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Swiss Federal Institute of Technology Zurich

Coherent spin and lattice dynamics studied with femtosecond x-ray diffraction

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Outline

- Why x-rays?
- Principles of scattering and diffraction
- Sources of short x-ray pulses
- Time-resolved scattering
- Time-resolved diffraction

Why x-rays?

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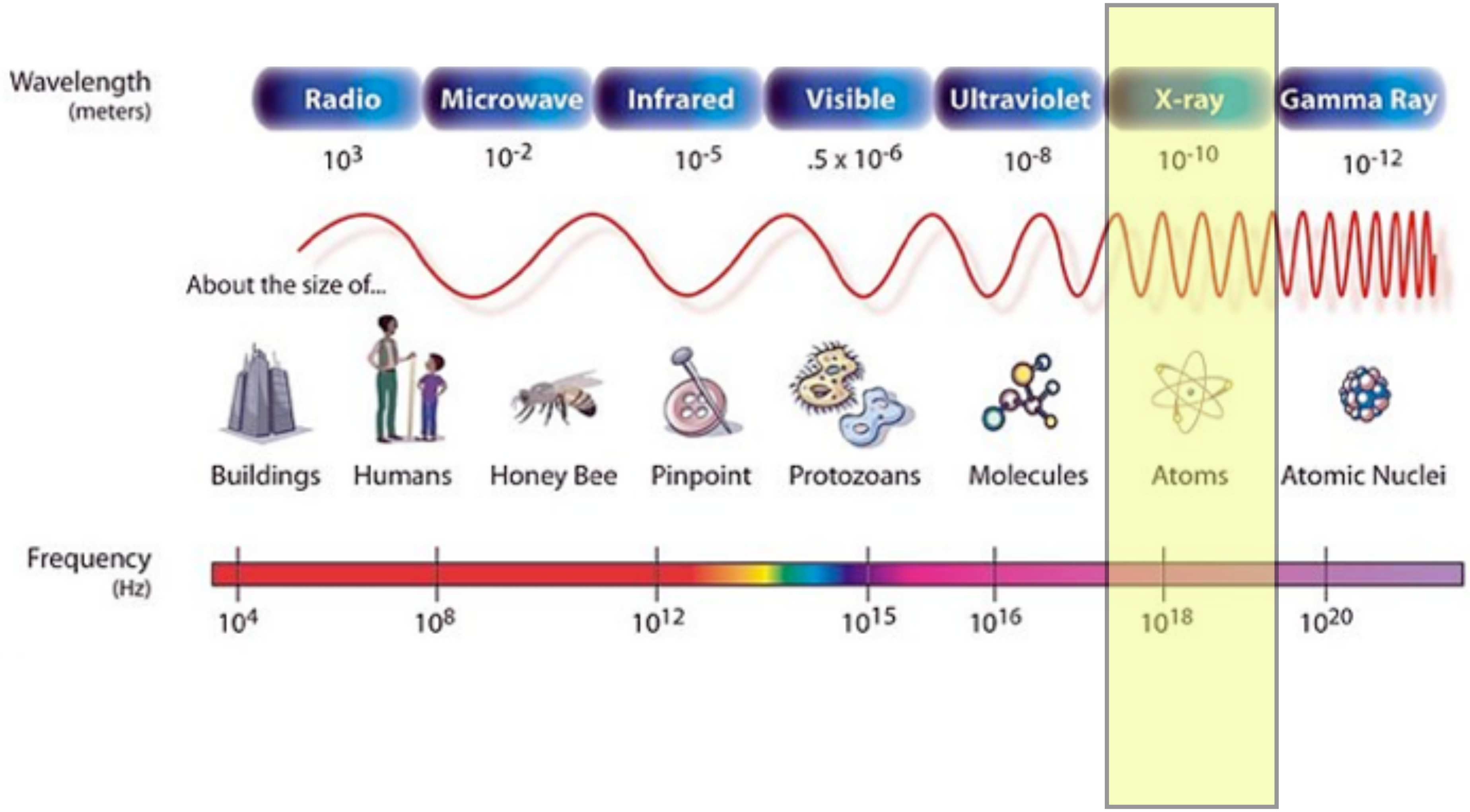
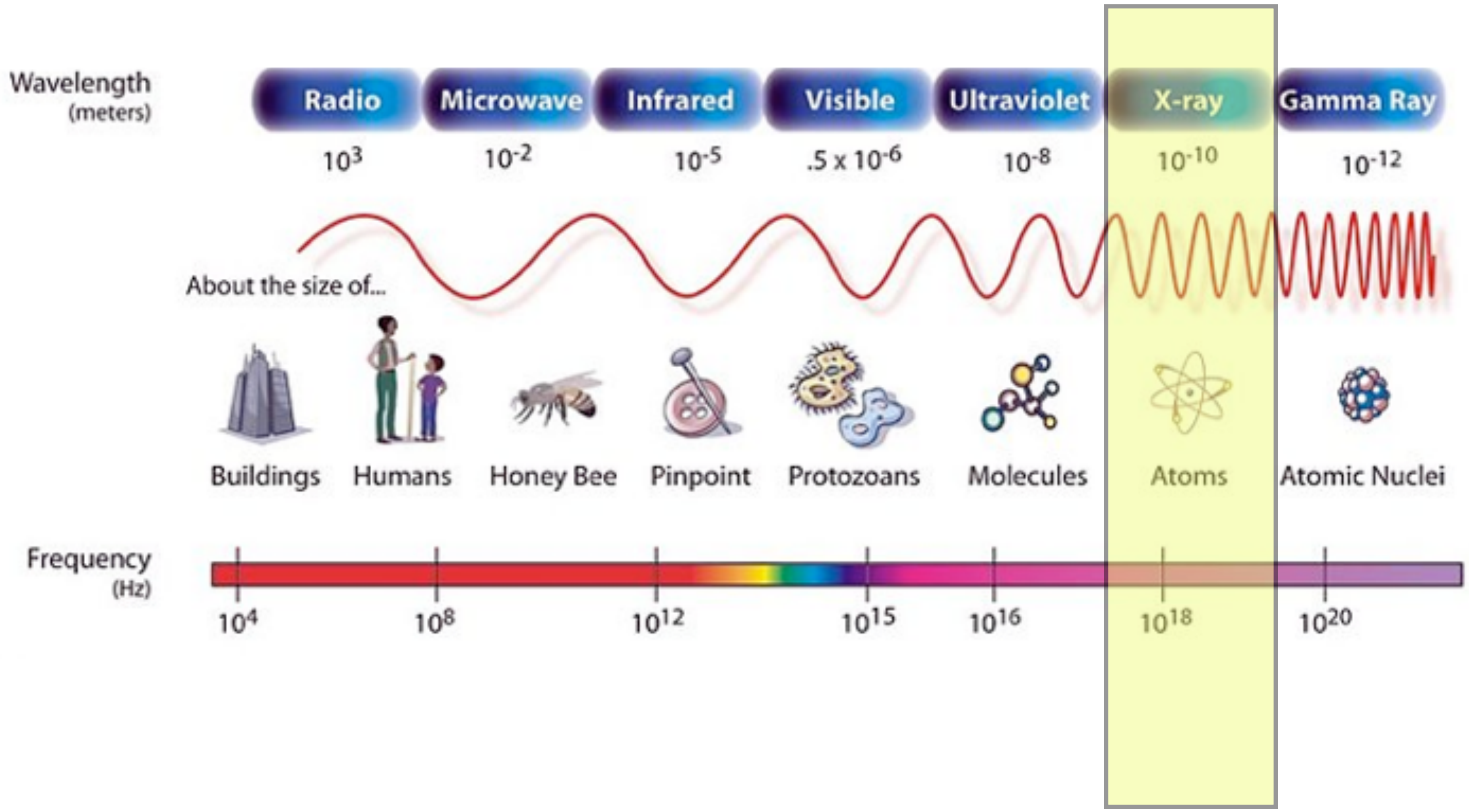
X-ray region of spectrum



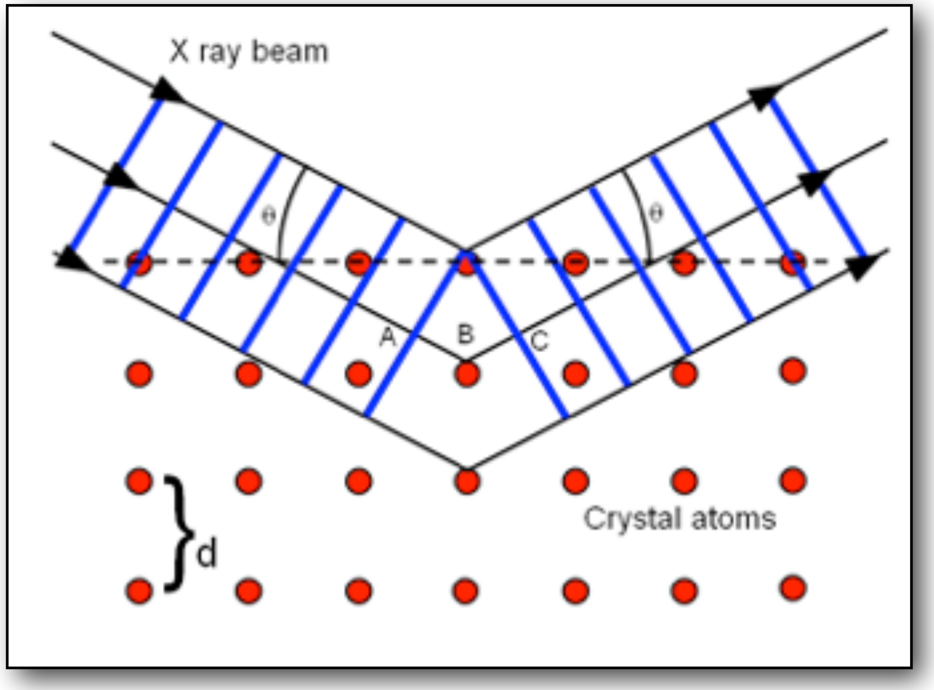
- Wavelength: 0.1-100 Å
- Photon energy: 100 eV - 100 keV

X-ray region of spectrum



- Wavelength: 0.1-100 Å
- Photon energy: 100 eV - 100 keV

X-ray scattering / diffraction



- Use interference of scattered radiation to infer electronic charge distribution, atomic structure
- Measure "cuts" of Fourier Transform space

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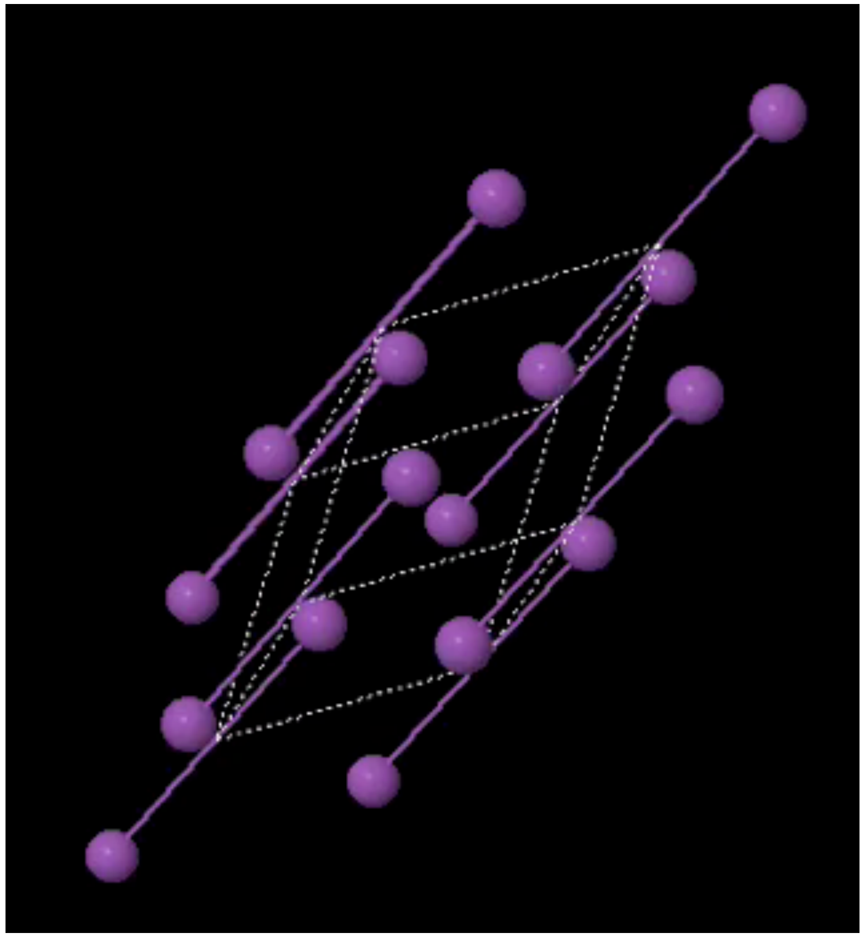
Ultrafast + scattering / diffraction

Vibrational dynamics



- Speed of sound (condensed media) ~ 2000 m / s
- Typical interatomic spacing ~ 1 Å
- $\Delta t \sim (1 \times 10^{-10}$ m)/(2000 m/s) = 50 fs

(tomorrow: spin and valence dynamics)

Principles of scattering and diffraction

Interactions: EM radiation & matter

- Hamiltonian for a free particle with mass m and charge q
  (non-relativistic)

$$H = \frac{|\mathbf{P} - q\mathbf{A}/c|^2}{2m} = \frac{|\mathbf{P}|^2}{2m} - \frac{q}{mc}(\mathbf{A}\cdot\mathbf{P} + \mathbf{P}\cdot\mathbf{A}) + \frac{q^2\,|\mathbf{A}|^2}{2mc^2}$$

KE term

inelastic (dipole, quad, etc.)

Thomson scattering

$$\mathbf{A} = \hat{\epsilon}\sqrt{\frac{\hbar}{2\epsilon_0 V\omega}}\left(a_k^{\dagger} e^{-i\mathbf{k}\cdot\mathbf{r}} + a_k e^{i\mathbf{k}\cdot\mathbf{r}}\right)$$

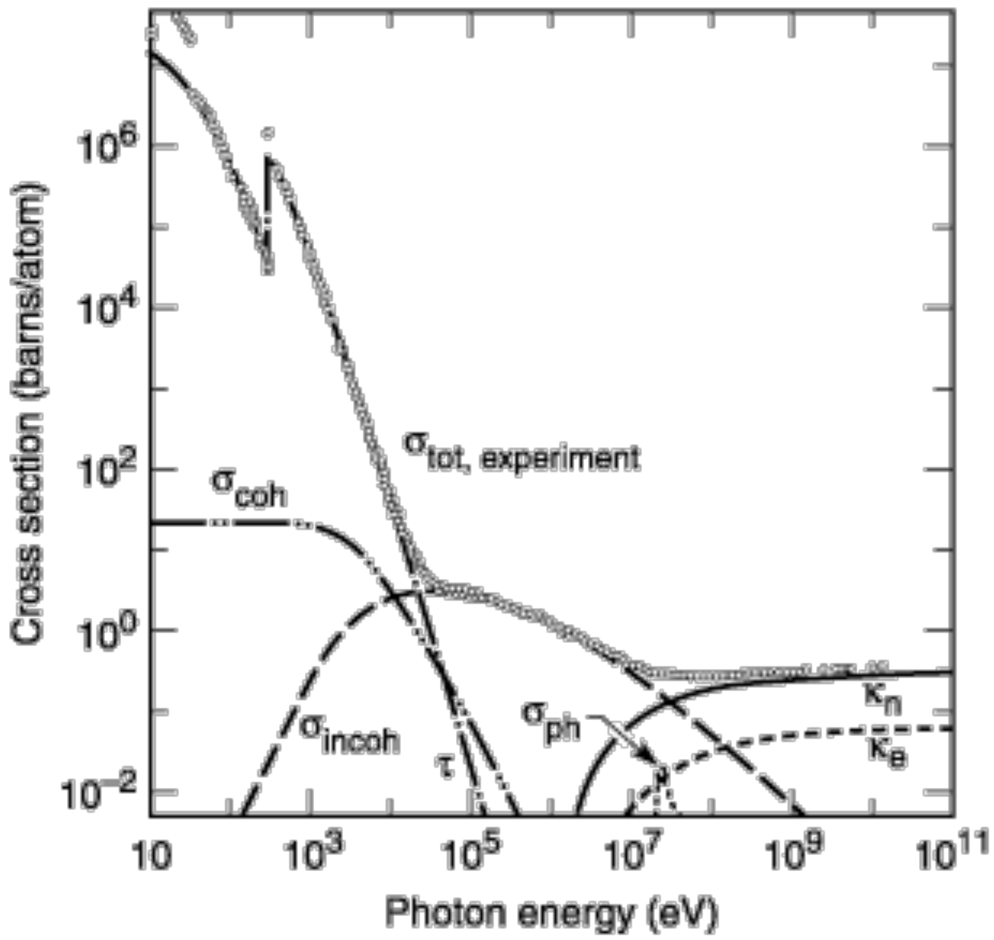
photon creation

photon annihilation

Interactions: EM radiation & matter

- Per atom elastic scattering weak, ~ 10^{-26} m^2
- Typically weaker than incoherent contributions…but maintains phase coherence

Carbon atom



J. H. Hubbell, H. A. Gimm, I. , “Pair, Triplet, and Total Atomic Cross Sections (and Mass Attenuation Coefficients) for 1 MeV–100 GeV Photons in Elements Z = 1 to 100,” J. Phys. Chem. Ref. Data 9, 1023 (1980).

Coherence

- To use interference as a probe, coherence essential
- What is coherence?

$$\mu(\mathbf{r}_1, \mathbf{r}_2) = \frac{\langle E(\mathbf{r}_1, t) E(\mathbf{r}_2, t)^* \rangle}{\sqrt{\langle |E(\mathbf{r}_1, t)|^2 \rangle \, \langle |E(\mathbf{r}_2, t)|^2 \rangle}}$$

Spatial coherence: "Complex coherence factor"

"Incoherent"

$\mu \rightarrow 0$

"Coherent"

$\mu \rightarrow 1$

Ability of waves at different locations to interfere

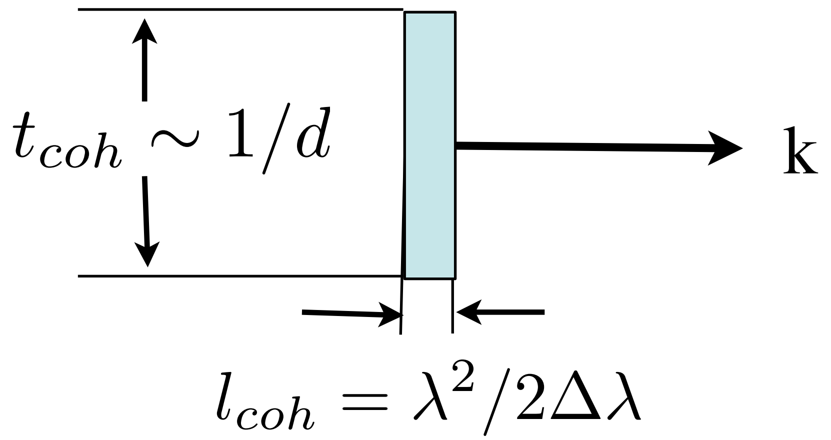
Coherence

$$\mu(\mathbf{r}_1,\mathbf{r}_2) = \frac{\langle E(\mathbf{r}_1,t)E(\mathbf{r}_2,t)^*\rangle}{\sqrt{\langle|E(\mathbf{r}_1,t)|^2\rangle\,\langle|E(\mathbf{r}_2,t)|^2\rangle}}$$

- Coherence volume: volume of space such that

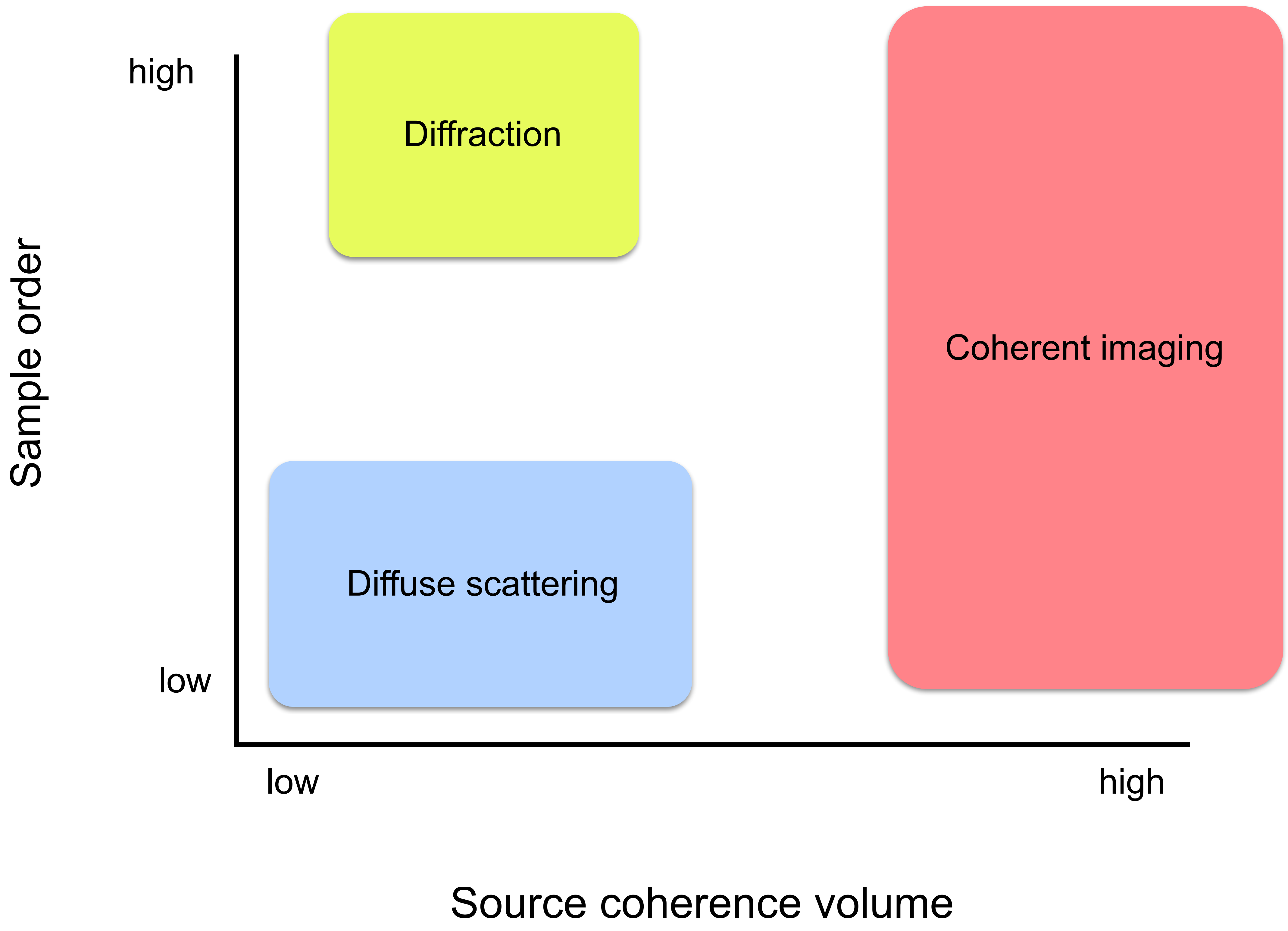
$$|\mu(0,\mathbf{r})| > 1/2$$

- Usually divided into “longitudinal” and “transverse”



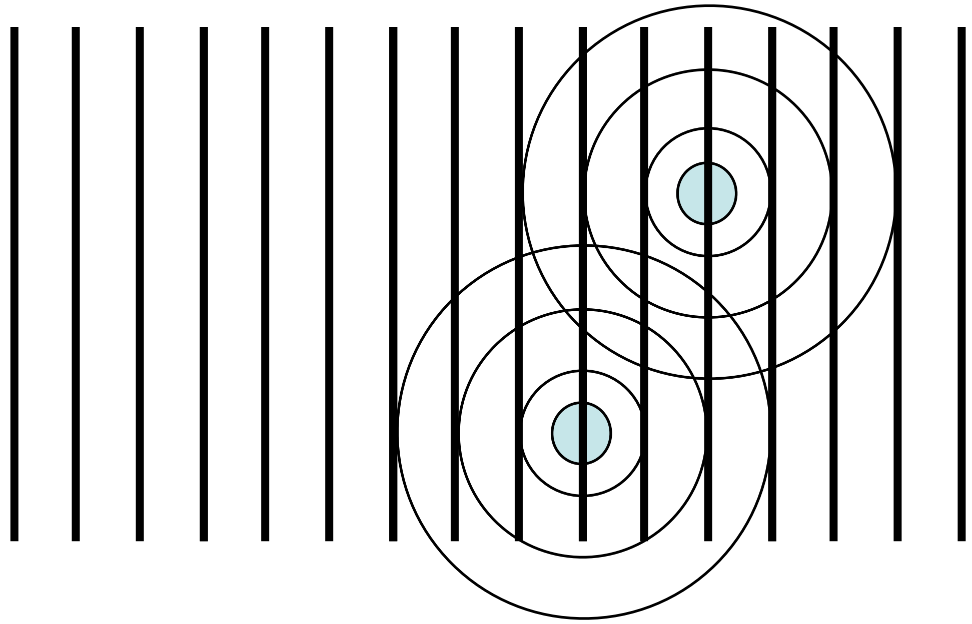
(d is an apparent source size)

Coherence and order



Diffuse scattering

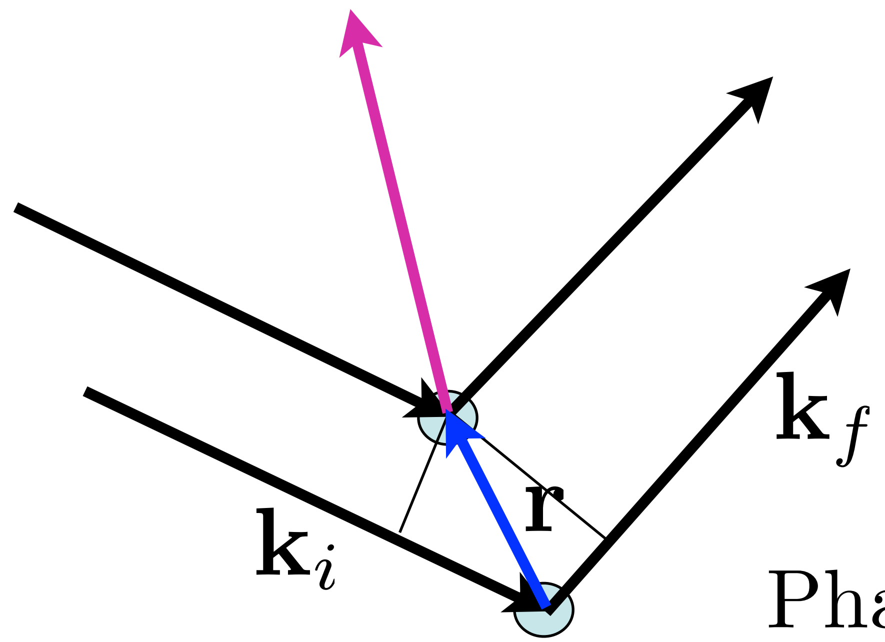
- Coherence volume small compared with illuminated sample volume
- Coherence volume large compared to interatomic spacings



Look at the *average* distances between atoms *within* the coherence volume dimensions

Diffuse scattering

Structure factor



$$\text{Phase difference} = \mathbf{r}\cdot\mathbf{k}_f - \mathbf{r}\cdot\mathbf{k}_i$$

$$\mathbf{Q} = \mathbf{k}_f - \mathbf{k}_i$$

$$F(Q) = \sum_j f_j e^{i\mathbf{Q}\cdot\tau_j}$$

... a Fourier transform

$$\frac{I_s}{I_0} = |F(\mathbf{Q})|^2$$

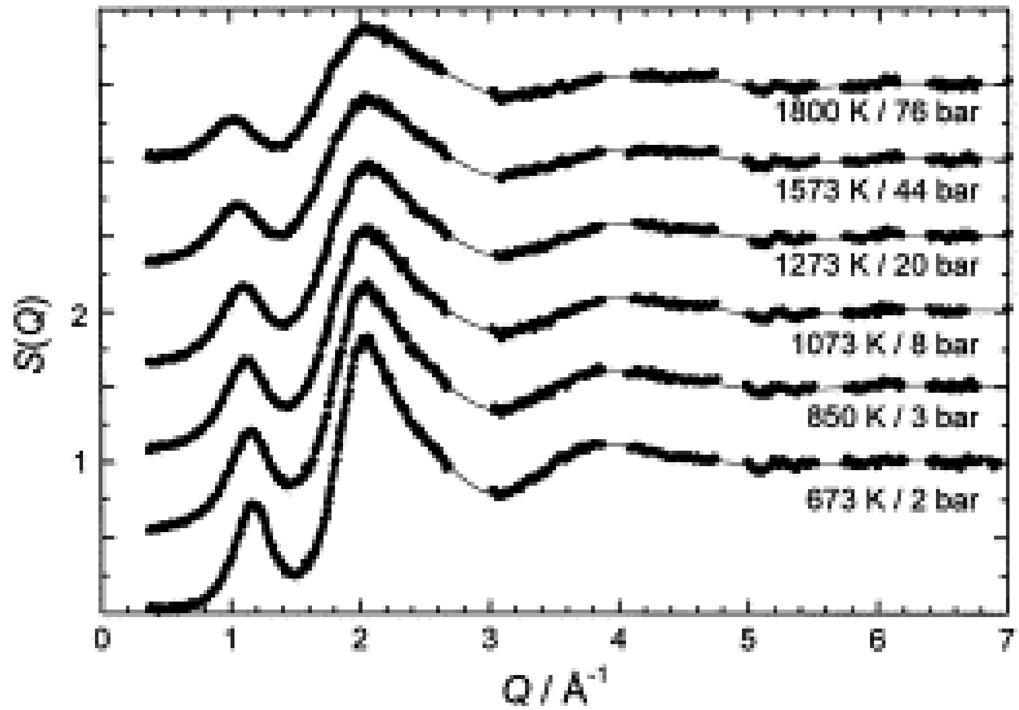
Diffuse scattering

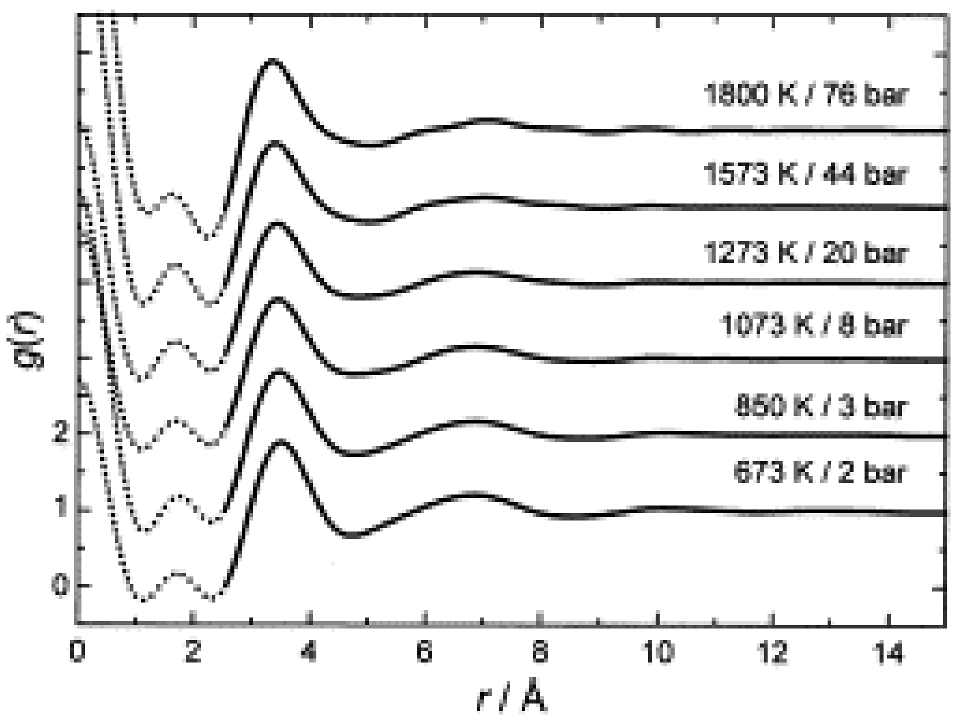
- How to get structure? (assume orientational disorder)

$$S(Q) = \sum_k N_k f_k(Q)^2 \sum_{l \neq k} N_k f_k(Q) N_l f_l(Q) \int 4\pi r^2 \rho_0 (g_{kl}(r) - 1) \frac{sin(Qr)}{Qr}$$

Pair correlation function

liquid K-Bi





[example from: Hochgesand, Physica B 276-278, 425 (2000)]

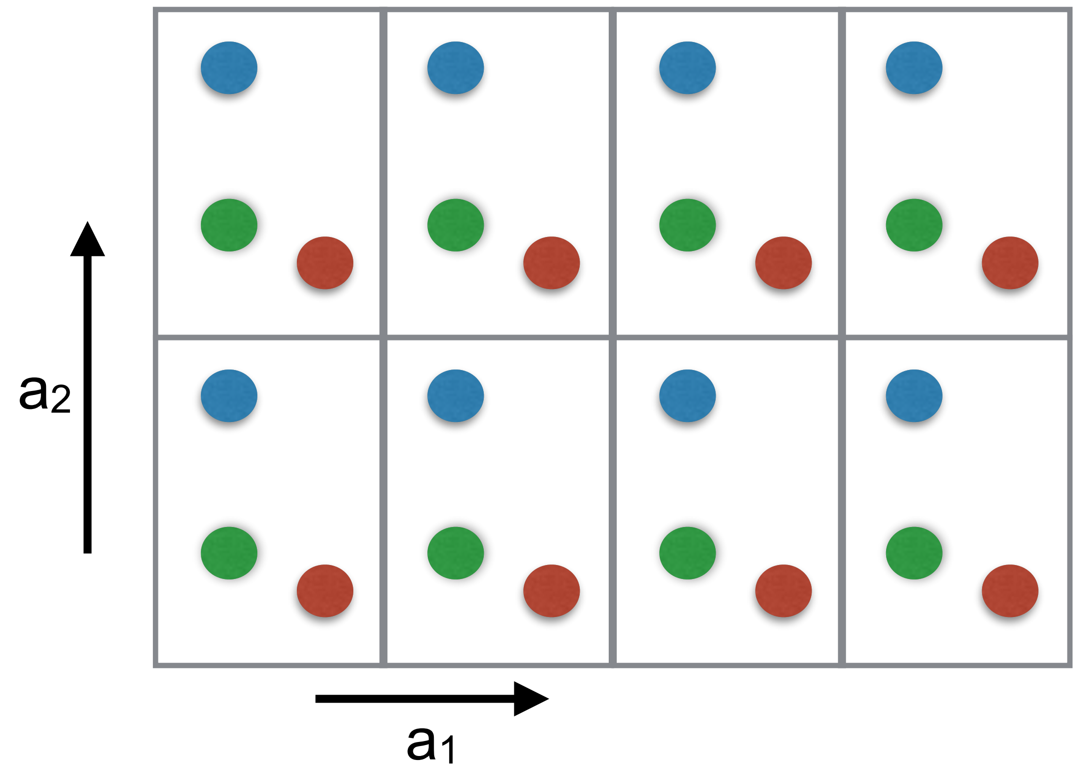
Diffuse scattering

- Advantages:
  - Given a structural model, easy to calculate diffraction
  - Selective, only sensitive to structure

- Disadvantages:
  - Requires a model (not invertible)
  - Interaction with all electrons in sample (solvents)
  - In normal use, just the pair correlation function (no higher orders)

Diffraction: crystals

- For now, we discuss systems with true long-range order (no quasicrystals or incommensurate superlattices)
- Unit cell: arrangement of atoms (basis)
- Vectors t describe translational symmetry, can be used to "build" the crystal from a unit cell

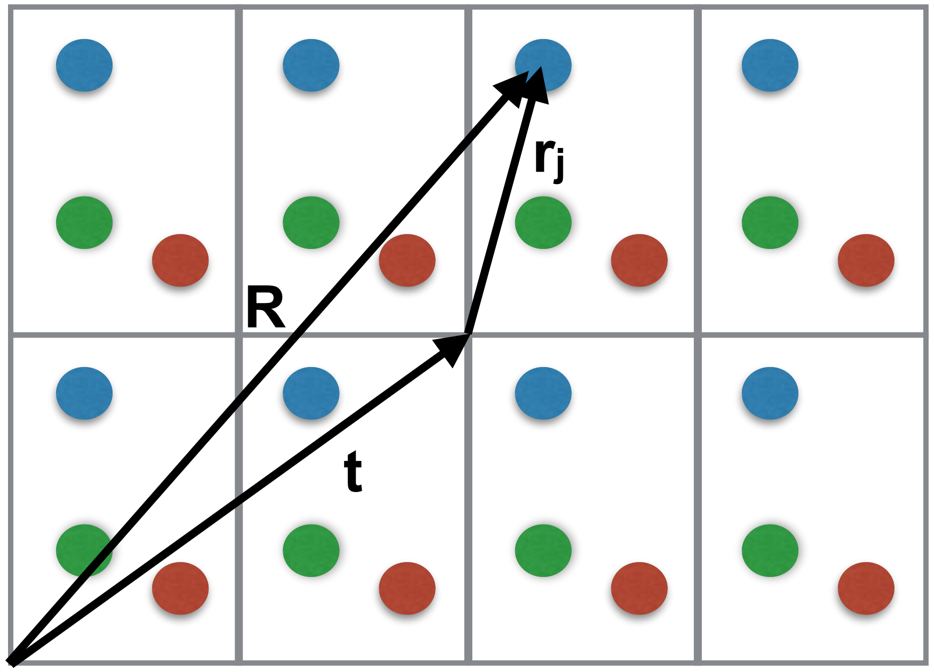


$$\mathbf{t} = n\mathbf{a}_1 + m\mathbf{a}_2$$

Diffraction: crystals

$$\frac{I_s}{I_0} = |F(\mathbf{Q})|^2 \qquad F(\mathbf{Q}) = \sum_{\mathbf{R}} f_R e^{i\mathbf{Q}\cdot\mathbf{R}}$$

$$F(\mathbf{Q}) = \sum_{\mathbf{t}} \left(\sum_j f_j e^{i\mathbf{Q}\cdot\mathbf{r}_j} \right) e^{i\mathbf{Q}\cdot\mathbf{t}} = \sum_{\mathbf{t}} F_c(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{t}}$$



$$F_c(\mathbf{Q}) = \sum_j f_j e^{i\mathbf{Q}\cdot\mathbf{r}_j}$$ Unit cell structure factor

Diffraction: crystals

$$\frac{I_s}{I_0} = |F(\mathbf{Q})|^2 \qquad F(\mathbf{Q}) = \sum_{\mathbf{t}} F_c(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{t}}$$

- For a large crystal (many unit cells), strong peaks when

$$\mathbf{Q} \cdot \mathbf{t}/2\pi \in I$$

- We call values of **Q** that satisfy this for all **t** *reciprocal lattice vectors* **G**

$$\mathbf{G} = h\mathbf{b}_1 + k\mathbf{b}_2 + l\mathbf{b}_3$$

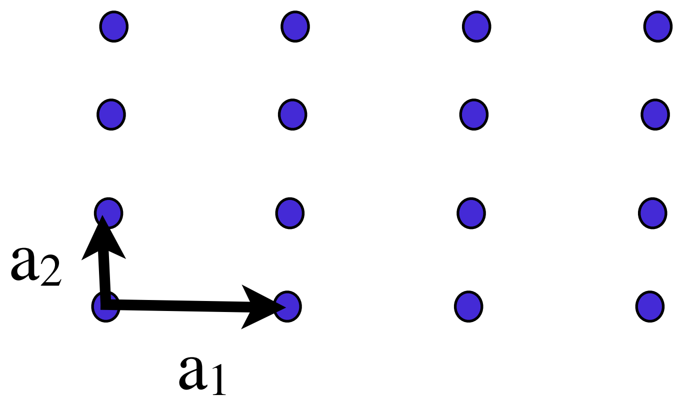
h, k, l integers; b_1, b_2, b_3 reciprocal primitive vectors

Diffraction: crystals

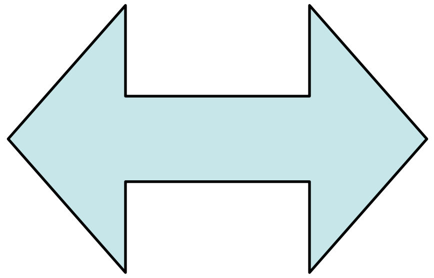
Reciprocal space

2D case (easily generalized)

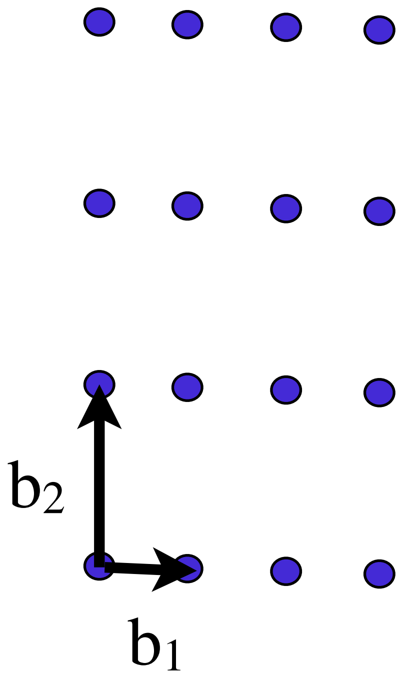
Direct space



$$\mathbf{a}_i = \sum_j a_{ij}\mathbf{x}_j$$



Reciprocal space



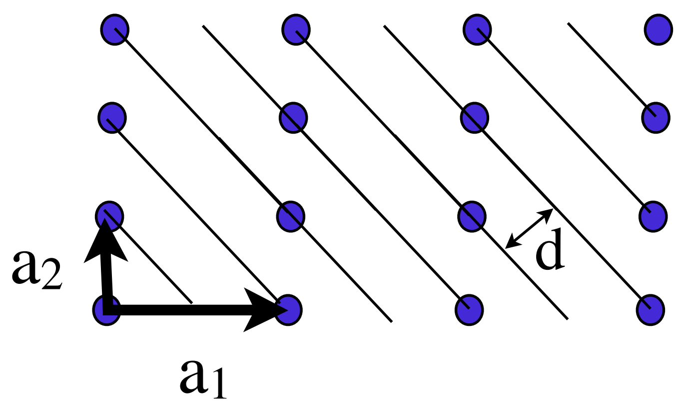
$$\mathbf{b}_i = \sum_j b_{ij}\mathbf{x}_j$$

$$\begin{bmatrix} b_{11} & b_{21} \\ b_{21} & b_{22} \end{bmatrix} = \left(2\pi \begin{bmatrix} a_{11} & a_{21} \\ a_{21} & a_{22} \end{bmatrix}^{-1} \right)^T$$

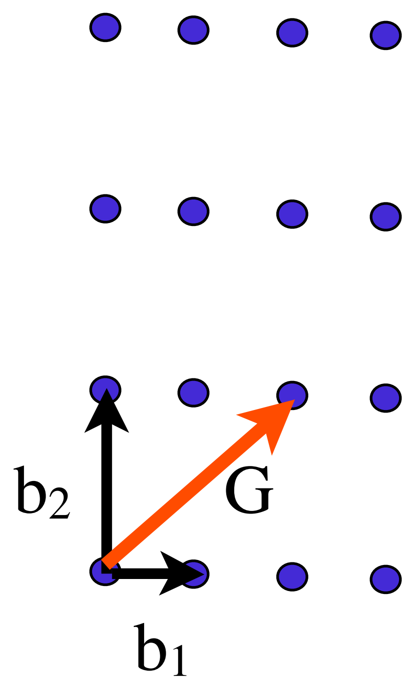
Diffraction: crystals

Reciprocal lattice

Direct space



Reciprocal space



Lattice planes represented by G:

$$\mathbf{G} = h\mathbf{b_1} + k\mathbf{b_2}$$

...where h, k are integers

Direction: orientation of plane

$$|\mathbf{G}| = 2\pi/d$$

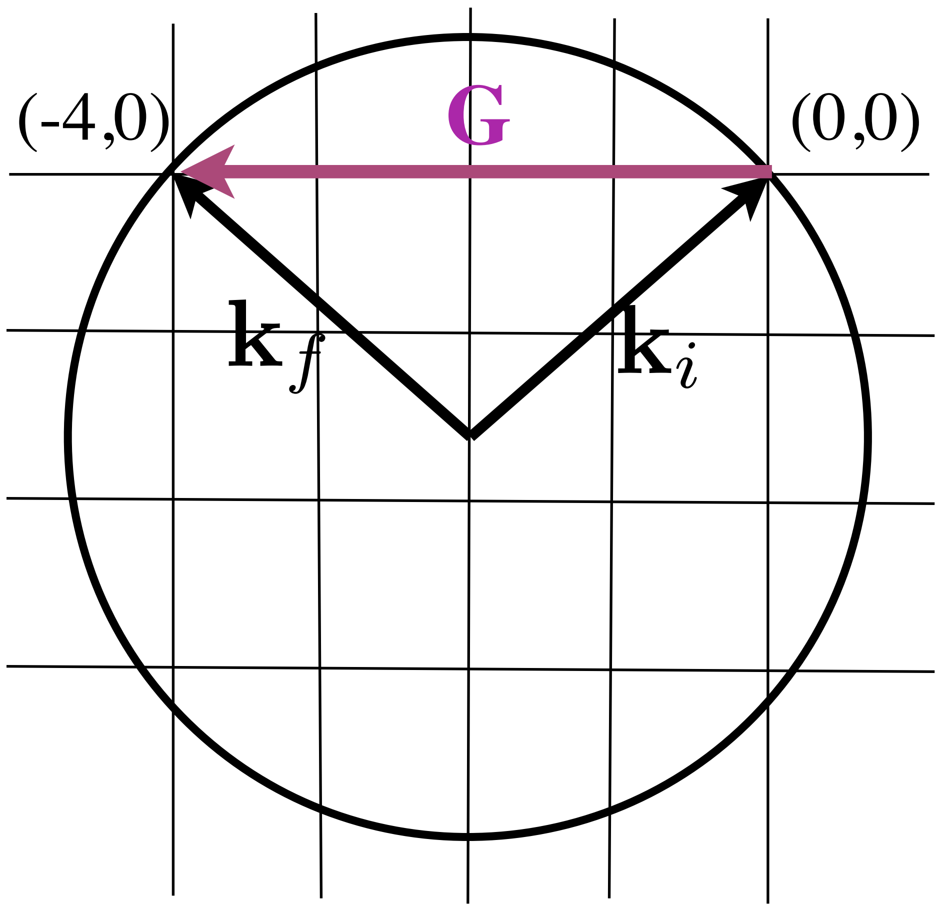
Diffraction: crystals

Ewald sphere (circle)

...A graphical way to predict where in reciprocal space Bragg peaks appear

$$\mathbf{k}_f = \mathbf{k}_i + \mathbf{G}$$

Determined *only* by long range translational order



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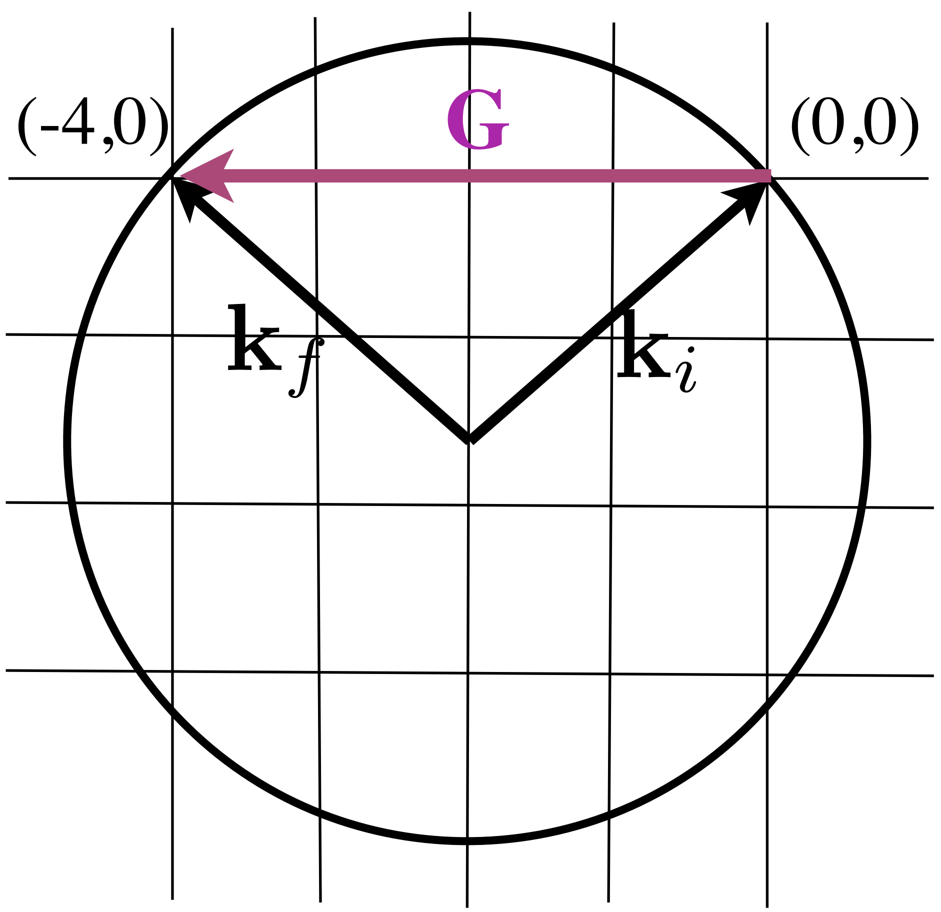
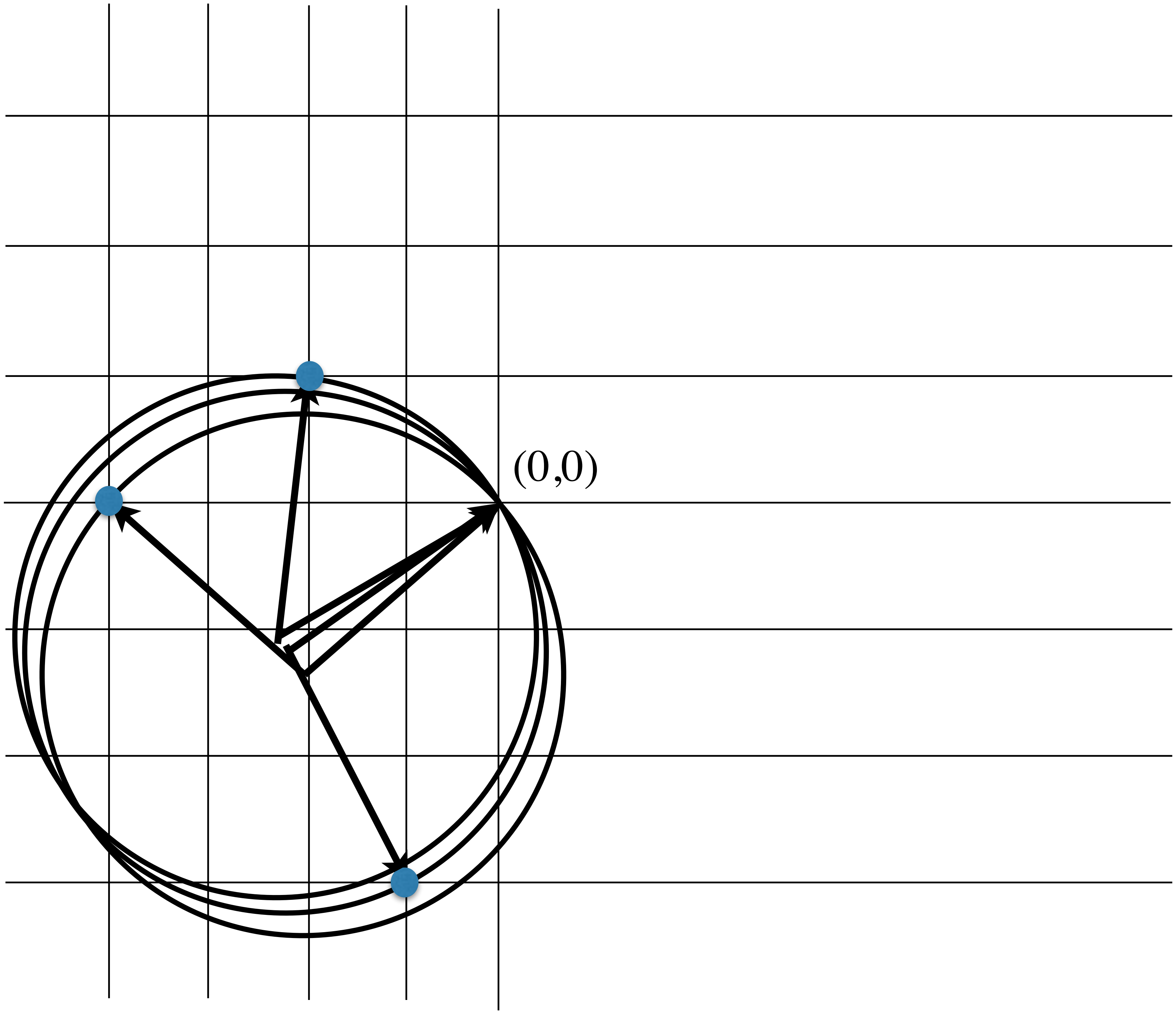
Diffraction: crystals

- Determining average structure from diffraction:
  - Find sets of Q that can lead to reflections
  - Practically, involves rotating crystal or changing x-ray wavelength to sweep the Ewald sphere around in reciprocal space



Diffraction: crystals



Diffraction: crystals

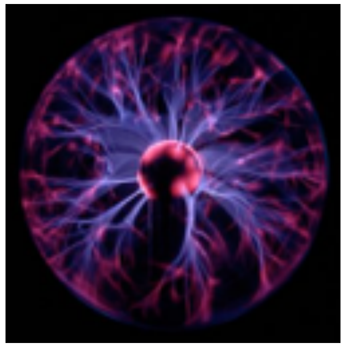
- Now we know the translational symmetry (shape of u.c.)
- For unit cell structure, need to measure $|F_c(\mathbf{G})|^2$ for several reflections
- “Systematic absences”: additional symmetries
- In principle, results in a system of nonlinear equations to solve
- Sometimes ambiguous, need tricks (e.g. anomalous diffraction, see tomorrow)

$$F_c(\mathbf{Q}) = \sum_j f_j e^{i\mathbf{Q}\cdot\mathbf{r}_j}$$ Unit cell structure factor

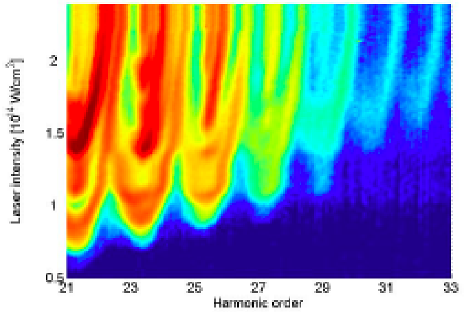
Short pulse x-ray sources

Overview of fs x-ray sources

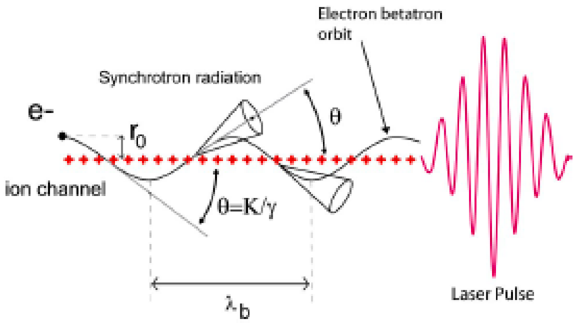
Laser-based



Plasma



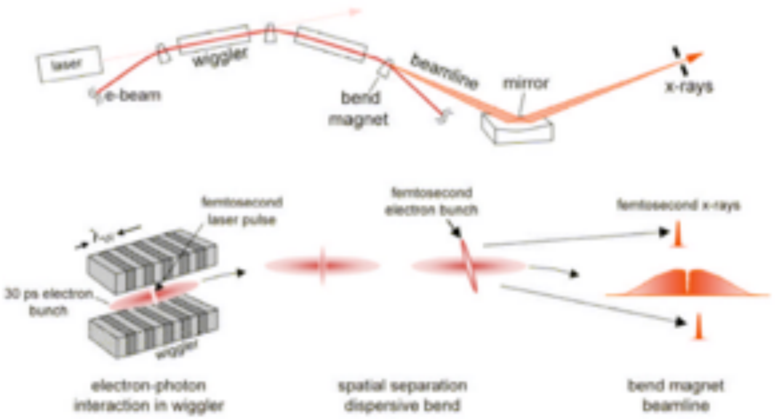
HHG



[Phuoc, *et al*. Phys. Plasmas 12, 023101 (2005)]

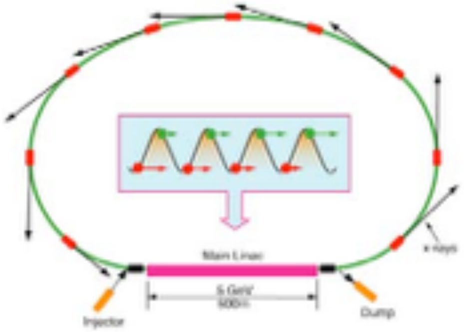
"Plasma-wiggler"

Accelerator-based

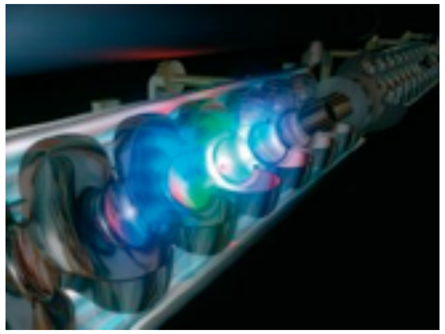


Zholents and Zolotorev, *Phys. Rev. Lett.*, **76**, 916,1996.

Slicing



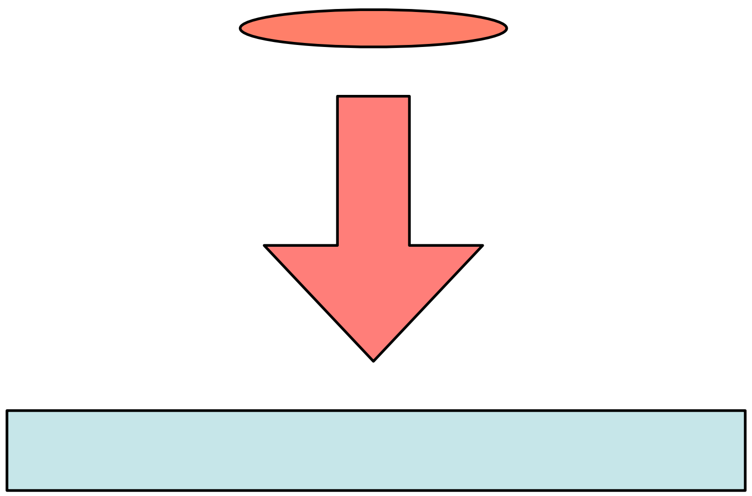
ERL



XFEL

Laser-produced plasmas

Basic idea: very high energy fs ablation

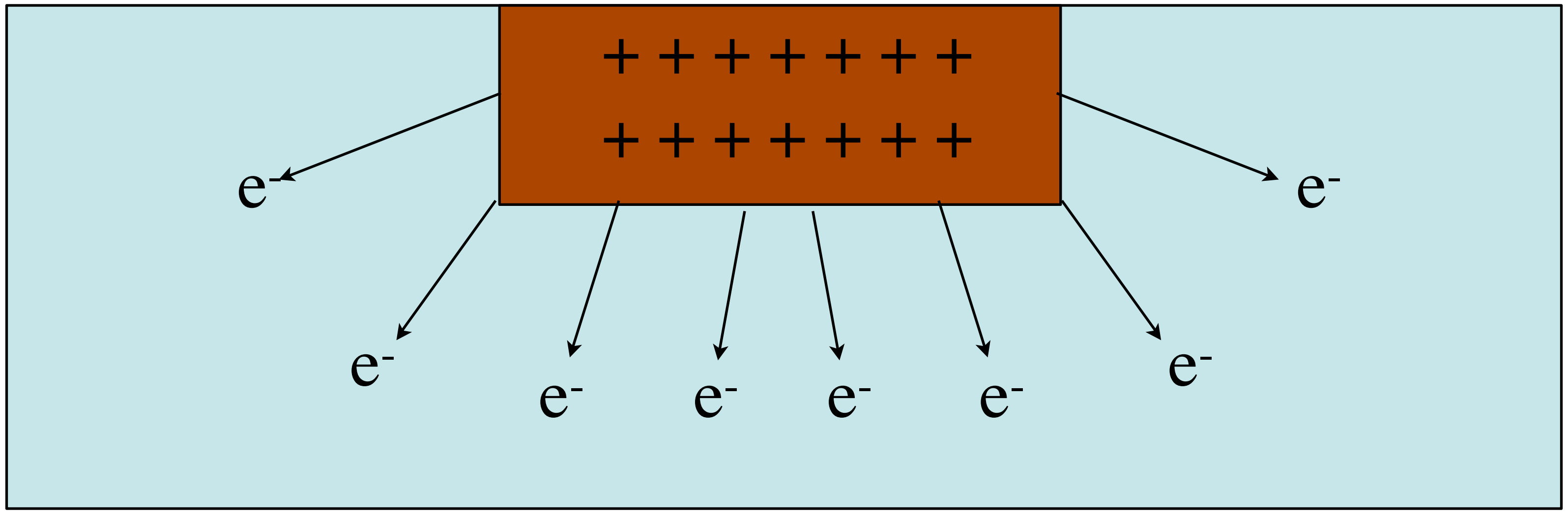


Laser-produced plasmas

Basic idea: very high energy fs ablation

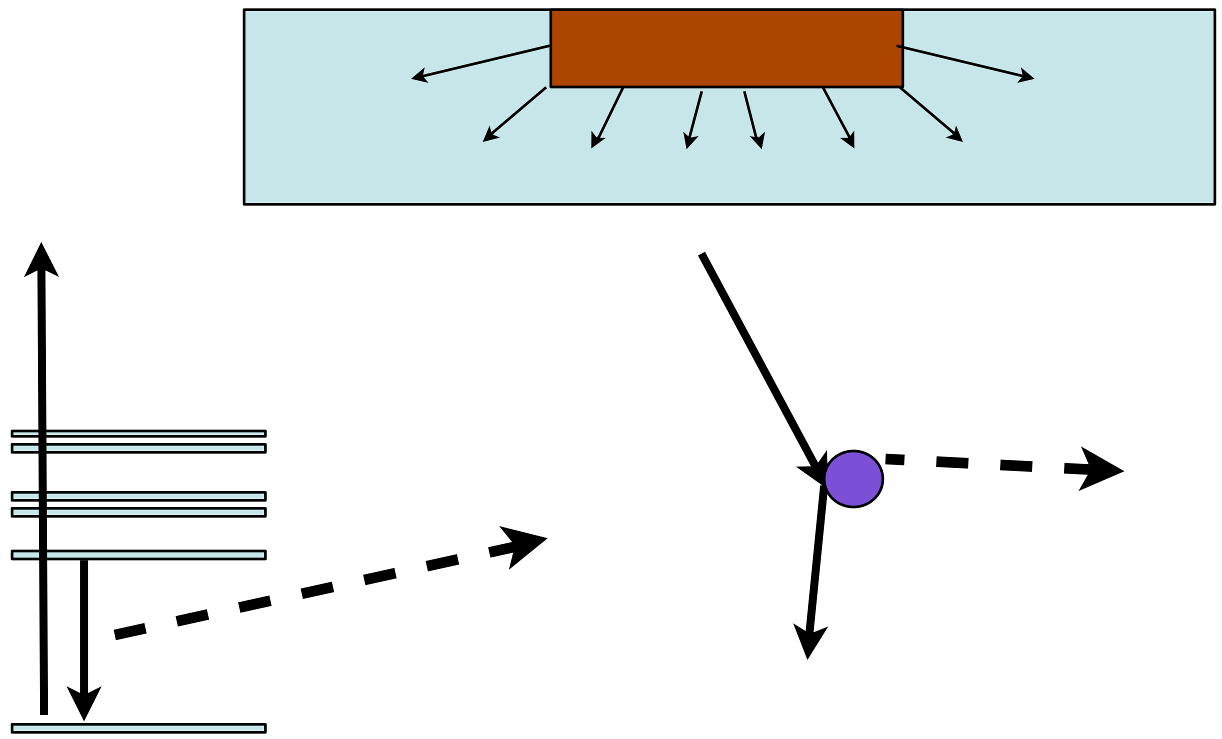
Laser-produced plasmas

High energy electrons sent into cold material



Laser-produced plasmas

Core level ionization of atoms causes x-ray line emission;
Bremsstrahlung radiation gives a continuum background



Laser-produced plasmas: properties

- Integrated flux: ~ 3 x 10^8/pulse at Ti Kα line (10 Hz system)
- Collimation: none (emits in all directions)
- Brilliance: ~ 5 x 10^4 photons/mm^2/$mrad^2$/0.1% BW/pulse
- Wavelength: Depends on target; most flux at atomic emission lines, but there is a continuum background esp. for high Z targets
- Pulse duration: ~300 fs (set by plasma dynamics)
- Rep rate: 10-1000 Hz (depends on laser)
- Stability: not formally characterized, but very sensitive to laser

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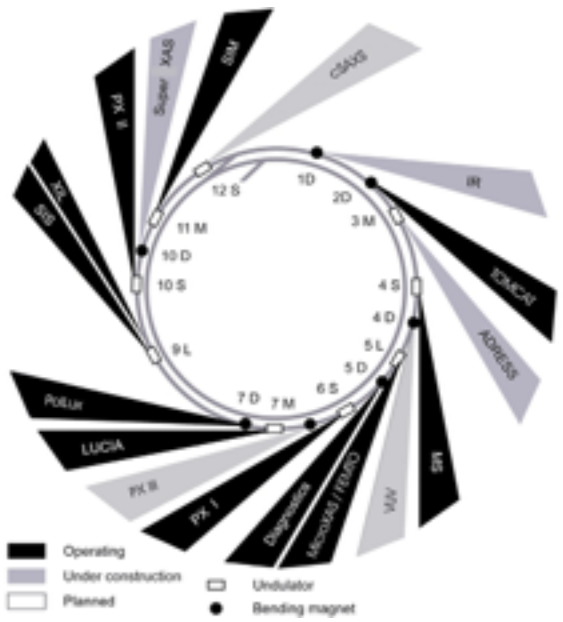
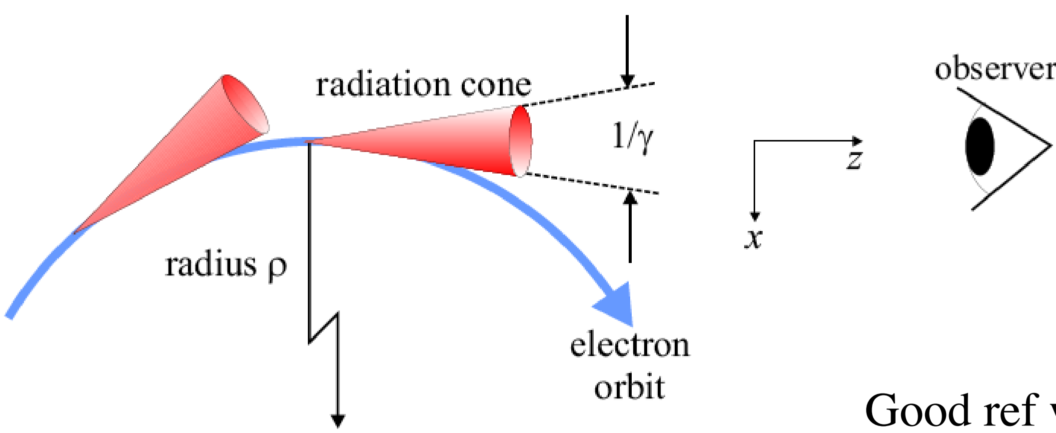
Synchrotron radiation



Light from accelerated relativistic electrons



Good ref with more math: K.-J. Kim, Nucl. Instrum. Methods Phys. Res. A246, 71 (1986)

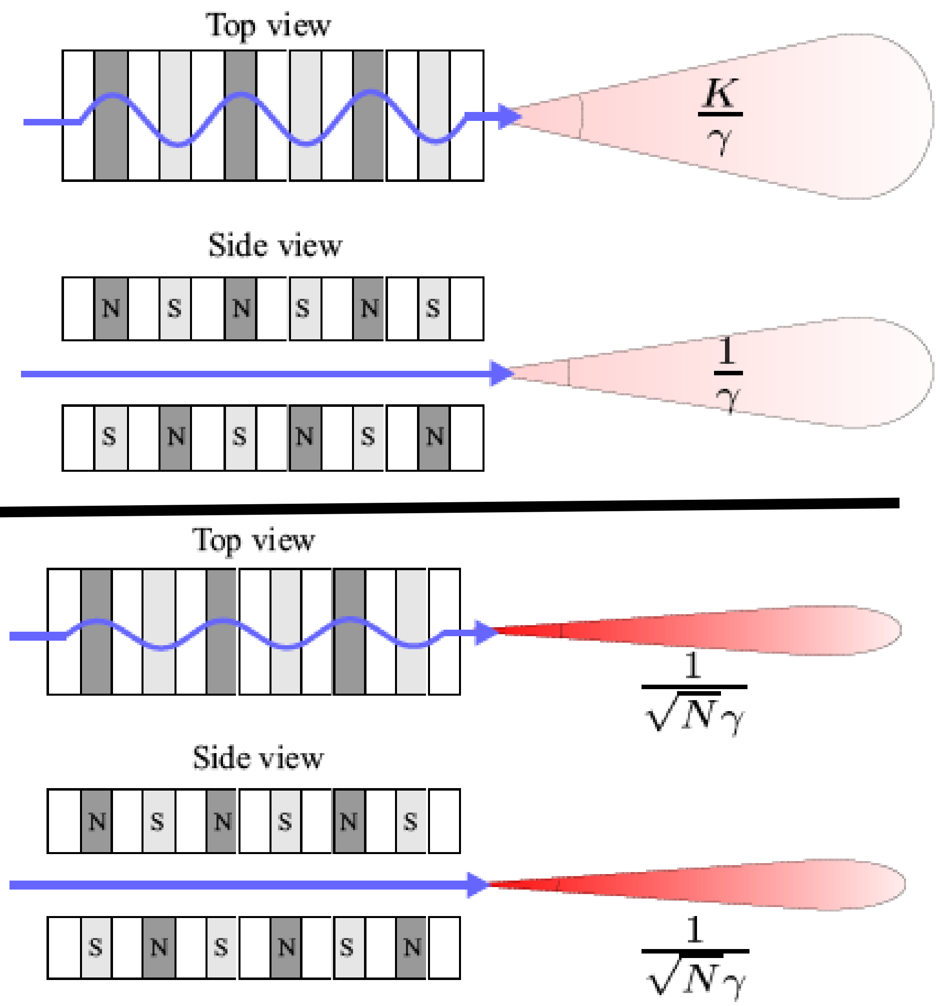
[Als-Nielsen & McMorrow, *Elements of Modern X-ray Physics*, John Wiley & Sons, Ltd, 2001]

Synchrotron radiation

Insertion devices: more bends for more light

Wiggler:
large angular excursions,
essentially a series of bends

Undulator:
small angular excursions,
inteference phenomena



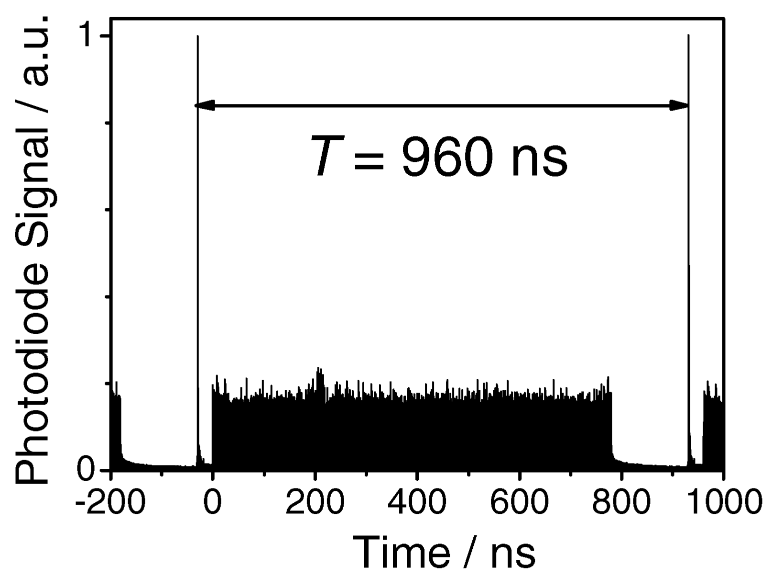
[Als-Nielsen & McMorrow, *Elements of Modern X-ray Physics*, John Wiley & Sons, Ltd, 2001]

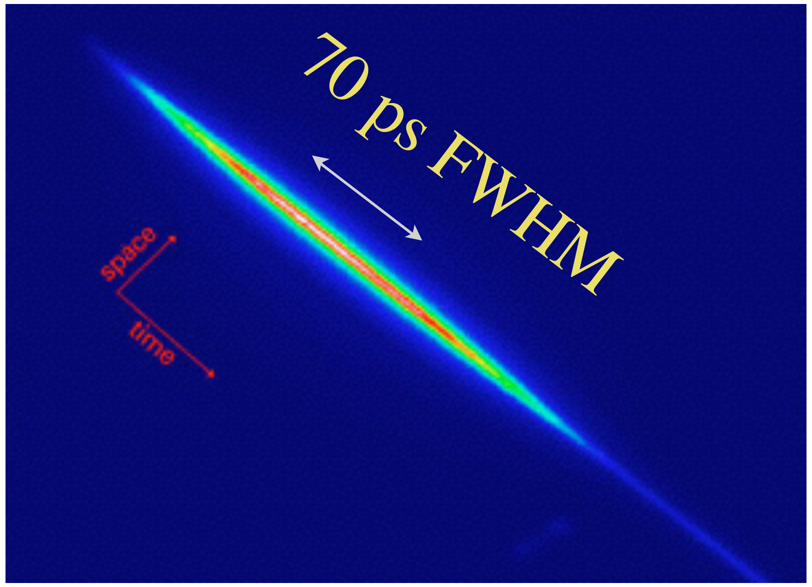
Synchrotron radiation

Insertion devices: more bends for more light

Synchrotron radiation

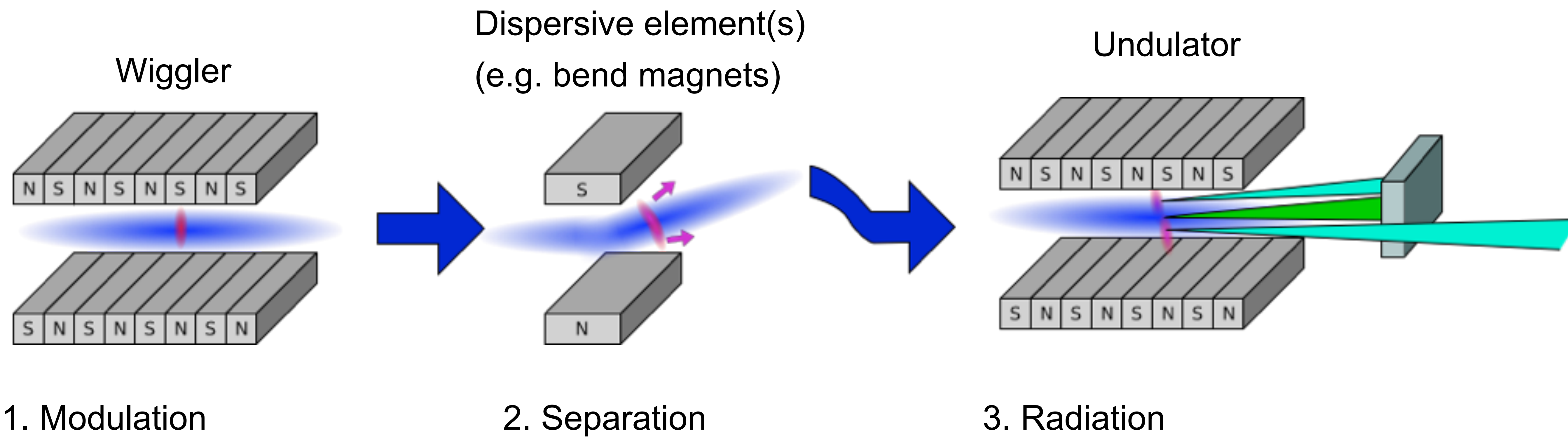
Time structure of synchrotron X-rays





- Electrons in bunches, spacing ~ 2 ns
- Stability of electron beam (e-e scattering) requires ~ 100 ps long bunches
- For femtosecond x-rays, create a transient short bunch...

Slicing

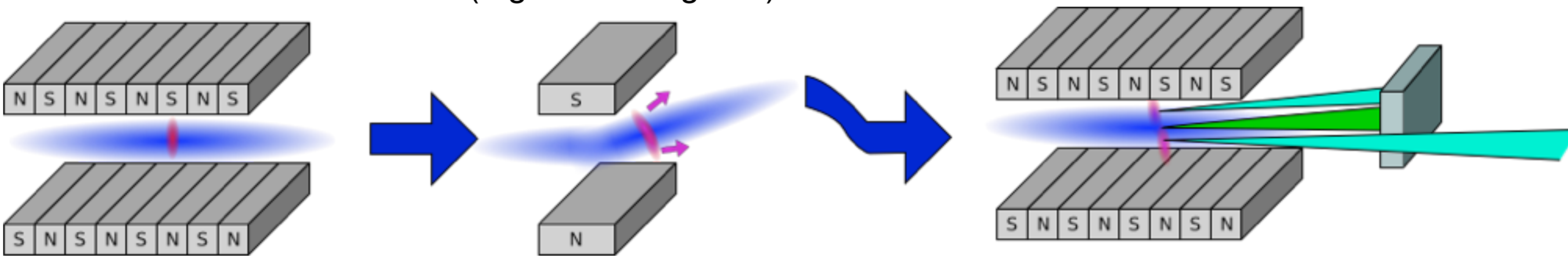

Wiggler
N S N S N S N S
S N S N S N S N
Dispersive element(s)
(e.g. bend magnets)
S
N
Undulator
N S N S N S N S
S N S N S N S N
1. Modulation
2. Separation
3. Radiation

Slicing

Dispersive element(s)
(e.g. bend magnets)

Wiggler

Undulator

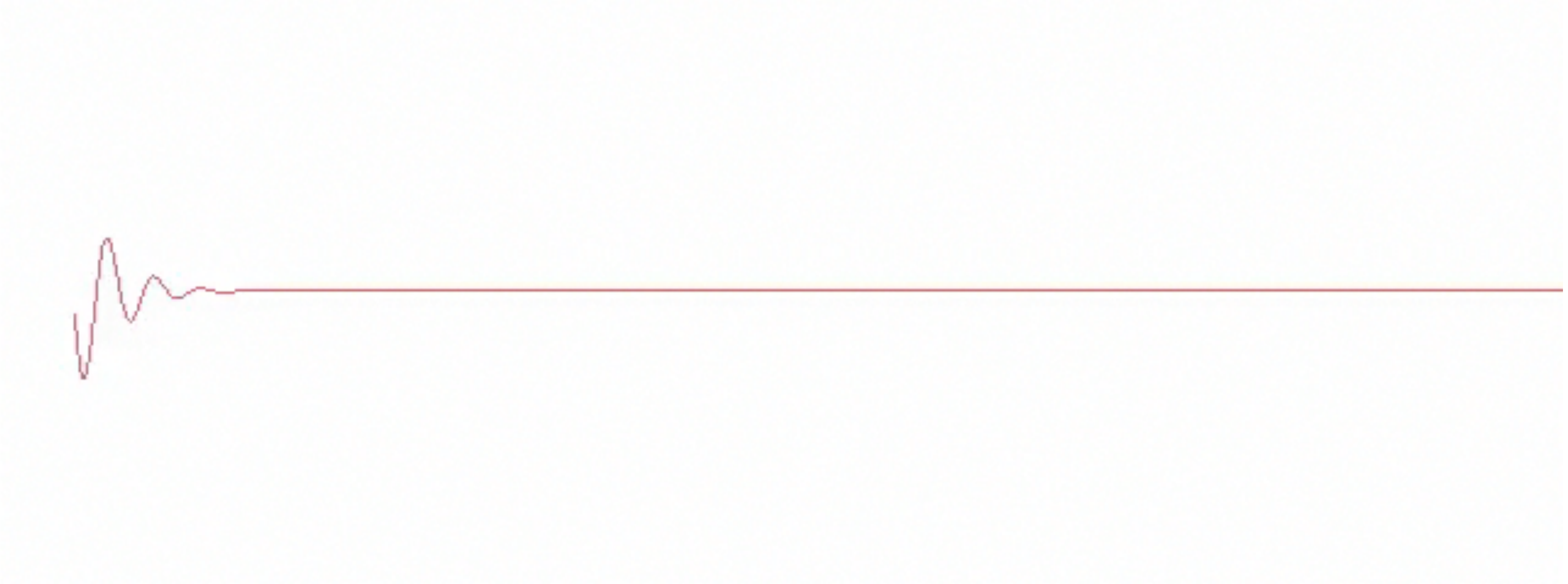


1. Modulation

2. Separation

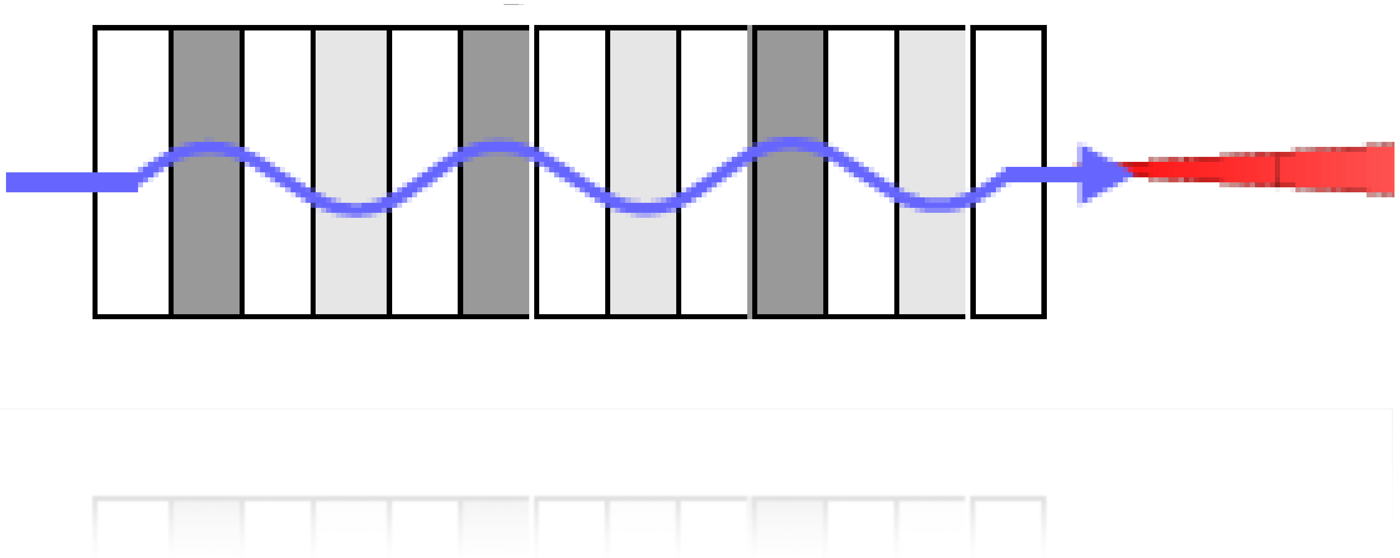
3. Radiation

Slicing



- E-field of laser transverse to direction of propagation
- Efficient energy exchange requires transverse component of electron momentum … undulator!

Slicing



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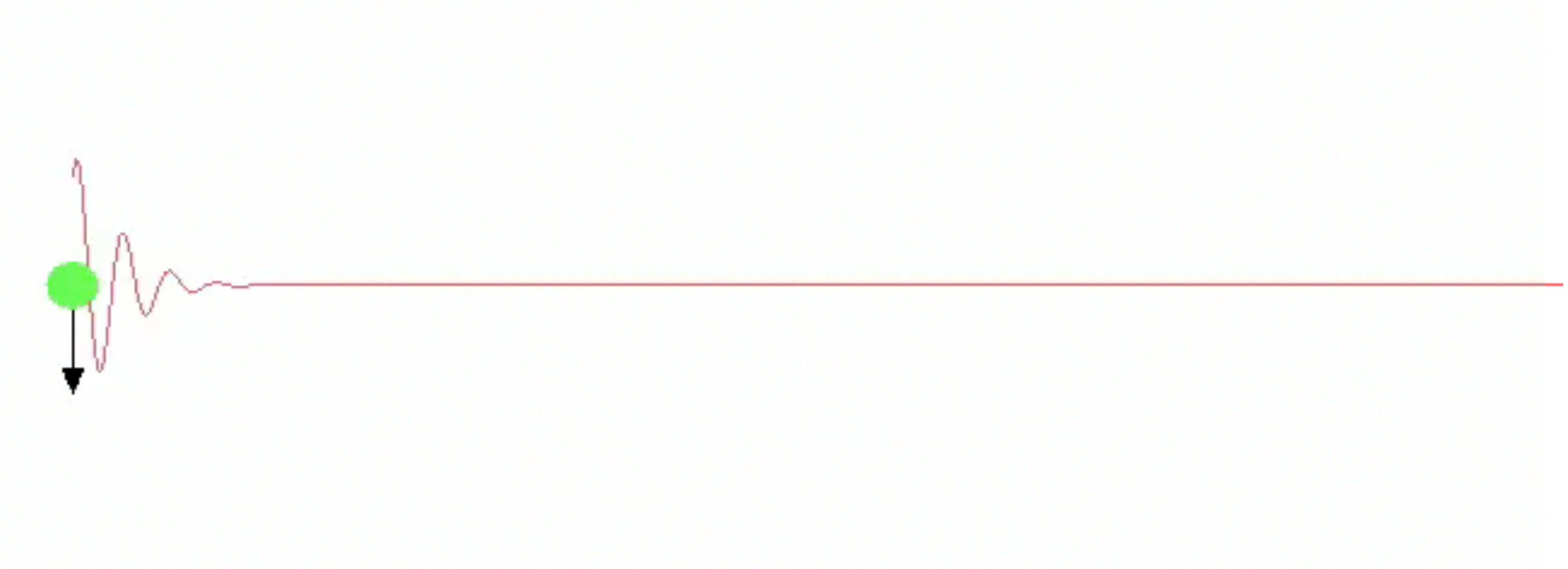
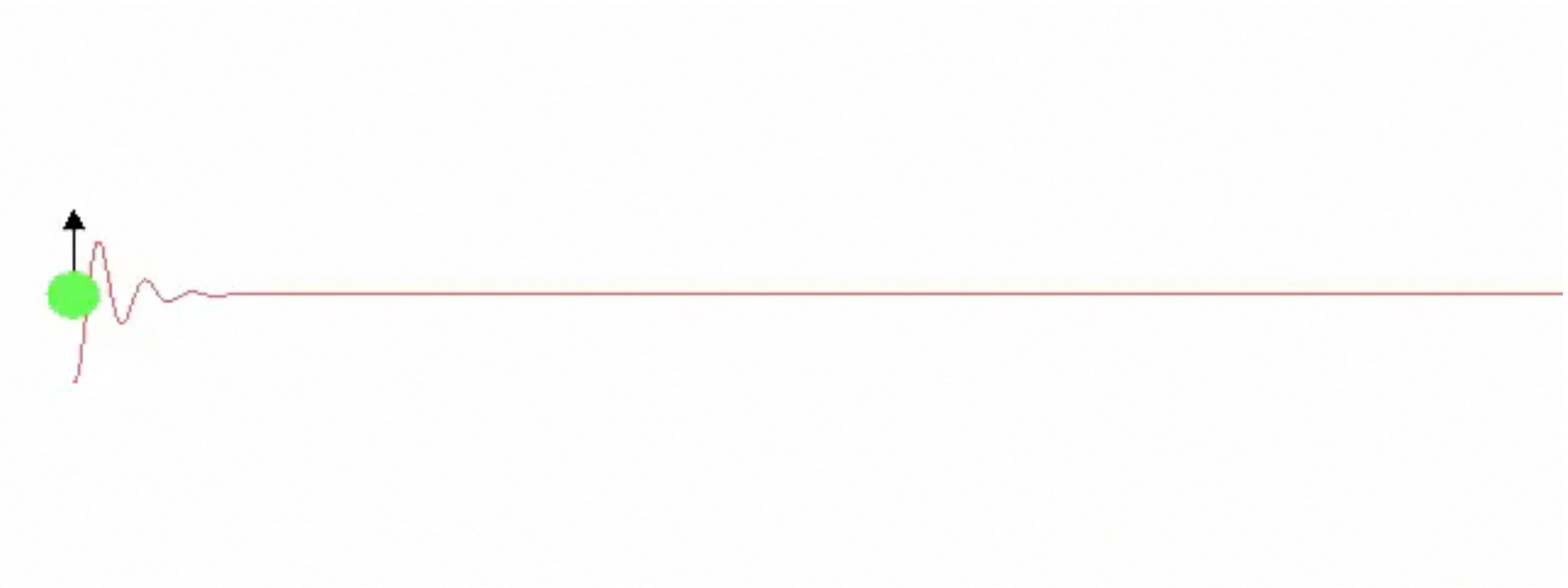
Slicing



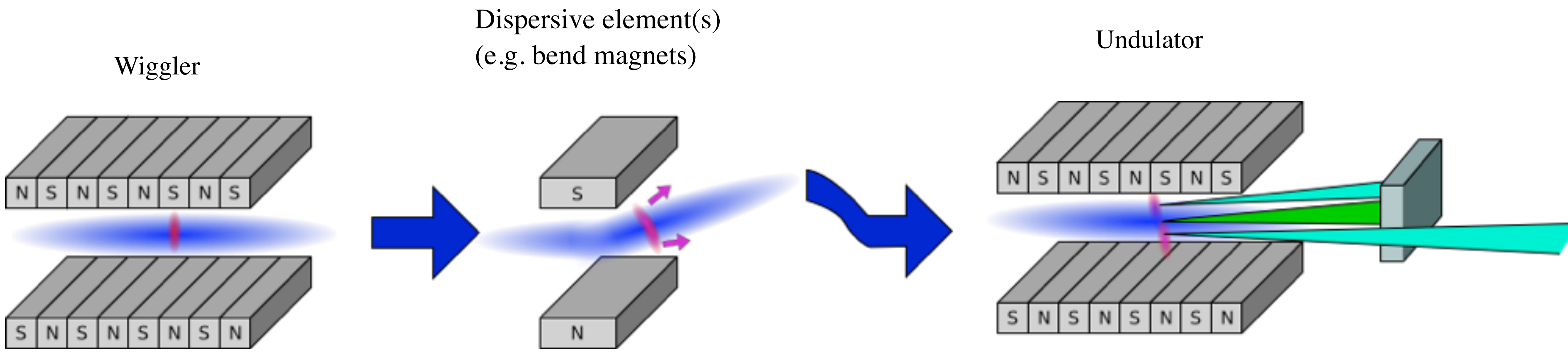
$$dE/dt = \mathbf{F} \cdot \mathbf{v} \geq 0$$

Slicing



$$dE/dt = \mathbf{F} \cdot \mathbf{v} \leq 0$$

Slicing

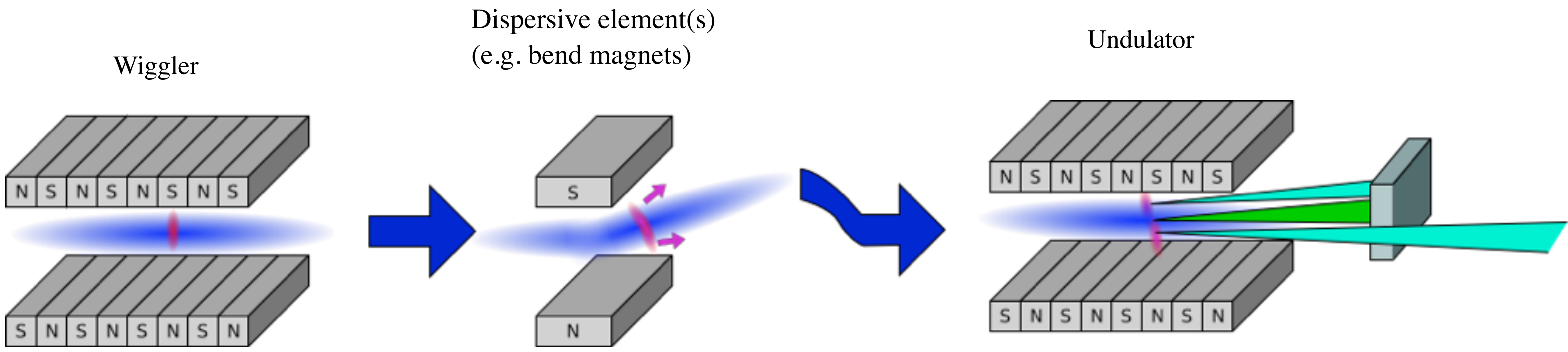


1. Modulation

2. Separation

3. Radiation

Slicing



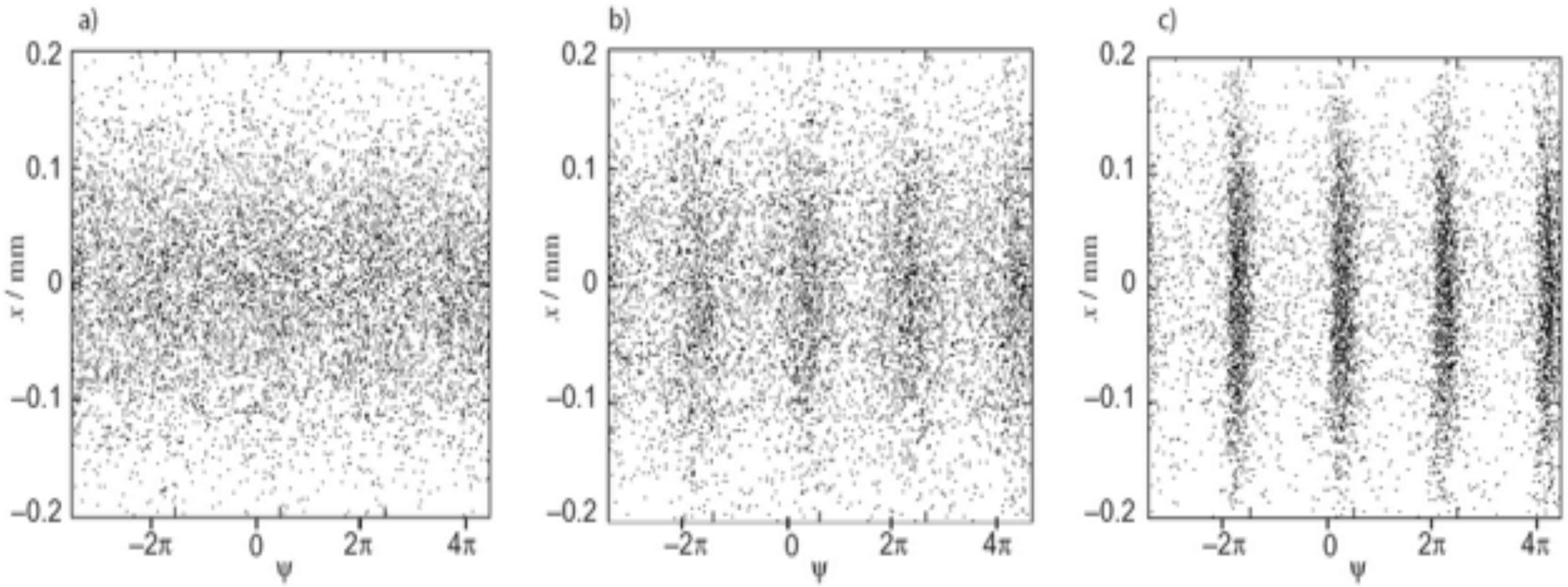
1. Modulation

2. Separation

3. Radiation

Free electron laser

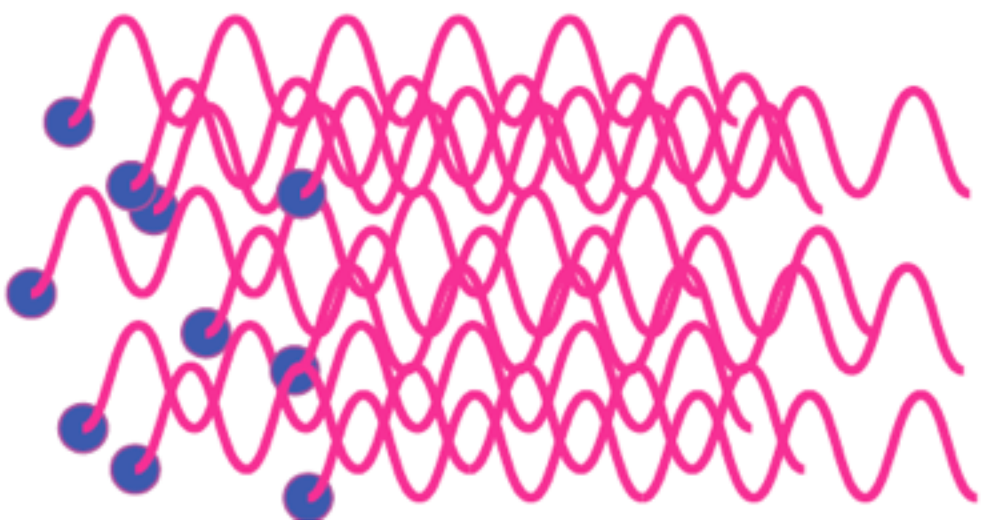
Like slicing, but long undulator → positive feedback → microbunching



Initial facilities (LCLS, SwissFEL, EU-XFEL, ...) seeded by noise

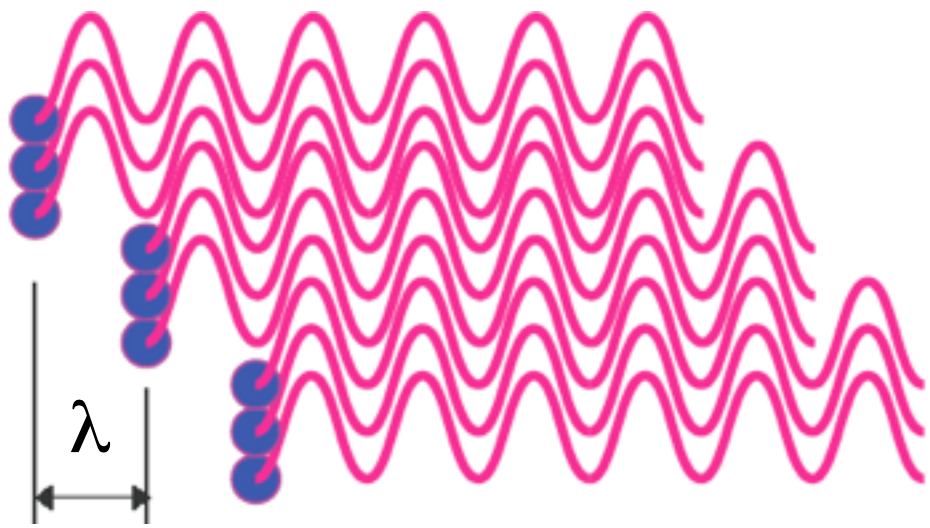
Free electron laser

Spontaneous



$$P = N\ P_1$$

Superradiant

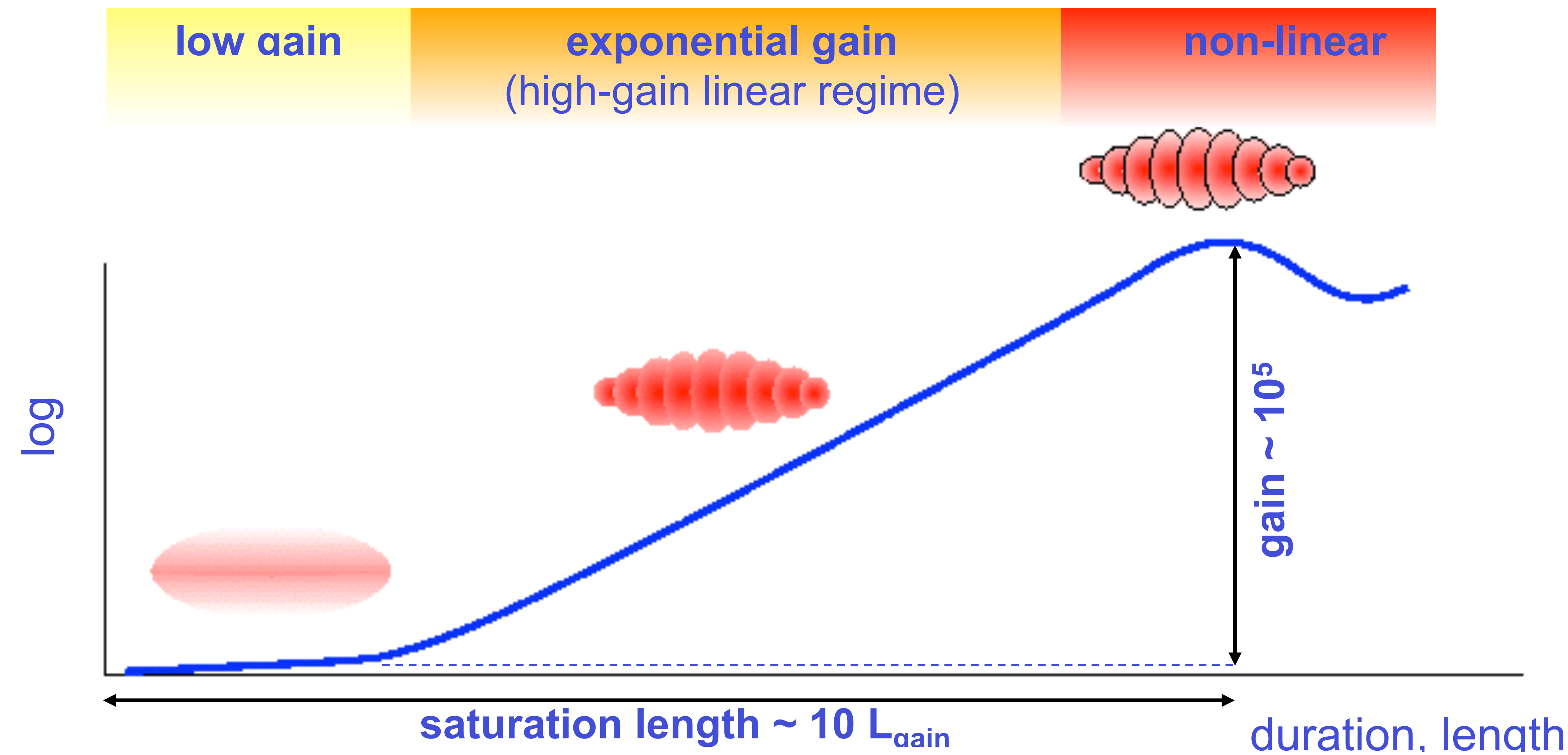


$$E = N\ E_1$$

$$P = N^2\ P_1$$

$N \approx 10^9$

Free electron laser



Result: coherent, bright, short (< 10 fs) x-ray pulses

Free electron laser

- Photons per pulse: ~10^{12}
- Wavelength: ~ 1-100 Å
- Pulse duration: ~ 10-100 fs (shorter "spikes")
- Rep rate: highly variable, from ~ 10 Hz to ~ 1 MHz "bursts"
- Collimation: ~ 1-10 μrad divergence
- Brilliance: ~ 10^{20} ph/mrad^2/mm2/0.1% BW/pulse
- Spatially coherent
- Stability poor (so far)

(recall: ~10^5 for plasma source!!)

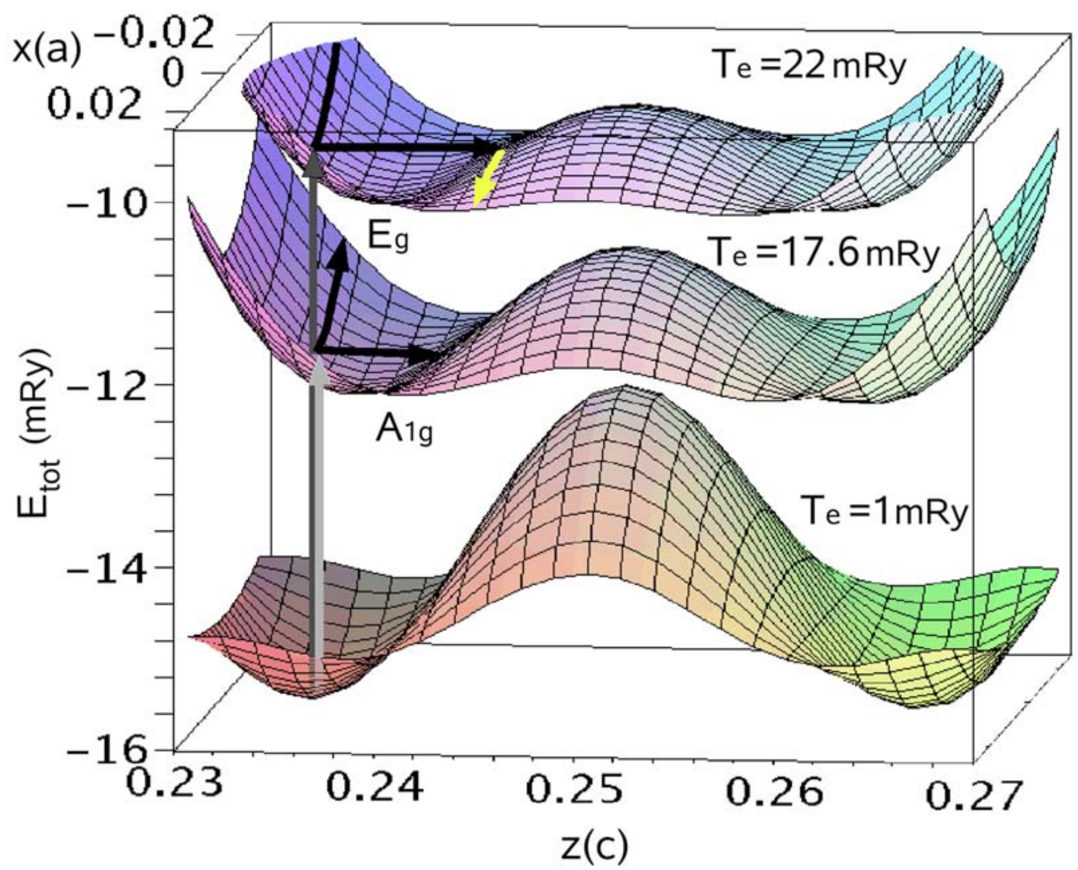
Time-resolved diffraction

“Indirect” control:

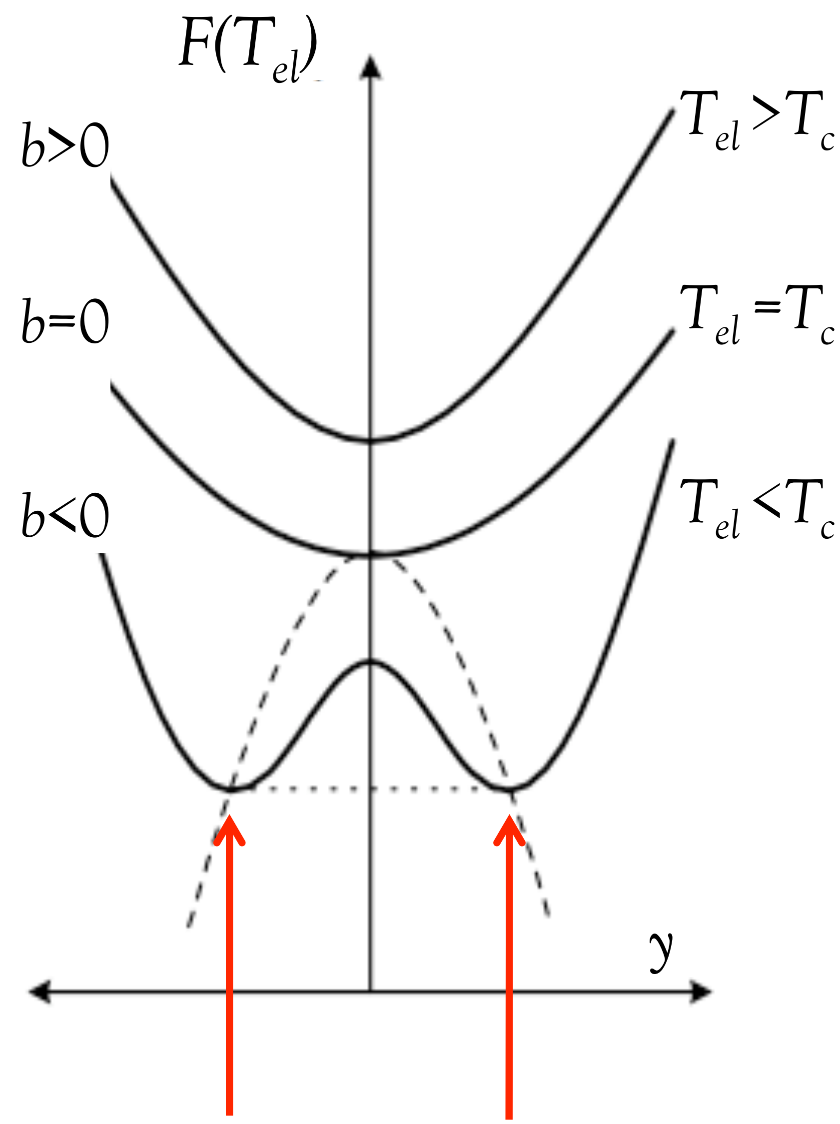
Electronically induced symmetry changes

Idea: driving symmetry changes

- Electronic excitation changes free energy surface for ions
- What if new surface has a different (lower) symmetry?



[Zijlstra, Tatarinova & Garcia, PRB 74, 220301 (2006)]

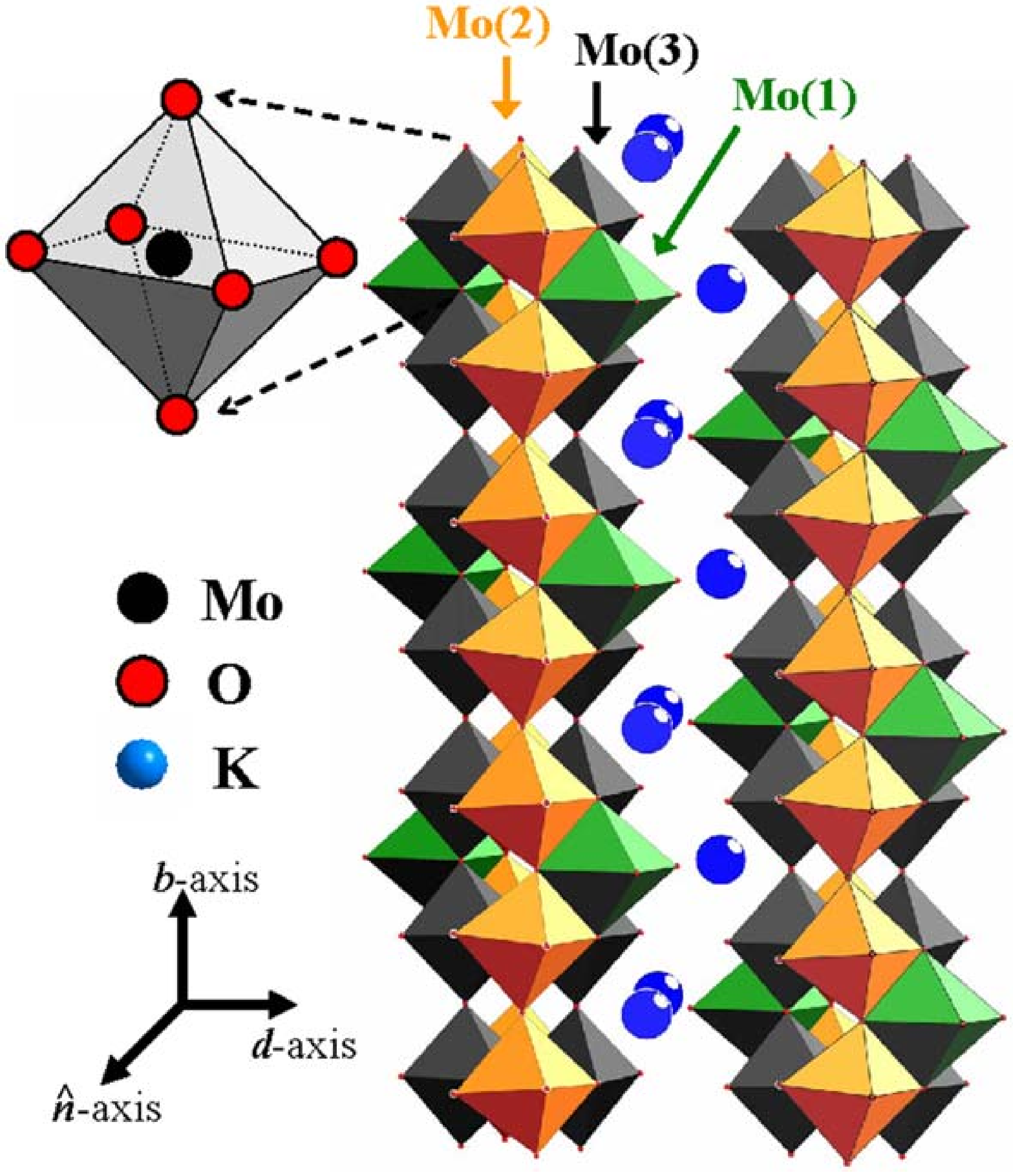


$$y = -\sqrt{-\frac{b}{2c}} \qquad y = +\sqrt{-\frac{b}{2c}}$$

$K_{0.3}MoO_3$

- Quasi 1D Peierls system
- Below 180 K incommensurate superlattice
- Optical excitation excites coherent phonons related to transition

e.g.: H. Schäfer et al. PRL 105, 066402 (2010)



[Tsai et al. Appl. Phys.Lett. 91, 022109 (2007)]

Experiment team: $K_{0.3}MnO_3$

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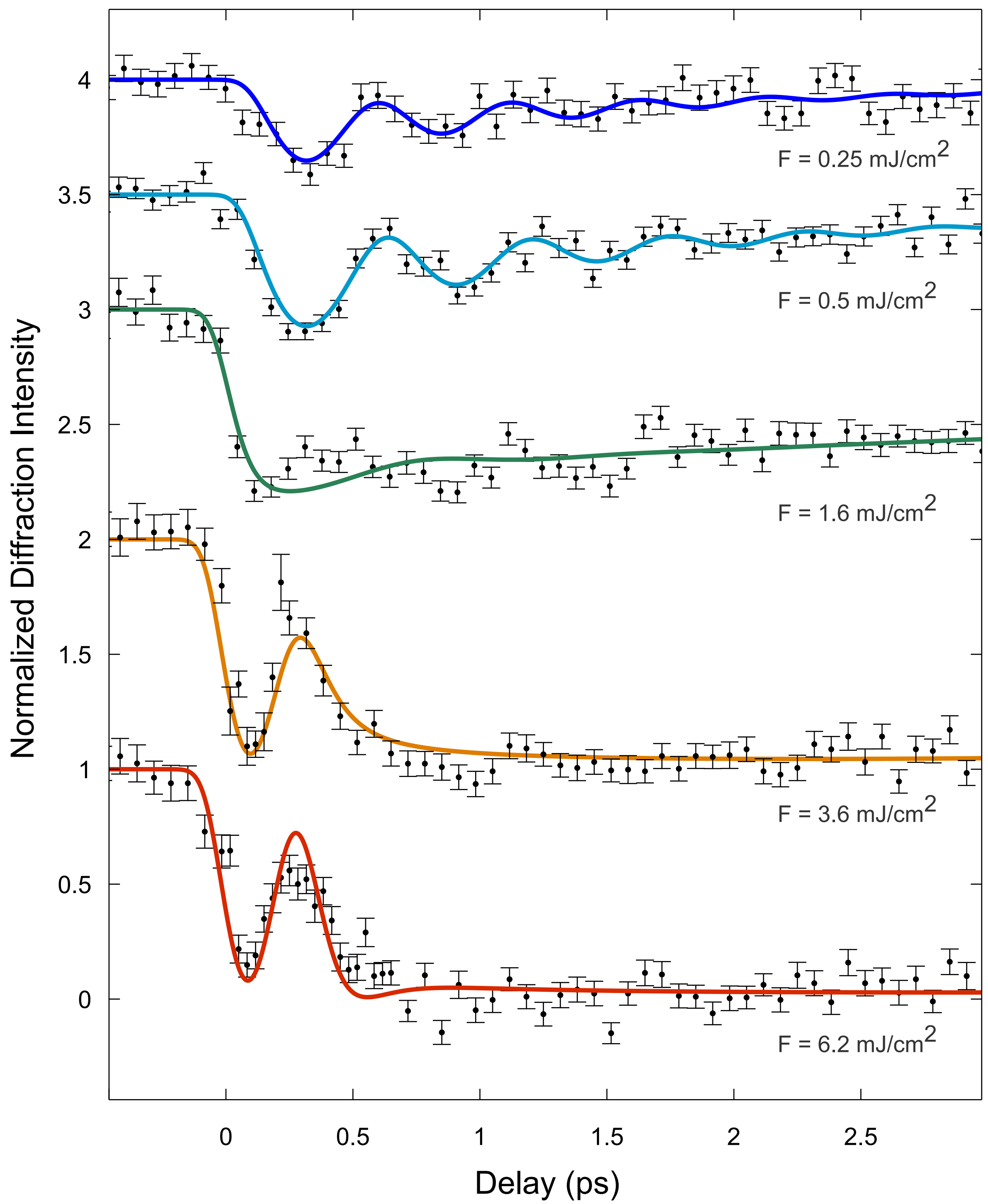
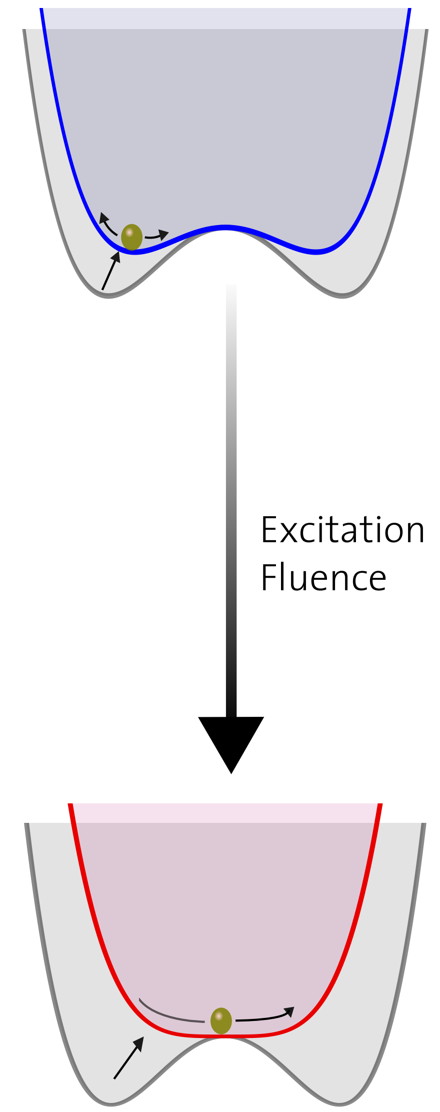
Dynamics of incommensurate modulation

- Low fluence: coherent phonon in low-symmetry potential
- High fluence: symmetry change
- Anomalous damping





[A. T. Huber et al. PRL 113, 026401 (2014)]

Dynamics of incommensurate modulation

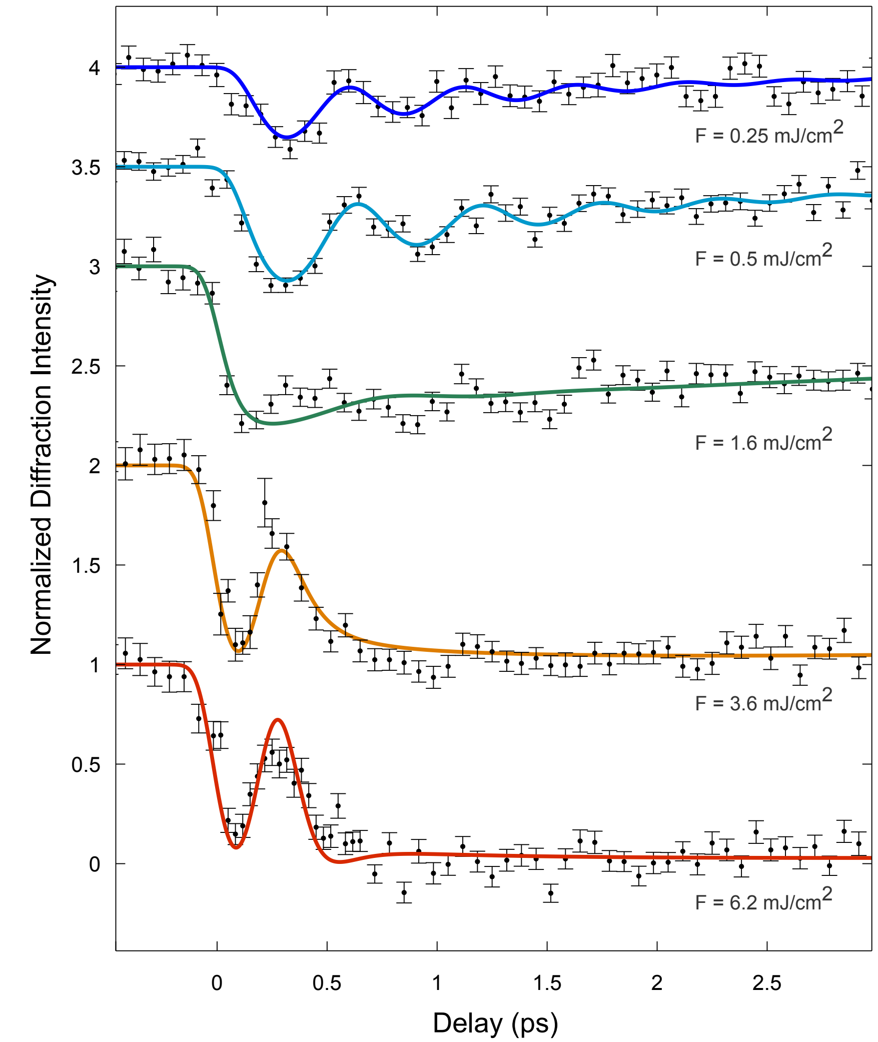
$$V(x) = \frac{1}{2}\left(\eta \exp\left(-\frac{t}{\tau_{\text{disp}}}\right) - 1\right)x^2 + \frac{1}{4}x^4$$

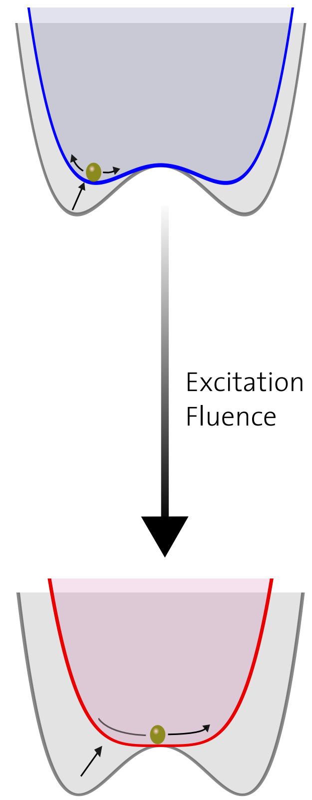
- Time-dependent potential surface, relaxes as electrons equilibrate with lattice
- Time-dependent damping rate

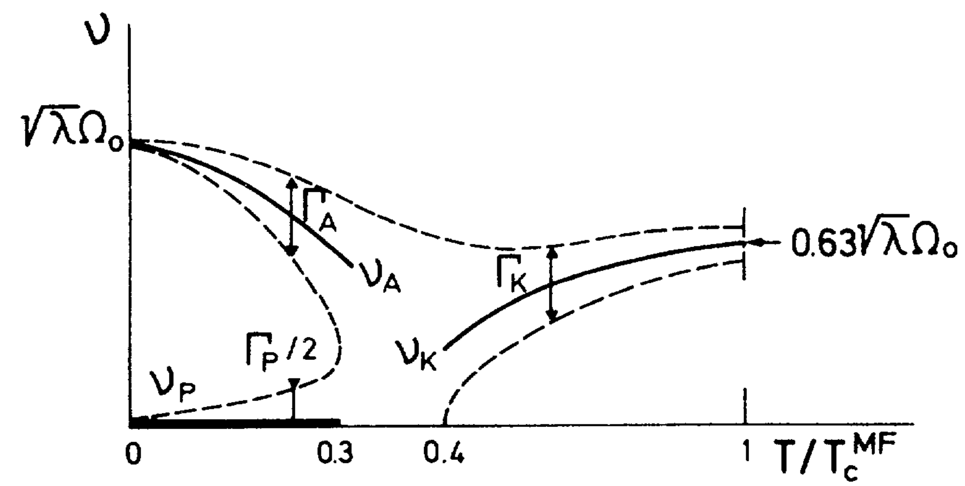
$$\frac{1}{\omega_{\text{DW}}^2}\frac{\partial^2}{\partial t^2}x - \left(1 - \eta \exp\left(-\frac{t}{\tau_{\text{disp}}}\right)\right)x + x^3 + \frac{2\gamma(t)}{\omega_{\text{DW}}^2}\frac{\partial}{\partial t}x = 0$$

$$\gamma(t) = \gamma_{\text{asym}}\left(1 - e^{-t/\tau_\gamma}\right)^2$$

[A. T. Huber et al. PRL 113, 026401 (2014)]







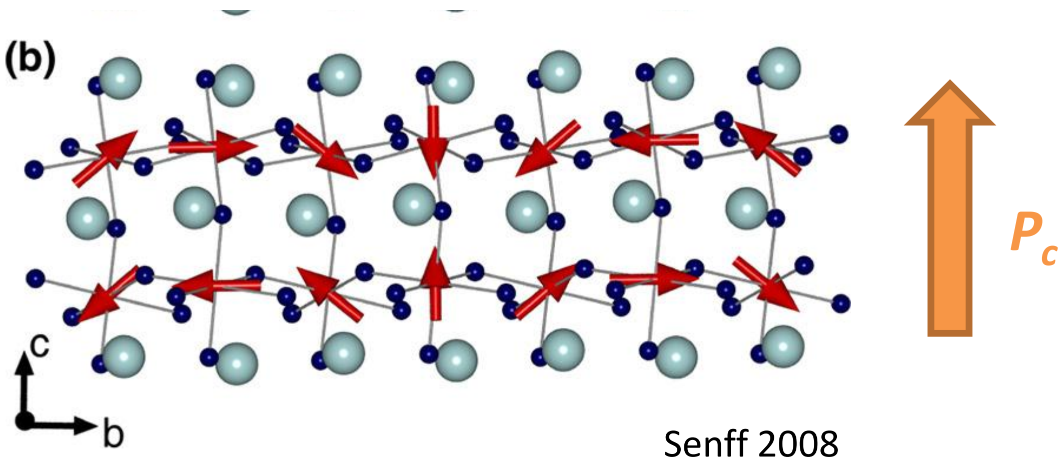
[Pouget et al. PRB 43, 8421 (1991)]

“Direct” control:

Spin dynamics of a large-amplitude coherent electromagnon

THz excitation: path to fast control of multiferroics?

TbMnO3

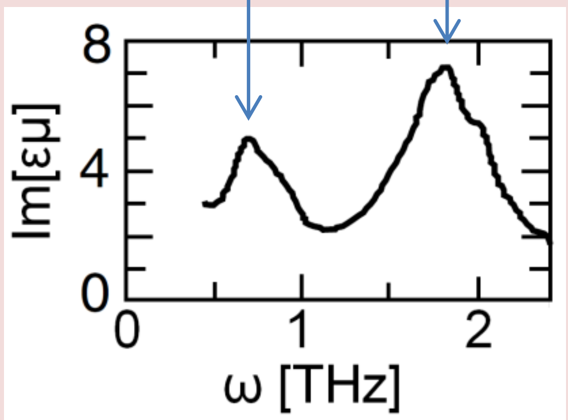


Senff 2008

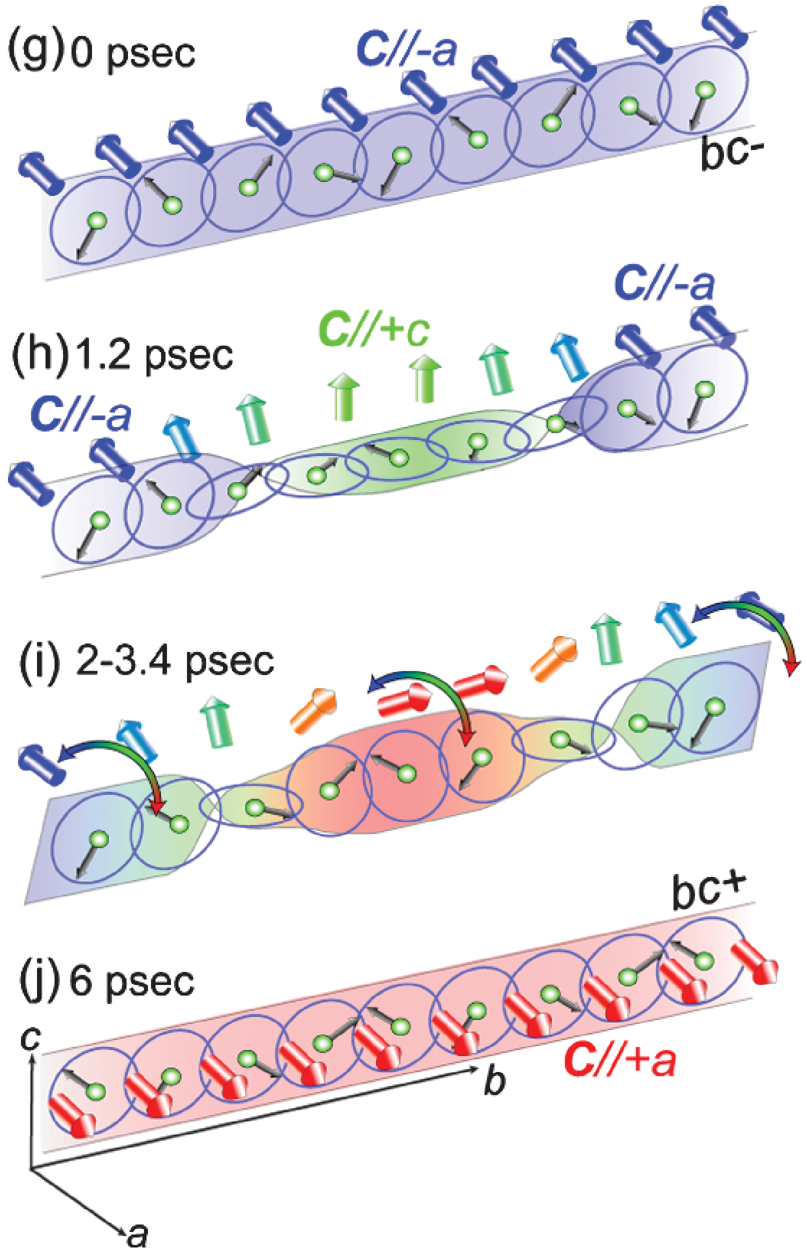
excitations due to the electromagnetic coupling:

higher-harmonic, ellipticity, phonons 0.7 THz

spin-spiral excitation, 1.8 THz

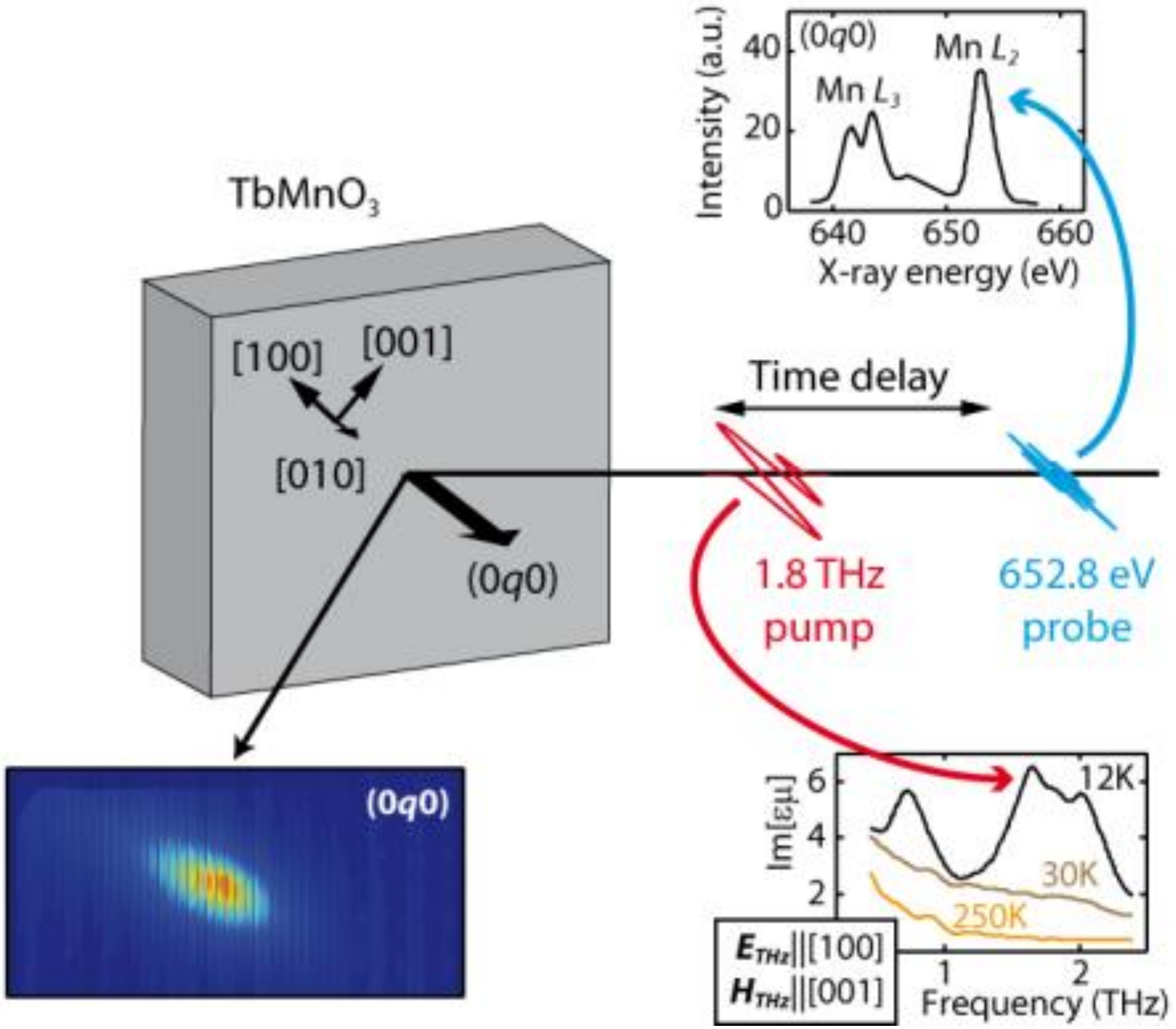


[Y. Takahashi et al., PRL **101**, 187201 (2008)]



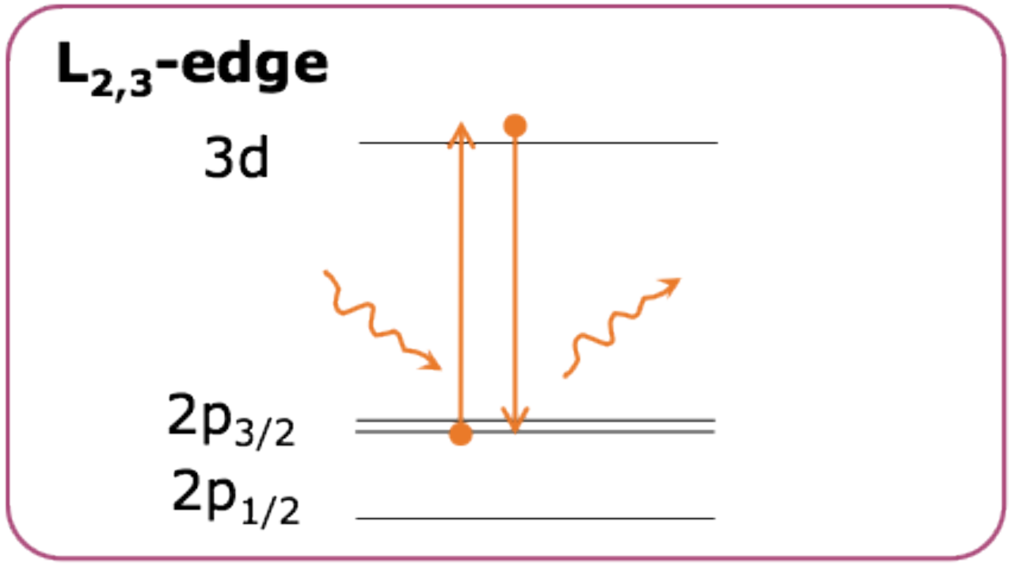
[Mochizuki & Nagaosa, PRL **105**, 147202 (2010)]

Experiment concept



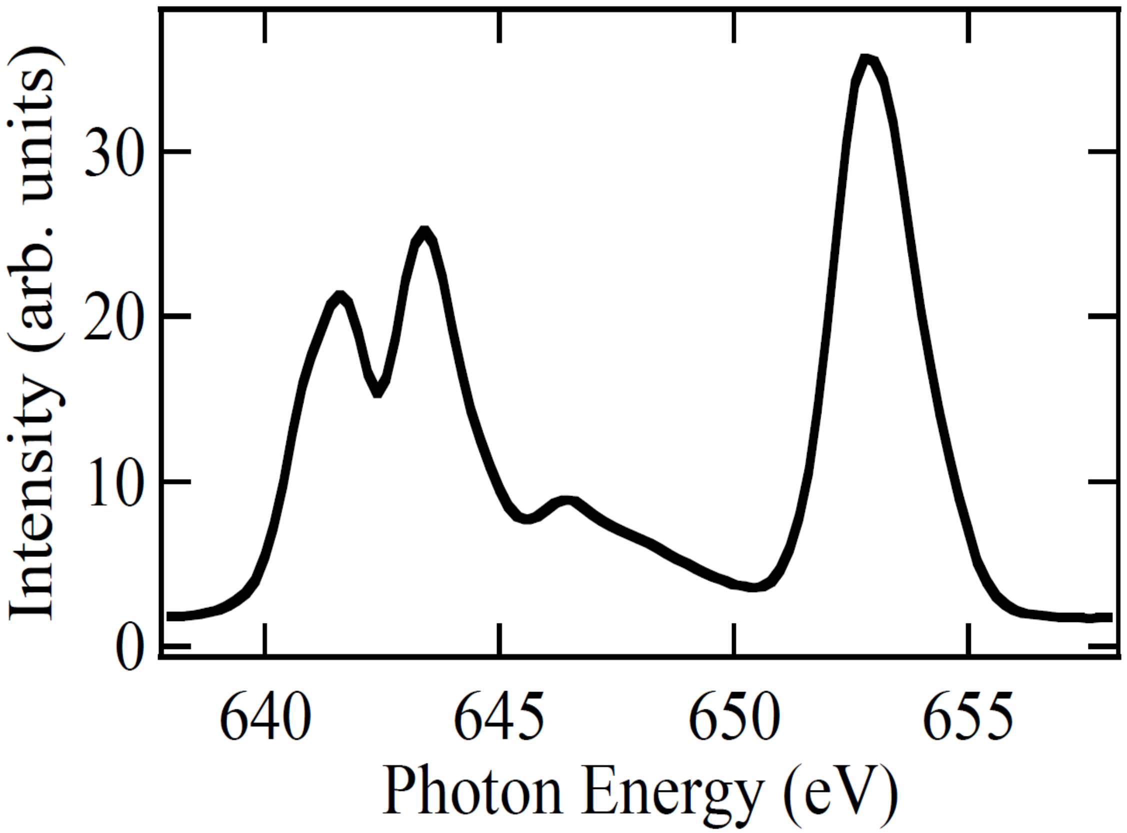
Pump electromagnon with THz, watch spins with resonant x-ray diffraction

X-ray pulses: probe spin order



$$\left\langle \mathbf{T}_q^k \right\rangle \propto \sum_n \frac{\langle g| O |n\rangle \langle n| O^* |g\rangle}{E_n - E_g - \hbar\omega + i\Gamma}$$

- (0q0) reflection at Mn L-edges: only magnetic order



- Experiment at LCLS
- Pulses of < 80 fs duration
- Time-stamping for < 250 fs resolution

[Beye et al. Appl. Phys. Lett. 100, 121108 (2012)]

Experiment team: $TbMnO_3$

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Stanford:

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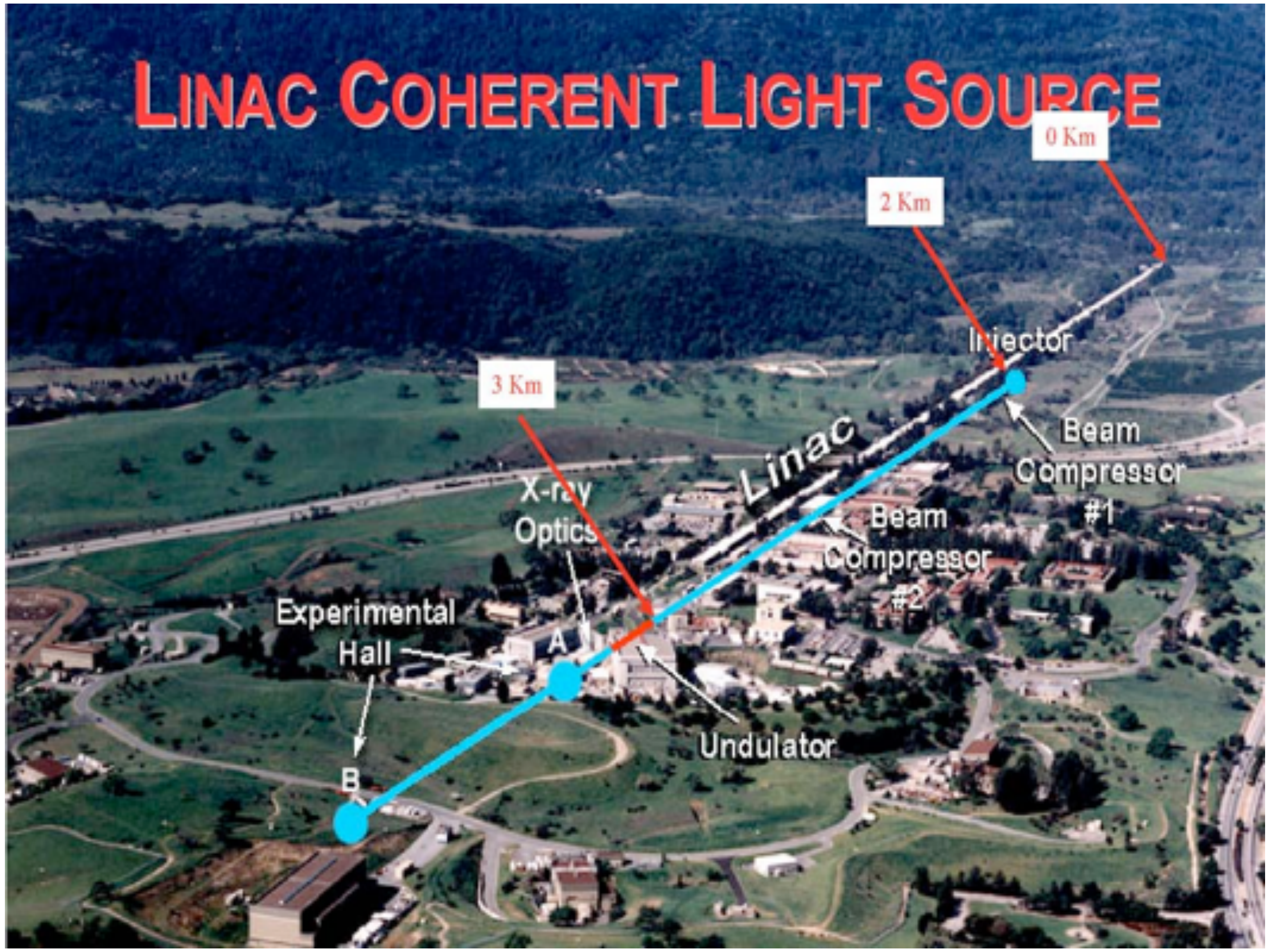
R. G. Moore

Johns Hopkins:

S. M. Koohpayeh



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must

LCLS

FNSNF
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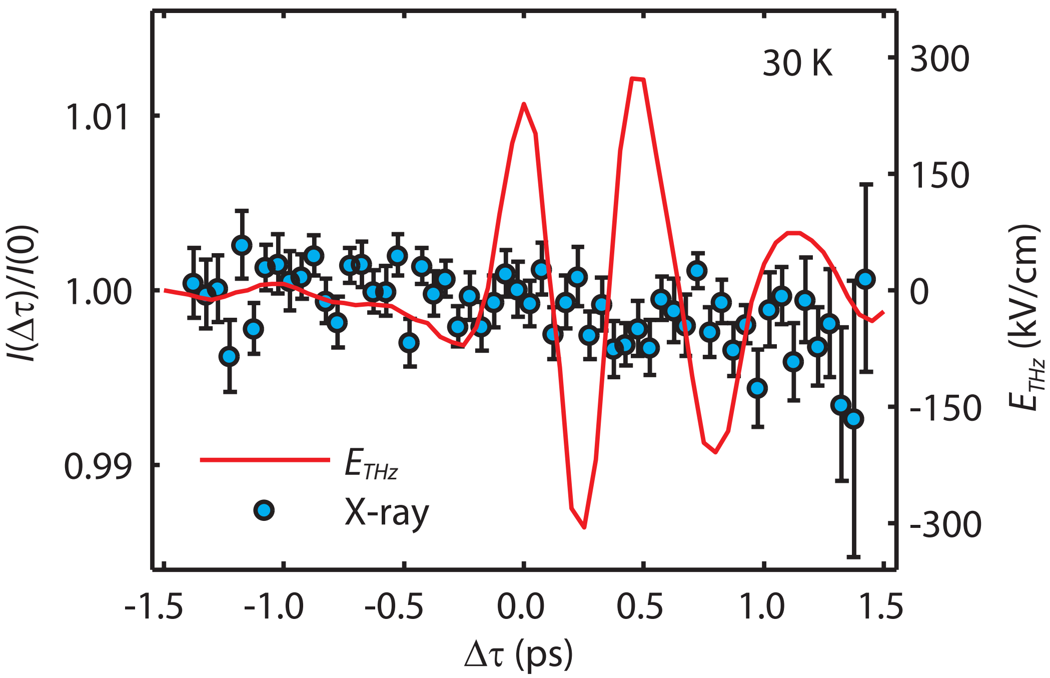
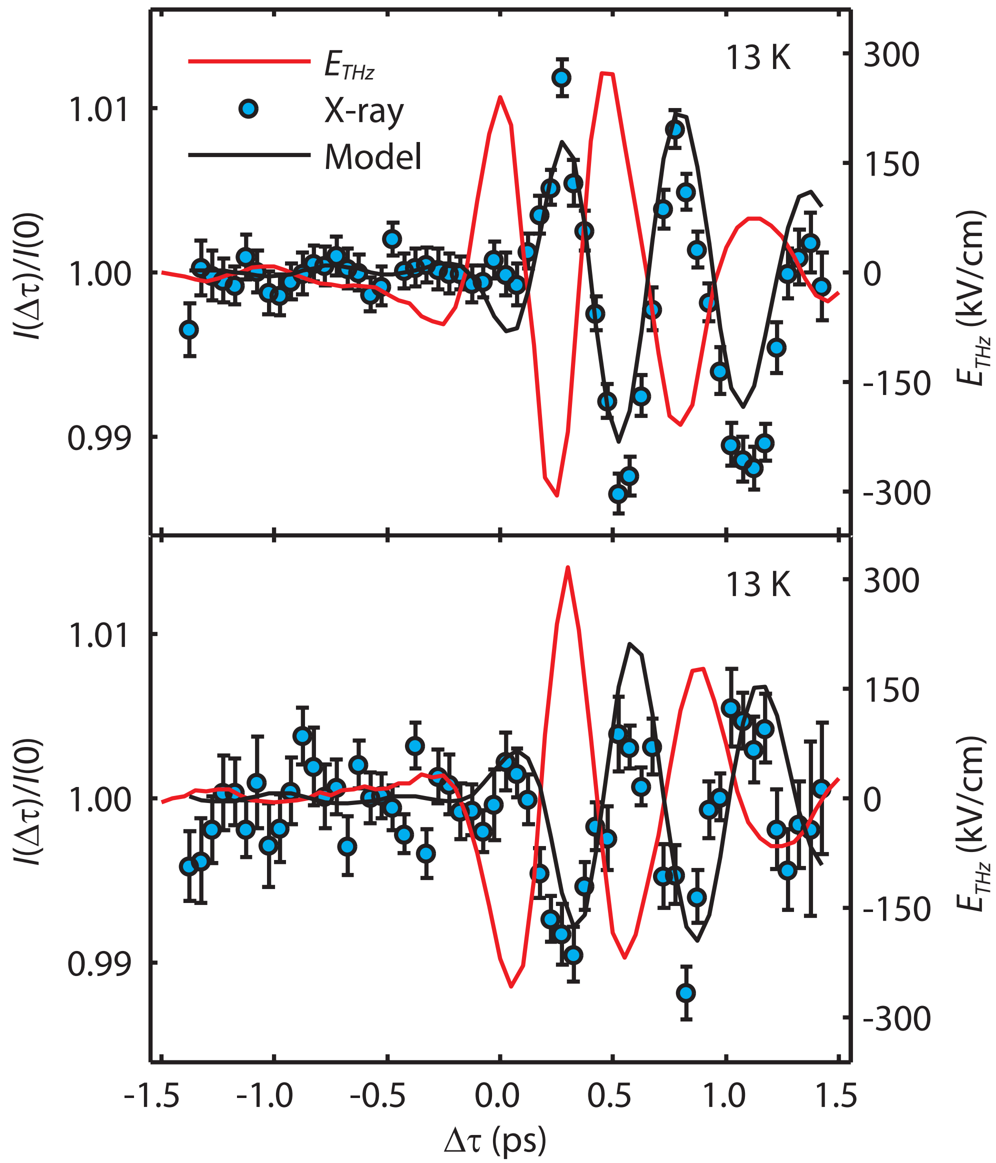
Results: coherent electromagnon

- E-field of THz → coherent spin response
- Measured spin response delayed by half cycle
- Response suppressed in non-multiferroic phase

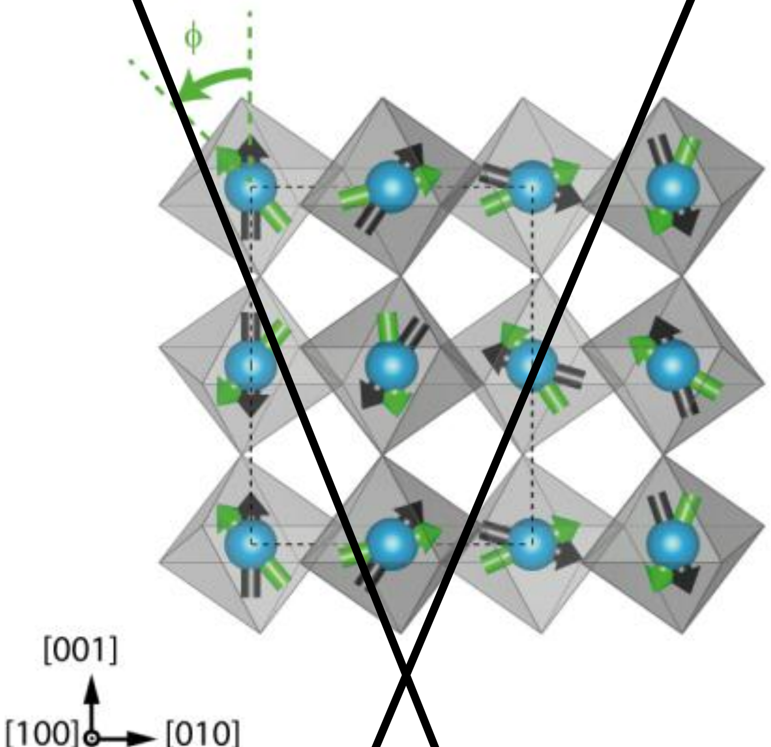


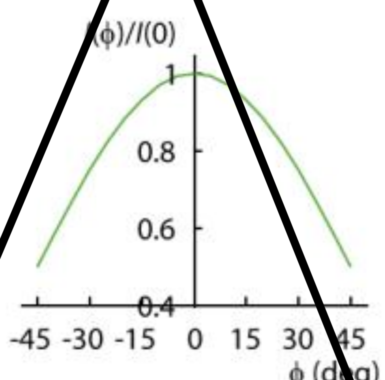


[T. Kubacka et al., Science **343**, 1333 (2014)]

Analyzing the motion

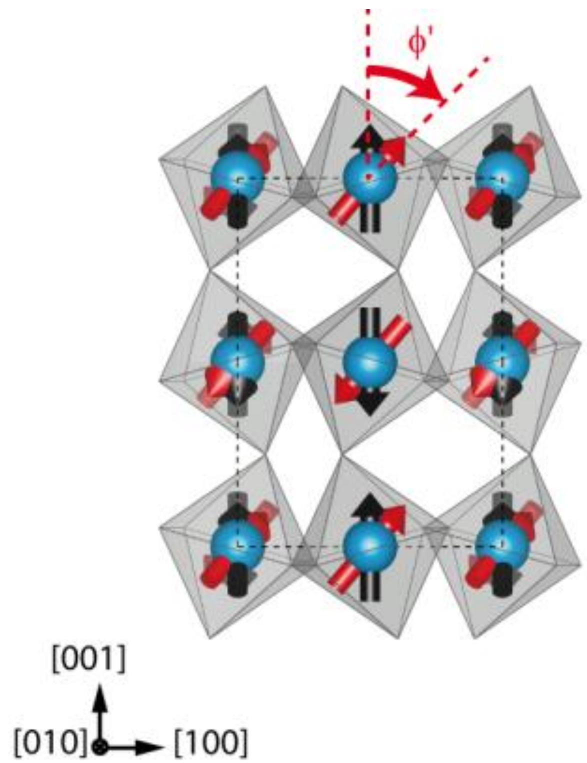
In-plane rotation

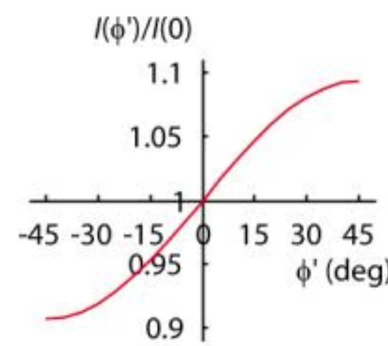




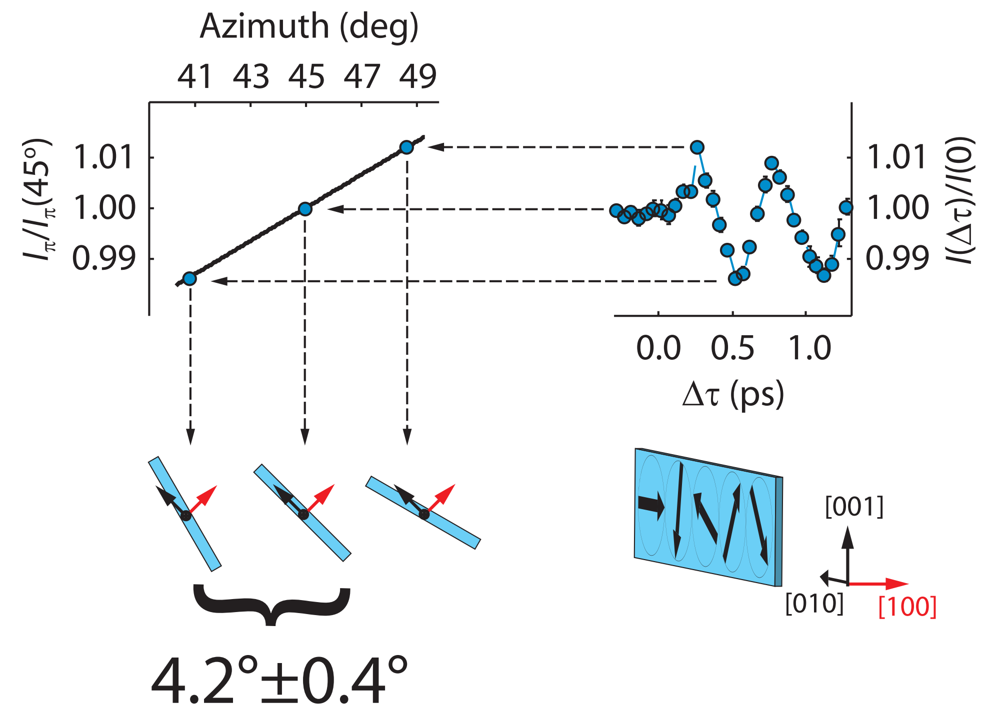
Even function

Rotation of spin planes





Odd function

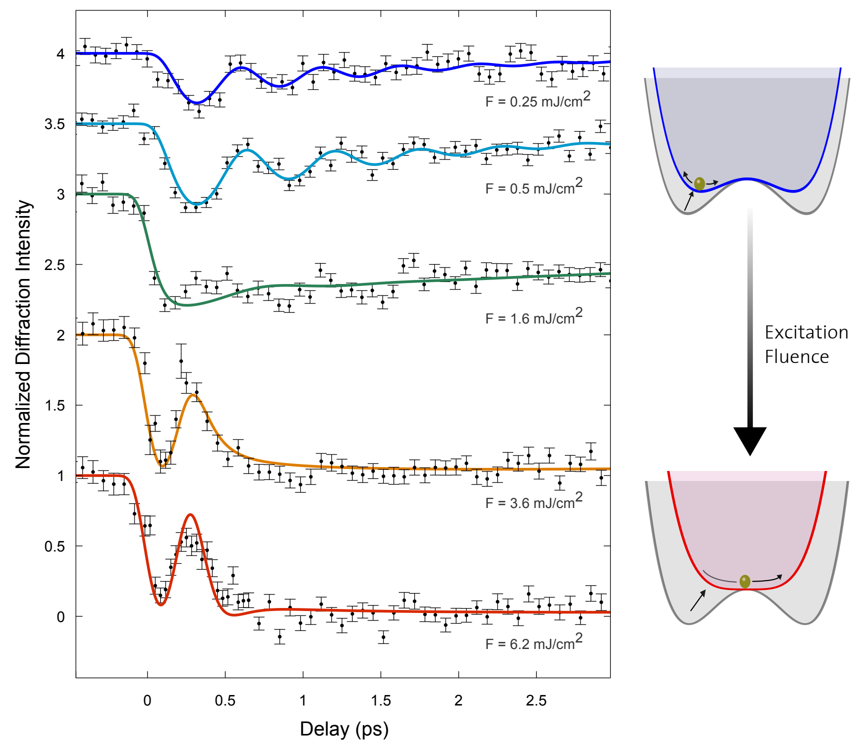


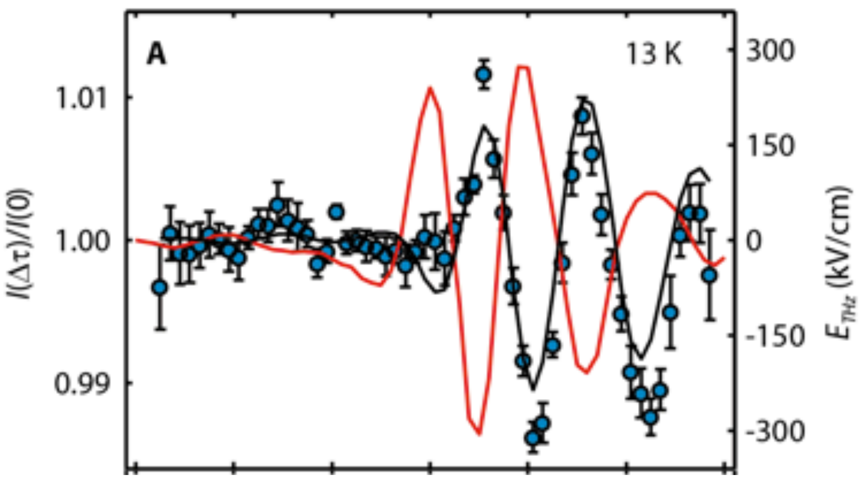
⇒ 4.2 ± 0.4 degree rotation of spin planes

[T. Kubacka et al., Science **343**, 1333 (2014)]

Summary

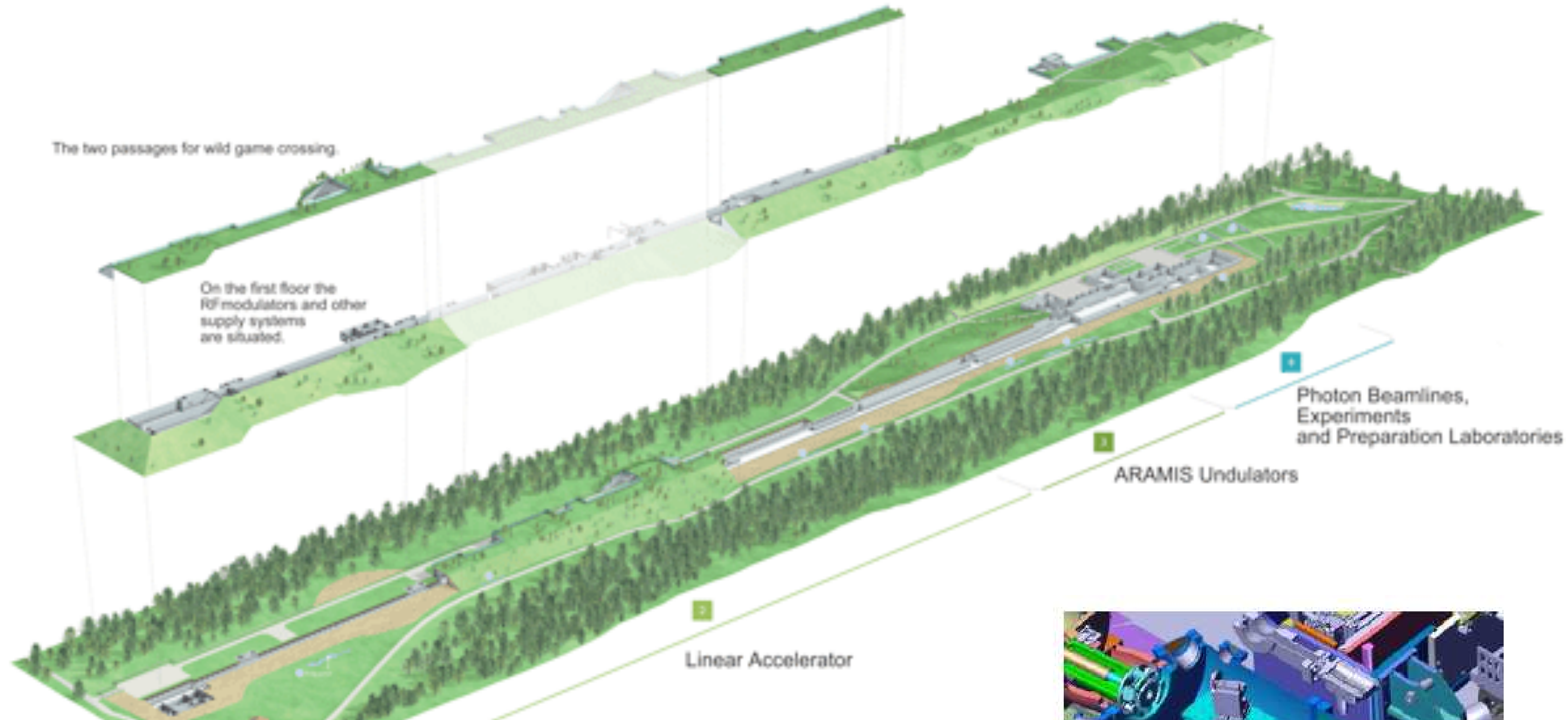
- Blue bronze: time-dependent free energy + damping

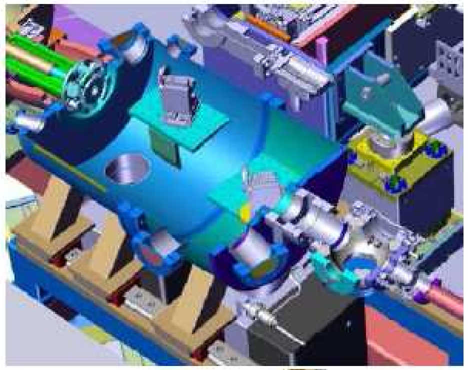


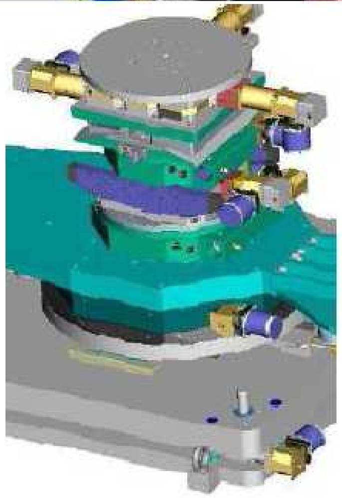


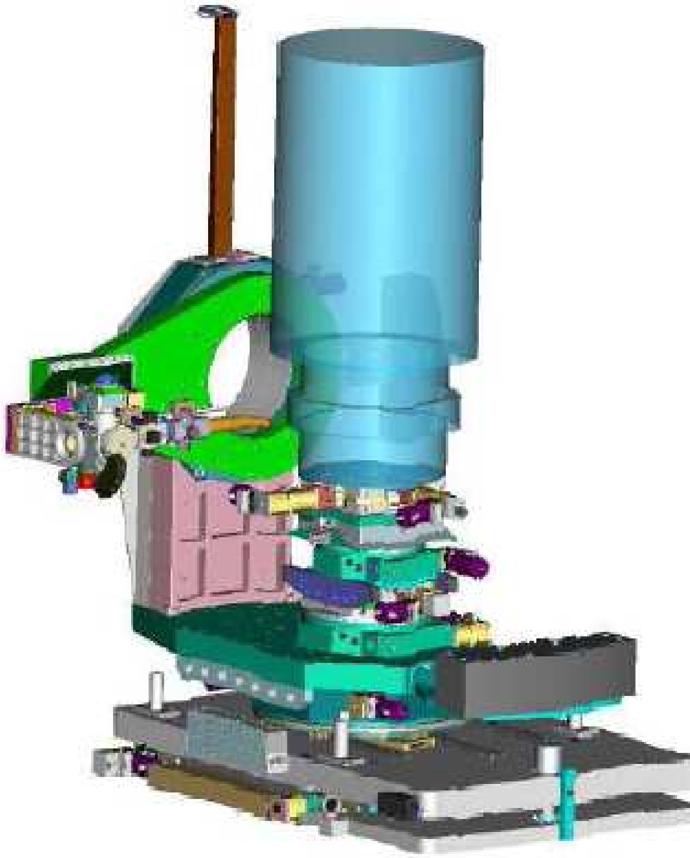
- $TbMnO_3$: Direct excitation of coherent electromagnon
  - See actual spin motions
  - Outlook: switching?

ESB station at SwissFEL









- Hard x-ray (4-12.4 keV)
- Time resolution to 10 fs
- Optimized THz pumping
- Support for low-T, high-B

[G. Ingold, P. Beaud]