The Mössbauer spectrometer MsAa-4

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Abstract. A description of the Mössbauer spectrometer MsAa-4 is given. The spectrometer has a modular design and it could be used as the γ-ray spectrometer as well. It has almost entirely digital design to assure stability and repeatability. It is powered via the rechargeable battery to make it immune to the power failures. All settings are performed remotely over the Ethernet link used also to transfer data and spectrometer functions. External CAN bus could be used to attach auxiliary equipment. The status of the external equipment could be used to control spectrometer functions. Internal digital oscilloscope monitors vital functions of the spectrometer over the Ethernet link. Driving software operates under Microsoft Windows® systems and it is fully compatible with the Mosgraf-2009 data processing suite.

Key words: γ-ray spectroscopy • Mössbauer spectroscopy • spectrometers

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Introduction

Several Mössbauer spectrometers are available commercially [6] and much more γ-ray spectrometers. More advanced Mössbauer spectrometers could be used as the medium or high class γ-ray spectrometers. The MsAa-4 Mössbauer spectrometer is a continuation of the previously made MsAa-x systems since the MsAa-1 design [9] – in particular the latest versions of the MsAa-3 high precision spectrometer [7]. It is a typical laboratory spectrometer of general purpose and of highest class either as the γ-ray spectrometer or as the Mössbauer spectrometer. The paper concentrates on the fundamental features of this spectrometer, while the earlier design stages are described in [3, 4]. Mössbauer spectra are collected in the multi-scaler mode (with the possible real time readdressing keeping track of the channel opening statistics) as the most reliable mode. There is a long time tradition of making Mössbauer spectrometers in Kraków, as the first Polish Mössbauer spectrometer was completed here in 1961 [1].

Functions of the system components

The main bin contains power supply for the essential parts of the system and data communication lines between boards and to the external equipment. The power supply is operated from the rechargeable battery the latter charged by a simple power supply connected to the standard power outlet. The bin accepts specialized boards. Currently there are three types of boards available, i.e., the nuclear board, a transducer board and the laser calibration board. The main bin is equipped with the external CAN link to the auxiliary equipment. The CAN units connecting to the external equipment are standardized. They are powered from the main bin via the CAN link. The nuclear board has the Ethernet port for the communication with the Ethernet/Internet network over the standard supplied category 5e cable. The spectrometer setup and data exchange is performed entirely via this link by using specialized software compatible with the Microsoft Windows® systems.

Nuclear board, detector heads and detectors

The nuclear board connects to the detector head powered from this board via the specialized cable used to exchange signals as well. It can accept also signals from the external nuclear electronics – just beyond the amplifier. The latter port is designed to accept signals from Si or Ge detectors equipped with standard or optically coupled amplifiers (miniature BNC 50 Ω with an adapter to the standard BNC). The board is equipped with an Ethernet link as above mentioned – 100 Mbit/s. It is used to store γ-spectrum of the detector and up to four multi-scaler Mössbauer spectra in the pre-selected windows of the γ-spectrum. Typical data banks have 4096 channels each with 32-bits per channel. There are two detector heads currently available. The first one accepts scintillation counters and it is equipped with the high voltage power supply and amplifier. The second one accepts either proportional or CEMS gas flow counters, and it has the high voltage power supply and amplifier, too. Both high voltage power supplies could provide up to +2000 V controlled in fine steps. All settings are performed from the nuclear board. Additional interchangeable board mounted directly on the nuclear board allows for significant flexibility while switching between various detectors. A detector signal is digitized with the 40 MHz frequency into 12-bits on the nuclear board. Such solution is extremely stable against the noise and it makes single channel analyzers obsolete. In order to operate the γ-ray spectrometer one can dispense with the remaining boards and systems.

Transducer board and transducer

A transducer board connects via the specialized cable to the transducer of the Mössbauer spectrometer. A separate multi-pin front connector could be used to extract (or set) various signals. The reference function is downloaded to this board via the Ethernet link of the nuclear board. The reference function is stored in 16 384 channels per full cycle with the 18-bit precision. A provision to load and use the correction function in the same number of channels per full cycle and with the 8-bit precision is provided. A small interchangeable board on the transducer board adds extra flexibility in the case of non-standard setups. The amplitude of the velocity and repetition frequency could be set in very fine steps. The sensitivity of the transducer pick-up coil is about 50 $mV \cdot s \cdot mm^{-1}$. A transducer is equipped with the position sensor allowing for compensation of the constant external forces. A bipolar compensation system is enclosed within the transducer board. A transducer utilizes hollow central tube of the 8-mm diameter. It serves as the access of the calibration optical beam to the prism just behind the source, and it might be very useful for the time domain setups on the synchrotrons utilizing motion of the one of the targets [2]. A transducer could be operated either horizontally or vertically – with the γ-ray beam outgoing either up or down. It could be enclosed as well within the cryostat space. A transducer is equipped with a γ-ray beam collimator mounted within the detachable protection tube. The maximum velocity with the standard springs could be approximately up to ±200 mm/s. Basic setup of the Mössbauer spectrometer consists of the nuclear and transducer systems.

Laser calibration board and laser calibration system

The laser calibration board is responsible for the operation of the laser velocity calibration system based on the Michelson-Morley interferometer [8, 10, 11]. The interferometer equipped with a metrological quality He-Ne laser connects directly to the back of the transducer, while the moving corner prism is attached just behind the source mounted on the transducer. The laser is operated from the dedicated power supply over the high voltage cable. This power supply is powered from the main battery and controlled via the specialized cable from the laser calibration board (RJ-11 connector on the laser calibration board). The interferometer is connected to the laser calibration board via the dedicated cable used to power the electronics of the interferometer and to exchange signals. A provision is made for several modes of calibration either hardware or software implemented covering all possible velocity ranges [5]. In principle, the optical beam could be passed to the room temperature area of the cryostat with the transducer enclosed within the cryostat by means of the vacuum tight (tilted) optical window. Spatially separated optical beams and the beam expander are used to assure extreme mechanical stability. The calibration data are stored on the laser calibration board. The board is accessible via the Ethernet link of the nuclear board. This system improves significantly performance of the spectrometer in particular for velocity ranges far beyond the typical ranges accessible by means of the standard absorbers.

Standard CAN unit

A standard CAN unit is operated and powered from the main bin. It is equipped with two connectors RJ-9 accepting three pre-selected voltages with the 12-bit resolution each and providing one 1-bit sensing port. Additional RJ-11 connector is set as the RS-232 port to accept and send signals to the external equipment. The spectrometer status could be changed depending on the signals from the CAN units. Practically unlimited number of such units could be connected to the main bin over distances covering a large laboratory room.

Other features

Essential signals within the spectrometer could be monitored with the help of the on-board digital oscilloscope having 12-bit vertical precision. The oscilloscope connects via the Ethernet/Internet link to the external computer running driving software. Each board (and the main bin) has own processor for the great flexibility of possible arrangements. The spectrometer is a stand-alone system within the Ethernet/Internet network operated to the full extent from remote. The spectrometer settings could be stored together with the data for the easy restoration of the previous conditions. All settings and data could be stored in the non-volatile spectrometer memory under emergency conditions. Many important signals are available on the respective pins of the particular board front connectors to either detector head or to the transducer. A transducer board is equipped for this purpose with a separate multi-pin front connector. Some of these signals are rather accepted by the spectrometer than delivered to the external systems. Standard setup contains rigid bench to mount transducer/laser system and detector head. All necessary accessories like cables and connectors are provided.

The Mosgraf-2009 data processing suite [5, 12] is provided with the Mössbauer spectrometer setups. The suite is fully compatible with the driving software of the spectrometer and it is designed to be operable under Microsoft Windows® systems (since the Windows-98 second edition inclusive) within the .NET Framework 3.5 (or higher) 32- or 64-bit environment. The suite requires installation on the particular computer, while the spectrometer driving system could be operated also without prior installation. A general view of the complete MsAa-4 spectrometer is shown in Fig. 1, while the view of the front panel of the main bin with all boards inserted is shown in Fig. 2.

Fig. 1. A general view of the complete MsAa-4 spectrometer with a proportional detector head and a Kr-filled proportional counter. The laser high voltage cable is not connected as well as the CAN unit shown. This version is operated directly from mains.

Fig. 2. A view of the front panel of the main bin with all the boards inserted, i.e., Ncl4 – nuclear board, Lsb4 – laser calibration board, and Vbd4 – transducer board. A lightning diode on the front panel of each board is an indicator of the operational conditions. The push-button marked as Fn and located close to the lightning diode enables software pre-selected internal functions.

Results of some tests

The best test of the Mössbauer spectrometer could be performed by collection of the spectrum under precisely defined conditions. Results of such test are reported below. A commercial high quality source of ^{57}Co(Rh) has been applied at room temperature. The source consisted of a Rh-foil having 6 μm thickness, nominal activity of 25 mCi, and an active diameter of 8 mm. The source was moved in the laboratory frame under ambient external pressure, and in the null external magnetic field. The absorber was placed perpendicular to the beam axis at room temperature. It was made of the α-Fe foil having natural isotope composition and 25 μm thickness (very high chemical purity α-Fe was used), diameter of 16 mm, and diameter exposed to the beam of 12 mm. The foil was obtained from AlfaAesar and used without further treatment. The absorber remained under ambient external pressure and in the null external magnetic field. Total (effective) beam divergence angle amounted to 11.70°. The nominal velocity range of the spectrometer was set to ±12 mm/s with 4096 channels per full cycle, and with the triangular reference function having 3% round-corner smoothing on edges. The velocity increased in the second half of the cycle. A repetition frequency of the spectrometer was set to 5.722 Hz. The self and standard laser calibrations were applied. Folded spectrum has been fitted within the standard transmission integral approximation. Results are summarized in Table 1 and in Fig. 3. The parameter B_0 stands for the number of counts (per folded data channel) far off the resonance. The parameter f_S/λ stands for the effective recoilless fraction of the source with the parameter λ describing background under resonant line within accepted window of the γ-ray spectrum. A photo-peak of the 14.41-keV pure M1 transition was chosen and a Kr-filled proportional counter was used. This parameter could be estimated from the γ-ray spectrum as the ratio of the total signal in the window to the signal due to the particular nuclear transition and it amounts here to $\lambda = 1.2099(3)$. Hence, the recoilless fraction of the source appears to be $f_S = 0.74(4)$. The parameter Γ_S stands for the source line width (broadened mainly due to the beam divergence). The parameter t_A denotes

Table 1. Fitted parameters of the test spectrum. The number of degrees of freedom amounts to 2039, as the number of the folded data channels equals 2047. The quality of the fit is measured by the χ^2 per degree of freedom and MISFIT criterion. Standard deviations are shown in parentheses. This convention applies for the whole paper

Parameter	Number	Value
B_0 (counts)	1	178 270(12)
f_S/λ	2	0.61(3)
Γ_S (mm/s)	3	0.204(7)
t_A	4	8.9532 – fixed
Γ_A (mm/s)	5	0.100(5)
B (T)	6	32.975(2)
S (mm/s)	7	–0.1100(2)
$g_{11}^{(111)}$	8	0.711(2)
$ecQ_eV_{zz}/4I_e(2I_e - 1)E_0$ (mm/s)	9	+0.00074(8)
χ^2 per degree of freedom		0.887
MISFIT (%)		0.020(5)

dimensionless resonant thickness of the absorber (kept fixed in the final fit to assure better numerical stability). The parameter B stands for the hyperfine field (value) acting on the resonant nuclei in the absorber, while the parameter S denotes total shift of the absorber resonant cross-section vs. source. The parameter $g_{11}^{(111)}$ accounts for the partial magnetization of the absorber within the plane of the foil [13, 14]. Finally, in order to improve the quality of the fit it was necessary to add some small axially symmetric electric quadrupole interaction with the main component of the electric field gradient tensor aligned with the hyperfine magnetic field. Quadrupole coupling constant, having the following form $A = [ecQ_eV_{zz}]/[4I_e(2I_e - 1)E_0]$ has been fitted to the data. Here, the symbol e stands for the positive elementary charge. The symbol c denotes the speed of light in vacuum, while the symbol Q_e stands for the (positive) spectroscopic quadrupole moment of the first excited nuclear state in ^{57}Fe. The symbol V_{zz} denotes the principal component of the electric field gradient tensor (the maximum component in the absolute terms), while the symbol $I_e = 3/2$ stands for the nuclear spin of the above-mentioned state. Finally, the symbol E_0 denotes resonant transition energy.

The above electric quadrupole interaction leads here to the splitting $\Delta = 6|A| = 0.0044(5)$ mm/s, i.e., much smaller than the natural line width of 0.096 mm/s. The presence of the dominant dipole magnetic interaction is essential to see this small effect. The splitting amounts to about 0.00018 of the total velocity range of about 24 mm/s. Hence, one can conclude that the linearity of the spectrometer is better than the factor of 0.00009 as the shift of the particular lines in the presence of magnetic interaction amounts to ½Δ.

The phase α-Fe has BCC structure and therefore the hyperfine electric quadrupole interaction possibly seen in the excited nuclear resonant state should vanish. This phase is extremely soft magnetically provided it is of high purity. However, some remnants of the orbital magnetism remain as the magnetic moment is due to the electronic spin polarization of the 3d band. Hence, some small magnetic anisotropy remains with the easy axis of magnetization being one of the principal crystal axes. Magnetic moment and subsequently the hyperfine field are aligned with one of the principal crystal axes. The same spurious spin-orbit coupling mechanism leads to the magnetostriction, and for the field (moment) aligned with the main crystal axis one observes elongation of the lattice constant along this axis (shortening in the perpendicular directions). Hence, the cubic symmetry is broken and one can expect some axially symmetric electric field gradient tensor on the iron nuclei (in the null external magnetic field). The main component of this tensor is aligned with the hyperfine magnetic field, and it has to be positive leading to the positive coupling constant as observed. The electric quadrupole interaction is enhanced on the iron nucleus by the (isotropic) polarization of the itinerant and core electrons. This interaction vanishes above the magnetic transition temperature, and it is extremely small otherwise. It could be seen in the sample having negligible strain and it strongly depends on the strain, i.e., on the thermal history of the sample. The question is: could it be observed by the transmission Mössbauer spectroscopy for the room temperature sample and for the resonant transition between the ground and first excited state of the ^{57}Fe nucleus? The answer is yes, provided the highest quality spectrometer is used, and the source is very good, too.

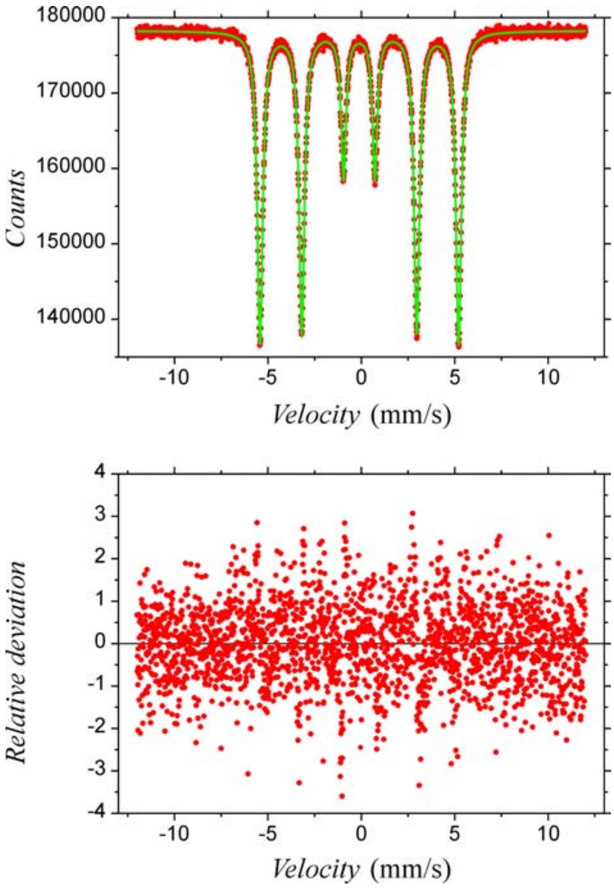

Fig. 3. Spectrum of the α-Fe foil with a calculated spectrum shape based on the fitted parameters. The relative residual deviations are shown in the lower part.

Conclusions

The MsAa-4 system is a high-end laboratory class fully digital and completely remotely controlled Mössbauer spectrometer of the general purpose. Actual functions could be easily changed by downloading/replacing on-board software. The system is fully rechargeable battery operated making it immune to the power failures. Downloadable reference functions allow for operation in various modes, e.g., for the constant acceleration or constant velocity modes. The laser velocity calibration system makes it independent of the available standard absorbers (sources). The external CAN bus is a flexible way to attach external sensors or actuators having feedback on the fundamental spectrometer functions. The spectrometer could be set as the independent unit of the Internet network, and hence it could be accessed from any point worldwide via the customized software.

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