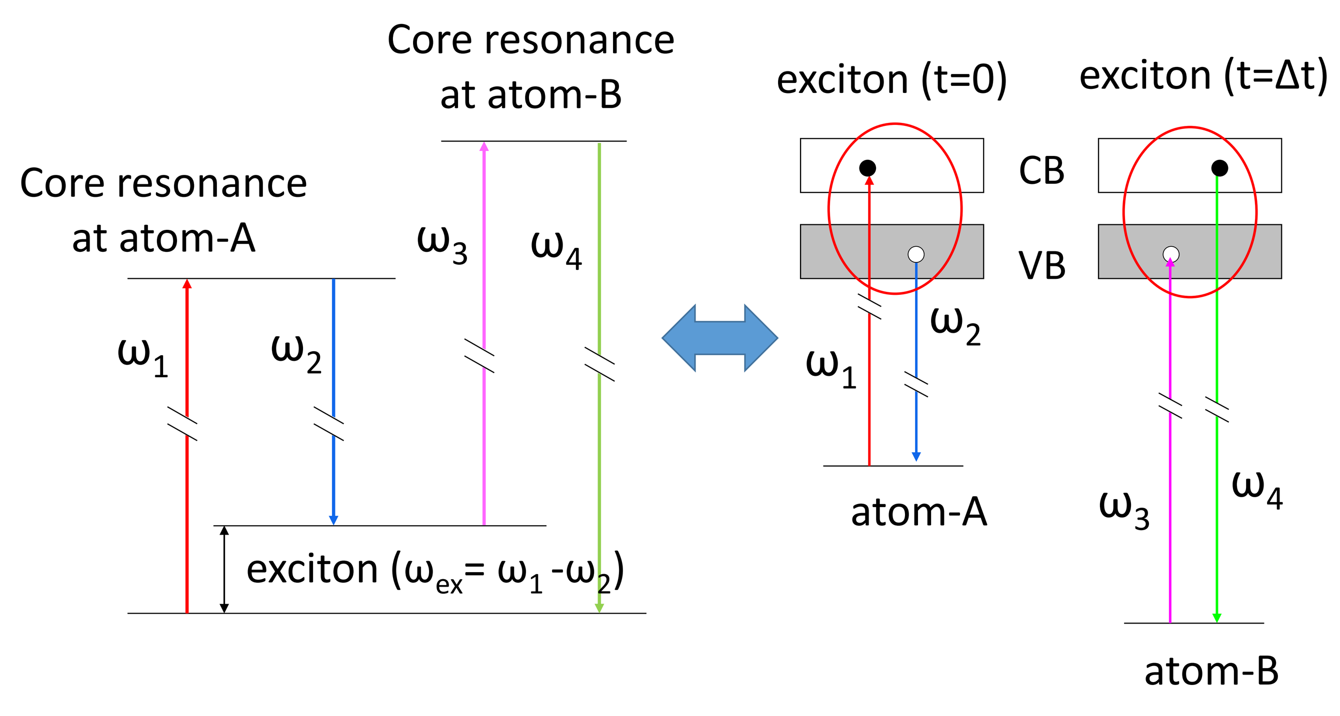
XUV/soft r-ray FWM[2]

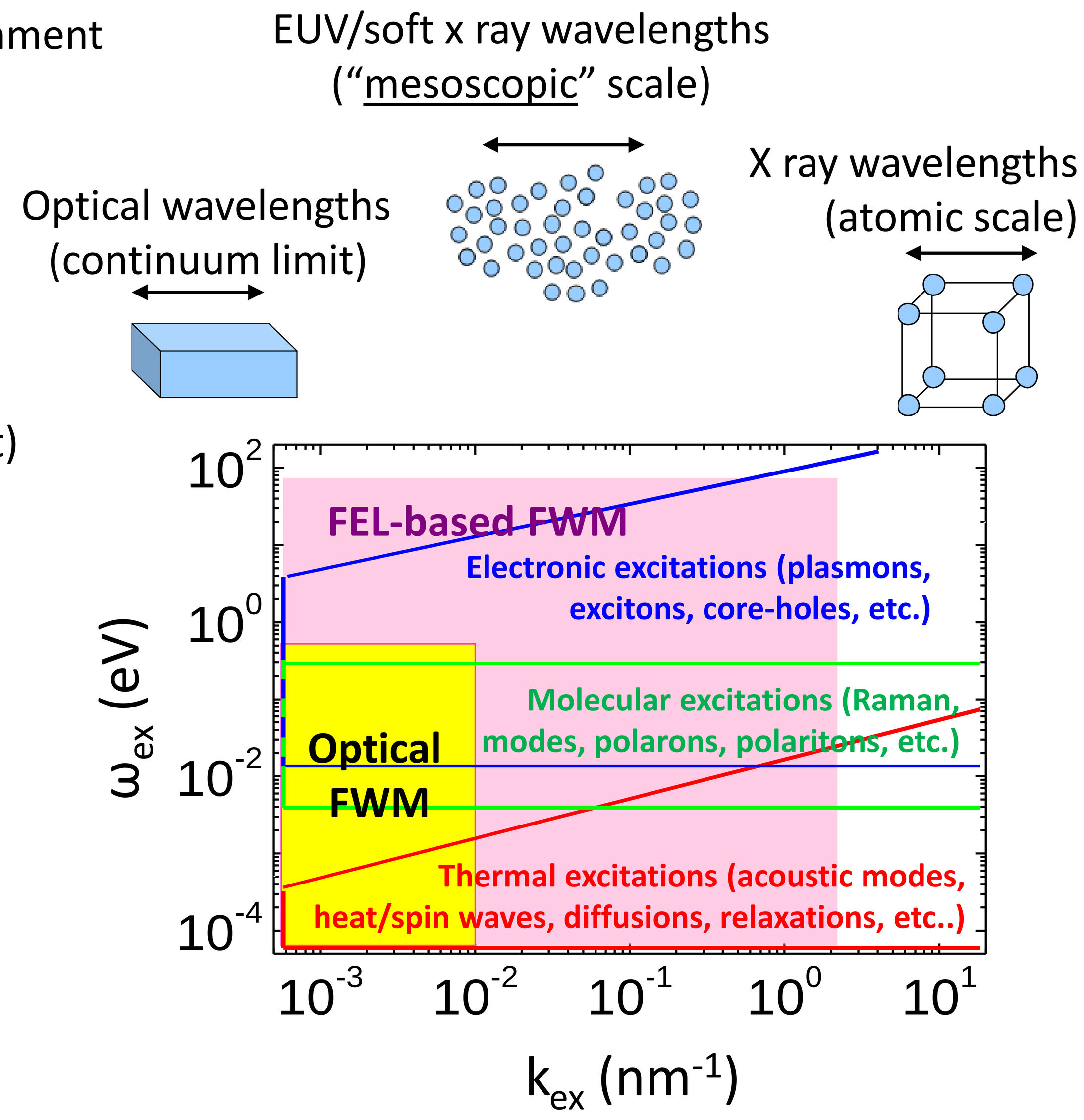
Larger $\mathbf{k}_{ex}$ and ω_{ex} ranges (up to several nm^{-1} and eV) with respect to optical FWM → high energy excitations, e.g. excitons, and unexplored "mesoscopic" k_{ex} range (0.1-1 nm^{-1})

Exploitation of core transitions → correlations between core resonances, localized on given and selectable atoms, and selected excitations (phonons, excitons, etc.)

Relaxed dipole selection rules and sensitivity to local structures/environment

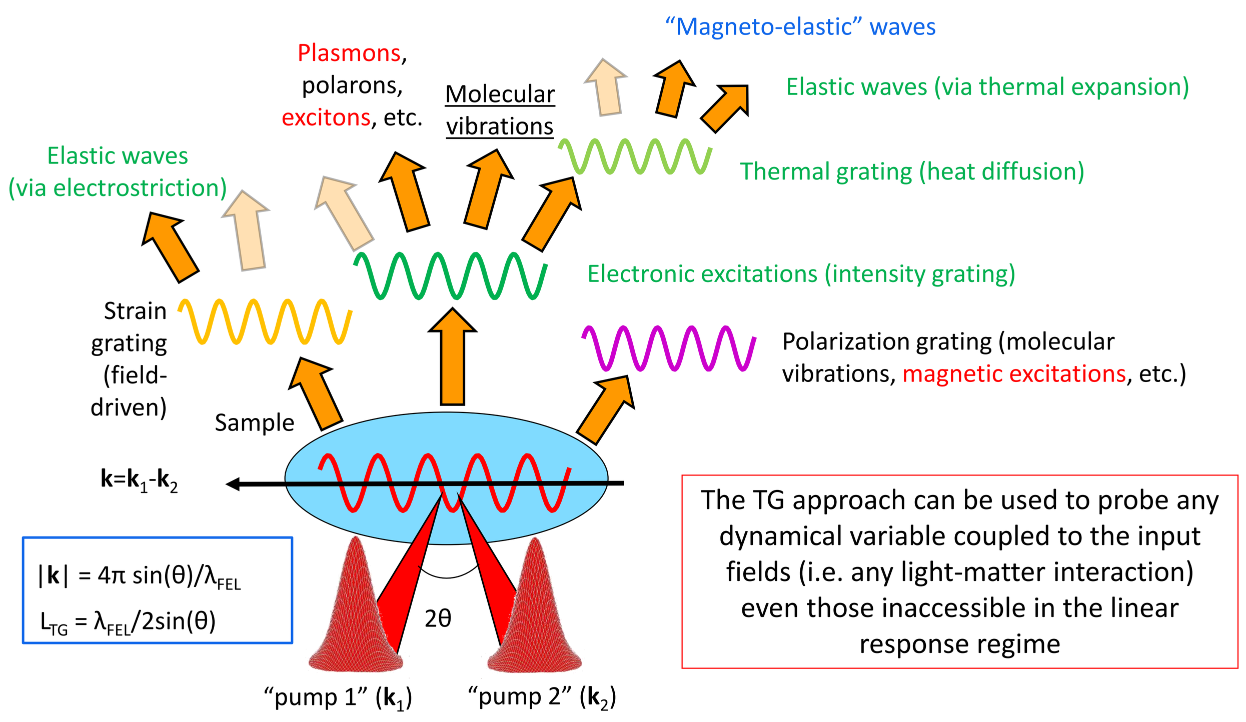
With respect to linear x-ray methods, where light-matter interaction take place in a single atomic site, the multi-wave nature of FWM may allow to monitor in real-time the dynamics between selected atoms



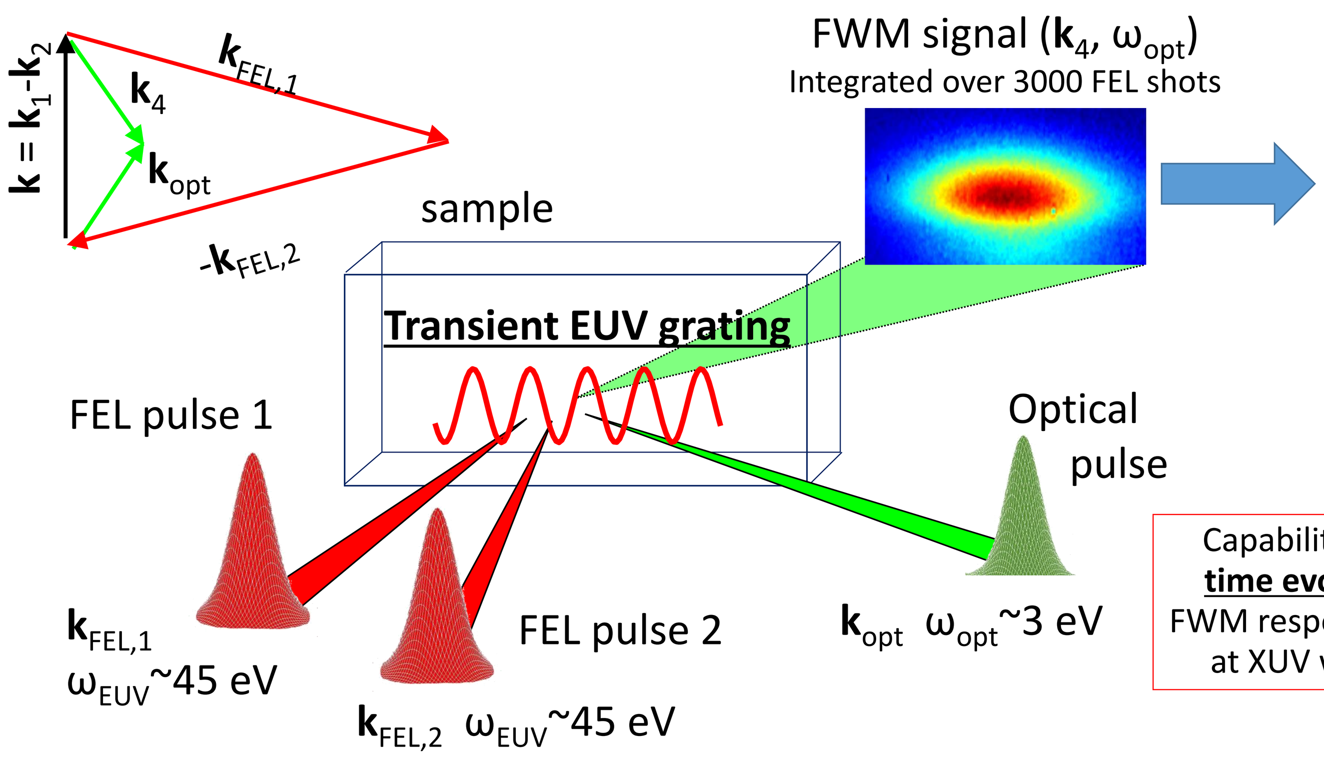


2) Tanaka and Mukamel, Phys Rev Lett 2002

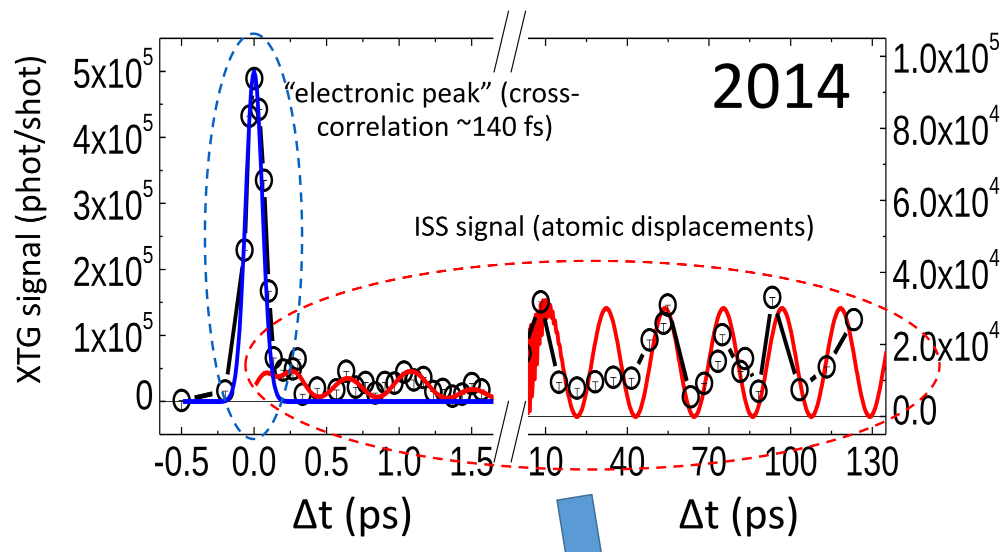
mini-TIMER (@DiProI): stimulated scattering by XUV transient gratings (X-TG)[3]

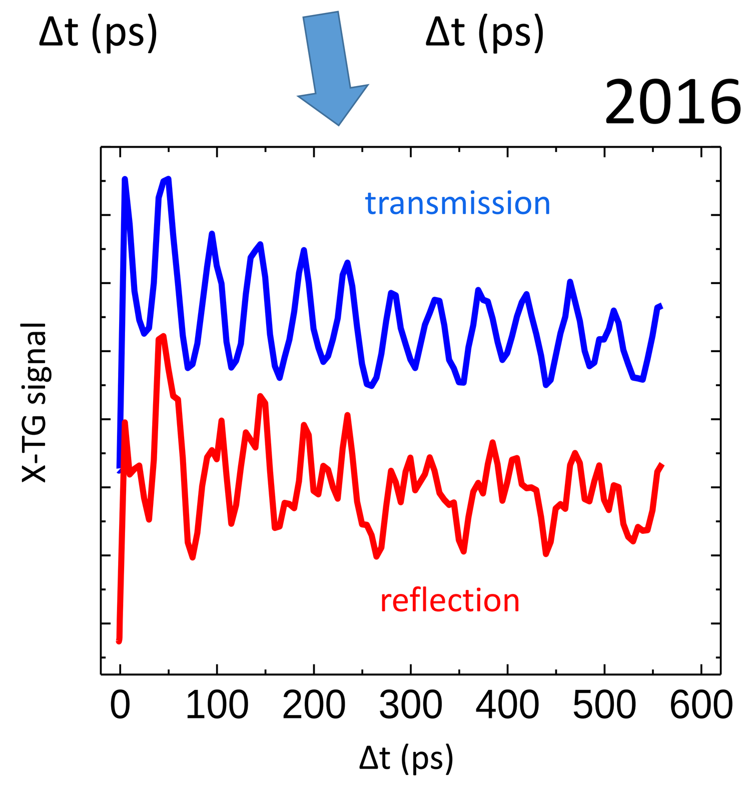


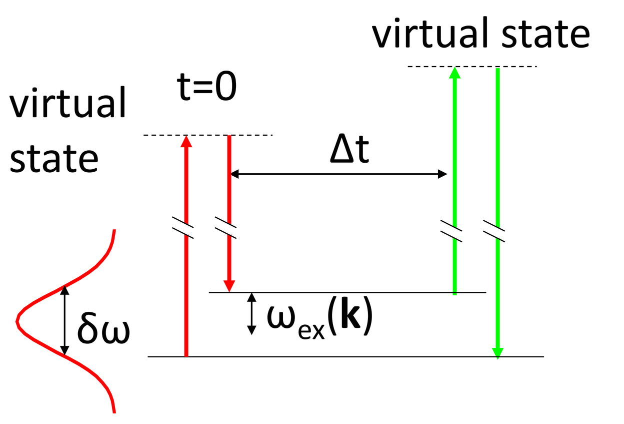
The TG approach can be used to probe any dynamical variable coupled to the input fields (i.e. any light-matter interaction) even those inaccessible in the linear response regime

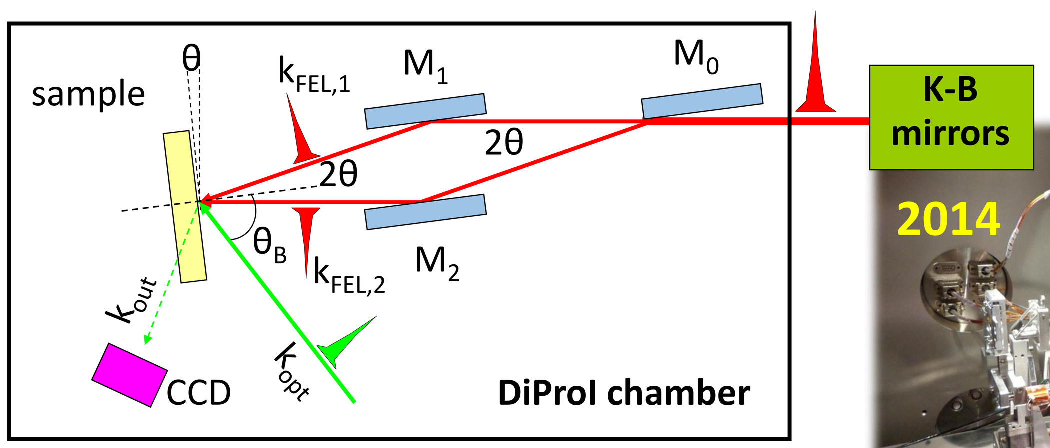


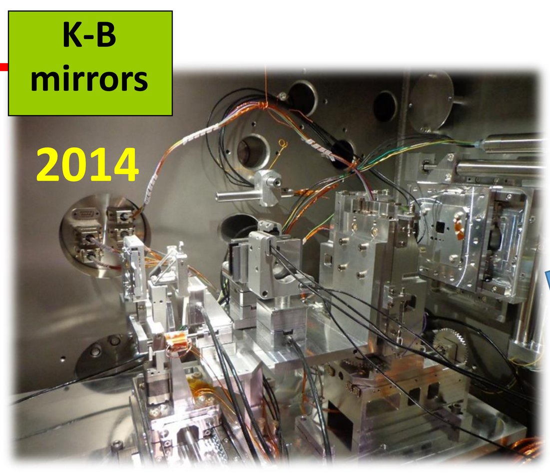
Capability to read the time evolution of the FWM response stimulated at XUV wavelengths[3]

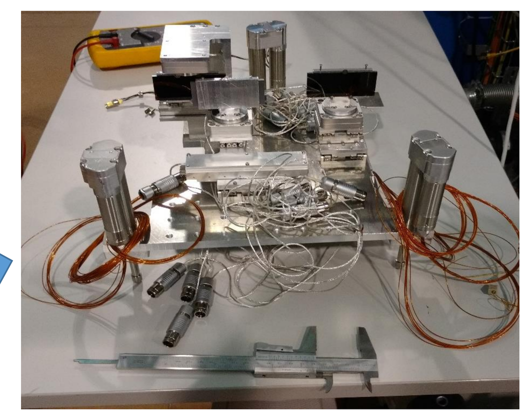












$\omega_{ex} < \delta\omega$ → low energy (slow) excitations, e.g. Brillouin, Raman and Rayleigh modes

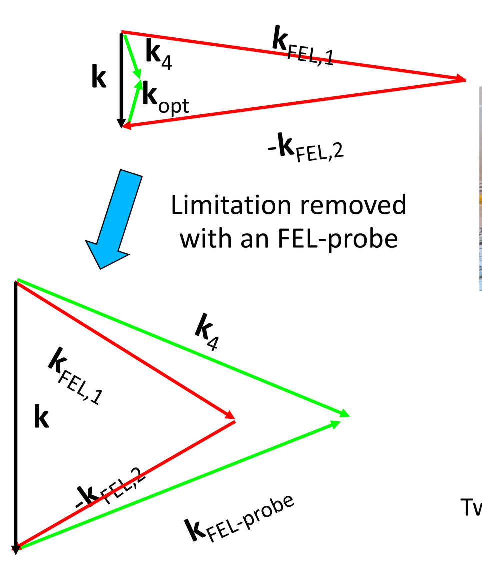
3) Bencivenga et al., Nature 2015

4) Collaboration with K. Nelson (MIT) and G. Knopp (PSI)

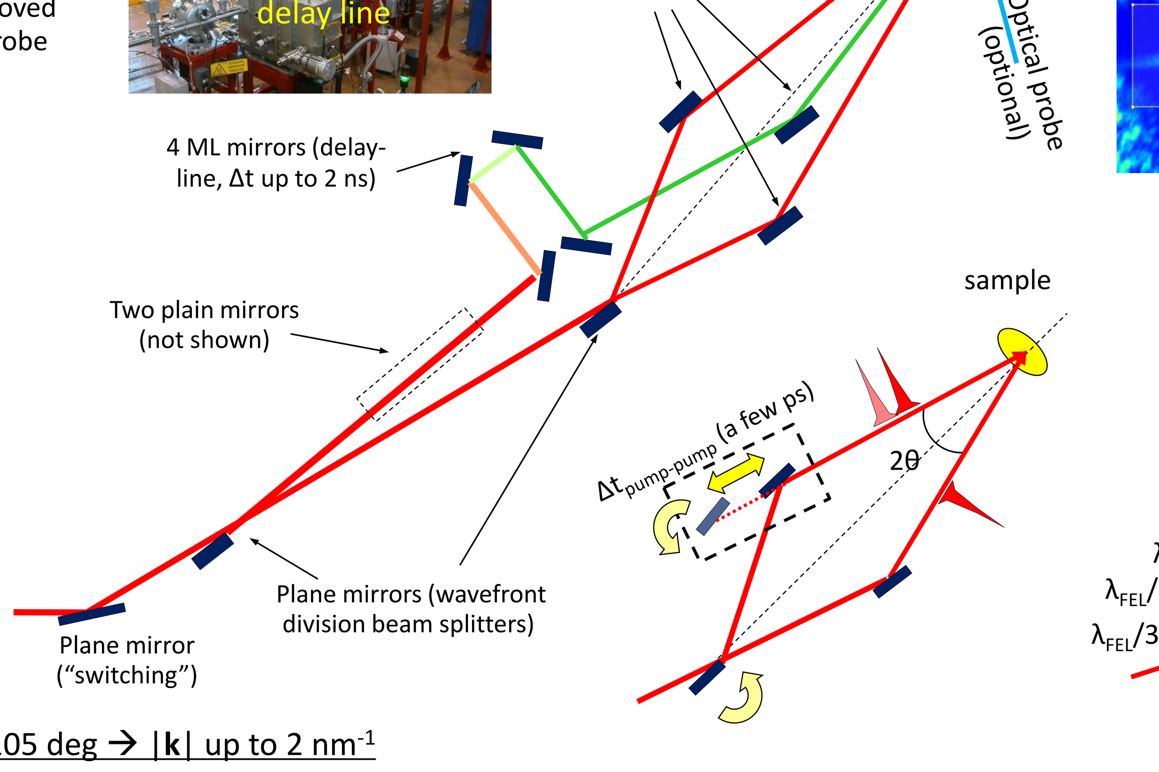
With mini-Timer v2 we greatly improved the data quality and exploitation of reflection geometry (opaque samples & surface sensitivity)[4]

EIS-TIMER[5] (all-FEL-based EUV/soft x-ray FWM)

Optical probe: $k_{ex} < 2k_{opt}$



5 plane mirrors (grazing)
4 multilayers (45° AOI)
3 focusing mirrors
to control:
3 beams
2 angles
2 delays



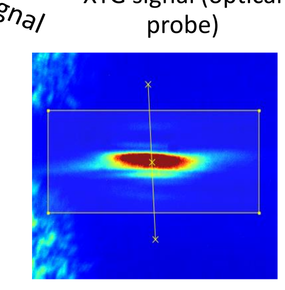
Crossing angles up to 105 deg → $|\mathbf{k}|$ up to 2 nm^{-1}

5) Bencivenga et al., J. Synchr. Rad. 2016

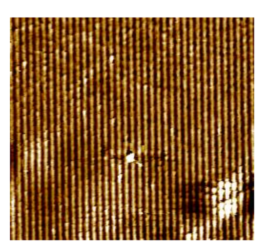
The main task for the next future: find the XTG signal from the FEL probe!

???

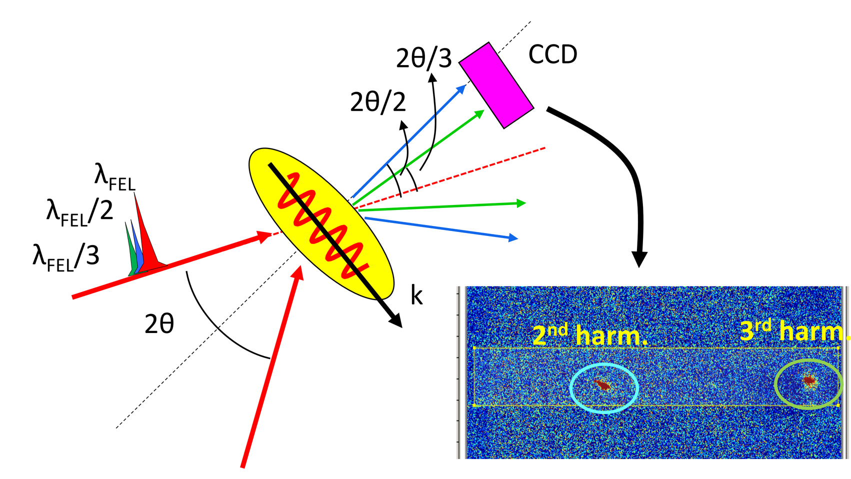
XTG signal (optical probe)



AFM topography (evidence of controlled FEL-FEL interference)

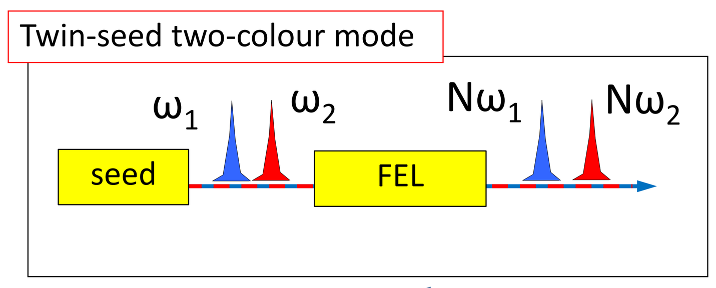


First **"all-XUV" XTG signal**: from 2nd and 3rd harmonics of the FEL, collinear with the pump beams → limited to Δt=0 (but possible to scan the photon frequencies and the phase!)



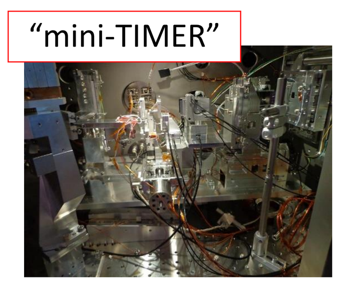
"color mini-TIMER" (two-colors XUV-FWM)[6]

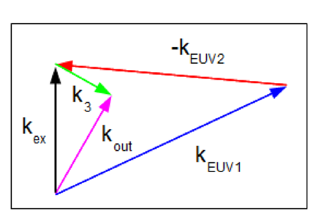
Twin-seed two-colour mode

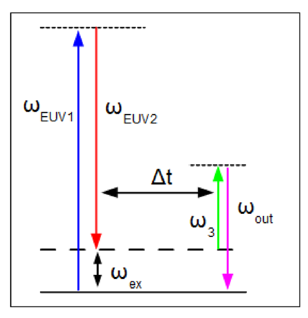


+

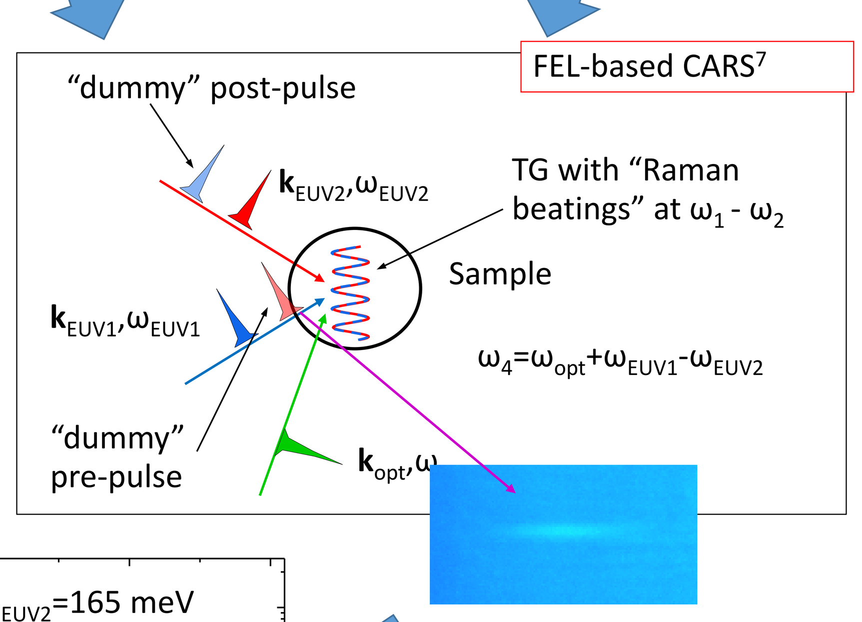
"mini-TIMER"

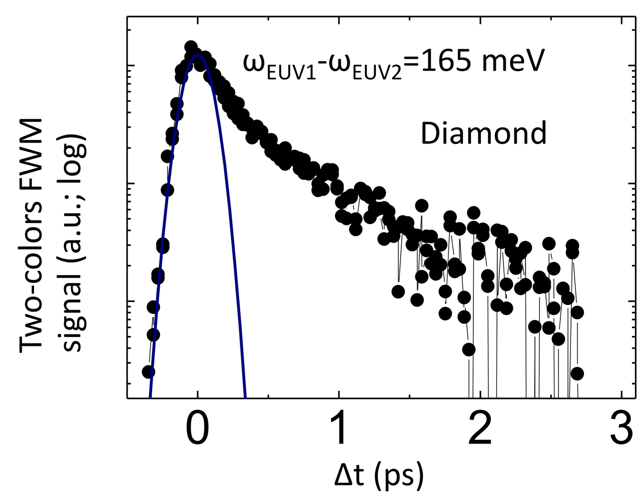






FEL-based CARS[7]

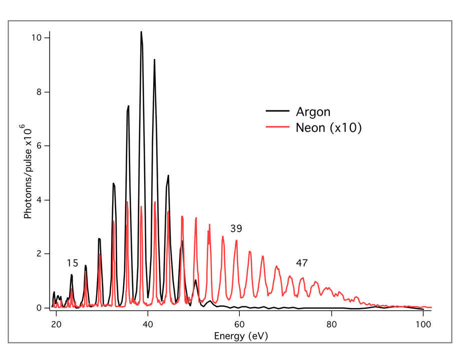




First evidence of a **two-colors XUV-FWM signal (XCRS)**!

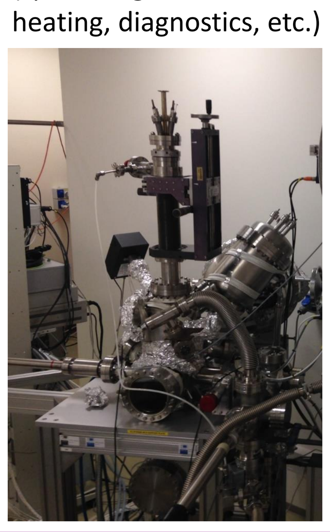
6) Bencivenga et al., in preparation; Faraday Discuss. 2015 (proposal)

FERMI experiments @CITIUS[7] (ankylography, magnetic TG, ultrafast molcular dynamics)

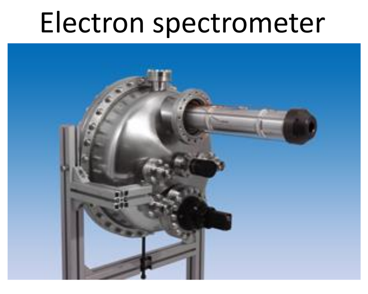


An ultrafast (10's fs) HHG source, to "simulate" FERMI, plus a suite of laser pumps, detectors etc. to prepare in advance some selected FERMI experiment and carry out complementary studies

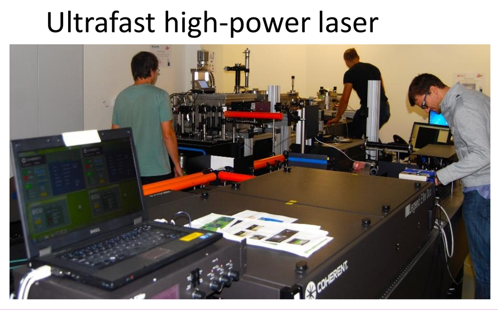
Sample preparation chamber (sputtering, electron beam heating, diagnostics, etc.)



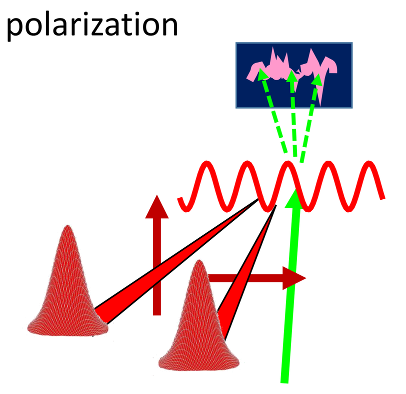
Electron spectrometer



Ultrafast high-power laser



Optical TG with crossed polarization



Plus resonant XUV scattering (Ni, Fe, Co edge) to probe spin grating induced by the TG

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Outlooks:

1) Exploitation of core-resonances (to date we did only non-resonant X-FWM experiments)
2) Implementation of FEL probing (EIS-TIMER)
3) Exploitation of the "double harmonic mode" ($\omega_{EUV1} - \omega_{EUV2} \approx$ eV's)
4) Magnetic materials (also tests @CITIUS)
5) Start to play with the "pump-pump" delay and signal spectrum