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UNUSUAL SHAPE OP THE MOSSBAUER SCATTERING LINE

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The influence of the Rayleigh and Compton contributions to a Mössbauer scattering line is studied. A Mössbauer scattering spectrum of absorption character is theoretically predicted and experimentally observed for iron impurities in beryllium.

1. Introduction

Conventionally, the Mössbauer scattering line is associated with an increase in intensity of gamma rays scattered at the resonance energy of the scatterer. However, in some cases the opposite phenomenon can also appear; that is, the intensity of scattered gamma rays can reach its minimum value at the resonance energy. Such an unusual shape of a Mössbauer scattering line can be expected for Mössbauer isotopes with large values of electron conversion coefficients contained in samples which have small cross section values for photoeffect as compared to those for Rayleigh and Compton scattering processes. The scattering line of absorption character is due to the internal conversion electron process and can be most easily observed for a very thick sample composed of very light elements with some impurities of resonant isotope. In such a case, at a velocity far from resonance most of the gamma quanta incident on the sample are scattered by Compton and Rayleigh scattering processes, whereas at the resonance velocity some of the quanta are lost due to an internal electron conversion process associated with the nuclear resonance interactions of gamma rays with Mössbauer nuclei. In this paper we report the results of the numerical analysis and the experimental investigations of the unusual shape of the Mössbauer scattering line.

2. Nuclear resonance, Rayleigh and Compton contributions to the scattering line

In order to evaluate the shape of a Mössbauer scattering line we have to consider the intensities of the fraction $f_0 I_0$ recoilless—recoillessly and recoilless— non-recoillessly scattered through nuclear resonance scattering, recoillessly and non-recoillessly scattered by Rayleigh scattering and non-elastically scattered by Compton scattering as well as the intensities of the fraction $(1 - f_0)I_0$ scattered both by Rayleigh and Compton scattering processes. I_0 and f_0 are the intensity and its recoilless fraction of Mössbauer gamma rays, respectively. At the resonance velocity of the scatterer the recoillcss-recoillcss and recoilless-non-recoilless nuclear resonance contributions have their maximum values and go to zero at velocities far from resonance. The recoilless and non-recoilless Rayleigh as well as non-elastic Compton contributions reflect, of course, the gamma ray intensity modulation in the scatterer by the nuclear resonance absorption and hence have their minimum values at the resonance velocity. The Rayleigh and Compton contributions originating from the source intensity $(1 - f_0)I_0$ are not velocity dependent.

Usually, the magnitude of the nuclear resonance contributions is much higher than that of Rayleigh plus Compton contributions. In this case the scattering line has its usual shape, that is, it is associated with an increase in gamma ray intensity at the resonance velocity. In some cases, however, the nuclear resonance contributions do not compensate, due to compensate, due to the internal electron conversion process, for the decrease in the value of Rayleigh plus Compton contributions at the resonance velocity. The corresponding scattering line is observed then to have its unusual shape of the absorption character.

In order to evaluate the shape of the background-corrected scattering line we have to separate the velocity dependent part from the total scattered

intensity and to correct it for electronic absorption. This may be done by calculating the ratio

$$\epsilon(S) = [N(S) - N(\infty)]/N(\infty) ,$$

where $N(S)$ and $N(\infty)$ are the numbers of counts at the Doppler energy shift S and at $S = \infty$, respectively. Thus, the shape of the background-corrected scattering line measured in the geometry of fig. 1 is described by the formula [1]

$$\epsilon(S) = a_r I_{x_r}(S) + a_n I_{x_n}(S) - a_R [1 - J_{x_r}(S)] - (a_N + a_C)[1 - J_{x_n}(S)] , \quad (1)$$

where

$$I_x(S) = \frac{1}{\pi[1 - \exp(-T)]} \int_{-\infty}^{\infty} dy\, Z_x(y, S) ,$$

$$J_x(S) = \frac{1}{\pi[1 - \exp(-T)]} \int_{-\infty}^{\infty} dy[(y - S_1)^2 + 1]\, Z_x(y, S) ,$$

$$Z_x(y, S) = \frac{1 - \exp\left[-T\,\frac{(y - S_1)^2 + x^2}{(y - S_1)^2 + 1}\right]}{[(y - S)^2 + 1][(y - S_1)^2 + x^2]} ,$$

while

$$a_r = \frac{f_0 f_1 \mu_r W_\gamma(\theta)}{(1 + \alpha)[\mu_R(\theta) + \mu_C(\theta)]} ,$$

$$a_n = \frac{f_0 (1 - f_1) \mu_r W_\gamma(\theta)}{(1 + \alpha)[\mu_R(\theta) + \mu_C(\theta)]} ,$$

$$a_R = \frac{f_0 f_R(\theta) \mu_R(\theta)}{\mu_R(\theta) + \mu_C(\theta)} ,$$

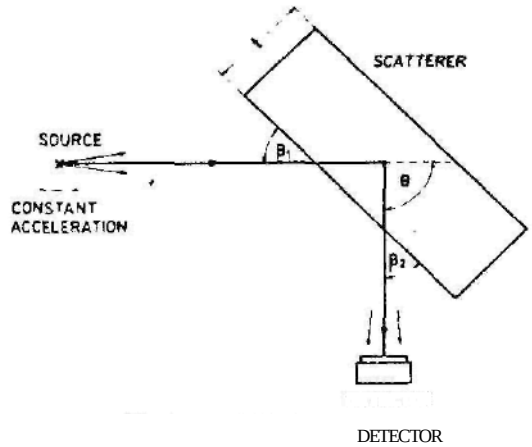

Fig. 1. Scattering geometry.

$$a_N = \frac{f_0[1 - f_R(\theta)]\,\mu_R(\theta)}{\mu_R(\theta) + \mu_C(\theta)} ,$$

$$a_C = \frac{f_0 \mu_C(\theta)}{\mu_R(\theta) + \mu_C(\theta)} , \quad x_r^2 = 1 + \mu_r/\mu ,$$

$$x_n^2 = 1 + \frac{\mu_r \operatorname{cosec} \beta_1}{\mu(\operatorname{cosec} \beta_1 + \operatorname{cosec} \beta_2)} ,$$

and $T = \mu t(\operatorname{cosec} \beta_1 + \operatorname{cosec} \beta_2)$. The energy of gamma rays γ and the isomer shift S_1 of the scatterer are given in respect to the energy of the Mössbauer transition in the source, y, S_t and the Doppler energy shift of the source S are expressed in $\Gamma/2$ units. θ is the scattering angle while β_1 and β_2 are the angles with respect to the scatterer surface of incoming the outgoing gamma rays, respectively. $W_\gamma(\theta)$ is the angular distribution function for the resonantly scattered gamma rays. μ_r is the maximal value of the nuclear resonance absorption coefficient of the scatterer while $\mu_R(\theta)$, $\mu_C(\theta)$ and μ are its Rayleigh, Compton and total electronic attenuation coefficients, respec-tively, t is the thickness of the scatterer f_1 and $f_R(\theta)$ are the recoilless fractions of (the scatterer for the nu-clear resonance and Rayleigh scattering processes, respectively, a is the total electron conversion coefficient for the Mössbauer transition.

The first two terms of eq. (1) originate from the recoillcss-recoilless and recoilless—non-recoilless nuclear resonance scattering, respectively. The third term is due to the recoilless Rayleigh, while the fourth one to non-recoilless Rayleigh plus non-elastic Compton scatterings.

Integration of eq. (1) over the energy can be performed explicitly both for thin and very thick samples. In the thick sample limit approximation the scattering line is described by two Lorentzian lines with different amplitudes A_r and A_n and different widths γ_r and γ_n:

$$\epsilon(S) = A_r L_{x_r}(S) + A_n L_{x_n}(S) , \quad (2)$$

where

$$A_r = \frac{f_0 \mu}{\mu_R(\theta) + \mu_C(\theta)} \left(1 - \frac{1}{x_r}\right) \times \left[\frac{f_1 W_\gamma(\theta)}{1 + \alpha} - \frac{f_R(\theta)\,\mu_R(\theta)}{\mu}\right] ,$$

$$A_n = \frac{f_0 \mu}{\mu_R(\theta) + \mu_C(\theta)} \left(1 - \frac{1}{x_n}\right) \times \left\{\frac{(1 - f_1)\, W_\gamma(\theta)(\operatorname{cosec} \beta_1 + \operatorname{cosec} \beta_2)}{(1 + \alpha) \operatorname{cosec} \beta_1}\right.$$

$$- \frac{[1 - f_R(\theta)]\,\mu_R(\theta) + \mu_C(\theta)}{\mu} \Bigg\},$$

$$L_x(S) = \frac{(x+1)^2}{(S - S_1)^2 + (x+1)^2} \;;$$

$$\gamma_r = x_r + 1 \quad \text{and} \quad \gamma_n = x_n + 1 \,.$$

In this approximation the scattering line has the amplitude $A = A_r + A_n$, while its width γ fulfills the relation $\gamma_n < \gamma < \tilde{\gamma}_r$

The signs of A_r and A_n depend on the values of α, μ $\mu_R(\theta)$ and $\mu_C(\theta)$, μ is the sum of total photoabsorption, Rayleigh and Compton attenuation coefficients Thus, the character of the scattering line strongly depends on the value of the electron conversion coefficient and the values of attenuation coefficients for photoabsorption and both Rayleigh and Compton scattering processes. A scattering line of absorption character can be observed in samples with small values of cross sections for photoefect, as compared to those for Rayleigh and Compton processes, which contain isotopes with Mössbauer transitions of large values of electron conversion coefficients. We have selected a beryllium scatterer with some iron impurities as a good sample for studying the influence of Rayleigh and Compton scatterings on the shape of the Mössbauer scattering line.

3. The results of numerical calculations

Eq. (1) was used to numerically calculate the shape of a scattering line for various scatterer parameters. Beryllium and carbon scatterers with iron impurities up to 1 at.% were considered in the calculations. For simplicity it was assumed that the scatterers possess a single Mössbauer line at room temperature. Some scattering spectra for the hypothetical beryllium scatterer are shown in figs. 2—5. The Mössbauer line can be transformed from its typical scattering shape to the absorption shape by verying the sample thickness (fig. 2), electronic and nuclear resonance absorption coefficients (figs. 3 and 4, respectively). The influence of the Rayleigh and Compton scatterings on the shape of the Mössbauer scattering line increases with the increase in scatterer thickness and

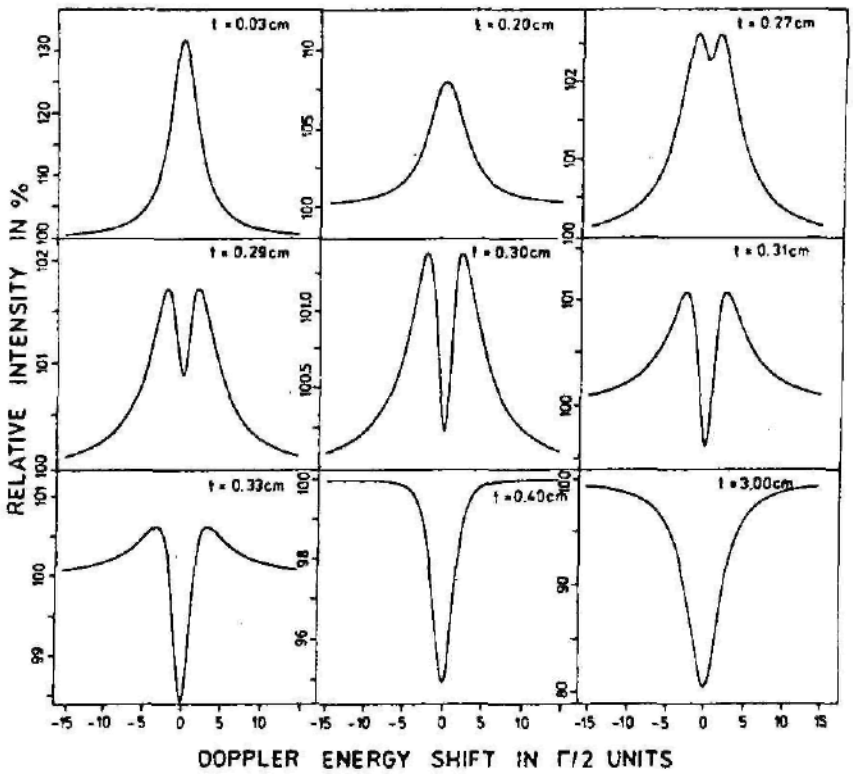

Fig. 2. Scattering lines calculated for hypothetical beryllium scatterers with 0.1 at.% of non-enriched iron impurities. Scatterers of various thicknesses *t* were considered in the calculations.

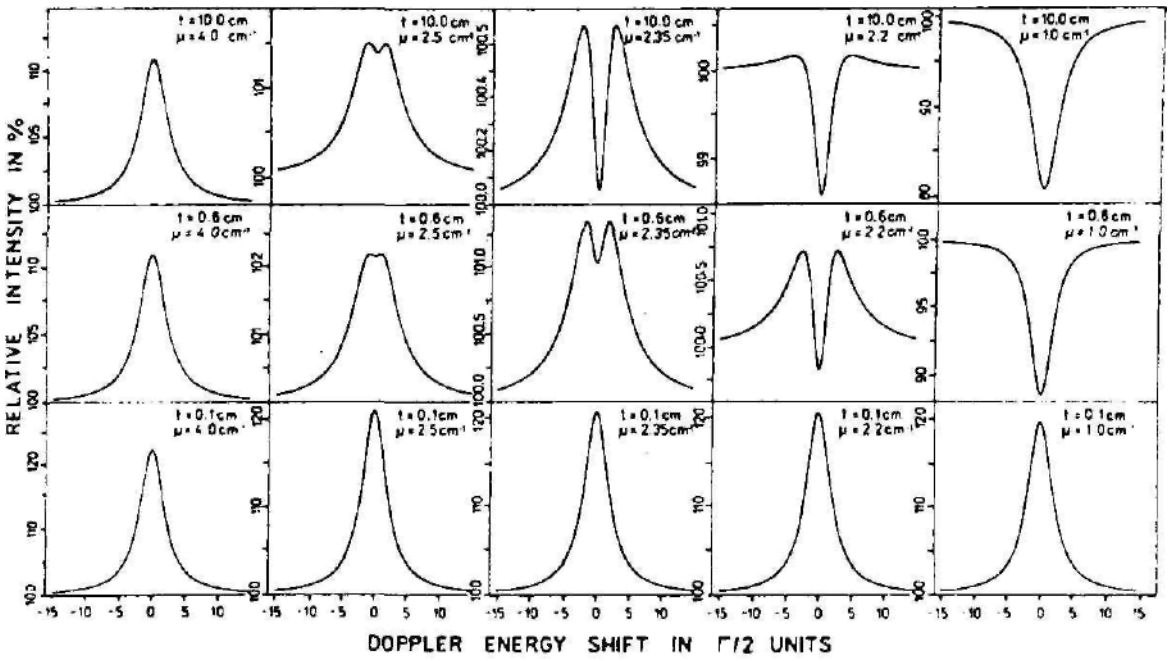

Fig. 3. Scattering lines calculated for hypothetical beryllium scatterers with 0.1 at.% of non-enriched iron impunities. 0.1 cm, 0.6 cm. and 10.0 cm thick scattcrers of various atomic attenuation coefficients μ were considered in the calculations.

the value of the nuclear resonance absorption coefficient μ_r and with the decrease in the value of the electronic absorption coefficient. In order to observe line inversion for the infinitely thick hypothetical beryllium and carbon scatters the iron concentration should not exceed 0.5 at.% and 0.8 at.%. respectively (fig. 6).

4. The experimental results

A 4cmx4cmx4cm beryllium scatterer with some non-enriched iron impurities of a natural origin was used in our preliminary experiments [2]. The influence of the sample thickness on the shape of the

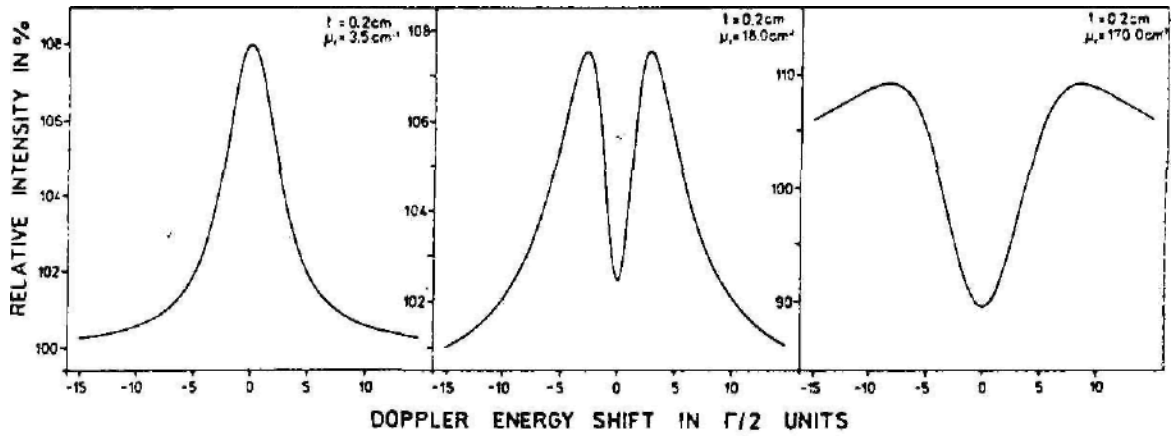

Fig. 4. Scattering lines calculated for hypothetical beryllium scatterers with 0.1 at.% of iron impurities. 0.1 cm thick scatterers with m = 1.0 cm^{-1} and various nuclear resonance absorption coefficients μ_r were considered in the calculations.

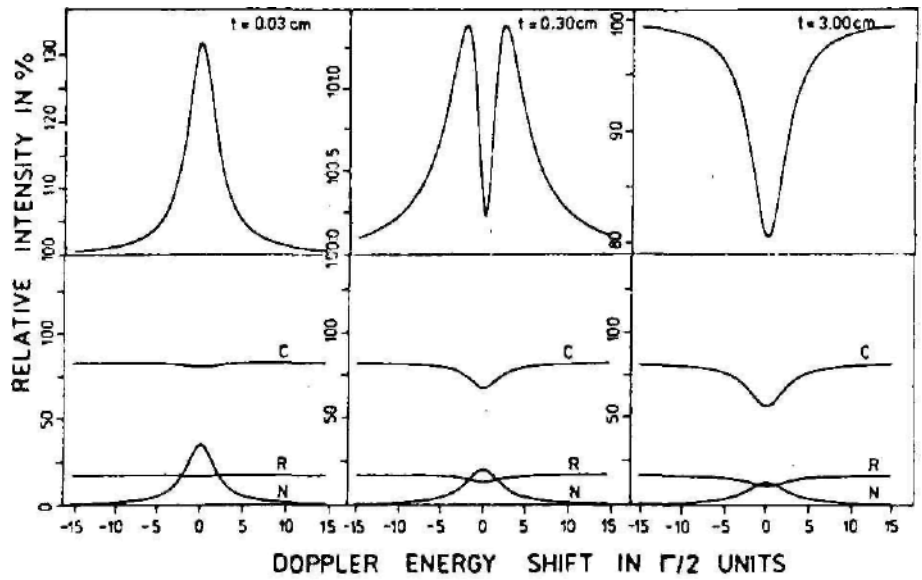

Fig. 5. Scattering lines and their nuclear resonance (N), Rayleigh (R) and Compton (C) contributions calculated for 0.1 at.% of non-enriched iron impurities in 0.03 cm, 0.3 cm and 3.0 cm thick hypothetical beryllium scatlcrcrs.

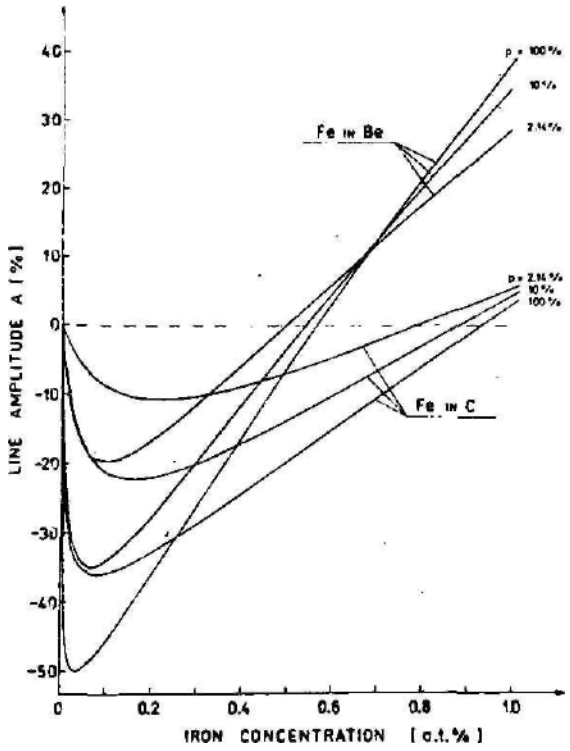

Fig. 6. The amplitudes *A* of Mössbauer scattering lines of infinitely thick hypothetical beryllium and carbon scatterers as function of iron concentration. The calculations were performed for 2.14'%,. 10% and 100%, ^{57}Fe abundances.

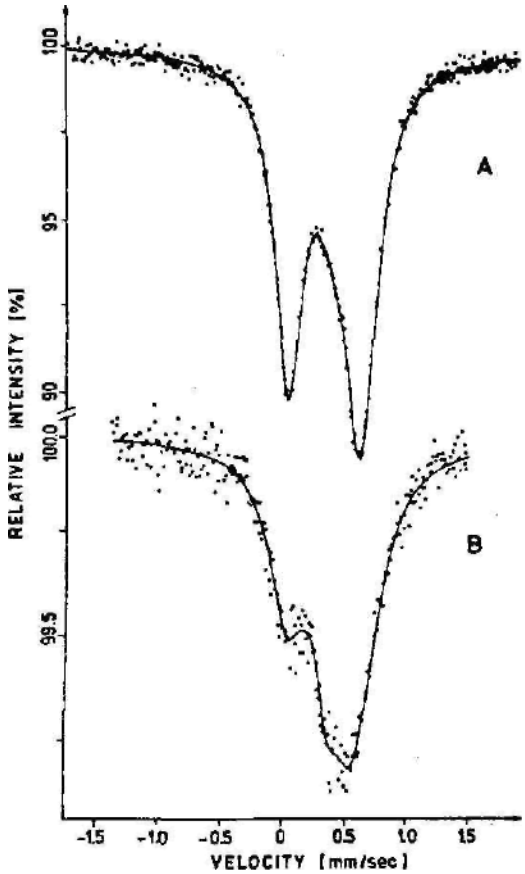

Fig. 7. The Mössbauer absorption spectra measured in transmission geometry for iron impurities in the 4 cm x 4 cm x 4 cm beryllium cube (A) and the set II of beryllium plates (B). The set I of beryllium plates possesses a Mössbauer absorption spectrum quite similar to that of the beryllium cub

scattering line was investigated for two sets (1 and II) of thin beryllium plates. The atomic attenuation coefficient of beryllium for the 14.4 keV gamma rays is very small and originates mostly from Compton and Rayleigh scattering processes. The experiments were performed at room temperature using a ^{57}Co(Cr) source and a constant acceleration Mössbaucr spectrometer of the Polon type. A proportional counter filled to 90% with kryptonium and 10% with methane under a pressure of 650 mm Hg was used as a detector for both 14.4 keV gamma rays and 6.47 keV X-rays. An ordinary 14.4 keV gamma ray Mössbauer absorption spectrum (measured in the transmission geometry) of iron impurities in beryllium is of the form of the very well known asymmetric doublet (fig. 7). Its origin was explained earlier by other investigators [3—6]. The ratio of line amplitudes of the doublet depends on the heat treatment of the sample.

The Mössbauer spectra measured for our samples in scattering geometries are shown in figs. 8-11. The scattered 14.4 keV gamma rays and the 6.47 keV conversion X-rays were detected in the same geometry and by the same counter, and two corresponding Mössbauer spectra (A and B) were simultaneously stored. The conversion X-ray doublets (A) have always a typical scattering shape. However, some of the corresponding spectra (B) obtained by registering scattered gamma radiation possess an unusual shape. With the increase in the scatterer thickness the 14.4 keV scattering spectra are transformed from their scattering shape to the absorption shape. The spectra have their usual scattering shape only for thin samples. The line inversion was observed in spectra of thick samples [figs. 8, 9 and 10 (B_4)]. It appears .earlier for Mössbauer lines corresponding to larger

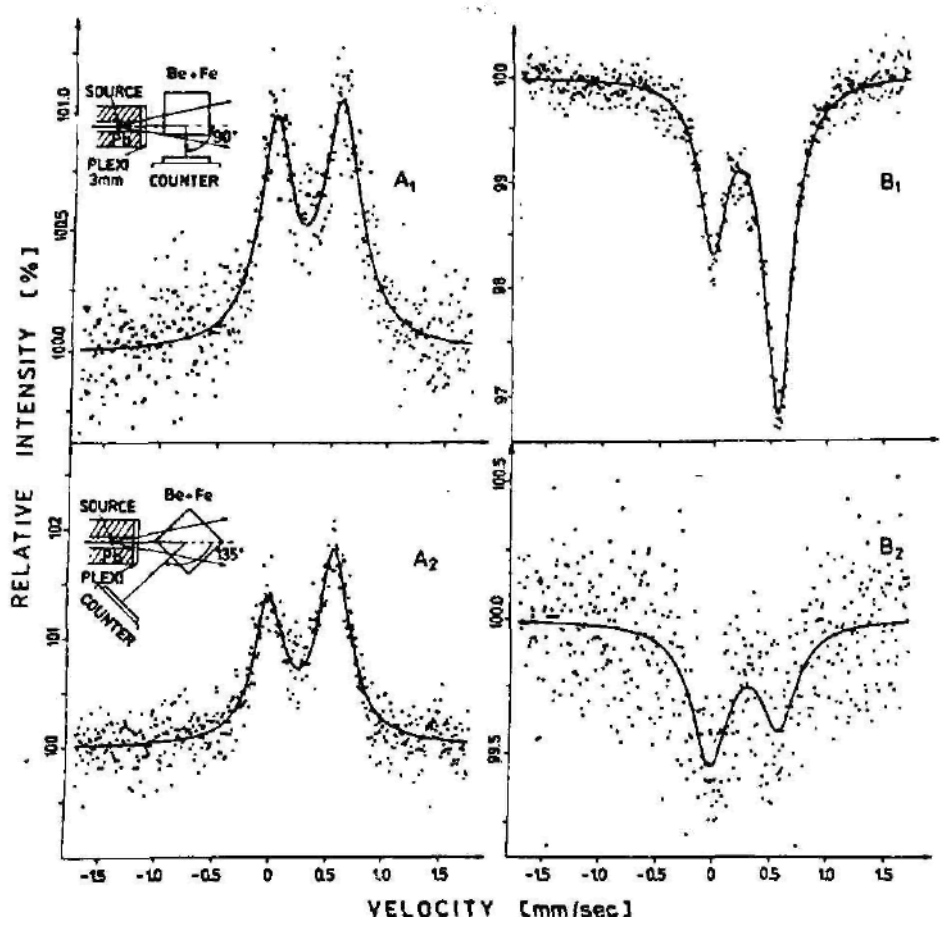

Fig. 8. Mössbauer X-ray (A_1, A_2) and gamma ray (B_1, B_2) spectra measured in the scattering geometries (indicated in the figure) for iron impurities in a 4cm x 4cm x 4cm beryllium cube. The A_1 (A_2) and corresponding B_1 (B_2) spectra were recorded simul-taneously in the same geometry and by the same counter.

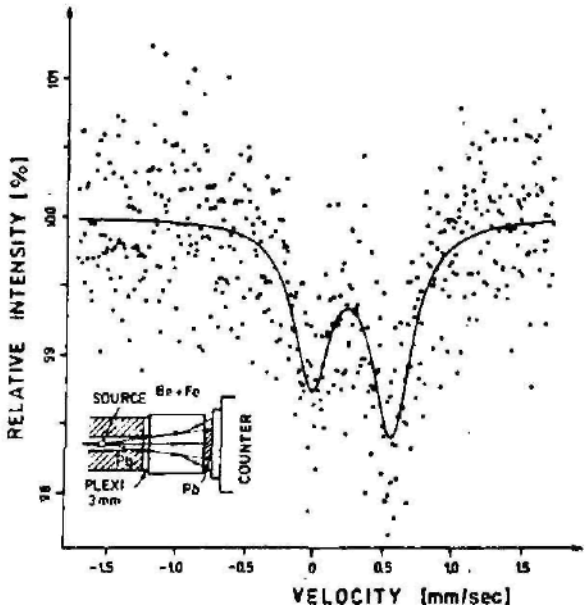

Fig. 9. A quadrupole doublet measured for the 4 cm x 4 cm x 4 cm beryllium cube with some iron impurities. The 14,4 keV gamma rays scttered forward were registered in the experiment.

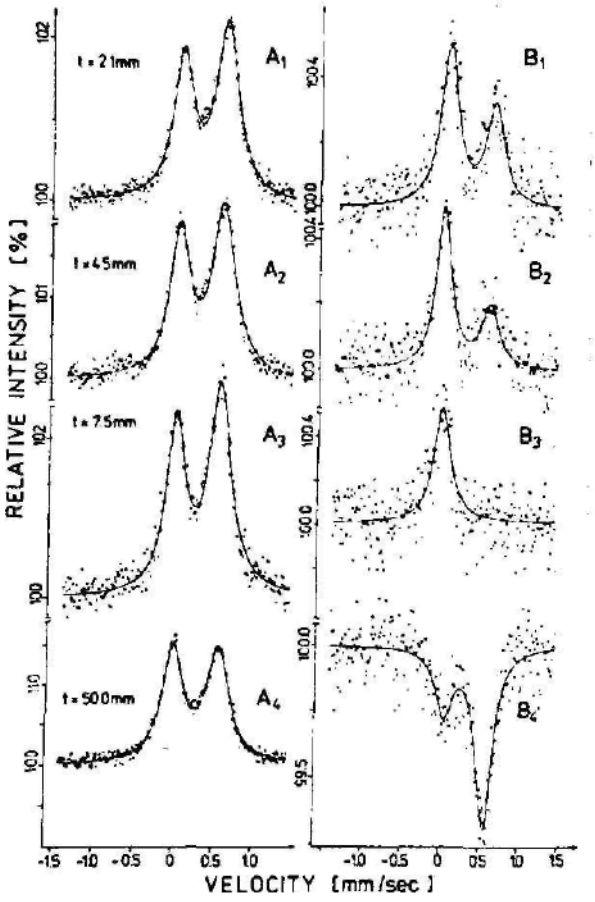

Fig. 10. Mössbauer X-ray (A_1, A_2, A_3, A_4) and gamma ray (B_1, B_2, B_3, B_4) scattering spectra measured at a scattering angle of 130° for iron impurities in set I of beryllium plates. The A_1, A_2, A_3, A_4 and corresponding B_1, B_2, B_3, B_4 spectra were recorded simultaneously in the same geometry and by the same counter.

values of the nuclear resonance attenuation coefficients (right hand side lines of spectra in fig. 11). These observations are in accordance with the results of the numerical calculations shown in figs. 2 and 4.

The shape of the 14.4 keV scattering spectra measured for a beryllium cube depends on its geometrical position with respect to the source and to the detec-tor. The X-ray scattering spectrum recorded for a beryllium cube in the forward scattering geometry was of a bad quality and is not presented. All spectra of set I and set II of beryllium plates (figs. 10 and 11) were recorded at a scattering angle of 130°. A pulse shape discriminator [7] was used to reduce the back-ground and thus to increase the quality of the 14.4 keV scattering spectra. The A_4 (fig. 10) and A_5 (fig. 11) X-ray scattering spectra were also recorded with the application of the pulse shape discriminator. The influence of the sample thickness on the shape of the gamma ray scattering spectra is clearly seen. It should be noticed that only the right hand side lines of the spectra B_3 to B_5 in fig. 11 are inverted. These lines correspond to a larger value of the nuclear resonance attenuation coefficient than the one responsible for the left hand lide lines. The set II of beryllium plates was specially selected to show such a case.

Usually the quality of a gamma ray Mössbauer scattering spectrum is much higher than that of conversion X-rays recorded simultaneously in the same geometry by the same counter. This is not the case when Rayleigh and Compton contributions compensate the nuclear resonance contribution to the gamma ray scattering line. For scatterers composed of light elements the Mössbauer spectrum of the required quality is much more quickly recorded when conversion X-ray instead of scattered 14.4 keV gamma rays are detected.

The inversion of the scattering line was observed earlier [8] at Bragg conditions in Mössbauer diffraction experiments but it was not discussed in detail.

The 14.4 keV scattering spectra of thick samples (figs. 8—11) indicate that the internal conversion electron process diminishes the intensity of gamma rays

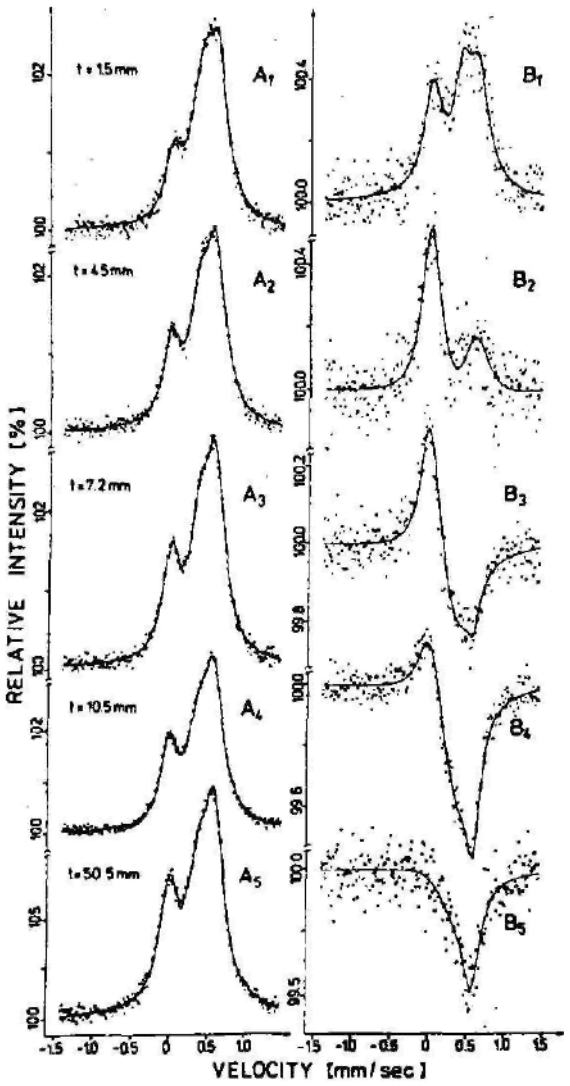

Fig. 11. Mössbauer X-ray (A_1, A_2, A_3, A_4, A_5) and gamma ray (B_1, B_2, B_3, B_4, B_5) scattering spectra measured at a scattering angle of 130° for iron impurities in set II of beryllium plates. The A_1, A_2, A_3, A_4, A_5 and corresponding B_1, B_2, B_3, B_4, B_5 spectra were recorded simultaneously in the same geometry and by the same counter.

scattered at the resonance velocity. Due to this the gamma ray scattering doublets of the absorption shape are observed for iron impurities in beryllium. This is not the case for the conversion X-ray scattering spectra which always have a scattering character.

It should be emphasized that all contributions to the total intensity of scattered gamma rays have to be taken into consideration in analysing the scattering spectra measured for samples composed of light elements. This is specially important in Mössbauer diffraction experiments.

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