

*Time-Resolved measurements by FEL spontaneous emission:  
A proposal for sub-picosecond pumps & probe structural and  
spectrometric investigations*

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# Diffraction mode: Energy Dispersive

## What is and why to use it?

A diffraction pattern represents the intensity of an X-ray radiation elastically scattered by a sample as a function of the momentum transfer  $\Delta p$ .

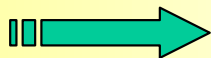
If the system under measurement is isotropic, the scattering depends no longer on the direction of  $\Delta p$  but on its magnitude  $q$  (scattering parameter) only:

$$q(E, \theta) = \alpha E \sin \theta \quad (\alpha = 1.014 \text{ \AA}^{-1} / \text{keV})$$

( $E$  = energy of an electromagnetic radiation,  $2\theta$  = scattering angle)

To scan a certain  $q$  range, two possibilities are available:

1. to make an *angular scan*, keeping  $E$  fixed  
Angular Dispersive mode (ADX).



2. to make an *angular scan*, keeping  $\theta$  fixed  
Energy Dispersive mode (EDX).



In the first case a *monochromatic* X-ray beam must be used, in the second, a continuous *polychromatic* (white) beam

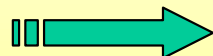
## Why to use the Energy Dispersive Mode?

1. Simpler geometric arrangement: no movement is required during the measurement



Prevention of systematic errors due to movements

2. Parallel collection of the experimental points at the various q-values



One-shot measurements

## *Intrinsic synchronization mode:*

*Consisting in splitting a laser pulse into two components, the first to be used as the **pump**, while the second to be converted into the X-ray **probe**:*

- **Thomson Scattering**
- **Laser Induced Plasma sources.**
- **X-ray Diodes.**

## *External synchronization mode:*

*for independent sources) making use of the synchrotron radiation*

*Intrinsic  
synchronization  
mode*

# Thomson Scattering

First proposed in 1963, a fast electron beam is used to **convert the second component of the split laser pulse into the X-ray probe** (via inelastic scattering). The resulting X-rays are short, highly directional, and tunable in terms of energy (photons up to 300 keV were obtained)

# Laser Induced Plasma sources

In this case, **flashes of monochromatic X-rays** (characteristic emission lines) can be generated by suprathermal electrons when a **laser pulse is focused onto a solid surface**. X-ray pulse duration close to 100fs can be obtained



# X-ray Diodes

X-ray tube photocathodes excited by short laser pulses generate picosecond X-ray pulses that have the same temporal characteristics as the photoemitted electron pulse impinging on the anode. This method is limited by competitive effects that do not allow high power short pulses to be obtained.

*External  
synchronization  
mode*

To-date Pump (optical laser pulse) & Probe (synchrotron X-ray pulse) for time resolved structural studies:

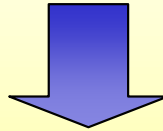
**Technique:** Monochromatic X-ray beam diffraction

**Relevant time quantities:**

**Laser** pulse duration : negligibly small ( $< 100$  fs)

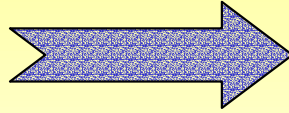
**Synchrotron** pulse duration  $\sim 100$  ps

Laser pulse to Synchrotron pulse **synchronization jitter**  $\sim 5$  ps



Two options

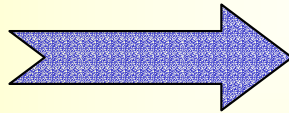
"**Low**" temporal resolution mode



Suitable for "**slow**" processes happening on the **time scale of ns** or longer

Equivalent to conventional optical pump & probe experiments: **delay time scan**

"**High**" temporal resolution mode



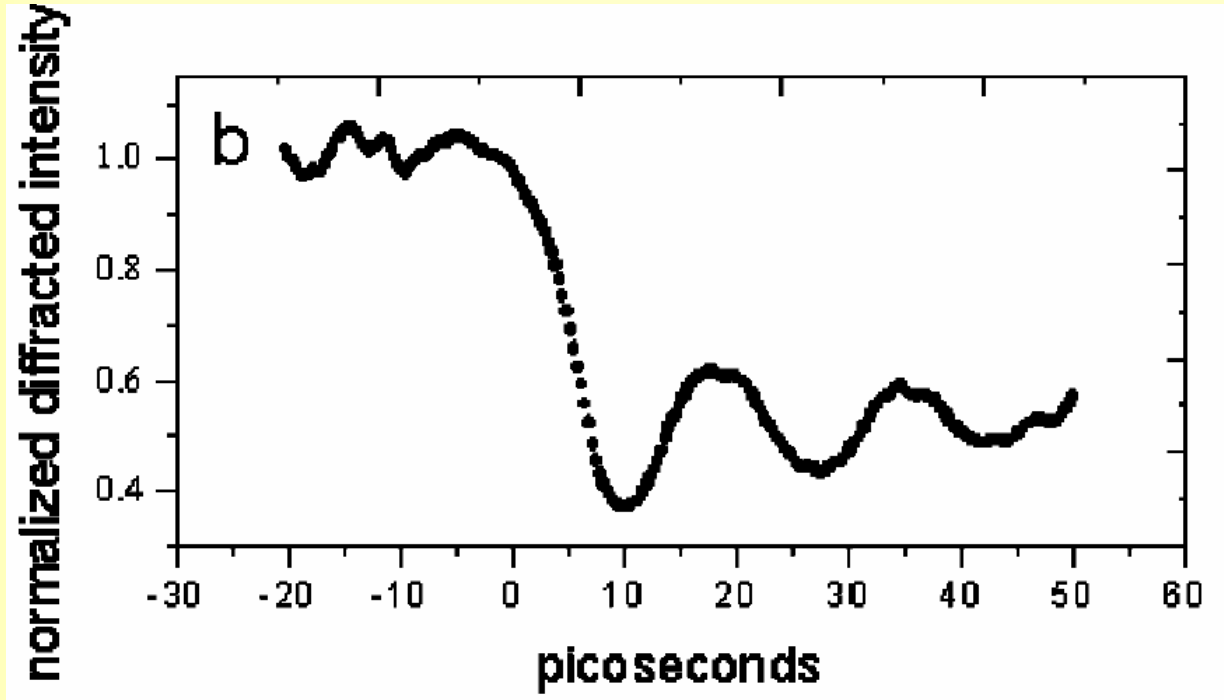
Suitable for "**fast**" processes happening on the **time scale of tens of ps**

Makes use of a Streak Camera whose **temporal resolution ~ 3ps @ 10 keV**.

A **single shot** measurement only is required to follow the whole evolution of the system (to be repeated several times to improve the statistics)

## Example:

Lattice coherent oscillations observed by measuring the intensity fluctuations of a Bragg peak



**Time-Resolved X-ray Diffraction from Coherent Phonons during a Laser-Induced Phase Transition**

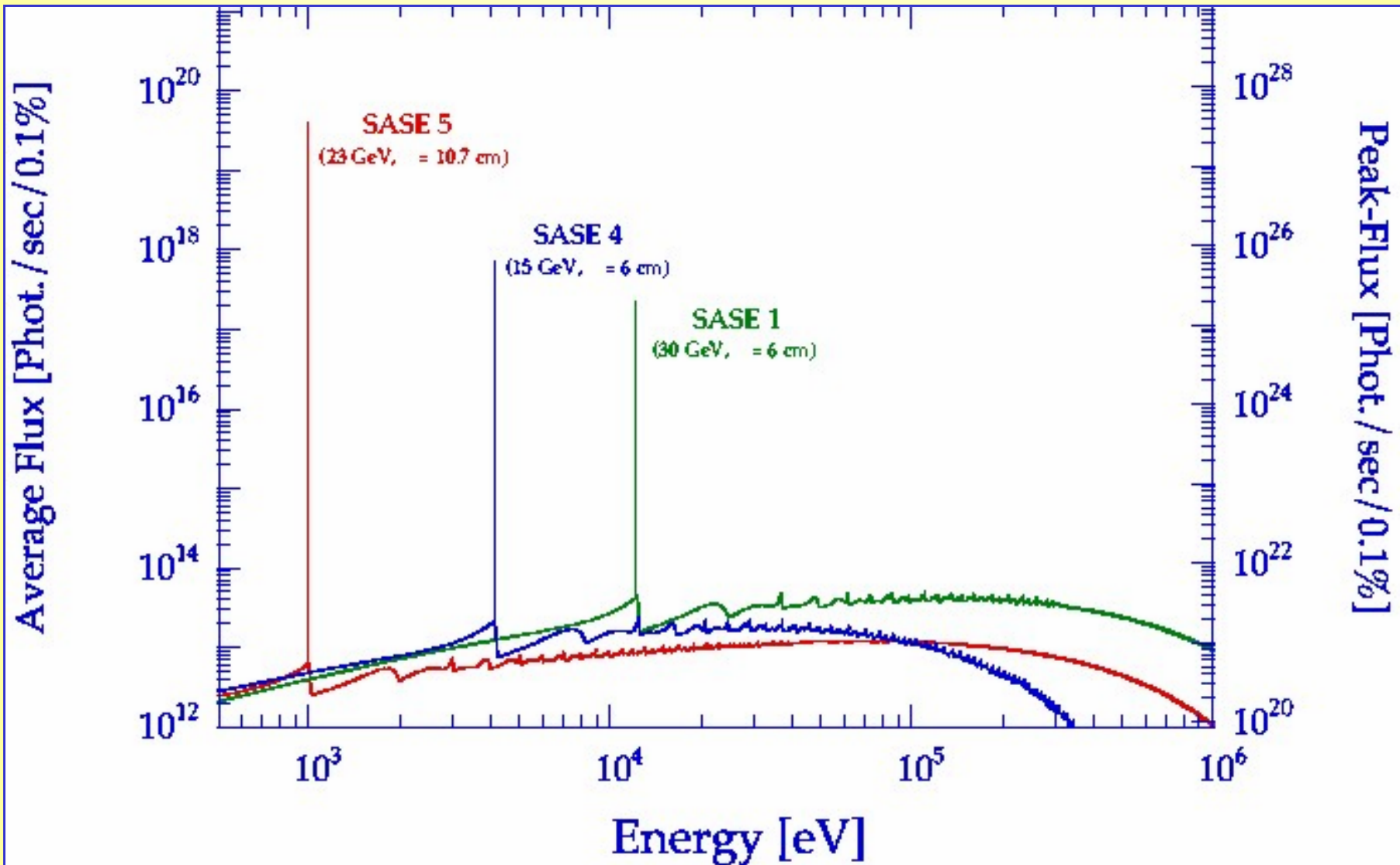
A. M. Lindenberg et al, PRL 84(1), 2000. (performed @ ALS, Lawrence Berkeley National Laboratory (Ca, USA))

Problem:

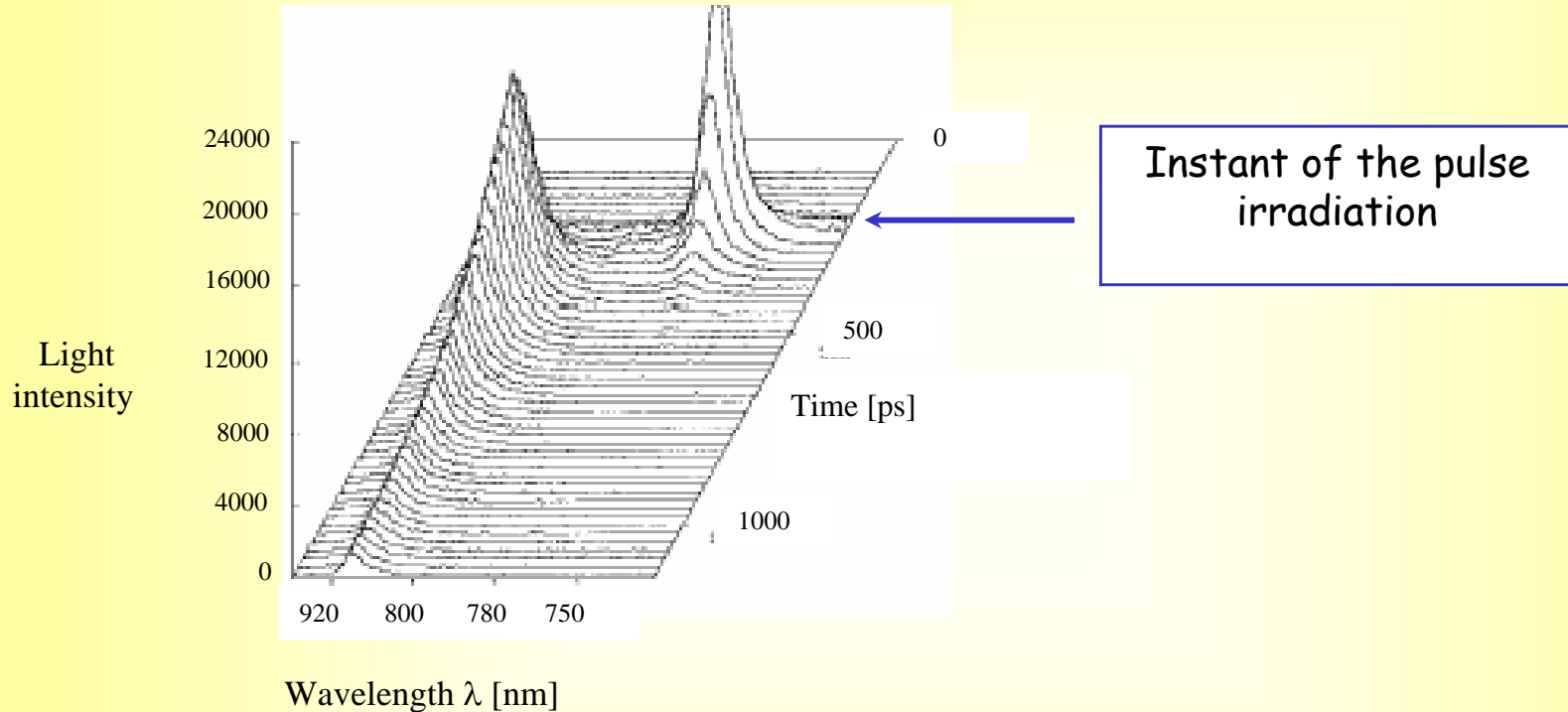
How to break the ps wall to take advantage from the unique time structure of a FEL?

Proposal: **Time-resolved Energy-Dispersive X-ray Diffraction**

FEL monochromatic lines are superimposed to a strong white radiation (graph from DESY website)

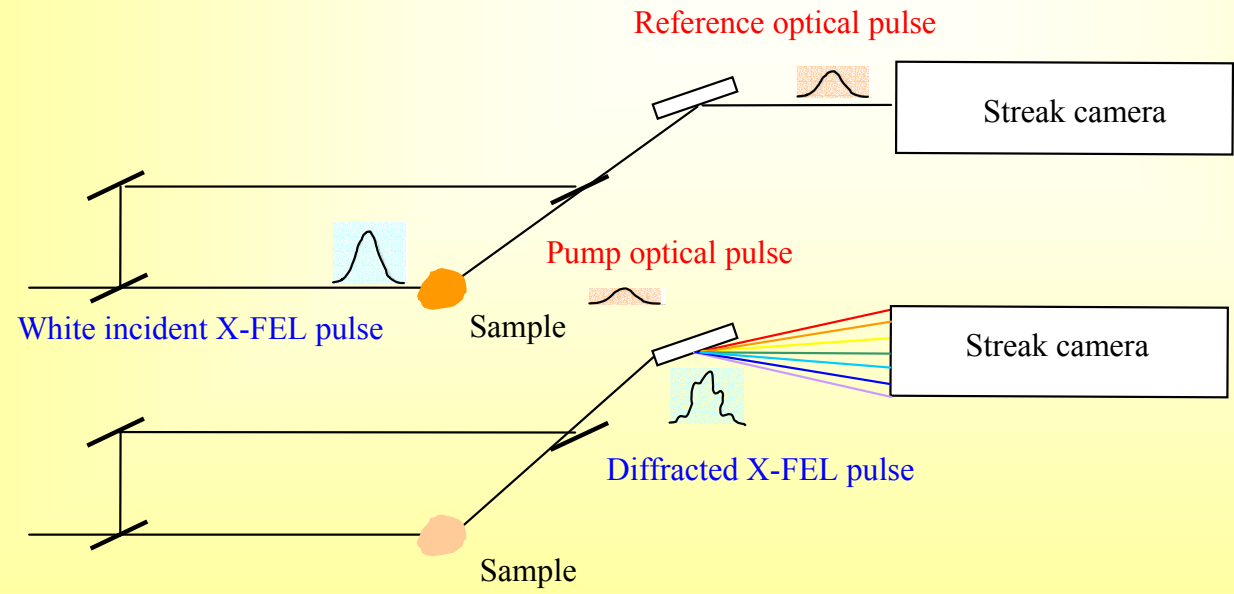
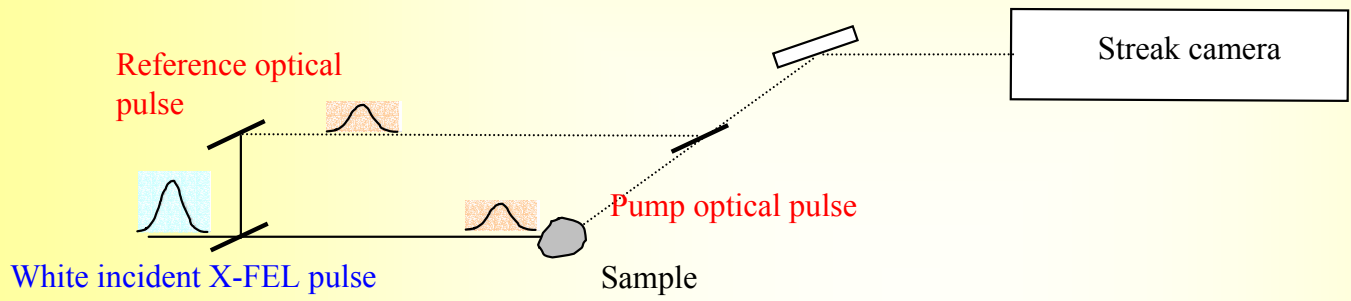
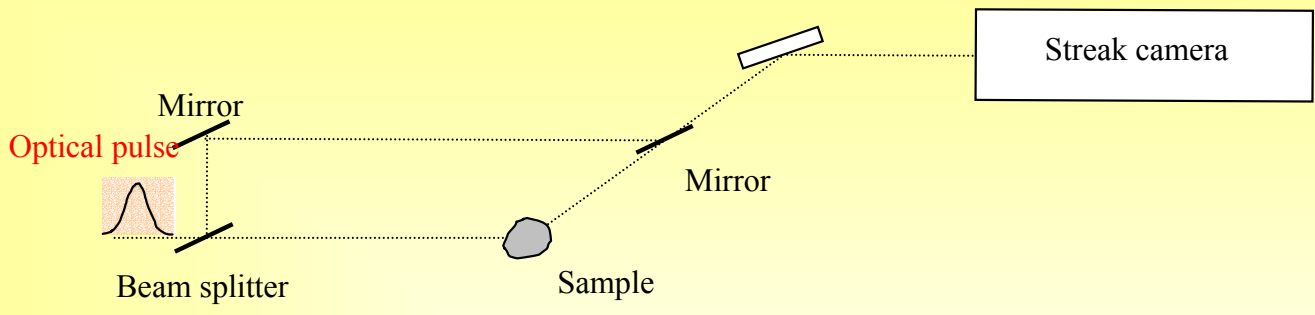


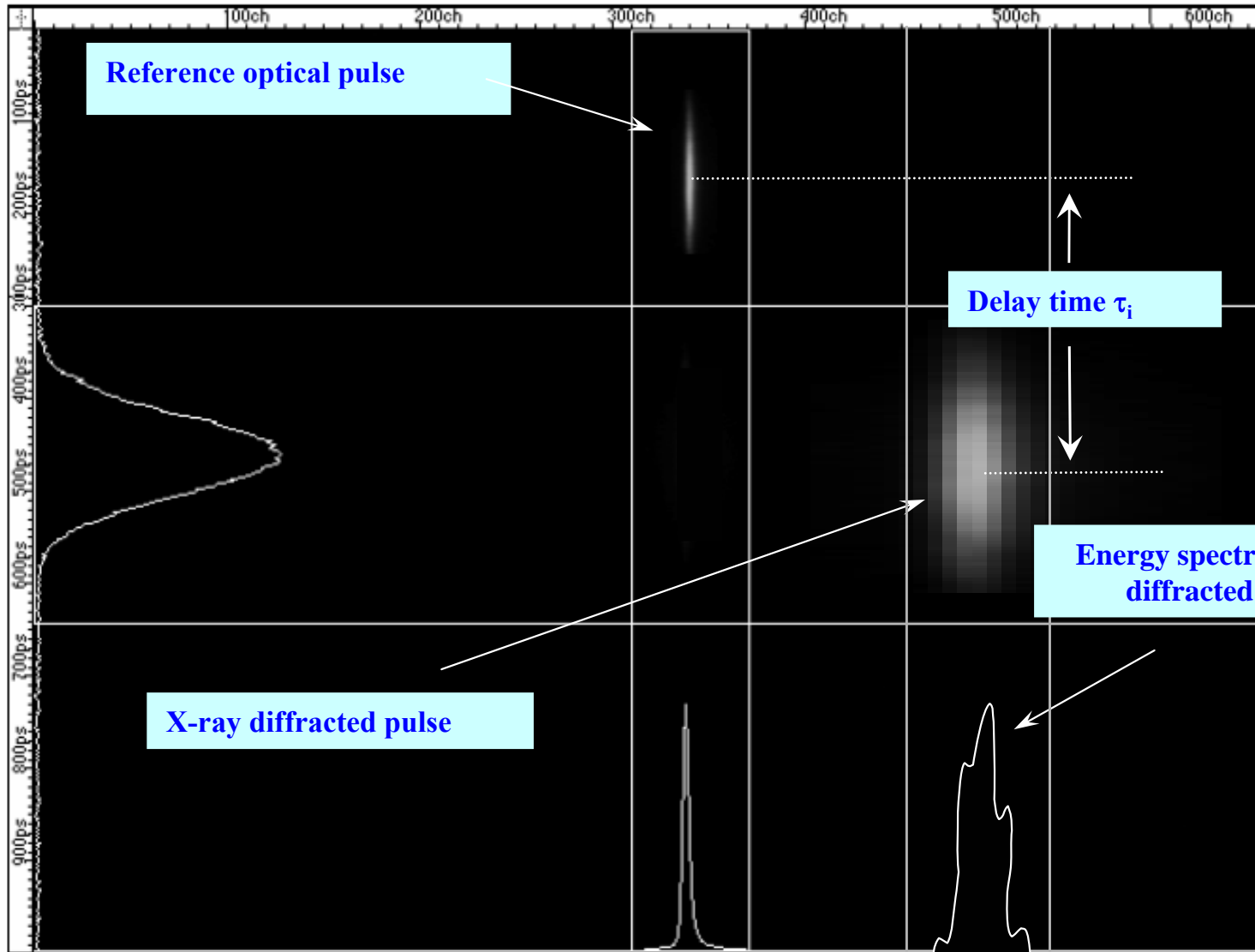
A **Streak Camera** could be used for time resolved data collection. In the following figure, a sequence of fluorescence spectra collected by a streak camera after irradiation of a sample with a short pulse is shown.



Alternatively, an **Energy Dispersive diffraction measurement** at a fixed angle using a polychromatic beam could be done using the same method. In this case, a 3D plot similar to that in figure would be acquired, but the sequence would represent the **time evolution** of the diffraction pattern (instead of the fluorescence spectrum) and, therefore, **of the sample structure**.







The delay time  $t$  is to be considered as a **random variable**.  
The only constrain is that  $t$  must be shorter than the sweep time of the streak camera (otherwise the two pulses cannot be visualized in a single scan).

Therefore, instead of fixing  $t$  **deterministically and a priori**, for instance through a delay line, it is **measured a posteriori as the sampling of a stochastic variable** (like in the Montecarlo method).

In this way, the  $t$  space is progressively populated by repeating many times the diffraction experiment (construction of the ensemble of events by many repetitions of the experiment).

By fitting or deconvolving the spots produced by the two pulses (shorter than the resolution time of the streak camera), a higher **temporal resolution** on  $t$  can be obtained.

## Other possible advantages:

Possibility of **first structural measurements** since, due to the wide energy spectrum (hardest components have an energy 2-3 orders of magnitude higher than that of the laser line), it could allow diffraction studies **before lasing in the keV range is reached**.

It would **use** one of the outstanding properties of the X-ray beam, namely its pulsed **time structure**, **preventing** the possible **damages** to the samples **due to** the high **intensity** (ablation, Coulombian explosion etc.)

Being based on the spontaneous emission, it is **independent of** laser **saturation**, which currently represents one of the major problems.

It would share the **same** experimental **apparatus** with **spectroscopy** measurements, minimal changes being required.

## Reference:

*“ Time-Resolved Energy Dispersive Diffraction from X-FEL spontaneous emission:  
A proposal for sub-picosecond pumps & probe structural investigations ”,*

**V. Rossi Albertini, B. Paci, P. Perfetti, NIM A 533, 584-590, 2004**