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Experimental observations on the resonant amplitude modulation of Mössbauer gamma rays

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Abstract. The spectral distribution of Mössbauer radiation, passed through a resonant medium, which performs additional ultrasonic vibrations is investigated experimentally for thick absorbers. The results are in quantitative agreement with the theory.

1. Introduction

The propagation of Mössbauer gamma radiation through a medium containing resonant nuclei which perform additional vibrations at megahertz frequencies is associated with certain alterations in the spectral distribution of the transmitted radiation. If the dispersion medium is 'thick', i.e. if its effective resonant thickness D is larger than unity, then the main effect modifying the spectrum is the periodic change in the absorption ability of the medium, which causes a periodic variation in the amplitude of the gamma radiation, analogous to the well known amplitude modulation of common electromagnetic waves.

This phenomenon (denoted below as 'resonant amplitude modulation of gamma quanta') has been investigated experimentally by Asher *et al* (1974). These authors observed a splitting of the single absorption line into an infinite set of equidistant lines, shifted from the single line by frequencies which are multiples of the modulation frequency. Asher *et al* (1974) also proposed a qualitative theoretical description of their results.

A more adequate theory of this phenomenon, based on classical dispersion theory, has been published recently (Tsankov 1980a, b, hereafter referred to as I and II); in this paper we report results from the experimental testing of this theory.

2. Theoretical

The theory in general is explained in I, but the formulae obtained in II are more convenient for practical calculations. When the radiation emitted from a source with resonant frequency ω_0 and width Γ is passed through a medium (modulator) with resonant frequency $\omega_0' = \omega_0$ and effective thickness D, which performs as a whole ultrasonic vibrations with amplitude A and frequency $\Omega \gg \Gamma$, then the spectral distribution of the transmitted radiation is described by

$$W(\omega) = \sum_{k=-\infty}^{\infty} W_k(\omega), \tag{1}$$

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$$W_k(\omega) = \frac{\frac{1}{4}\Gamma^2}{(\omega-\omega_0-k\Omega)^2+\frac{1}{4}\Gamma^2} J_0^2(\kappa A) J_k^2(\kappa A)\left[1+\exp\left(-\frac{\frac{1}{4}D\Gamma^2}{(\omega-\omega_0-k\Omega)^2+\frac{1}{4}\Gamma^2}\right)\right.$$
$$\left. -2\exp\left(-\frac{\frac{1}{8}D\Gamma^2}{(\omega-\omega_0-k\Omega)^2+\frac{1}{4}\Gamma^2}\right)\cos\frac{\frac{1}{4}(\omega-\omega_0-k\Omega)D\Gamma}{(\omega-\omega_0-k\Omega)^2+\frac{1}{4}\Gamma^2}\right] \qquad (2)$$

if $k \neq 0$,

$$W_0(\omega) = \frac{\frac{1}{4}\Gamma^2}{(\omega-\omega_0)^2+\frac{1}{4}\Gamma^2}\left[(1-J_0^2(\kappa A))^2 + J_0^4(\kappa A)\exp\left(-\frac{\frac{1}{4}D\Gamma^2}{(\omega-\omega_0)^2+\frac{1}{4}\Gamma^2}\right)\right.$$
$$\left. +2(1-J_0^2(\kappa A))J_0^2(\kappa A)\exp\left(-\frac{\frac{1}{8}D\Gamma^2}{(\omega-\omega_0)^2+\frac{1}{4}\Gamma^2}\right)\cos\frac{\frac{1}{4}(\omega-\omega_0)D\Gamma}{(\omega-\omega_0)^2+\frac{1}{4}\Gamma^2}\right];$$

here $\kappa = 2\pi/\lambda$ stands for the photon wavenumber.

The main effect expected is the splitting of the resonant gamma line. This effect depends on the vibration amplitude of the modulator, and has a maximum in the region of low modulation indices κA. This might be seen from figure 1, where the intensities at the maxima for the unshifted line W_{00} and for the first sideband W_{10} are plotted as functions of the amplitude A for $D = 5$. The amplitude region in which splitting is

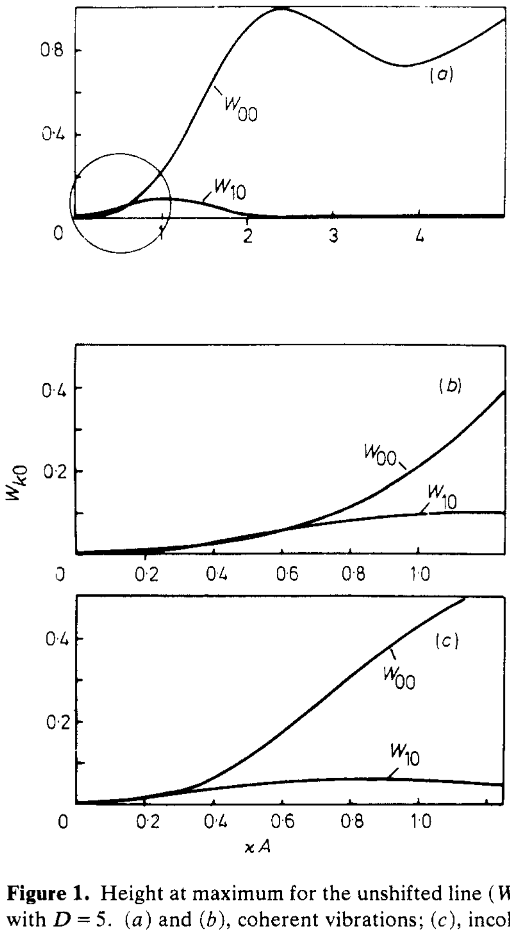

Figure 1. Height at maximum for the unshifted line (W_{00}) and for the first sideband (W_{10}) with $D = 5$. (*a*) and (*b*), coherent vibrations; (*c*), incoherent vibrations.

visible is bounded by a circle in figure 1(*a*); this region is shown in figure 1(*b*) on a larger scale.

The curves shown in figure 1 are valid assuming coherent vibrations of the modulator. However, in most experiments performed so far on frequency modulation of gamma rays, the Mössbauer spectrum observed corresponds to incoherent vibrations of the modulator. Mishory and Bolef (1968) have explained this fact by assuming a very short (10^{-11} s) relaxation time for ultrasonic phonons in crystals. Other reasons for the incoherent vibrations of the sample are connected with the non-homogeneity in the contacting surfaces of the transducer and the absorber, and especially that in the acoustic glue between them. These problems have been investigated recently by Mkrtchyan *et al* (1979), the only authors who have obtained coherent vibrations.

The incoherency of vibrations of the absorber causes a considerable reduction in the splitting effect; this may be seen from figure 1(*c*), where the same dependence as shown in figures 1(*a*) and (*b*) is presented, but this time calculated (by a numerical integration of (2) multiplied by the Rayleigh distribution) for the case of incoherent vibrations of the modulator. The relation W_{10}/W_{00} at $\kappa A = 1{\cdot}0$ is four times smaller in the incoherent case.

Therefore, a preliminary estimation of the degree of coherency of the vibrations of a concrete modulator is rather important; this was the purpose of the first experiment.

3. Determination of the degree of coherency of the vibrations of the modulator and its calibration in terms of amplitudes

The calibration of a modulator in terms of amplitudes consists of determining its mechanical reaction (vibration amplitude A) excited by a given external action (e.g. the amplitude U of the electric field applied to it). This can be obtained, together with the degree of coherency of the vibrations performed, in an experiment analogous to that of Mishory and Bolef (1968).

The experimental set-up consists of a source, modulator and detector. The dependence

$$y(A) = \frac{N_\infty - N_A}{N_\infty - N_0} \tag{3}$$

is measured where N_∞ is the counting rate at 'infinite' Doppler velocity, N_A is the counting rate for the source at rest and the modulator vibrated with amplitude A, and N_0 is the same quantity for both the source and the modulator at rest. In the framework of the theory of frequency modulation it is easy to prove that $y(A) = J_0^2(\kappa A)$ for coherent vibrations and $y(A) = \exp(-\kappa^2 A^2) I_0(\kappa^2 A^2)$ for incoherent vibrations of the modulator. The same result may be deduced from the theory of amplitude modulation if one supposes that N_∞ refers to a system consisting of a source at rest and a modulator vibrated at 'infinite' amplitude (which is, obviously, the same with respect to destruction of the resonance). In the last case, the substitution of equation (17) from II into equation (3) here gives

$$y(A) = \frac{\frac{1}{2}\pi\Gamma(1 - \exp(-\frac{1}{2}D) I_0(\frac{1}{2}D)) J_0^2(\kappa A)}{\frac{1}{2}\pi\Gamma(1 - \exp(-\frac{1}{2}D) I_0(\frac{1}{2}D))} = J_0^2(\kappa A), \tag{4}$$

and analogously for the incoherent case using equation (18) from II.

The important conclusion arising from identity (4) is that $y(A)$ does not depend on the effective thickness of the modulator D. Hence, $y(A)$ is exactly the amplitude behaviour of the central line intensity in a frequency modulated spectrum. This conclusion could not be obtained in the framework of the theory used by Mishory and Bolef (1968) (which is an approximation to the theory of II, valid for thin, 'linear' modulating absorbers, i.e. within $O(D^2)$); however, this is the explanation for the good consistency of their experimental results.

The simplest way to account for the degree of coherency (however, see also Sadikov 1977) is to compose the superposition

$$y(A) = \alpha J_0^2(\beta U) + (1-\alpha)\exp(-\beta^2 U^2) I_0(\beta^2 U^2) \tag{5}$$

where α measures the coherent contribution and $\beta = \kappa A/U$ is the necessary calibration coefficient (assuming a linear transducer).

In our experiments we used a 10 mCi $^{57}Co(Pd)$ source mounted on an electromechanical vibrator for driving with 'infinite' velocity. The modulating absorber is a layer of about 4000 Å of ^{57}Fe, diffused into a 20 μm thick Pd foil. The absorber is glued to an AT-cut piezoelectric quartz transducer. The operating frequency was fixed at 32·0 MHz, almost the same as the third resonance of the crystal (31·7 MHz).

The electric signal is obtained from a C4-31 frequency synthesiser (frequency drift 0·01 Hz) and is then amplified by a home-made resonance amplifier. The maximal value of the output voltage was monitored by a Tektronix 465 oscilloscope using a capacitive attenuator.

A xenon proportional counter was used to register transmitted radiation. The commutation of the source velocity 'zero-infinity' was repeatedly processed using a home-made device, aimed at avoiding the linear part of the equipment drift.

The results of the measurements performed were fitted to the function (5) with α and β as free parameters. The best approximation corresponds to $\alpha = -0{\cdot}006 \pm 0{\cdot}04$ and $\beta = 0{\cdot}0290 \pm 0{\cdot}0003$ Å V^{-1}; hence the oscillations of the modulator constructed are purely incoherent. This is to be expected, since no efforts, such as polishing, choice of the acoustic glue, etc., have been made to improve the acoustic contact between the surfaces of the quartz and the absorber (see Mkrtchyan *et al* 1979).

4. Spectrum of the amplitude modulated radiation

The experimental arrangement now consists of a source at rest, a modulator, a secondary (analysing) absorber and detector. The analysing absorber is a foil of $^{57}Fe(Pd)$, mounted on a light frame, and connected to the vibrator of a precision Mössbauer spectrometer (Ormandjiev *et al* 1979) via an aluminium tubule.

Certain preventive measures were taken to remove the proper mechanical vibrations arising in the connection system, and it was verified that as a result the linearity of the velocity scale of the spectrometer did not change.

The geometry just described is completely equivalent in its theoretical aspects with that used by Asher *et al* (1974), who used an analysing absorber at rest, with the source and modulator connected together with the Mössbauer vibrator. However, in the last case it is possible that there is partial acoustic transmission of ultrasonic vibrations from the modulator to the source through the (necessarily) strong packing between them; therefore it is possible that a splitting in the spectrum due to the frequency modulated source radiation may be observed. This possibility is supported by the fact that the splitting effect in the experiments of Asher *et al* does not fade away when the amplitude applied to the transducer increases. The possibility of similar side effects is completely eliminated in the case of a source at rest, since then the source and the modulator are distant from each other.

The spectrum of the amplitude modulated radiation is shown in figures 2(*a*) and (*b*) for several values of the vibration amplitude A. The splitting effect is very poor, due to the incoherency of vibrations. Nevertheless, the results are in general agreement with the theory. This is better seen from figure 3, where the maximum height of the first sideband W_{10} is plotted as a function of κA. The experimental points refer to the area of the first satellite (the results for the left- and right-hand lines are averaged), normalised to the area of the central line corresponding to the maximum value of κA used. The full curve is the theoretical one for incoherent vibrations, calculated for the thickness parameter $D = 3{\cdot}92$, the value obtained from the previous experiment.

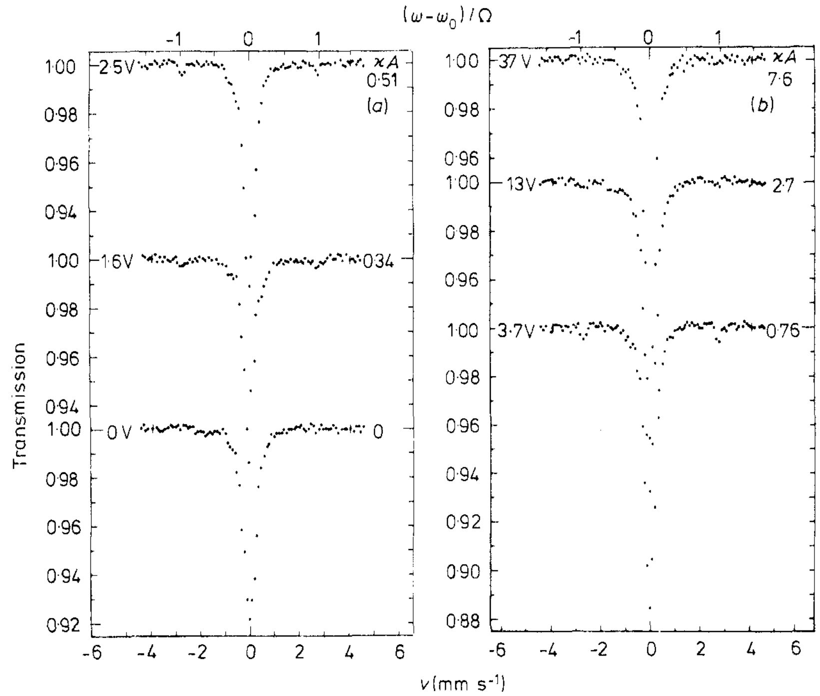

Figure 2. Spectrum of the amplitude modulated radiation for several values of vibration amplitude A.

Attempts to observe the higher-order sidebands in the spectrum were not successful.

The statistical reliability of the results, plotted in figure 3, is obviously insufficient to evaluate any quantitative agreement between the experiment and the theory. Therefore a new experiment was performed, aimed at determining the relative transmission of the modulator in the region of the unshifted Mössbauer line.

The experimental set-up in this case is the same as that used in the previous experiment (i.e. the source is at rest); however, in the present case the analysing

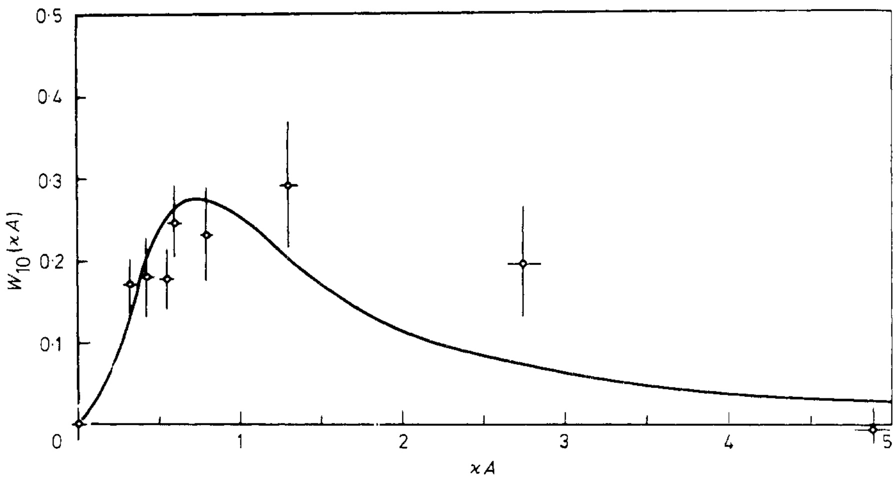

Figure 3. Intensity of the first satellite as a function of the amplitude of vibration of the modulator; the full curve is a theoretical curve calculated for $D = 3{\cdot}92$.

absorber is either moved at 'infinite' Doppler velocity, or it is at rest. The measured quantity is

$$y(A) = (N_\infty - N_0)_A \tag{6}$$

where A is the vibration amplitude of the modulator. Because of the weak splitting effect, $y(A)$ may be fitted to a good approximation by an expression proportional to the

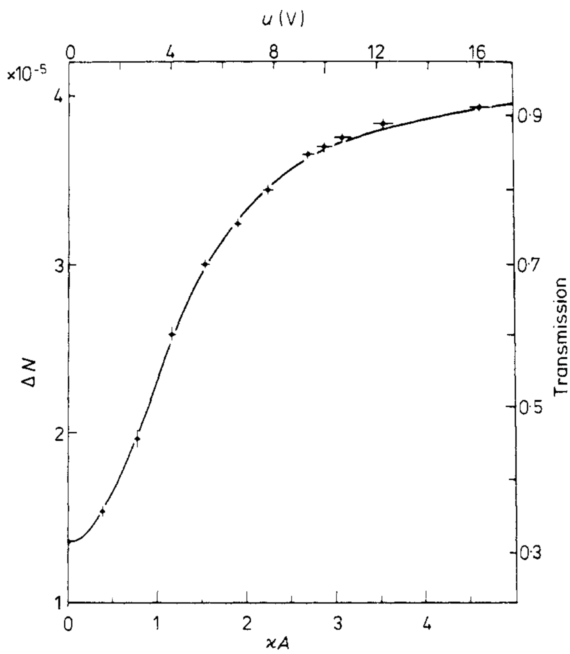

Figure 4. Resonant transmission of the modulator in the region of the unshifted Mössbauer line as a function of vibration amplitude A. $D = 3{\cdot}92 \pm 0{\cdot}046$.

area of the modulated spectrum (see II, equation (18)):

$$y(A) = C[1 - \exp(-\kappa^2 A^2) I_0(\kappa^2 A^2)(1 - \exp(-\tfrac{1}{2}D) I_0(\tfrac{1}{2}D))]; \qquad (7)$$

here C is a normalising constant, and D is the effective thickness of the modulator.

The results are shown in figure 4. The full curve is derived from a fit of the experimental points according to (7) with the free parameters C and D, and using the calibration coefficient obtained from the first experiment. The corresponding value of the functional χ^2 is 8·81 for ten degrees of freedom; therefore one may consider the results from the experiments performed to be in quantitative agreement with the theory proposed for the resonant amplitude modulation of Mössbauer gamma quanta.

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