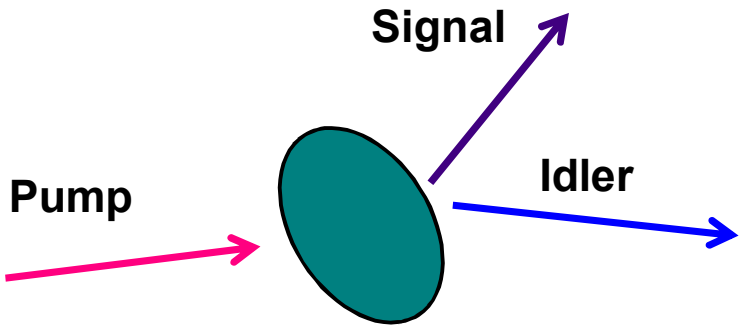
X-ray nonlinear optics with XFELO

Kenji Tamasaku (RIKEN)

1. X-ray PDC
2. Nonlinear spectroscopy

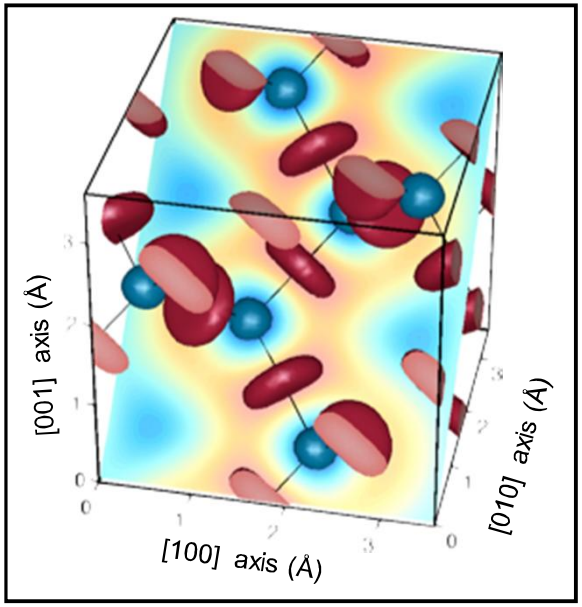
X-ray parametric down-conversion into SX, EUV, VUV, UV, visible, infrared regions

Parametric Down-conversion
(2nd order nonlinear process)



PDC can reveal optical response at longer wavelength with the atomic resolution. (Sum-frequency generation with synchronized Ti:Sa laser also gives the same information for UV,V and IR.)

Example)
Structure of linear susceptibility, χ(r), at 207 Å with 0.54-Å (=λ/380) resolution.



Sample: diamond
E_1(pump): 11.107 keV
E_3(idler): 60 eV

K.Tamasaku et al., Nat. Phys. 7, 705 (2011)

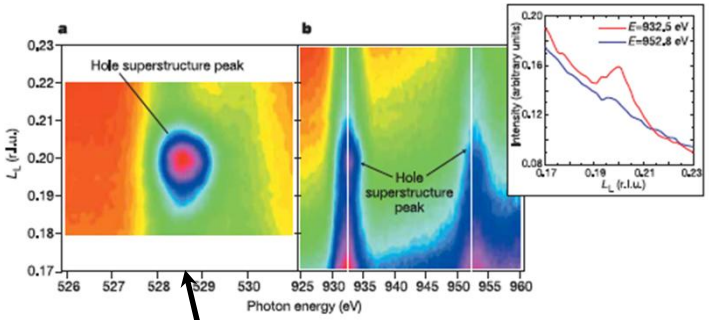
X→X+SX PDC for materials science

Ex) Investigation of charge order

- X-ray and neutron diffraction sees mainly lattice distortion (not charge order).
- SX diffraction is powerful tool.

But,

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

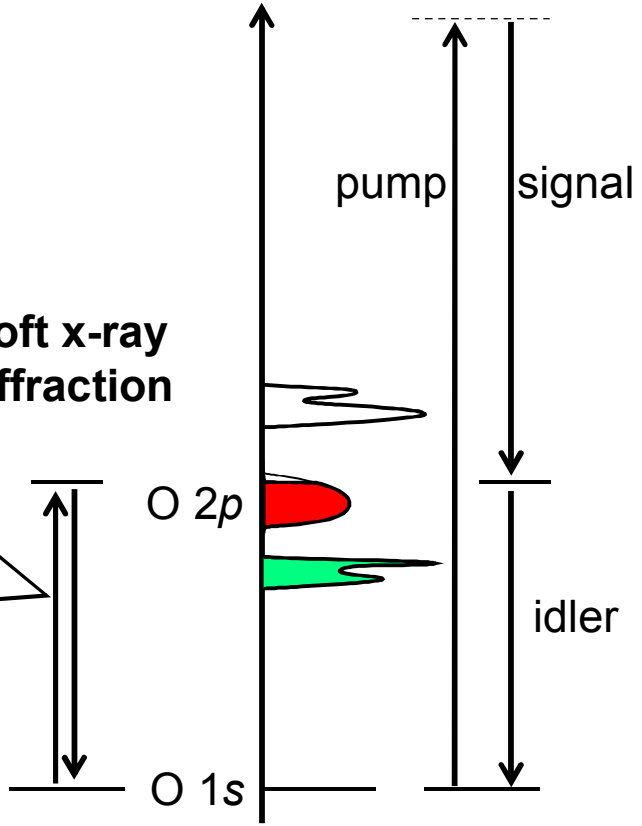


Oxygen *K*-edge

23 Å : *not enough resolution*

Sample environment is not flexible due to absorption.

X-ray PDC
can realize Å resolution
at O 1s→2p resonance



We need spectral brightness of XFELO!

Is nonlinear spectroscopy possible with XFELO?

SACLA
1-μJ & 5-fs pulse w 1-μm^2 focusing
Two-photon absorption is measurable.

XFELO:
10^9 ph/pulse, 5 keV ~ 1 μJ
500-fs pulse w 100-nm^2 focusing
may allow observation of TPA.

We can consider nonlinear spectroscopy with XFELO

- X-ray two-photon absorption spectroscopy
  to access the 1s-3d transition.
- Stimulated Raman scattering(?)
  to measure XAS of lighter materials.
  Need for two-color X-rays (XFELO+Spontaneous X-rays).