NUCLEAR FORWARD SCATTERING OF SYNCHROTRON RADIATION

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1. Introduction

In 1974, Ruby has designed a new method for investigation of hyperfine interactions of nuclei in time domain using nuclear forward scattering of synchrotron radiation [1]. Since the 90-ties of the last century, extremely high brilliance of the third generation of synchrotron X-ray sources has enabled to measure Mössbauer spectra in time domain [2, 3]. This was allowed by using high-resolution monochromators with the resolution of the ordered of several meV for 14,4 keV photons at the synchrotron facilities of *ESRF* (France)*, APS* (USA)*, Spring-8* (Japan), and since 2011 also at *PETRA III* (Germany). The aim of this work is to compare conventional transmission Mössbauer spectra in energy domain and nuclear forward scattered spectra in time domain. Using selected examples of typical types of conventional Mössbauer spectra of some calibration materials we demonstrate the resulting interferences between de-excitation transitions among nuclear splitted levels (*quantum beats*) as well as the effects of the samples thickness (*dynamical beats*).

2. Experimental Details

Samples of 11,5 μm (enriched to ^{57}Fe) and 25 μm thick stainless steel foils; sodium nitroprusside pressed powder; 12,5 μm thick bcc-Fe foil, and powder magnetite (Fe_3O_4) were measured by Mössbauer spectrometry in transmission geometry using ^{57}Co(Rh) source at the *Institute of Nuclear and Physical Engineering, FEI STU. Nuclear forward scattering* (*NFS*) time spectra were recorded at *ESRF* Grenoble and *DESY-Hamburg* at *PETRA III* facility. Samples were put at the beamline and incoming monochromatized 14,4 keV photons with the energy resolution of 6 meV (*ESRF)* and 5 meV (*PETRA III*) excited all ^{57}Fe nuclei at the same time. The de-excitation processes were detected in a 175 ns time window produced by a 16 electron bunch mode of the *ESRF* synchrotron radiation source and in a 192 ns time window provided by a 40 electron bunch mode at *PETRA III.* NFS spectra were measured at room temperature by avalanche photodiods.

Mössbauer and NFS spectra were evaluated with the CONFIT [4] and CONUSS [5] codes, respectively.

3. Results

Mössbauer spectrum of stainless steel in energy domain, which is shown in Fig. 1a, consists of a single absorption line. The corresponding NFS pattern in Fig. 1b exhibits a straight line in semi logarithmic scale that characterises a de-excitation process governed by simple exponential time decay. Experimental line width of the Mössbauer line is about 0.32 mm/s as determined from a calculated fit to the experimental data. Using this quantity as well as physical thickness of the sample (25 μm) and total content of Fe (55 % of natural Fe) we can determine the so-called effective thickness that is related to the overall amount of ^{57}Fe nuclei contained in the sample.

If we measure sample with high effective thickness, i.e. relatively high content of ^{57}Fe nuclei (note that natural Fe contains only 2.17 % of ^{57}Fe) the Mössbauer lines are significantly broadened. Mössbauer spectrum of stainless steel in Fig. 2a corresponds to a foil with a physical thickness of only 11 μm. Its line width is, however more than factor of two broader (0.73 mm/s) than in the previous case. This sample was prepared from Fe enriched to ^{57}Fe. The corresponding NFS pattern in Fig. 2b has changed drastically. It shows the so-called *dynamical beats* which indicate that self absorption is taking place in the sample due to high concentration of ^{57}Fe nuclei.

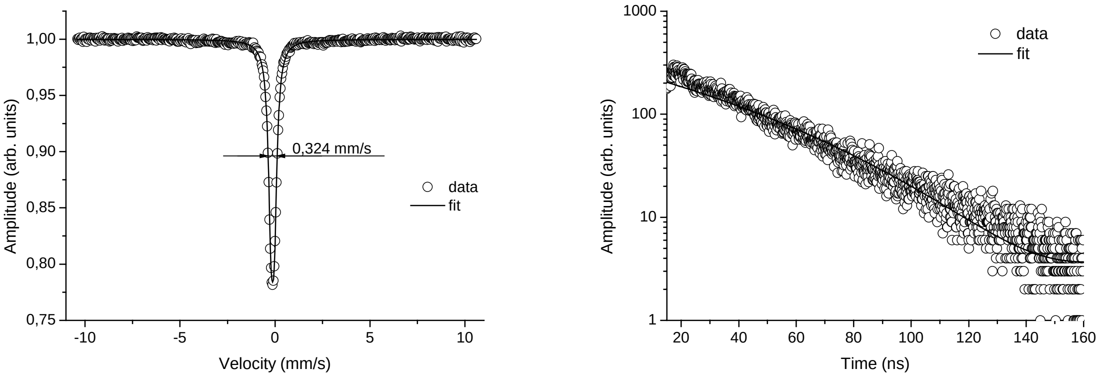
(a) (b)

Fig.1: *Stainless steel (25 μm): Mössbauer spectrum in energy domain (a) and corresponding NFS pattern in time domain (b).*

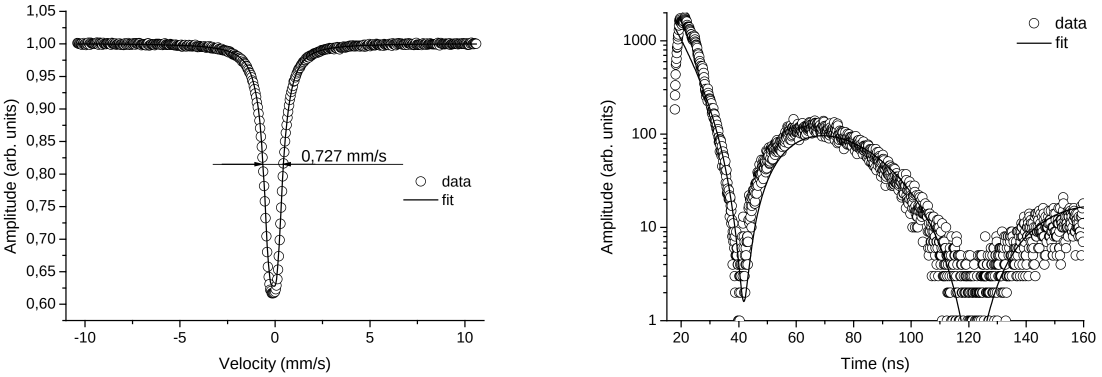
(a) (b)

Fig.2: *Stainless steel (11 μm, enriched to ^{57}Fe): Mössbauer spectrum in energy domain (a) and corresponding NFS pattern in time domain (b).*

Sodium nitroprusside is a calibration material for low velocity ranges. It exhibits pure electric quadrupole interactions that are characterized in the Mössbauer spectrum by a quadrupole doublet as seen in Fig. 3a. Corresponding NFS pattern in Fig. 3b shows similar features as *dynamical beats* in samples with high effective thickness. Here, however, the origin of beats is directly related to the hyperfine interactions and they are called *quantum beats*. They are quite distinct for electric quadrupole and magnetic dipole hyperfine interactions. Examples of the latter are shown in below.

Natural (metallic) iron (bcc-Fe) is the basic material for Mössbauer spectroscopy. The sextuplet of its spectral lines (sextet) in Fig. 4a is due to transitions among excited and ground nuclear states that are split because of magnetic dipolar interactions. Consequently, NFS pattern in Fig. 4b clearly indicates their presence by rapidly oscillating *quantum beats*.

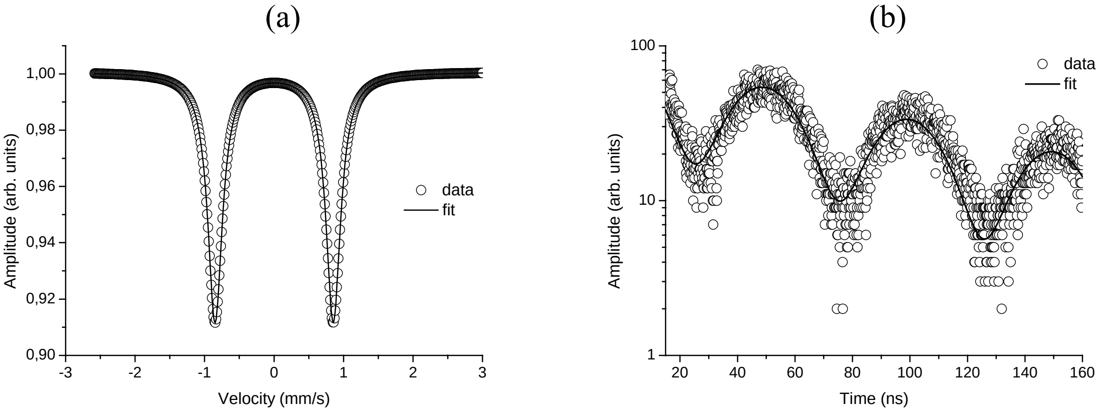

Fig. 3: *Sodium nitroprusside: Mössbauer spectrum in energy domain (a) and corresponding NFS pattern in time domain (b).*

In conventional Mössbauer spectrometry (energy domain), the absorption lines are recorded sequentially as the energy of gamma photons sweeps over the resonances for individual transmissions. On the other hand, pulsed structure of synchrotron radiation excites all nuclear transitions at the same time. Thus, in a NFS experiments the de-excitation photons constitute an interferogram in time domain.

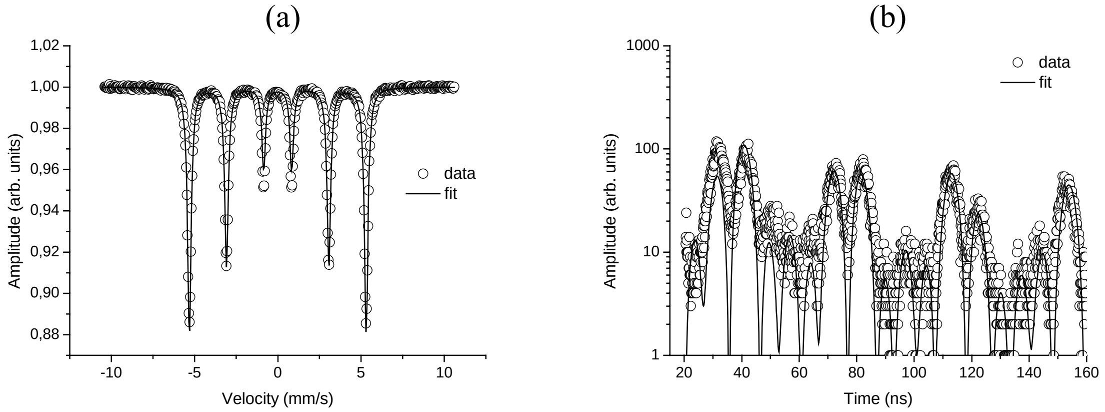

Fig. 4: *Natural iron (bcc- Fe): Mössbauer spectrum in energy domain (a) and corresponding NFS pattern in time domain (b).*

Quantum beats often show rather complicated patterns and that is why it is not easy to distinguish from how many components they consist of. Their number could be reconstructed only via appropriate evaluation software [5]. From this point of view, the amount of spectral components can be identified in energy domain Mössbauer spectra without major difficulties. A typical example is demonstrated in Fig. 5, where Mössbauer and NFS spectra of magnetite (Fe_3O_4) are presented. While two sextets are recognized in a conventional Mössbauer spectrum in Fig. 5a, the corresponding NFS pattern in Fig. 5b is similar to that from Fig. 4b. Nevertheless, high diagnostic potential of NFS and possibility to acquire the data from *in situ* experiments under extreme (high temperature, high pressure) makes this method attractive for materials research.

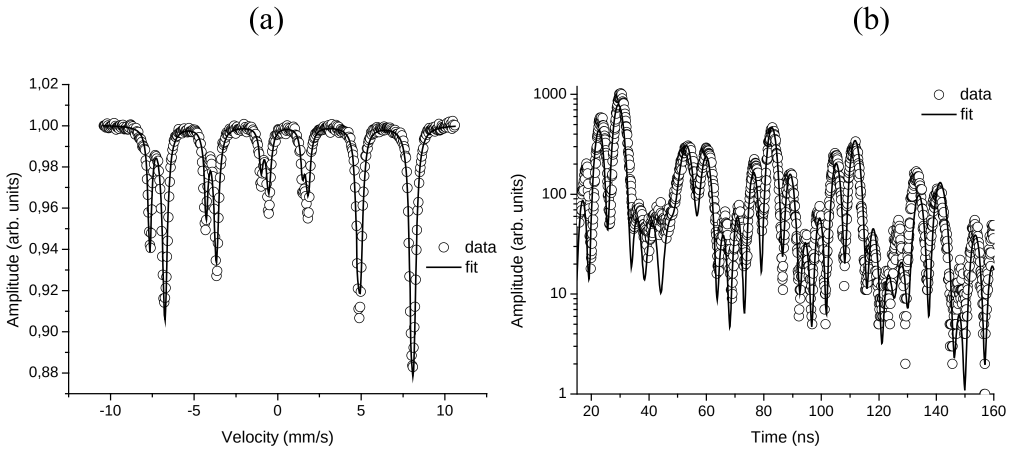

Fig. 5: *Magnetite (Fe_3O_4): Mössbauer spectrum in energy domain (a) and corresponding NFS pattern in time domain (b).*

4. Conclusion

Nuclear forward scattering is a powerful method for investigation of phase transformations and hyperfine fields in samples with small dimensions, like for example thin films, or nanoparticles measured under extreme conditions (high pressure produced by diamond anvil cells up to 100 GPa [6], high temperatures and magnetic field), or dynamical experiments. In this work we have presented nuclear forward scattering measurements in time domain for selected samples. The NFS patterns were compared with conventional Mössbauer spectra. Using typical siglet-, doublet- and sextet-like Mössbauer spectra we have presented the differences between quantum and dynamical beats in the corresponding NFS patterns.

Acknowledgement

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