Stimulated Resonant Inelastic X-Ray Scattering

SwissFEL

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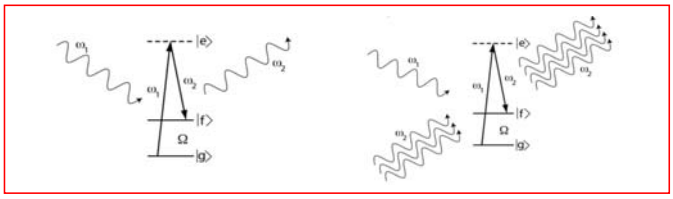
Introduction

Resonant Inelastic X-ray Scattering (RIXS) is good:
- element specific
- photon-in / photon-out
- may violate dipole-selection rule

but it has a very low counting rate.

Stimulated RIXS [1]:
- is analogous to stimulated Raman
- may be orders of magnitude more efficient
- may make t-dependent RIXS possible
- requires high brightness photon beams (XFEL) with multiple λ

1. spontaneous vs. stimulated RIXS



The stimulated cross-sections dominates for an incoming ω_2 φλυξ δενσιτψ

$$F(\omega_2) > \frac{\omega_2^2}{32\pi^3 c^2}$$

This corresponds to only of order 10^9 ph/XFEL pulse. (LCLS: 10^{12} ph/pulse)

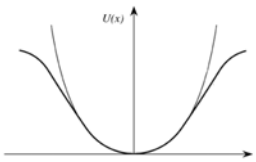
2. X-ray non-linearity

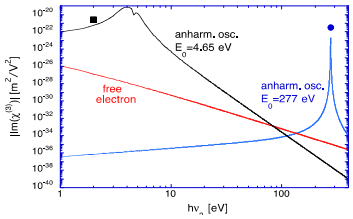
Stimulated RIXS cross-section is proportional to the 3rd order susceptibility:

$$\frac{d^2\sigma_{stim}}{d\Omega_2 d\omega_2} = \frac{32\pi^2\hbar\omega_1\omega_2}{\varepsilon_0 c^2} F(\omega_2)\,\mathrm{Im}\left(\chi^{(3)}\right)/N$$

which can be estimated using an anharmonic oscillator model [2]:

$$\chi^{(3)}\left(\omega_2 = \omega_2 + \omega_1 - \omega_1\right) = \frac{6Ne^4}{\varepsilon_0 m^3 r_a^2}\frac{\omega_0^2}{\left|\omega_1^2 - \omega_0^2 - 2i\gamma\omega_1\right|^2\left(\omega_2^2 - \omega_0^2 - 2i\gamma\omega_1\right)^2}$$

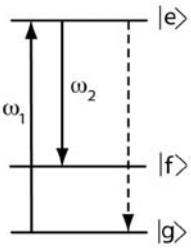




$$\omega_1 = \omega_0 = \omega_2 + \Omega;\ \gamma \approx \Omega << \omega_0;\ \hbar\gamma = 1\,eV$$

$$\chi^{(3)}_{res,X-ray}/N \approx \frac{e^4}{8\varepsilon_0 m^3 r_a^2 \omega_0^2 \gamma^4}$$

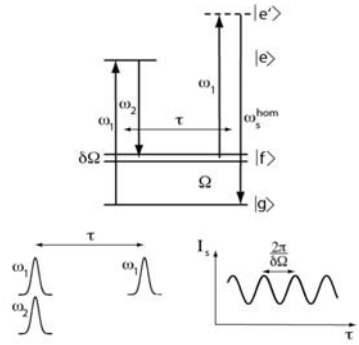
3. Stimulated RIXS pumping and FWM



$$\frac{\partial\Delta}{\Delta} \approx \frac{-32\pi I_1 I_2 \tau}{\varepsilon_0 \hbar c^2}\mathrm{Im}\left(\frac{\chi^{(3)}}{N}\right)$$

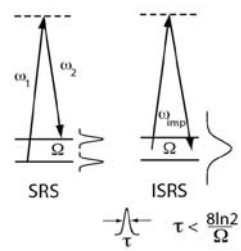
Stimulated RIXS can be used to efficiently excite an intermediate state $|f\rangle$
This is visible via a decrease in fluorescence (dashed arrow).

For a typical case, full inversion ($\partial\Delta/\Delta = -1$) occurs for incident peak powers $P_1 = P_2 = 30$ MW. (LCLS: 10 GW!)



A second stimulated scattering events (⇒ "Four-Wave Mixing") can be used to detect coherent quantum beats in the intermediate states.

4. Impulsive stimulation

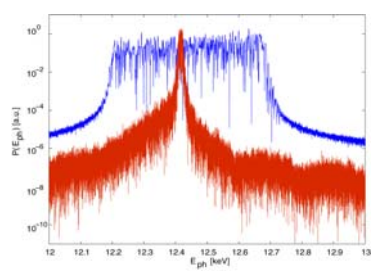


A single broadband pulse (e.g., lifetime broadened) can deliver both the components ω_1 and ω_2.

5. SwissFEL possibilities

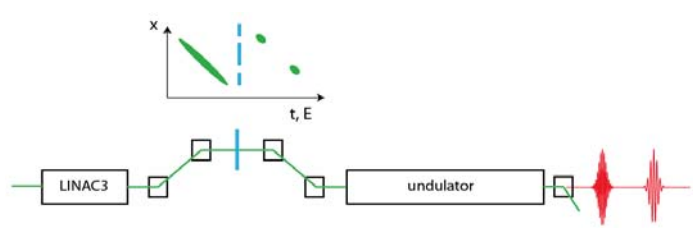
A) overcompressed broadband mode

For impulsive stimulation, the pulse bandwidth limits the RIXS excitation energy.



By taking advantage of strong wake fields in the C-band accelerator structures, a 4% FWHM broadband mode is possible (blue curve), i.e., much broader than normal SASE (red) [4]. (LCLS: 1.5%)

B) 2-color pulses from masked spoiler



Slits in a thin "spoiler foil" inserted in the LINAC bunch compressor only allow lasing to occur during two temporally-separated slices [5].

Variation of the slit spacing and bunch compressor parameters may allow tuning of the timing and wavelength offsets of the resulting photon pulses [3].

References

[1] BD Patterson, *SLAC Tech. Note* **SLAC-TN-10-026** (2010)
[2] RW Boyd, *Nonlinear Optics*, Elsevier (2008)
[3] S Reiche, *private communication* (2010)
[4] P Emma, et al, *private communication* (2010)