Parametric x-rays in crystals: prospects for research and applications

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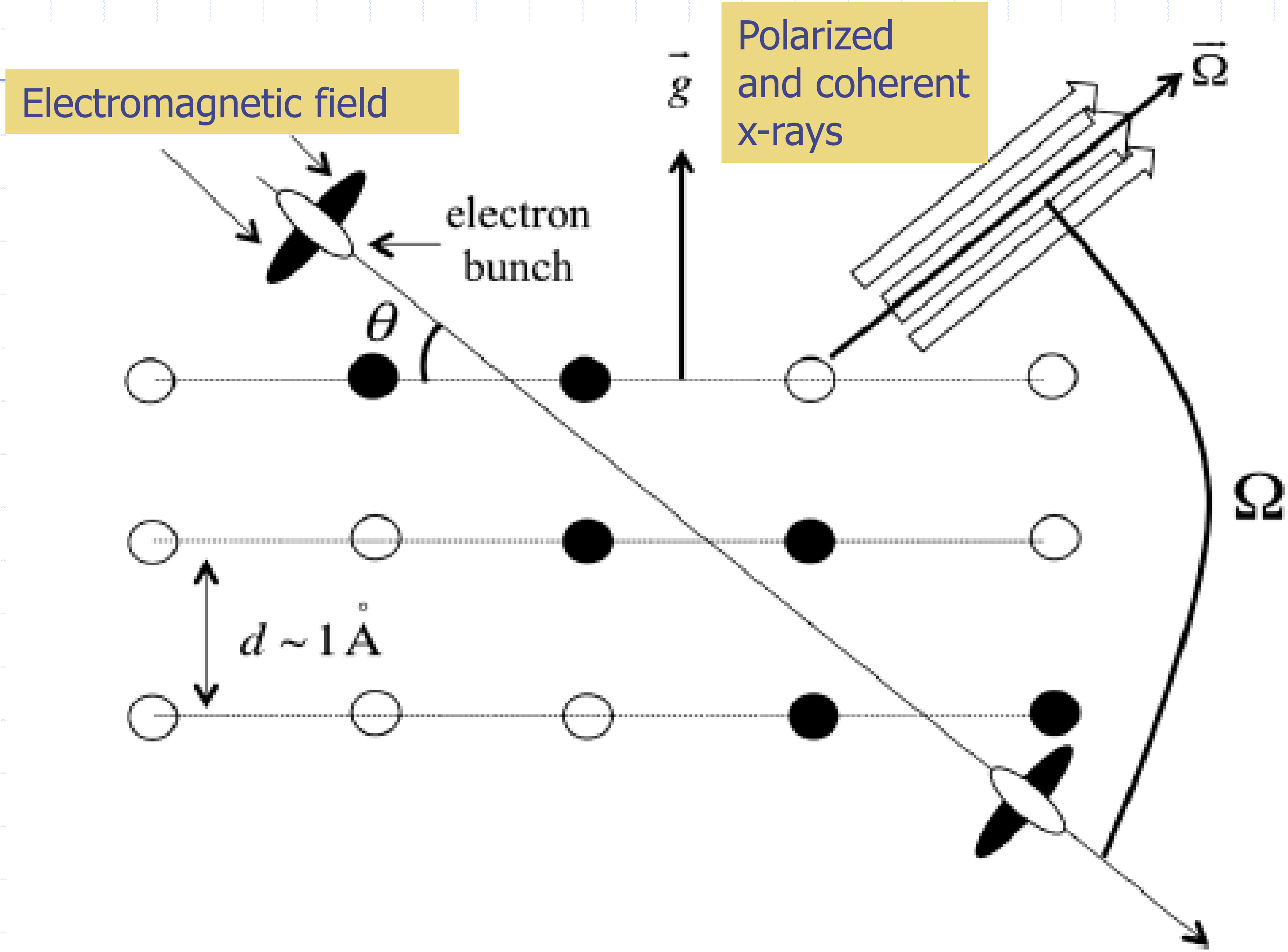
PARAMETRIC X-RAYS

- BASIC PROPERTIES
- PROSPECTS FOR BIOMEDICAL APPLICATIONS
- TOPICS FOR RESEARCH
- PRECISE SPECTROSCOPY
- CONCLUSIONS

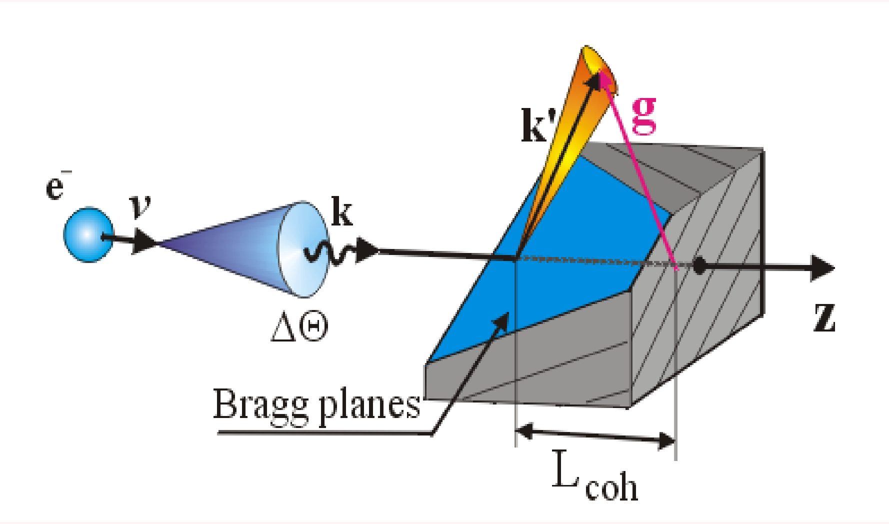
BOOK V.G. Baryshevsky, I.D. Feranchuk, A.P. Ulyanenkov, Parametric X-Ray Radiation in Crystals: Theory, Experiment and Applications, vol. 213, Springer-Verlag, Berlin Heidelberg, 2005.

REVIEW V.G. Baryshevsky Spontaneous and induced radiation by electrons/positrons in natural and photonic crystals. Volume free electron lasers (VFELs): From microwave and optical to X-ray range // Nuclear Instruments and Methods B 355 (2015) 17–23

PXR visual presentation - 1



PXR Visual Presentation - 2



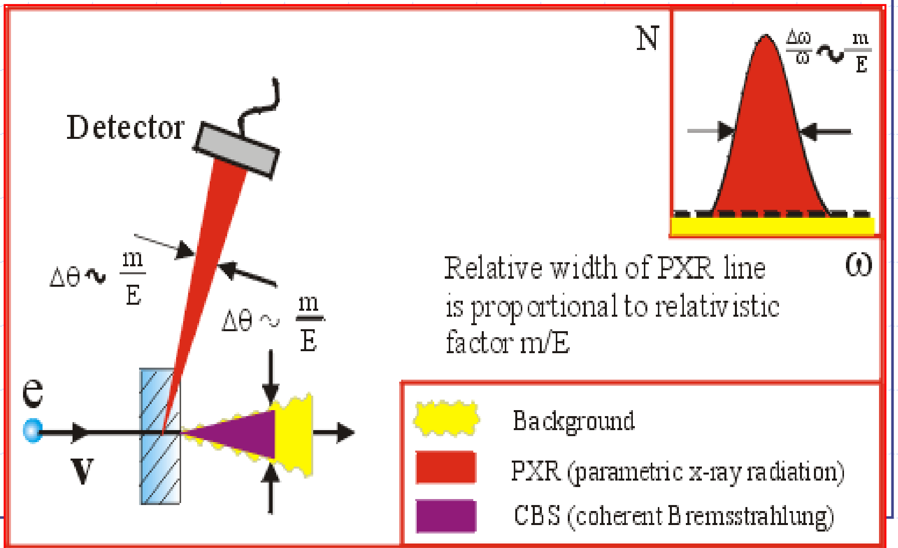
$$\vec{k} = \frac{\omega}{v^2}\vec{v} \qquad \vec{k}' = \vec{k} + \vec{g}$$

$$L_{coh} = \frac{1}{q_z} = \frac{c}{\omega(1 - v/c\cos\theta + \chi_g)}$$

$$I_{PXR} \to \max, \quad 1 - v/c\cos\theta + \chi_g \to 0, \quad L_{coh} \to \max$$

$$E_{opt} \approx \frac{mc^2}{\sqrt{|\chi_g|}} \approx 50\,MeV$$

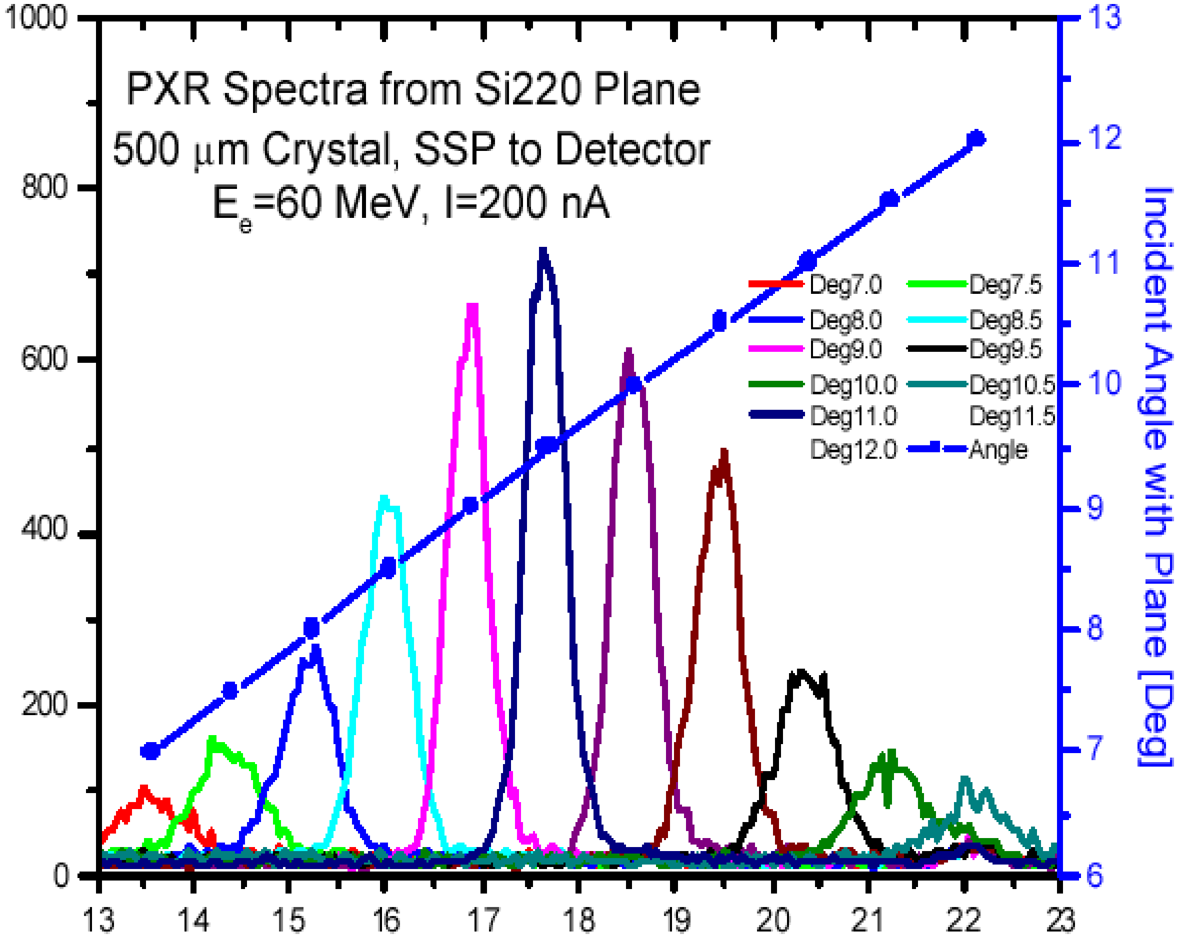
Relativistic electrons: the background radiation caused by effects other than PXR, e.g., coherent and incoherent *Bremsstrahlung*, is concentrated within the narrow cone along the direction of particle beam



Motivation for application – PXR properties

- PXR are quasi-monochromatic x-rays;
- PXR energy can be changed continuously by crystal rotation;
- PXR are directed, and polarized, and partially coherent x-rays;
- PXR energy does not depend on energy of incident charged particles but only on their Lorenz factor;
- PXR radiation angle can be as large as 180 degrees;
- PXR has no theoretical limits for charged particle energy, i.e. this radiation mechanism exists at any, low or high as possible, beam energy.

PXR frequency tunability



About 40% tunability around 17.7 keV Bragg condition

B. Sones et al.
ANS Transactions, v. 88 (2003), RPI Linac, USA

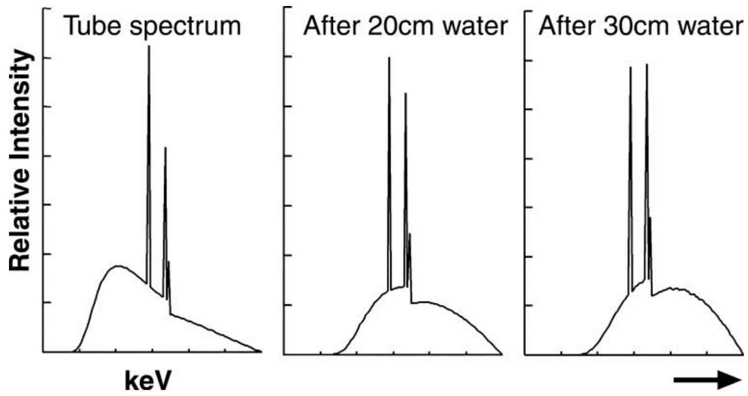
Rensselaer Polytechnical Institute

Comparison of some x-ray generation processes at accelerators

Type of radiation	Yield, photon/e	E, MeV
BR	1.2*(-8)	500
CBR	1.7*(-7)	0.2
SR	1.2*(-5)	**3 *(+3)**
TR	1.0*(-9)	125
RR	1.6*(-7)	50
PXR	1.3*(-5)	**50**

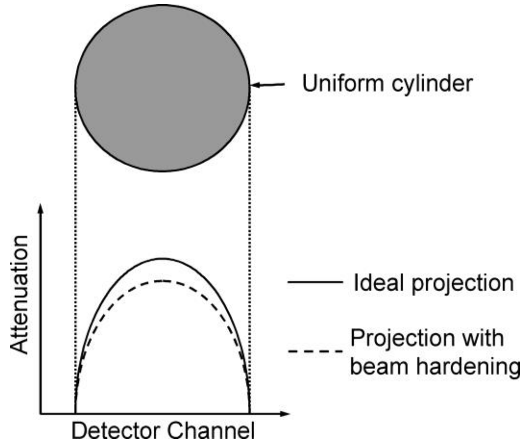
V.G. Baryshevsky, I.D. Feranchuk // NIM 228 (1985) 490

Ideal case: Monochromatic source tunable for the particular imaging situation



Requirements:

- - high contrast
- - low patient dose
- - maximum SNR
- - no beam hardening artefacts



Variants:

- → use a synchrotron X-ray source (100 m)
- → improving of X-ray tubes (10 cm) based technology
- → search for alternatives (table-top): parametric X-rays, channeling X-rays, etc.....

Biomedical Applications

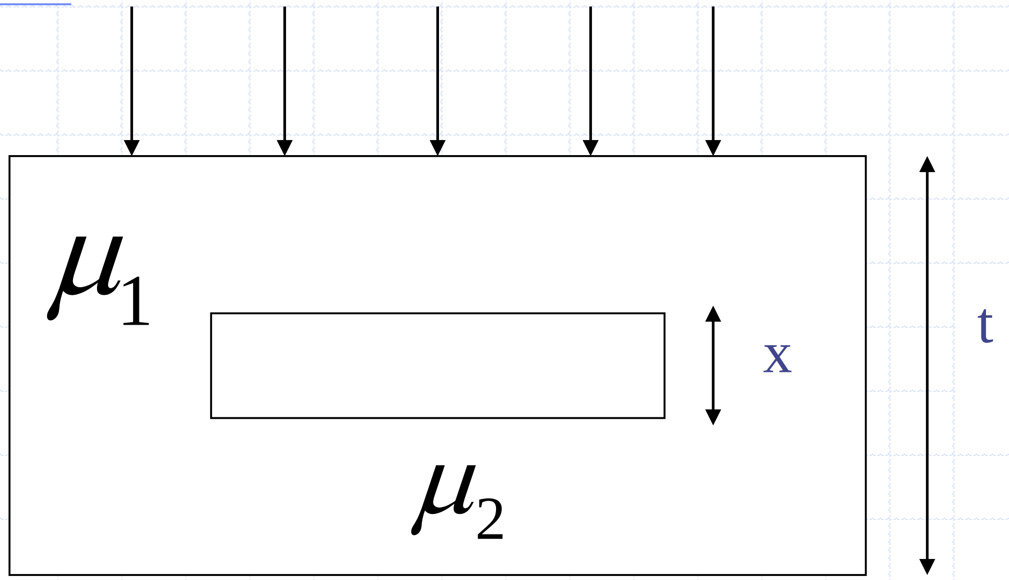
- An x-ray source with a narrow frequency range, ideally at a single frequency, would be advantageous because it would allow x-rays only at the desired frequency to be produced.
- By eliminating x-ray frequencies outside of the range required for a specific application, the dose may be significantly reduced. It has been estimated that **mammographic** examinations performed with near mono-energetic x-rays will deliver a dose to the patient that will be from **one-tenth** to **one-fiftieth** of the dose delivered by a conventional x-ray system. This estimate is similarly applicable to **angiography** and other radiography studies.

Optimal X-Ray Energies for Medical Imaging

- mammography, soft tissues - 15-25 keV;
- radiography of chest, extremities and head - 40-50 keV;
- abdomen and pelvis radiography - 50-70 keV;
- digital angiography - ~33 keV.

X-Ray Flow Evaluation

The Physics of Medical Imaging / S. Webb (ed), Bristol: Hilger, 1978.



$$N = k^2(1+R)\exp(\mu_1 t)/(\varepsilon(\Delta\mu x)^2 x^2)$$

Number of x-ray quanta needful to visualize 1.0 mm^3 of biological tissue at 1% contrast is 2.6x10^{13} quanta/m^2.

What do we need for *in vivo* quality X-RAY imaging?

Evaluating radiological examination in general context, number of x-ray quanta needed to visualize 1.0 mm^3 of biological tissue at 1% contrast is **~10^6 photons/mm^2** in the case of *digital* detection

What do we exactly need for *in vivo* quality X-RAY imaging?

Due to heart beat and breathing, above mentioned photon flux must be provided within ~1 second.

Photons must penetrate considerable field of vision,

say 10x10 cm.

Thus, we need, *at least*

$\sim 10^6\ mm^{-2} * 100x100\ mm^2 / 1.0\ s =$

$\sim 10^{10}$ photons/s

with tunable x-ray energy in 10-70 keV range.

Mono-chromaticity could be of $\sim 10^{-2}$ and less for a patient's dose reduction.

Radiation background should be low.

All these conditions together can be met today only at SRS

Number of the PXR photons regarding to parameters of a realistic electron accelerator

1 A = 6*10^18 electrons/s

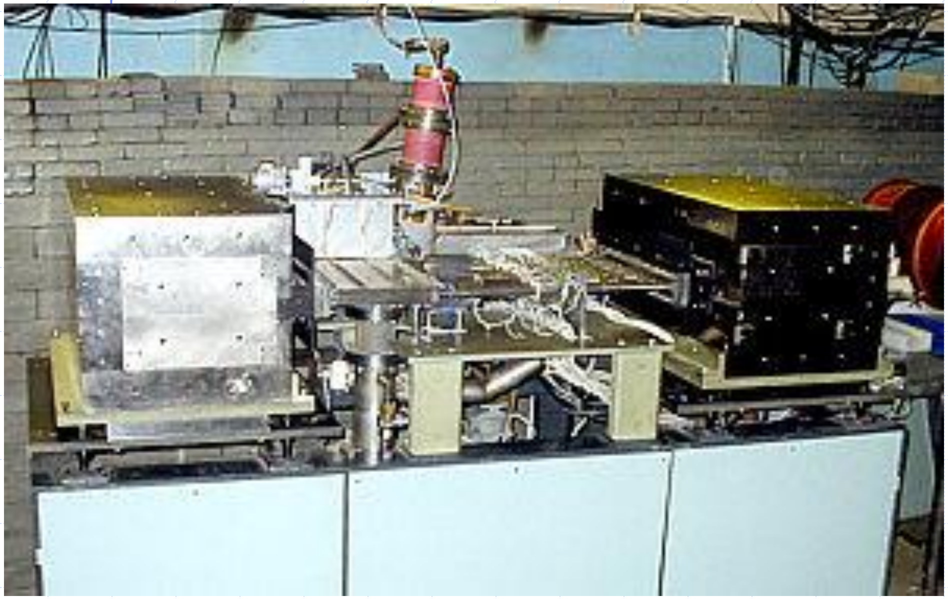
PXR photons/s = 10^-5 per electron/s

Thus,

10^10 PXR photons can be generated by ~some mA beam

Electron Beam Laboratory SINP, Moscow, Russia

Parameters of a pulsed racetrack microtron



Injection energy: 48 keV
Energy gain: 4.8 MeV / orbit
Orbits: 14
Output energy: 14.8 - 68.3 MeV
Output current at 68.3 MeV: 10 mA
Orbit circumference increase: 1l/orbit
Operating frequency: 2,856 MHz
Klystron power pulsed: 6 MW
End magnet field induction: 0.963 T
RTM dimensions: 2.2x1.8x0.9 m3

http://vserv.sinp.msu.ru/ebl/

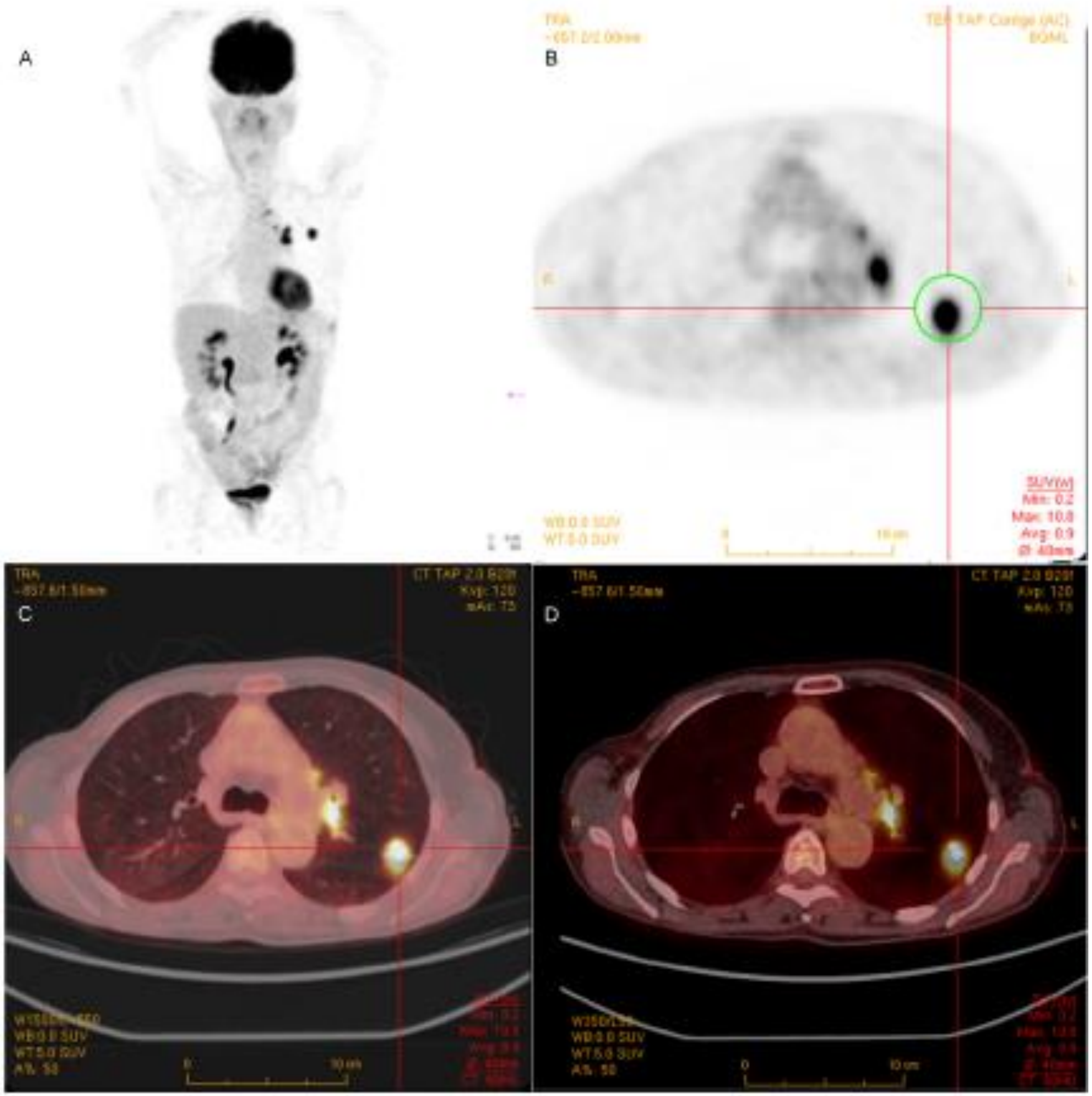
Limitations of medical x-ray imaging

The quality of the x-ray images is ultimately limited by **two** opposing factors, one is that increased image spatial and contrast resolution requires an increased number of detected x-ray photons per image resolution unit, and the second is the direct dependence between the increased dose deposited by x-rays and the increased risk of tissue damage and further cancer development.

Some ways of optimization:

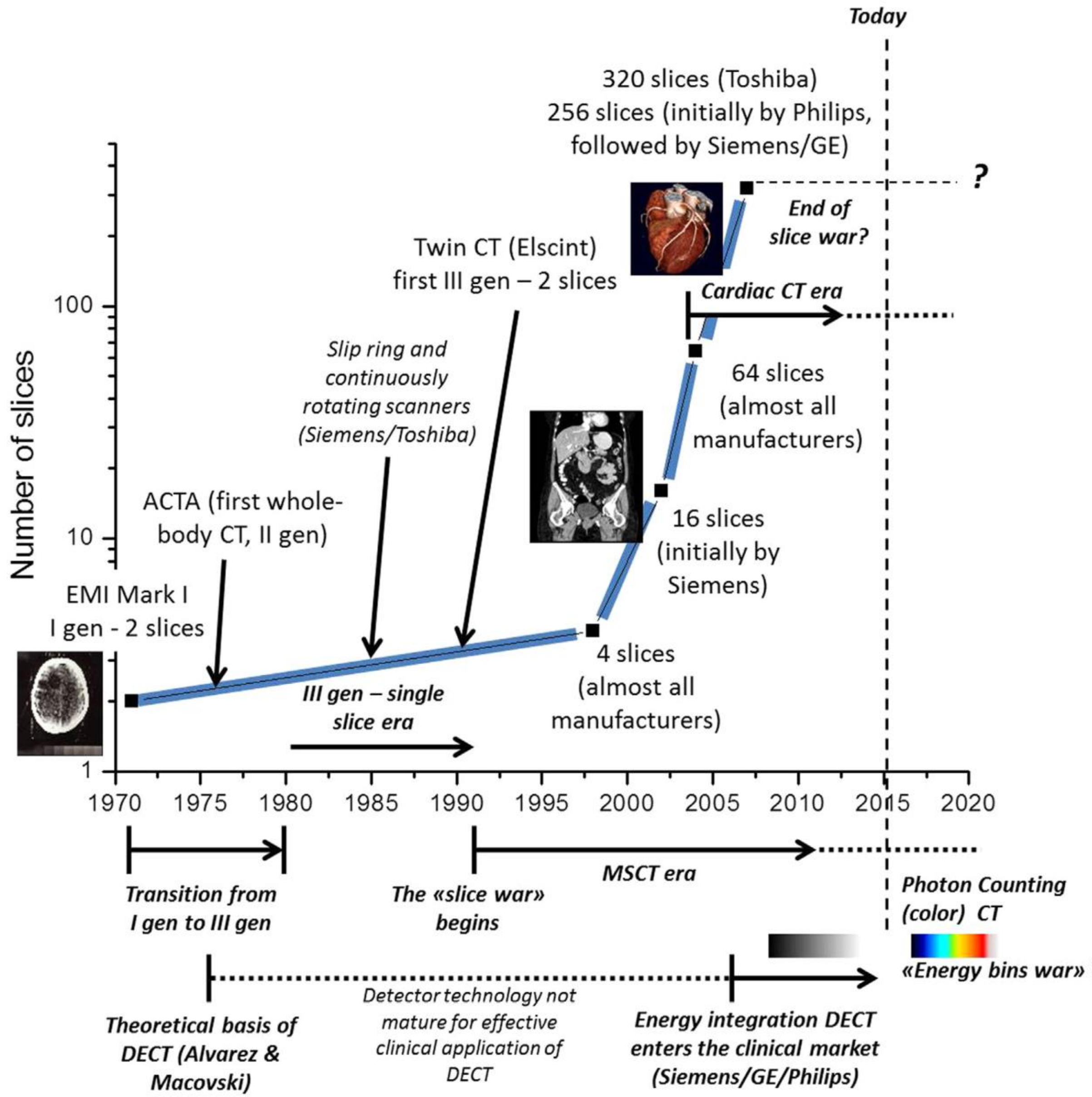
- Monochromatic x-rays instead of *Bremsstrahlung*
- Utilization of x-ray refractive index – phase sensitive imaging

Multi-modality imaging



PET/CT images of glucose metabolism.
(A) Maximum intensity projection image representing the whole-body bio-distribution of FDG. (B) shows an axial PET slice positioned on a lung lesion highlighted by the circled region. (C,D) show two fusion images obtained with two different HU windows.

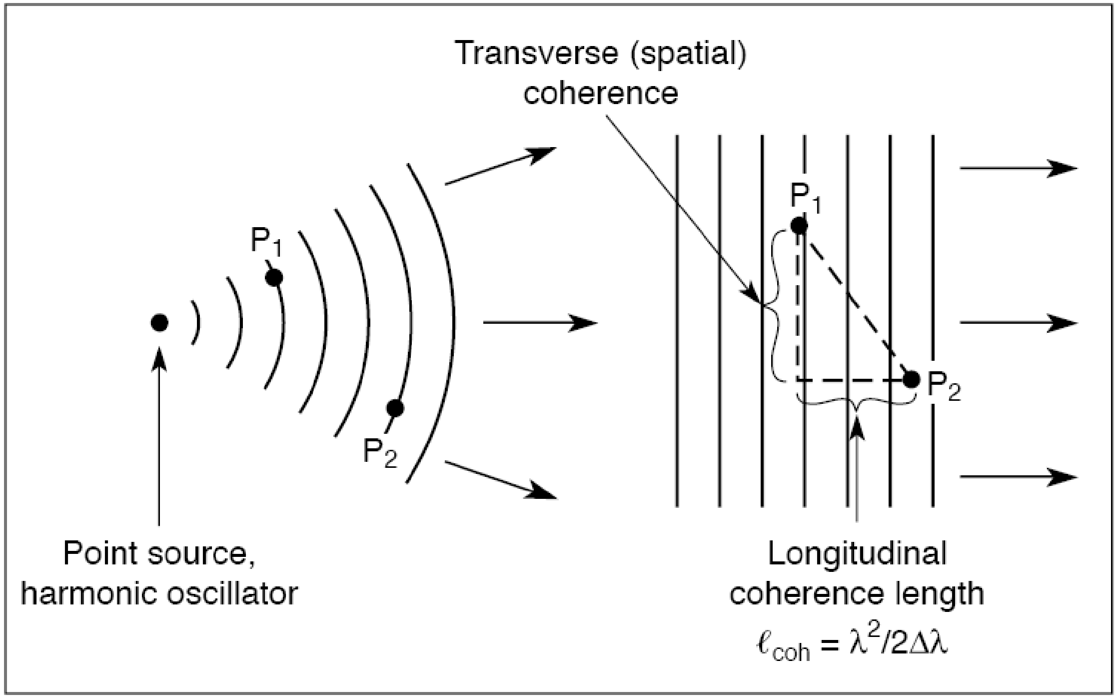
The market has reached a plateau in terms of number of CT slices due to patient dose, image quality and cost considerations



D. Panetta Advances in X-ray detectors for clinical and preclinical Computed Tomography // NIM A809 (2016) 2–12

As the slice war has come to an end by some years, manufacturers seems now following a different direction for next generation of CT scanners for which the number of energy bin is to be increased instead of the number of slices. Extending the capability of CT scanners beyond the physical limitation posed by energy integrating detectors (EID) offers several advantages. Bremsstrahlung spectra generated by Xray tubes are such that all images obtained by energy integrating detectors are unavoidably affected by beam hardening artifacts (besides several more or less fancy software techniques for artifact reduction); hence, objects with same composition and density can have different reconstructed CT numbers (expressed in Hounsfield Units, or HU) depending on the depth and composition of surrounding materials. This limitation prevents material differentiation and quantitative imaging, unless (i) the incident Xray spectrum is narrowed enough to be considered quasimonochromatic, or (ii) the attenuation data is selectively recorded in different data bins as a function of the photon energy.

Phase sensitive imaging requires coherent x-rays



$$\mu_{12} = \frac{\langle E_1(t)E_2^*(t)\rangle}{\sqrt{\langle|E_1|^2\rangle}\sqrt{\langle|E_2|^2\rangle}} \qquad (8.12)$$

A high degree of coherence ($\mu \rightarrow 1$) implies an ability to form a high contrast interference (fringe) pattern. A low degree of coherence ($\mu \rightarrow 0$) implies an absence of interference, except with great care.

In general, radiation is partially coherent

Longitudinal (temporal) coherence length

$$\ell_{coh} = \frac{\lambda^2}{2\,\Delta\lambda} \qquad (8.3)$$

Full spatial (transverse) coherence

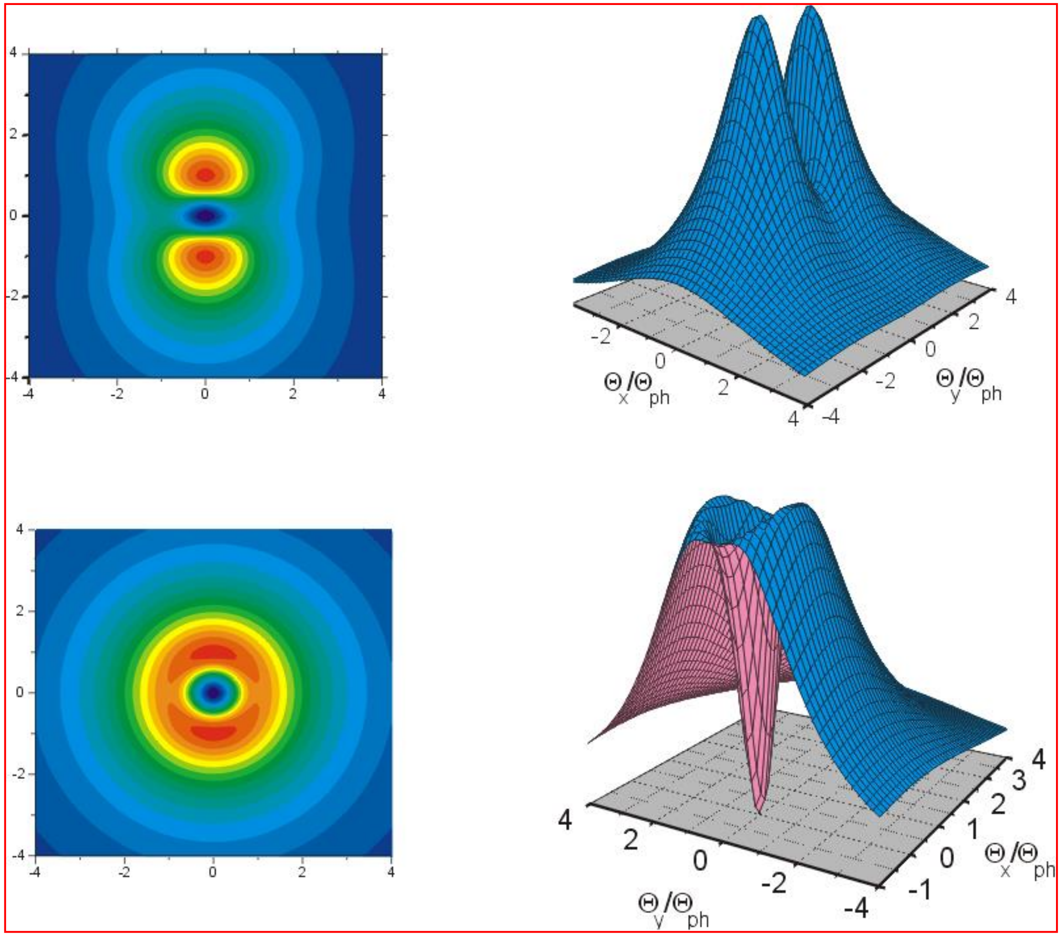
$$d \cdot \theta = \lambda/2\pi \qquad (8.5)$$

$$\bar{P}_{coh,\lambda/\Delta\lambda} = \underbrace{\eta}_{\text{beamline efficiency}} \underbrace{\frac{(\lambda/2\pi)^2}{(d_x\theta_x)(d_y,\theta_y)}}_{\text{spatial filtering}} \cdot \underbrace{N\frac{\Delta\lambda}{\lambda}}_{\text{spectral filtering}} \cdot \bar{P}_{cen}$$

D. Attwood http://www.coe.berkeley.edu/AST/srms

Coherence of PXR

PXR may provide combination of monochromatic absorption imaging and phase contrast imaging. Parametric x-rays are coherent to a considerable extent. That is its ***intrinsic*** property.



PXR angular distributions at Bragg angles of 45 and 9 arc deg.

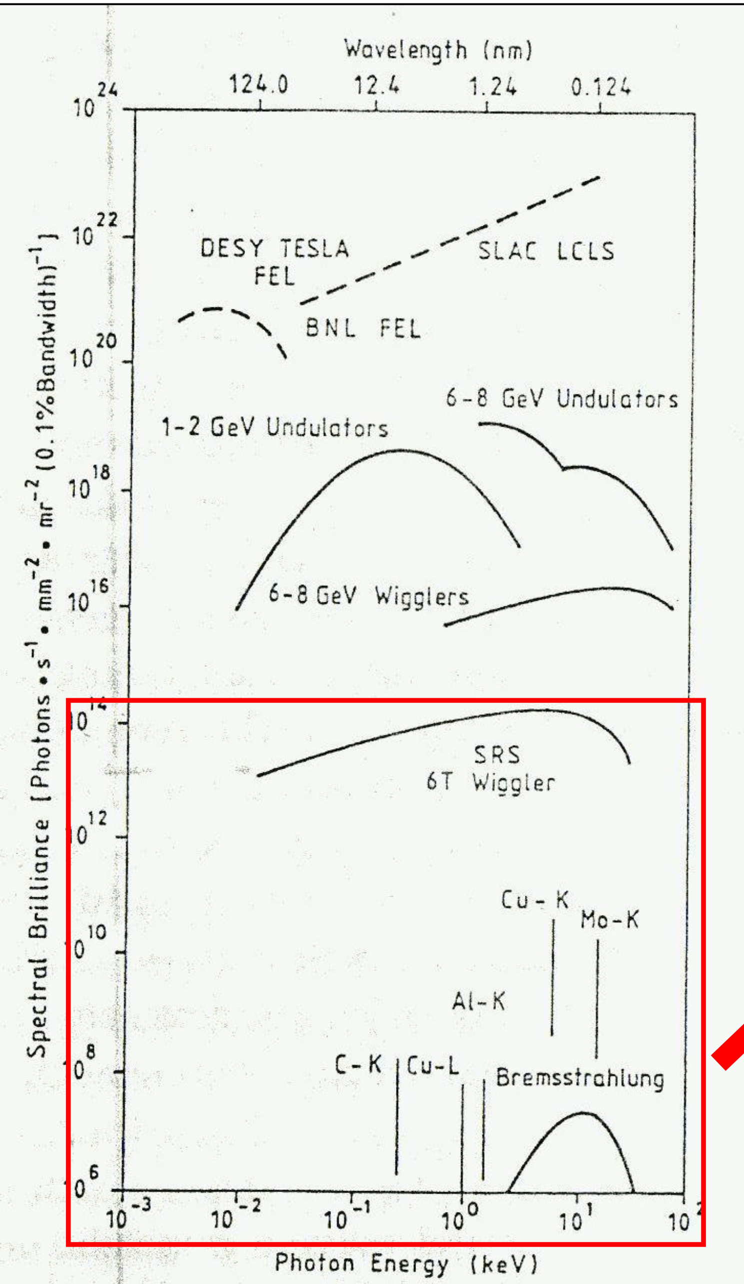
Shape of PXR angular distribution strongly depends on the Bragg angle and on the quantity of diffracted transition radiation and *Bremsstrahlung*.

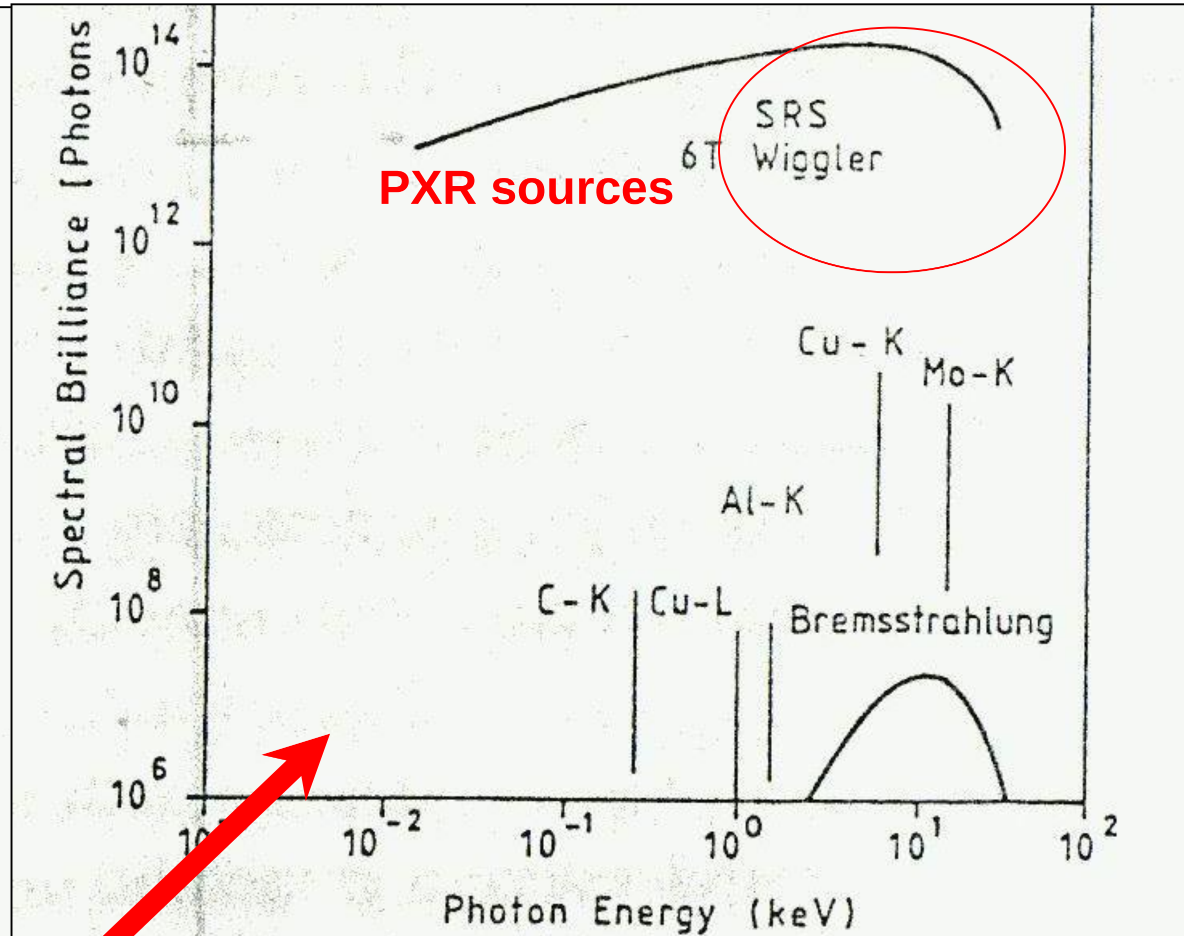
Proportion between coherent and partly coherent components depends on generation geometry, i.e. Bragg, or Laue, or extremely asymmetric diffraction.

When crystal target is rotating to tune PXR frequency, proportion between coherent (*pure* PXR) and partly coherent components (diffracted radiations), as well as shape of the “source” changes considerably.

Thus, PXR spatial (and temporal) coherence is the value, highly variable with crystal target rotation.

Spectral brilliance of x-ray sources





PXR spectral brilliance comparable with that of bending magnet, but its flux is coherent on a great extent

Prospects

- PXR is the *prospective* source of coherent hard x-rays obtainable at a relatively low energy electron accelerator
- Application of PXR for bio-med imaging can provide *combination* of monochromatic and phase sensitive images, thus reducing dose while improving contrast (First PXR-DEI image was taken at LEBRA-PXR source)
- Coherence properties of a *real* PXR beams are the matter of further research, both theoretically and experimentally
- It is *important* to develop techniques of PXR coherence metrology and/or in-line monitoring

Spectroscopy

Mössbauer spectroscopy is a powerful and well established method employed for conducting research in various fields such as physical, chemical, biological, earth, and fundamental physical sciences.

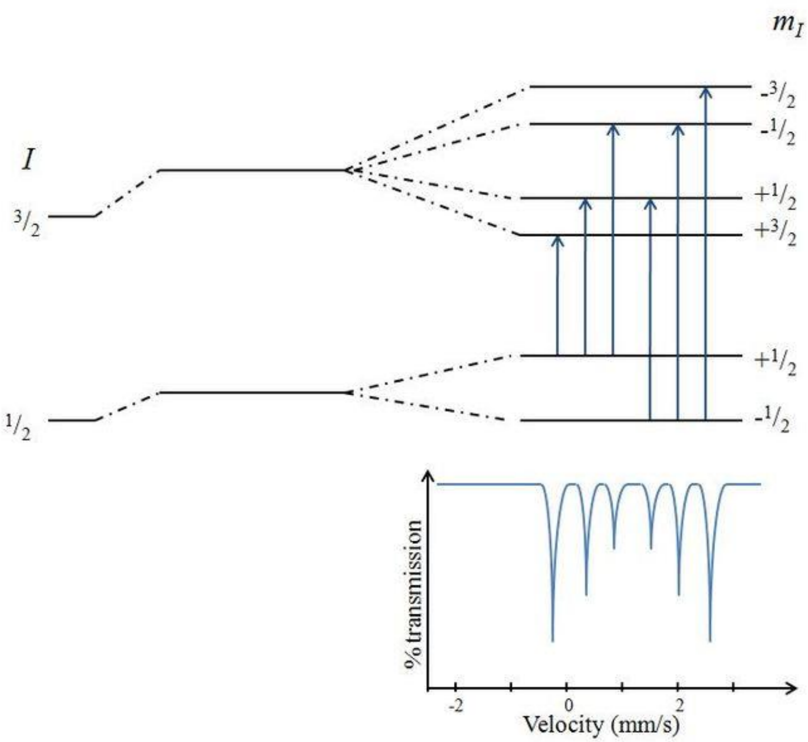
Mössbauer spectroscopy provides element-specific information on surrounding electronic states and magnetism, which is required in modern materials science and in complex systems such as biological substances.

Mössbauer spectroscopy probes tiny changes in the energy levels of an atomic nucleus in response to its environment. Typically, three types of nuclear interaction may be observed: an isomer (chemical) shift; quadrupole splitting; and magnetic or hyperfine splitting (Zeeman effect).

Due to the high energy and extremely narrow line widths of gamma rays, Mössbauer spectroscopy is one of the most sensitive techniques in terms of energy (and hence frequency) resolution, capable of detecting change in just a **few parts per 10^{11}**.

Mössbauer effect

In 1957, Rudolf Mössbauer discovered that, in some circumstances, if the nucleus is bound in a crystal lattice, the whole crystal recoils rather than the individual nucleus. Due to the much greater mass involved in recoil the energy of the emitted ray is very close to that of the difference in energy between the nuclear energy levels, and resonant absorption is possible.



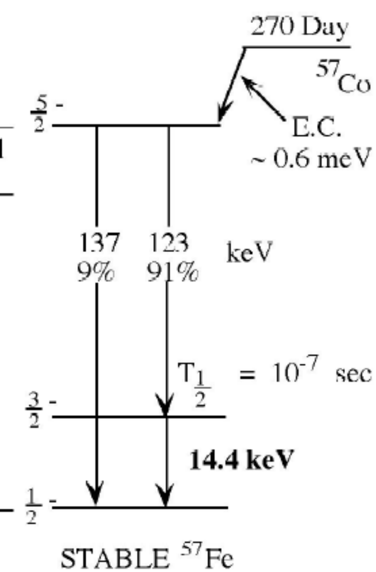
Magnetic splitting of the nuclear energy levels and the corresponding Mössbauer spectrum

Mössbauer spectroscopy is limited by the need for a suitable gamma-ray source. And samples must be solid (frozen)

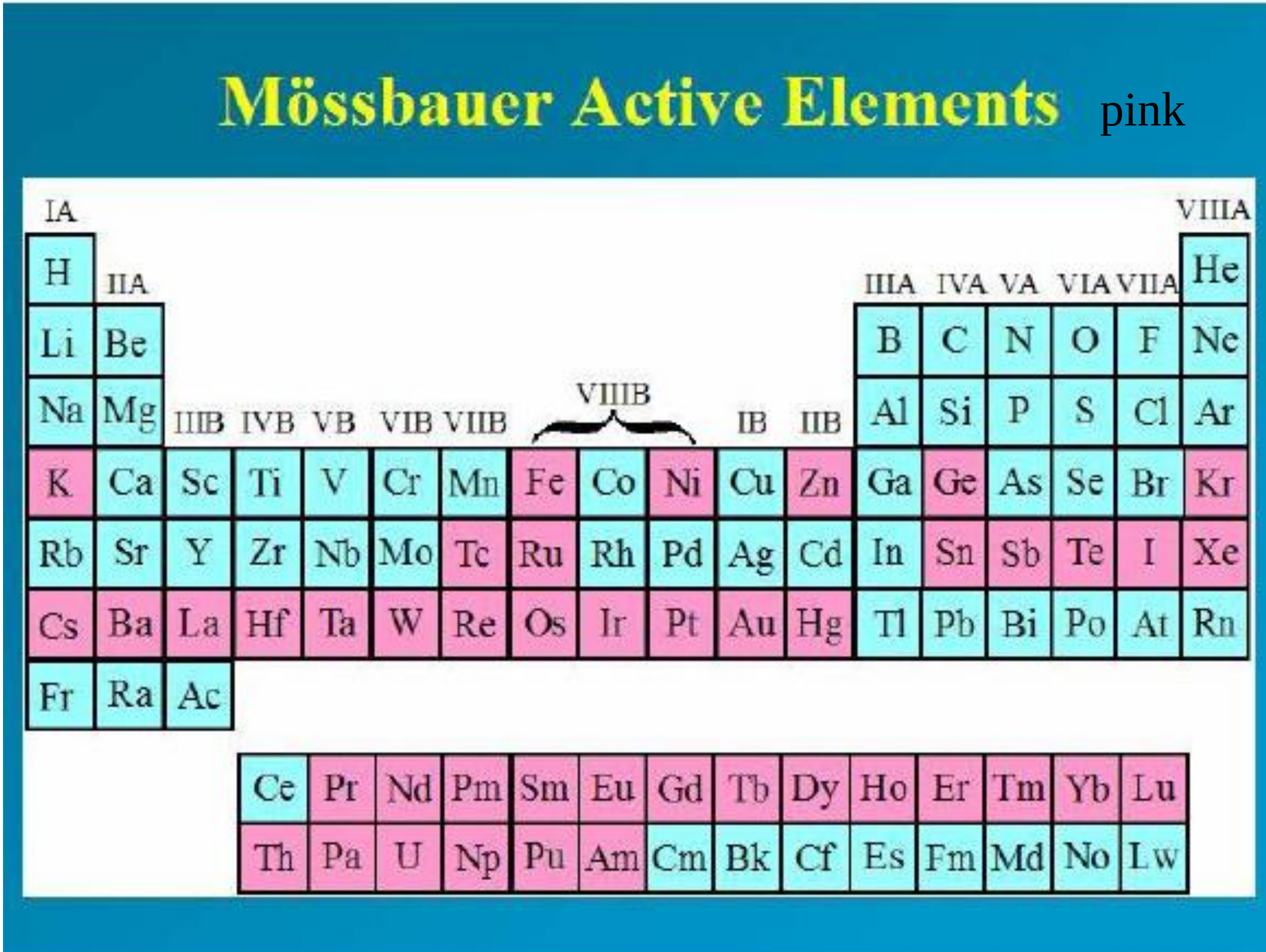
PROPERTIES OF ^{57}Fe α

	Ground state	First excited state
Energy (keV)	0	14.36
Spin and parity	$\frac{1}{2}$ -	$\frac{3}{2}$ -
Magnetic moment (nm)	0.0903	-0.153
Quadrupole moment (barns)	0	0.29
Mean life (sec)	Stable	1.4×10^{-7}

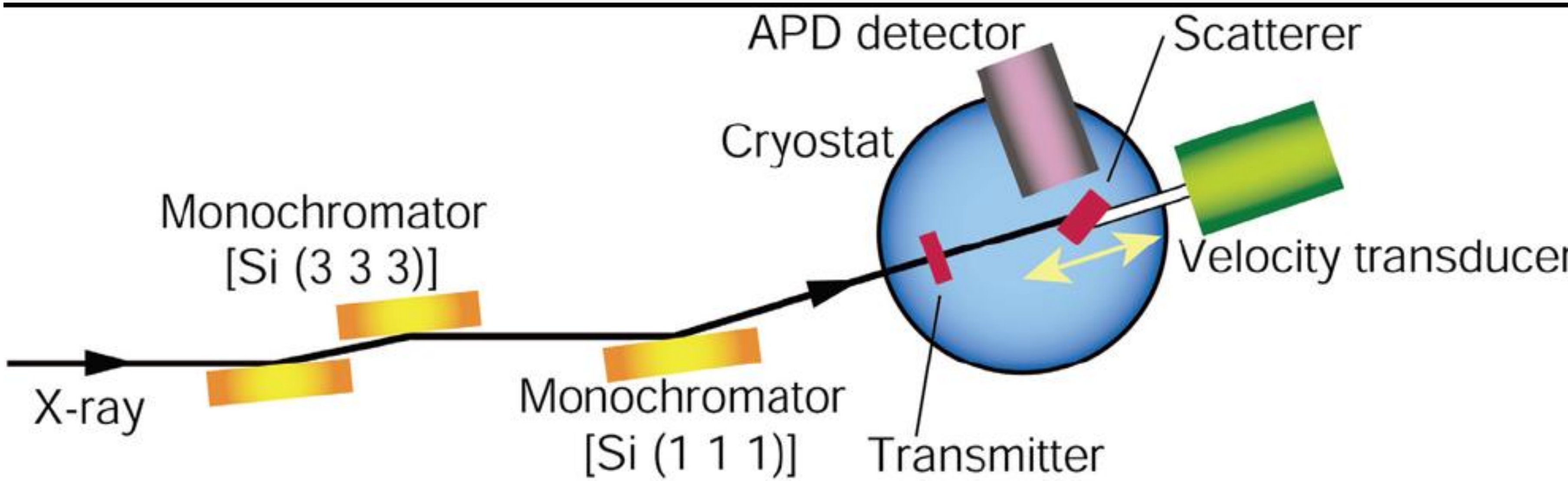
α Internal conversion coefficient: 9.7 ± 0.2.



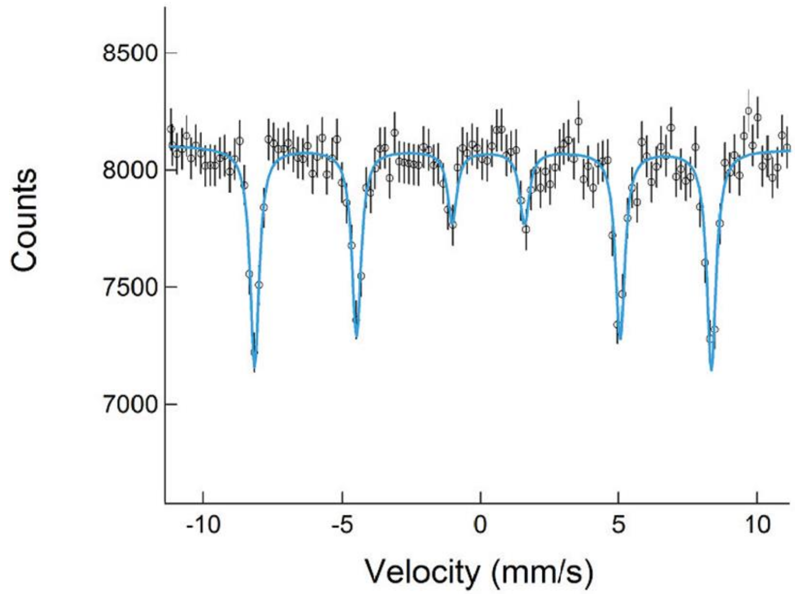
Mössbauer effect



Synchrotron-Radiation-Based Mossbauer Spectroscopy



The applicability
of SR-based Mossbauer spectroscopy
increasing.



M. Seto at al. PRL 102, 217602 (2009)

Synchrotron-Radiation-Based Mossbauer Spectroscopy

TABLE I. Comparison between SR-based Mössbauer spectroscopy and Mössbauer spectroscopy using radioactive sources.

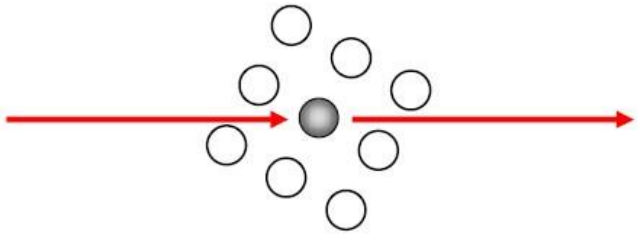
	Synchrotron radiation [proposed energy domain method (E) and time domain NFS method (T)]	Radioactive sources
Surface, interfaces, multilayers	Clean, unclean, and nanostructured samples (E); limited to clean surfaces (T)	Clean, unclean, and nanostructured samples
Multiple extreme conditions [a], imaging	Relatively easy, sub-μm resolution is possible (E,T)	Usually not easy, limited to μm resolution
Applicable isotopes	Short- and extremely long- lived and high-energy isomers are possible (E); efficient for low-energy and sufficiently long-lived isomers (T)	Isotopes for which appropriate sources are available

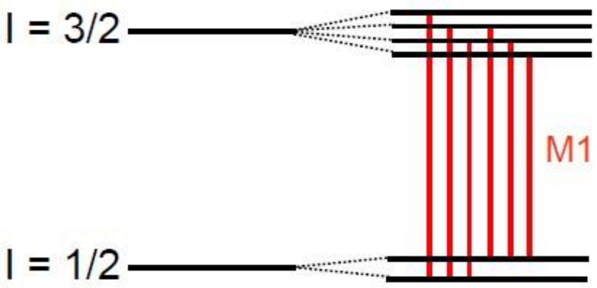
[a]High pressure (~200 GPa), applied magnetic field (~10 T), high (~1000 K) or low (~ mK) temperature.

The spectral density of PXR in the peaks exceeds the analogous value for the SR to one electron. It is also important that the PXR frequencies are defined solely by the crystal parameters and do not depend on the electron energy. It allows generate hard X-rays from electrons with essentially less energy than in the case of SR.

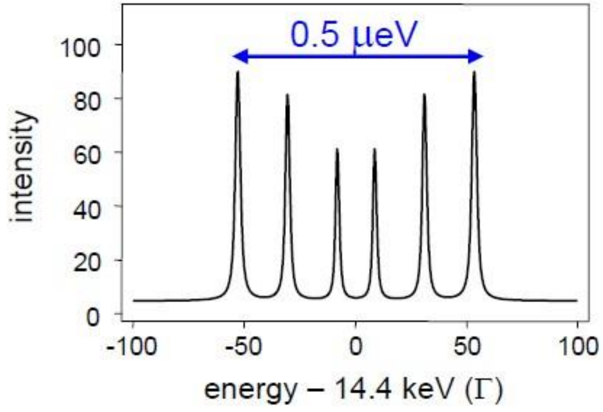
Mossbauer spectra in energy and time domains

Nucleus embedded in lattice:

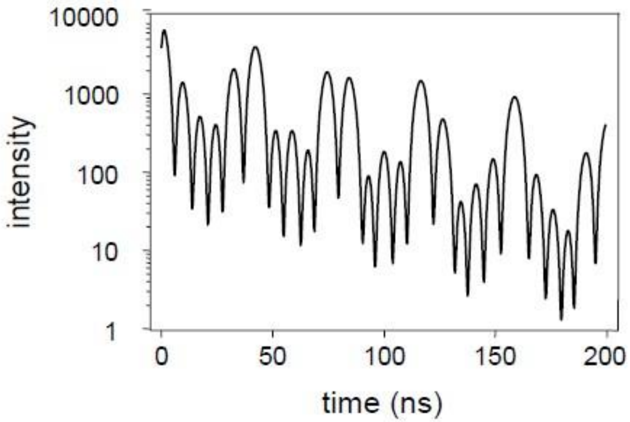




Energy domain :



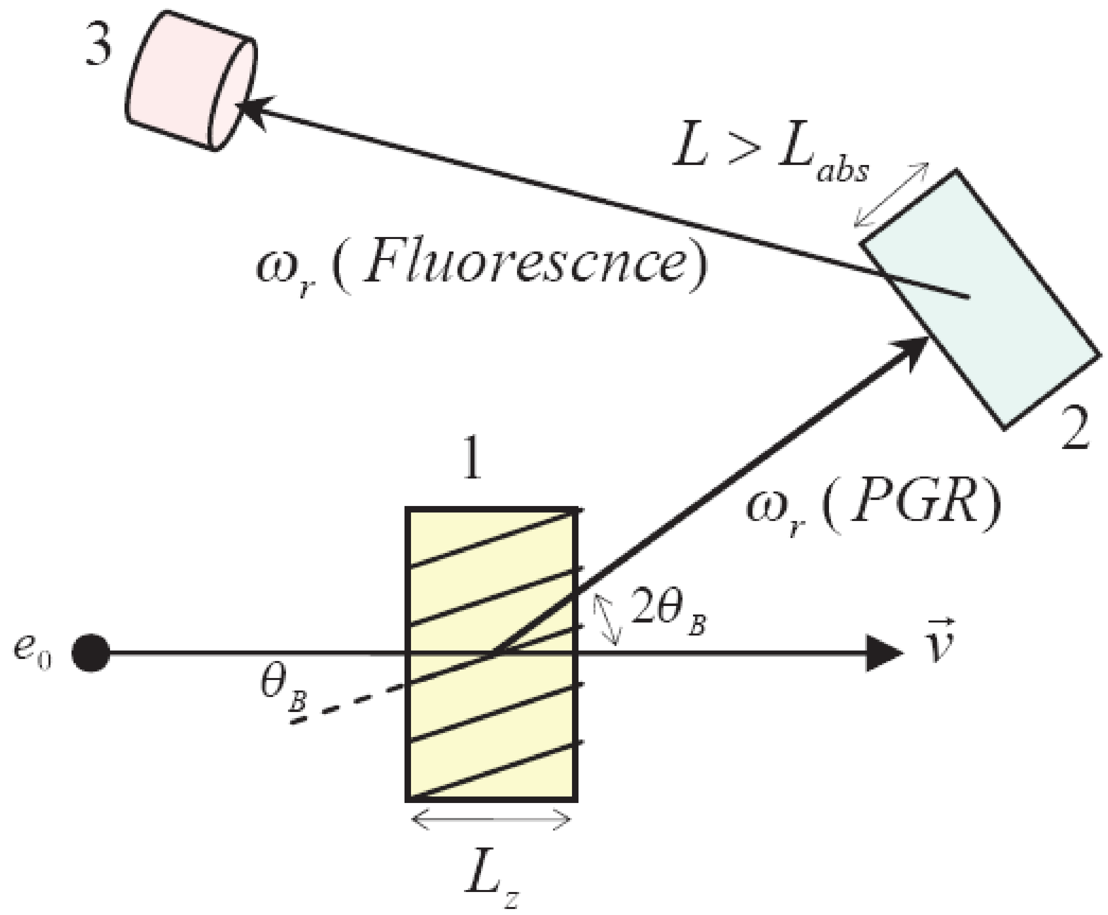
Time domain :



quantum beats

due to hyperfine splitting of nuclear states

EXPERIMENTAL SETUP FOR PGR OBSERVATION



Sketch of the experimental setup for the observation of PGR:

1) Mossbauer crystal-radiator; 2) Mossbauer absorber;

3) detector-counter of the fluorescence photons from the absorber

PROSPECTS

Mossbauer spectroscopy can probe the hyperfine fields, measuring local properties of materials (structural and magnetic)

Hyperfine interactions: interactions between a nucleus and its environment (isomer shift, quadrupole splitting, magnetic splitting)

Conversion electron Mossbauer spectroscopy allows to study magnetic properties of mono-layer thick nanostructures

Accelerator-based Mossbauer spectroscopy can provide spectra in time domain to addition of energy domain (standard MS)

Accelerator-based Mossbauer spectroscopy can provide polarized photons necessary for magnetic measurements

Accelerator-based Mossbauer spectroscopy can deal with non-conventional Mossbauer isotopes

DRO is the coherent process of x-ray photon emission by the relativistic oscillator (e.g. electron channeling in a crystal) and its diffraction within the single crystal target. It sometimes called diffracted channeling radiation – **DCO**.

DRO formation was firstly considered in [**Baryshevsky V.G., Dubovskaya I.Ya. // Doklady Akad. Nauk SSSR, 231 (1976) 1336, (in Russian)**] and detailed review and references may be found in [**Baryshevsky V.G., Dubovskaya I.Ya. Diffraction phenomena in spontaneous and stimulated radiation by relativistic particles in crystals, Lawrence Berkeley Laboratory, LBL-31695 (1991) 122 p.**].

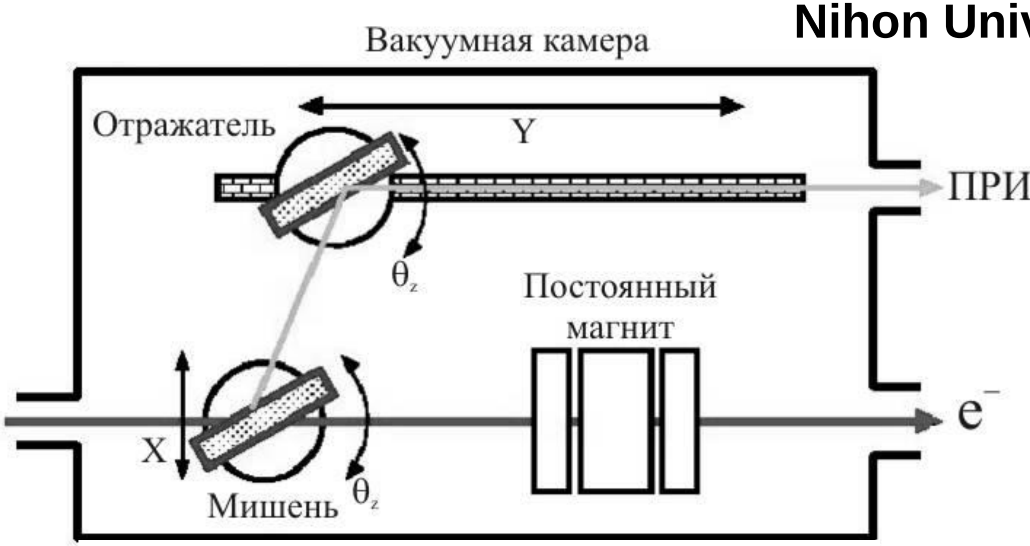
Diffracted channeling radiation

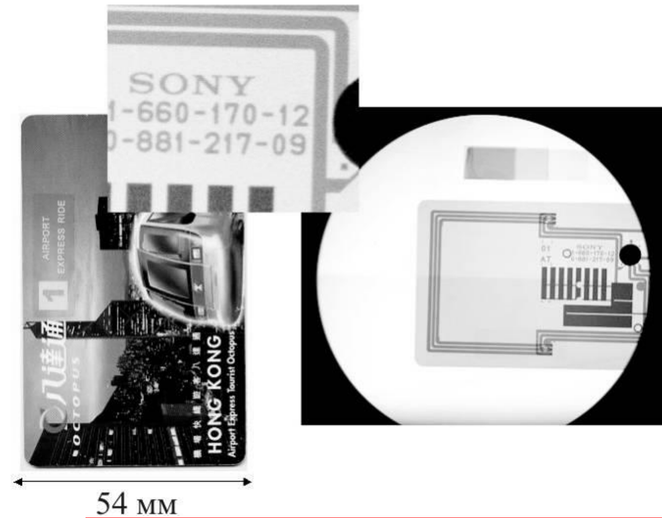
$$R = \frac{I_{DRO}}{I_{PXR}} \cong Q_{nn} \left(\frac{\theta_L}{4 \dfrac{\Delta\gamma}{\gamma} \theta_D} \right)^2 \frac{\left(1 + \theta_D^2 \gamma^2 + \gamma^2 \gamma_n^{-2}\right)\left(\sin^2 \psi + \cos^2 \psi \cos^2 2\theta_B\right)}{\left(\sin^2 \varphi + \cos^2 \varphi \cos^2 2\theta_B\right)}$$

For example, for *Si* target this ratio is estimated as **R~5** for electrons of 34 MeV energy channeled between (100) planes and radiation diffracted by (220) planes.

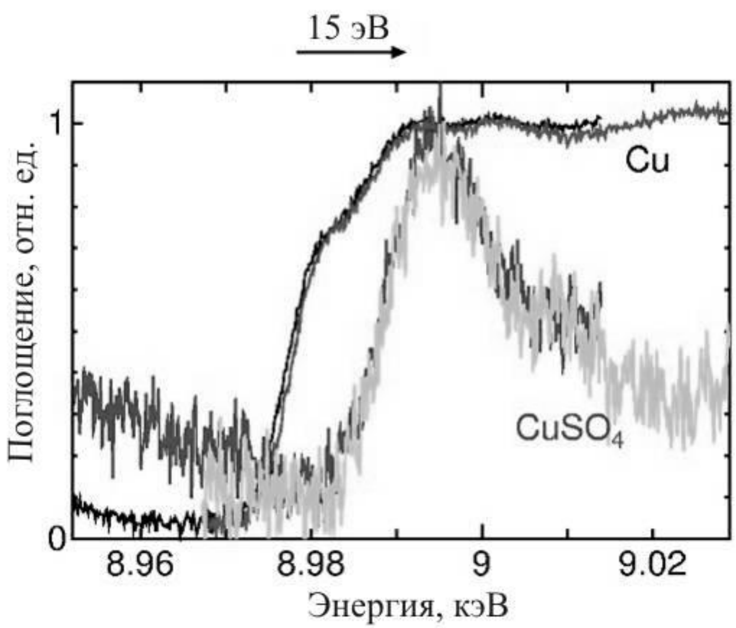
Practical examples of PXR applications

Nihon University, Japan

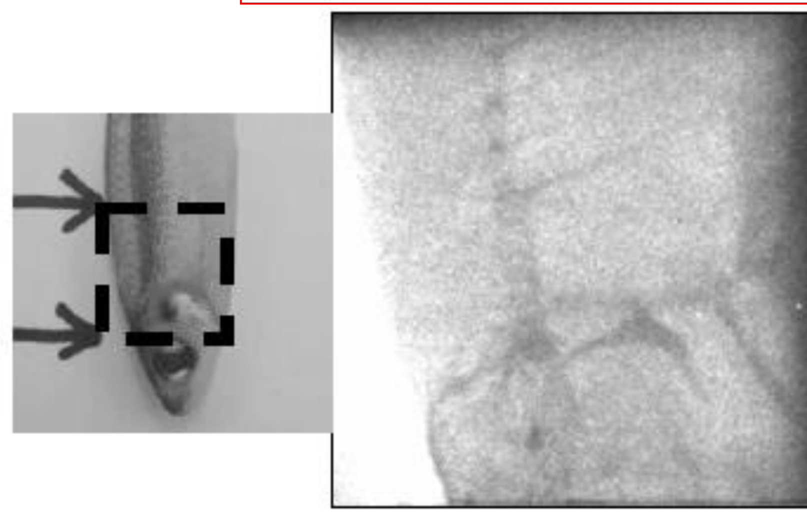




enhanced contrast images



EXAFS



Rensselaer Polytechnic Institute, NY, USA

Dose reduction

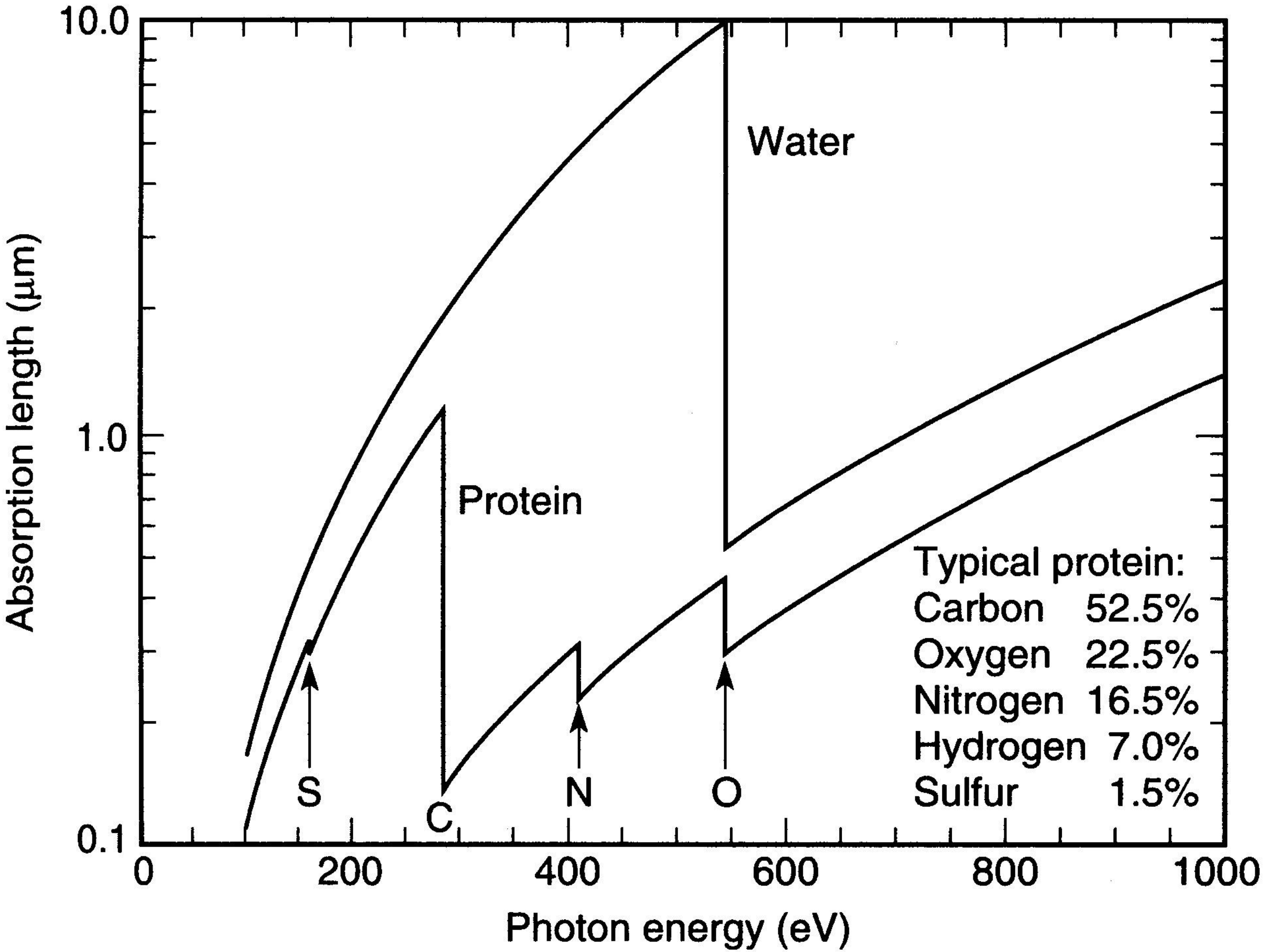
Possibility to reduce absorbed dose by PXR use:

- **Choice of the PXR frequency within “water window”**
- **Selective action on the specific molecular bonds**

$$D_{int}(G) \simeq \frac{1}{k}\int_{\omega_{min}}^{\omega_{max}} d\omega I(\omega)\sigma_{ph}(\omega)Y_f(Z)n_S L,$$

$$D_{res}(G) \simeq \int_{\omega_{min}}^{\omega_{max}} d\omega I_{res}(\omega)\sigma_{res}(\omega)Y_f(Z)n_S L,$$

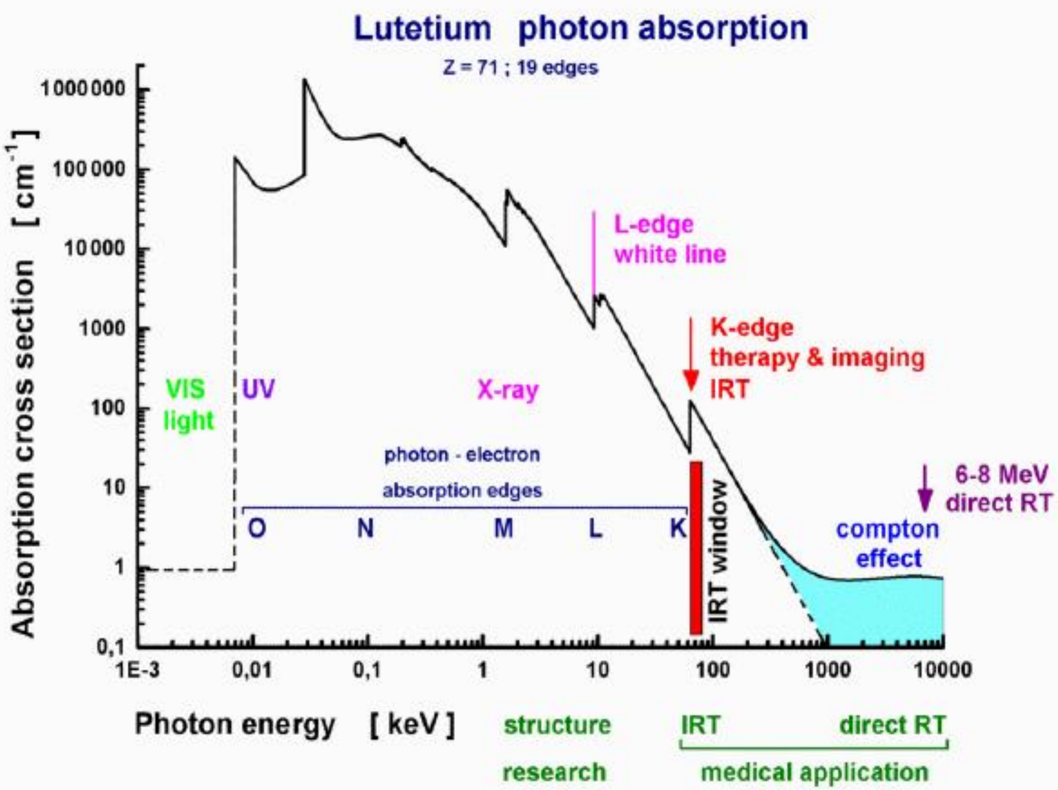
Soft x-ray water window



Photon activation therapy

Photon activation therapy PAT:

PAT / PXT : Lu, Gd + γ ⇒ $n \cdot e^-$ Auger-electrons (secondary radiation) ⇒ DNA inactivation @ tumor



A) Photon activation therapy PAT inactivates cancer cells by absorption of X-ray photons at the target K-edge and subsequent local production of toxic products, e.g. free radicals.

B) The photon absorption spectrum of Lutetium is the key for research and medical applications.

During indirect radiation therapy IRT photons of 63.3 keV are absorbed at K-electrons (1:1).
In the direct radiation therapy RT the compton effect of 8 MeV photons produces some compton electrons.

www.mpsd.de/irt

PXR effect on protein molecule

Deactivation of cysteine enzyme (ionization of the sulfur bridge) results in destruction of some molecules in cancer cells

$$NH_3 - CH - COO$$
$$|$$
$$CH_2$$
$$|$$
$$SH$$

Cysteine is sulfur-containing enzyme, which determines bonds and activity of many bio-active molecules

$$\sigma_{res}^{(S)} \approx 8.2\sigma_{non}^{(tot)}$$

Use of resonant radiation from PXR source could provide eight times reduction of total radiation dose for getting the equivalent radiation effect

For some cancer tumors number of sulfur atoms 3 times exceeds this number for normal cells

Soft X-Ray Imaging

Methods: Soft x-ray imaging, photoelectron emission microscopy (PEEM), scanning transmission x-ray microscopy (STXM), full-field microscopy, x-ray diffraction imaging (XDI), x-ray tomography, computer-aided tomography (CAT).

Problems:

Cell biology

Nano-magnetism

Environmental science

Soft matter, polymers

The tunability of radiation is absolutely essential for the creation of contrast mechanisms.

Soft X-Ray Spectroscopy

Methods: Soft x-ray absorption spectroscopy (XAS), near-edge x-ray absorption fine structure (NEXAFS) spectroscopy, soft x-ray emission spectroscopy (SXES), resonant inelastic x-ray scattering (RIXS), x-ray magnetic circular dichroism (XMCD), x-ray photo-emission spectroscopy (XPS), Auger spectroscopy.

Problems:

Complex materials

Magnetic materials

Environmental science

Catalysis

The photon energy <u>tunability</u> and its brilliance for some above listed applications are essential.

Soft X-Ray Scattering

Methods: Soft x-ray emission spectroscopy (SXES), inelastic x-ray scattering (IXS), resonant x-ray inelastic scattering (RIXS), speckle patterns, small-angle x-ray scattering (SAXS).

Problems:

Strongly correlated materials

Magnetic materials

Environmental science

Catalysis

The tunability of radiation and its brilliance for some above listed applications are essential.

Applications to the life sciences

- Potential to form high spatial resolution images in hydrated bio-material
- Ability to identify atomic elements by the coincidence between photon energy and atomic resonances of the constituents of organic materials

Concern

Radiation-induced damage: photon energy deposited per unit mass (dose) can cause observable changes in structure

CONCEPT OF PXR SOURCE

PHYSICAL REVIEW ACCELERATORS AND BEAMS 21, 014701 (2018)

Compact and intense parametric x-ray radiation source based on a linear accelerator with cryogenic accelerating and decelerating copper structures

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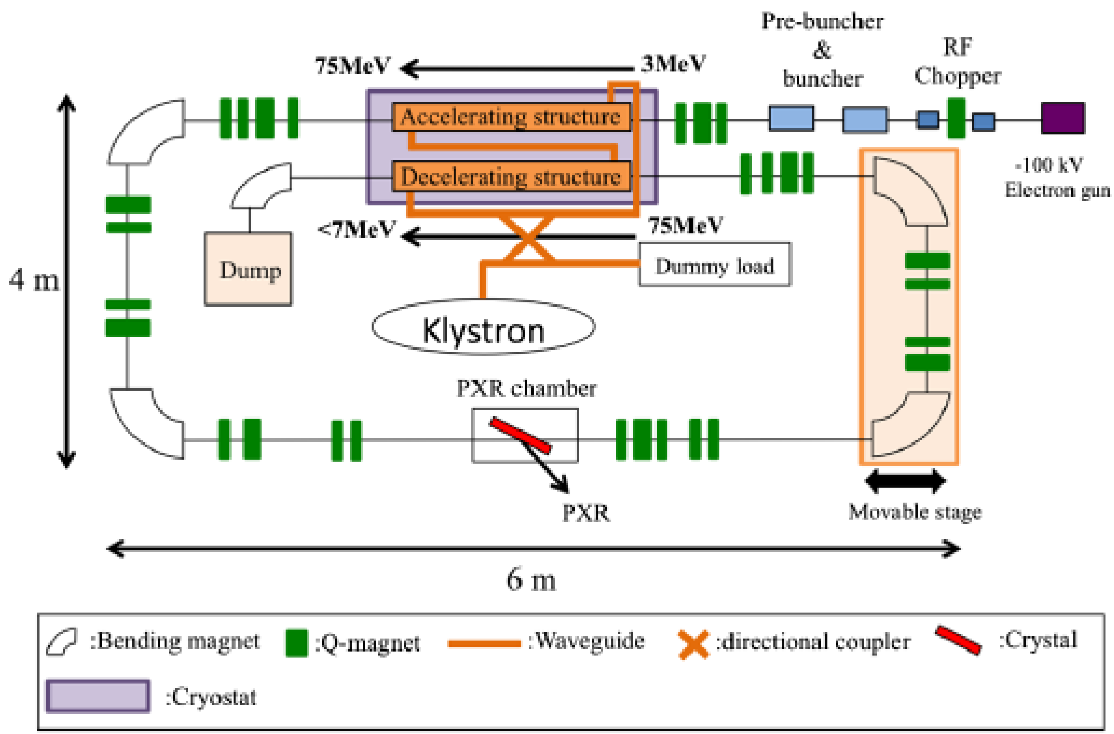
T. Tanaka,[3] K. Hayakawa,[3] I. Sato,[3] and K. Endo[4]



TIME DURATION OF THE PARAMETRIC X-RAY RADIATION

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Time evolution of the parametric X-Ray radiation, produced by a relativistic charged particle passing through a crystal, is studied. The most attention is given to the cases when the radiation lasts much longer ($t_{PXR} \sim 0.1$ ns) than the the time t_p of the particle flight through the crystal ($t_p \sim$ 1 ps). It is shown that such long duration of the radiation makes possible the detailed experimental investigation of the complicated time structure of the parametric X-ray pulses, generated by electron bunches, which are available with modern acceleration facilities.

Parametric (quasi-Cherenkov) cooperative radiation produced by electron bunches in natural or photonic crystals

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New targets

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Parametric X-rays from a polycrystalline target

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A B S T R A C T

A theoretical description of parametric X-ray radiation (PXR) from a nanocrystal powder target is presented in terms of the orientation distribution function (ODF). Two models of ODF resulting in the analytical solution for the PXR intensity distribution are used and the characteristic features of this distribution are considered. A promising estimate of the number of the emitted photons is obtained for the case of a nanodiamond powder target using the parameters of ASTA Facility at Fermilab. The PXR spectra from polycrystal and single crystal targets are compared. The application scenarios of PXR from nanocrystals are discussed.

PROSPECTS

We have fabricated homogeneous 500 nm-thick films made of single crystal HPTP nano-diamonds which demonstrated good x-ray diffraction patterns. Next step is to develop methods to produce sandwich-type ND targets of thickness suitable for the accelerator x-ray experiments, i.e. tens of microns.

We consider that targets made of nanodiamonds may provide good prospects in generating crystal-assisted radiations by the relativistic charged particles, such as **PXR, diffracted transition radiation, diffracted *Bremsstrahlung***, etc.

New Geometries

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Radical increase of the parametric X-ray intensity under condition of extremely asymmetric diffraction

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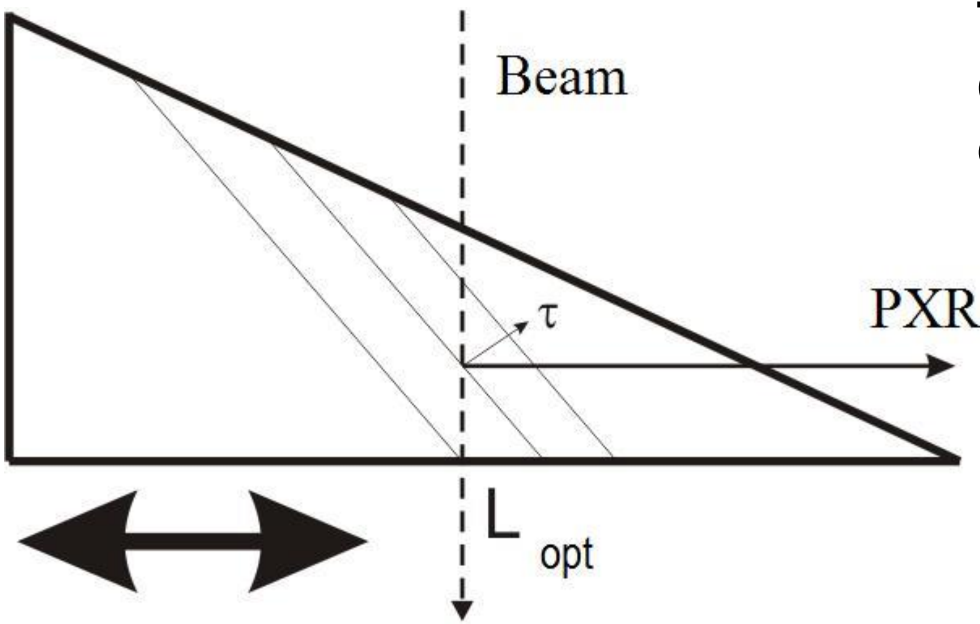
ABSTRACT

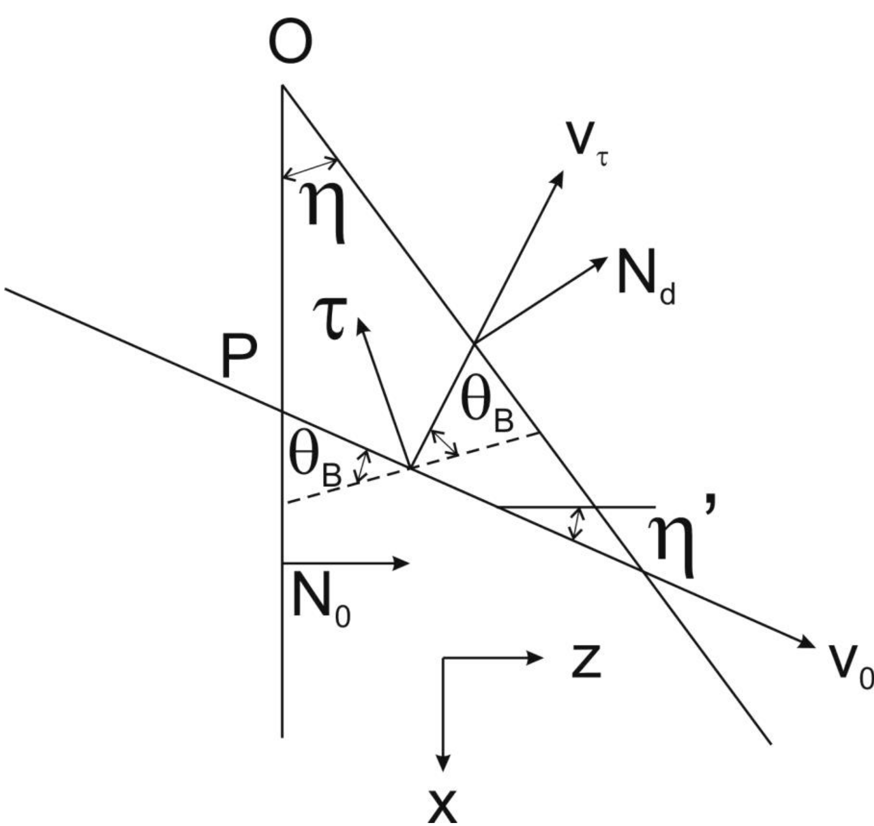
Parametric X-ray radiation (PXR) from relativistic electrons moving in a crystal along the crystal-vacuum interface is considered. In this geometry the emission of photons is happening in the regime of extremely asymmetric diffraction (EAD). In the EAD case the whole crystal length contributes to the formation of X-ray radiation opposed to Laue and Bragg geometries, where the emission intensity is defined by the X-ray absorption length. We demonstrate that this phenomenon should be described within the dynamical theory of diffraction and predict a radical increase of the PXR intensity. In particular, under realistic electron-beam parameters, an increase of two orders of magnitude in PXR-EAD intensity can be obtained in comparison with conventional experimental geometries of PXR. In addition we discuss in details the experimental feasibility of the detection of PXR-EAD.

Optimal PXR shape of crystal target - wedge



To calculate optimal asymmetric geometries and wedge configurations – dynamical theory required

$$\frac{\partial N}{\partial\omega\partial\Omega} \sim \exp\left(-\frac{L}{L_a}\right)$$



Ideas for the future research

- PXR Yield optimization (target material, Laue and Bragg, symmetric/asymmetric, wedge-shaped, X ray energy range, polarization, angular distribution, etc.) on the base of exact dynamical theory
- Analysis of experimental data
- Account of coherent *Bremsstrahlung* and PXR interference
- Development of dedicated targets
- Calculations of heat deposit and its dependence on PXR properties

Ideas for the future research

- Theory and simulations of PXR generation in bent crystals
- X ray optics (conventional, poly-capillary, bent crystals)
- Possibilities of biomedical applications
- Application of photonic crystals for radiation generation in THz, UV and soft X ray ranges
- Feasibility study of PXR collective generation