New Horizon of Accelerator based Molecular Physics

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DESY

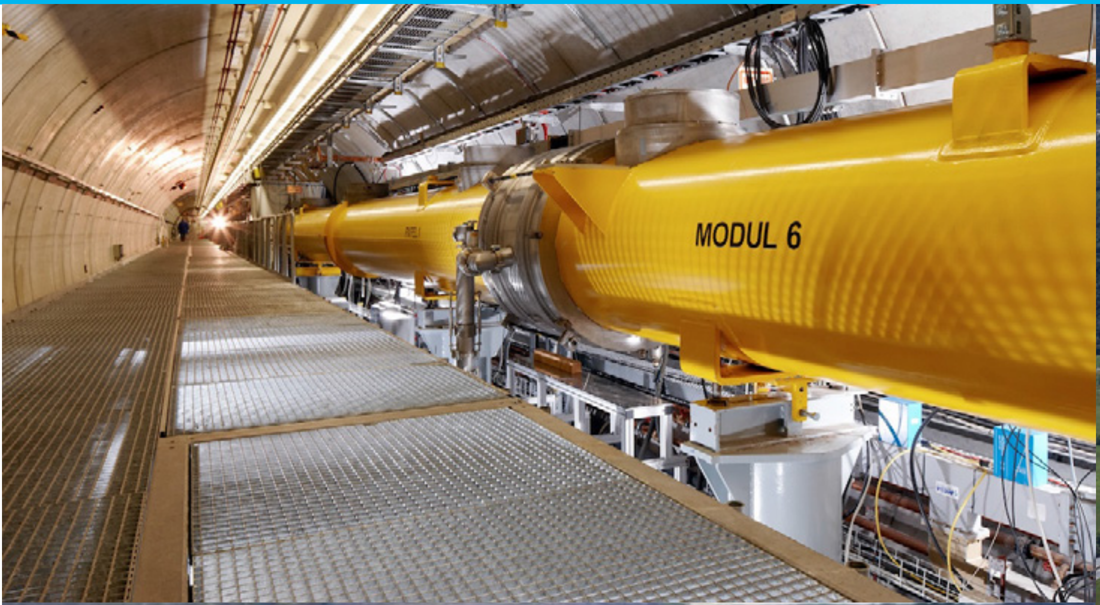
Aug. 19, 2019, Taoyuan

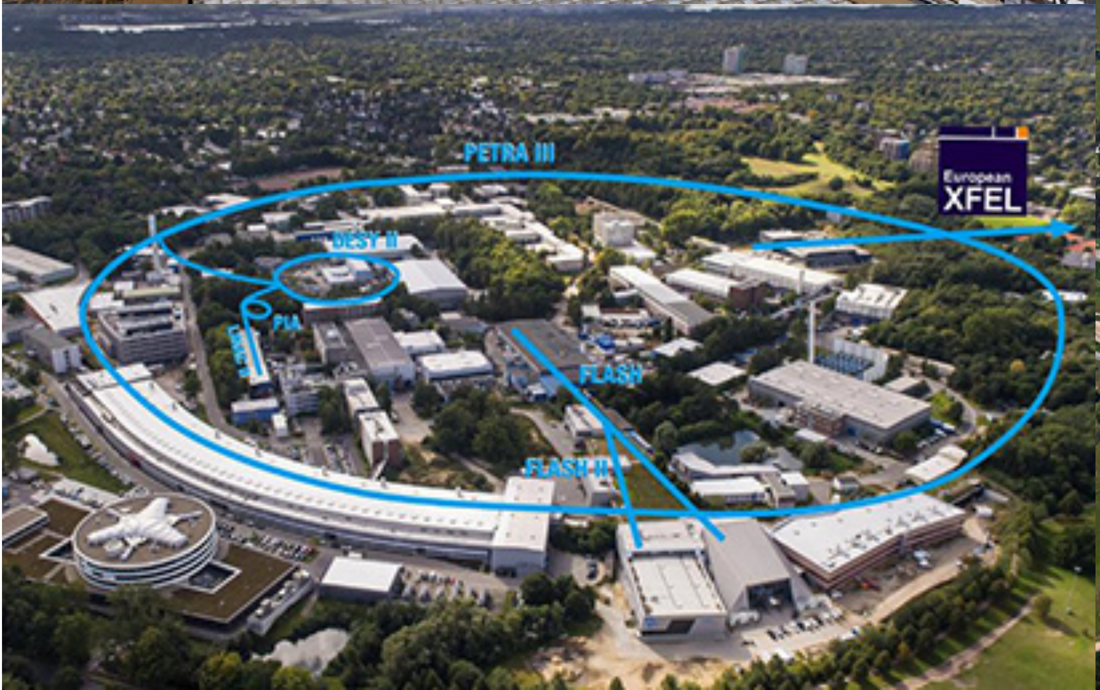
Zheng

Research Field

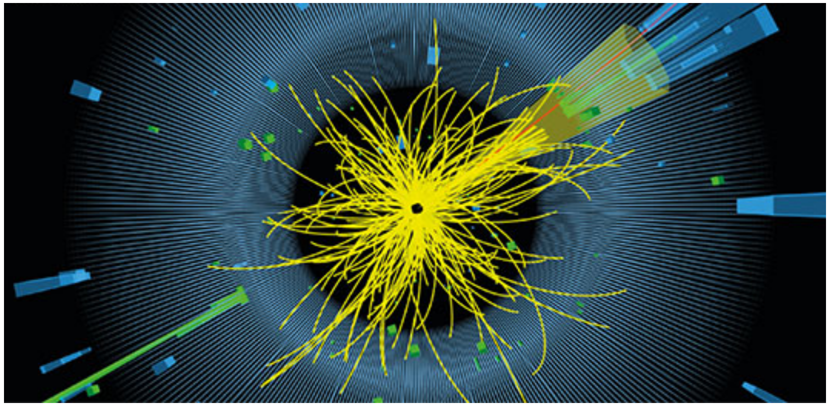
Atomic, Molecular and Optical Physics

Accelerators @ DESY and SLAC

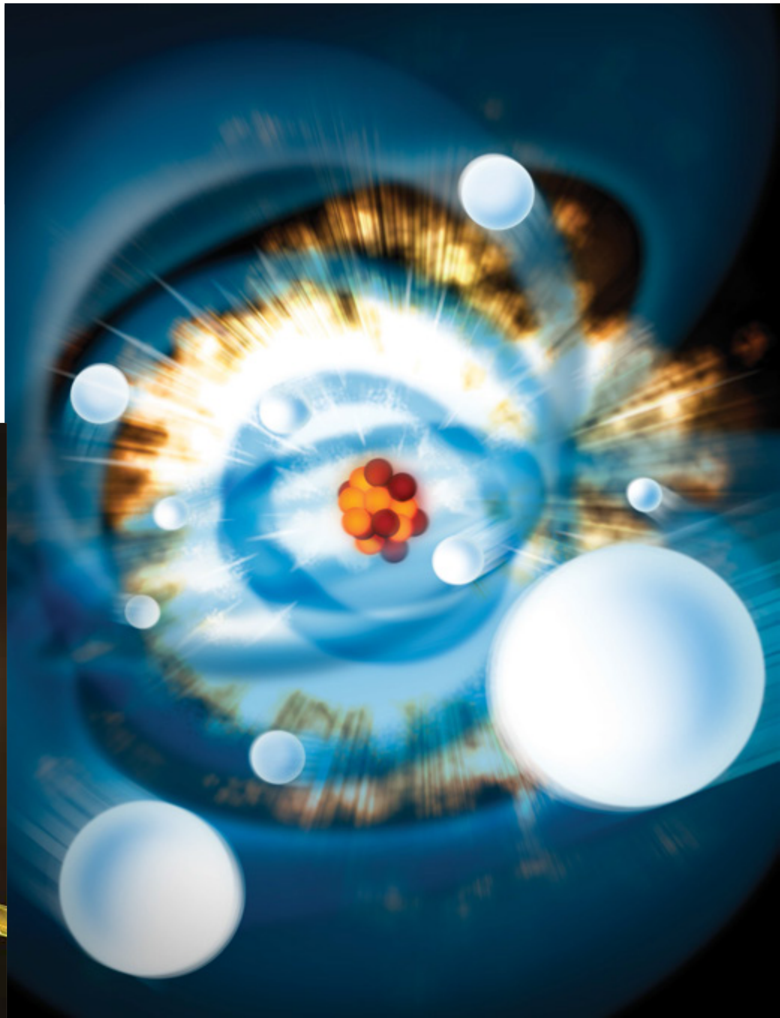




Accelerator physics: from particles to molecules

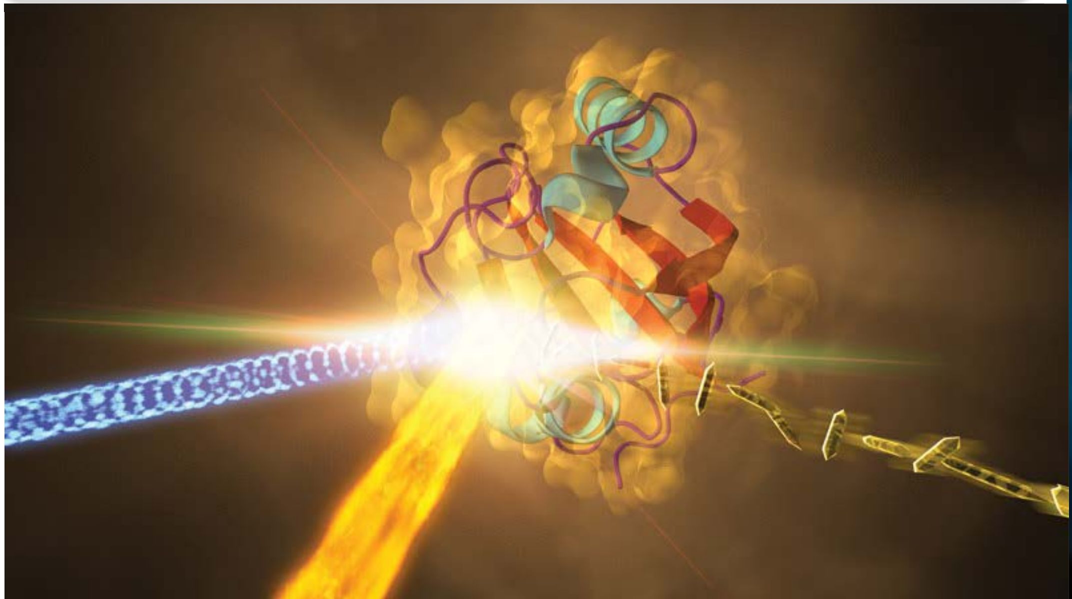


Elementary particles

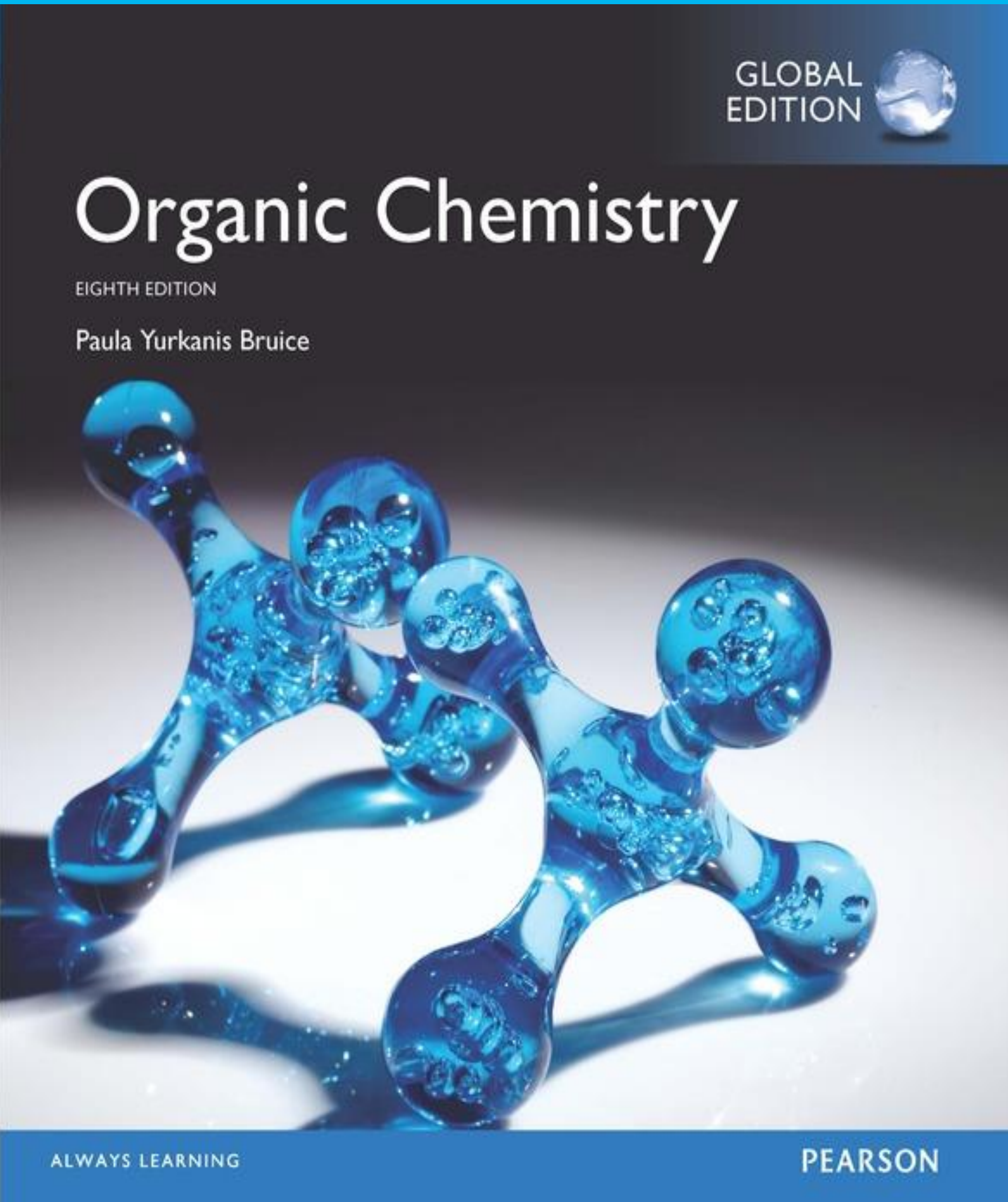


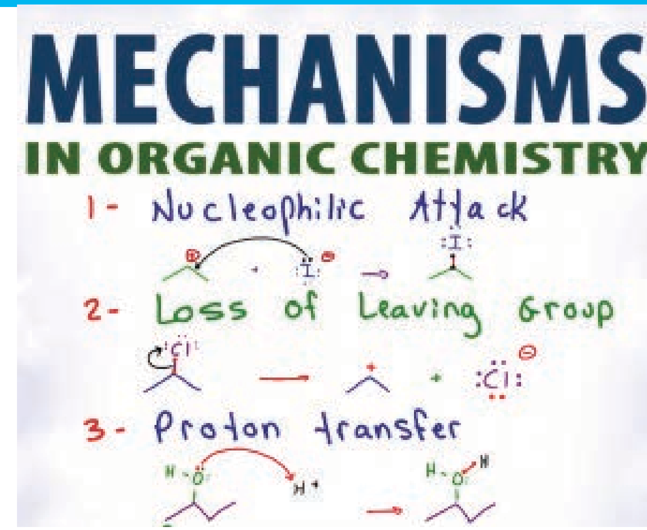
Molecules

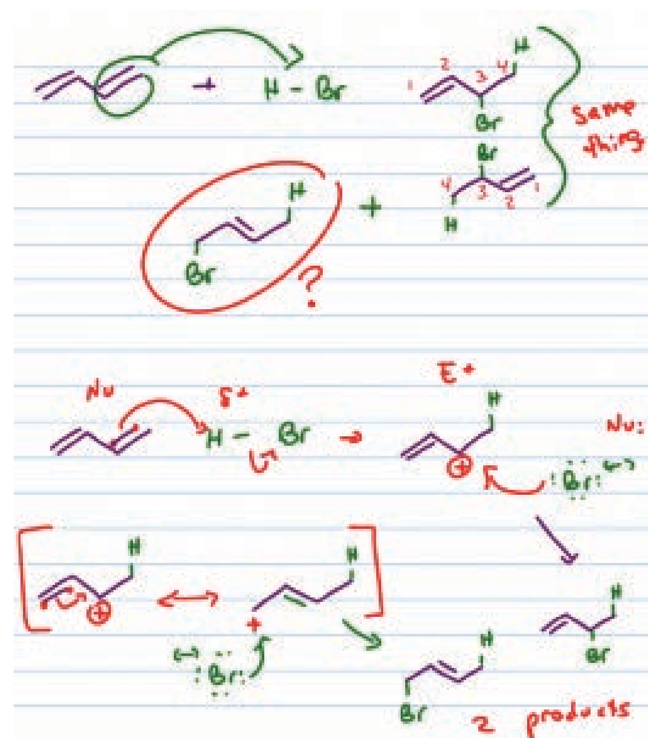
What can accelerator do for molecular physics?



Prelude:- Are these arrows real? just imagination?





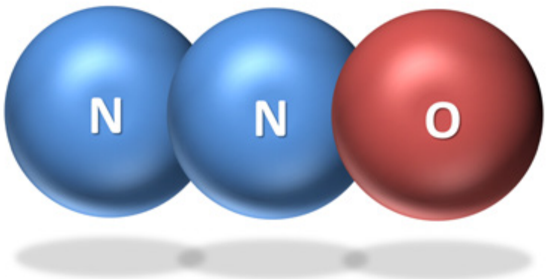


A centenary dream comes true

Arrhenius law

$$k = Ae^{-\frac{E_a}{k_B T}}$$

$$N_2O \rightarrow N_2 + O$$



$$A = 6.1 \times 10^{14}\ \text{s}^{-1}$$

$$\tau \sim A^{-1} = 1.6 \times 10^{-15}\ \text{s}$$

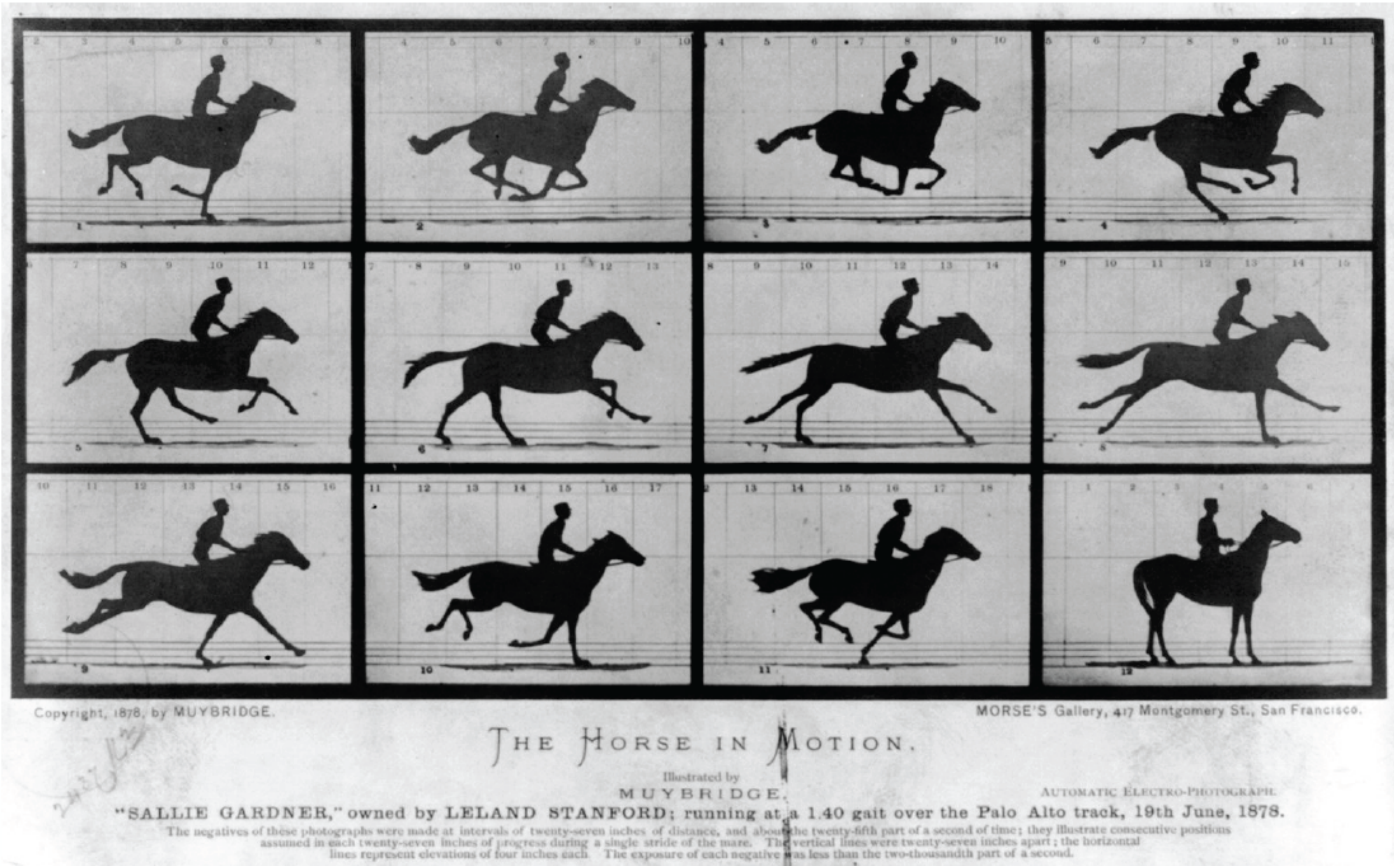
1.6 femtosecond

S. A. Arrehnius, Z. Phys. Chem. 4, 96 (1889)

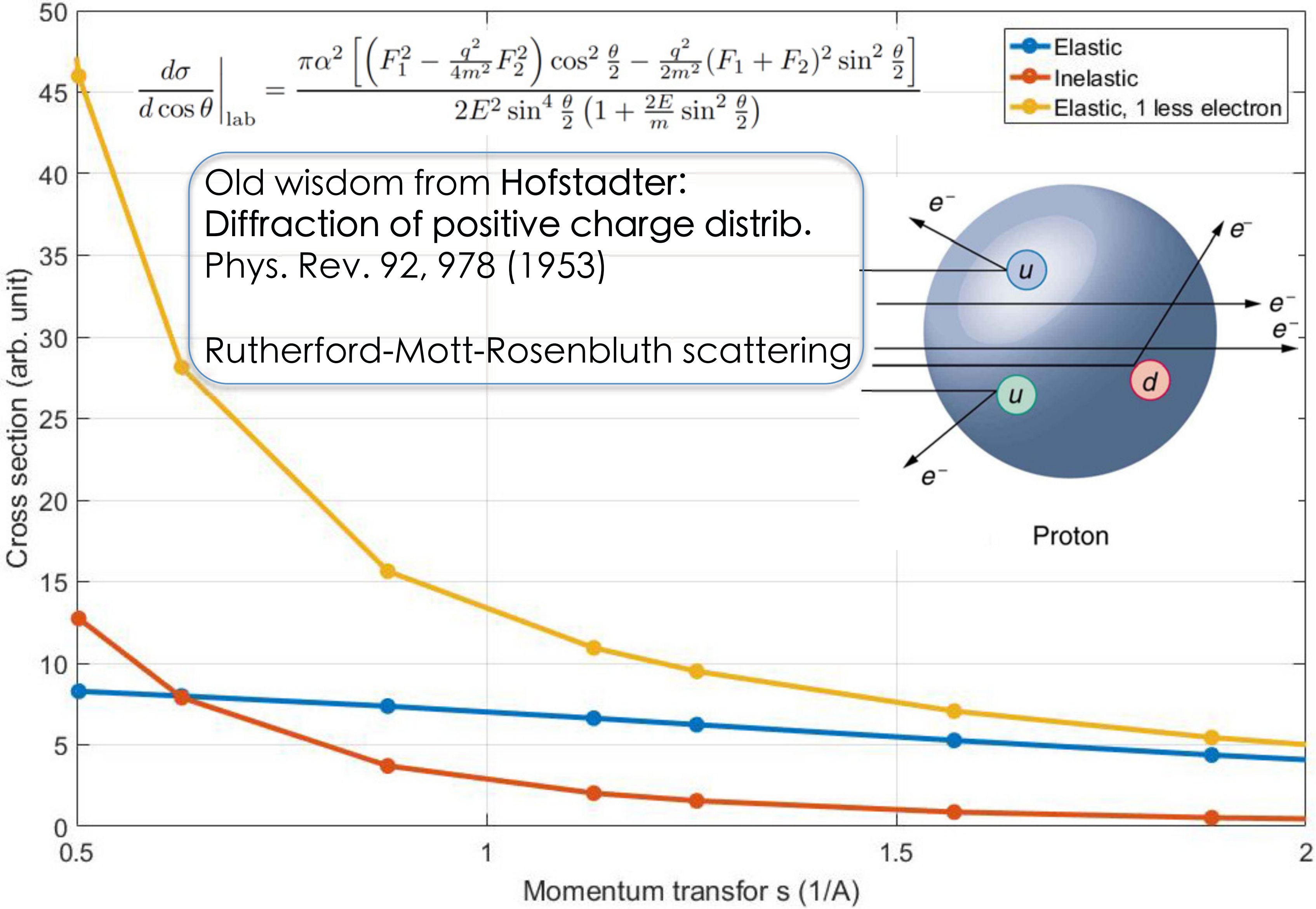
S. R. Logan, J. Chem. Educ. 59, 279 (1982)

K. J. Laidler, J. Chem. Educ. 61, 494 (1984)

Time resolved measurements of dynamics

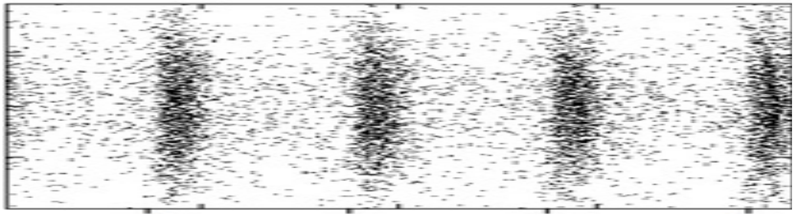


Diffraction:- from proton to molecule



J. Yang, X. Zhu, T. Wolf, ZL, M. Centurion et al. Science, (2018)

Ultrafast Molecular Diffraction

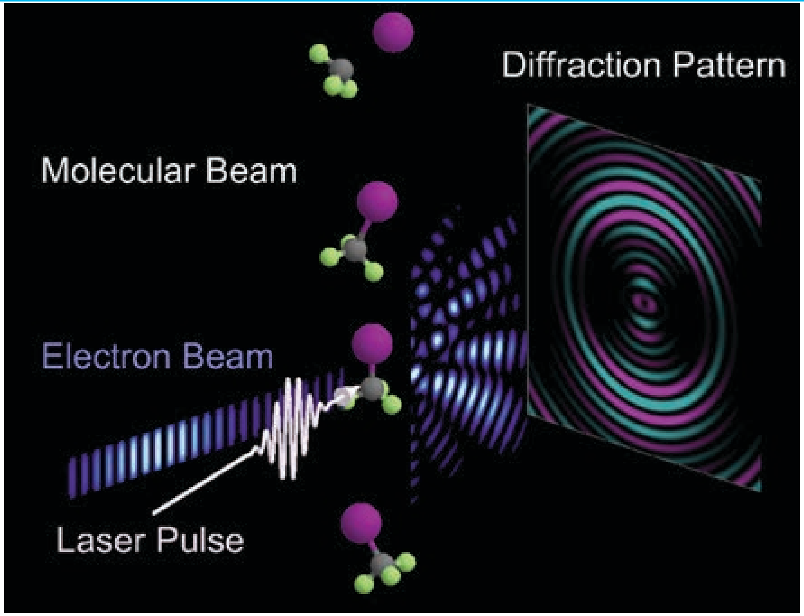


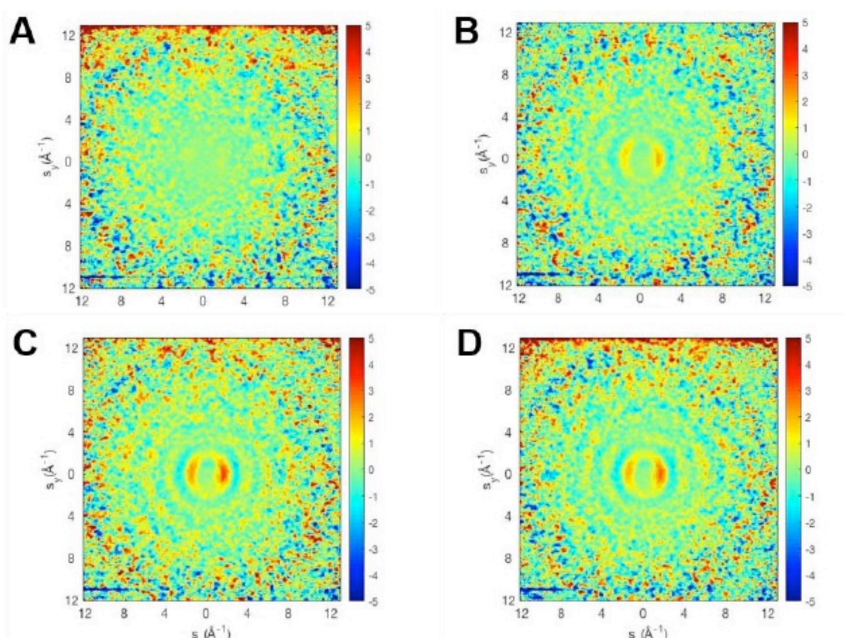
Ultrafast Diffraction

The motion of molecules is on femtosecond ($1fs=10^{-15}s$) time scale.

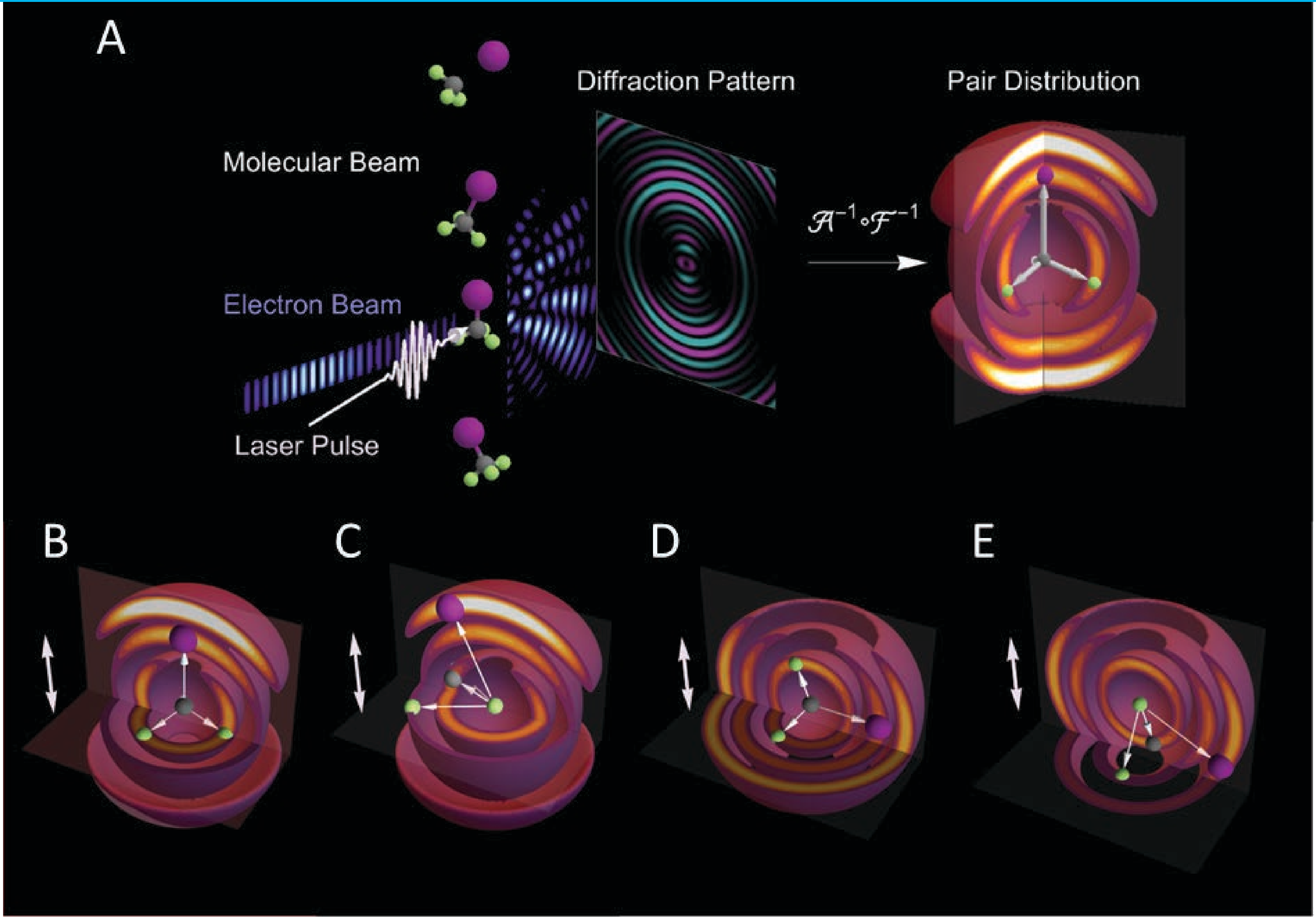
The compressed electron bunches can have <50fs pulse width.

We can see the motion pictures of single molecule



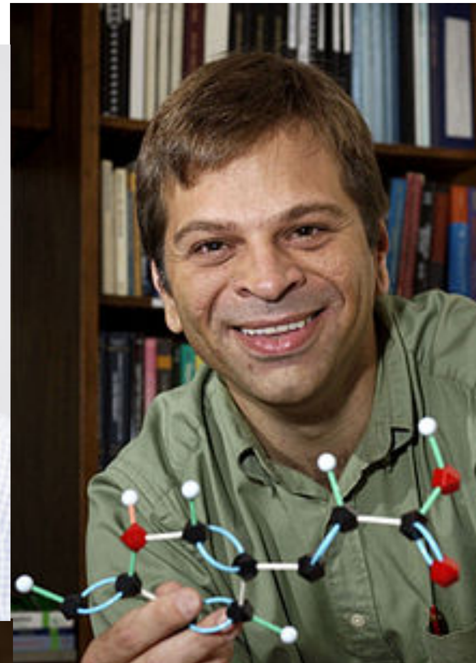


CF_3I Photodissociation (3.7MeV electrons)

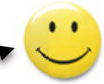


J. Yang, X. Zhu, T. Wolf, ZL et al. Science, 361, 64 (2018)

Dwayne Miller



Zheng

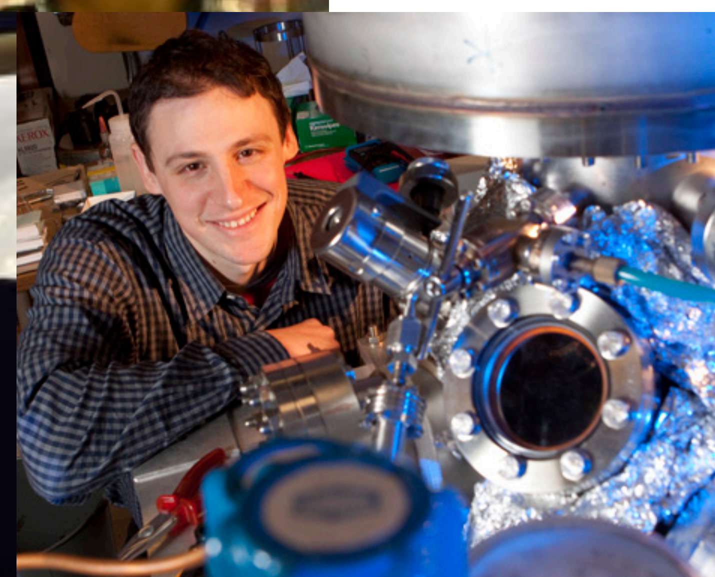


Todd Martinuez

Jie Yang

Jim Cyran

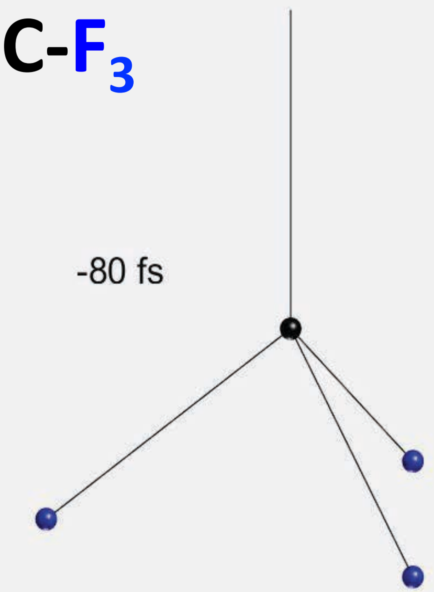
Xiaolei Zhu

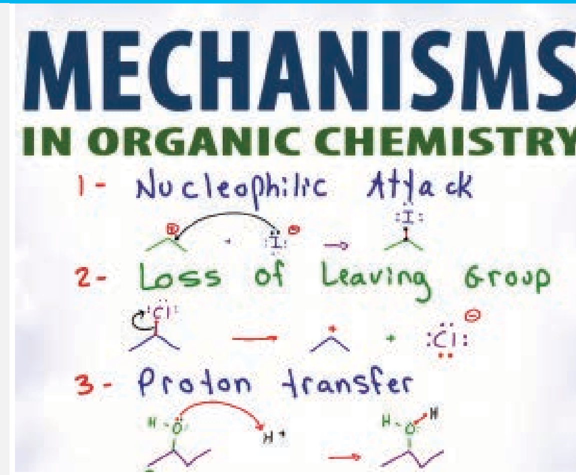


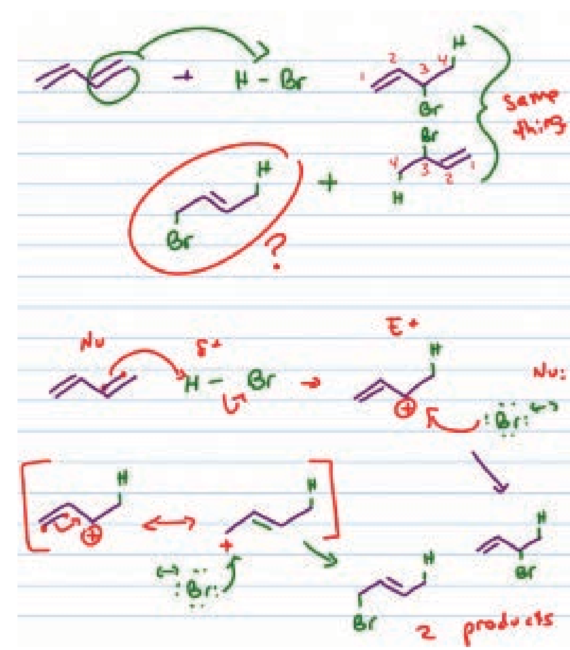
Martin Centurion

The Molecular Movie: Seeing is believing

Recoil of C-F_3

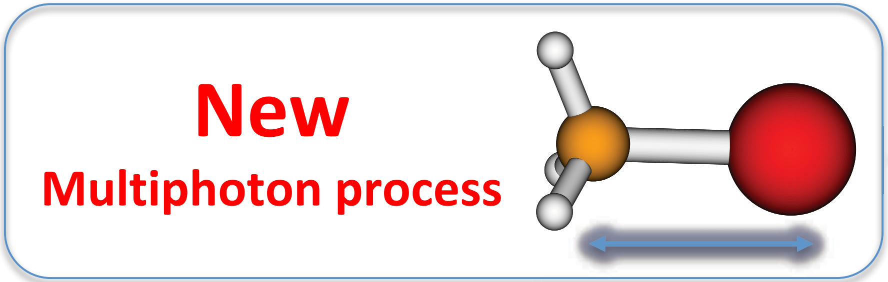


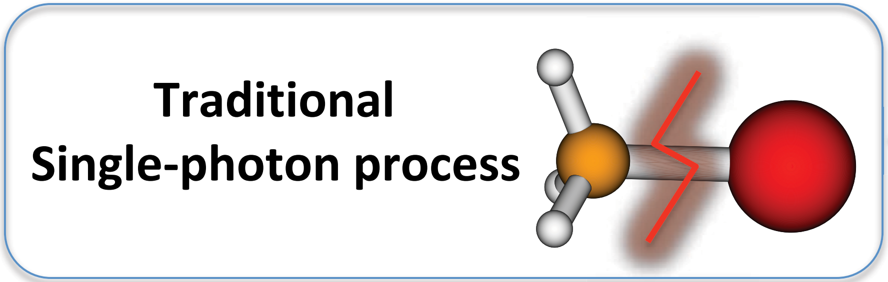


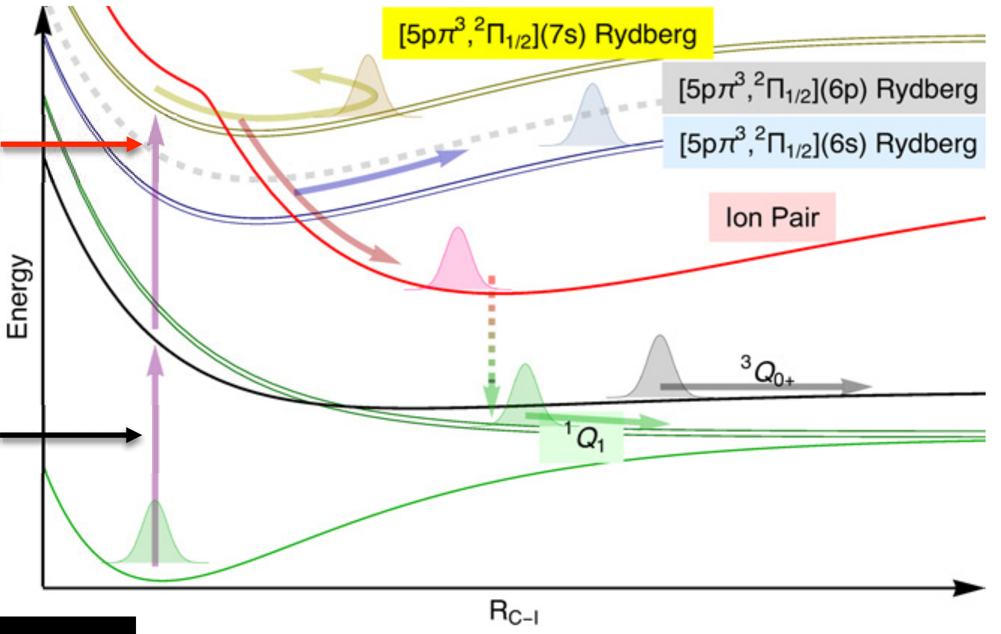


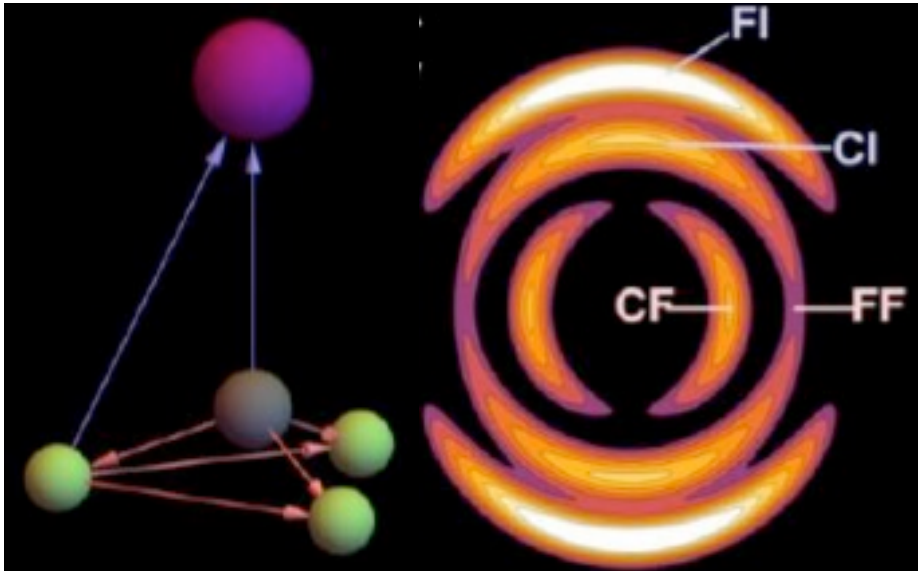
Seeing the once imaginary molecular motion in organic chemistry textbook.

Mysterious Oscillation – New Channel



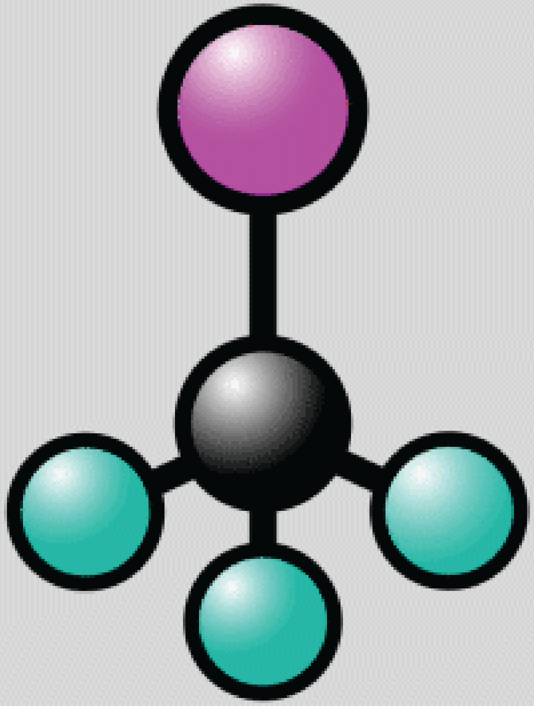






J. Yang, X. Zhu, T. Wolf, <u>ZL</u> et al. Science, 361, 64 (2018)

New Channel



J. Yang, X. Zhu, T. Wolf, ZL et al. Science, 361, 64 (2018)

Quantum tomography by TRED diffraction

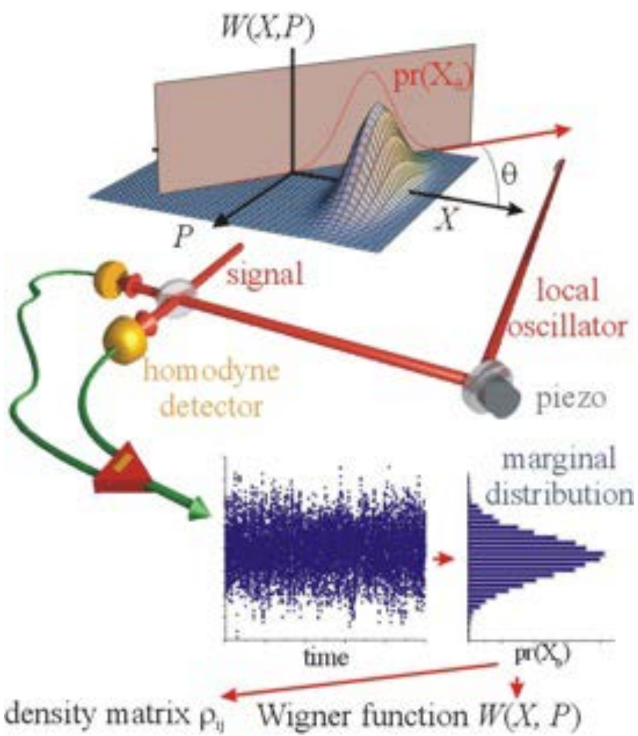
The time in diffraction pattern I(Q;t) does not only offer us the molecular motion in time, but also unveils the complete quantumness of the molecule.

Pauli Problem (Pauli, 1933): could we retrieve Ψ(x) from observable $P(x)=|\Psi(x)|^2$?

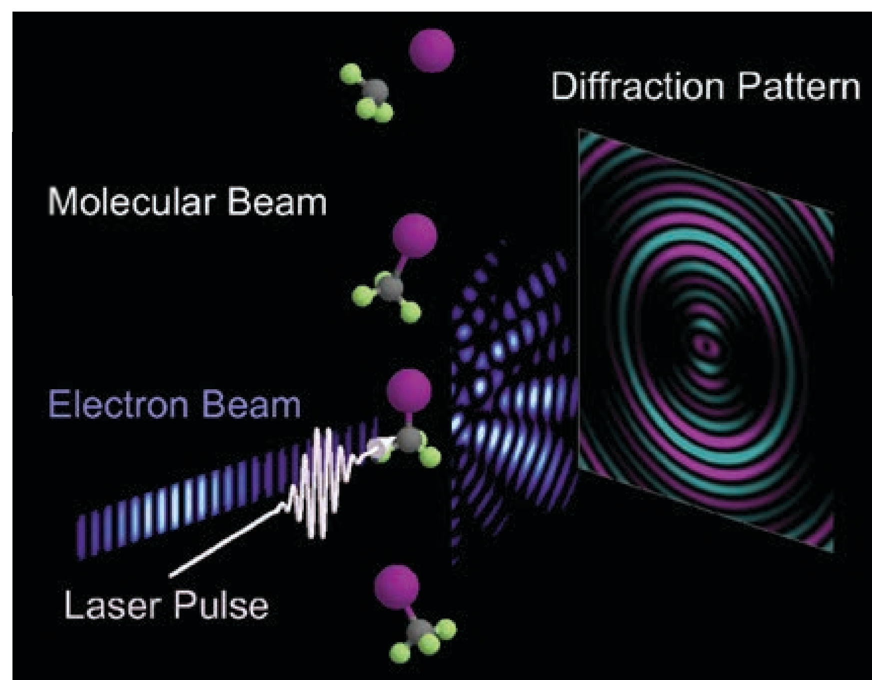
The Wigner function W(x,p) and density matrix ρ_{mn} can be retrieved from I(Q;t) !

Quantum optics:- Homodyne detection
RMP, 81, 299 (2009)

TRED :- Diffraction as unitary evolution t≅θ
$|\Psi(x;\Theta)|^2 \cong |\Psi(x;\omega t)|^2$

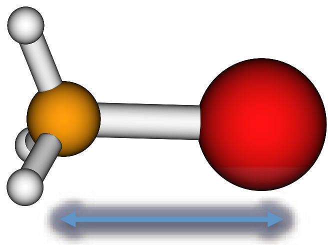


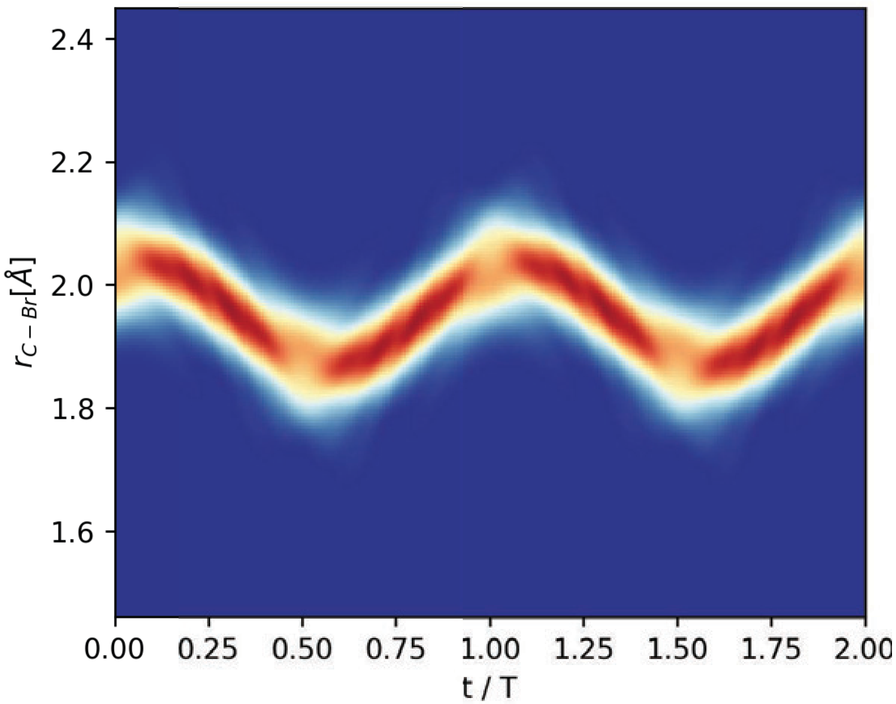
θ≅t

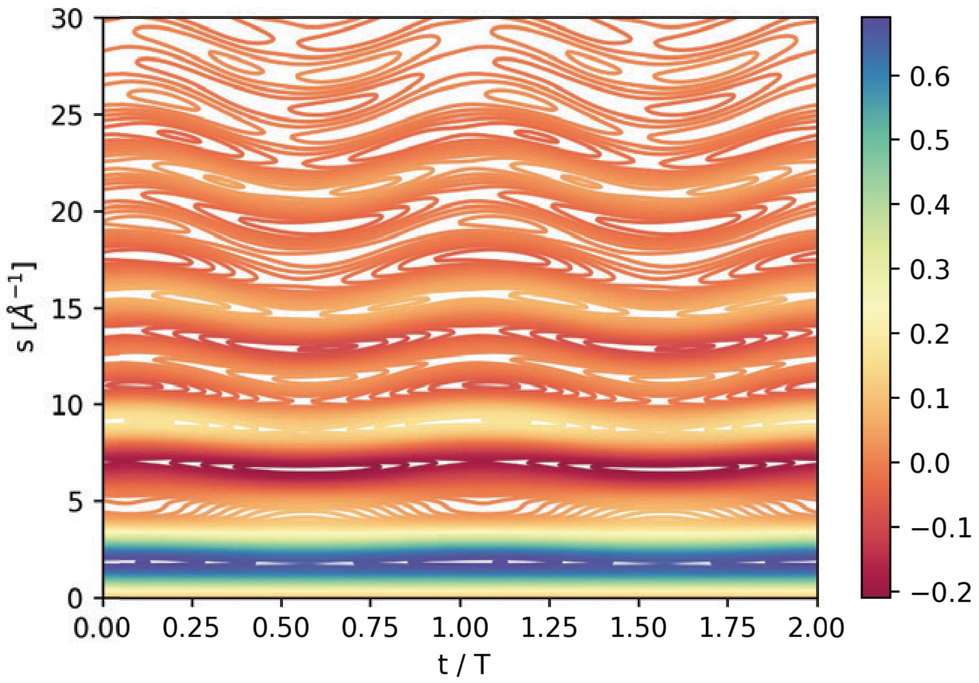


Quantum tomography by TRED diffraction

Diffraction from time dependent vibration wave packet of CH_3Br molecule



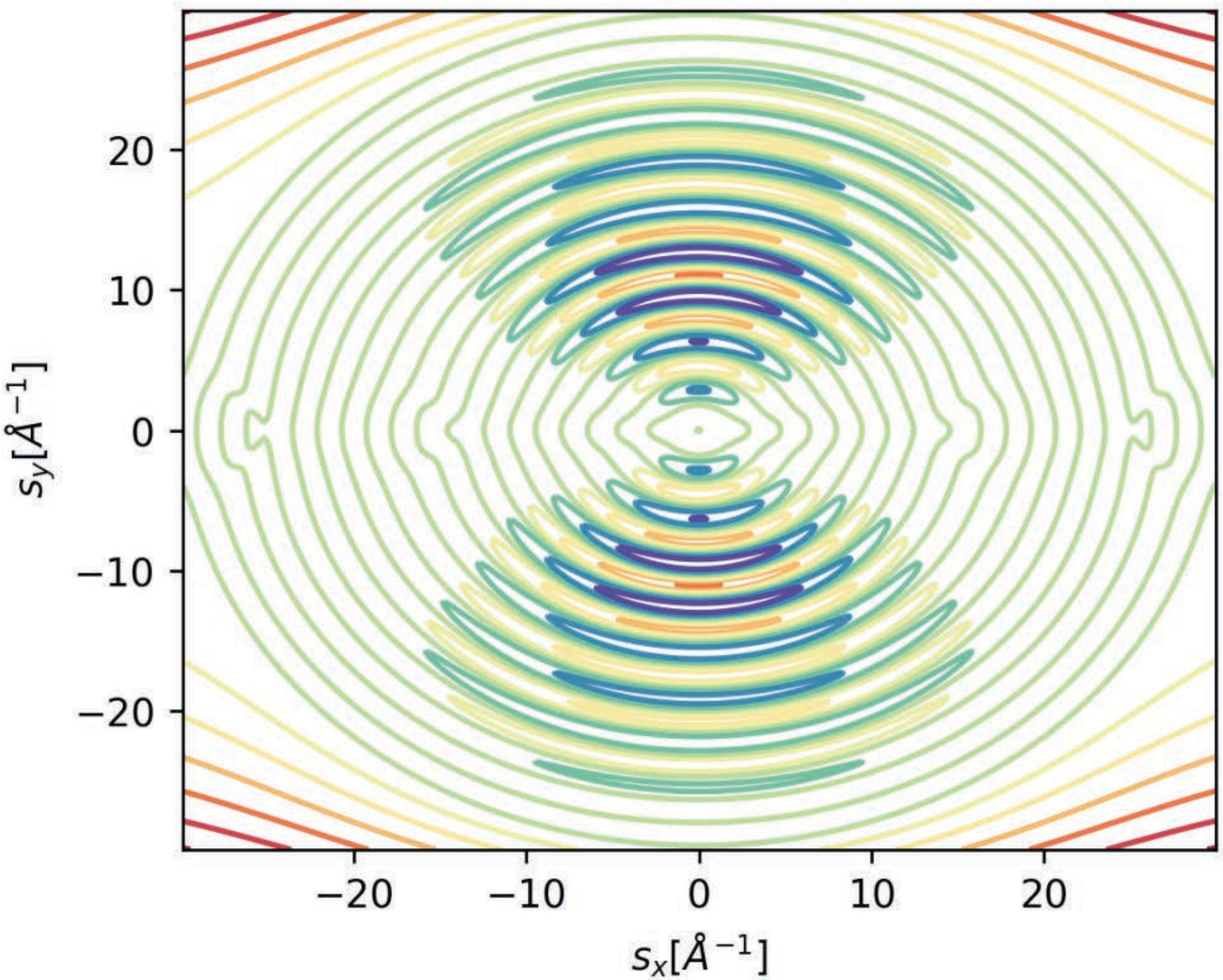


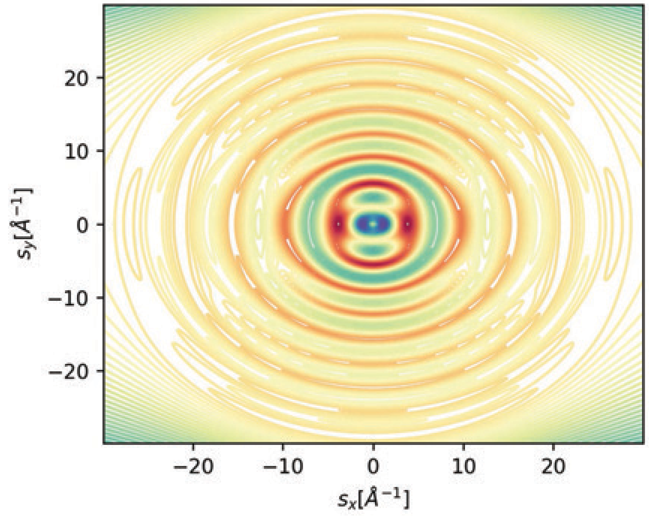


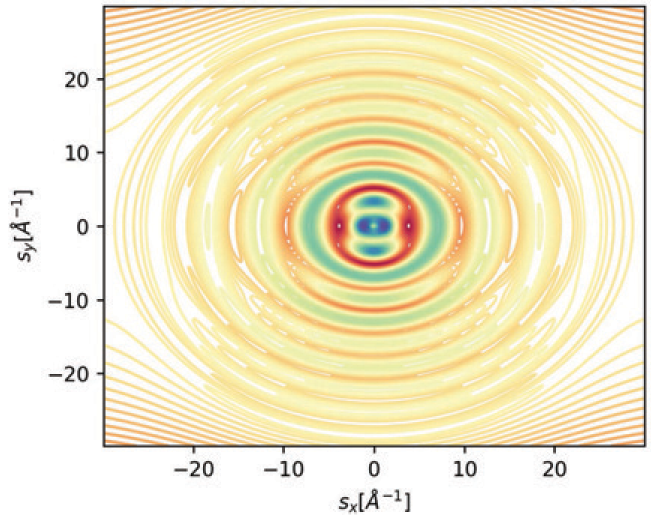
H. Timmers*, X. Zhu*, <u>ZL*</u>, D. Neumark, S. Leone et al. Nature Commun. 10, 3133 ('19)

<u>ZL*</u>, S. Gyawali*, A. Ischenko, S. Hayes, R. J. D. Miller, submit to ACS Photonics ('19)

Difference of diffraction intensity t=0 & t=T/2



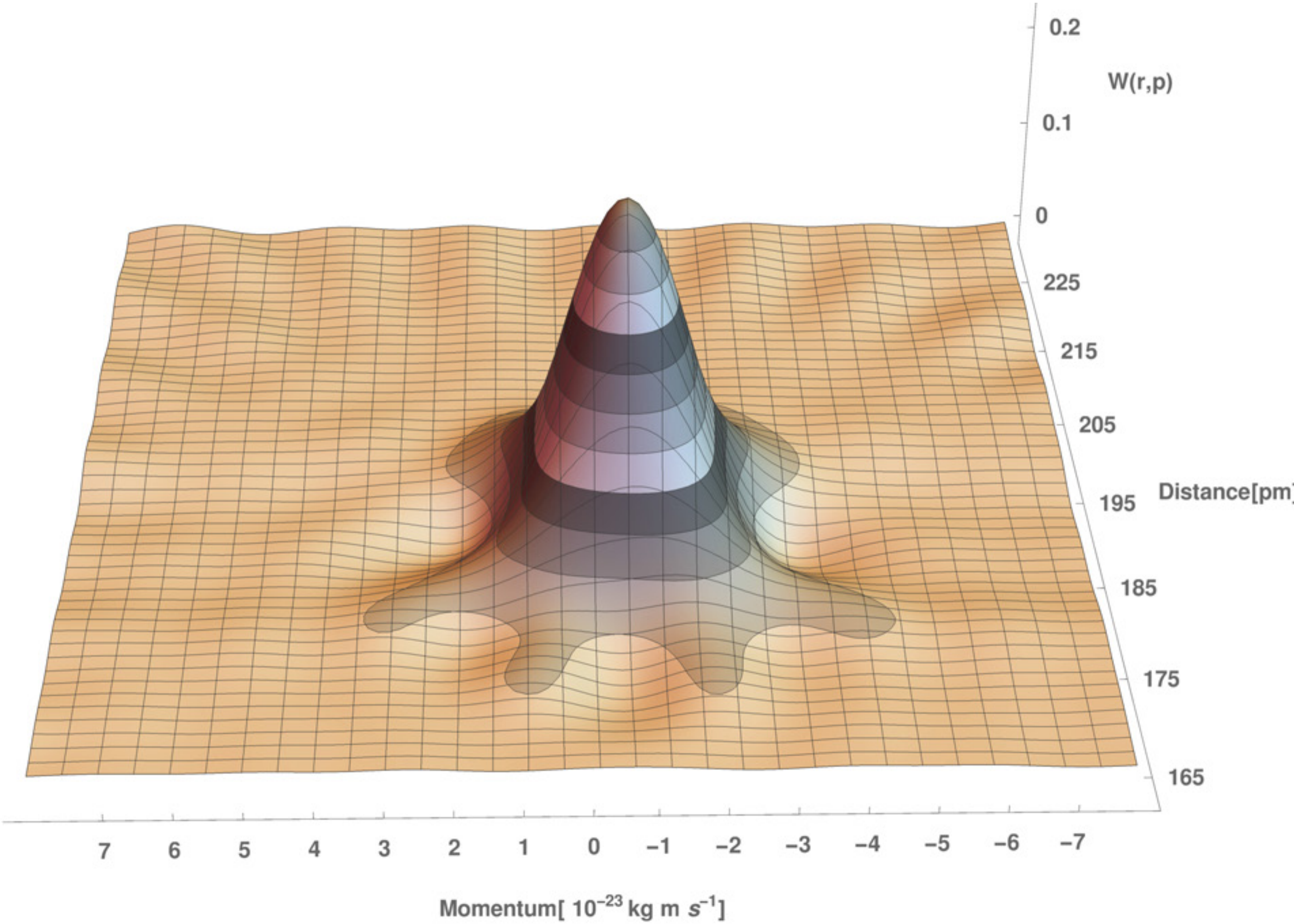




<u>ZL</u>, S. Gyawali, A. Ischenko, S. Hayes, R. J. D. Miller, submit to ACS Photonics (2019)

Wginer function

$$W(r,p) = \frac{1}{2\pi^2}\int_{-\infty}^{\infty} dx \int_0^{\pi} d\theta \, \Pr(x,\theta) K(r\cos\theta + p\sin\theta - x)$$

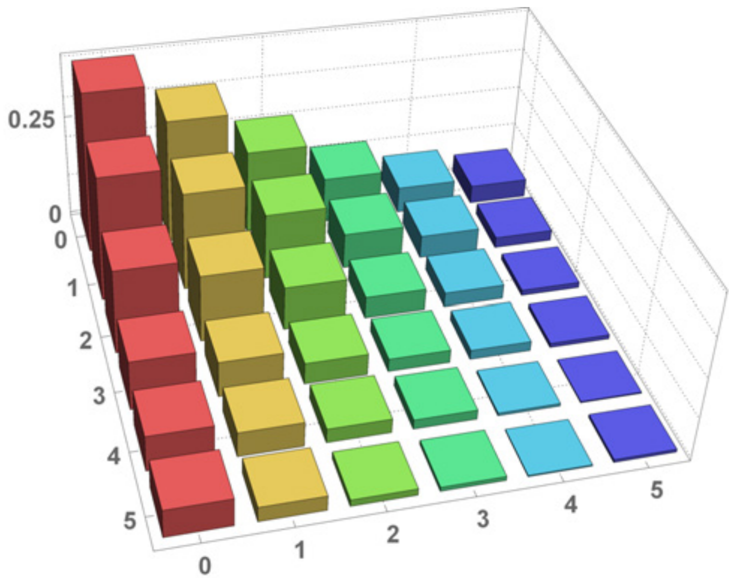


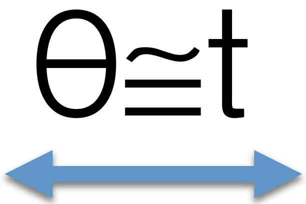
<u>ZL</u>, S. Gyawali, A. Ischenko, S. Hayes, R. J. D. Miller, submit to ACS Photonics (2019)

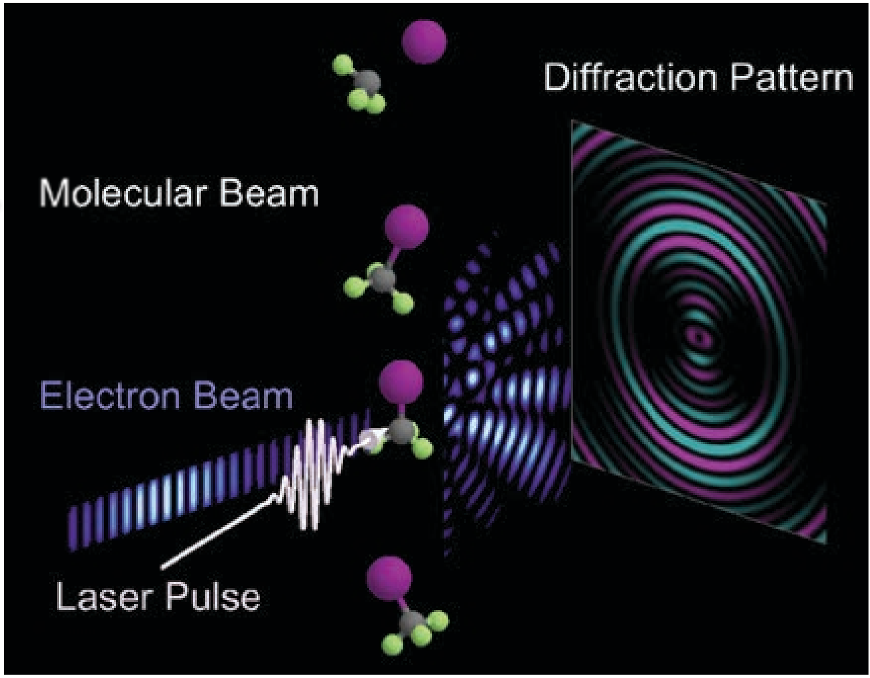
Density matrix of vibrational states

Density Matrix of coherent wavepacket
vibrational motion

TRED :- Diffraction as unitary evolution t≅θ
$|\Psi(x;\Theta)|^2 \cong |\Psi(x;\omega t)|^2$

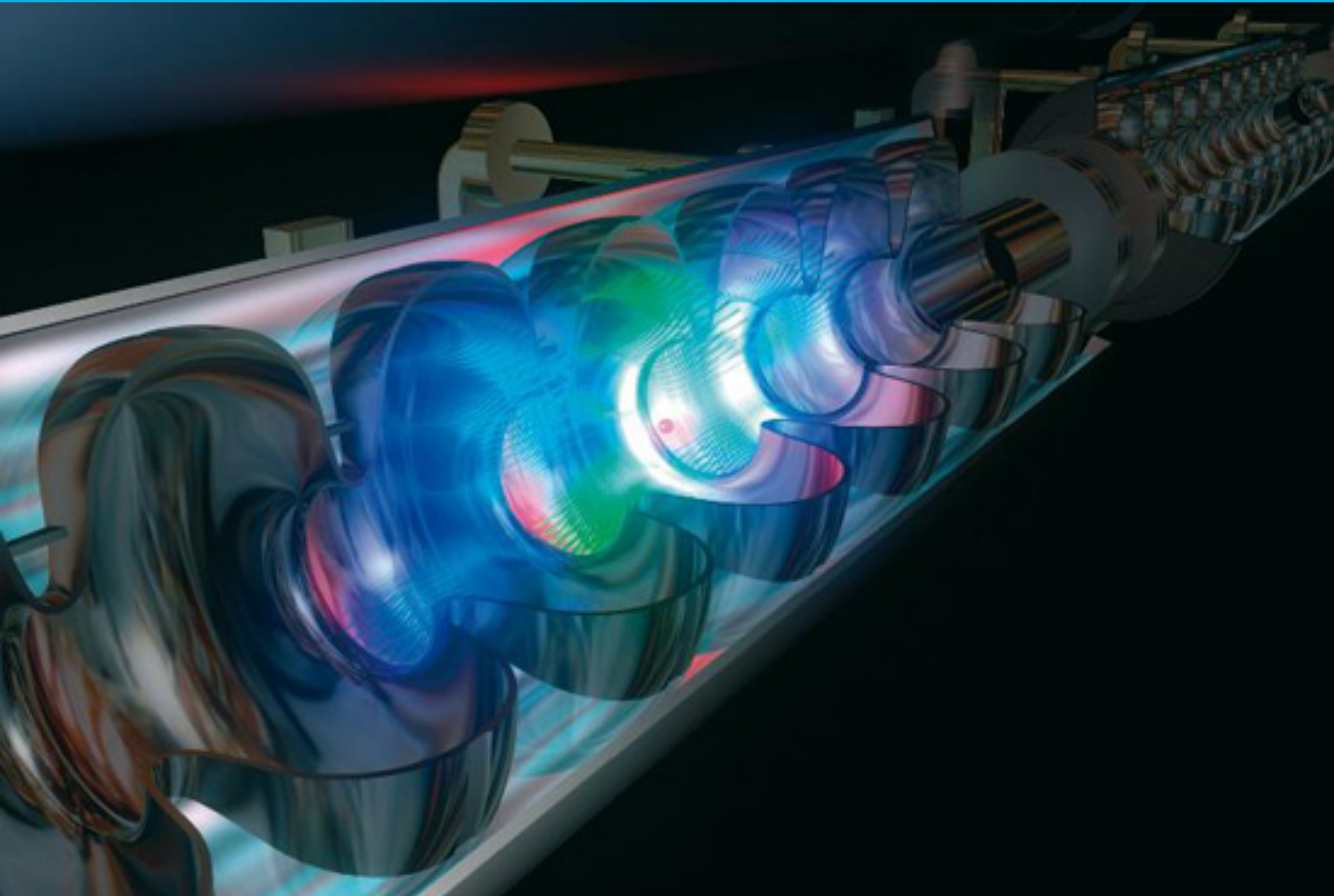






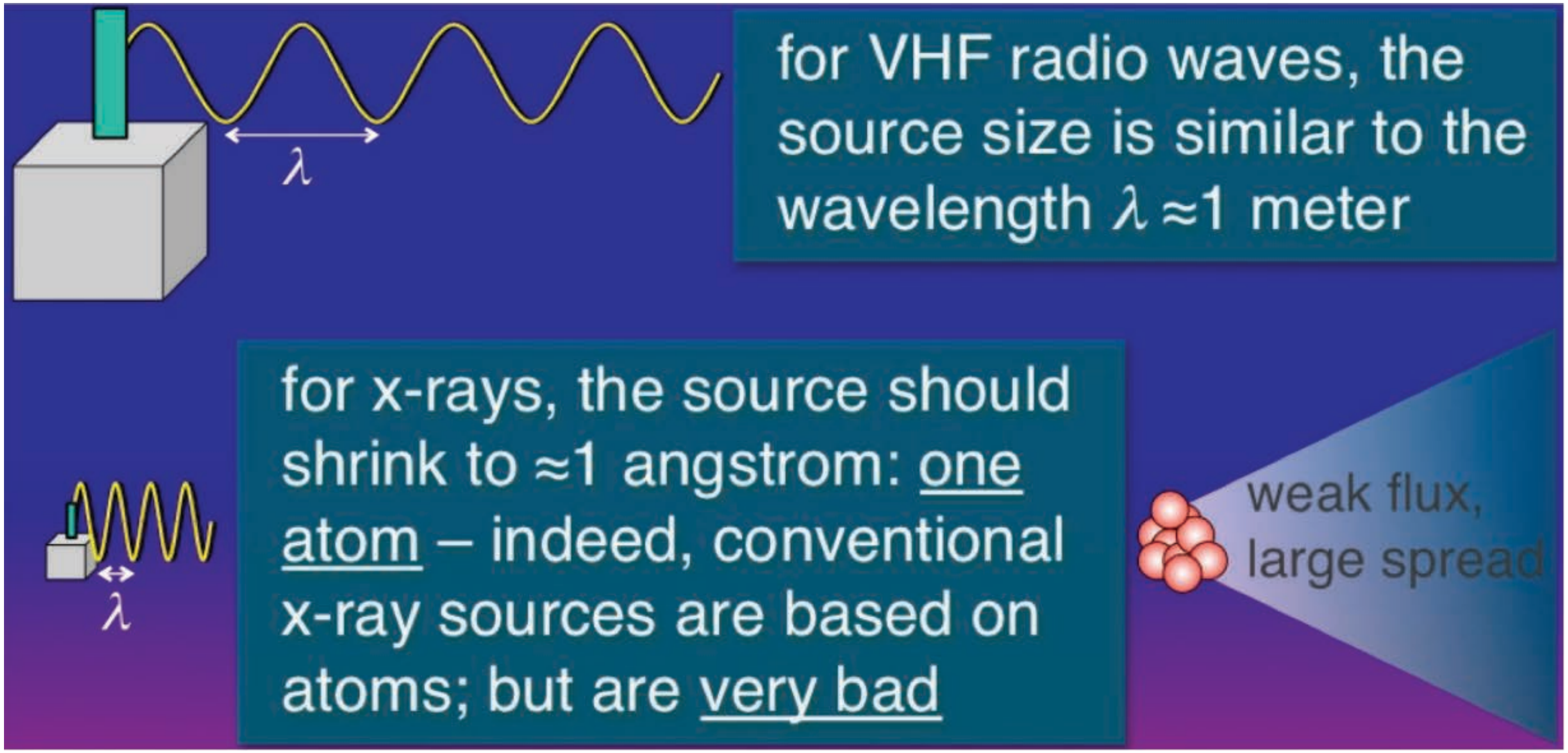
ZL, S. Gyawali, A. Ischenko, S. Hayes, R. J. D. Miller, submit to ACS Photonics (2019)

XFEL:- X-ray Free Electron Lasers (GeV electrons)



Paradox for a good X-ray source

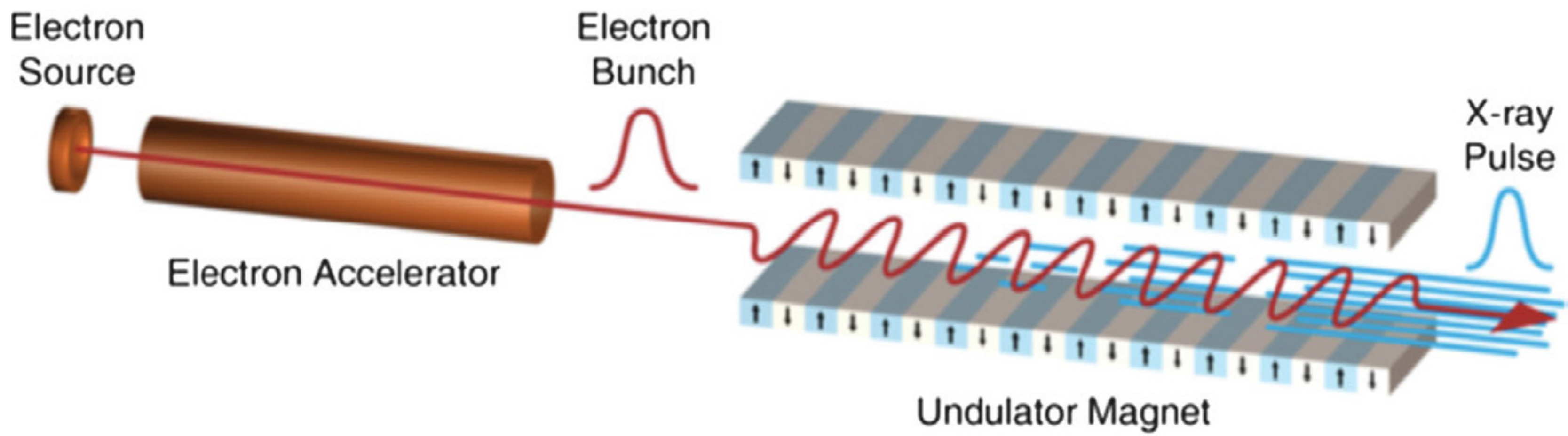
Femtosecond | **X-ray** | **Coherent Laser**



To improve them, we have to have artificial source with size ~1 angstrom: no way!

How does XFEL work?

Femtosecond | **X-ray** | **Coherent Laser**



Must the theory be so formal and complicated?

RMP, 88, '16

$$\vec{r}(t) = \left(\rho \sin \frac{\beta c}{\rho} t, \rho \left(1 - \cos \frac{\beta c}{\rho} t\right), 0\right).$$

In the limit of small angles we compute

$$\hat{n} \times \left(\hat{n} \times \vec{\beta}\right) = \beta \left[-\vec{\varepsilon}_{\parallel} \sin\left(\frac{\beta c t}{\rho}\right) + \vec{\varepsilon}_{\perp} \cos\left(\frac{\beta c t}{\rho}\right) \sin\theta\right]$$

$$\omega\left(t - \frac{\hat{n} \cdot \vec{r}(t)}{c}\right) = \omega\left[t - \frac{\rho}{c} \sin\left(\frac{\beta c t}{\rho}\right) \cos\theta\right]$$

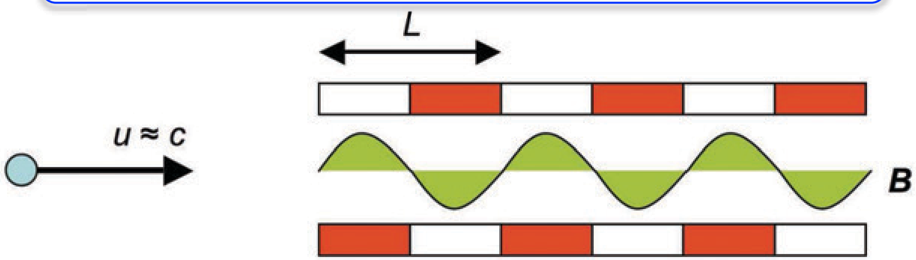
Substituting into the radiation integral and introducing

$$\xi = \frac{\rho\omega}{3c\gamma^3}\left(1 + \gamma^2\theta^2\right)^{3/2}$$

$$\frac{d^3W}{d\Omega d\omega} = \frac{e^2}{16\pi^3\varepsilon_0 c}\left(\frac{2\omega\rho}{3c\gamma^2}\right)^2 \left(1 + \gamma^2\theta^2\right)^2 \left[K_{2/3}^2(\xi) + \frac{\gamma^2\theta^2}{1 + \gamma^2\theta^2} K_{1/3}^2(\xi)\right]$$

Femtosecond X-ray Free electron lasers (GeV electrons)

Tunable wavelength:- from THz to X-ray



GeV e⁻ periodic magnet undulator

$$\lambda \simeq \underbrace{\frac{L}{\gamma}}_{\text{Lorentz contraction}} \times \underbrace{\frac{1}{2\gamma}}_{\text{Doppler}} = \frac{L}{2\gamma^2}; \quad B_{\text{lab}} \xrightarrow{\bar{F}=LFL^{-1}} \text{TEM}_{\text{electr}}$$

$$u^2 \to u^2 - v_T^2; \; 1/\gamma^2 \to 1/\gamma^2\left(1+\frac{v_T^2\gamma^2}{c^2}\right); \; v_T \sim BL$$

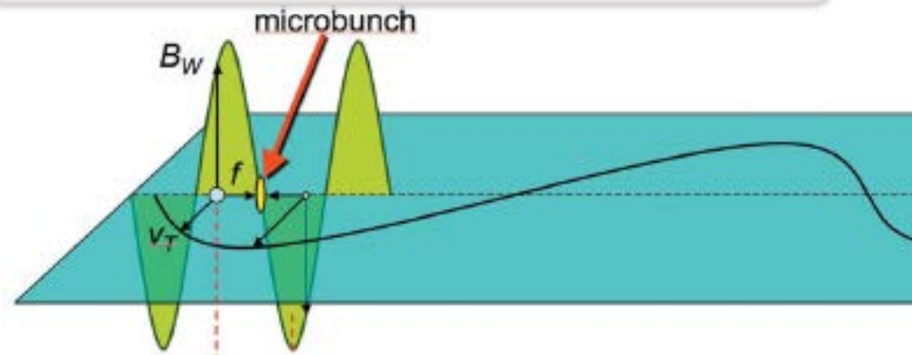
$$\lambda \simeq \frac{L}{2\gamma^2}[1+\underbrace{\gamma^2(\tilde{\kappa}BL/c)^2}_{K^2/2}] = \frac{L}{2\gamma^2}(1+K^2/2)$$

Wavelength tunable through *B, L, u*
e.g.:

$$\gamma = E/m_e = 5\text{GeV}/0.511\text{MeV} \simeq 1\times10^4, \; L = 7\text{cm}$$

$$\lambda \simeq 0.07\text{m}/[2\times(1\times10^4)^2] \simeq 3.5\text{Å} \; (\text{X-ray})$$

as/fs pulses from microbunches



Electrons form microbunches in space due to Lorentz force.

The emitted pulse duration is thus

$$\Delta t = N_u \lambda / c$$

These are as/fs photon pulses
e.g.:

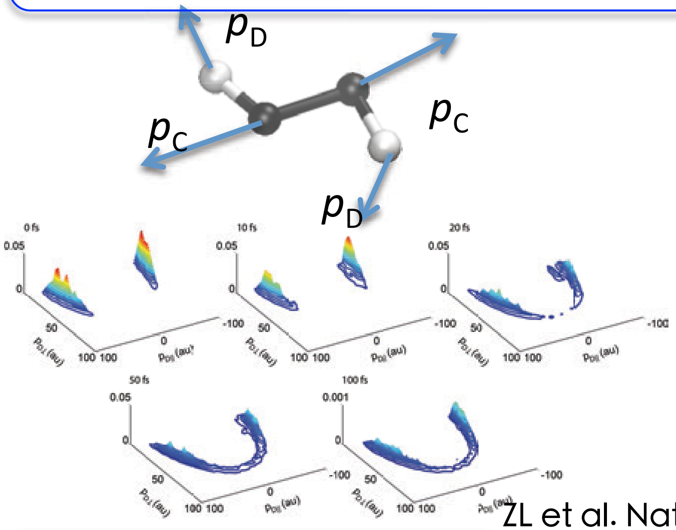
$$N_u = 1000, \; \lambda = 3.5\text{Å}$$

$$\Delta t = 1.2 \text{ fs}$$

JSR, 18, '11

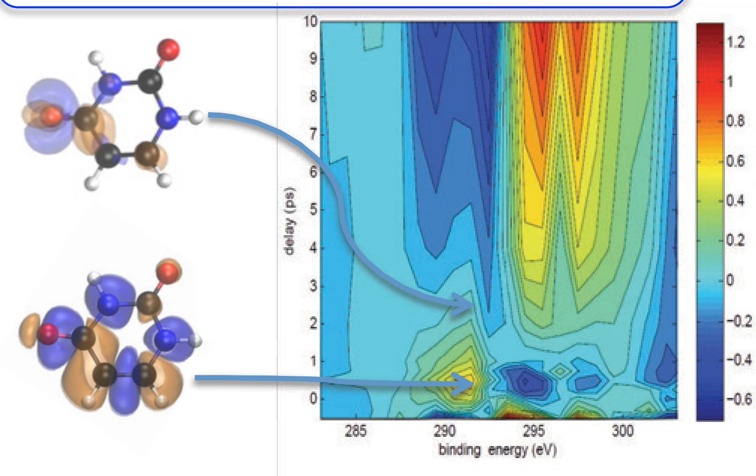
XFEL Applications

FEL as probe
Transient molecular structure



ZL et al. Nature Commun. 8, 453 (2017)

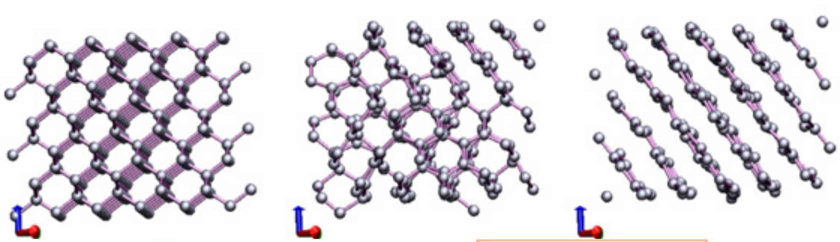
FEL as probe
Transient electronic structure

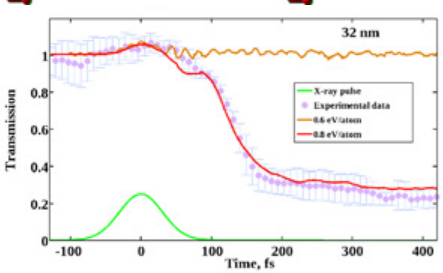


M. Ilchen,...,ZL et al. Phys. Rev. A, 95, 053423 (2017)

FEL as pump
Warm dense matter

-100fs
150fs
400fs

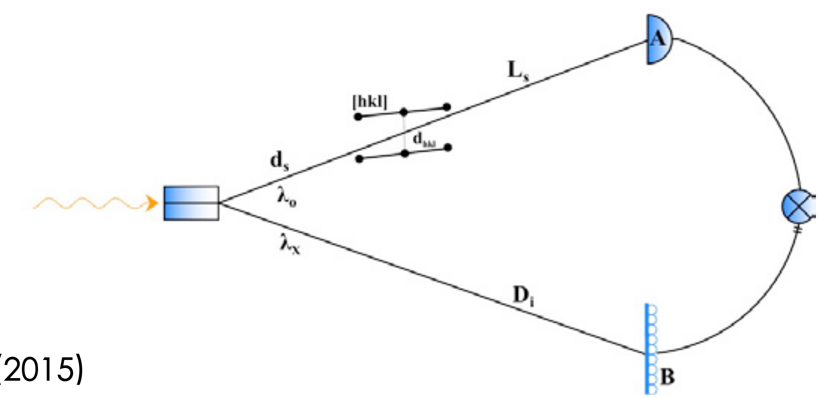




Absorbed dose = 0.8 eV/atom
FEL pulse duration = 51 fs
FEL photon energy = 47.4 eV
Probe pulse duration = 32.8 fs
Probe wavelength = 630 nm
Angle of pulse incidence = 20°

ZL, et al. Phys. Rev. Lett. 115, 143002 (2015)
Phys. Rev. B 91, 054113 (2015)
Phys. Rev. B 93, 144101 (2016)
Phys. Rev. B 95, 014309 (2017)

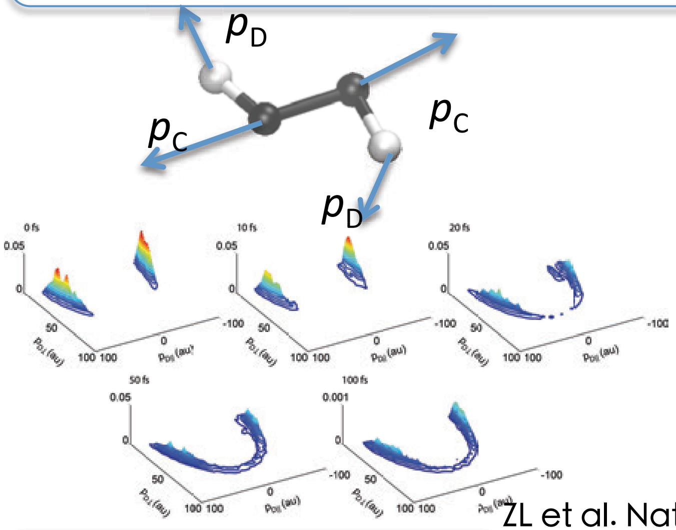
FEL for diffraction
Quantum diffraction



ZL et al., J. Phys. B (2018)
ZL et al., Europhys. Lett. (2017)

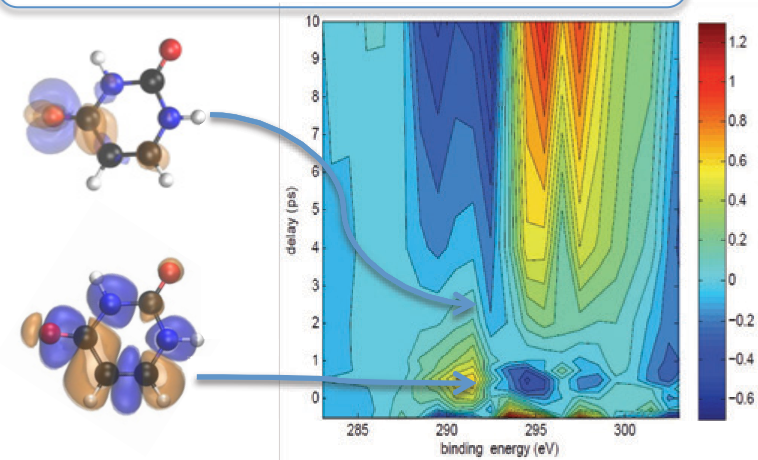
XFEL Applications

FEL as probe
Transient molecular structure



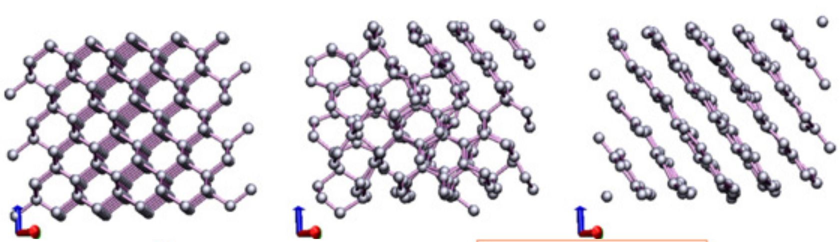
ZL et al. Nature Commun. 8, 453 (2017)

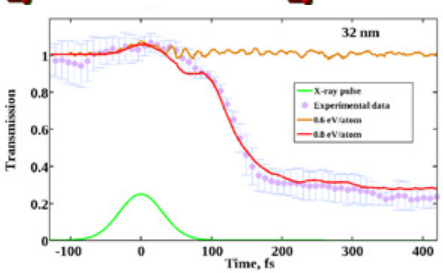
FEL as probe
Transient electronic structure



M. Ilchen,⋯,ZL et al. Phys. Rev. A,95,053423(2017)
ZL, T. Wolf et al., Submitted (2019)

FEL as pump
Warm dense matter





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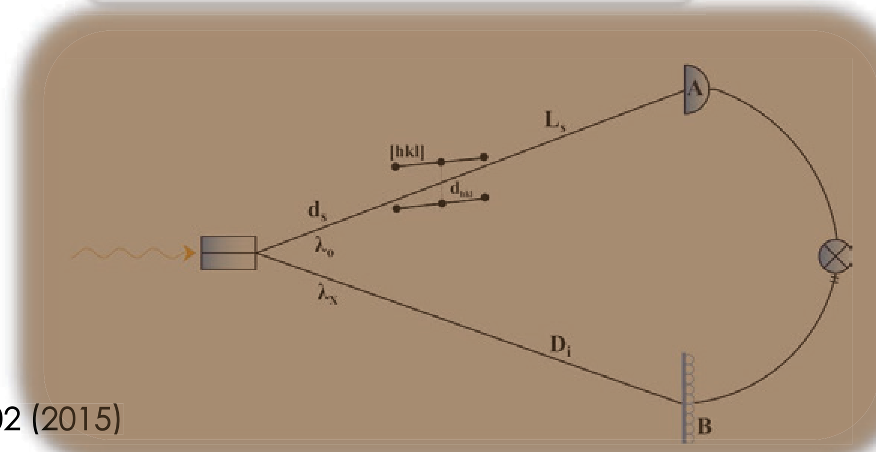
FEL for diffraction
Quantum diffraction



ZL et al., J. Phys. B (2018)
ZL et al., Europhys. Lett. (2017)

Quest for Ideal Microscope

Cryogenic Imaging

Cryo-TEM
Nobel Prize '2017

✓ Resolution: ~0.1nm
⊗ Not for living organism

Cryo X-ray diffraction
Nobel Prize '2009

✓ Resolution: ~0.1nm
⊗ Not for living organism

Room-Temperature Imaging

Superresolving microscope
Nobel Prize '2014

⊗ Resolution: ~20nm
✓ Radiation damage free

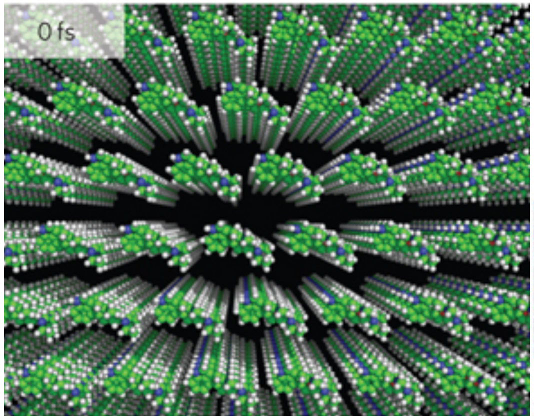
S. Hell *et al.* Opt. Lett. (1994); Opt. Lett. (1999)

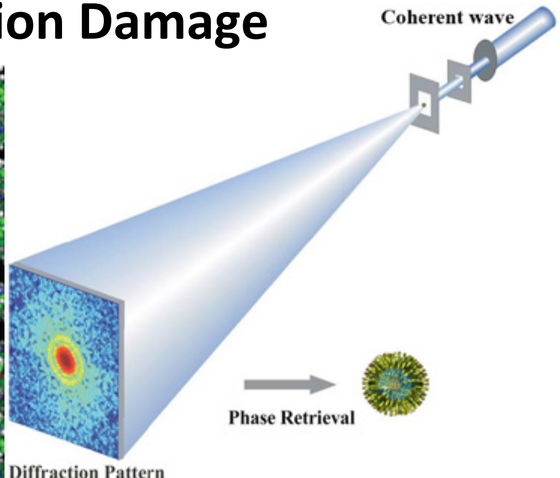
XFEL diffraction

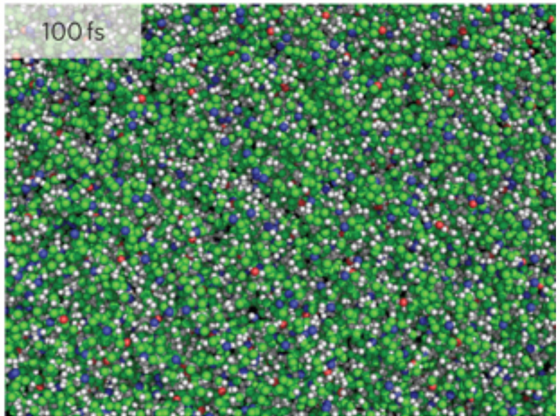
✓ Resolution: ~0.1nm
⊗ Serious radiation damage

H. Chapman *et al.*, Nature (2011)

Culprit for ⊗'s: Radiation Damage



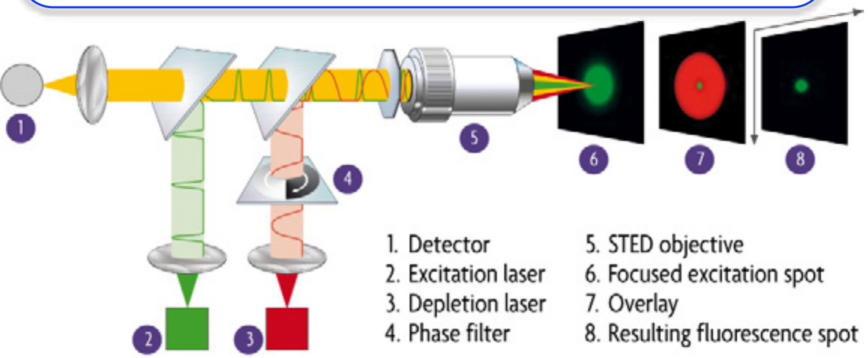




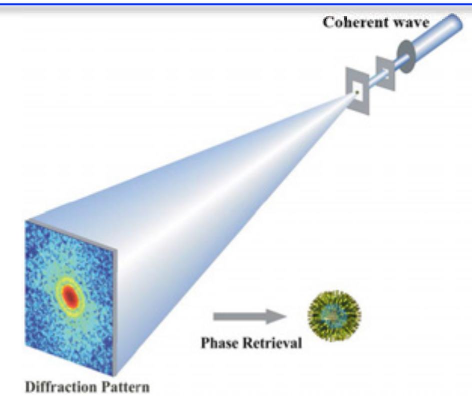
The Ideal Microscope

Superresolving microscope :
- ✓ Radiation damage free
- ✗ Resolution: ~20nm .

XFEL diffraction:
- ✓ Resolution: ~0.1nm
- ✗ Serious radiation damage .



Active Motif Inc.



An ideal microscope
- ✓ Resolution: ~0.1nm
- ✓ Radiation damage free
- ☺ For living organism
- ☺ For single molecules

Quantum Diffraction:- An Ideal Microscope?

Nikita Medvedev, Yanhua Shih and Henry Chapman

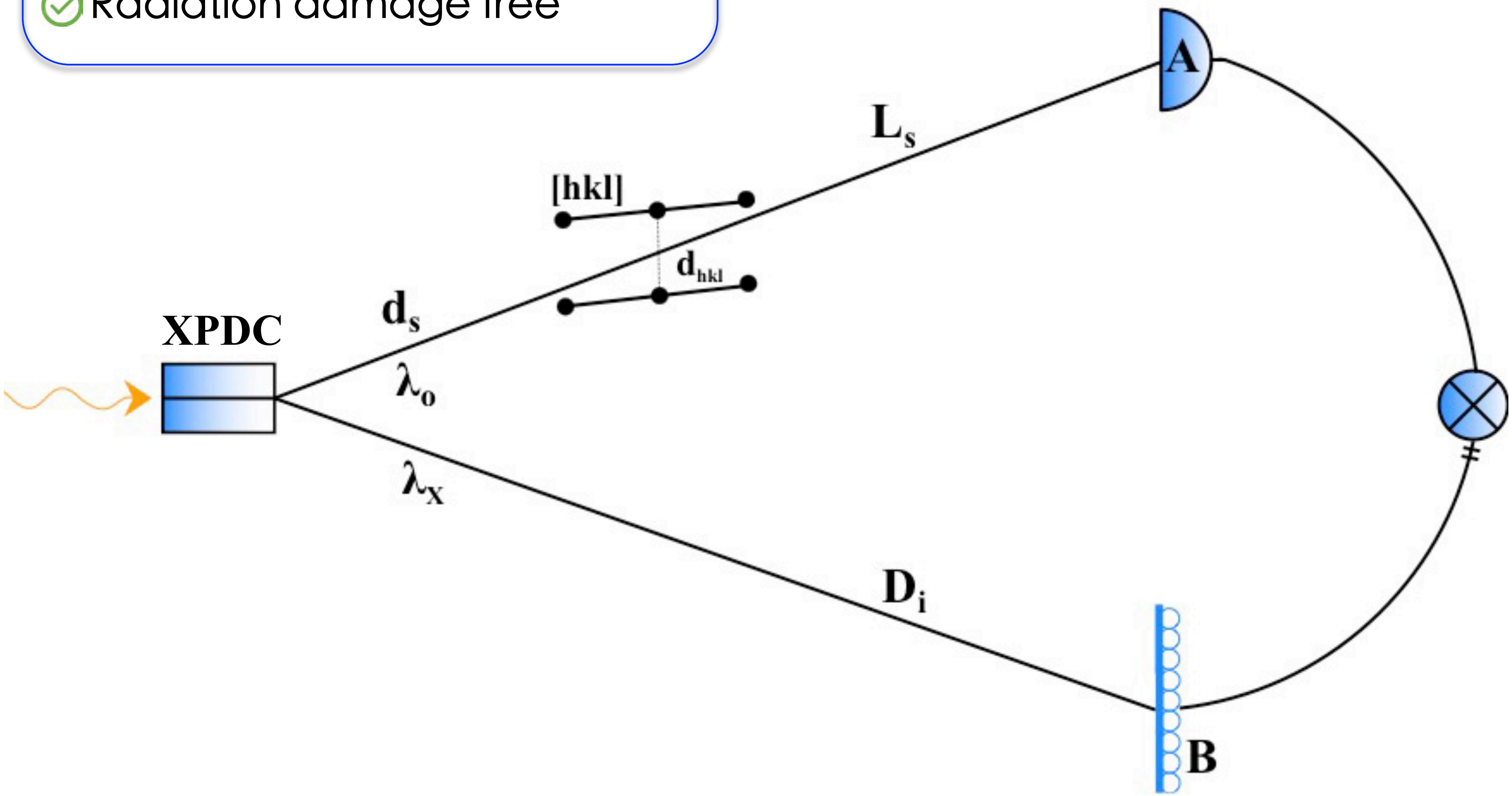
ZL, N. Medvedev, H. Chapman, Y. Shih, J. Phys. B (2018)
ZL, et al., Europhys. Lett. (EPL) (2017)

Is there more exotic quantumness we can exploit using FEL?

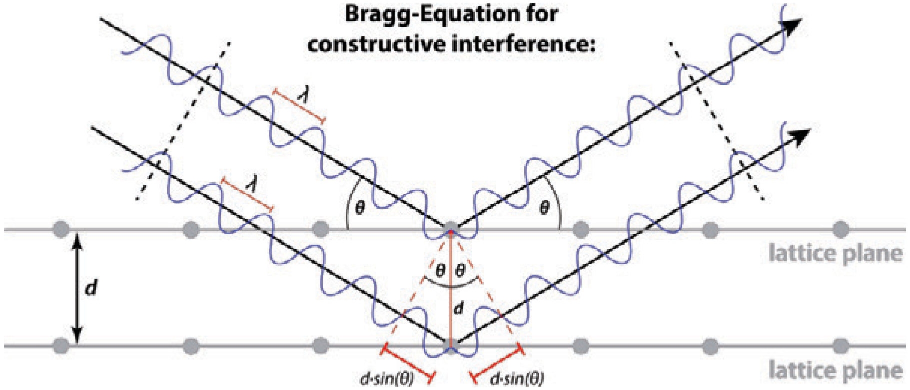
XFEL quantum diffraction

XFEL quantum diffraction

- ✓ Resolution: 0.1nm
- ✓ Radiation damage free



Modified Bragg condition for quantum diffraction



The Bragg condition can only be satisfied when light wavelength λ<2d.

$$2d\sin\theta = n\lambda$$

From 2nd order coherence function, we obtain the modified Bragg condition

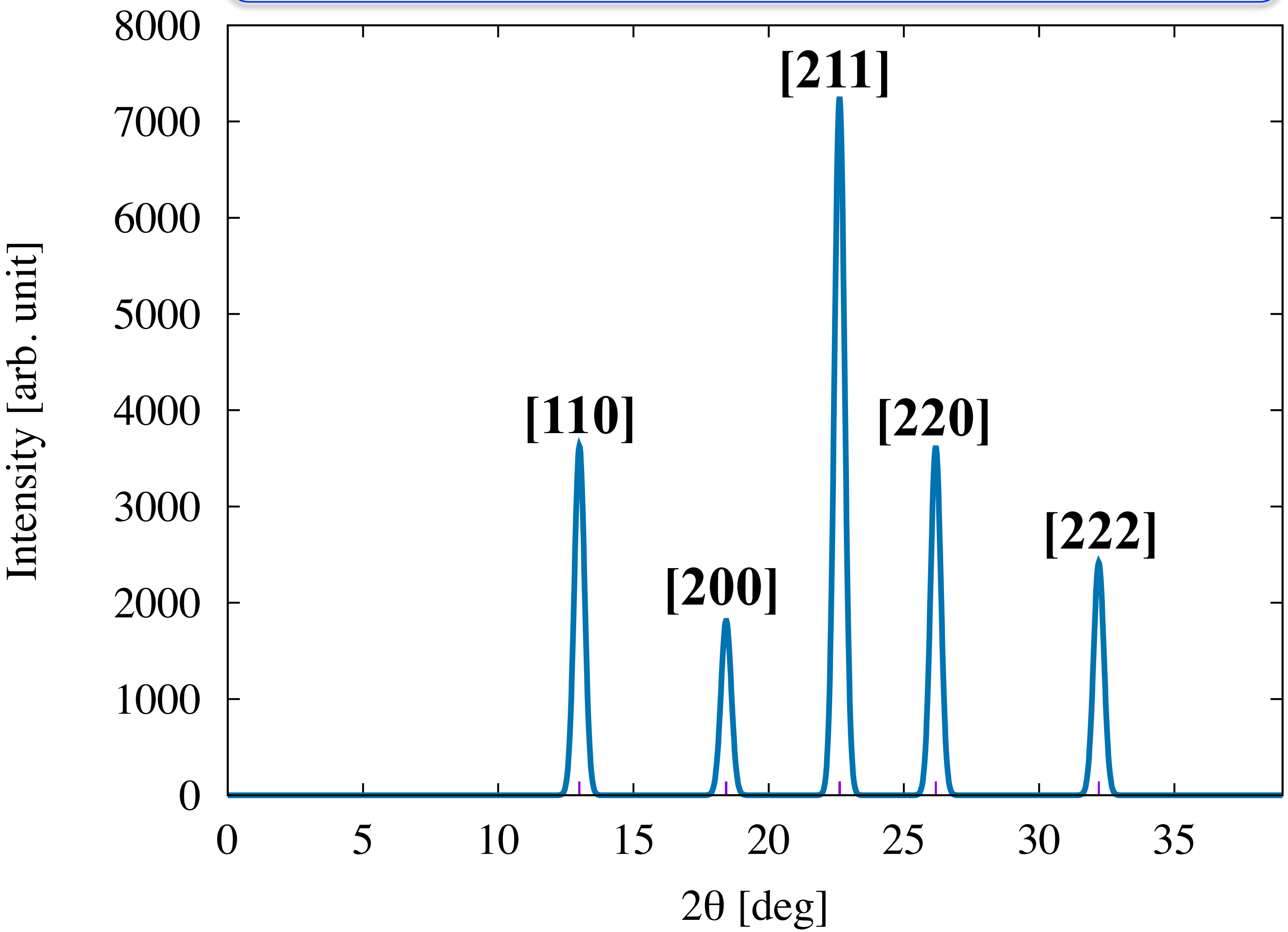
$$R_c(\rho_B) = \frac{1}{T}\int dt_A dt_B S(t_B,t_A)\int_{\sigma_A} d^2\rho_A\,\sigma_B \mathrm{tr}\left[E_A^{(-)}E_B^{(-)}E_B^{(+)}E_A^{(+)}\rho\right]$$

$$2d\sin\theta\left\{1+\frac{|\rho_B-d|^2}{\left(\frac{d_s}{D_i}+\frac{\lambda_X}{\lambda_o}\right)^2 D_i^{\,2}}\right\}=n\lambda_o$$

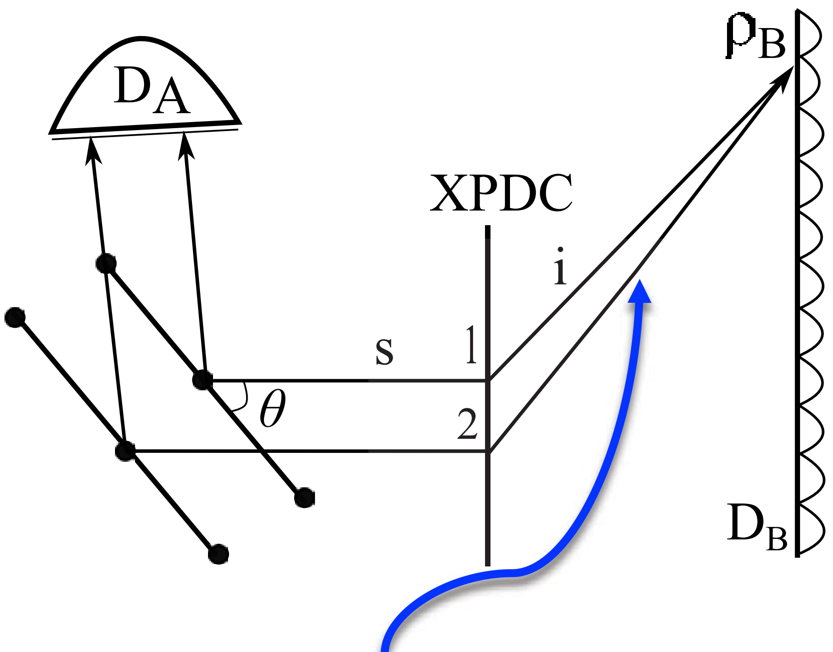
Magnification factor >10^3

Quantum diffraction (3.1eV photon)

bcc nanocrystal a=b=c=4Å
Optical photon 3.1eV, X-ray photon 3.1keV



Physical picture



$$2d\sin\theta + \frac{\lambda_{s,\mathrm{o}}}{\lambda_{i,\mathrm{X}}}\left(r_{B1} - r_{B2}\right) = n\lambda_{s,\mathrm{o}}$$

<u>Two-photon diagram</u>

$$\hat{E}_A^{(+)} = \hat{a}_{1s} e^{ik_s r_{A1}} + \hat{a}_{2s} e^{ik_s r_{A2}}$$

$$\hat{E}_B^{(+)} = \hat{a}_{1i} e^{ik_i r_{B1}} + \hat{a}_{2i} e^{ik_i r_{B2}}$$

$$G_{AB} = \mathrm{Tr}\left[\hat{E}_A^{(-)} \hat{E}_B^{(-)} \hat{E}_B^{(+)} \hat{E}_A^{(+)} \hat{\rho}\right]$$

$$\simeq \left| e^{ik_s r_{A1} + ik_i r_{B1}} + e^{ik_s r_{A2} + ik_i r_{B2}} \right|^2$$

Due to entanglement, we have

$$\Delta(x_s - x_i)\Delta(k_s + k_i) = 0 \quad \Rightarrow$$

we can thus concacenate optical paths at positions 1 and 2.

The phase is compensated by an optical path difference magnified $\frac{\lambda_{s,\mathrm{o}}}{\lambda_{i,\mathrm{X}}}$ times,

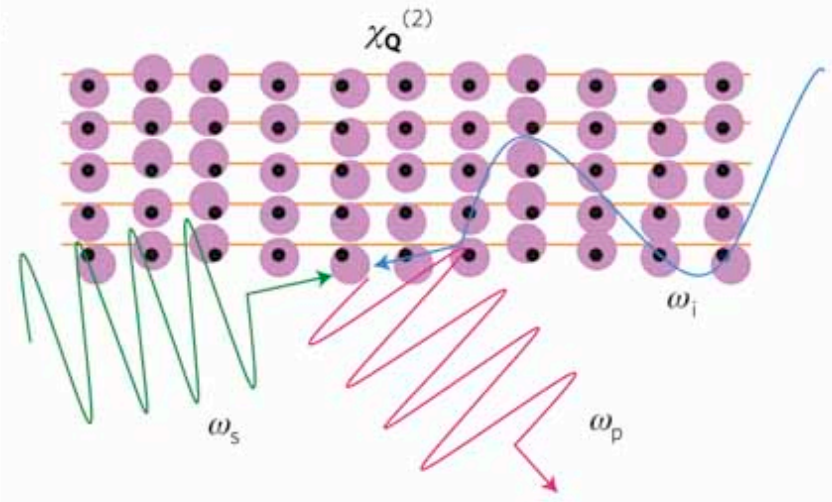
The modified Bragg condition can be satisfied though $d \ll \frac{\lambda_{s,\mathrm{o}}}{2}$.

D. N. Klyshko *et al.*, Usp. Fiz. Nauk (1988); JETP (1994)
D. V. Strekalov *et al.*, Phys. Rev. Lett. (1994)

Do we have sufficient such photon pairs?

$$\frac{\partial \vec{v}}{\partial t} + (\vec{v}\cdot\vec{\nabla})\vec{v} = -\frac{e}{m}(\vec{E} + \frac{1}{c}\vec{v}\times\vec{B})$$

$$\frac{\partial \rho}{\partial t} + \vec{\nabla}\cdot(\rho\vec{v}) = 0\,.$$



$$\begin{aligned} \vec{J}^{(2)}(\omega_3) &= \rho^{(0)}\vec{v}^{(2)} + \rho^{(1)}\vec{v}^{(1)} \\ &= \rho^{(0)}\frac{e^2}{m^2}\left[\frac{\vec{E}_1\times(\vec{k}_2\times\vec{E}_2)}{\omega_1\omega_2\omega_3} + i\frac{(\vec{E}_1\cdot\vec{\nabla})\vec{E}_2}{\omega_1^2\omega_3}\right] \\ &+ \frac{ie^2}{m^2}\frac{(\vec{\nabla}\rho\cdot\vec{E}_2)\vec{E}_1}{\omega_1^2\omega_2} + \text{terms with interchanged index 1 and 2}\,, \end{aligned}$$

K. Tamasaku *et al.*, Nature Phys. (2011)
S. Shwatz *et al.*, Phys. Rev. Lett. (2012)

$$eJ_i^{(2)}(\omega_3) \propto R_{ijk}^{(2)}(\omega_3;\omega_1,\omega_2)A_j(\omega_1)A_k(\omega_2)$$

$$\chi^{(n)}(\omega;\omega_1,\omega_2,\cdots,\omega_n) = i^{1-n}\frac{c^n}{\omega_1\omega_2\omega_n}R^{(n)}(\omega;\omega_1,\omega_2,\cdots,\omega_n)$$

$$\frac{d\sigma}{d\Omega}^{(2)} = \frac{\omega_3\omega_2^3\omega_1^3}{288\pi^3c^7}\left|\chi^{(2)}\right|^2$$

XPDC from diamond crystal

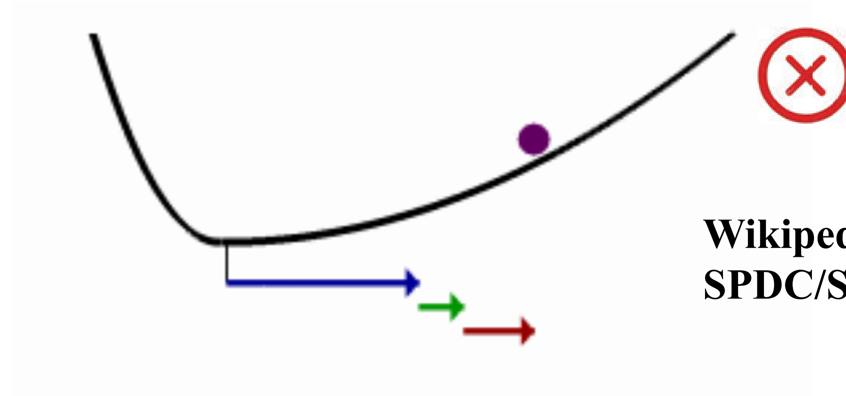
$$\frac{d\sigma}{d\Omega}^{(2)} \sim 1.9\times10^3\,\text{fm}^2 = 19\,\text{b} \quad \text{XPDC}\ \omega_3(=3.1\,\text{keV}) \to \omega_2(=3096.9\,\text{eV}) + \omega_1(=3.1\,\text{eV})$$

ZL, N. Medvedev, H. Chapman, Y. Shih, J. Phys. B (2017)

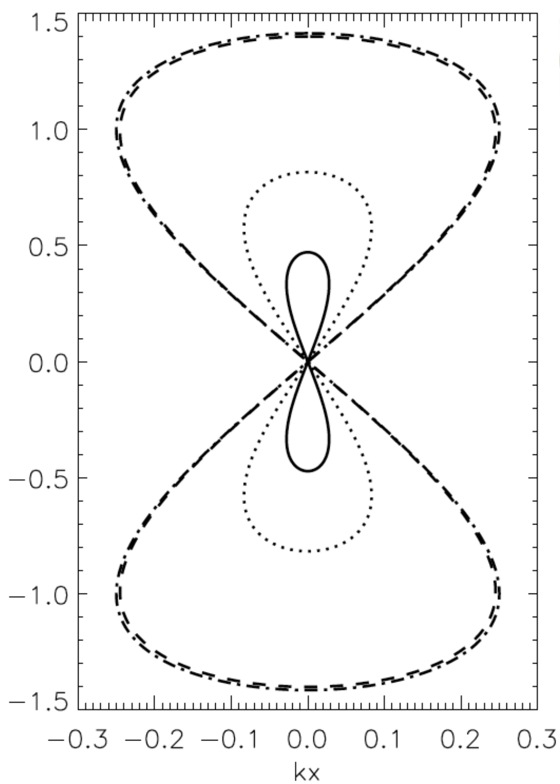
Single electron as nonlinear medium for XPDC

$$\frac{J^{(2)}_{\text{anharmonic}}}{J^{(2)}_{\text{figure}-8}} = \frac{\lambda}{2\pi d}\frac{\omega_0^2}{\omega^2} \leq 10^{-6}$$

for $\omega_0 \sim 1\text{eV}$ and $\omega \sim 1\text{keV}$



Wikipedia: SPDC/SHG



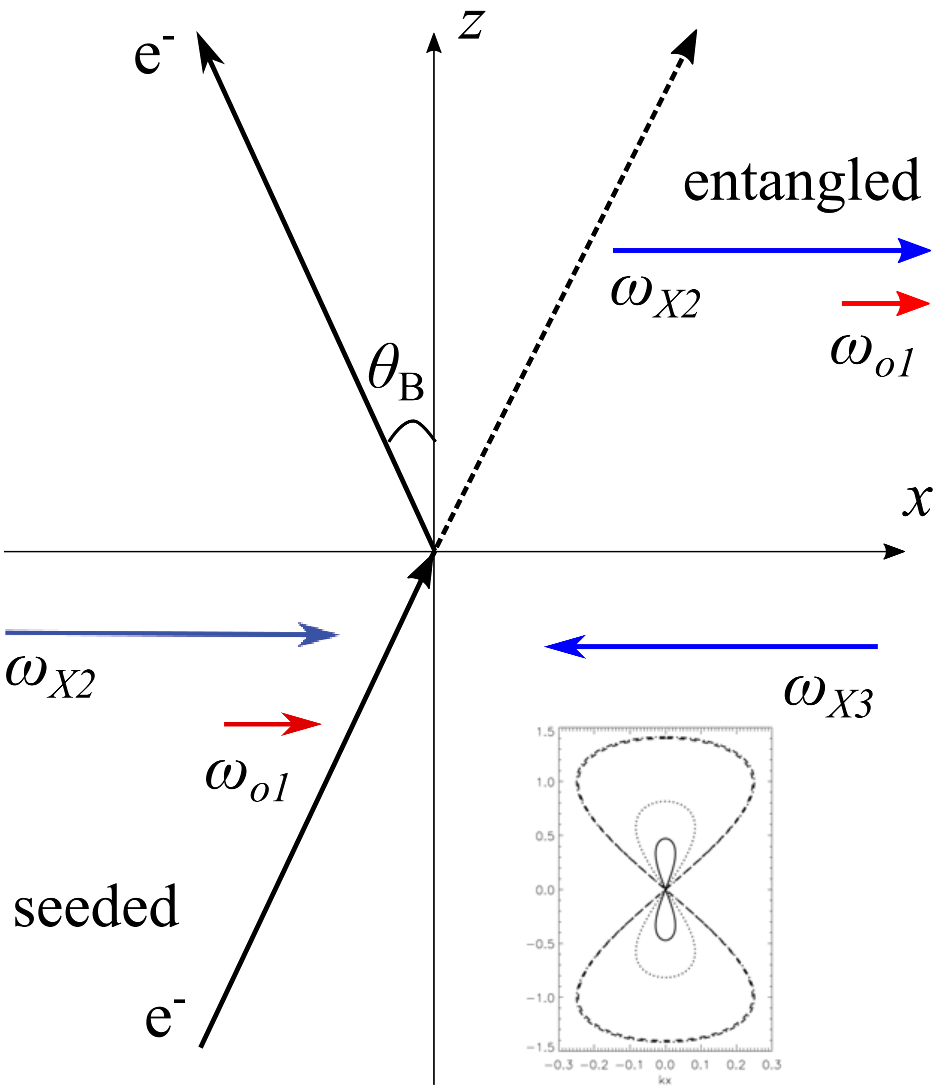
$$x = -\frac{a^2}{8k(1+a^2/2)}\sin 2\phi$$

$$z = \frac{a}{k\sqrt{1+a^2/2}}\cos\phi\,,$$

$$\phi = \omega t - kx \quad a = \frac{A}{c}$$

We can do XPDC without crystal!

Single electron XPDC:- Kapitza-Dirac-like process



XPDC $\omega_{X3} \rightarrow \omega_{o1} + \omega_{X2}$

using single electron as nonlinear medium.

The electron is deflected by θ_B.

Using Kapitza's method of pondermotive decomposition, we obtain

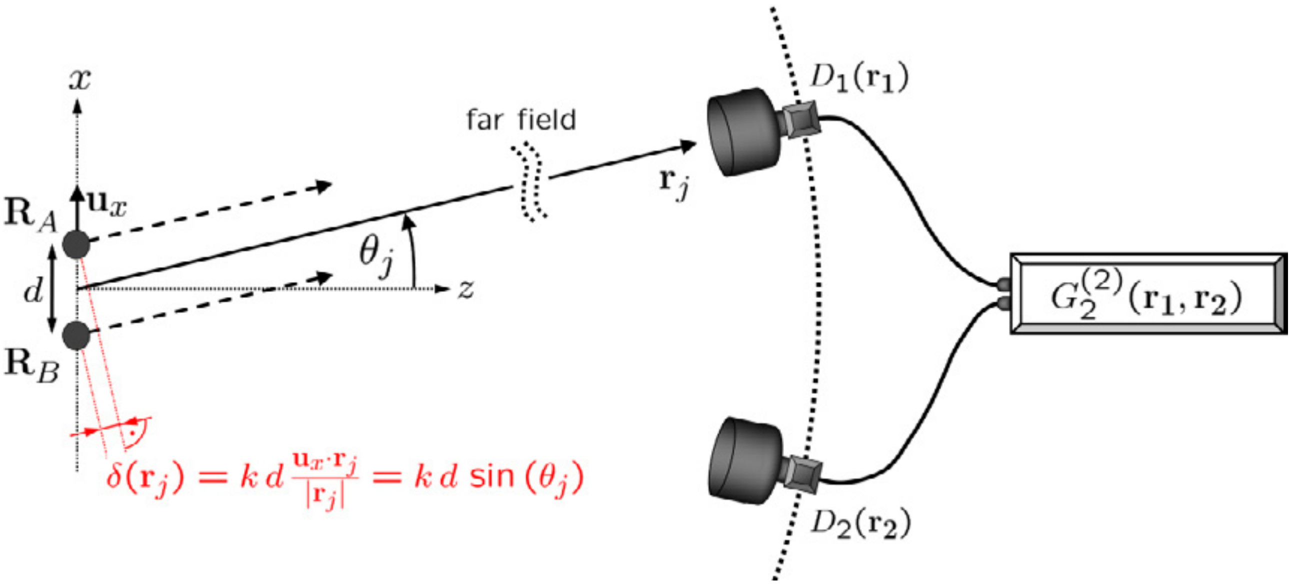
$$P(\omega_1, \omega_2, \omega_3) = \left| \frac{v_z E_1 E_2 E_3}{2c^2 \omega_1 \omega_2 \omega_3} \right|^2 \delta(\varepsilon_{fi})$$

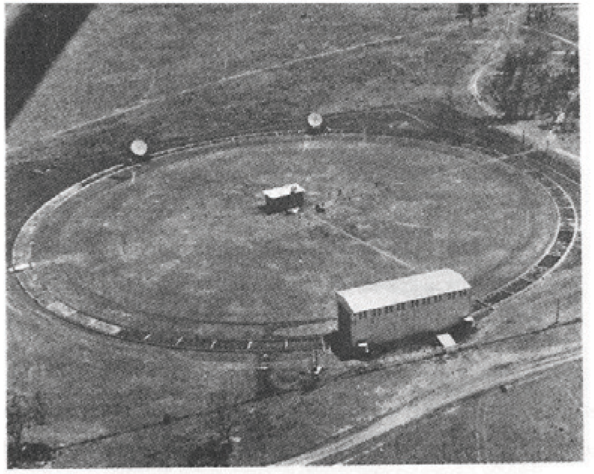
P could eventually reach 10^{-5} with laser intensity up to 10^{18} W/cm^2.

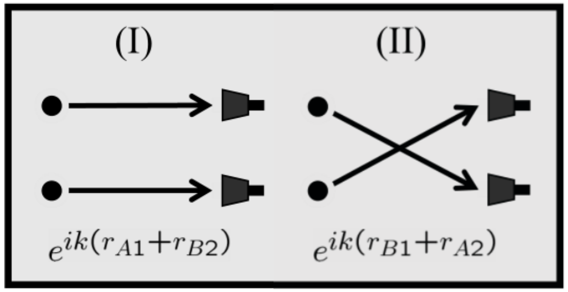
ZL, N. Medvedev, H. Chapman, Y. Shih, J. Phys. B (2017)

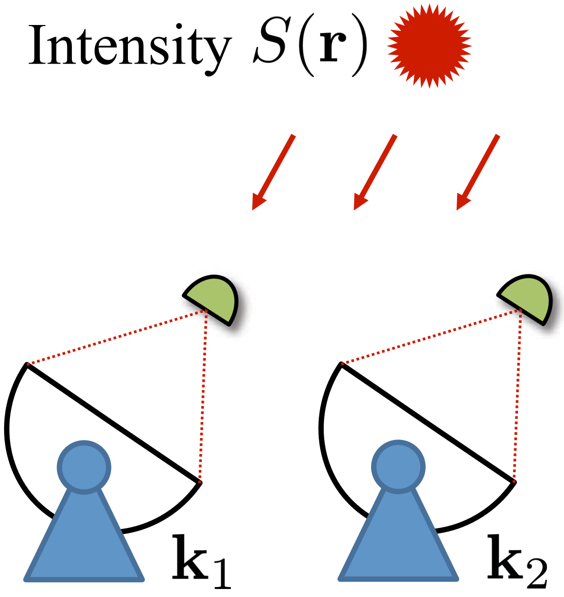
M. V. Fedorov, Electrons in a Strong Field, Nauka, Moscow (1991)
O. Smirnova *et al.*, Phys. Rev. Lett. (2004)
D. Bauer, Vorlesungsskript, MPI-K

Incoherent molecular diffraction





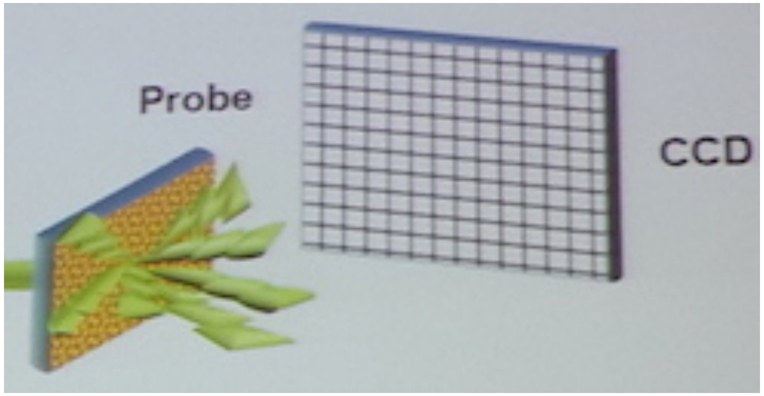




$$g^{(2)}(\mathbf{k}_1, \mathbf{k}_2) \sim |e^{ik(r_{A1}+r_{B2})} + e^{ik(r_{A1}+r_{B2})}|^2$$

Max von Laue's original experiment

tion experiment. "Because we believed at first that we had to deal with fluorescence radiation, a crystal had to be chosen that contained a metal with a considerable atomic weight," Friedrich and Knipping presented as the argument why they chose copper sulphate as a crystal [3, p. 314]. In other words: The crystal was not imagined to act as a three-dimensional diffraction grating for the *primary* beam of X-rays, but as an emitter of the so-called characteristic X-rays. Laue apparently expected that if this characteristic radiation originates from the regularly arranged points of the crystal's space lattice, then they should be the photographic plates on which the diffraction pattern was recorded were placed left and right and in the back of the crystal [3, Fig. 1], so that



Eckert, Ann. Phys. '12

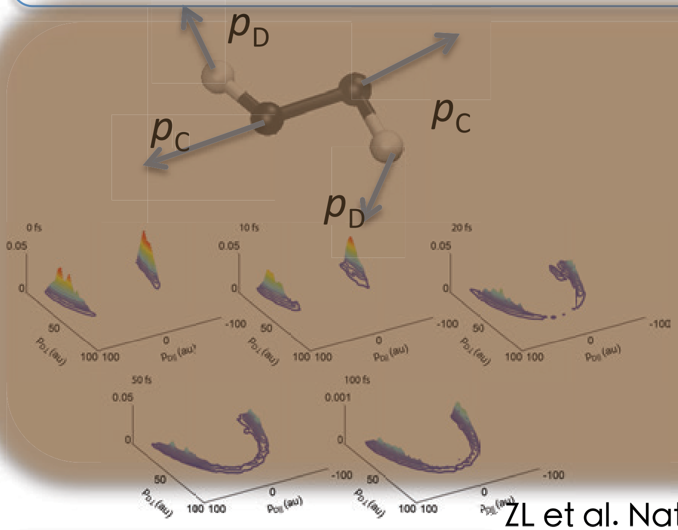
Background-free - different wavelength
Superresolving - covering larger Ewald Sphere (two photon wave vec. $q=k_1-k_2$)
Free of wave-front distortion

ZL, Anton Classen, et al. arXiv (2016); Europhys. Lett. (2017)
Anton Classen, Kartik Ayyer, Henry Chapman et al. PRL (2017)

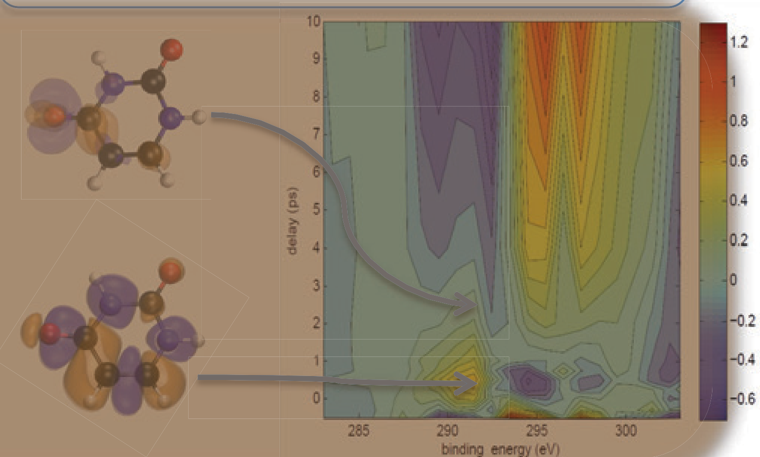
XFEL Applications

FEL as probe
Transient molecular structure

FEL as probe
Transient electronic structure



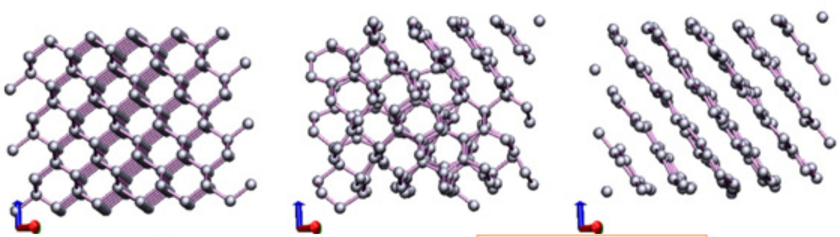
ZL et al. Nature Commun. 8, 453 (2017)

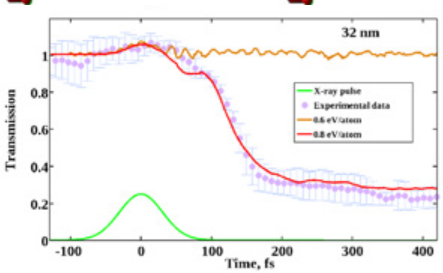


M. Ilchen,···,ZL et al. Phys. Rev. A,95,053423(2017)
ZL, T. Wolf et al., Submitted (2019)

FEL as pump
Warm dense matter

-100fs
150fs
400fs

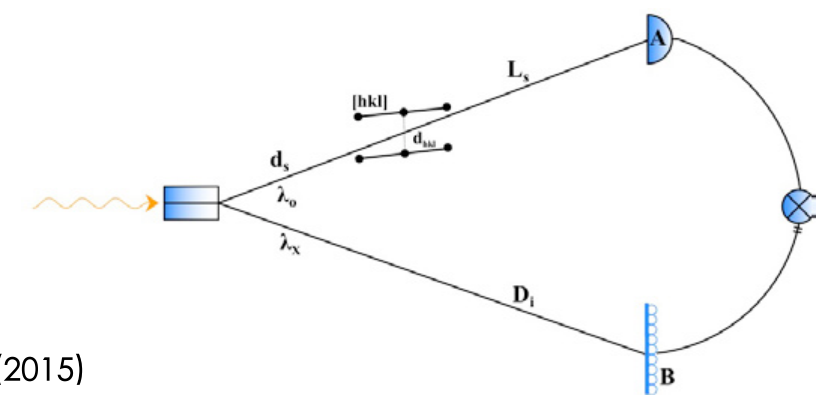




Absorbed dose = 0.8 eV/atom
FEL pulse duration = 51 fs
FEL photon energy = 47.4 eV
Probe pulse duration = 32.8 fs
Probe wavelength = 630 nm
Angle of pulse incidence = 20°

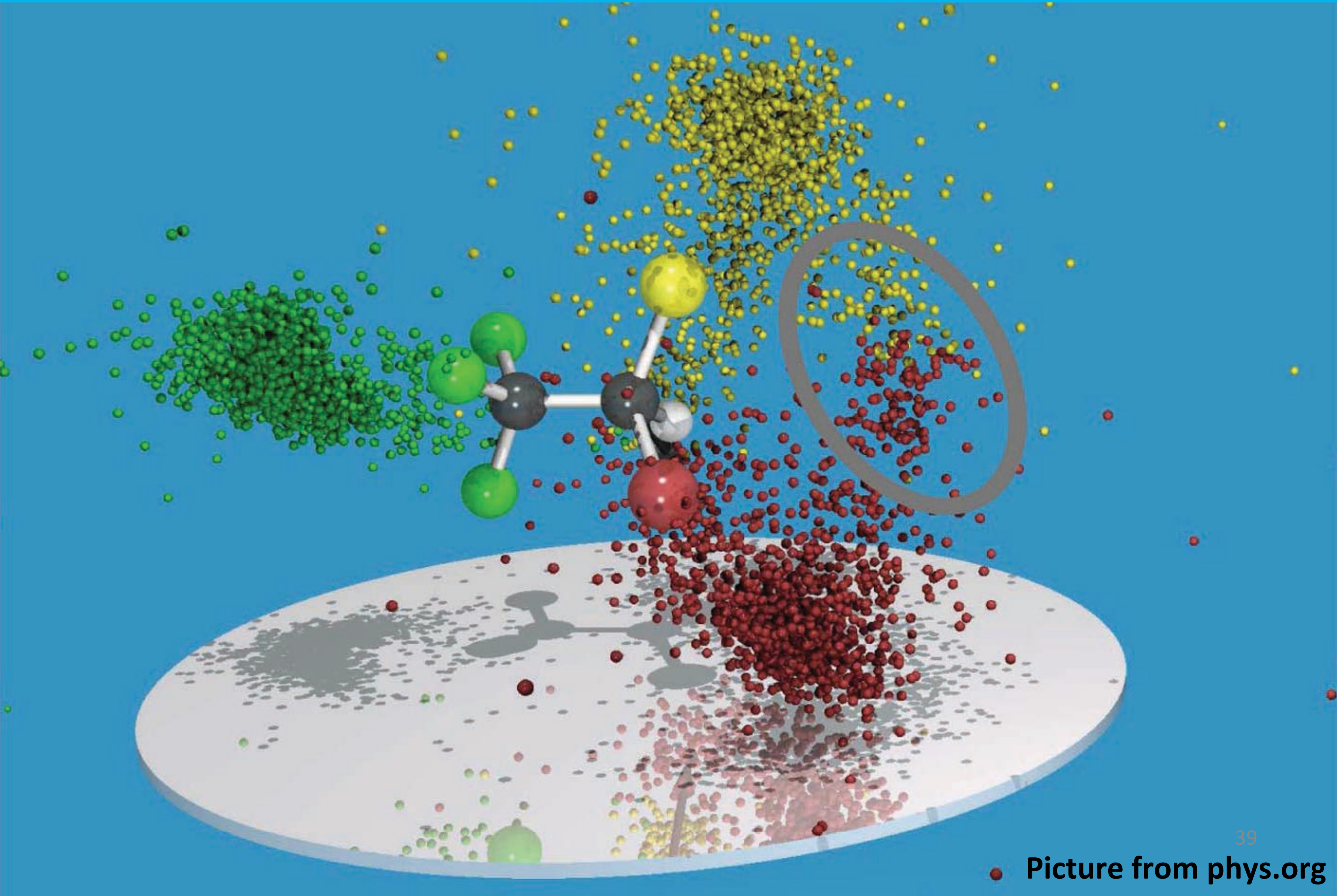
ZL, et al. Phys. Rev. Lett. 115, 143002 (2015)
Phys. Rev. B 91, 054113 (2015)
Phys. Rev. B 93, 144101 (2016)
Phys. Rev. B 95, 014309 (2017)

FEL for diffraction
Quantum diffraction



ZL et al., J. Phys. B (2018)
ZL et al., Europhys. Lett. (2017)

Alternative of X-ray imaging:- Coulomb explosion imaging



- Picture from phys.org

Basile, Ludger, Todd, Stefan, Lamine, Sang-Kil, Oriol, Robin

ZL et al. Nature Commun. 8, 453 (2017)

X-ray Coulomb explosion imaging

X-CEI

Old method (CEI since 1980's) - New questions
Coulomb explosion imaging now in X-ray femtosecond regime

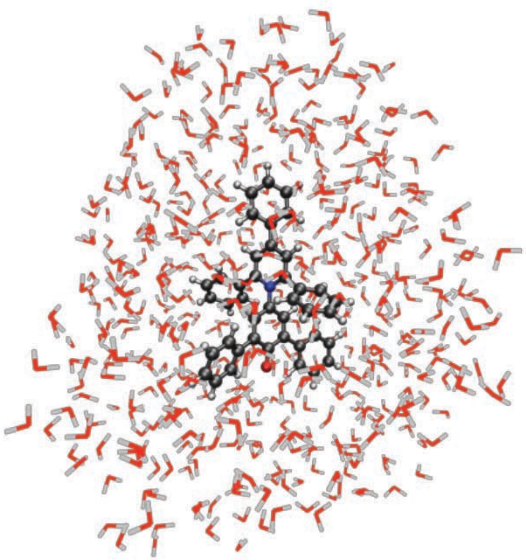
Examine:
Is Coulomb explosion imaging trustworthy in every aspects?
⊗ **No!**

The difference:
? **X-ray core ionization CEI vs. UV/VUV valence ionization CEI.**

Ab initio molecular dynamics of core-excited molecules:
? **Unconventional methodologies.**

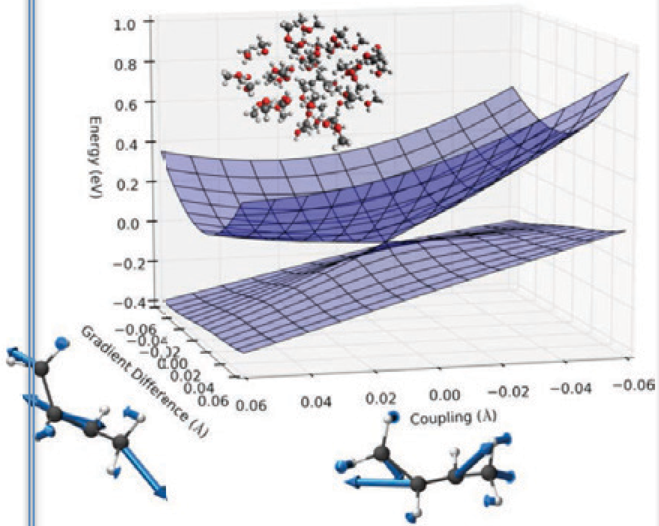
GPU accelerated ab initio molecular dynamics

Energy Calculations:
1000-2000 atoms



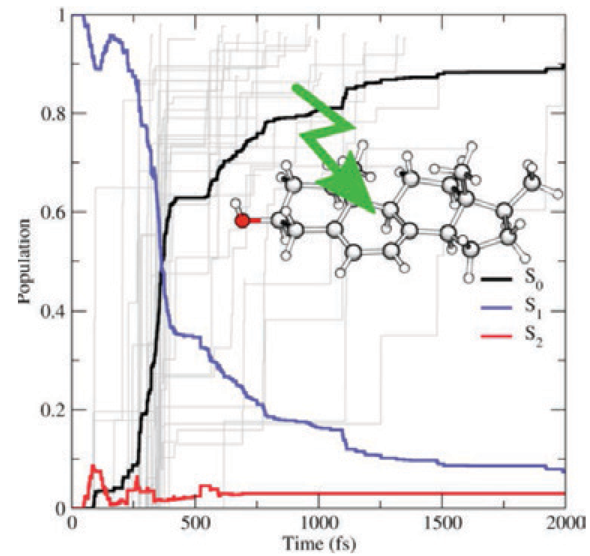
Betaine-30
Microsolvated in
500 Water Molecules

Conical Intersection:
250-750 atoms



Butadiene
Microsolvated in
50 Methanol Molecules

Excited State Dynamics:
Up to 100 atoms

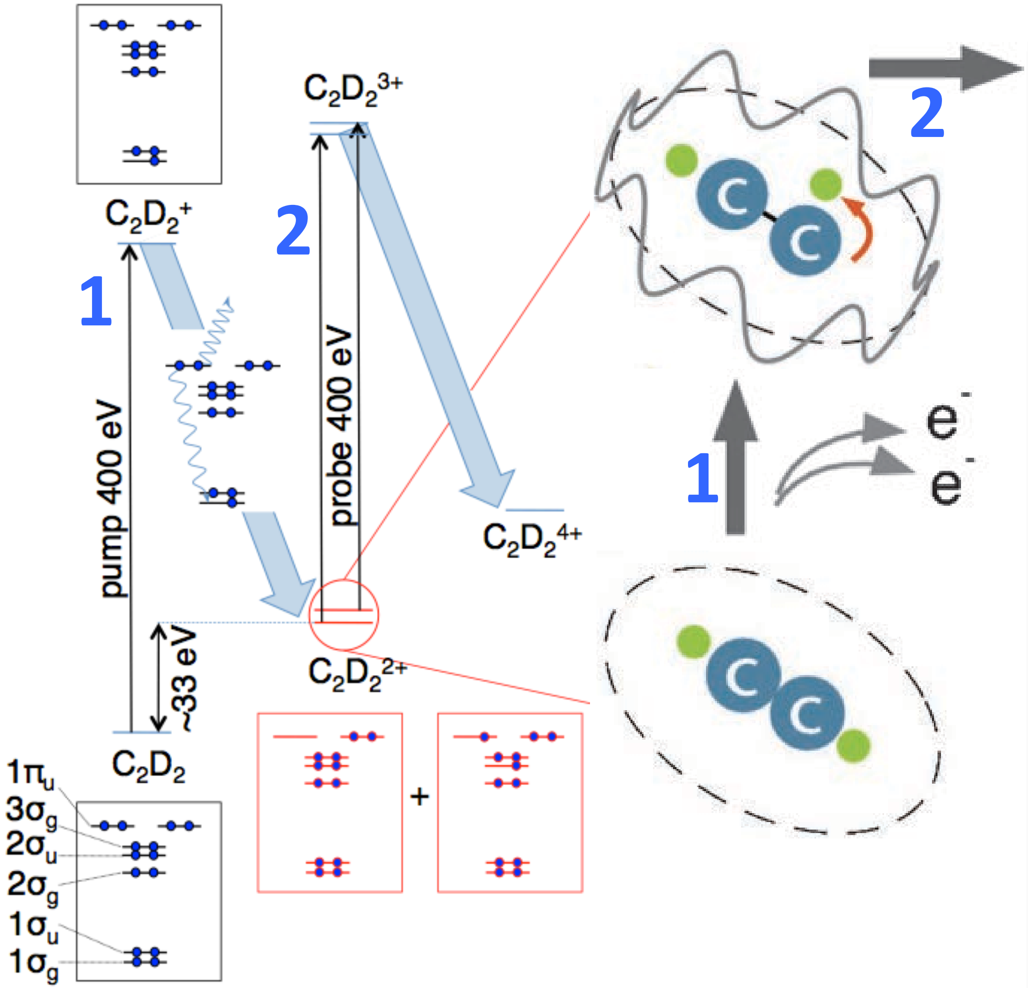


Provitamin D_3

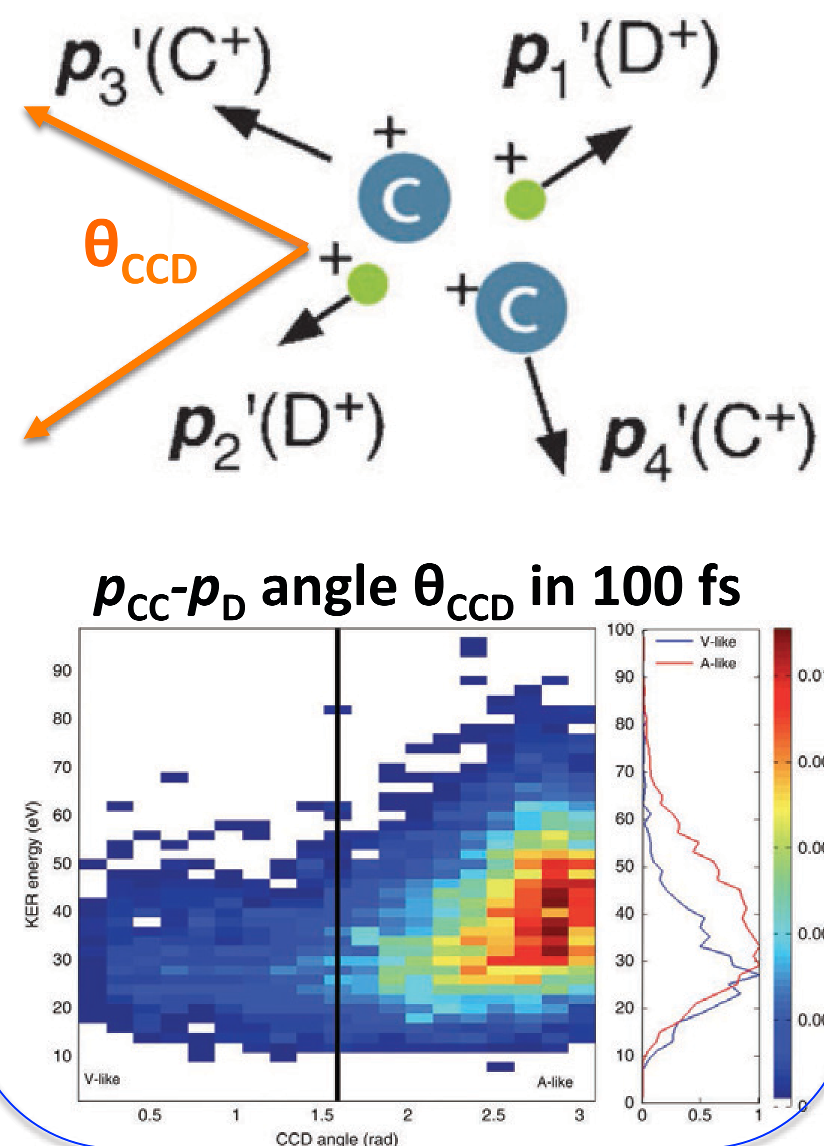
Fully GPU-based TeraChem@SLAC

Isomerisation of acetylene:- X-CEI@SLAC

X-ray ionization – Auger decay

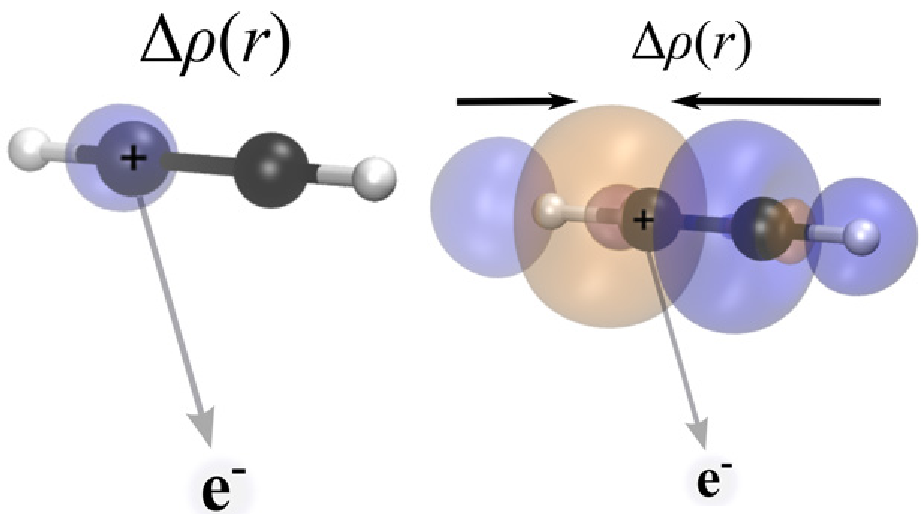


X-ray CEI



T. Osipov *et al.*, Phys. Rev. Lett., 90, 233002 (2003)
A. Matsuda *et al.*, PCCP, 13, 8697 (2011)
C. Liekhus-Schmalz *et al.*, Nature Commun., 6, 8199 (2015)

Isomerization of X-ray core-ionized acetylene?



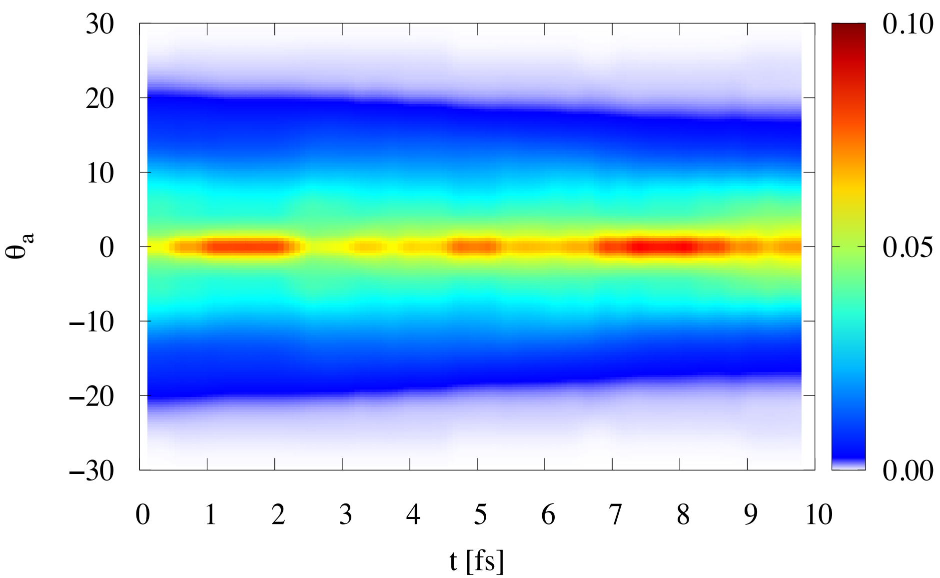
Paradox

Bonds strengthened after X-ray ionizing a bonding electron ?!

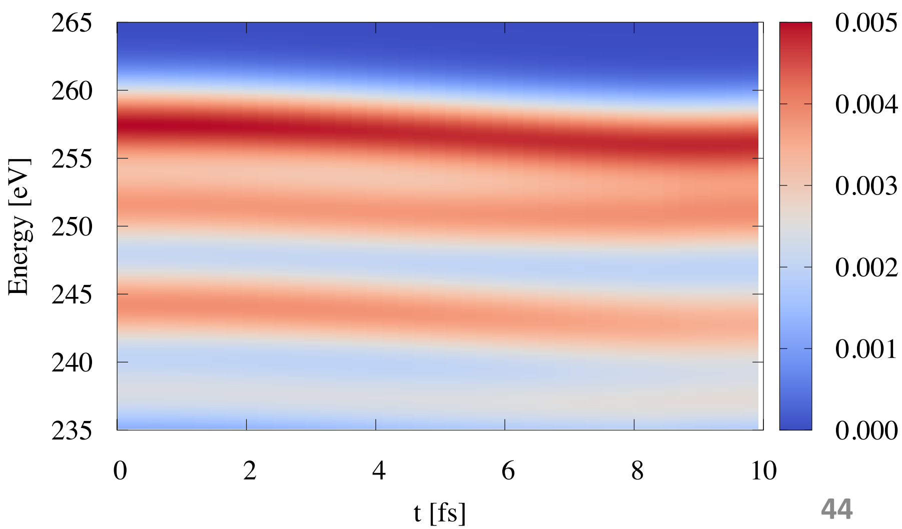
No isomerization :-(

Counterexample to L. Cederbaum et al. JMS '1996

Angular distribution of hydrogen



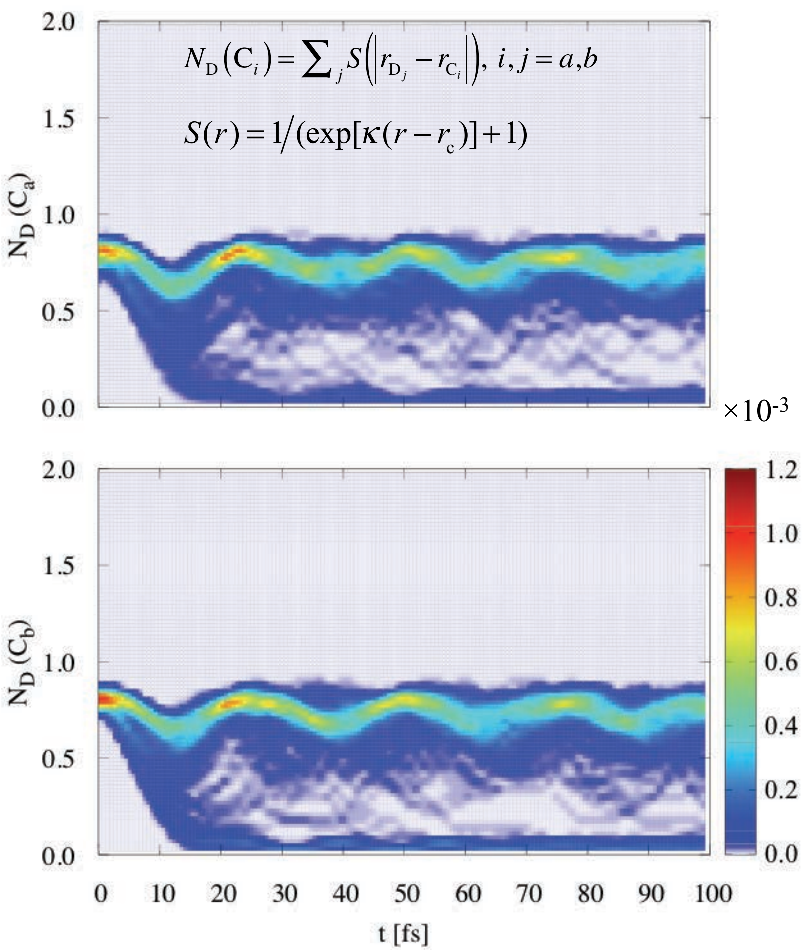
Time dependent Auger spectra

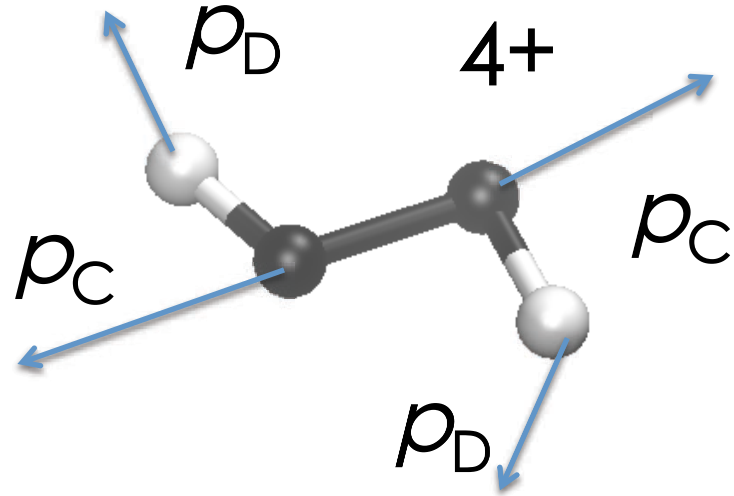


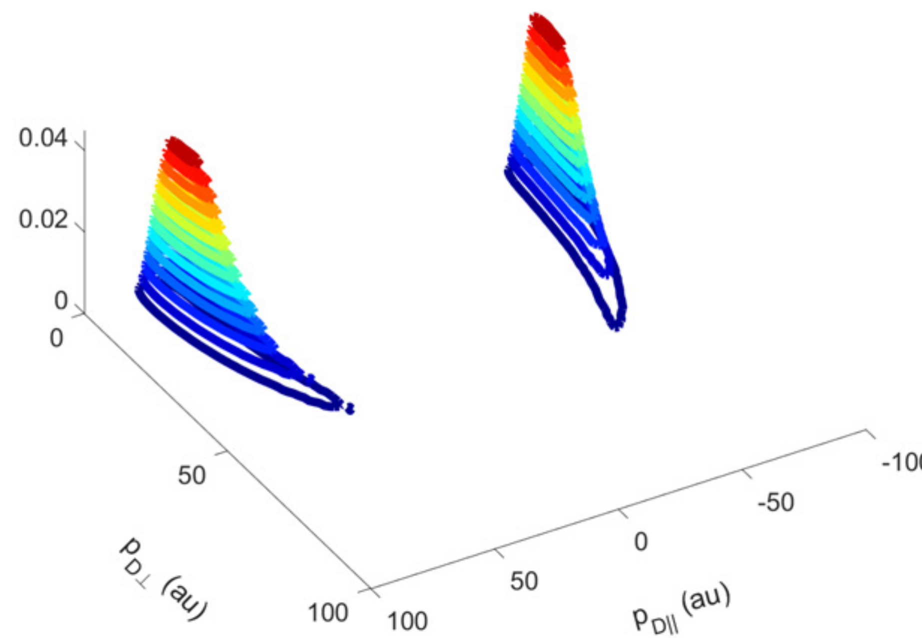
Isomerization of doubly-ionized acetylene?

Potential barrier >2 eV
D coordination number N_D<1

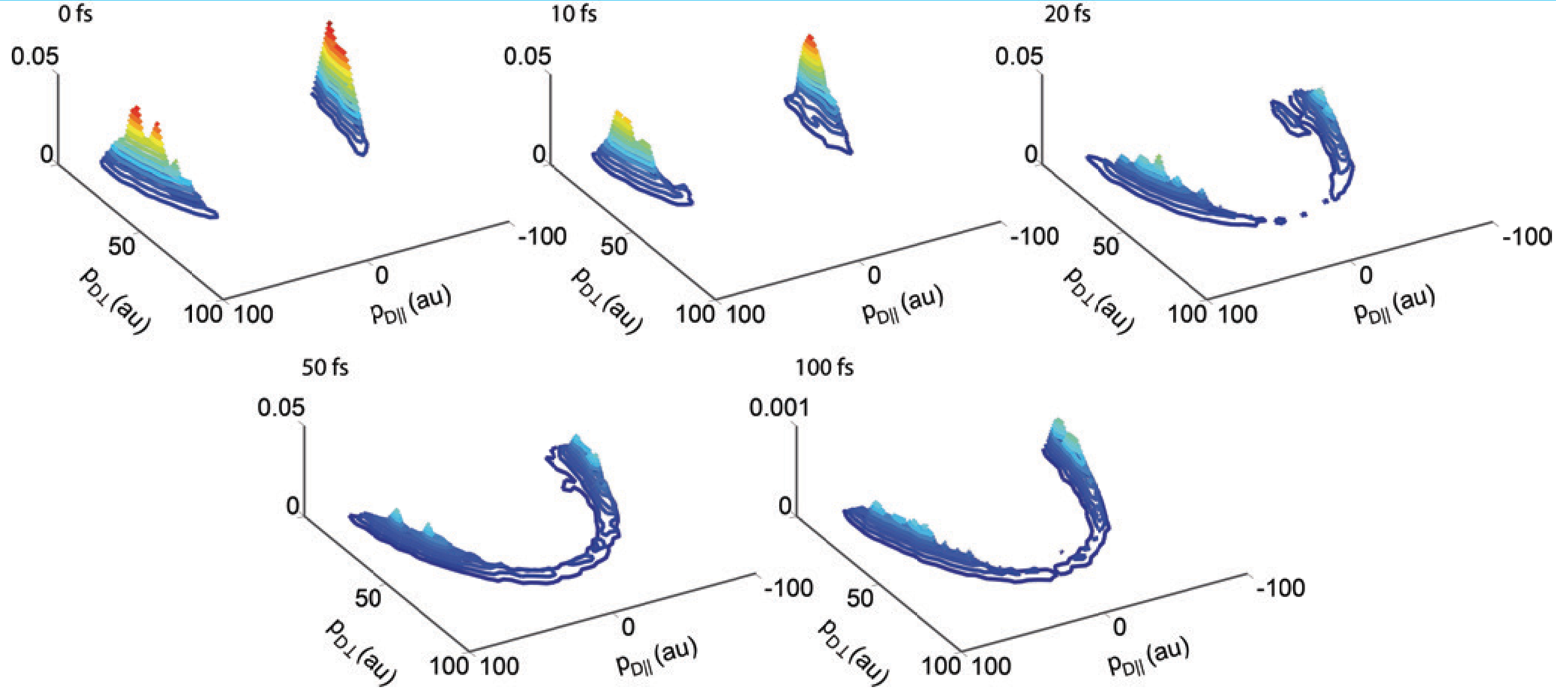
Still no isomerization :-(

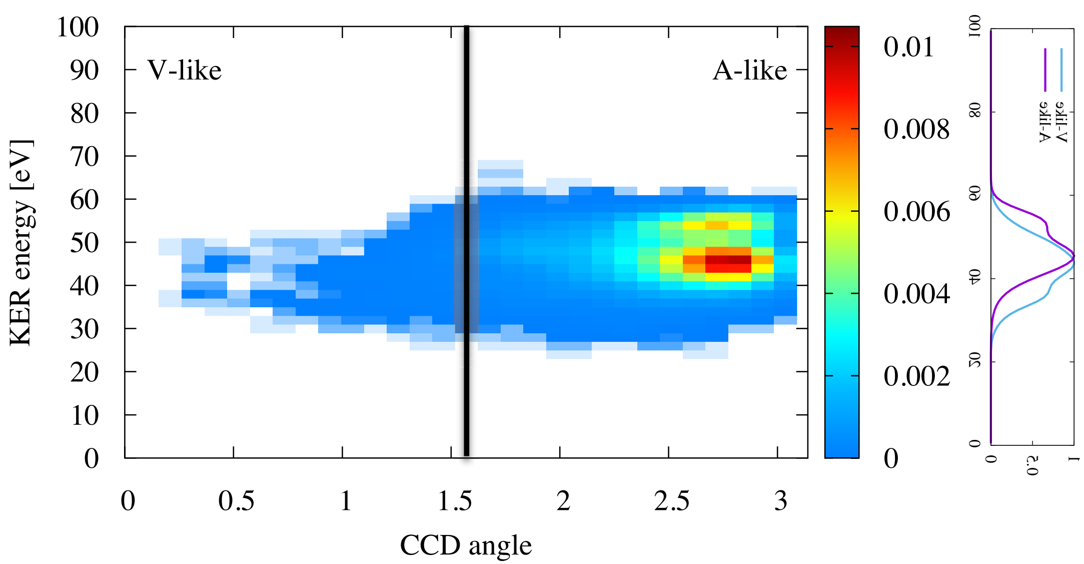


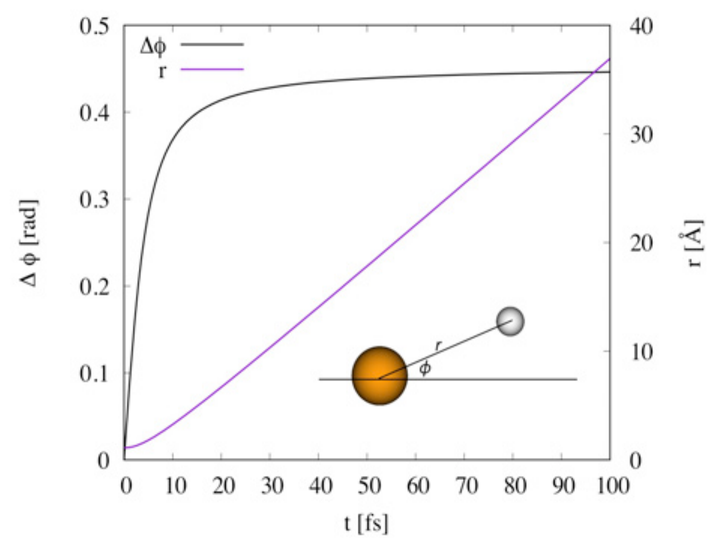




Reproduce Coulomb explosion imaging observation without isomerization







ZL, L. Inhester, C. Liekhus-Schmalz, T. Osipov, P. Bucksbaum, T. Martinez *et al*. Nature Commun. (2017)

Phase space at ultrafast time scale with REMI

Reaction Microscope
REMI has weakness

☹

REMI has to be amended before it could be used for angular motion.

☹

Eventually not amendable.

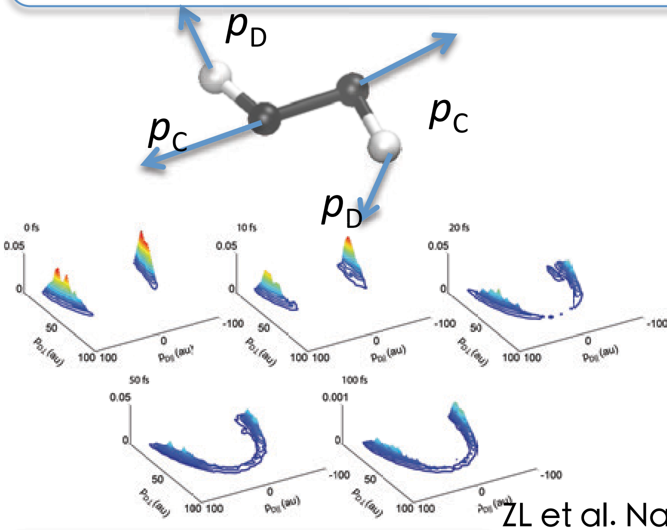
☺

We can still measure transient angular momentum distribution with REMI.

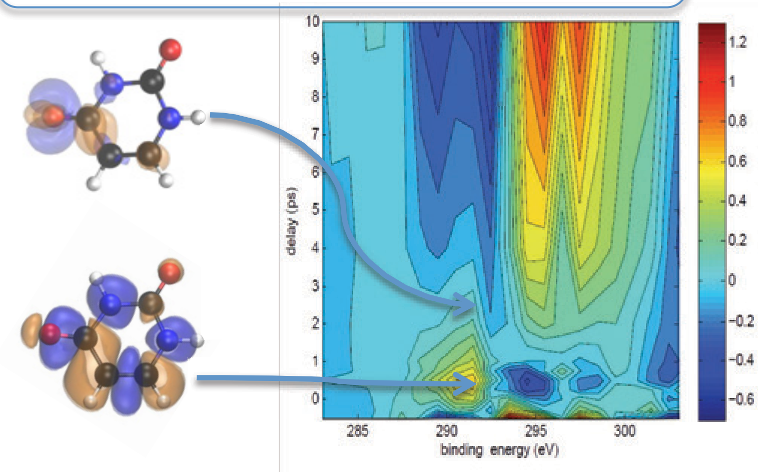
'14-'17 SLAC/DESY, Ewald Fellow for Free Electron Laser Physics

FEL as probe
Transient molecular structure

FEL as probe
Transient electronic structure

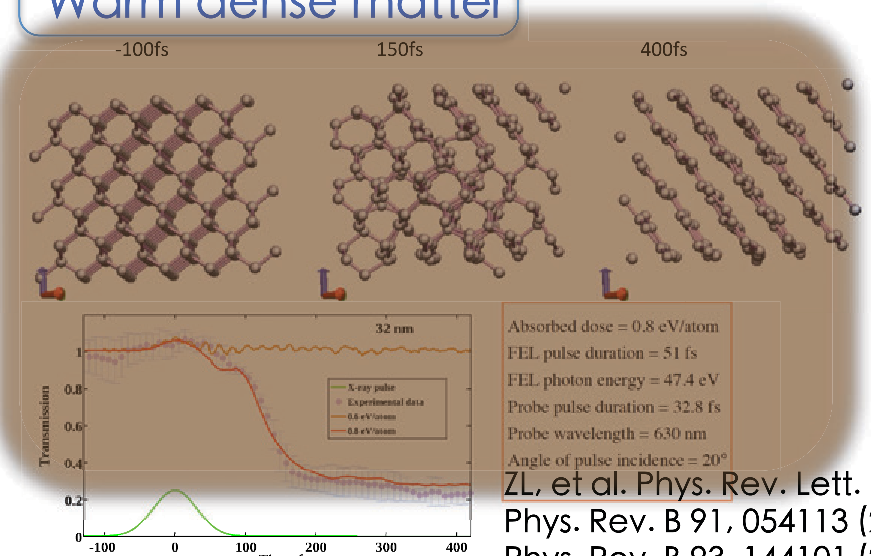


ZL et al. Nature Commun. 8, 453 (2017)



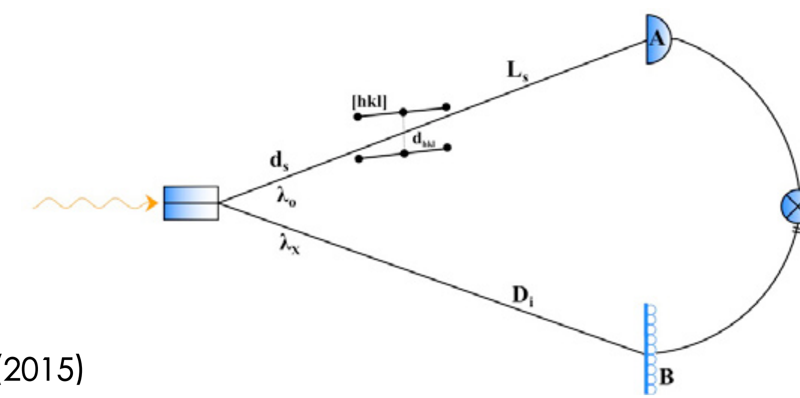
M. Ilchen,⋯,ZL et al. Phys. Rev. A,95,053423(2017)
ZL, T. Wolf et al., Submitted (2017)

FEL as pump
Warm dense matter



ZL, et al. Phys. Rev. Lett. 115, 143002 (2015)
Phys. Rev. B 91, 054113 (2015)
Phys. Rev. B 93, 144101 (2016)
Phys. Rev. B 95, 014309 (2017)

FEL for diffraction
Quantum diffraction



ZL et al., J. Phys. B in print (2017)
ZL et al., Europhys. Lett. in print (2017)

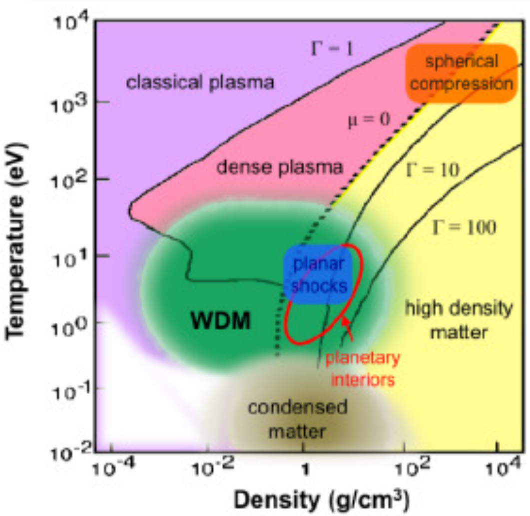
XFEL pump of solid state materials

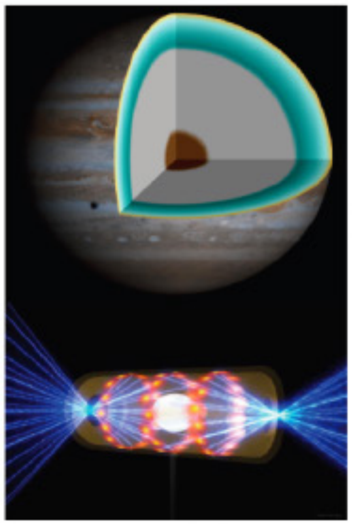
Nikita Medvedev, Victor Tkachenko, Beata Ziaja-Motyka, Ryan Coffee

N. Medvedev, ZL et al.,
Phys. Rev. B 91, 054113 (2015)
Phys. Rev. B 93, 144101 (2016)
Phys. Rev. B 95, 014309 (2017)
Phys. Rev. B 99, 144101 (2019)

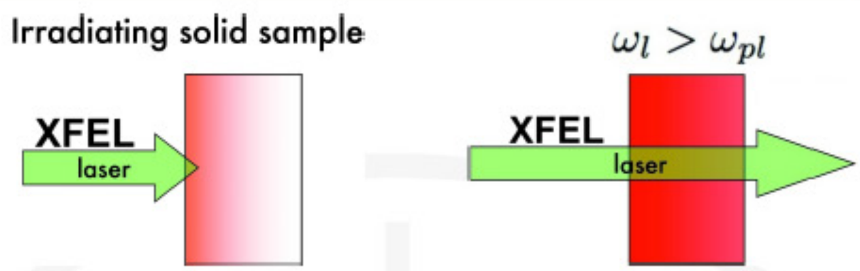
Astrophysics lab on earth:- warm dense matter

Warm $T \simeq 10^4 - 10^5 K$ (T_{Fermi})
Dense $\rho \simeq 10^{22} \text{cm}^{-3}$
$\Gamma = V_{\text{Coulomb}}/E_{\text{kinetic}} \simeq 1$



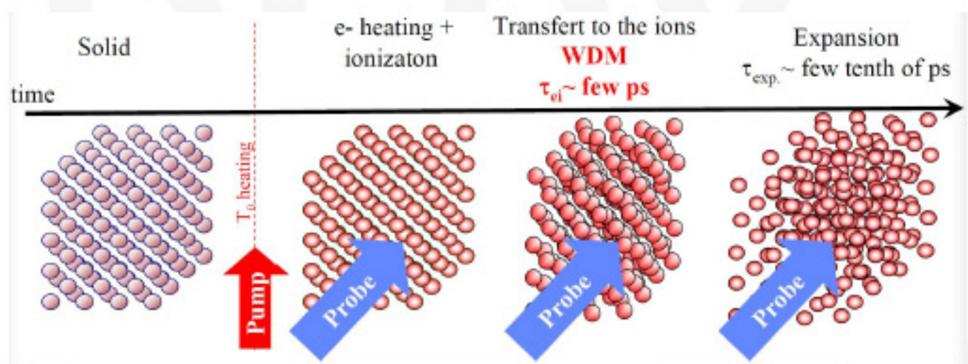


Create Warm Dense Matter with XFEL



- No expansion during heating ⇒ ultrafast pulse ⇒dense
- Warm in temperature ($10^5 K$) ⇒ high intensity ⇒warm

Pump–Probe Spectroscopy of WDM



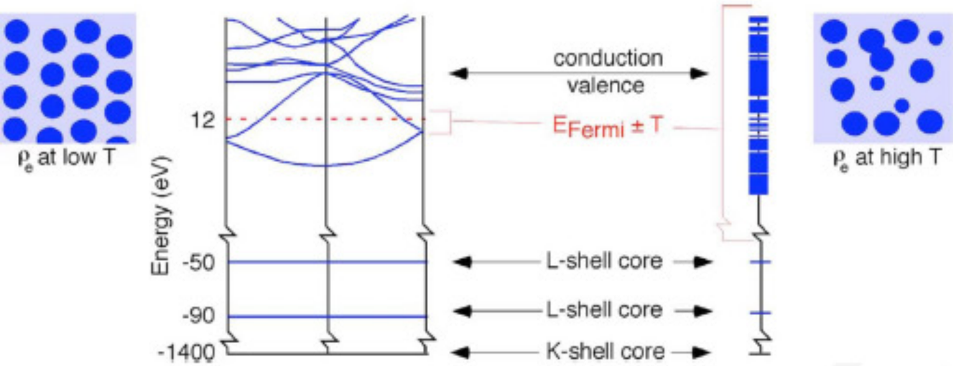
Warm dense matter occurs in

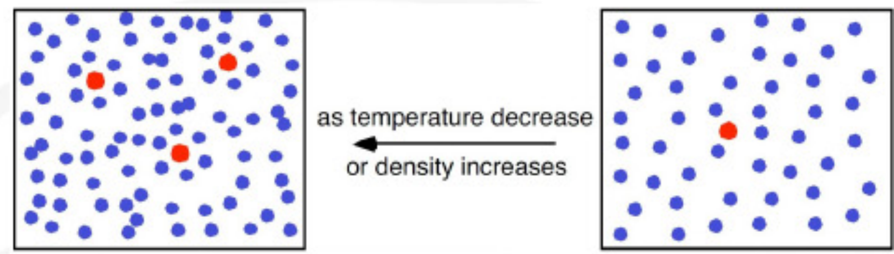
- Cores of giant planets
- Shock-compressed solids
- Inertial confined fusion · · ·

Warm dense matter:- between solid and plasma

WDM - Regime between **cold condensed matter** / **hot plasma**

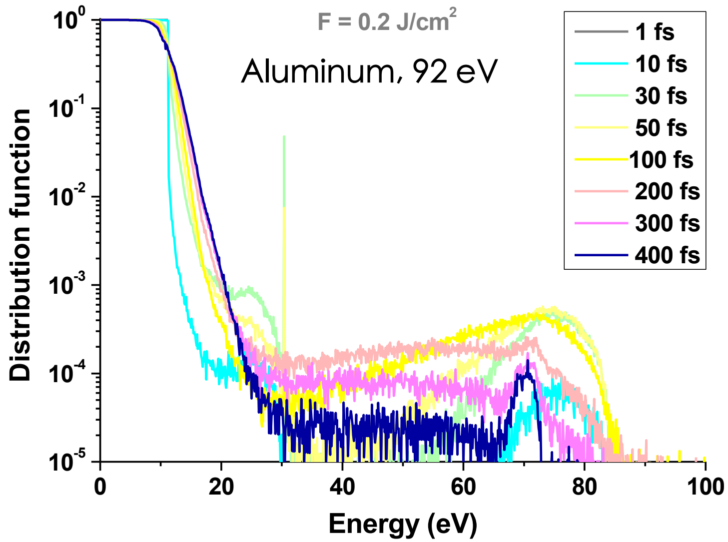
Cold condensed matter $\Longrightarrow$ **WDM** | **WDM** $\Longleftarrow$ Hot plasma



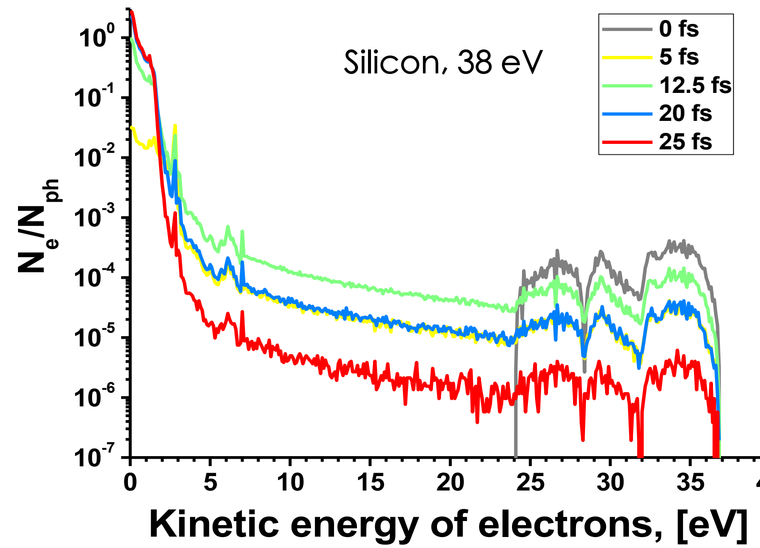


- Fermi energy is the max e^- energy level in cold condensed matter
- $T \ll E_{Fermi}$ (Aluminum, $E_{Fermi} = 11.7$ eV, 1.4×10^5 K) standard cold condensed matter theories work.
- $T \simeq E_{Fermi}$ Warm dense matter ■ Huge and extremely excited molecules (warm dense *superionic* **water** in cores of Neptune and Uranus)
- $T \gg E_{Fermi}$ hot plasma are weakly coupled, classical point particles

Really high excited states:- keV above GS

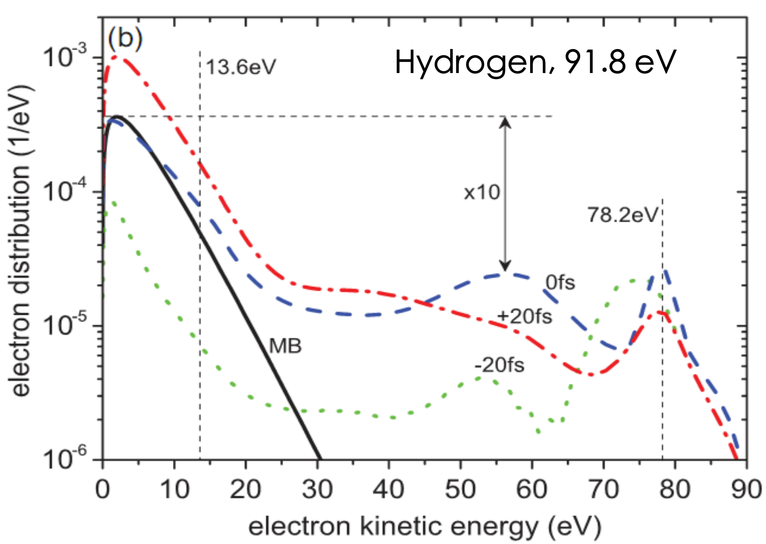


N. Medvedev *et al.*, PRL 107 (2011)

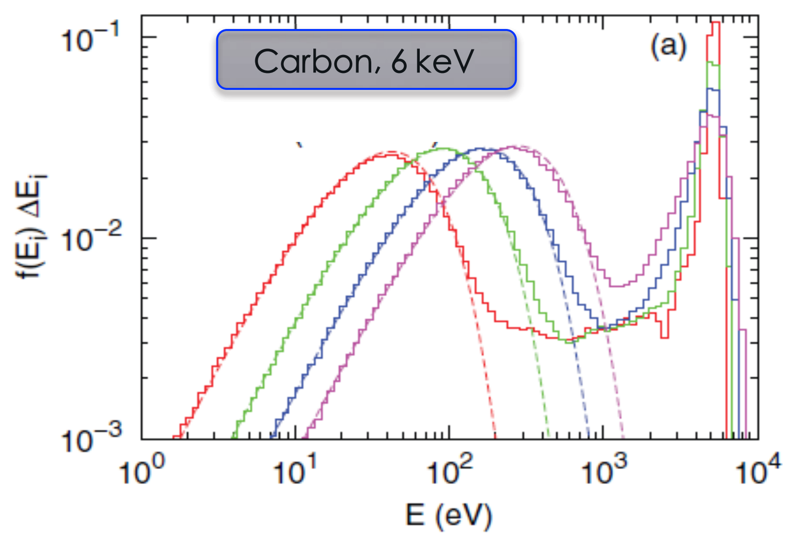


N. Medvedev, B. Rethfeld, NJP 12 (2010)

R.R. Faustlin, B. Ziaja *et al.*, PRL 104 (2010)



S. P. Hau-Riege, PRE 87 (2013)



Challenges of simulating warm dense matter

Warm Dense Matter

is not solid

Conventional energy band theory insufficient

Too high excitation
Too many excited electrons

is not plasma

Conventional plasma theory insufficient

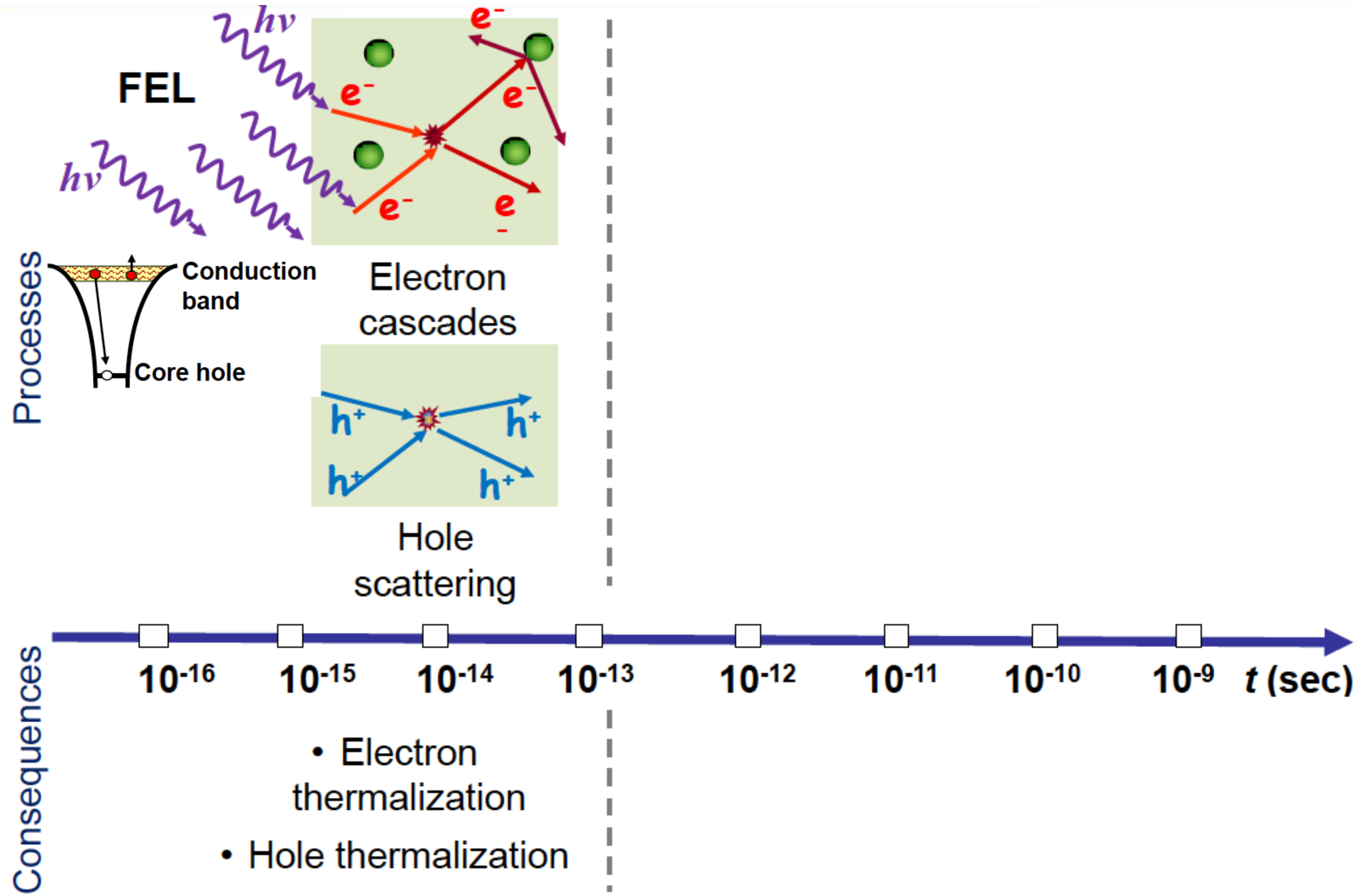
Low energy electrons close to Fermi level
Dispersion different from free particles

A theory that is able to describe

Energy band like dispersion of low energy electrons around Fermi level

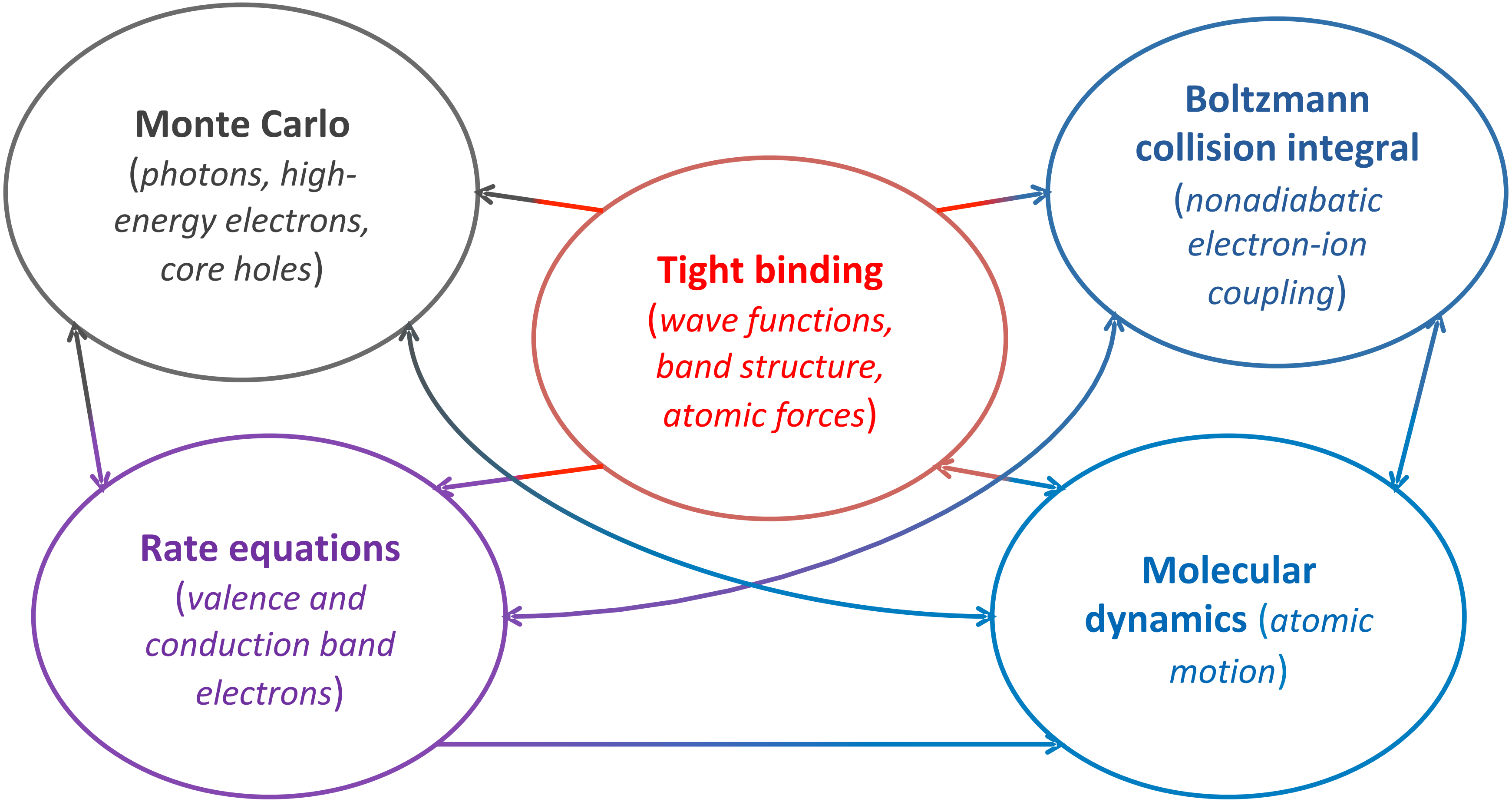
Kinetic collision/recombination of quasi-free high energy electrons

Ab initio simulation:- X-ray creation of WDM

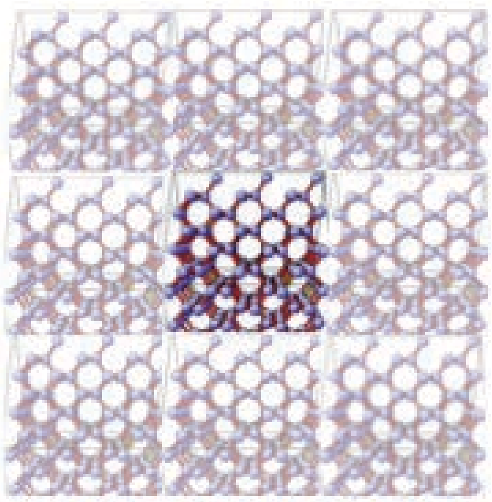


XTANT Model

XTANT: X-ray-induced Thermal And Nonthermal Transitions



Parrinello MD in a supercell

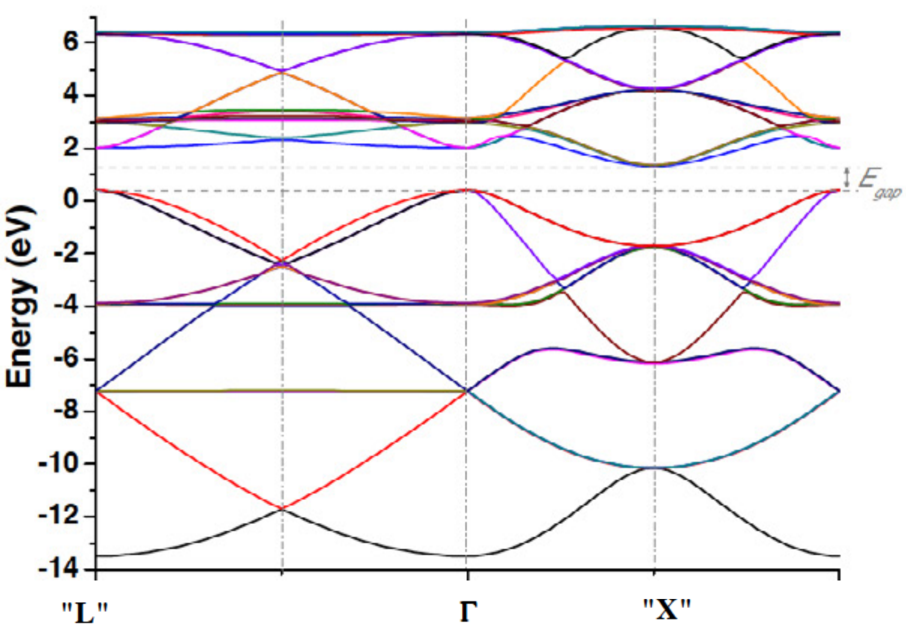


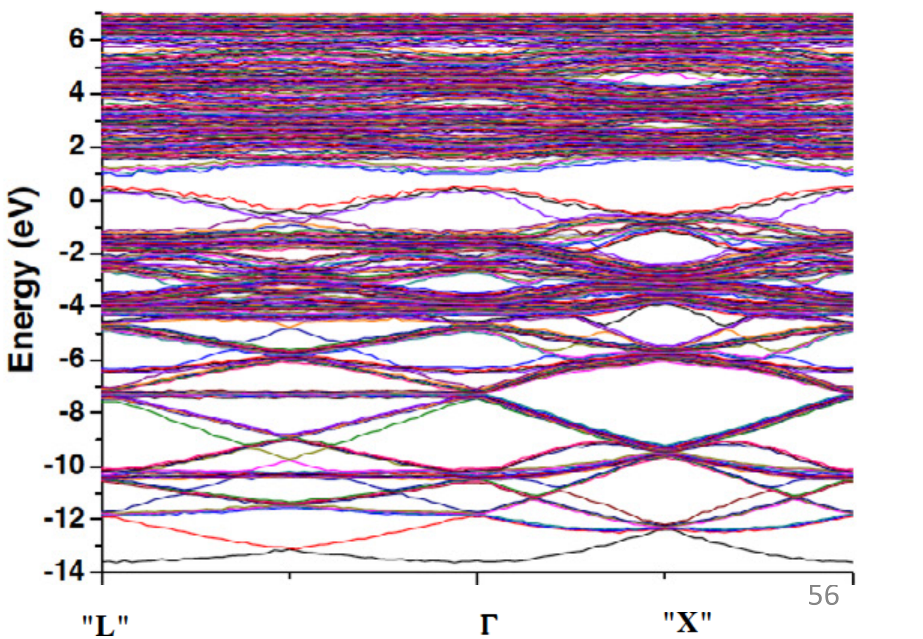
$$L = \sum_{i=1}^{N} \frac{M_i}{2} \dot{\mathbf{s}}_i^{\mathrm{T}} h^{\mathrm{T}} h \, \dot{\mathbf{s}}_i - \Phi(\{r_{ij}\}, t) + \frac{W_{\mathrm{PR}}}{2} \mathrm{Tr}(\dot{h}^{\mathrm{T}} \dot{h}) - P_{\mathrm{exp}} \Omega,$$

$$\mathbf{r}_i = h(\mathbf{s}_i + \mathbf{z}), \;\; z_\alpha \in \{-1, 0, 1\},$$

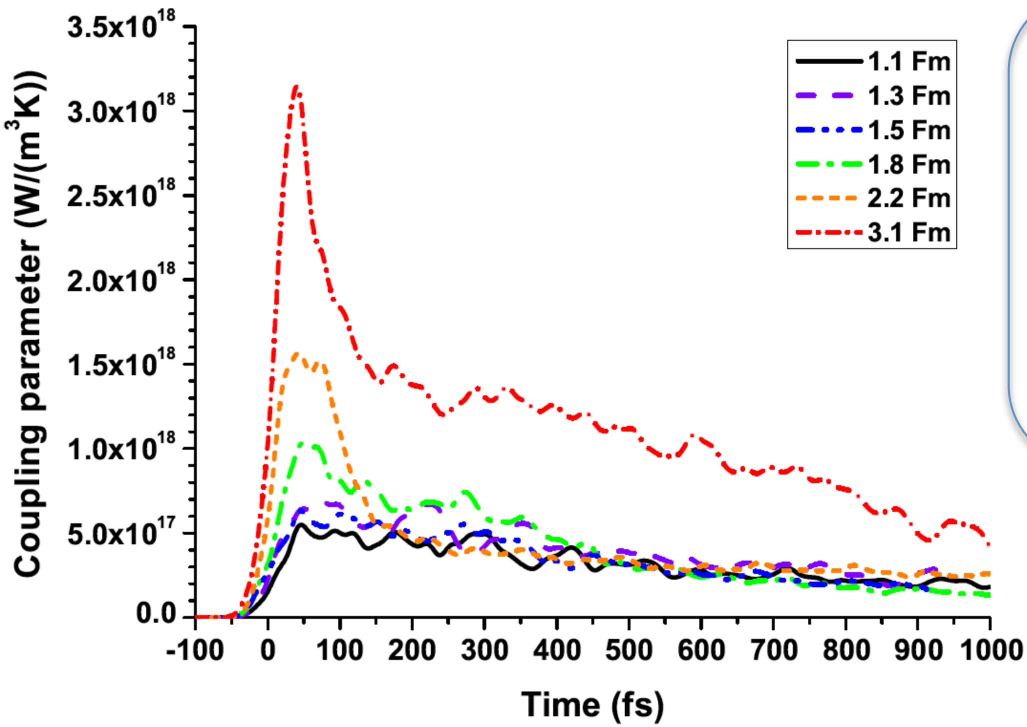
$$\ddot{\mathbf{s}}_i = -\frac{1}{M_i} g^{-1} \frac{\partial \Phi}{\partial s_i} - g^{-1} \dot{g} \, \dot{\mathbf{s}}_i, \quad i = 1, \ldots, N,$$

$$\ddot{h}_{\alpha\beta} = \frac{1}{W_{\mathrm{PR}}} \left(\sum_{i=1}^{N} M_i \, \dot{\mathbf{s}}_i^{\mathrm{T}} (h \dot{\mathbf{s}}_i) - P_{\mathrm{ext}} \sigma - \frac{\partial \Phi}{\partial h_{\alpha\beta}} \right)$$





Electron-ion coupling in WDM:- a puzzle



Among

"outstanding questions of warm dense matter physics"

Experiments have disagreement with theoretical predictions by 10^2-10^3.

T. White *et al.*, Phys. Rev. Lett. (2014)

$$C_e \frac{\partial T_e}{\partial t} = \nabla(K_e \nabla T_e) - g(T_e - T_i) + S_e \delta(t),$$

$$C_i \frac{\partial T_i}{\partial t} = \nabla(K_i \nabla T_i) + g(T_e - T_i),$$

Electron-ion coupling parameter $g \rightarrow g(t)$

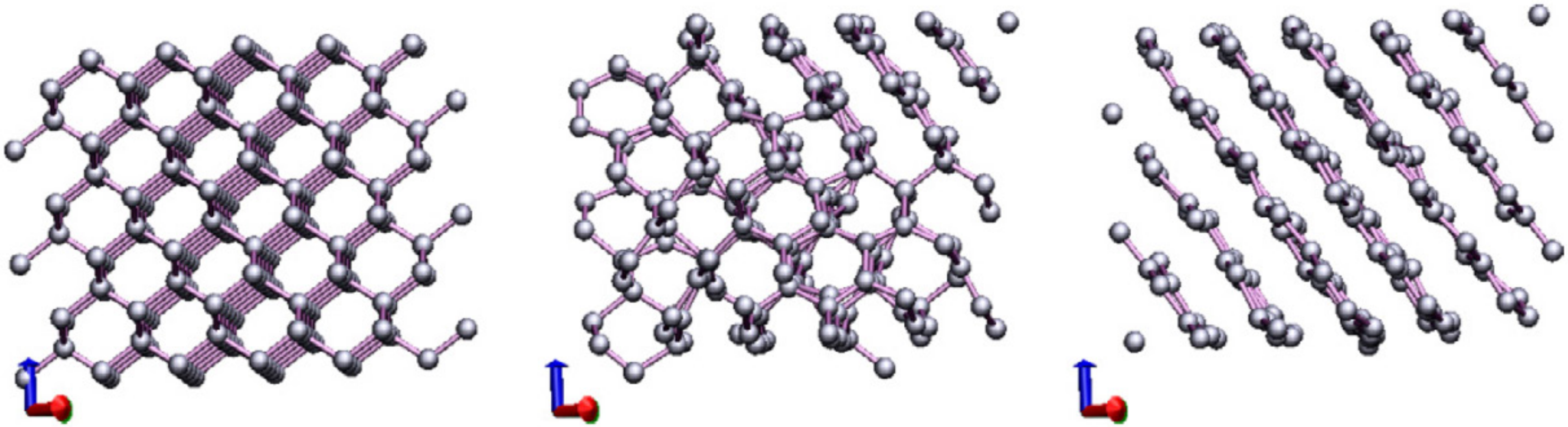
- g is not a constant in time (WDM is crystal-like).
- Phonon picture breaks down (WDM is plasma-like).
- g(t) can be well calculated now.

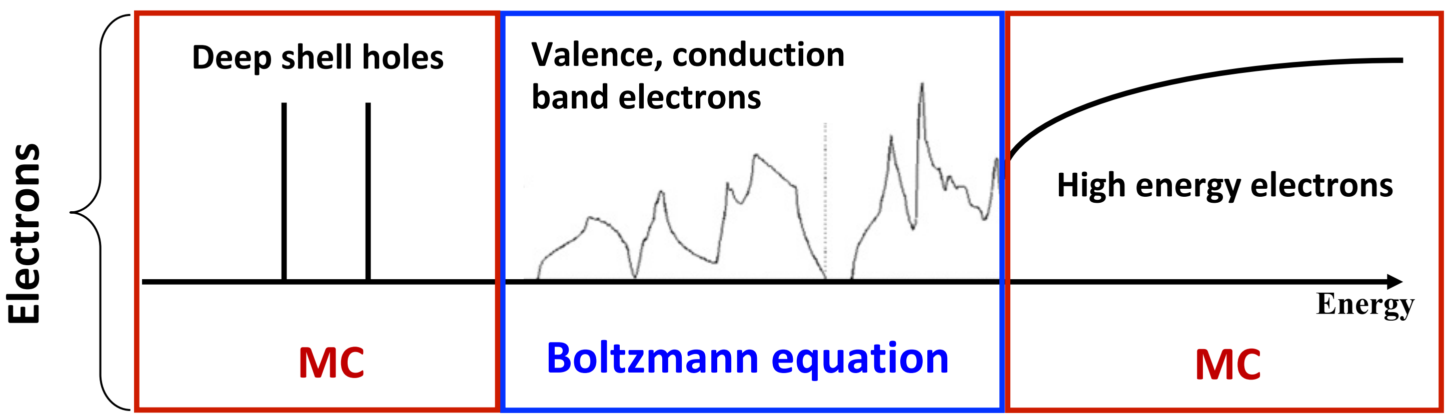
N. Medvedev, ZL, V. Tkachenko and B. Ziaja, Phys. Rev. B 95, 014309 (2017)

Simulation of X-ray damage to solids

Accurate prediction of phase transition @ high X-ray dose

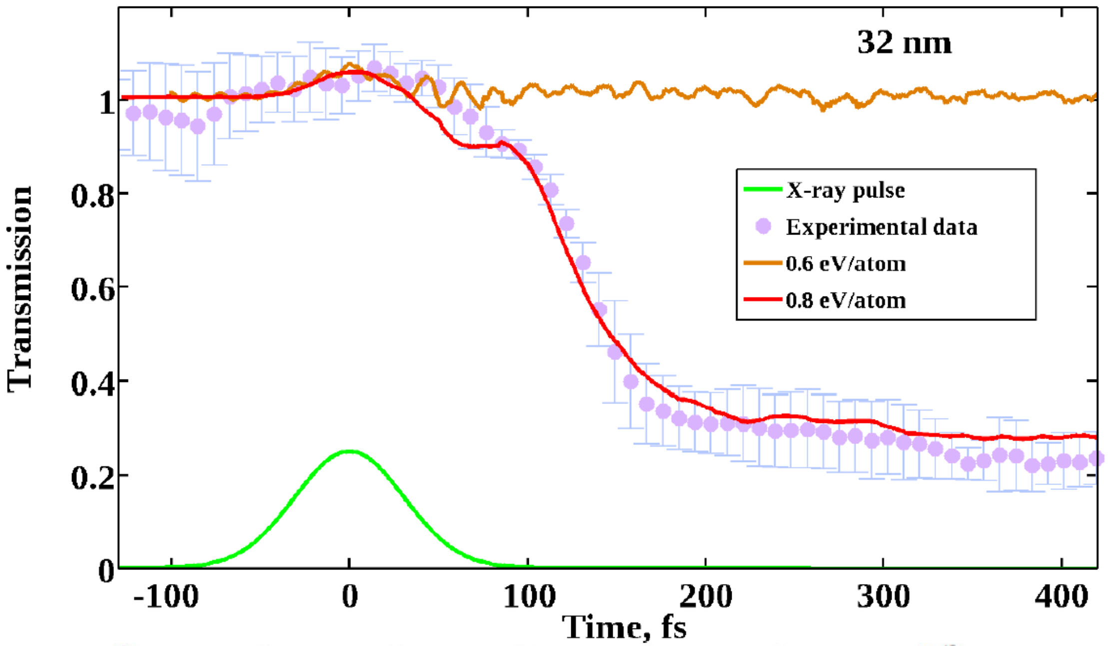
-100fs 150fs 400fs





Optical characterization of phase transition

$$\epsilon^{\alpha\beta}(E) = \delta_{\alpha,\beta} + \frac{4\pi e^2\hbar^2}{m\Omega}\sum_{n,n'}(\eta_{n'}-\eta_n)\frac{F^{\alpha\beta}_{n,n'}}{E_{n,n'}}\left[\frac{1}{E-E_{n,n'}+i\gamma}\right]$$



Absorbed dose = 0.8 eV/atom

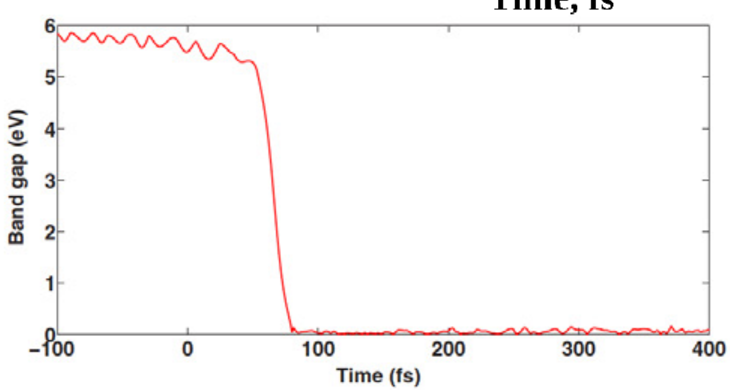
FEL pulse duration = 51 fs

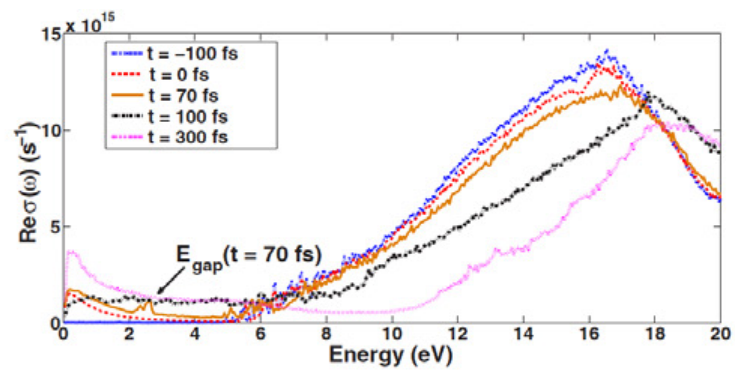
FEL photon energy = 47.4 eV

Probe pulse duration = 32.8 fs

Probe wavelength = 630 nm

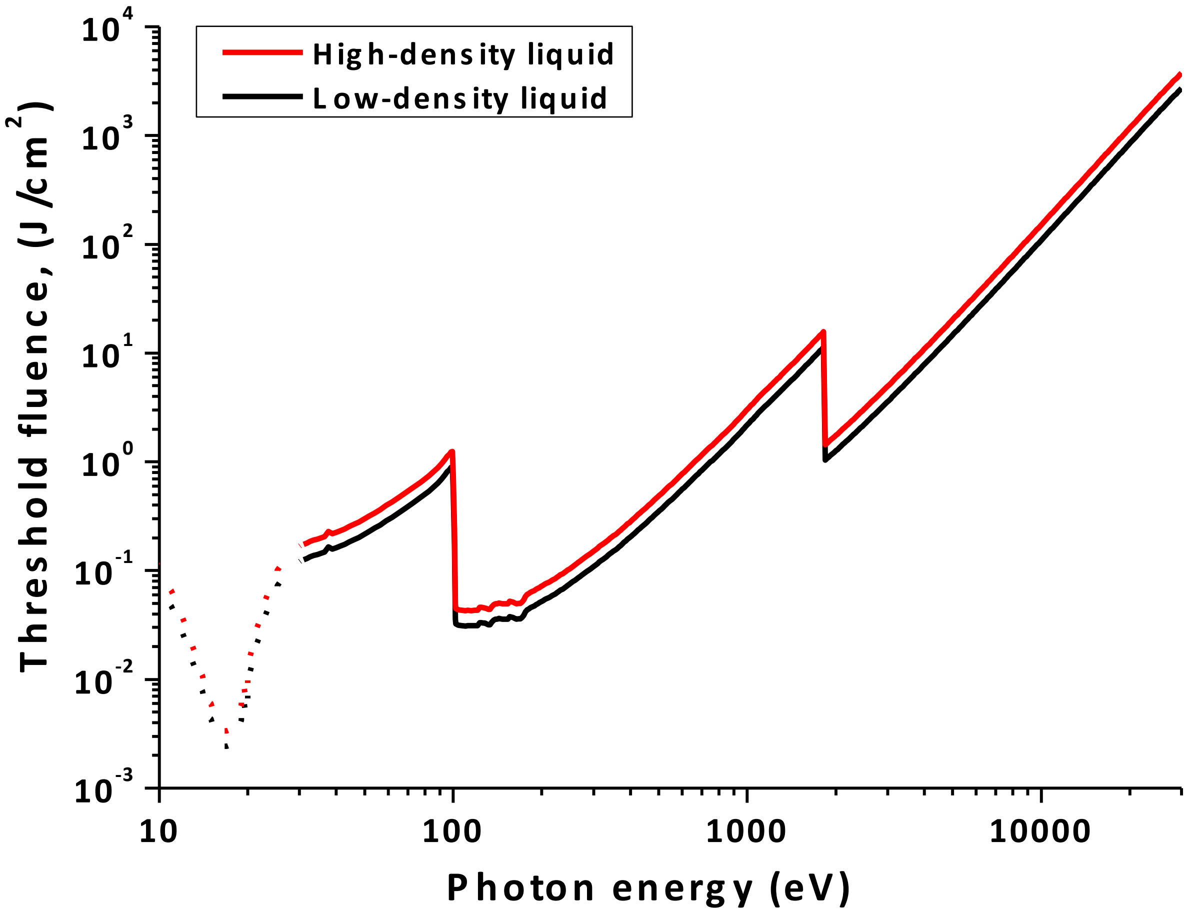
Angle of pulse incidence = 20°





V. Tkacheko, N. Medvedev, ZL, P. Piekarz, B. Ziaja, Phys. Rev. B 93, 144101 (2016)

Application to semiconductor detector engineering



Silicon damaging threshold in terms of incoming photon fluence

corresponding to absorbed dose of 0.6 and 0.9 eV/atom

N. Medvedev, ZL, B. Ziaja, Phys. Rev. B 91, 054113 (2015)

Academic Collaboration

Theory

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Prof. Jochen Küpper

Aarhus University
Prof. Oriol Vendrell
(PhD supervisor,
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Stanford University
Prof. Todd Martinez
(PostDoc supervisor)

University of Maryland, Baltimore County
Prof. Yanhua Shih

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Max-Planck Institute
Prof. Dwayne Miller, FRSC
(PostDoc supervisor)

SLAC
Prof. Phil Bucksbaum
Dr. Thomas Wolf
Dr. Andreas Kaldun

University of California, Berkeley
Prof. Stephen Leone

Funding

VolkswagenStiftung

Theory and Experiment

Theory is when you know everything but nothing works.

Experiment is when everything works but no one knows why.

In our lab rules the perfect combination of theory and experiment:

Nothing works and no one knows why.

We hope and work for the opposite!

Thank you very much

DESY

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