Wave-mixing experiments with multi-colour seeded FEL pulses

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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
TIMER: aim of the project

UNSOLVED PROBLEMS IN PHYSICS

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 instabilities** (phonon softening)
- **Electron-phonon coupling**
- **Thermodynamics** ($c_v, \lambda, \Theta_D, S_D$, 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**
Transient grating method

|Q| = 4π sin(θ)/λ

θ_B = sin^{-1}(θλ_2/λ)
EUV/x-ray transient grating

\[ |Q| = \frac{4\pi \sin(\theta)}{\lambda} \]

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

\[ Q = 0.03 - 1 \text{ nm}^{-1} (\sim 3 \text{ nm}^{-1}) \]

\[ L^* \sim \frac{2\pi}{Q^*} \]

- disordered systems \((L^* \sim 10 \text{ nm})\)
- nanostructures \((L^* \sim 1-100 \text{ nm})\)

1) F. Bencivenga and C. Masciovecchio, NIMA (2008)
Non-linear (wave-mixing) signal

\[ E_{\text{out}} \sim \sum p(i,j,k,\ldots) \left[ \chi E_i \chi^{(2)} E_j \chi^{(3)} E_k + \ldots \right] \]

Driving forces in the wave equation: non-linear emission at \( \omega_{\text{out}} = \sum p(i,j,k,\ldots) \pm \omega_i \), not necessarily equal to any \( \omega_i \). For TG: \( \omega_{\text{out}} = \omega_1 - \omega_1 + \omega_3 = \omega_3 \)

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

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

\[ \chi^{(3)} E_j^2 / \chi \ll 10^{-4} \]
\[ \Rightarrow I_{\text{fwm}} / I_{\text{lin}} \ll 10^{-8} \]

\[ \chi^{(n)} \text{ decreases on increasing } \omega_i \text{ [2]} \]

1) T. E. Glover et al., Nature (2012)
2) B. D. Patterson, SLAC-TN (2011); F. Bencivenga et al., NJP (20013)

“Textbook equation” (optical)
Non-linear (wave-mixing) signal

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

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

**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 $\delta k^{-1}$ → increase in $N$ → $N^2$ increase of $I_{\text{fwm}}$ along $k_{\text{fwm}}$
“mini-TIMER” (@DiProI)

K-B

Padres
(I₀, spectrum, filters, beam position, etc.)

DiProI chamber

Phase matching

k_FEL₁

k_FEL₂

2θ

M₀

M₁

M₂

 CCD

θ₁

θ_B

λ_opt

λ_FEL

k_wm

2θ = 6°

θ_B = 48.2°

λ_opt = 392.8 nm

λ_FEL = 27.6 nm
Alignement (reference pinholes + screens) $\rightarrow \delta \theta \approx 0.2^\circ$

Degrees of freedom: $M_{1,2,3}$ pitch-roll-Z-Y; $M_{1,2}$ X; sample/sample pinhole X,Y,Z,pitch,roll

$2\theta = 2-9^\circ$ at $\Delta t_{\text{FEL-FEL}}$ = constant

$\Delta t_{\text{FEL-FEL}} = \pm 0.2$ ps at $2\theta$ = 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 ($2\theta > 0$).
Inprints on PMMA (and SiO$_2$) $\rightarrow 2\theta = 3.16^\circ$
Grating visibility after multi-shot exposure $\rightarrow$ FEL$_1$-FEL$_2$ optical path difference $< \lambda_{FEL}$ ($< 27.6$ nm)
Quantitative analysis (also single/multi-shot on PMMA) is running

Permanent gratings on SiO$_2$ (@ FEL flux $> 50$ mJ/cm$^2$)

Clean sample

FEL2014 – Basel – filippo.bencivenga@elettra.eu
"mini-TIMER" (@DiProI)

\[
\begin{align*}
2\theta &= 6.16^\circ \\
\theta_B &= 49.9^\circ \\
\lambda_{\text{opt}} &= 392.8 \text{ nm} \\
\lambda_{\text{FEL}} &= 27.6 \text{ nm}
\end{align*}
\]

Phase matching

\[\theta_B = 47-51^\circ; \Delta t = -20 +300 \text{ ps}\]

Ideal phase matching

\[\delta \lambda_{\text{FEL}} = \delta \lambda_{\text{opt}} = \delta \theta = \delta \theta_B = 0\]

\[
sinc^2(\delta k L_{\text{int}}) \Rightarrow 0.1-0.15 \text{ nm}
\]

0.03 nm

\[
\begin{align*}
\lambda &= 27.60 \\
\lambda &= 27.65 \\
\lambda &= 27.70 \\
\lambda &= 27.75
\end{align*}
\]
FEL-stimulated EUV-FWM signal

Phase matching

$2\theta = 6.16^\circ$
$\theta_B = 49.9^\circ$
$\lambda_{opt} = 392.8 \text{ nm}$
$\lambda_{FEL} = 27.6 \text{ nm}$

A “well defined” coherent beam propagates along $k_{out} \rightarrow \text{FWM}$
Vertical dimension fits with the divergence of input beams
Horizontal dimension is larger due to thin grating effects ($L_{abs} \approx 30 \text{ nm} < \lambda_{opt}$)

$v$-$\text{SiO}_2$ sample
$\lambda_{FEL}$
$M_1$
$M_0$
$\theta_B$
$\lambda_{opt}$
$M_2$
$\lambda_{FEL}$

$\theta_B = 47-51^\circ ; \Delta t = -20 +300 \text{ ps}$

$5-7 \text{ mm}$
$1-1.5 \text{ mm}$

Set of 5 signal-background images (total 3000-3000 shots with-without FEL, acqu. time $\approx 12 \text{ min.}$)

$F. \text{ Bencivenga (unpublished)}$
FEL-stimulated EUV-FWM processes

\[ \lambda_1 = \lambda_2 = 27.6 \text{ nm}; \ Q = 25 \mu m^{-1}; \ \lambda_3 = 392.8 \text{ nm} \]

\[ \Delta t \approx 0: \text{ sharp TG peak (} R_{cc}(\Delta t) ; \approx 130-140 \text{ fs FWHM, resolution limited)} \rightarrow \text{ electronic response} \ (\text{coherent spike}) \]

TG signal extends up to \( \Delta t \approx 100 \text{ ps} \) \( \rightarrow \text{ Longitudinal acoustic mode} \) at (almost) the expected frequency \( (\omega_{LA} = c_s Q \approx 0.145 \text{ THz}) \) and lifetime > 1 ns

TG efficiency \( (I_{TG} / I_{opt}) \) at \( \Delta t = 0 \approx 10^{-7} \) (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 \( \approx 10^{-5} \))

1) R. Cucini et al., Opt. Lett. 2011
FEL-stimulated EUV-FWM processes

\[ \frac{I_{TG}}{I_{opt}} - R_{cc}(\Delta t) \]

\[ \Delta t = 0\text{-}1.5 \text{ ps: } \text{two oscillations} \text{ ("optic modes") at } \omega_1 \approx 7.2 \text{ THz (} F_1 \text{ hyper-Raman mode } \rightarrow \text{ tetrahedral rotations) and } \omega_1 \approx 26 \text{ THz (} \nu_{2b} \text{ Raman mode } \rightarrow \text{ tetrahedral bendings)}. \]
FEL-stimulated EUV-FWM processes

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

A = \text{bkg (no FEL)} - \text{bkg*}  \quad \text{B = signal-bkg*}  \quad (\text{bkg*} \rightarrow \text{no FEL signal taken in another (nearby) portion of the sample})

A = |E_L|^2 \rightarrow \Delta t\text{-independent signal} \ (“\text{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

Q = 25 \mu m^{-1}

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

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Two-colour FEL-pump/FEL-probe

“twin-seed” two-colour\(^1\)

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

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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)

→ δ 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

\(\delta, \beta\)

\(\varepsilon = (1-\delta+i|\beta|^2)\)
Standing wave (TG) with "beatings" at $\omega_1 \pm \omega_2$

$k_3, \omega_3 = \omega_1$

"dummy pulse" (@ $\omega_2$)

$k_2, \omega_2$

Two-colour excitation pulses

$k_{\text{out}} = k_1 - k_2 + k_3$

$\omega_{\text{out}} = \omega_1 - \omega_2 + \omega_1$

M. Zangrando (FEL conference 2013)

"All-FEL" coherent (anti-stokes) Raman scattering
Transient grating + two-colour

Optical input fields ($\omega \sim \text{eV}$)
$\rightarrow \omega_{\text{ex}} < 0.1$’s eV (vibrations)
EUV/x-ray fields ($\omega_i > 100$’s eV)
$\rightarrow \omega_{\text{ex}} \sim 1-10$ eV’s (excitons)

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

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

$\omega_1 - \omega_2 = 5$ eV

$\chi^{(3)}$ (m$^2$ V$^{-2}$)

F. Bencivenga et al., Faraday Discus. (2014)
Transient grating + three-colour

\[ \omega_{\text{out}} = \omega_1 - \omega_2 + \omega_3 \]

Core resonance at atom-A

Core resonance at atom-B

\[ \omega_{\text{ex}} > \delta \omega \]

Transient grating + three-colour

FWM measures the **coherence between two atoms**: tuning $\omega_i$'s (to $\omega_{\text{res}}$'s of selected elements) and $\Delta t$ 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 $\lambda_i$'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

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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

"twin-seed" two-colour

Possible to achieve a three-colour seeded FEL emission at FERMI, but the tunability in $\omega_i$’s is limited by the FEL gain bandwidth →
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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• 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
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