A New Era of X-Ray Science: Beyond one-photon-at-a-time

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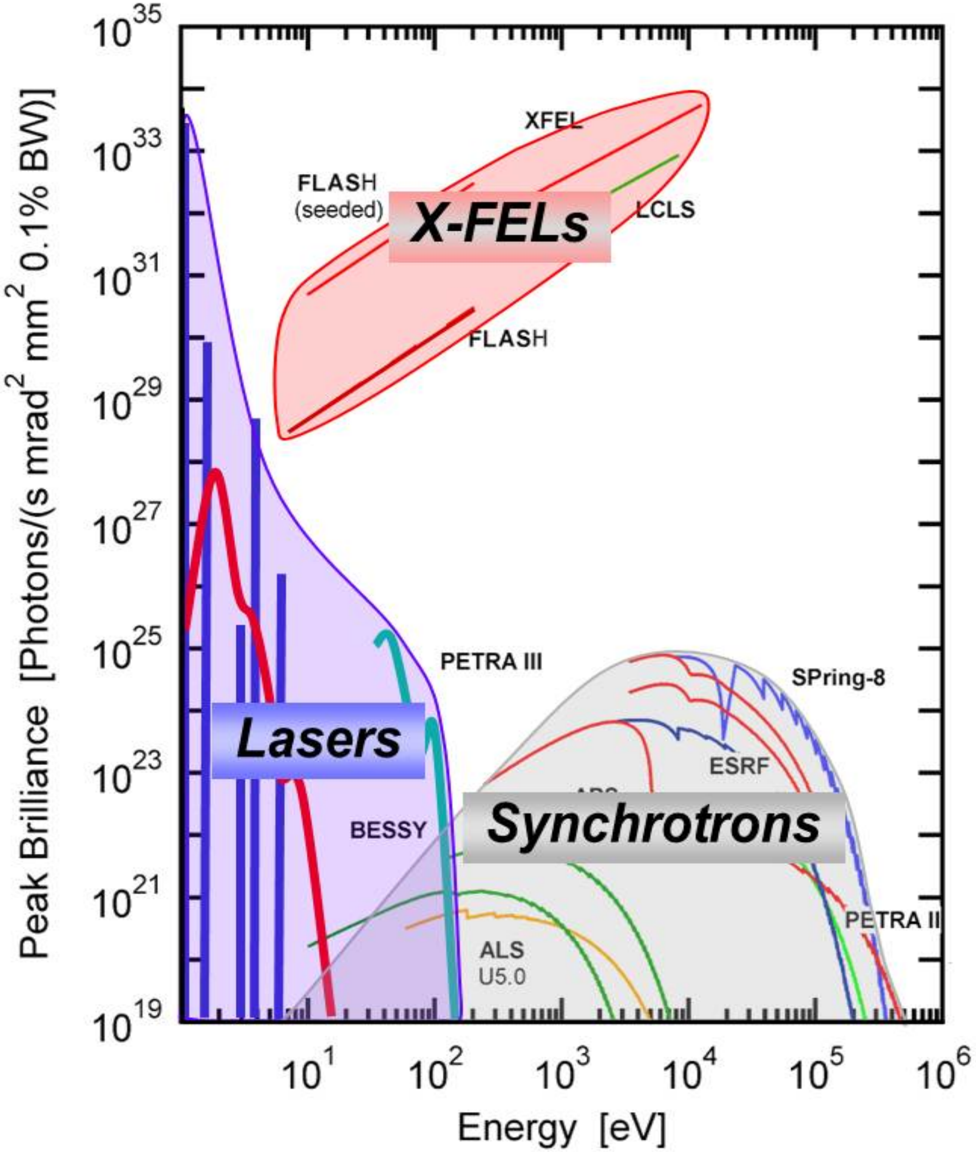
Science at FELs, PSI September 2014

Categories of Lightsources

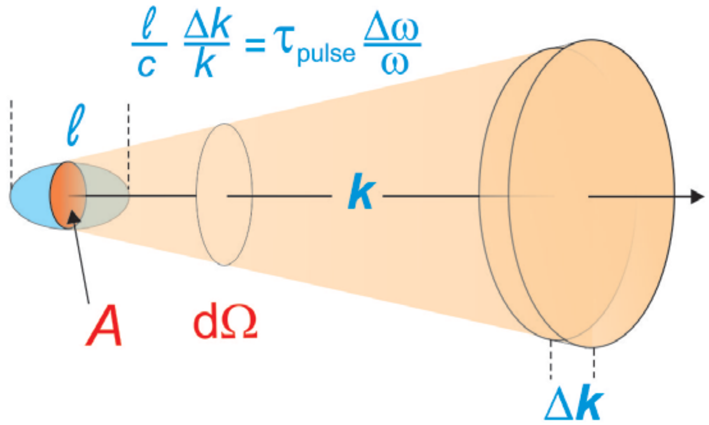


Synchrotron & X-FEL sources are based on electron accelerators

J. Ullrich, A. Rudenko, R. Moshammer
Ann. Rev. Phys. Chem. 63, 635 (2012)

Toward the perfect x-ray source

Source brightness (or brilliance)

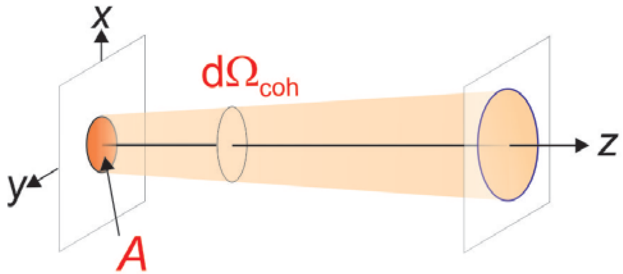


Peak brightness of a source

$$\mathcal{B} = \frac{n_{ph}}{\underbrace{A\, d\Omega}_{\Delta r\, \Delta p \geq \hbar/2} \;\; \underbrace{\tau_{pulse}\Delta\omega/\omega}_{\Delta t\, \Delta \mathcal{E} \geq \hbar/2}}$$

space-momentum uncertainty — time-energy uncertainty

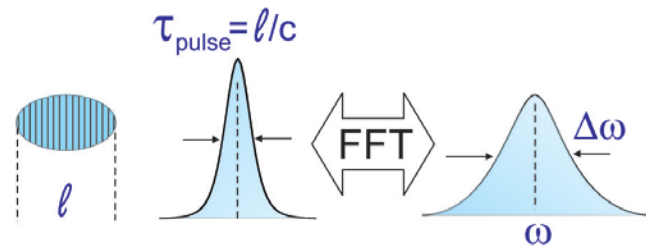
optimum synchrotron source



$A\, d\Omega_{coh} = \lambda^2$

"diffraction limited source"

optimum X-FEL source



$\tau_{pulse}\frac{\Delta\omega}{\omega} = \frac{\lambda}{c}$

"transform limited source"

If these conditions are satisfied, x-rays are completely coherent

Brightness, coherence & degeneracy parameter

Peak brightness of a source

$$\mathcal{B} = \frac{n_{ph}}{A\,d\Omega\;\tau_{pulse}\Delta\omega/\omega}$$

$$\tau_{pulse}\frac{\Delta\omega}{\omega} = \frac{\lambda}{c}$$

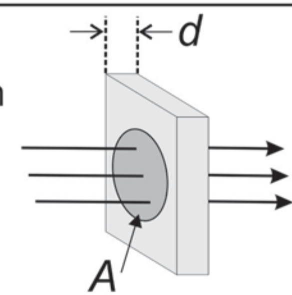
$$A\,d\Omega_{coh} = \lambda^2$$

Peak brightness of a coherent source

$$\mathcal{B} = \frac{n_{ph}^{coh}\,c}{\lambda^3}$$

flux of x-ray beam

$$\Phi_0 = \frac{n_{ph}\,c}{V} = \frac{n_{ph}}{A\,t_d}$$

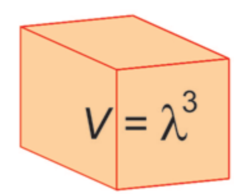


$$c = \frac{d}{t_d}$$

Peak brightness of coherent source = coherent flux

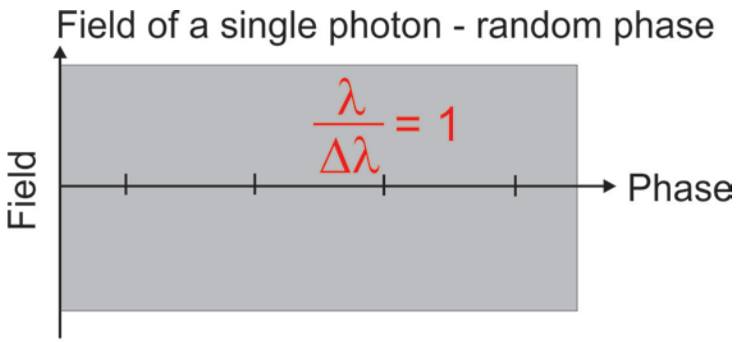
Degeneracy parameter = number of photons in λ^3

$$n_{ph}^{coh} = \frac{\lambda^3}{\mathcal{B}\,c}$$

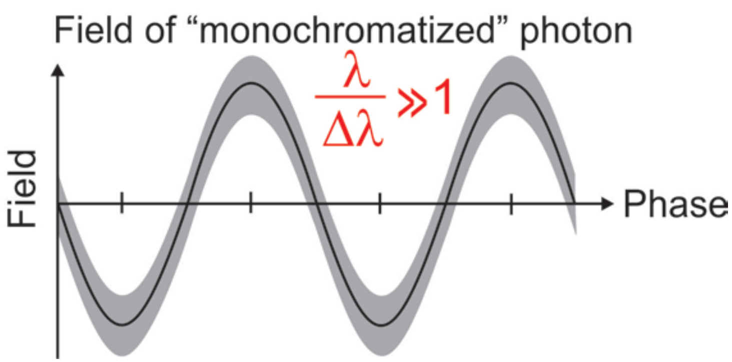


coherence volume
or "size" of one photon

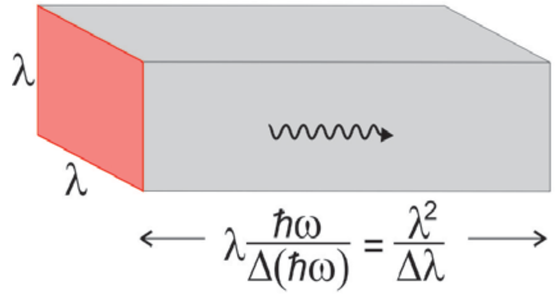
The effect of increased monochromaticity



$$\frac{\Delta k}{k} = \frac{\Delta\omega}{\omega} = \frac{\Delta\lambda}{\lambda} = 1$$

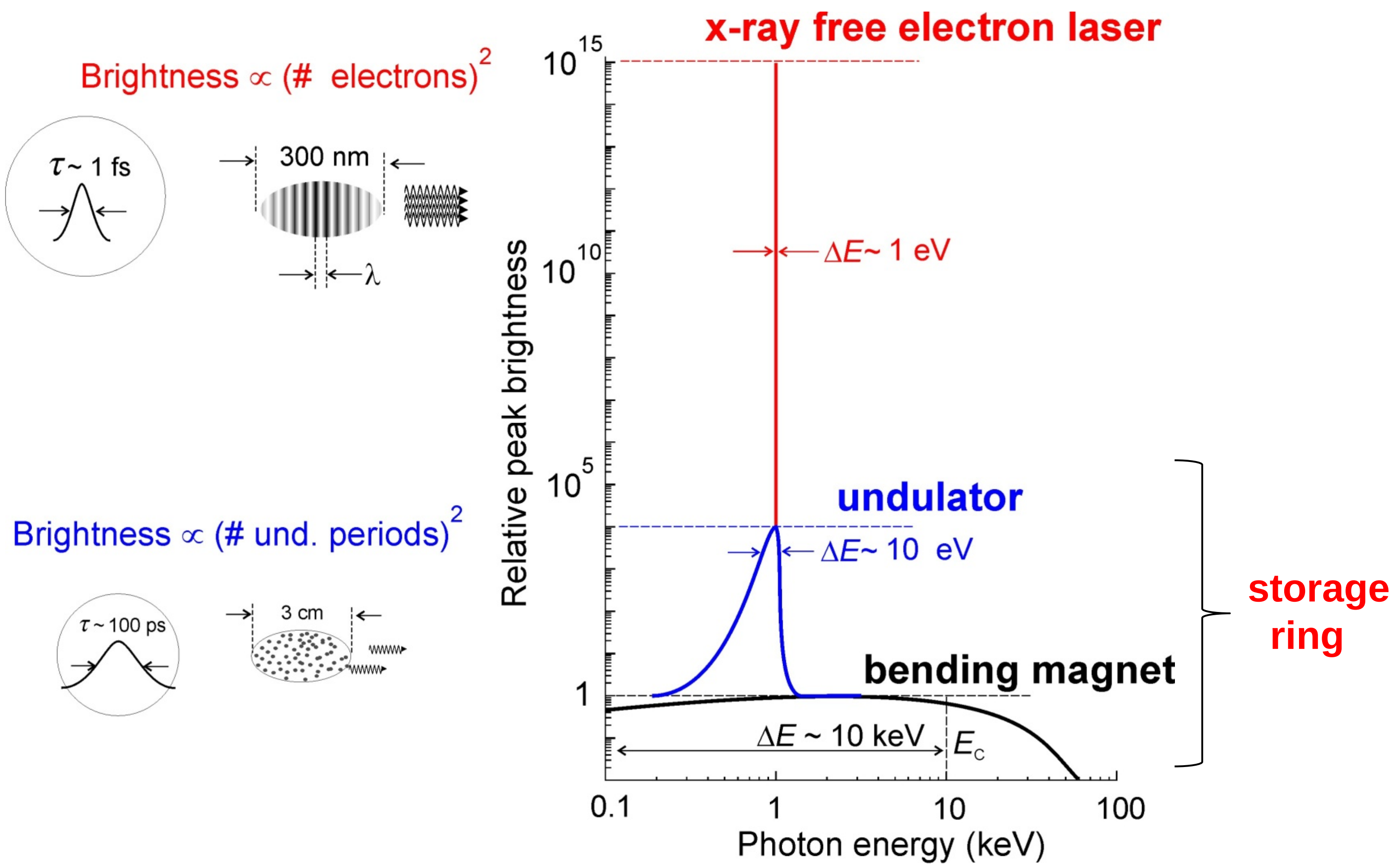


generalized photon coherence volume

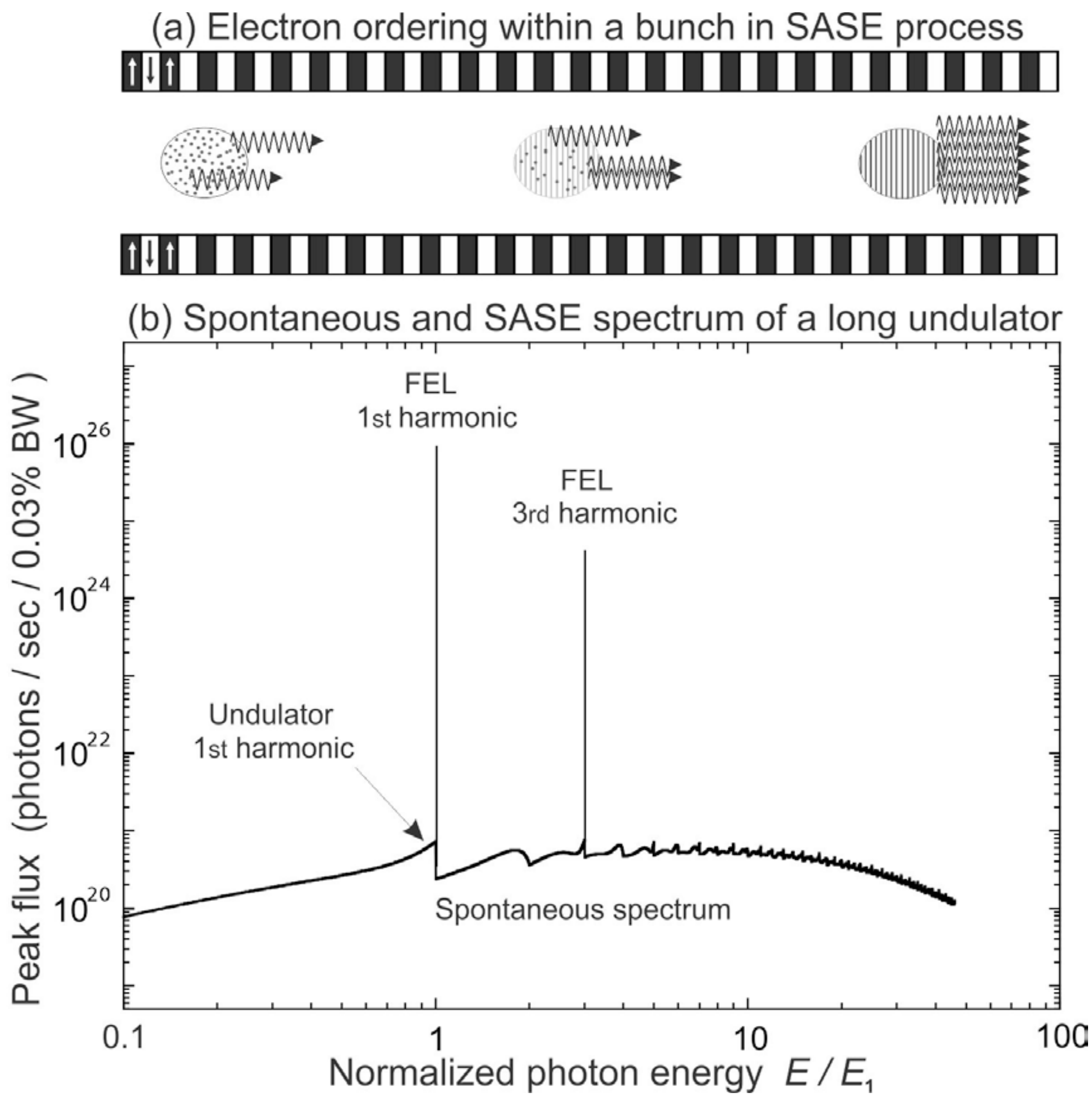


$$V_{\boldsymbol{pk}} = \lambda^3 \frac{\hbar\omega}{\Delta(\hbar\omega)}$$

Overview of x-ray source spectra

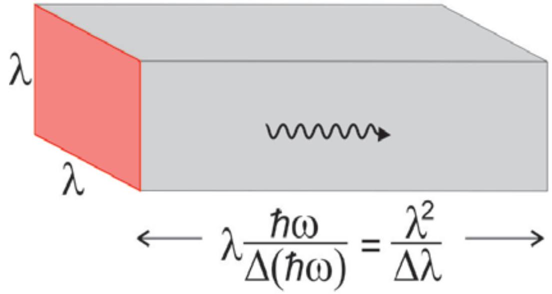


Calculated X-FEL spectrum



Example: Photons within coherence volume

photons in beam coherence volume:



$$n_{\mathrm{coh}} = B\,\frac{\lambda^3}{(2\pi)^3 c}$$

Storage ring: $n_{coh} < 1$ photon → "one photon at a time"

X-Ray laser: $n_{coh} \sim 10^9$ photons

Who needs peak brightness?

Three examples:

- **Coherent Imaging**
  Laterally coherent (diffraction limited) photons

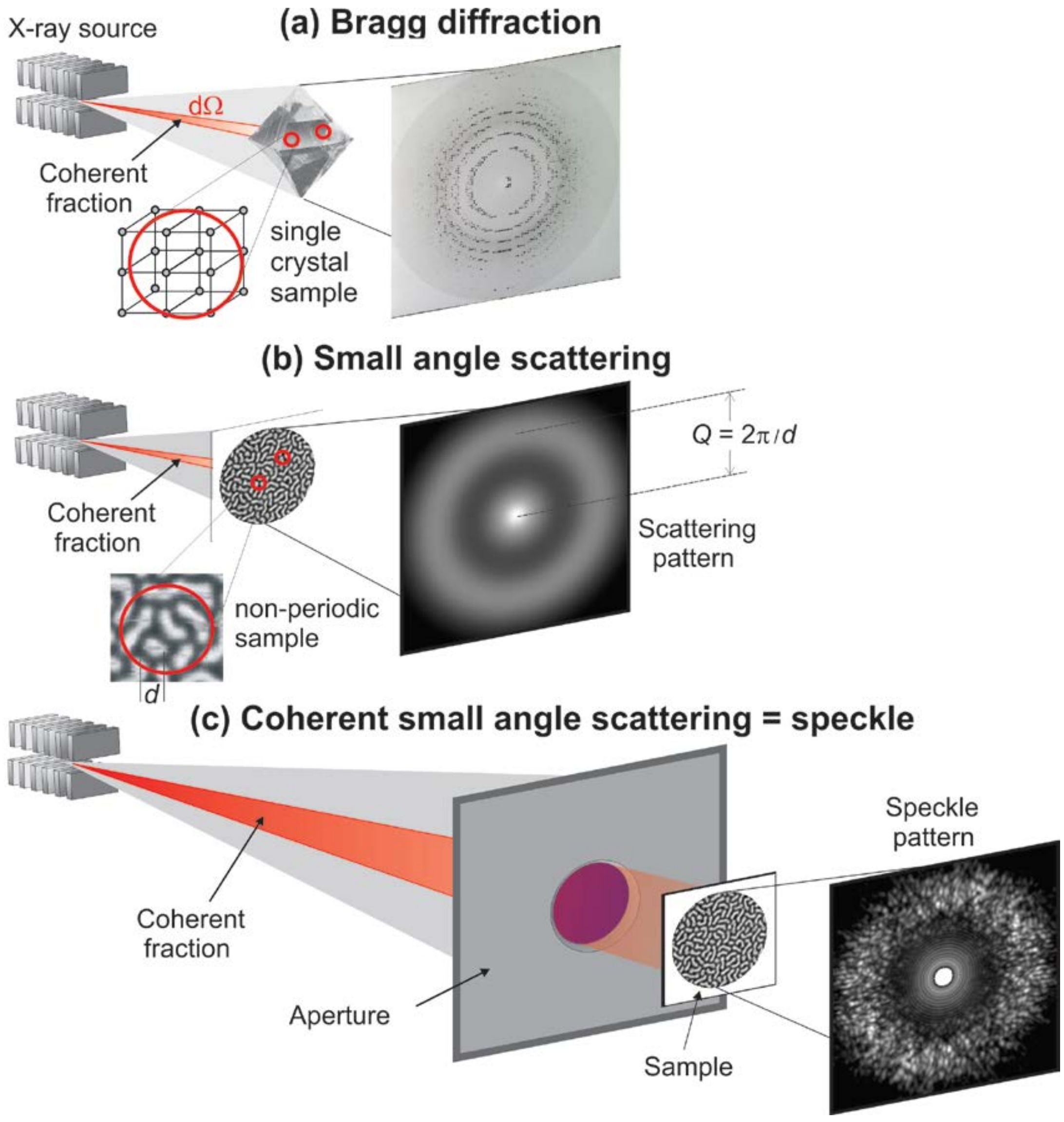
- **Ultrafast Science**
  photons in ultrashort pulse

- **Non-linear x-ray science**
  Laterally and longitudinally coherent photons

- **Coherent Imaging**

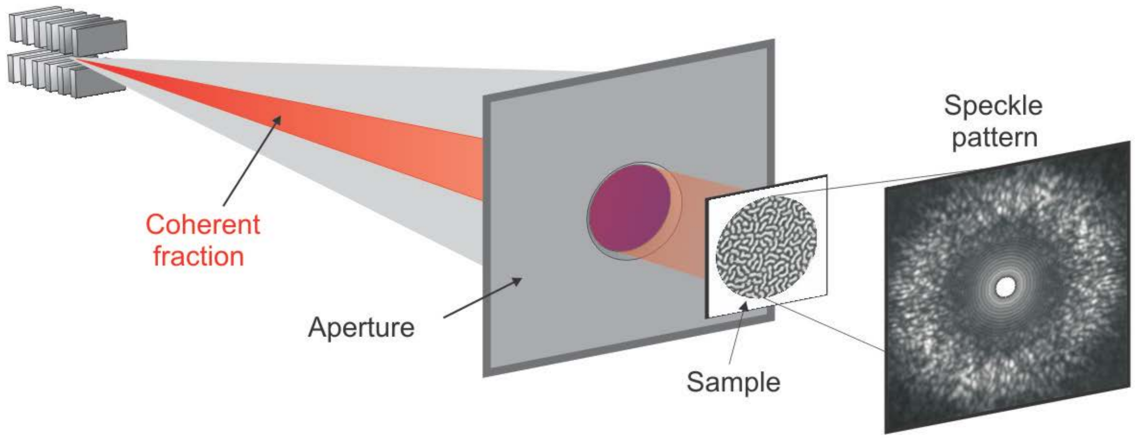
  Laterally coherent (diffraction limited) photons

Importance of a "diffraction limited source" or laterally coherent source

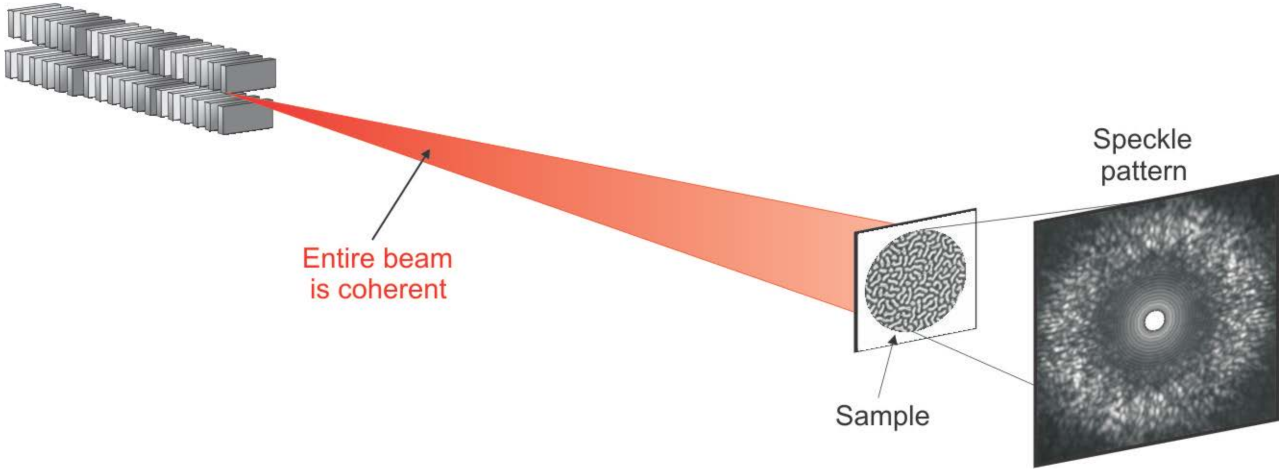


X-FEL x-rays are diffraction limited allow single shot diffraction imaging

Storage ring: undulator

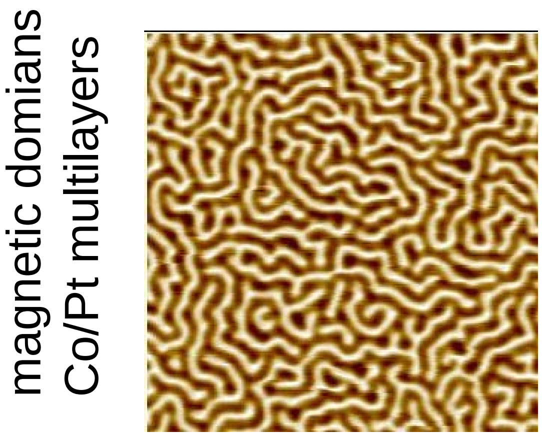


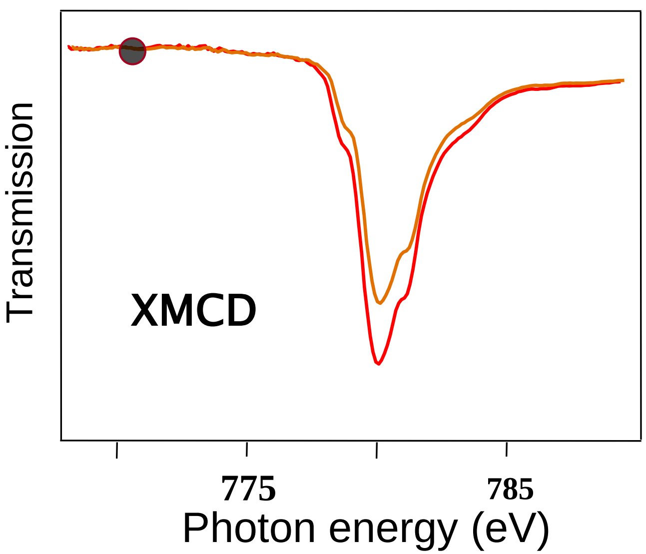
X-ray Free Electron Laser

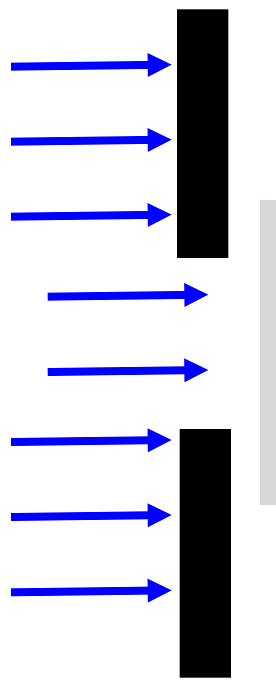


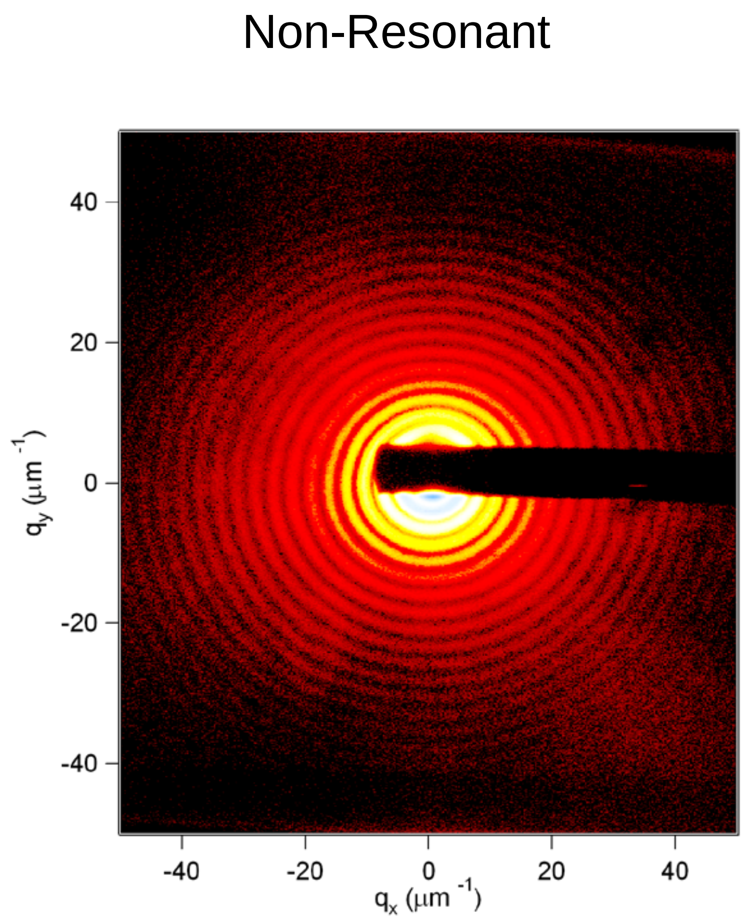
The importance of resonant excitation

- magnetic nanostructure “speckles” only seen on resonance



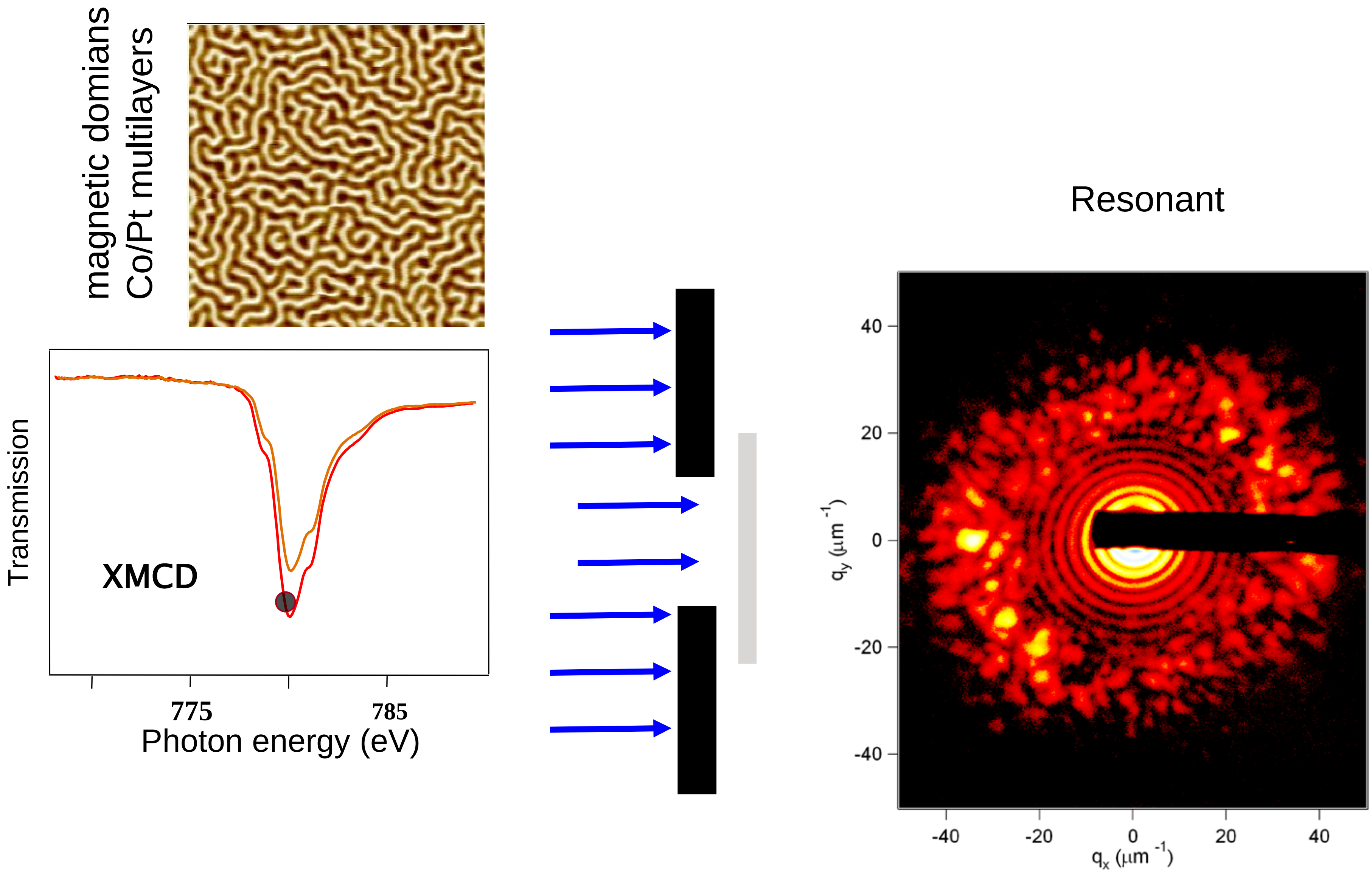






The importance of resonant excitation

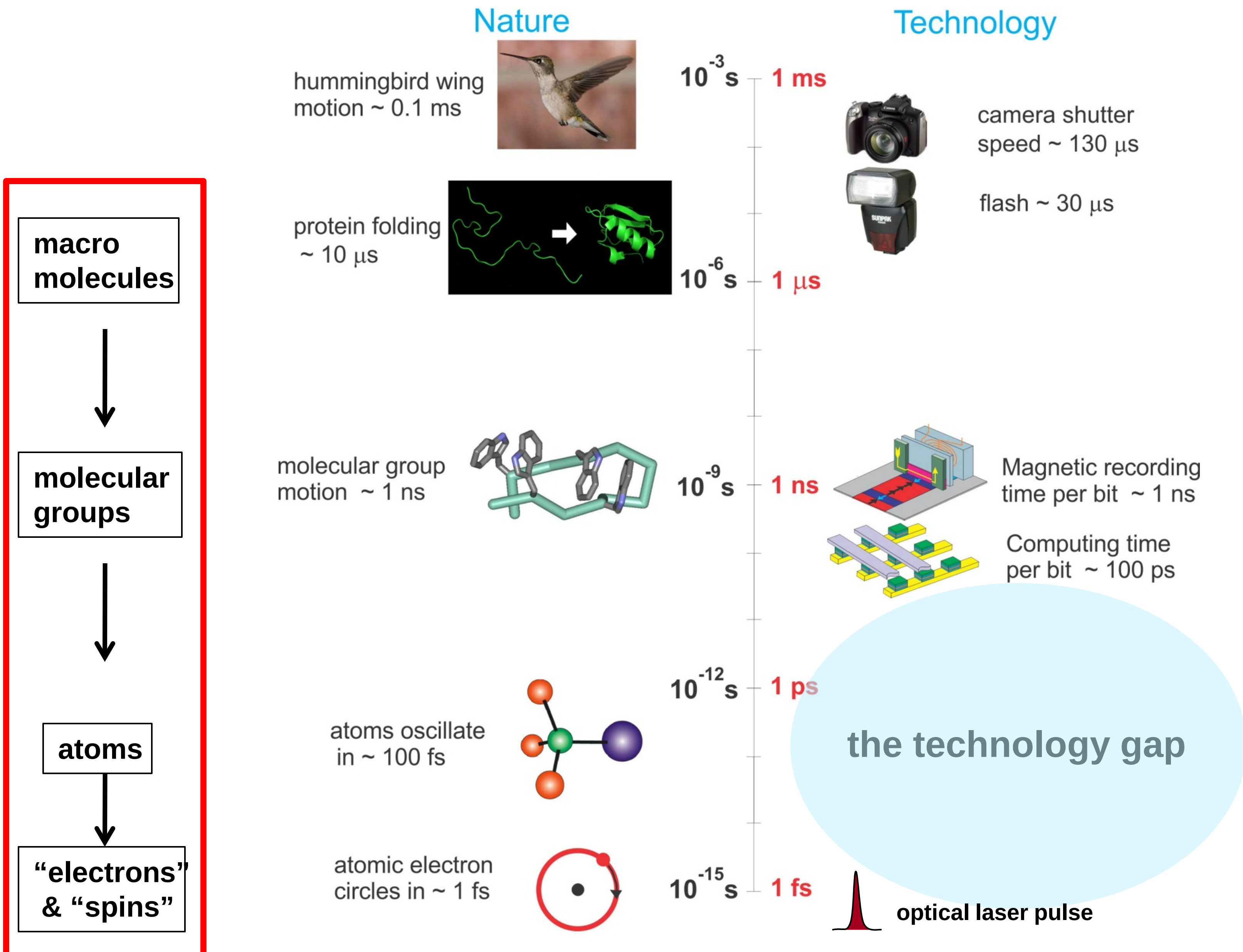
- magnetic nanostructure “speckles” only seen on resonance



- **Ultrafast Science**

  photons in ultrashort pulse

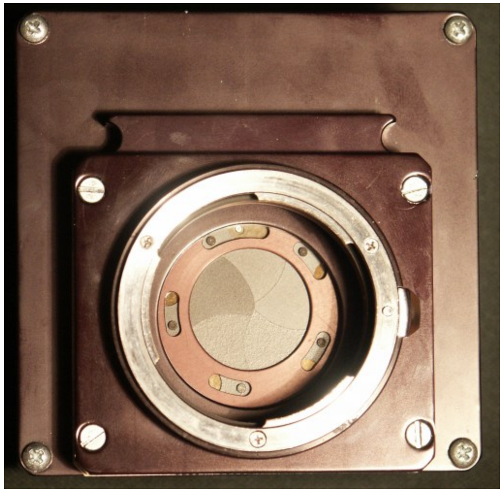
The speed of things – the smaller the faster



Characteristic speeds of atoms and electrons

• Atoms :	speed of sound:	1 nm / 1 ps
• Electrons:	Fermi velocity:	1 nm / 1 fs
• Light:	speed of light:	1 nm / 3 as

Learn from fast photography with visible light



Fastest camera has shutter speed of 0.2 ms

- hummingbird has blurry wings
- picture typically dark because exposure is too short

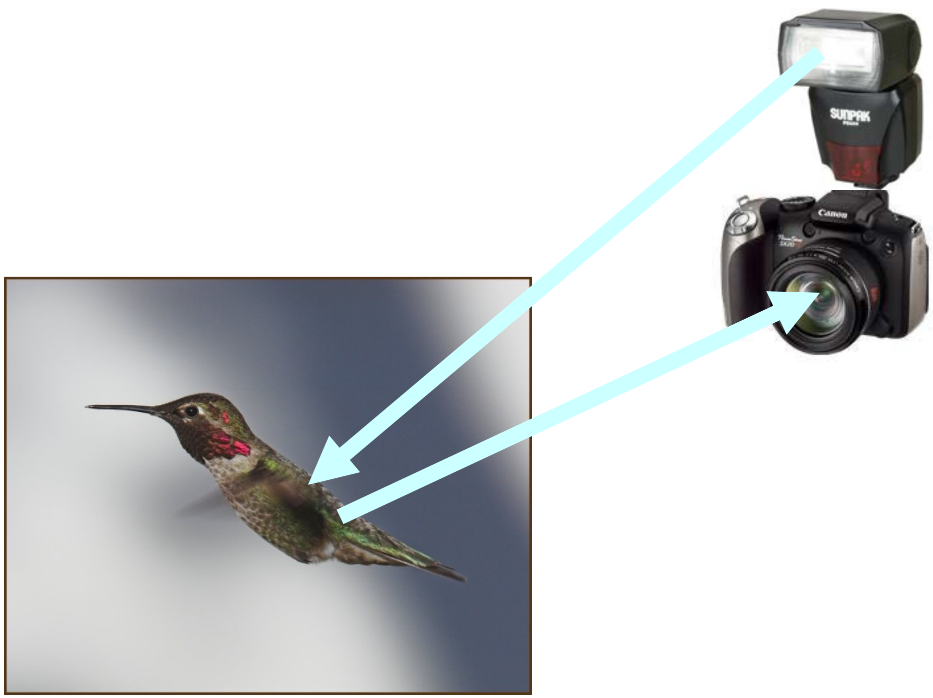
The trick to recording ultrafast pictures

light flash duration and intensity determines picture quality

The trick to recording ultrafast pictures

- Use a bright flash, faster than existing shutter speed
- Capture bright reflected light flash with camera

  leave shutter open, flash light is stronger than background light



ultrafast flash

20 µs

shutter speed

200 µs

light flash duration and intensity determines picture quality

Elements of nanoscale movies

- **X-rays**

  for atomic resolution

- **ultrafast flash**

  to overcome camera speed problems

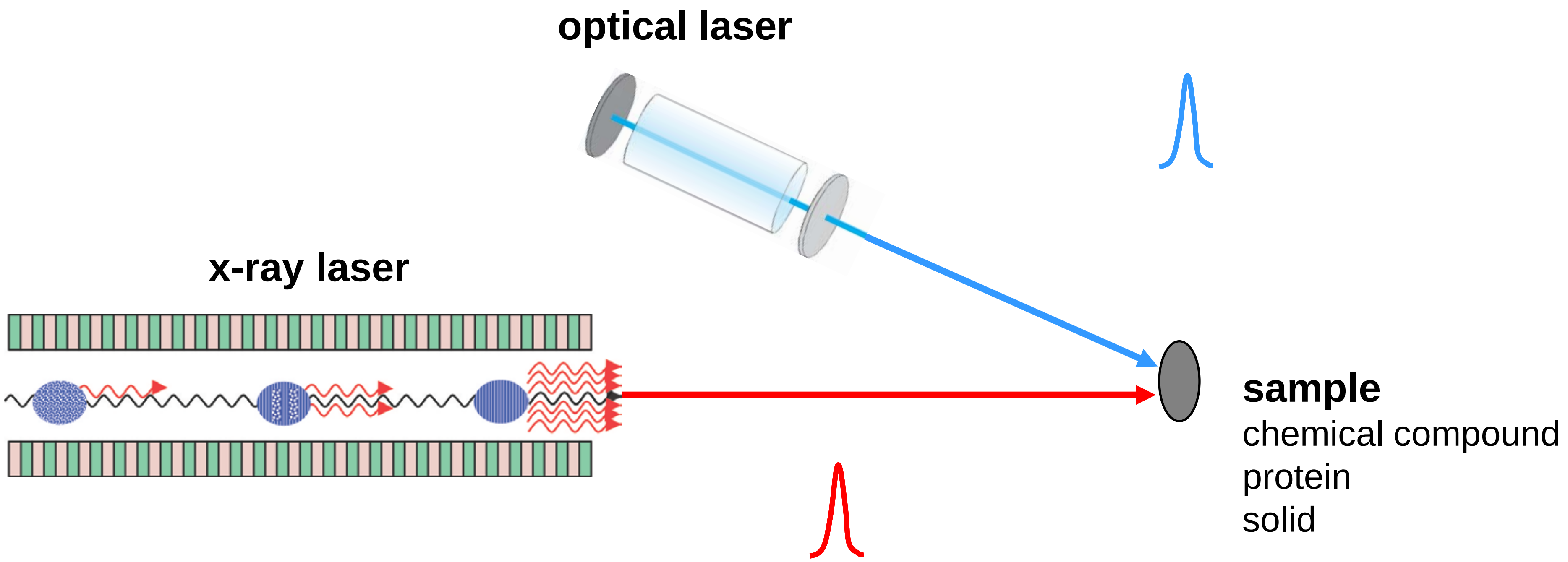
- **ultrabright flash**

  to overcome intensity problem

X-FELs

In structural biology, fast & intense pulses beat atomic motion and damage

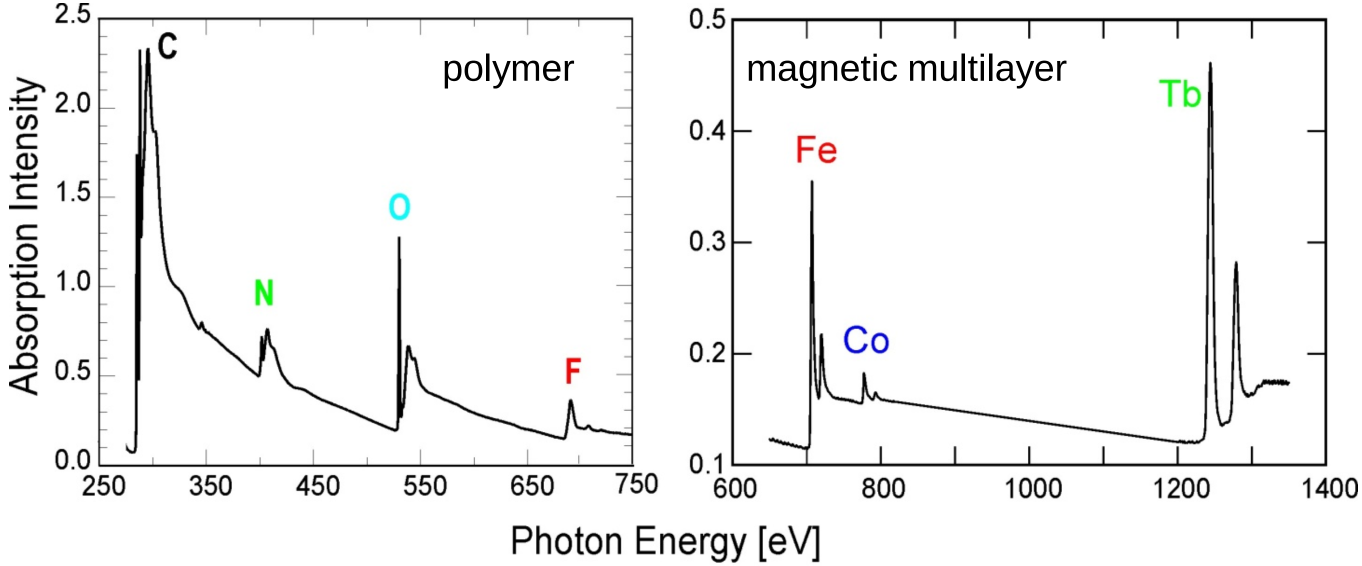
The arrangement for most ultrafast studies – “pump-probe”

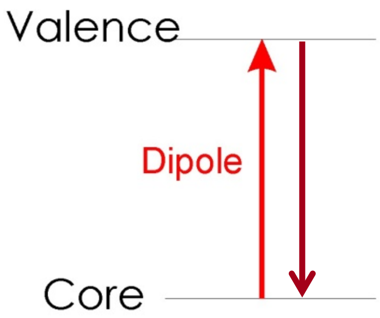


- **Non-linear x-ray science**

Laterally and longitudinally coherent photons

The goal is to control resonant transitions in x-region



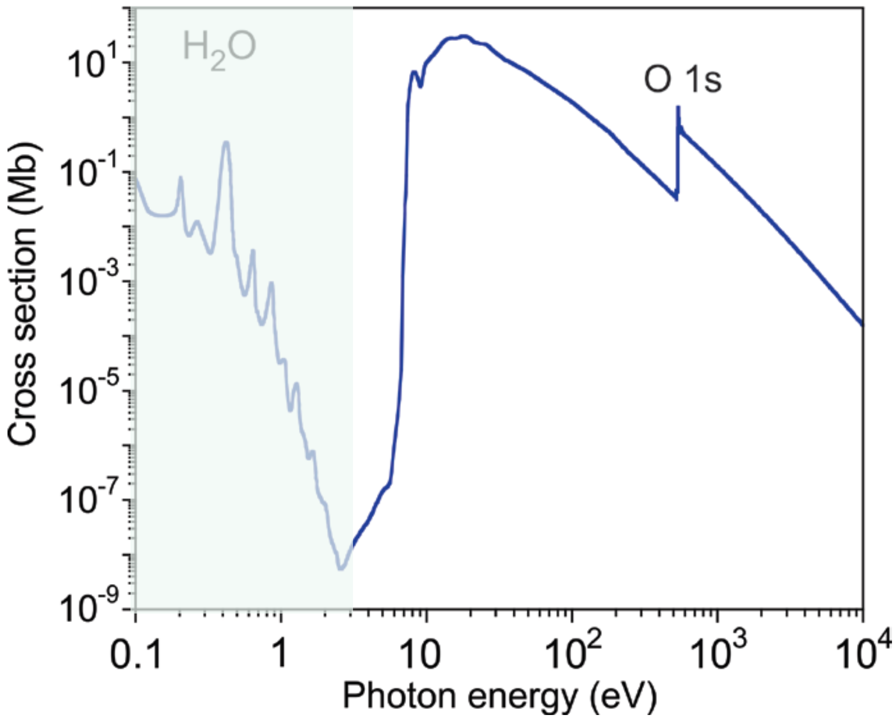


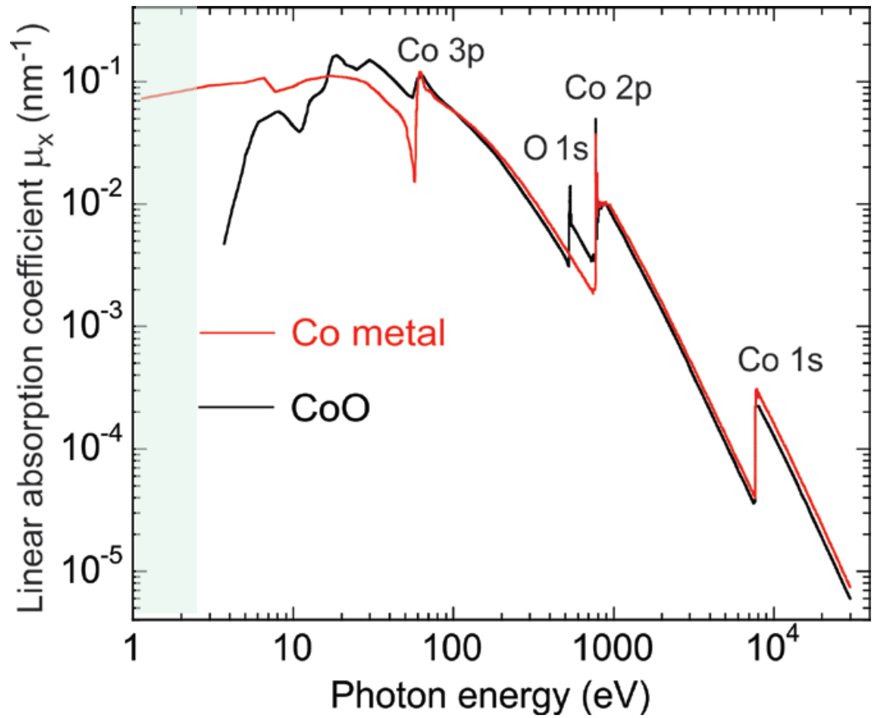
Molecules, Polymers: p-orbitals
Transition metals : d-orbitals
Rare Earths: f-orbitals

1s K-edge
2p L edge
3d M edge

Element specificity, Chemical specificity, Valence properties

X-ray response is simpler than optical response





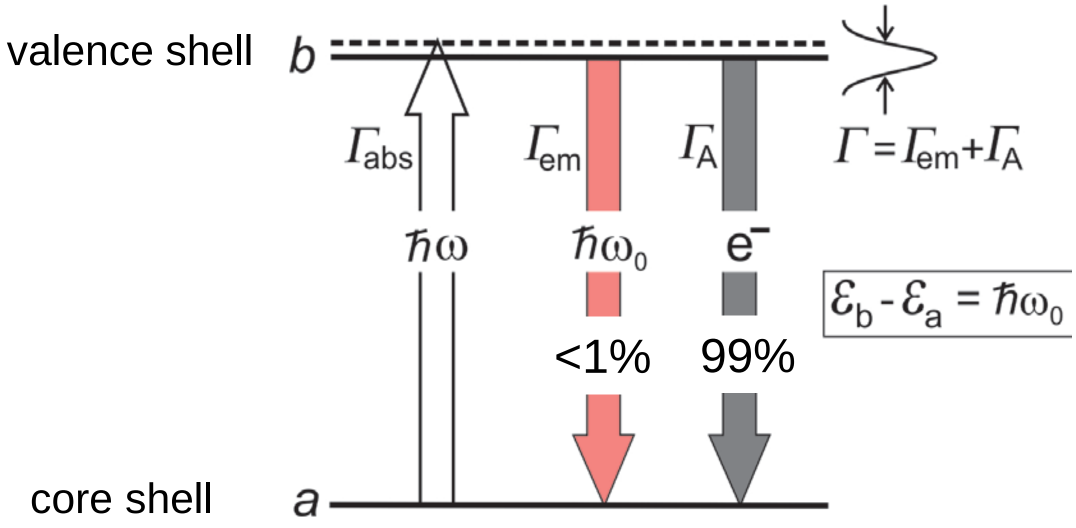
Tune to absorption resonance – strong transitions

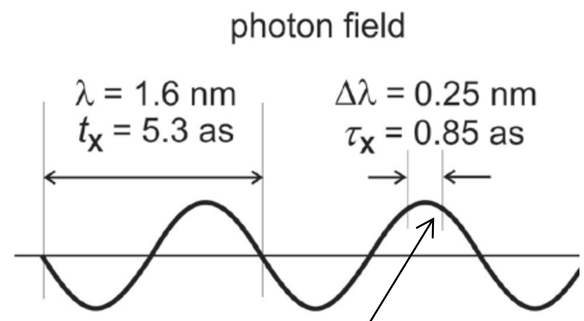
Soft x-ray resonant cross sections similar to optical region

first experiments:

Rohringer *et al*, Nature, **481**, 488 (2012), Beye *et al.* Nature, **501**, 191 (2013)

X-Rays in versus x-rays out: the soft x-ray problem



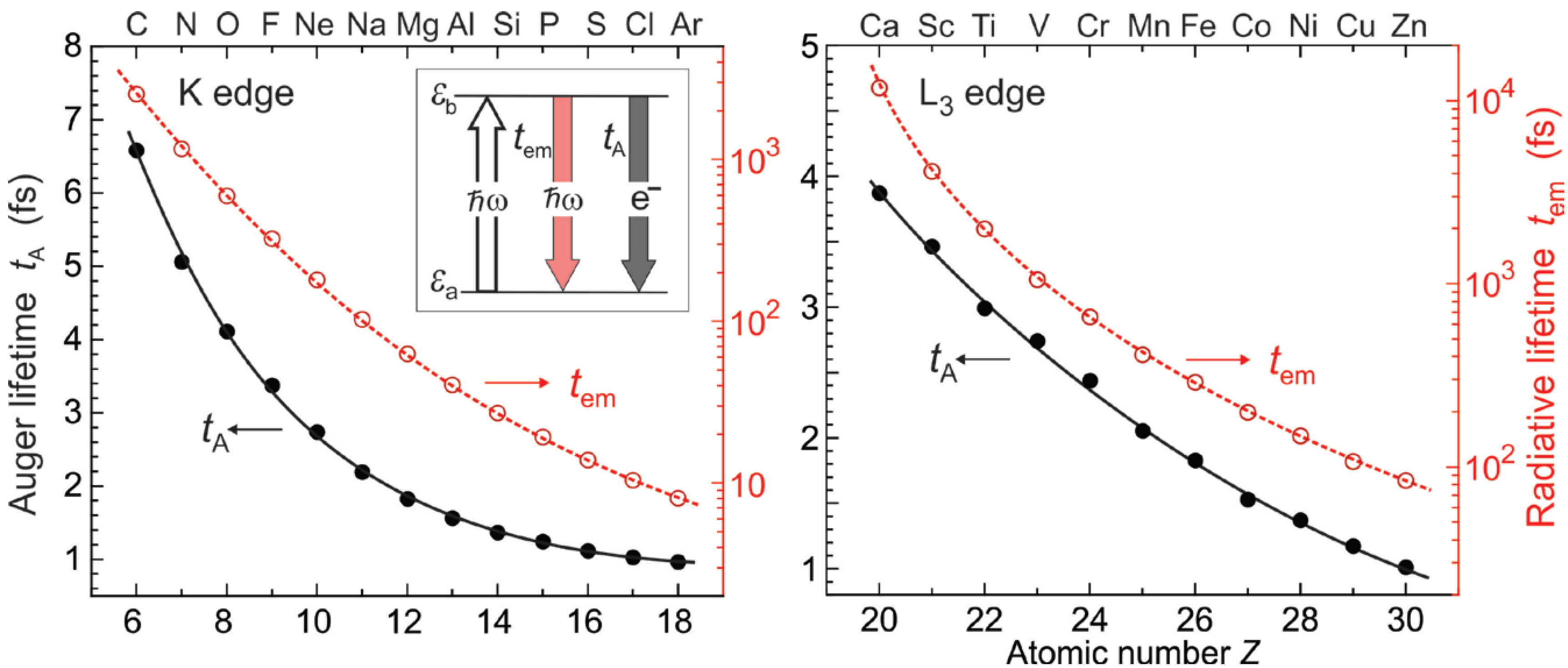


- **Excitation** time
- spontaneous decay via **Auger** or **x-ray emission**

Auger decay completely dominates

X-ray decay very weak

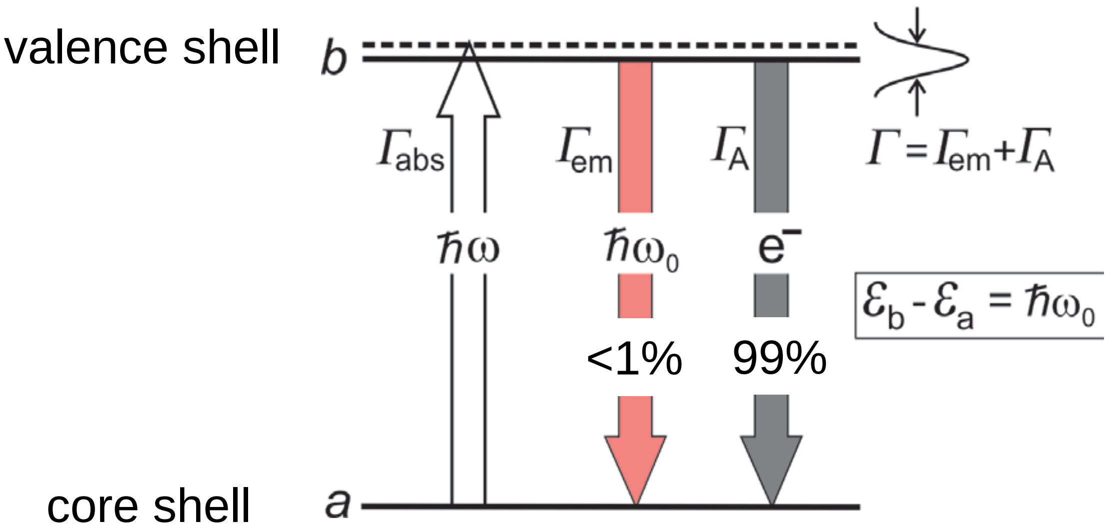
Excited state decay times for important elements



In soft x-ray region Auger decay completely dominates (is faster)

- it occurs on fs timescale
- x-ray emission is much slower
- this is a problem for resonant inelastic x-ray scattering (RIXS)

How can we get more photons out ?



Soft x-ray example

Auger decay completely dominates

X-ray decay very weak

$$\mathcal{W}_{b\to a}^{\mathrm{em}} = \frac{\Gamma_x}{\hbar}(1+n_\Gamma)$$

$$= \frac{\Gamma_x}{\hbar}\frac{\epsilon_0\lambda^3}{\pi^2\Gamma}\left(|E_{\mathrm{ZP}}^{\Gamma}|^2 + \underbrace{n_\Gamma\,|E_{\mathrm{ZP}}^{\Gamma}|^2}_{|E_{\mathrm{in}}^{\Gamma}|^2}\right)$$

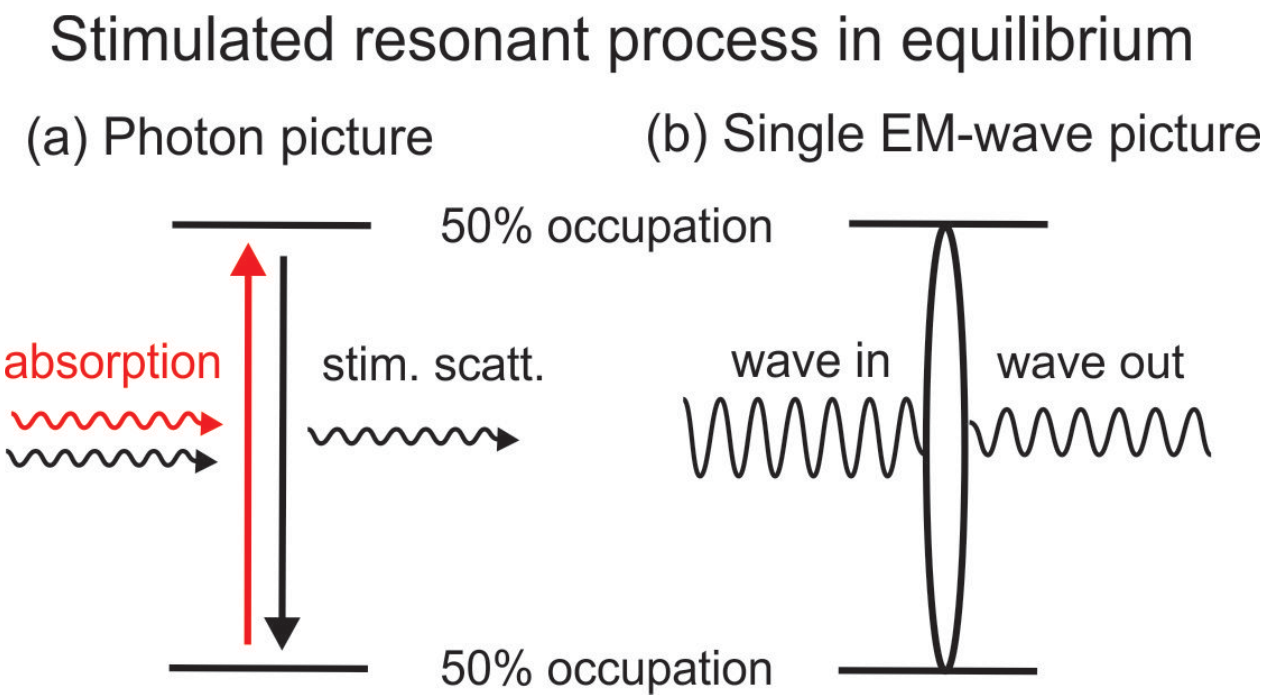
$$|E_{\mathrm{ZP}}^{\Gamma}|^2 = \frac{\pi^2\Gamma}{\epsilon_0\lambda^3}$$

spontaneous= zero-point field

stimulated= incident field

When the incident field becomes stronger than the zero-point field stimulated decays begin to dominate over spontaneous decays

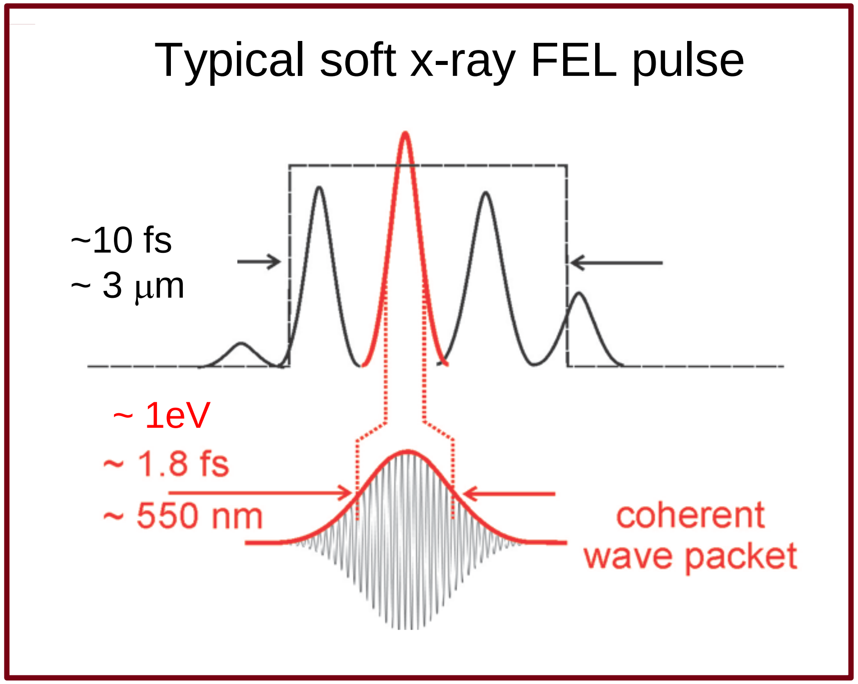
Stimulation requires many photons in the coherence volume



More than one photon at-a-time = strong classical field

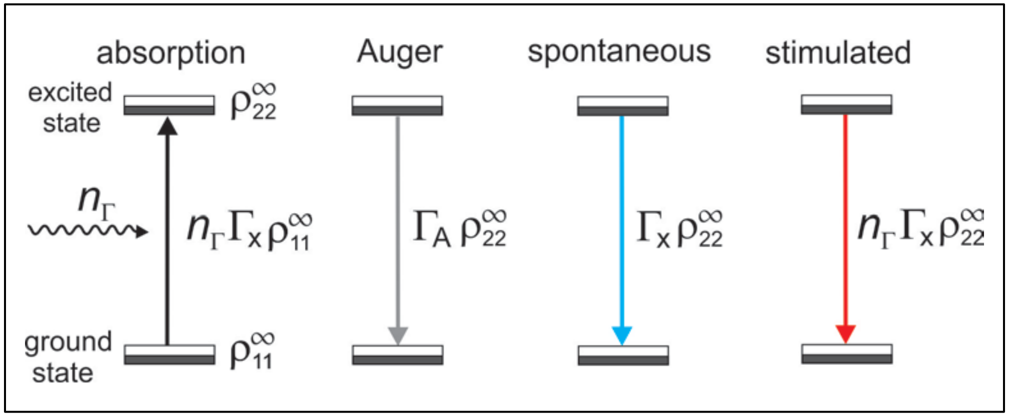
offers complete "up-down" control

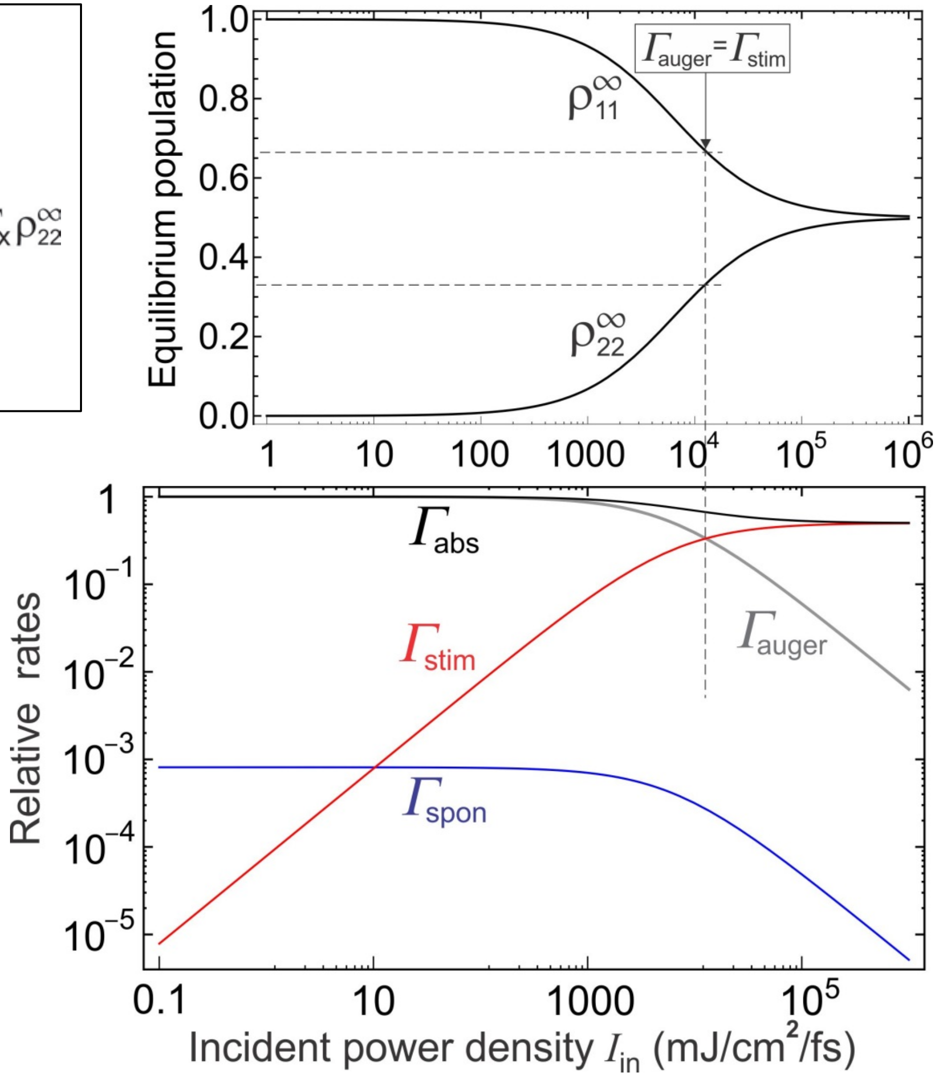
Want transform limited X-FEL pulse – single spike



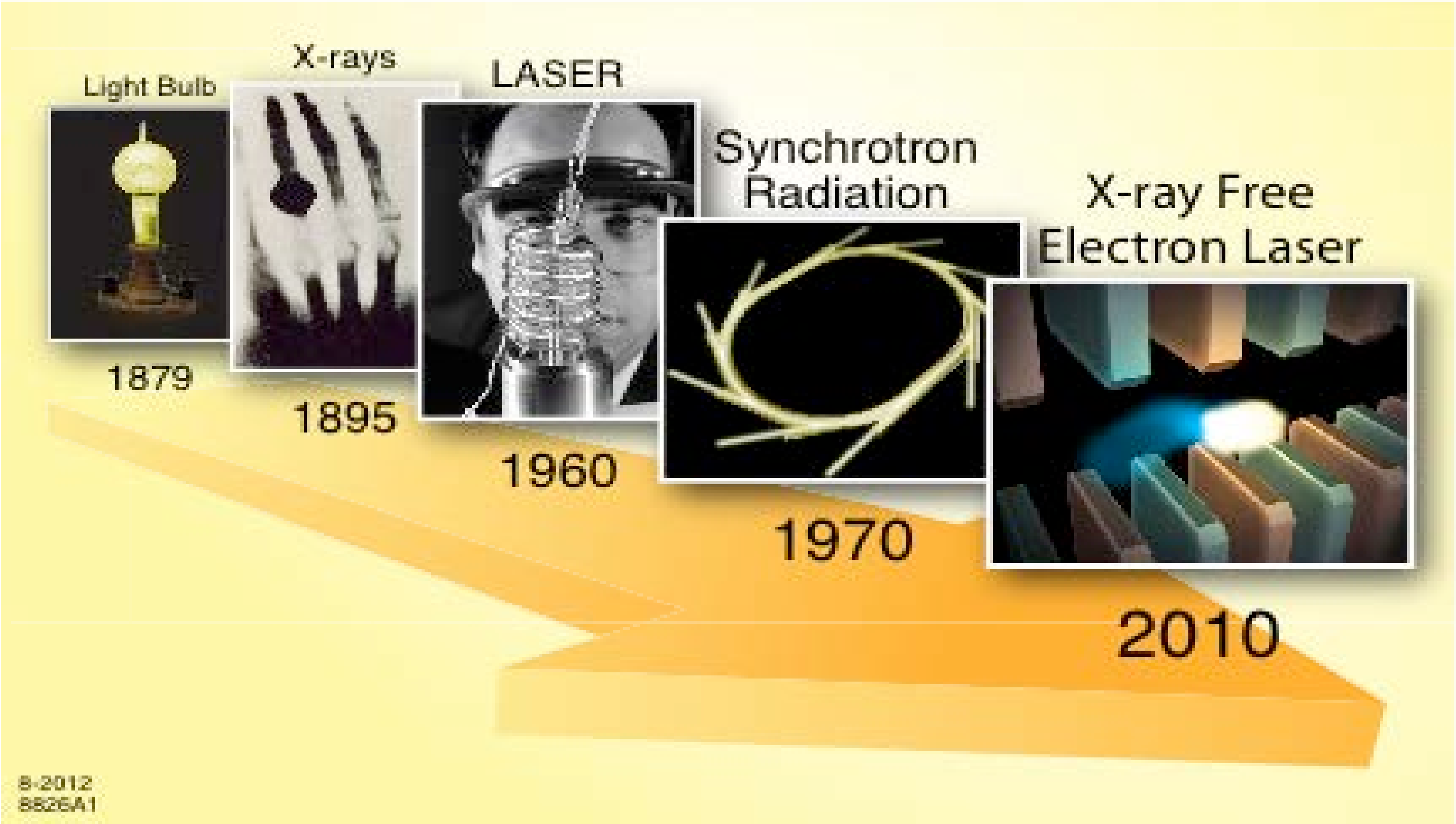
10^9 photons per spike (eV)

Population and transition rates of atoms as function of incident intensity





Light revolutions



The end