Light-matter interactions using ultrashort ultra-bright X-ray pulses

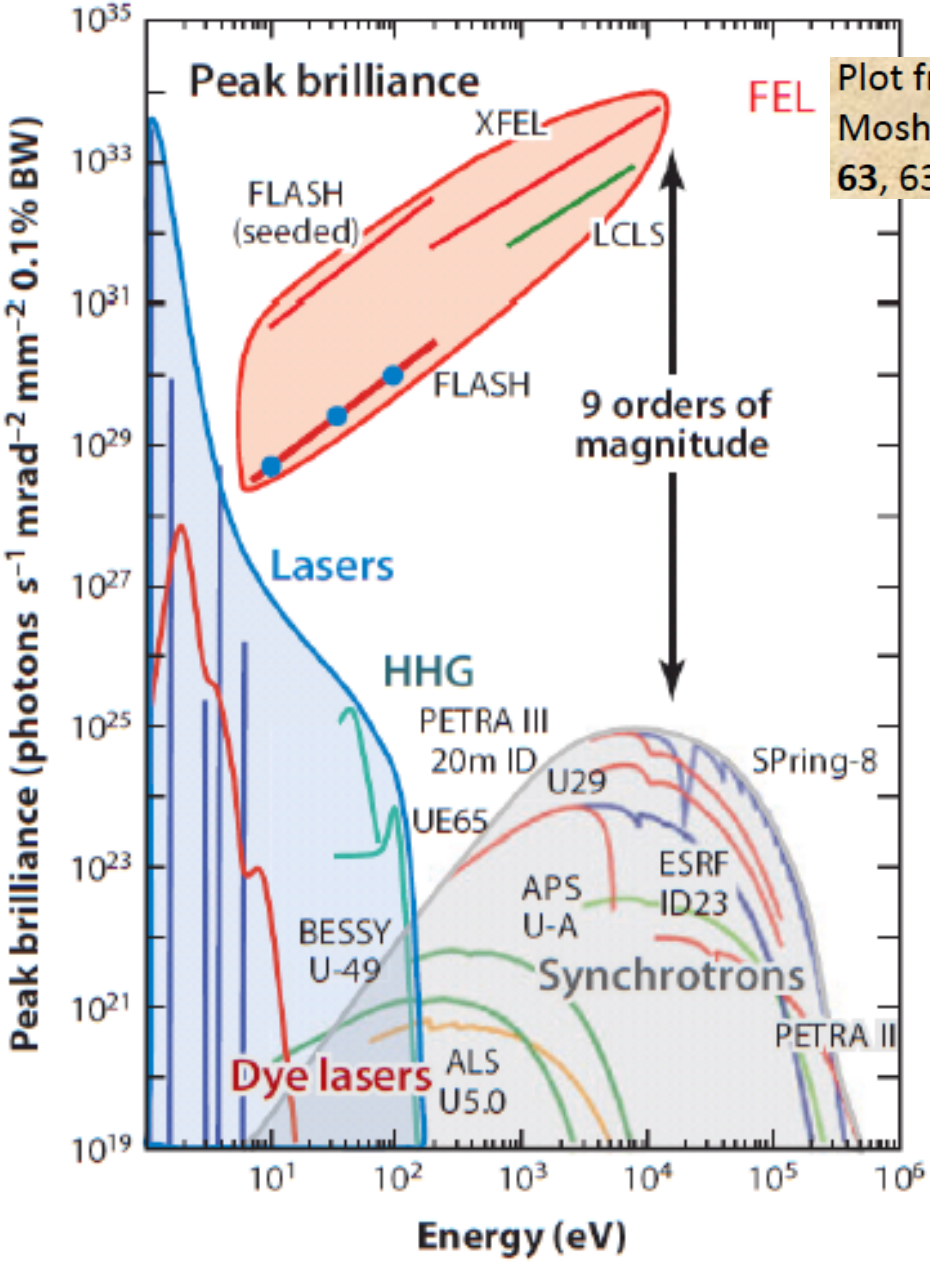
Massimo Altarelli

Max-Planck Institute for Structure and Dynamics of Matter
Hamburg, Germany

Overview

- Introduction to the worldwide X-ray Free-Electron-Laser facilities
- Examples of scientific applications of ultrabright, ultrashort (<100 fs) pulses
- Towards non-linear X-ray physics
- Conclusions

The promise of Free-Electron Lasers



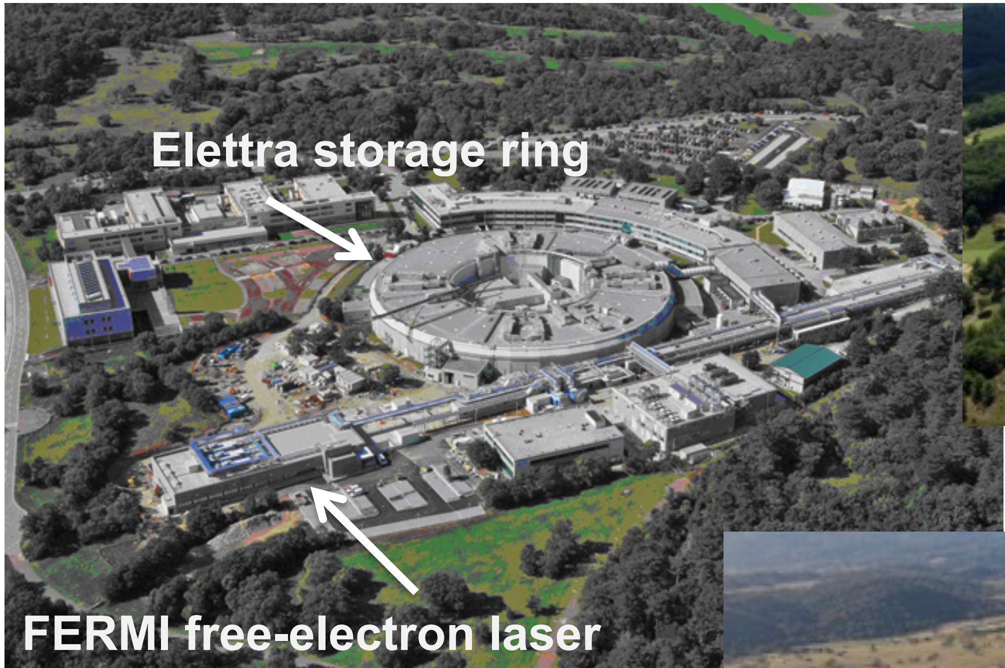
Plot from J. Ullrich, A. Rudenko, R. Moshammer, Ann. Rev. Phys. Chem. 63, 635 (2012)

Unprecedented peak brilliance

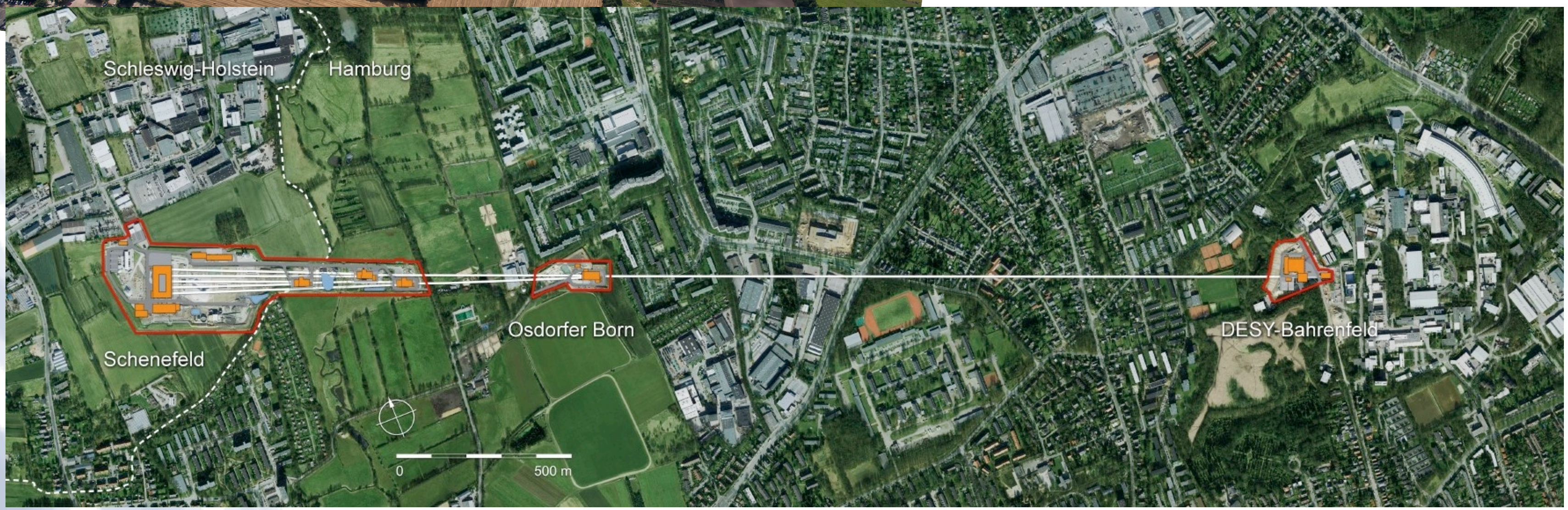
Delivered in pulses with ~10 to ~100 fs duration, with 10^{10} to 10^{14} photons / pulse

Revolution in time-dependent x-ray experiments

The promise of Free-Electron Lasers



The European XFEL, Hamburg region



X-ray FEL's worldwide I

Facility	FLASH, GERMANY	LCLS I, US	SACLA, JP	FERMI, ITALY	PAL-XFEL, KR	DCLS, DALIAN, China
Max. electron energy (GeV)	1.25	14.3	8.5	1.5	10	0.3
Wavelength range (nm)	3-55	0.1–4.4	0.06–0.3	4 - 100	0.06–10	50 - 150
Photons/pulse	~ 3 X 10^{13}	~ 10^{12}	2 x 10^{11}	10^{13} - 10^{14}	10^{11}–10^{13}	1.4 X 10^{14}
Peak brilliance	1 X 10^{31}	2 x 10^{33}	1 x 10^{33}	10^{31}	1.3 x 10^{33}	
Pulses/second	5000 - (8000)	120	60	10 (50)	60	1 - 50
Date of first beam	2000	2009	2011	2011	2016	2016/2017

X-ray FEL's worldwide, II

Facility	European XFEL	SWISS-FEL, CH	Shanghai FEL I, China	Shanghai FEL II, China	LCLS II, US	Shanghai SCRF
Max. electron energy (GeV)	17.5	5.8	0.8 GeV (1.6)	1.6	4 (=>8?)	8 ?
Wavelength range (nm)	0.05–4.7	0.1 – 7	3 – 10 (2 – 40)	1.2 – 10	0.25 – 4.7	
Photons/pulse	~ 10^{12}	~3.6 X 10^{10}			2 10^{11} – 2 10^{10}	
Peak brilliance	5 x 10^{33}	7 X 10^{32}				
Pulses/second	27 000	100	10 - 50	10 - 50	10^5 - 10^6	10^6
Date of first beam	2017	2017/18	2019	2019	2021	?

Free-electron laser “zoology”

- Besides the obvious difference in wavelength range, there are other important criteria differentiating the existing or projected FEL facilities
- Normal conducting vs. superconducting linac RF technology (low vs. high repetition rate)

Normal conducting vs superconducting RF Linacs

- Linacs with normal conducting RF have typically a repetition rate of order of 100 pulses / s
- Linacs with superconducting RF can have repetition rates from 10^3 up to 10^6 pulses/s
- Typical average brilliance:

Pulses/s

~ Peak Brilliance X Pulse duration X N/s

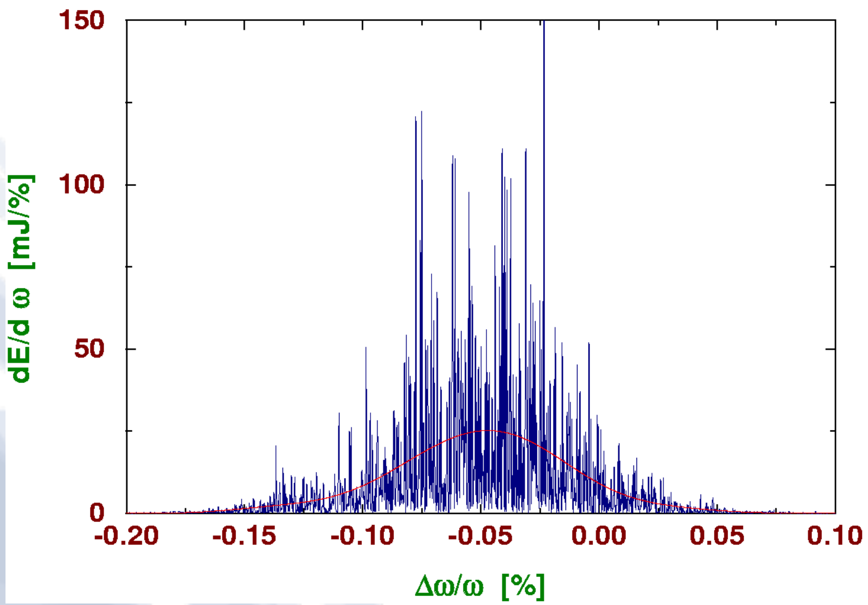
~ 10^{32} X 10^{-13} X N/s photons/mm^2/mrad^2/0.1%BW

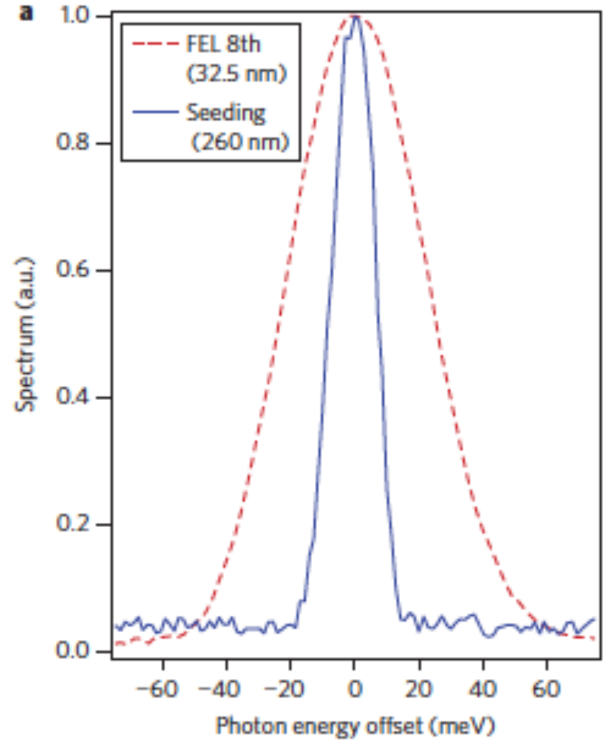
Free-electron laser "zoology", cont.

- SASE (Self-Amplified Spontaneous Emission) vs. "Seeding": longitudinal coherence, shot to shot reproducibility.

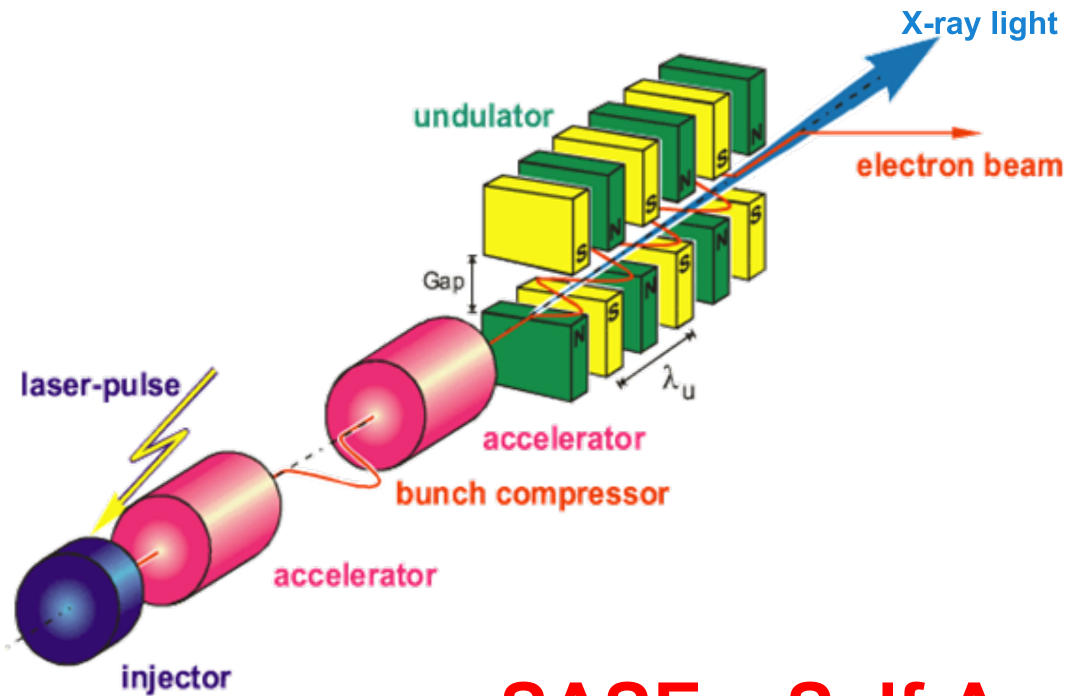
Seeded

SASE





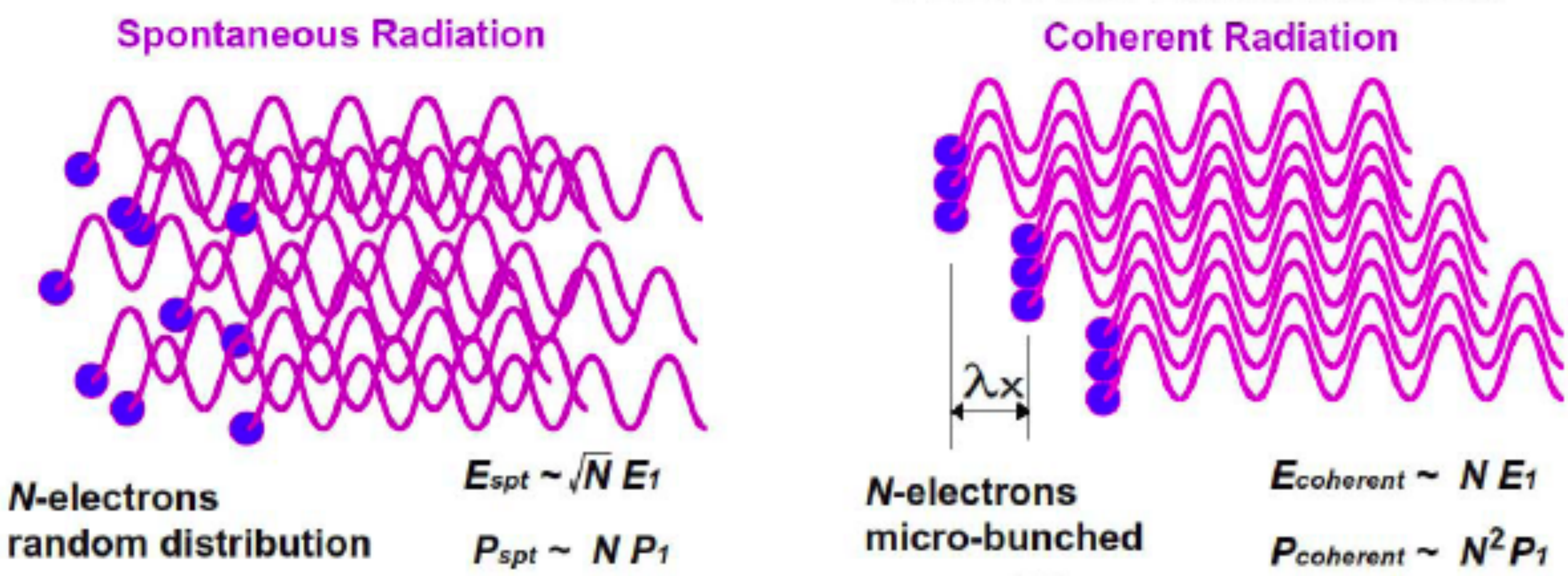
Free Electron Lasers



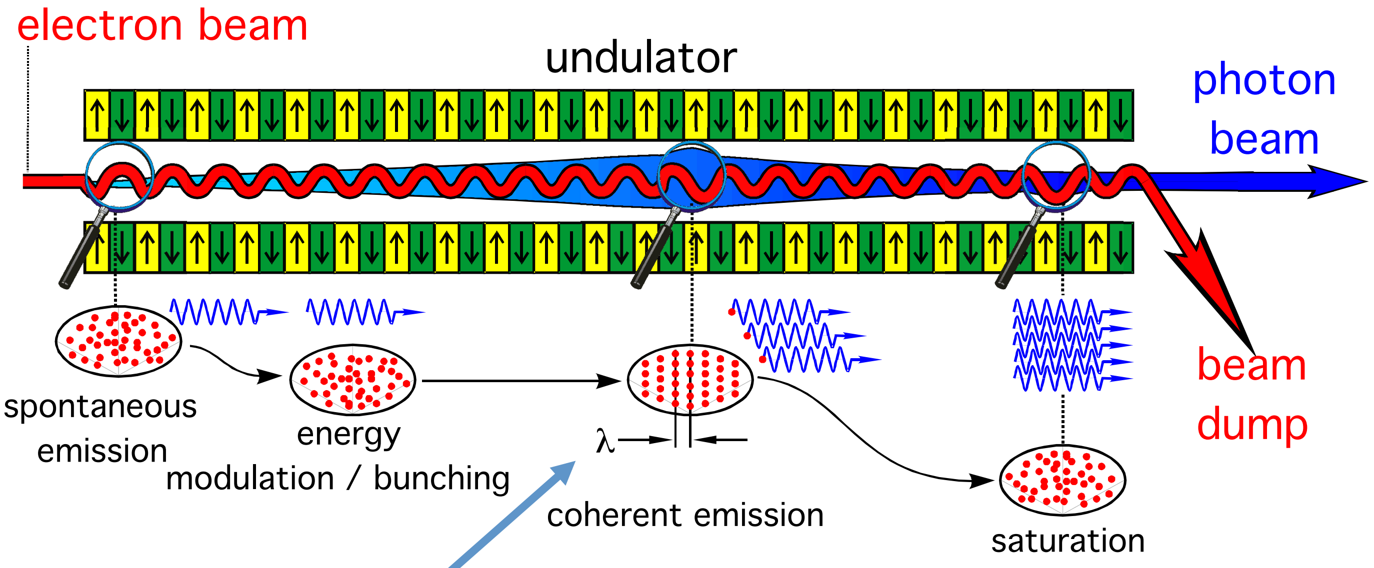
SASE – Self-Amplified Spontaneous Emission

Kondratenko, Saldin (1979)
Bonifacio, Pellegrini, Narducci (1984)

Spontaneous (synchrotron) vs coherent (FEL) radiation



Self-Amplified Spontaneous Emission (SASE)

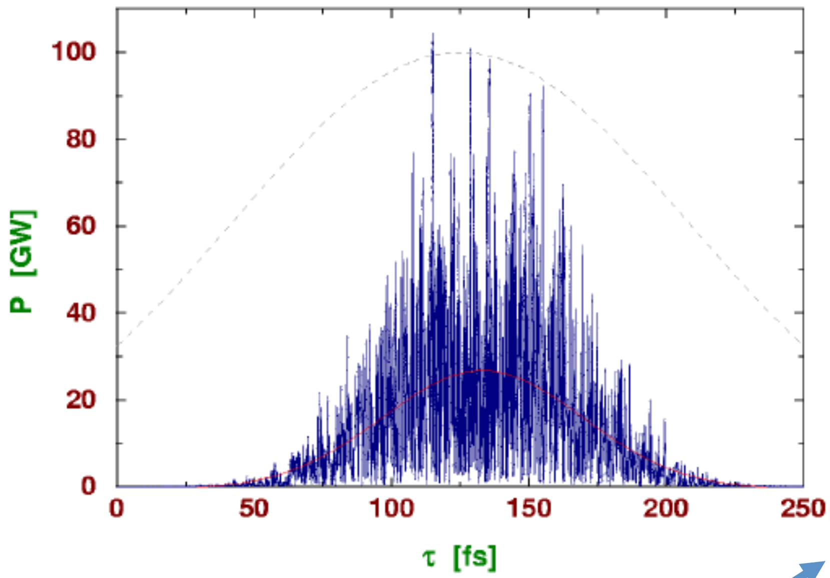


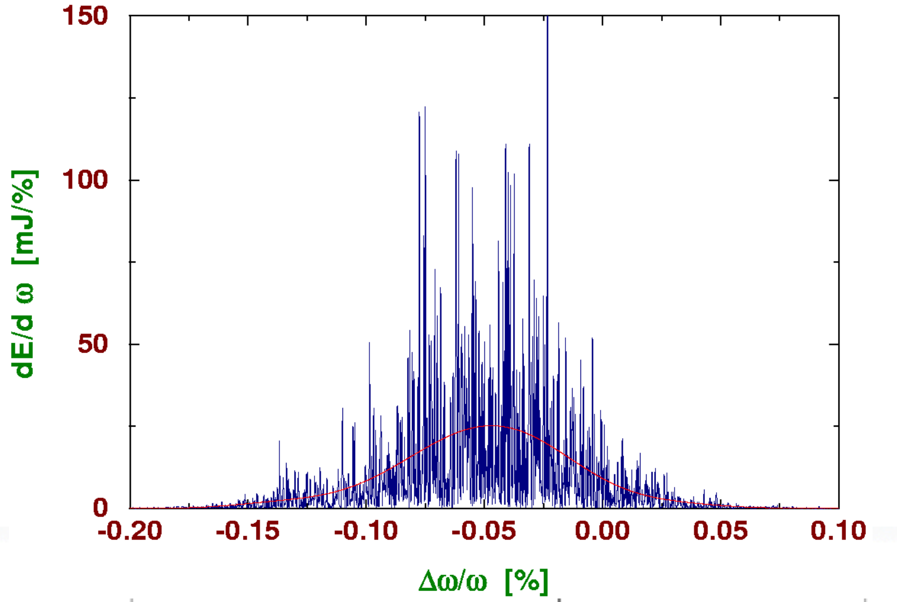
This cartoon is too optimistic! Cannot have a single coherent domain. Reason: light from the back of the bunch travelling throughout undulator can only influence electrons ahead over a „cooperation length" N_{und} λ, where N_{und} is the number of periods in undulator (typically several thousands) and N_{und} λ << bunch length.

European XFEL: N_{und} ~ 4 000, bunch length up to 45 μm, λ ~ 0.1 nm

So, number of incoherent domains ~ 45 μm / (4 000 0.1 nm) ~ 100 domains

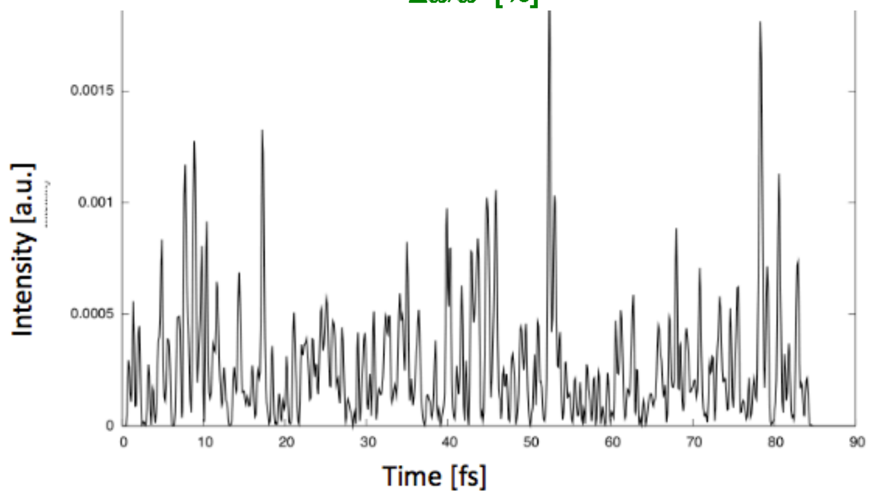
Simulations of SASE FEL's





European XFEL at
$\lambda \sim 0.1$ nm (M. Yurkov)

LCLS at $\lambda \sim 1$ nm



LCLS bandwidth at 1keV photon energy: $\Delta\omega$ = 6-9 eV
Coherence time: 0.3 - 0.5 fs

Properties of SASE and of seeded FEL

- SASE FEL radiation has excellent transverse coherence properties but very poor longitudinal coherence properties

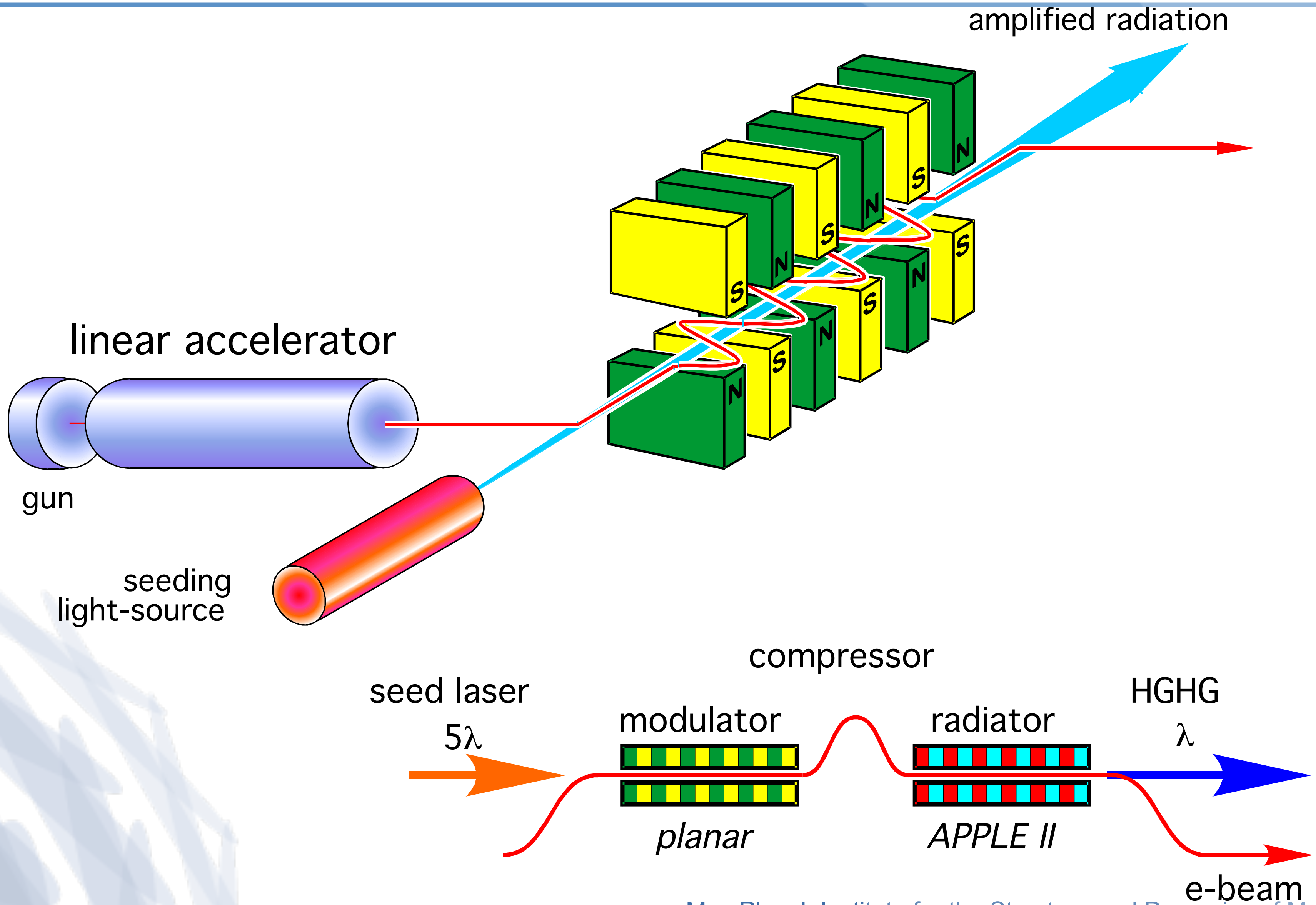
  (longitudinal coherence = monochromaticity)

- Poor reproducibility shot to shot (intensity, spectral distribution, spike structure, pointing direction)

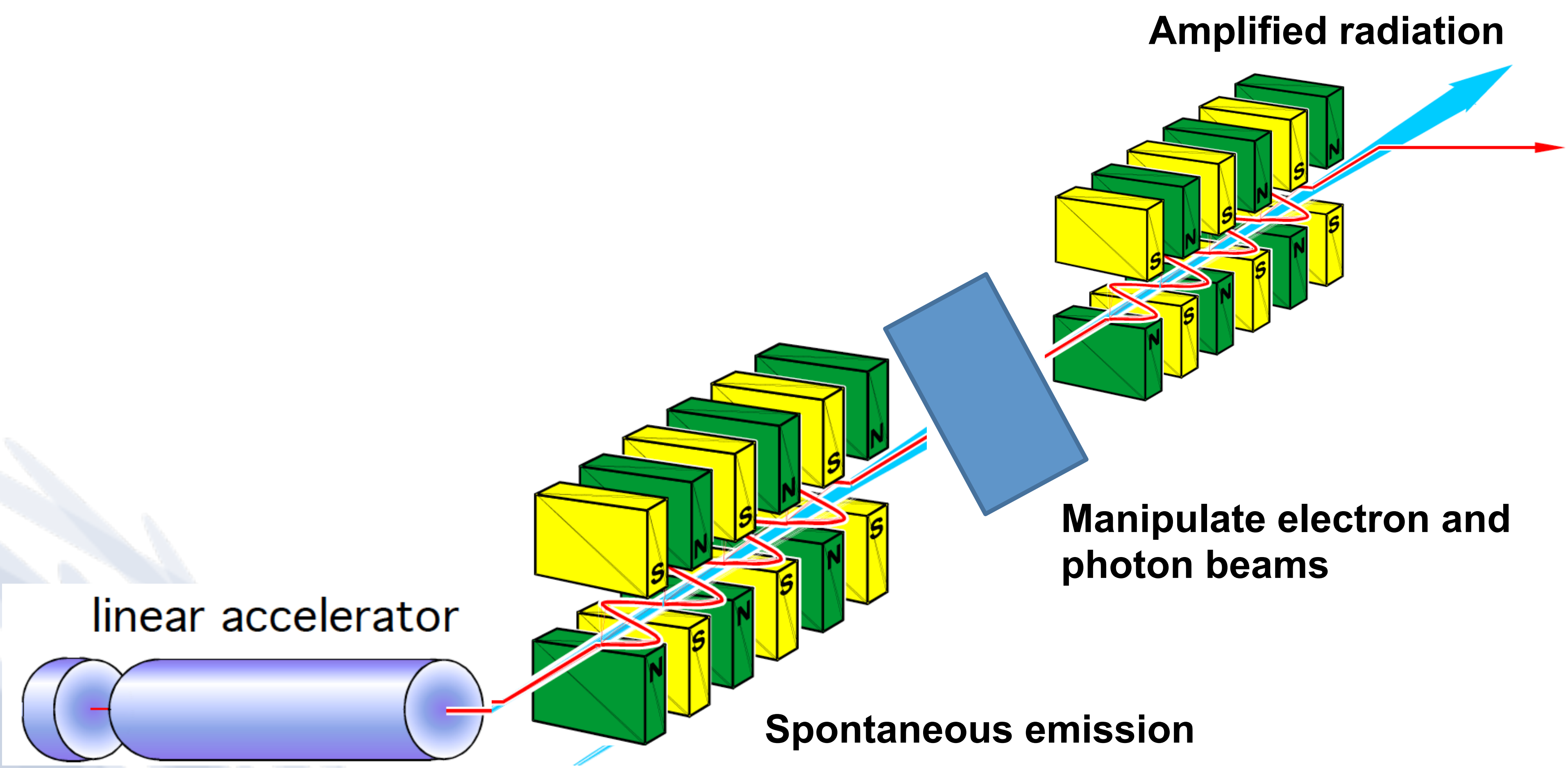
- Cure for longitudinal coherence = “Seeding” by external laser source (VUV to very soft x-rays) or by “Self seeding” (harder x-rays). Produces high spectral stability, intensity fluctuations still present

„Seeding“ by High Gain Harmonic Generation

mpsd



„Self-seeding“ for shorter wavelengths x-rays



Seeding with 260 nm laser

mpsd

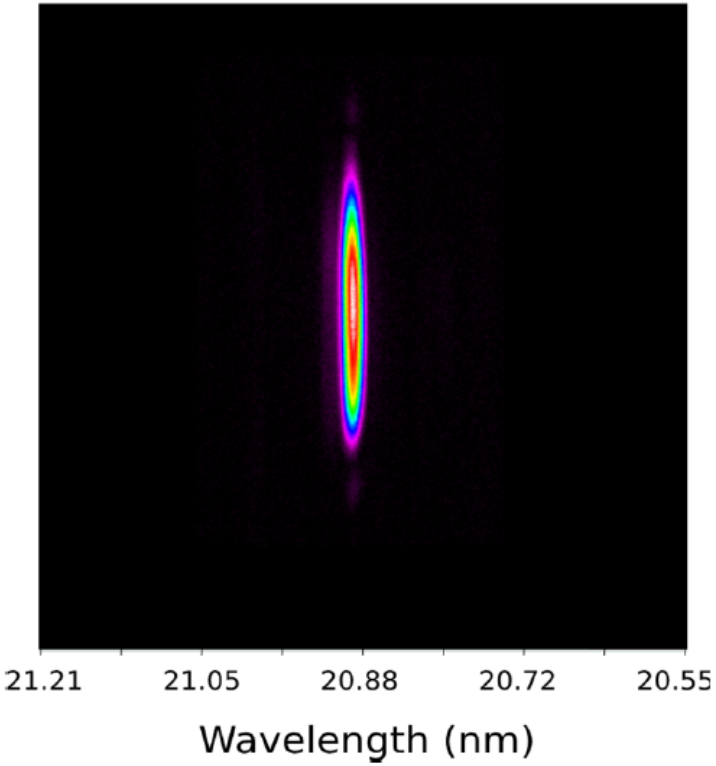
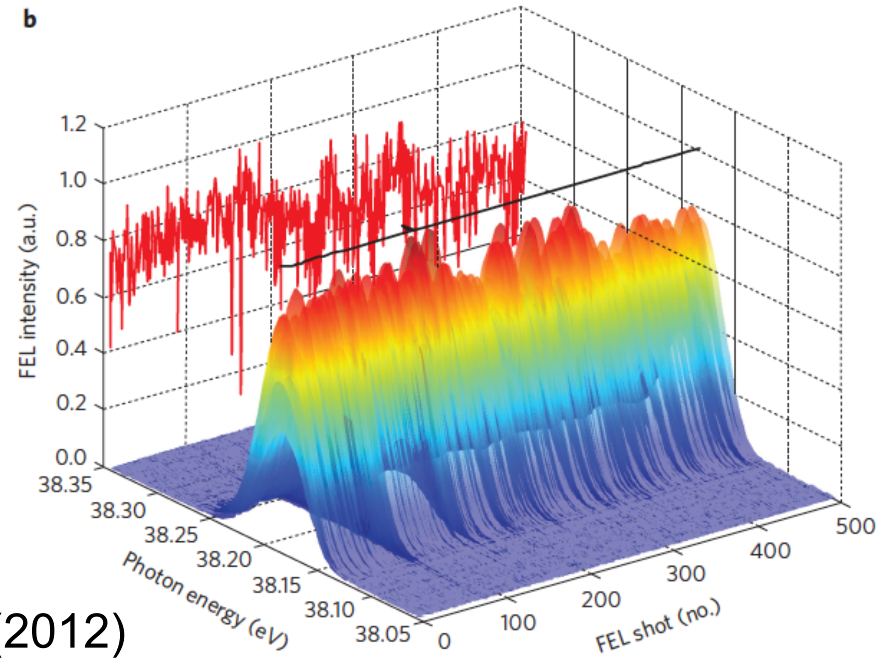


Figure 1: Spectral Line of FERMI FEL-1 at Harmonic 12.

FERMI@ Elettra, Trieste

L. Giannessi *et al*.,
Proceedings of FEL2017
Paper MOD04



E. Allaria *et al*., Nature Photonics **6**, 699 (2012)

X-ray FEL's, present and future

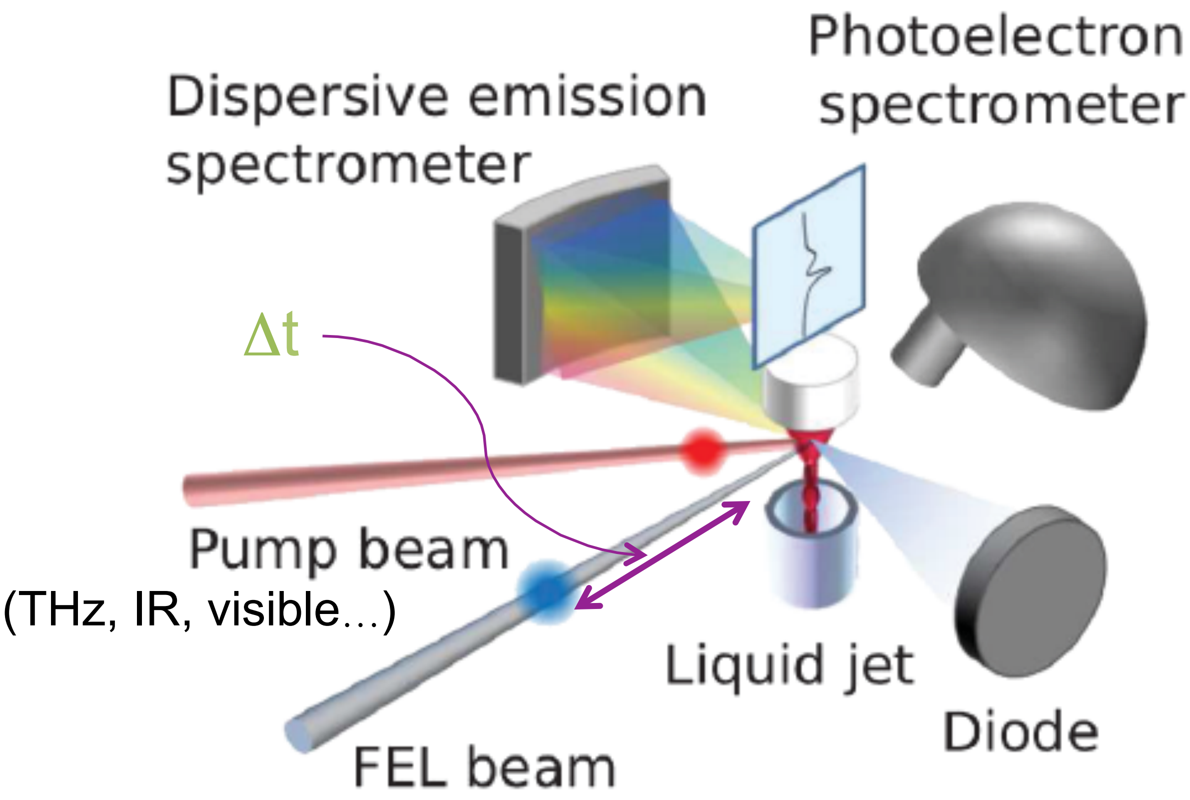
	SASE, possibly self-seeded	Seeded
Normal Cond.	LCLS I, SACLA, PAL-XFEL, Swiss-FEL	FERMI@Elettra, Dalian CLS, Shanghai FEL I Shanghai FEL II?
Supercond.	FLASH, European XFEL, LCLS II, LCLS II (HE), Shanghai SCRF-FEL	FLASH upgrade?

Scientific interest of ultrabright, ultrashort (<100 fs) pulses

- “Molecular movies”
- Beating radiation damage: ”Diffraction before Destruction”
- “Snapshot” view of the liquid state

"Pump – probe" experiments

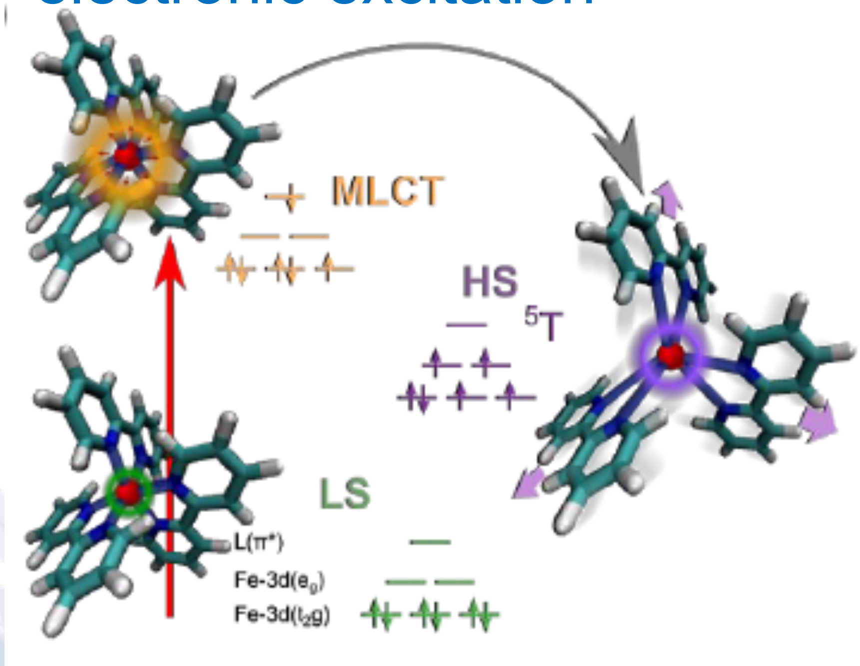
- Interrogate system by the XFEL beam, at time Δt after triggering a process by the pump optical laser beam.
- By varying pump-probe delay Δt record a "molecular movie"



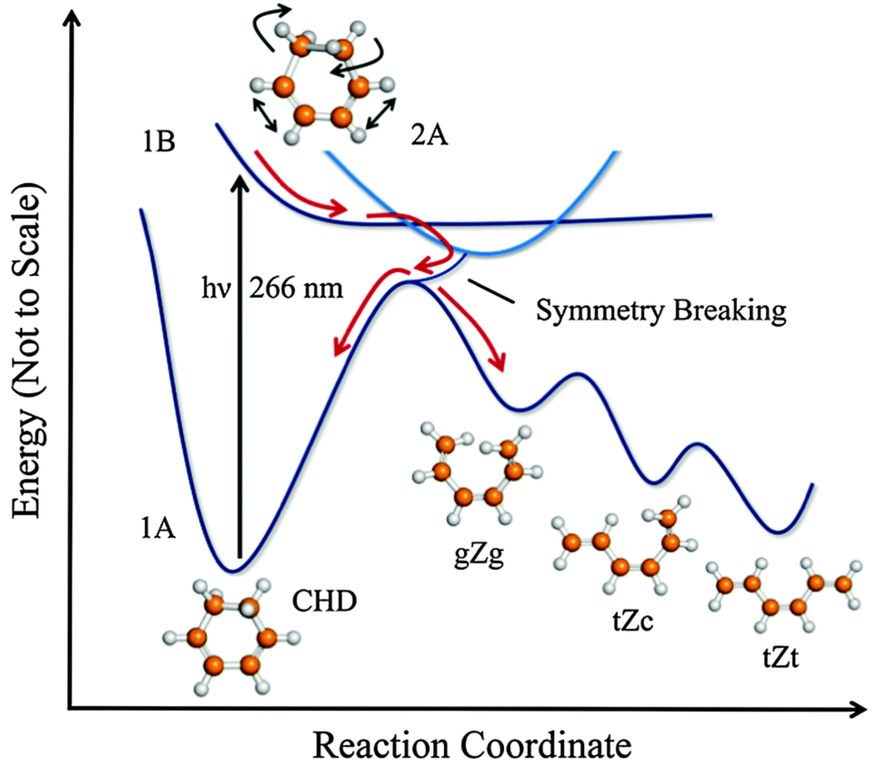
(After C. Bostedt et al., Rev. Mod. Phys. ***88****, 015007 (2016))*

Molecular Movies

Low-spin – High-spin transition in TM bypiridines via electronic excitation



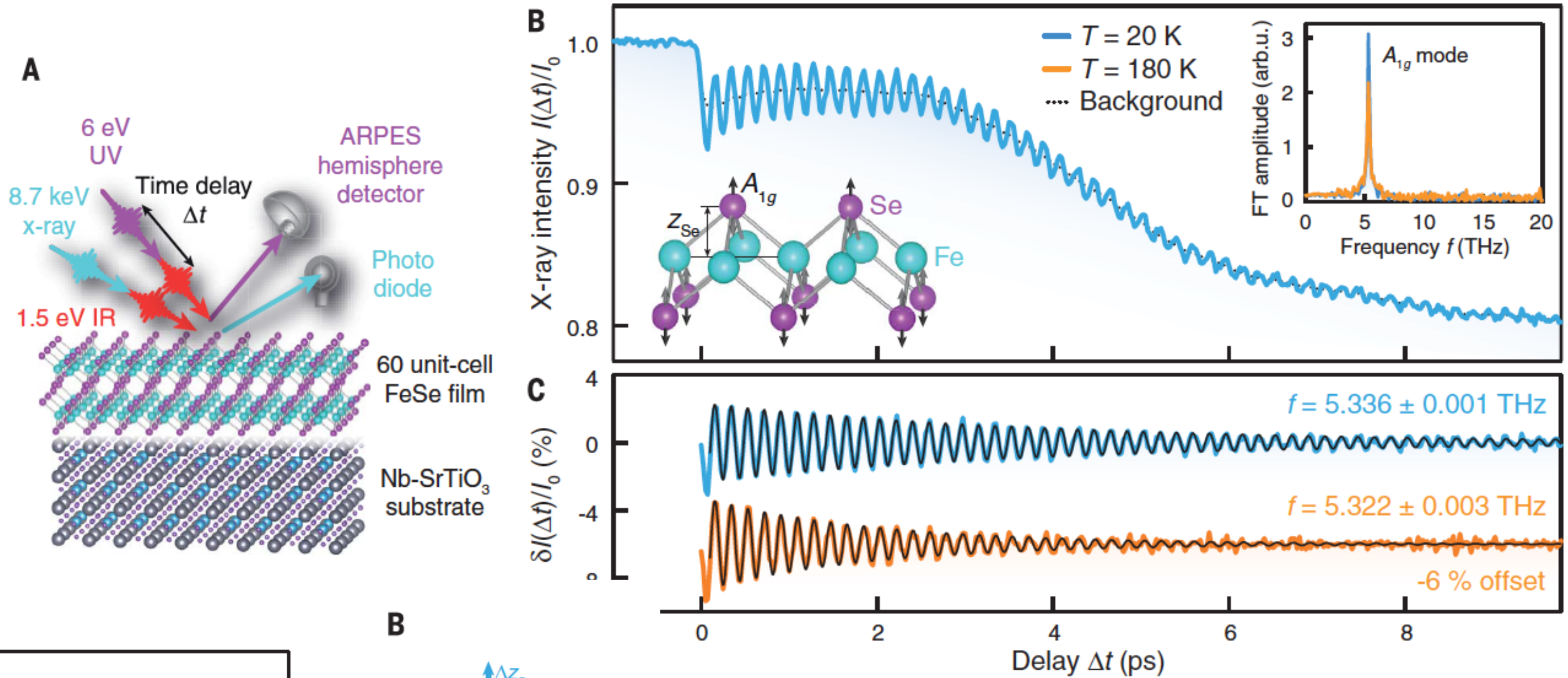
Laser induced isomerization of cyclohexadyene (CHD)



Time resolution affected by a) pulse duration

b) synchronization & jitter

Realistic limit ~ 50 fs



Femtosecond electron-phonon lock-in by photoemission and x-ray free-electron laser

S. Gerber,[1,2]* S.-L. Yang,[1,3]* D. Zhu,[4] H. Soifer,[1] J. A. Sobota,[1,5] S. Rebec,[1,3] J. J. Lee,[1,3] T. Jia,[1,3] B. Moritz,[1] C. Jia,[1] A. Gauthier,[1,3] Y. Li,[1] D. Leuenberger,[1] Y. Zhang,[6] L. Chaix,[1] W. Li,[1] H. Jang,[7] J.-S. Lee,[7] M. Yi,[8] G. L. Dakovski,[4] S. Song,[4] J. M. Glownia,[4] S. Nelson,[4] K. W. Kim,[9] Y.-D. Chuang,[5] Z. Hussain,[5] R. G. Moore,[1] T. P. Devereaux,[1] W.-S. Lee,[1]† P. S. Kirchmann,[1]† Z.-X. Shen[1,3]†

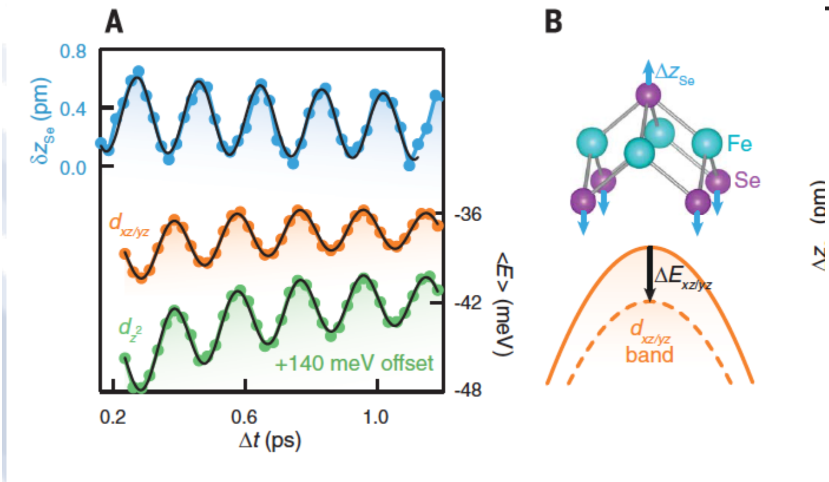


Fig. 3. Coherent lock-in at the A_{1g} frequency. (**A**) Oscillations of the selenium displacement δz_{Se} (blue) and the momentum-averaged energy shift

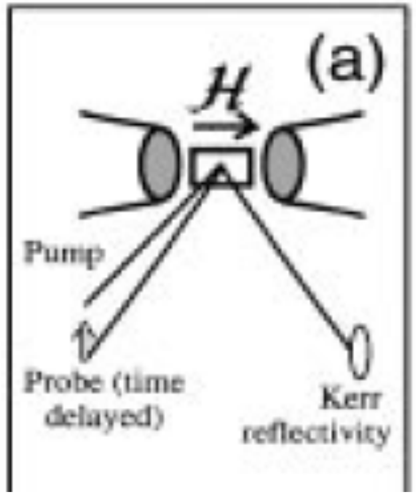
Ultrafast demagnetization by fs laser pulses: a puzzle since more than 20 years

mpsd

VOLUME 76, NUMBER 22 PHYSICAL REVIEW LETTERS 27 MAY 1996

Ultrafast Spin Dynamics in Ferromagnetic Nickel

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot
Institut de Physique et Chimie des Matériaux de Strasbourg, Unité Mixte 380046 CNRS-ULP-EHICS, 23, rue du Loess, 67037 Strasbourg Cedex, France
(Received 17 October 1995)

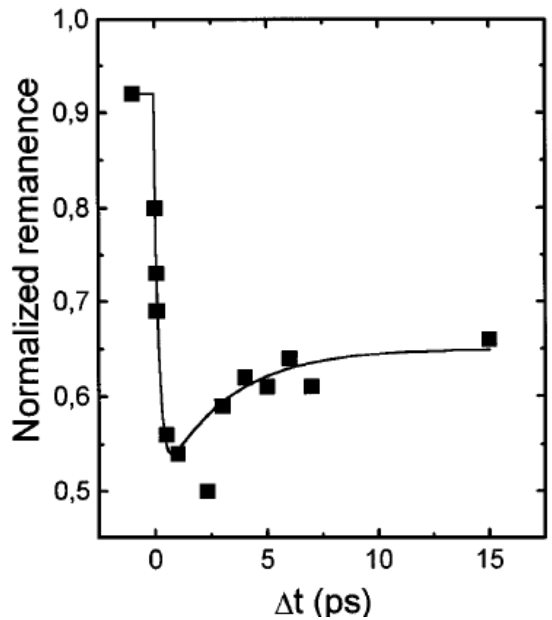


Pump: 60 fs laser

Two time scales:

One is << *1 ps*

One is ~ *10 ps*



Ferrimagnetic Iron-Gadolinium

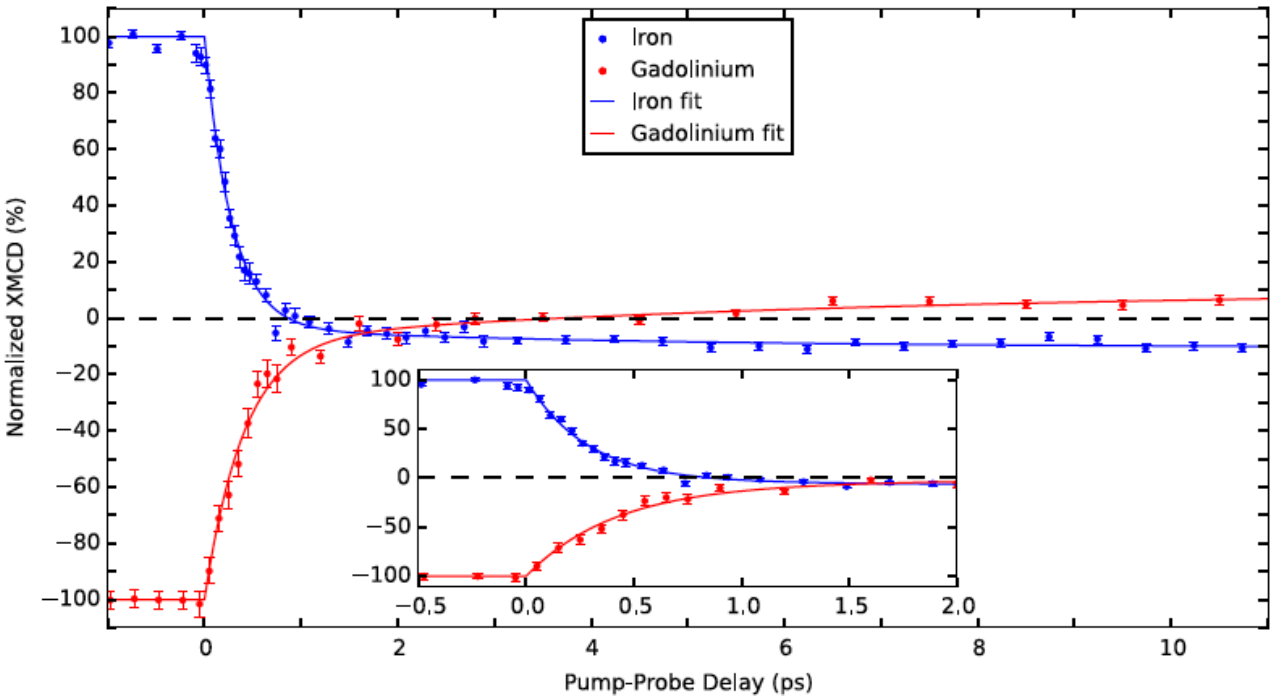
mpsd

LCLS, SLAC
Stanford

REVIEW OF SCIENTIFIC INSTRUMENTS **87**, 033110 (2016)

Femtosecond X-ray magnetic circular dichroism absorption spectroscopy at an X-ray free electron laser

Daniel J. Higley,[1,2,a)] Konstantin Hirsch,[1] Georgi L. Dakovski,[1] Emmanuelle Jal,[1] Edwin Yuan,[1,2] Tianmin Liu,[1,3] Alberto A. Lutman,[1] James P. MacArthur,[1,3] Elke Arenholz,[4] Zhao Chen,[1,3] Giacomo Coslovich,[1] Peter Denes,[4] Patrick W. Granitzka,[1,5] Philip Hart,[1] Matthias C. Hoffmann,[1] John Joseph,[4] Loïc Le Guyader,[1,6,7] Ankush Mitra,[1] Stefan Moeller,[1] Hendrik Ohldag,[1] Matthew Seaberg,[1] Padraic Shafer,[4] Joachim Stöhr,[1] Arata Tsukamoto,[8] Heinz-Dieter Nuhn,[1] Alex H. Reid,[1] Hermann A. Dürr,[1] and William F. Schlotter[1]



$Gd_{24}Fe_{66.5}Co_{9.5}$

410 fs, 6.5 ps

280 fs, 4.1 ps

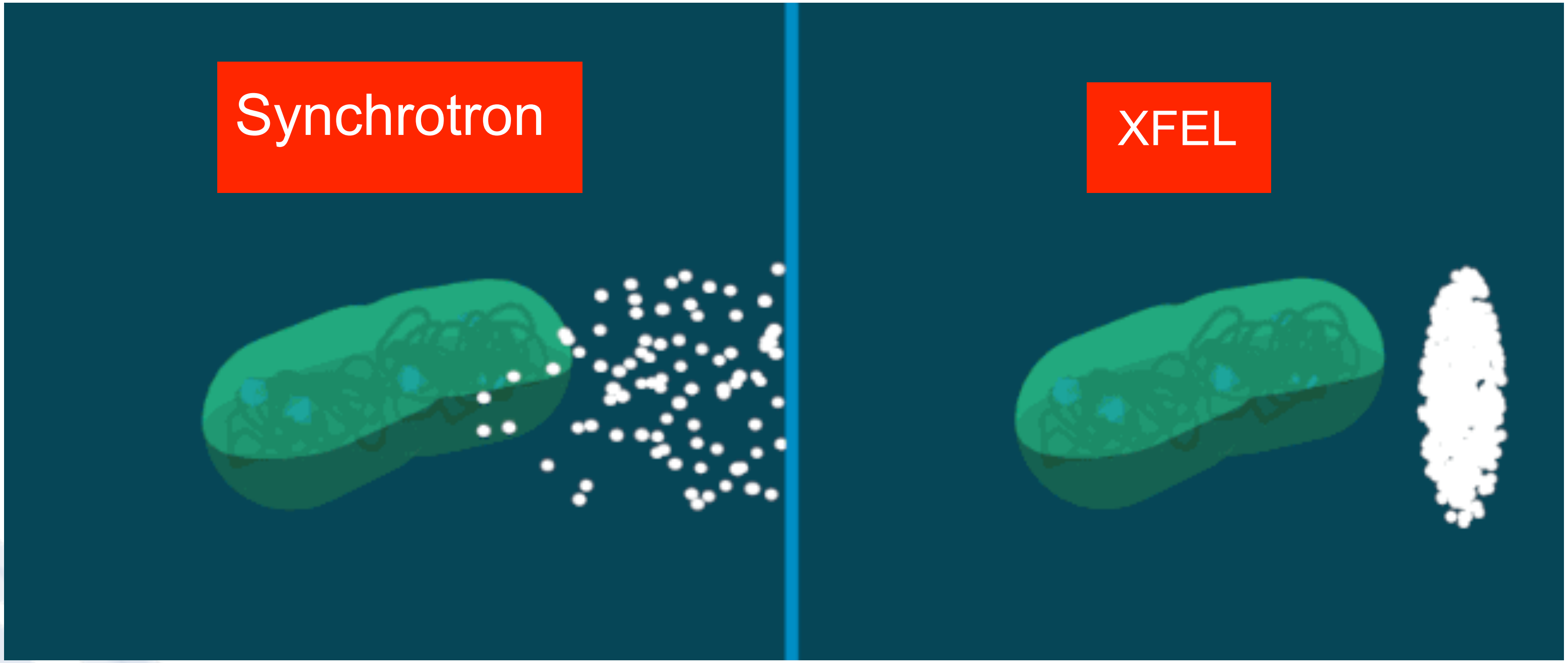
the Structure and Dynamics of Matter

Beating radiation damage: Diffraction before Destruction

mpsd

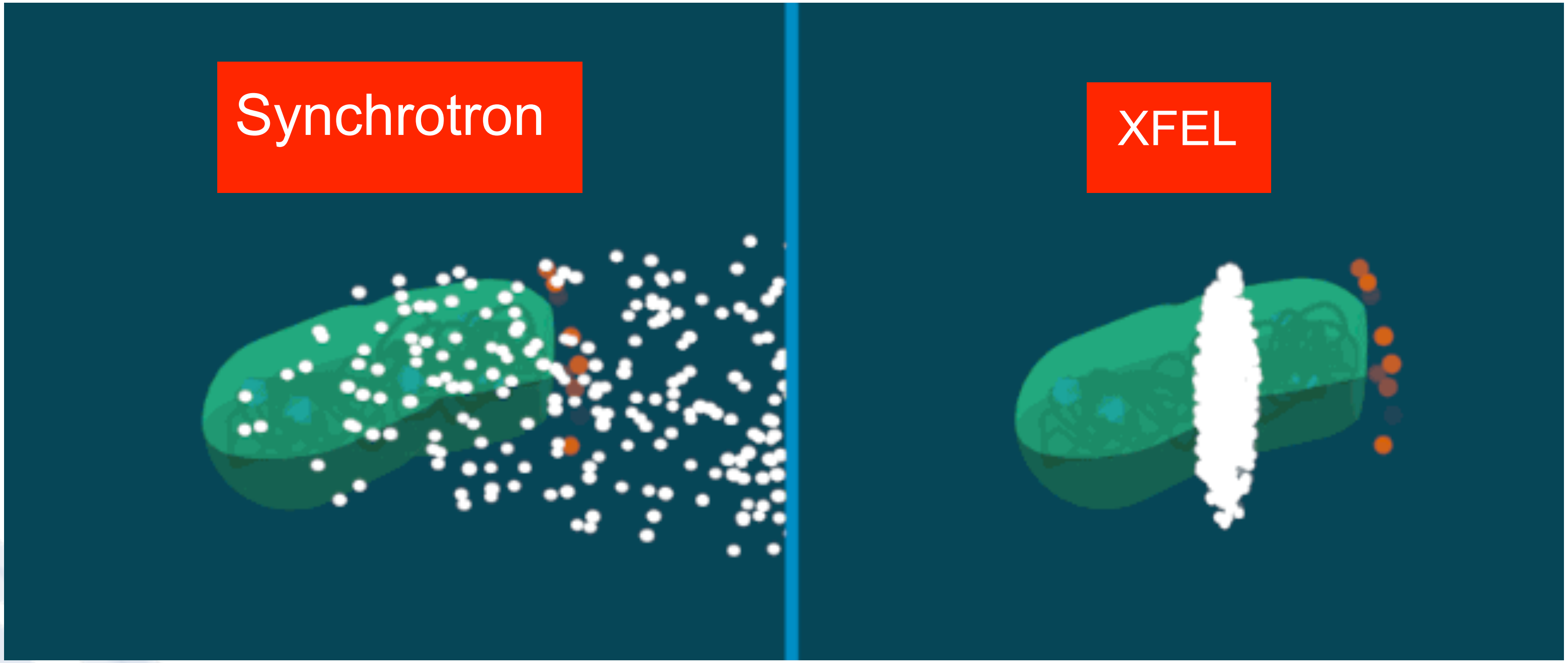
Credits: www.slac.stanford.edu

Beating radiation damage: Diffraction before Destruction



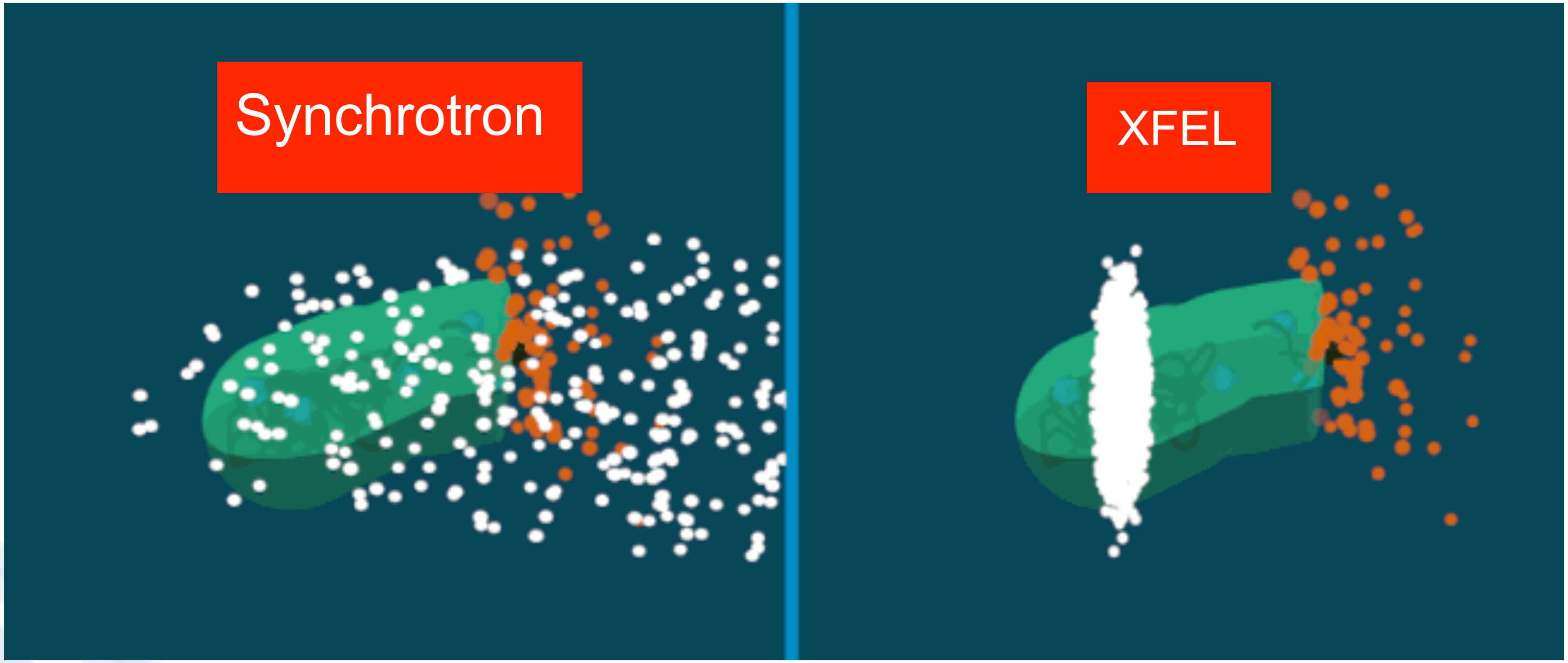
Credits: www.slac.stanford.edu

Beating radiation damage: Diffraction before Destruction



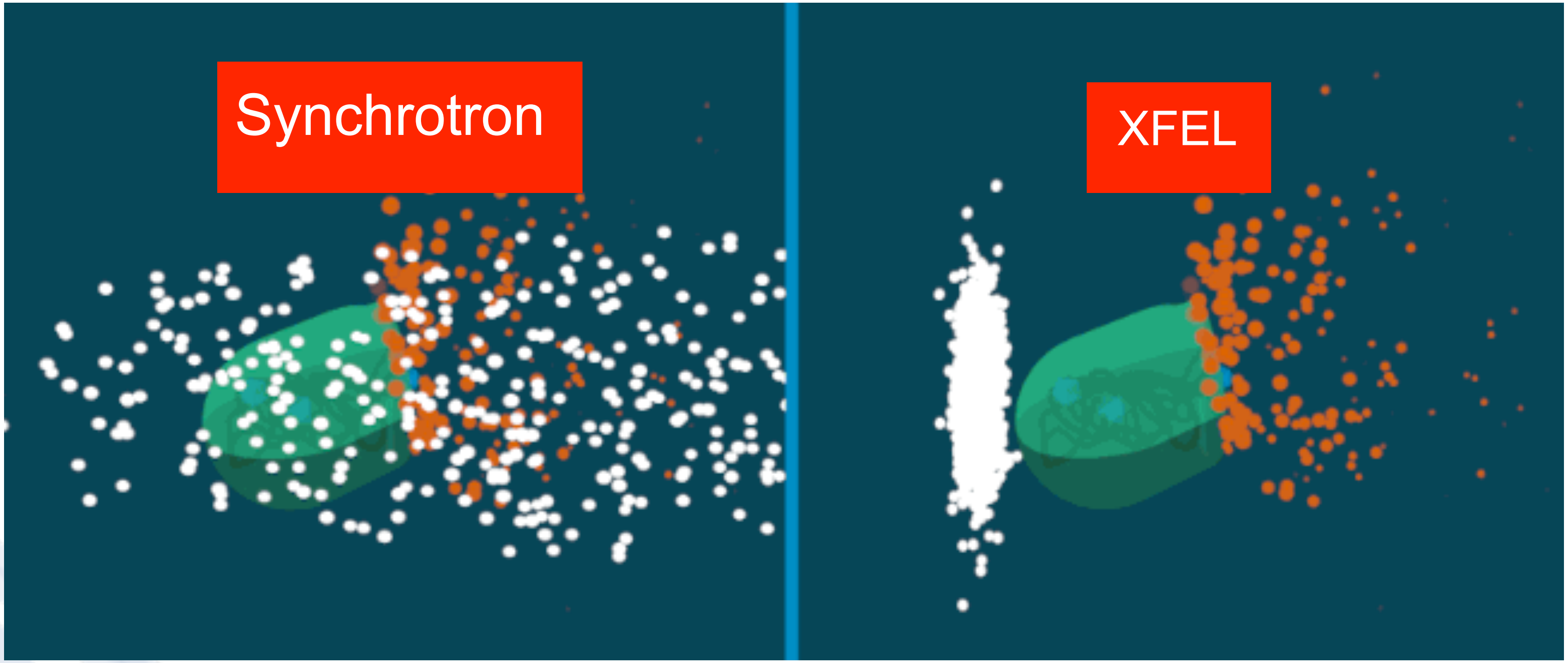
Credits: www.slac.stanford.edu

Beating radiation damage: Diffraction before Destruction

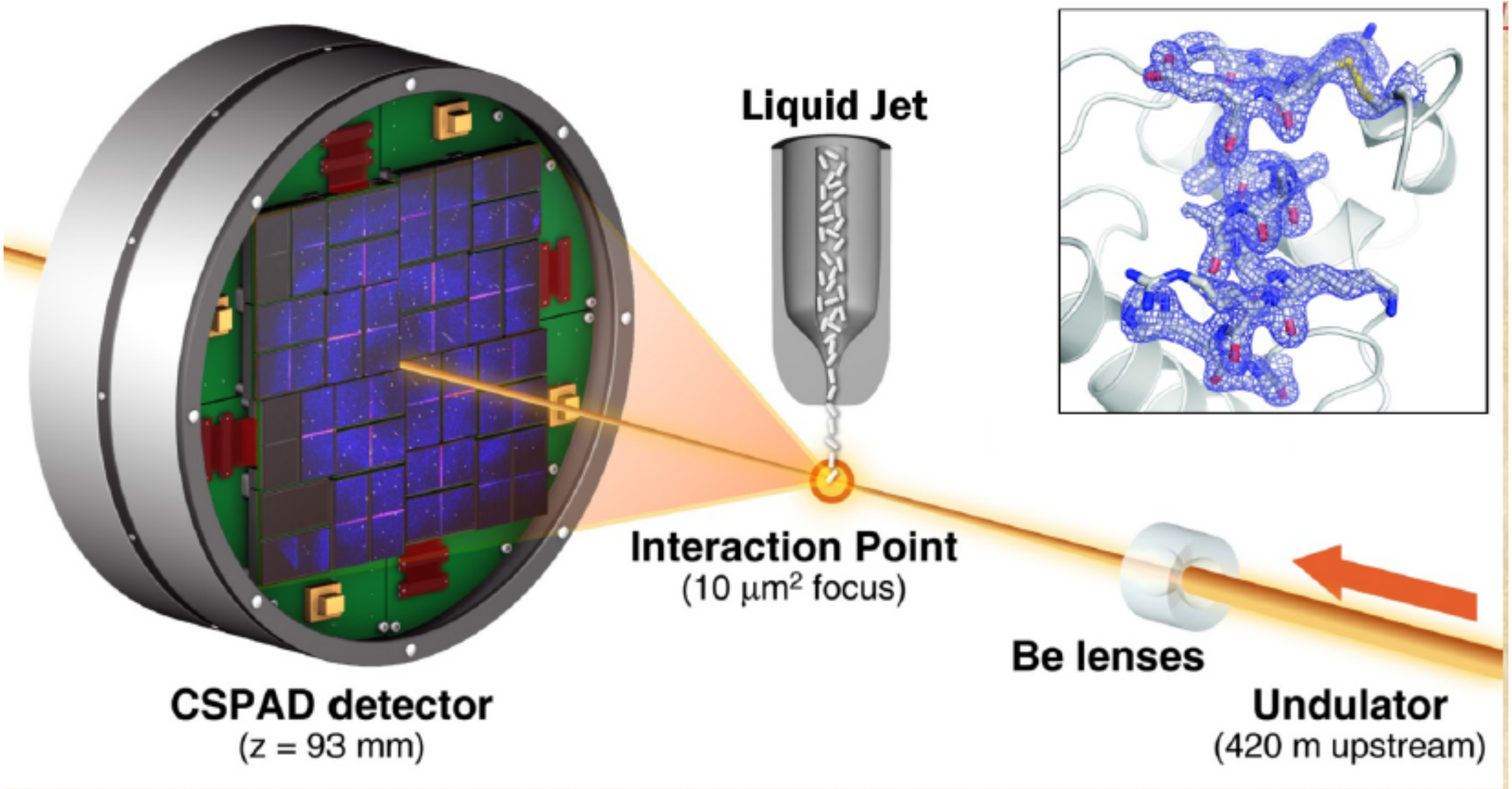


Credits: www.slac.stanford.edu

Beating radiation damage: Diffraction before Destruction

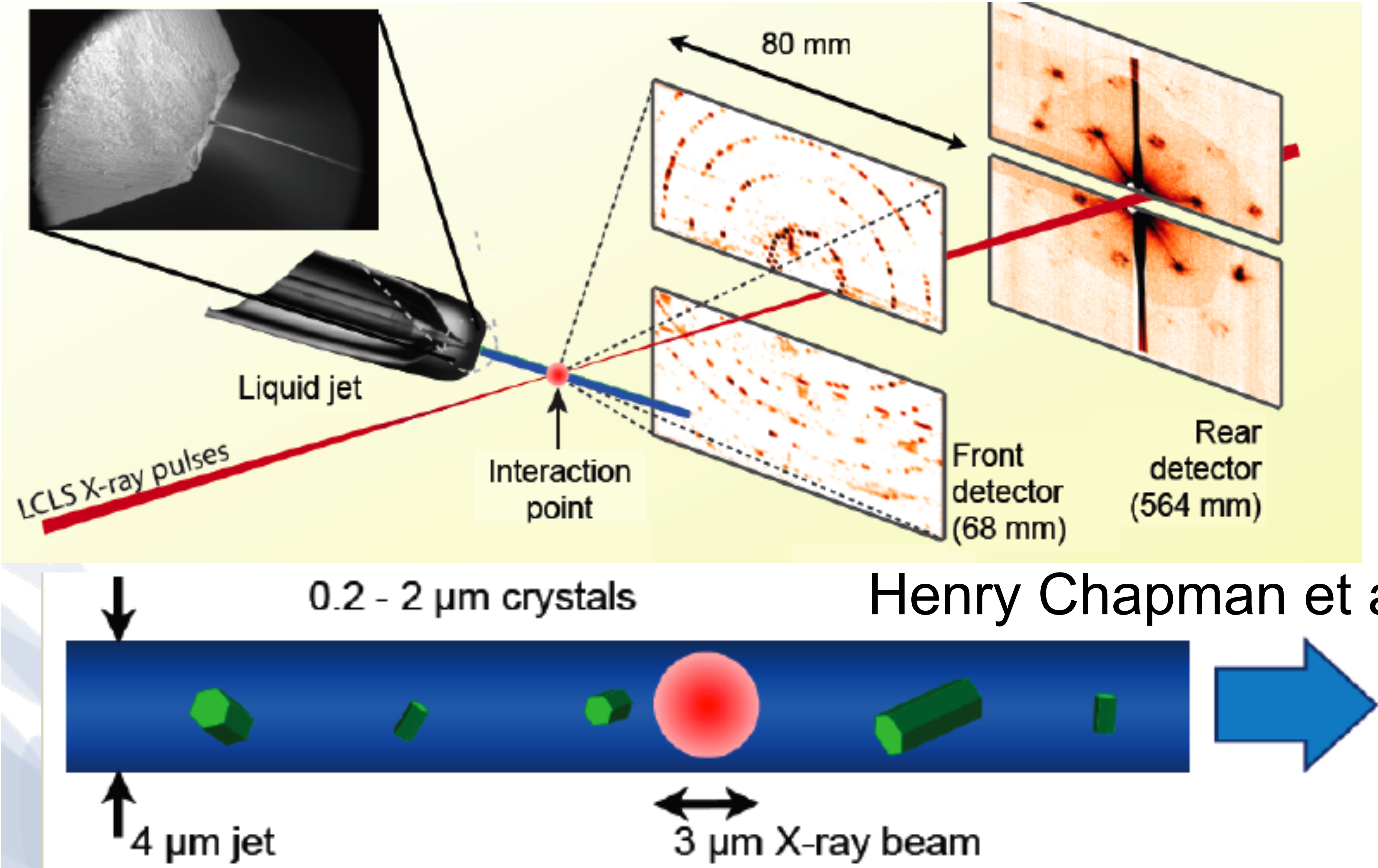


Credits: www.slac.stanford.edu



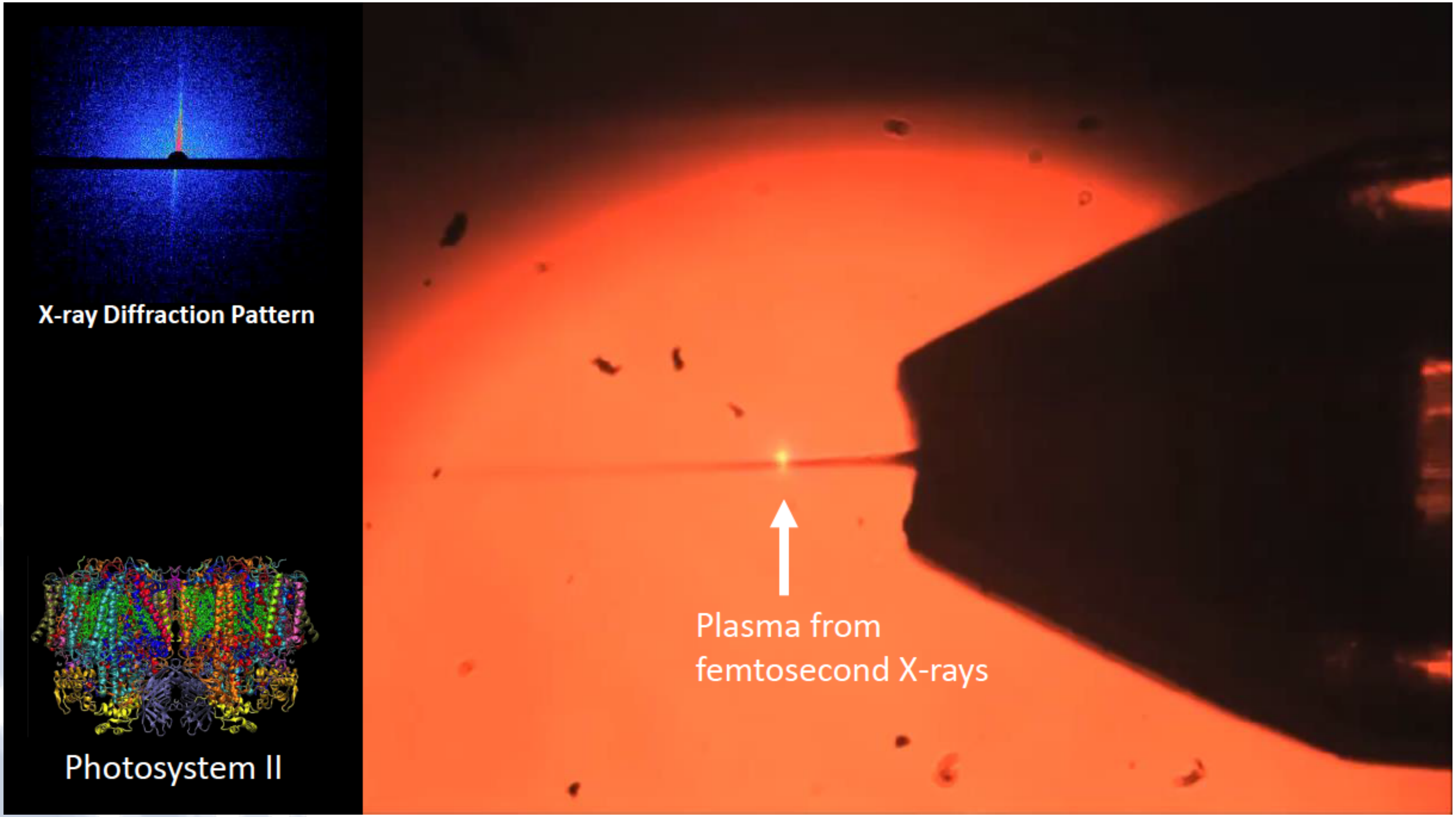
Boutet et al., *Science*, 337, 362 (2012)

Serial Femtosecond crystallography



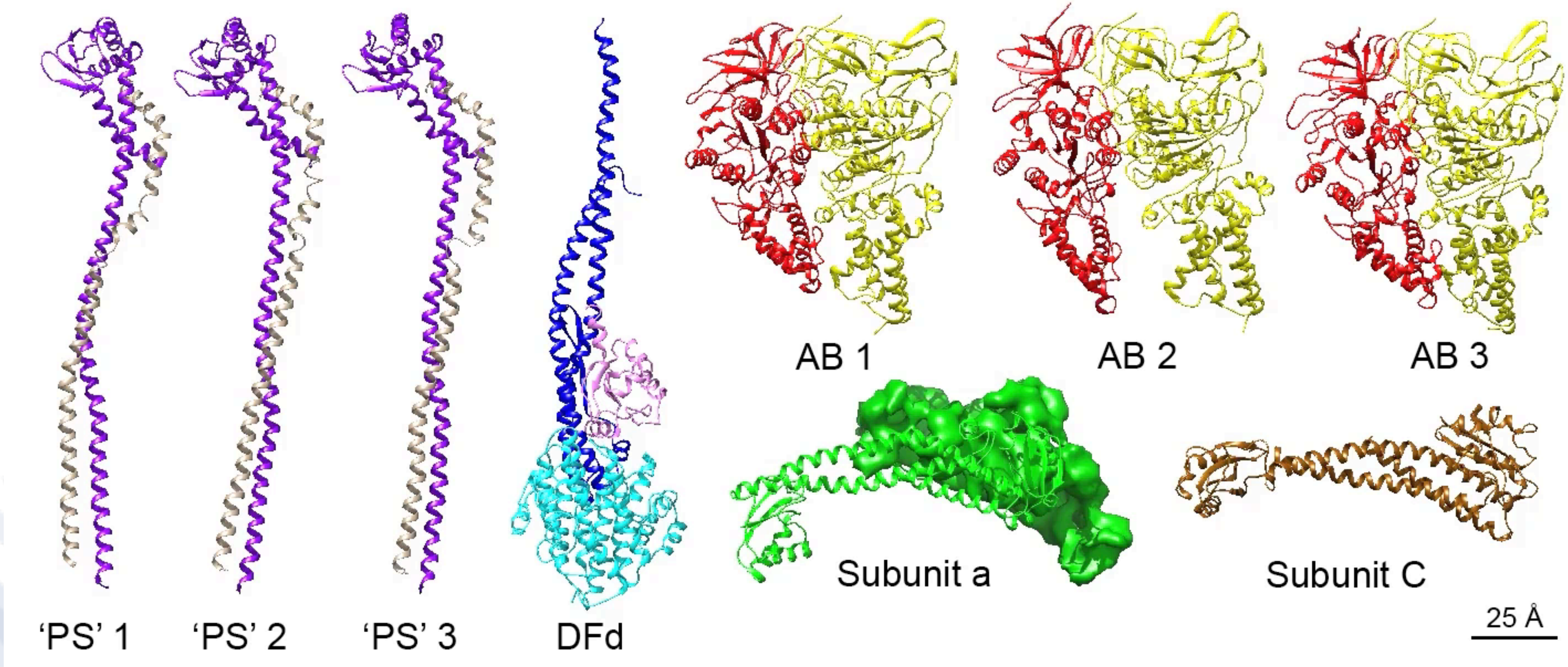
Henry Chapman et al.

Actual LCLS beamtime photo



Credit: Josè M. Martin Garcia, ASU

“Movie” of V-ATPase



Zhao, J., Benlekbir, S. & Rubinstein, J. L. *Nature* **521**, 241–245 (2015)

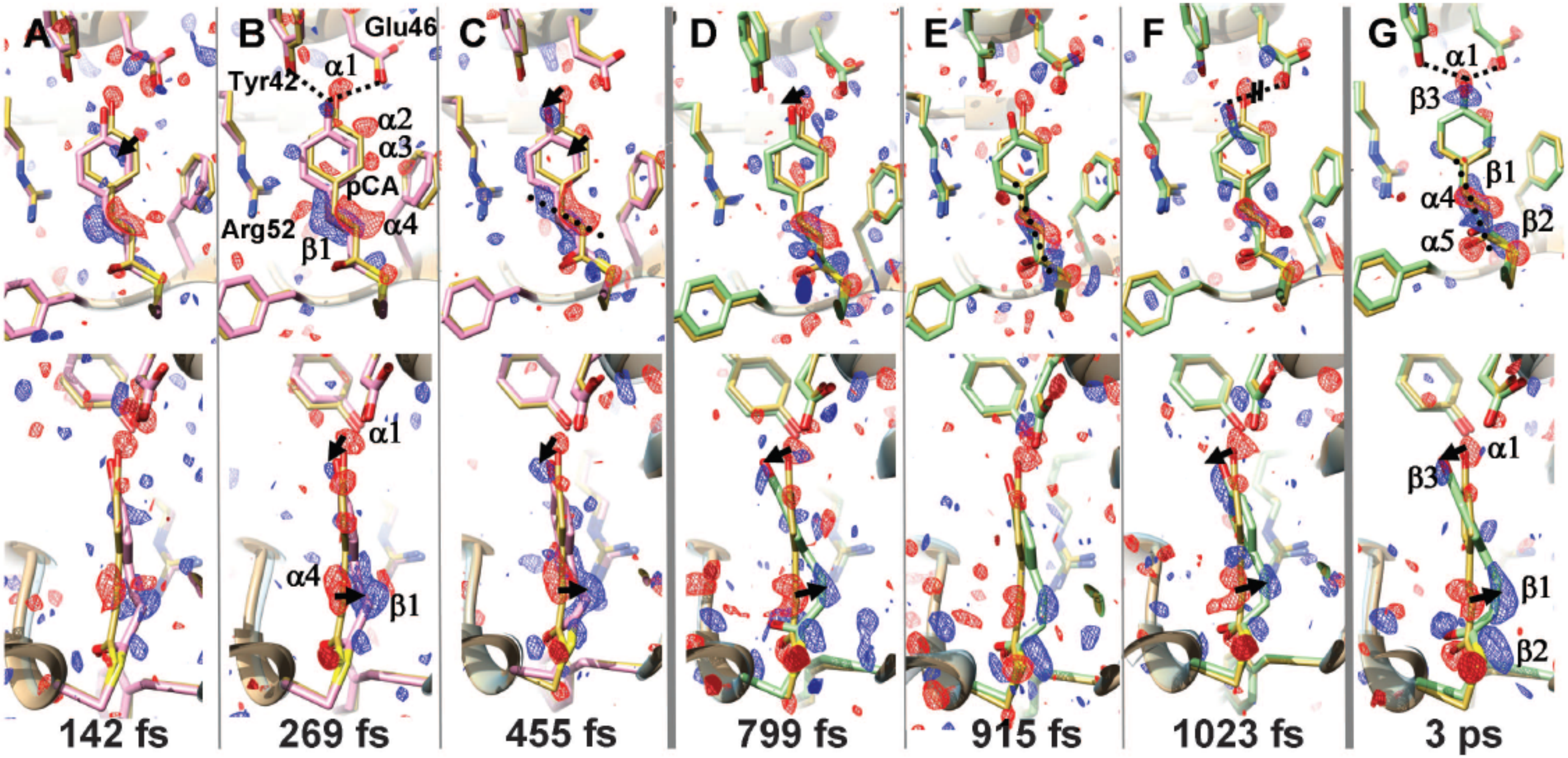
Science 352, 725 (2016): molecular movie

PROTEIN STRUCTURE

Femtosecond structural dynamics drives the trans/cis isomerization in photoactive yellow protein

Kanupriya Pande,[1,2] Christopher D. M. Hutchison,[3] Gerrit Groenhof,[4] Andy Aquila,[5] Josef S. Robinson,[5] Jason Tenboer,[1] Shibom Basu,[6] Sébastien Boutet,[5] Daniel P. DePonte,[5] Mengning Liang,[5] Thomas A. White,[2] Nadia A. Zatsepin,[7] Oleksandr Yefanov,[2] Dmitry Morozov,[4] Dominik Oberthuer,[2] Cornelius Gati,[2] Ganesh Subramanian,[7] Daniel James,[7] Yun Zhao,[7] Jake Koralek,[5] Jennifer Brayshaw,[1] Christopher Kupitz,[1] Chelsie Conrad,[6] Shatabdi Roy-Chowdhury,[6] Jesse D. Coe,[6] Markus Metz,[2] Paulraj Lourdu Xavier,[2,8] Thomas D. Grant,[9] Jason E. Koglin,[5] Gihan Ketawala,[6] Raimund Fromme,[6] Vukica Šrajer,[10] Robert Henning,[10] John C. H. Spence,[7] Abbas Ourmazd,[1] Peter Schwander,[1] Uwe Weierstall,[7] Matthias Frank,[11] Petra Fromme,[6] Anton Barty,[2] Henry N. Chapman,[2,12] Keith Moffat,[10,13] Jasper J. van Thor,[3] Marius Schmidt[1]*

PYP molecular movie



Top: front view; bottom: sideview

Photosynthesis, from solar photons to hydrocarbons

Photosystem II is a 700 kDa protein complex, through which the following reaction takes place in plants, algae, bacteria..:

$$2\ H_2O + 4\ h\nu \Rightarrow O_2 + 4H^+ + 4\ e^-$$

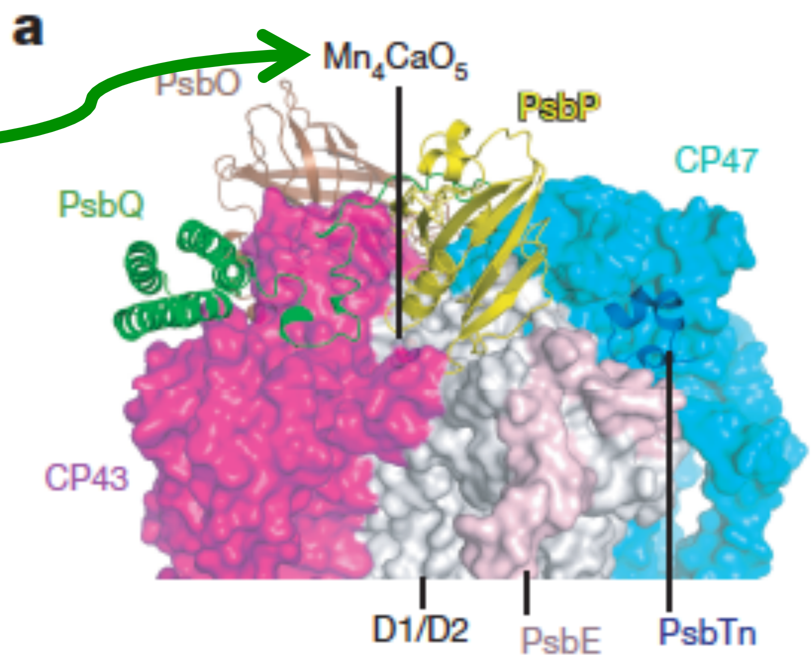
Travel separately to recombine elsewhere and power chemical reactions

Evolves into the atmosphere

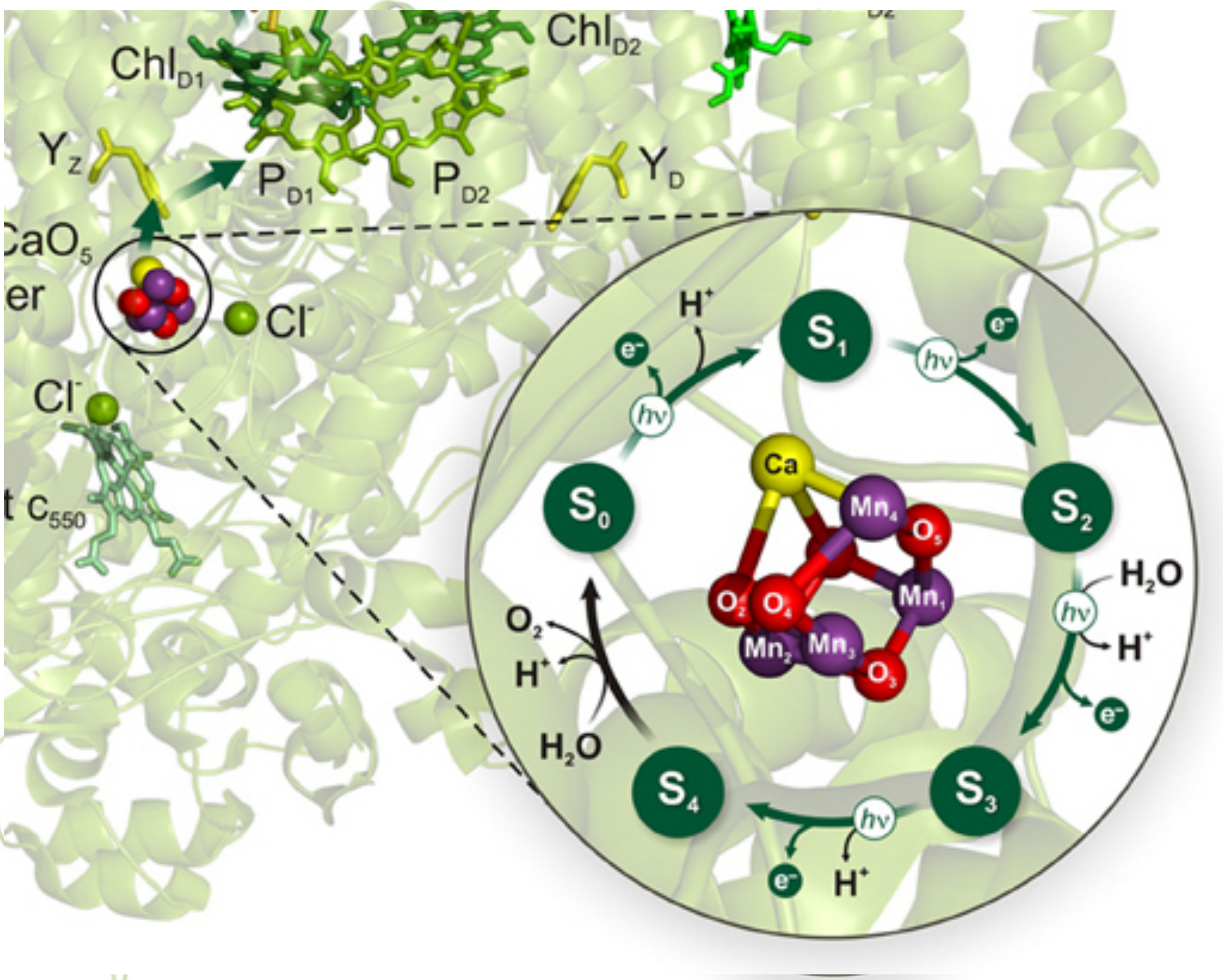
The energy of the four photons is delivered to the OEC "oxygen evolving complex", Mn_4CaO_5, where most of the action seems to take place

X. Wei et al., Nature 2016,

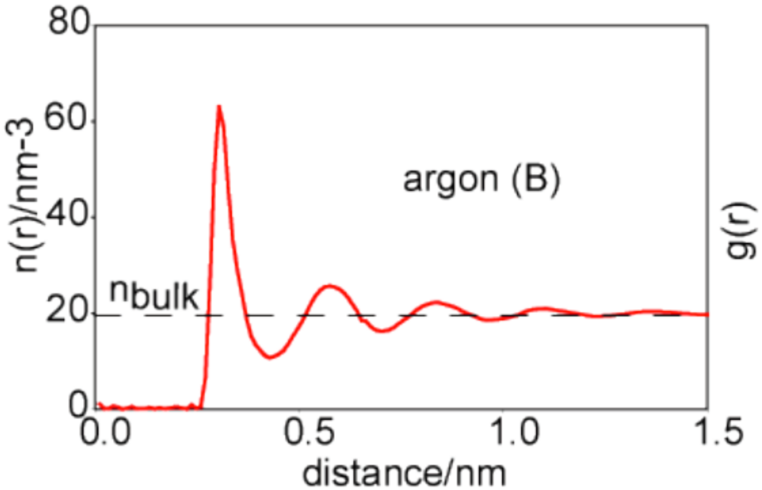
doi:10.1038/nature18020

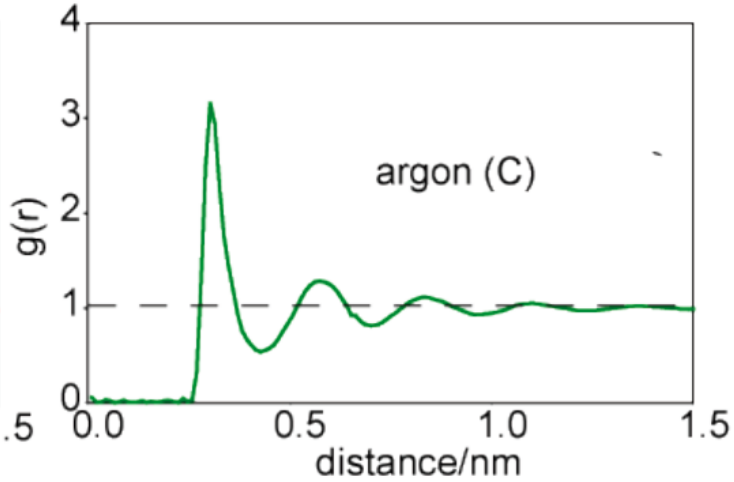


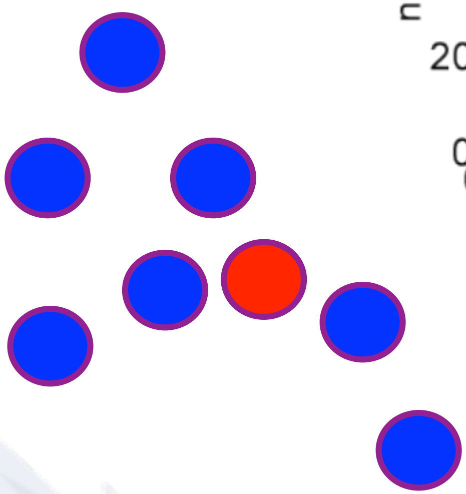
The Kok Cycle: how does it work?



X-ray Diffraction from liquids





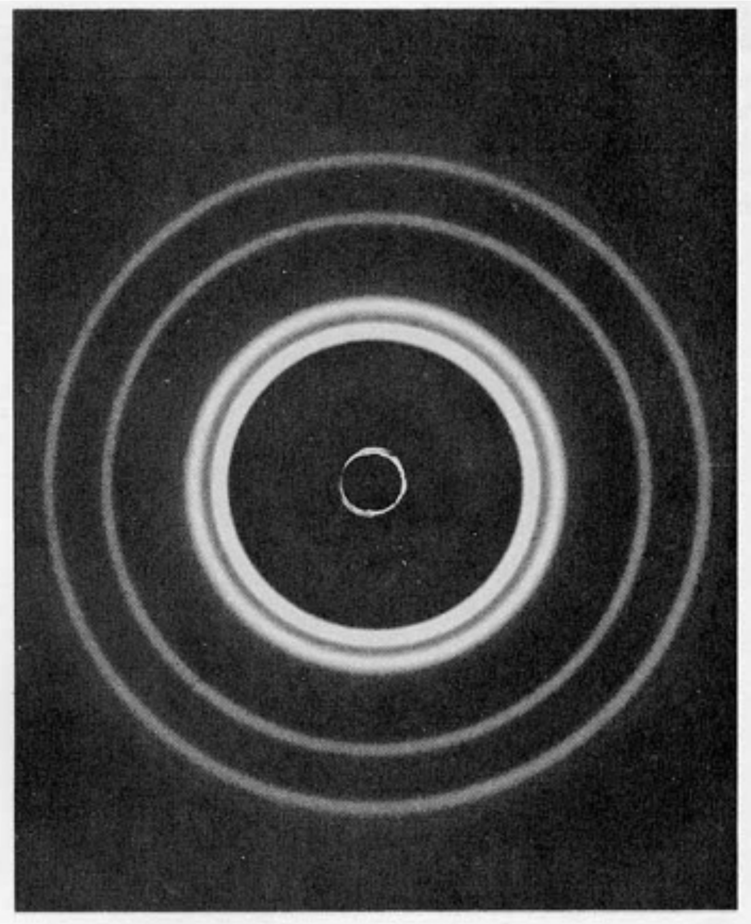


$$S(\boldsymbol{q}) - 1 = \rho_o \int [g(\boldsymbol{r}) - 1]\, e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\, d\boldsymbol{r}$$

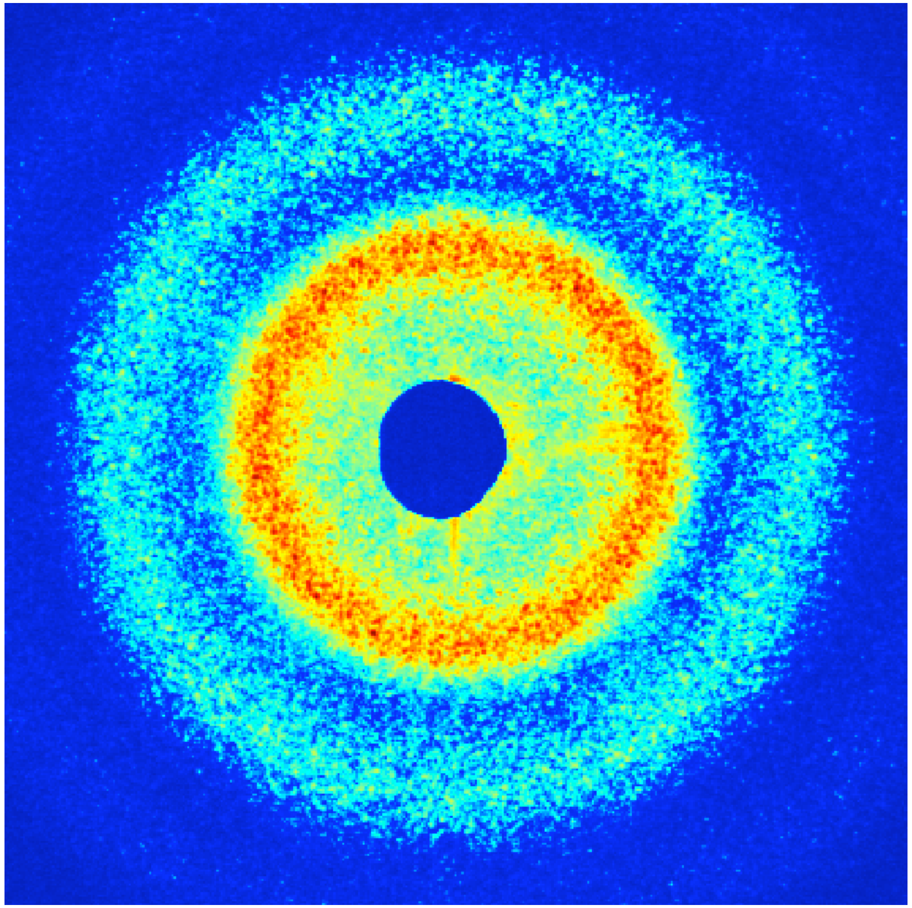
$$g(\boldsymbol{r}) - 1 = \frac{1}{\rho_o (2\pi)^3} \int [S(\boldsymbol{q}) - 1]\, e^{-i\boldsymbol{q}\cdot\boldsymbol{r}}\, d\boldsymbol{q}$$

Liquid

X-ray diffraction from liquids…



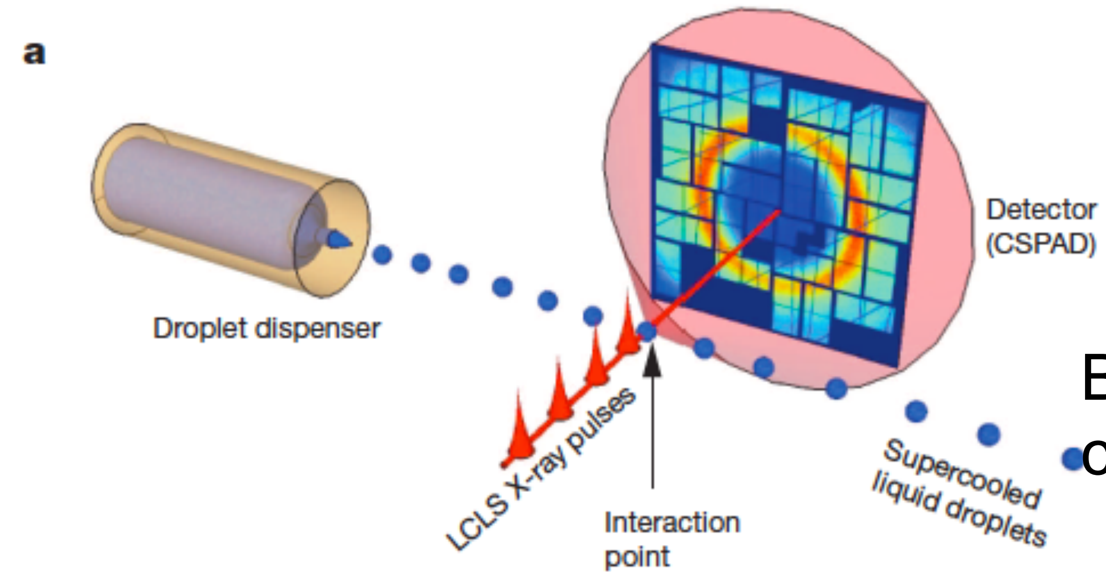
Slow, incoherent…



Fast, coherent => Speckles!
"Hologram"

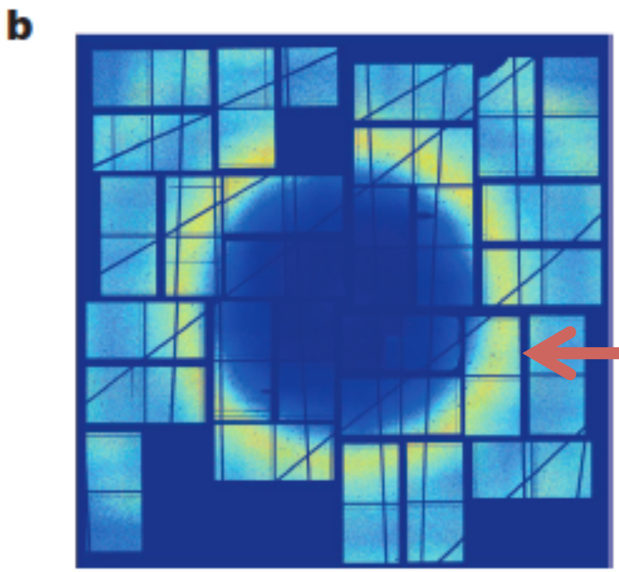
J. Sellberg et al., Nature 510, 381 (2014)

mpsd

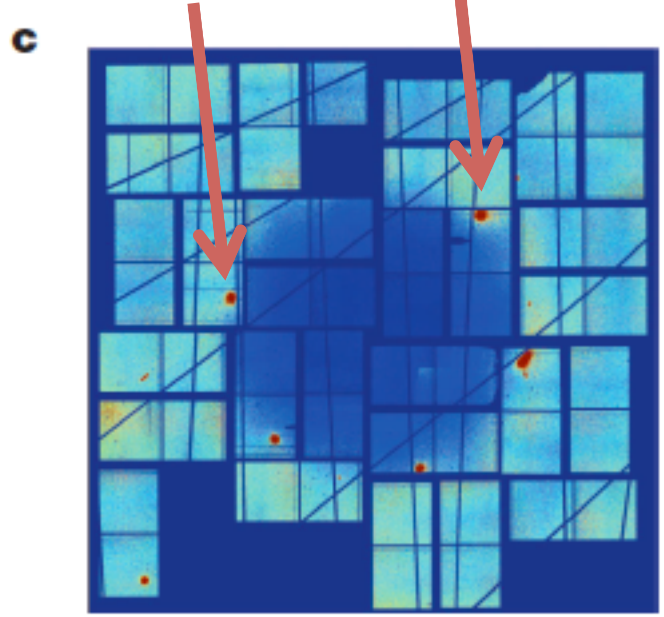


LCLS

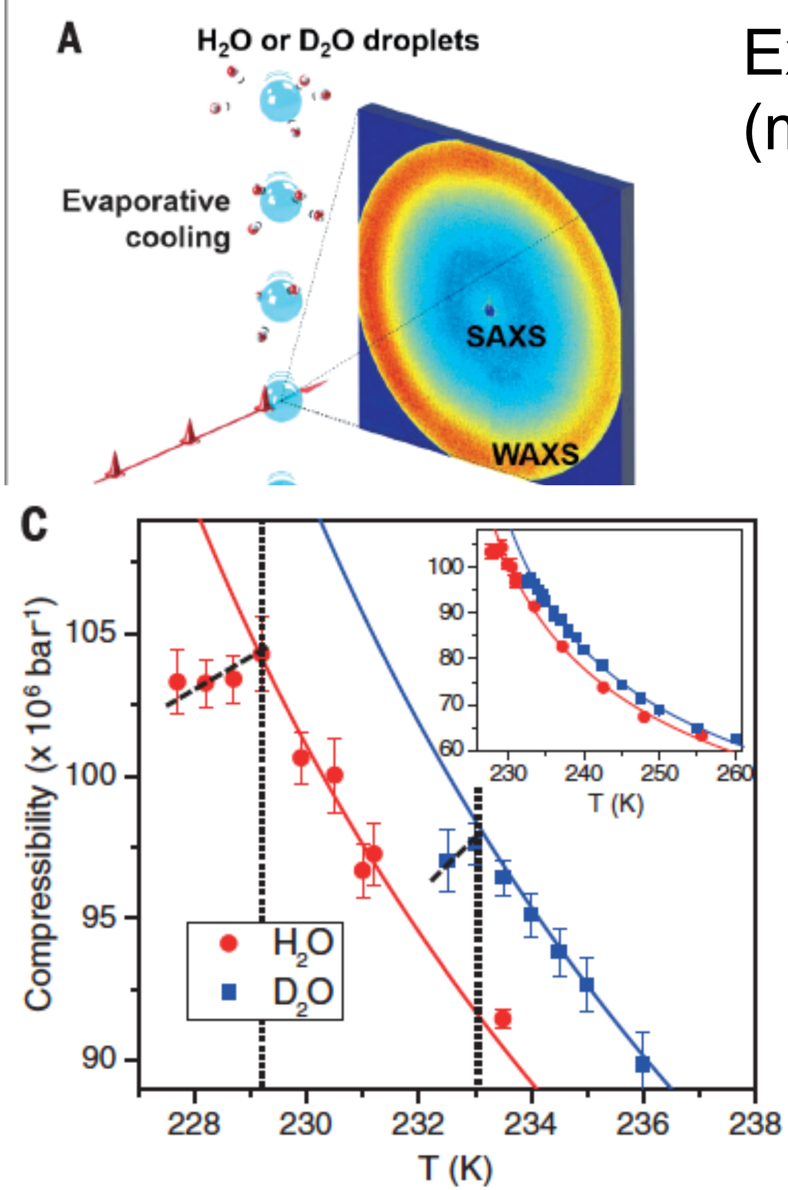
Droplet size ~9 to 37 µm
Bragg peaks from ice crystallites down to ~12 nm



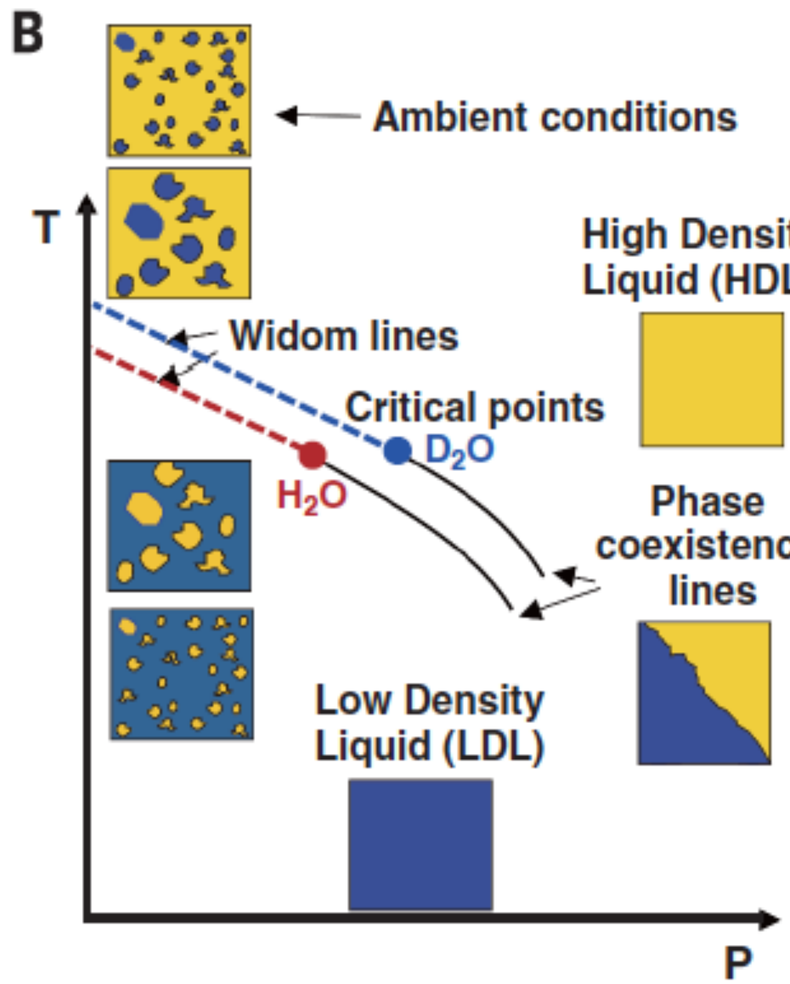
Diffuse scattering from liquid



K.H. Kim *et al.*, Science 358, 1589 (2017)



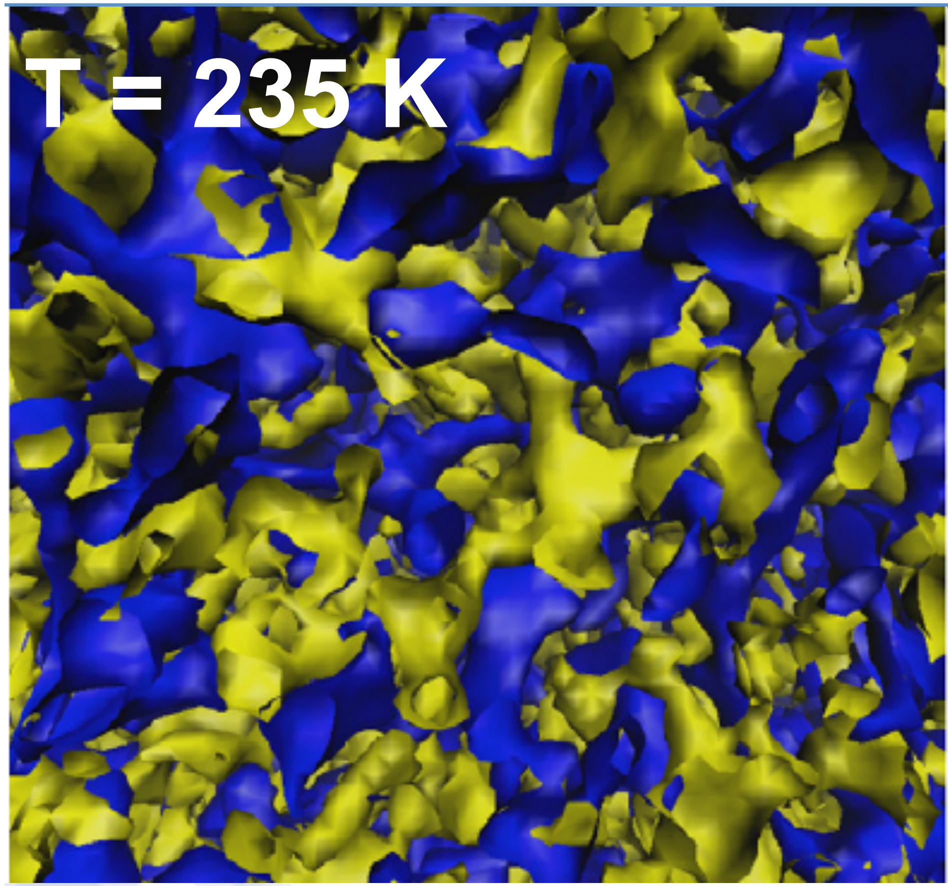
Exploring the supercooled (metastable) water phase diagram

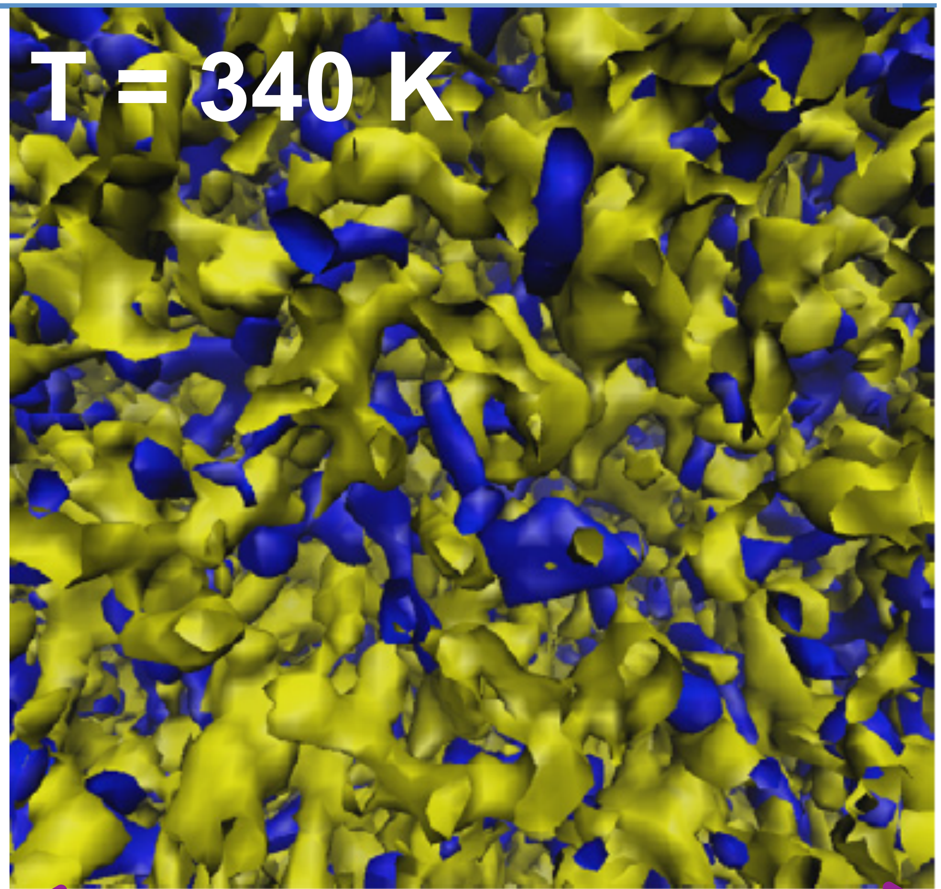


Matter

Simulations of Water Structure

mpsd





10.6 nm

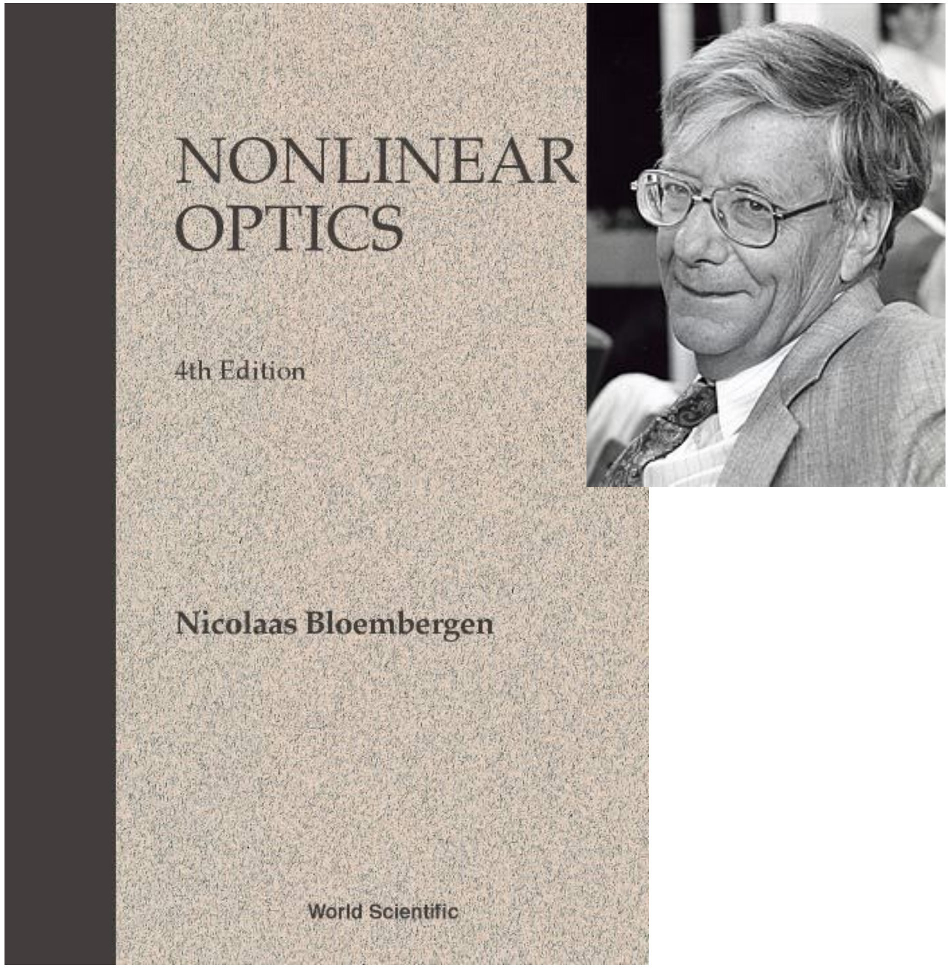
High-density structure

Low-density structure

Wikfeldt J. et al., Chem. Phys. 134 (2011) 214506

Towards Non-linear X-ray Physics

Nicolaas Bloembergen, Physics Nobel Laureate in 1981



Non-linear effects in IR and visible detected after invention of lasers

It is clear that the familiar notion of a linear optical response with a constant index of refraction, that is, an induced polarization proportional to the amplitude of the light field, should be dropped at much less extreme intensities. There is a nonlinearity in the constitutive relationship which may be expanded in terms of a power series in the electric field components

$$P_i = \chi_{ij}^{(1)} E_i + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \ldots \quad (1)$$

Such nonlinearities have been familiar at lower frequencies for over a century.

From N. Bloembergen, Rev. Mod. Phys. 54, 685 (1982)

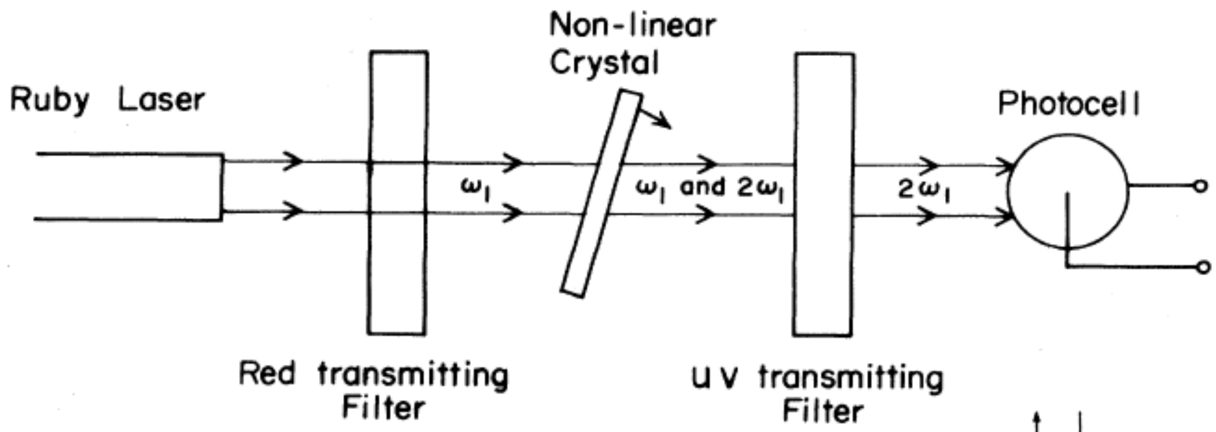


FIG. 3. Second harmonic generation of light.

Examples of non-linear processes

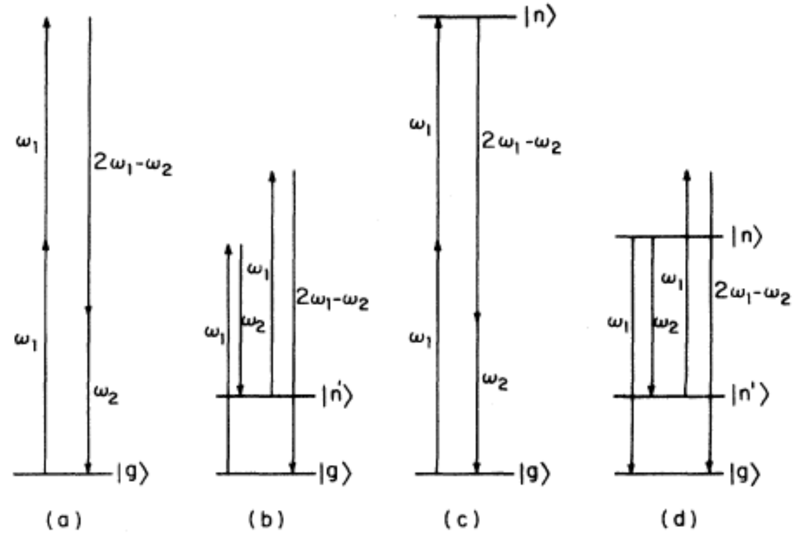
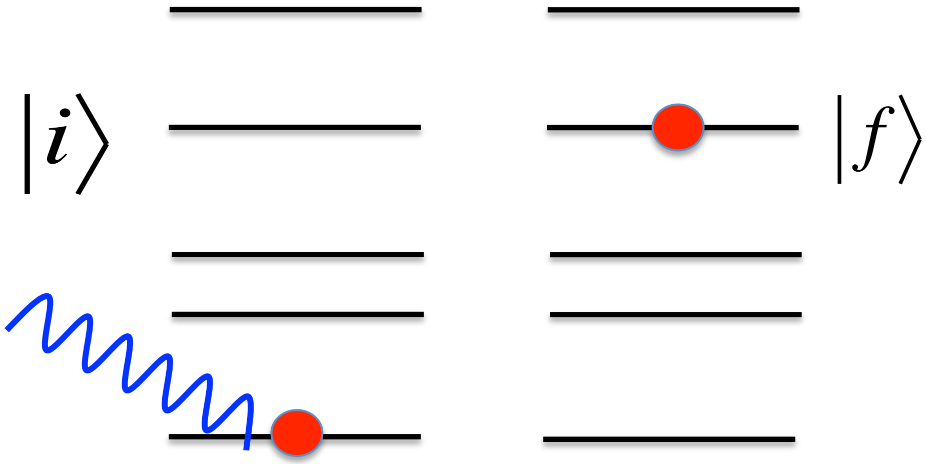


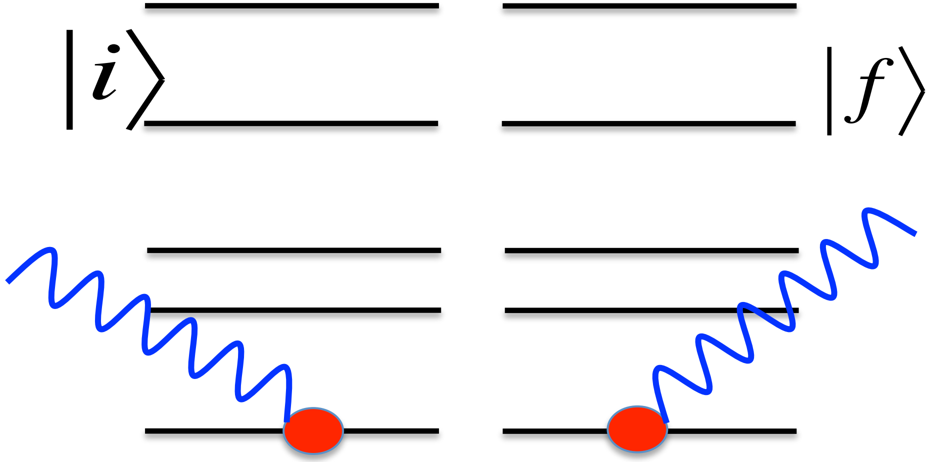
FIG. 8. The creation of a new beam at $2\omega_1 - \omega_2$ by two incident beams at ω_1 and ω_2, respectively, according to the geometry of Fig. 7(b). (a) Nonresonant mixing. (b) Intermediate Raman resonance (coherent anti-Stokes Raman scattering, or CARS). (c) Intermediate two-photon absorption resonance. (d) One-photon resonantly enhanced CARS.

Basic scattering processes

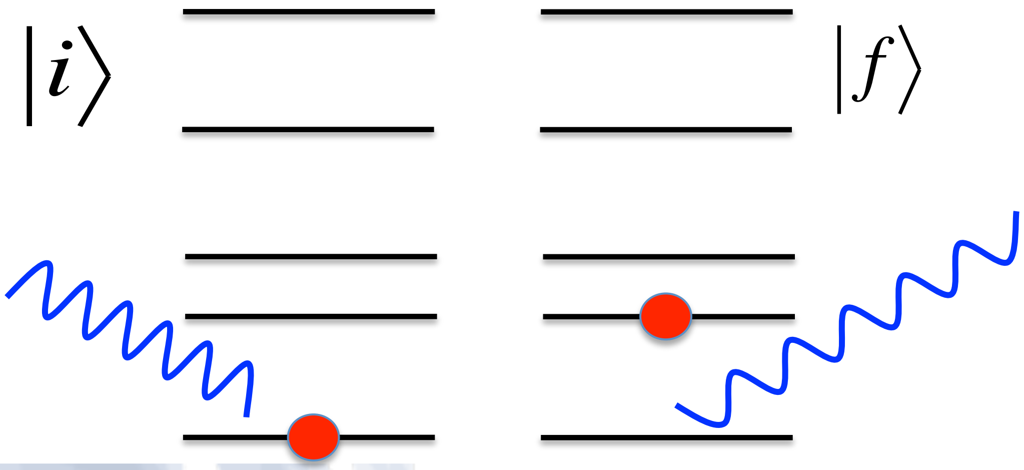
Absorption



Elastic scattering



Inelastic Scattering



Calculating transition probabilities

- **The way forward:**

Assume we have, *in principle*, a description of the energy levels and the eigenstates of electronic matter: H_{el}

Assume we have, in principle, a description of the energy levels and the eigenstates of the free E.M. field: H_{rad}

We can try to calculate the probability of transitions between different states of these two systems induced by their interaction: H_{int}

A practical implementation

mpsd

$$H = H_{el} + H_{rad} + H_{int} \ ,$$

$$H_{el} = \sum_{i=1}^{N} \left[\frac{\mathbf{p}_i{}^2}{2m} + V(\mathbf{r}_i) + (e\hbar/2m^2c^2)\mathbf{s}_i \cdot (\nabla V(\mathbf{r}_i) \times \mathbf{p}_i) \right]$$

$$H_{rad} = \sum_{\mathbf{k},\lambda} \hbar\omega_{\mathbf{k}} \left(a^\dagger(\mathbf{k},\lambda)a(\mathbf{k},\lambda) + 1/2 \right) \ ,$$

H'_1 H'_2

$$H_{int} = \sum_{i=1}^{N} [(e^2/2mc^2)\mathbf{A}^2(\mathbf{r}_i) - (e/mc)\mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i$$

$$-(e\hbar/mc)\mathbf{s}_i \cdot (\nabla \times \mathbf{A}(\mathbf{r}_i)) \quad H'_3$$

$$+(e\hbar/2m^2c^3)\mathbf{s}_i \cdot [(\partial\mathbf{A}(\mathbf{r}_i)/\partial t) \times (\mathbf{p}_i - (e/c)\mathbf{A}(\mathbf{r}_i))]]$$

H'_4

Perturbative calculation of transition rates

- Fermi's "golden rule":

$$w = \frac{2\pi}{\hbar} \left| \langle f | H_{\text{int}} | i \rangle + \sum_n \frac{\langle f | H_{\text{int}} | n \rangle \langle n | H_{\text{int}} | i \rangle}{E_i - E_n} + \ldots \right|^2 \delta(E_f - E_i)$$

1st order — 2nd order — Higher orders

The idea behind is that H_{int} is "small", that 1st order is (either zero or) bigger than 2nd, that is bigger than 3rd ….

Linear physics: stop at the first (non-vanishing) order

	Elastic, non-res	Inelastic, Non-res.	Elastic Resonant	Inelastic, Resonant	
H'_1	Thomson/ Bragg scattering	Compton, Raman, $S(\mathbf{q},\omega)$			
H'_2	Orbital magnetic scattering		Resonant Elastic (REXS: charge, spin, orbital order…)	Absorption, XMCD, Emission, Res. Inelastic (RIXS)	
H'_3	Spin Magnetic				
H'_4	Spin Magnetic				

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Ultra-intense, coherent very short pulses

- Examine how the implementation of this program is affected by the unconventional features of X-ray free-electron laser pulses:

  – High intensity
  – Time duration in the $\Delta t \ll 1\ ps$ regime
  – Spatial coherence

Perturbative calculation of transition rates

- Fermi's "golden rule":

$$w = \frac{2\pi}{\hbar}\left| \langle f | H_{\text{int}} | i \rangle + \sum_n \frac{\langle f | H_{\text{int}} | n \rangle \langle n | H_{\text{int}} | i \rangle}{E_i - E_n} + \ldots \right|^2 \delta(E_f - E_i)$$

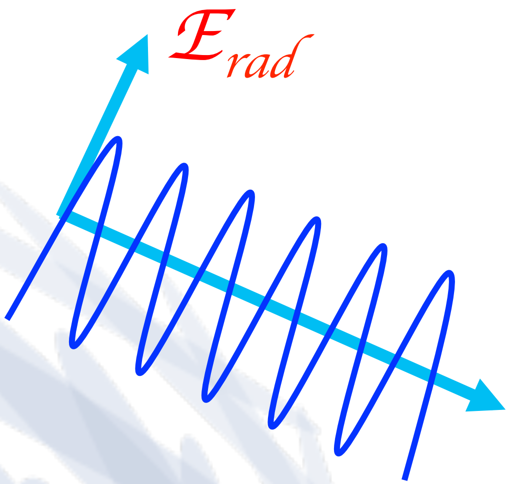
1st order — 2nd order — Higher orders

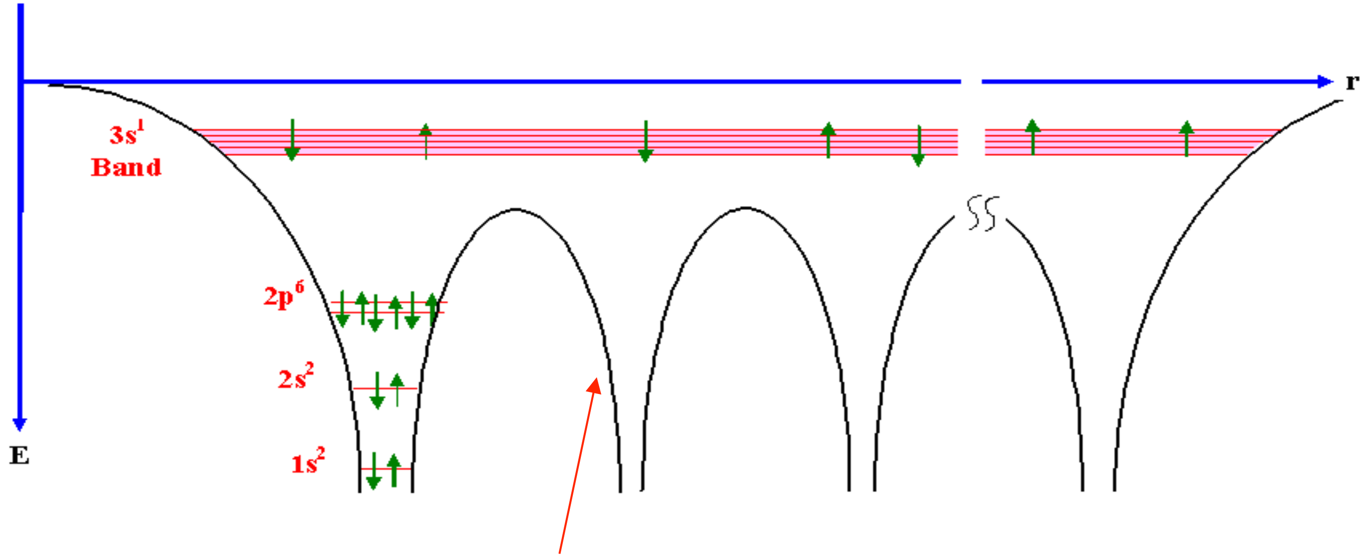
The idea behind is that H_{int} is "small", that 1st order is (either zero or) bigger than 2nd, that is bigger than 3rd ….

Linear physics: stop at the first (non-vanishing) order

What does it mean “the perturbation is small”?

- Physically, verify that the physical action of the radiation on the electronic system is small, and cannot affect the structure of the energy levels drastically.
- Compare the electric field of the radiation to the atomic electric field.





$$\mathcal{E}_{at} = -grad\ V$$

Estimate of the atomic electric field

Field inside an atom:

$\mathcal{E}_{at} \sim e/a^2_B = 27.2\ V/a_B \sim 5.2\ X10^9\ V/cm$

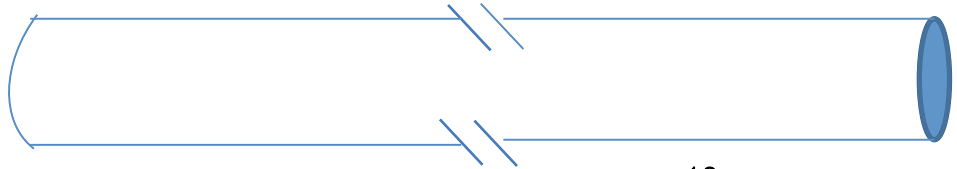
(Remember: $a_B \sim 5\ X\ 10^{-9}$ cm

$e^2 / a_B \sim 2$ Ryd = 1 Hartree ~ 27.2 eV)

Let's compare this with the radiation field in relevant cases

Radiation electric field: synchrotron case

- Suppose your synchrotron beamline delivers 10^{11} ~ 1 keV photons/s on a μm^2 spot, typically in ~10^6 pulses, each ~10 ps long:
- I = energy/unit area & unit time = 10^5 X 10^3 eV/ 10^{-19} cm^{2}s= 10^{27} eV /cm^2 s.



1 cm^2

c X 1 s = 3 X 10^{10} cm

- Energy/unit volume: = I / c = (10^{27} eV /cm^2 s) / 3 10^{10} cm/s

= 3.3 10^{16} eV/cm^3 → $(1/4\pi) \langle \mathcal{E}^2 \rangle$ (gaussian units)

= 3.3 10^{16} (e/a_B) (V/cm^3) a_B

27.2 V

$\langle \mathcal{E}_{rms} \rangle \sim 10^5$ V/cm $\ll$ Atomic field ~ 5 X 10^9 V/cm

LCLS Stanford, experimental results (2010)

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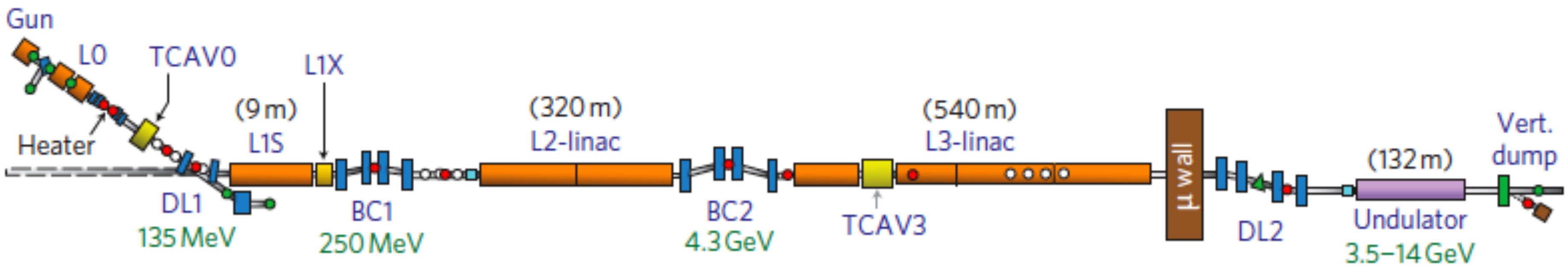
Table 1 | Design and typical measured parameters for both hard (8.3 keV) and soft (0.8–2.0 keV) X-rays. The 'design' and 'hard' values are shown only at 8.3 keV. Stability levels are measured over a few minutes.

Parameter	Design	Hard	Soft	Unit
FEL gain length	4.4	3.5	~1.5	m
Radiation wavelength	1.5	1.5	6-22	Å
Photons per pulse	2.0	1.0-2.3	10-20	10^{12}
Energy in X-ray pulse	1.5	1.5-3.0	1-2.5	mJ
Peak X-ray power	10	15-40	3-35	GW
Pulse length (FWHM)	200	70-100	70-500	fs
Bandwidth (FWHM)	0.1	0.2-0.5	0.2-1.0	%
Peak brightness (estimated)	8	20	0.3	10^{32} *
Wavelength stability (r.m.s.)	0.2	0.1	0.2	%
Power stability (r.m.s.)	20	5-12	3-10	%



Radiation electric field: X-ray FEL case

- Synchrotron 10^5 ~1 keV photons/ 10^{-11}s on a μm^2 spot
- X-ray FEL 10^{13} ~1 keV photons / 7 10^{-14} s on a μm^2 spot

Synchrotron:

$< E_{rms} > \sim 10^6$ V/cm

X-ray laser:

$< E_{rms} > \sim 10^{10}$ V/cm

$<\mathcal{E}_{at}> \sim 5 \times 10^9$ V/cm

High-field ionization regime

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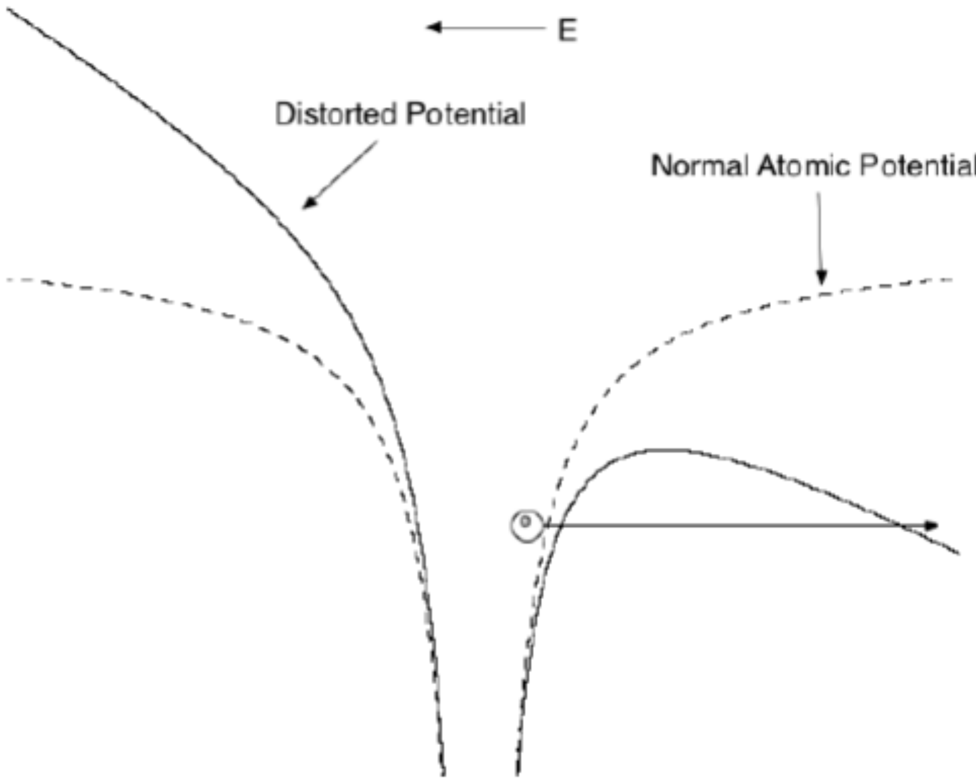
- Atomic Potential is severely distorted ⇒ field ionization ?



Well known from ultra-high power lasers!

Strong-field ionization theory of Keldysh, 1964

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SOVIET PHYSICS JETP VOLUME 20, NUMBER 5 MAY, 1965

IONIZATION IN THE FIELD OF A STRONG ELECTROMAGNETIC WAVE

L. V. KELDYSH

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor May 23, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 47, 1945-1957 (November, 1964)

Expressions are obtained for the probability of ionization of atoms and solid bodies in the field of a strong electromagnetic wave whose frequency is lower than the ionization potential. In the limiting case of low frequencies these expressions change into the well known formulas for the probability of tunnel auto-ionization; at high frequencies they describe processes in which several photons are absorbed simultaneously. The ionization probability has a number of resonance maxima due to intermediate transition of the atom to an excited state. In the vicinity of such a maximum the ionization cross section increases by several orders of magnitude. The positions and widths of the resonances depend on the field strength in the wave. It is shown that for optical frequencies the mechanism under consideration, of direct ionization by the wave field, may be significant in the case of electric breakdown in gases, and especially in condensed media.

No tunnel ionization at x-ray frequencies!

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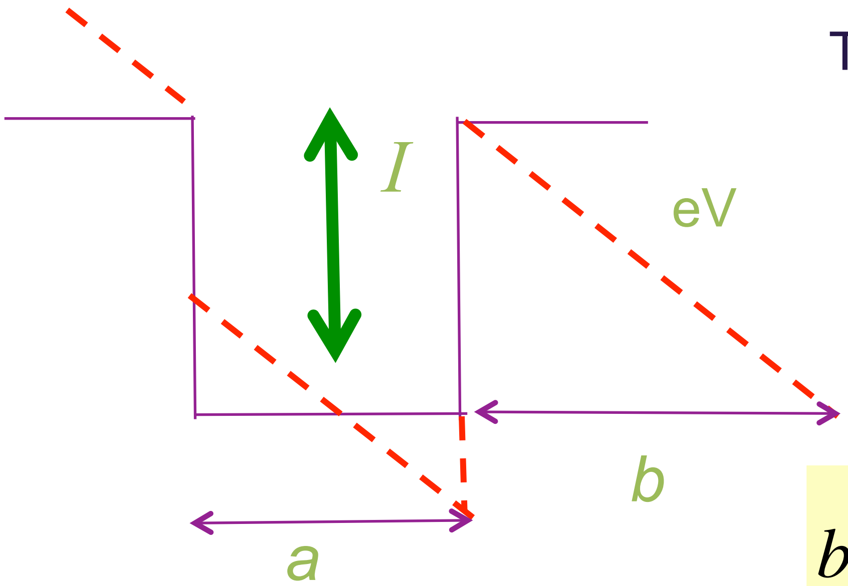


To tunnel, the electron takes a time ~ τ, because it must go through a distance *b* and its velocity is of order:

$$v \sim \frac{\hbar}{ma} \sim \sqrt{\frac{I}{m}}$$

$$b \sim \frac{I}{eE_{rms}}$$

$$\tau \sim b/v \sim \frac{\sqrt{mI}}{eE_{rms}} \equiv \frac{2\pi}{\omega_{tunn}}$$

For $I \sim 10\ eV$, $e\mathcal{E}_{rms} \sim 10^{11}$ V/cm ➔ $\tau_{tunn} \sim 10^{-18}$ s

X-ray ½ period at 1 keV: $\tau_X \sim 10^{-18}$ s!)

…no time to tunnel. **Keldysh parameter:**

$$\gamma \approx \frac{\omega_X}{\omega_{tunn}} \begin{cases} > \sim 1 & \text{multi-photon} \\ << 1 & \text{tunneling} \end{cases}$$

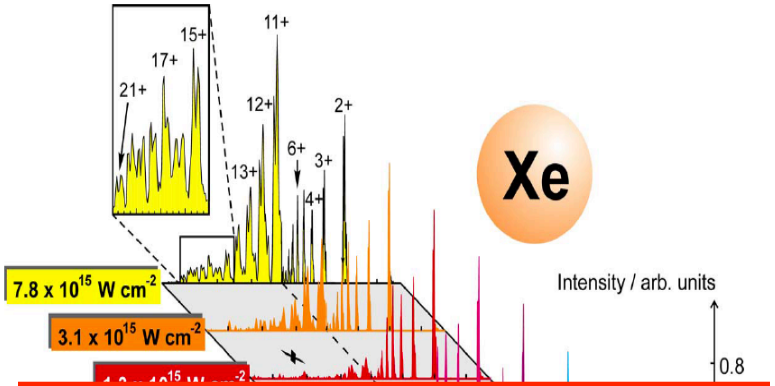
Multi-photon Multiple Ionization

Multi-electron system: Xe

(Courtesy Michael Meyer)

λ (FLASH) = 13.3 nm Sorokin et al., PRL 99, 213002 (2007)

hν = 93 eV

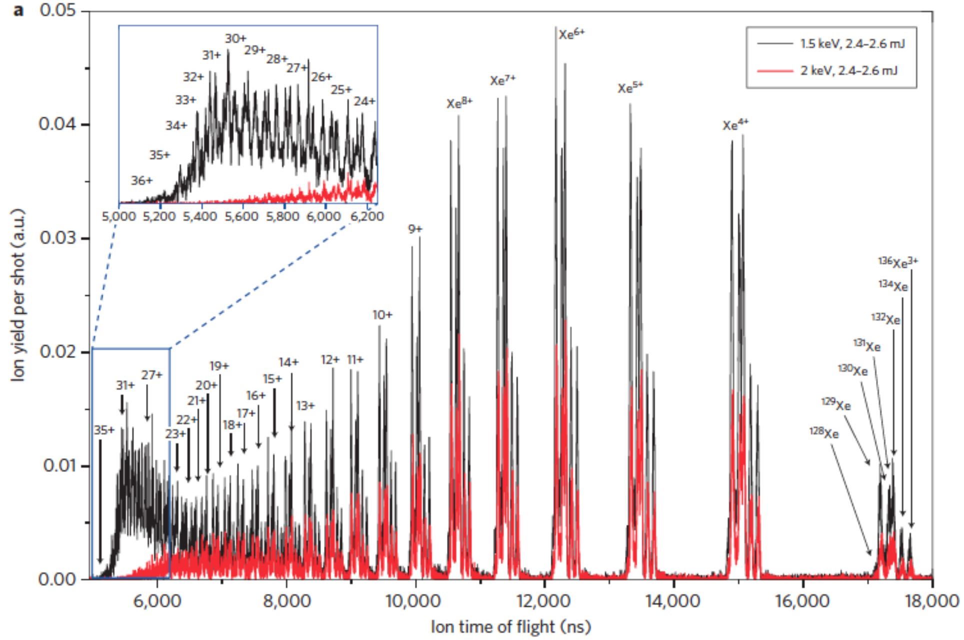


Direct and/or sequential processes ??

IP(Xe 21+) ≈ 5 keV

λ (LCLS) = 0.8 nm Rudek et al., Nat.Phot. 6, 858 (2012)

hν = 1.5 keV



Xe 35+

Saturable absorption in Al at higher intensity

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nature physics

ARTICLES

PUBLISHED ONLINE: 26 JULY 2009 | DOI: 10.1038/NPHYS1341

Turning solid aluminium transparent by intense soft X-ray photoionization

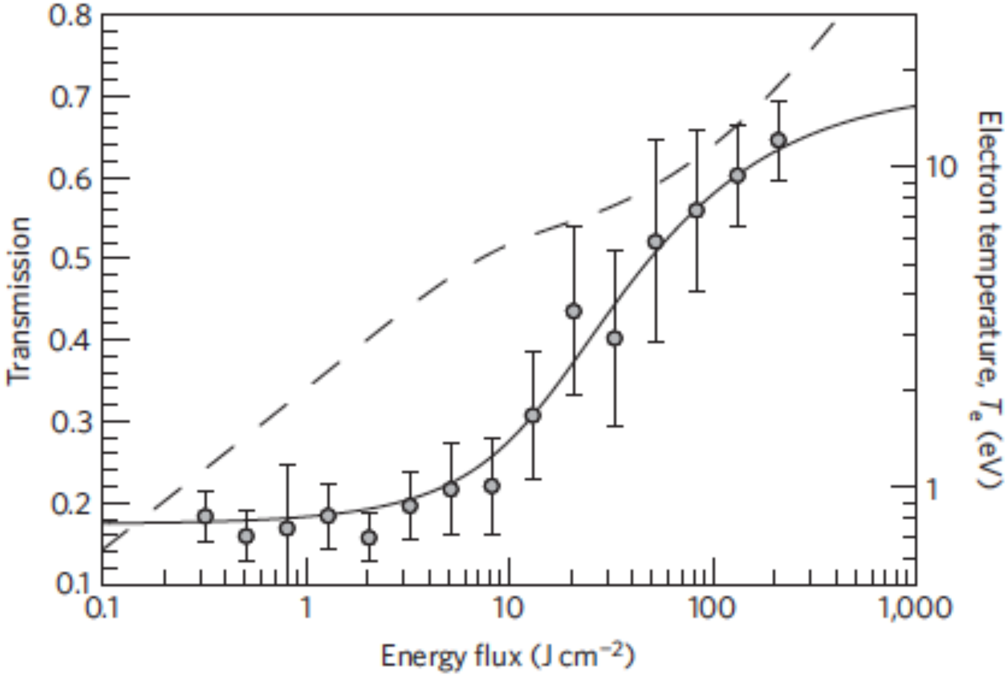
Bob Nagler *et al.**

FLASH

92.5 eV photons, 15 fs pulses

Minimum spot ~ 1.5 μm

Al L_2, L_3 edges: 72.7, 73.1

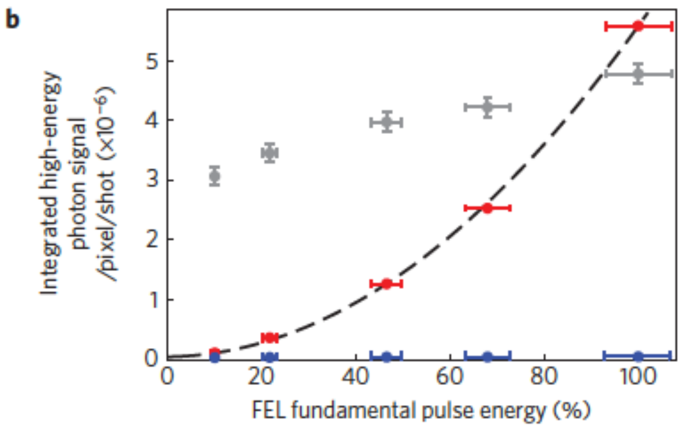


Saturable absorption in Al at higher intensity

- Depletion of L_2, L_3 electrons during pulse depending on intensity and Auger lifetime (~ 40 fs)

- Edge shift to >93 eV for second L-edge ionization

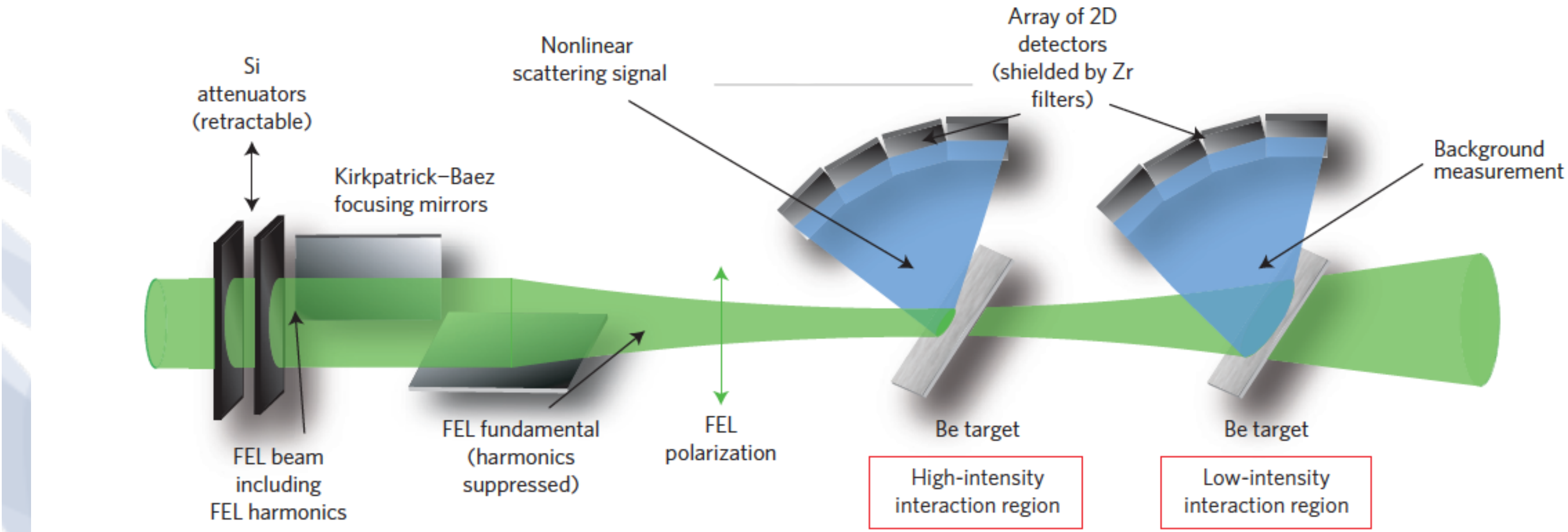
$\mathcal{E} \sim 5\ 10^{11}$ V/cm, ~ 10^{20} W /cm^2



ARTICLES
PUBLISHED ONLINE: 31 AUGUST 2015 | DOI: 10.1038/NPHYS3452
nature physics

Anomalous nonlinear X-ray Compton scattering

Matthias Fuchs[1,2*], Mariano Trigo[2,3], Jian Chen[2,3], Shambhu Ghimire[2], Sharon Shwartz[4], Michael Kozina[2,3], Mason Jiang[2,3], Thomas Henighan[2,3], Crystal Bray[2,3], Georges Ndabashimiye[2], Philip H. Bucksbaum[2], Yiping Feng[5], Sven Herrmann[6], Gabriella A. Carini[6], Jack Pines[6], Philip Hart[6], Christopher Kenney[6], Serge Guillet[5], Sébastien Boutet[5], Garth J. Williams[5], Marc Messerschmidt[5,7], M. Marvin Seibert[5], Stefan Moeller[5], Jerome B. Hastings[5] and David A. Reis[2,3]

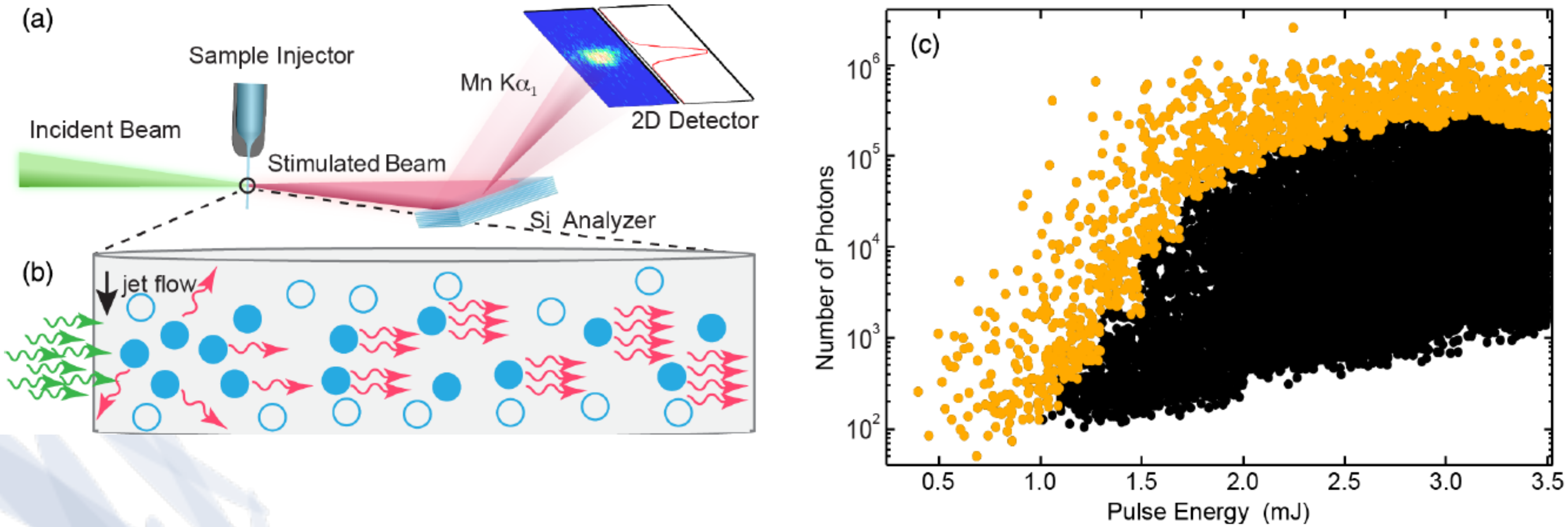


Phys. Rev. Lett. **120**, 133203 (2018)

mpsd

Stimulated X-Ray Emission Spectroscopy in Transition Metal Complexes

Thomas Kroll,[1,2,*] Clemens Weninger,[3,1,†] Roberto Alonso-Mori,[1] Dimosthenis Sokaras,[2] Diling Zhu,[1] Laurent Mercadier,[3] Vinay P. Majety,[3] Agostino Marinelli,[4] Alberto Lutman,[4] Marc W. Guetg,[4] Franz-Josef Decker,[4] Sébastien Boutet,[1] Andy Aquila,[1] Jason Koglin,[1] Jake Koralek,[1] Daniel P. DePonte,[1] Jan Kern,[1,5] Franklin D. Fuller,[5] Ernest Pastor,[5] Thomas Fransson,[6] Yu Zhang,[6] Junko Yano,[5] Vittal K. Yachandra,[5] Nina Rohringer,[3,7,8,‡] and Uwe Bergmann[1,6,§]



Detected number of photons in the Mn $K\alpha_1$ region (5 eV integration window) as a function of the nominal incoming XFEL pulse energy for the 5M $MnCl_2$ solution. The actual pulse energy on target is ~20% of the nominal pulse energy shown in the figure. The 50 strongest shots in each 0.1 mJ interval are shown in orange, all other shots in black.

More examples of non-linearities…

- …see lecture by C. Masciovecchio on Friday.

Nonlinearities, sample damage, etc…

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- …can be a nuisance in many cases!

78 | NATURE | VOL 520 | 2 APRIL 2015

Orbital-specific mapping of the ligand exchange dynamics of $Fe(CO)_5$ in solution

Ph. Wernet[1], K. Kunnus[1,2], I. Josefsson[3], I. Rajkovic[4]†, W. Quevedo[4]†, M. Beye[1], S. Schreck[1,2], S. Grübel[4]†, M. Scholz[4], D. Nordlund[5], W. Zhang[6]†, R. W. Hartsock[6], W. F. Schlotter[7], J. J. Turner[7], B. Kennedy[8]†, F. Hennies[8], F. M. F. de Groot[9], K. J. Gaffney[6], S. Techert[4,10,11], M. Odelius[3] & A. Föhlisch[1,2]

- “The employed average fluence of the incident monochromatized x-ray pulses on the sample was 30 mJ/cm2 per pulse ($1.6 \cdot 10^{10}$ photons per pulse). In order to achieve this, the LCLS beam was attenuated by a factor of 10 with a gas attenuator installed upstream of the SXR beamline monochromator… In this way, spectral distortions observed in the $Fe(CO)_5$ RIXS spectra at high peak fluences could be avoided ”

Nonlinearities, sample damage,…

PHYSICAL REVIEW B **95**, 121105(R) (2017)

Nonequilibrium lattice-driven dynamics of stripes in nickelates using time-resolved x-ray scattering

W. S. Lee,[1,*] Y. F. Kung,[1,2] B. Moritz,[1,3,4] G. Coslovich,[5,6] R. A. Kaindl,[5] Y. D. Chuang,[7] R. G. Moore,[1] D. H. Lu,[8] P. S. Kirchmann,[1] J. S. Robinson,[5,6] M. P. Minitti,[6] G. Dakovski,[6] W. F. Schlotter,[6] J. J. Turner,[6] S. Gerber,[1] T. Sasagawa,[9] Z. Hussain,[7] Z. X. Shen,[1,†] and T. P. Devereaux[1,‡]

"… the front-end nitrogen gas attenuator (set at approximately 20% transmission) was used to avoid radiation damage from the LCLS beam…"

→ High repetition rate (high average brilliance) can be very important in such cases!

Conclusion

- X-ray Free-Electron Lasers open new avenues for research in physics, chemistry and structural biology
- The promise of a revolution in the investigation of time-dependent phenomena, of non-linear x-ray effects, etc. raises great expectations and explains the worlwide interest in these facilities
- There is however a steep „learning curve“ to climb, for experiments and also for theory, in order to exploit these potentialities in full