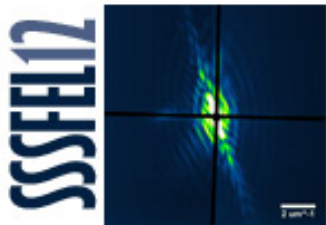


A proposal for a mode-locked hard x-ray free-electron laser

Ryan Lindberg and Alexander Zholents
Argonne National Laboratory



Seeding and Self-seeding at New FEL Sources

ICTP, Adriatico Guesthouse / Trieste, Italy / 10-12 December 2012



X-ray FELs enable new science

LETTER

Femtosecond X-ray protein nanocrystallography

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X-ray crystallography provides the way to atomic-resolution structure, but the time needed to collect a diffraction pattern is so long that it is not possible to study fast processes. X-ray free-electron lasers (XFELs) provide a solution: they produce a very bright, very short X-ray pulse that can be used to collect a diffraction pattern in a few femtoseconds. This is particularly challenging to do in a protein, which is a very delicate structure. However, the importance of this field is growing. We present a method for structure determination where single-pulse X-ray diffraction 'snapshots' are collected from a fully hydrated stream of nanocrystals using femtosecond pulses from a hard-X-ray free-electron laser, the Linac Coherent Light Source. We discuss this concept with nanocrystals of photosynthetic reaction center protein as an example. More than 2,000 diffraction patterns were collected in 1.5 s, and a three-dimensional data set was assembled from individual photosynthetic reaction center (RC) units in 20 s. We highlight the problem of nanocrystal damage to crystallography and the necessity of a nanosecond-scale process. This offers a new approach to the structure determination of macromolecules that do not tolerate long exposure times at a synchrotron source or a conventional radiation source or are particularly sensitive to radiation damage.

Science
High-Resolution Protein Structure Determination by Serial Femtosecond Crystallography
Sébastien Boutin et al.
Science 337, 362 (2012)
DOI: 10.1126/science.1217373

High-Resolution Protein Structure Determination by Serial Femtosecond Crystallography

Sébastien Boutin^{1,2}, Lukas Lomb³, Gert J. Williams⁴, Thomas M. B. Meyer^{5,6}, Andrew Aguiar⁷, R. Bruce Doak⁸, David P. DeRosier⁹, Jan Steinhilber¹⁰, Robert L. Shoeman¹¹, Marc Messerschmidt¹², Anton Barty¹³, Thomas A. White¹⁴, Stephen Kusnerik¹⁵, Richard A. Kistner¹⁶, R. Marvin Abt¹⁷, Paul A. Hergenrother¹⁸, Clark Roemer¹⁹, Philip Han²⁰, Jack Fromm²¹, Gert J. Williams²², Sol M. Grzesek²³, Hugh T. Wilson²⁴, Mark M. Teare²⁵, Marianne Hübner²⁶, Lars J. Koster²⁷, Niklas van Belle²⁸, John Moran²⁹, Wilfried Grossehans³⁰, David Arslan³¹, Michael J. Bogan³², Carl Caspar³³, Ralfred Probst³⁴, Christina Y. Hwang³⁵, Mark S. Hunter³⁶, Linda C. Johnson³⁷, George Wilson³⁸, Christopher Beemann³⁹, Mingming Liang⁴⁰, Andrew V. Martin⁴¹, Kent Walz⁴², John Beedee⁴³, Niclas Svanberg⁴⁴, Ramon Knebel⁴⁵, Douglas Wang⁴⁶, Nadia A. Zatsepin⁴⁷, Donald Schaefer⁴⁸, James DeWetter⁴⁹, Richard Neenan⁵⁰, Petra Fromme⁵¹, John C. H. Spence⁵², Henry N. Chapman⁵³, Hans Schlichting⁵⁴

Structure determination of proteins and other macromolecules has historically required the growth of high-quality crystals sufficiently large to diffract x-rays efficiently while withstanding radiation damage. We applied serial femtosecond crystallography (SFX) using an x-ray free-electron laser (XFEL) to obtain high-resolution structural information from microcrystals (less than 1 micrometer by 1 micrometer by 1 micrometer) of well-characterized model protein lysozyme. The agreement with synchrotron data demonstrates the immediate relevance of SFX for analyzing the structure of the large group of difficult-to-crystallize molecules.

PHYSICAL REVIEW LETTERS

Ultraintense X-Ray Induced Ionization, Dissociation, and Frustrated Absorption in Molecular Nitrogen

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 (Received 24 May 2010; published 23 June 2010)

LETTER

Single mimivirus particles intercepted and imaged with an X-ray laser

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X-ray lasers offer new capabilities in understanding the structure of biological systems, complex materials and matter under extreme conditions. Very short and extremely bright x-ray pulses can be used to overexpose key damage processes and obtain a single diffraction pattern from a large macromolecule, a virus or a cell before the sample explodes and turns into plasma. The continuous diffraction pattern of non-crystalline objects permits circumventing the exact phase problem. We show that high-quality diffraction data can be obtained with a single X-ray pulse from a non-crystalline biological sample, a single infectious particle, which was intercepted and imaged with an X-ray free-electron laser, the Linac Coherent Light Source. Calculations indicate that the x-ray diffracted into the lens by the plasma lens of the particle is only 10% of the total pulse had it been fully transmitted. The reconstructed data show that approximately 50% of the data needed to define the virus. We expect that significantly higher resolutions will be obtained in such experiments with shorter and brighter x-ray pulses focused on a cell core. The resolution in such experiments can be further extended for samples available in multiple identical copies.

PHYSICAL REVIEW LETTERS

Direct Measurements of the Ionization Potential Depression in a Dense Plasma

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ARTICLES

Femtosecond electronic response of atoms to ultra-intense X-rays

L. Young¹, E. P. Kanter², B. Krässig³, Y. Li⁴, A. M. March⁵, S. T. Pratt⁶, R. Santra⁷, S. H. Southworth⁸, N. Rohringer⁹, L. F. DiMauro¹⁰, G. Doumy¹¹, C. A. R. Rongé¹², M. Berrnhil¹³, P. H. Bucksbaum¹⁴, J. P. Cryan¹⁵, S. Ghimire¹⁶, J. M. Glownia¹⁷, D. A. Reis¹⁸, J. D. Bozack¹⁹, C. Bostedt²⁰ & M. Messerschmidt²¹

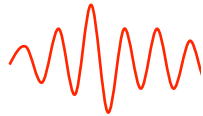
An era of exploring the interactions of high-intensity, hard X-rays with matter has begun with the startup of a hard-X-ray free-electron laser, the Linac Coherent Light Source (LCLS). Understanding how electrons in matter respond to ultra-intense X-ray radiation is essential for all applications. Here we reveal the nature of the electronic response in a free atom to unprecedented high-intensity, ultra-short-wavelength, high-fluence radiation (on spectrally 10¹⁸ W cm⁻², 15–0.6 nm⁻¹, ~10⁻¹⁵ s X-ray photon per Å²). At this fluence, the atoms rapidly ionize during the course of a single femtosecond-duration X-ray pulse—by means of a field-induced electron-to-proton Auger-decay process. Fully developed near-threshold absorption of the X-ray pulse photoelectron to free-electron conversion produces 'holes' of atoms and an ionization-induced X-ray transparency. Such transparency, due to the presence of free-particle vacancies, can be induced in all atomic, molecular and condensed matter systems at high intensity. Quantitative comparisons with theory allow us to test next-generation LCLS fluences and pulse duration. Our successful modeling of X-ray/atom interactions using a straightforward rate equation approach favorably for extension to complex systems.

... and this proposal ... adding to XFELs new functional capabilities customarily for atomic lasers.

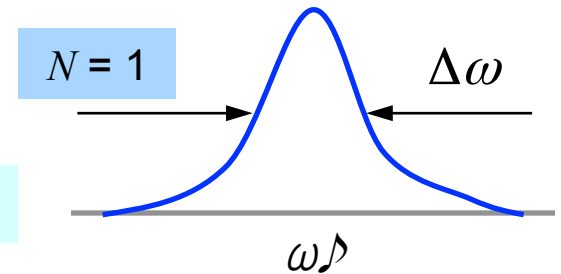


Cavity modes and frequency comb*

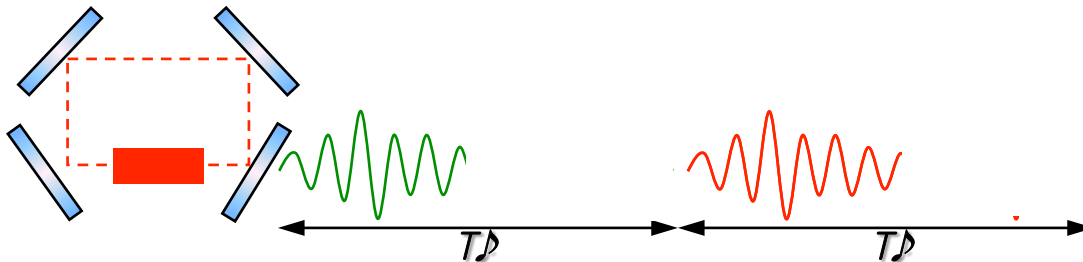
Single pulse \mathcal{D}



- $\Delta\omega$ is defined by atomic linewidth or gain bandwidth

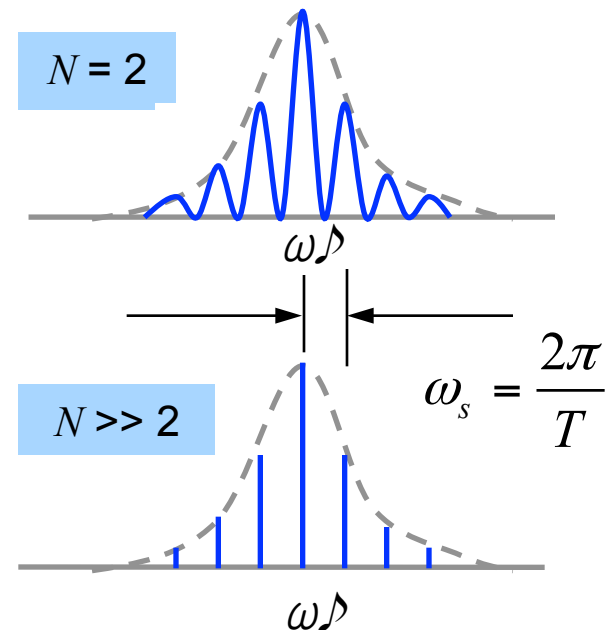


Cavity with a round trip time = $T\mathcal{D}$



A repeated waveform generates a spectral comb

- Mode spacing $\omega_s = 2\pi/T$
- Number of modes: $M = \Delta\omega/\omega_s$
- Linewidth of a mode $\sim 1/N$



* A.E. Siegman, *Lasers* (University Science Books, Sausalito, USA, 1986). See Chap. 27.

Signal

$$\varepsilon^{(N)}(t) = \sum_{n=0}^{N-1} \varepsilon(t - nT)$$

Spectral intensity

$$I^{(N)}(\omega) \sim |\tilde{\varepsilon}^{(N)}(\omega)|^2$$

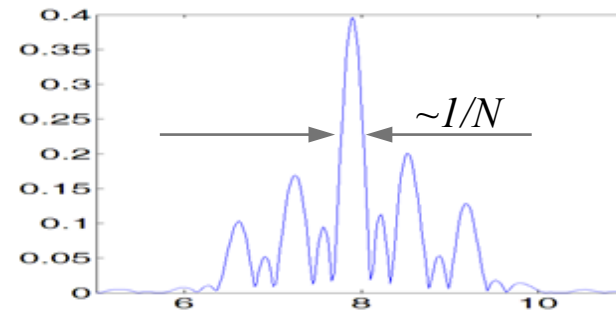
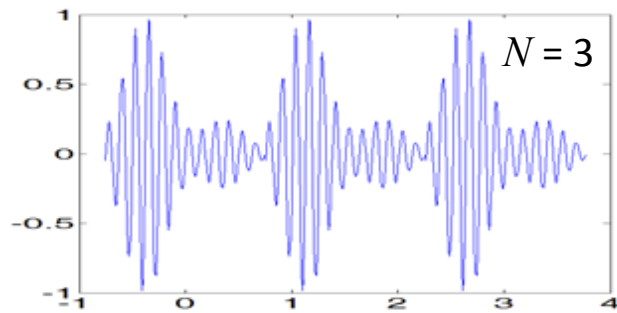
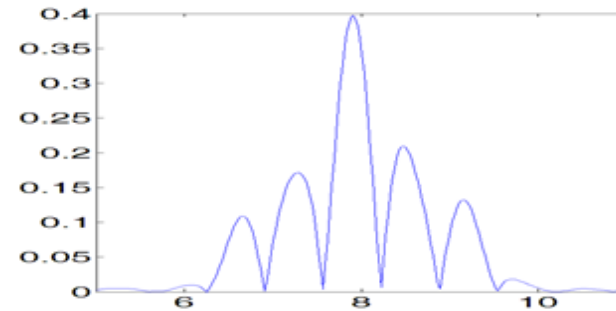
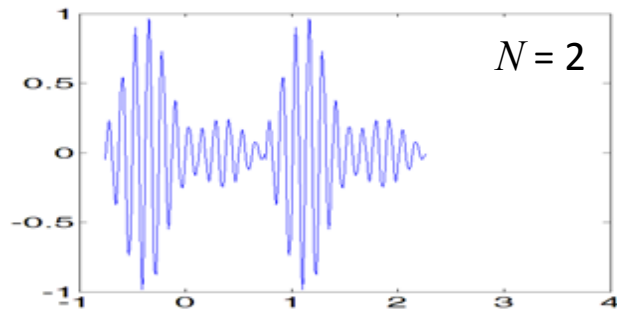
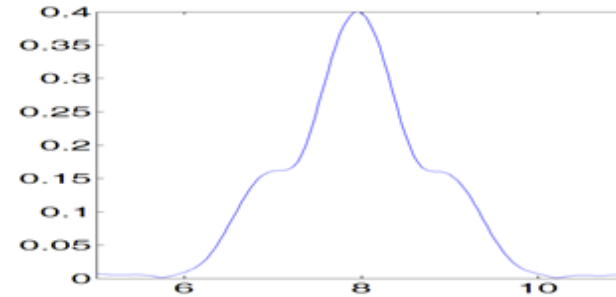
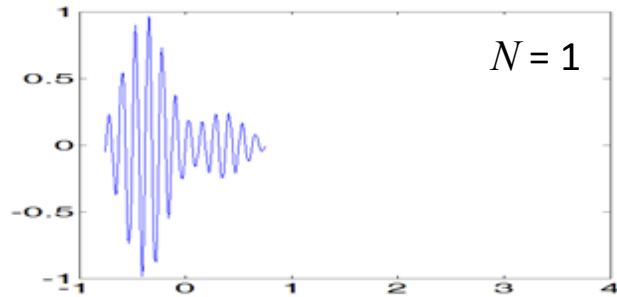
Applying Fourier time shifting identity and summation

$$F\{\varepsilon(t - nT)\} = e^{-in\omega T} F\{\varepsilon(t)\} \quad \sum_{n=0}^{N-1} e^{in\omega T} = (1 - e^{iN\omega T}) / (1 - e^{i\omega T})$$

One obtains:

$$I^{(N)}(\omega) = \left(\frac{\sin(N\omega T / 2)}{\sin(\omega T / 2)} \right)^2 I^{(0)}(\omega)$$

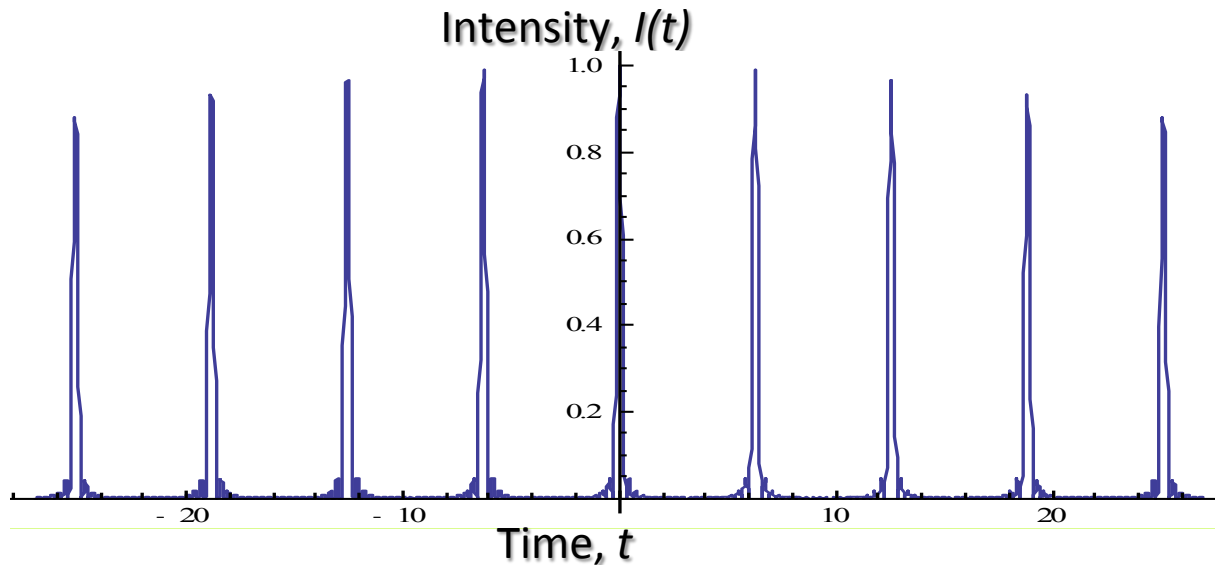
$$I^{(N)}(\omega) = \left(\frac{\sin(N\omega T / 2)}{\sin(\omega T / 2)} \right)^2 I^{(0)}(\omega)$$



When the axial modes are all in phase, then the time-domain signal has a pulsed pattern, closely resembling the comb structure of the frequency domain:

$$\varepsilon^{(N)}(t) = \varepsilon_0 \sum_{n=0}^{N-1} e^{-i(\omega_0 + n\omega_s)t} = \varepsilon_0 e^{-i\omega_0 t} \frac{1 - e^{-iN\omega_s t}}{1 - e^{-i\omega_s t}} \quad (\text{equal field amplitudes are assumed for simplicity})$$

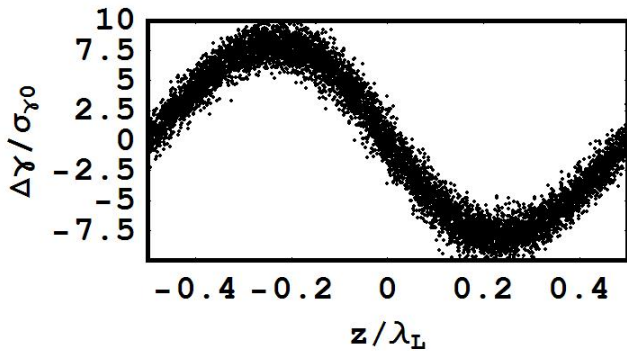
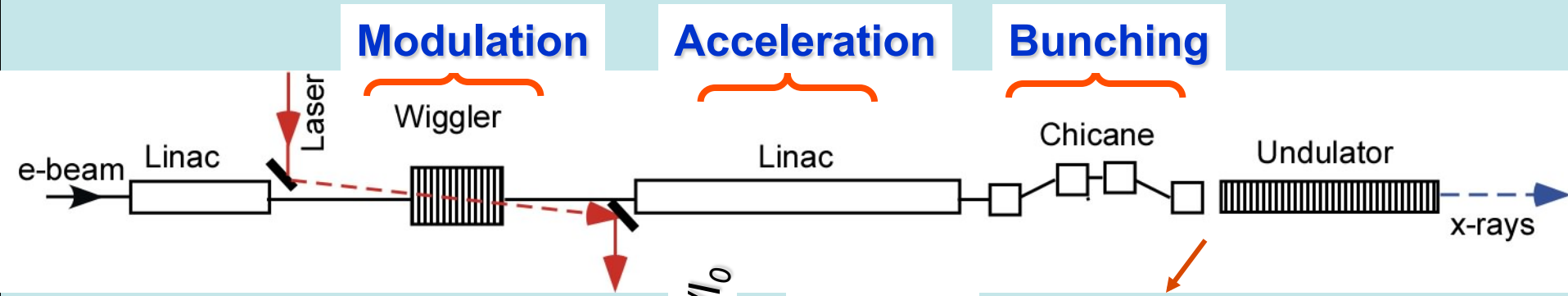
$$I^{(N)}(t) = \left(\frac{\sin(N\omega t / 2)}{\sin(\omega t / 2)} \right)^2 I^{(0)}(t)$$



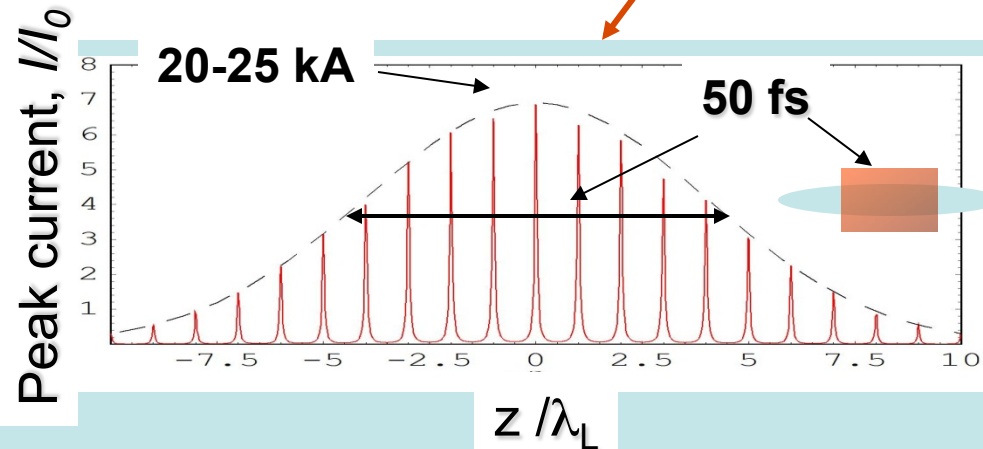
Modulation at round trip frequency or gain modulation causes mode-locking. A periodic pulse structure of sufficiently short pulses can be obtained when mode phase lock. Often locking only neighboring modes is sufficient.

FEL gain modulation through peak current modulation*

Optical manipulation of electrons to obtain a “train” of microbunches

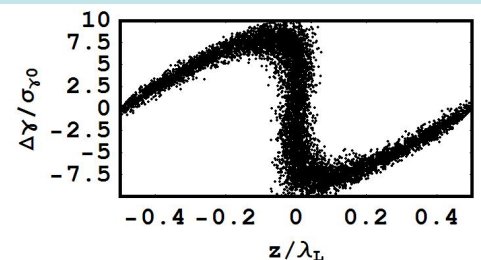


Only one optical cycle is shown



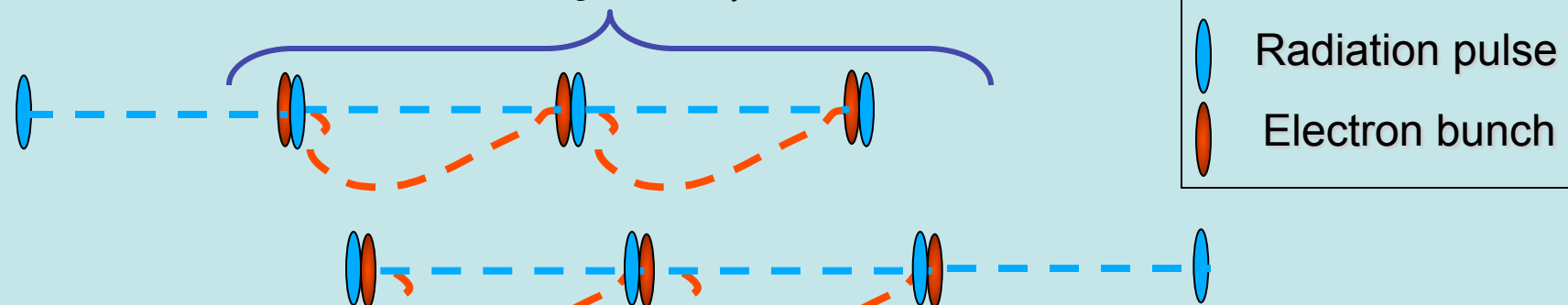
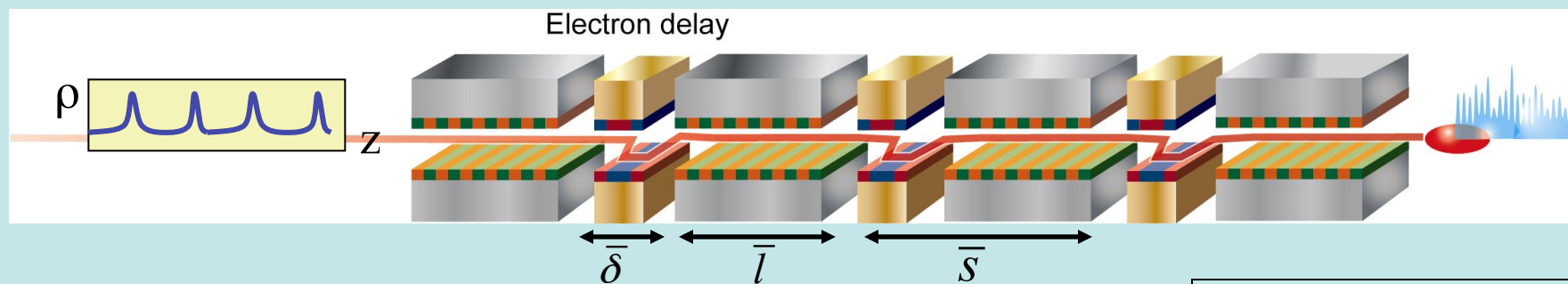
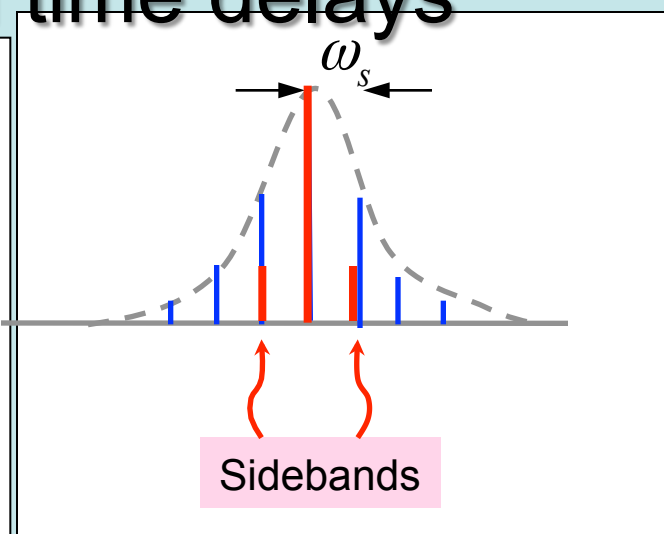
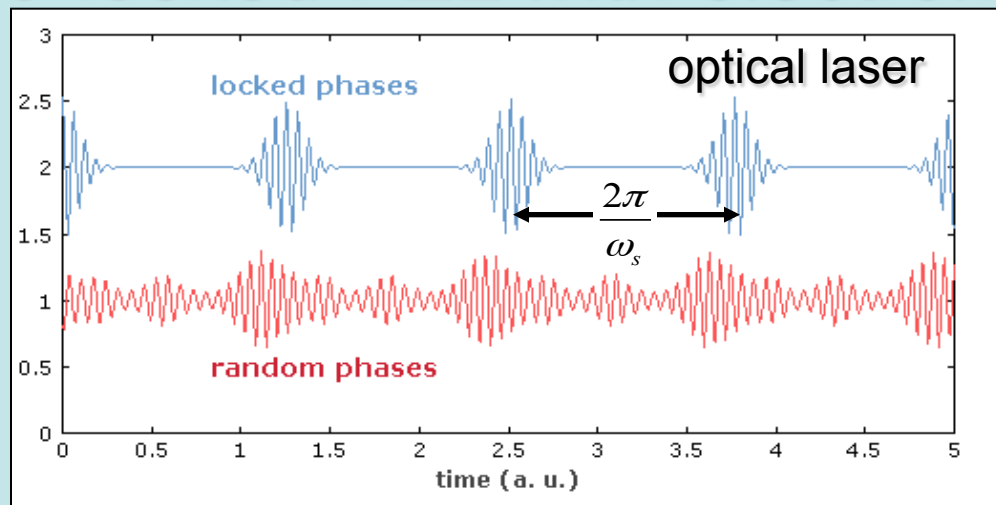
• Electron beam after bunching at optical wavelength

Region with large peak current gives the dominant x-ray radiation synchronized to laser source

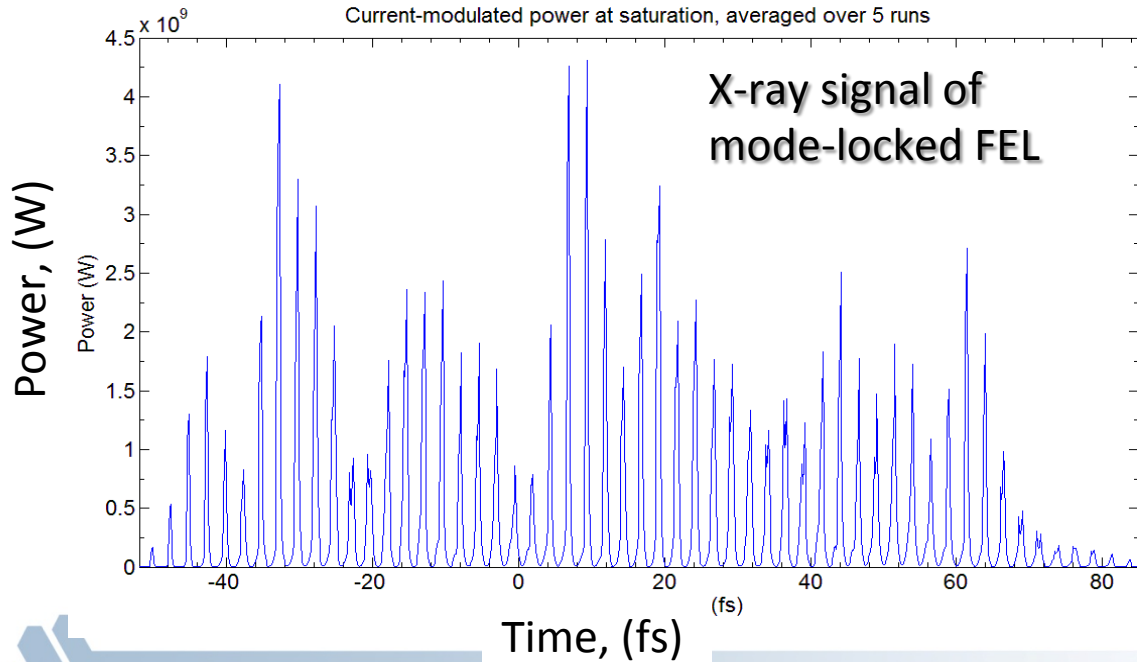
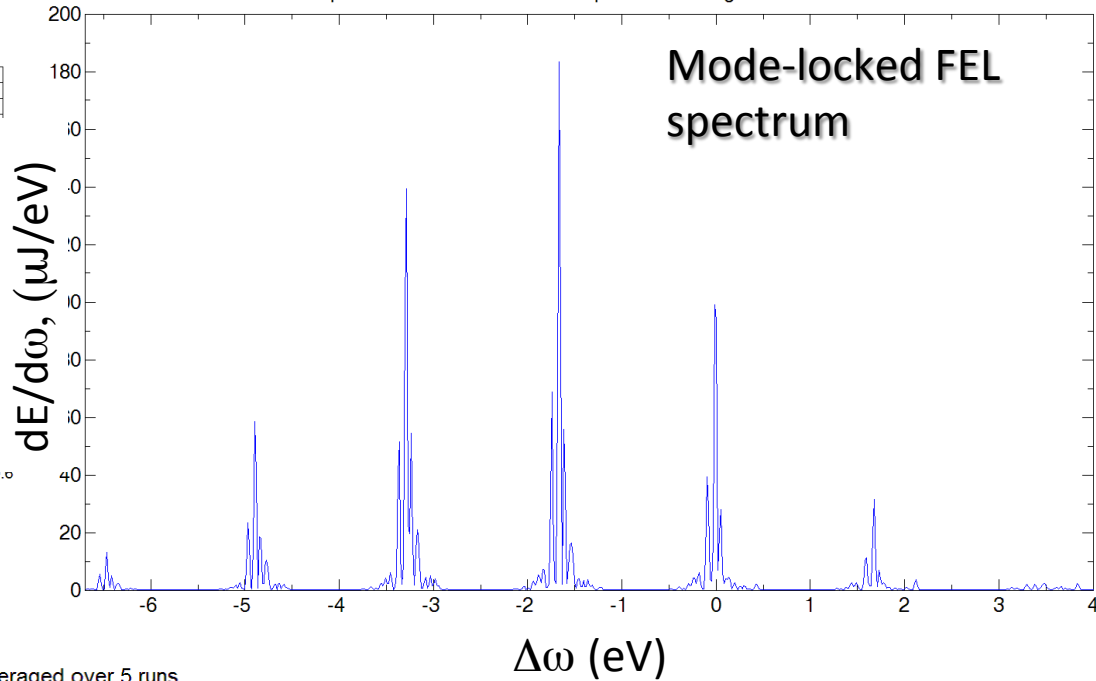
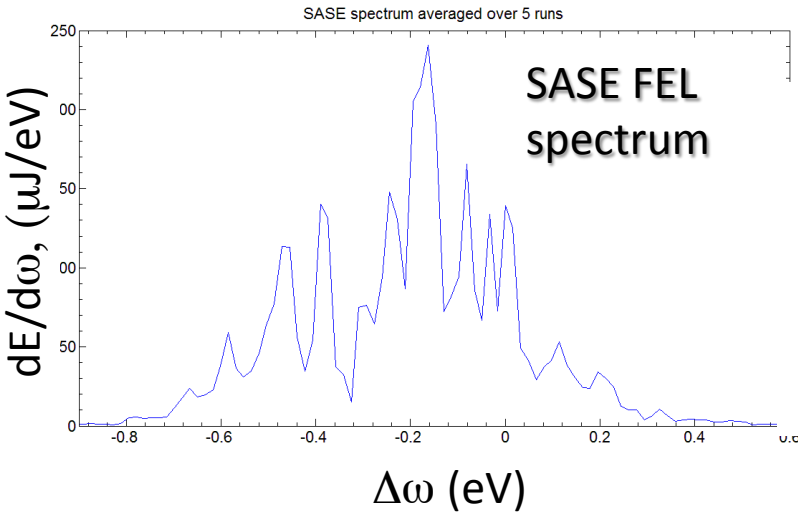


* A. Zholents, PRST-AB, 8, 040701(2004).

Mode-locked FEL with electron time delays*



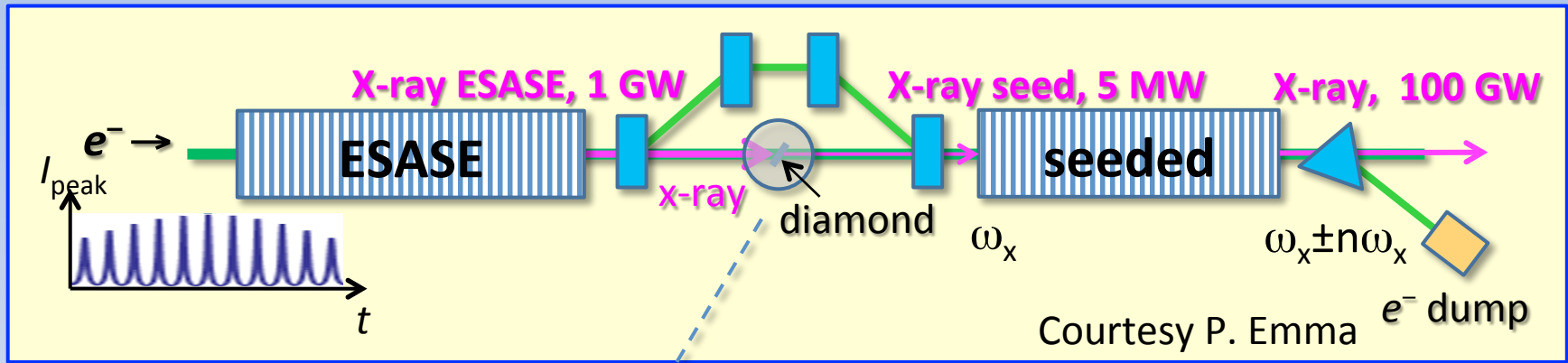
* N.R.Thompson and B.W.J.McNeil, PRL, 100, 203901(2008);
 E. Kur, et al., New Journal of Phys., 13, 063012(2011);
 C. Feng, et al., Phys. Rev. ST – AB, 15, 080703(2012).



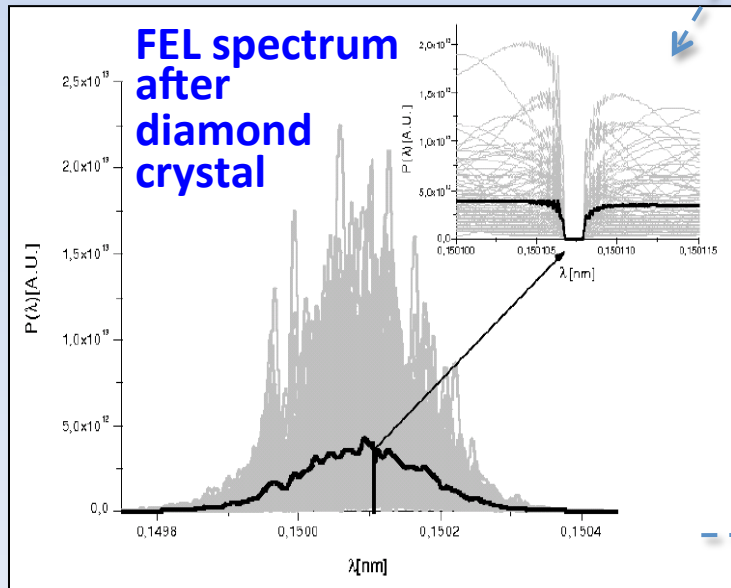
Generation of multiple frequency lines may enhance the capability of FELs in some specific applications:

- transmission and reflectivity spectra of various materials;
- resonant inelastic x-ray scattering (RIXS);
- ab-initio* phasing of nanocrystals.

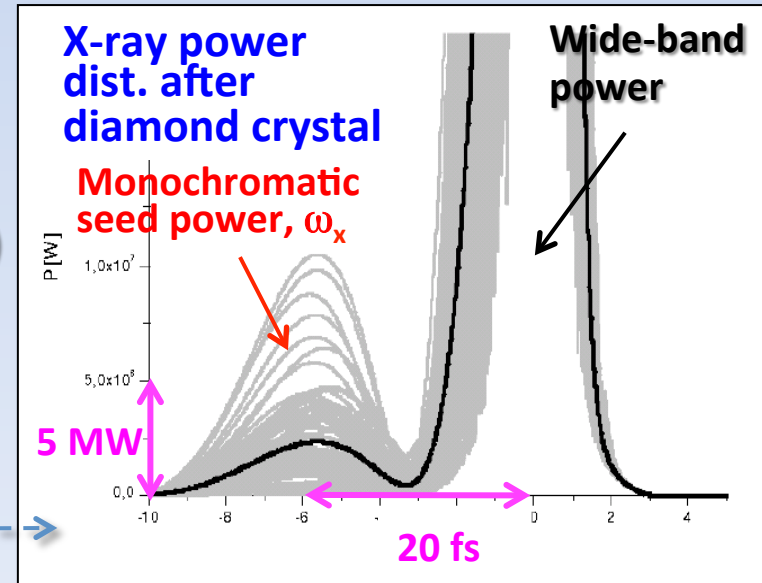
Self-seeding is a new path to a mode-locked FEL*



*) self-seeding idea by Geloni, Kocharyan, Saldin

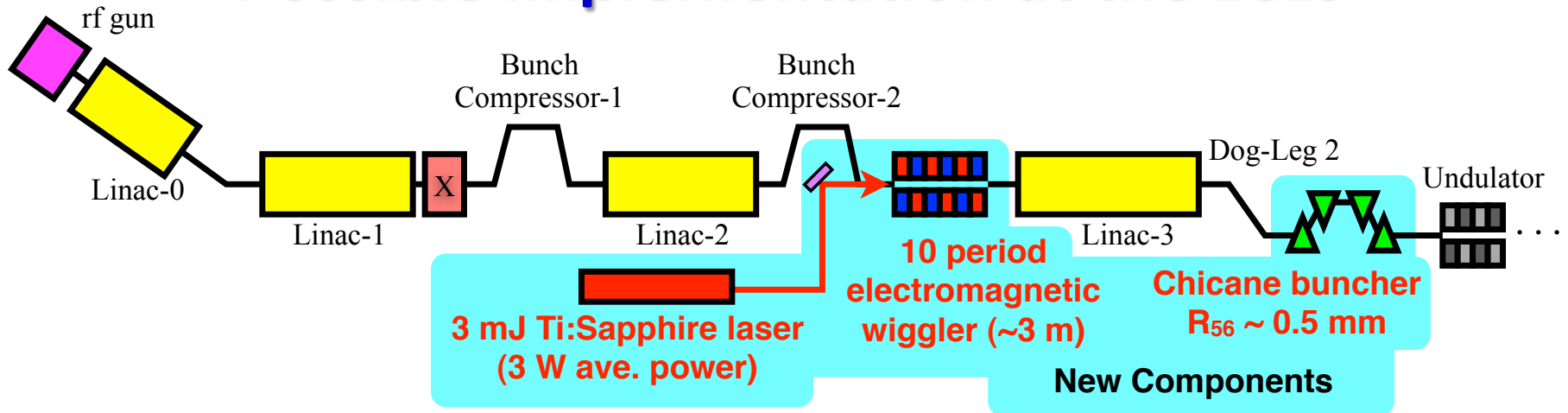


Use short, low-charge bunch to self-seed at 1.5 Å (20-40 pC)

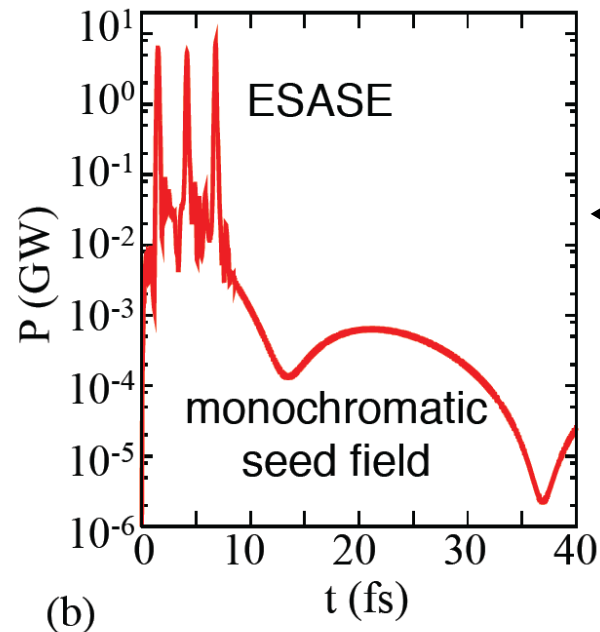
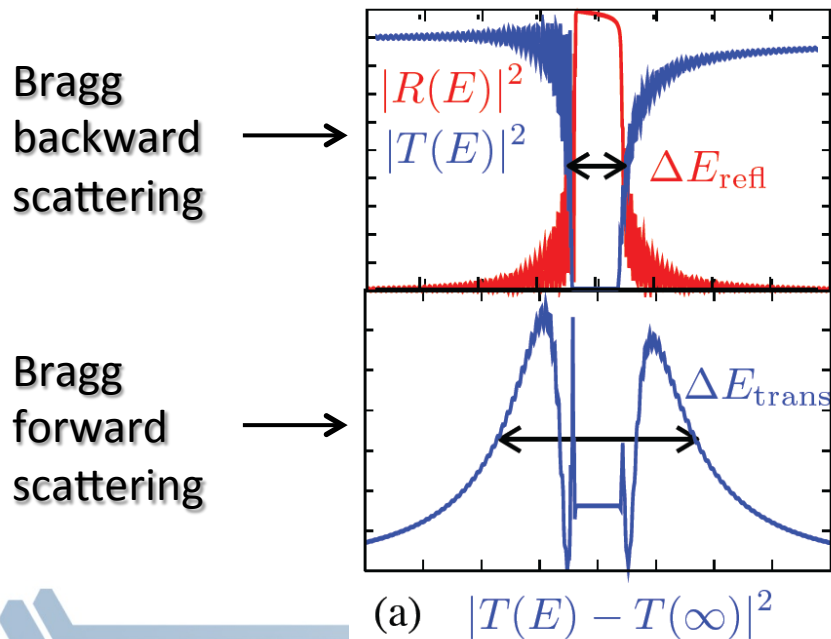


*D. Xiang, et al., Phys. Rev. ST-Accel. Beams, 1050707(2012).

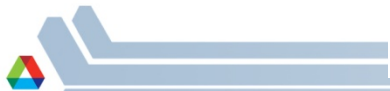
Possible implementation at the LCLS



Schematic of equipment additions within current LCLS layout.
Items highlighted in cyan are the new components.



Example of the seeding field for the proposed system

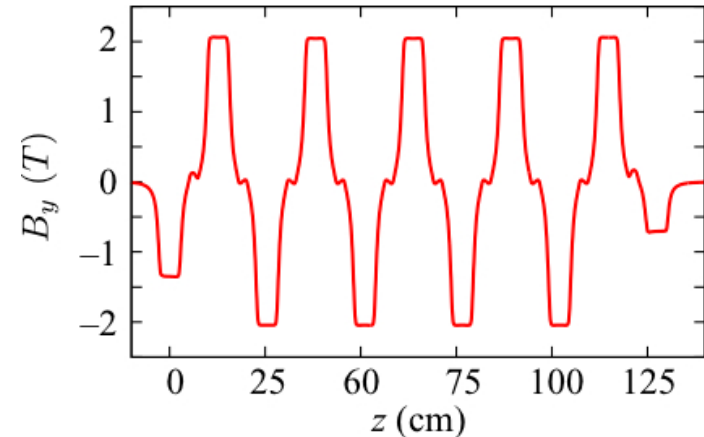


Possible implementation at the LCLS (2)

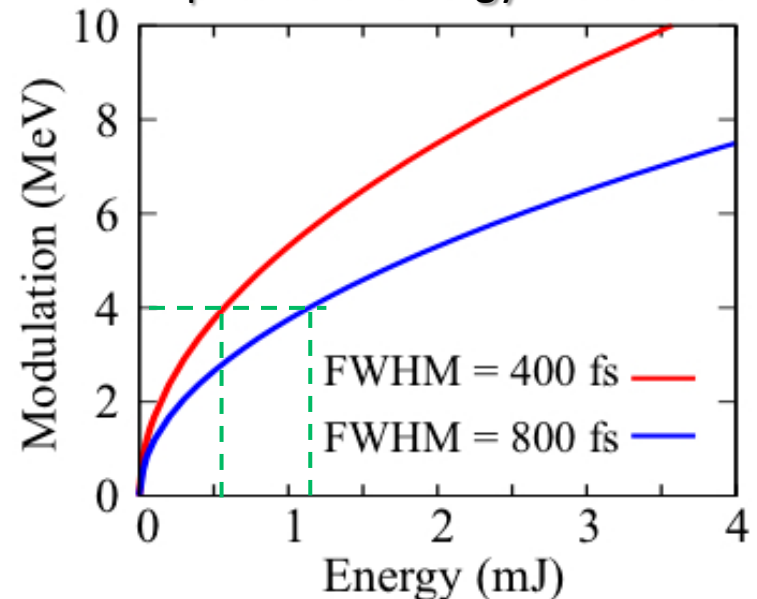
Nominal parameters used in simulations

Parameter	Symbol	Value
Peak laser power		1.3 GW
Electron beam energy spread	σ_γ	2 MeV
Wiggler length	L_u	2.5 m
Wiggler period	λ_u	25.5 cm
Wiggler parameter value		31
Energy modulation amplitude	$\Delta\gamma$	4 MeV
Electron energy at wiggler	$\gamma_r mc^2$	4.5 GeV
Electron energy at buncher	$\gamma_0 mc^2$	13.6 GeV
Momentum compaction of buncher	R_{56}	0.55 mm
Total length of buncher		$\lesssim 10$ m
Final spike peak current	I_{peak}	10 kA
Current spike duration (FWHM)		0.5 fs

Wiggler magnetic field



Amplitude of energy modulation



Possible implementation at the LCLS (3)

Bragg reflection condition from the (004) atomic planes of diamond

Length over which the field is reflected from the crystal $\sim 23 \mu\text{m}$

Bragg angle $\sim 54^\circ$, 8 keV x-ray

$$\Delta t_{wake} \sim \frac{\Lambda^2 \sin \theta}{cd} \approx 15 \text{ fs}$$

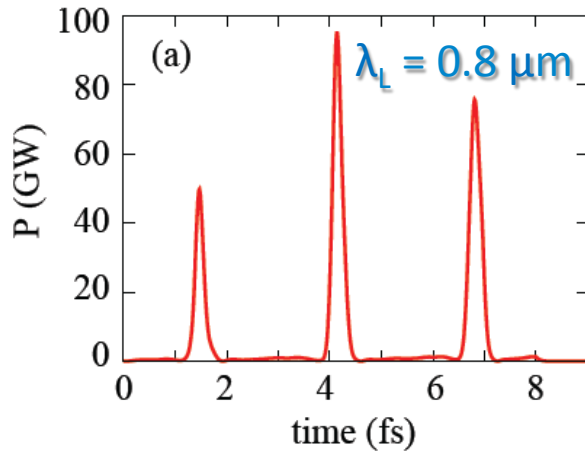
Duration of the “wake”

Diamond crystal thickness, $100 \mu\text{m}$

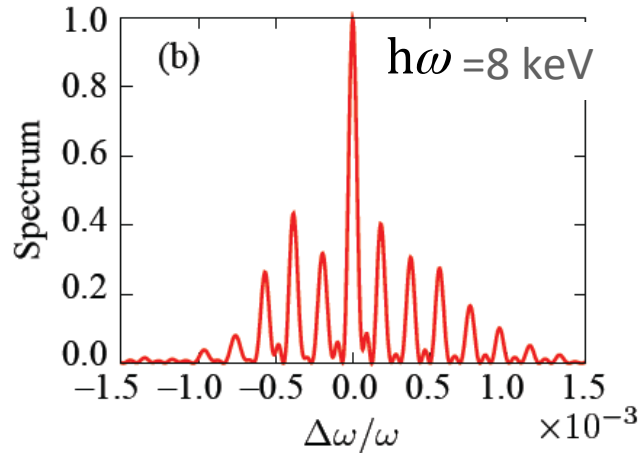


Possible implementation at the LCLS (4)

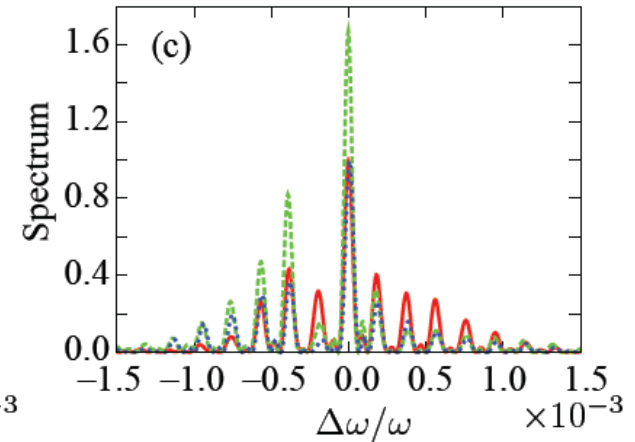
Simulation results



Single shot x-ray power profile



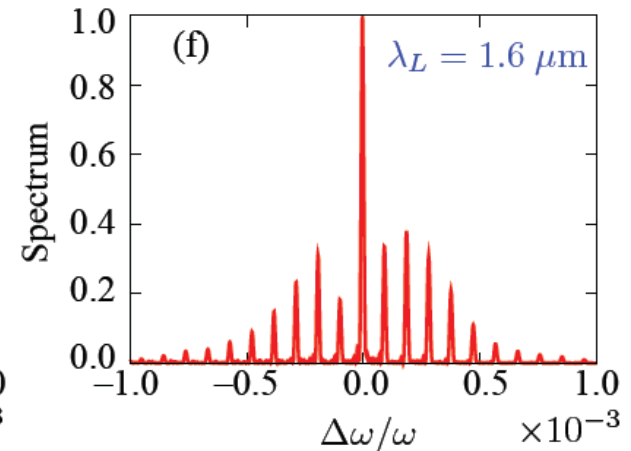
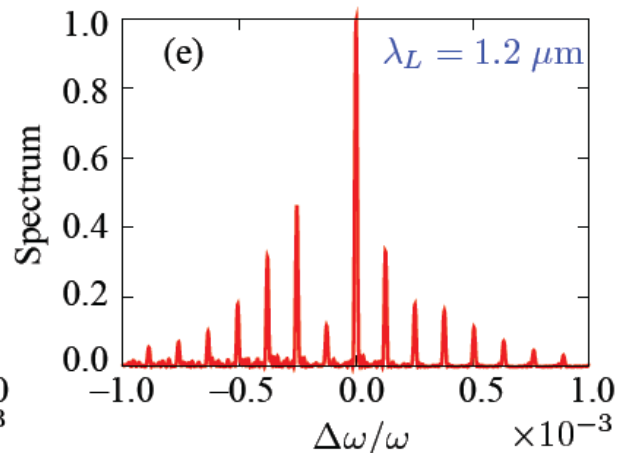
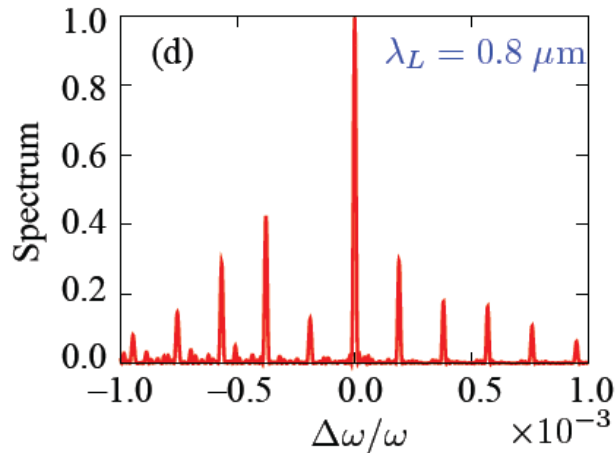
Single shot x-ray spectrum



Three statistically independent trials showing the fluctuation level due to SASE

Possible extension to a better mode-locked FEL

Using reflection from 331 atomic planes in diamond for 7.6 keV x-rays leads to a seeding wake structure ≥ 50 fs and ≥ 40 fs long bunch train*



Single shot x-ray power profile as before, but with 5 times narrower spikes

Changing laser modulation frequency allows control over spike separation

* longer delay chicane than is presently available at the LCLS self-seeding monochromator is required

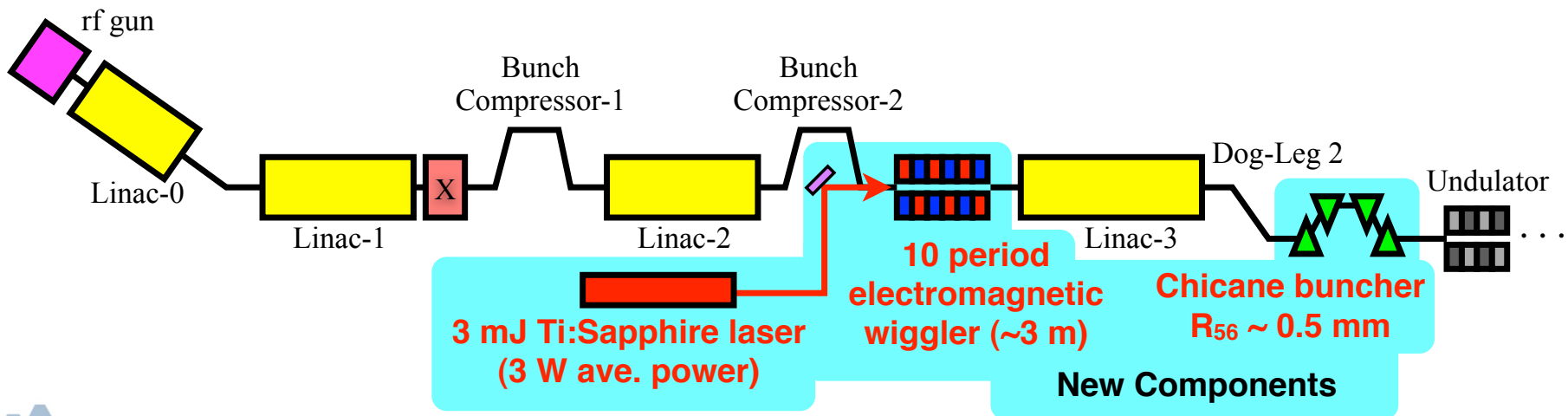


Summary

1)



2) Implementation of mode-locking at LCLS is possible with minimal perturbation to current operation



Thank you for your attention

