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The spectrum of Mössbauer radiation passed through a vibrating resonant medium

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Abstract. The spectrum of the Mössbauer gamma-radiation, passed through a resonant absorber vibrated at ultrasonic frequencies, is studied in a new approach. An analytical expression is derived for the line profile, which consists of an infinite series of equidistant satellites. Their intensities depend on both the vibration amplitude and the absorber thickness. Certain ideas appearing in the investigations so far are critically discussed. Most of the phenomena considered can be explained by the classical waves dispersion theory.

1. Introduction

It is well known that the additional RF vibrations, when applied to either the source or the absorber in conventional Mössbauer experiments, modify the line shape of the resonantly transmitted radiation, resulting in a splitting of the original single line into an infinite set of sidebands, equidistantly spaced from it by frequencies which are multiples of Ω, the frequency of the external perturbation (Ruby and Bolef 1960). This phenomenon can be explained adequately from either a wave point of view, in terms of a frequency modulation of the photon wave (see the review of Makarov and Mitin (1976)), or from a corpuscular one—in terms of creation and annihilation of acoustic phonons which interact with the gamma-radiation field (Mishory and Bolef 1968). Both approaches lead to the same results, which is a remarkable development of the corpuscle–wave dualism.

Extending this analogy, certain experimental investigations have appeared in the last few years (Asher *et al* 1974, Cashion and Clark 1979), concerning another phenomenon that might be classified as an 'amplitude modulation of gamma quanta due to time-dependent resonant processes'. In the experiments of Asher *et al*, Mössbauer gamma-radiation, irradiated by a single line source, is passed through a filter containing resonant nuclei to which certain constrained RF oscillations could be applied by a piezo-electric transducer, and the emerging radiation is analysed by a second absorber using conventional techniques. In such a case the vibrating filter changes its resonant absorption ability periodically for a time which is comparable with the lifetime of the excited Mössbauer state, causing a substantial change in the spectral distribution of the radiation passed in a manner analogous to the amplitude modulation of the common electromagnetic waves.

In the experiments of Cashion and Clark, the source is also performing ultrasonic vibrations with respect to the lab frame, in-phase or in anti-phase with the modulating absorber. Thus, in this case both the amplitude and the phase modulation are responsible for the spectrum observed.

A splitting of the spectrum into a set of equidistant satellites has been observed in both experiments. The results of Asher *et al* have been explained by the authors qualitatively using the optical theorem, applied to the forward-scattered component of the radiation. However, their approach yields an infinite set of coupled differential equations for the partial amplitudes, which can be solved in a few special cases, in particular when the modulating absorber is thin. At the same time, it is beyond doubt that the expected splitting effect should be increasing with the modulator thickness.

This difficulty may be avoided, and an attempt is proposed in the present work to describe quantitatively the phenomena just discussed.

The necessary results may be obtained using the theory developed by Lynch *et al* (1960) and Harris (1961) for the transmission of the Mössbauer radiation through a stationary resonant absorber. Lynch *et al* have performed a simple classical treatment, assuming the absorber to be composed of a collection of single-level damped harmonic oscillators; then each frequency component of the source radiation field is changed in amplitude and phase as it passes through the absorber, according to a frequency-dependent index of refraction. The pure quantum-mechanical calculations of Harris confirm perfectly the results from the classical theory. An explanation of the surprising circumstance that it should be possible to provide a macroscopic description of the absorber in this case is suggested by Thieberger *et al* (1968) by the fact that the average contribution modifying the incident wave at a point inside the absorber is due to the mutually in-phase, coherently forward-scattered waves from the oscillators.

2. Theory

So, we are interested in the spectral distribution $W(\omega)$ of the gamma radiation from a single line Mössbauer source, at rest in the lab frame, which emerges through a resonant absorber (modulator) with an effective thickness D, performing as a whole constrained harmonic oscillations at frequency $\Omega/2\pi$ and amplitude A.

It is convenient to consider for a time a frame at rest with the resonant absorbing nuclei of the modulator. In this frame, the time dependence of the source radiation would be

$$a(t)=\mathrm{e}^{\mathrm{i}\omega_0 t-\Gamma t/2}\,\mathrm{e}^{\mathrm{i}\varkappa A\sin\Omega(t+\eta)},\tag{1}$$

where $\varkappa=2\pi/\lambda$ is the wavenumber of the photon, ω_0 and Γ are the resonant frequency and the halfwidth of the irradiating nuclear state, and η is the phase of the source-to-modulator vibrations at $t=0$.

The Fourier decomposition of (1) is

$$a(\omega)=\sum_{k=-\infty}^{\infty}\mathrm{J}_k(\varkappa A)\frac{\mathrm{e}^{\mathrm{i}k\Omega\eta}}{\Gamma/2+\mathrm{i}(\omega-\omega_0-k\Omega)}.\tag{2}$$

The modulating filter is at rest in the current frame, so that each Fourier component is modified, as it passes through it, in accordance with the theory of Lynch *et al*:

$$a'(\omega)=a(\omega)\exp\left(\frac{\mathrm{i}D\Gamma/4}{\omega-\omega_0'-\mathrm{i}\Gamma/2}\right),\tag{3}$$

where $\hbar\omega_0'$ denotes the absorber's resonant energy.

The corresponding amplitude in the time domain is

$$a'(t)=\int_{-\infty}^{\infty}\mathrm{d}\omega\, a'(\omega)\,\mathrm{e}^{\mathrm{i}\omega t};$$

this integral may be solved analogously to that of Hamermesh (Lynch *et al* 1960):

$$a'(t)=\sum_{k=-\infty}^{\infty}\mathrm{J}_k(\varkappa A)\,\mathrm{e}^{\mathrm{i}k\Omega\eta}\exp(\mathrm{i}\omega_0't-\tfrac{1}{2}\Gamma t)\times\sum_{n=0}^{\infty}\left[\frac{\mathrm{i}(\omega_0-\omega_0'+k\Omega)}{D\Gamma/4}\right]^n(D\Gamma/4t)^{n/2}\mathrm{J}_n[2(D\Gamma/4t)^{1/2}],\tag{4}$$

or, alternatively,

$$a'(t)=\sum_{k=-\infty}^{\infty}\mathrm{J}_k(\varkappa A)\,\mathrm{e}^{\mathrm{i}k\Omega\eta}\Bigg[\exp[\mathrm{i}(\omega_0+k\Omega)t-\tfrac{1}{2}\Gamma t]\exp\left(\frac{\mathrm{i}D\Gamma/4}{\omega_0-\omega_0'+k\Omega}\right)-\exp(\mathrm{i}\omega_0't-\tfrac{1}{2}\Gamma t)\sum_{n=1}^{\infty}\left(\frac{\mathrm{i}D\Gamma/4}{\omega_0-\omega_0'+k\Omega}\right)^n(D\Gamma/4t)^{-n/2}\mathrm{J}_n[2(D\Gamma/4t)^{1/2}]\Bigg].\tag{4'}$$

The choice between (4) and (4′) is determined by convergence requirements for the sum over n.

At this stage it is possible to return in the lab frame:

$$a''(t)=a'(t)\exp[-\mathrm{i}\varkappa A\sin\Omega(t+\eta)].\tag{5}$$

Now, we need the Fourier transform of (5). It has the form

$$a''(\omega)=\int_0^{\infty}\mathrm{d}t\,\mathrm{e}^{-\mathrm{i}\omega t}a''(t)=\sum_{l=-\infty}^{\infty}\mathrm{J}_l(\varkappa A)\,\mathrm{e}^{-\mathrm{i}l\Omega\eta}\int_0^{\infty}\mathrm{d}t\,\mathrm{e}^{-\mathrm{i}(\omega+l\Omega)t}a'(t).\tag{6}$$

Denoting in (6)

$$x=(D\Gamma/4t)^{1/2},\qquad \alpha=\frac{\Gamma/2+\mathrm{i}(\omega-\omega_0'+l\Omega)}{D\Gamma/4},$$

one comes to the integral

$$A_n=\frac{2}{D\Gamma/4}\int_0^{\infty}\mathrm{d}x\,\mathrm{e}^{-\alpha x^2}x^{n+1}\mathrm{J}_n(2x).\tag{7}$$

This integral may be solved and the result is (Gradsteyn and Ryžhik 1963)

$$A_n=\frac{2}{D\Gamma/4}\frac{2^n\Gamma(n+1)}{2^{n+1}\Gamma(n+1)\alpha^{n+1}}\,{}_1F_1\left(n+1;n+1;\frac{-4}{4\alpha}\right),\tag{8}$$

where ${}_1F_1(\alpha;\mu;z)$ is the confluent hypergeometric function. Using the relation ${}_1F_1(\alpha;\alpha;z)=\exp(z)$, one obtains

$$a''(\omega)=\sum_{k=-\infty}^{\infty}\sum_{l=-\infty}^{\infty}\mathrm{J}_k(\varkappa A)\mathrm{J}_l(\varkappa A)\,\mathrm{e}^{\mathrm{i}(k-l)\Omega\eta}[\Gamma/2+\mathrm{i}(\omega-\omega_0'+l\Omega)]^{-1}\times\exp\left(-\frac{D\Gamma/4}{\Gamma/2+\mathrm{i}(\omega-\omega_0'+l\Omega)}\right)\sum_{n=0}^{\infty}\frac{[\mathrm{i}(\omega_0-\omega_0'+k\Omega)]^n}{[\Gamma/2+\mathrm{i}(\omega-\omega_0'+l\Omega)]^n}.\tag{9}$$

The sum over n appearing in (9) is a geometric progression, which is convergent for each ω if $|\omega_0 - \omega_0' + k\Omega| < \Gamma/2$. If this condition is not satisfied, it is necessary to use the alternative expression (4′) for $a'(t)$. Then the corresponding integral would have the form

$$B_n = \frac{2}{D\Gamma/4} \int_0^\infty \mathrm{d}x \, \mathrm{e}^{-\alpha x^2} x^{-n+1} \mathrm{J}_n(2x)$$

$$= \frac{2}{D\Gamma/4} \frac{2^n \Gamma(1)}{2^{n+1}\Gamma(n+1)\alpha} \, {}_1F_1\left(1; n+1; \frac{-4}{4\alpha}\right). \tag{10}$$

Using the power series expansion of the function ${}_1F_1$,

$${}_1F_1(1; n+1; -\beta) = 1 - \frac{1}{n+1}\frac{\beta}{1!} + \frac{1\,.\,2}{(n+1)(n+2)}\frac{\beta^2}{2!} - \ldots,$$

it is easy to prove that the following is true:

$$\frac{1}{n!}\, {}_1F_1(1; n+1; -\beta) = \sum_{p=0}^{\infty} \frac{(-\beta)^p}{(n+p)!},$$

where $\beta = 1/\alpha$.

Combining (9) and (10), one may write

$$a''(\omega) = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} \mathrm{J}_k(\varkappa A)\mathrm{J}_l(\varkappa A)\, \mathrm{e}^{\mathrm{i}(k-l)\Omega\eta} C_{kl}, \tag{11}$$

where

$$C_{kl} = \{\Gamma/2 + \mathrm{i}[\omega - \omega_0 - (k-l)\Omega]\}^{-1} \exp\left(-\frac{D\Gamma/4}{\Gamma/2 + \mathrm{i}(\omega - \omega_0' + l\Omega)}\right)$$

$$\text{if } |\omega_0 - \omega_0' + k\Omega| < \Gamma/2,$$

$$C_{kl} = \{\Gamma/2 + \mathrm{i}[\omega - \omega_0 - (k-l)\Omega]\}^{-1} \exp\left(\frac{\mathrm{i}D\Gamma/4}{\omega_0 - \omega_0' + k\Omega}\right)$$

$$- \sum_{n=1}^{\infty} \left(\frac{\mathrm{i}D\Gamma/4}{\omega_0 - \omega_0' + k\Omega}\right)^n \sum_{p=0}^{\infty} \frac{(-D\Gamma/4)^p}{(n+p)!\,[\Gamma/2 + \mathrm{i}(\omega - \omega_0' + l\Omega)]^{p+1}}$$

$$\text{if } |\omega_0 - \omega_0' + k\Omega| \neq 0. \tag{12}$$

Finally, the spectrum of interest may be obtained by squaring the modulus of (11) and averaging the result over the initial phase η. The last operation yields certain additional restrictions of the form

$$\frac{1}{2\pi} \int_0^{2\pi} \mathrm{d}y \, \exp[\mathrm{i}(k - l - k' + l')y] = \delta_{k-l,k'-l'}. \tag{13}$$

3. Results and discussion

Results from the numerical calculations performed are listed in figures 1 to 4. All the spectra are convoluted by a single natural width Lorentzian in order to demonstrate what may be seen in a real experiment.

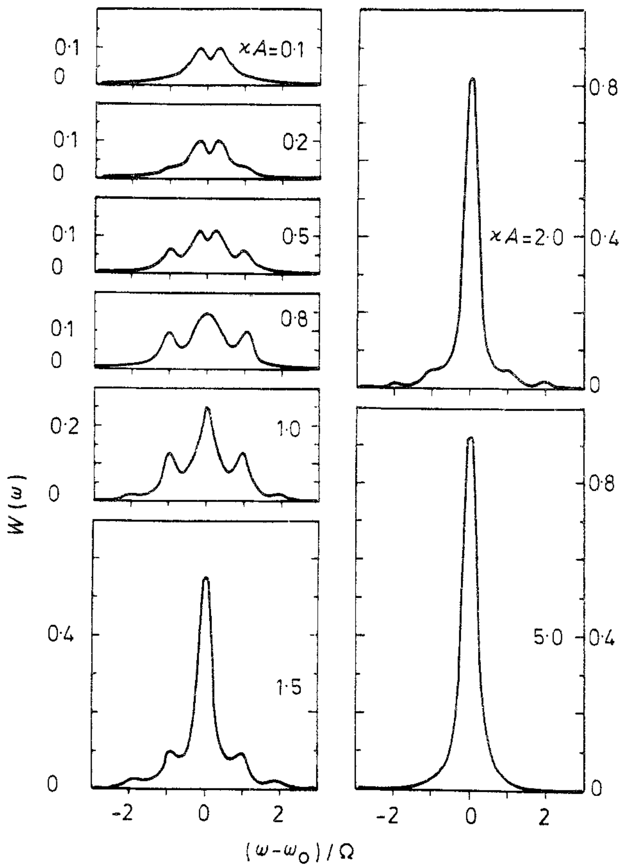

Figure 1. The spectrum of the transmitted radiation for $D = 5$, $\omega_0' - \omega_0 = 0$ and for several amplitudes of vibration.

It is seen from (11) that the spectrum consists of an infinite set of satellite lines, shifted from the source line in frequency by multiples of Ω. We would like to discuss here some essential features of the spectrum.

(a) If $\omega_0' - \omega_0 = 0$ (i.e. no isomer shift between the source and the modulating absorber), the spectrum is symmetric with respect to ω_0. In fact, if one writes (11) in the general form

$$a''(V) = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} D_{kl}, \tag{14}$$

where $V = \omega - \omega_0 = \omega - \omega_0'$, then, replacing V by $-V$, k by $-k$, l by $-l$, and using the well known property $\mathrm{J}_{-n}(z) = (-1)^n \mathrm{J}_n(z)$, one obtains

$$a''(-V) = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} D_{kl}^*(-1)^{k+l}.$$

Then, obviously,

$$W(V) = \sum_k \sum_l \sum_{k'} \sum_{l'} D_{kl} D_{k'l'}^* \delta_{k-l,k'-l'}$$

$$W(-V) = \sum_k \sum_l \sum_{k'} \sum_{l'} D_{kl}^* D_{k'l'} (-1)^{k+l+k'+l'} \delta_{k-l,k'-l'}.$$

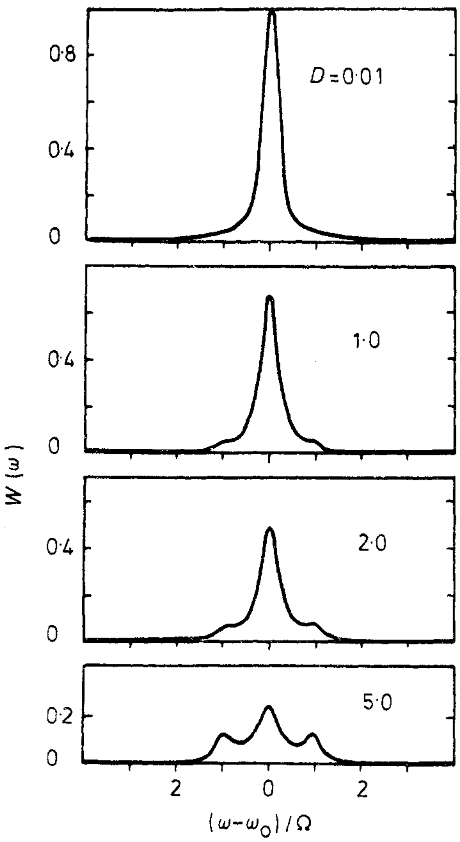

Figure 2. The spectrum of the transmitted radiation for amplitude $\varkappa A = 1$, $\omega_0' - \omega_0 = 0$ and for several thickness parameters.

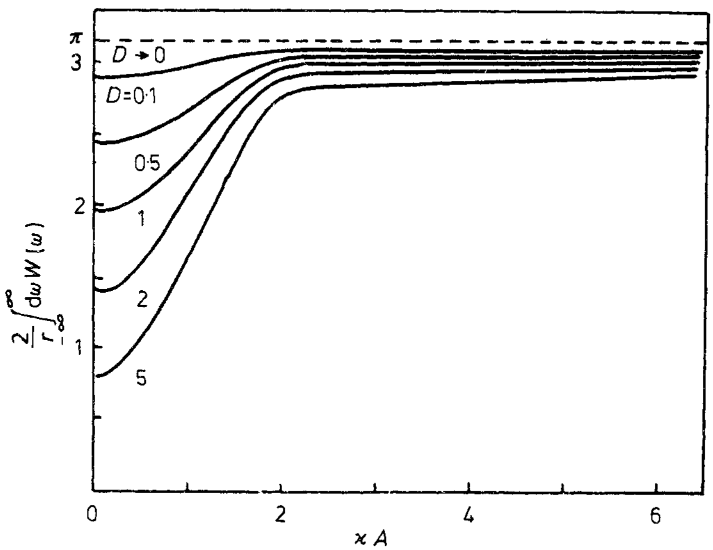

Figure 3. The spectrum area for $\omega_0' - \omega_0 = 0$ against the vibration amplitude $\varkappa A$ and the absorber thickness D.

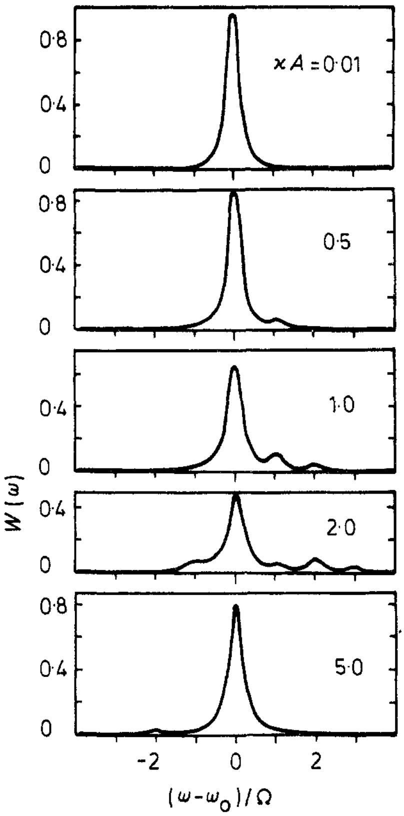

Figure 4. The spectrum of the transmitted radiation for $D = 5$, $\omega_0' - \omega_0 = \Omega$ and for several amplitudes of vibration.

The quantity $k + l + k' + l'$ is always an even number due to the presence of the δ symbol; thus $W(-V) = W(V)$.

(b) If the modulating filter is at rest (i.e. $A = 0$), then (11) describes the well known spectrum of the radiation passed through a thick stationary absorber:

$$W(\omega)|_{\varkappa A=0} = [(\omega - \omega_0)^2 + \Gamma^2/4]^{-1} \exp\left(-\frac{D\Gamma/4}{(\omega - \omega_0')^2 + \Gamma^2/4}\right). \tag{15}$$

In this case a typical fallout of the line profile (which is clearly observable in figure 1 for $\varkappa A < 1$) appears at the absorber's resonant frequency ω_0', because of the resonance self-absorption of the radiation. Its depth increases with the absorber thickness D.

If $\varkappa A \to \infty$, then the spectrum is reduced to a single natural width Lorentzian, centred at ω_0 (see figure 1, $\varkappa A = 5$). Therefore, a region in the vibration amplitudes exists wherein the expected splitting effect is maximal. This prediction is in agreement with the experiments of Asher *et al.* Numerical calculations indicate that this region is at $\varkappa A \simeq 1{\cdot}0$.

(c) When the effective thickness of the absorber is small, $D \to 0$, then the spectrum tends to a single Lorentzian line:

$$a''(\omega)|_{D=0} = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} \mathrm{J}_k(\varkappa A)\mathrm{J}_l(\varkappa A) \frac{1}{\Gamma/2 + \mathrm{i}[\omega - \omega_0 - (k-l)\Omega]} = \frac{1}{\Gamma/2 + \mathrm{i}(\omega - \omega_0)}. \tag{16}$$

The effect of the amplitude modulation of the gamma radiation increases with the modulator thickness D. This is seen from figure 2, where the spectra at the same amplitude ($\varkappa A = 1$) are presented for several values of D, as well as from figure 3, where the spectrum area is plotted against the vibration amplitude A for several values of D. The last diagram might serve for evaluating the modulator thickness or its vibration amplitude, if one of these characteristics and the area under the experimental line are known.

(d) The spectrum of Mössbauer radiation, transmitted through a vibrated filter which has a non-zero isomer shift with respect to the source, $\omega_0' - \omega_0 = \Omega$, is shown in figure 4. This case refers to the second experiment of Asher *et al*, when the source radiation resonantly excites the modulator substate, corresponding to annihilation of one phonon. The spectrum is asymmetric, which is more significant at low modulation indices, $\varkappa A < 2$. However, the relative intensities of sidebands with respect to the unshifted line are considerably smaller in this case, making the splitting effect difficult to observe.

(e) The experiments of Cashion and Clark can also be described in the framework of the formalism proposed. Since the source has also been vibrated ultrasonically in this case, then the spectrum of the registrated radiation would always consist of additional lines irrespective of the vibration state of the absorber, due to the frequency modulation. If the source and the filter are vibrated in-phase, then the line shape of the radiation passed would be

$$W(\omega)_{\text{in}} = \sum_{k=-\infty}^{\infty} \mathrm{J}_k^2(\varkappa A)[(\omega - \omega_0 - k\Omega)^2 + \Gamma^2/4]^{-1} \exp\left(-\frac{D\Gamma^2/4}{(\omega - \omega_0' - k\Omega)^2 + \Gamma^2/4}\right), \qquad (17)$$

and in the case of an anti-phase motion the spectrum would have the form

$$W(\omega)_{\text{anti}} = \left\langle \left| \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} \mathrm{J}_k(2\varkappa A)\mathrm{J}_l(\varkappa A)\, \mathrm{e}^{\mathrm{i}(k-l)\Omega\eta} C_{kl} \right|^2 \right\rangle, \qquad (18)$$

where C_{kl} is defined by (12), and the symbol $\langle \ldots \rangle$ stands for the necessary averaging over the initial phase η.

It is important to stress that the spectrum in this case would also be symmetric, if there is no isomer shift between the source and the modulator ($\omega_0' - \omega_0 = 0$). This assertion may be proved in an analogous manner to that in (a). The asymmetry of the spectrum, observed by Cashion and Clark, may be explained quite naturally by the presence of a non-zero isomer shift. The source ^{57}Co(Cu) has an isomer shift of $+0{\cdot}31$ mm s^{-1} with respect to the stainless steel absorber; hence, the negative sidebands of the source radiation are absorbed, in general, more significantly than the positive ones. This qualitative conclusion might be checked by experimenting with a source and absorber without isomer shift, when the spectrum would be symmetric, as well as by using a source with a negative isomer shift with respect to the absorber, when the negative sidebands of the spectrum would be dominant, in the framework of the present approach (see equation (18)).

(f) The theory developed in this work may be extended to describe the actual experiments of Asher *et al*, considering a combination of a source at rest and two absorbers vibrating in anti-phase. It may be proved that there is no asymmetry in the first of Asher's experiments (using Pd(Fe) filters). So, one may deduce that the presence of a certain isomer shift between the components of the considered system is the reason for observation of asymmetry in the spectrum rather than the retention of phase coherence.

As we have assumed that the source nuclei are being vibrated as a whole, then the effects of reduction of the resonance self-absorption of the radiation due to standing waves in the (eventually) vibrating source could not be of account in the present approach. These effects are only significant at high acoustic powers of the ultrasonic field applied (see Mishory and Bolef (1968) and Mitin (1978)).

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