



# Cristallographie et Grands Equipements

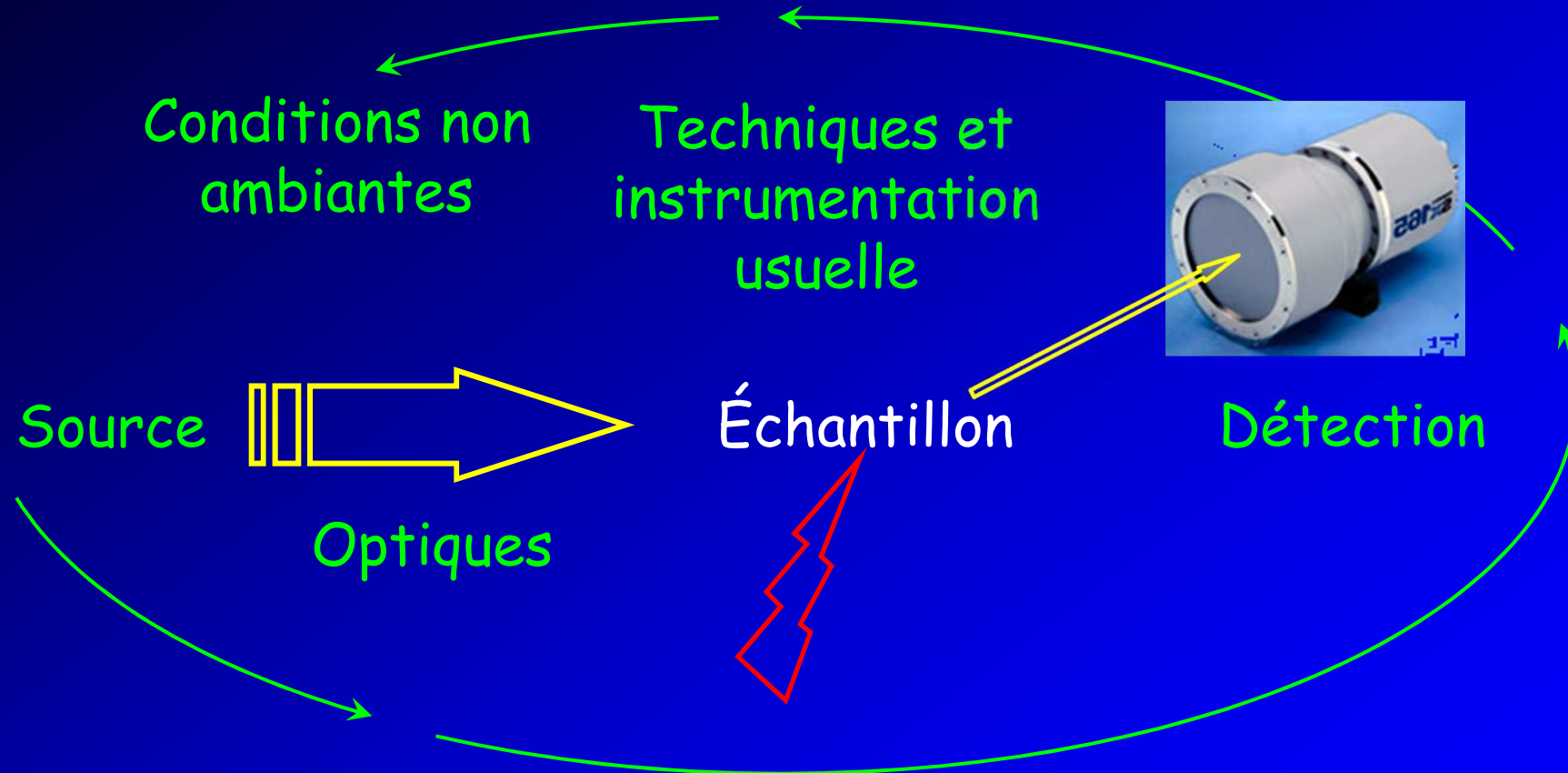
## Méthodes expérimentales : Rayons X



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# Sommaire



# Les sources de Rayons X

comparer différentes sources

produire des Rayons-X

Source de laboratoire

Grands Instruments

# Accès

Lab. source 0.3 m



Synchrotron 100 m



X-fel 1000 m

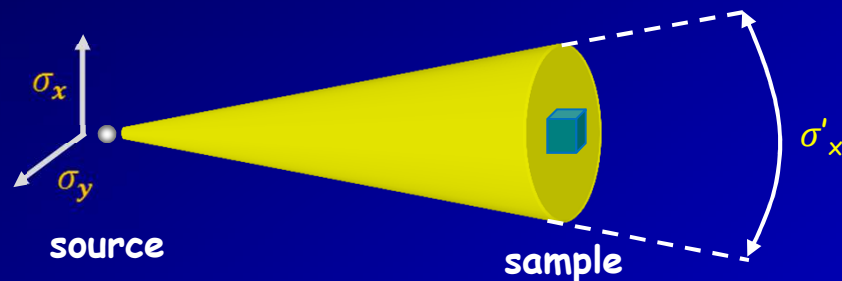


# How to compare X-ray sources?

## spectral brilliance/brightness:

Photon flux based on the source size and divergence  
per  $\Delta E/E = 0.1\%$  bandwidth

ph/s/mrad<sup>2</sup>/mm<sup>2</sup>/0.1%BW



Flux (ph/s)

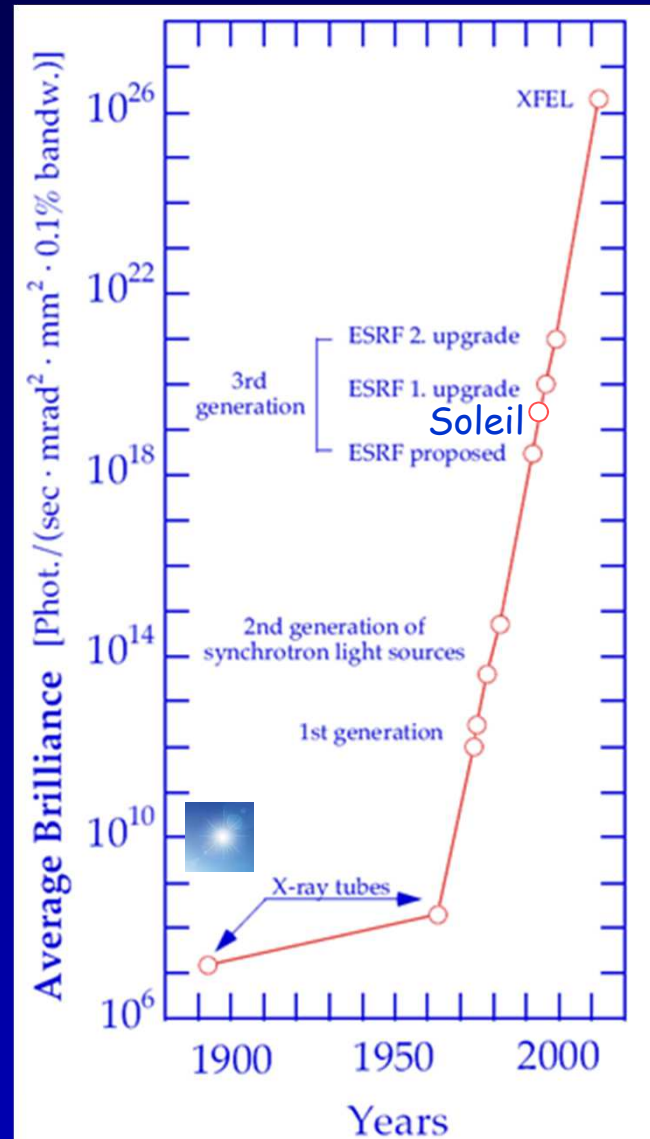
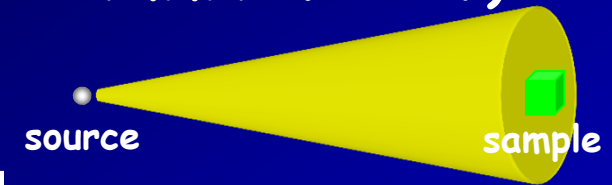
Energy (keV) or Wavelength ( $\text{\AA}$ )

Source size  $\sigma_x, \sigma_y$  (mm)

Divergence  $\sigma'_x, \sigma'_y$  (mrad)\*

\*(1 mrad  $\sim$  0.06 $^\circ$ )

# Source de rayons X: brillance (ph/s/mrad<sup>2</sup>/mm<sup>2</sup>/0.1%BW)



**Sources de laboratoire**

# Sources de laboratoire

## Tubes à rayons X

principe:

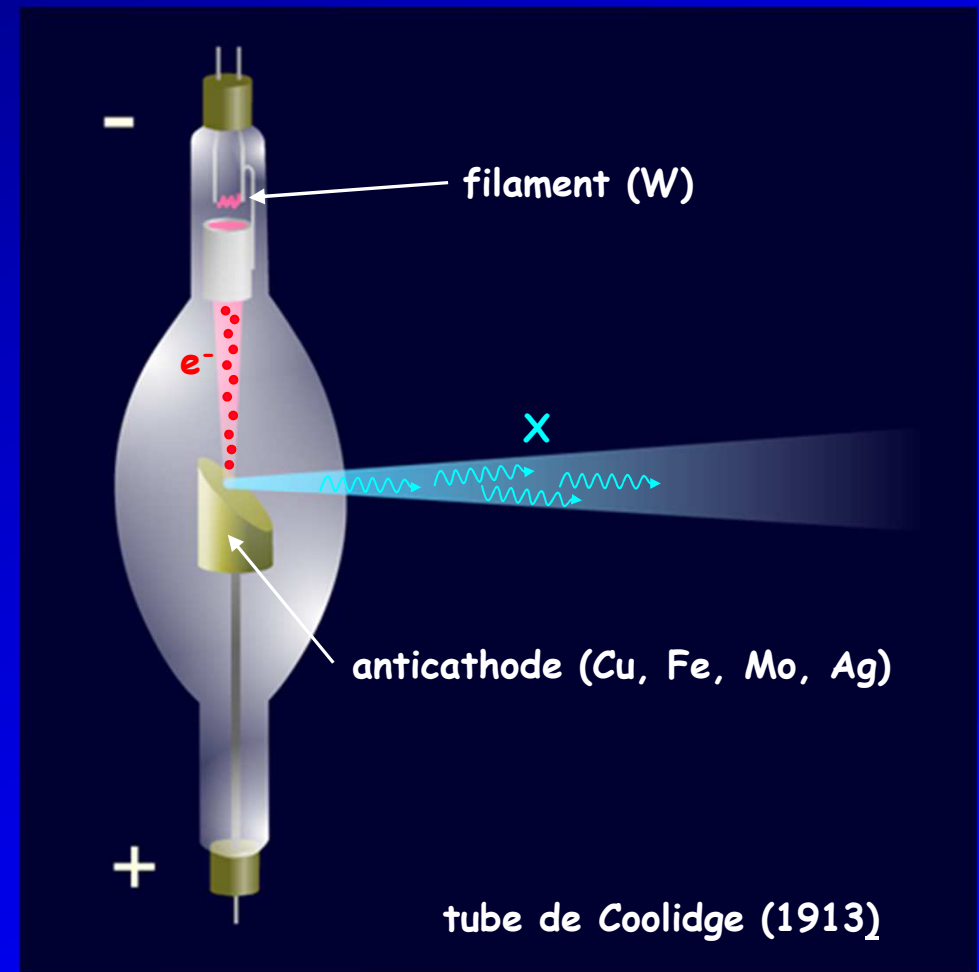
$$\eta = \frac{E(X)}{E(e^-)} \sim 10^{-6} Z V(\text{kV}) < 1\%$$

Tube scellé

Anode tournante

Micro-source

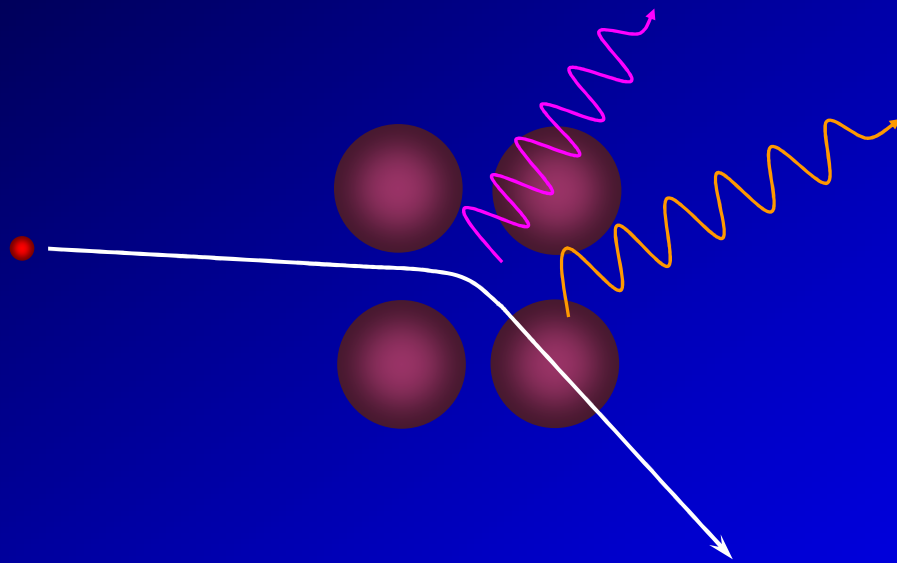
Jet de métal liquide



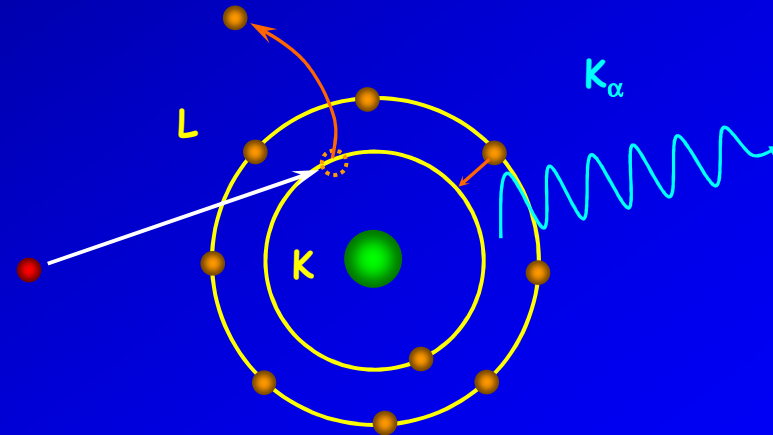


# Mécanisme de production des rayons X « de laboratoire »

rayonnement blanc + raies caractéristiques



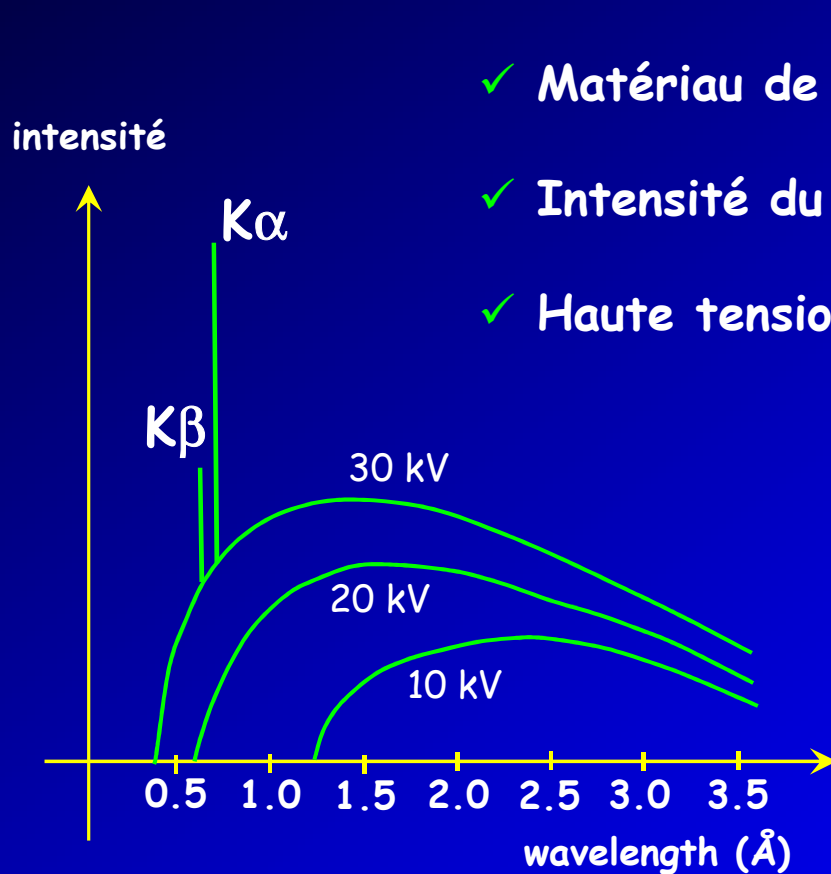
rayonnement de freinage  
Bremsstrahlung



effet photoélectrique

# Sources de laboratoire

## Paramètres des sources de laboratoire



✓ Matériau de la cible ( $V_K \leftrightarrow \lambda$ )

✓ Intensité du courant  $i$

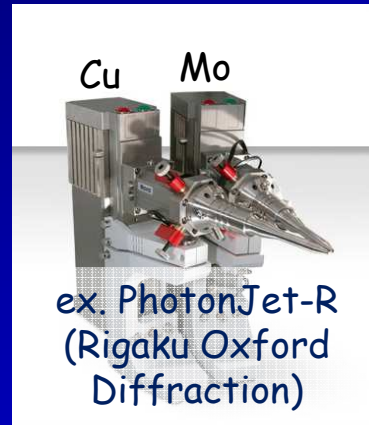
✓ Haute tension  $V$

	$\lambda(\text{Å})$		
	K $\alpha$ 1	K $\alpha$ 2	K $\beta$
Cr	2.294	2.290	2.085
Fe	1.940	1.936	1.757
Co	1.793	1.789	1.621
Cu	1.544	1.540	1.392
Mo	0.7135	0.7093	0.6323
Ag	0.5638	0.5594	0.4970

ex.: tube scellé: Mo (2kW)  $V_K = 20$  kV,  $i = 40$  mA,  $V = 40$  kV

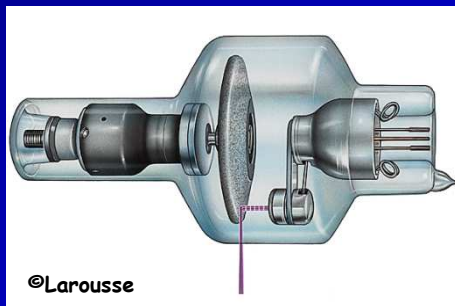
# Sources de laboratoire

## Tube scellé $\mu$ source



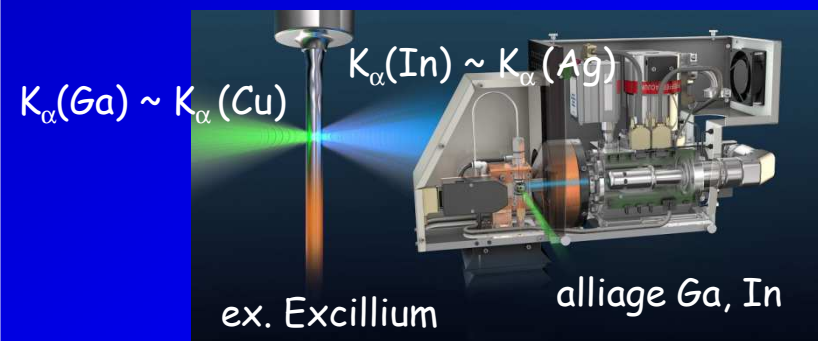
Flux  $4 \cdot 10^8$  ph/s  
Taille de source : 70-150  $\mu\text{m}$   
Brillance spectrale :  $3 \cdot 10^8$  UB

## Anode tournante



Flux  $4 \cdot 10^9$  ph/s  
Taille de source : 70  $\mu\text{m}$   
Brillance spectrale :  $2 \cdot 10^9$  UB

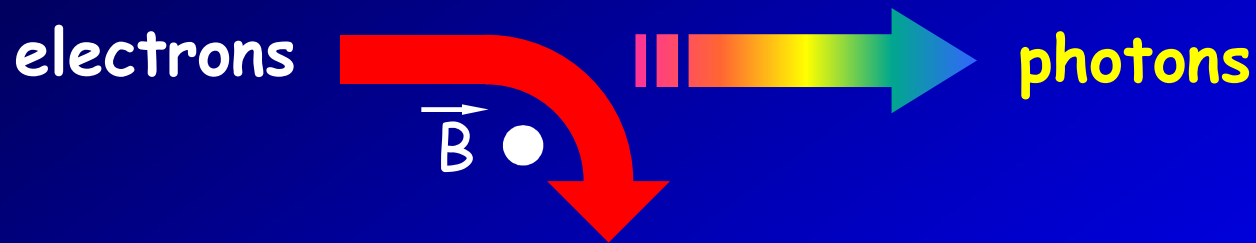
## Jet métal liquide



Flux / qqs  $10^9$  ph/s  
Taille de source : 5-30  $\mu\text{m}$   
Brillance spectrale :  $2-10 \cdot 10^{10}$  UB

# Les sources synchrotron

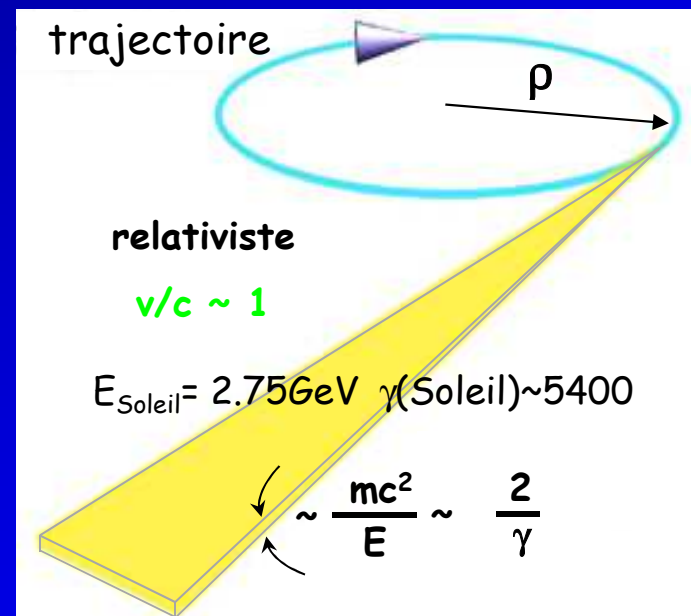
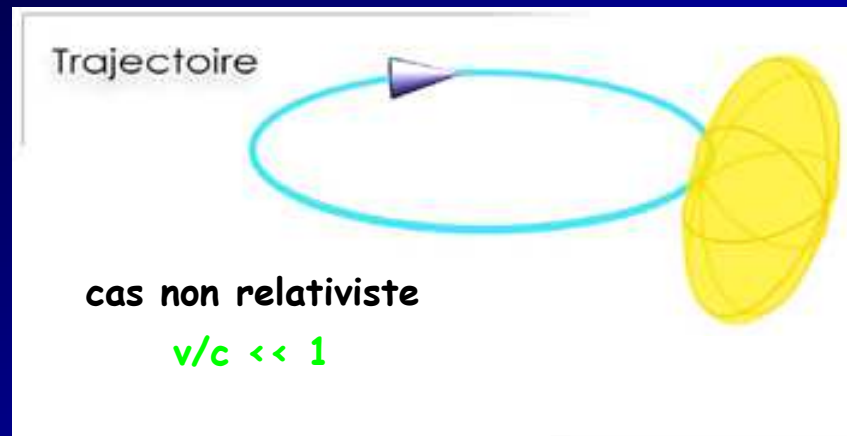
# Le rayonnement synchrotron



- ✓ forte brillance  $> 10^{10}$  x brillance (source labo)!!
- ✓ rayonnement blanc: des IR lointains aux rayons X durs
- ✓ lumière pulsée et polarisée (linéairement, circulairement...)

# Synchrotron

**Principe** : une particule chargée soumise à une accélération rayonne!



$$P_{\text{ray/s}} \sim \frac{E^4}{m^4 \rho^2} \longrightarrow e^- \text{ ou } e^+$$

# Synchrotron

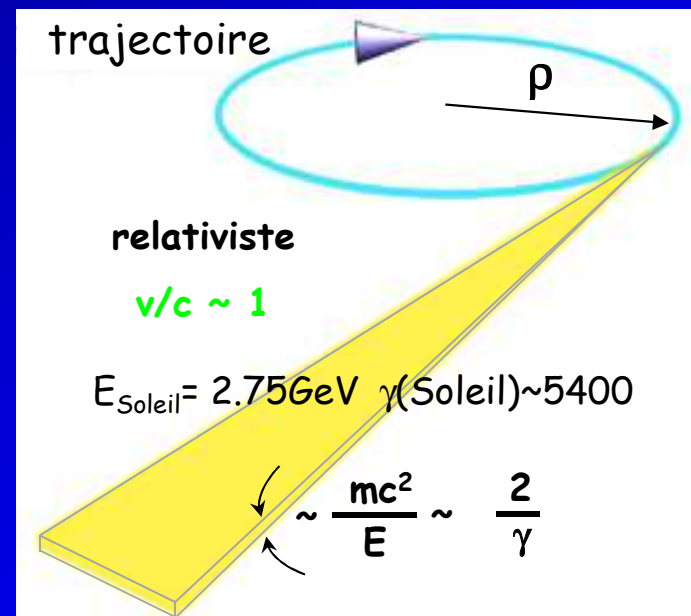
**Principe** : une particule chargée soumise à une accélération rayonne!

Source Lab      Vert. Div.  
                         '5' mrad

Aimant de courbure    0.2 mrad

Élément d'insertion    0.01 mrad

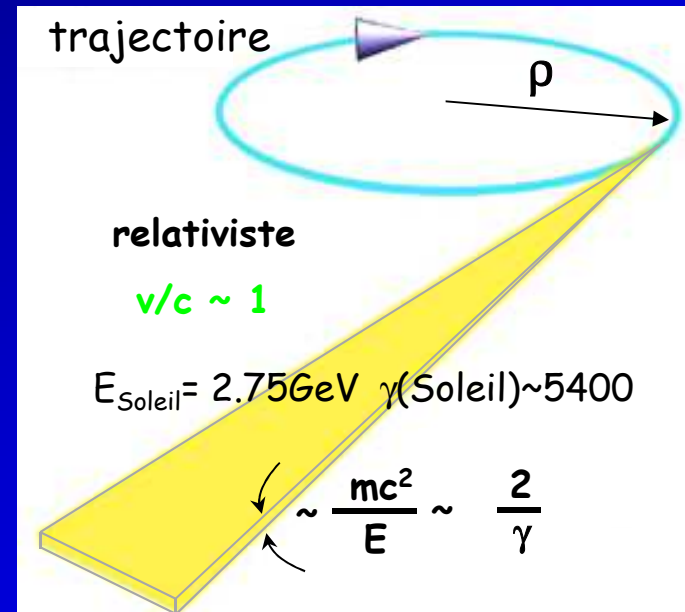
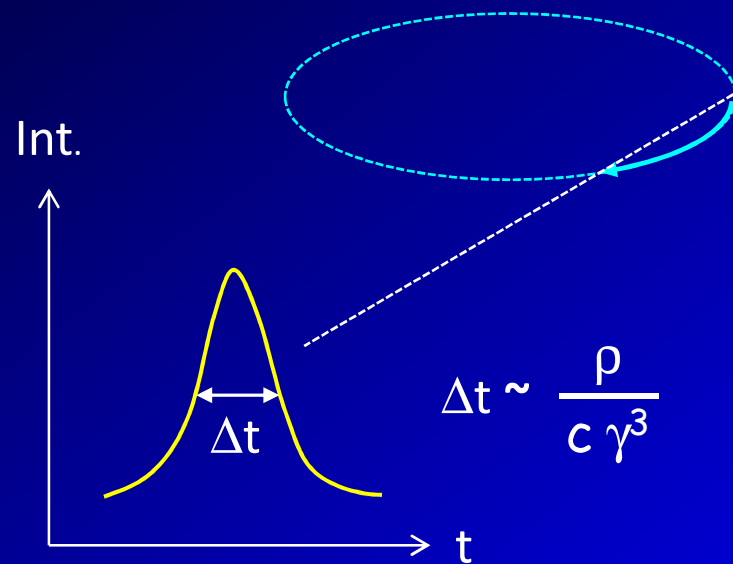
(1 mrad  $\sim$  0.06°)



➔ { accélérer les  $e^-$  pour émission de lumière: trajectoire courbe  
régime relativiste ( $v \sim c$ ) : collimation

# Synchrotron

## Un rayonnement blanc

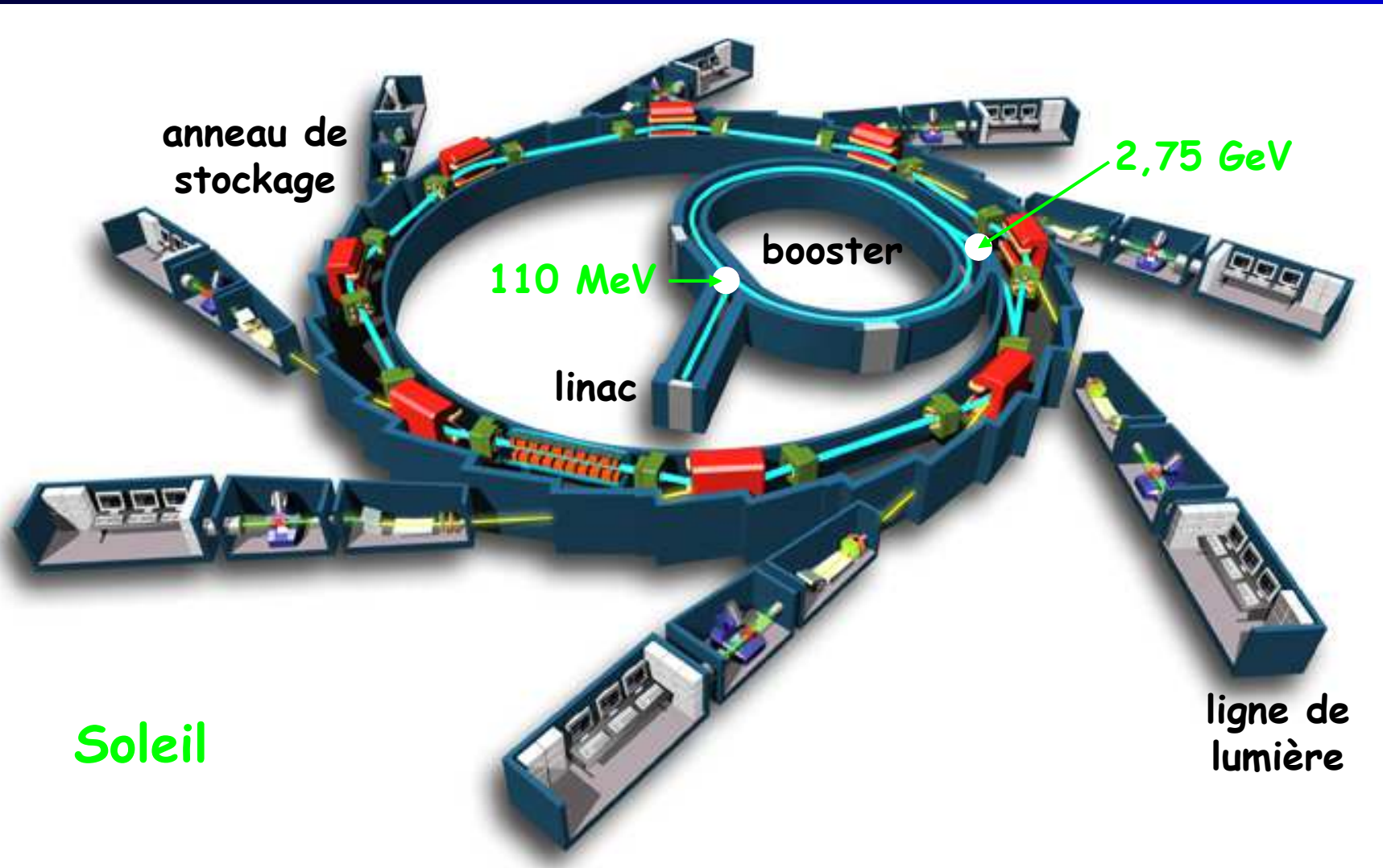


$\Delta t \sim 10^{-20} \text{ s}$   $\rightarrow$  composantes fréquentielles jusqu'à  $1/\Delta t$

 rayonnement blanc  $\rightarrow$  des IR jusqu'au rayons X!



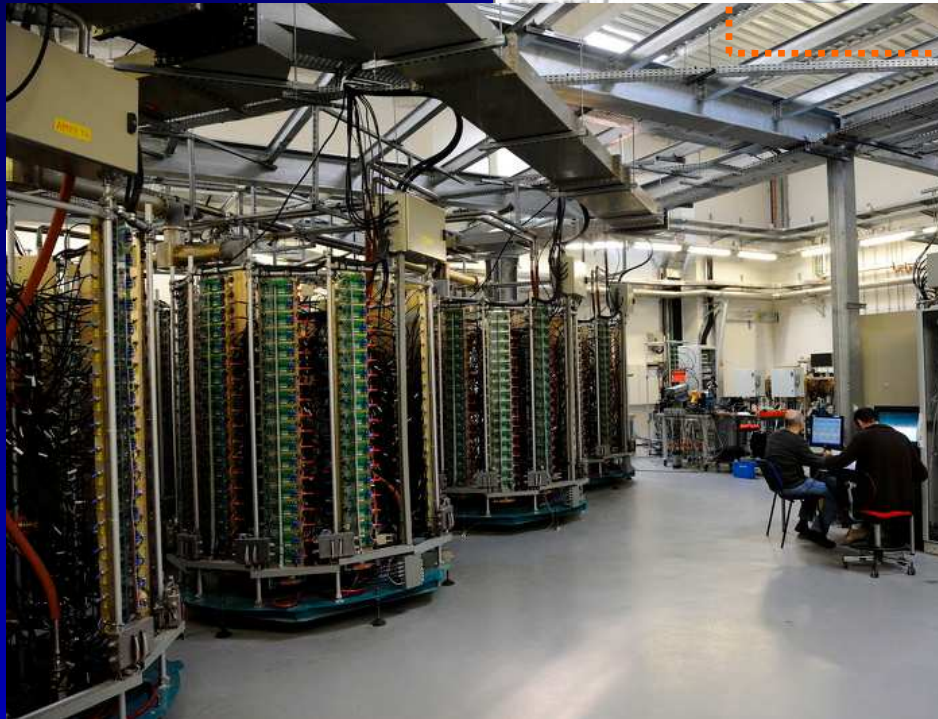
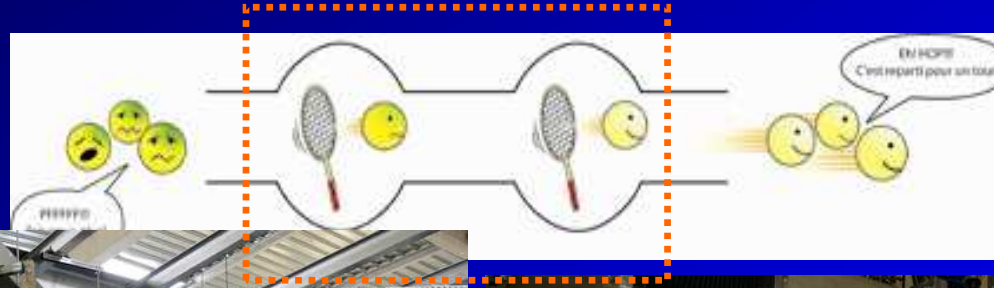
# Synchrotron: la machine



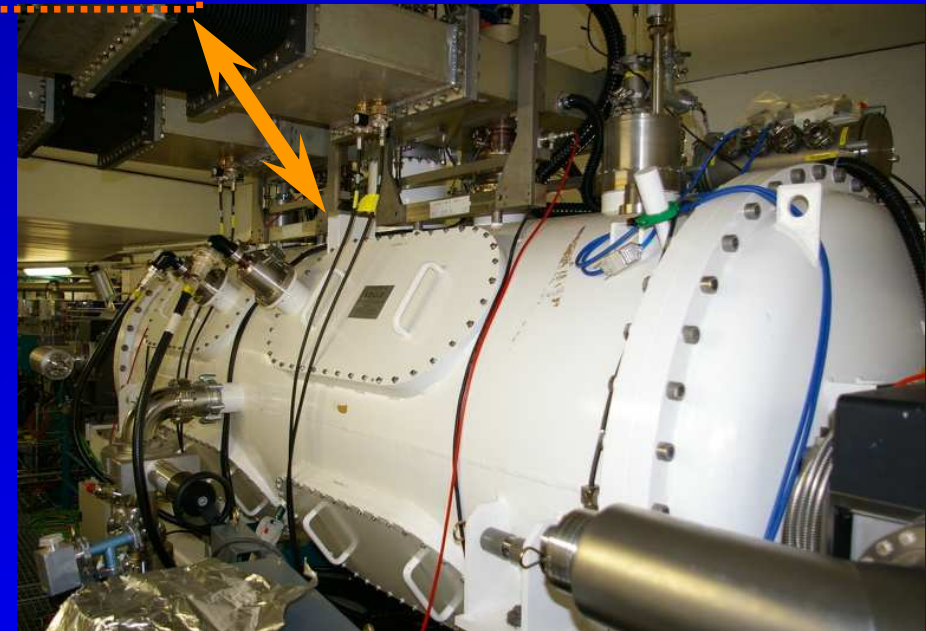
# Cavité Radiofréquence (352 MHz)

compenser l'énergie perdue par rayonnement au cours d'une révolution

si non 2.75 GeV perdus en 3 ms (~2 400 t) !! 1150 keV ~ 0.04%

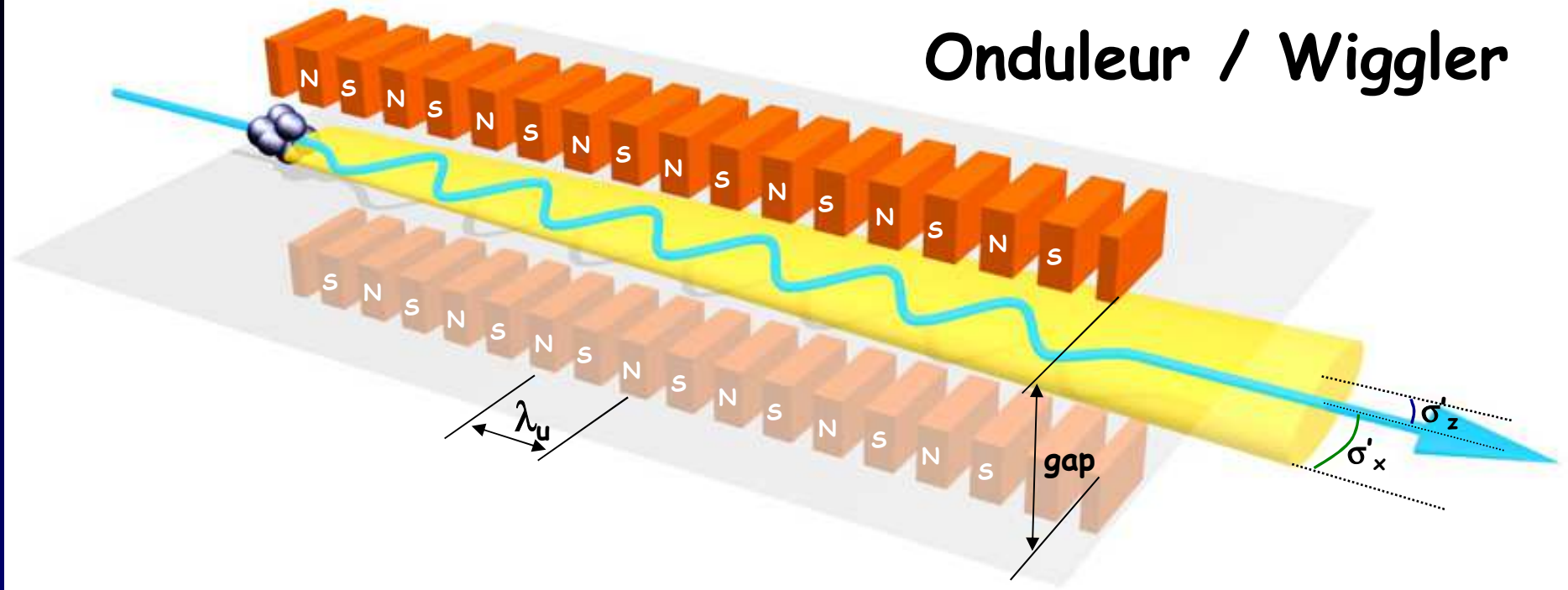


Générateur radiofréquence



1<sup>er</sup> cryomodule (cavité RF refroidie à 4K)

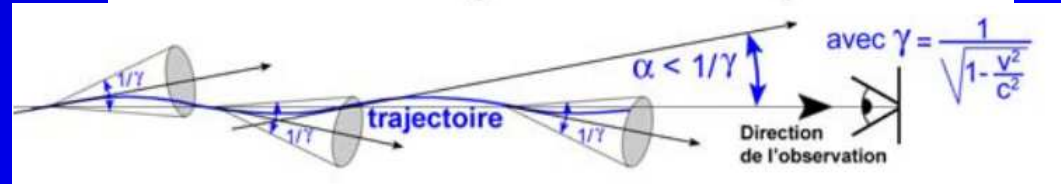
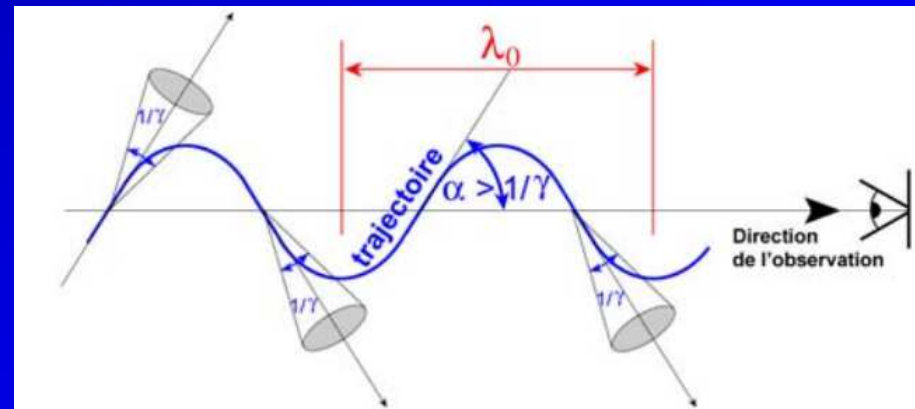
# Onduleur / Wiggler



amplitude d'oscillations  $\longleftrightarrow K \sim \lambda_u [\text{cm}] B_0 [\text{T}] \longleftrightarrow \text{gap (i)}$

$K \gg 1$  wiggler  $I \sim 2N I(\text{wiggle})$

$K \sim 1$  onduleur  $I \sim N^2 I(\text{wiggle})$



# Onduleur à aimants permanents ex. U20

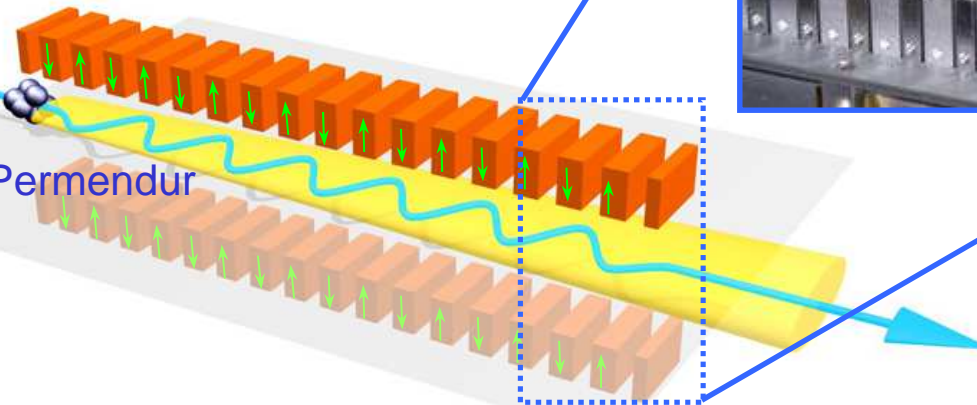
Système magnétique: Hybride  
Aimant permanent:  $\text{Sm}_2\text{Co}_{17}$   
Pôle magnétique: Vanadium Permendur

Gap = 5.5 mm – 30 mm

Période = 20 mm

Nper = 98, Longueur = 2 m

$B_0 = 0.97 \text{ T}$



# Onduleur électromagnétique

ex. HU640

$\frac{1}{2}$  machoire supérieure



Gap = 19 mm  
Période = 640 mm  
Nper = 14, Longueur = 9 m  
 $B_{x,z} \sim 0.1$  T



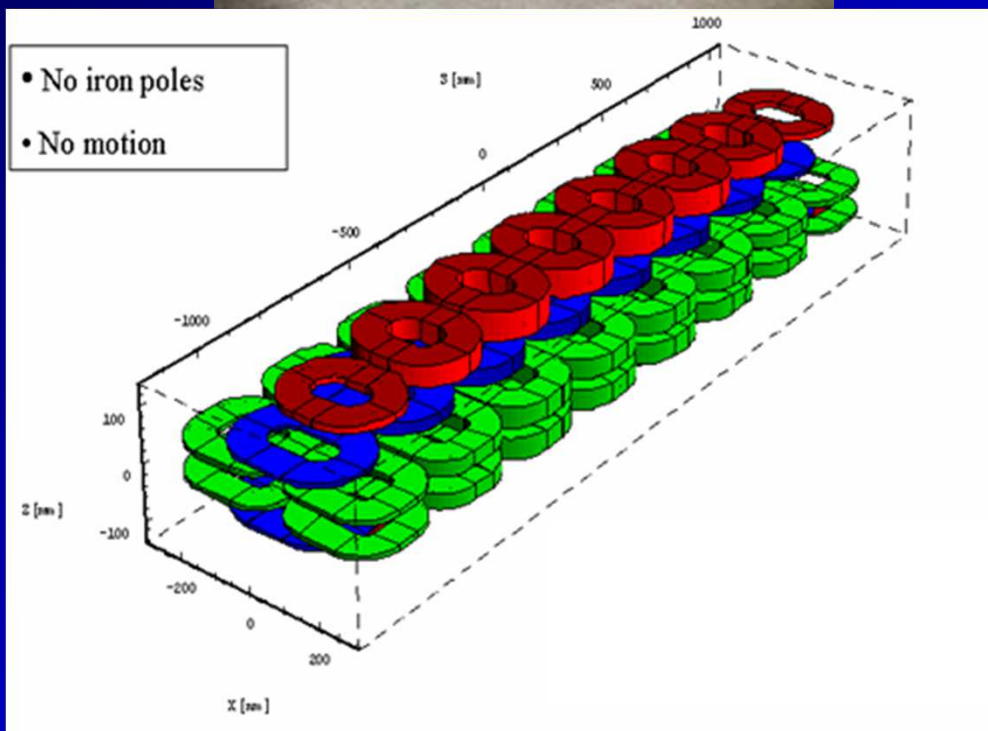
Energie: 5 - 40 eV  
(~VUV)

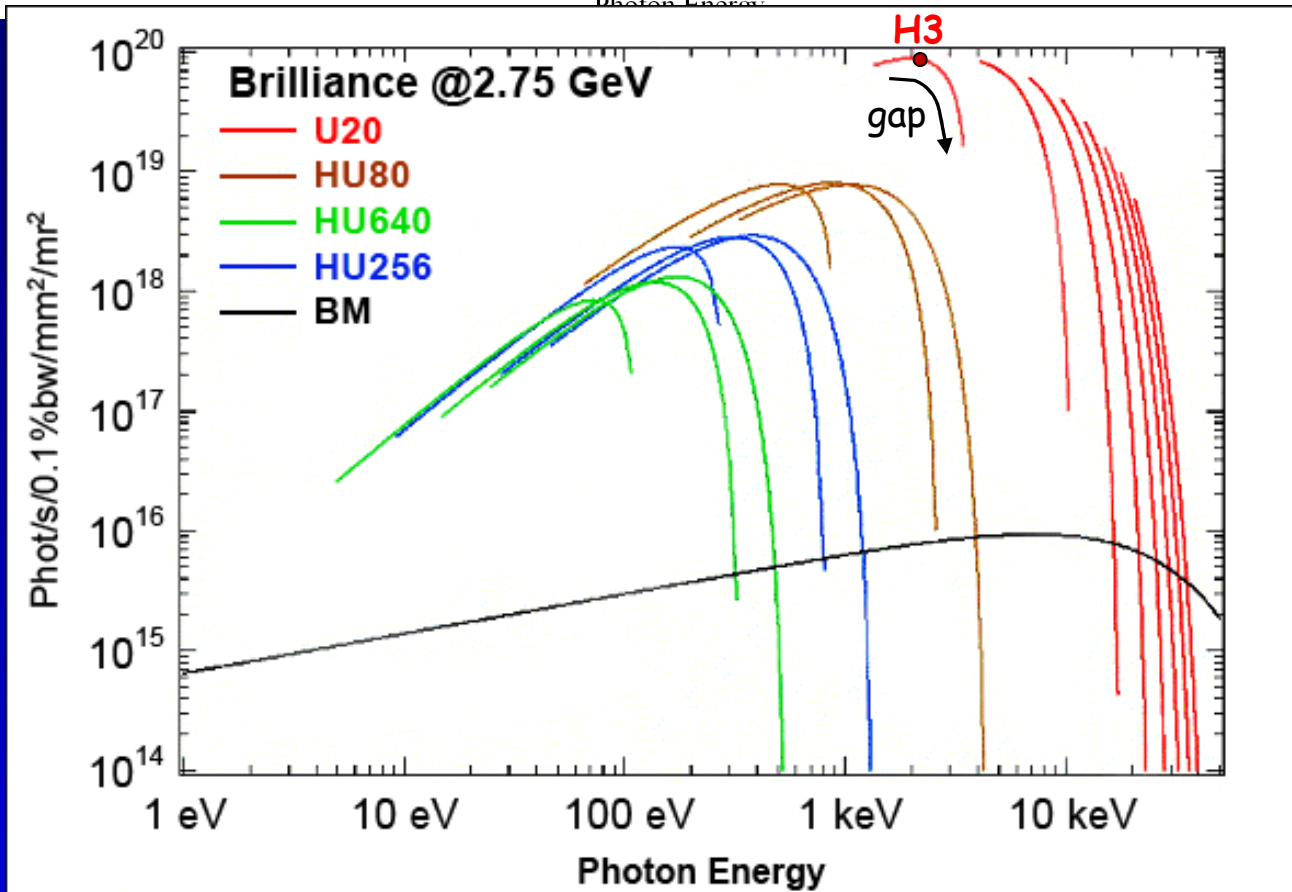
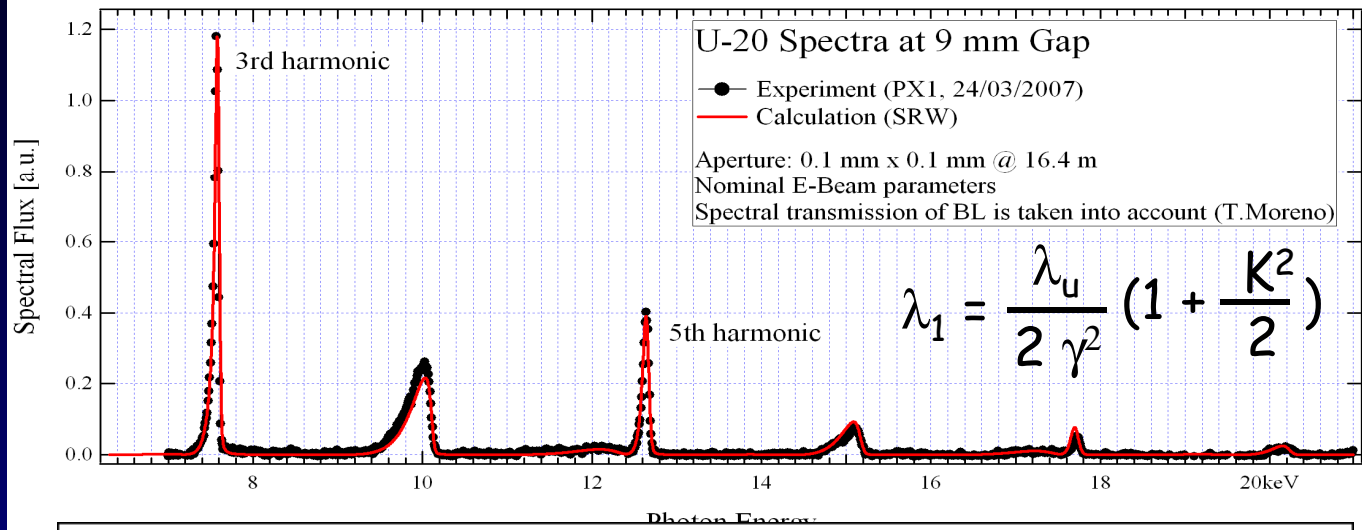
ajustement de la polarisation du faisceau

3 jeux de bobines (RVB)

R et B ( $B_z$ )

V ( $B_x$ )





2 exemples :

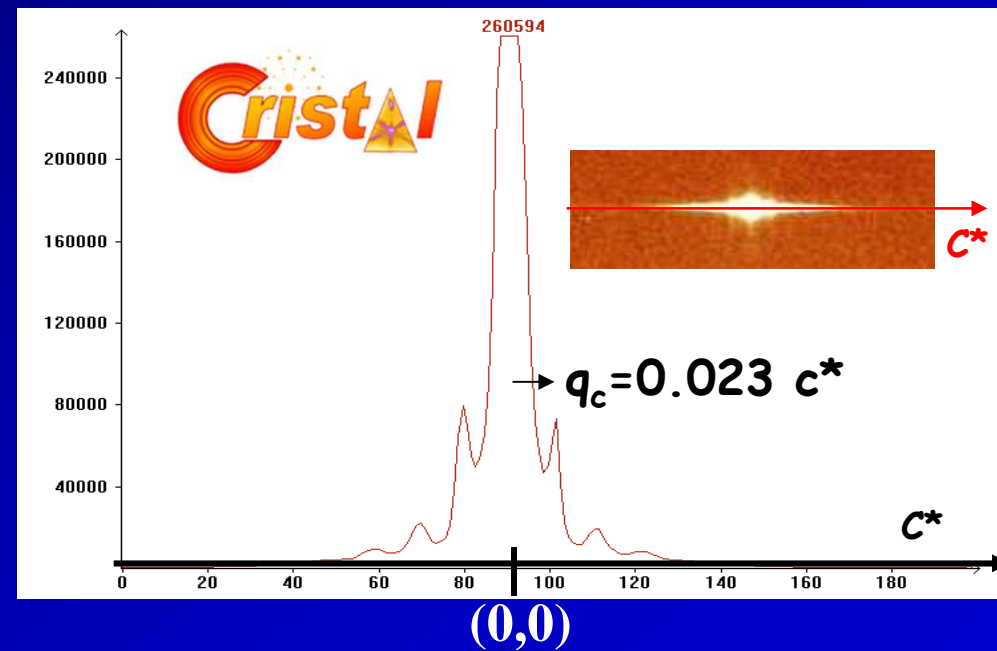
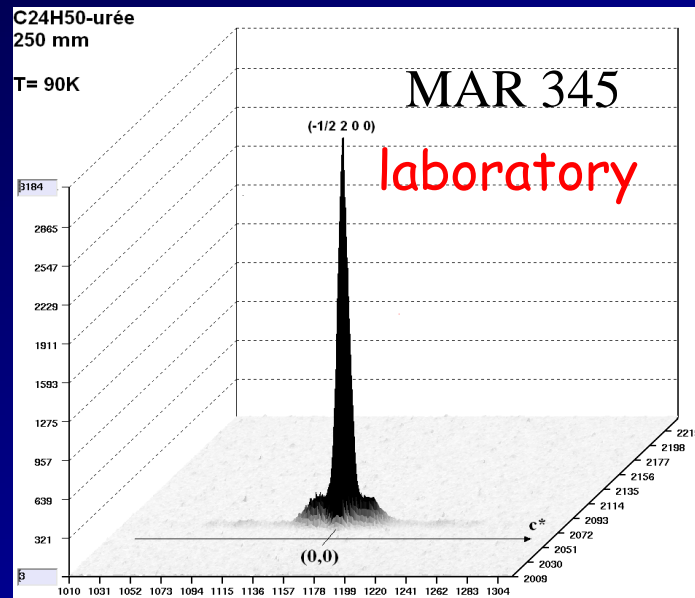
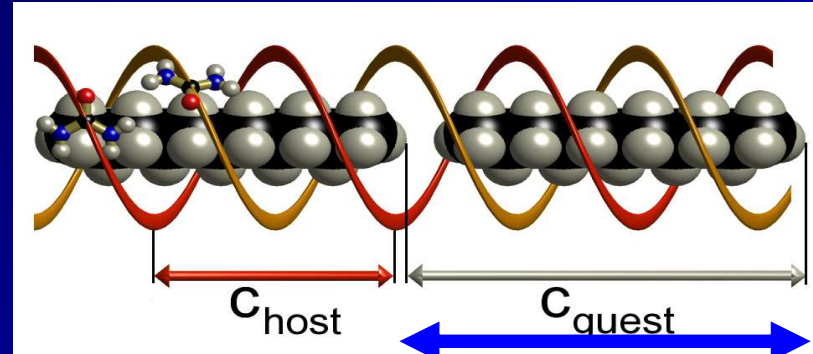
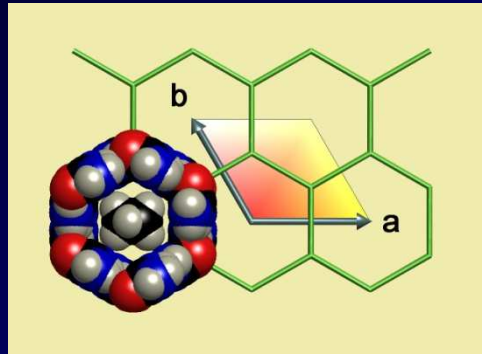
→ Parallelisme du faisceau

→ Accordabilité de la longueur d'onde

# Synchrotron source: a low divergent x-ray beam

Example: Phase transition in Tetracosane-urea  $C_{24}H_{50}$ -urea

Guerin et al., Phys. Rev. B91, 184101 (2015)



Discovery of a new characteristic length  $\sim 270 \text{ \AA}$  at low T

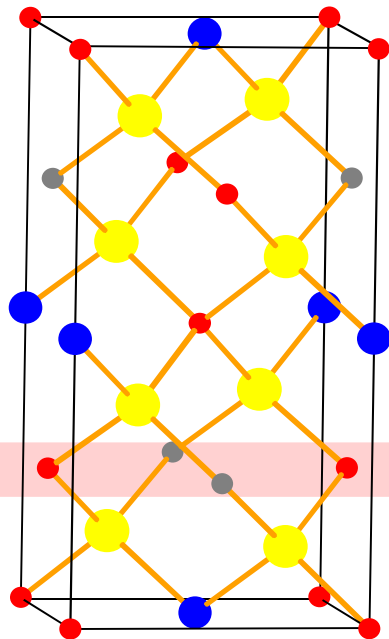


# Synchrotron source: wavelength tunability - example

## Cation distribution in photovoltaic $\text{Cu}_2\text{ZnSnS}_4$ derivatives: A single crystal anomalous diffraction investigation

Lafond et al., *Acta Cryst. B*70, 390 (2014)

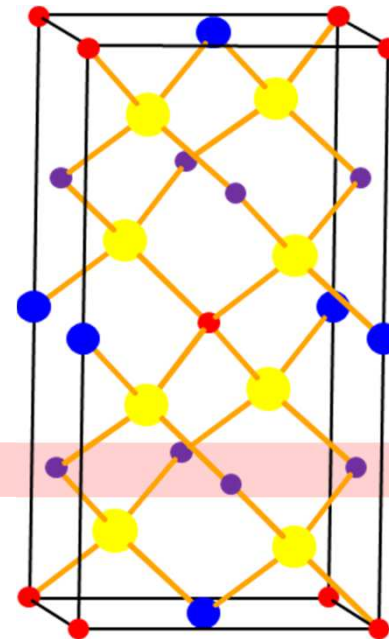
kesterite I-4



Cu & Zn



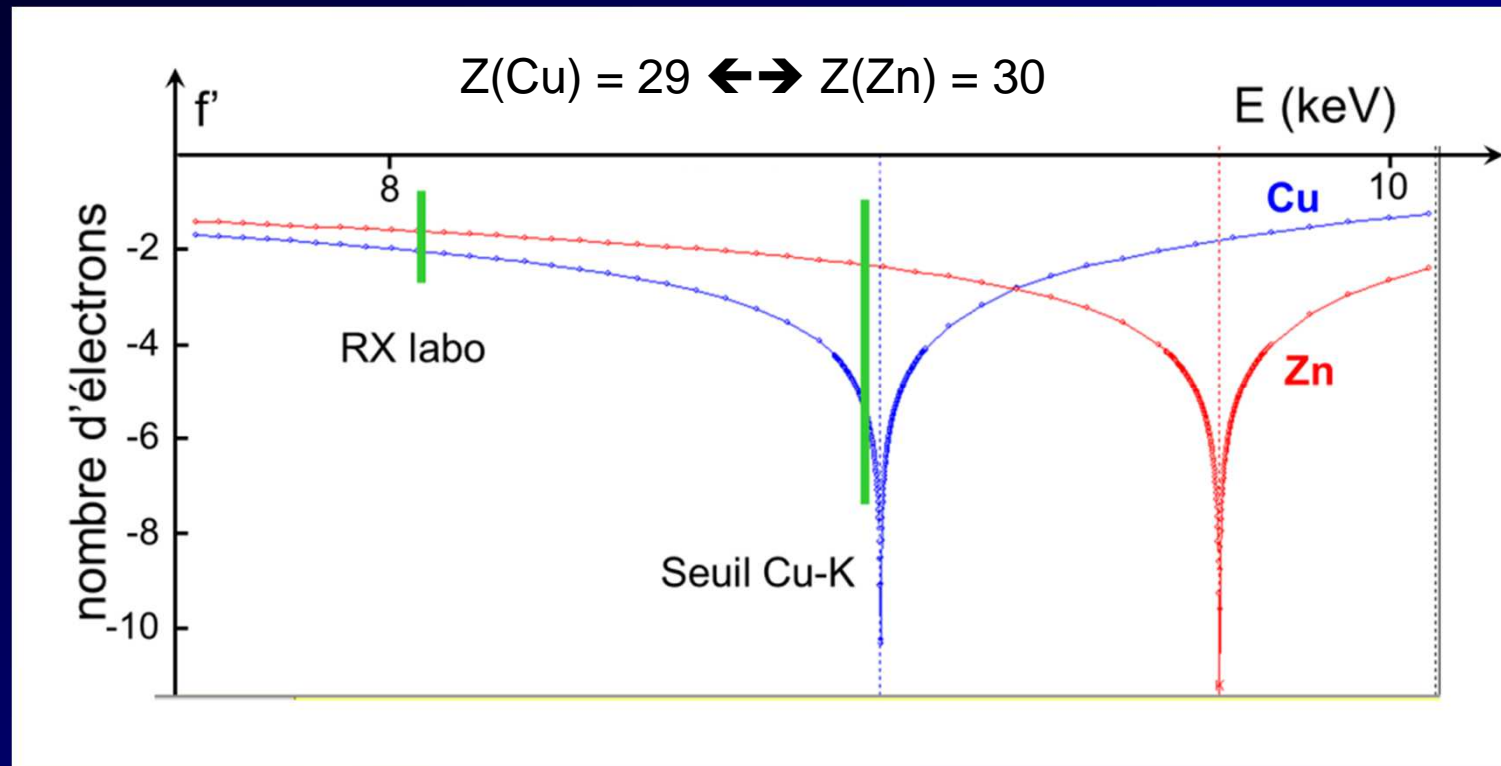
disordered-  
kesterite I-42m



Cu/Zn

# Synchrotron source: wavelength tunability - example

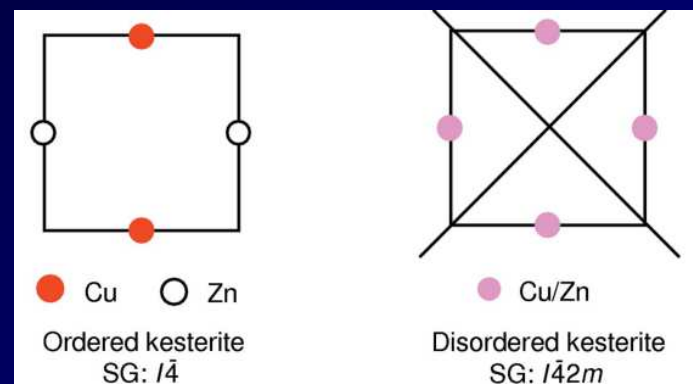
Atomic form factor:  $f = f_0(\sin(\theta)/\lambda) + f'(\lambda) + i f''(\lambda)$



Anomalous diffraction @ Cu  $K\alpha$  edge : **enhancement of the Cu/Zn contrast**

$E = 8.968 \text{ keV (} 1.382 \text{ \AA)}$  :  **$Z(\text{Zn}) - Z(\text{Cu}) \sim 4 e^-$**

# Synchrotron source: wavelength tunability - example



Structural model	kesterite	modified kesterite	disordered kesterite
2c	Cu	Zn	Cu/Zn (50/50)
2d	Zn	Cu	Cu/Zn (50/50)
U(2c) ( $\text{\AA}^2$ )	-0.002(1)	0.034(1)	0.017(1)
U(2d) ( $\text{\AA}^2$ )	0.031(1)	-0.005(1)	0.014(1)
R/wR(obs)	4.12/7.45	4.08/7.33	4.05/7.28
Fourier-difference ( $e^{-}/\text{\AA}^3$ )	1.48/-1.68	1.44/-1.66	1.46/-1.68

Unphysical Debye Waller-factors ←

→ Realistic Debye Waller-factors  
Slight improvement of the agreement factors

# Source de rayons X synchrotrons: une lumière pulsée

Mode de remplissage: (ex. Soleil)

uniforme: 1 bouffée  $\sim 40$  ps toutes les  $\sim 3$  ns

8 paquets:  $\sim 70$  ps toutes les  $\sim 150$  ns

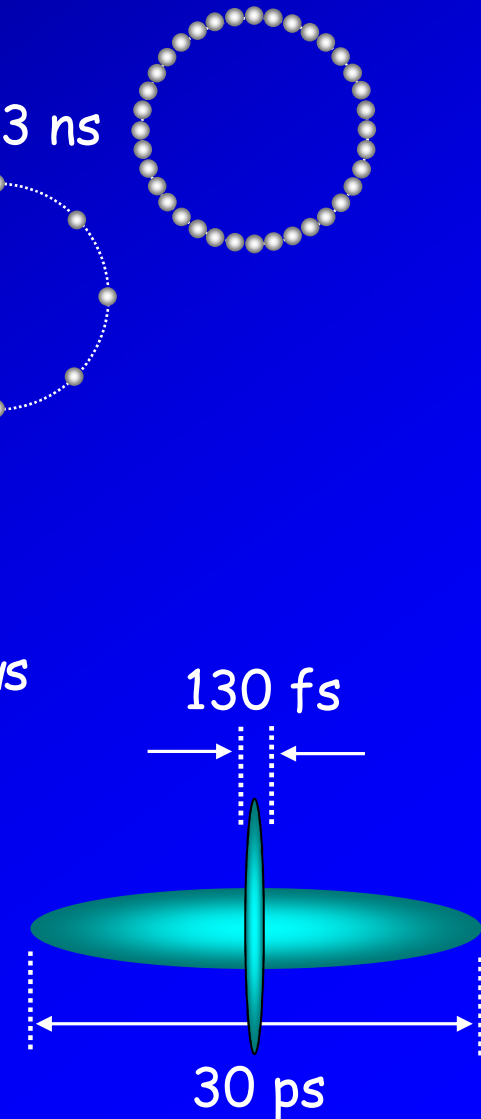
hybride: ex. 312 + 1

vers des bouffées encore plus courtes...

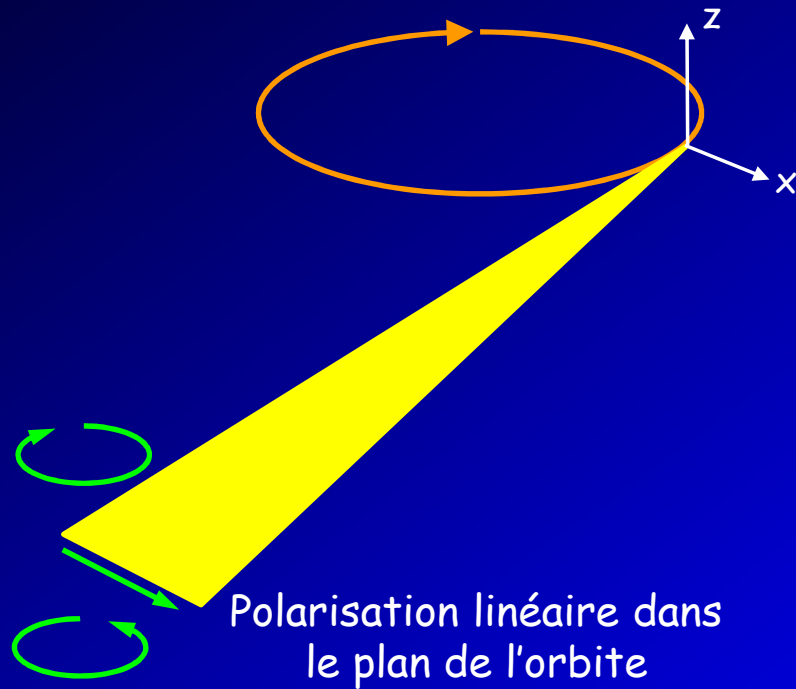
1 paquets « tranchés »:  $\sim 130$  fs toutes les  $\sim 1.2$   $\mu$ s

Chaque paquet est « tranché »  
par une impulsion laser pour  
créer un paquet plus court

Expériences résolues en temps

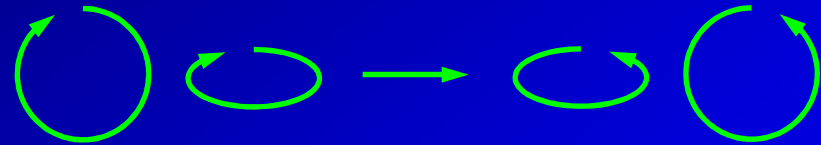


# Source synchrotron: une lumière polarisée

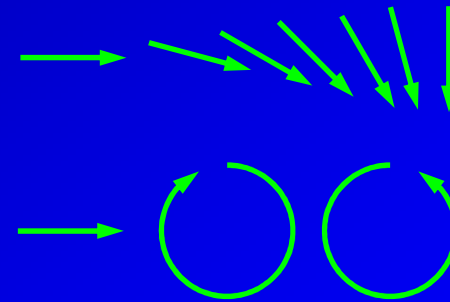


Onduleur avec  $B_z + B_x$

Polarisation accordable



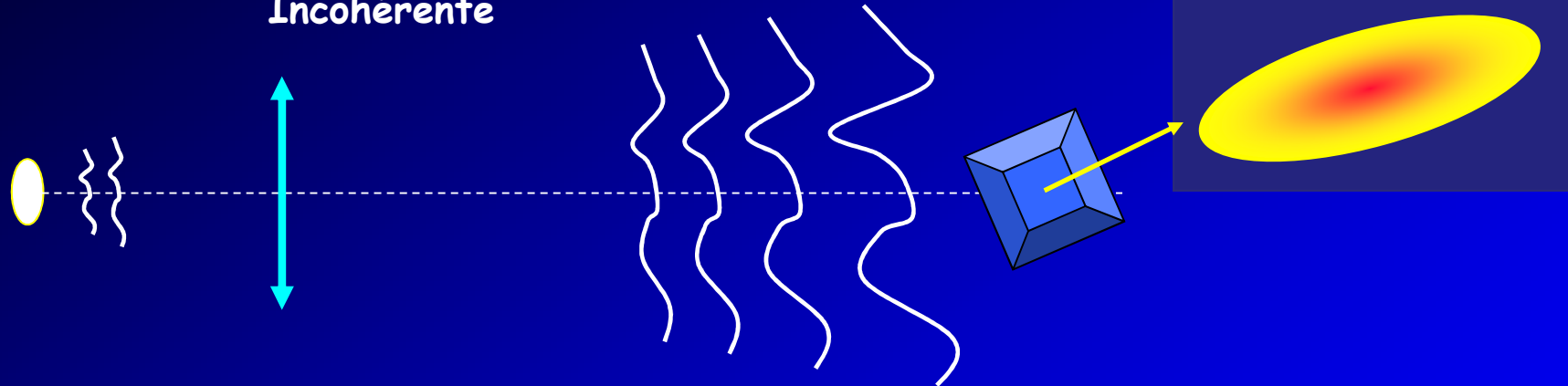
Onduleur + « phase plate »



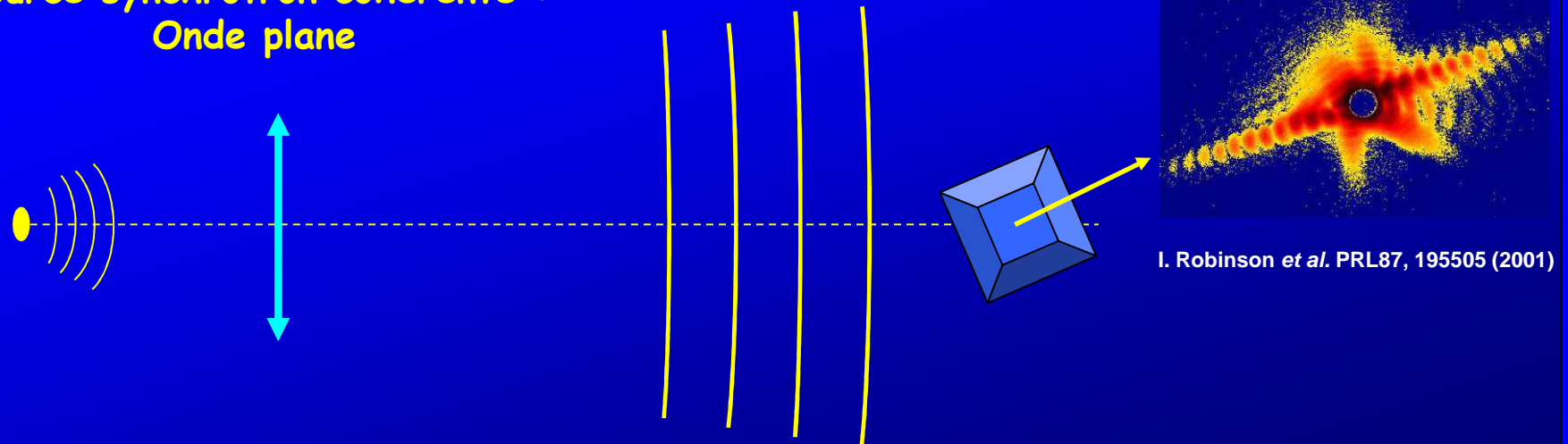
⇒ Diffraction magnétique  
ex. domaines magnétiques,  
structures magnétiques

# Source synchrotron: une source cohérente

Source classique ne donne pas d'onde plane :  
Incohérente



Source synchrotron cohérente :  
Onde plane



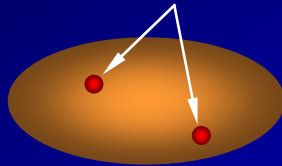
*(cf. cours de D. Le Bolloc'h vendredi après-midi)*

... encore plus de photons ?

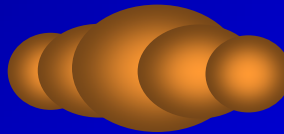
# Les sources de 4<sup>ème</sup> génération XFEL

**Principe** : après une certaine distance parcourue dans un onduleur, les  $e^-$  sont sensibles au champ électrique rayonné par les autres  $e^-$

rayonnement incohérent



pas de cohérence intrinsèque  
 $e^-$  émettent indépendamment



modulation spatiale de la distribution  
des  $e^-$  dans le paquet

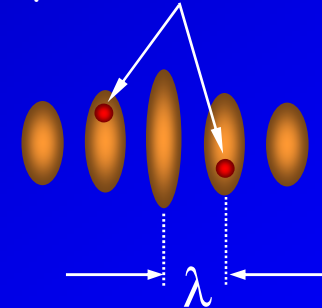
si densité d' $e^-$  suffisante



mécanisme de self-amplification  
(laser à  $e^-$  libres)

**Self Amplified Stimulated Emission**

rayonnement cohérent



réduire la taille des paquets!  
~100 $\mu$ m, 100fs



# Les sources de 4<sup>ème</sup> génération XFEL

- ✓ Linac Coherent Light Source (Stanford, USA)
- ✓ XFEL/Spring 8 (Hyogo, Japon)
- ✓ E-XFEL (Hamburg, Allemagne)

2.5 - 20 GeV, 3.4 km

$\lambda \sim 1 - 60 \text{ \AA}$

brillance moyenne  $> 10^{25}$ , pic  $\sim 10^{33}$

$\sim 10^{12}$  photon/impulsion

Taux de répétition 30 kHz

$\sigma \sim 100 \mu\text{m}$ ,  $\sigma' \sim 0.8 \mu\text{rad}$

durée de l'impulsion  $\sim 100 \text{ fs}$

✓ ...

(X)FEL: liste exhaustive cf. [http://sbfel3.ucsb.edu/www/vl\\_fel.html](http://sbfel3.ucsb.edu/www/vl_fel.html)



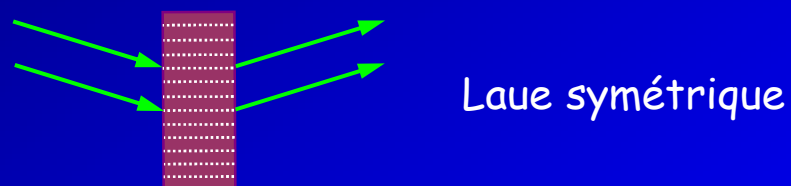
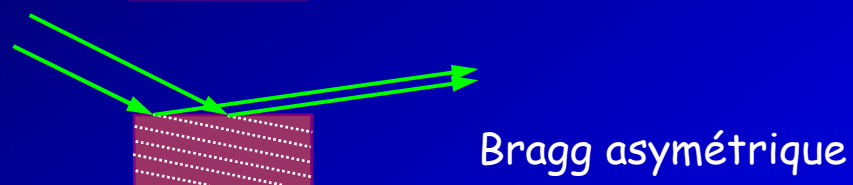
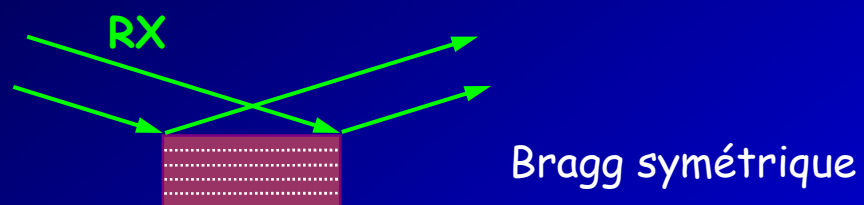
# Optiques pour les Rayons X

- ✓ Faisceau monochromatique
- ✓ Faisceau focalisé

# Optiques pour les Rayons X

**Monochromateurs** extraire un faisceau monochromatique du faisceau blanc

**Principe** : réflexion du faisceau blanc sur une famille de plans réticulaires



$$2d_{hkl} \sin \theta = n\lambda$$

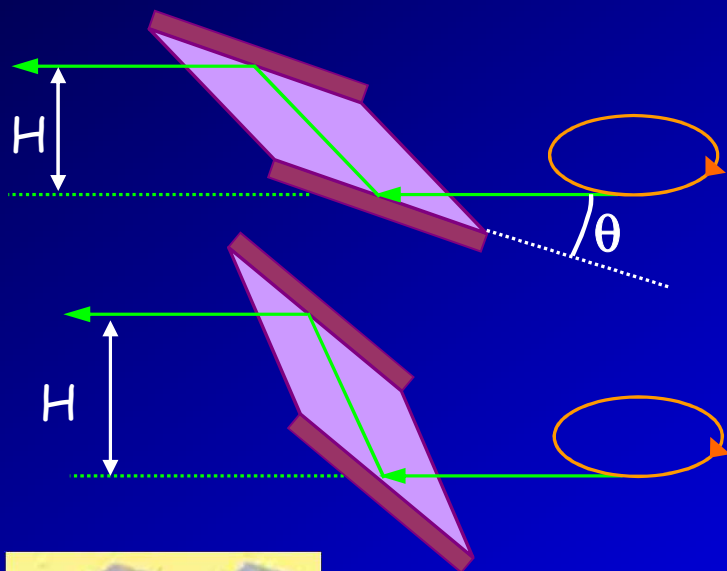
Plans réticulaires = miroirs

Cristaux parfaits → **Si**, Ge, C<sub>diamant</sub>... C<sub>graphite</sub>

$$\lambda (\text{\AA}) = \frac{12.39842}{E (\text{keV})}$$

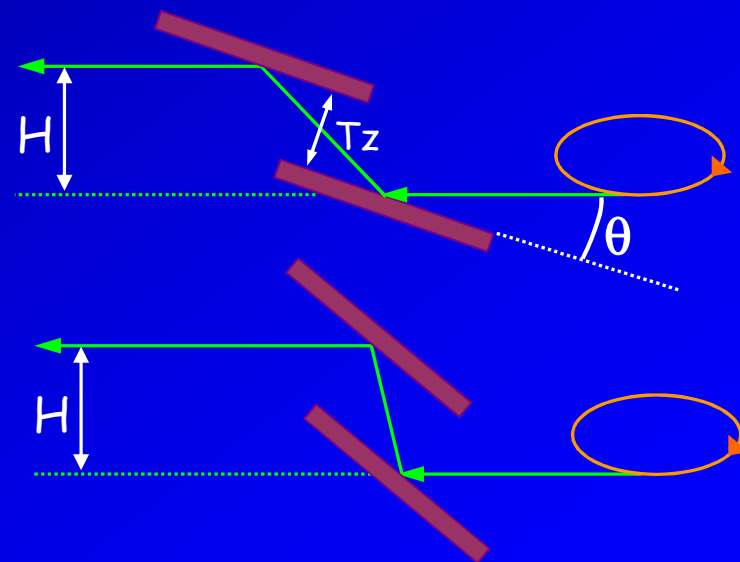
# Optiques pour les Rayons X

Monochromateur  
channel cut



H variable

Monochromateur  
double cristaux



sortie fixe (H fixe)

# DCM @ CRISTAL

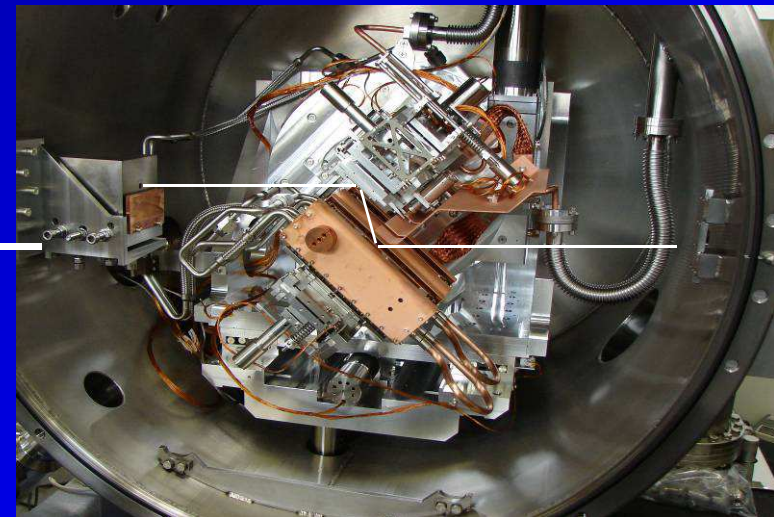
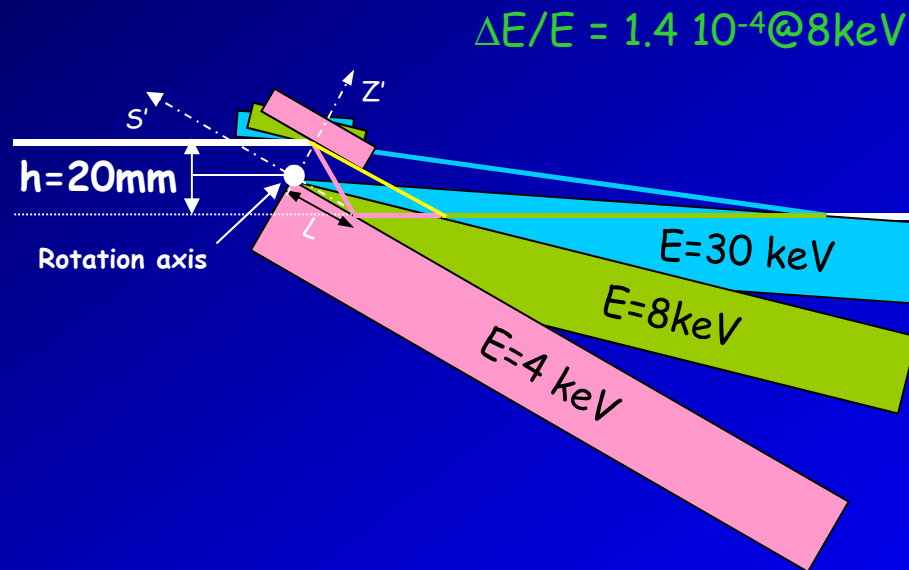
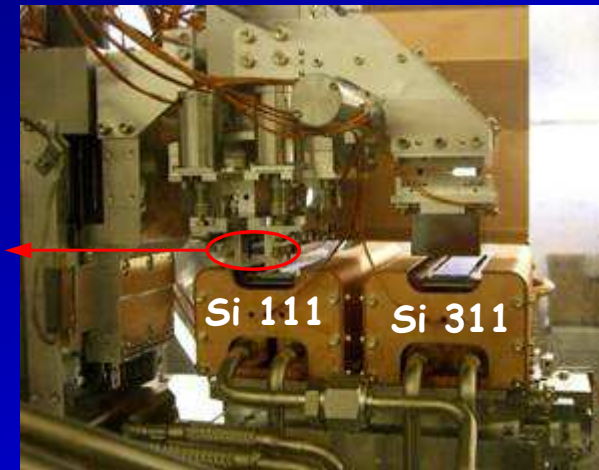
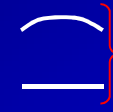
Deux paires de cristaux Si(111) et Si(311)

Sortie fixe

Focalisation sagittale

Impact fixe sur 2<sup>nd</sup> cristal

4 keV - 30 keV ( $4^\circ < \theta < 30^\circ$ )



Refroidissement à l'azote liquide ( $P_{\text{Max}} = 135 \text{ W}$ ;  $D_{\text{Max}} = 28 \text{ W/mm}^2$ )

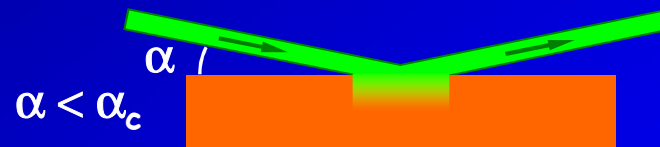
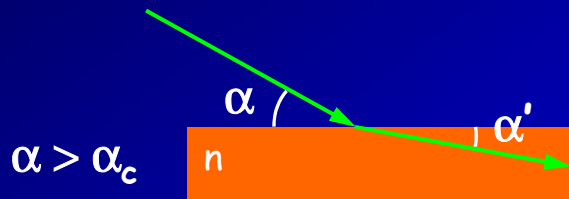
# Optiques pour les Rayons X

## Miroirs (focalisants)

Rayons X

$$n = 1 - \delta + i\beta$$

$n \lesssim 1$  réflexion totale quand  $\alpha < \alpha_c$



$$\delta = \frac{1}{2\pi} \rho r_0 \lambda^2 \quad \boxed{\delta \sim 10^{-5}}$$

$$\alpha_c = \sqrt{2\delta} \quad \boxed{\alpha_c \sim \text{mrad}}$$

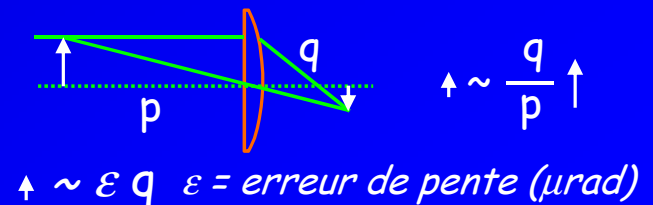
$$\alpha_c (\text{mrad}) = 1.6 \lambda_{(\text{\AA})} \sqrt{\rho_{(\text{g/cm}^3)}}$$

réflexion totale + miroir courbé

(courbure idéale = ellipse)



rejection d'harmonique + focalisation



(1 mrad  $\sim$  0.06°)

# Optiques pour les Rayons X

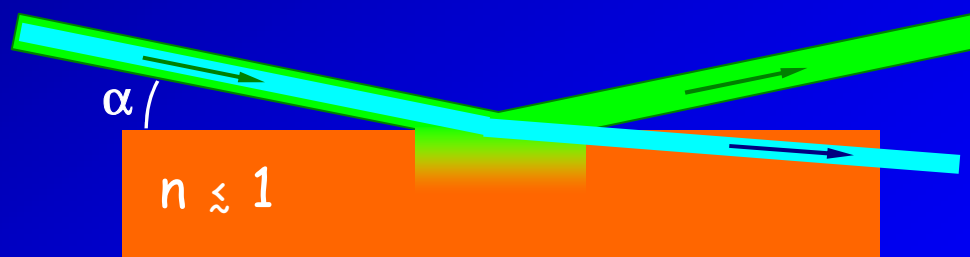
## Miroirs (focalisants)

Rejection d'harmoniques

$$\lambda_2 < \lambda_1$$

$$\alpha < \alpha_c(\lambda_1)$$

$$\alpha_c(\lambda_2) < \alpha$$



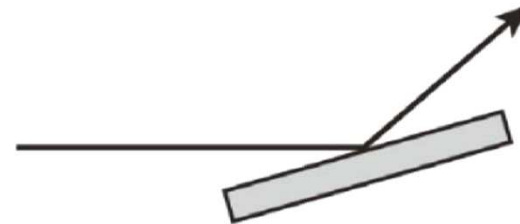
$$\alpha_c \sim \sqrt{\rho \lambda}$$

pour couvrir un large domaine d'énergie:  
plusieurs pistes de densité différente

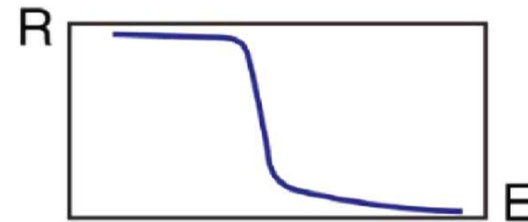
# Optiques pour les Rayons X

## Miroirs (focalisants)

- Deflecting the beam



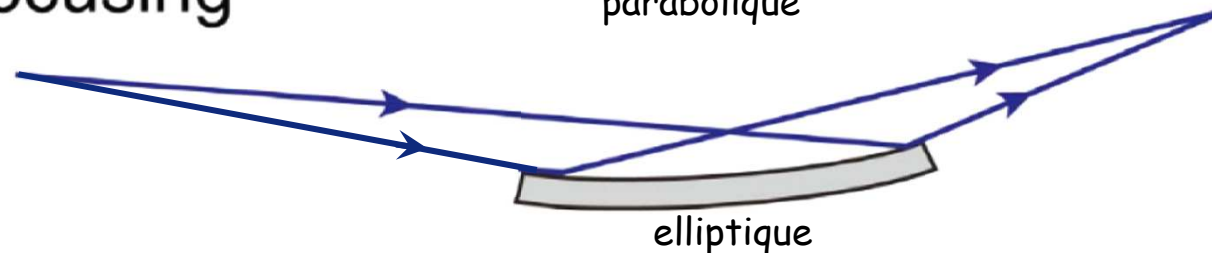
- Low pass filter



- Collimation



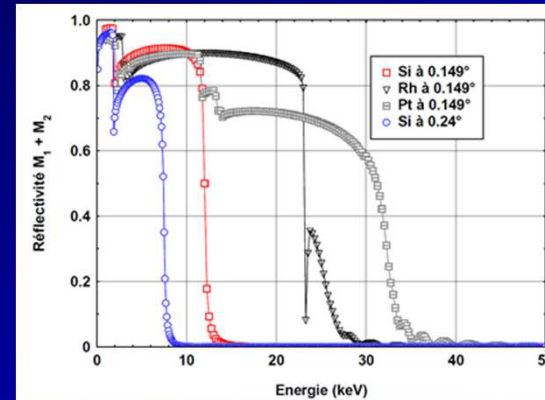
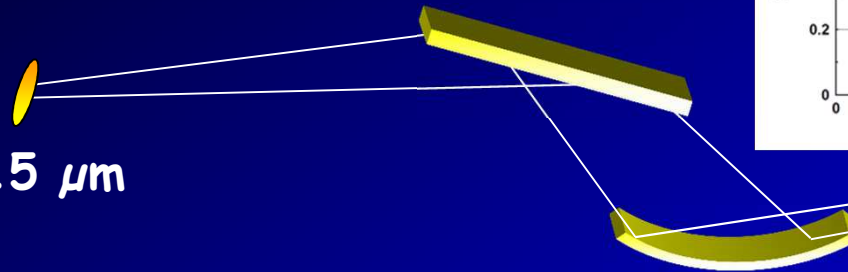
- Focusing





# ex: Miroirs de Cristal

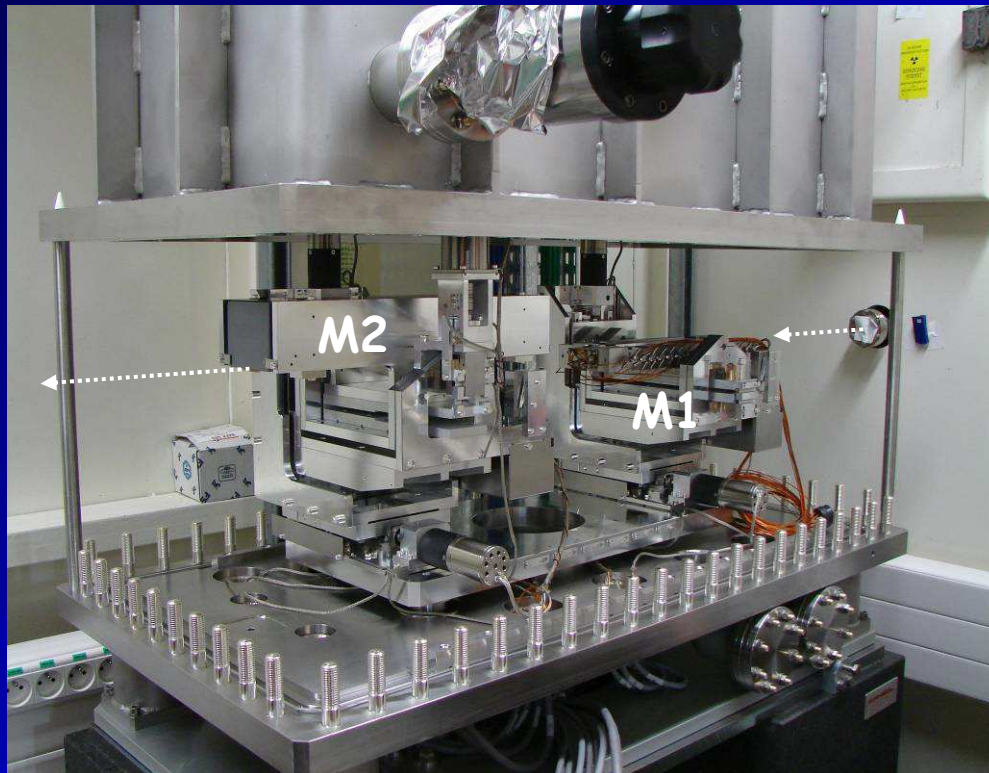
$V \sim 15 \mu\text{m}$



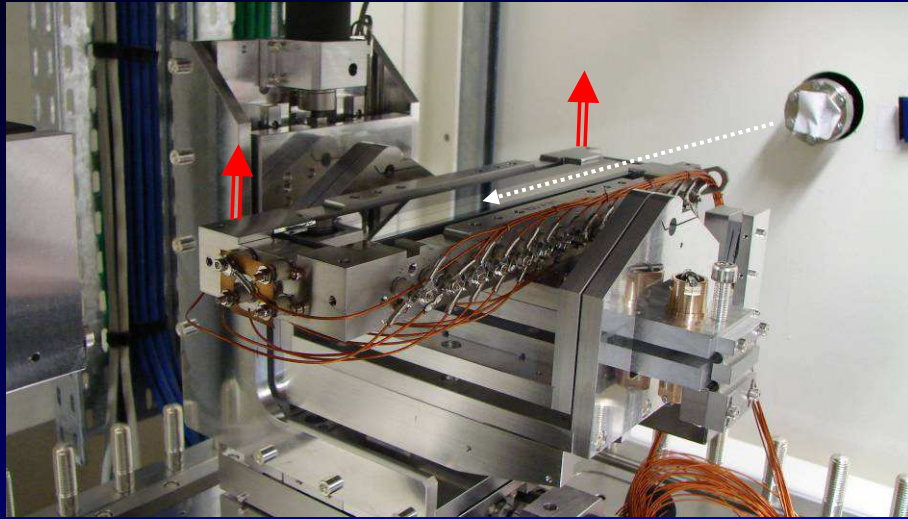
source

$V \sim 20 \mu\text{m}$

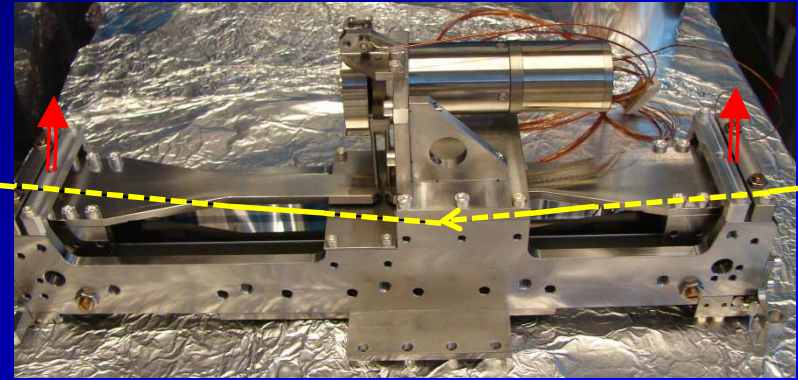
Miroir déformable (piézoélectrique)  
+ miroir plat  
3 pistes: Si, Rh, Pt



Total length	300 mm
Optical length	260 mm
Total width	45 mm
Electrodes	16/8
Slope error (long.)	1.5 $\mu\text{rad}$
Slope error (trans.)	10 $\mu\text{rad}$
Roughness	0.3 nm
Bands	Rh/Pt (10mm * 50+/-10 nm)

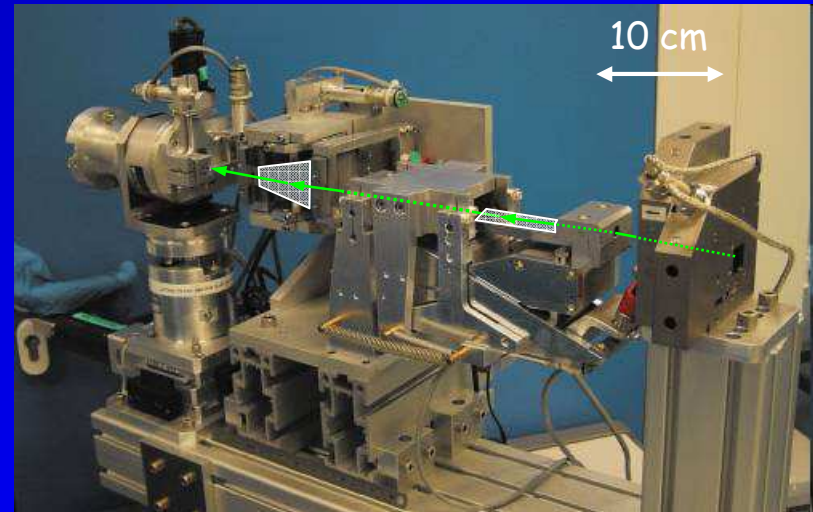
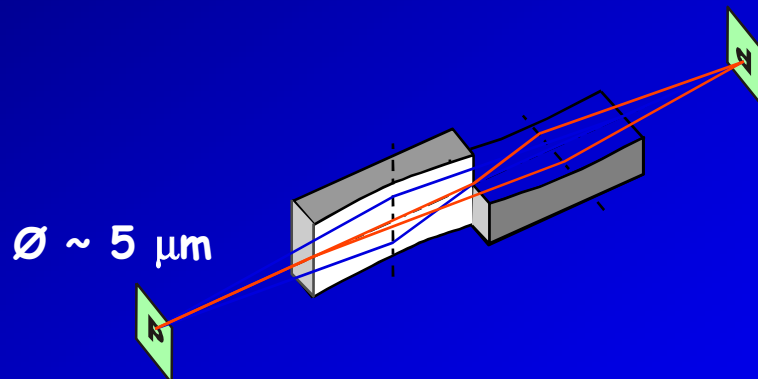


Miroir bimorphe déformable (piézoélectrique)  
3 pistes: Si, Rh, Pt



Miroir déformable (mécanique)  
3 pistes: Si, Rh, Pt

géométrie type Kirkpatrick-Baez (1948)



## Un compromis intéressant: les miroirs multicouches

accroissement de la réflectivité au delà de  $\alpha_c$  par addition cohérente de la réflectivité de surface empilée

$$n \lambda \sim 2 \cdot \Lambda \cdot \sin \theta$$



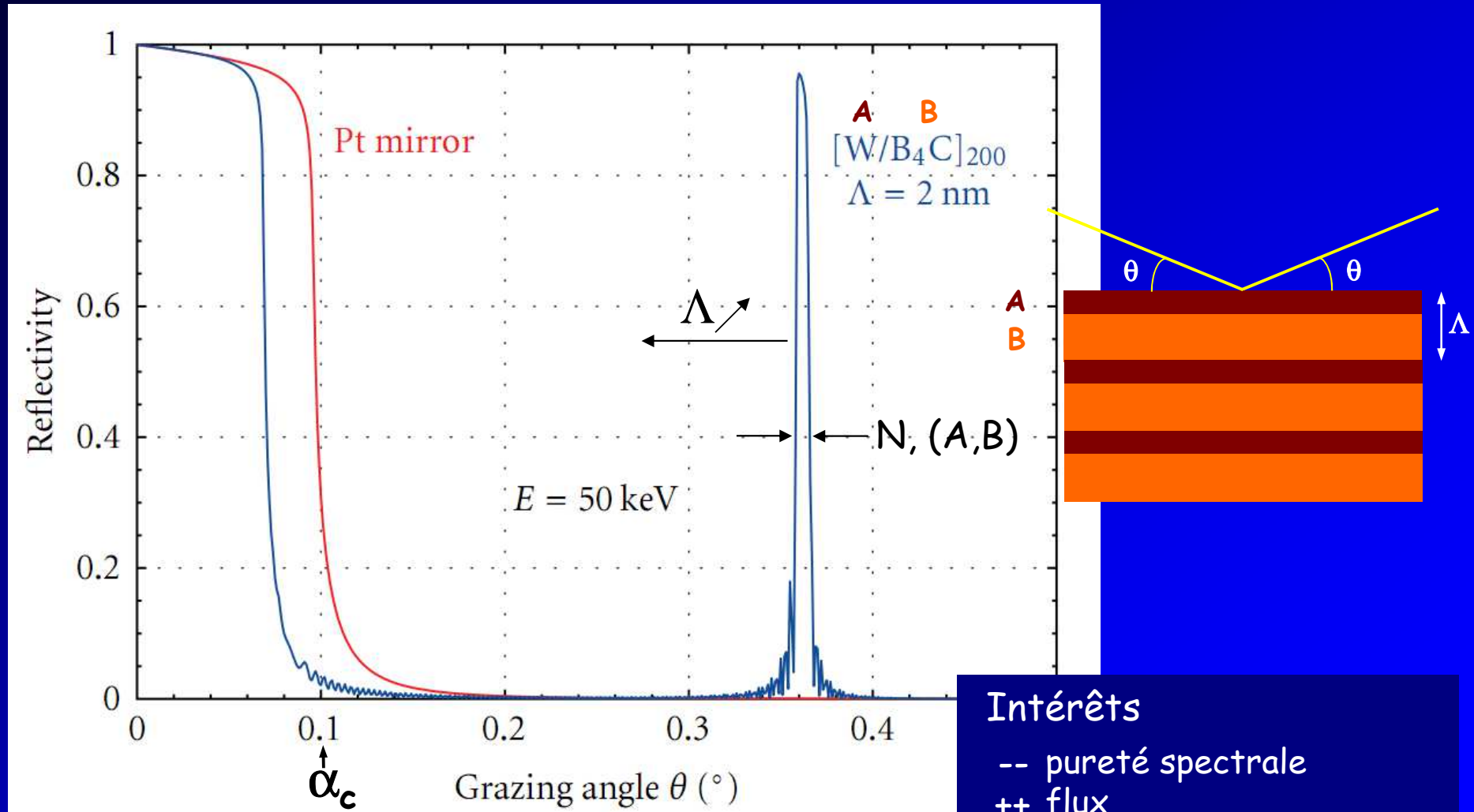
faisceau monochromatique

+ courbure = focalisation

faisceau monochromatique + focalisé

# Miroirs multicouches rendre le faisceau monochromatique + focalisation

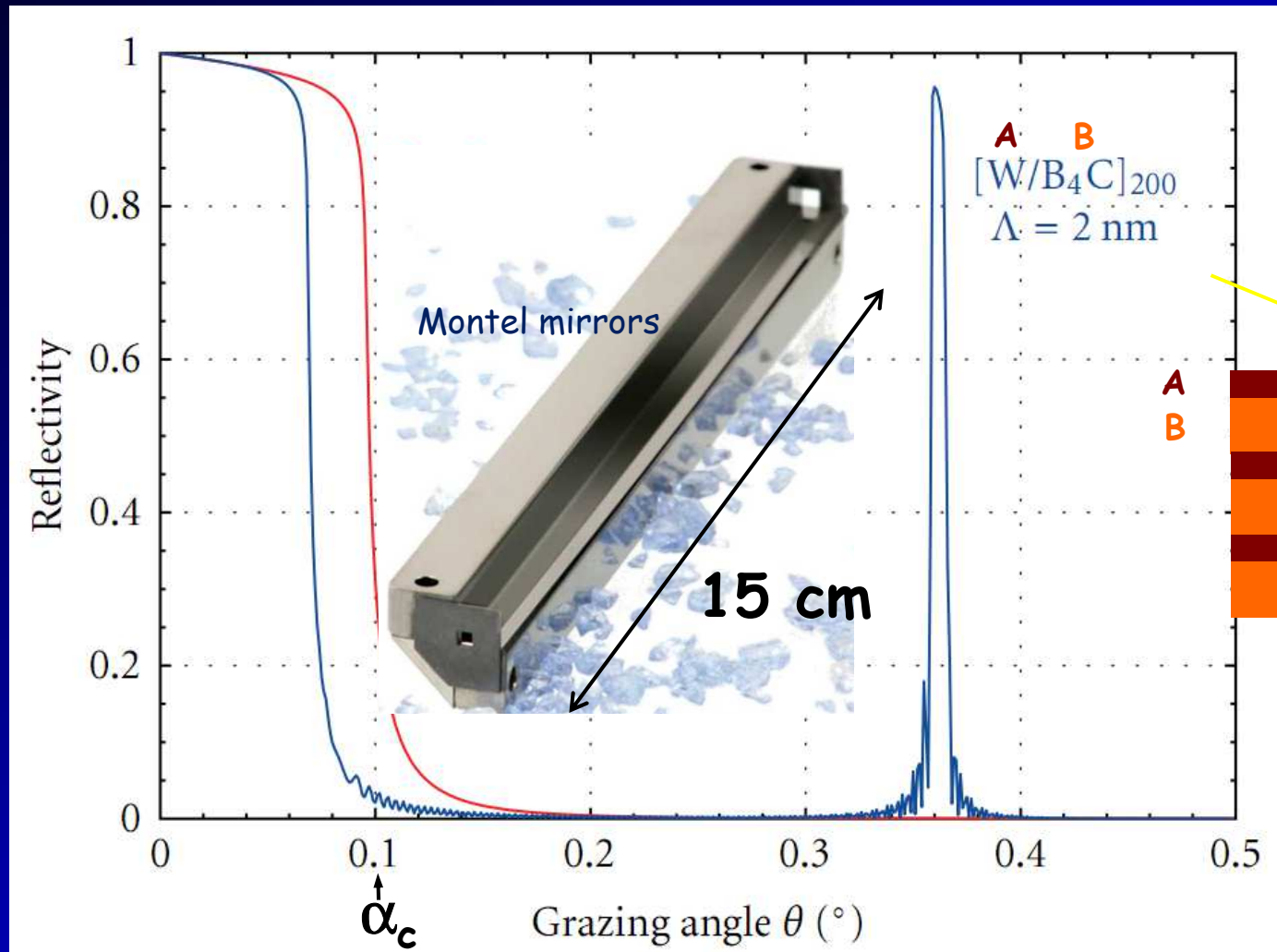
$$\lambda \sim 2 \cdot \Lambda \cdot \sin \theta$$



## Intérêts

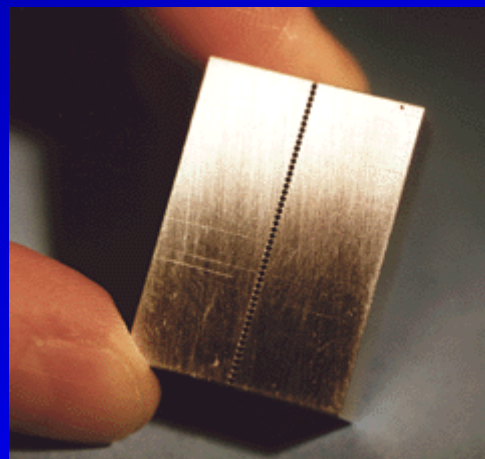
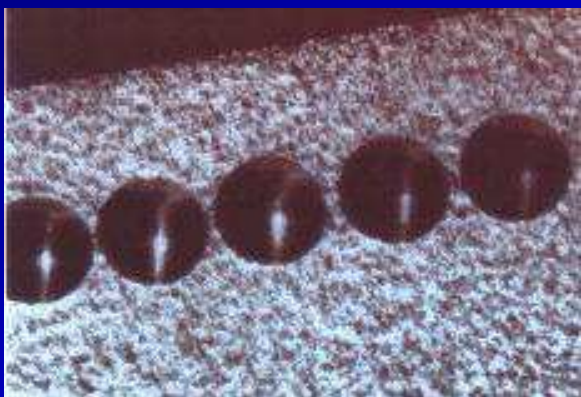
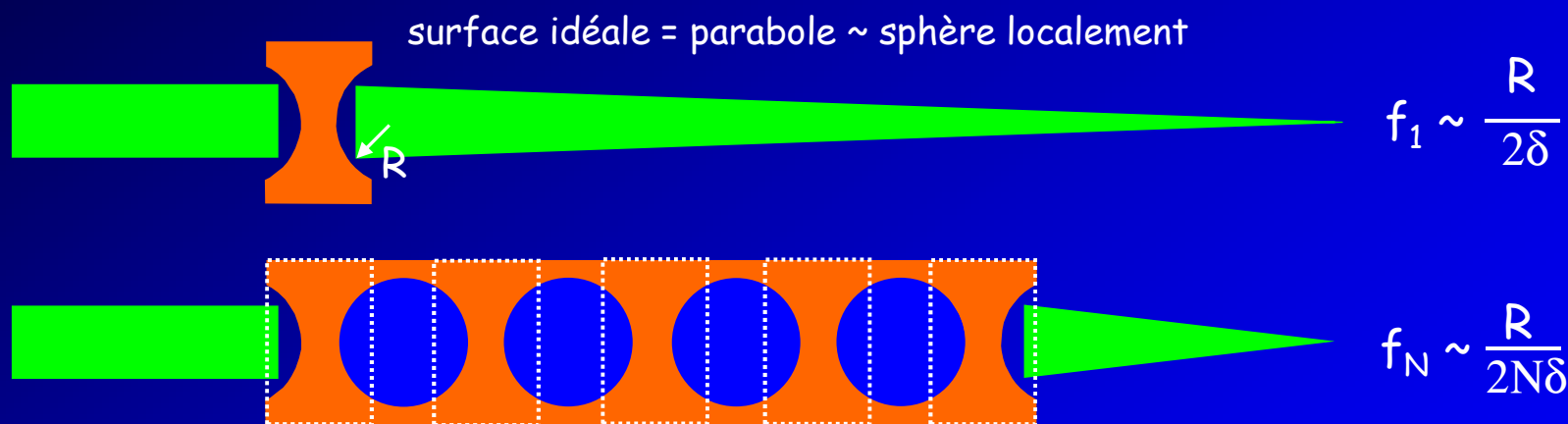
- pureté spectrale
- ++ flux
- ++ flexibilité de fabrication

# Miroirs multicouches rendre le faisceau monochromatique + focalisation



# Optiques pour les Rayons X

## Lentilles réfractives



Ex. Aluminium

$\lambda = 0.9 \text{ \AA}$

$n = 1 - \delta_{Al} = 1 - 2.8 \cdot 10^{-6}$

$R = 300 \text{ }\mu\text{m}$  30 holes

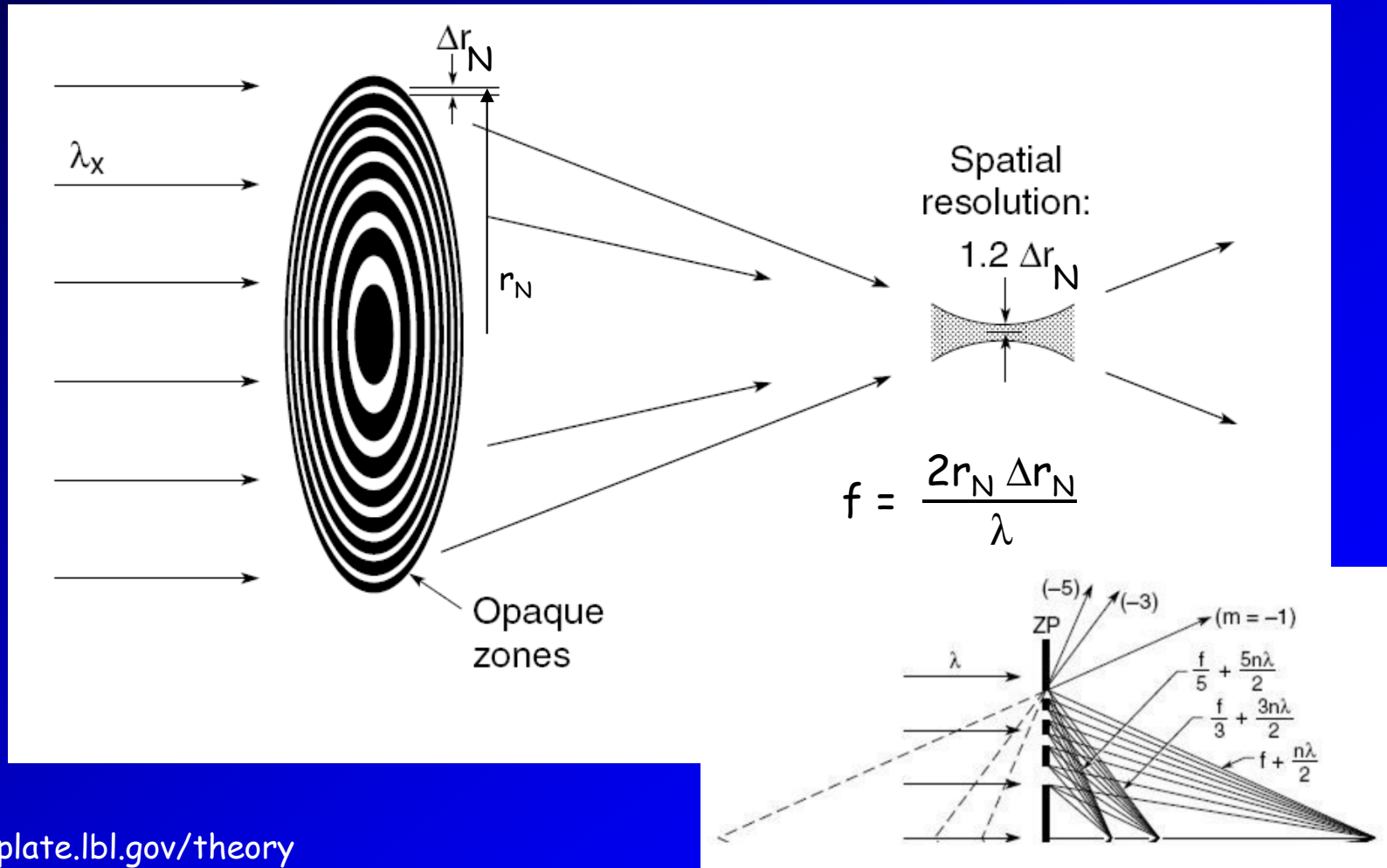
$f_1 = 54 \text{ m}$

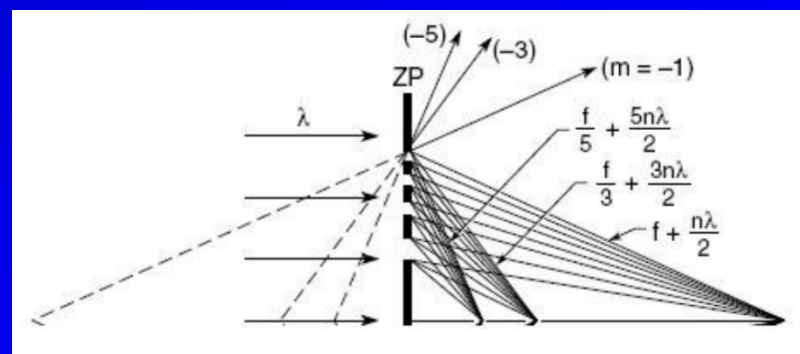
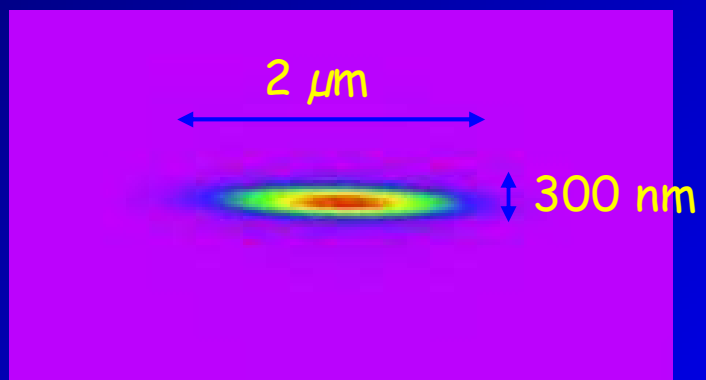
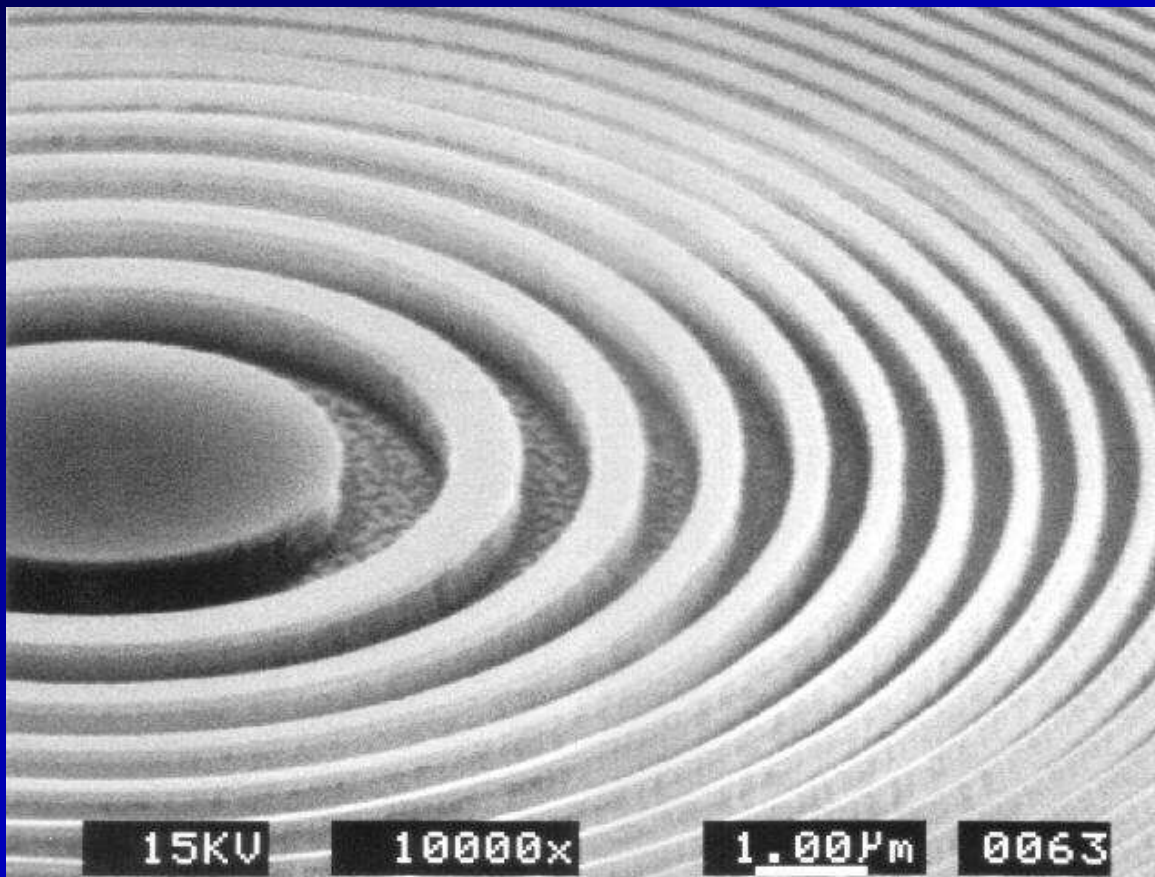
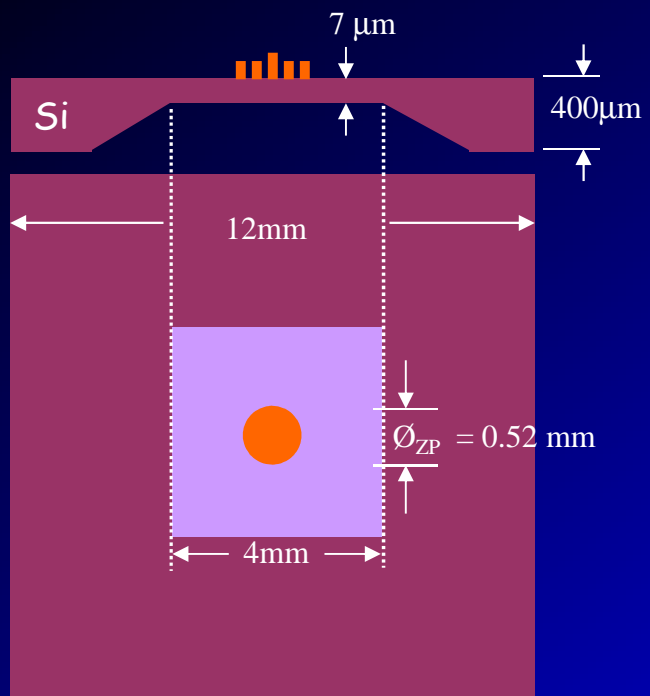
$f_{30} = 1.8 \text{ m}$

Snigirev et al. Nature

# Optiques pour les Rayons X

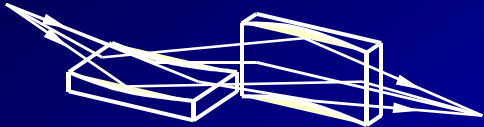


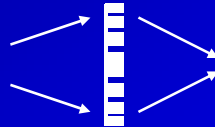

## Lentilles de Fresnel







# Des faisceaux toujours plus petits...

	REFLECTIVE			DIFFRACTIVE	REFRACTIVE	
	Kirkpatrick Baez systems		Capillaries	Waveguides	Fresnel Zone plates	Refractive lenses
	mirrors Kirkpatrick Baez, 1948	multilayers Underwood Barbee, 1986	Kreger 1948	Feng <i>et al.</i> 1993	Baez 1952	Snigirev <i>et al.</i> , 1996
						
<b>E</b>	< 30 keV	< 80 keV	< 20 keV	< 20 keV	< 30 keV (80)	<1 MeV
<b>ΔE/E</b>	wide band	10 <sup>-2</sup>	wide band	10 <sup>-2</sup> – 10 <sup>-3</sup>	10 <sup>-3</sup> - 10 <sup>-4</sup>	10 <sup>-3</sup> - 10 <sup>-4</sup>
<b>resolution/ min. spot size</b>	<b>25 nm 15keV Mimura (2006)</b>	<b>41×45 nm<sup>2</sup> 24keV Hignette (2005)</b>	<b>50 nm Bilderback (1994)</b>	<b>40×25 nm<sup>2</sup> Salditt (2004)</b>	<b>30 nm 20keV Kang, (2006) ~15nm &lt;1keV</b>	<b>50 nm@20keV Schroer (2004) 150 nm@50keV Snigirev (2006)</b>
<b>spot-size</b>	+++	+++	+++	+++	+++	+++
<b>flux</b>	+++	+++	---	---	++	+
<b>achromatic</b>	YES	NO	YES	NO	NO	NO <i>but f(N,E)</i>
<b>coherence</b>	+	+	+/-	+++	++	+/-
<b>in-line</b>	NO	NO	YES	YES	YES	YES
<b>long-f</b>	YES	YES	NO	NO	YES	YES
<b>easy to use</b>	+/-	+/-	++	+/-	++	++
<b>clean-spot</b>	+++	++	+++	+	+	++

# La détection des Rayons X

# La détection

- ✓ Compteurs à scintillations
- ✓ Compteurs à semi-conducteur
- ✓ Image plate
- ✓ Couplage de charge
- ✓ Pixels

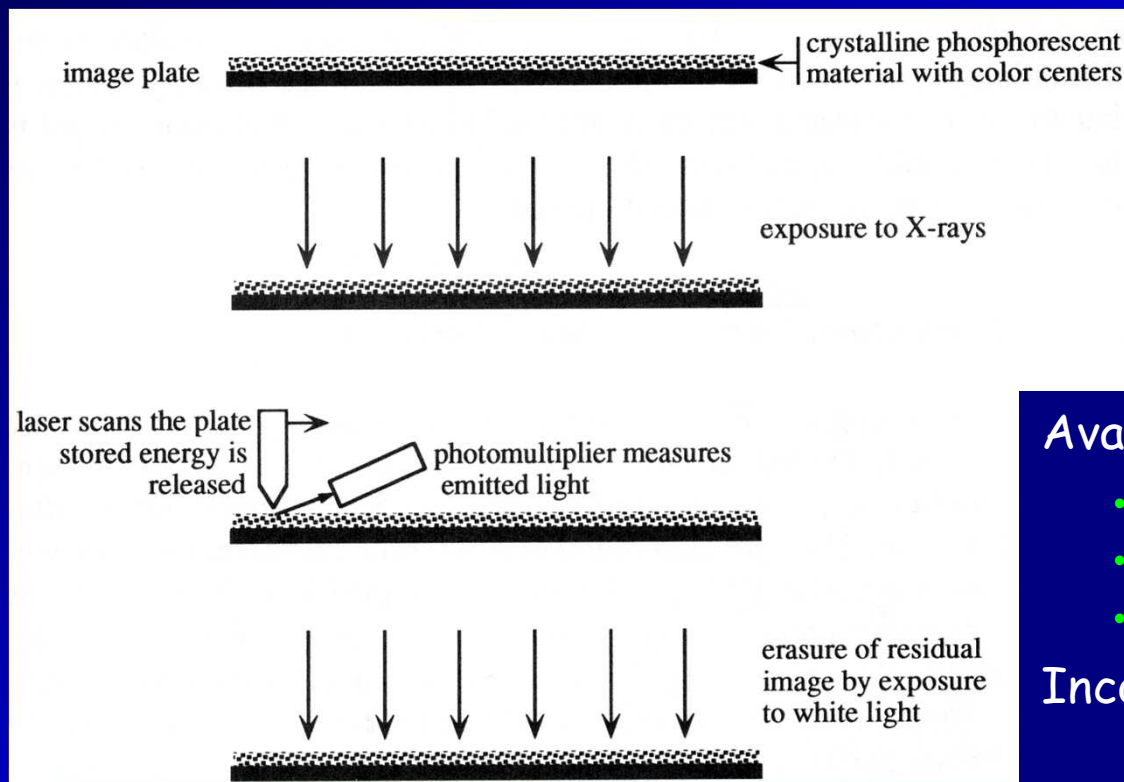
} détecteur bi-dimensionnel 😊

paramètres importants : {  
Efficacité  
Dynamique  
Résolutions (spatiale/énergie)

# Détection des Rayons X

## Image Plate

**Principe** : Plaque avec un mélange de BaFBr:Eu<sup>2+</sup>  
cristaux photo-stimulables = centres photosensibles =  
stockent une partie de l'énergie des rayons X qui les  
touchent ( $t_{1/2} \sim 8h$ )



### Avantages:

- grande surface
- bonne dynamique ( $> 10^5$ )
- résolution acceptable ( $\sim 100\mu m$ )

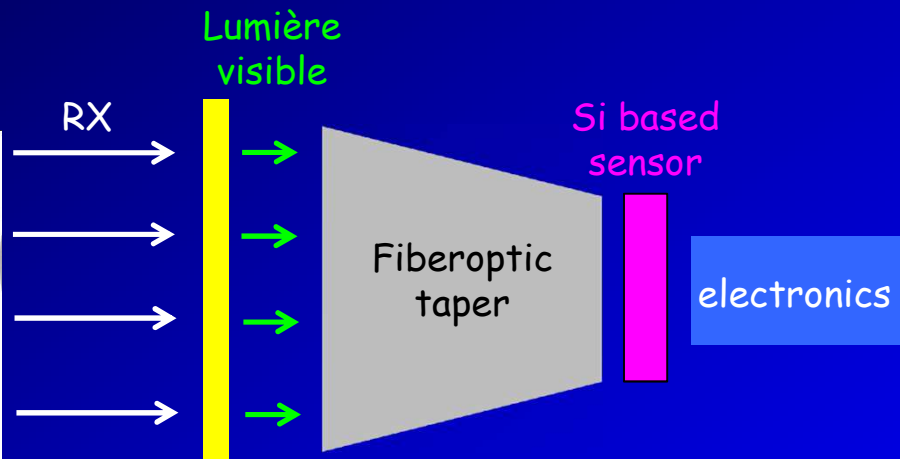
### Inconvénient:

temps de lecture ( $\sim 1-2$  min)

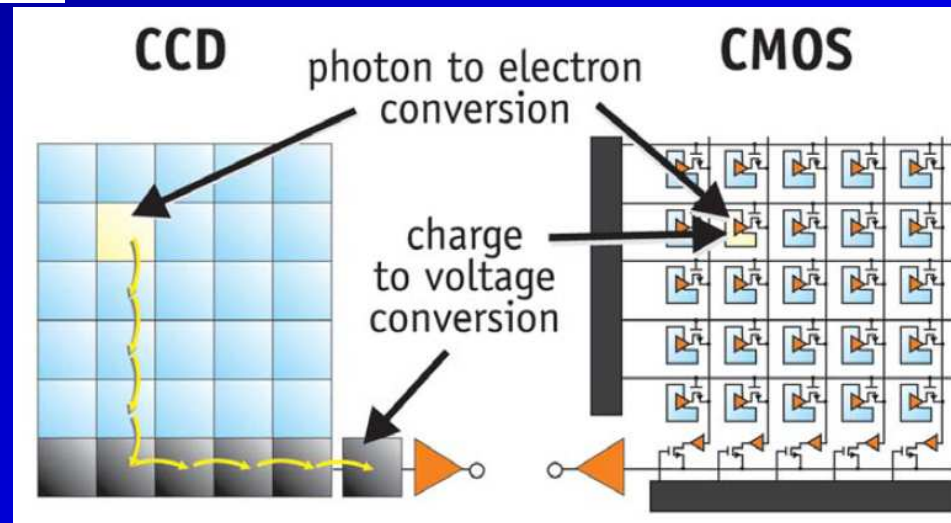
# X Ray detection

## Détecteur à intrégration de charges (CCD/CMOS)

Principe :



Charge Coupled Device

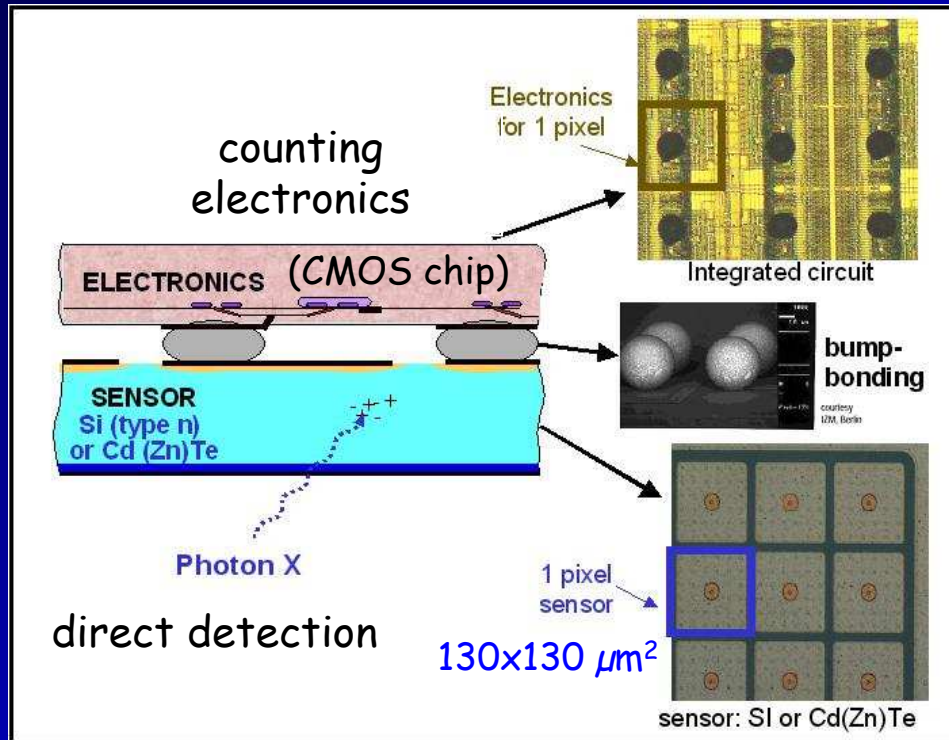


Complementary Metal-Oxide Semiconductor

# Détection des Rayons X

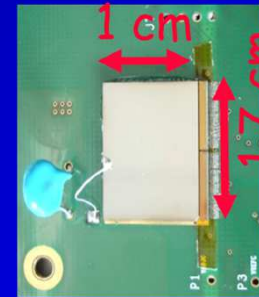
## Détecteur compteur de photon (Hybrid Pixel Detectors)

**Principe** : chaque pixel est un petit détecteur indépendant, ayant sa propre chaîne de décision (amplification/seuil) et son système de lecture

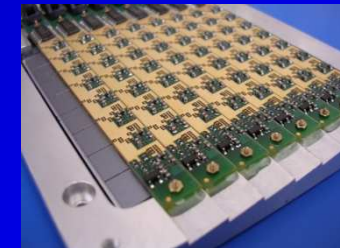


**XPAD**

$7 \times 12 \text{ cm}^2$



$80 \times 120$  pixels



$8 * (7 \text{ chips})$



autre ex. : Medipix, Pilatus ...

# X Ray detection

	CCD	CMOS	Hybrid pixels
type	charge integrating	charge integrating	photon counting
signal out of pixel	e- packet	Voltage	Voltage
signal out of chip	Voltage (analogue)	Bits (digital)	Bits (digital)
signal out of camera	Bits (digital)	Bits (digital)	Bits (digital)
sensor complexity	+	-	--
pixel size	+ (~ 50 $\mu\text{m}$ )	+ (~ 50 $\mu\text{m}$ )	- (~ 130 $\mu\text{m}^*$ )
dynamic range	+ (15-18 bits)	- (15-18 bits)	++ (32 bits*)
uniformity (dark/illumination)	+/+	-/+	++/++
speed	- (5Hz)	+ (500 Hz)	++ (1 kHz)
windowing (Region Of Interest)	--	++	++
antiblooming	-	++	++
continuous scans (shutter free)	-	+	+
dead zones	+	+	--

(\* ImXPAD detector)

Techniques usuelles de diffraction

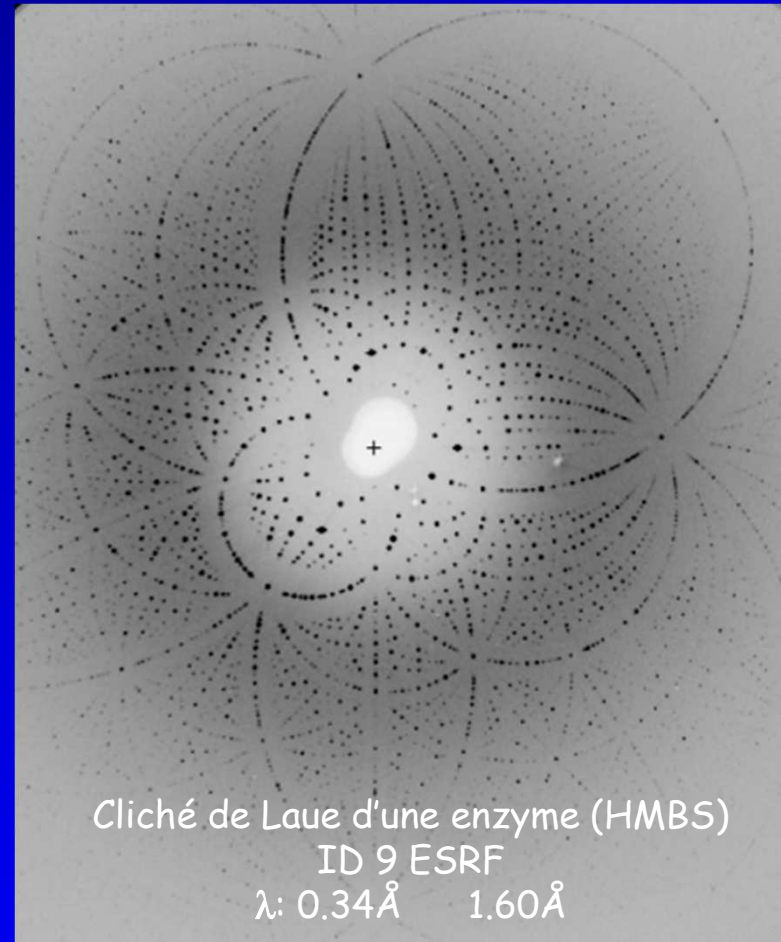
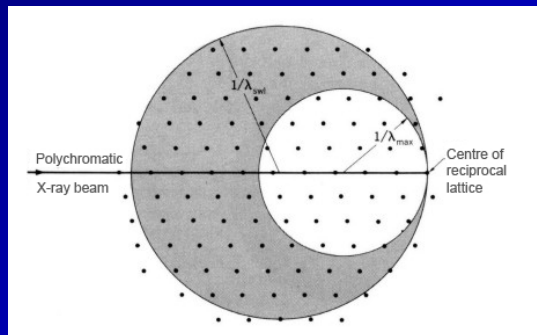
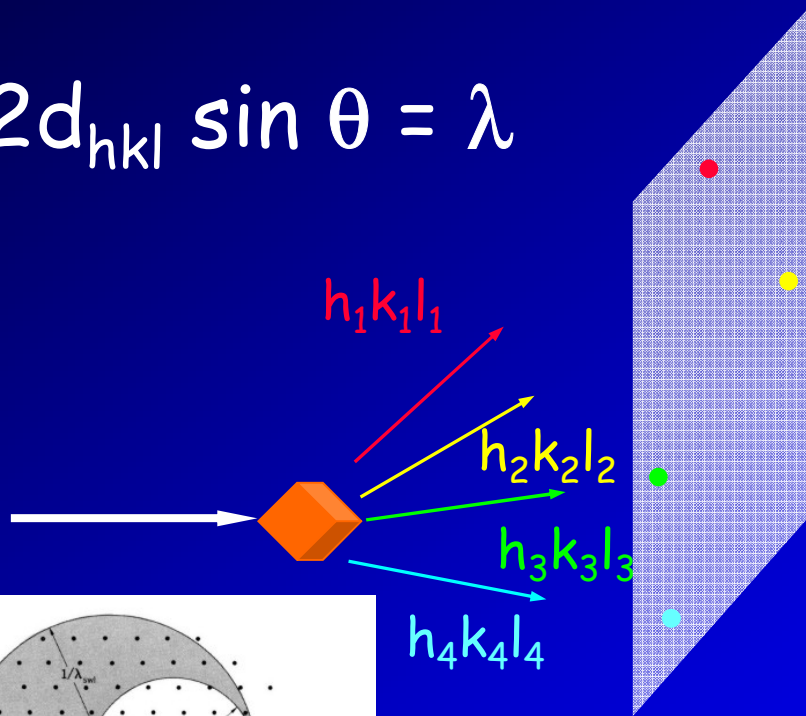
Diffraction sur monocristal



# Diffraction sur monocristal

**Méthode de Laue** faisceau blanc, capturer tous les  $\theta$

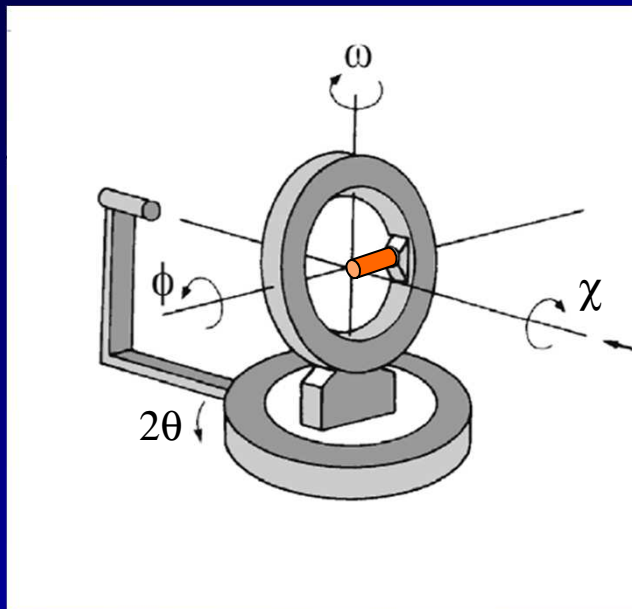
$$2d_{hkl} \sin \theta = \lambda$$



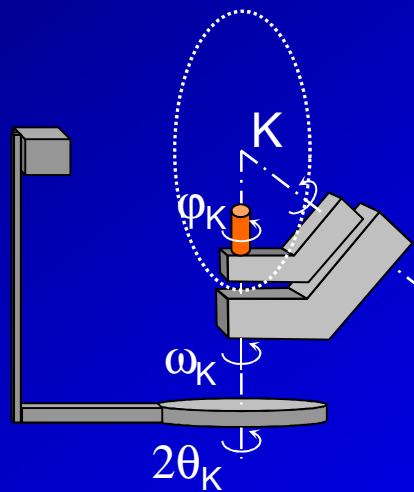
➡ Enregistrement simultané d'un grand nombre de raies de Bragg

# Diffraction sur monocristal

**Diffractomètre 4 cercles** faisceau monochromatique  
Orienter le cristal dans une direction quelconque  
Mesurer l'intensité des taches de Bragg

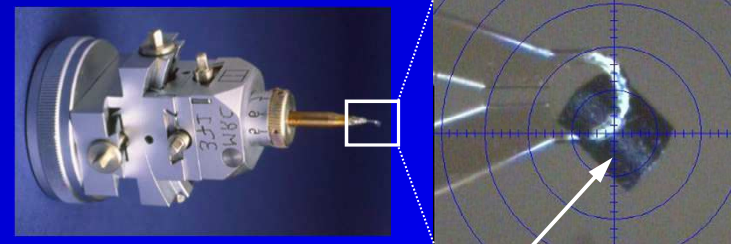


Géométrie eulérienne



Géométrie « kappa »

( le plus grand nombre possible  
avec la meilleure précision )

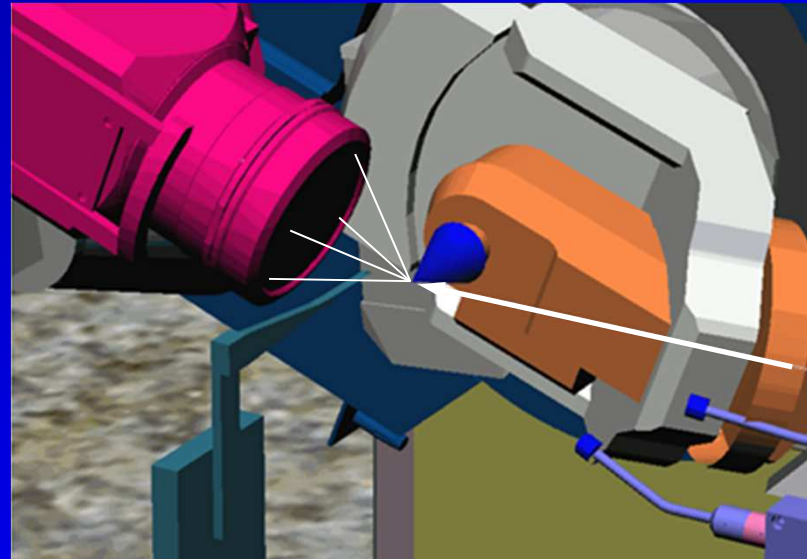
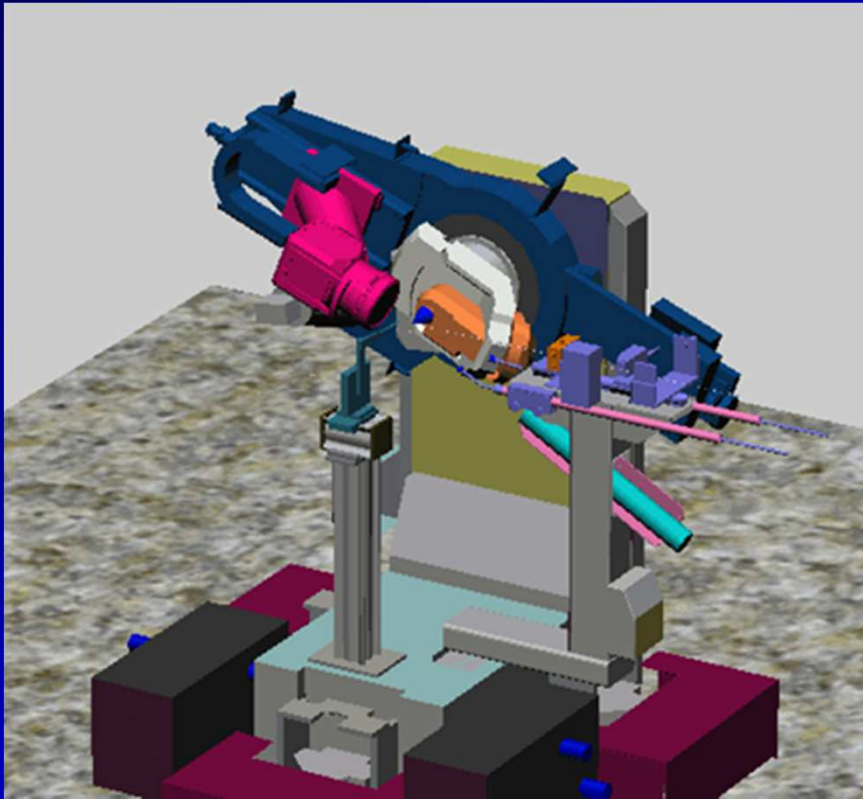


échantillon:  $\text{dim}_{\text{max}} \sim 150 \mu\text{m}$

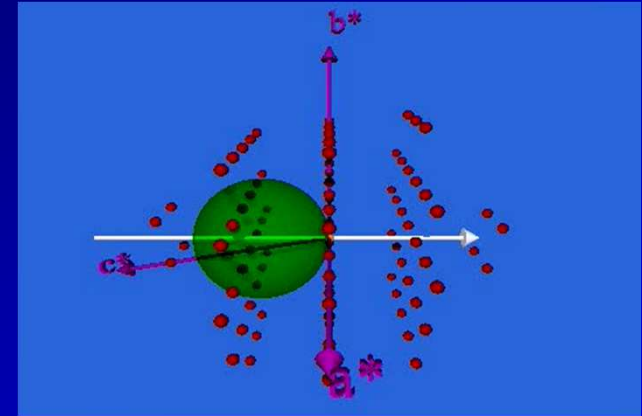
➔ Géométrie « kappa » pour réduire les effets d'ombrage  
Détecteur 2D  $\chi =$  combinaison des rotations  $\omega$ ,  $\kappa$  et  $\varphi$

# Méthode d'oscillation

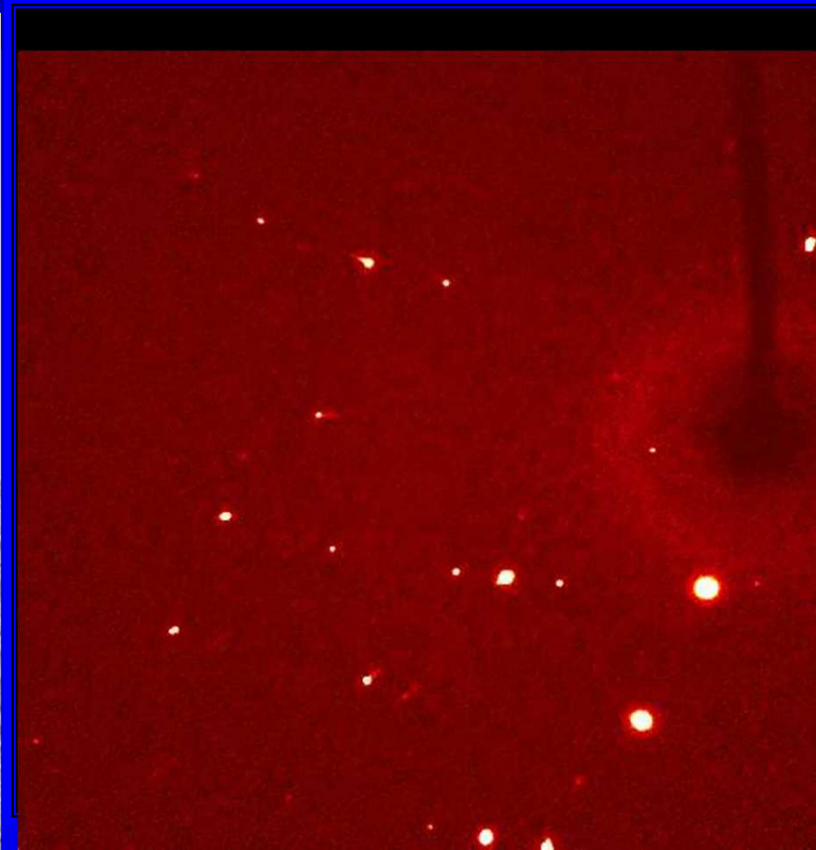
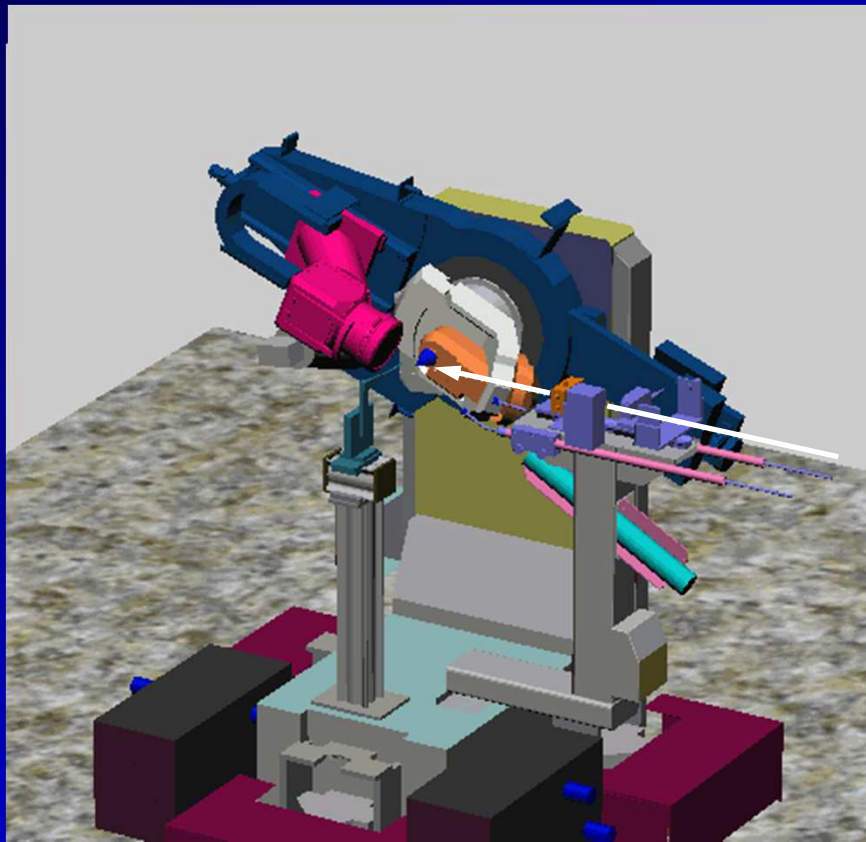
rotation du cristal (typiquement  $1^\circ$ ) et collecte simultanée des intensités



# Méthode d'oscillation



rotation du cristal (typiquement  $1^\circ$ ) et collecte simultanée des intensités



Techniques usuelles de diffraction

Diffraction par les poudres

# Méthode des poudres

**Poudre** : ensemble de monocristaux (<1-10  $\mu\text{m}$ ) orientés aléatoirement.

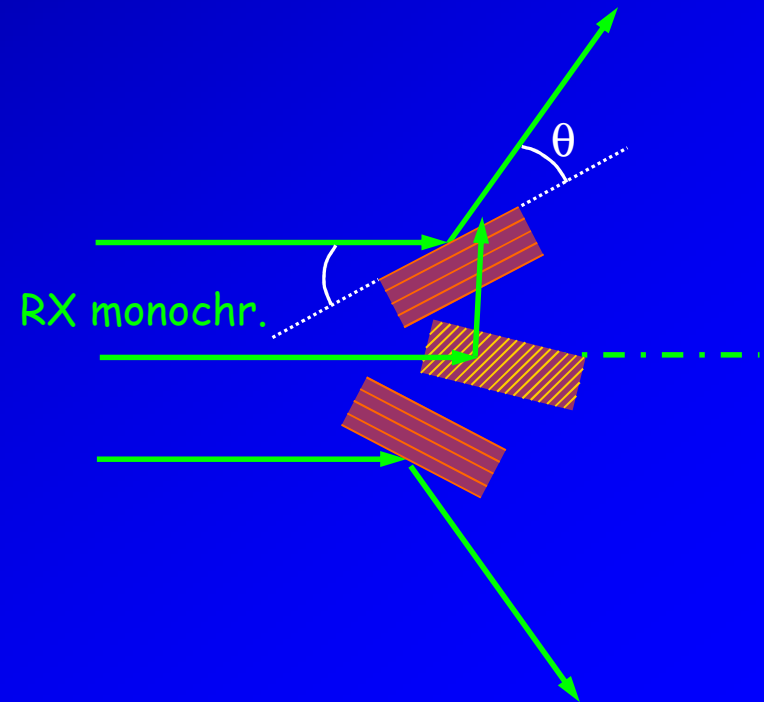
➡  $2d \sin \theta = \lambda$     satisfaite  $\forall d$

✓ dispersion angulaire     $\lambda = \text{fixed}$

$$d_{hkl} \Rightarrow \theta_{hkl}$$

✓ dispersion d'énergie     $\theta = \text{fixed}$

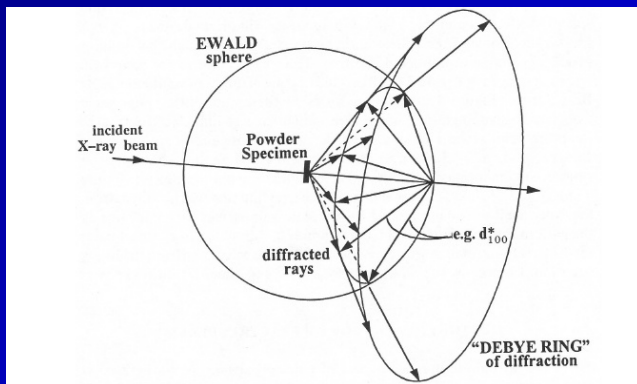
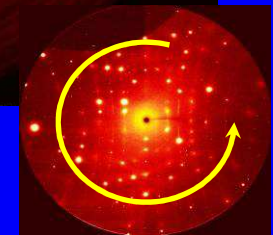
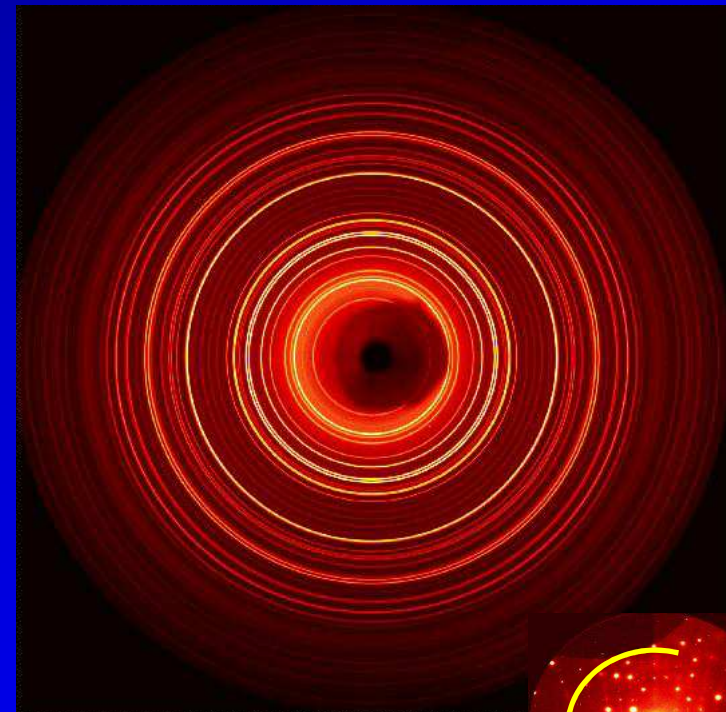
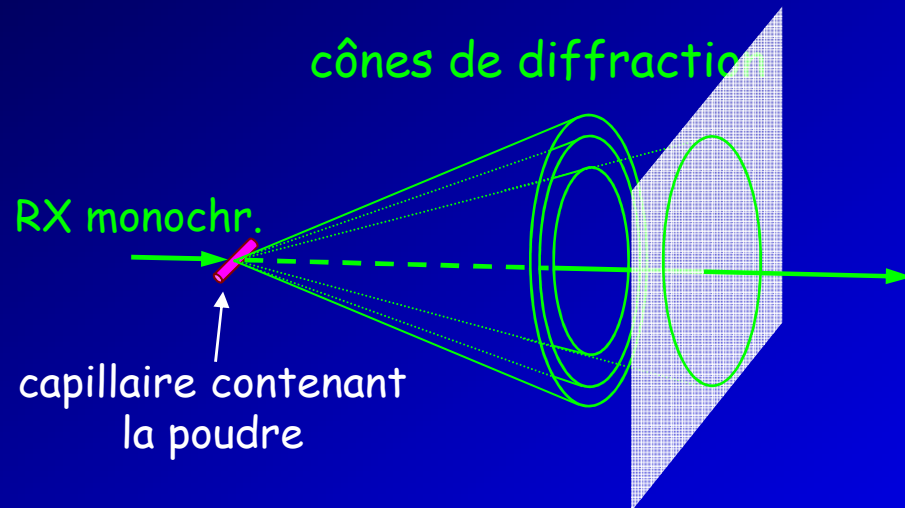
$$d_{hkl} \Rightarrow \lambda_{hkl}$$



# Méthode des poudres

**Poudre** : ensemble de monocristaux (<1-10  $\mu\text{m}$ ) orientés aléatoirement.

➔  $2d \sin \theta = \lambda$  satisfaite  $\forall d$

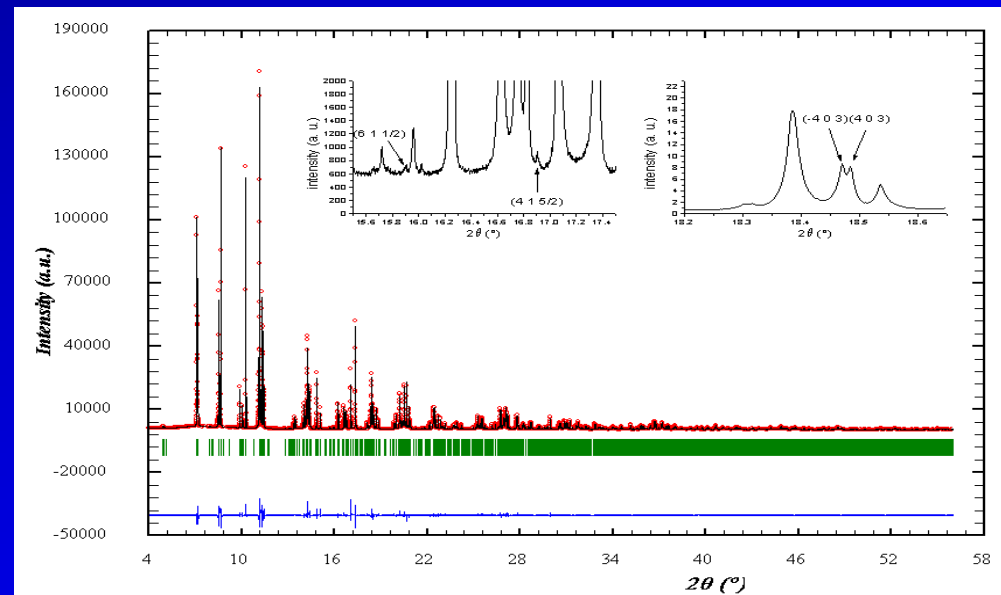
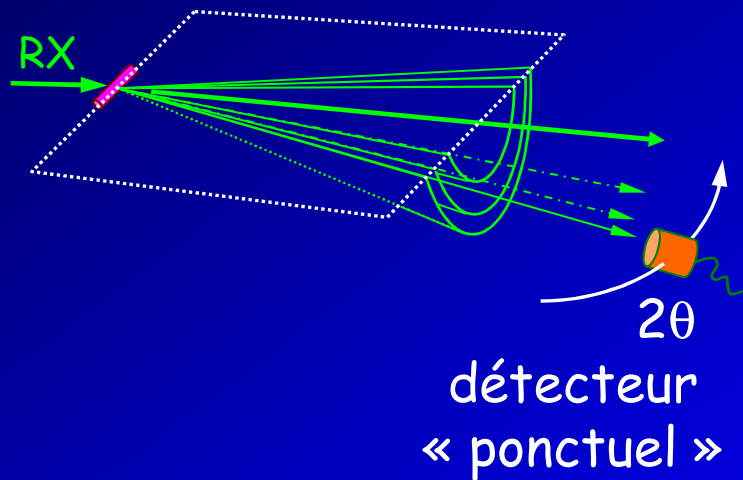
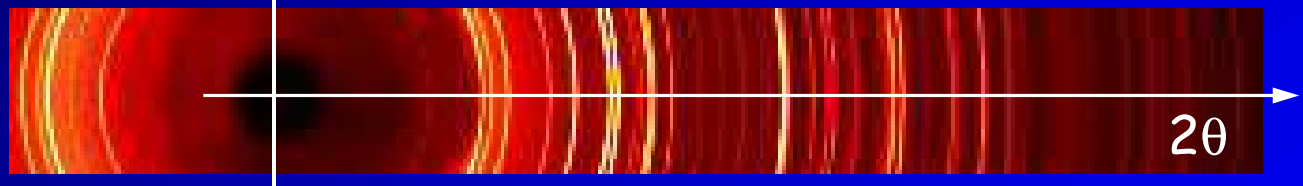


détecteur 2D  
Image plate  
CCD  
Détecteur pixels

# Méthode des poudres

**Poudre** : ensemble de monocristaux (<1-10  $\mu\text{m}$ ) orientés aléatoirement.

➔  $2d \sin \theta = \lambda$     satisfaite  $\forall d$

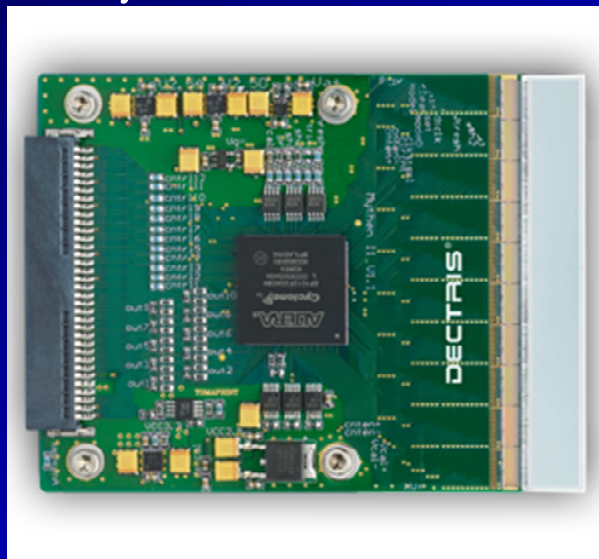




# Méthode des poudres

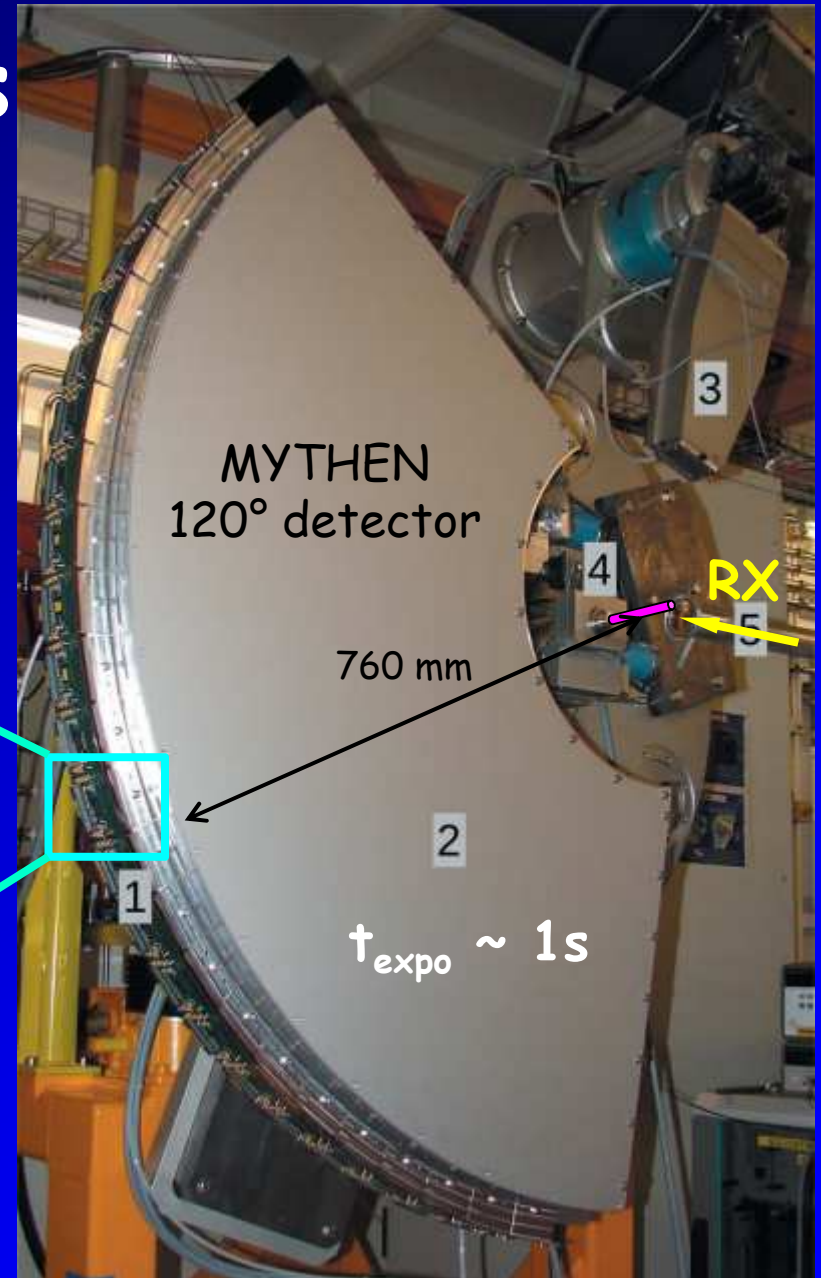
Materials Science @ Swiss Light Source

*pixel detector*



1280 strips

1 strip HxV = 8mm x 55  $\mu$ m

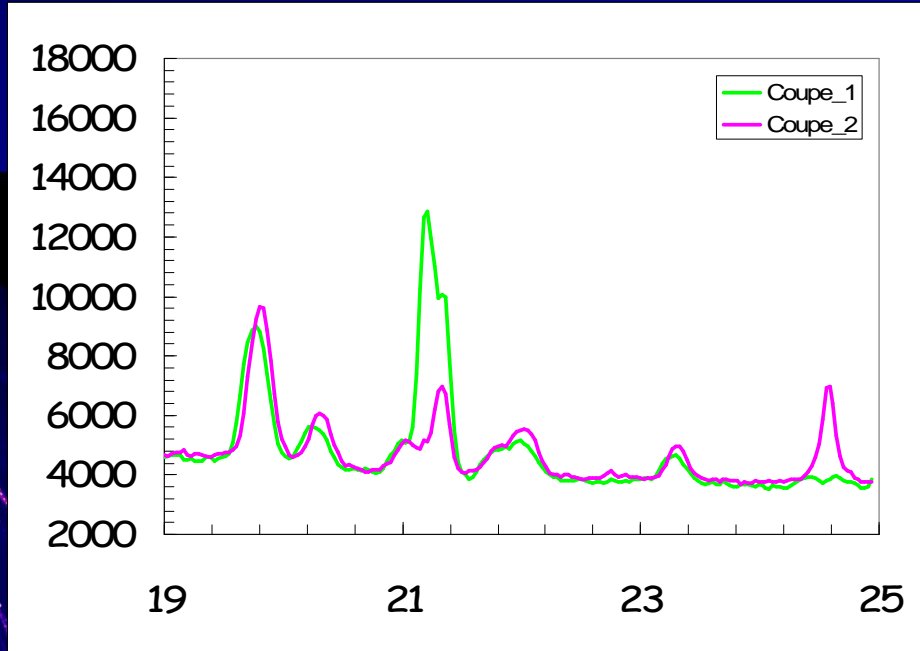
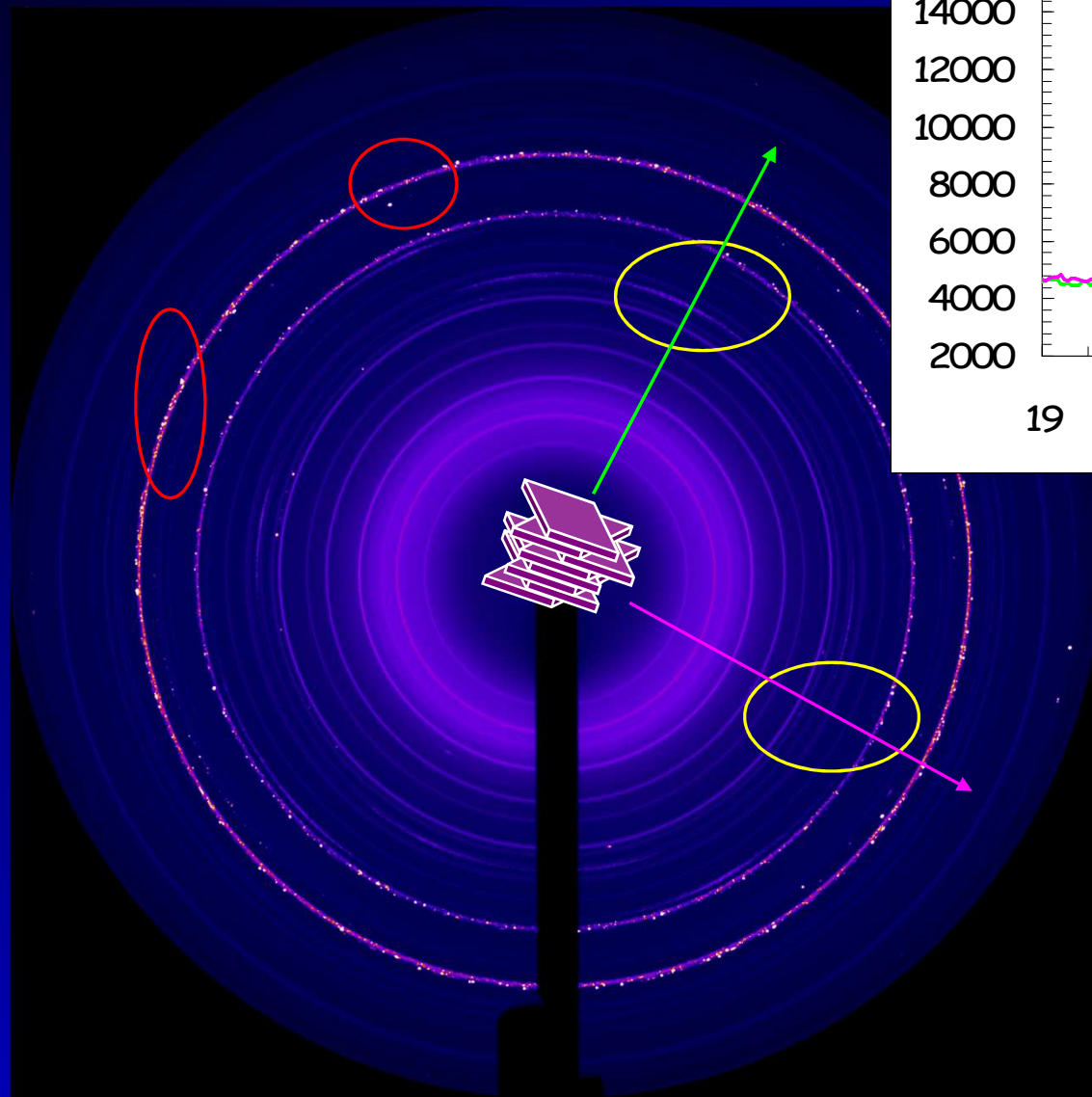


MYTHEN  
120° detector

760 mm

$t_{\text{expo}} \sim 1\text{s}$

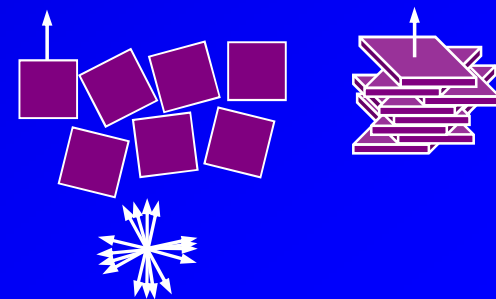
RX



anneaux +/- continus

effet du nombre de grains

texturation de l'échantillon



# Méthode des poudres

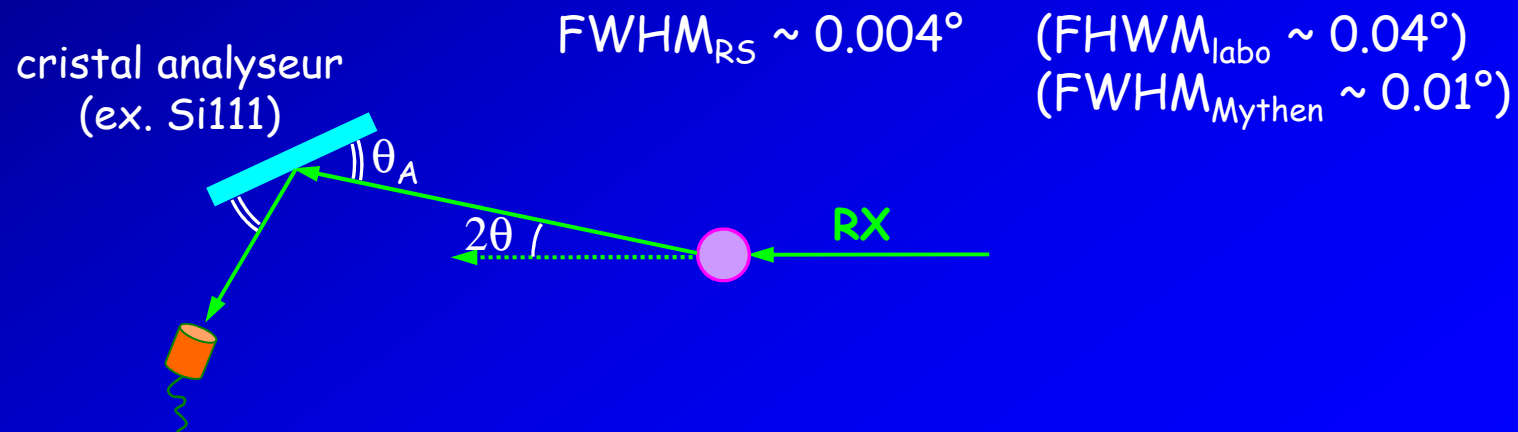
## Haute résolution

améliorations :

✓ du rapport signal/bruit

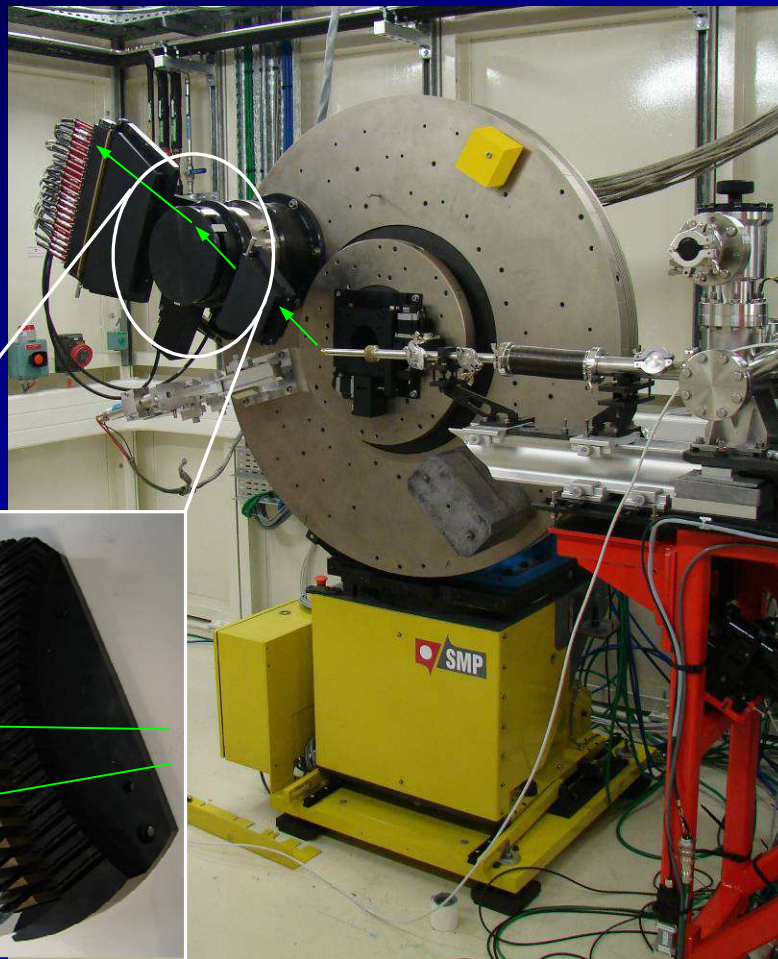
$$I_{\max} / \text{bruit} \sim 1000$$

✓ de la largeur des raies

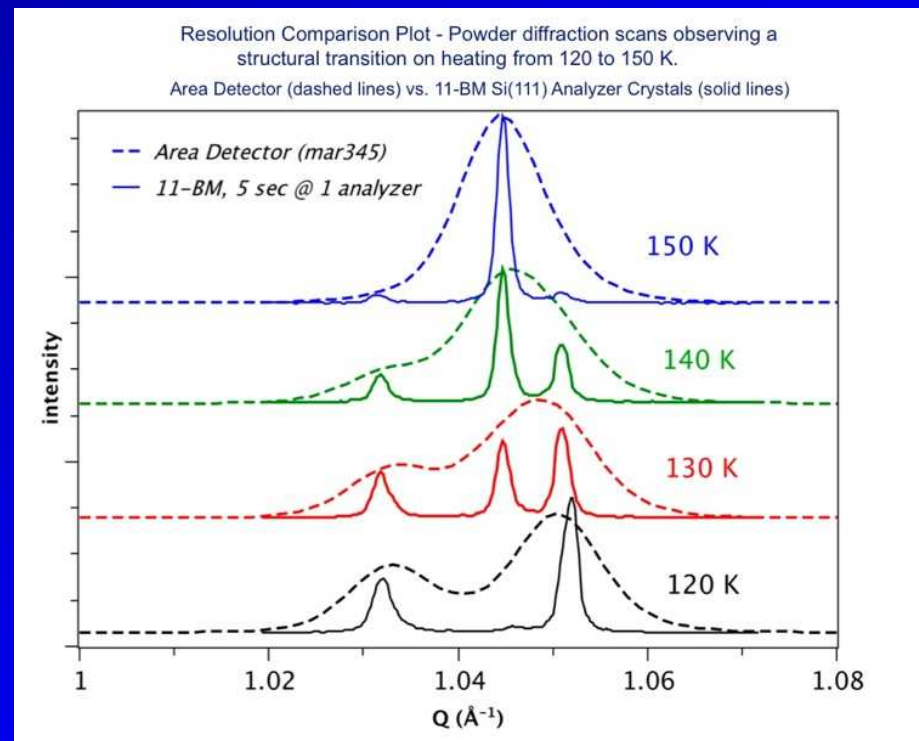


# Méthode des poudres

## Diffractomètre 2 cercles @ CRISTAL



exemple: transition de phase structurale



# Méthode des poudres: intérêts

- ✓ si pas de (« gros ») monocristal, fonction de distribution de paires (pdf)
- ✓ identification de phase
- ✓ rapide
- ✓ ...

**mais** info 3D perdue!!!  Raies symétriquement équivalentes superposées  
Raies à  $d_{hkl}$  proches se recouvrent...

méthode dispersive

environnement échantillon contraignant (étude sous P)

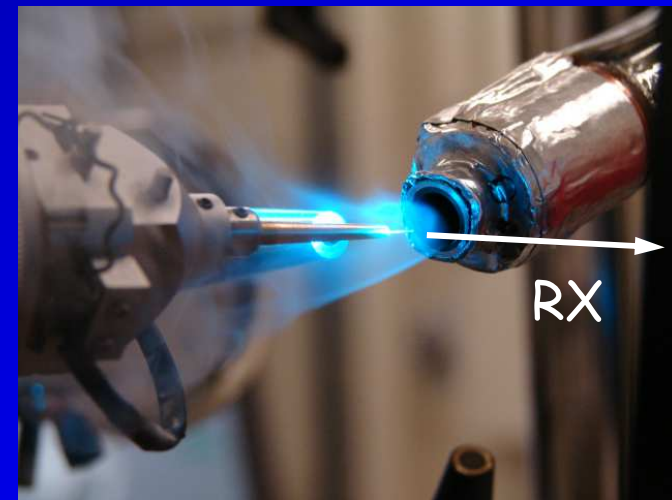
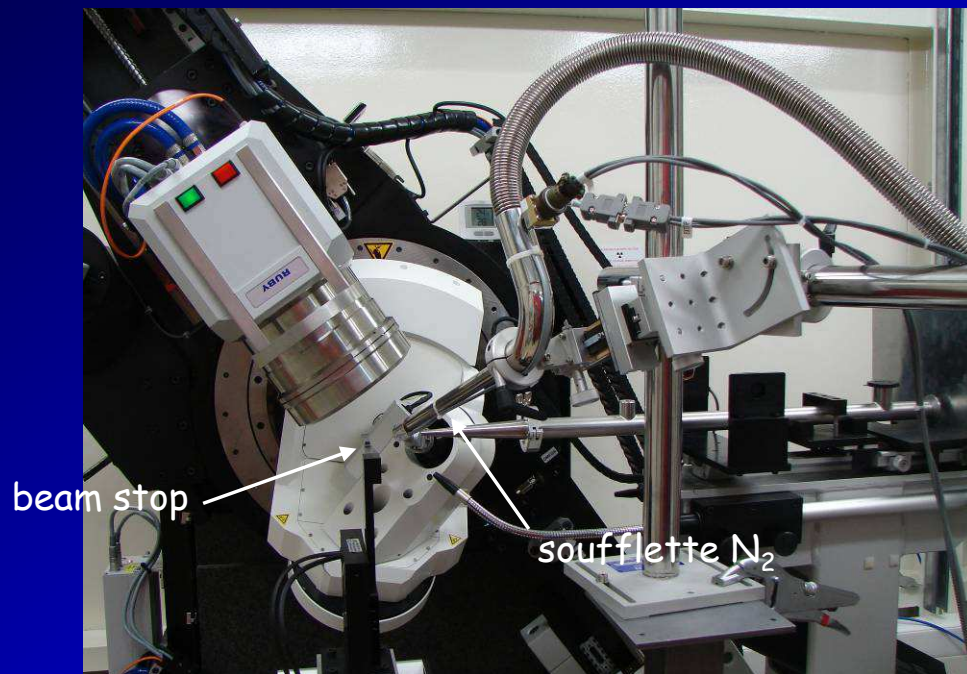
détecteur résolu en énergie

**Diffraction sous conditions  
non ambiantes**

# Conditions non ambiantes

## Basse (haute) température

- 1) souffler un gaz froid (chaud) ( $N_2$ , He) sur l'échantillon

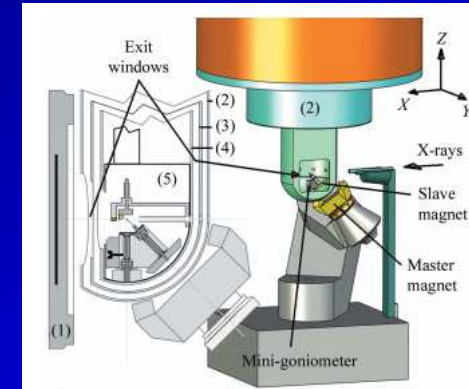


$11K < T < 500K$

# Conditions non ambiantes

Basse (haute) température

2) placer l'échantillon dans un cryostat (four)

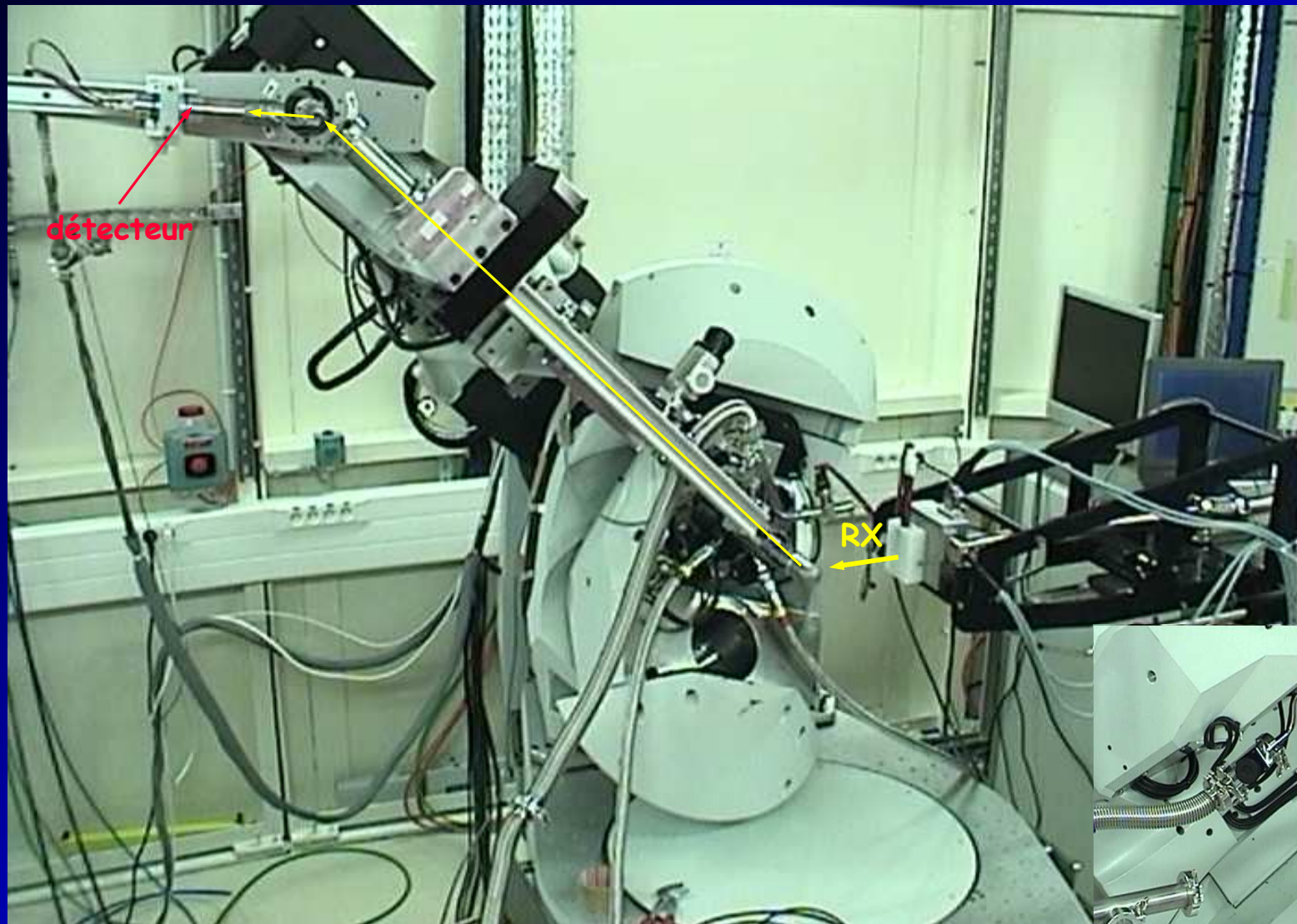


Cryostat à bain d'He:  $T_{\min} \sim 3\text{K}$

Fenêtre/dôme de Be



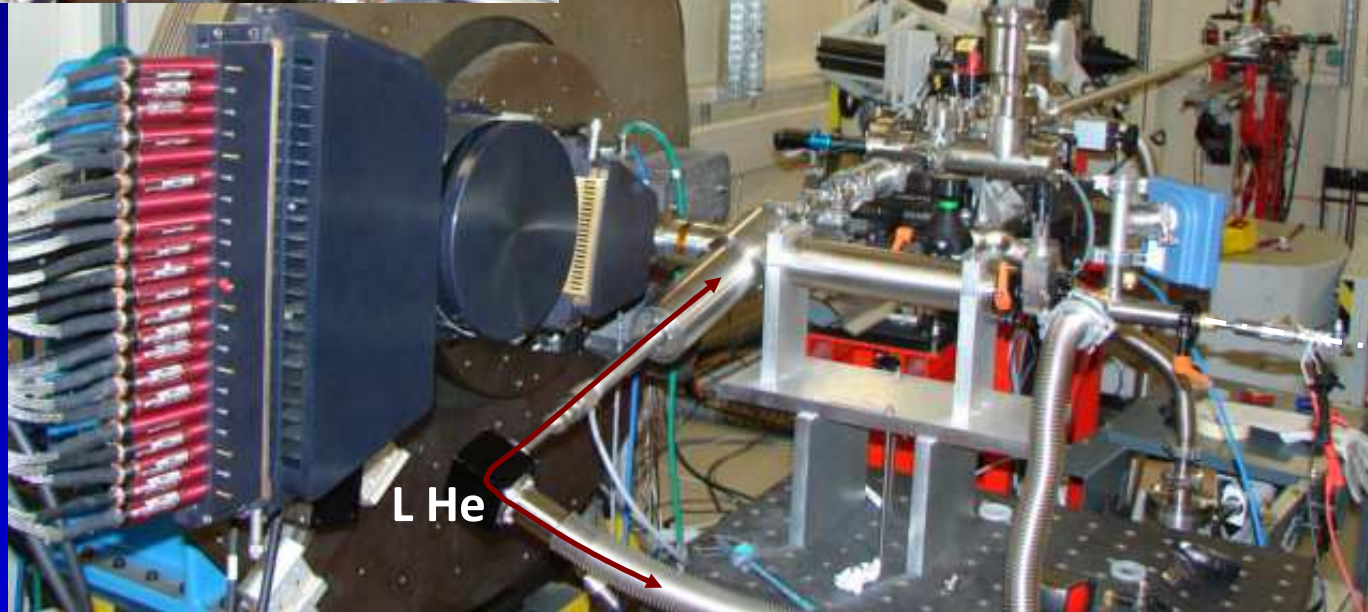
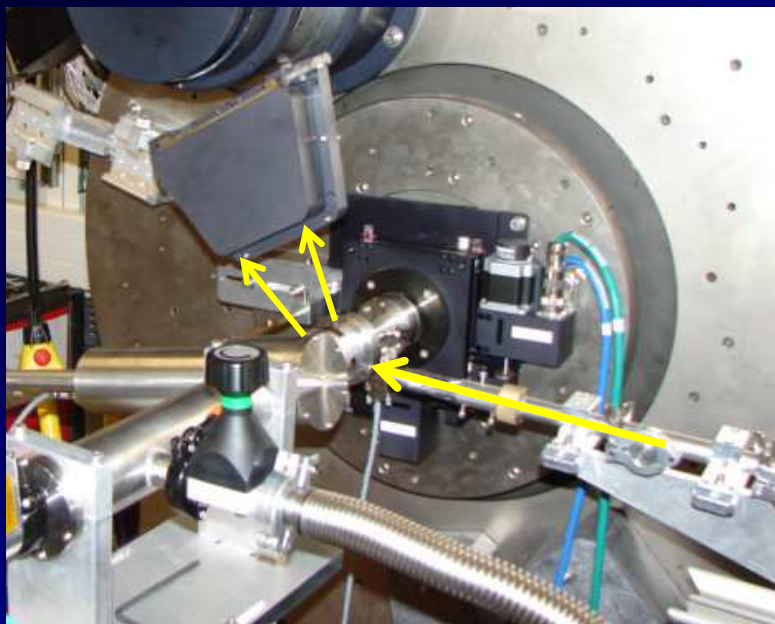
# Conditions non ambiantes



échantillon dans un cryostat (four)  
supporté par le diffractomètre

# Conditions non ambiantes

Diffraction de poudres à basse température

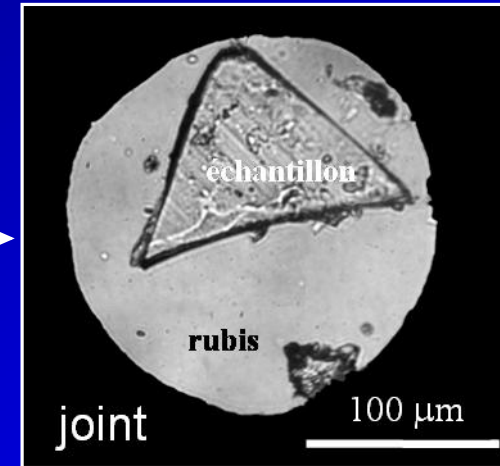
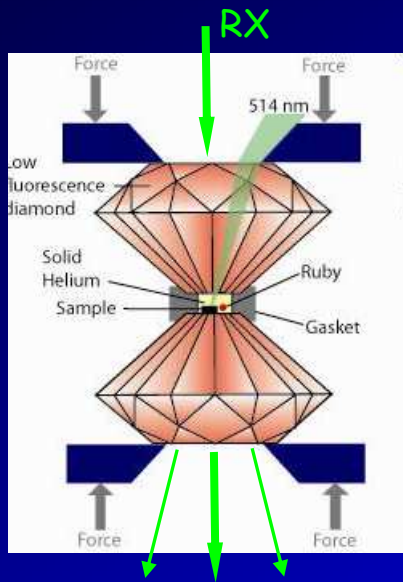


L He

rotation  
 $\omega$  t/s

# Conditions non ambiantes

## Pression



cellule à enclume de diamant

transparente RX/visible

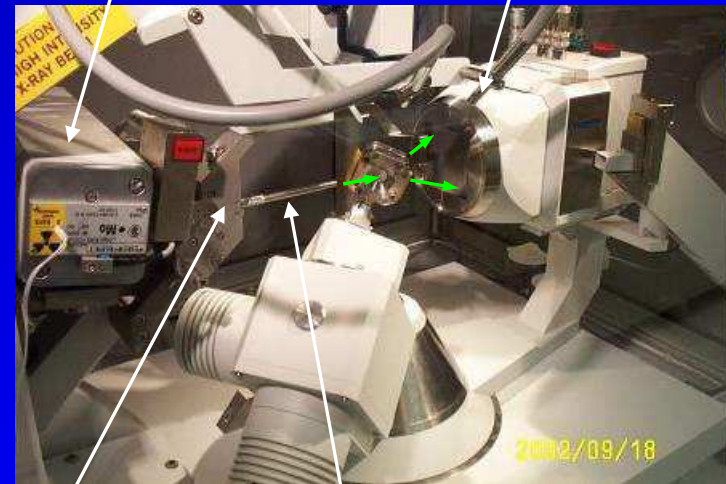
$P_{\max} \sim 5 - 300 \text{ GPa}$

faible encombrement (implantable dans cryostat)

angle d'ouverture limité

source RX

détecteur

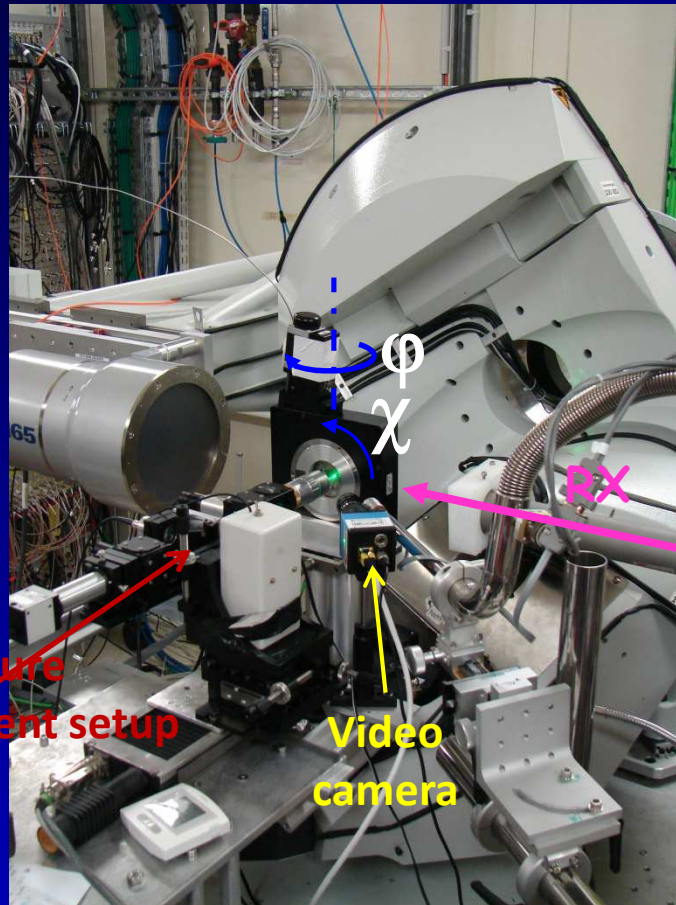


monochromateur collimateur

1 GPa = 10 kbars

# Conditions non ambiantes

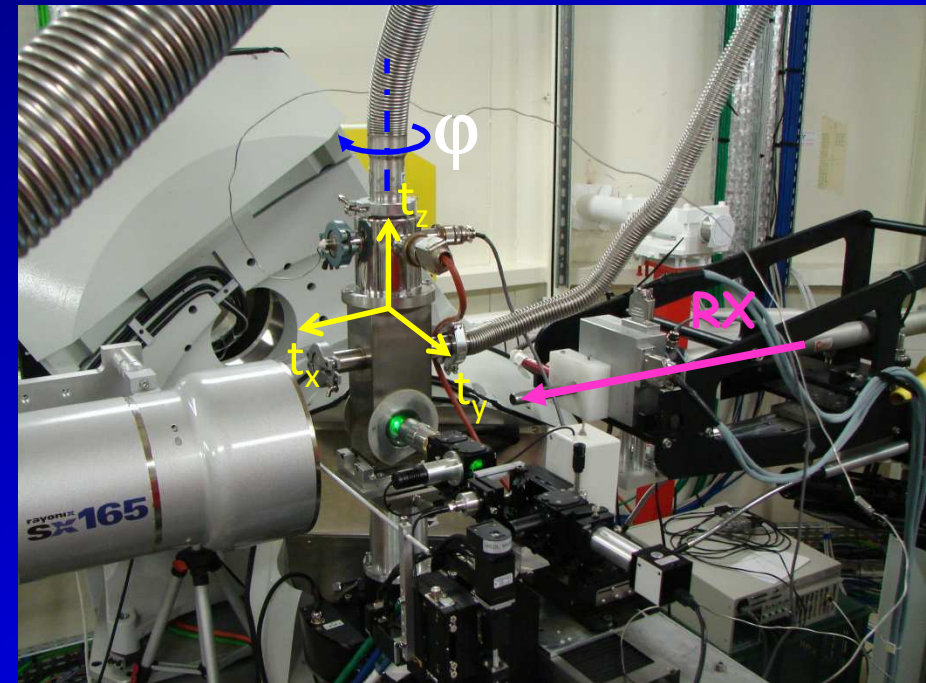
Ajustement in-situ de la pression in-situ (Membrane Diamond Anvil Cell)



Pressure measurement setup

Video camera

Température Ambiante  
(configuration mesure de pression)



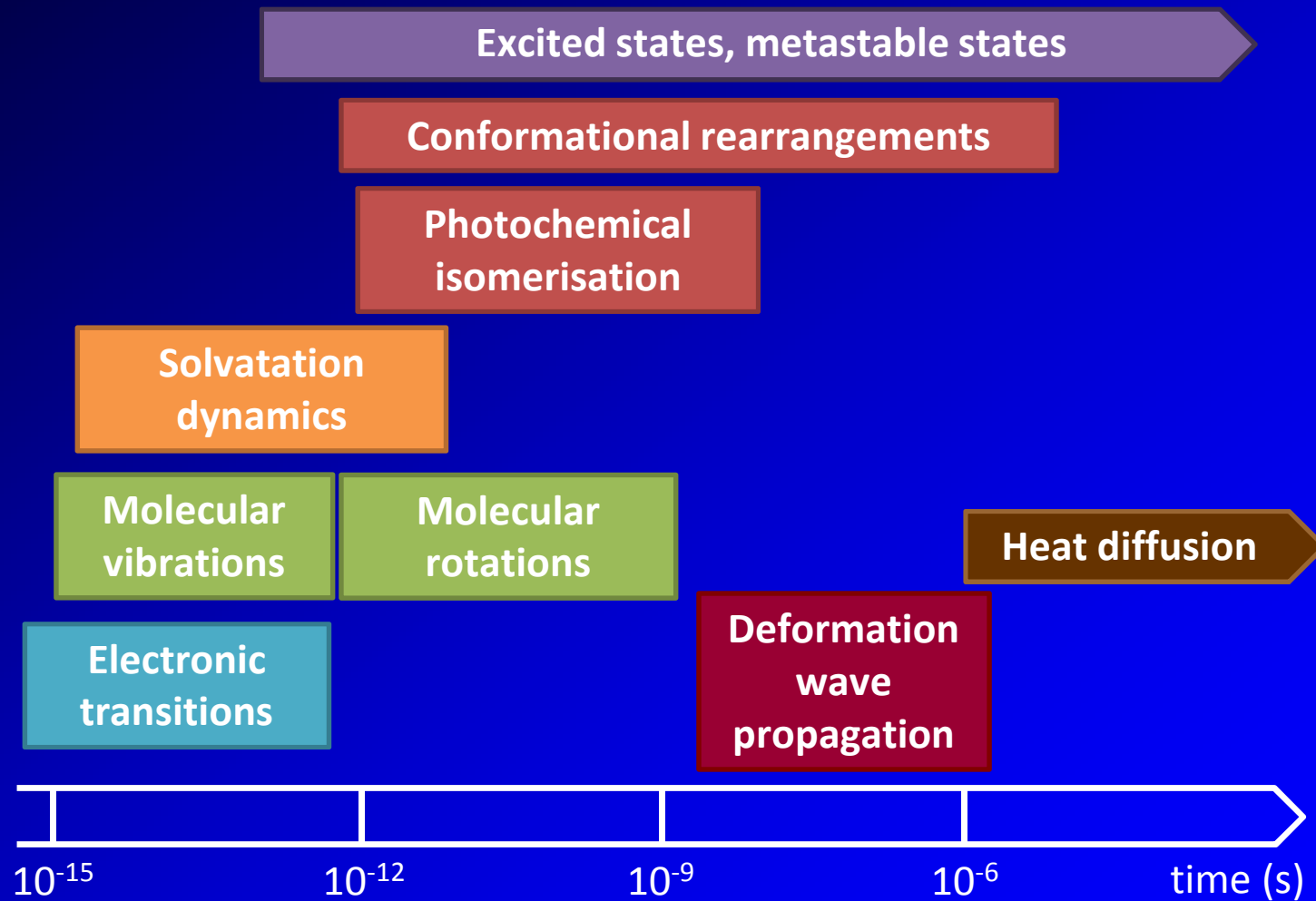
Basses Températures

$T_{\min} = 4 \text{ K}$

Experimental setup @ 6-circle.cristal.soleil

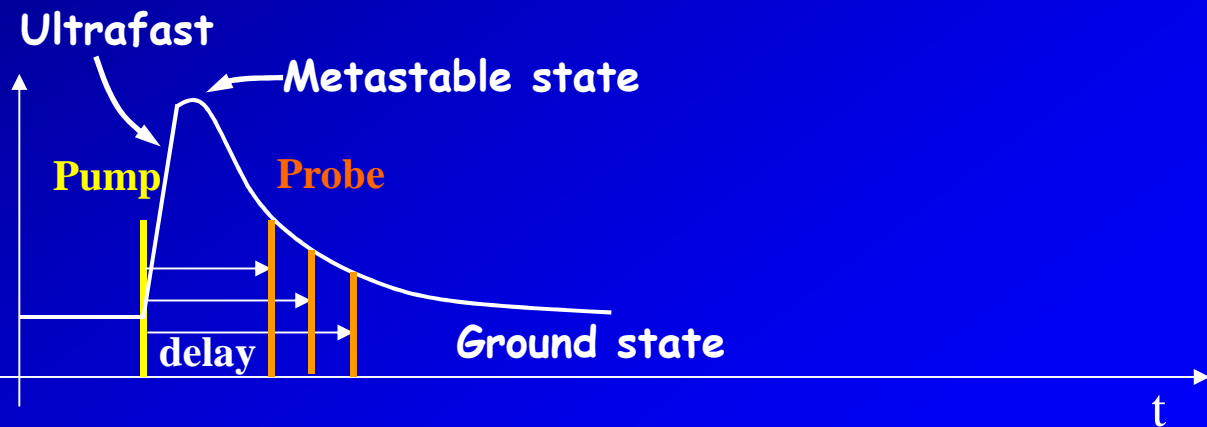
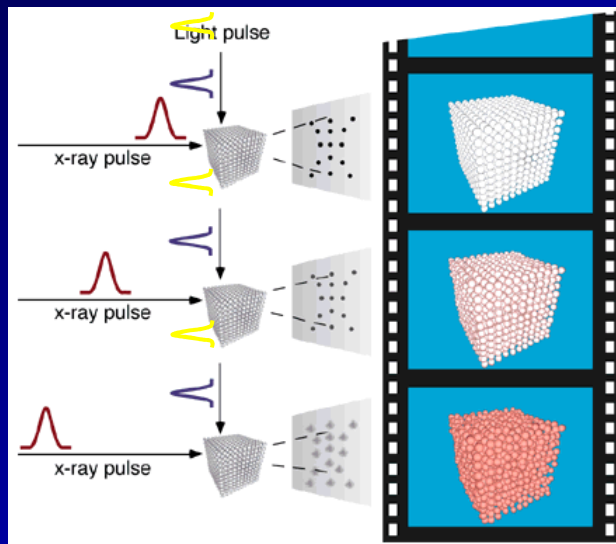
# Non ambient Conditions

Fast (ultra-fast) Time resolved experiments



# Non ambient Conditions

## Time resolved experiments



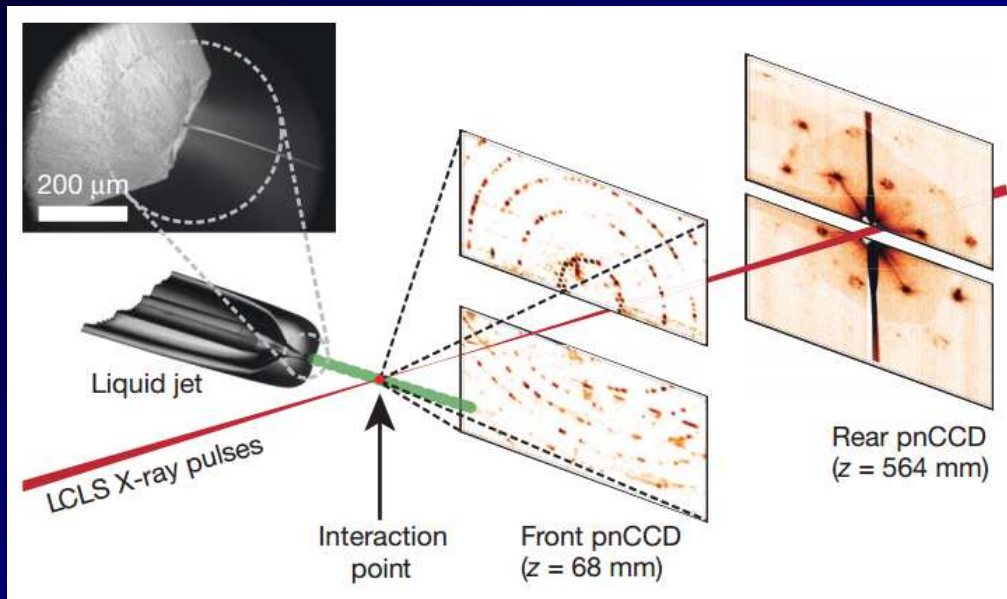
"single shot" method very brillante source(cf. XFEL)



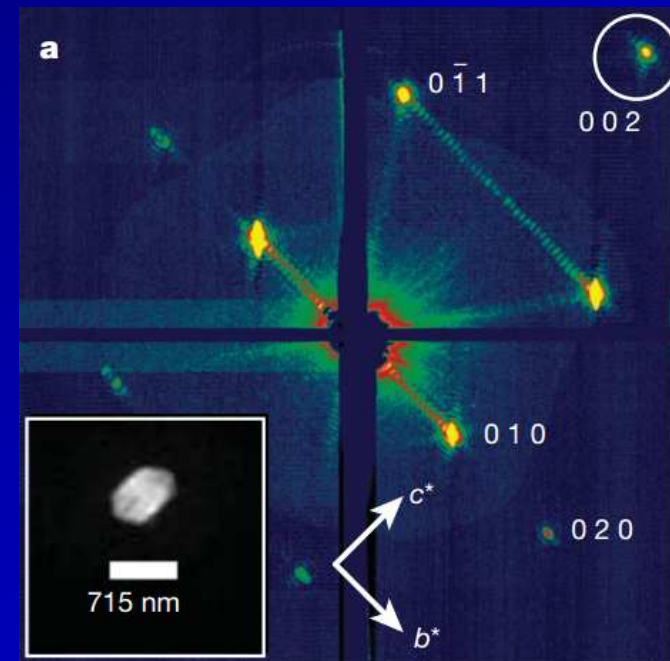
sample

# XFEL source - ex. of a « single shot » experiment

## Protein crystallography lysozyme @SFX.LCLS.eu

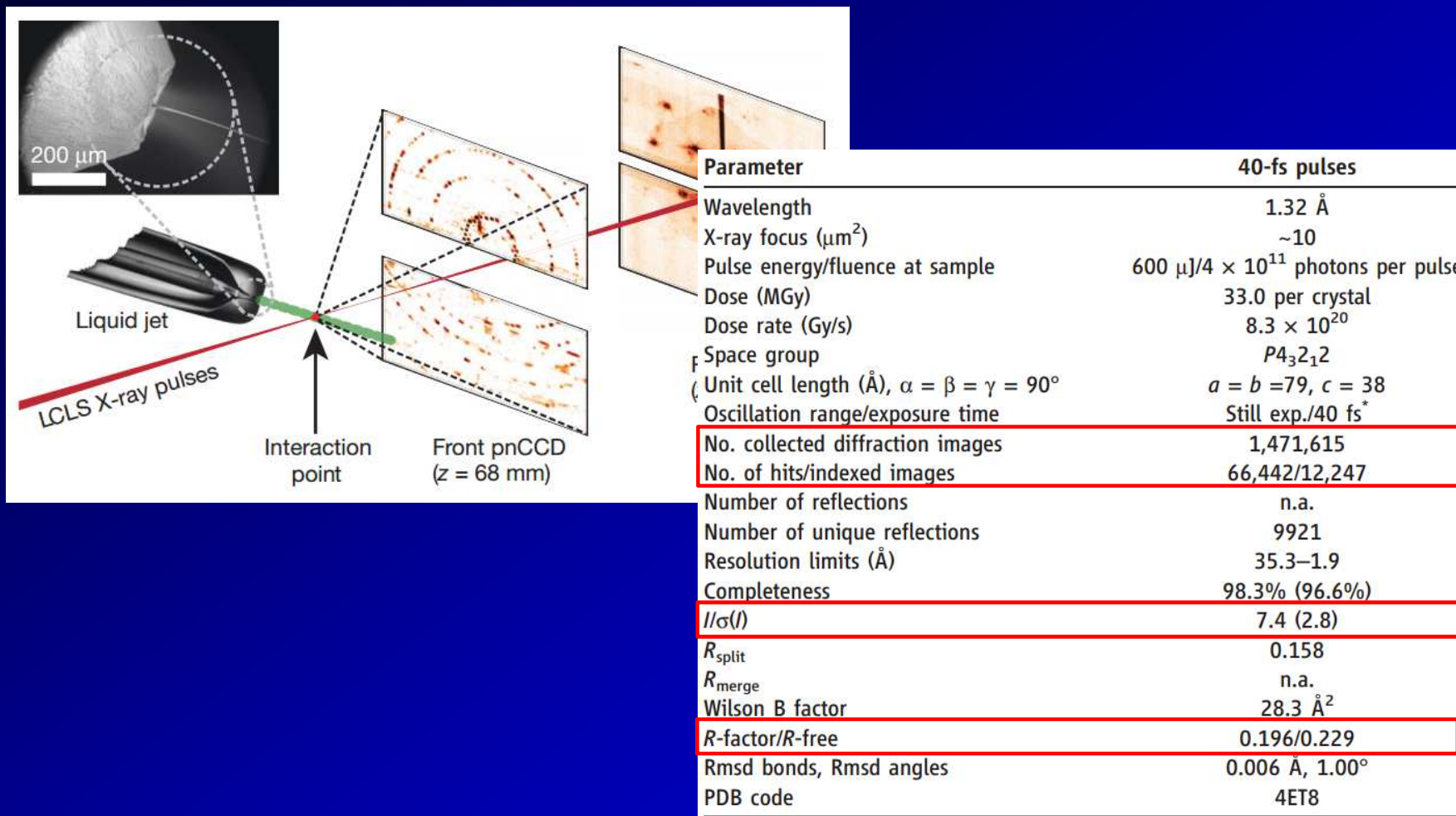


**Figure 1 | Femtosecond nanocrystallography.** Nanocrystals flow in their buffer solution in a gas-focused, 4- $\mu\text{m}$ -diameter jet at a velocity of  $10 \text{ m s}^{-1}$  perpendicular to the pulsed X-ray FEL beam that is focused on the jet. Inset, environmental scanning electron micrograph of the nozzle, flowing jet and focusing gas<sup>30</sup>. Two pairs of high-frame-rate pnCCD detectors<sup>12</sup> record low- and high-angle diffraction from single X-ray FEL pulses, at the FEL repetition rate of 30 Hz. Crystals arrive at random times and orientations in the beam, and the probability of hitting one is proportional to the crystal concentration.



# XFEL source - ex. of a « single shot » experiment

## Protein crystallography lysozyme @SFX.LCLS.eu



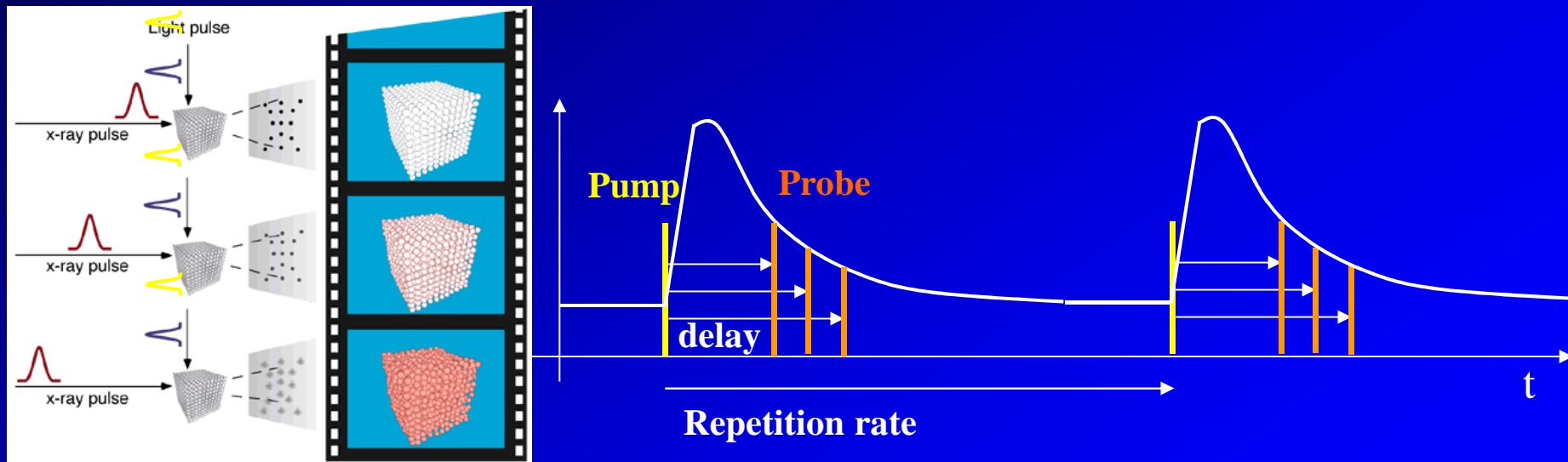


# Non ambient Conditions

## Time resolved experiments

### pump-probe method (stroboscopic method)

reversible processus time relaxation adapted with excitation frequency

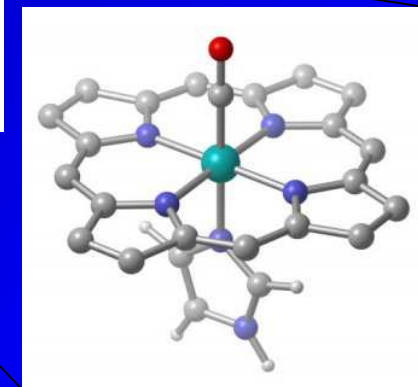
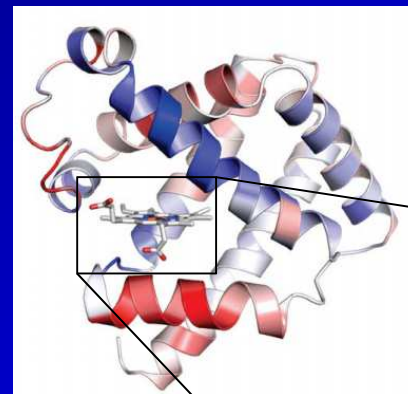
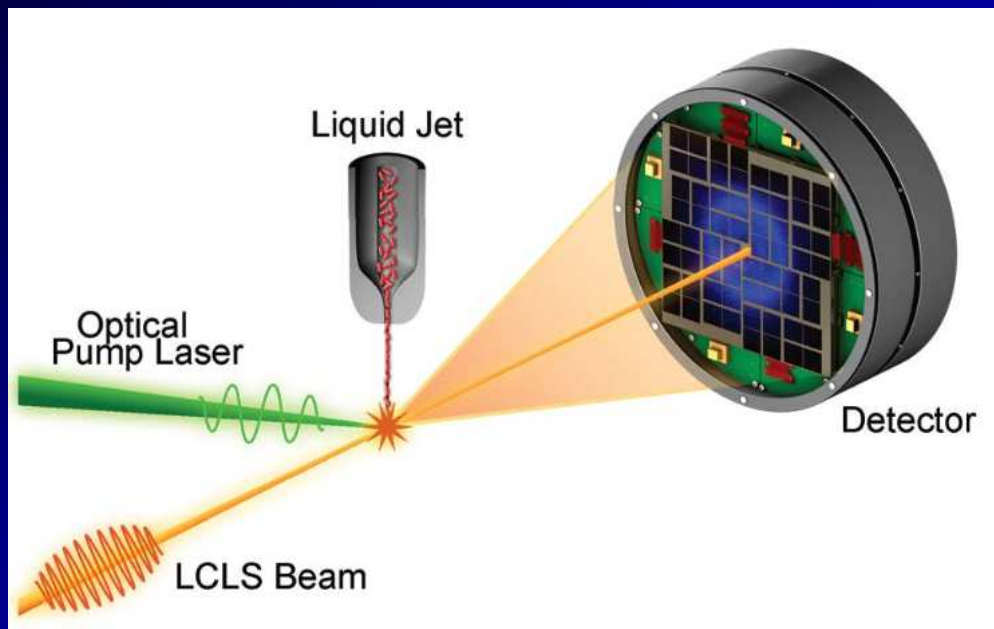


Pump : laser pulse (~ 40 fs)

Probe : X-ray pulse (ESRF 50 ps, SOLEIL 30/10 ps, 130 fs, XFEL!!)

# XFEL source - ex. of a « single shot » experiment

Time resolved Serial Protein crystallography@SFX.LCLS.eu

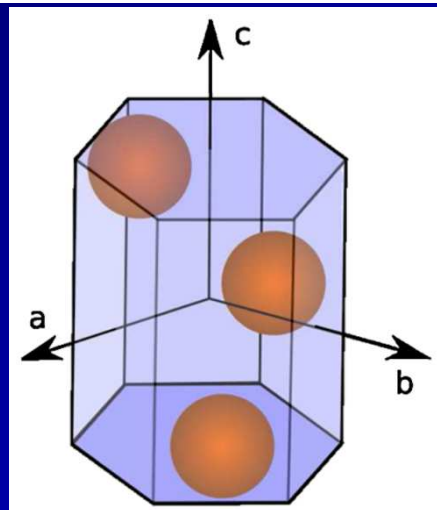
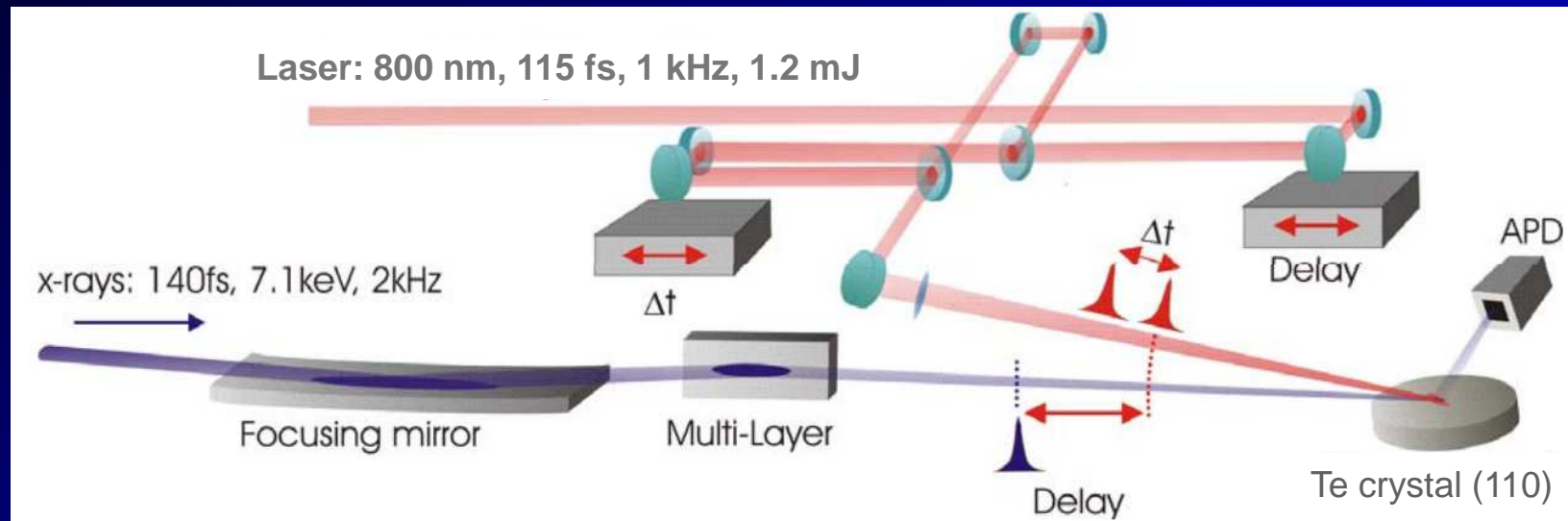


dissociation photo-induite du complexe myoglobin-CO  
photolyse de la liaison Fe-CO ~ 500 fs

# Non ambient Conditions

Time resolved experiments: example of a pumb-probe experiment

Te @ Swiss Ligth Source



$P3_1 2 1$   
 $a = 4.456 \text{ \AA}$   
 $c = 5.921 \text{ \AA}$

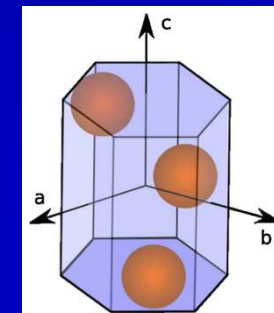
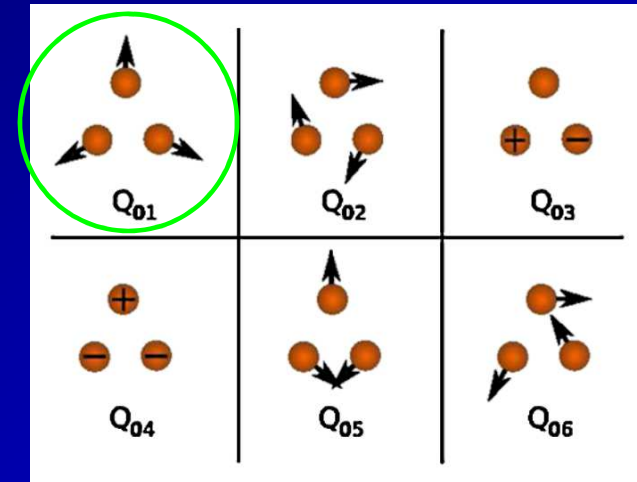
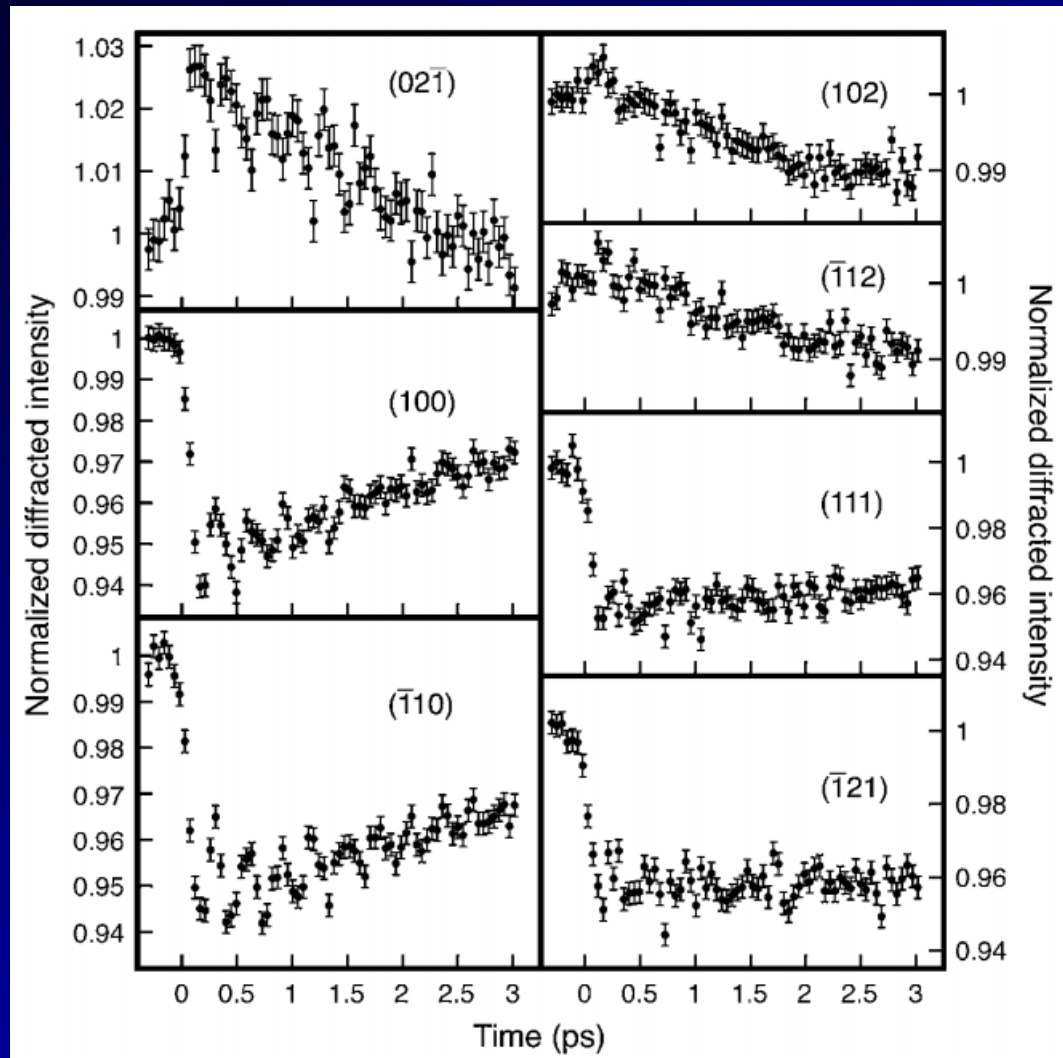
Incidences:

RX: (grazing):  $0.45^\circ$   
laser :  $9.4^\circ$

# Non ambient Conditions

Time resolved experiments: example of a pump-probe experiment

Bi @ Swiss Light Source



sudden increase of the  
helix  $\varnothing$  (0.7 pm)

# Conclusion: intérêts des Sources de RX synchrotrons

## Brillance : Intensités faibles

- Structures complexes
- Petits cristaux ( $< 20 \mu\text{m}$ )
- Densité électroniques
- Expériences résolues en temps
- Diffraction magnétique...

## Faible divergence : Précisions des mesures

- Séparation de phases
- Résolution de structures poudres (ab initio)
- Diffraction cohérente...

## Accordabilité

- Longueur d'onde optimale (cf. bio-cristallographie)
- Diffraction résonnante

## Cohérence

Polarisation « accordable » (nature et/ou direction)

Lumière pulsée Expériences résolues en temps (ps, qq 10 fs)

