Influence of Gamma rays collimation on Mössbauer lines

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(recibido el 5 noviembre 1987; aceptado el 1 de julio de 1988)

Abstract. We investigate the effect of gamma rays collimation in Mössbauer absorption spectra when thin absorbers are used. A simple model is proposed to study shift and broadening of spectral lines. This model along with some approximations, makes the calculations easily done. The results are in good agreement with those in the literature.

PACS: 33.45.+x

1. Introduction

Lack of collimation, as it has been long known, affects the width and position of Lorentzian lines in Mössbauer spectroscopy [1,2,3,4,5]. Even where good collimation exists, gamma rays incident upon the detector window produce, at finite angles with respect to the direction of relative motion, the well-known cosine effect [4].

Corrections for this effect have been proposed in the past [4,5,6] in the form of angular distribution functions of photons inciding upon the detector window. These approaches in general involve tedious calculations, with the exceptions of Refs. [3] and [5] which yield results for immediate use. However, a shortcoming of Ref. [3] is sits failure to mention broadening, and of Ref. [5] is the fact that the gamma ray's distribution function as suggested cannot be used on its complete domain for $R/d \ll 1$, where R and d are the collimator's radius and length respectively.

It is the aim of this paper to introduce a simple model for calculating shift and broadening of the spectral lines produced by non-collimated gamma rays incident upon the detector, that allow calculations to be easily performed and give results that are in good conformity with those in the literature [5]. For working purposes it will be assumed in this paper that the distance between source and detector window is much greater than source and detector radii.

2. Point source

A point source located on the perpendicular axis of a detector window emits gamma rays that pass through a thin sample and produce an absorption peak having a

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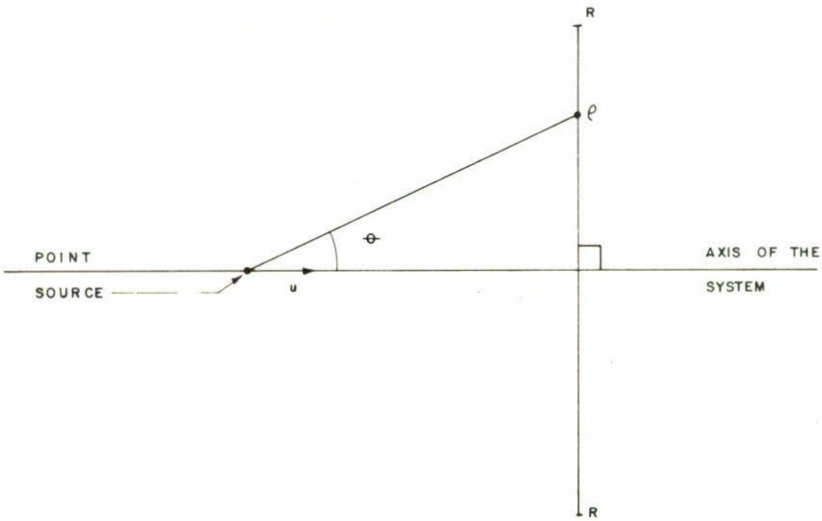

FIGURE 1. Point source located on the perpendicular axis of the detector window of radius R.

Lorentzian shape

$$f(v) = \frac{\Delta}{\pi} \frac{1}{\Delta^2 + (v - v_0)^2}, \tag{1}$$

where $v = u\cos\theta$ is the component of source-absorber velocity u along the gamma ray propagation path (Fig. 1), v_0 the velocity of maximum absorption at the perpendicular incidence, Δ the mid-width at half maximum of the peak, and Δ/π a normalization constant.

3. Extended source

To find the line shape in the case of an extended source, it is first necessary to obtain a distribution function which takes into account the gamma rays emitted by each element of area of the extended source and which incide on each element of area of the detector window. A given element of source area located at a distance r from the source-detector axis will illuminate, according to Fig. 2, each element of detector area with a distinct intensity. The difference is due both to the inverse square and to the cosine laws of illumination.

Considering gamma rays emitted in a direction parallel to the axis of the system, for example, along line d or any other line which is parallel, no effect appears capable of modifying the line shape. However in the case of rays travelling along lines d' and

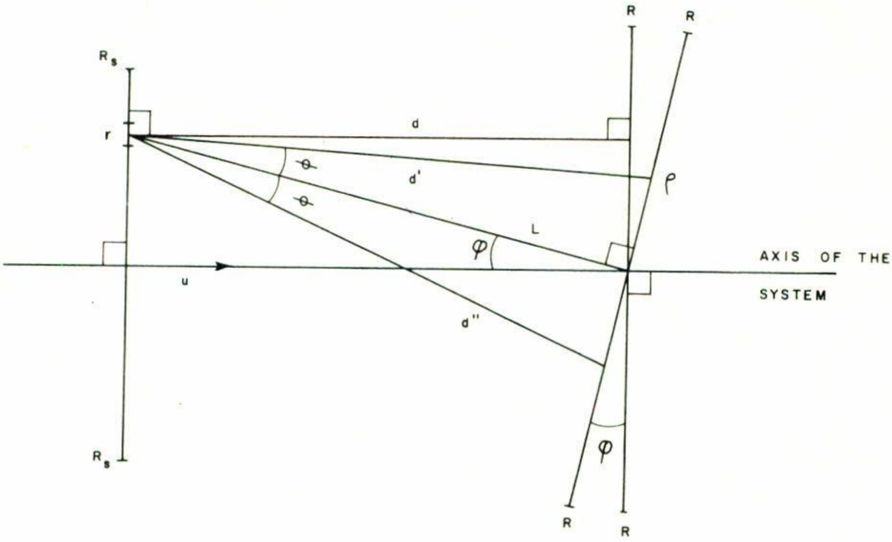

FIGURE 2. Element of source area located at distance r from the axis of the system. This same element can be assumed, however, to be located on the perpendicular axis of the detector window which (as seen from the location of the element of source area) appears to be rotated by an angle ϕ.

d'', where d' and d'' are symmetrical with respect to direction L, we may assume that the detector has rotated in such a way that its perpendicular L now forms an angle ϕ with the axis of the system. The component of the source speed along this new axis is $u\cos\phi$, which by the cosine law of illumination projects onto rays d' and d'' as

$$u\cos\theta\cos\phi \tag{2}$$

and for each element of source area analogously. The expressions for $\cos\theta$ and $\cos\phi$ based on Fig. 2 can be written

$$\cos\theta = \frac{\sqrt{d^2+r^2}}{d'} \qquad \text{and} \qquad \cos\phi = \frac{d}{\sqrt{d^2+r^2}} \tag{3}$$

respectively. The product $\cos\theta\cos\phi$ then becomes

$$\cos\theta\cos\phi = \frac{d}{d'} = \frac{d}{\sqrt{d^2+r^2+\rho^2}}, \tag{4}$$

which is equal to the ratio of distances for parallel and non parallel emissions to the system axis. The denominator in the last member of Eq. (4), is based on Fig. 2. When constructing the distribution function of gamma rays incident upon the detector, the correction accounting for the inverse square law of illumination which must be taken into consideration is therefore

$$\left(\frac{d}{d'}\right)^2 = \cos^2\theta\cos^2\phi. \tag{5}$$

Passing on next to the radial aspects of the distribution function, and for purposes of simplicity, consider the physical fact that the gamma rays emitted from elements of source area located at large values of r, as well as those incident on the detector at large values of ρ will contribute most in the spectral lines distortion. Therefore, the radial part of the distribution function should be written as

$$2\pi\rho d\rho 2\pi r dr. \tag{6}$$

Finally, using Eqs. (5) and (6) the distribution function may be formulated as

$$F(r,\rho)drd\rho = K4\pi^2\cos^2\theta\cos^2\phi r dr\rho d\rho, \tag{7}$$

where the normalization constant K is found through performing the integral

$$\int_0^{R_S}\int_0^{R} F(r,\rho)drd\rho = 1,$$

being

$$K = \frac{1}{\pi^2 R_S^2 R^2\left[1 - \frac{R_S^2+R^2}{2d^2}\right]}, \tag{8}$$

where R_S and R are the source's and detector's radii respectively. The distribution function thus becomes

$$F(r,\rho) = \frac{4\rho r\cos^2\theta\cos^2\phi}{R_S^2 R^2\left[1 - \frac{R_S^2+R^2}{2d^2}\right]}. \tag{9}$$

It will be seen in time that distribution (9) contributes to simplify integrations and affords results which are in good agreement with those obtained in Ref. [5], in which a more complicated distribution function is given. For the condition $R/d \ll 1$, where R and d are respectively the radius and length of the collimator used, the distribution proposed in Ref. [5] cannot be extended over its complete domain.

4. Line shape

The observed line shape produced by an extended source is obtained by averaging Eq. (1) with $v = u\cos\theta\cos\phi$ and using (7) as a weighing function

$$I(u) = \frac{\Delta}{\pi} K \int_0^{R_S} \int_0^{R} \frac{4\pi^2 \cos^2\theta \cos^2\phi \, r dr \rho d\rho}{\Delta^2 + (u\cos\theta\cos\phi - v_0)^2}. \tag{10}$$

The integration of (10) becomes straightforward when the product $\cos\theta\cos\phi$ is expanded into a series and terms above the second order in $(r^2 + \rho^2)/d^2$ for small R^2/d^2 and R_S^2/d^2, discarded.

Maintaining the same degree of approximation, the result of the integration is

$$I(u) = \frac{\Delta}{\pi} \frac{1}{B^2} \left[1 + \frac{R^2 + R_S^2}{2d^2} \frac{A^2}{B^2} \right], \tag{11}$$

where

$$A^2 = u(u - v_0); \qquad B^2 = \Delta^2 + (u - v_0)^2. \tag{12}$$

The explicit form of (11) can be obtained by introducing into it the expressions (12)

$$I(u) = \frac{\Delta}{\pi} \frac{1}{\Delta^2 + (u - v_0)^2} \left[1 + \frac{R^2 + R_S^2}{2d^2} \frac{u(u - v_0)}{\Delta^2 + (u - v_0)^2} \right]. \tag{13}$$

For having a correction term which depends upon the geometry of the system source-detector, this profile is no longer a Lorentzian one. The correction term is proportional to a factor that depends upon the linear dimensions of source detector and the distance d. It may be noted that when R and R_S tend to zero or when d tends to infinity the effect disappears and a Lorentzian term remains. Predictably, this new line may lose its normalization. Performing the integration

$$\int_{-\infty}^{\infty} I(u) du = 1,$$

the following normalization constant is found which, as could be expected, depends upon the geometry of source and detector

$$\frac{1}{1 + \frac{R^2 + R_S^2}{4d^2}}. \tag{14}$$

Expanding this expression into a series in which terms up to the first order in

$(R^2 + R_S^2)/4d^2$ are conserved, and inserting the result in (13) the new line becomes:

$$I(u) = \frac{\Delta}{\pi} \frac{1}{\Delta^2 + (u - v_0)^2} \left[1 + \frac{R^2 + R_S^2}{2d^2} \frac{u(u - v_0)}{\Delta^2 + (u - v_0)^2}\right] \left[1 - \frac{R^2 + R_S^2}{4d^2}\right]. \quad (15)$$

It is to be noticed that the spectral line (15) has a height reduction equal to the factor

$$1 - \frac{R^2 + R_S^2}{4d^2}. \quad (16)$$

This reduction, as before, is an effect attributable to the finite size of source and detector and finite distance d.

Since the unitary area of the line remains unchanged, not only can we expect a broadening of the line itself but also, in all probability, a shift in position such as will be treated in the next section.

5. Shift and broadening of the spectral line

From Eq. (15), the point of maximum absorption can be found by obtaining

$$\frac{dI(u)}{du} = 0.$$

Solving it gives the third degree equation

$$u_{\text{MAX}} = v_0 - \alpha \left[\frac{2u_{\text{MAX}}(u_{\text{MAX}} - v_0)^2}{\Delta^2 + (u_{\text{MAX}} - v_0)^2} - \left(u_{\text{MAX}} - \frac{v_o}{2}\right)\right], \quad (17)$$

where

$$\alpha = \frac{R^2 + R_S^2}{2d^2}. \quad (18)$$

To find the solution of (17), iteration will be used in which by observing that as α tends to zero, $u_{\text{MAX}} = v_0$. Now substituting the value derived in (17), the next approximation is obtained

$$u_{\text{MAX}} = v_0 + \alpha\frac{v_0}{2} = v_0\left(1 + \frac{\alpha}{2}\right).$$

Substitution of (18) in the last member of this expression yields

$$u_{\text{MAX}} = v_0 \left[1 + \frac{R^2 + R_S^2}{4d^2}\right] \qquad \text{or} \qquad \frac{u_{\text{MAX}} - v_0}{v_0} = \frac{R^2 + R_S^2}{4d^2}. \quad (19)$$

The above equation indicates that the line has been shifted by the same fractional amount (16) by which its height has been diminished.

Next, the broadening of the line will be evaluated by finding the points at which the height of the profile is half that of its maximum. From Eq. (15) these points are found by solving the fourth degree equation

$$\frac{1}{1+\dfrac{(u-v_0)^2}{\Delta^2}}\left[1+\alpha\frac{u(u-v_0)}{\Delta^2+(u-v_0)^2}\right]=\frac{1}{2},$$

where α is defined in (18). The latter equation may be written as

$$\xi^2-2\xi\Delta^2-2\Delta^2\alpha u(u-v_0)=0, \tag{20}$$

where

$$\xi=\Delta^2+(u-v_0)^2. \tag{21}$$

Eq. (20) can also be solved by iteration. A first approximation to the solution is obtained for $\alpha=0$ in which case

$$u^{(1)}=v_0\pm\Delta. \tag{22}$$

On the other hand, the solution of (20) is

$$\xi=\Delta^2\pm\Delta\sqrt{\Delta^2+2\alpha u(u-v_0)}. \tag{23}$$

By substituting (22) in (23), the second approximation is obtained

$$u_{\substack{\text{half}\\\text{height}}}=v_0\pm\Delta\left[1+\frac{\alpha}{2}\left(1\pm\frac{v_0}{\Delta}\right)\right]. \tag{24}$$

To find this solution, an expansion in series keeping terms up to the first order in α must be made.

The half width of the observed line is obtained using the definition

$$\Delta^*=\pm\left[u_{\substack{\text{half}\\\text{height}}}-u_{\text{MAX}}\right]. \tag{25}$$

By substituting (18), (19) and (24), the result

$$\Delta^*=\Delta\left[1+\frac{R^2+R_S^2}{4d^2}\right] \tag{26}$$

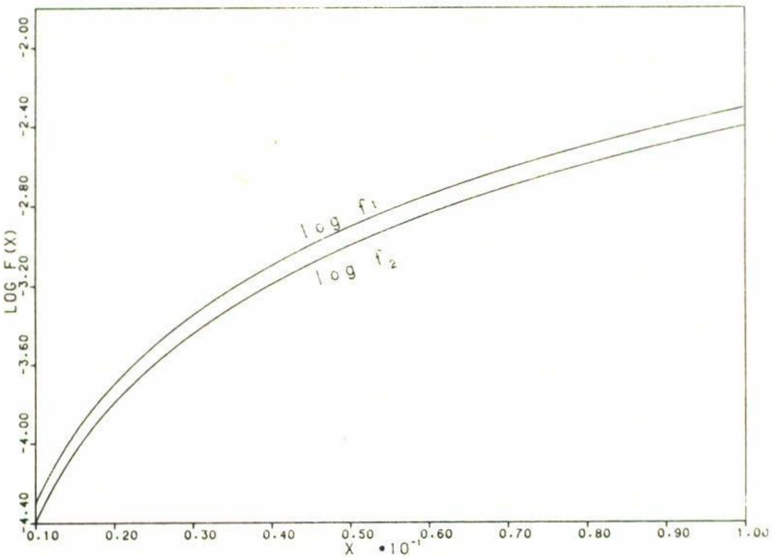

FIGURE 3. Comparison of $\log f_1(x)$ and $\log f_2(x)$ as functions of $x = R/d$.

is obtained. This expression reflects that the line has been broadened by the same fraction by which its height has been diminished and its position shifted based on (16) and (19) respectively.

The geometrical factor

$$\frac{R^2 + R_S^2}{4d^2}, \tag{27}$$

derived under the approximations introduced in this work, accounts for the modification of the Mössbauer line when lack of collimation is present.

6. Discussion

Calculations result in a simple and symmetrical formula for evaluating the fraction by which the height, position and shape of a Mössbauer line are modified when lack of collimation of gamma rays is present. To increase the counting rate upon handling a weak source, the experimenter often reduces the distance between source and detector; at that time the problem arises as to how near the detector can be approximated to the source and still satisfy the conditions $R/d \ll 1$, $R_S/d \ll 1$ before the line becomes appreciably modified. As an example, Eq. (27) is applied to an experimental setup in which the parameters have the following values: $R_S = 0.4$ cm, $R = 1.27$ cm and $d = 10$cm. The resultant 0.44% is less than the experimental error for usual measurements.

Hence, to increase the counting rate by locating the source at a point nearer the detector may give a value for Eq. (27) which becomes comparable to experimental error, and the ratios $R/d, R_S/d$ are not longer much less than unity. To illustrate, experiments for $d = 3, 4, 5cm$ were performed for stainless steel as an absorber. Mössbauer spectra were obtained in which the modifications in height, shift and broadening were about 20%. However, this figure is not conclusive because for those distances the model presented in this paper breaks down.

Finally, we observe from the geometrical factor (27) that for the particular case $R_S = R$ we have

$$f_1(x) = \frac{1}{2}x^2, \tag{28}$$

where $x = R/d$. This expression can be compared with its homologous formula

$$f_2(x) = \frac{2\pi}{9\sqrt{3}}x^2, \tag{29}$$

in equations (16) and (21) of Ref. [3], in an interval such that both models are valid; that is $x^2 \ll 1$. Fig. 3 shows the behaviour of log $f_1(x)$ and log$f_2(x)$ versus x, in the interval of interest. On the other hand, if the geometrical factor (27) is calculated for $R_S = 0$, the result coincides with that obtained from Eq. (6), Ref. [1] when an expansion for $x^2 \ll 1$ is made.

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Resumen. Se investiga el efecto de la colimación de rayos gamma sobre los espectros de absorción Mössbauer cuando se usan absorbedores delgados. Se propone un modelo sencillo para estudiar corrimiento y ensanchamiento de líneas espectrales. El uso de este modelo y de algunas aproximaciones permite realizar cálculos fácilmente. Los resultados muestran buena semejanza con los reportados en la literatura.