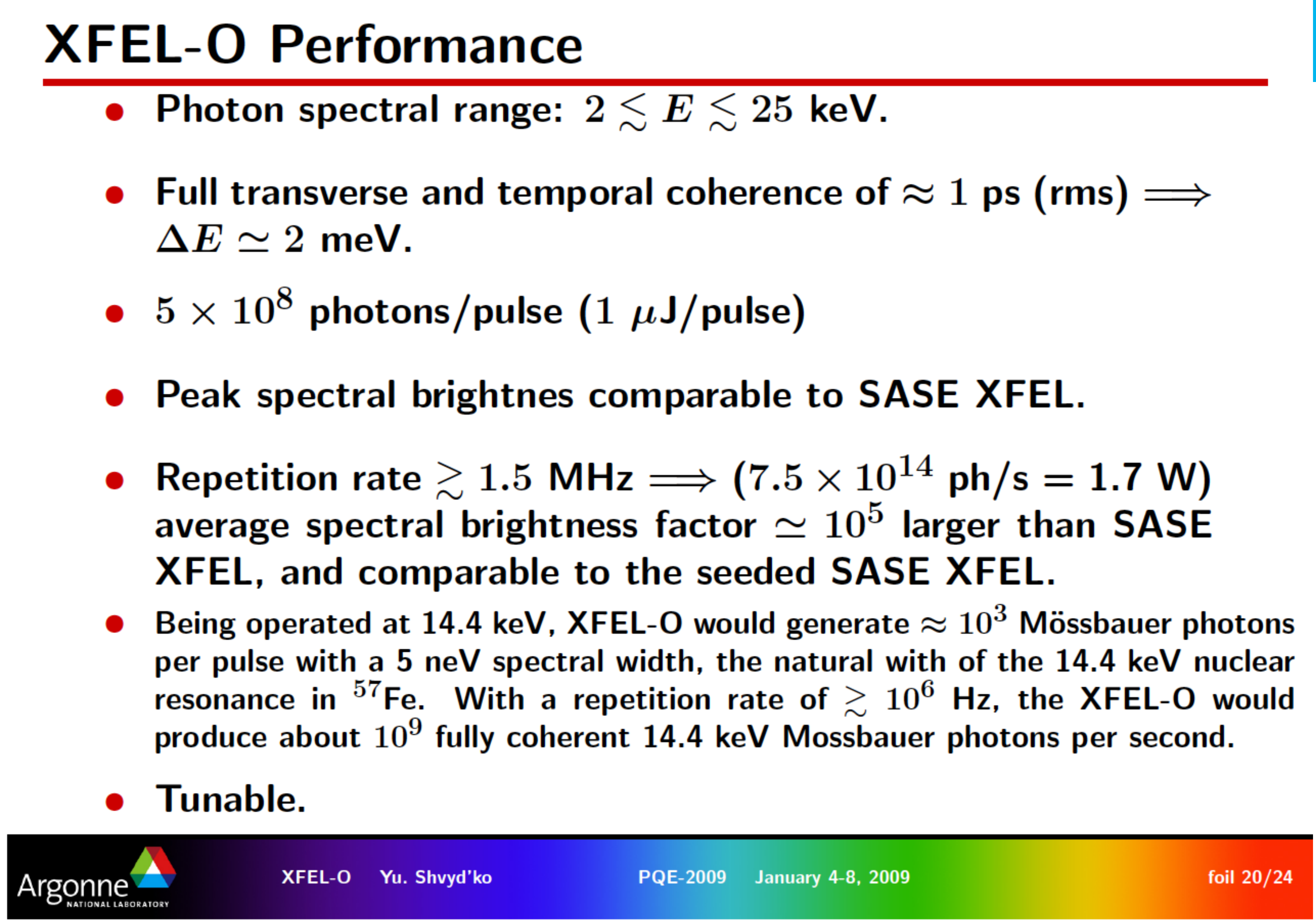
XFELO Retreat, SLAC, 28 June – 1 July 2016

Nuclear Resonant Scattering with an XFELO

Ralf Röhlsberger

Deutsches Elektronen Synchrotron DESY, Hamburg



XFEL-O Performance

- Photon spectral range: $2 \lesssim E \lesssim 25$ keV.
- Full transverse and temporal coherence of ≈ 1 ps (rms) $\Longrightarrow$ $\Delta E \simeq 2$ meV.
- 5×10^8 photons/pulse ($1\ \mu$J/pulse)
- Peak spectral brightnes comparable to SASE XFEL.
- Repetition rate $\gtrsim 1.5$ MHz $\Longrightarrow$ (7.5×10^{14} ph/s = 1.7 W) average spectral brightness factor $\simeq 10^5$ larger than SASE XFEL, and comparable to the seeded SASE XFEL.
- Being operated at 14.4 keV, XFEL-O would generate $\approx 10^3$ Mössbauer photons per pulse with a 5 neV spectral width, the natural with of the 14.4 keV nuclear resonance in ^{57}Fe. With a repetition rate of $\gtrsim 10^6$ Hz, the XFEL-O would produce about 10^9 fully coherent 14.4 keV Mossbauer photons per second.
- Tunable.

Argonne NATIONAL LABORATORY XFEL-O Yu. Shvyd'ko PQE-2009 January 4-8, 2009 foil 20/24

Outline

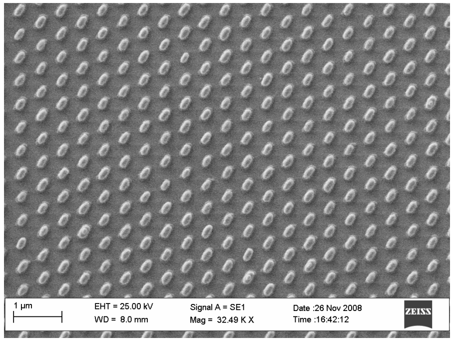
- Micro-eV resolved spectroscopy for dynamics in mesoscopic/artificially structured materials

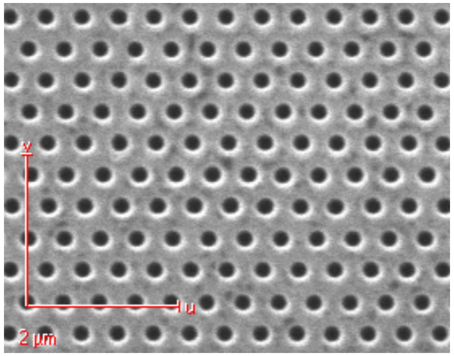
- Pump-probe NRS for non-equilibrium dynamics

- NRS ptychography in the time-energy domain for high-resolution spectroscopy of hyperfine interactions

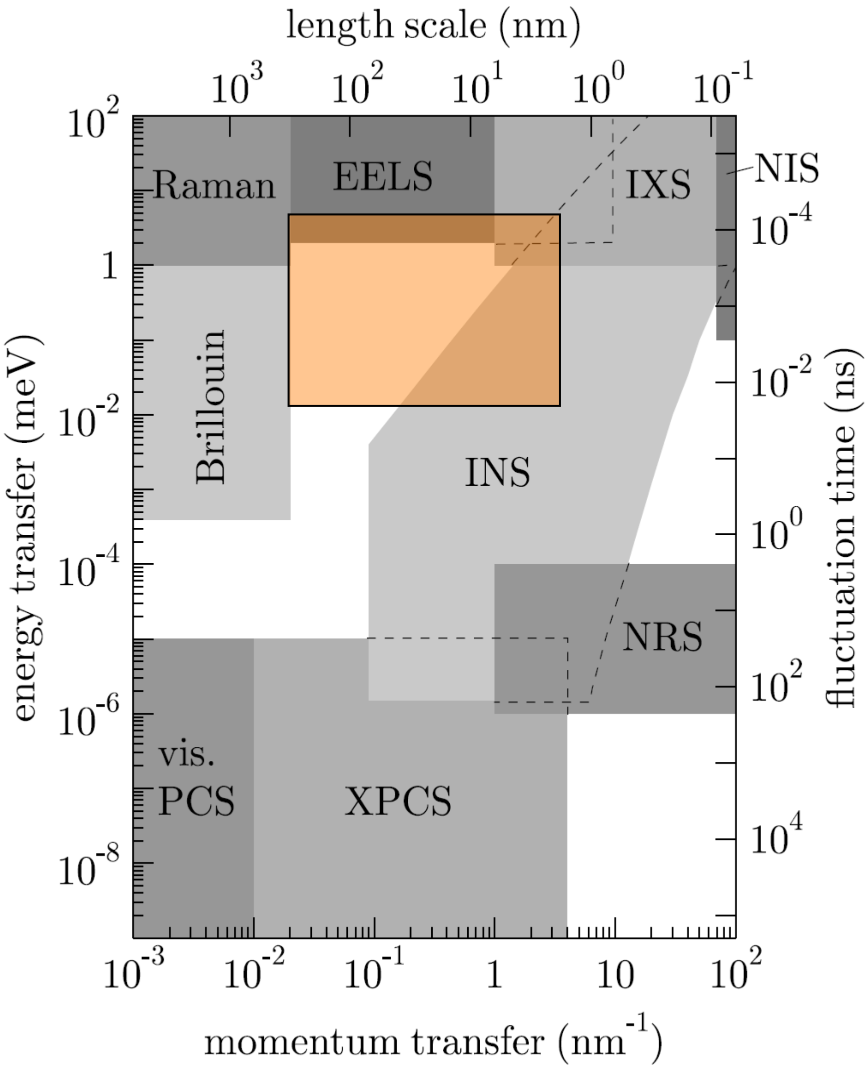
(1) Dynamics on mesoscopic length scales (1 nm – 100 nm)

Artificially structured materials

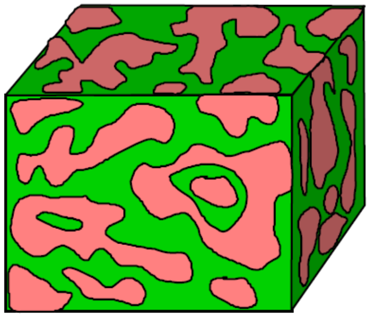


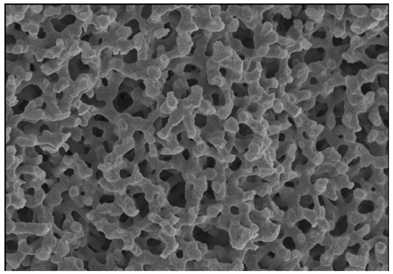


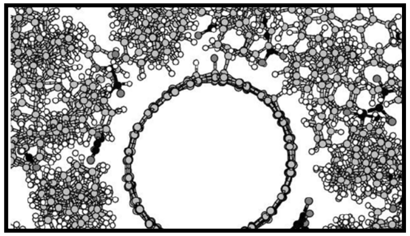
Photonic and phononic crystals



Nanocomposites







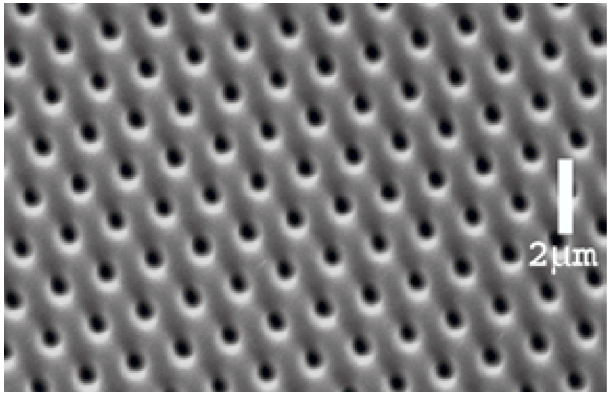
Dynamics of artificially structured materials

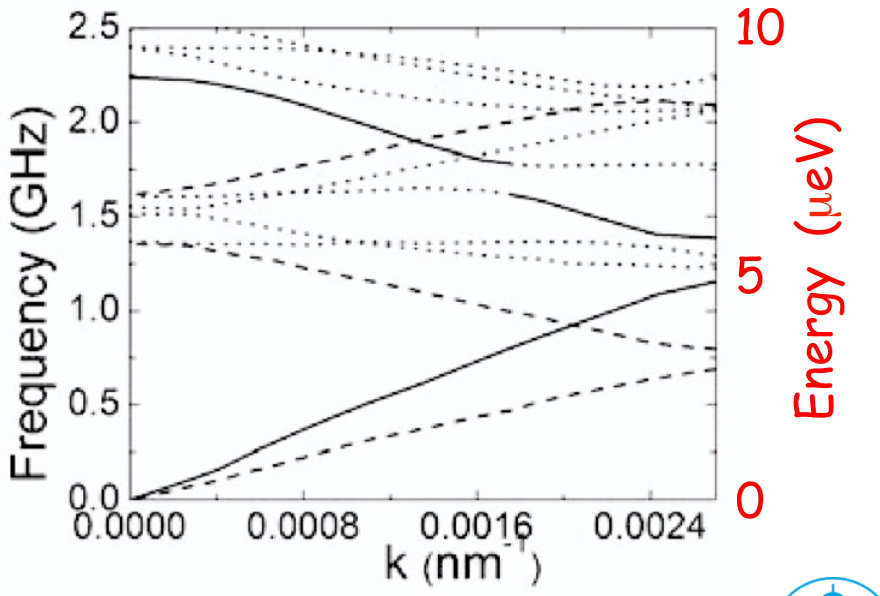
Dynamical properties are modified due periodic variation of elastic properties

This allows to tailor the vibrational properties of new materials by adjusting their structure

→ Nanocomposites (e.g. metal/polymer, amorphous/crystalline)

Phononic crystal

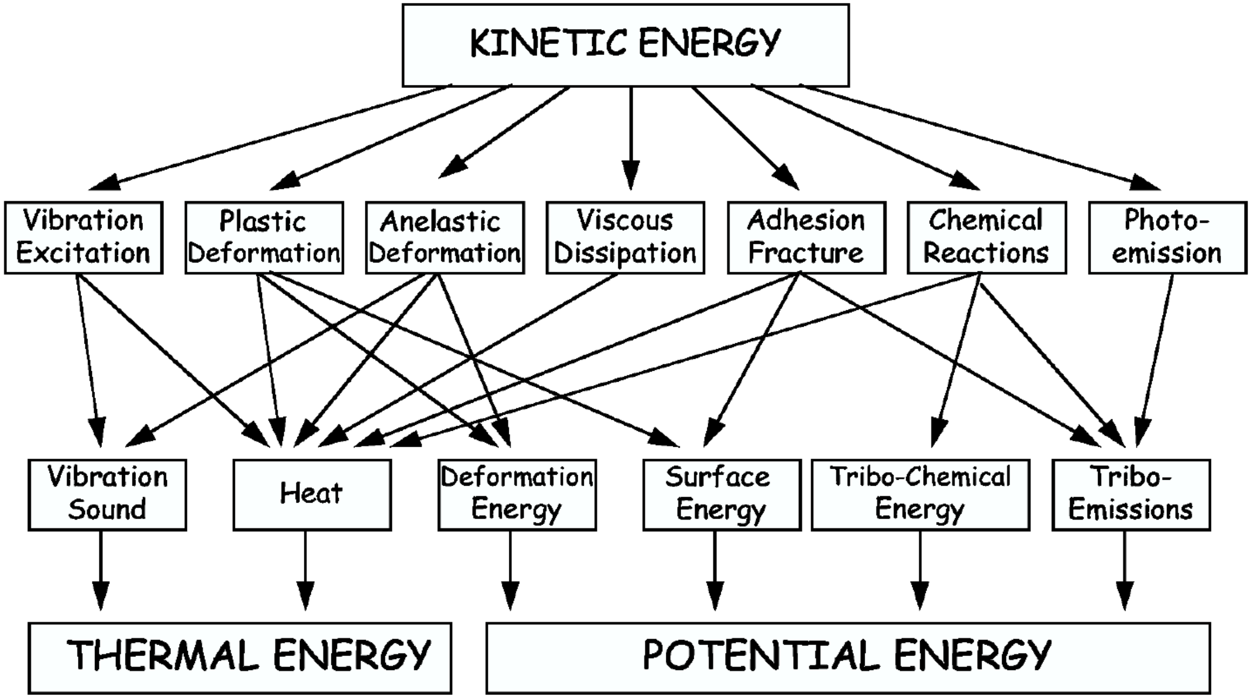




T. Gorishnyy et al., Phys. Rev. Lett. 94, 115501 (2005)

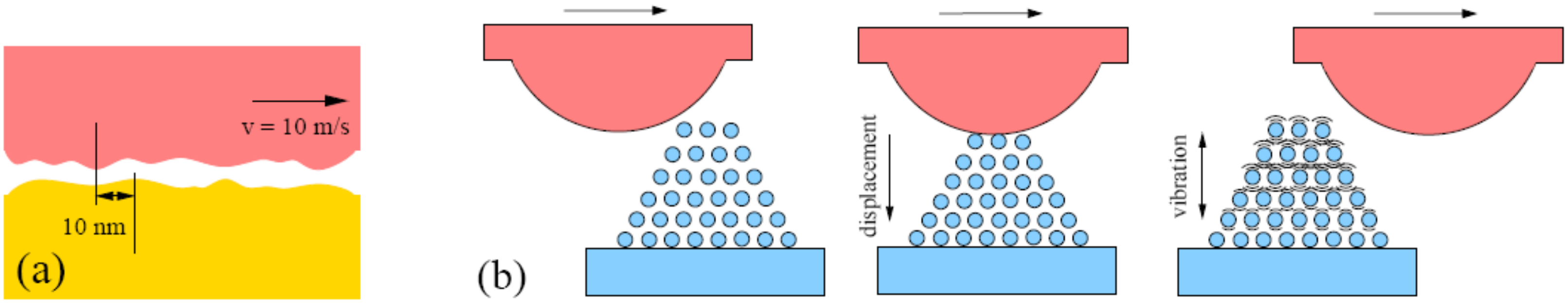
Friction in (micro)mechanical devices

Tailoring dynamical properties of materials
→ understanding the microscopic origin of energy dissipation



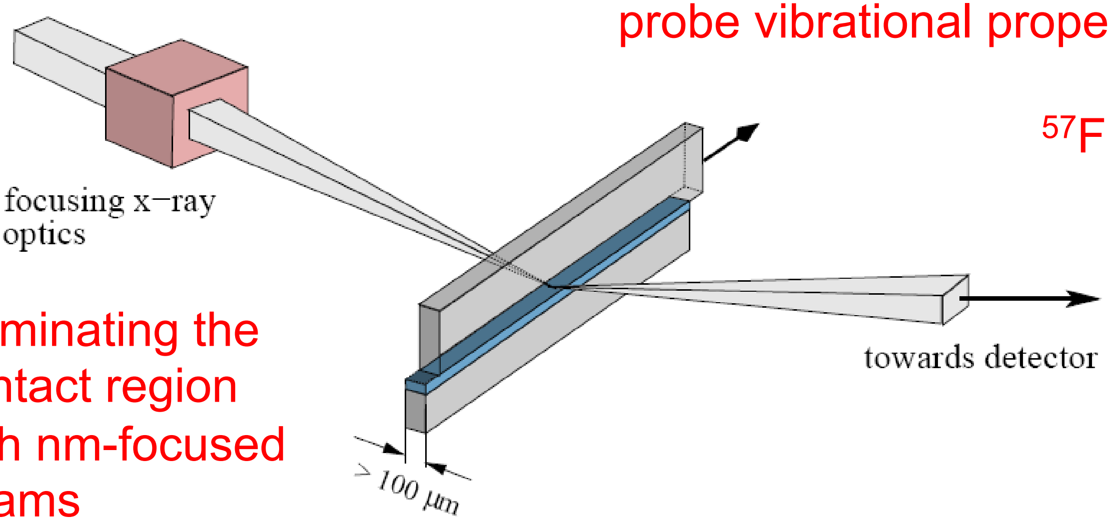
Investigation of kinetic friction with synchrotron radiation

Microscopic view of sliding friction

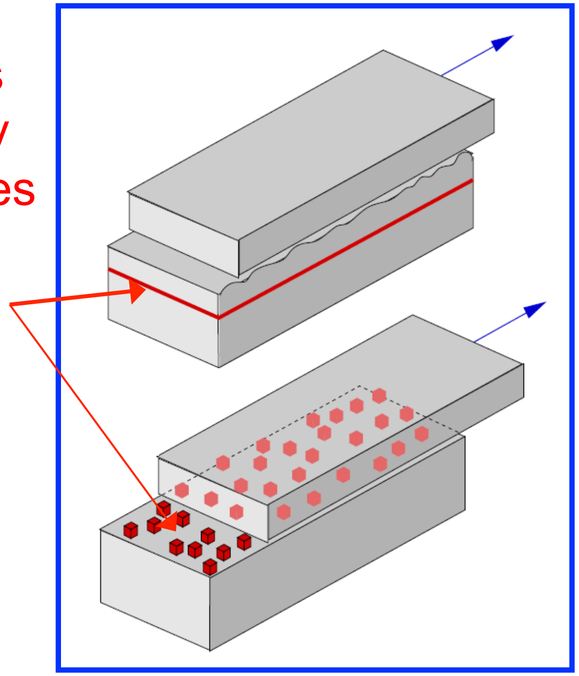


Experimental scheme

Employing ultrathin layers or nano particles to locally probe vibrational properties



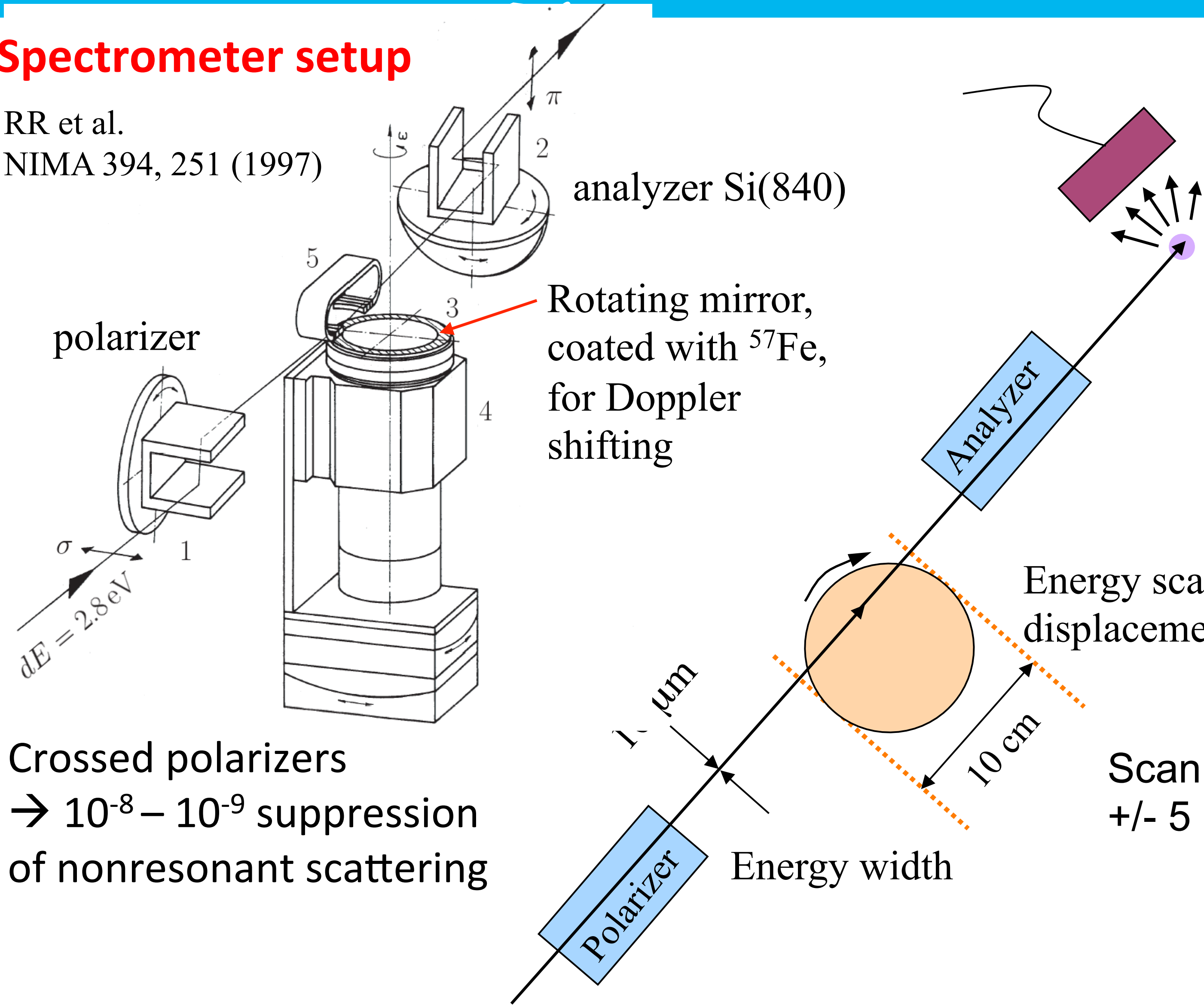
Illuminating the contact region with nm-focused beams



Nuclear inelastic spectroscopy with μeV-resolution

Spectrometer setup

RR et al.
NIMA 394, 251 (1997)



polarizer

analyzer Si(840)

Rotating mirror, coated with ^{57}Fe, for Doppler shifting

Monitoring the delayed fluorescence from ^{57}Fe analyzer nuclei in the sample

Energy scan by transverse displacement of disk

Energy width

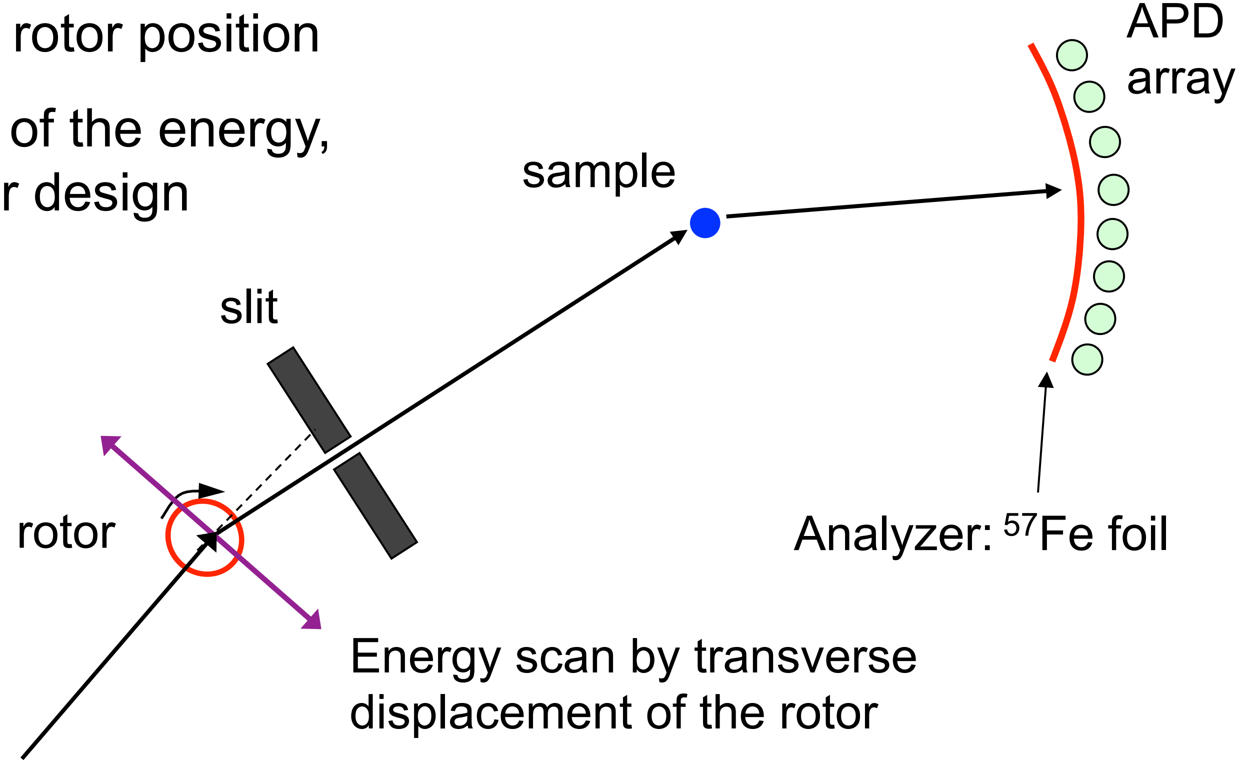
Scan range appr. +/- 5 meV

Crossed polarizers
→ $10^{-8} – 10^{-9}$ suppression of nonresonant scattering

A μeV spectrometer for IXS

Focusing of the beam to < 10 μm spotsize at the rotor position

Easy scanning of the energy, simple analyzer design



APD array

sample

slit

rotor

Analyzer: ^{57}Fe foil

Energy scan by transverse displacement of the rotor

Flux at sample position:

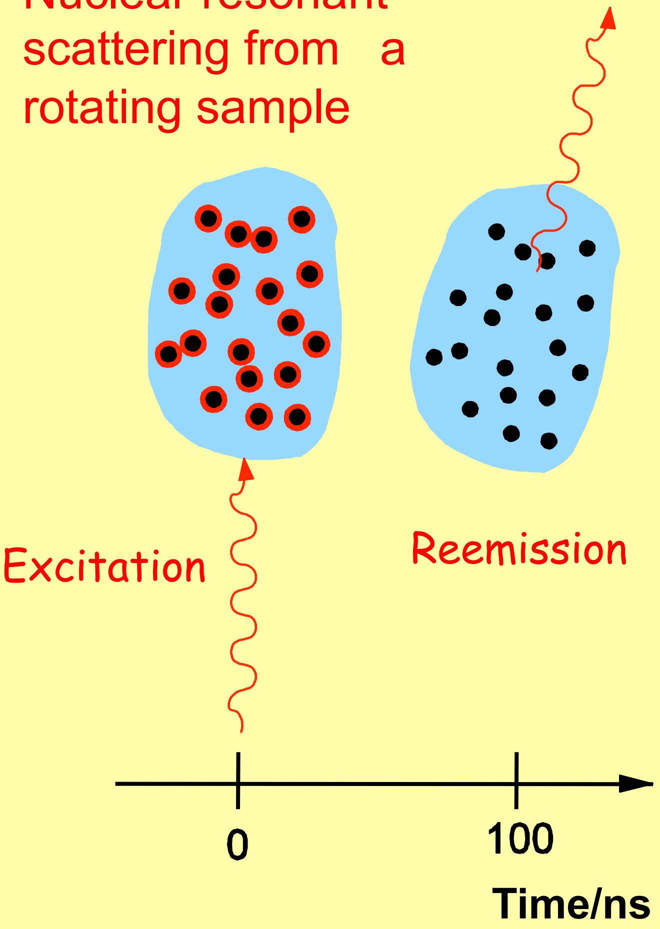
$I_0 = 10^5$/s existing sources, measured

$I_0 = 10^{10}$/s XFELO, expected

A μeV spectrometer, alternative version

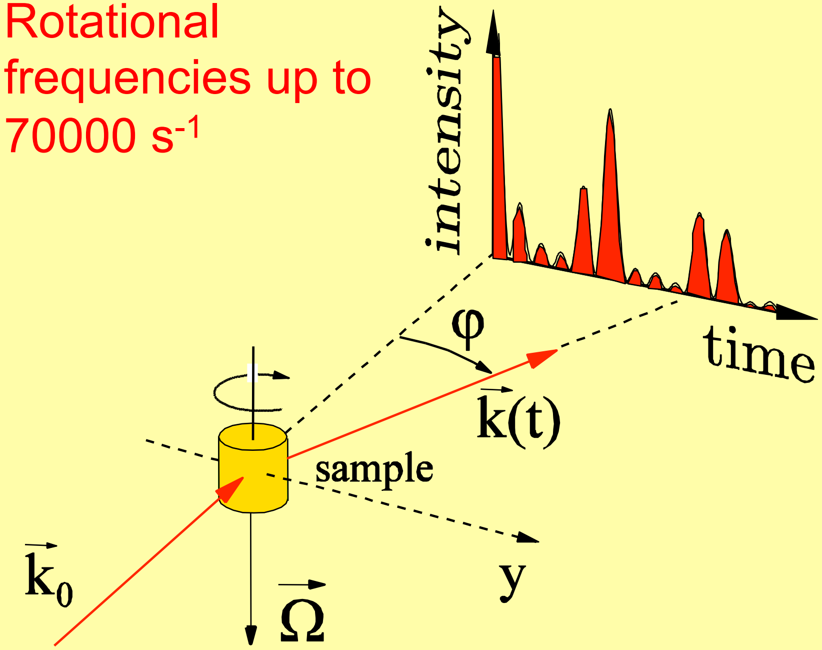
The nuclear lighthouse effect

Nuclear resonant scattering from a rotating sample



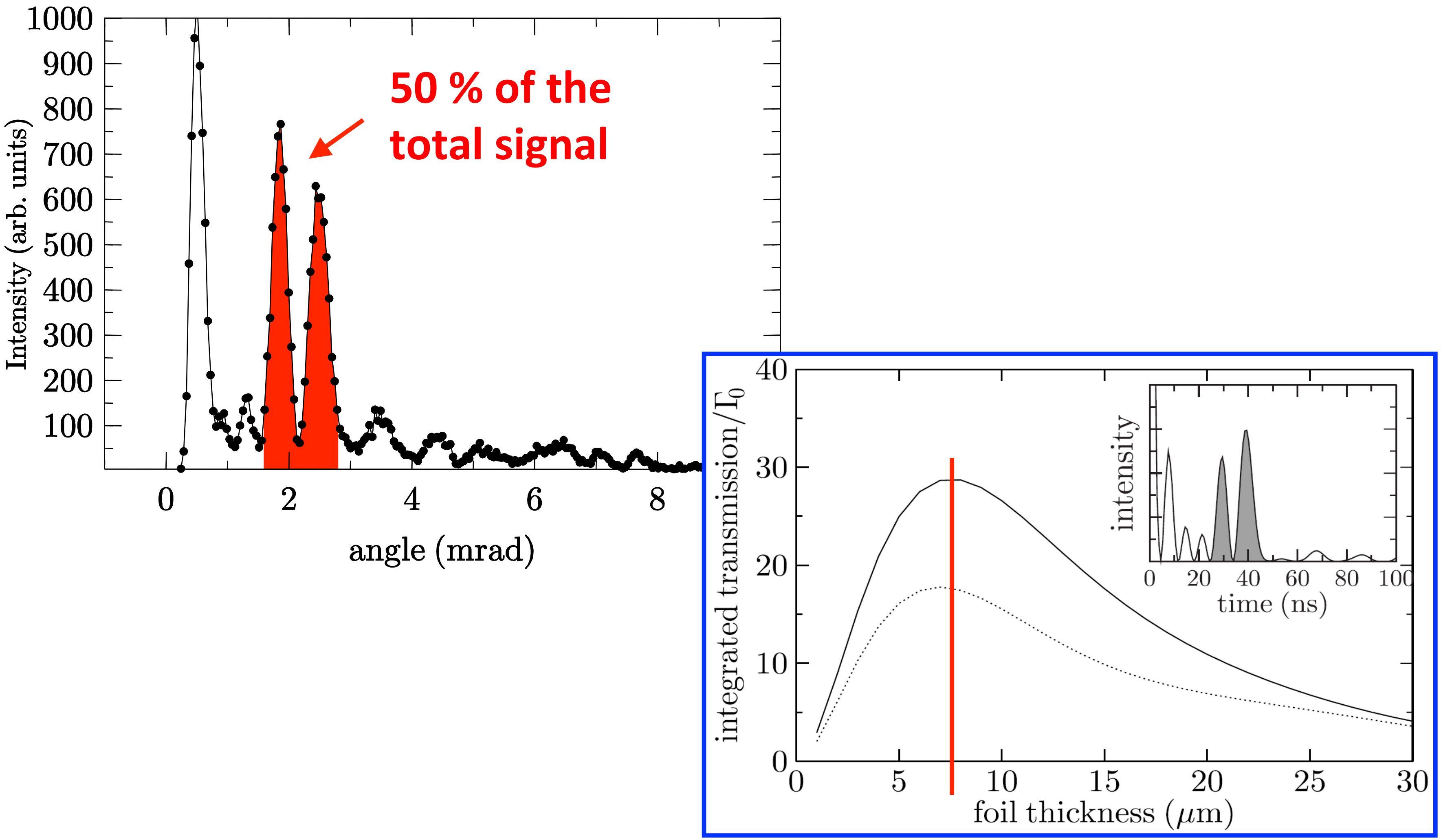
The time spectrum of the nuclear decay is mapped to an angular scale

Rotational frequencies up to 70000 s^{-1}



Phys. Rev. Lett. 84, 1007 (2000)

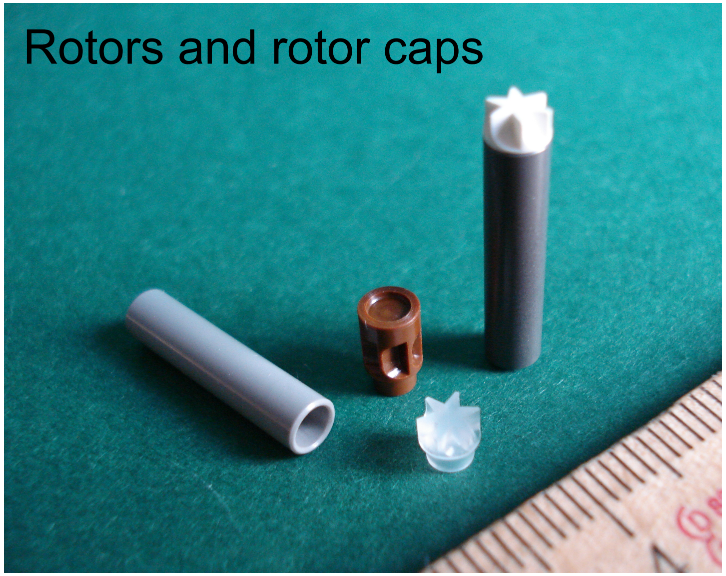
NRS from a polycrystalline ^{57}Fe foil

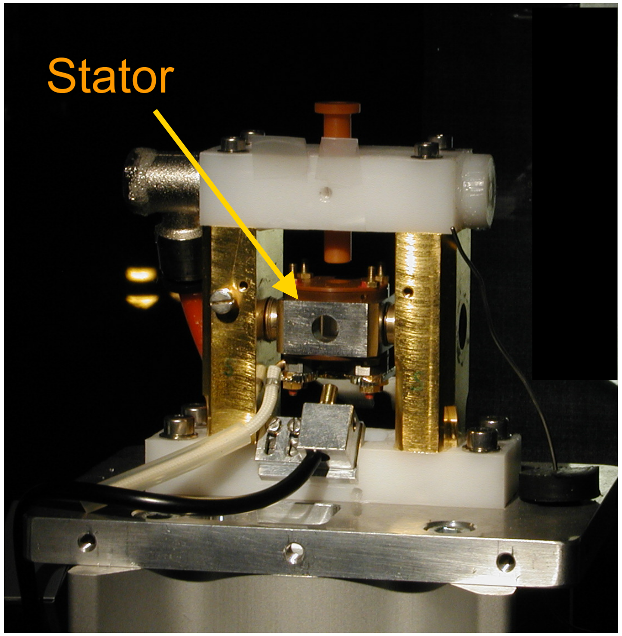


50 % of the total signal

Optimum thickness: 7.5 µm

Magic angle spinning (MAS) technique of NMR spectroscopy


Rotors and rotor caps



Rotational freq.	Diameter	V_{max} (m/s)	Material
7 kHz	7 mm	132	Si_3N_4
15 kHz	4 mm	141	Si_3N_4, ZrO_2
35 kHz	2.5 mm	165	ZrO_2
70 kHz	1.3 mm	110	ZrO_2

(2) Nonequilibrium dynamics

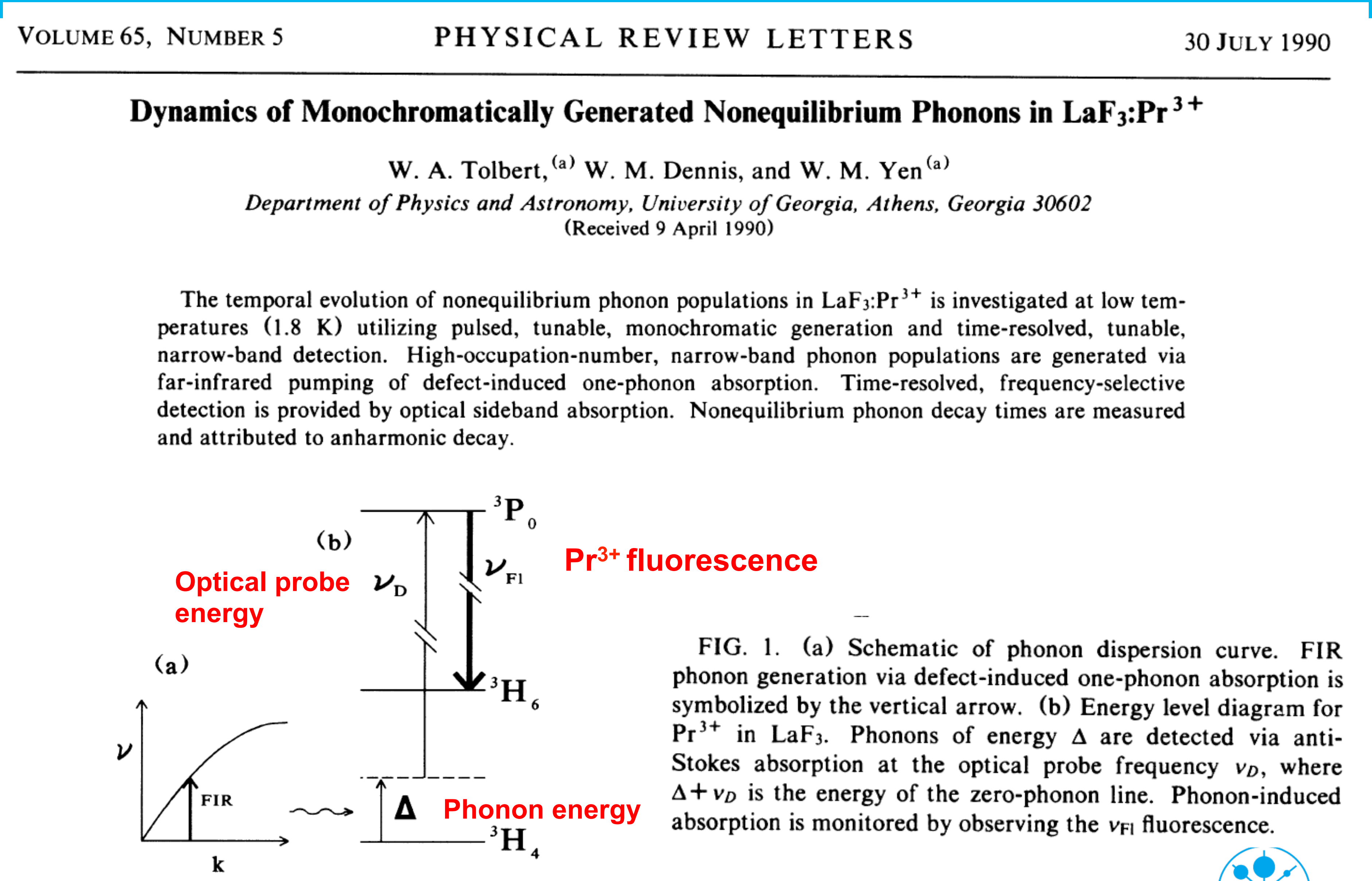
How do materials react on impulsive excitations ?

→ Short-pulse electric, magnetic and optical stimuli form the basis for high-speed data processing

→ Relaxation mechanisms important to understand, simultaneously on spatial and temporal scales

→ Probe the relaxation mechanism via nuclear resonant scattering (elastic and inelastic)

Probing non-equilibrium phonons via impurity atoms



VOLUME 65, NUMBER 5 PHYSICAL REVIEW LETTERS 30 JULY 1990

Dynamics of Monochromatically Generated Nonequilibrium Phonons in $LaF_3:Pr^{3+}$

W. A. Tolbert,(a) W. M. Dennis, and W. M. Yen(a)
Department of Physics and Astronomy, University of Georgia, Athens, Georgia 30602

(Received 9 April 1990)

The temporal evolution of nonequilibrium phonon populations in $LaF_3:Pr^{3+}$ is investigated at low temperatures (1.8 K) utilizing pulsed, tunable, monochromatic generation and time-resolved, tunable, narrow-band detection. High-occupation-number, narrow-band phonon populations are generated via far-infrared pumping of defect-induced one-phonon absorption. Time-resolved, frequency-selective detection is provided by optical sideband absorption. Nonequilibrium phonon decay times are measured and attributed to anharmonic decay.

Optical probe energy

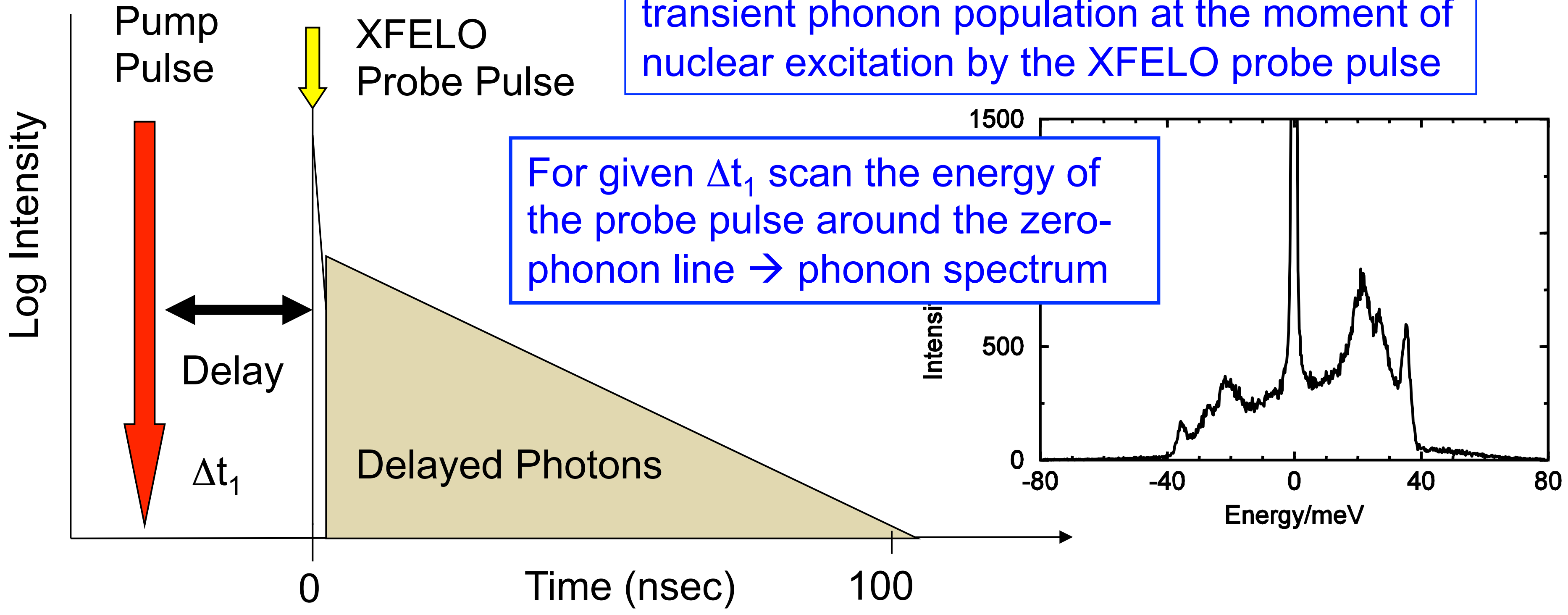
Pr^{3+} fluorescence

Phonon energy

FIG. 1. (a) Schematic of phonon dispersion curve. FIR phonon generation via defect-induced one-phonon absorption is symbolized by the vertical arrow. (b) Energy level diagram for Pr^{3+} in LaF_3. Phonons of energy Δ are detected via anti-Stokes absorption at the optical probe frequency ν_D, where $\Delta+\nu_D$ is the energy of the zero-phonon line. Phonon-induced absorption is monitored by observing the ν_{Fl} fluorescence.

Probing nonequilibrium phonons via nuclear excitation

Yield of delayed photons measures the transient phonon population at the moment of nuclear excitation by the XFELO probe pulse



For given Δt_1 scan the energy of the probe pulse around the zero-phonon line → phonon spectrum

- → Probing non-equilibrium phonon populations
- → Spatial resolution: Nanofocusing or ultrathin probe layers

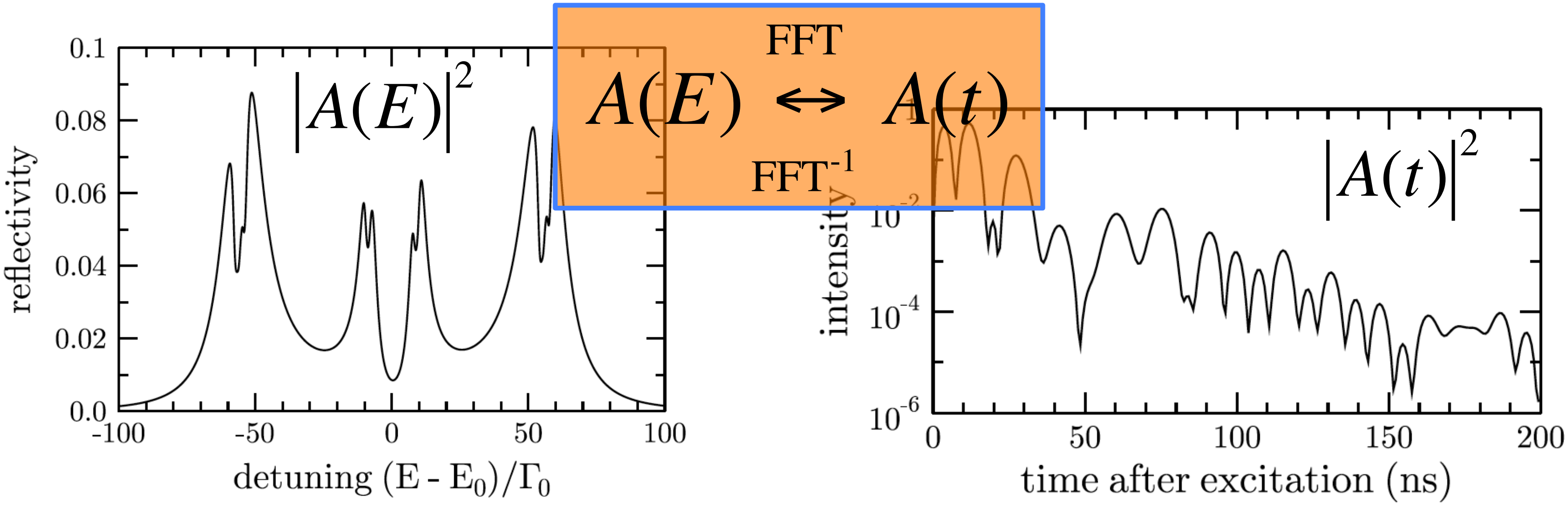
G. K. Shenoy and RR, Hyperfine Interact. 182, 157 (2008)

Application: Ptychographic Spectroscopy

Task: High-resolution spectroscopy of nuclear hyperfine interactions

Approach: Determine energy spectrum from diffraction pattern in the time domain

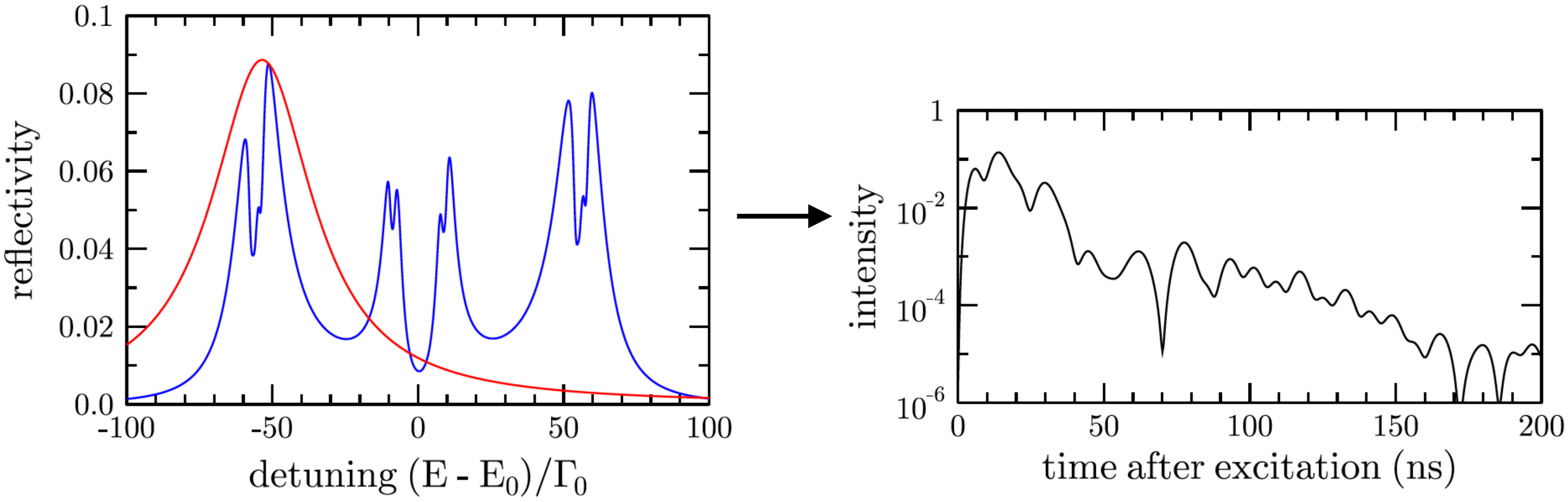
Problem: Phase is lost in the detection process



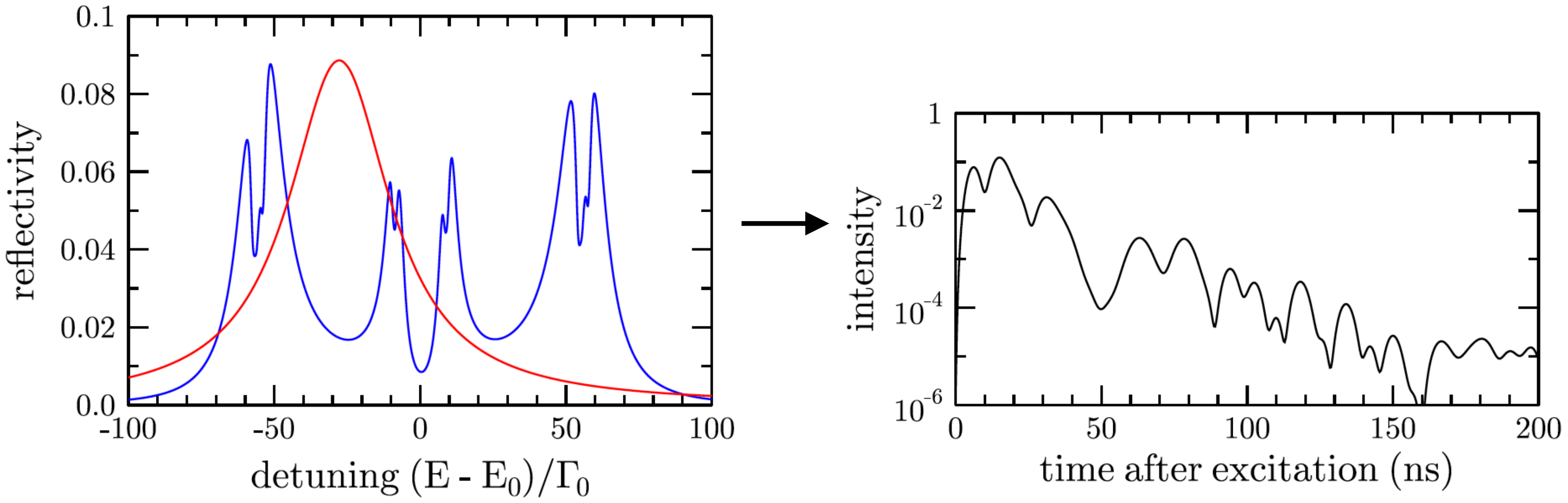
$$A(E) \xleftrightarrow[\mathrm{FFT}^{-1}]{\mathrm{FFT}} A(t)$$

Solution: Taking temporal beat patterns with overlapping illumination functions (as in Scanning Diffraction Microscopy a.k.a. Ptychography)

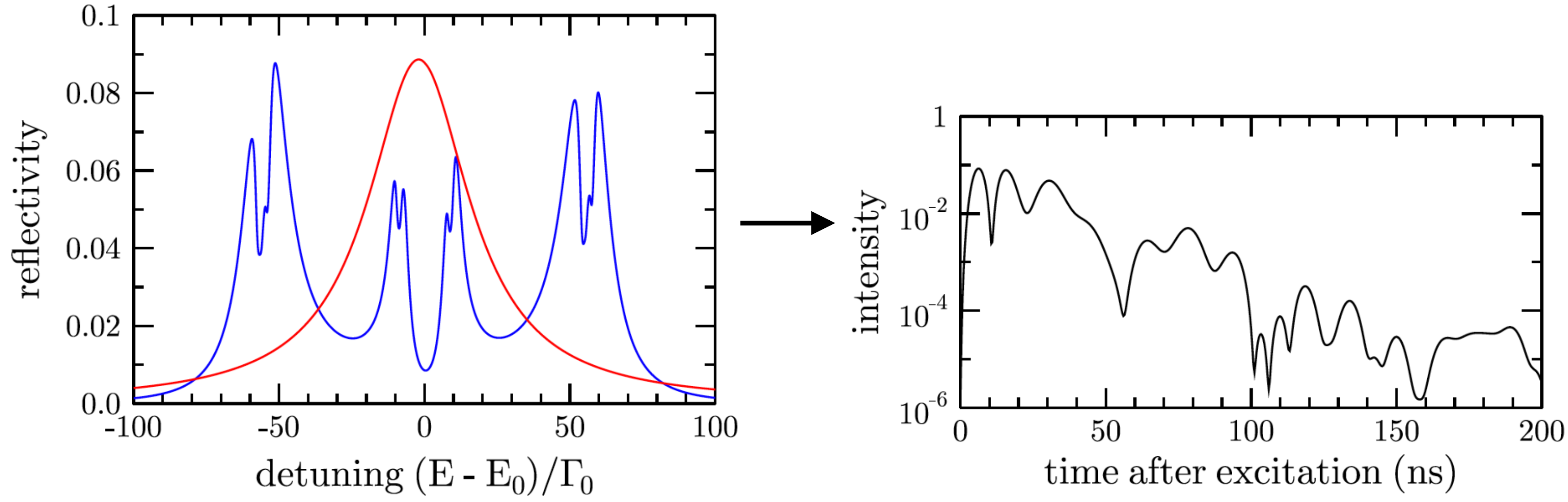
Ptychography in the Time – Energy Domain



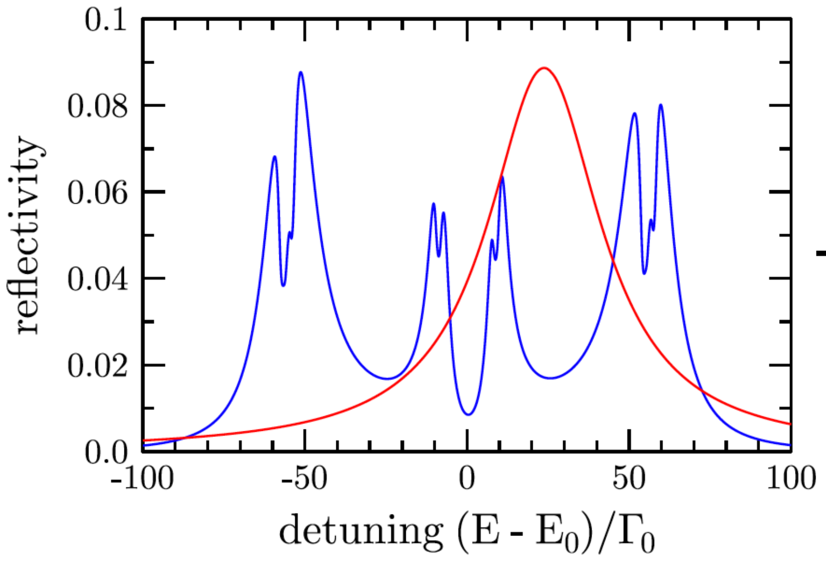
Ptychography in the Time – Energy Domain

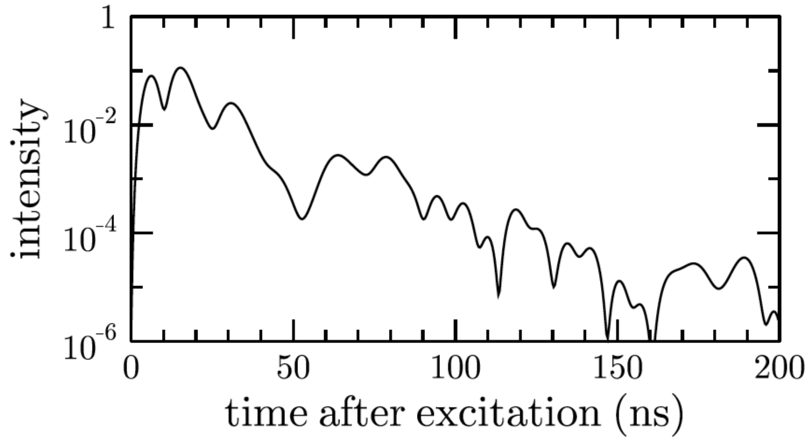


Ptychography in the Time – Energy Domain

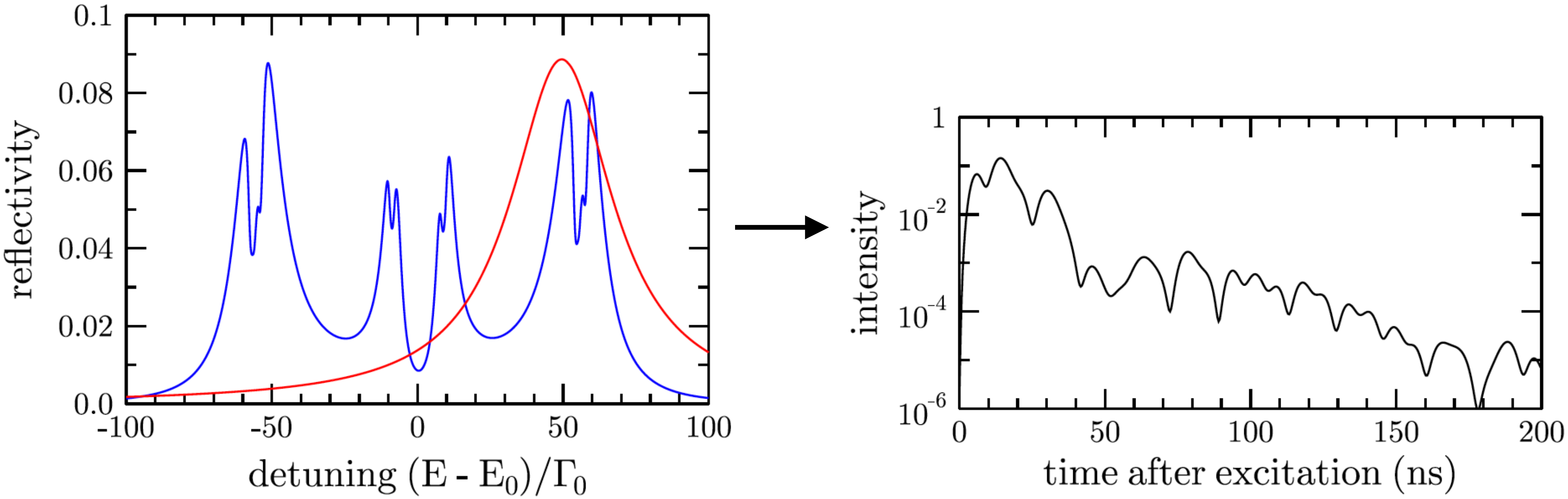


Ptychography in the Time – Energy Domain





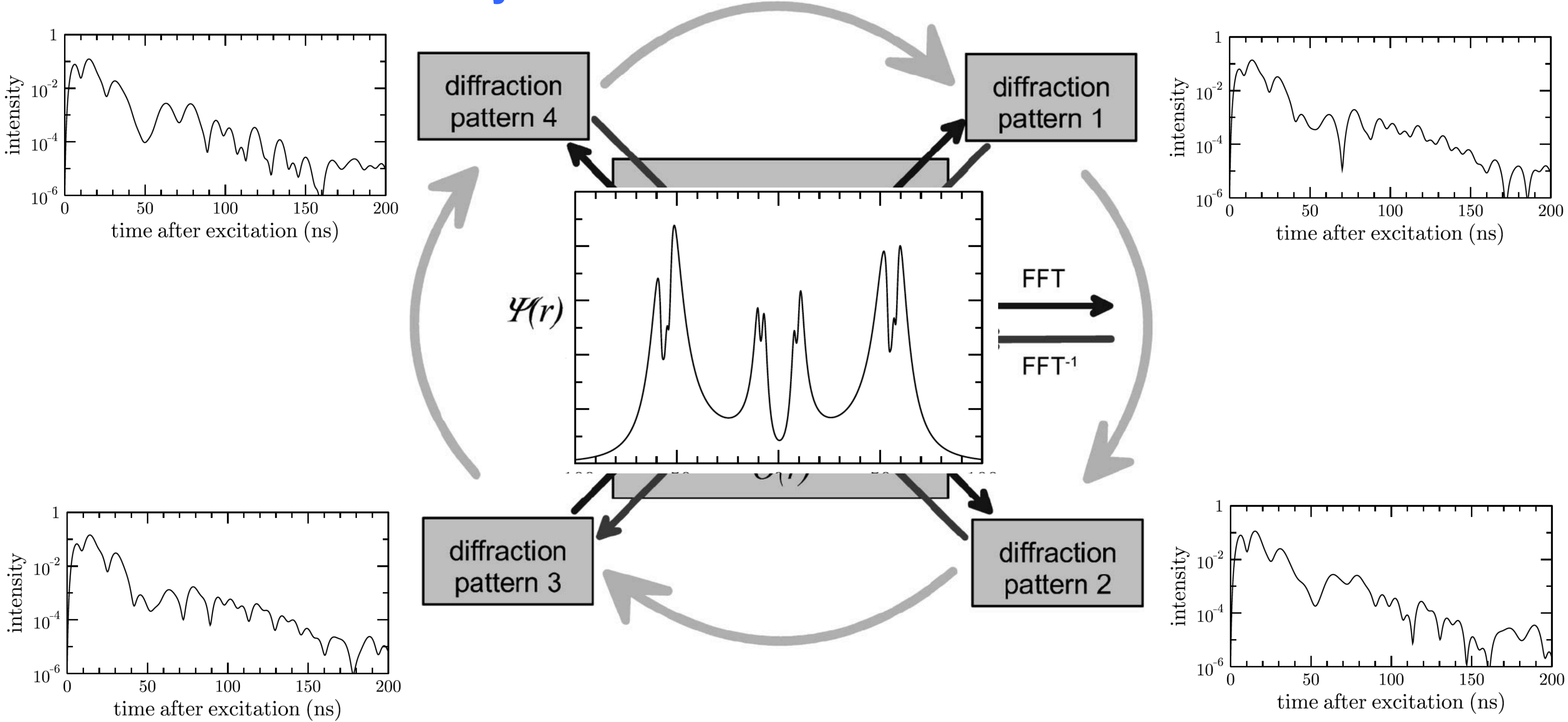
Ptychography in the Time – Energy Domain



Ptychography: Iterative Phase Retrieval Algorithm

Iterative image retrieval via back projection of diffraction patterns into the image plane

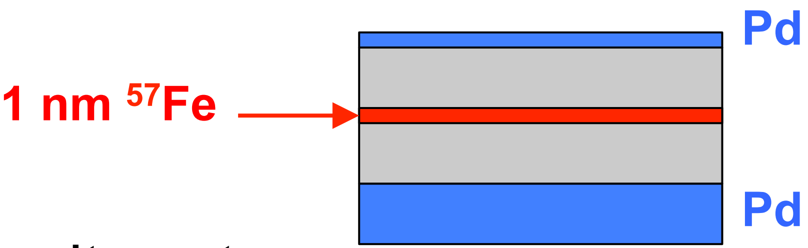
Fast converging and stable procedure due to high redendancy

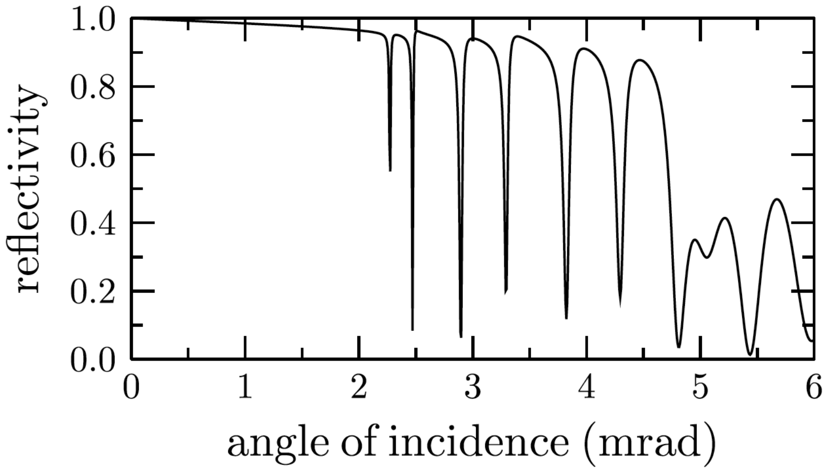


Method takes advantage of the long pulse separation of the XFELO (μs)

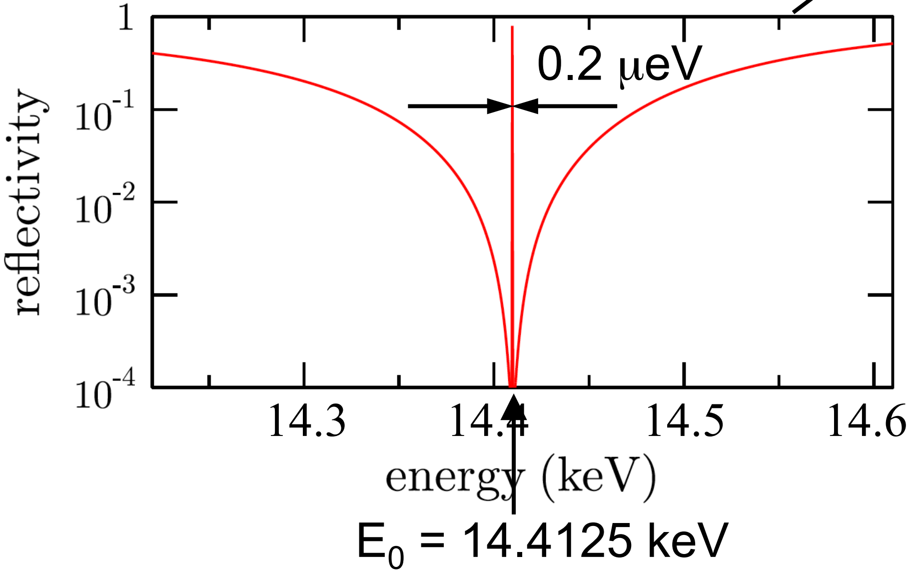
→ Precision spectroscopy of hyperfine interactions

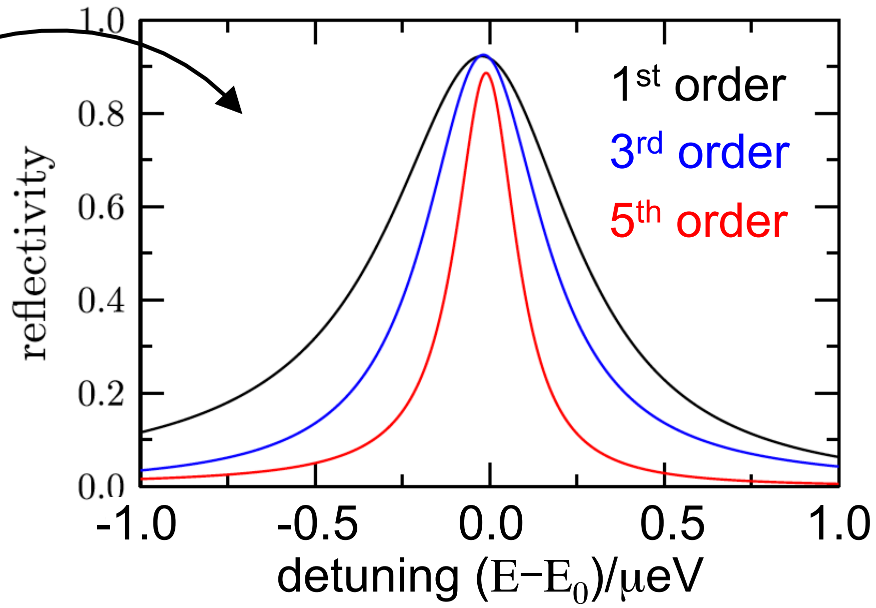
Ultranarrow Bandpass Filters for X-rays





Cavity acts as **Antireflection coating** for non-resonant radiation, but **Reflector** for resonant radiation





Summary

Nuclear resonant scattering with an XFELO

Scientific fields

- Dynamics of mesoscopic materials
- Non-equilibrium dynamics
- High – resolution hyperfine spectroscopy

Methods

- Phonon microscopy with μeV-resolution
- Nuclear resonant pump-probe experiments
- Time – energy domain ptychography