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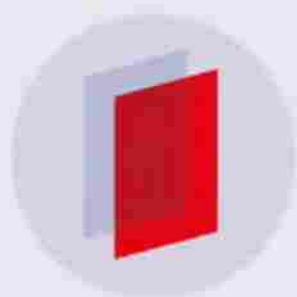 M Nakada, J Wang, T Kitazawa et al.

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Optimal configuration for Programmable Mössbauer Experiments

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Abstract. Based on channel independency of recently developed Mössbauer instrumentation an approximation to optimal configuration of experiments is presented. The analysis relies on the presumption that all the available channels of the spectrum are not equally efficient for a given experimental application. A quantification of this concept is presented and a method for different channel layout comparison is proposed. The optimization of recorded spectra is important in dynamic experiments where efficiency in data taking imposes feasibility limits as well as in static applications as a way of reducing experimental time.

1. Introduction

Mössbauer effect spectra are usually taken in constant acceleration, constant velocity or sinusoidal modes. Recently a Programmable Velocity Mode (PVM)[1], based on a constant velocity mode strategy, was introduced as an alternative to the conventional modes. PVM opens new possibilities on the art of Mössbauer experimentation. The ability of measuring different spectral zones with different time weights is one of the important applications of this technique. Another recently developed application is the tracking of interest regions as they change due to the variation of an external and controlled parameter [2], technique that we have called Mössbauer Line Tracking (MLT).

In both cases there is a region of interest that is measured and further analyzed. Up to now, with the conventional spectrometers that use uniform velocities distribution, the question of which is the best set of velocities (sampling distribution) that most efficiently acquire a spectrum region was inconsequent. However with the liberty of freely choosing velocities and time of acquisition, this question becomes important if we are looking to improve the experiments. Furthermore in the case of MLT the success of the experiments strongly depends on a careful choice of the velocities.

In the following, we will refer as Region Of Interest (ROI) to the set of velocity positions and acquisition times,

$$\{(v_i, t_i), i = 1, .., N\}$$

where the measurements will be done. In both static and dynamic experiments the optimal ROI selection is constrained to the total time per ROI ($T_{ROI} = \sum t_i$). In the case of dynamic experiments is obvious that T_{ROI} is defined by the rate of change on the spectral profile. In

1

fact, as fast are the spectrum changes as short must T_{ROI} be in order to reproduce quasi-static information of the spectrum.

MLT procedure exploits the programmable attribute of the PVM. The objective of the technique is to apply a dynamic ROI to follow an evolving spectrum using a feedback algorithm. The efficiency of this algorithm might depend on different factors and is not part of the discussion of this work. However the knowledge on the goodness of the ROI selection is a valuable tool at the moment of designing the tracking algorithm.

2. Theory
2.1. Variation of measured parameters due to statistical dispersion
Let $F(v, t; p_1^0, p_2^0, ...)$ be the Intensity of the Transmission Spectrum (ITS) at a velocity v when the acquisition time is t. The characteristic parameters of the absorption profile (the centers of the lines, the FWHM, etc.) are represented by the quantities $p_1^0, p_2^0, ...$ With the supra index 0 we are indicating that they are the true parameters.

For each v_i and t_i of the ROI a counting y_i will be measured, according to a Poisson distribution with mean $F(v_i, t_i; p_1^0, p_2^0, ...)$. In the following the reduced notation: $F_i(p_1, p_2, \cdots) \equiv F(v_i, t_i; p_1, p_2, \cdots)$, where the index i indicates the ROI element, will be used.

For a given set of experimental data $\{(v_i, t_i, y_i), i = 1, ..., N\}$ the spectrum parameters p are estimated by minimizing the Chi-Square function[3],

$$\chi^2(p_1, p_2, ..p_n) = \sum_{i=1}^{N} \left(\frac{y_i - F_i(p_1, p_2, ...)}{\sigma_i} \right)^2 \tag{1}$$

where σ_i is the variance of the measurement y_i. Since the measurements follow a Poisson distribution, the square variance coincides with the mean value, $\sigma_i^2 = F_i(p_1^0, p_2^0, ...)$.

We will simplify the analysis and suppose that there is an unique unknown parameter, p. The corresponding results can be used as a first approximation to understand the dependence of the ROI selection on the searched parameter. The parameter p that minimizes the Chi-Square function must satisfy,

$$\sum_{i=1}^{N} \frac{1}{\sigma_i^2} (y_i - F_i(p)) F_i'(p) = 0 \tag{2}$$

where F_i' denotes $\frac{\partial F_i}{\partial p}$. Assuming small differences between the data and the correspoding ITS, the expected change in p, $\Delta p = p - p^0$, should also be small. Therefore both F_i and F_i' could be approximated at first order around p^0,

$$\sum_{i=1}^{N} \frac{1}{\sigma_i^2} \left(y_i - F_i(p^0) - F_i'(p^0)\Delta p \right) \left(F_i'(p^0) + F_i''(p^0)\Delta p \right) = 0 \tag{3}$$

Neglecting terms with square orders on Δp, this quantity can be isolated:

$$\Delta p = \frac{\sum \frac{1}{\sigma_i^2} \Delta y_i F_i'(p^0)}{\sum \frac{1}{\sigma_i^2} (F_i'(p^0)^2 - \Delta y_i F_i''(p^0))} \simeq \frac{\sum \frac{1}{\sigma_i^2} \Delta y_i F_i'(p^0)}{\sum \frac{1}{\sigma_i^2} F_i'(p^0)^2} \tag{4}$$

where for the last approximation it was assumed that in almost all the cases $\sum F_i'(p^0)^2 \gg \sum \Delta y_i F_i''(p^0)$. In the following we work under this hypothesis.

The last equation gives the change on the estimated parameter due to the fact that the measured values y_i do not exactly match the transmission values.
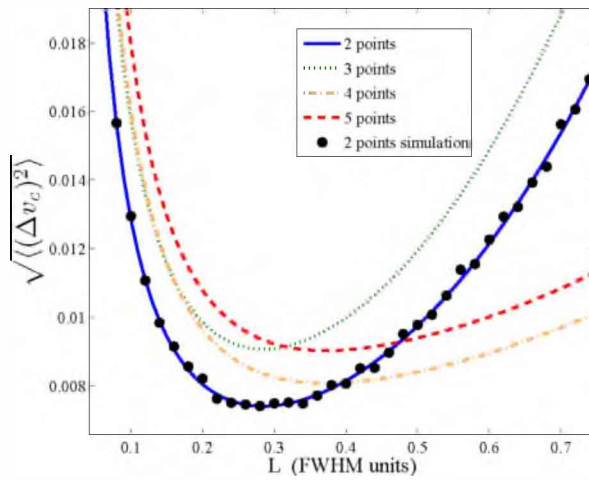
Figure 1. Variance of the center estimation $\sqrt{\langle (v_c - v_c^0)^2 \rangle}$ for different distributed ROIs. The curves correspond to 2, 3, 4 and 5 points ROIs, all with $T_{ROI} = 100$ s, a counting rate of 10^4 counts/s and velocities witin $-L$ and L. All the curves were calcluted with equation 5. The points correspond to 3000 events simulations for the case of the two points ROI.

2.2. Goodness of the selected ROI

The goodness of the selected ROI is given by the confidence on the estimation of the corresponding parameter. The mean square $\langle (\Delta p)^2 \rangle$ is a measure of this confidence. Note that the mean value is made over a the set of results obtained from infinity identical experiments. In that sense $\langle \Delta y_i \rangle = 0$ and $\langle (\Delta y_i)^2 \rangle = F_i(p^0)$. Whith these identities the mean square value over eq. 4 results on,

$$\langle (\Delta p)^2 \rangle = \frac{1}{\sum_{i=1}^{N} \frac{F_i'(p^0)^2}{\sigma_i^2}} = \frac{1}{\sum_{i=1}^{N} \frac{F_i'(p^0)^2}{F_i(p^0)}} \tag{5}$$

Therefore the goodness of the ROIs can be measured with the last expression. To minimize $\langle (\Delta p)^2 \rangle$ and increase the ROI goodness the velocities must be selected near the maximum of derivate respect to the unkown parameter.

The last expression is in good agreement with results obtained by simulations. Figure 1 shows a comparison of the root square mean value obtained by both methodologies.

3. Applications

It is well known that the theoretical shape of a Mössbauer absorption line for thin absorbers is Lorentzian. Moreover the Lorentzian shape is accepted to be a good approximation for thick absorbers. Then, as a practical example, we will apply the presented results to this case. The normalized absorption line is given by,

$$f(v_i; \Gamma, ef, v_c) = 1 - \frac{ef}{4\frac{(v - v_c)^2}{\Gamma^2} + 1} \tag{6}$$

where Γ is the full width at medium height (FWHM), ef is the maximum of absorption, while v_c is the center of the Lorentzian. The transmission function depending on the acquisition time is related with the last trough the next equality,

$$F(v_i, t_i; \Gamma, ef, v_c) = rt_i f(v_i; \Gamma, ef, v_c) \tag{7}$$

where r is the counting rate at background velocities (where the Mössbauer absorption is null).

In the Figure 1 the $\langle (\Delta v_c)^2 \rangle$ was calculated for different uniform ROIs centered on the true center. All the presented results correspond to a time per ROI of 100 s, a counting rate of 10000 counts/s and an absorption effect of 0.1. The figure shows that the best uniform ROI to obtain

the center corresponds to two points positioned on $\pm 0.28\Gamma$. These positions correspond to the points of maximum derivate of the ITS respect to the paramter v_c.

But the maximum derivative velocity position depends on the involved parameter. When looking for v_c the positions of maximum derivate are $v_c \pm \frac{1}{2\sqrt{3}}\Gamma \sim v_c \pm 0.28\Gamma$. If the unkown parameter is Γ, the maximum derivative velocities are $v_c \pm \frac{\Gamma}{2}$, while if ef is unkown then the velocities at the center of the absorption line are more efficient.

The two points ROI has been implemented in a real MLT experiement [2] with succesfull results. In the cited case, the counting rate and the effect were so low that a fast ROI was required to assure a good tracking. ITS parameters different from line center (Γ, ef and counting rate r) were carefully measured with a previous conventional Mössbauer spectrum and assumed constant during the tracking.

MLT is a technique that can benefit from this kind of analysis. The tracking is more likely to succeed if the ROI is selected within these criteria. But the results are not restricted to better finding the center of an absorption line. Any variable of the spectrum can be considered as target of the analysis and as a result an optimum ROI can be selected to better determine it.

4. Conclusions

The possibility of individually selecting energy and live time for each channel of the spectrum highly improves the performance of a Mössbauer spectrometer. The resulting spectrum must not necessarily be centered in null velocity and its channels are not required to be equally spaced. These facts enable the experimentalist to focus the observation in a region of interest of the spectrum where the information is more concentrated.

But frequently the ROI selection is not straightforward and depends on the complexity of the experiment. We have presented a mathematical tool that aids in this selection, providing a comparison factor between different channel sets. This tool is based on the assumption that every channel of the spectrum is not equally efficient for a given experimental application. It was demonstrated that channels closer to the maximum derivative of the studied parameter contribute better to the quality of parameter determination. This principle was quantified for several practical situations (i.e. line center, effect and width determination) taking a Lorentzian as line shape approximation.

Results show that optimal ROI selection depends on the searched parameter (e.g. the ROI for best line effect resolution is not the same that the ROI for best line center resolution). When more than one parameter is being studied, as can be the case of a complicated spectrum, there will be a compromise between a great number of variables to be determined, being responsibility of the experimentalist to find an appropriate balance between them. The present approach can be analytically extended to these cases.

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