Istituto di Struttura della Materia

Consiglio Nazionale delle Ricerche

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

1° LIFE Meeeting
ENEA Frascati,
19-20 Febbraio 2009

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Outline

1) The Energy Dispersive X-ray Diffraction:
   What is and why to use it?

2) Synchronization in Pump&Probe experiments

3) A proposal for high temporal resolution Pump&Probe experiments by X-FEL spontaneous emission

Diffraction modes:
Angular vs. Energy Dispersive

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

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

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

(E = energy of the electromagnetic radiation, 2θ=scattering angle)

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

1. *to make an angular scan, keeping E fixed*

→ *Angular Dispersive mode (ADXD).*

2. *to make an angular scan, keeping ϑ fixed*

→ *Energy Dispersive mode (EDXD).*

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 ⇒ Easy one-shot measurements

Intrinsic synchronization mode:

Consisting of 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. Examples:

- **Thomson Scattering**
- **Laser Induced Plasma sources.**

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

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 ~ **100 ps**

Laser pulse to Synchrotron pulse **synchronization jitter ~ 5 ps**

⇓

Two options

1. "*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**

2. "*High*" temporal resolution mode

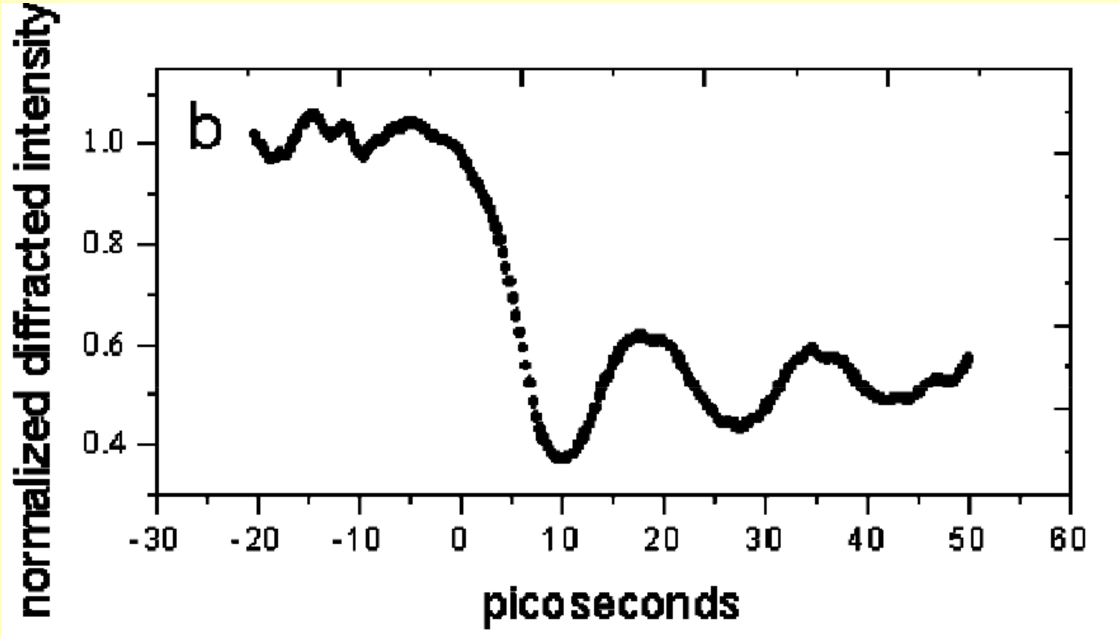
Suitable for "fast" processes happening on the time scale of tens of ps

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



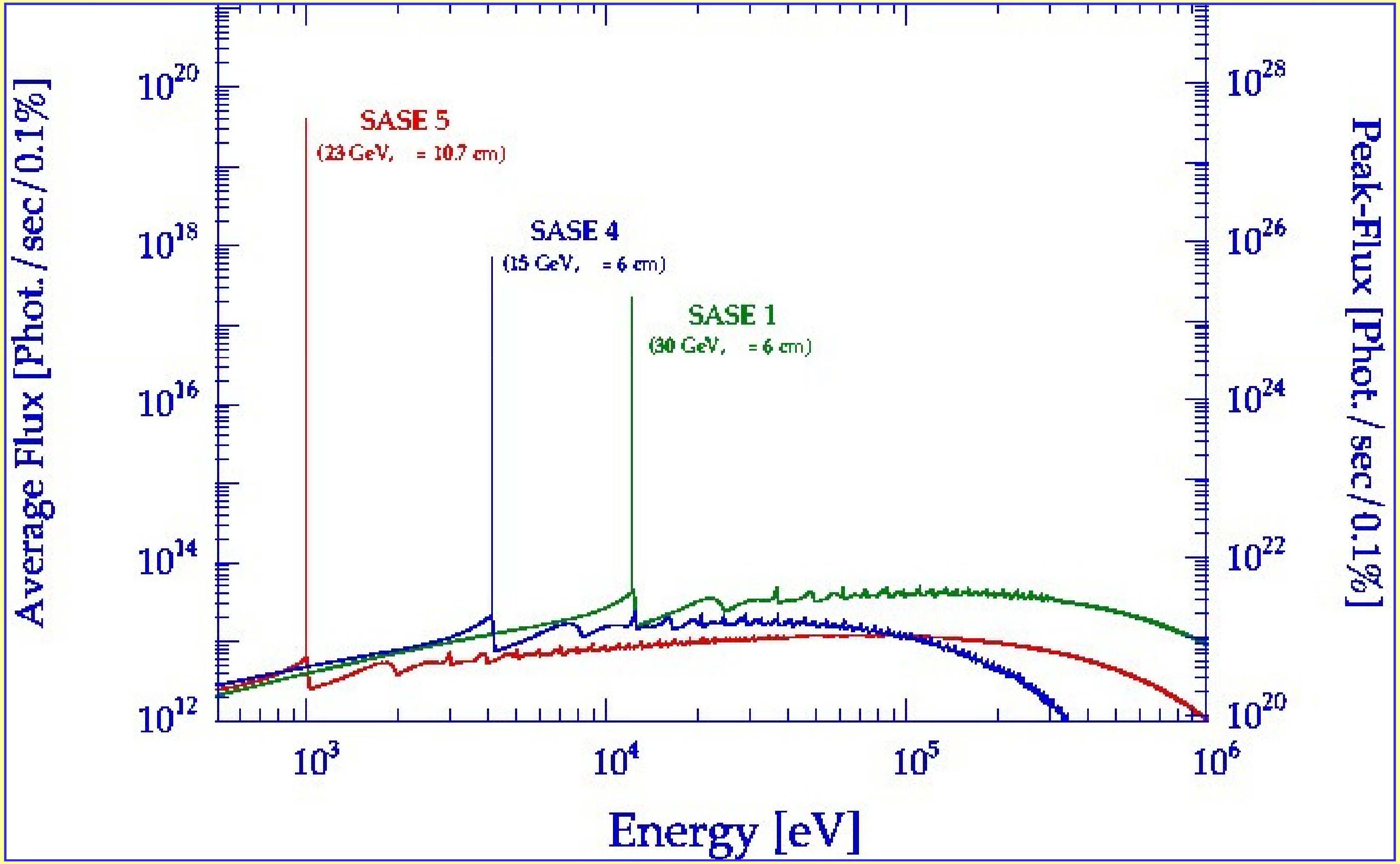
In “**Time-Resolved X-ray Diffraction from Coherent Phonons during a Laser-Induced Phase Transition**” by A. M. Lindemberg et al, PRL 84(1), 2000. (performed @ ALS, Lawrence Berkeley National Laboratory (Ca, USA))

Problem:

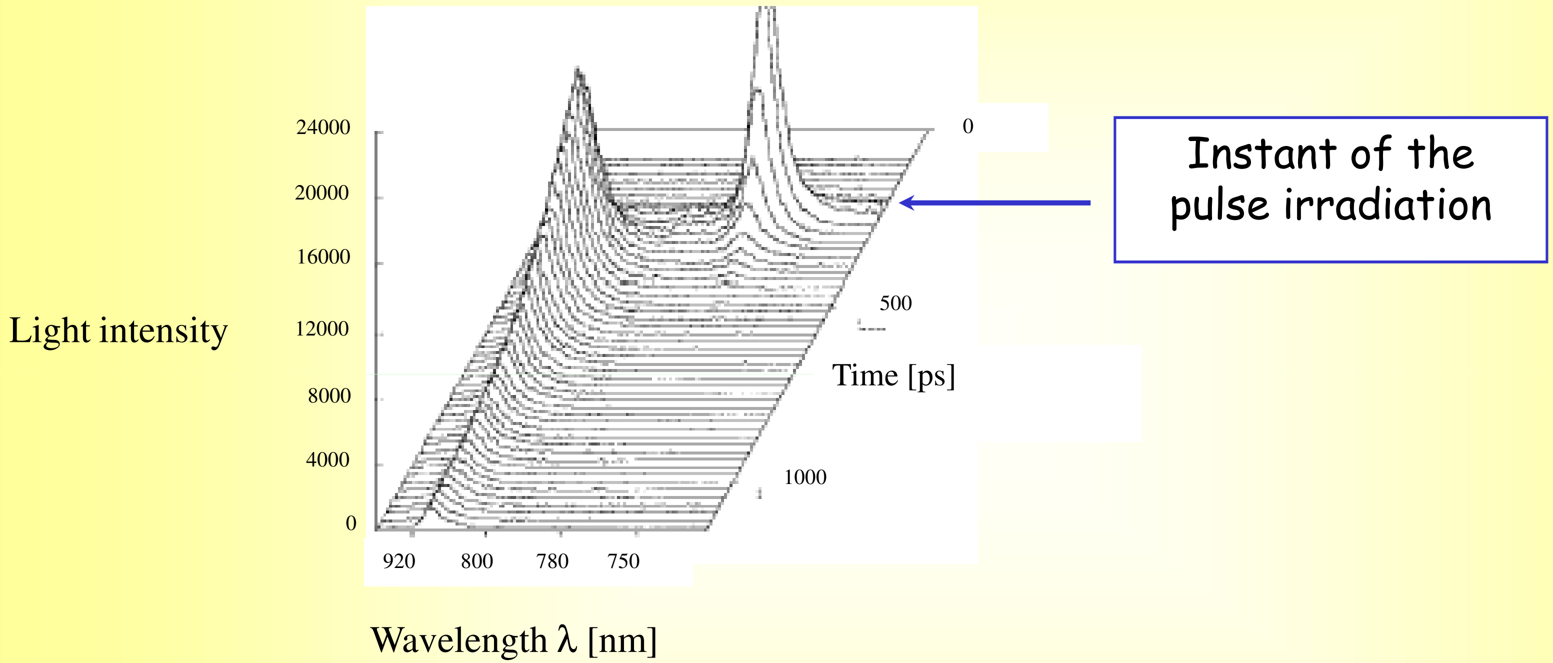
How to break the ps wall to take advantage from the unique time structure of a FEL?

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

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

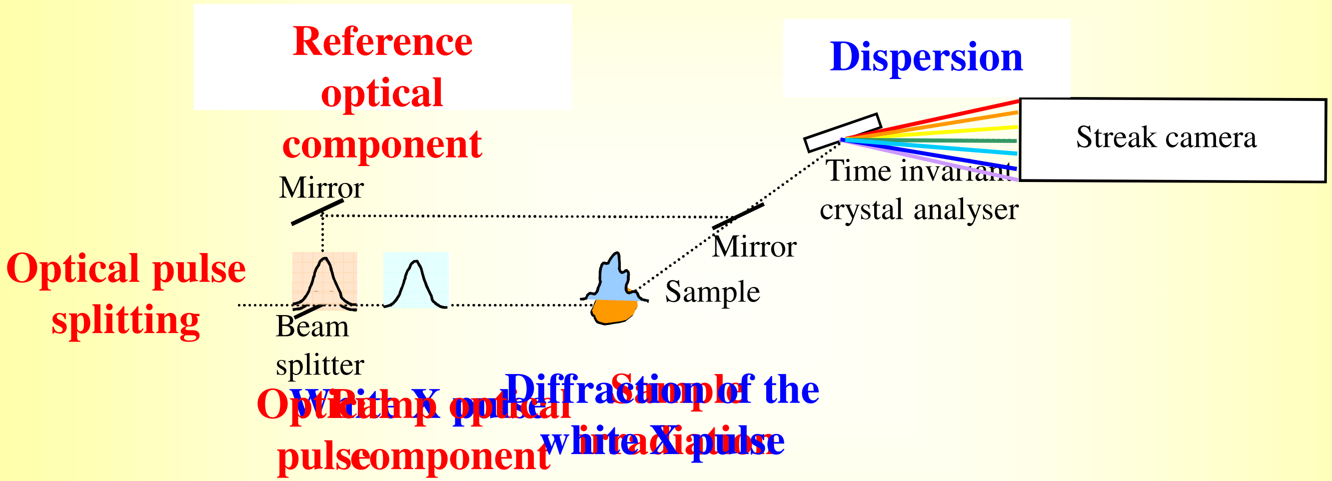


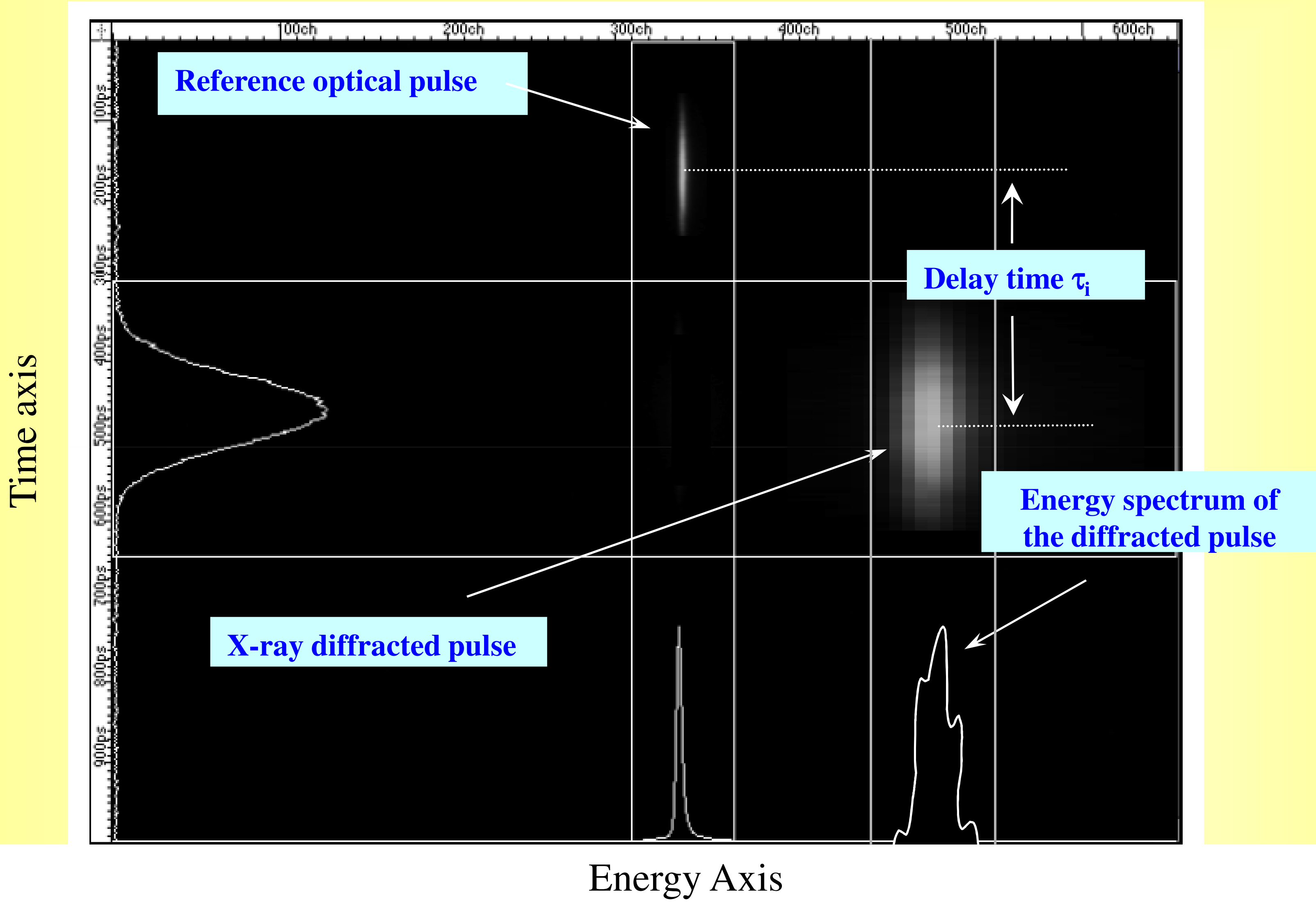
A **Streak Camera** can be used for time resolved data collection. In the following figure, a sequence of fluorescence spectra collected by a streak camera after irradiation of a sample with a short pulse is shown.

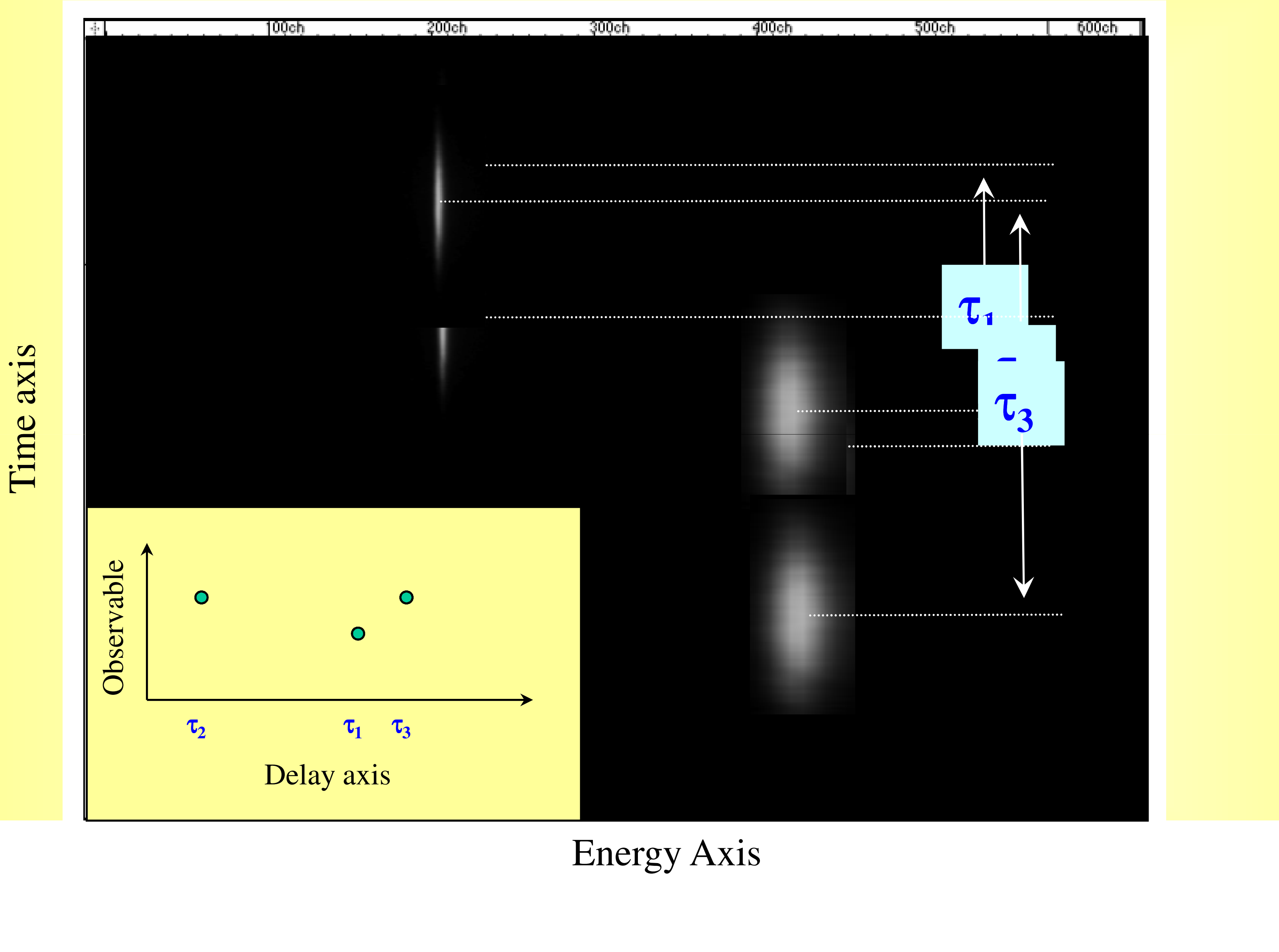


Analogously, an Energy Dispersive **diffraction measurement** at a fixed angle using a polychromatic beam could be done using the same method. In this case, a 3D plot similar to that in figure would be acquired, but the sequence would represent the **time evolution** of the diffraction pattern (instead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

Schematic setup of the experiment




Reference optical pulse
Delay time τ_i
Energy spectrum of the diffracted pulse
X-ray diffracted pulse
100ch
200ch
300ch
400ch
500ch
600ch
100ps
200ps
300ps
400ps
500ps
600ps
700ps
800ps
900ps
Time axis
Energy Axis


100ch
200ch
300ch
400ch
500ch
600ch
Time axis
τ1
–
τ3
Observable
τ2
τ1
τ3
Delay axis
Energy Axis

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

Therefore, instead of fixing τ deterministically and *a priory*, 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 τ-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 τ than that associated to the streak camera can be obtained!!!

Where's the trick???

We already know the FEL pulse shape in the time domain: It does not have to be measured by the streak camera…

The pulse distribution in the time domain is δ-like. Therefore, what actually appears on the streak camera monitor is the transfer function of the camera itself, which can be fully characterized prior to the experiment

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, in many cases also 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.

- This kind of experiments requires preliminary knowledge and practice with the detecting system, which can be gained by working at lower energies.

- The tests can be carried out at SPARC, by using a conventional (optical) steak camera and suitable samples for diffractometric and spectroscopic investigations.

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)