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Improving detector signal processing with pulse height analysis in Mössbauer spectrometers

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Abstract A plenty of different programming techniques and instrument solutions are used in the development of Mössbauer spectrometers. Each of them should provide a faster spectrum accumulation process, increased productivity of measurements, decreased nonlinearity of the velocity scale, etc. The well known virtual instrumentation programming method has been used to design a computer-based Mössbauer spectrometer. Hardware solution was based on two commercially-available PCI modules produced by National Instruments Co. Virtual Mössbauer spectrometer is implemented by the graphical programming language LabVIEW 7 Express. This design environment allows to emulate the multichannel analyzer on the digital oscilloscope platform. This is a novel method based on Waveform Peak Detection function which allows detailed analysis of the acquired signal. The optimal treatment of the detector signal from various detector types is achieved by mathematical processing only. As a result, the possibility of an increase of signal/noise ratio is presented.

Keywords Mössbauer spectrometers · Waveform Peak Detection · Digital signal processing

1 Introduction

At present, various programming languages have been used for the software implementation of Mössbauer spectrometers. A few years ago, the first application of current well-known virtual instrumentation programming method in Mössbauer spectrometry was published [1–3]. The advanced use of the graphical programming language LabVIEW [4] in Mössbauer spectrometry has been described in [5], where the hardware solution is based on two commercially-available PXI or PCI modules produced by National Instruments Co [6]. Data acquisition is realized via NI 5102 digital oscilloscope that is used as a multichannel analyzer, and NI 5401 function generator which is used as a velocity

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generator. Virtual instrument (VI) working as a Mössbauer spectrometer is implemented by LabVIEW 7 Express, and this novel method is based on Waveform Peak Detection (WPkD) function that allows detailed analysis of the acquired signal in energy and time dimensions. This function is a software equivalent of the electronic pulse height analyzer device, so that the optimal treatment of the detector signal from the various detector types is achieved by mathematical processing only.

The purpose of this paper is to report on a new design of Mössbauer spectrometer, where the application of novel programming techniques in Mössbauer spectrometry is used. We experimentally show an increase in the signal/noise ratio by digital signal processing (DSP) before Mössbauer spectrum accumulation. The experimental results show an ability to use various detectors in the transmission and in the backscattering modes, and to tune proper signal acquisition from measuring the pulse height spectra.

2 Waveform peak detection and scintillation detectors

The negative pulses from the detector are acquired by a suitable sampling frequency, and locations and magnitudes of their amplitudes can be obtained. In Fig. 1, the signal, acquired from photomultiplier tube with an NaI:Tl scintillation crystal and a ^{57}Co source, and sampled by 5 MS/s sampling frequency, is depicted. The 8-bit binary representation of the voltage detector signal is used.

The amplitude selection process is controlled by several input parameters of WPkD (see Fig. 2). This function finds the location, magnitude of amplitude and the second derivatives of the peaks in the detector signal. The threshold and width input parameters serve as separation tools of true detector pulses from the noise. The threshold determinates the minimum value of the peak amplitude and the width determinates the minimum peak width according to a number of samples over the threshold.

The amplitude values of the detected peaks, which WPkD generates, are used for the pulse height analysis. The locations output contains positions of valid peaks found in the current block of data. The amplitude values of the detected peaks are used, together with the location values and proper low and high discrimination levels, for an accumulation of Mössbauer spectra. The procedure to set the optimum number of velocity channels was published in [5].

2.1 Transmission Mössbauer spectrometry with NaI:Tl and YAP:Ce scintillators

With this Mössbauer spectrometer, a proper use of NaI:Tl and YAP:Ce scintillators, unwinding from different activities of radioactive source, was tested. The negative pulses from these scintillators were acquired by 5 MS/s sampling frequency.

Two Mössbauer spectra of α-^{57}Fe$_2$O$_3$ (>90% enrichment of ^{57}Fe) were measured in the same geometry and with the same high voltage on the photomultiplier tube for a time of 1 h. For both detectors, the amplitude analysis was performed for proper setting of the discrimination levels. For NaI:Tl (thickness of 0.15 mm) scintillator, an input range of ±0.25 V with a discrimination window of 33.47 mV was used, and for YAP:Ce (thickness of 0.35 mm) scintillator, an input range of ±0.05 V with an discrimination window of 7.87 mV was used.

If the ^{57}Co source with an activity, A, of 50 mCi has been used, the resonant effect was the same, but the number of counts was twice as high as those observed for YAP:Ce. The same measurements were carried out with a source of 20 mCi activity. In Table 1, the

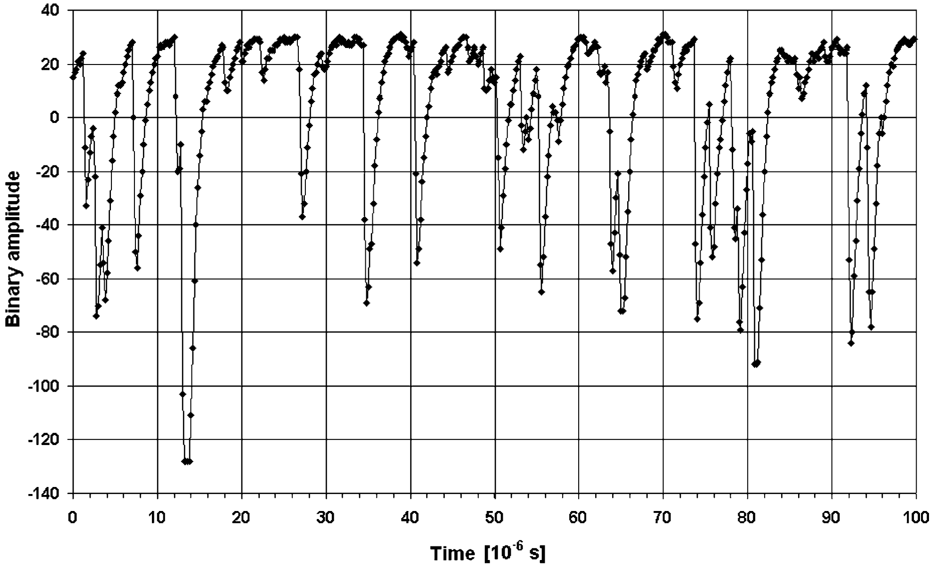
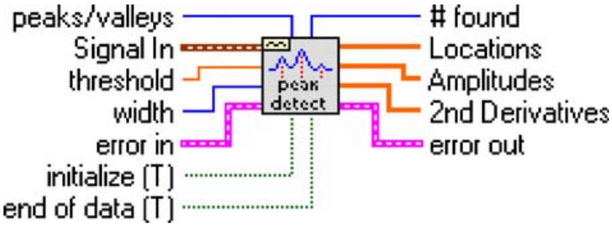


Fig. 1 The NaI:Tl detector signal acquired by 5 MS/s

Fig. 2 Icon of Waveform Peak Detection VI



results of these comparing measurements are listed. The productivity, Q, was calculated according to the equation in a form of

$$Q = \varepsilon^2 I_{\mathrm{out}}(\infty),$$

where $\varepsilon<1$ is the resonant effect, and $I_{\mathrm{out}}(\infty)$ is the background intensity in the Mössbauer spectrum [7].

Counts, being twice as high for YAP:Ce scintillator than for NaI:Tl scintillator, confirm that by using high activity sources, NaI:Tl scintillator is overloaded. YAP:Ce is able to registrate a higher activity because of its shorter light scintillation and short-impulse overlapping. From the Q_{50}/Q_{20} ratio, one can see how the productivity of the measurement increases with the high activity source with different scintillators.

2.2 Conversion X-ray Mössbauer spectrometry with NaI:Tl detector

Measuring the conversion X-ray Mössbauer spectrum represented the next motivation for showing the flexibility of the presented system. In this technique, the conversion 6.3 keV X-rays are detected in backscattering geometry. In our configuration, thanks to a high

Table 1 Results of comparing measurements with NaI:Tl and YAP:Ce scintillators

Parameter	*A* [mCi]	NaI:Tl	YAP:Ce
Background	50	143,176	284,631
	20	80,983	145,682
Resonant effect [%]	50	26.98	27.25
	20	28.07	26.38
Productivity Q	50	10,422	21,135
	20	6,381	10,138
Ratio Q_{50}/Q_{20}		1.6	2.1

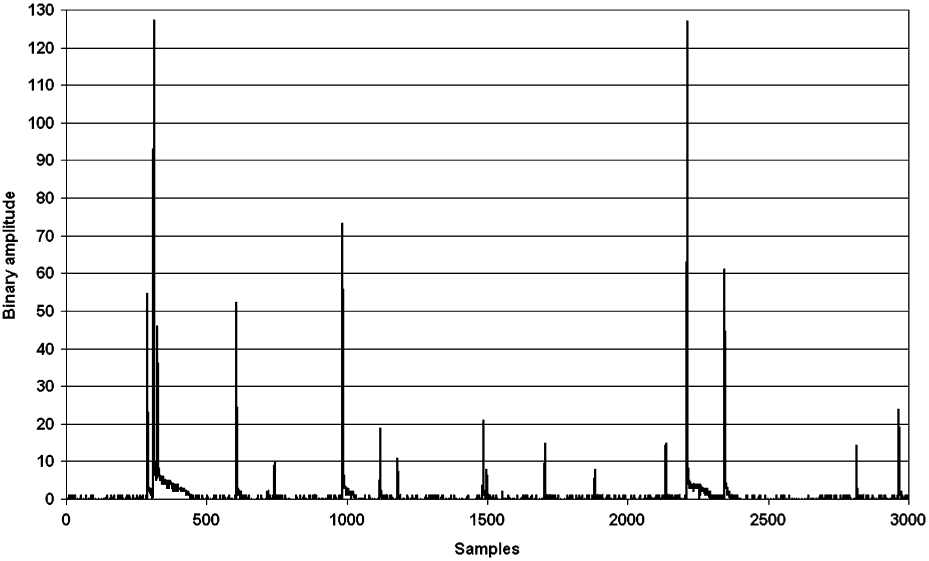


Fig. 3 Gas-flow proportional counter (CEMS mode) signal

activity source (50 mCi) and thin calibration sample, it was possible to registrate spectrum in the transmission mode. The measurement was performed with NaI:Tl scintillator for 1 h with a resonant effect of 51%.

3 WPkD and gas-flow conversion electron detectors

It is also possible to measure conversion electron Mössbauer spectra with presented spectrometer system. In this situation, we used a gas-filled conversion electron counter (90%He + 10%CH_4). The pulse-height spectra of the acquired signal from this counter show a typical sharp peak in the number of low energy electrons, but there are counted noise impulses too. The electron energy depends also on the depth from which the electron is emitted.

The typical signal, acquired on the output of this proportional counter, is shown in Fig. 3. This signal was acquired by 1 MS/s sampling frequency, as this value is sufficient for a quality data processing without a loss of any signal information. The acquiring with a higher sampling frequency is possible, but without obtaining any new information on the signal.

In Fig. 3, it is seen that impulses are, in fact, a combination of two components, high-fast (electron) and low-slow (ions) ones.

Furthermore, when applying WPkD in this design, we arrive at the next disadvantage. In Fig. 4a), a detail of the detector signal is shown, and in Fig 4b), amplitudes and locations of founded peaks, declared as a valid by WPkD, are shown. One can see that there are "false" impulses on the bottom of high-energy impulses that can be of higher amplitudes than some impulses for low-energy electrons. It negatively influences the quality of the Mössbauer spectrum when they are counted into the spectrum.

The process of reducing these artificial peaks in the acquired signal is based on the use of the second derivative values (WPkD output) of each founded peak (see the inset of Fig. 4c). In this situation, the magnitude of these "false-peak" second derivatives does not reach a value of −1.

The software "noise-reducing" procedure was created as a SubVI (subprogram in main VI), and used before the Mössbauer spectrum accumulation. The effects and results of using this procedure are depicted in Fig. 4d), where only true peaks, belonging to the electron detection are present in the final signal. The −1 value as a reducing parameter can be too high sometime and consequently, some true impulses can be deleted.

Various pulse-height spectra of the detector signal, acquired with a different width of WPkD and the second derivative parameters, are shown in Fig. 5. The value of threshold of WPkD was set to 1 in all cases.

In order to choose the best combination of parameters, several Mössbauer spectra, with the same level of background ($I_{\mathrm{out}}(\infty)\approx 2.5\times 10^6$) were measured, see Fig. 6.

The spectra were measured with different width parameter (4, 5, and 6) as an input parameter for WPkD, and different second derivative parameters, used in software procedure before spectra accumulation, in energy window from 5 to 110 channel. Each spectrum was fitted by Recoil 1.0 software to find spectra with the highest statistical quality. The results of the fitting procedures are presented in Table 2, where the values of HWHM (half width at half of maximum) for w_3 of the spectral lines 3 and 4 are also listed.

It can be seen that the time of measuring increases with decreasing value of the second derivative. This is in accordance with the shape of the amplitude spectra in Fig. 5 for all widths, where the total number of counts decreases when low-energy impulses are not counted after the reducing process is applied. For reaching the same level of the background (approximately the same statistical quality of the spectrum), we thus need a longer measuring time.

From comparison of χ^2 values (χ^2 represent parameter of correspondence of the fit with Lorentz line shape), we can conclude that with using the second derivative parameter as a noise-reducing tool, χ^2 value decreases with decreasing value of the second derivative.

The Mössbauer spectra were recorded into 750 velocity channels; it means that one channel has the width of about 67 μs. The width of the fast component of the typical impulse in the detector signal is from 20 to 50 μs, and thus, every impulse is correctly recorded into the relevant velocity channel. As the slow component of the impulse can be about 1 ms long and the false impulses generated by WPkD are counted into following channels, their intensities are higher than without this counting, thus increasing their widths (HWHMs). This fact is seen in Table 2, where, based on our noise-reducing process, w_3 value decreases, and thus a better resolution of the sub-spectra is possible. However, with

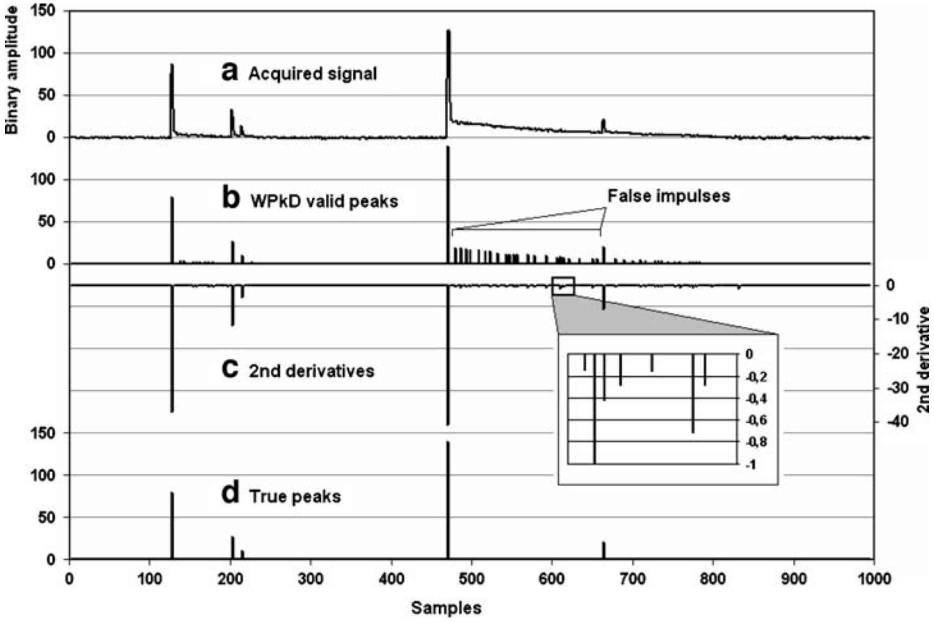


Fig. 4 Detail of the CEMS signal with overlap impulses

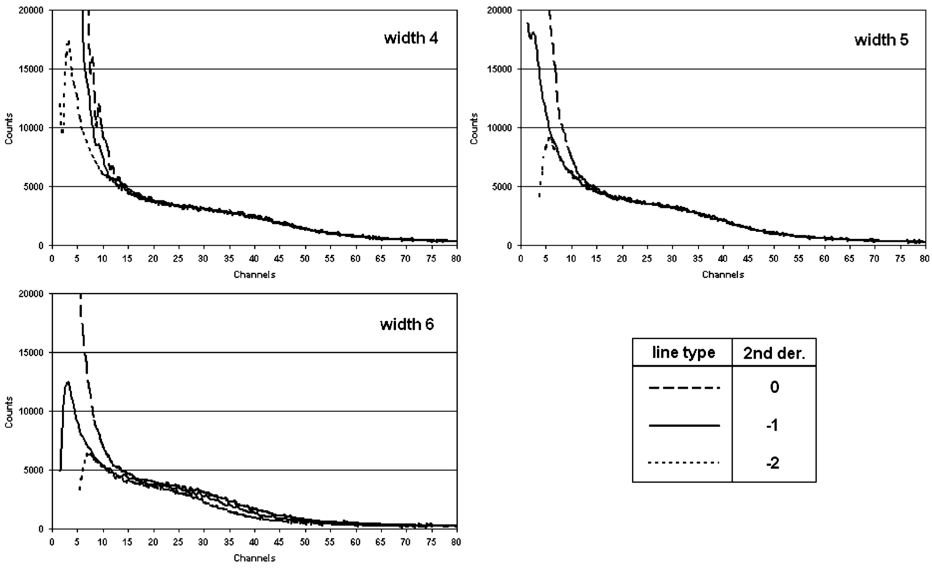


Fig. 5 Pulse-height spectra for various width and the second derivative parameters

Fig. 6 Measured Mössbauer spectrum of α-Fe sample

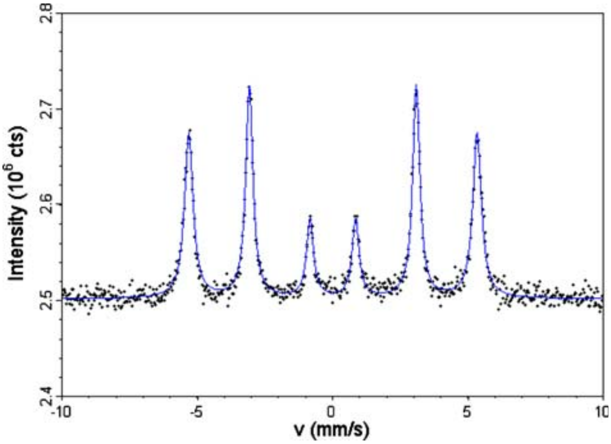


Table 2 Measurements with various combinations of parameters

i	Width	Window	2nd	ε [%]	Q	Time [h]	$\chi 2$	w_3 [mm/s]
1	4	5–110	0	8.4	17,693	34	59.1	0.21
2	4	5–110	−1	7.1	12,626	44	25.7	0.15
3	4	5–110	−2	7.3	13,283	51	19.1	0.16
4	5	5–110	0	7.6	14,451	43	27.9	0.16
5	5	5–110	−1	7.6	14,632	44	19.2	0.14
6	5	5–110	−2	7.3	13,271	65	20.1	0.15
7	6	5–110	0	7.7	14,920	46	24.7	0.16
8	6	5–110	−1	7.7	14,857	50	18.6	0.13
9	6	5–110	−2	7.8	15,236	54	19.5	0.15

too high value of the reducing parameter (−2 in this case), χ^2 values and widths w_3 increases and useful signal is deleted.

4 Conclusion

The novel design, significantly improving the efficiency of the Mössbauer measurements and the precision of their results, was constructed. Virtual Mössbauer spectrometer, based on LabVIEW graphical programming environment and Waveform peak detection function, is new tool in Mössbauer spectroscopy and offers various combinations of detectors and measurement geometries.

The use of this novel embodiment of Mössbauer spectrometer, based on WPkD, offers a further combination of input parameters for a better signal acquisition, actually depending on the situation. One can, for instance, use a higher sampling frequency for faster detectors, which involve number of samples in one impulse in acquired signal, or change the width and threshold as a WPkD input parameters for moving the level for noise reduction in the signal. It is possible to registrate more quality spectra with a higher sampling frequency. However, one has to bear in mind that this results in too much data for DSP analyzing and therefore, a higher computation time.

Disadvantage of this solution lies on the fact that the performance of such a spectrometer is given by the performance of the computer used (PC or PXI). Detecting the gamma-ray with this system, there are no evident troubles, however, when used for detecting the conversion electrons, there are naturally false impulses generated and then other DSP routine is necessary to be developed.

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