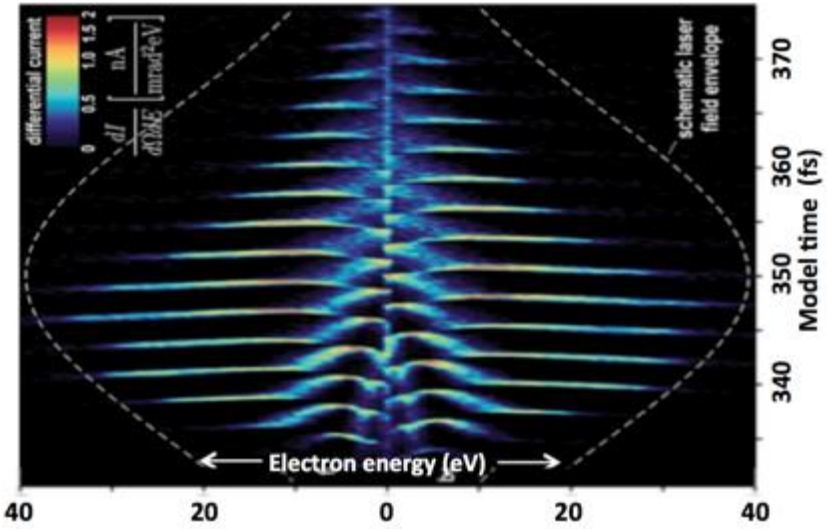
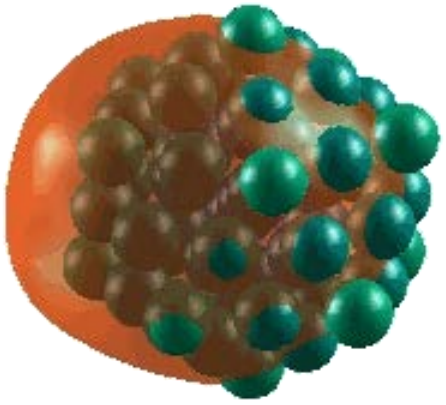




Fennel, Doeppner et al., PRL 98, 0143401 (2007)

Resolving collective electron dynamics in nanoplasmas with time resolution of 1 femtosecond

LH13@AMOLCLS, 3 – 8 July 2015

T. Doeppner, T. Fennel, C Bostedt

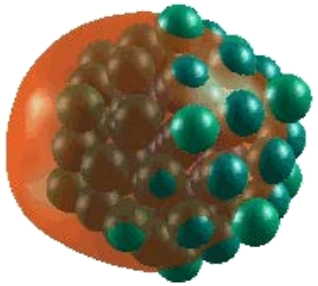
B. Bachmann, M. Bucher, S. Carron, R. Coffee, P. Drake, K. Ferguson, L. Fletcher, E. Gamboa, S. Glenzer, S. Goede, T. Gorkhover, S. Hau-Riege, D. Kraus, A. Levitan, M. MacDonald, KH Meiwes-Broer, T. Osipov, T. Pardini, C. Peltz, S. Skruszewicz

SLAC NATIONAL ACCELERATOR LABORATORY

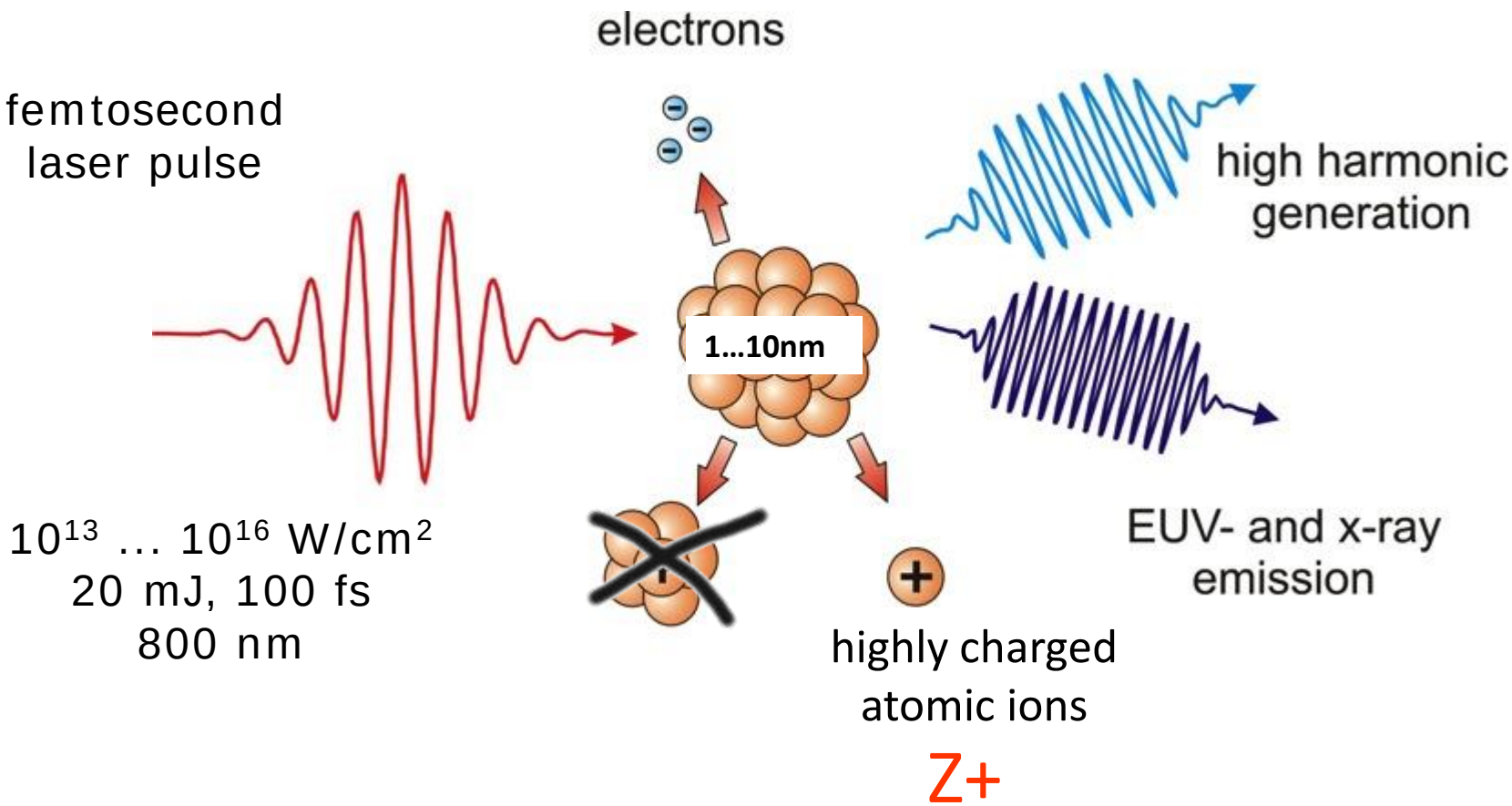
Berkeley UNIVERSITY OF CALIFORNIA

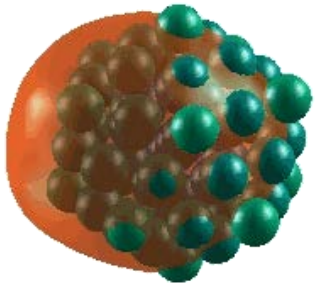
UNIVERSITÄT ROSTOCK gegründet 1419

UNIVERSITY OF MICHIGAN



Atomic Clusters are model systems for laser-matter interaction





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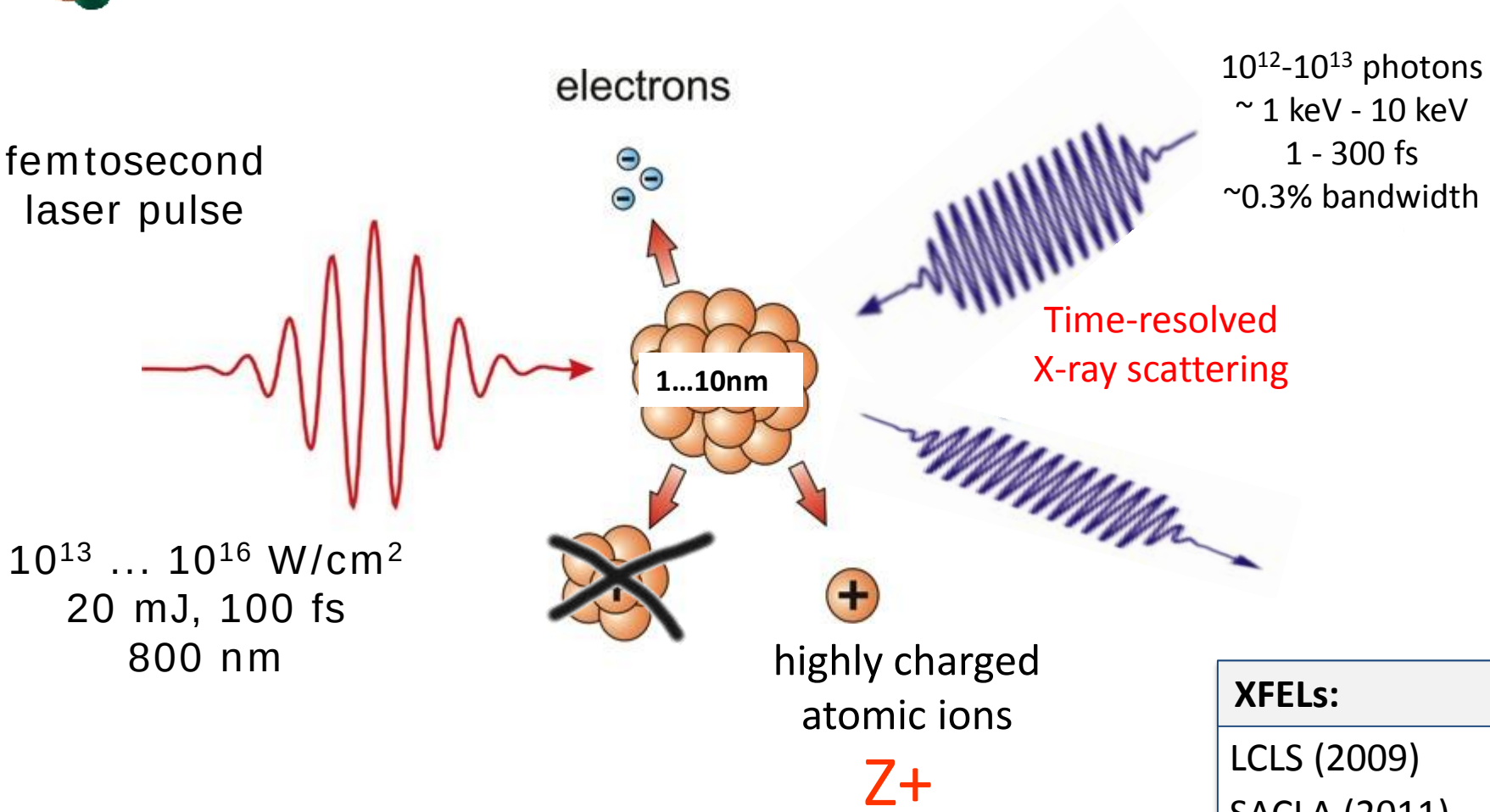
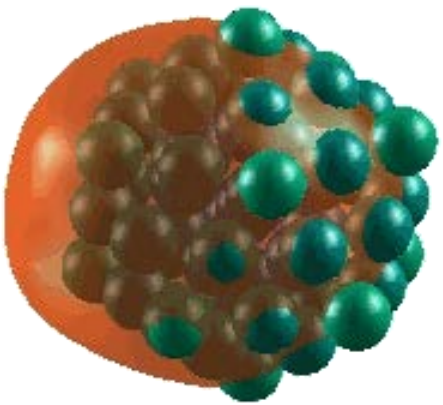


X-ray Free Electron Lasers, for the first time, allow to study transient properties of the nanoplasmas

XFELs:
LCLS (2009)
SACLA (2011)
Eur. XFEL (2016)
SwissFEL (2016)

Optical response in nano-sized particles is dominated by the surface plasmon

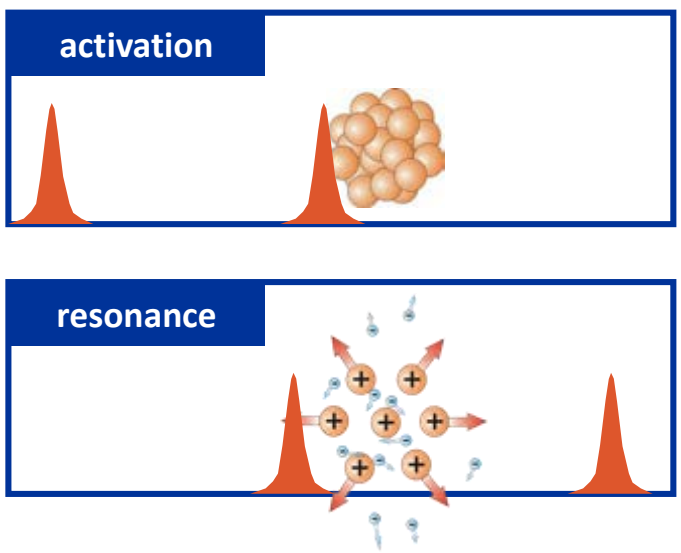
Idea

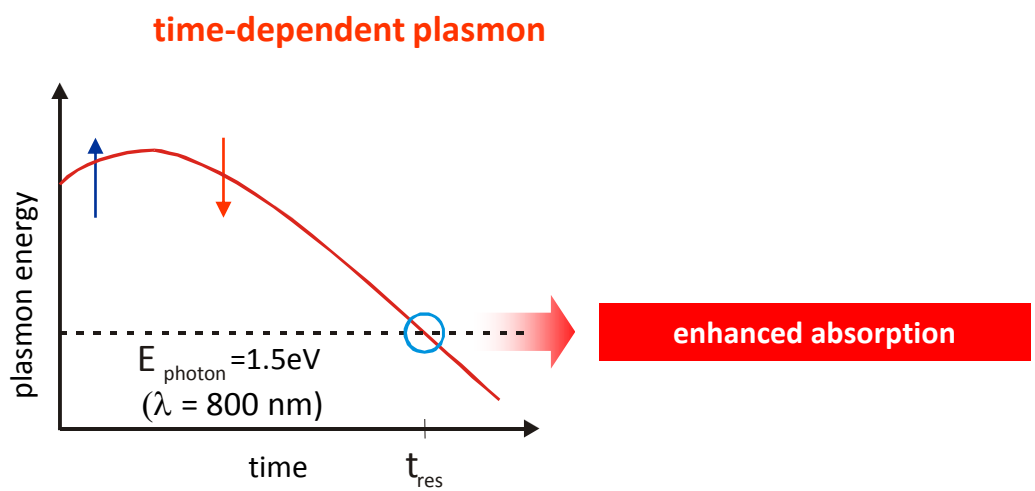


classical analogon: Mie-plasmon

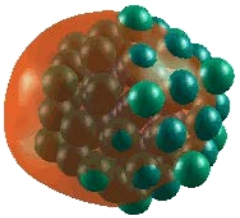
$$\omega_{Mie} = \sqrt{\frac{n_{bg} e^2}{3 m \varepsilon_o}} = \frac{\omega_p}{\sqrt{3}}$$

G. Mie (1908)

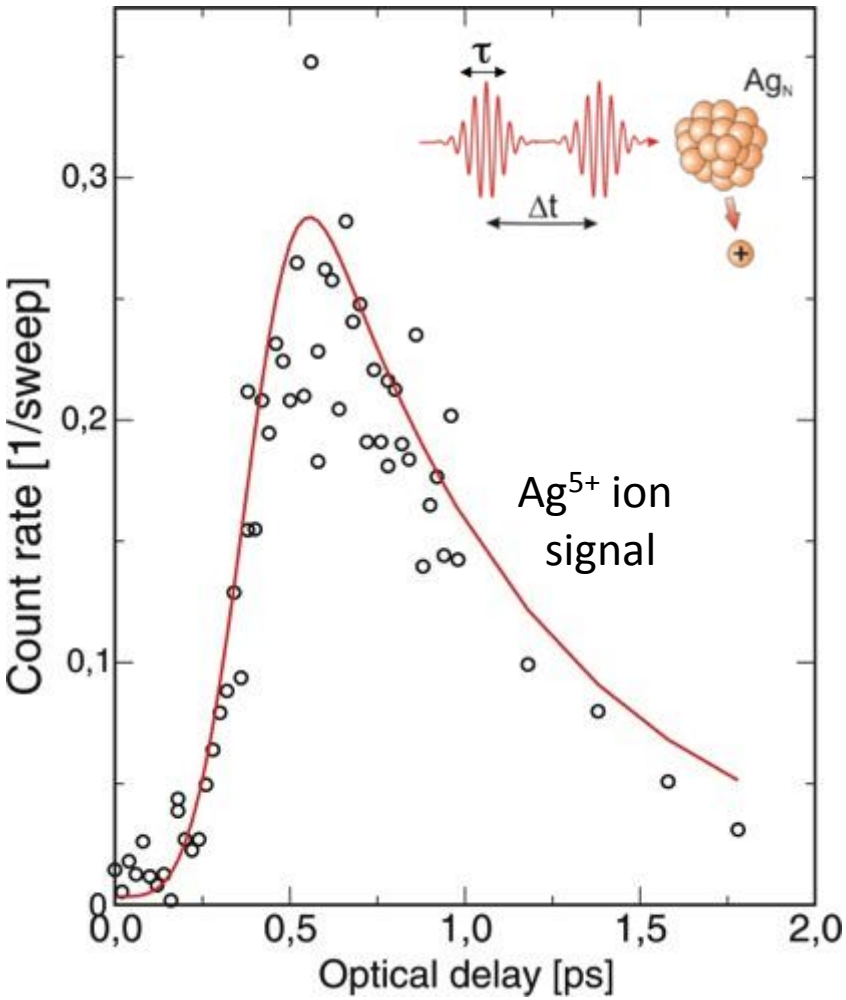




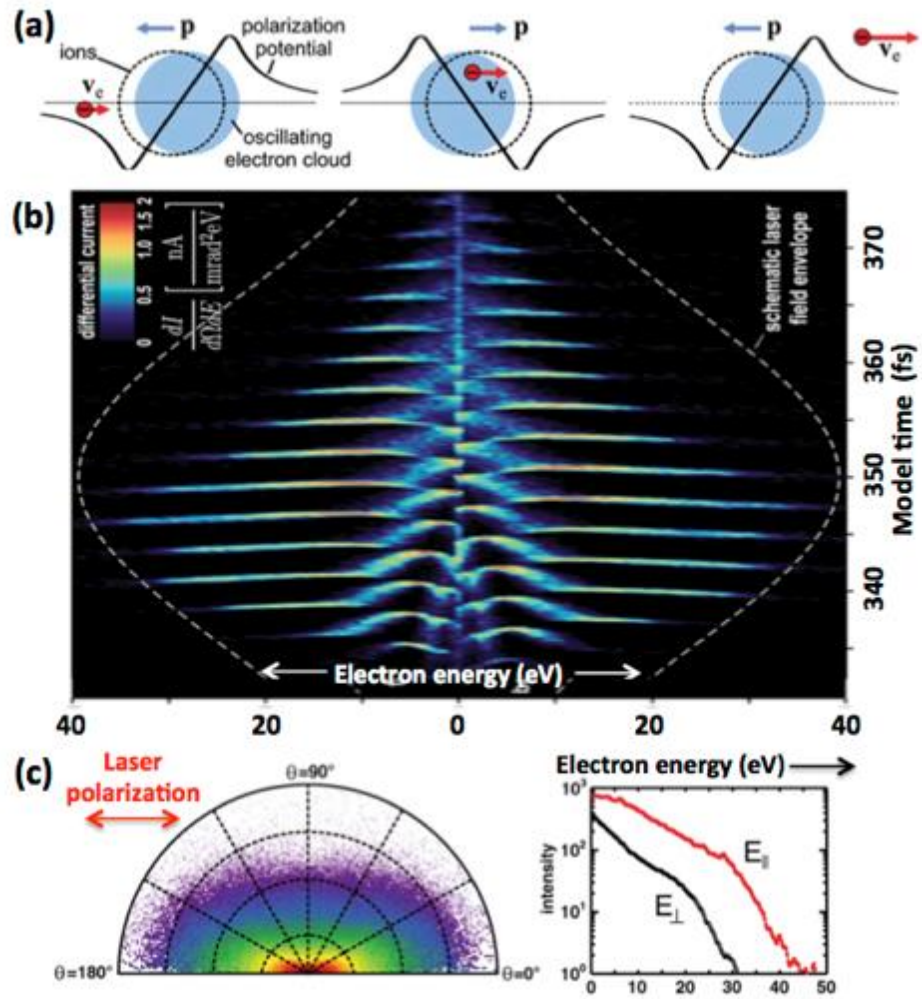
Döppner, Fennel et al., Phys. Rev. Lett. **94**, 013401 (2005).



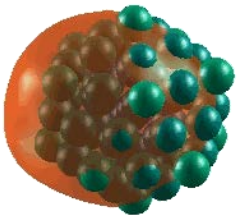
Resonant plasmon excitation results in enhanced ionization and directed electron emission



Doeppner, Fennel et al., PRL 94, 013401 (2005)



Fennel, Doeppner et al., PRL 98, 0143401 (2007)



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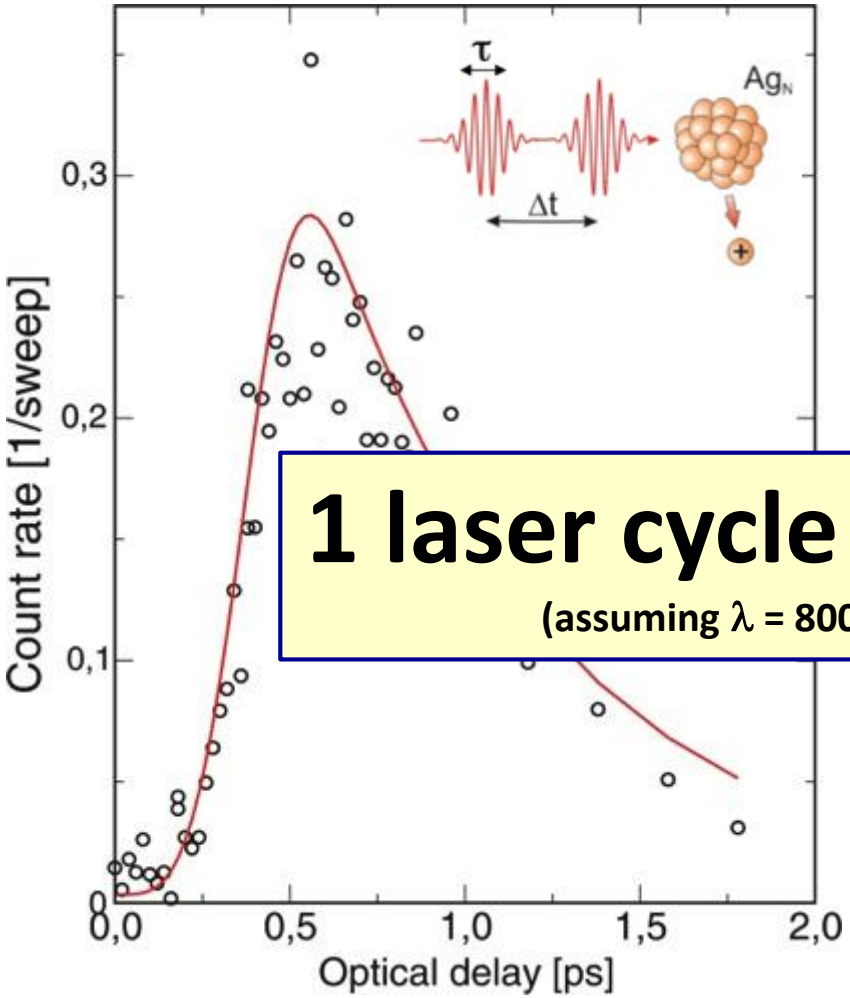
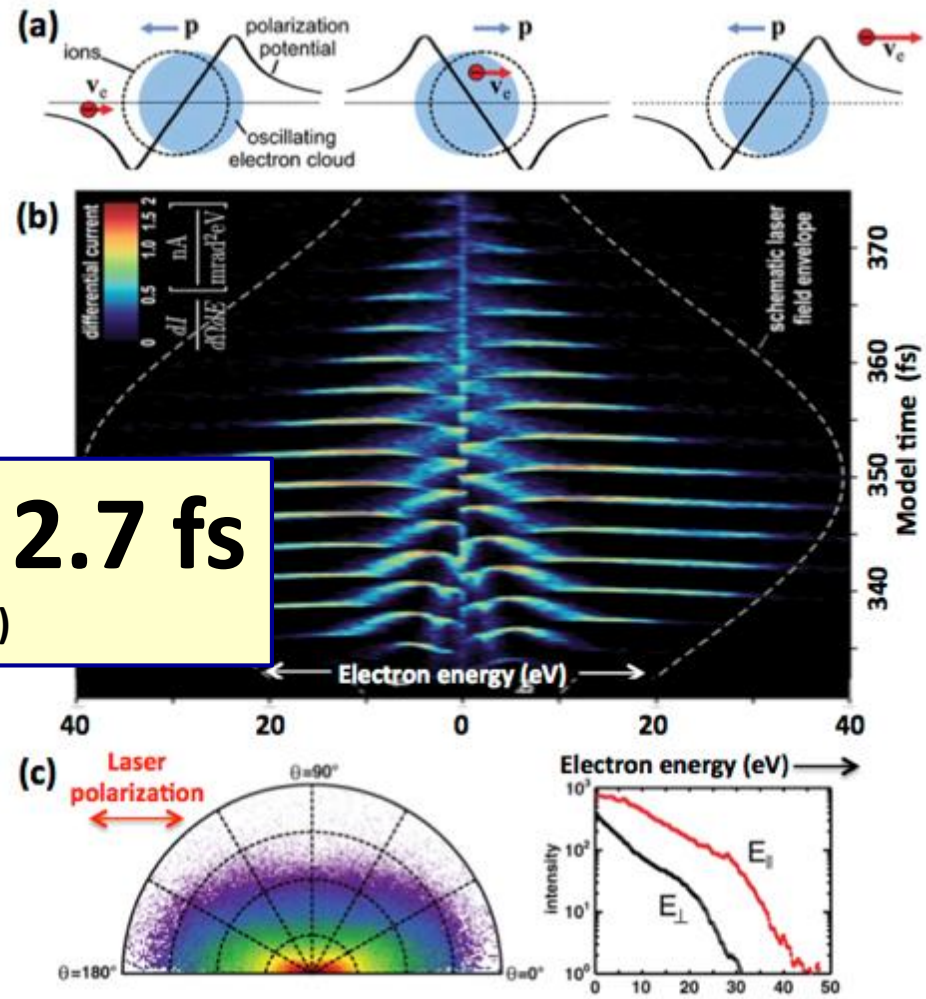


1 laser cycle = 2.7 fs

(assuming λ = 800 nm)



Doeppner, Fennel et al., PRL 94, 013401 (2005)

Fennel, Doeppner et al., PRL 98, 0143401 (2007)

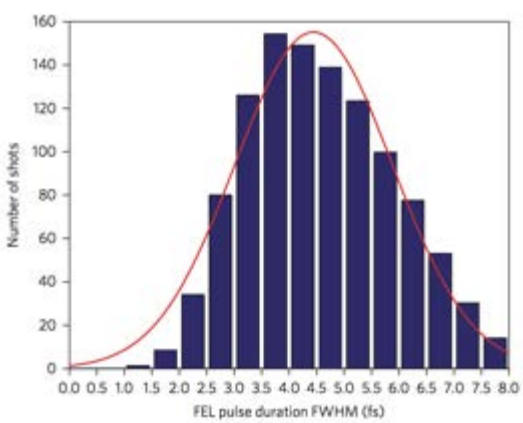
Generation and Measurement of ~ 1 fs short x-ray pulses has been demonstrated at LCLS/ SLAC

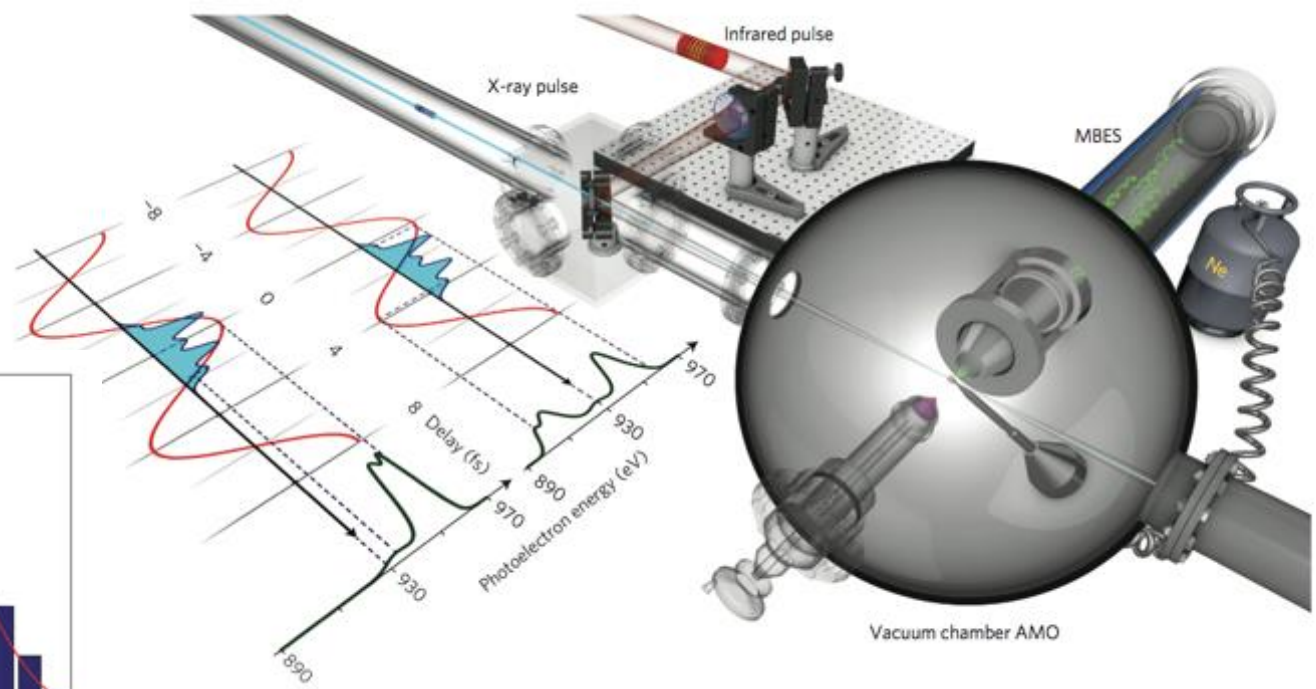
nature photonics

Vol. 8, p950 (2014)

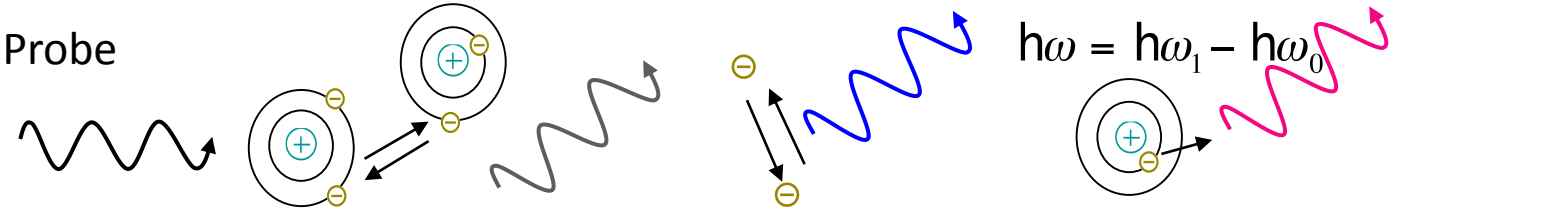
Measuring the temporal structure of few-femtosecond free-electron laser X-ray pulses directly in the time domain

W. Helml[1,2†], A. R. Maier[3,4†], W. Schweinberger[2], I. Grguraš[3,5], P. Radcliffe[6], G. Doumy[7,8], C. Roedig[8], J. Gagnon[2], M. Messerschmidt[9], S. Schorb[9], C. Bostedt[9], F. Grüner[3,4], L. F. DiMauro[8], D. Cubaynes[10], J. D. Bozek[9], Th. Tschentscher[6], J. T. Costello[11], M. Meyer[6,10], R. Coffee[9], S. Düsterer[12], A. L. Cavalieri[3,5] and R. Kienberger[1,2*]



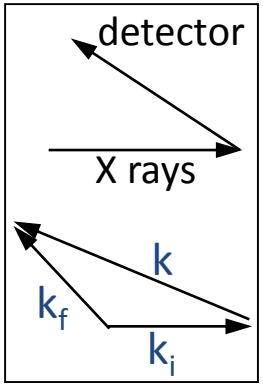


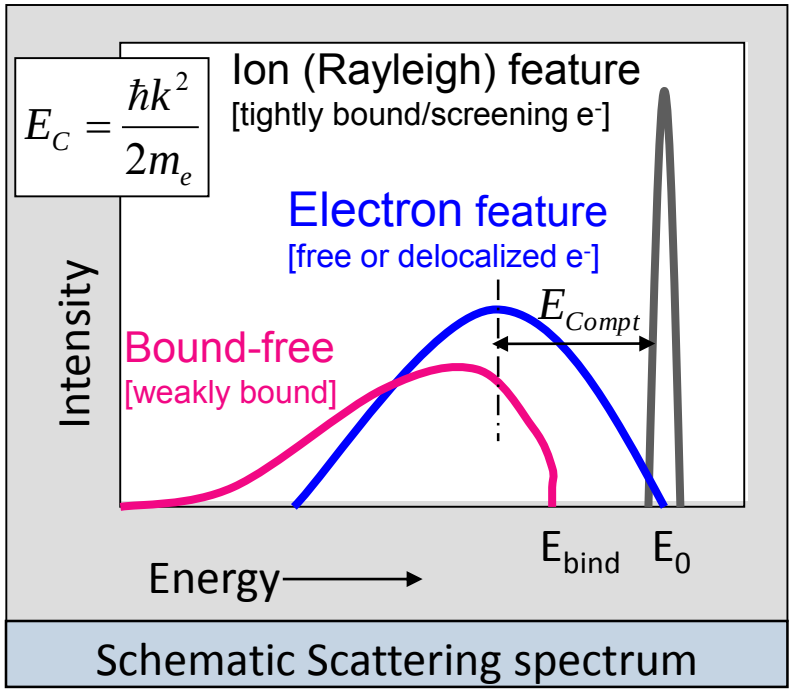
Spectrally resolved x-ray Thomson scattering has been developed as a reliable diagnostic for dense plasma parameters (T_e, n_e, S_{ii} etc.) on larger laser facilities (Omega, JLF@LLNL, NIF)



$$S(k,\omega) = \left|f_I(k) + q(k)\right|^2 S_{ii}(k,\omega) + Z_f S^0_{ee}(k,\omega) + Z_b \int \tilde{S}_{ce}(k,\omega-\omega')S_s(k,\omega')d\omega'$$

Ion feature | Electron feature | Bound-free

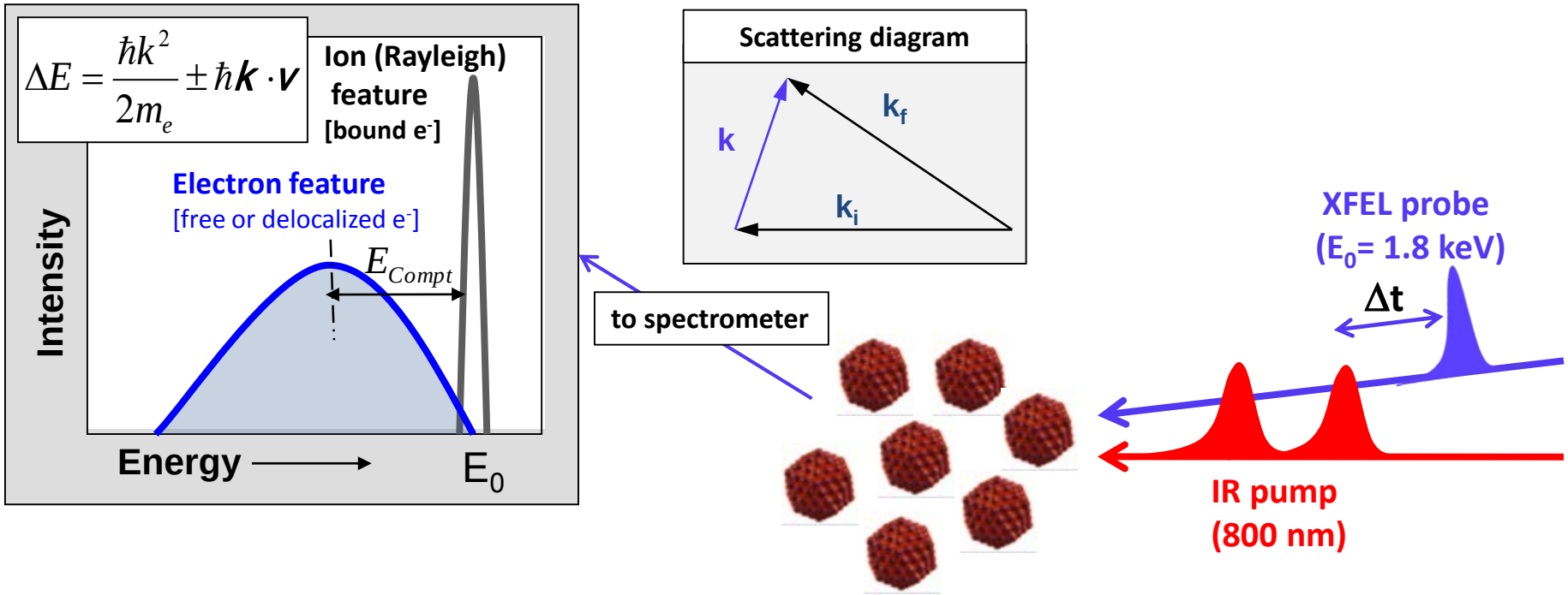




- Free or delocalized electrons result in the Compton down-shifted line, $Z_f\, S_{ee}\,(k,\omega)$
- Compton profile is Doppler-broadened through relation $\pm\hbar\boldsymbol{k}\bullet\boldsymbol{v}$; → maps electron velocity distribution along scattering vector $\boldsymbol{k}$
- Bound-free contribution also results into down-shifted spectrum $Z_b\, S_{ce}\,(k,\omega)$
- The ion feature describes elastic scattering $S_{ii}(k,\omega)$

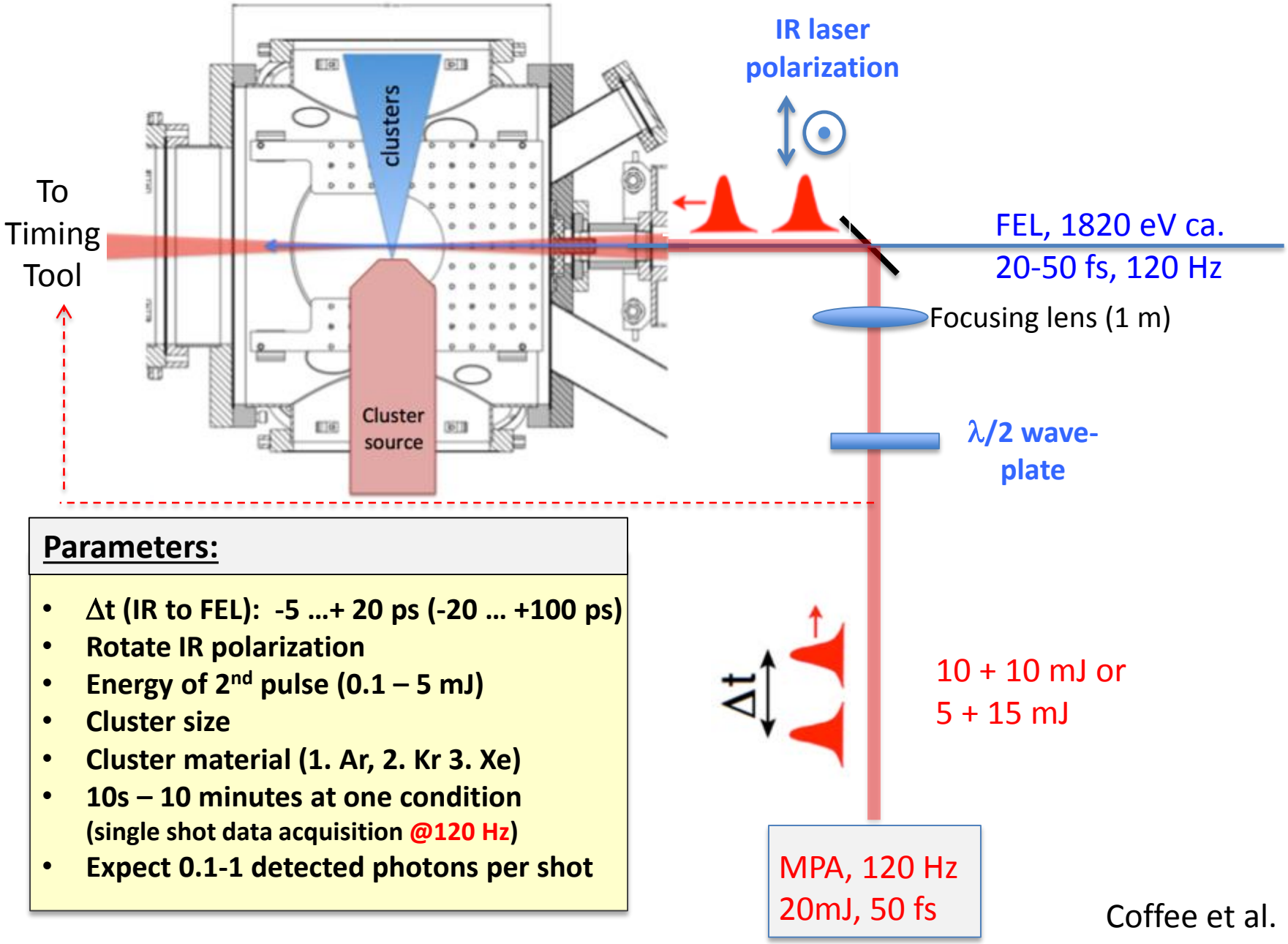
Chihara, JPCM (2000); Gregori et al., PRE (2003), Glenzer & Redmer, RMP (2009)

Anisotropic collective electron dynamics can be measured in forward scattering geometry



The IR pump polarization can be varied to scan the scattering vector (k) orientation with respect to the collective electron motion

First implementation at AMO@LCLS in July 2015



Parameters:

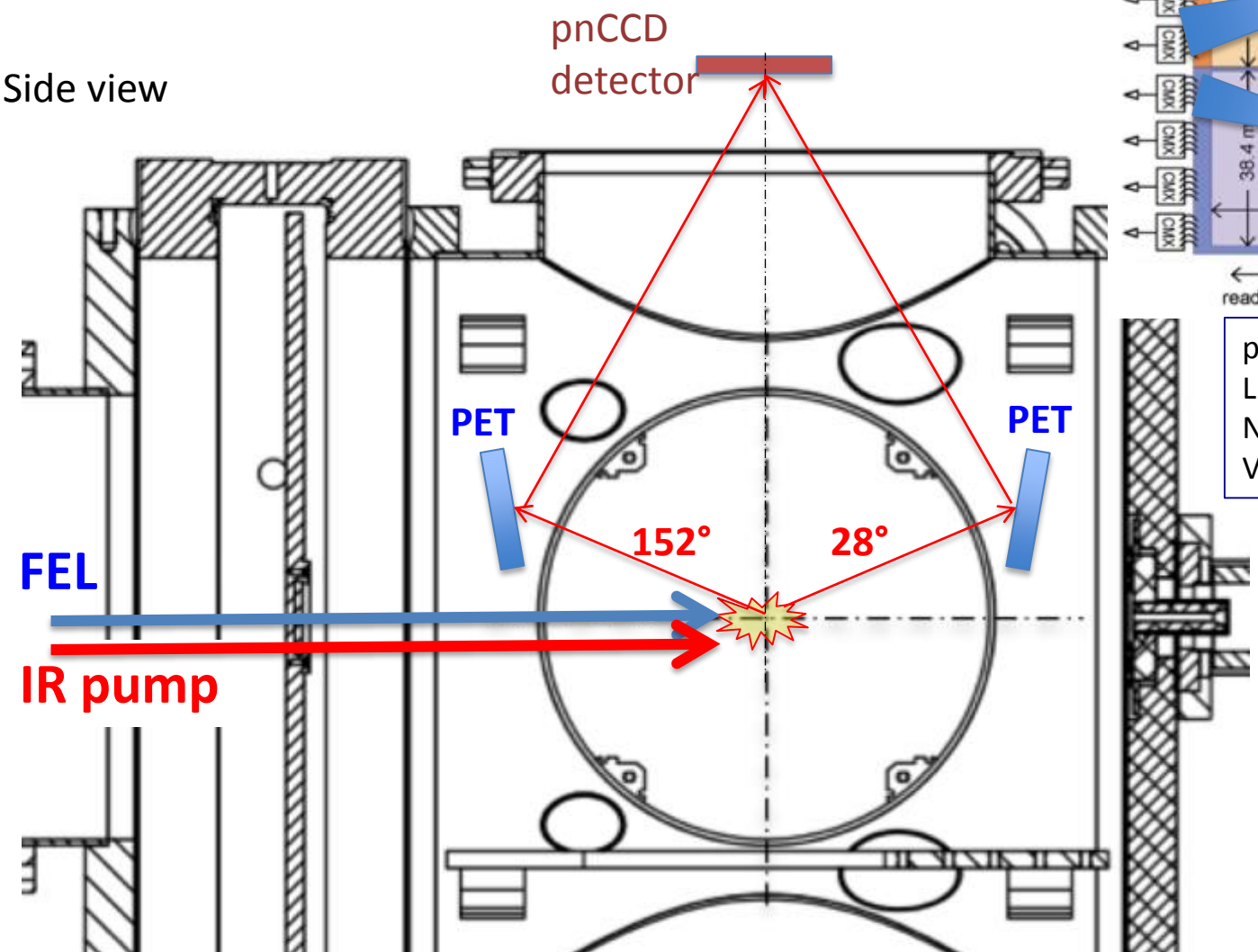
- **Δt (IR to FEL): -5 …+ 20 ps (-20 … +100 ps)**
- **Rotate IR polarization**
- **Energy of 2nd pulse (0.1 – 5 mJ)**
- **Cluster size**
- **Cluster material (1. Ar, 2. Kr 3. Xe)**
- **10s – 10 minutes at one condition (single shot data acquisition @120 Hz)**
- **Expect 0.1-1 detected photons per shot**

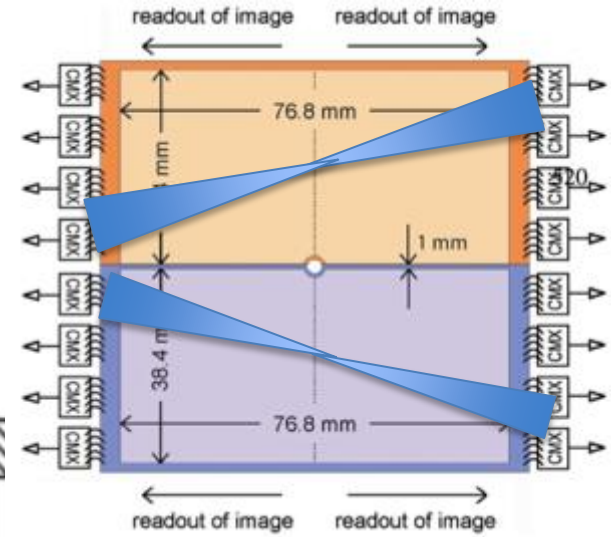
Coffee et al.

X-ray spectra at 2 scattering angles can be simultaneously recorded on large area pnCCD detector at 120 Hz

Center Bragg angle at 51 deg (PET@1820eV)

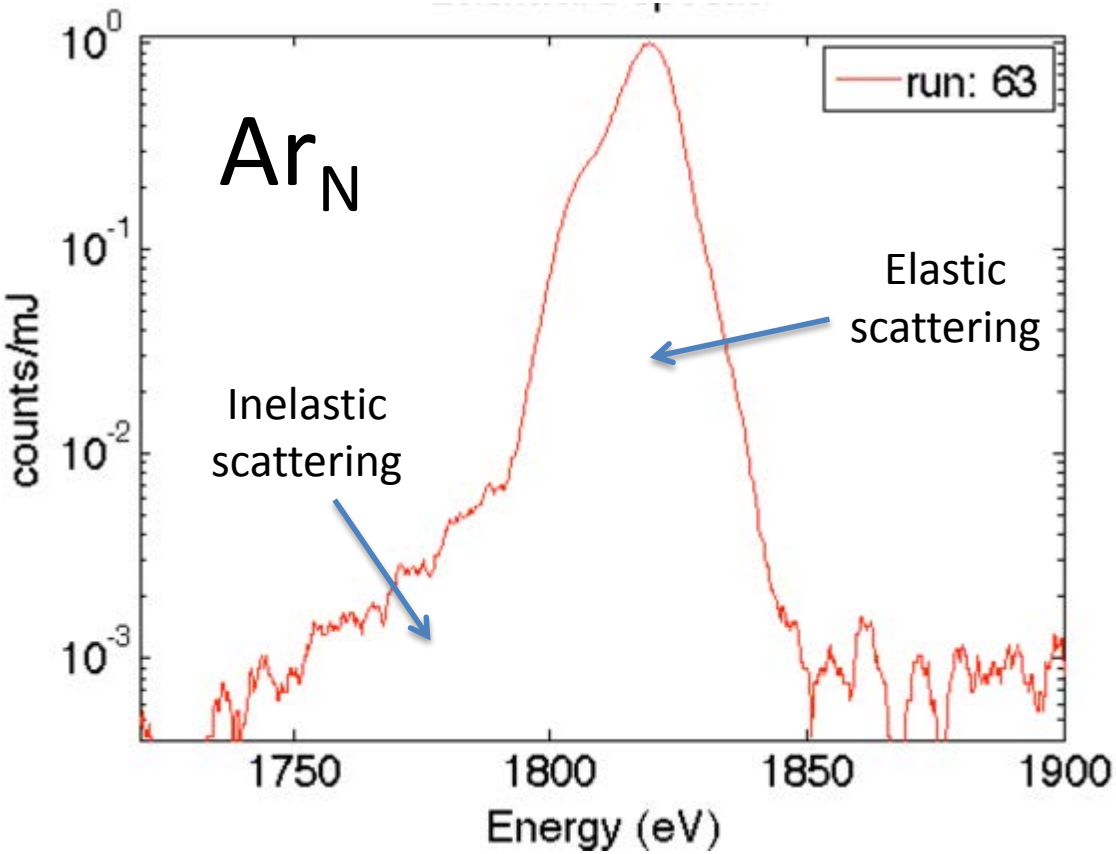
Side view





pnCCD detector:
Lueders et al.
Nucl. Instrum. & Methods A
Vol. **614**, 483-496 (2010)

During our recent AMO@LCLS beam time we have demonstrated the measurement of high dynamic-range x-ray scattering spectra



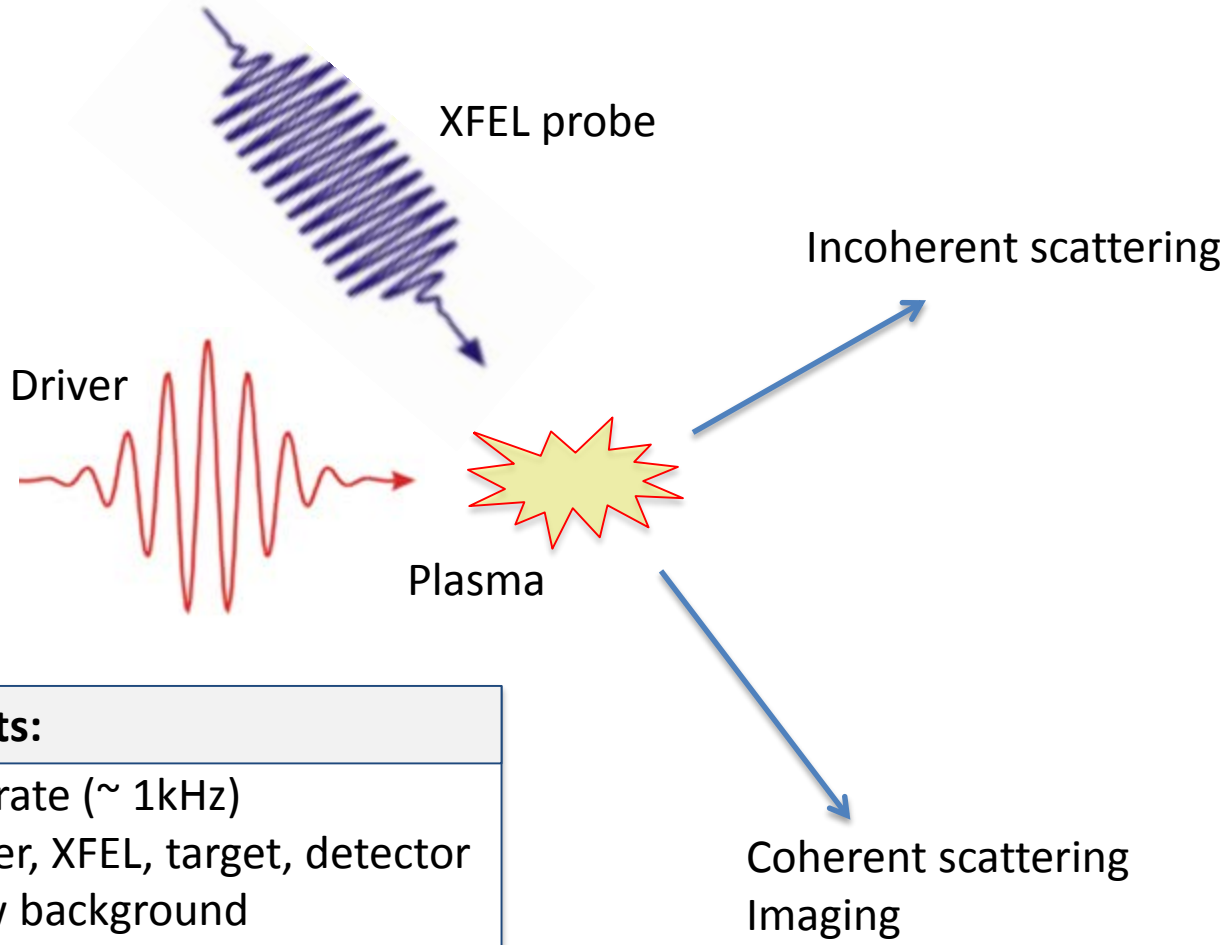
- Ar_N target (N ~ 10,000)
- FEL only
- FEL parameter: 1.8 keV, 60 fs, 2 mJ/ pulse
- Integral over 150,000 shots (ca. 20 min. at 120 Hz)
- detected ~ 1 photon/ shot

Scattering spectra from undriven Ar clusters shows clear inelastic shoulder – likely mix of bound-free and free-free contributions – analysis in progress

Summary

- We have successfully demonstrated proof-of-principle experiments to measure high-quality x-ray scattering spectra from driven nanoplasmas with ~ 20 fs time resolution at AMO/LCLS
- Challenges:
  - Operate XFEL with ~ 1 mJ at ~ 1 fs pulse length
    (scientific requirements for LCLS-II upgrade are currently being discussed)
  - Increase rep rate to 1-10 kHz (XFEL and IR laser)
  - Improve synchronization IR to XFEL
  - Develop mono-disperse particle source
  - Develop high-readout rate detectors, consistent with laser rep rate,
    with single photon counting capability
- Improved experiments will enable studying fundamental plasma processes and parameters
  - Resolve individual plasmon oscillations in the time-domain
  - Measure electron-ion collision rates
  - Measure equilibration dynamics

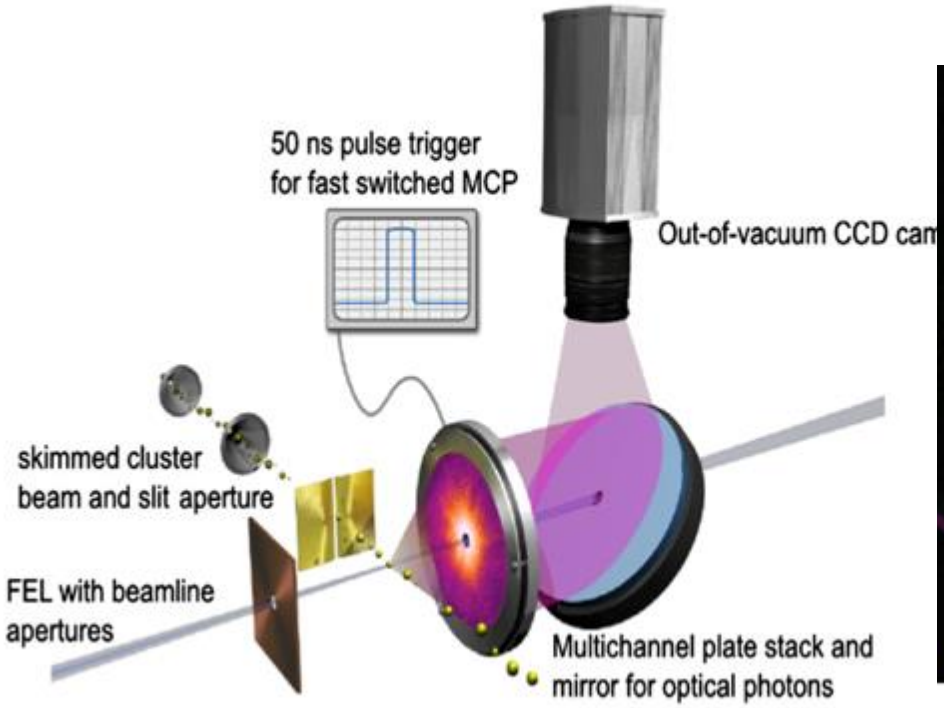
The concept of integrating over many laser shots to achieve high dynamic-range signals can be generalized for other applications

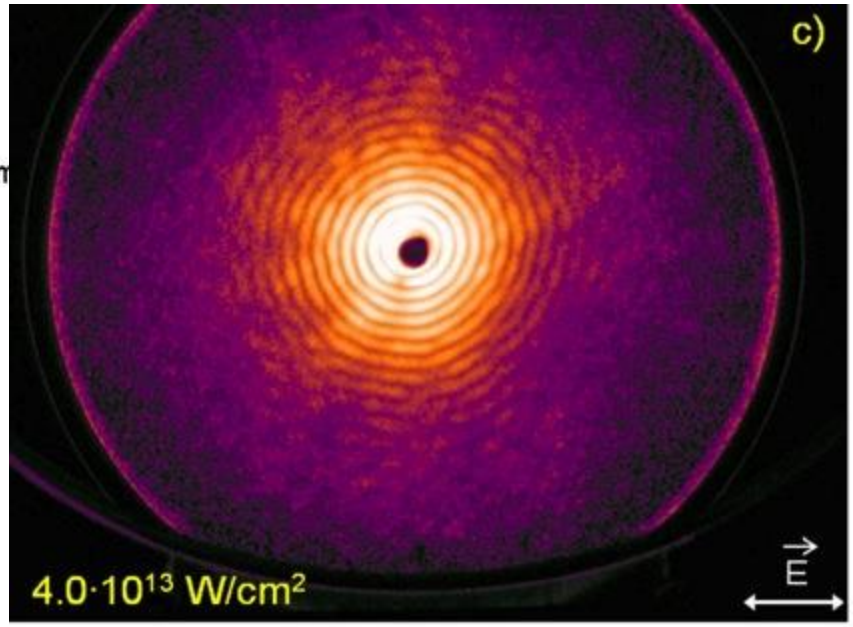


requirements:

- High rep rate (~ 1kHz)
  - Driver, XFEL, target, detector
- No or low background
- Accurate synchronization of driver with XFEL
- Shot-to-shot stability

Coherent x-ray scattering from individual particles has been developed as well to study transient nanoplasma properties





Bostedt et al., PRL 108, 093401 (2012)
Gorkhover et al., PRL 108, 245005 (2012)

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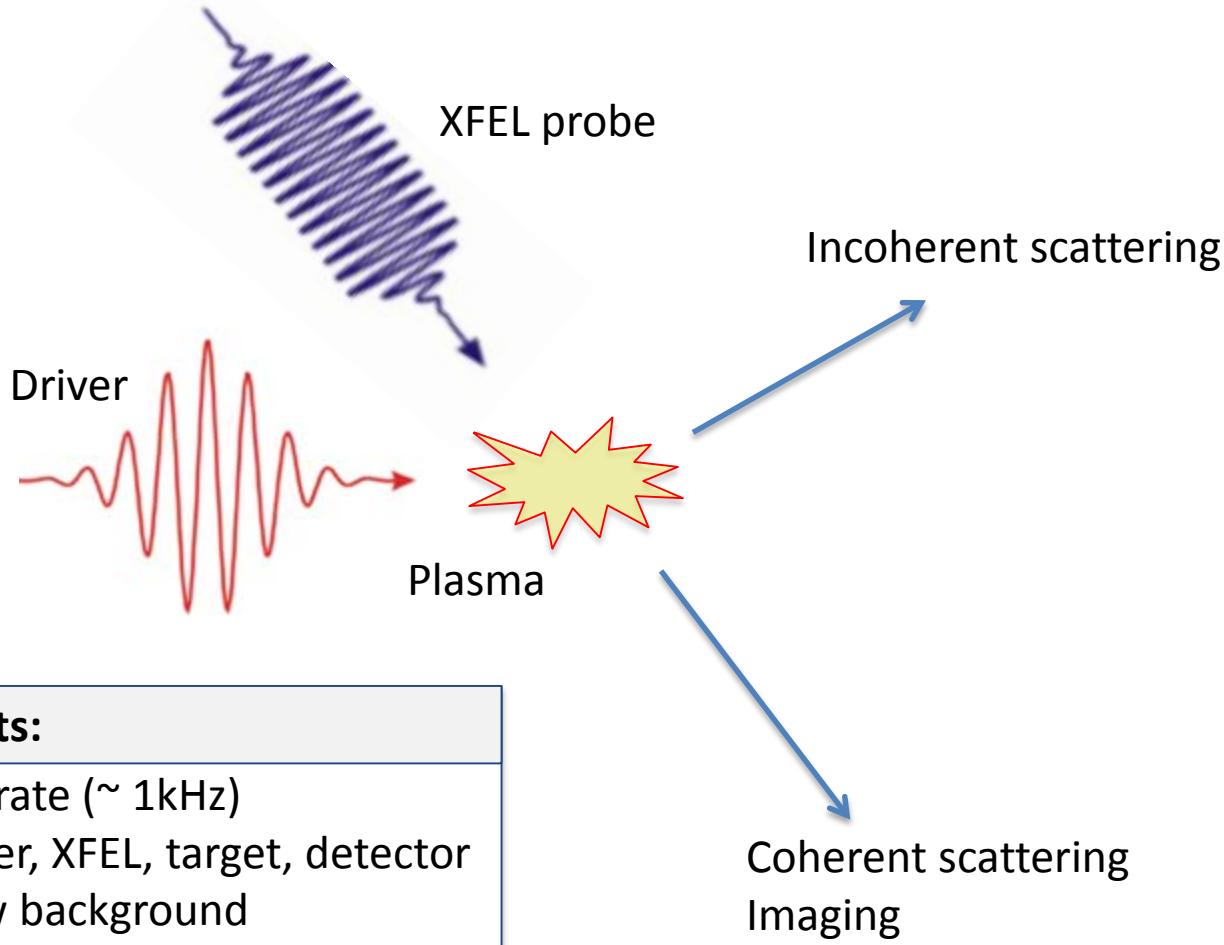

requirements:

- High rep rate (~ 1kHz)
  - Driver, XFEL, target, detector
- No or low background
- Accurate synchronization of driver with XFEL
- Shot-to-shot stability

Questions

1) What can we learn when resolving the plasmon oscillation in the time domain?
   - The time delay between the plasmon excitation and the XFEL probe is variable. Scanning this delay allows to observe the actual excitation process and the subsequent decay into single particle excitations.
2) Are there any quantum objects or processes that one could observe?
   - On the driver side, conceptually one can separate the excitation into tuning the system in resonance (1st IR pulse) and the plasmon excitation (2nd IR pulse). Then, the 2nd IR pulse can in principle be turned down so that the plasmon excitation remains in the linear regime.
3) Can one observe quadrupole oscillations
   - yes

Significance for fundamental science

Nanoplasmas at extreme conditions (T ~ 100s eV, near solid density) can be produced with very repeatable with table-top laser systems. Since they are free from interaction with their surrounding they are ideal model systems for light – matter interactions. Fundamental processes like ultrafast collective electron dynamics can be studied with x-ray Thomson Scattering in high-repetition rate experiments, that ultimately can approach a time resolution of ~ 1 femto-second, ie fundamental time scales like electron- electron collision times. In particular, this time scale is shorter than a single cycle of the infra-red laser driver (2.7 fs for a 800 nm laser pulse).

Such measurements with unprecedented time-resolution allow studying fundamental aspects of light-matter interaction, for example the role of collective excitations, and fundamental equilibration processes over a wide range of plasma conditions. The results can be used to benchmark high-energy density physics models in the challenging regime of non-LTE plasmas.

Facilities

This science case is taking advantage of new exciting capabilities provided by Free Electron Lasers like LCLS at SLAC in Stanford, CA. Such facilities enable this new class of x-ray scattering experiments that benefit (1) from high-brilliance, and highly collimated x-ray beams, (2) ultra-short pulse lengths, and (3) high repetition rate operation, which allows to accumulate data until the required data quality is obtained.