# X-ray nonlinear optics

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Outline

***1. X-ray nonlinear optics***

2. Parametric down-conversion of x-rays

3. Nonlinear susceptibility of diamond

4. Future perspectives

# X-ray specific features: $A^2$-interaction

$$\mathcal{H}' = \sum_k \left[ -\frac{e}{mc}\vec{p}_k \cdot \vec{A}_k + \frac{e^2}{2mc^2}\vec{A}_k \cdot \vec{A}_k \right]$$

(pA) (A$^2$)

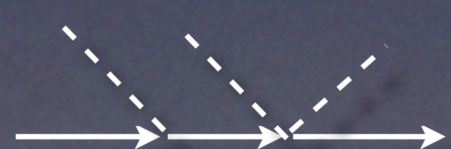
@ Coulomb guage

Ex. 2nd order nonlinear process = 3 photon process (need 3 A)

(pA)(pA)(pA) → "nonlinear optics"

(pA)($A^2$) → X-ray nonlinear optics

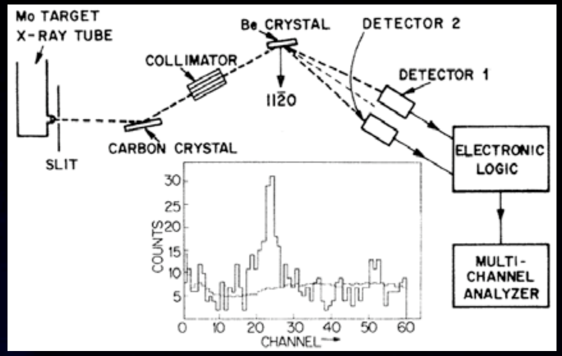
ignored term in "nonlinear optics"



# XNLO - scince 1969

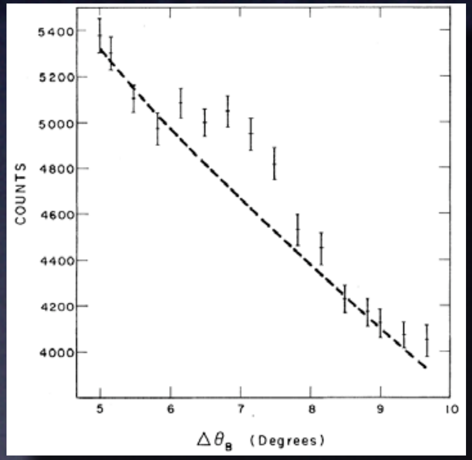
Theory & Experiment

Parametric down-conversion into x-rays




Eisenberger, PRL 1972

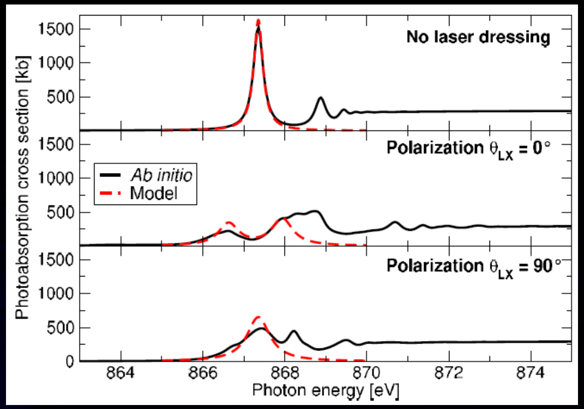
Parametric down-conversion into EUV




Danino, PRL 1981

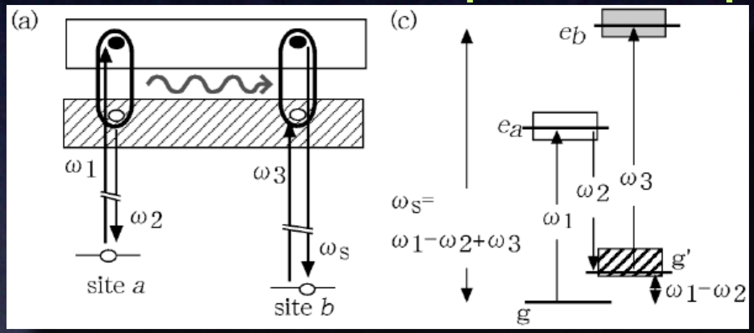
- Observable or not?

EIT




Buth, PRL 2007

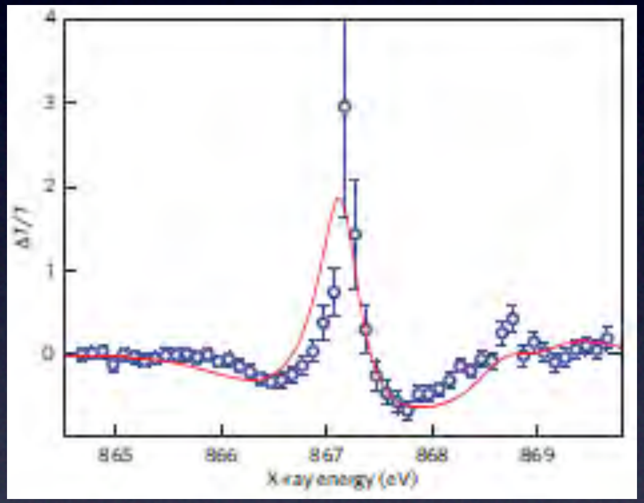
Coherent Raman spectroscopy




Tanaka, PRL 2002

Pulse control of x-rays

EIT



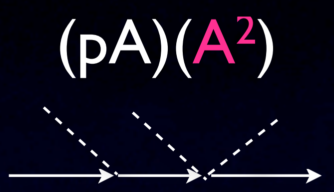

Glover, NPhys. 2010

Local probe of electronic excitation

- *Scientific interest*
- *Application with XFEL*

Outline

1. X-ray nonlinear optics




**2. *Parametric down-conversion of x-rays***

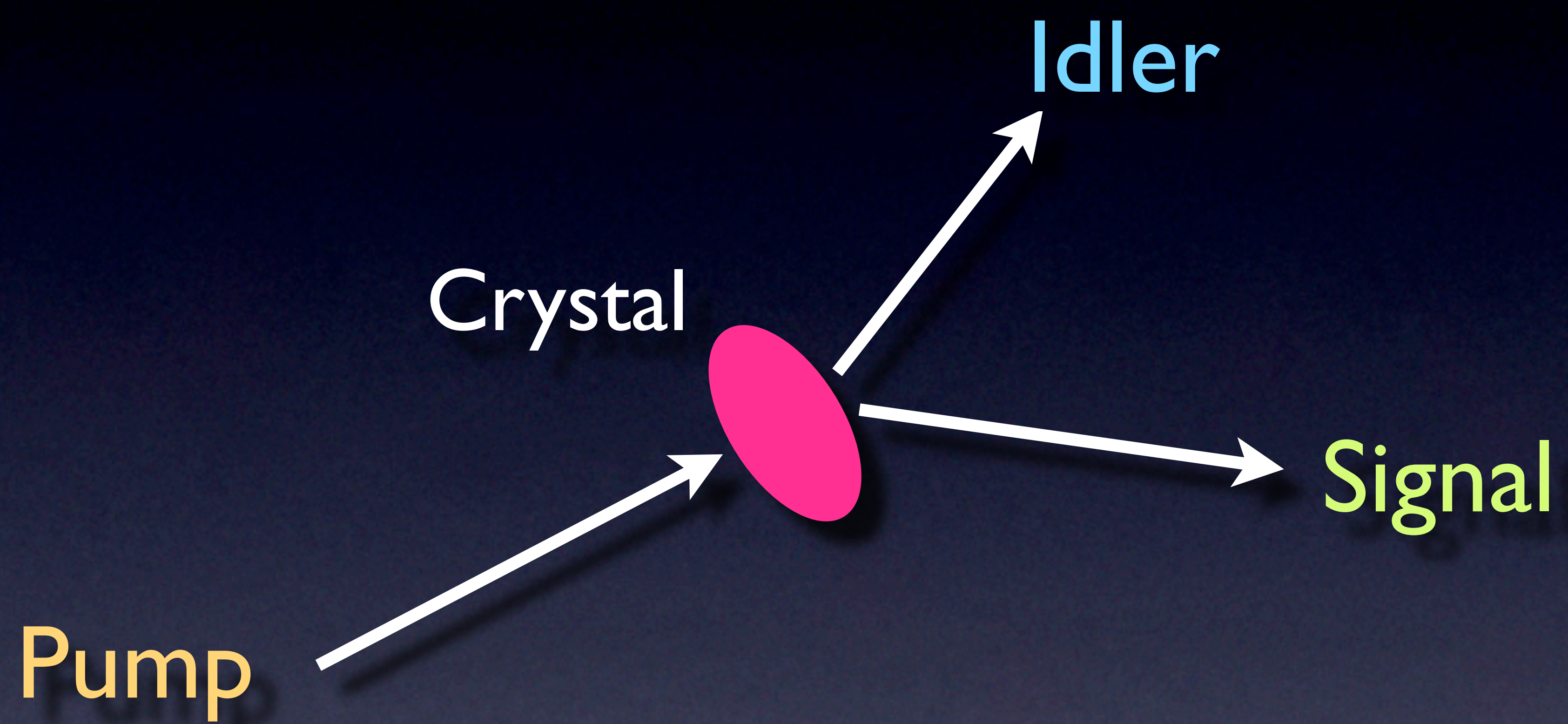
3. Nonlinear susceptibility of diamond

4. Future perspectives


XFEL (x-ray laser)
under construction
30 Gw
~800 m
~500 m
SPring-8 (x-ray flash lamp)
< 2 kW

# X-ray parametric down-conversion (PDC)

*- $2^{nd}$ order nonlinear process*

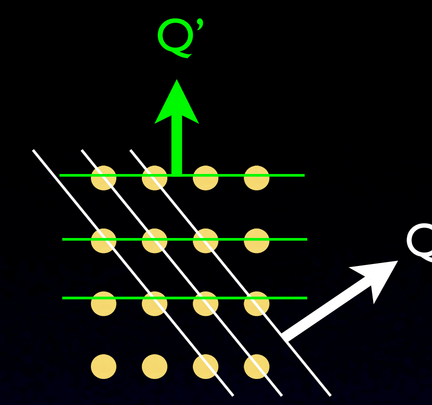



✓ Momentum conservation
✓ Energy conservation

Unique application of x-ray PDC

# Momentum conservation (phase matching)

Phase matching with reciprocal lattice vector; Q

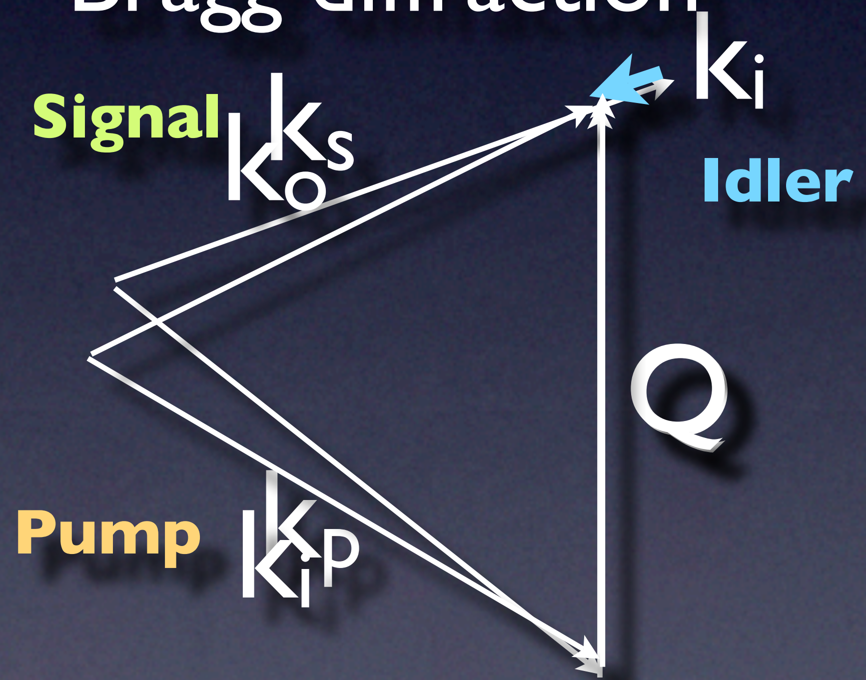



→ "Nonlinear diffraction"

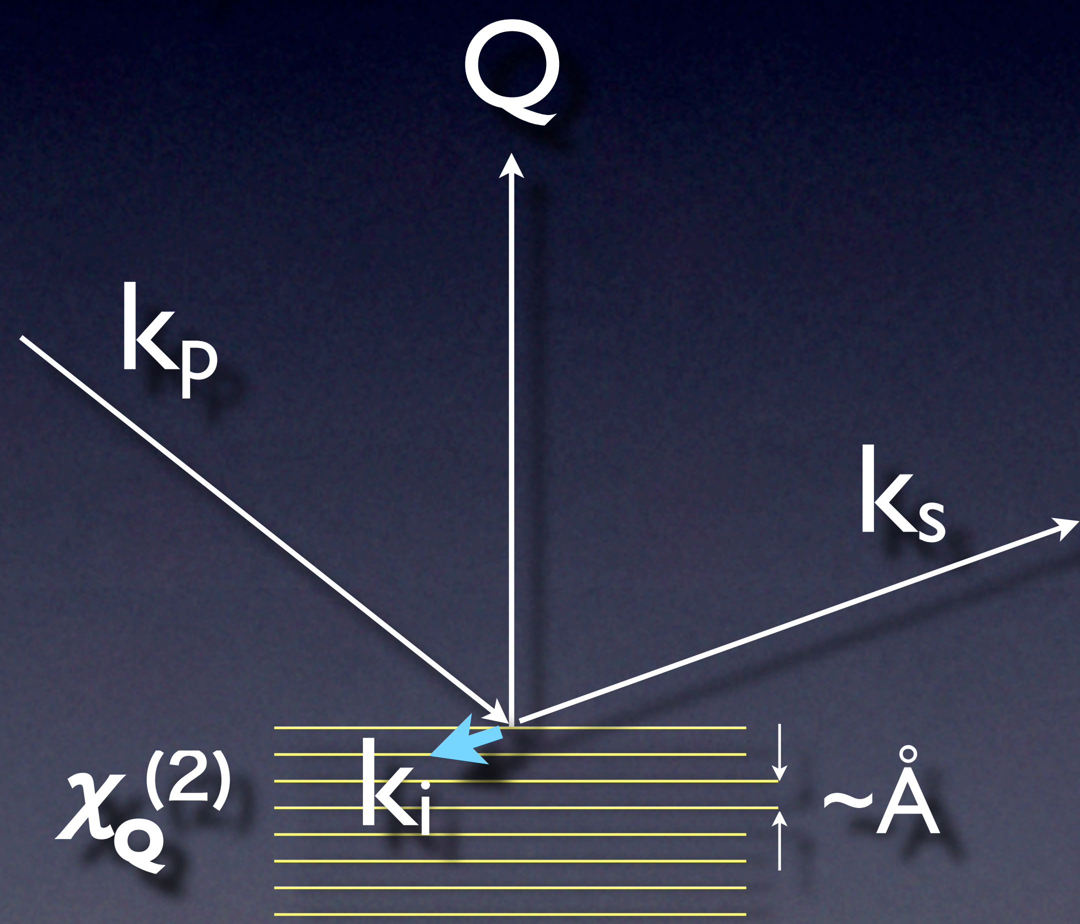
*Momentum space*

$k_p+Q=k_s+k_i$

Bragg diffraction




*Real space*

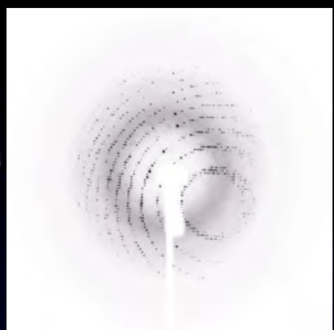


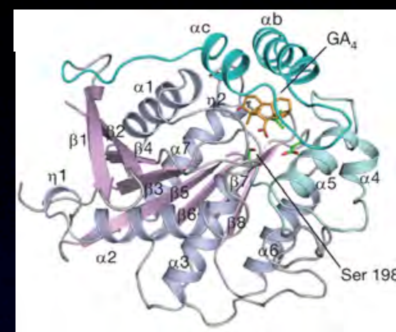

Grating of nonlinear polarizability

# X-ray nonlinear diffraction

## Usual (linear) diffraction

structure of $\chi$





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

***Total charge density***

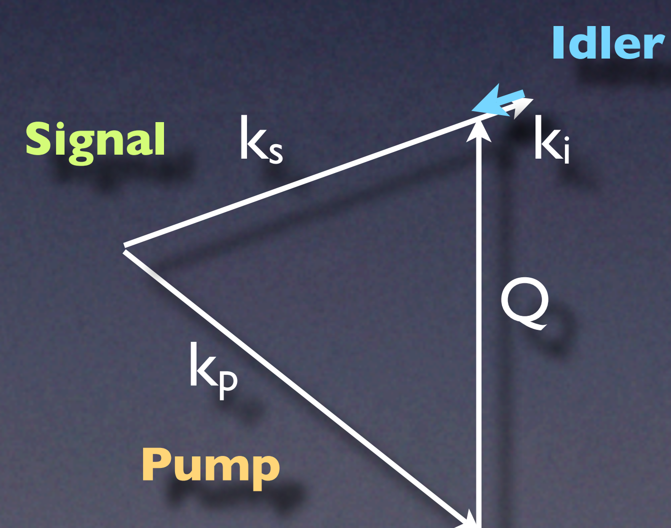
Susceptibility

Linear: $P = \chi E$

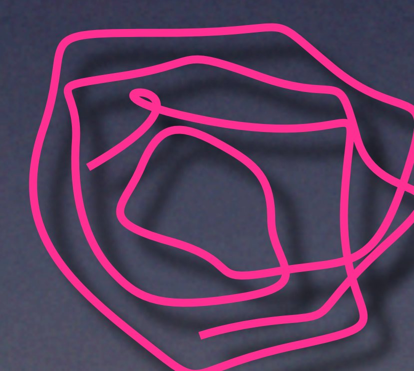
$2^{nd}$ order NL: $P_p = \chi^{(2)} E_s E_i$

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

## Nonlinear diffraction

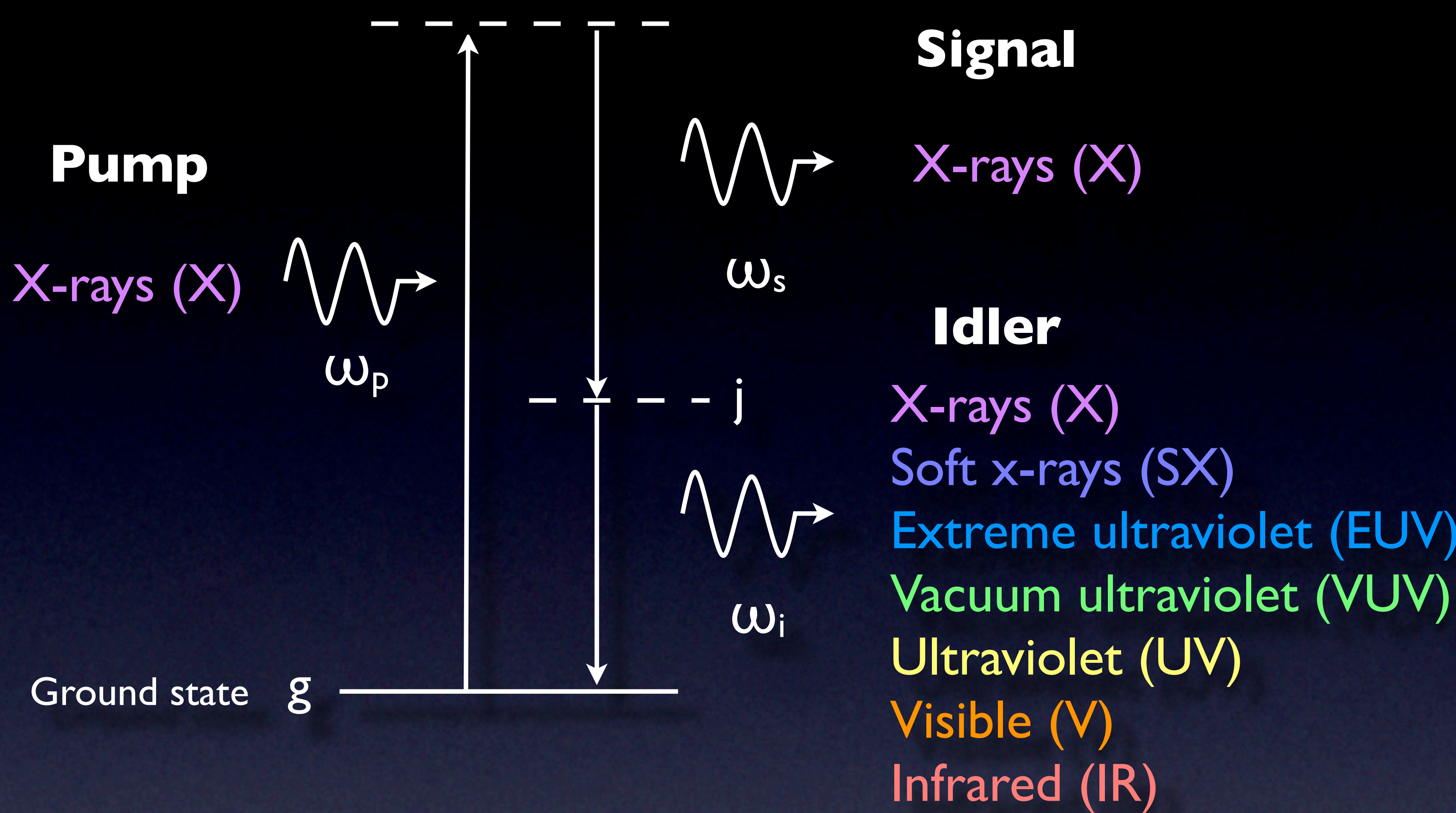



structure of $\chi^{(2)}$



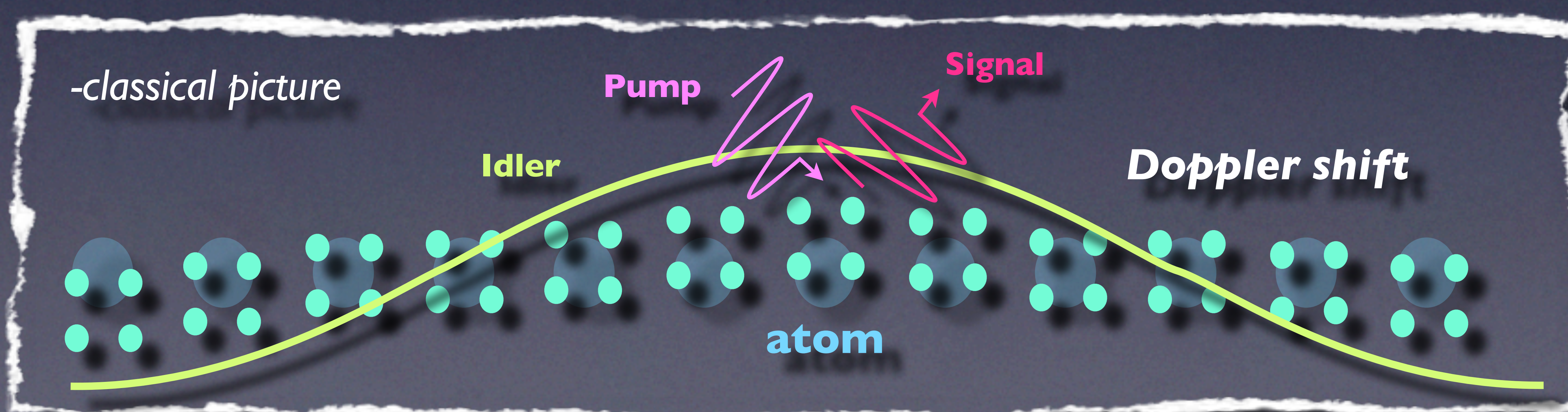
with Å resolution

# Energy conservation



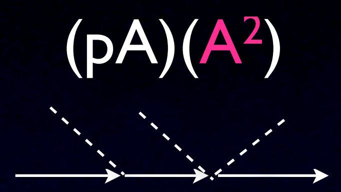

QM description:

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

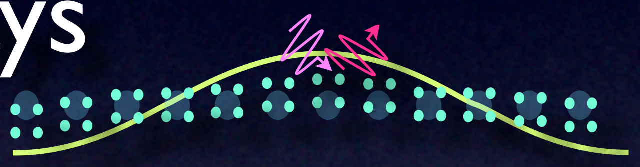


Outline

1. X-ray nonlinear optics




2. Parametric down-conversion of x-rays



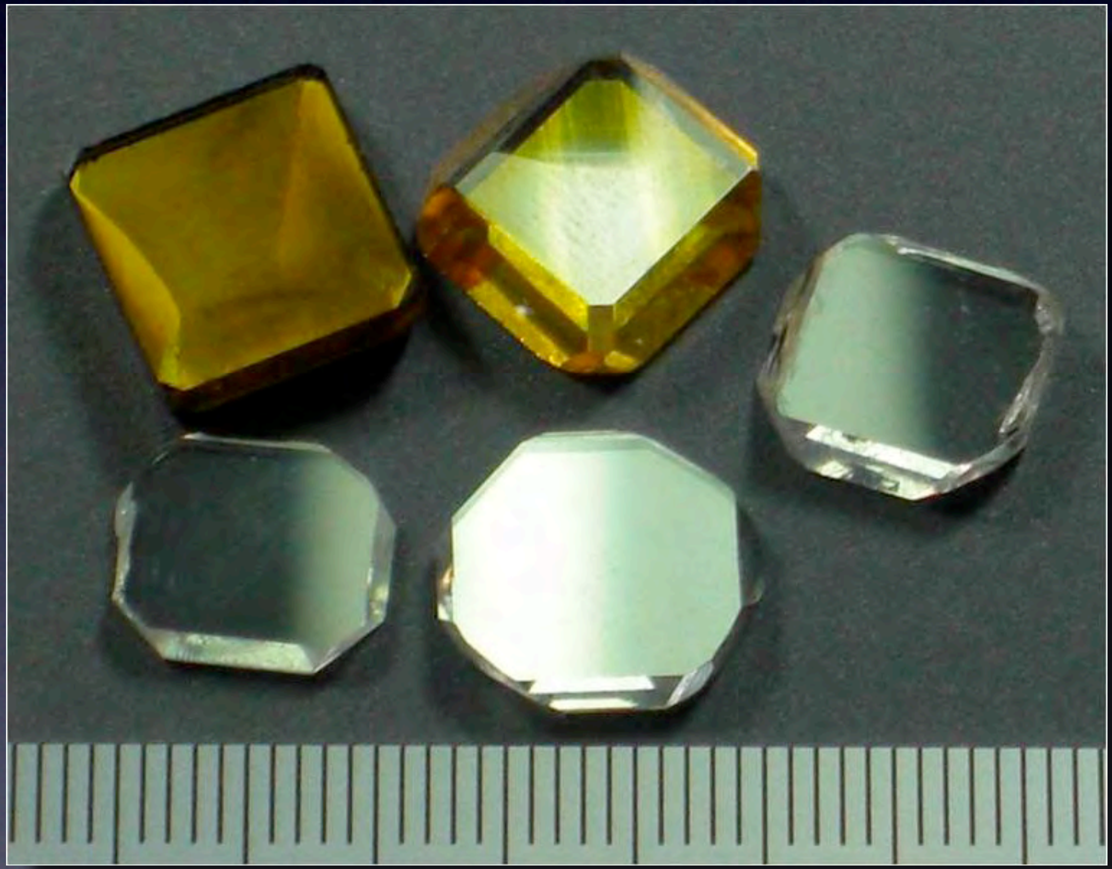
***3. Nonlinear susceptibility of diamond***

4. Future perspectives

# Experiments

Very different from optics...

Nonlinear medium: diamond



4/6 electrons : Bond
2/6 electrons : Core

# 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)**

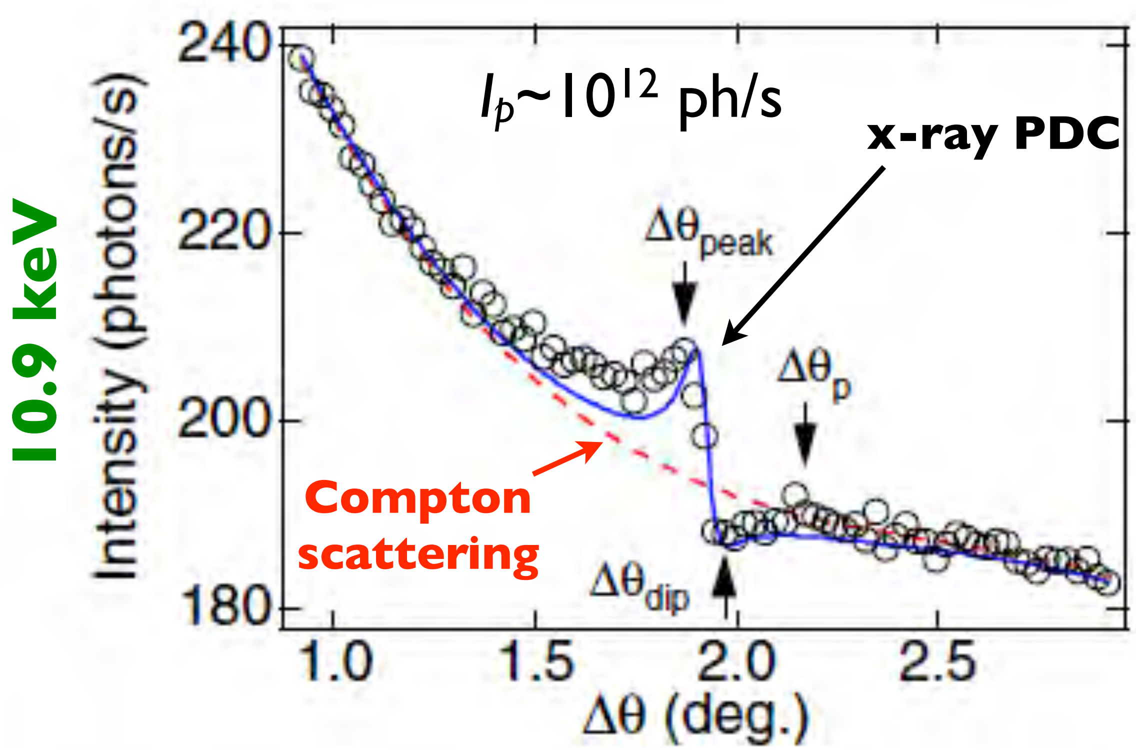
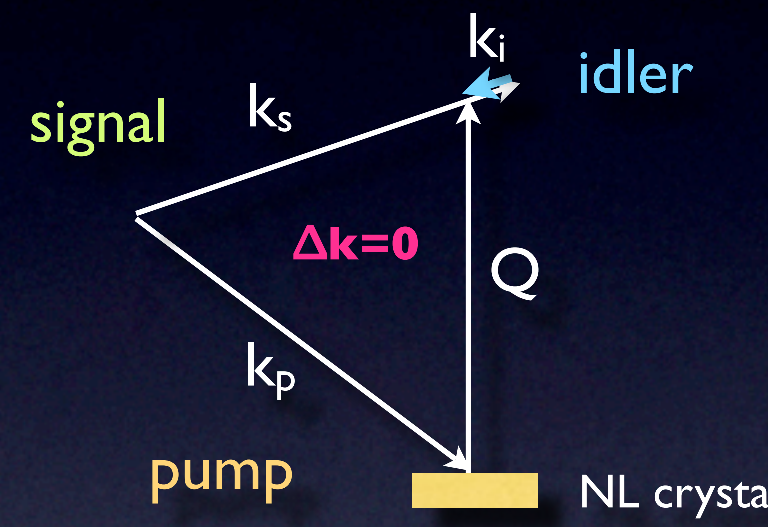
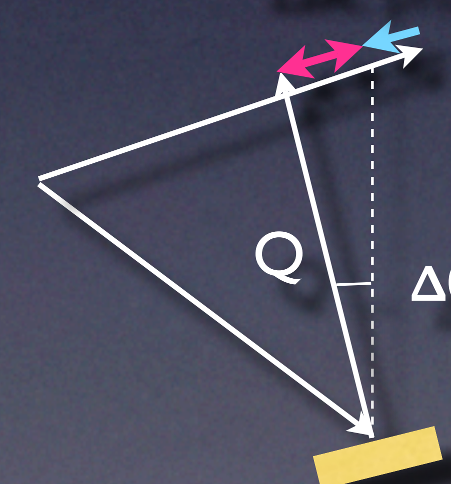



FIG. 3 (color online). The rocking curve of the nonlinear diffraction measured at $\Delta E = -100$ eV. Dashed line is polynomial fitting to the Compton background. The solid line is fitting with (7).

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**

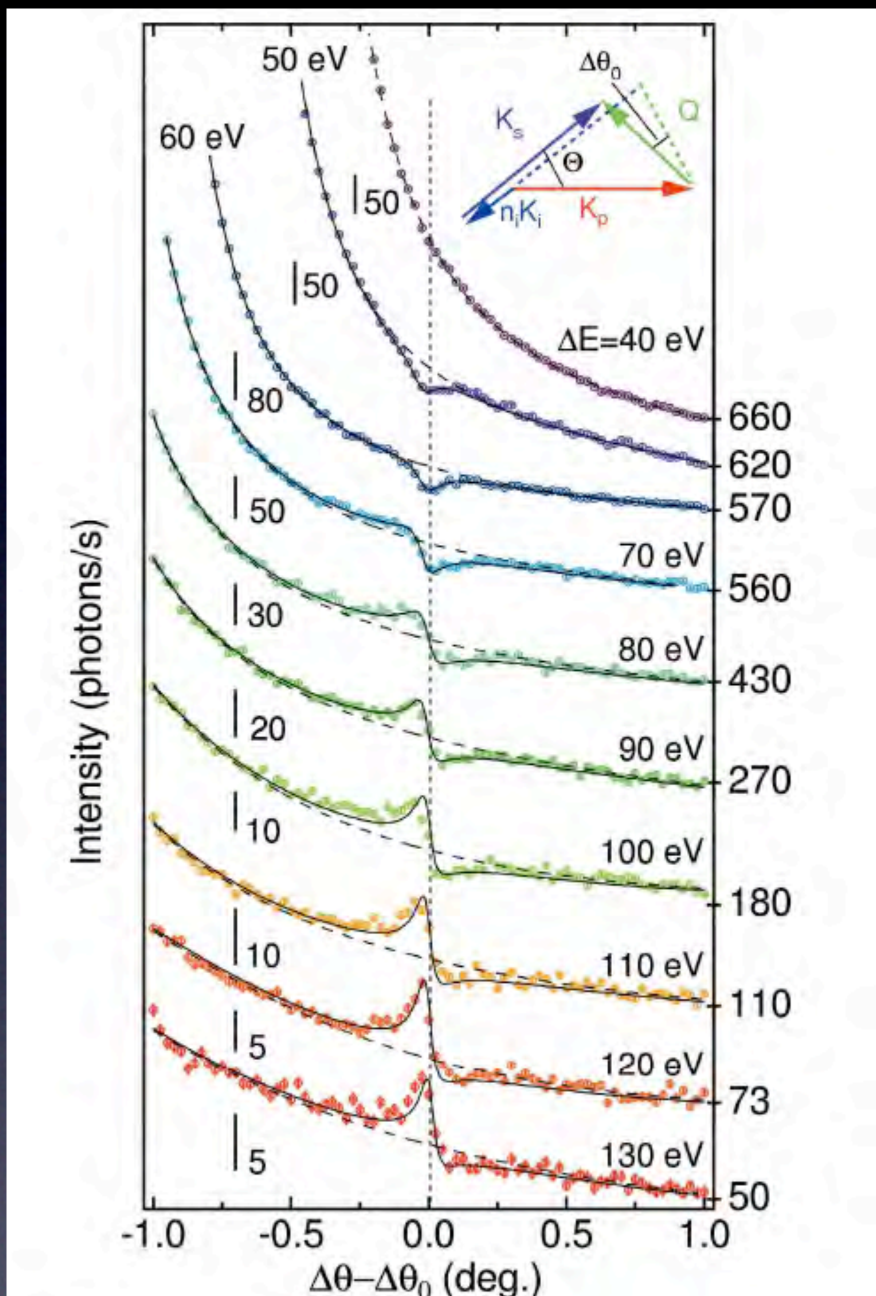
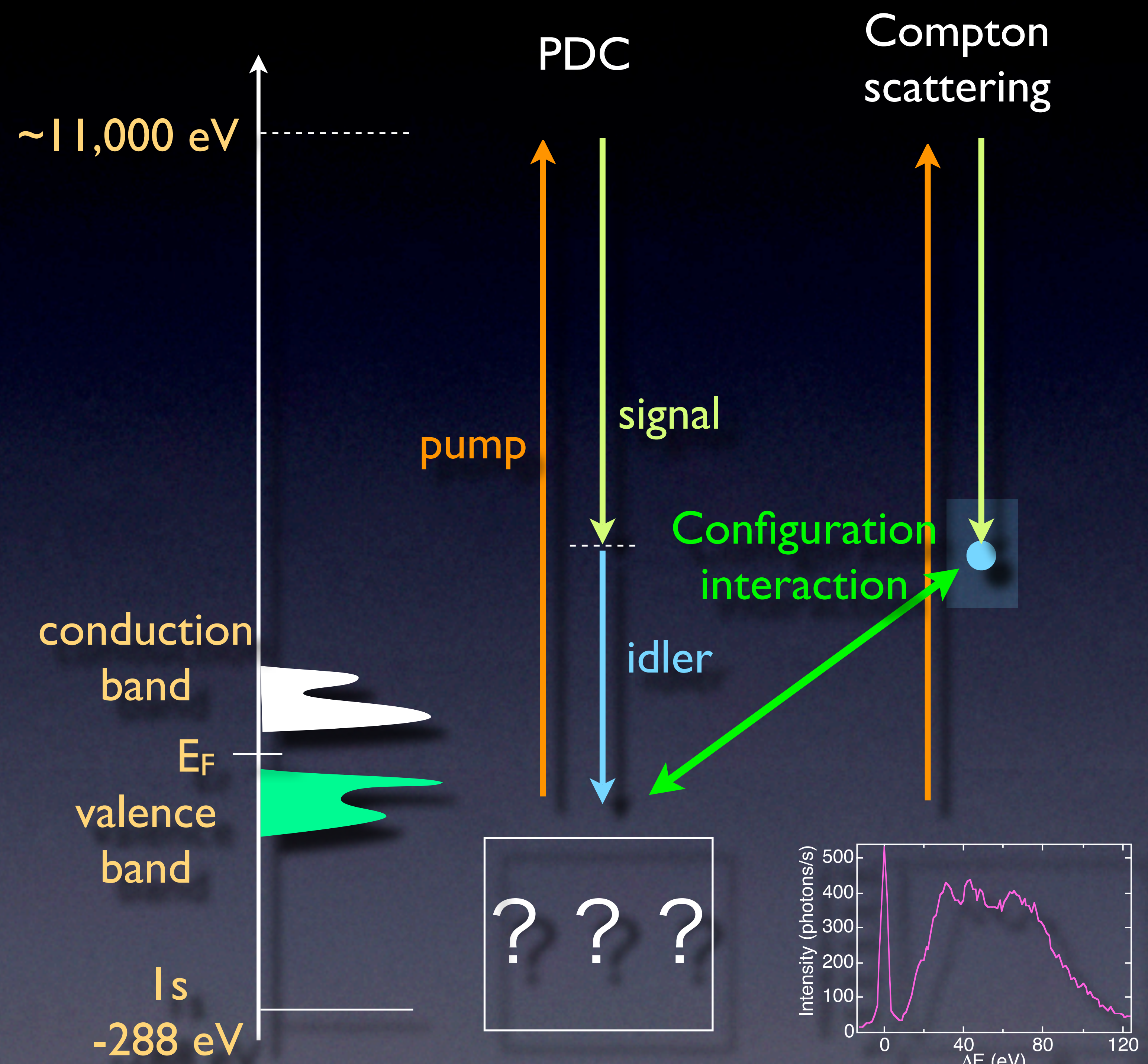
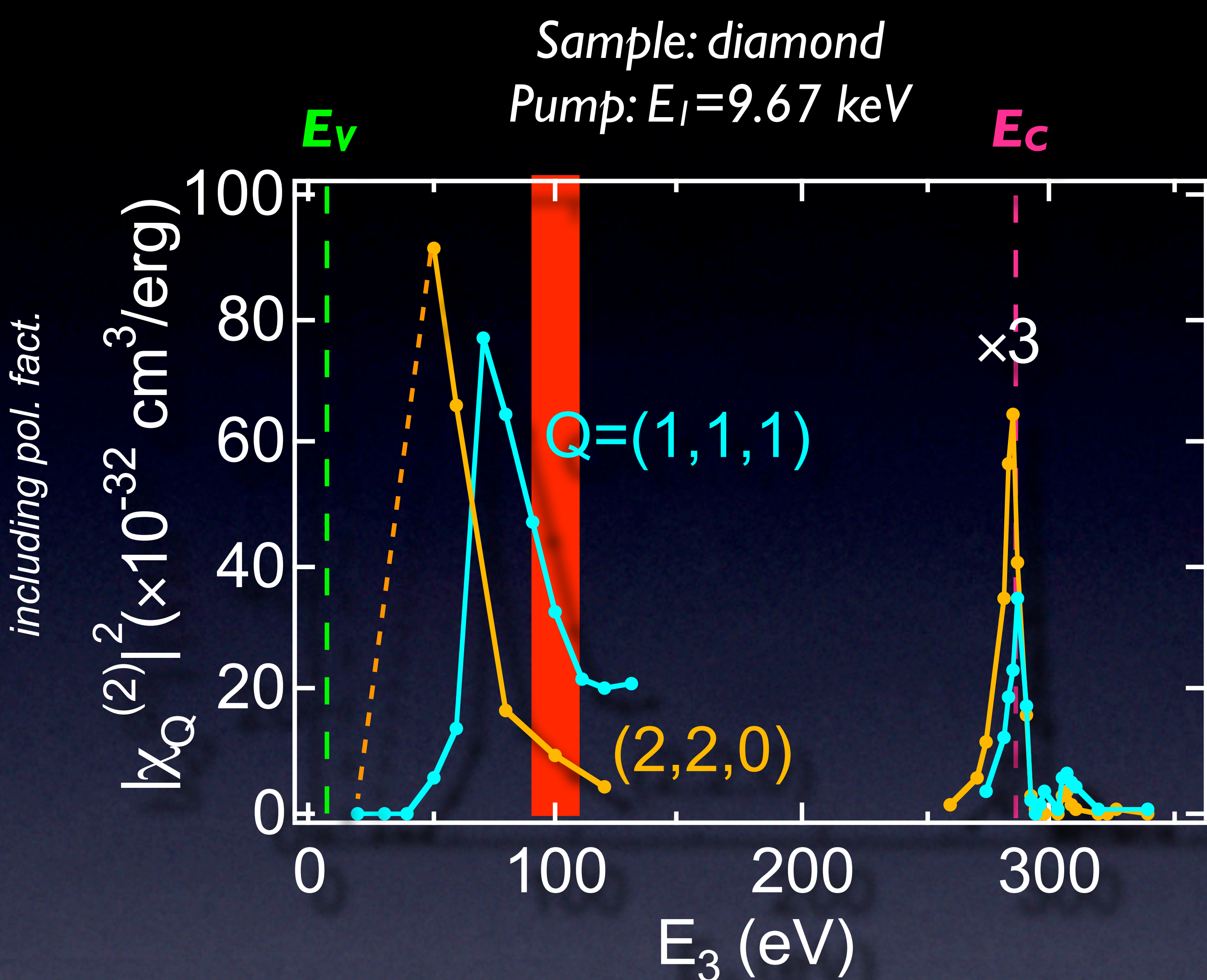


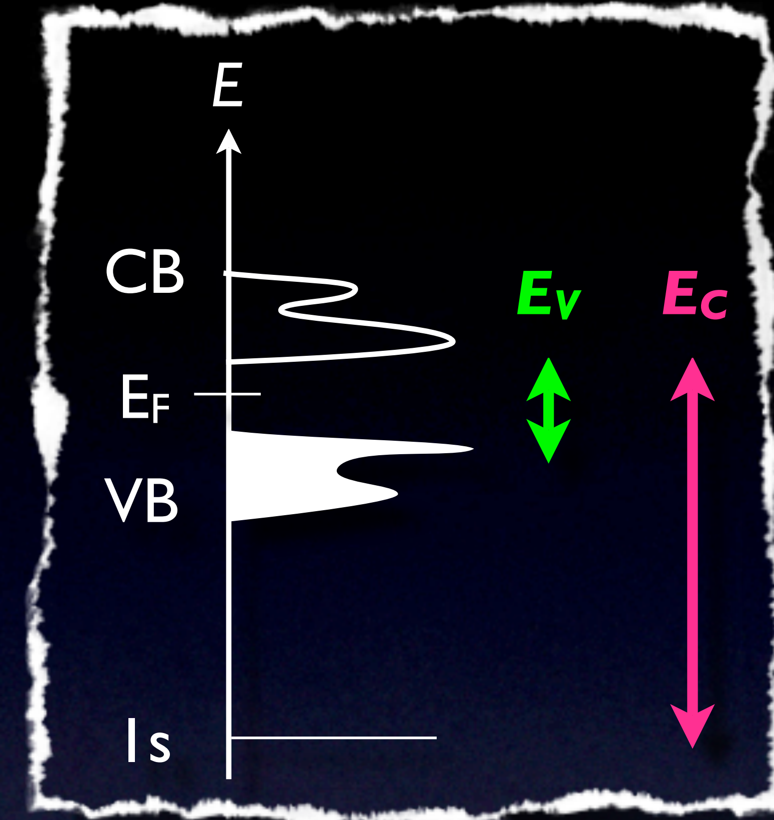

**Figure 1**
Rocking curves of nonlinear diffraction measured at various phase-matching conditions from $\Delta E = 40$ to 130 eV. The solid line is fitting with (3). The dashed line is the estimated background with (4). The vertical bar indicates the scale for each curve. The inset shows the schematic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



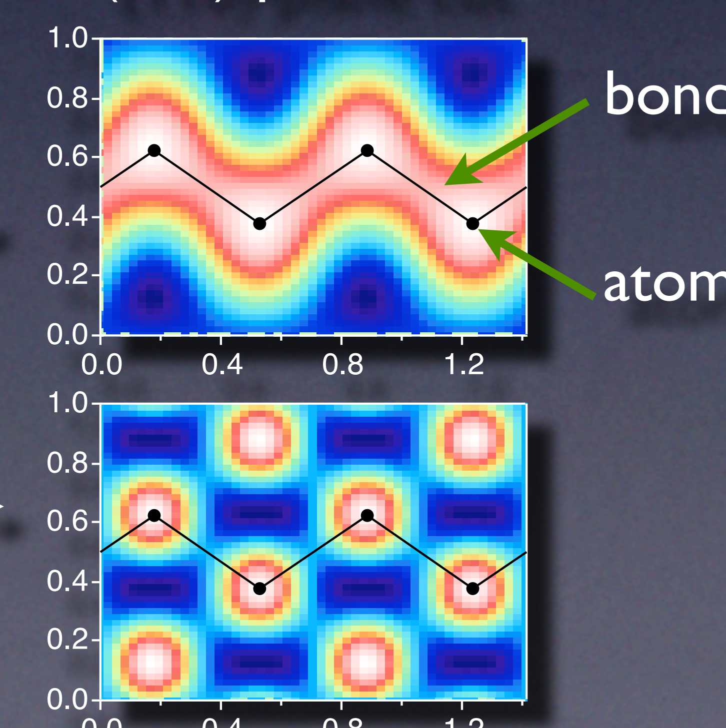

K.Tamasaku and T.Ishikawa, Acta Cryst. **A63**, 437 (2007).

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




Q component
in real space
(110)-plane cut



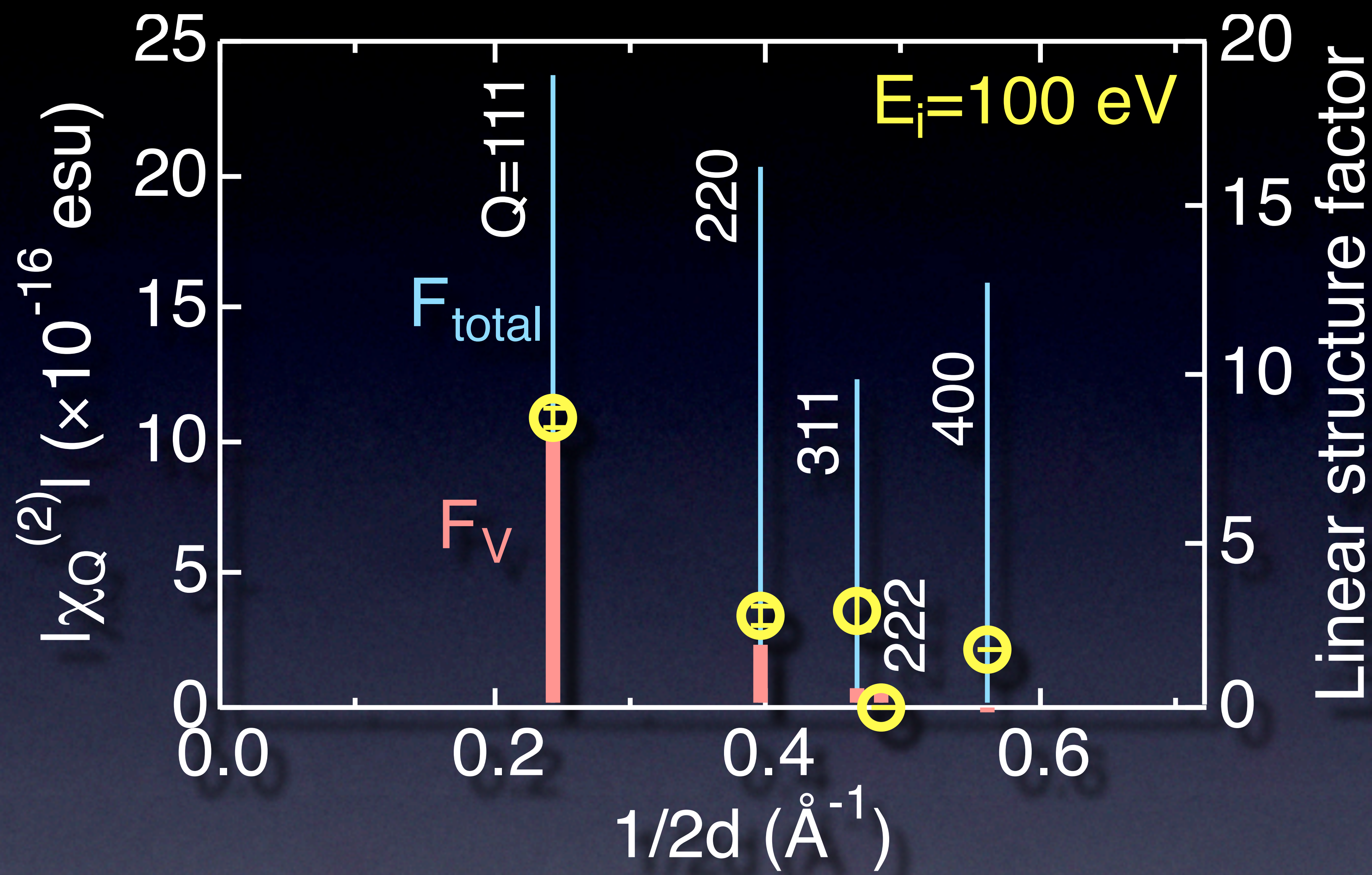

*For diamond structure Q samples*

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

(2,2,0): atomic site

# $\chi_Q^{(2)}$ @ $E_i$=100 eV

Diamond
$E_p$=11.107 keV




Similar Q-dependence to the valence charge density.

→ *Reasonable*

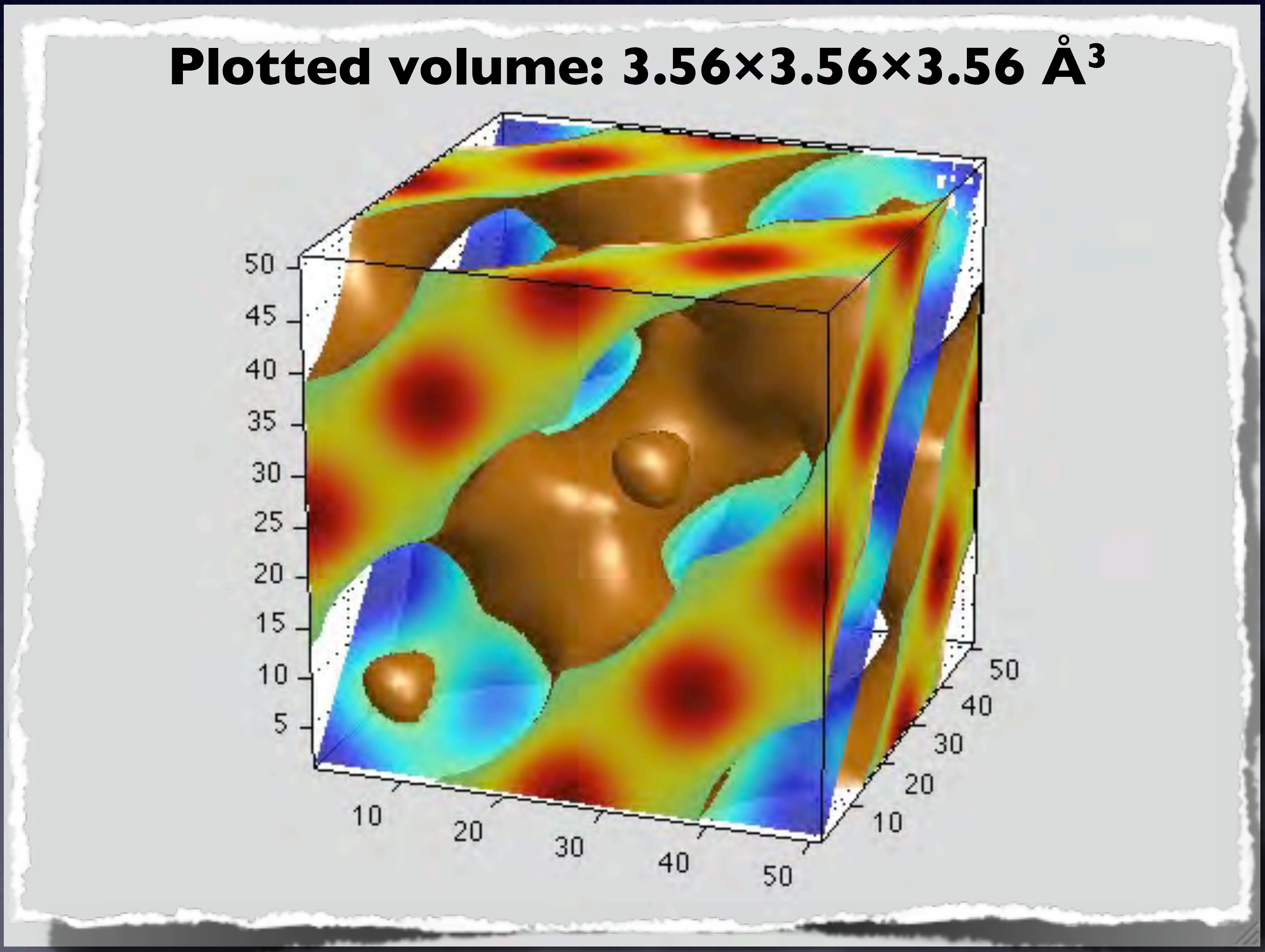
*cf. optical response at 100 eV*

# Reconstruction of $\chi^{(2)}(r)$

$|\chi_Q^{(2)}|$ : measured

$\arg(\chi_Q^{(2)})$ : phase of valence charge ← *Fano effect (interference with Compton scattering by valence electrons)*

$$\chi^{(2)}(r) = \sum_Q \chi_Q^{(2)} \exp(-iQ \cdot r)$$




Real space structure of **linear** susceptibility, $\chi(r)$, at 100 eV (124 Å) with angstrom resolution!

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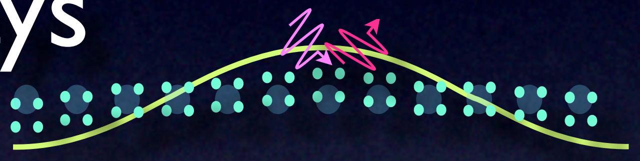
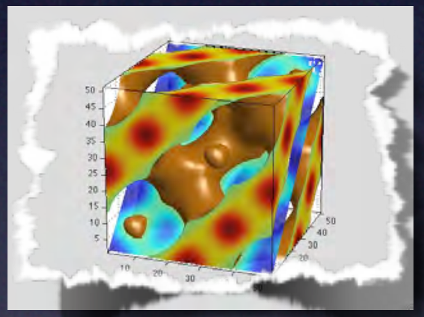
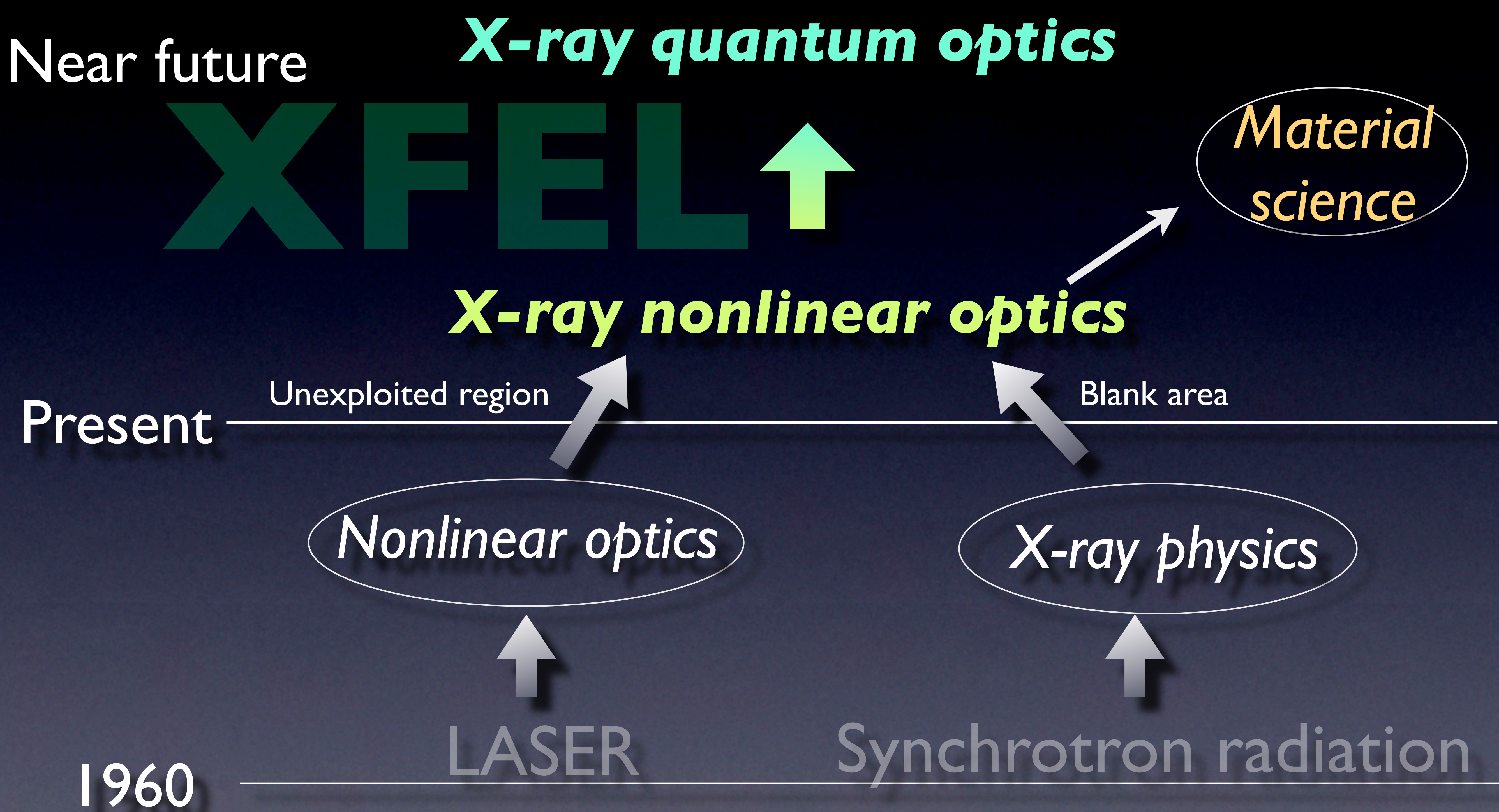


3. Nonlinear susceptibility of diamond



***4. Future perspectives***

# Perspective of XNLO



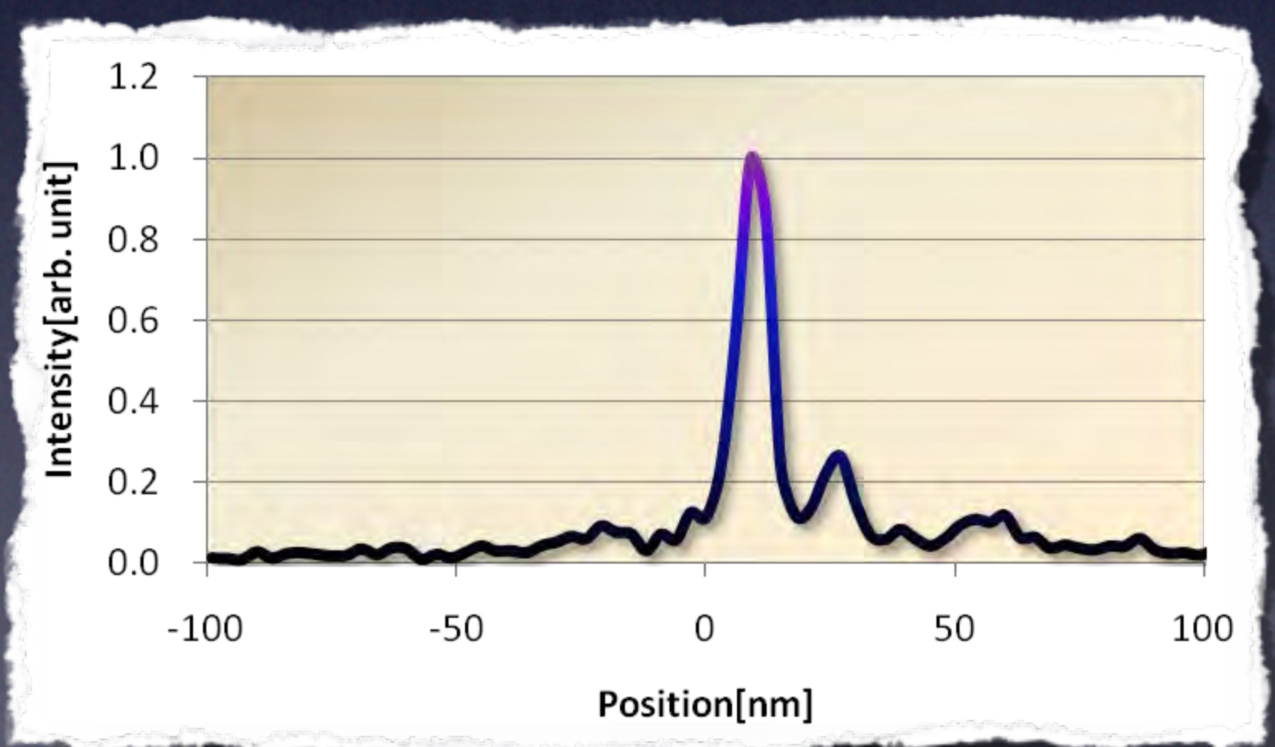
# Nonlinear response of vacuum - Ultimate NLO

Schwinger limit : $I_{QED}=4\times10^{29}$ W/cm$^2$

*Highest electric field in vacuum.*

0.1 nm fucusing ⇒ 40 TW is sufficient to **boil vacuum**.

cf. XFEL : 30 GW (2011)




Smallest focal spot :
7 nm @ 2010
(Osaka Univ. Yamauchi lab.)