Single-bunch Operation, Generation of Ultra-short Pulses at Storage Ring
And their Applications *(KEK International Center, Feb.28-Mar.1, 2005)*

A proposal for new pump-probe by combination of high-intensity fs laser with synchrotron radiation

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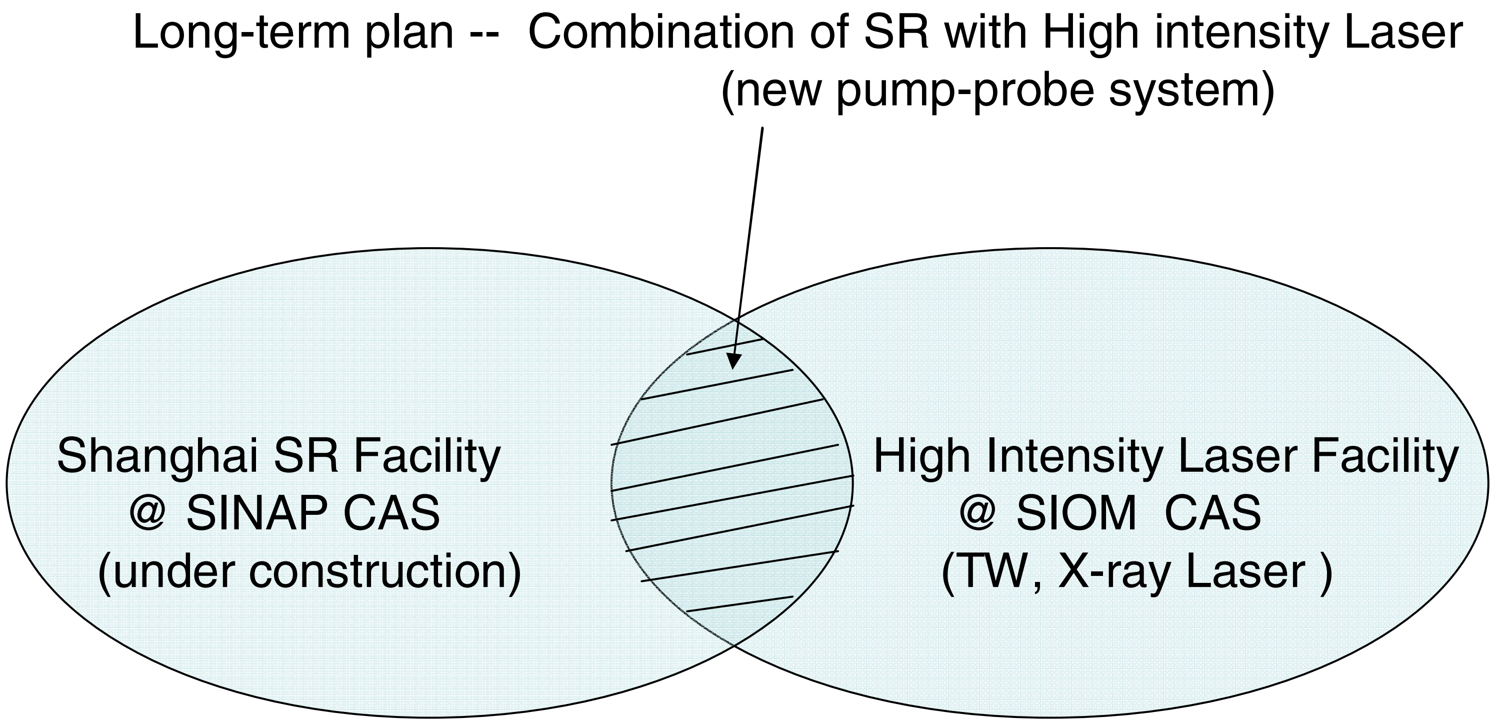
Speckles

Intensity correlation spectroscopy (JST/CREST)

Inner Shell 2-hole excitation (JST/CREST)

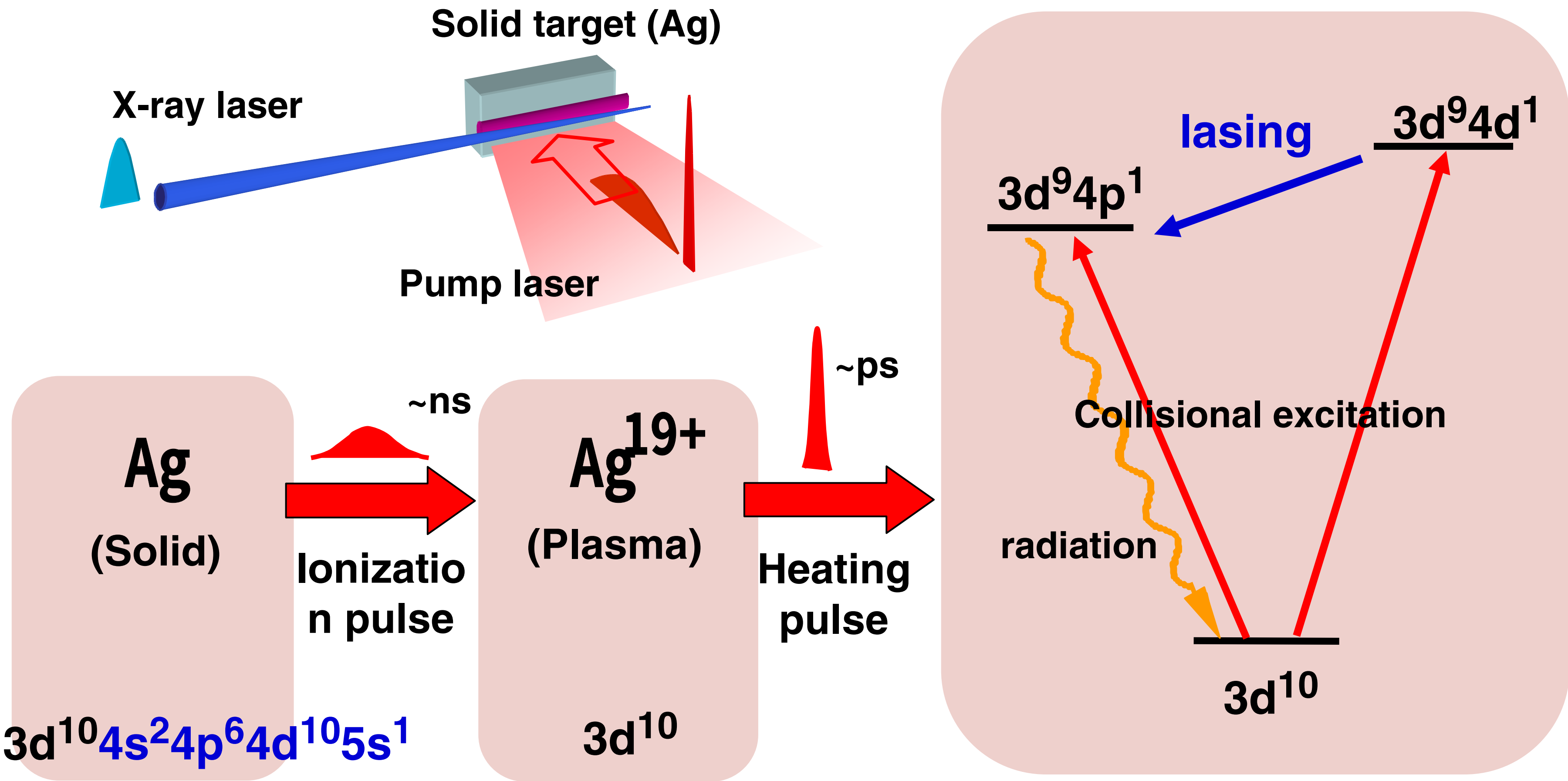
○ New Pump-probe system

Two Big CAS Facilities in Shanghai



Plasma based X-ray Laser

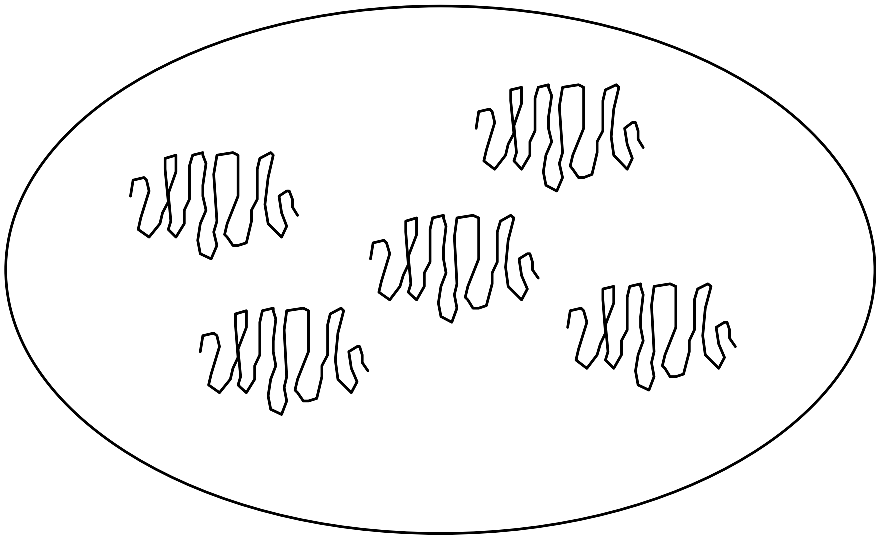
X-ray laser is generated with transient collisional excitation scheme



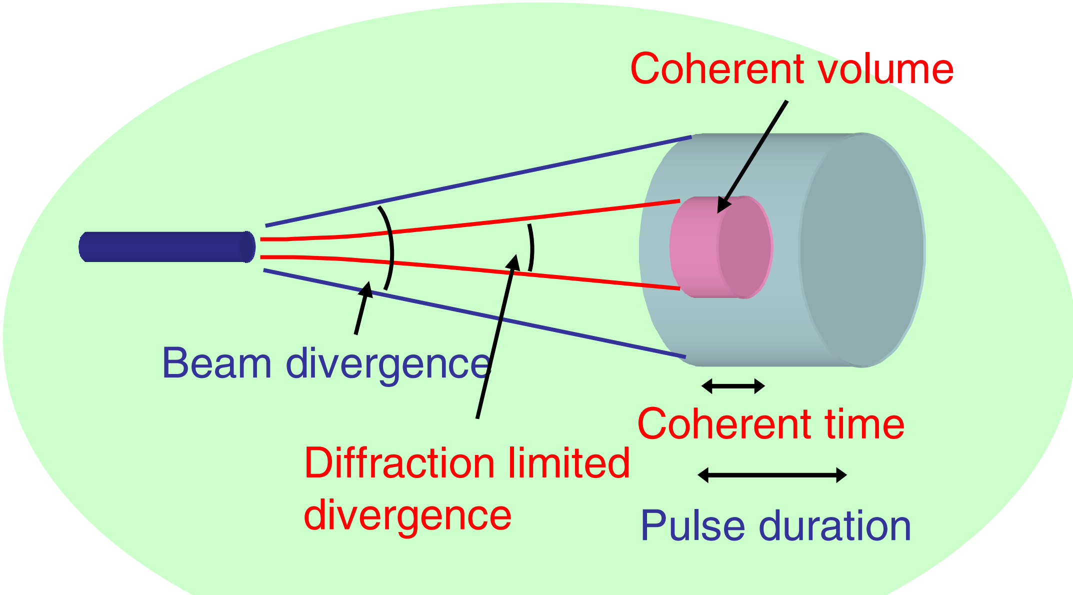
Two pulse is used for pumping pulse to generate high gain transiently.

Plasma based X-ray Laser

Similar to Single pulse of SASE FEL



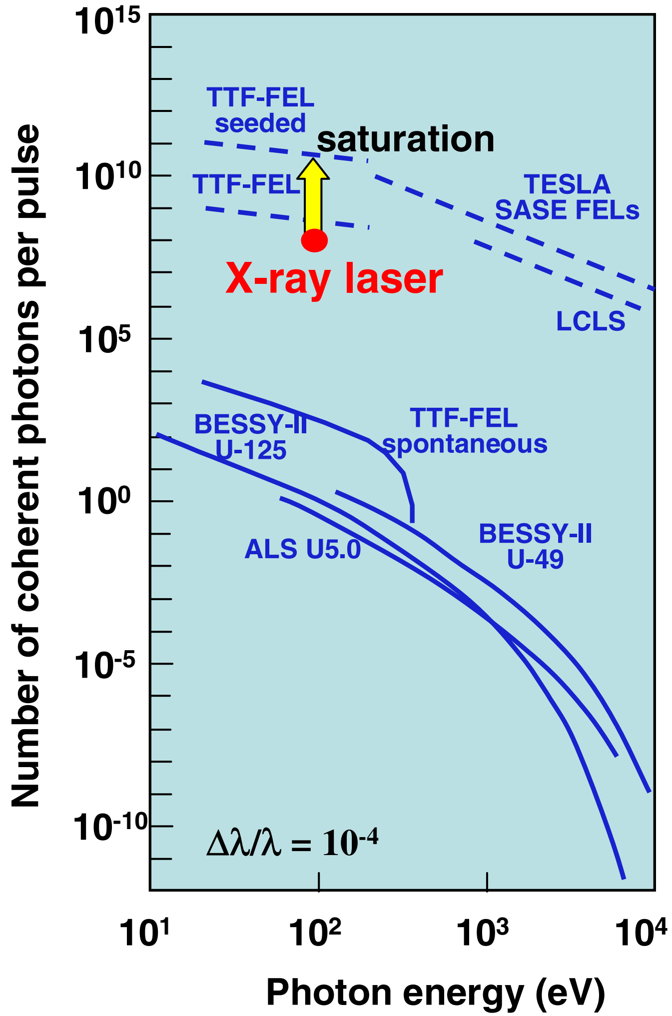
Number of Coherent photon



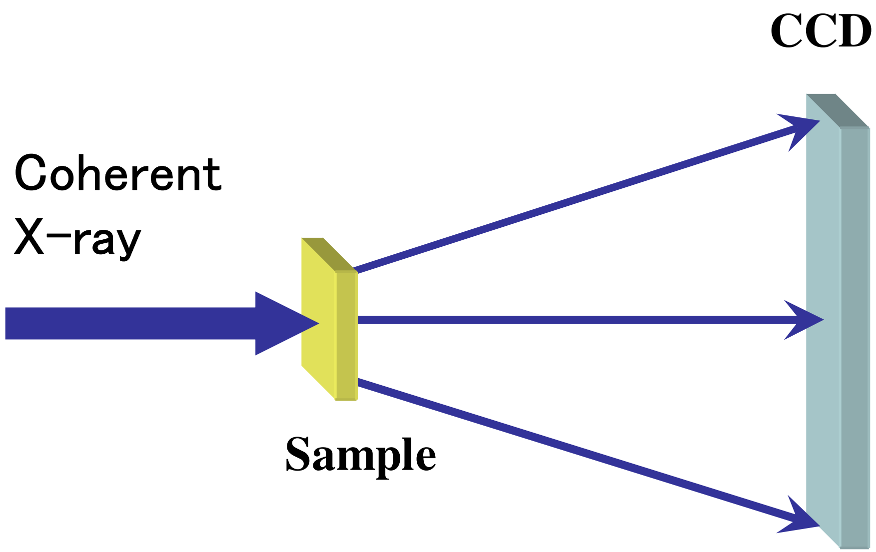
Number of coherent photons
= Number of the photons in the coherent volume

10^8 coherent photons / pulse

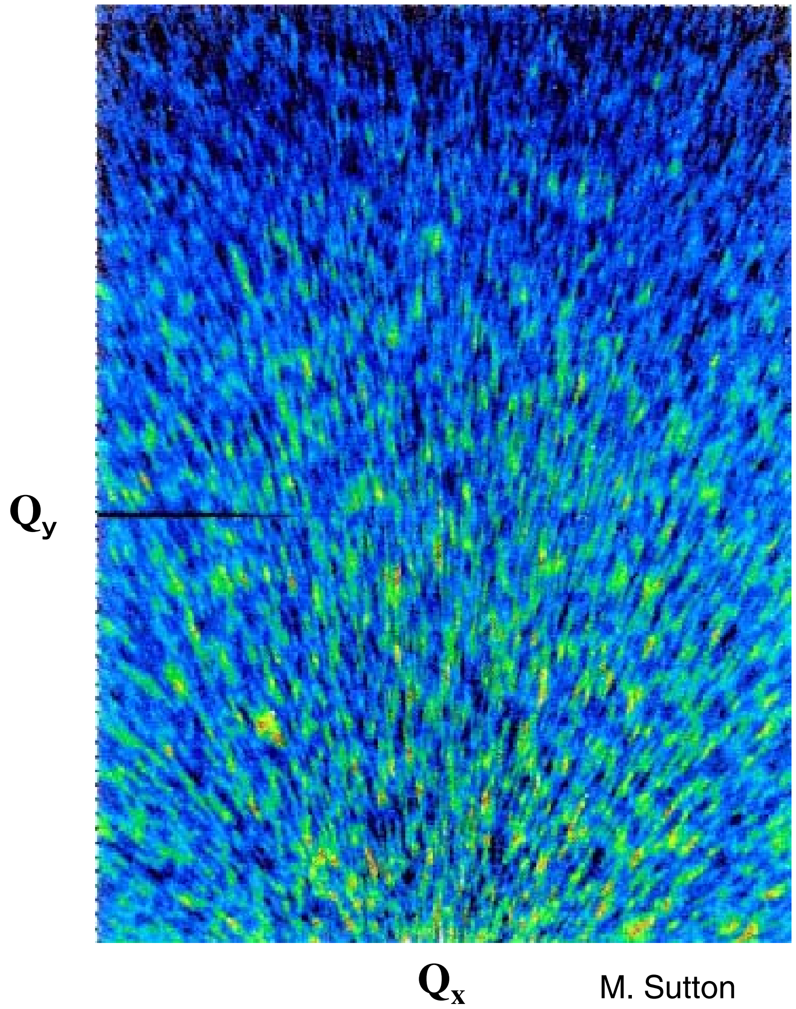
Higher than the SOR x-rays !



X-ray Speckle



Inhomogeneous structure
In space and time



M. Sutton

Space time correlation function

$$G(r,t)=\frac{1}{N}\left\langle \sum_{i,j}^{N}\int_{-\infty}^{\infty}dr'\,\delta(r+r_i(0)-r')\,\delta(r'-r_j(t))\right\rangle$$

Dynamic structure factor

$$S(Q,\omega)=\int_{-\infty}^{\infty}dr\int_{-\infty}^{\infty}dt\,\exp i(Qr-\omega t)G(r,t)$$

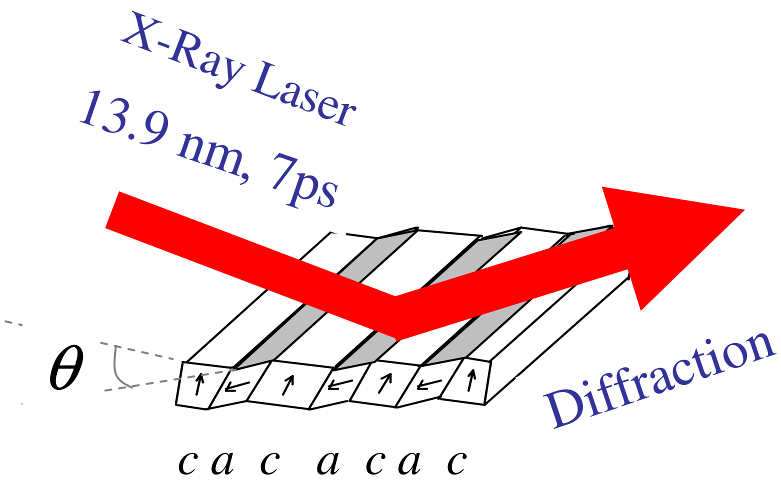
$$S(Q,t)=\frac{N}{2\pi\hbar}\int_{-\infty}^{\infty}\exp(iQr)G(r,t)dr$$

Speckle

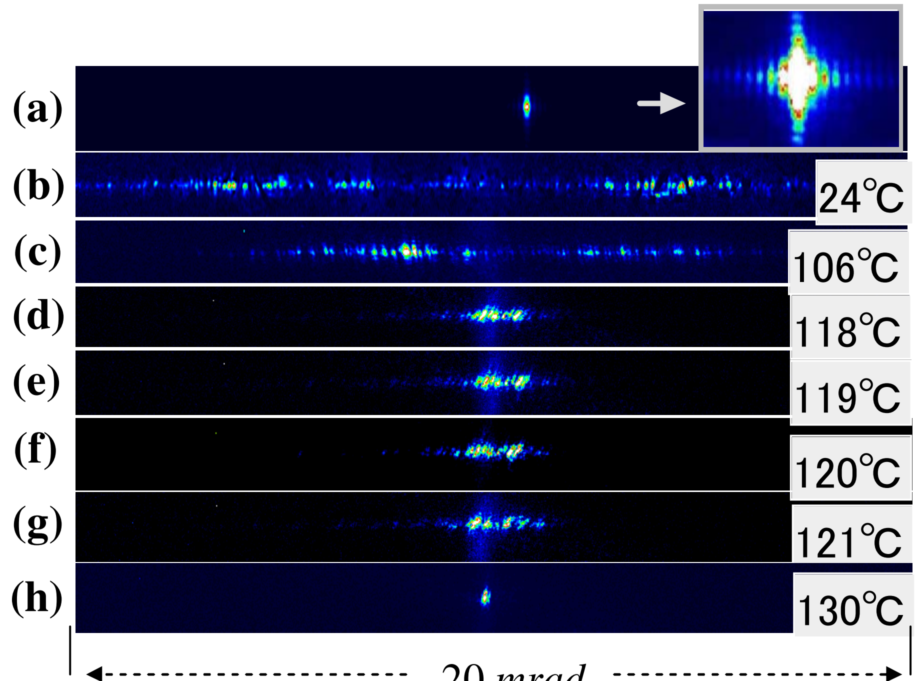
$$S(Q,0)=\frac{N}{2\pi\hbar}\int_{-\infty}^{\infty}\exp(iQr)G(r,0)dr$$

Picosecond X-Ray Speckles below T_C

$T_c = 122^oC$



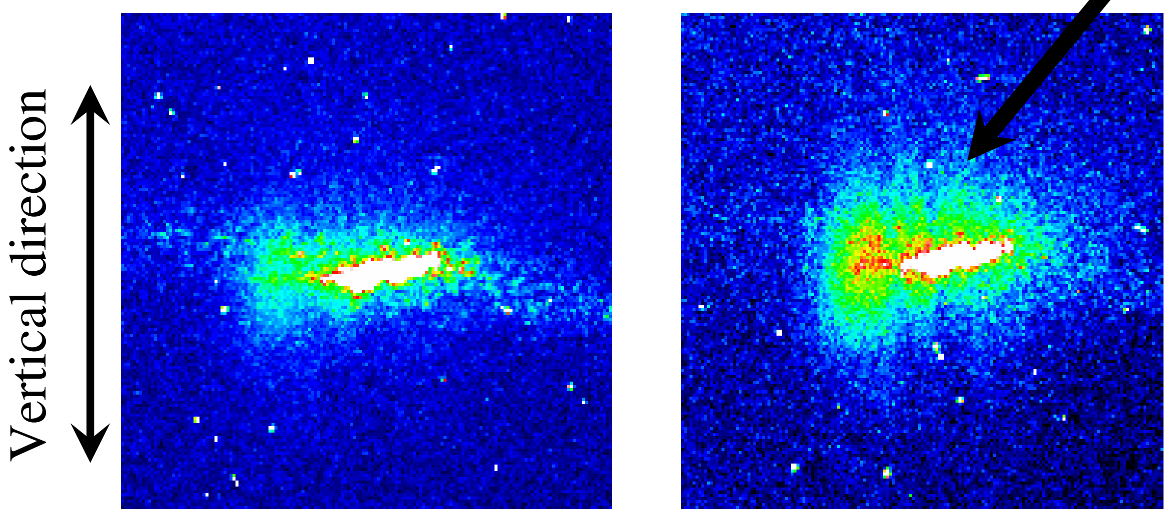
$BaTiO_3$ with *a/c* 90^o domains



Tai et. al., Phys. Rev. Lett. 89, 257602 (2002)

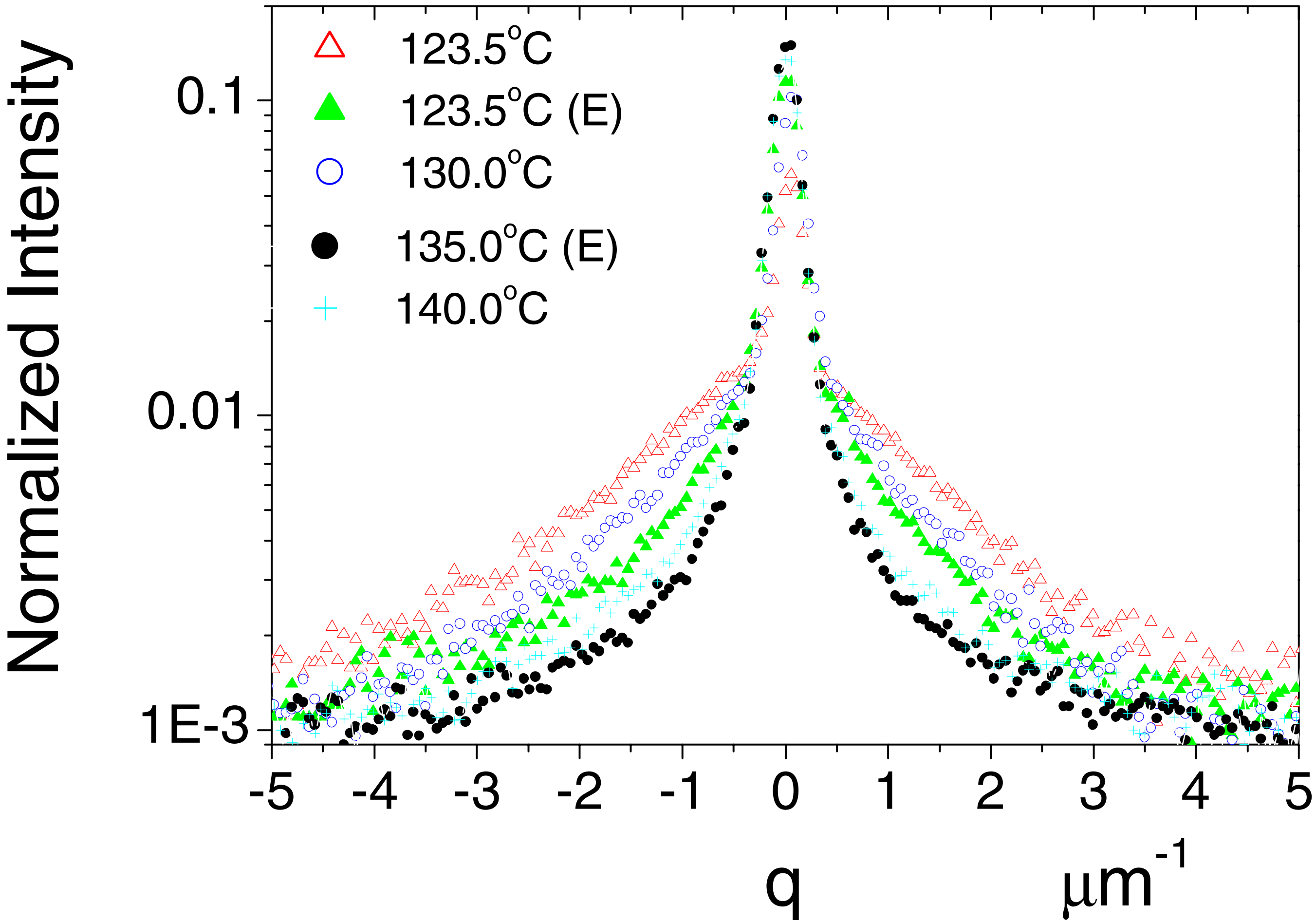
Picosecond X-Ray Speckles above T_C

$T_c = 122^oC$

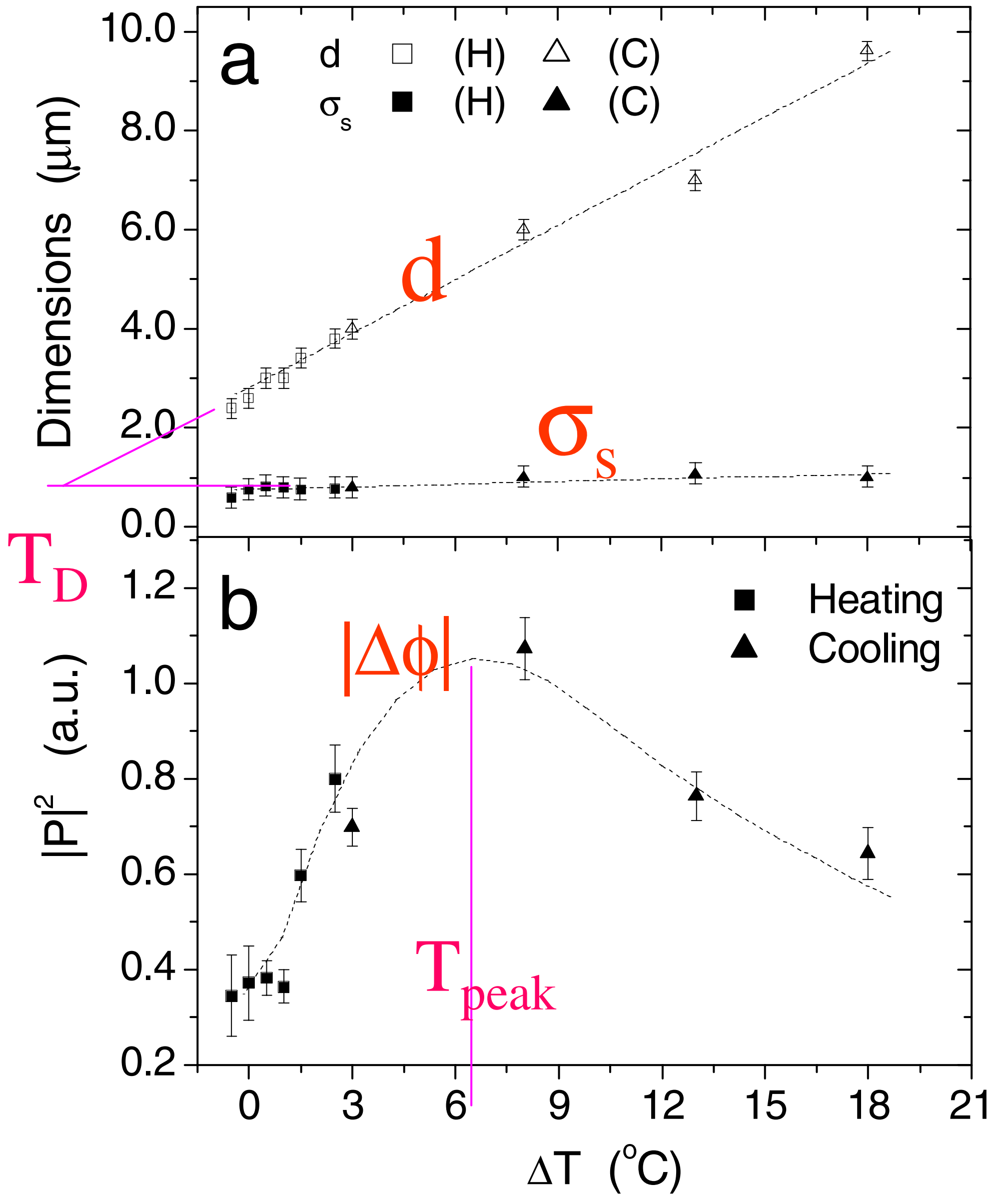


140 °C   130 °C

Profile of the x-ray beam scattered in the vertical direction



Characteristic Cluster Parameters above T_c



(1) T_D: a temperature where dynamic Clusters have condensed into the the ferroelectric domains

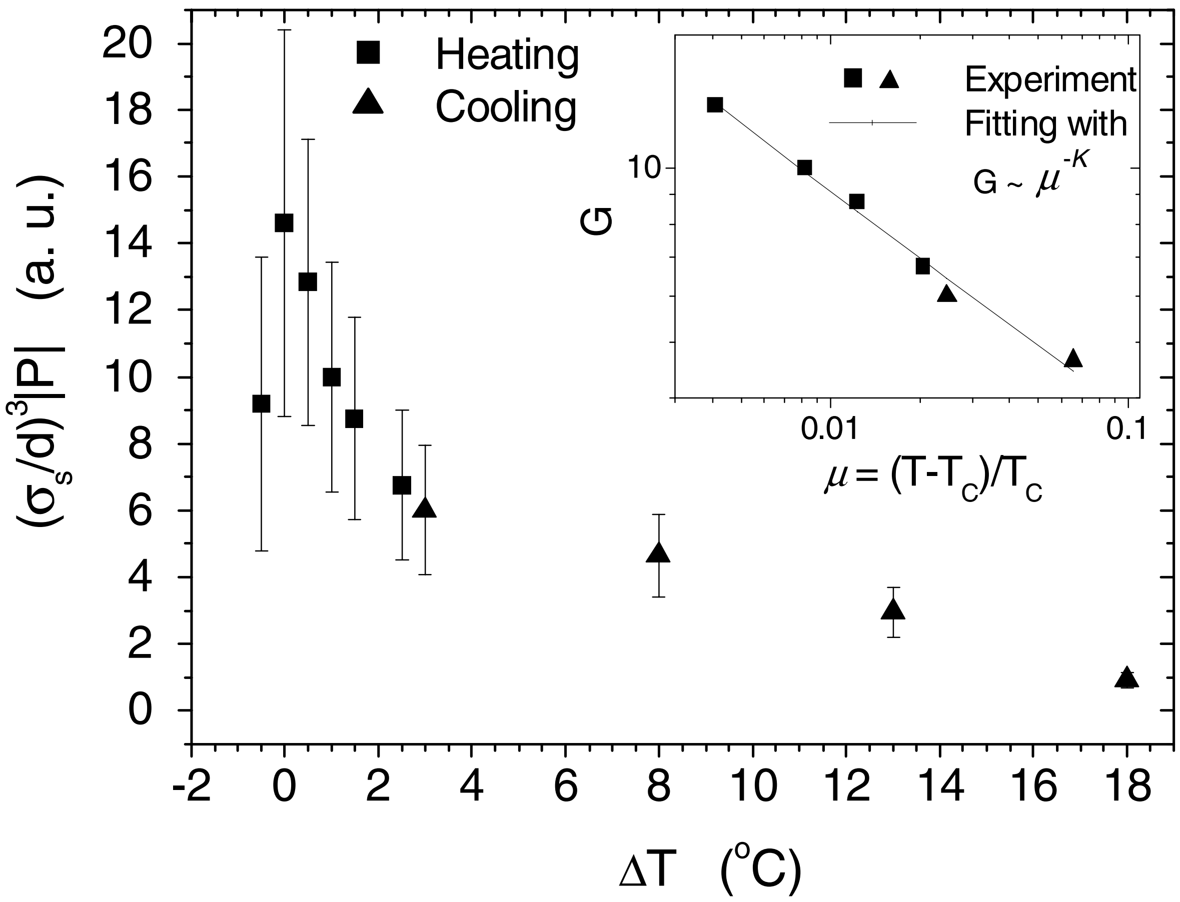
(2) The increase of $|P|^2$ indicates the increase of coherent motion among Ti ions.

(3) T_{peak} indicates a temperature where crossover from displacive to relaxational phase transition occur, since the increase of the fluctuation among off-center sites will decoherence the polarization.

Quadratic Kerr effect

$|P|^2 \sim |\Delta\phi| = |\Delta n|$

Macroscopic Polarization Fluctuations
(Clusters' Short-Range Correlation Strength)



- ★ Macroscopic average: $(\sigma_s/d)^3\,|P|$

⇕

$G = (1/d)^3\,(\sigma_s\,|P|)$

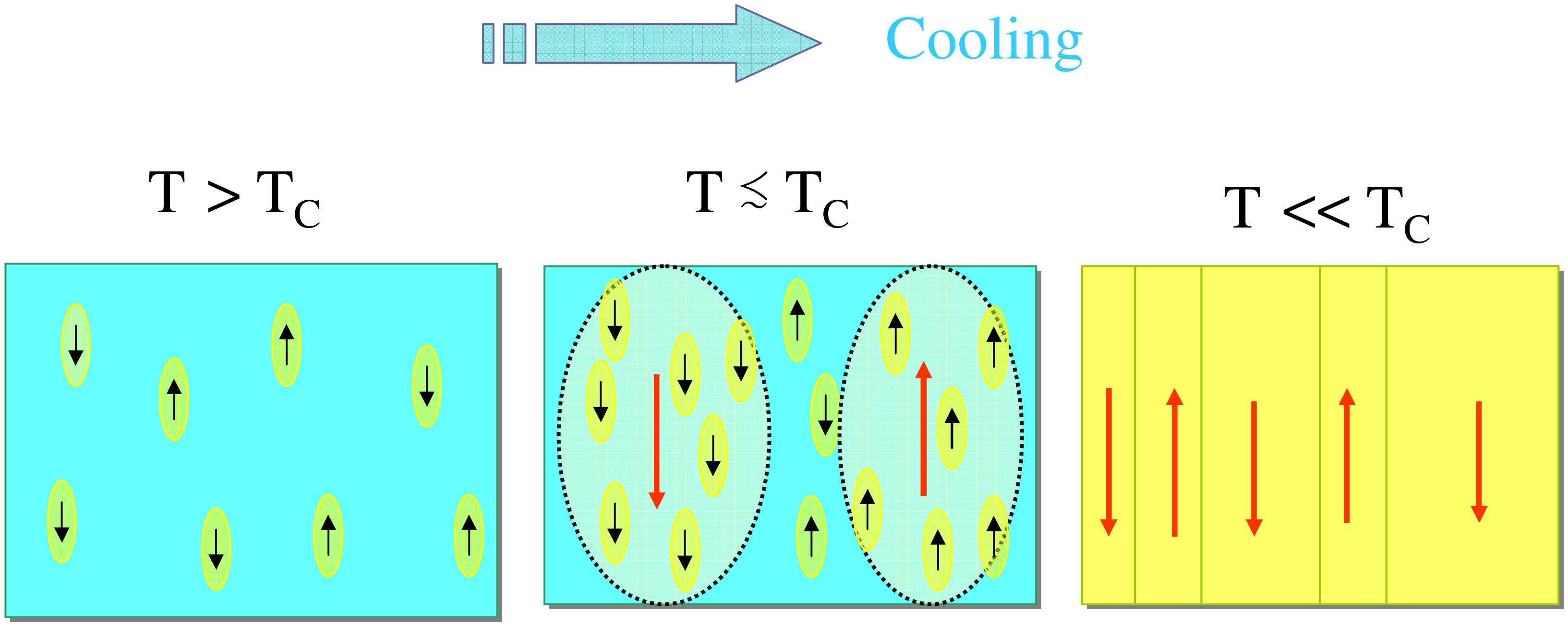
- ★ Cluster' dipole potential

- ★ From fitting $G \propto (T-T_c)^{-0.41\pm0.02}$

Proposed Image of Phase Transition for $BaTiO_3$

Cooling

$T > T_C$ $\qquad$ $T \lesssim T_C$ $\qquad$ $T << T_C$



$< P >_t = 0$ $\qquad$ $< P >_t \neq 0$

Clusters in Paraelectric Phase

Bunch of Clusters

Ferroelectric domain

Intensity correlation Spectroscopy

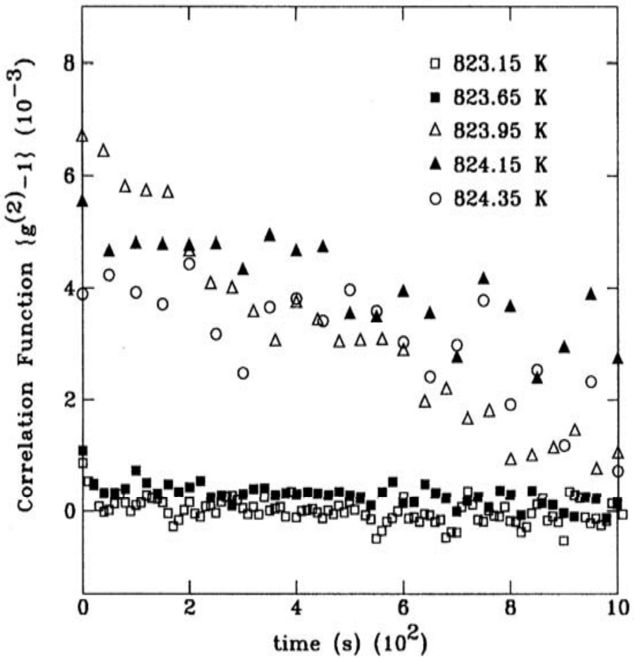
Intensity correlation

$$g^{(2)}(Q,t)=\frac{\left\langle I(Q,t'+t)I(Q,t')\right\rangle_{t'}}{\left\langle I(Q,t')\right\rangle_{t'}^{2}}$$

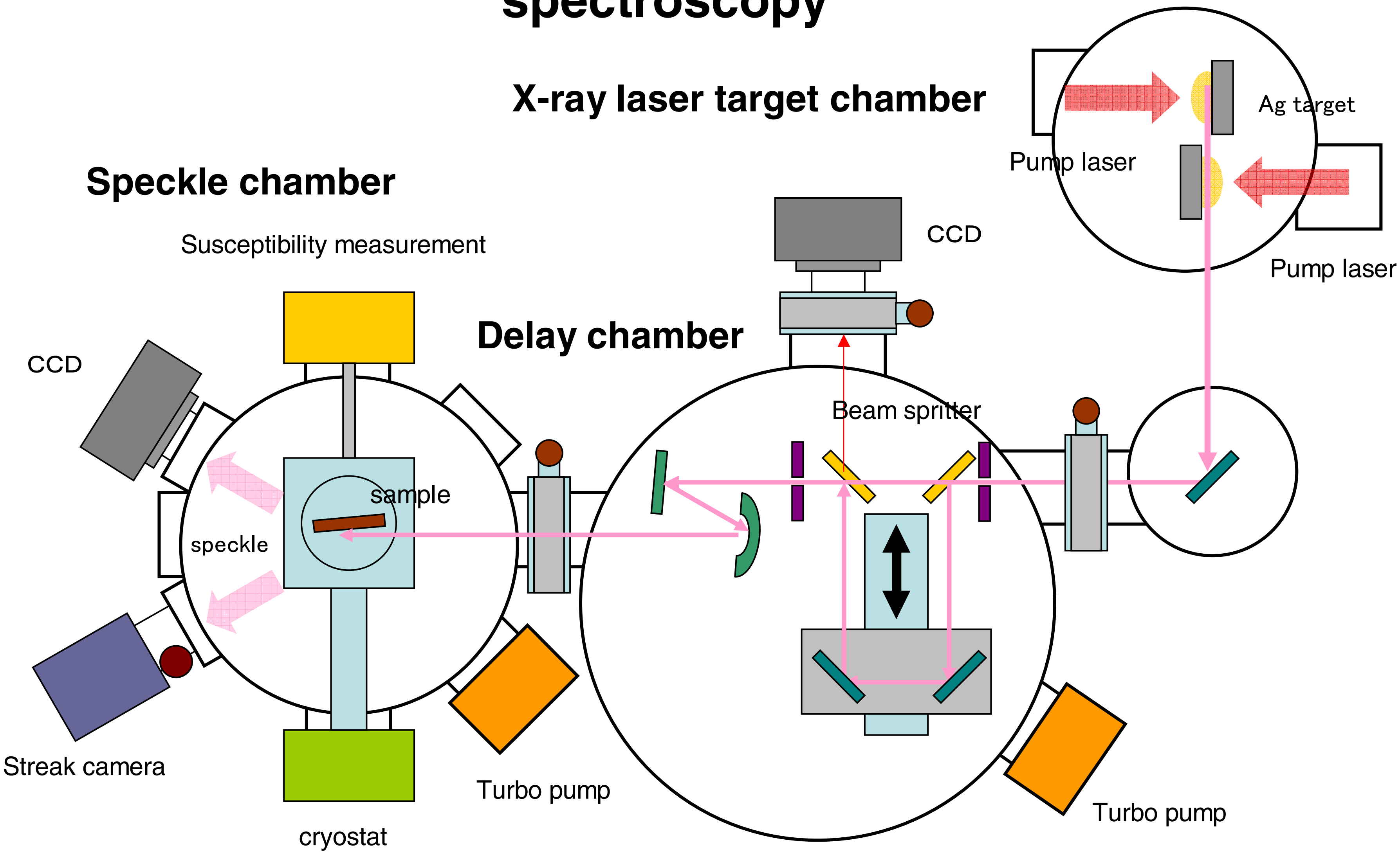
Simple case

$$g^{(2)}(Q,t)=1+\beta\left|\exp\left(-\frac{t}{\tau(Q)}\right)\right|^{2}$$

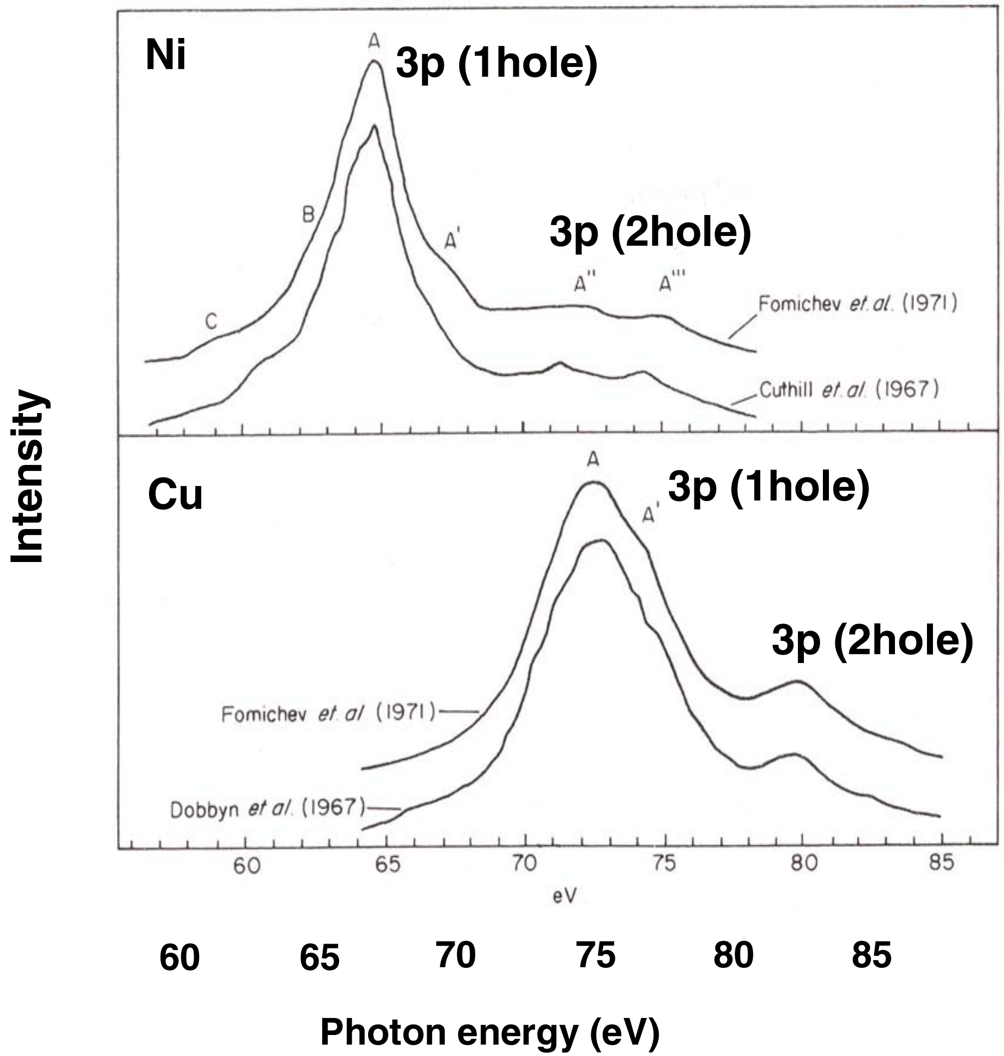
Fe_3Al

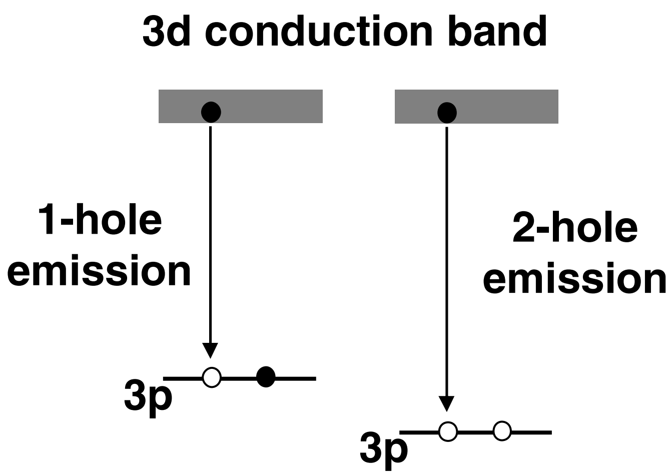


Experimental equipments for Intensity correlation spectroscopy

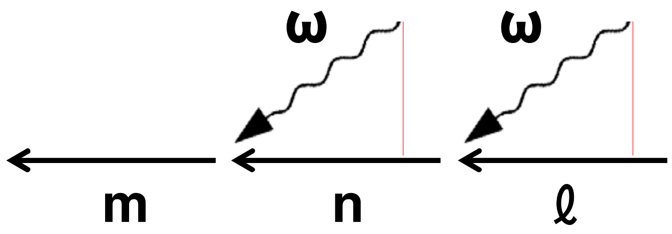


Inner shell Two hole excitation by electron impact





Two hole excitation by 2 photons

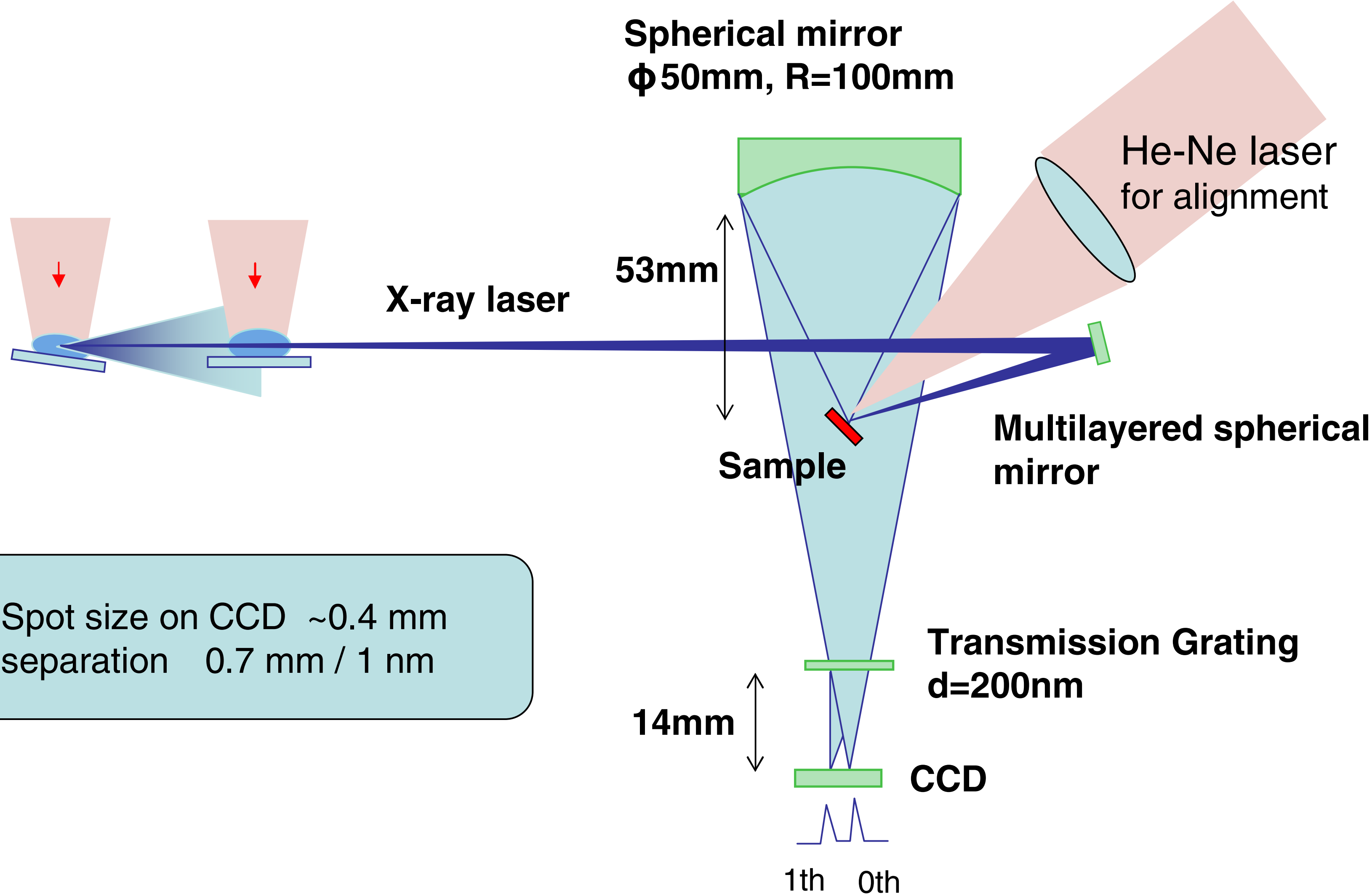


$$\frac{w_2}{w_1} = 4\alpha\left(\frac{\hbar\omega}{\Gamma_n}\right)^2 \lambda |\boldsymbol{x}|^2_{mn} \frac{n}{V_c}$$ **= 10^{-3}**

$$\alpha = \frac{e^2}{4\pi\varepsilon_0\hbar c}$$

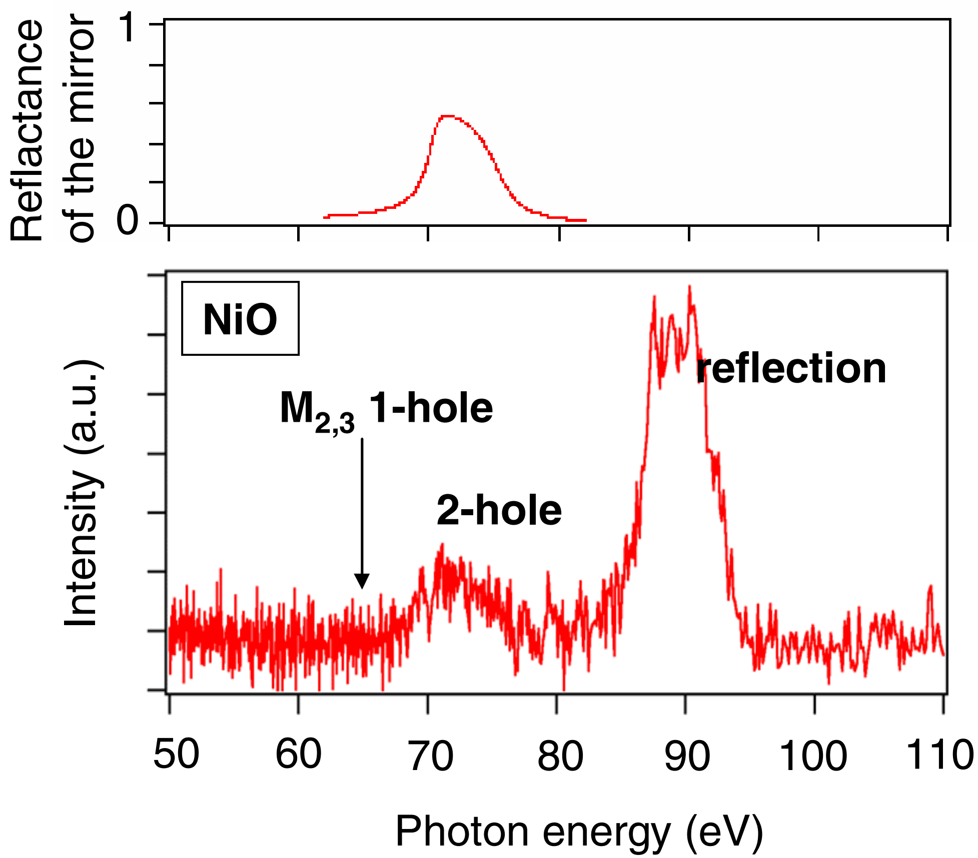
n coherent photon 10^8個
Vc coherent volume $(100\mu m)^3$
Γ n level width 0.1eV
|x| atomic radious 1nm

Experimental set up

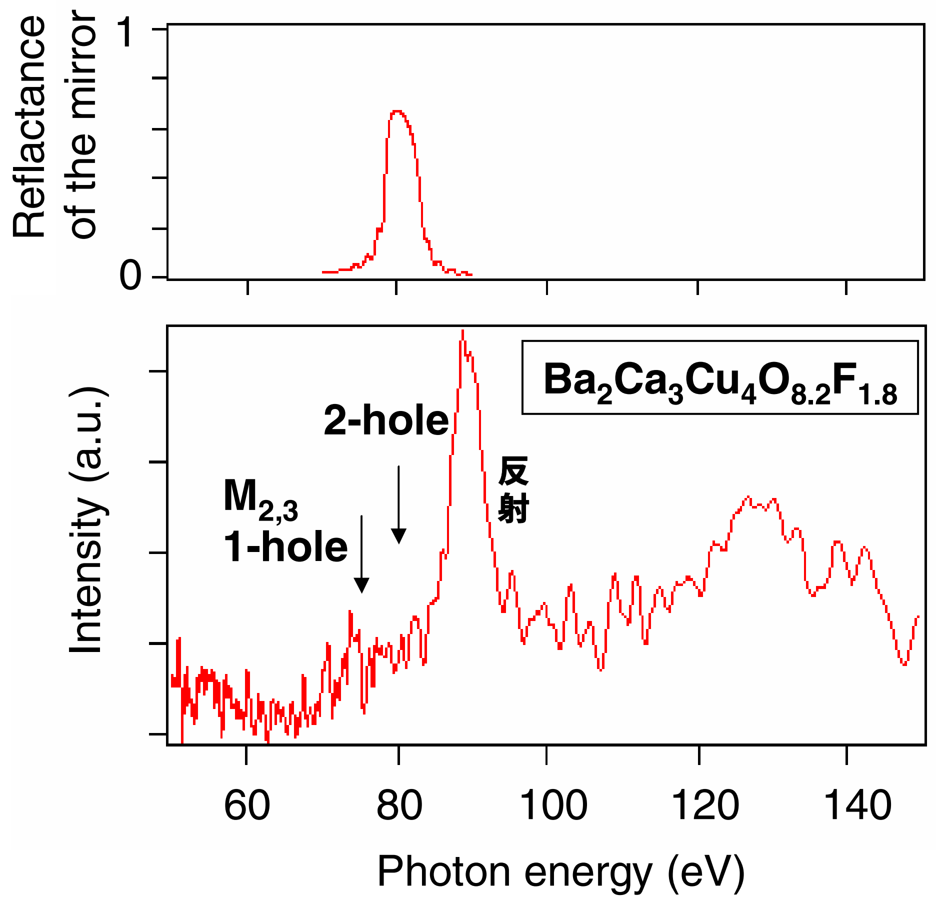


Spot size on CCD ~0.4 mm
separation 0.7 mm / 1 nm

Fluorescence spectrum after 2 hole excitation
(Strongly correlated electron system)

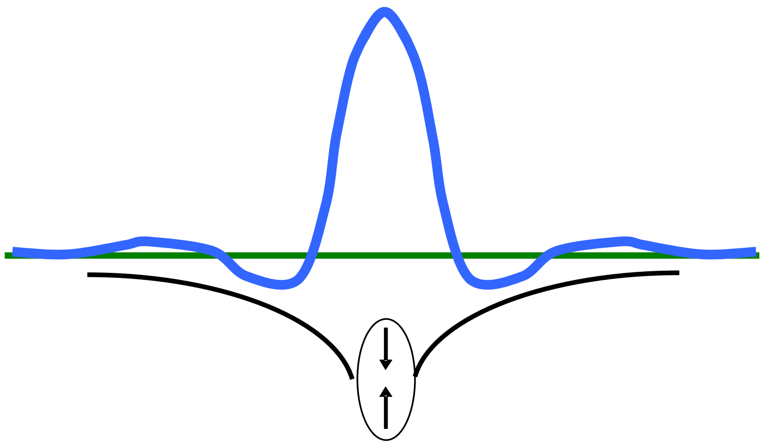


Fluorescence spectrum after 2 hole excitation (High Tc superconductor)



Polarization of 3d electrons due to two holes in 3p orbital

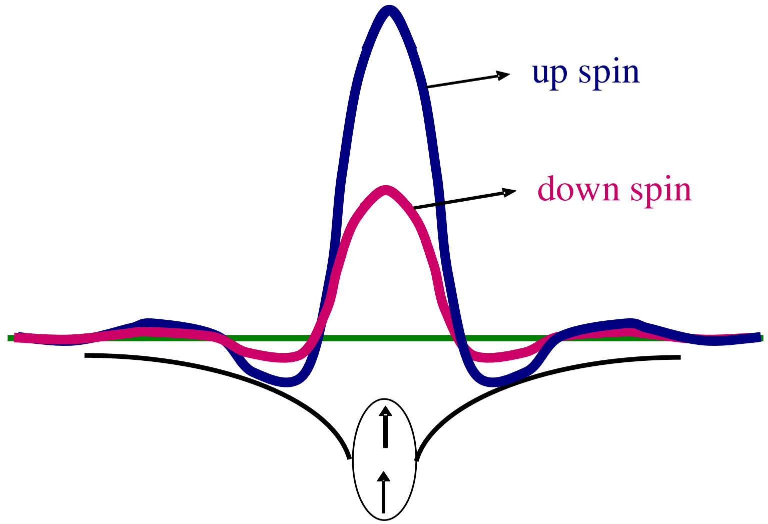
Polarization in Charge



2e+ Coulomb attractive force

Singlet two 3p holes

Polarization in Charge and spin



2e^{+} Coulomb attractive force

Triplet two 3p holes

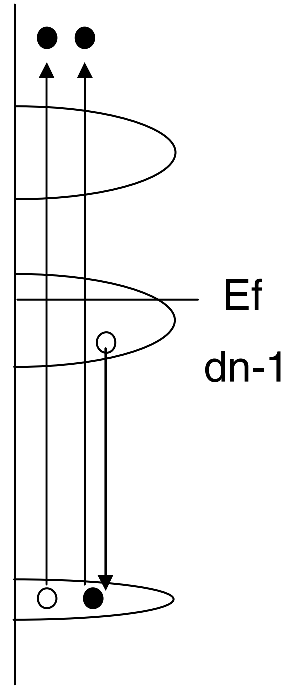
2-holes are created in an inner shell of the same atom

New developments by means of 2 hole excitation spectroscopy

Difference in emission spectrums from triplet state and from singlet state reflects the exchange parts of 3d hole and 3p hole at final state.

From this difference, we can understand the orbital distribution in 3d LHB.

（These information are inaccessible by 1hole spectroscopy)



New Pump-probe system
- combination of SR and High intensity Laser-

What kinds of new science will come ?

Basic Considerations

(1) X-ray Laser: pump

Excitation to inaccessible states by SR or visible laser
Example: Inner shell 2 hole excitation

SR: probe

Physics, Chemistry, … of excited state.
Relaxation process etc.

(2) SR: pump

Non thermally excited states
Use of resonance

fs Laser: probe

Instantaneous observation of dynamic process

Possible Combinations

XRL Pump - SR Probe
Highly-excited state physics (chemistry, material science)

SR Pump - fs laser Probe
Ultra-fast excited state physics (chemistry, material science)

Thank you for your attention !