Recent progress in x-ray nonlinear optics and future perspectives

Kenji Tamasaku RIKEN/PRESTO

Workshop on Evolution and Control of Complexity
Key Experiments Using Sources of Hard X-rays
@ APS 10/12/2010

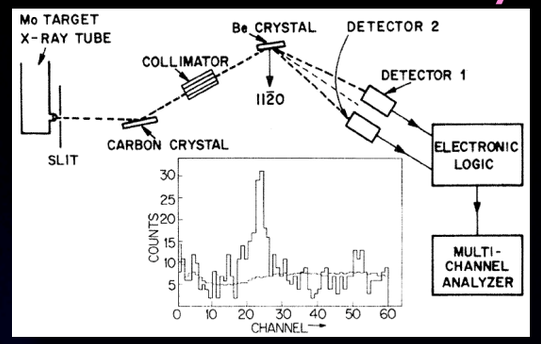
Outline

1. ***X-ray nonlinear optics***
2. Parametric down-conversion of x-rays
3. Nonlinear susceptibility of diamond
4. Future perspectives

XNLO - scince 1969

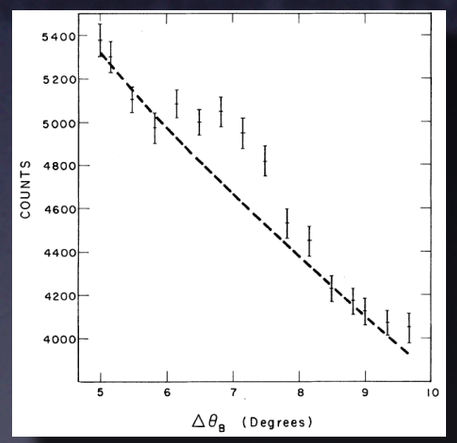
Theory & Experiment

Parametric down-conversion into x-rays



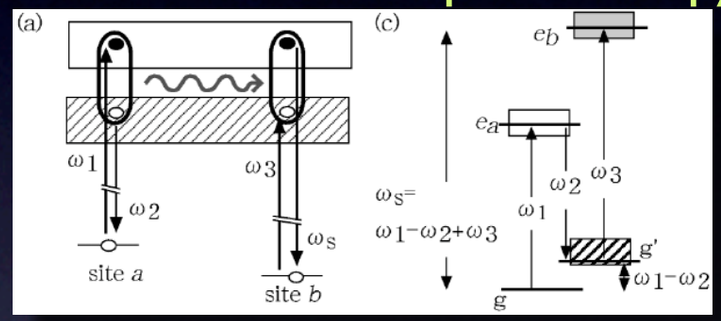
Eisenberger, PRL 1972
(cf. Yoda, JSR 1998,
Adams, JSR 2000)

Parametric down-conversion into EUV



Danino, PRL 1981

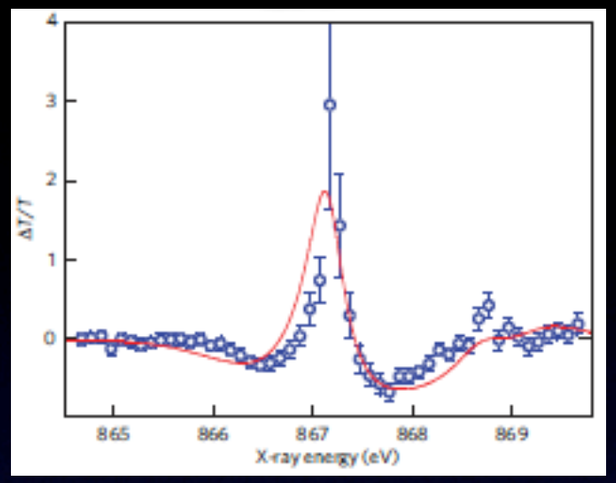
Coherent Raman spectroscopy



Tanaka, PRL 2002

Local probe of
electronic excitation

EIT



Glover, NPhys. 2010
(cf. Buth, PRL 2007)

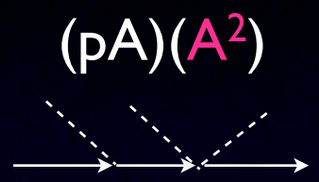
Pulse control of
x-rays

- Observable or not?

- *Scientific interest*
- *Application with XFEL*

Outline

1. X-ray nonlinear optics



2. Parametric down-conversion of x-rays

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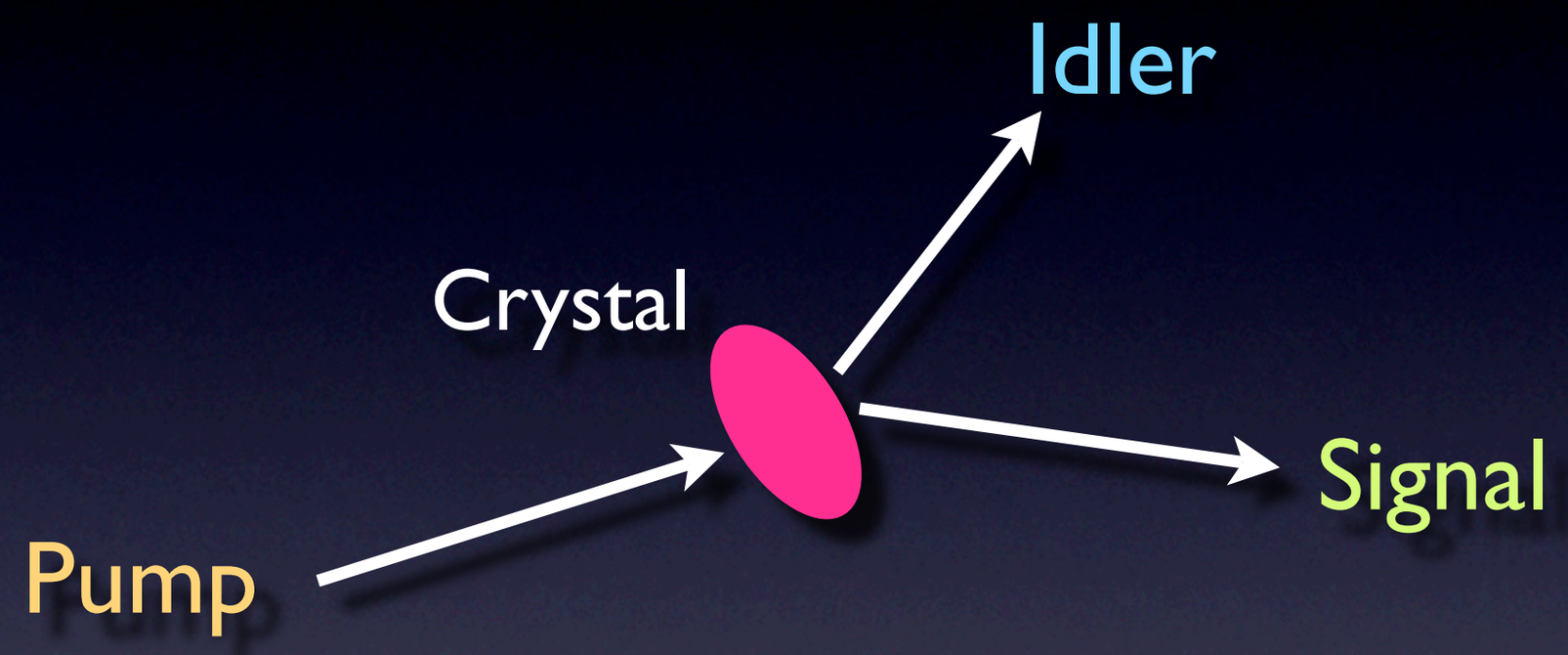
4. Future perspectives

X-ray parametric down-conversion (PDC)

- 2nd order nonlinear process

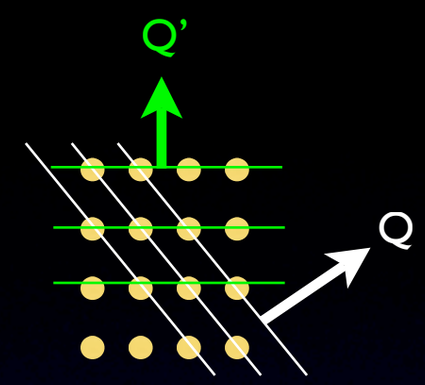


✓ Momentum conservation
✓ Energy conservation

Momentum conservation (phase matching)

Phase matching with reciprocal lattice vector; Q

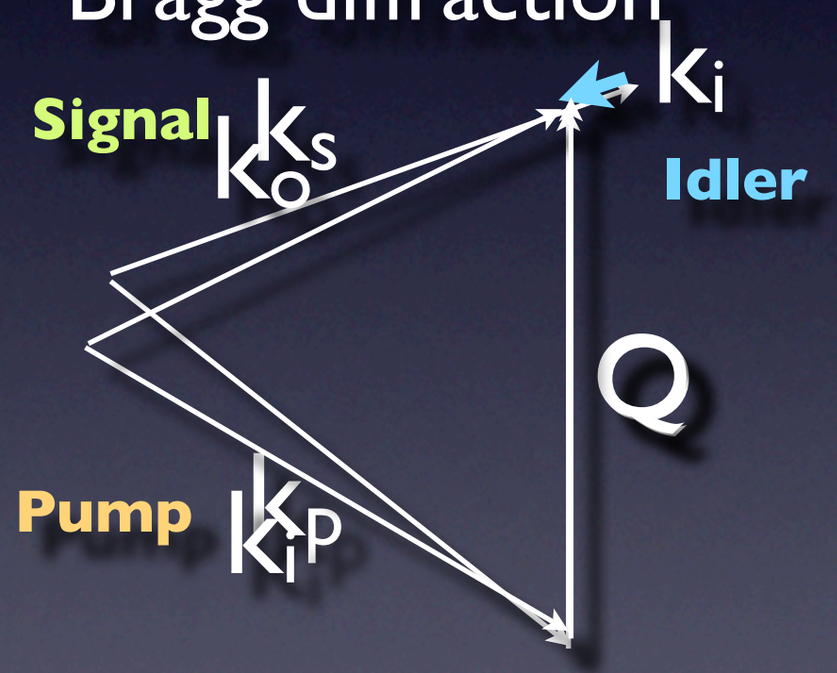
➡ "Nonlinear diffraction"



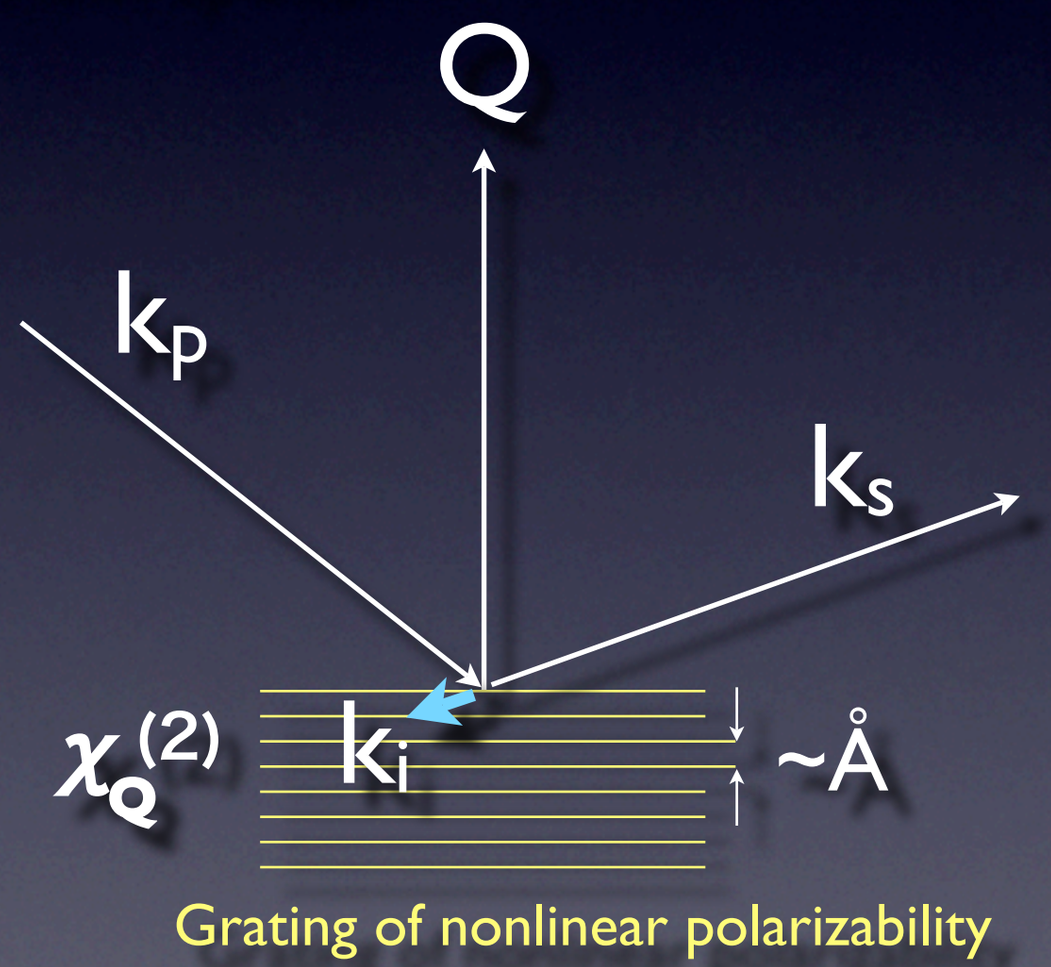
Momentum space

$k_p+Q=k_s+k_i$

Bragg diffraction



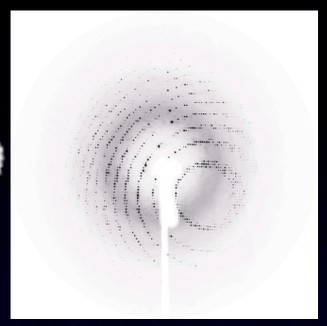
Real space

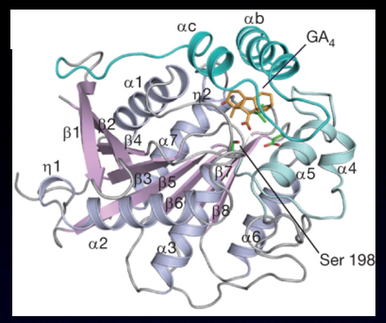


X-ray nonlinear diffraction

Usual (linear) diffraction

structure of χ





Nature **456**, 520 (2008).

Total charge density

Susceptibility

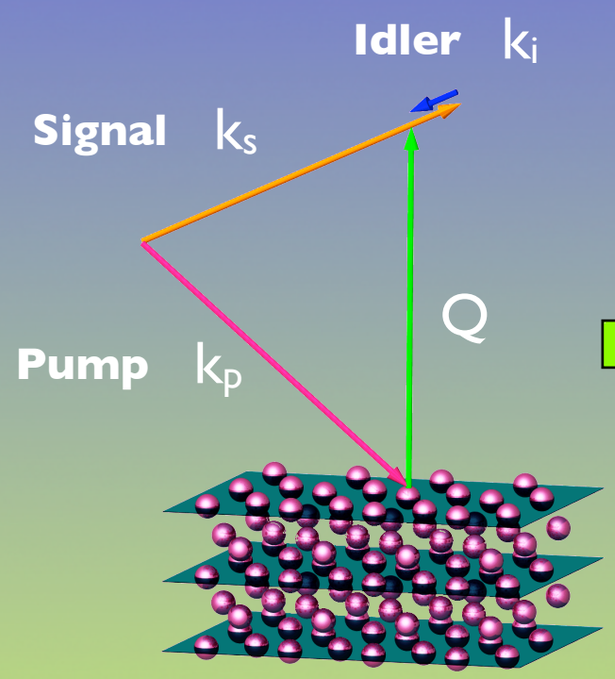
Linear: $P = \chi E$

2nd order NL: $P_p = \chi^{(2)} E_s E_i$

3rd order NL: $P_4 = \chi^{(3)} E_1 E_2 E_3$

Output Input

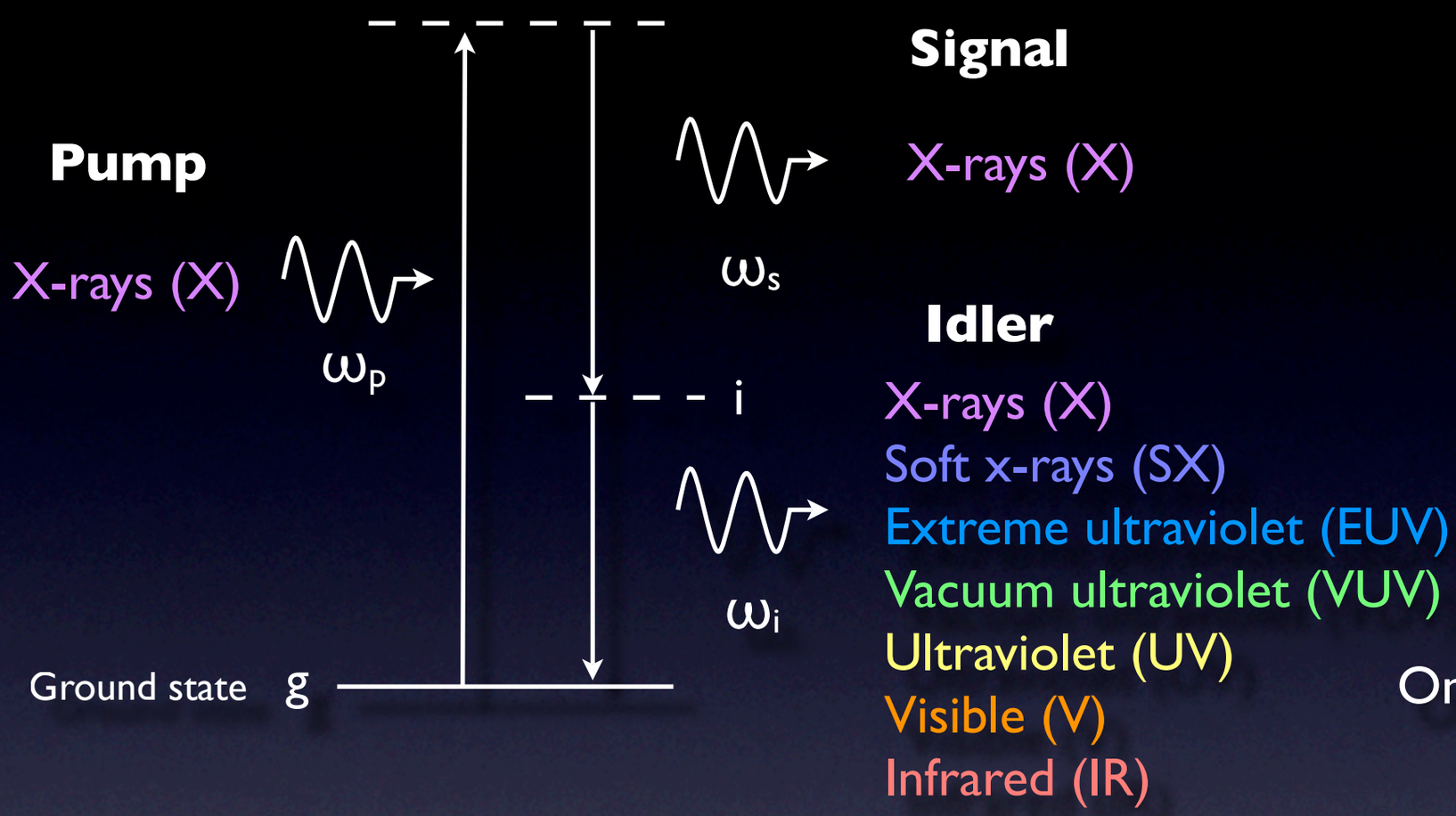
Nonlinear diffraction



structure of $\chi^{(2)}$

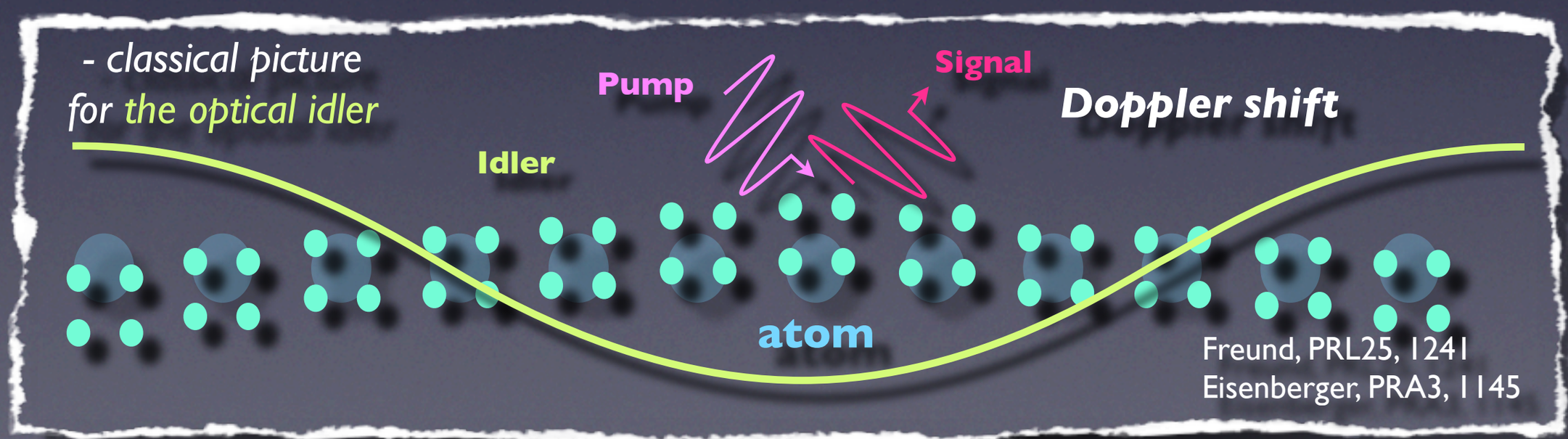
with Å resolution

Energy conservation



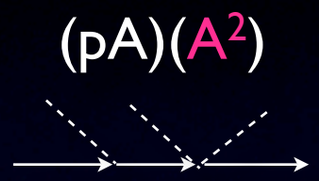
One of matrix elements:

$$\frac{\langle g|\, A^2\, |j\rangle \langle j|\, pA\, |g\rangle}{E_g - E_j - \hbar\omega}$$

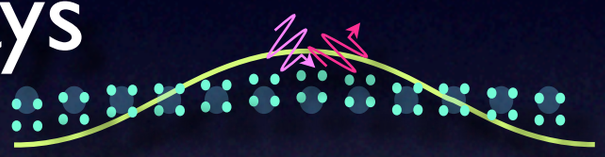


Outline

1. X-ray nonlinear optics



2. Parametric down-conversion of x-rays



3. Nonlinear susceptibility of diamond

4. Future perspectives

Δk-dependence of signal intensity

Diamond, Q=(1,1,1)

X → X + EUV

11.0 keV 10.9 keV 100 eV

Rocking curve of nonlinear diffraction
(Δk-dependence of signal intensity)

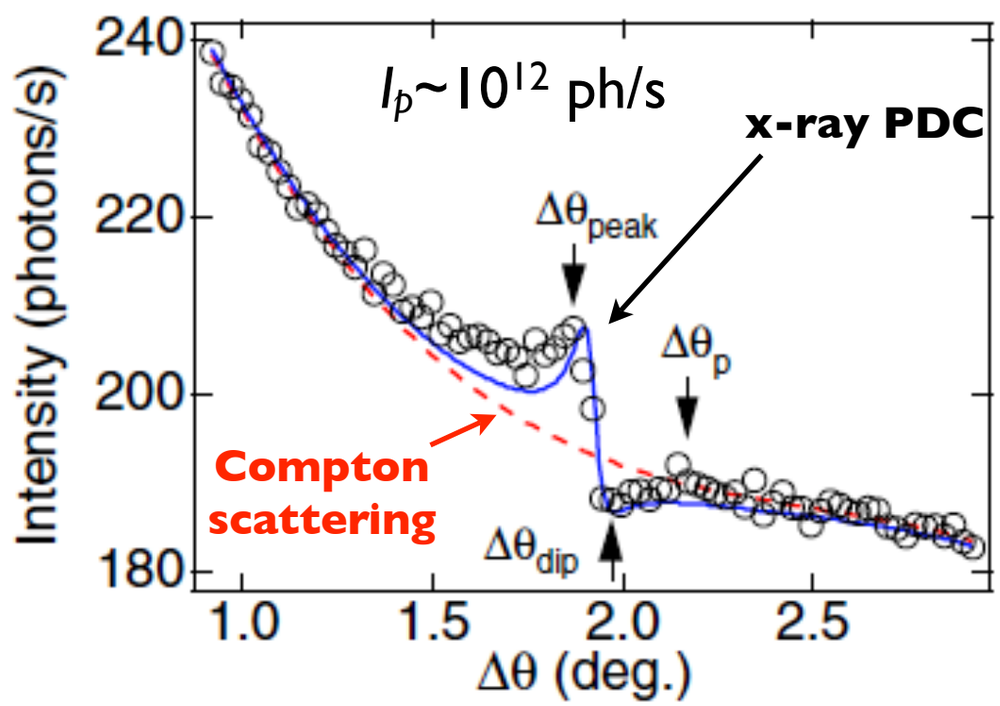
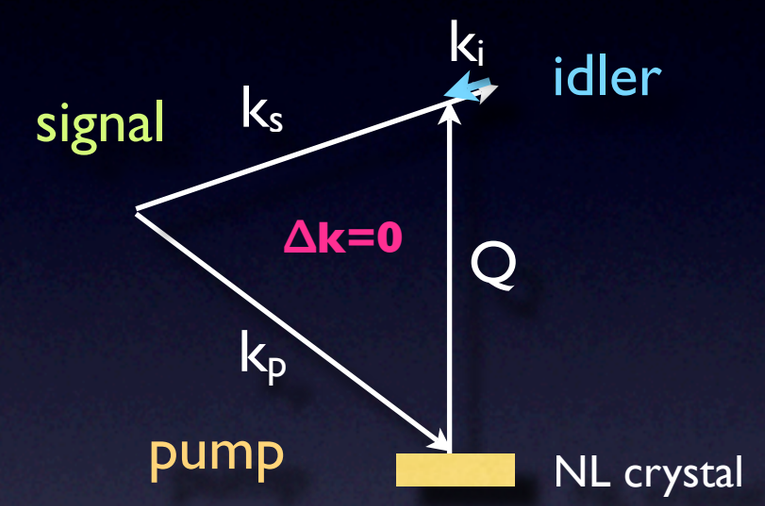
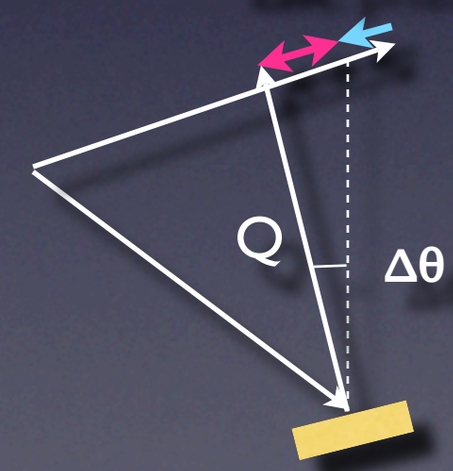


FIG. 3 (color online). The rocking curve of the nonlinear diffraction measured at $\Delta E = -100$ eV. Dashed line is polynomial fitting to the Compton background. The solid line is fitting with (7).

K.Tamasaku and T.Ishikawa, PRL **98**, 244801 (2007).



Δk: phase mismatch



Fano effect: PDC vs Compton scattering

K. Tamasaku et. al., Phys. Rev. Lett. **103**, 254801 (2009)

Idler: 40-130 eV

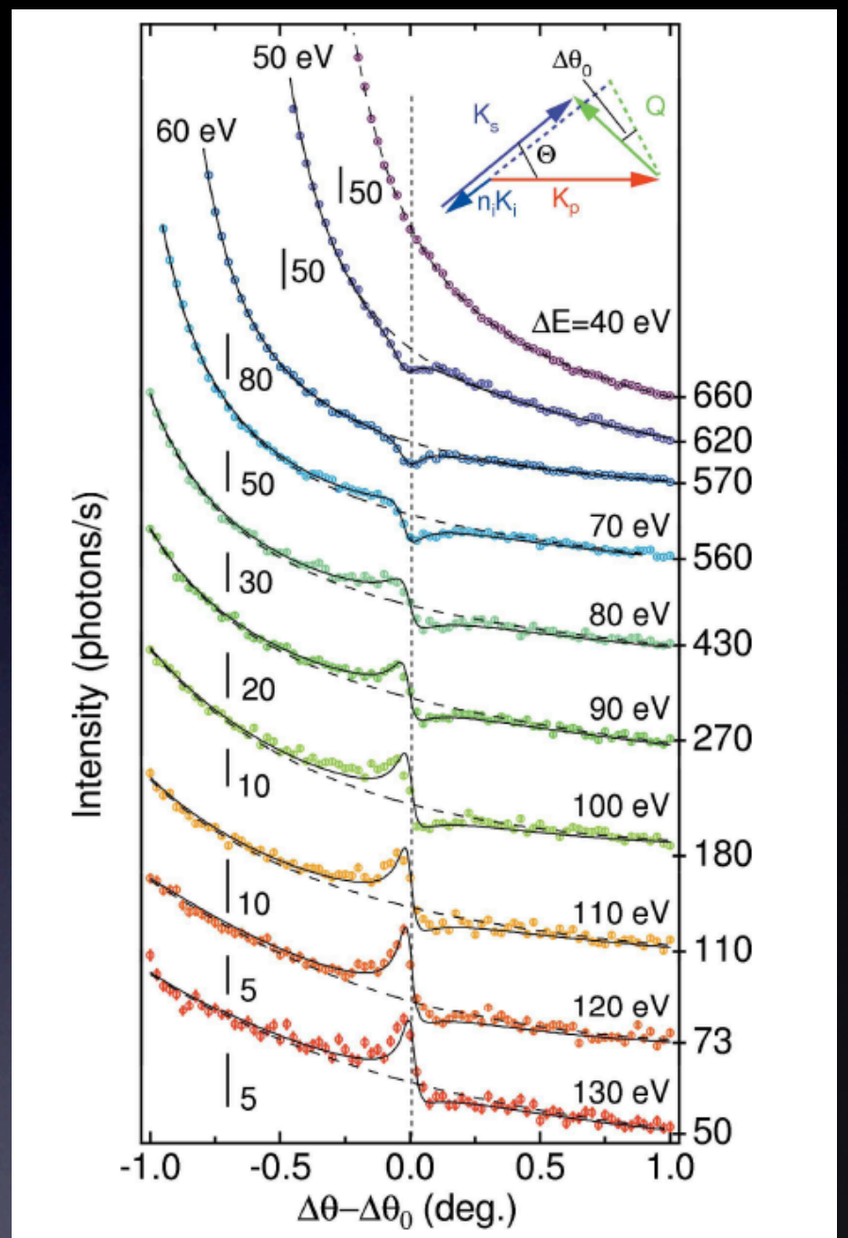
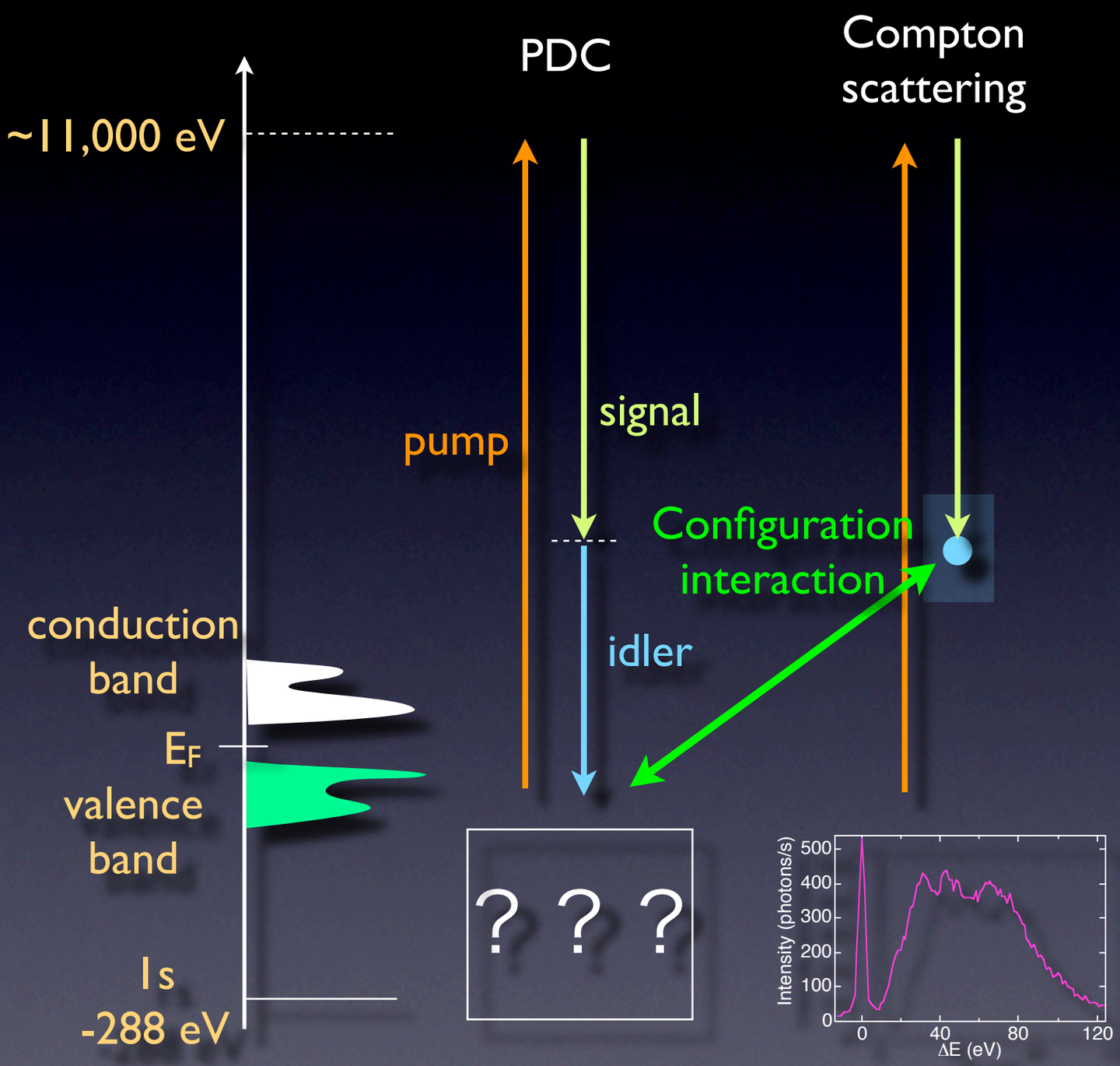


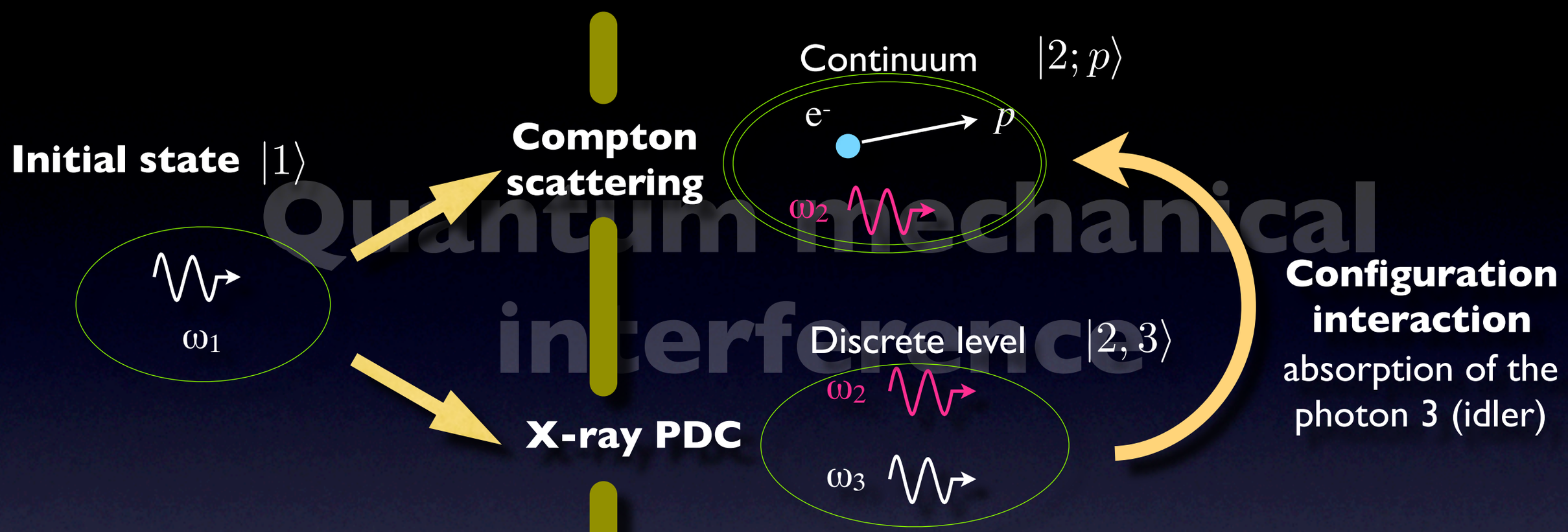
Figure 1
Rocking curves of nonlinear diffraction measured at various phase-matching conditions from $\Delta E = 40$ to 130 eV. The solid line is fitting with (3). The dashed line is the estimated background with (4). The vertical bar indicates the scale for each curve. The inset shows the schematic phase-matching geometry. The broken lines correspond to the Bragg diffraction ($E_i = 0$).



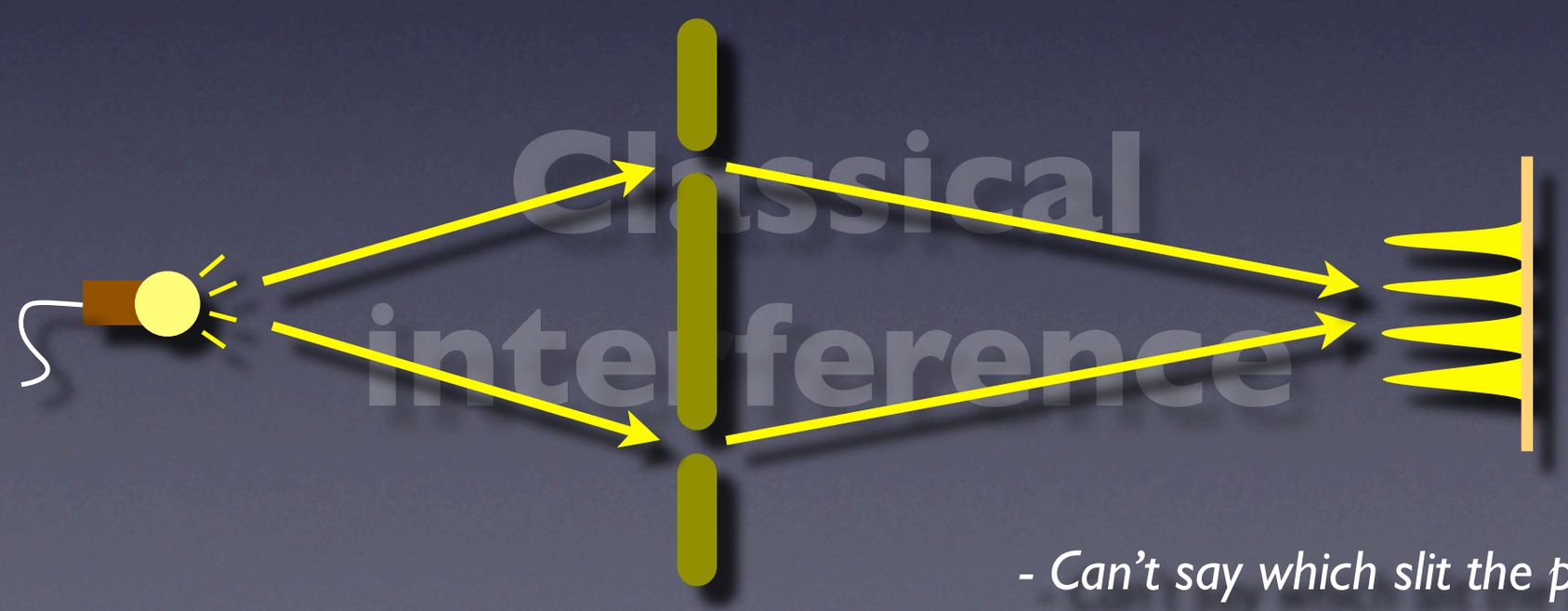
K. Tamasaku and T. Ishikawa, Acta Cryst. **A63**, 437 (2007).

Fano effect & Young's slits

K.Tamasaku *et al.*, PRL **103**, 254801 (2009).

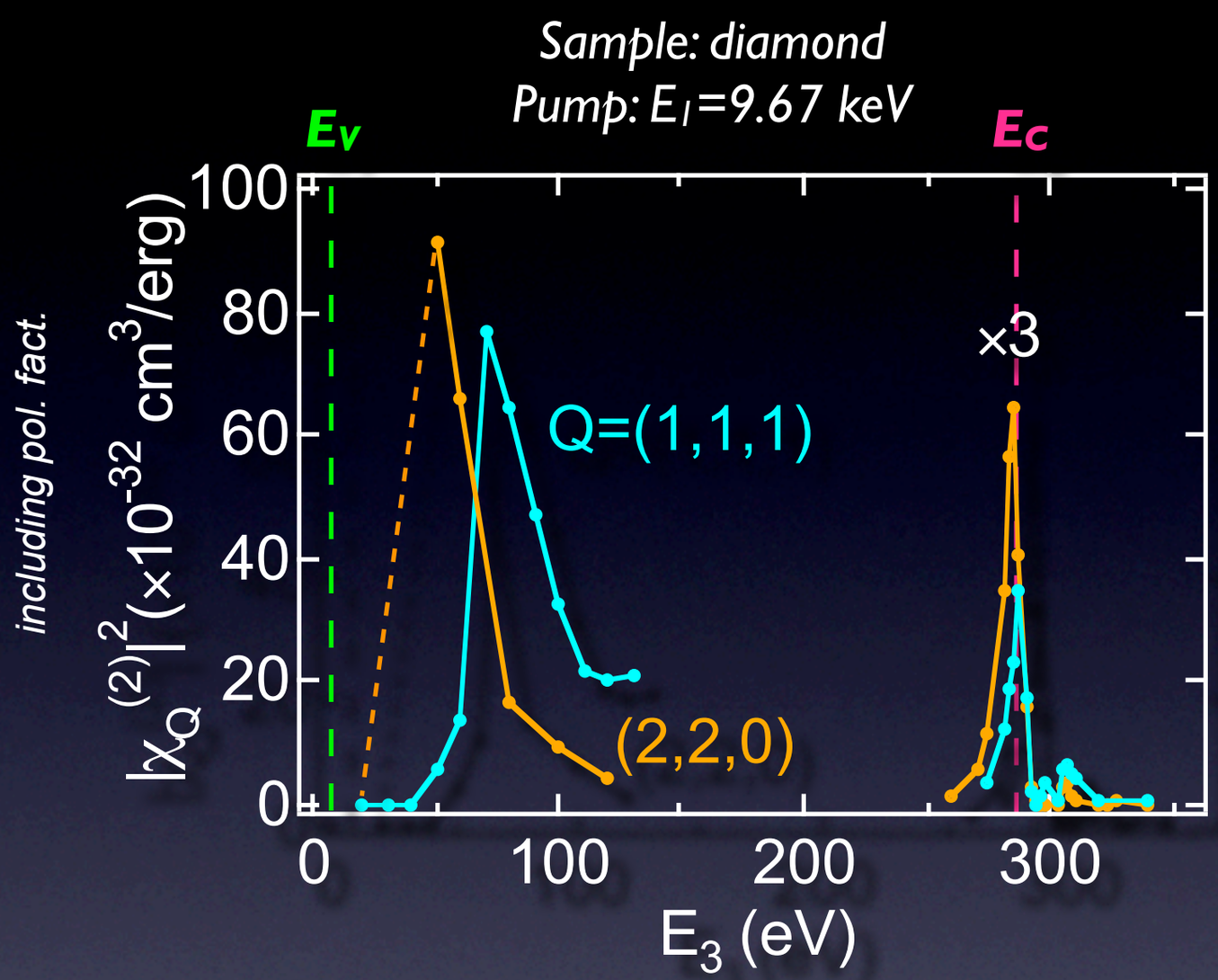


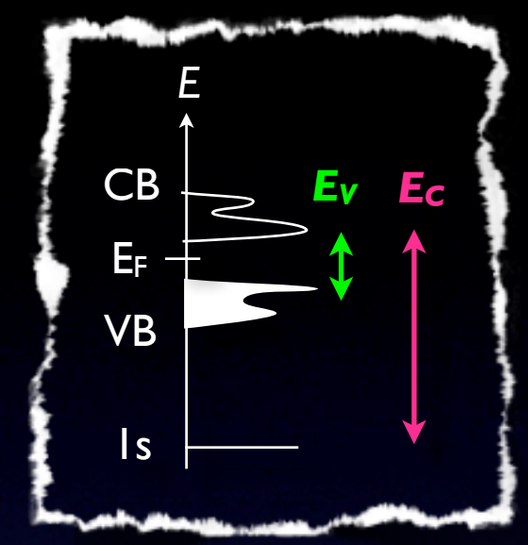
- Can't say which process produced the photon 2 (signal).



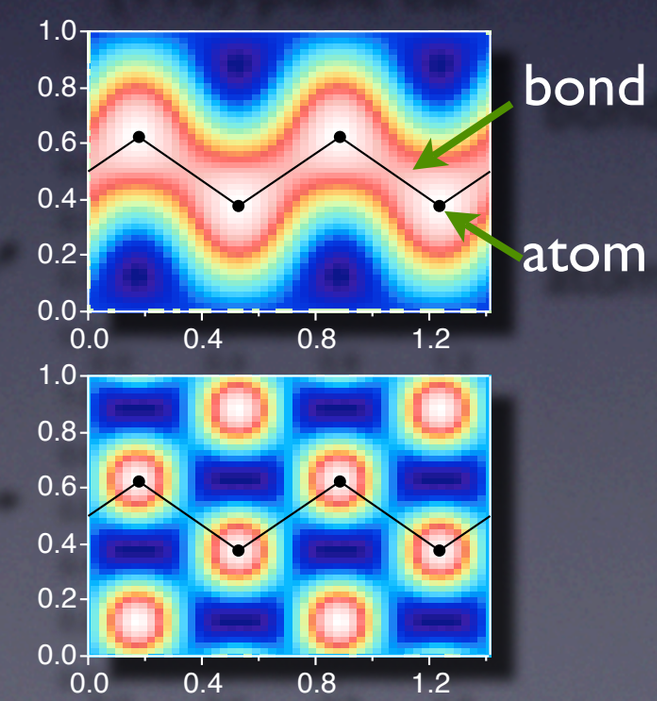
- Can't say which slit the photon passed through.

E_3- & Q-dependence of $\chi^{(2)}$





Q component
in real space
(110)-plane cut



For diamond structure Q samples

(1,1,1): atomic site + bonding site

(2,2,0): atomic site

Outline

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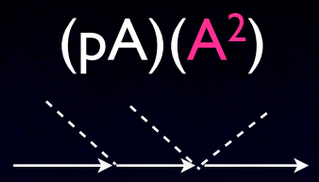

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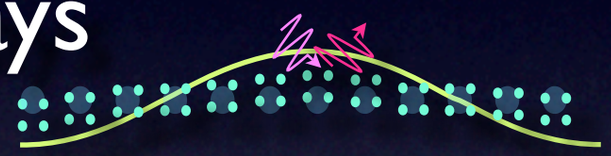

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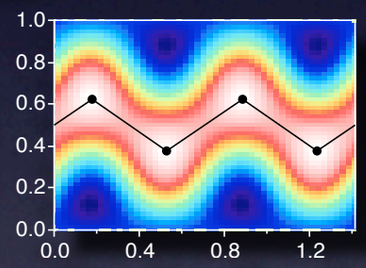
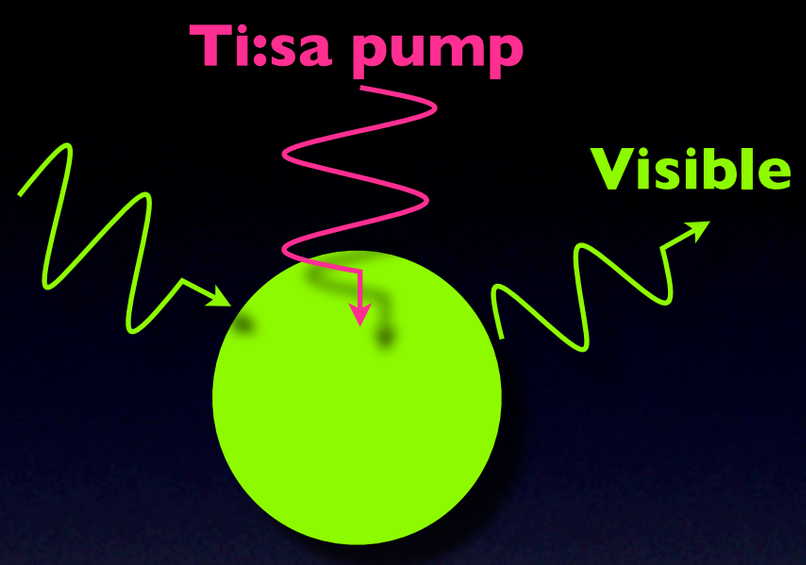


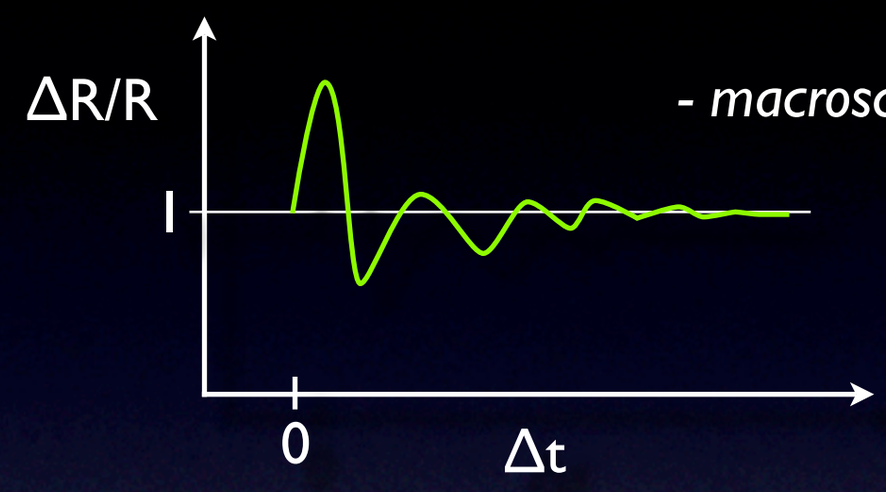
4. Key experiments

Pump-probe & x-ray PDC

- Quick & simple application of x-ray PDC

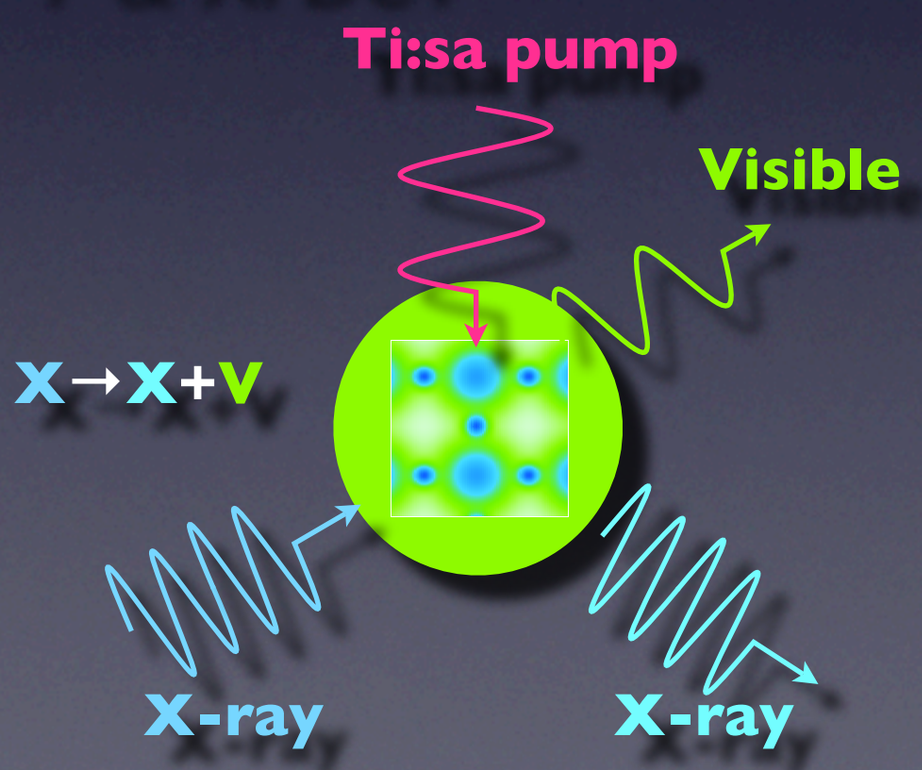
Conventional P & P

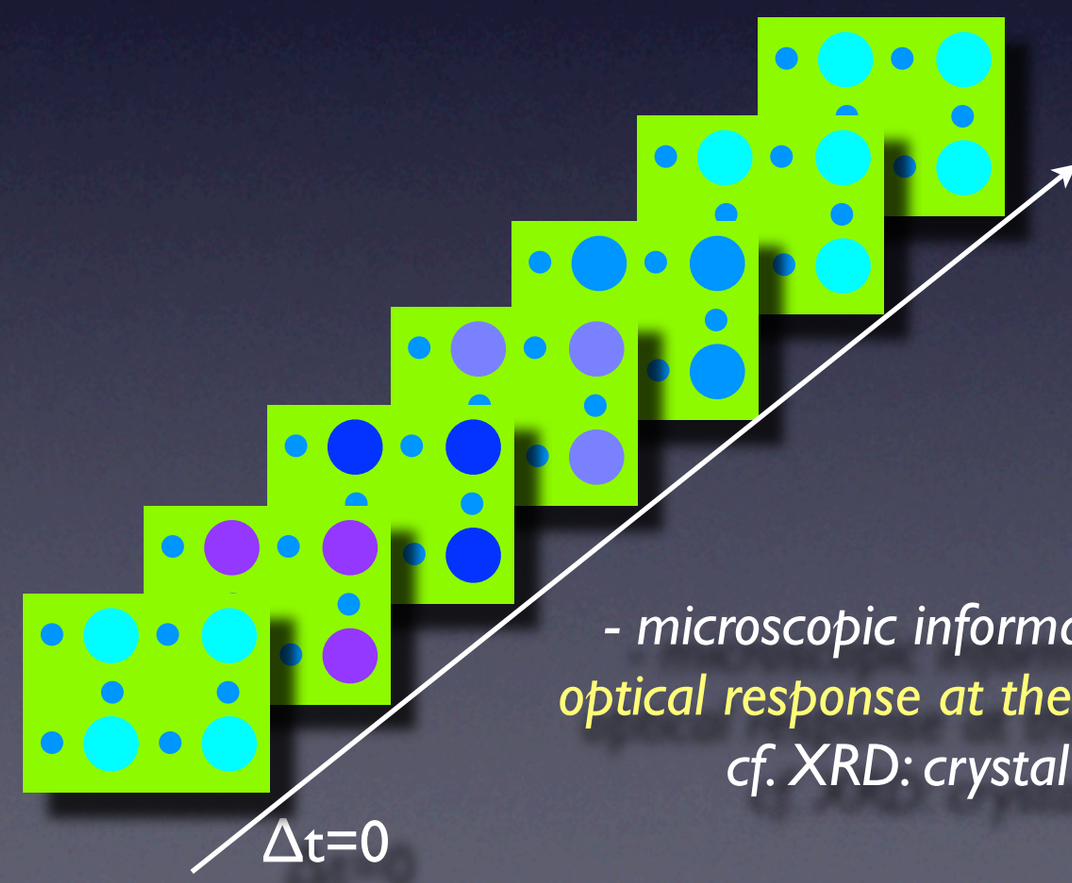




- macroscopic information

P & XPDC-P





- microscopic information about the optical response at the idler wavelength

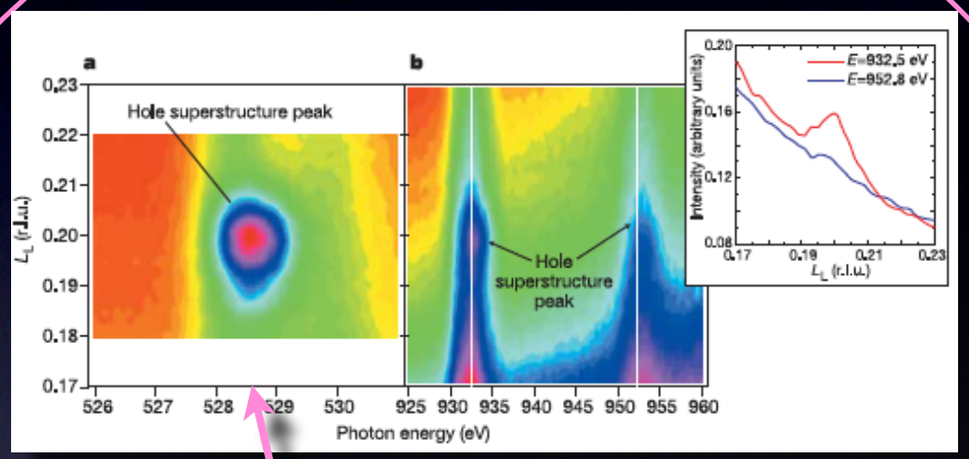
cf. XRD: crystal structure

X→X+SX for material science

- Easily combined with pump & probe method to investigate dynamics

SX diffraction is powerful tool, but...

Abbamonte, *Nature* **431**, 1078 (2004).



Oxygen *K*-edge

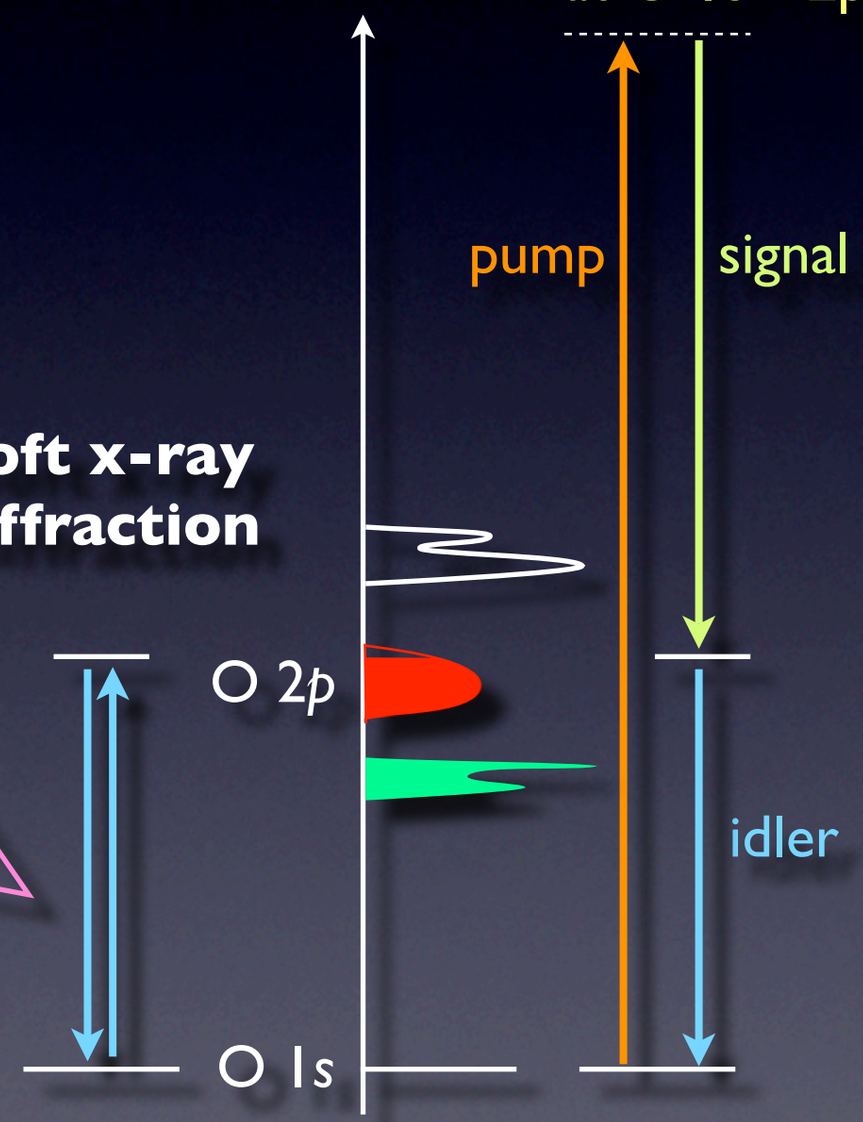
23 Å : *not enough resolution to investigate within the unit cell*

Sample environment is very limited due to absorption.

Soft x-ray diffraction

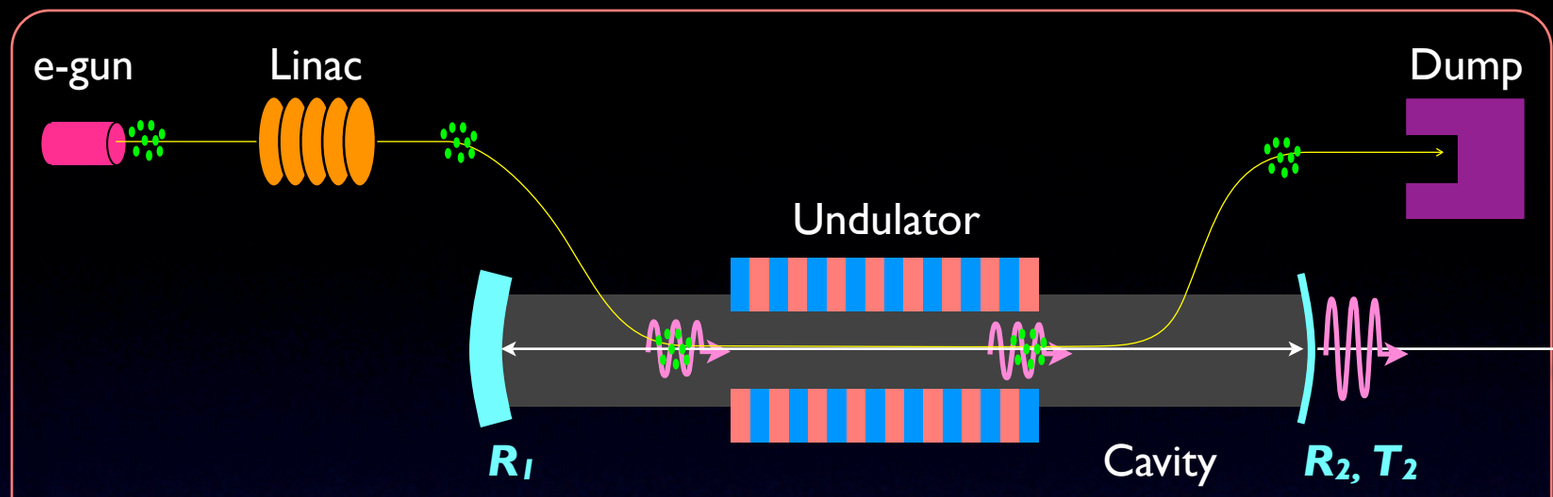
X-ray PDC

can realize Å resolution at O 1s→2p resonance



Higher resolution & flexible sample environment

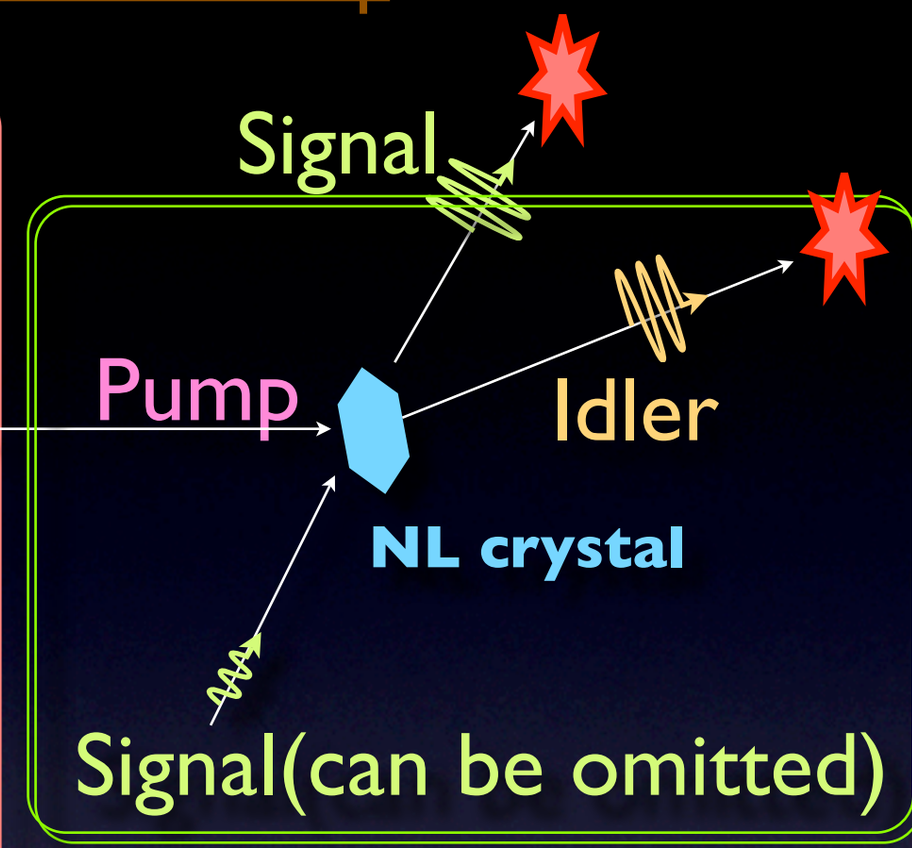
Optical parametric amplifier



- ★Fully coherent x-rays
- ★10^9 photons/pulse
- ★1 MHz
- ★$\Delta E \sim$ meV
- ★$\Delta t \sim$ ps
- ✓**But, operate at a fixed wavelength**

XFEL Oscillator

K. J. Kim et al., PRL100, 244802 (2008).



Optical Parametric Amplifier

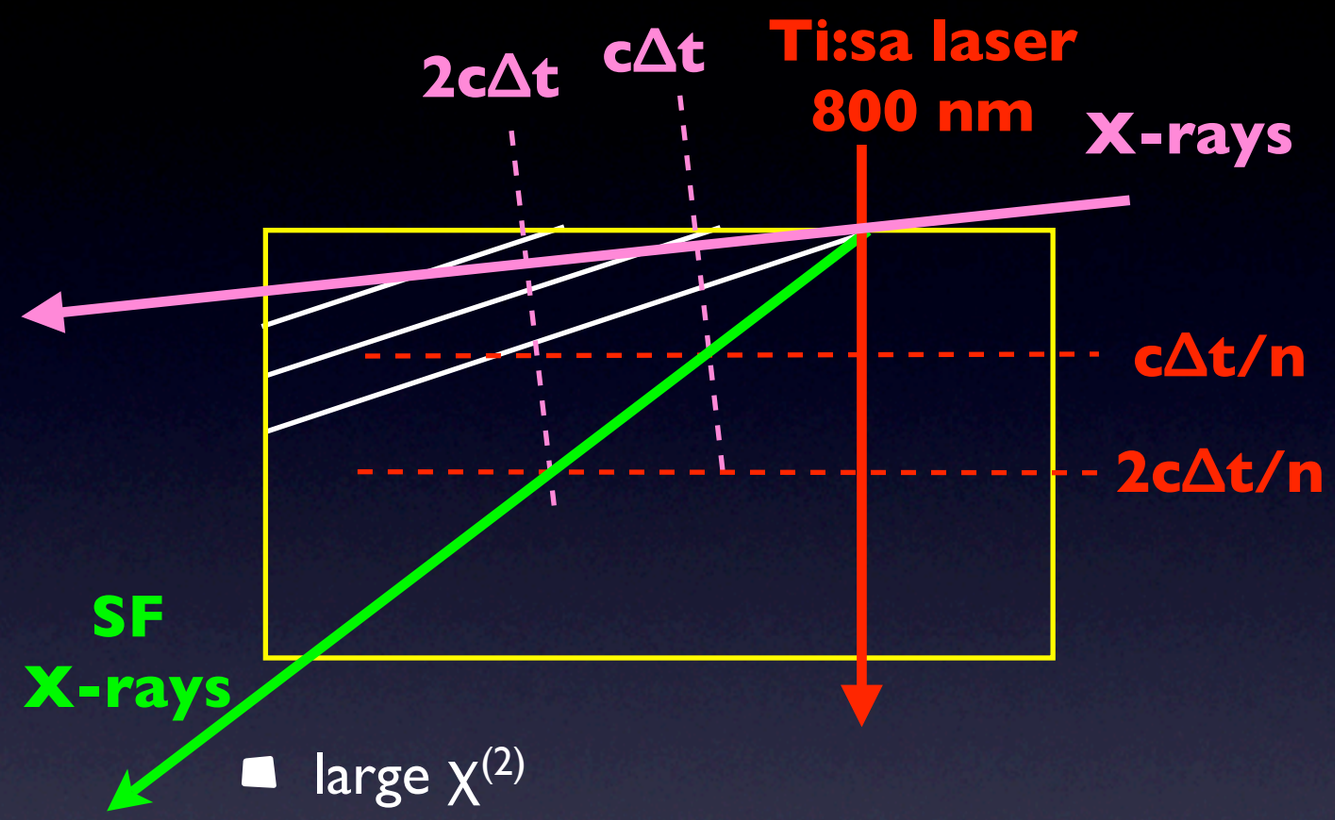
-wavelength conversion-

Spectroscopic application of XFEL-O

Cross-correlator by sum-freq. generation (SFG)

- Another method to measure pulse width

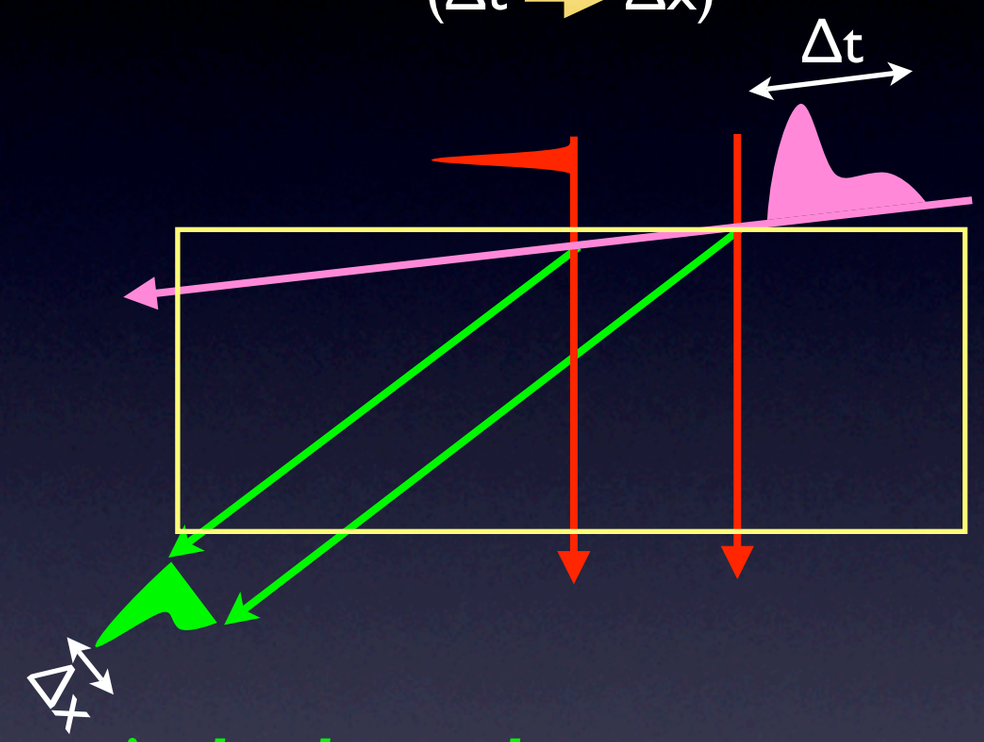
Group velocity matching



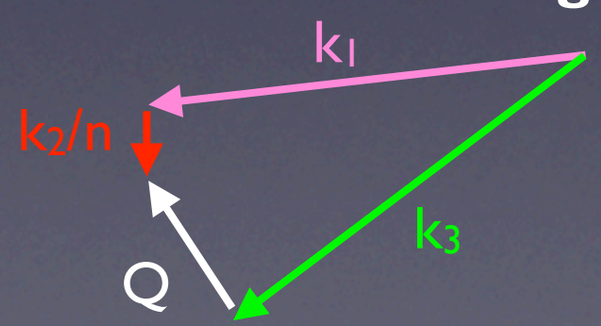
- large $\chi^{(2)}$
- n: cost. around 800 nm & smaller is easier

Cross-correlator

(Δt → Δx)



single-shot pulse measurement

To measure the temporal resolution of XFEL

Phase matching