New Horizon of Accelerator based Molecular Physics

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MAX-PLANCK-GESELLSCHAFT

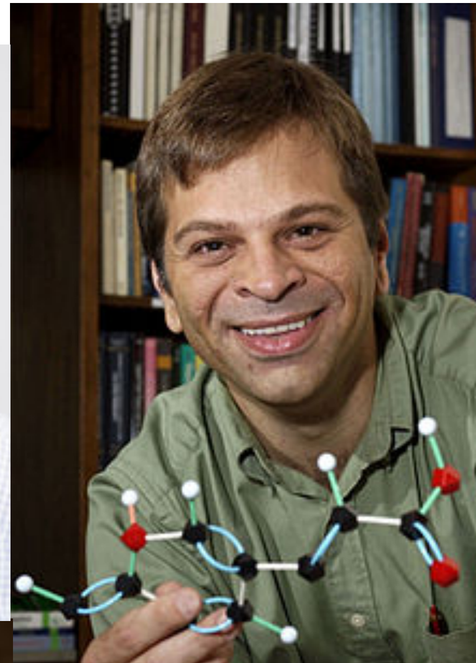
DESY

SLAC NATIONAL ACCELERATOR LABORATORY

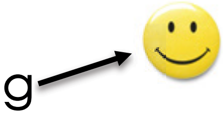
25th March 19, New scientific capabilities at EuXFEL, Hamburg

Dwayne

Todd



Zheng

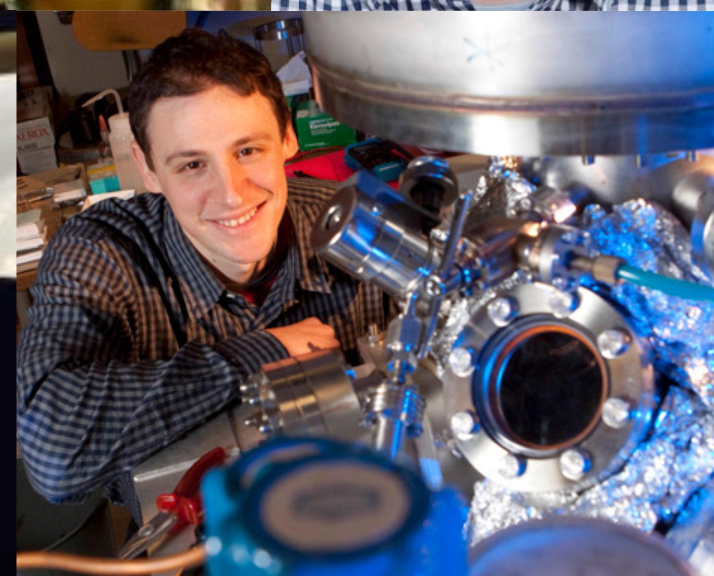


Jie

Ryan

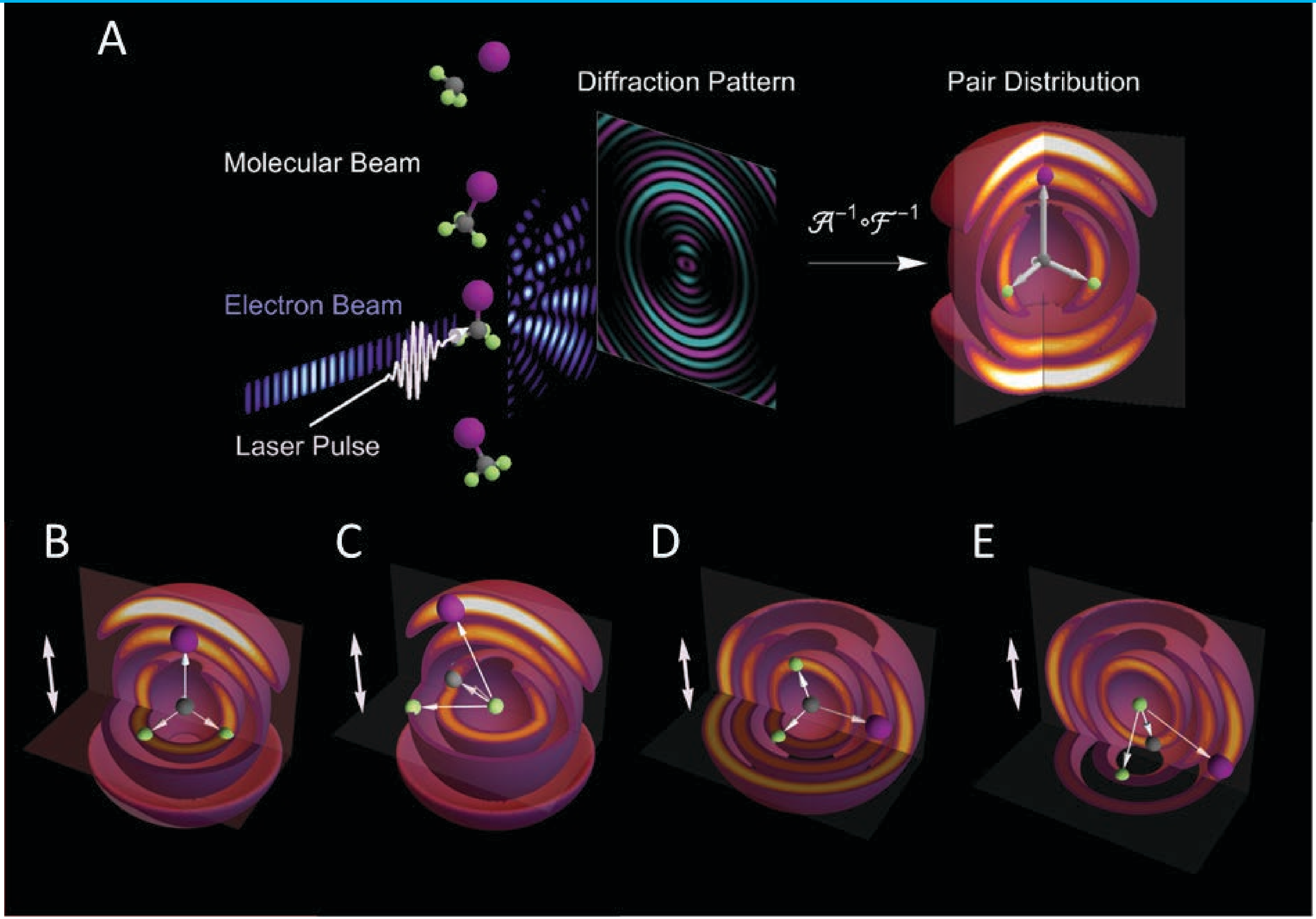
Jim

Xiaolei



Martin

CF_3I Photodissociation (3.7MeV electrons)



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

Quantum tomography

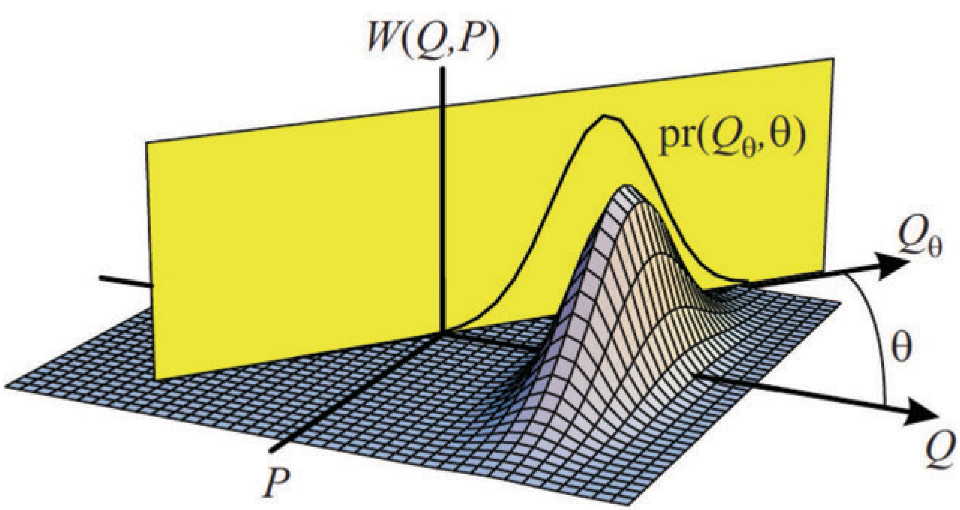
The time in diffraction pattern I(Q;t) does not only offer us the molecular motion in time, but also unveils the complete quantumness of the molecule.

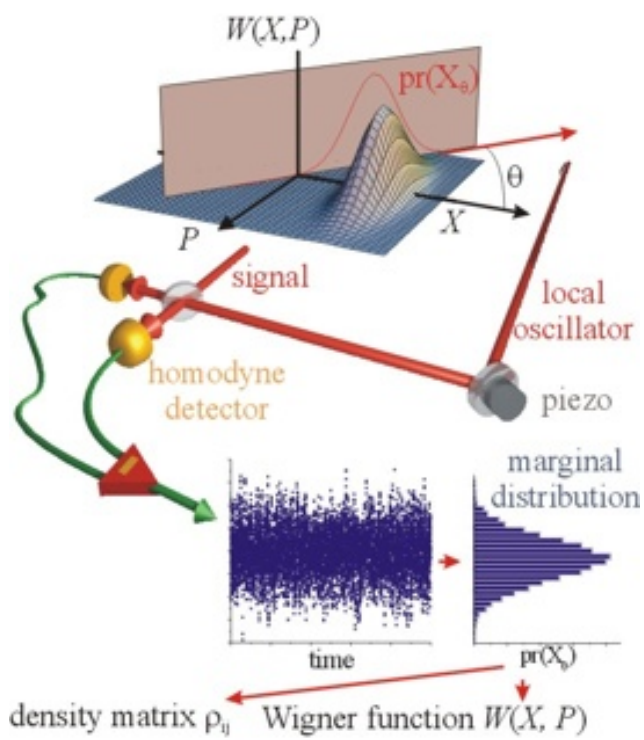
Pauli Problem (Pauli, 1933): could we retrieve Ψ(x) from observable $P(x)=|\Psi(x)|^2$?

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

Quantum tomography from quantum optics to ultrafast diffraction

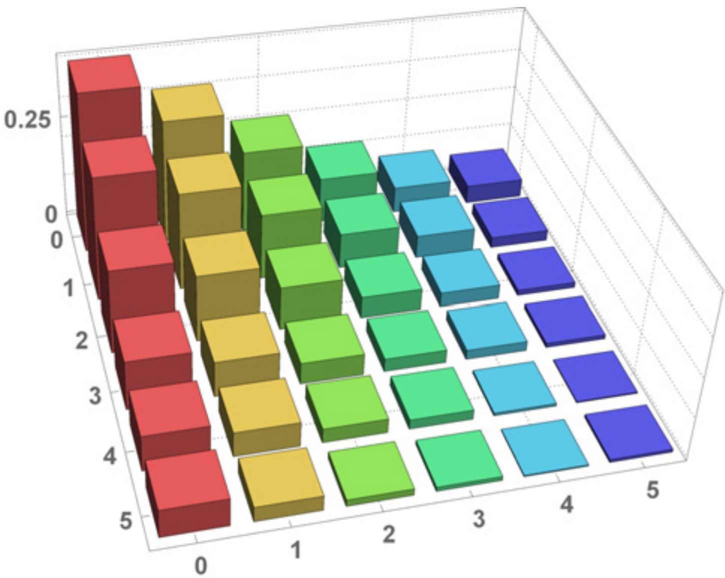
Quantum optics - Homodyne detection
Rev. Mod. Phys. 81, 299 (2009)





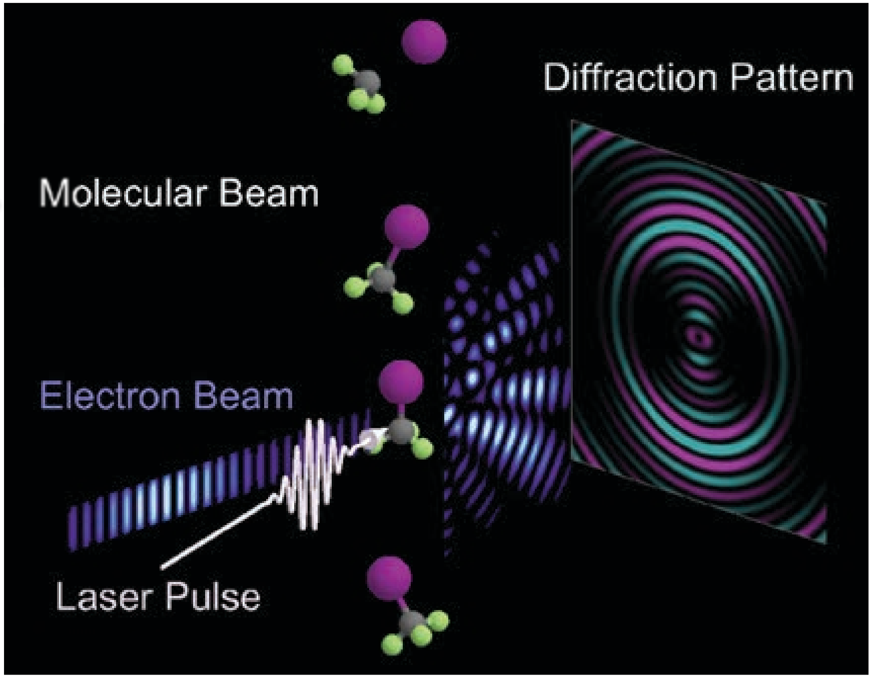
Quantum tomography by TRED diffraction

Density Matrix of coherent wavepacket
vibrational motion



θ≅t

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)

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

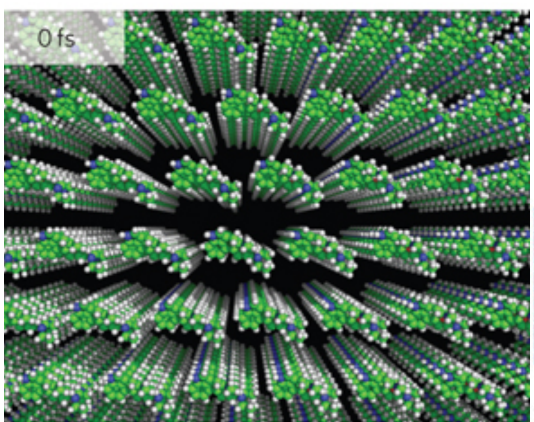
S. Hell *et al.* Opt. Lett. (1994); Opt. Lett. (1999)

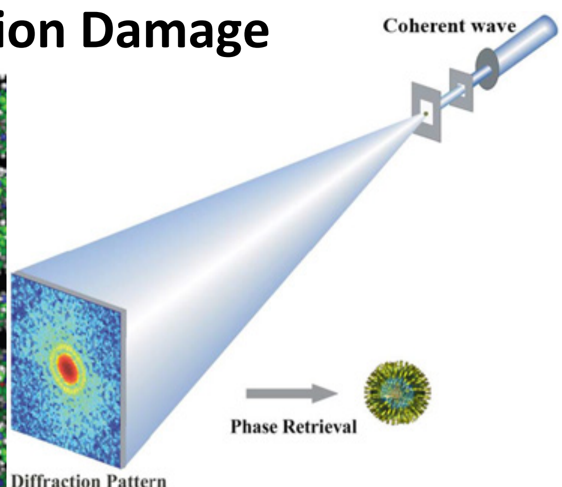
XFEL diffraction

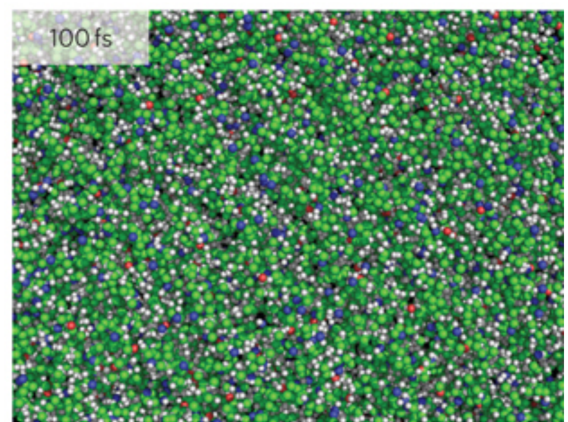
✓ Resolution: ~0.1nm
⊗ Serious radiation damage

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

Culprit for ⊗'s: Radiation Damage



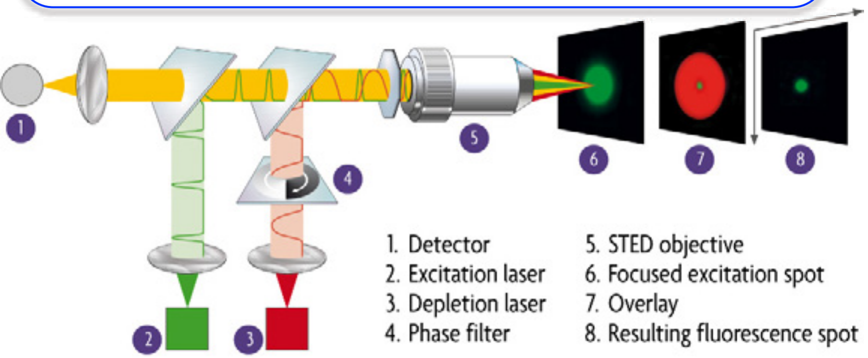




The Ideal Microscope

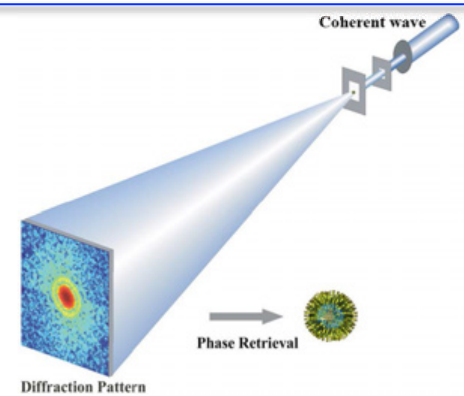
Superresolving microscope：
- ✓ Radiation damage free
- ✗ ~~Resolution: ~20nm~~

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



Active Motif Inc.

+



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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 51, 025503 (2018)
ZL, et al., Europhys. Lett. (EPL) 120, 16003 (2017)

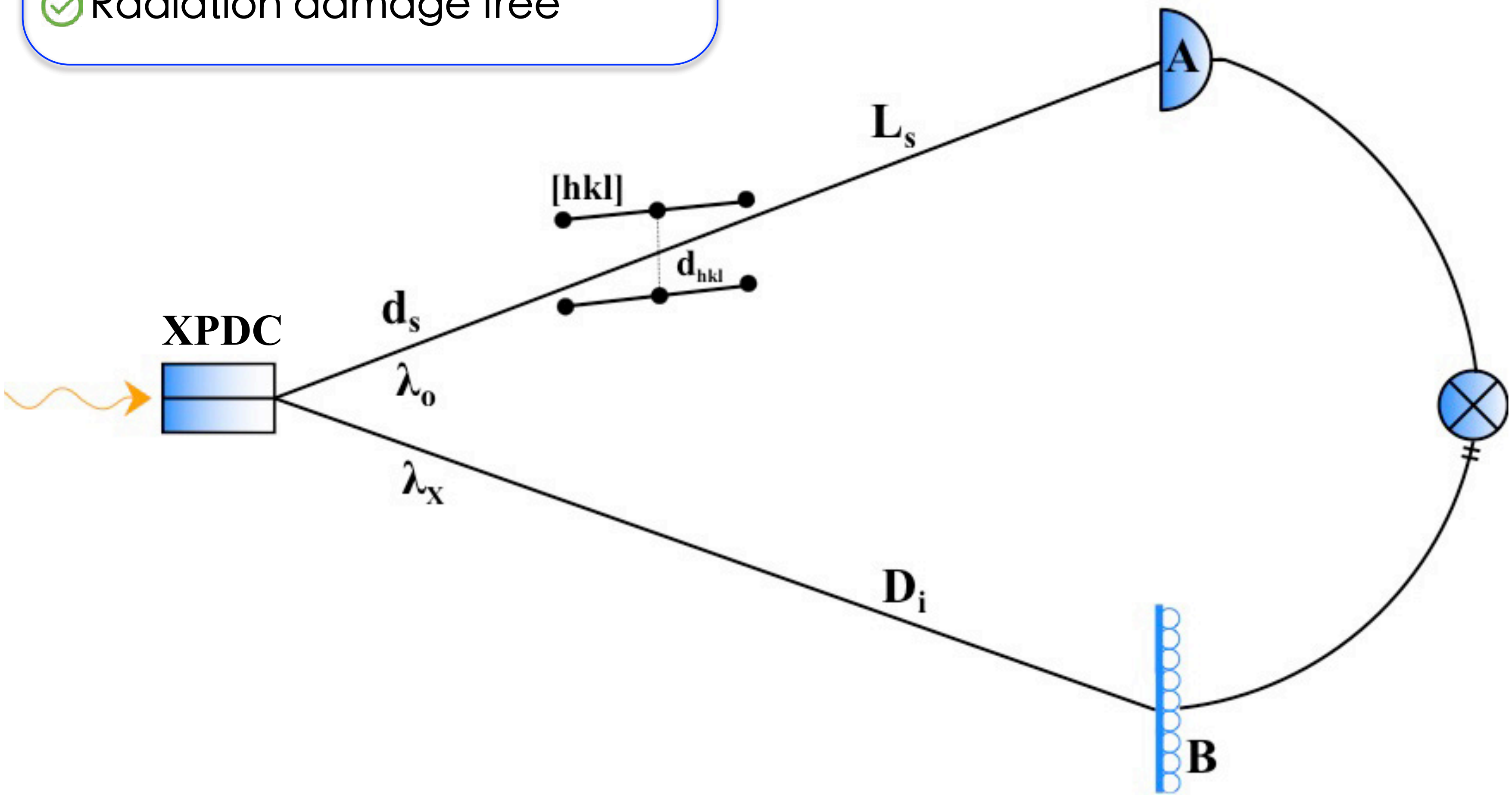
Is there more exotic quantumness we can exploit using FEL?

Referee Comment:
This is an exciting, comprehensive, seminal paper that breaks a significant amount of new scientific ground.

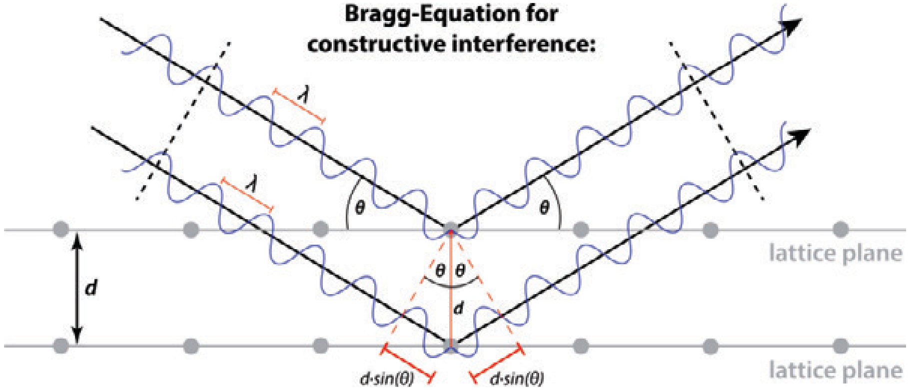
XFEL quantum diffraction

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

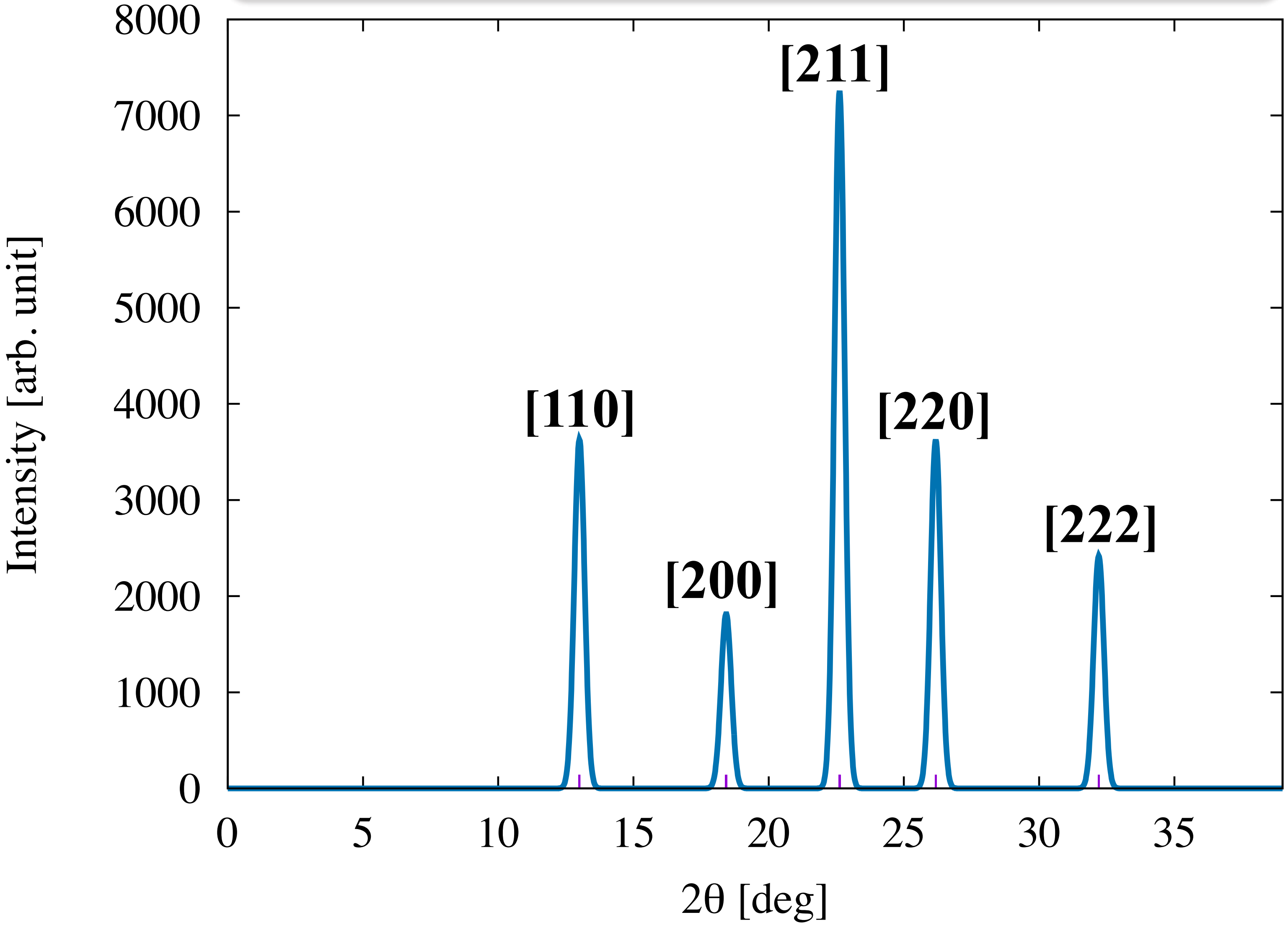
$$R_c(\rho_B) = \frac{1}{T}\int dt_A dt_B S(t_B, t_A)\int_{\sigma_A} d^2\rho_A\, \sigma_B \mathrm{tr}\left[E_A^{(-)}E_B^{(-)}E_B^{(+)}E_A^{(+)}\rho\right]$$

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

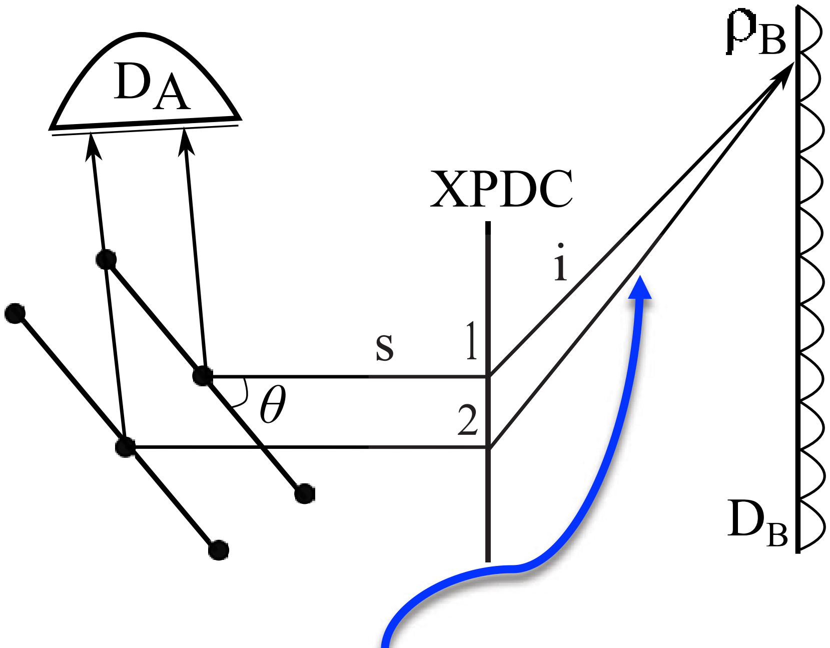
Magnification factor >10^3

Quantum diffraction (3.1eV photon)

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



Physical picture



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

Two-photon diagram

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

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

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

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

Due to entanglement, we have

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

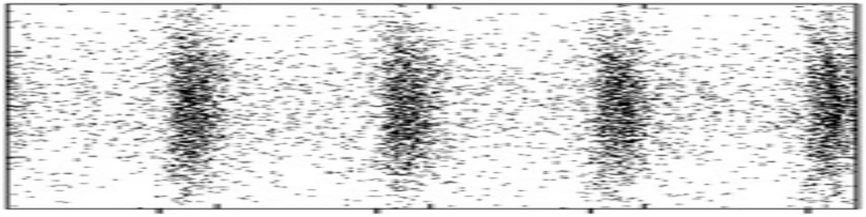
we can thus concacenate optical paths at positions 1 and 2.

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

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

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

Prelude:- The Making of a Molecular Movie

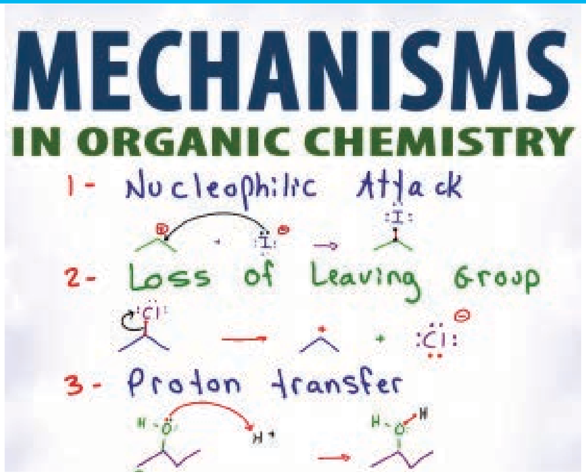


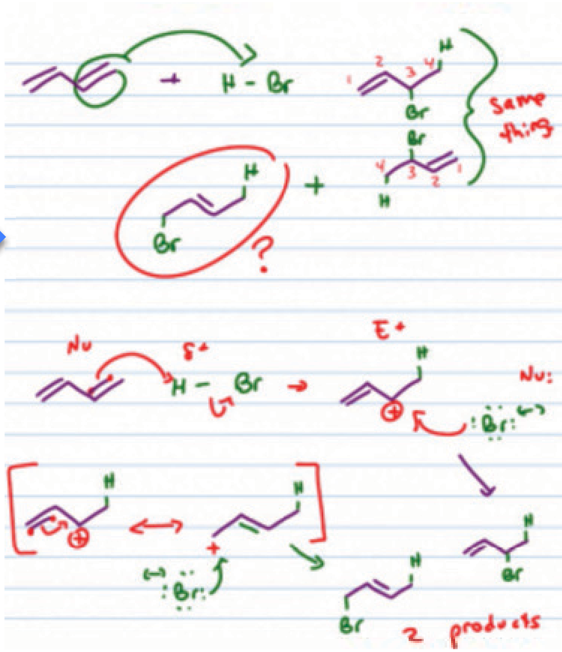
Electron Microscope in time

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

The compressed electron bunches from electron accelerator can be as short as femtoseconds!

We can see the motion pictures of molecule directly!

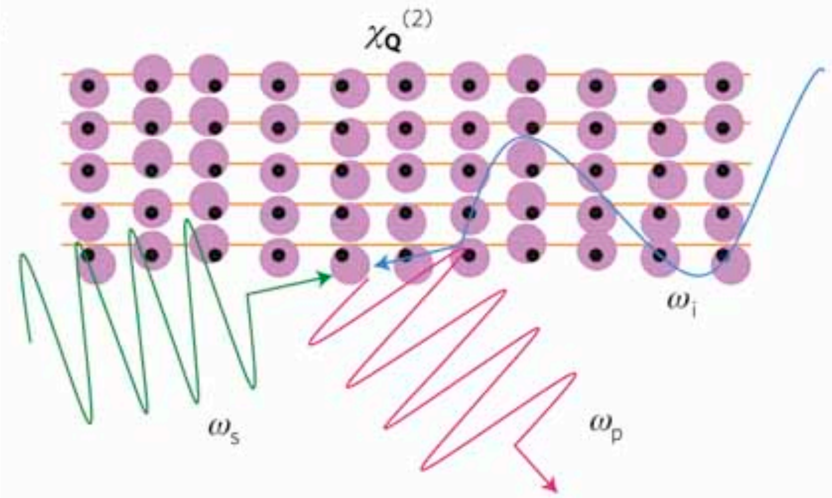




Do we have sufficient such photon pairs?

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

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



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

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

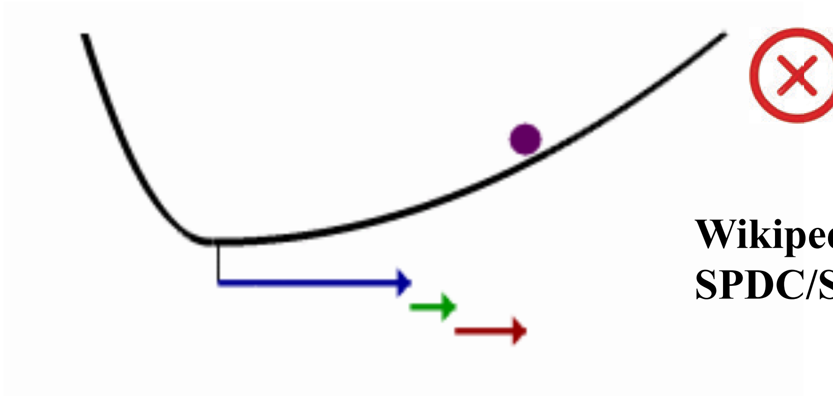
$$\frac{d\sigma}{d\Omega}^{(2)} \sim 1.9\times10^3\,\text{fm}^2 = 19\,\text{b} \quad \text{XPDC}\ \omega_3(=3.1\,\text{keV}) \rightarrow \omega_2(=3096.9\,\text{eV}) + \omega_1(=3.1\,\text{eV})$$

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

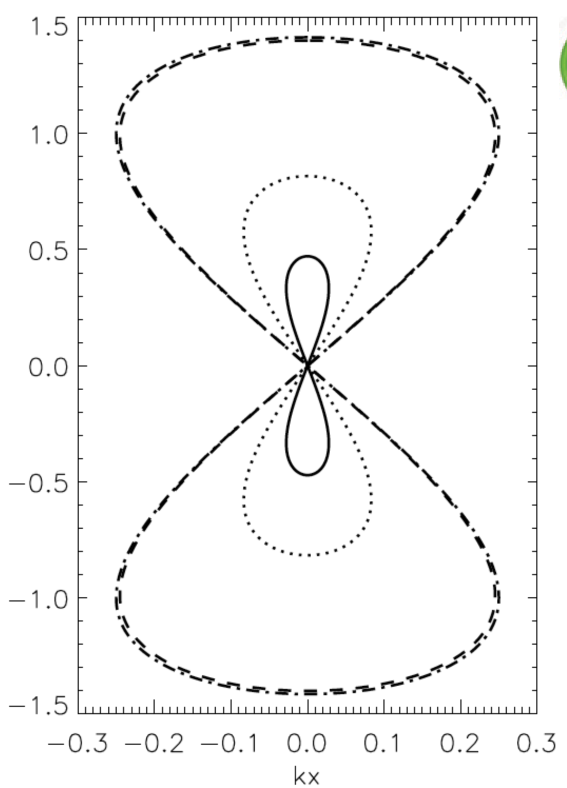
Single electron as nonlinear medium for XPDC

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

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



Wikipedia: SPDC/SHG



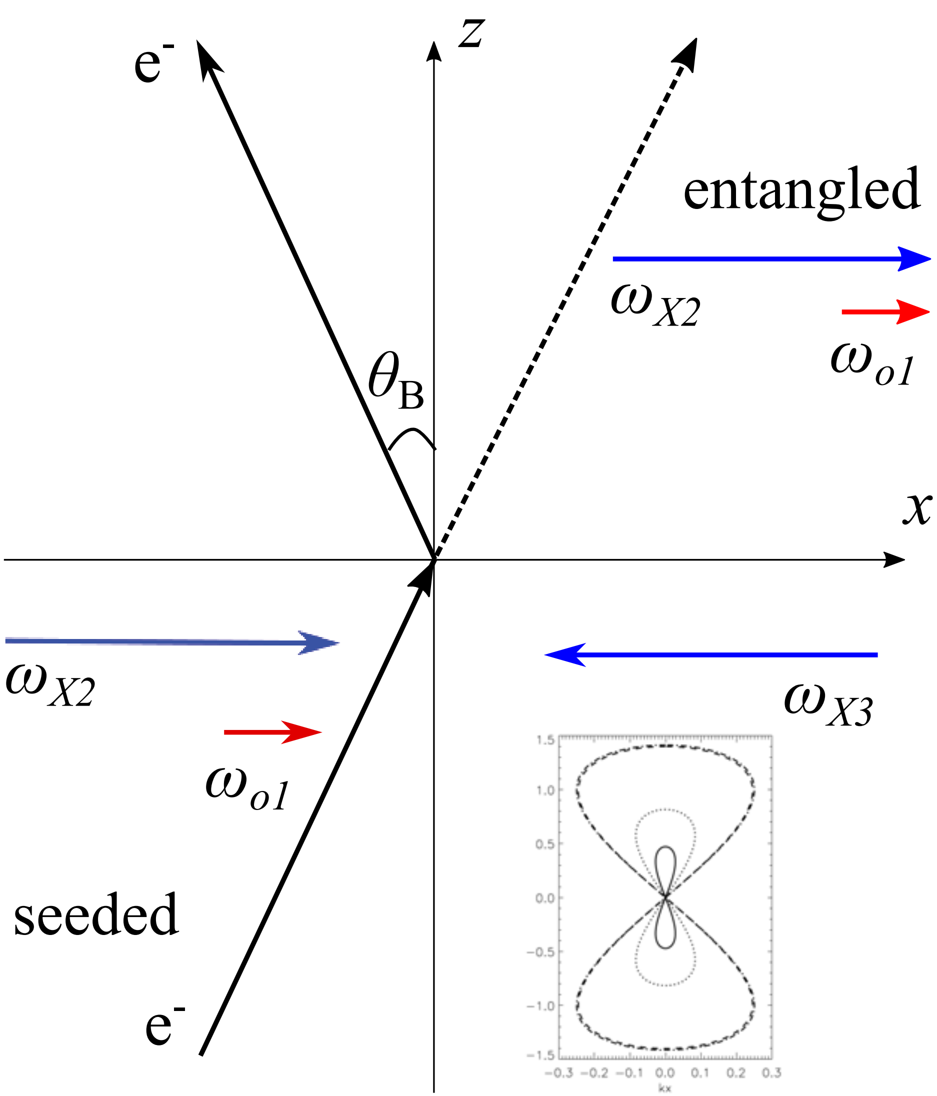
$$x = -\frac{a^2}{8k(1+a^2/2)}\sin 2\phi$$

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

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

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 the probability of entangled pair production P

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

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