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Techniques for Inelastic X-ray Scattering with μeV-Resolution

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We introduce a novel type of spectrometer that provides a μeV bandpass together with a tunability over a few meV. The technique relies on nuclear resonant scattering (Mössbauer effect) of synchrotron radiation at the 14.4-keV resonance of ^{57}Fe. Energy tuning is achieved by the Doppler effect in high speed rotary motion. The resonantly scattered monochromatic radiation is extracted by a polarization filtering technique or by spatial separation due to the 'nuclear lighthouse effect'.

Keywords: Mössbauer effect, monochromatization, spectrometer, μeV-resolution

Inelastic x-ray scattering studies with high energy resolution have gained momentum in recent years, particularly due to the high brilliance of third-generation, undulator-based synchrotron radiation sources. Vibrational excitations in solids and liquids are studied with meV-resolution by using backscattering monochromators and analyzers [1,2]. A different approach has been introduced to measure the vibrational density of states (VDOS) via inelastic nuclear resonant scattering [3–5]. In that case, Mössbauer nuclei in the sample are used as energetic analyzers. This method relies on detection of time-delayed fluorescence photons emitted by decaying nuclei that were excited by synchrotron radiation. The yield of those fluorescence photons, as a function of energy gives a direct measure of the VDOS of the ^{57}Fe atoms in the sample. Energy resolutions from a few meV down to the sub-meV range are achieved in this kind of spectroscopy by using high-order Bragg reflections [6–8]. However, there is currently no x-ray spectroscopic technique available that covers the range from a few μeV to a few meV with μeV resolution. Vibrational excitations in this energy range have attracted considerable attention recently. For instance, vibrational spectra of disordered solids exhibit a universal feature in the range of 1–10 meV, the so-called boson peak, its origin still being subject of debate [9]. Further examples are magnons, two-level systems, phasons in quasicrystals, rotational excitations in liquids, soft phonons, etc.

The first type of spectrometer to be discussed here is shown in fig.1 [10]. It employs a grazing incidence reflection from a thin film of ^{57}Fe magnetized parallel to the incident beam (Faraday geometry). In this scattering geometry, strong orthogonal scattering from incident σ - polarization into π - polarization occurs in a very narrow (μeV) range around the 14.4 keV resonance of ^{57}Fe. This narrow-band π - polarization component can be separated from the broad nonresonant charge scattering by polarization filtering. This is accomplished by two Si(840) channel-cut crystals that serve as a polarizer/analyzer pair in crossed setting. In this geometry the π - polarization is filtered out with a rejection ratio of up to 10^{-8} against the σ - polarization.

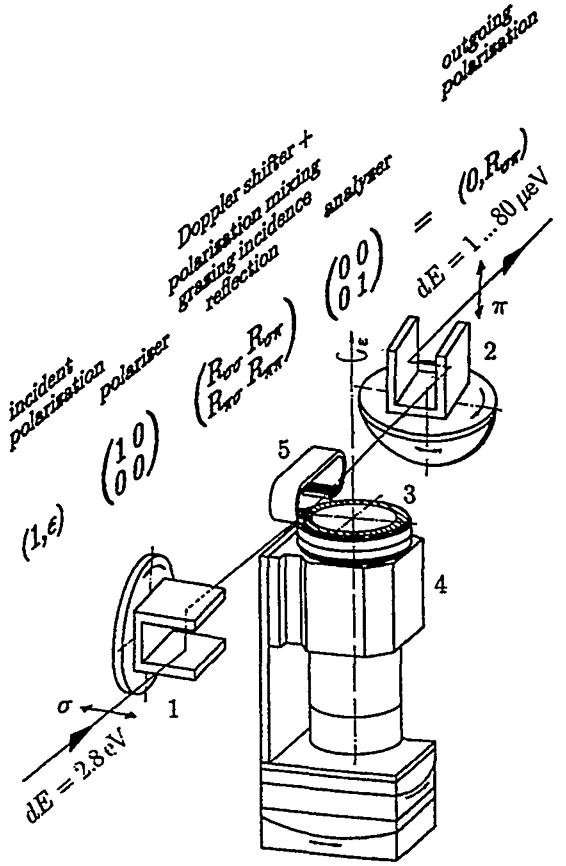

FIG. 1. Schematic setup of a spectrometer with μeV-resolution based on elastic nuclear resonant scattering. 1,2: Si(840) polarizer/analyzer pair in crossed setting, 3: Superpolished mirror coated with ^{57}Fe on Pd, 4: Motor, 5: Permanent magnet

Tunability is achieved by coating the film on the surface of a fast rotating disk of 15 cm diameter that acts as a Doppler shifter. When the disk rotates, nuclear resonant reflection takes place at an energetic position that is shifted relative to the resonance energy E_0. The energy shift ΔE is determined by the disk-velocity component $v_{\|} = \omega\, x$ of the disk parallel to $\vec{k}_0$, i.e. $\Delta E = (\omega\, x/c)\, E_0$, where x is the transverse displacement of the beam relative to the center of rotation. Therefore, ΔE can be varied either by changing the frequency ω or the displacement x. For example, at 14.4 keV, Doppler shifts up to ±3 meV are reached at rotational frequencies $\nu = \omega/2\pi$ of up to 150 Hz.

The maximum transmission through the spectrometer is observed when the ^{57}Fe film is coated on a total reflecting substrate layer (here: Pd) and the angle of incidence coincides with the critical angle of the substrate layer. The angular dependence of the transmitted intensity is shown in fig.2. The inset shows the energy spectrum of the transmitted intensity with a FWHM of appr. 0.7 µeV.

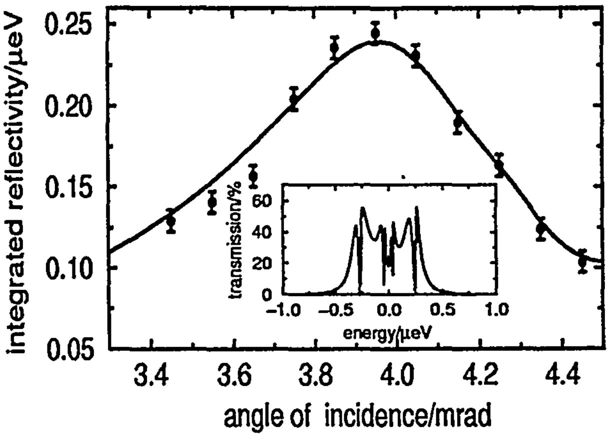

FIG. 2. Transmission through the spectrometer, as a function of incidence angle on the ^{57}Fe/Pd film, recorded with resonantly scattered, i.e. time-delayed quanta. (Experiment carried out at ESRF, Grenoble)

Another approach to ultra-high resolution monochromatization is introduced by nuclear resonant scattering from samples rotating with very high frequencies of several kHz. Due to the long lifetime of the ^{57}Fe resonance of 141 ns, the resonantly scattered radiation is deflected off the intense primary beam by an angle that is proportional to the decay time of the excited nuclear state. Accordingly, the time spectrum of the nuclear decay is mapped to an angular scale as shown on the left hand side of fig.3. At a rotational speed of 15 kHz, e.g., the angular deflection is appr. 0.1 mrad/ns. This 'nuclear lighthouse effect' [11] allows to extract a µeV wide band out of synchrotron radiation simply by angular selection. The setup is shown schematically in fig.3. The rotor is a hollow cylinder of 3 mm in diameter that contains an α - ^{57}Fe foil. The tunability is demonstrated on the right hand side of fig.3, where the energy of the transmitted radiation was measured for various displacements Δx.

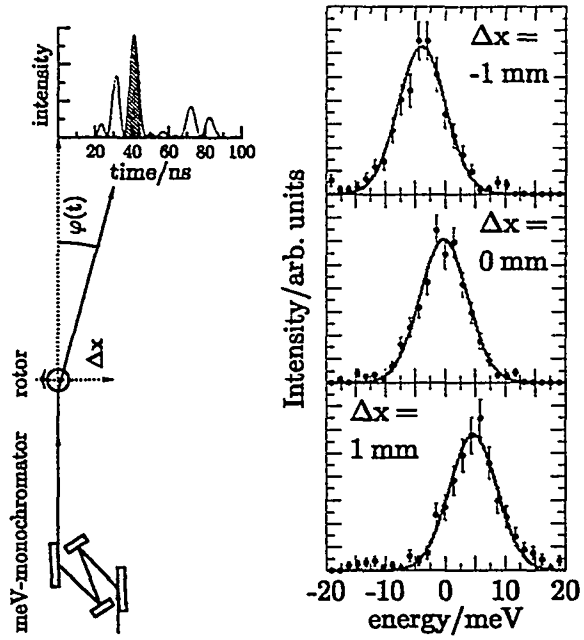

FIG. 3. µeV bandpass filtering and meV energy tuning synchrotron radiation by the nuclear lighthouse effect. (periment carried out at HASYLAB, Hamburg)

In future experiments, samples will be placed downstream of the spectrometer. Flux values on the sample close to 10^5 s^{-1} are expected at third generation synchrotrons like ESRF,APS and SPring8. This should lead to inelastic countrates in the order of 0.1 s^{-1} for samples containing Mössbauer isotopes, which is quite comparable to countrates in neutron inelastic scattering.

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