

Line shape of Mössbauer spectra obtained with the use of a resonant detector

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Abstract It is shown that the spectrum registered by a resonant detector can be described as a linear combination of the Lorentzian line and the square of the Lorentzian line.

Keywords Mössbauer spectroscopy · Resonant detector · Shape of line

1 Introduction

Resonant detectors can be used for the registration of Mössbauer radiation. Their operation is based on the transformation of absorbed resonance radiation into conversion electrons and the registration of conversion electrons arising from the transition of resonant nuclei from the excited state to the ground one. The main characteristic of a resonant detector in comparison with a scintillation detector is that it registers only the resonant gamma rays emitted by the source [1]. However, a resonant detector introduces some distortions in the line shape of the absorption spectrum [2–4]. Existing fitting programs usually use a sum of Lorentzian lines. Fitting of a spectrum obtained with a resonant detector with existing known fitting programs gives a specific difference spectrum. For an iron source [4], the difference spectrum is presented in Fig. 1a. The area of such difference spectrum can be up to

This article is part of the Topical Collection on *Proceedings of the International Conference on the Applications of the Mössbauer Effect (ICAME 2015), Hamburg, Germany, 13–18 September 2015*

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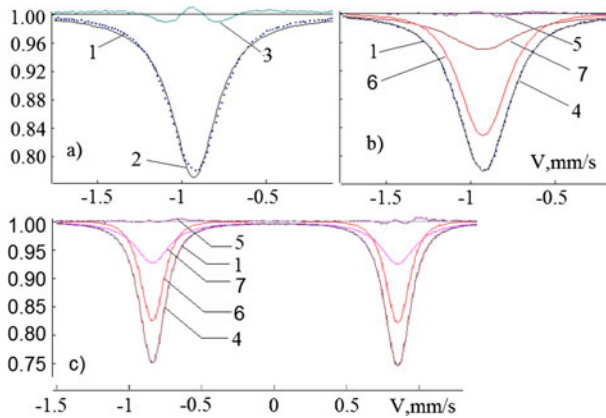


Fig. 1 Mössbauer spectra of iron (line #3 of sextet) (a, b) and sodium nitroprusside (c), obtained with a resonant detector: 1 – experimental spectrum; 2 – Lorentzian line; 3 – difference spectrum in case of fitting with the Lorentzian line; 4 – linear combination of the Lorentzian line (6) and its square (7); 5 – difference spectrum for the case of fitting with the linear combination of the Lorentzian line and its square

10 % of the total area of the fitting spectrum. In part where the resonance peak approaches the baseline and near the maximum of the resonance peak the difference spectrum is positive due to the excess of the theoretical values over the experimental ones (positive area). Between those parts the difference spectrum is negative (negative area). Such specific difference spectra are observed both for all investigated samples, including thin samples, and standard samples. It is noteworthy that such specific difference spectra are not obtained as the result of fitting spectra registered with scintillation detectors in spite of using the same fitting programs. We have fitted spectra obtained with a resonant detector using other functions, for example, the Lorentzian-Gaussian, the Voigt profile [5, 6], taking into account the additional component decomposition of the exponent in the function of the energy distribution of the absorbed gamma rays in the sample [3]. These steps reduce the difference spectrum, but do not change its shape.

In the last few years works which take into account the nature of the resonant detector and its influence on the spectrum were done by various researchers [2, 7, 8]. But the proposed models are complex to implement.

In this work we found a simpler model for describing spectra obtained with a resonant detector and we applied it to the spectra obtained with an iron detector.

2 Theoretical deduction

A spectrum registered with a scintillation detector represents an energetic distribution of gamma rays passed through an absorber. It can be represented as a convolution of the line of resonant emission of gamma quanta with a source $WS(E)$ and the line of resonant absorption of gamma quanta with energy E in the studied sample $(1 - Wa(E))$ [9, 10]. A resonant detector registers only resonant gamma quanta. Thus, a resonant detector imposes

the $W_{RD}(E)$ line to the absorption line and the registered spectrum has a form discussed in [11, 12].

$$F_{RD}(v) = \int_0^{+\infty} W_s(E)[1 - W_a(E, v)]W_{RD}(E)dE - \int_0^{+\infty} W_s(E)W_a(E, v)W_{RD}(E)dE. \tag{1}$$

The value of the first integral in (1) doesn't depend on the velocity of the relative movement and gives the number of gamma quanta registered far from the resonance in the absorber. The second integral in (1) gives the line of the absorption spectrum detected using a resonant detector.

The following assumptions are made: the distribution function of absorbed quanta in a resonant detector from energy $W_{RD}(E)$ is similar to the absorption function in the sample; all atoms in the detector are in an equivalent state and the detector doesn't move. It is supposed that each of the $W_s(E)$, $W_a(E)$ and $W_{RD}(E)$ lines has the form of the Lorentzian line [9, 10]. Then for a separate line of absorption in a sample the second integral in (1) has the following view:

$$\epsilon_{RD}(v) = \frac{f_s f_a n_k f_{RD} \sigma_0^2 \Gamma_s^2 \Gamma_a^2 \Gamma_{RD}^2}{(2\pi)^3} \times \int_0^{+\infty} \frac{dE}{\left((E - E_s^0)^2 + \left(\frac{\Gamma_s}{2}\right)^2 \right) \left((E - E_0^a (1 - \frac{v}{c}))^2 + \left(\frac{\Gamma_a}{2}\right)^2 \right) \left((E - E_0^{RD})^2 + \left(\frac{\Gamma_{RD}}{2}\right)^2 \right)}, \tag{2}$$

where f_s – the probability of the Mössbauer effect in the source, f_a and f_{RD} – the probabilities of resonant absorption in the sample and in the detector, respectively, σ_0 – the maximum cross section of resonant absorption, E_s^0, E_0^a, E_0^{RD} – the energies of the resonant transition in the source, the sample and the detector, respectively, $\Gamma_s, \Gamma_a, \Gamma_{RD}$ – the widths of the line of the resonant transition in the source, the sample and the detector, respectively. In ideal crystals $\Gamma_s, \Gamma_a, \Gamma_{RD}$ are equal to the natural width, but in practice they can exceed the natural width because of crystal lattice imperfection and/or due to diffusion of Mössbauer atoms via vacancies [13] and/or due to thickness effects.

Further mathematical calculations are carried out within the following model of the spectrum registration process. In practice, in a resonant detector there is the same energy of transition as in the source. Equality of the widths of the lines of resonant transitions in the source, the absorber and the detector is assumed.

As a result the line of the absorption spectrum registered with use of a resonant detector can be presented with the following formula:

$$\epsilon_{RD}(v) = \frac{A_1}{\pi} \frac{\Gamma/2}{(v - \delta) + (\Gamma/2)^2} + \frac{(\Gamma/2)^2}{((v - \delta)^2 + (\Gamma/2)^2)^2} \tag{3}$$

Here A_1 and A_2 – normalizing coefficients, Γ – width of the Lorentzian line on a half-height in mm/s, δ – isomer shift of the line in mm/s. Normalizing coefficients are chosen so that the area under the spectrum was equal to their sum.

As one can see, this function includes a function of the Lorentzian line and the square of the Lorentzian line. The presence of square of the Lorentzian function is due to the specifics resulting from formula (1) which describes a resonant detector operation.

The formula (3) has been compared with the formula deduced by Odeurs et al. [7, p.6155,#56], which was successfully tested for fitting of the spectrum of tin [2]. Analyze of Odeurs' formula shows that it can be converted to a form involving the square of the Lorentzian function and under certain assumptions the Lorentzian line.

Based on the formula (3), taking into account possible conversion of the formula deduced by Odeurs, we can assume that in order to fit the actual spectra a linear combination of the Lorentzian line and its square should be applied.

The full registered spectrum is described with the sum of lines from all nonequivalent atom states of the studied element:

$$F_{RD}(v) = B - \sum_{k=1}^N \left[\frac{A_{1k}}{\pi} \frac{\Gamma_k/2}{(v - \delta_k) + (\Gamma_k/2)} + \frac{A_{2k}\Gamma_k}{\pi} \frac{(\Gamma_k/2)^2}{((v - \delta_k)^2 + (\Gamma_k/2)^2)} \right], \quad (4)$$

where B – number of the quanta recorded at a large distance from resonance, N – number of lines of a spectrum.

3 Experimental check

To verify the obtained theoretical curve (4) the spectra of sodium nitroprusside and alpha-iron obtained with a resonant detector are fitted. So, we focus on a number of practical important spectra obtained on an iron detector.

Only one line of the alpha-iron spectrum is presented in Fig. 1a,b for illustration purposes. The case of fitting with the Lorentzian function is presented in Fig. 1a and the case of fitting with the sum of a Lorentzian function and its square is presented in Fig. 1b. Spectrum of quadrupole splitting which corresponds to sodium nitroprusside fitted with the sum of Lorentzian function and its square is presented in Fig. 1c. The difference spectrum in the case of fitting with (4) (Fig. 1b, c) often has a random variation relative to the baseline. This means the specific difference spectrum is lost and the sum of the squares of the standard deviations is less than half when fitting with a sum of Lorentzian lines (Fig. 1a). In case of alpha-iron the χ^2 value for the fit with (4) is 1.3, while it is 4.3 for the Lorentzian fit. In case of sodium nitroprusside the χ^2 value for the fit with (4) is 1.2.

The Lorentzian line width in (4) is of the order of 0.3 mm/s, which is more than double the natural line width. It is important to note that in case of fitting with the Lorentzian line the calculated line width is approximately equal to 0.17 mm/s. The area of the Lorentzian line square is approximately 30 % of the total area of the spectrum. The square of the Lorentzian function allows to describe the top of the peak quite well. The Lorentzian line in case of fitting a spectrum with (4) has a smaller amplitude than it has in case of fitting a spectrum with just the Lorentzian line. Thus, positive area of the difference spectrum is lost. The negative area of the difference spectrum is lost due to the greater line width.

4 Discussion

Therefore, the deduced function (4) describes the experimental points fairly well while the fitting procedure remains relatively simple at the same time. The fitting with use of this function (4) removes the characteristic distortions of the difference spectrum. The width of the Lorentzian line in (4) is bigger than the double natural line width. Other researchers noted the narrow experimental spectra registered with resonant detector [1, 2, 14]. This phenomenon can be explained (as we see) with the presence of not only a Lorentzian line but also of the square of Lorentzian line.

As a result, for fitting the spectra obtained with the use of a resonant detector, it is necessary to consider the particular form of the spectral lines, the description of which should be selected based on the physics of the experiment.

5 Conclusions

The function describing a spectrum registered with the use of a resonant detector is theoretically deduced taking into account the operation of a resonant detector.

This function is a linear combination of the Lorentzian line and the square of the Lorentzian line.

The deduced curve accurately describes the experimental points removing the characteristic distortions of the difference spectrum.

The width of the Lorentzian line entering in function is larger than twice the natural line width.

As can be seen from the above, the proposed method can be used for successful fitting of the spectra obtained with a resonant detector in a realistic setting. It is necessary to take into account a particular form of spectra registered with a resonant detector.

Acknowledgments This study was supported by the RF Ministry of Education and Science, project no. 14.575.21.0086 unic project identifier RFMEFI57514X0086.

References

1. Irkayev, S.M., Kuzmin, R.N., Opalenko, A.A.: *Yadernii gamma resonanc. (Apparatura I metodika)*. M.: MGU, 208 p. (Rus) (1970)
2. Odeurs, J., Hoy, G.R., L'abbé, C., Koops, G.E.J., Pattyn, H., Shakhmurov, R.N., Coussement, R., Chiodini, N., Paleari, A.: *Hyper Inter* **139/140**, 685–690 (2002)
3. Gladkov, V.P., Kashcheev, V.A., Kuskov, A.H., Petrov, V.I.: *J. Appl. Spectrosc.* **71**(5), 668–671 (2004). (Rus)
4. Gladkov, V.P., Martinenko, S.S., Petrov, V.I.: *J. Appl. Spectrosc.* **78**(12), 316–320 (2011). (Rus)
5. Armstrong, B.H.: *Quant. Spectrosc. Radiat* **7**, 61–88 (1967)
6. Ida, T., Ando, M., Toraya, H.: *J. Appl. Cryst.* **33**, 1311–1316 (2000)
7. Odeurs, J., Hoy, G.R., L'abbé, C., Shakhmurov, R.N., Coussement, R.: *Phys. Rev. B* **62**(10), 6148–6157 (2000)
8. Gladkov, V.P., Martinenko, S.S., Petrov, V.I., Filippov, V.P.: *Bull. Russ. Acad. Sci. Phys.* **77**(6), 779–783 (2013)
9. Goldansky, V.I., Herber, R.H.: *Chemical Applications of Mossbauer Spectroscopy*. Acad Press, London (1968)
10. Vertheim, G.: *Mossbauer Effect: Principles and Applications*. Academic press, New York and London (1964)

11. Spijkerman, J.J., Travis, J.C., Pella, P.A., DeVoe, J.R.: Preliminary Study on the Characteristics and Design Parameters for a Mössbauer Resonant Detector. Nat. Bur. Stand. (U.S.), Technical note 541 (1971)
12. Irkayev, S.M.: Mnogomernaya parametricheskaya messbauerovskaya spectrometriya. SPb., 35(Rus) (1994)
13. Bender, O., Schroeder, K.: Phys. Rev. B **19**(7), 3400–3413 (1979)
14. Mitrofanov, K.P., Illarionova, N.V., Shpinel, V.S.: Prib. Tekh. Eksp. **3**, 49 (1963)