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TRANSMISSION AND LINE BROADENING OF RESONANCE RADIATION INCIDENT ON A RESONANCE ABSORBER†

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The transmission of resonance radiation emitted from a source of finite thickness and passing through an external resonance absorber is discussed The transmission integral is examined for both a linear and a Gaussian distribution of radioactive atoms in the source Several special cases are presented, and the general case is evaluated by numerical integration Over the range of values considered, the transmitted line shape closely approximates a Breit-Wigner curve whose width is greater than the natural width of the transition This line broadening, caused by resonance absorption in the source and in the external absorber, is presented graphically as a function of source and absorber thickness

1. Introduction

R. L. Mossbauer's discovery that recoilless emission and absorption of nuclear gamma radiation can occur[1]) has stimulated a host of recent investigations[2]). Interest in this process has grown rapidly because the Mossbauer effect allows the direct observation of many phenomena formerly thought unmeasurable. A terrestial measurement of the gravitational red-shift[3]), a test of the equivalence principle for rotating systems[4]), and the observation of the Zeeman splitting of excited nuclear levels[5]), are but a few of the experiments made possible by the Mössbauer effect. In addition, the effect is finding many applications in the measurement of the internal fields in solids.

When recoilless emission and absorption of gamma radiation occurs, the conditions for nuclear resonance fluorescence are inherently satisfied. The numerous applications of the Mössbauer effect follow from the fact that, in such cases, the very narrow lines resulting from transitions from metastable nuclear levels can actually be observed: resonance lines with widths in the range 10^{-10} to 10^{-5} eV, which correspond respectively to gamma transitions from levels whose half-lives vary between 10^{-5} and 10^{-10} sec, have been measured.

Although experiments involving the Mossbauer effect can be performed with either a transmission or a scattering geometry, most of the work done so far has employed the former approach[6]). A typical transmission experiment consists of measuring the resonance radiation passing through a resonance absorber, as a function of the relative velocity between source and absorber. In this manner, a resonance line shape is traced out. Since the great utility of the Mössbauer effect depends upon the measurement of such lines, a consideration of the line shape is of interest. In what follows, we will be concerned with the transmission

† This work has been supported in part by the joint program of the U S Office of Naval Research and the U S Atomic Energy Commission

[1]) R L Mossbauer, Z Physik 151 (1958) 124

[2]) For a listing of recent experiments, see the bibliography contained in Proceedings of the Allerton Park Conference on Mossbauer Effect, University of Illinois, Urbana, Illinois, June 5–7, 1960 (unpublished)

[3]) R V Pound and G A Rebka, Jr, Phys. Rev Letters 4 (1960) 337

[4]) H J Hay, J P Schiffer, T E Cranshaw and P A Egelstaff, Phys. Rev Letters 4 (1960) 165

[5]) R. V Pound and G A Rebka, Jr, Phys Rev Letters 3 (1959) 554,
G DePasquali, H Frauenfelder, S Margulies and R N Peacock, Phys Rev Letters 4 (1960) 71,
S. S Hanna, J Heberle, C Littlejohn, G J Perlow, R S Preston and D. H Vincent, Phys Rev Letters 4 (1960) 177

[6]) H Frauenfelder, D R F Cochran, D E Nagle and R D. Taylor, Nuovo Cim 19 (1961) 183

of resonant gamma radiation emitted from a source of finite thickness†, and passing through a finite resonance absorber. In particular, we will consider two types of sources, one in which the radioactive atoms are distributed uniformly and one which has a Gaussian distribution of emitting nuclei.

2. General Formulation

In our calculation of the transmitted intensity, we will assume that a fraction f of all decays occur without recoil energy loss[7]). The resonance radiation resulting from these decays will be taken to have an emission and absorption spectrum of Breit-Wigner shape. The remaining fraction of the radiation is non-resonant, and is subject only to ordinary electronic absorption. We will consider a source having arbitrary area, and extending in depth from $x = 0$ to $x = \infty$. The distribution of emitting atoms along the x-axis will be denoted by $\rho(x)$. We will deal only with the radiation emitted normal to the area of the source, as shown in fig. 1. The distribution of the absorbing atoms in both the source and absorber will be taken to be uniform.

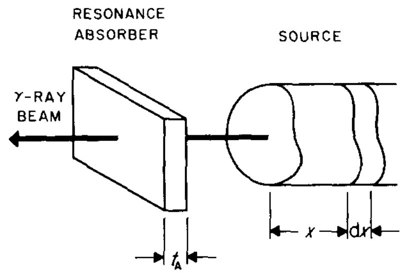

Fig 1 Geometry used to calculate the transmission of γ-radiation through a resonance absorber moving with a velocity v relative to the source

Under these circumstances, the transmission through a resonance absorber of thickness t_A, moving with a velocity v relative to the source, is given by

$$p(\mathscr{S}) = e^{-\mu_A t_A} \left\{ (1-f) \int_0^\infty dx\, \rho(x) e^{-\mu_S x} + f \frac{\Gamma}{2\pi} \int_{-\infty}^{\infty} dE \exp\left[-f'_A n_A a_A \sigma_0 t_A \frac{\Gamma^2/4}{(E-E_0)^2 + \Gamma^2/4} \right] \right.$$
$$\left. \times \int_0^\infty \frac{dx\, \rho(x)}{(E - E_0 + \mathscr{S})^2 + \Gamma^2/4} \exp\left[-\left(f'_S n_S a_S \sigma_0 \frac{\Gamma^2/4}{(E-E_0+\mathscr{S})^2 + \Gamma^2/4} + \mu_S \right) x \right] \right\}. \quad (1)$$

In this equation, Γ is the full width at half-height of both the emission and absorption lines which are centered about E_0, and σ_0 is the absorption cross-section at resonance††. The subscripts S and A identify the following source and absorber quantities

f' = probability of resonance absorption without recoil,
n = number of atoms per cubic centimeter of volume,
a = fractional abundance of the atoms which can absorb resonantly,
μ = ordinary mass attenuation coefficient, evaluated at E_0.

The quantity $\mathscr{S} = (v/c)E_0$ characterizes the Doppler shift between the source and absorber.

The first term in (1) represents the transmission of the non-resonant fraction of the radiation, and is independent of the Doppler shift $\mathscr{S}$. In the second term, which is the resonant contribution, the x-integral represents the emission and self-absorption in the source. We will neglect the μ_S appearing in the exponential of this integral, since the mass absorption is usually much smaller than the resonance absorption. The remaining factors in the second term represent the absorption in the external resonance absorber. The lower limit on the energy integral has been taken as $-\infty$ instead of zero for convenience.

[7]) W Marshall and J P. Schiffer, The Debye-Waller Factor in the Mossbauer Effect, A.E R.E , Harwell (1960), (unpublished)

† The case of a beam of γ-radiation, having a Breit-Wigner energy spectrum, passing through a resonance absorber, has been considered by W M Visscher in The Evaluation of the Transmission Integral, Los Alamos Scientific Laboratory (1959) (unpublished) This corresponds, in effect, to the non-resonant absorbing source described in Section 5

†† The absorption cross-section at resonance is given by

$$\sigma_0 = 2\pi \lambda\!\!\!^{-2} \frac{2I^* + 1}{2I + 1} \frac{1}{1+\alpha}$$

where $\lambda\!\!\!^{-}$ is the wavelength of the γ-ray, I^* and I are the nuclear spins of the initial and final states, respectively, and α is the conversion coefficient for the transition

Equation (1) can be seen to be independent of the sign of the Doppler shift $\mathscr{S}$. Since only relative motion between source and absorber is pertinent, $\mathscr{S}$ can be included in the absorber part of the transmission integral instead of the source part, if desired.

Equation (1) can easily be generalized for the case when the emission and absorption lines consist of more than one component (as when electric or magnetic splitting exists) by forming appropriate sums. In this paper we will limit ourselves to the overlap of a single emission line with a single absorption line, each centered about E_0. For convenience, we will translate the energy axis so that both lines are centered about $E = 0$ in the absence of any Doppler shift; that is, $(E - E_0)$ will be replaced by E.

Sources for Mossbauer experiments are generally prepared in either of two ways†

1. The activity is electroplated or otherwise deposited on a source backing. The activity is then diffused into the host lattice by heating[8]).

2. The activity and the host atoms are co-plated on a backing, thereby building up a lattice containing the radioactive atoms as integral parts[9]).

The second method produces a source in which the emitting atoms are uniformly distributed in depth. The same type of distribution results from the first method if the backing is very thin and the diffusion time is very long. On the other hand, if either the backing is thick or the diffusion time is short, then the first method produces an activity distribution which is approximately Gaussian. Both types of distribution will be discussed below.

3. Uniform Source Distribution

We first consider a source of thickness t_S, having N radioactive atoms per unit length

$$\rho(x) = \begin{cases} N \text{ atoms/cm}, & t_S \geq x \geq 0 \\ 0 \quad , & x > t_S \end{cases} \tag{2}$$

Since Nt_S is the total number of radioactive atoms and $e^{-\mu_A t_A}$ represents the non-resonant electronic absorption in the external absorber, we will deal with a normalized transmission $P(\mathscr{S})$ defined by

$$P(\mathscr{S}) = p(\mathscr{S})/(e^{-\mu_A t_A} N t_S) \tag{3}$$

For the distribution given in (2) the x part of the transmission integral can easily be evaluated, and we find

$$P(\mathscr{S}) = (1-f)\left[\frac{1-e^{-\mu_S t_S}}{\mu_S t_S}\right] + f\frac{2}{\pi\Gamma}\frac{1}{T_S}\int_{-\infty}^{\infty} dE \exp\left(\frac{-T_A\Gamma^2/4}{E^2+\Gamma^2/4}\right) \times \left[1-\exp\left(\frac{-T_S\Gamma^2/4}{(E+\mathscr{S})^2+\Gamma^2/4}\right)\right] \tag{4}$$

Here, $T_S = f'_S n_S a_S \sigma_0 t_S$ and $T_A = f'_A n_A a_A \sigma_0 t_A$ are effective source and absorber thicknesses, respectively. The first term in this equation is the non-resonant transmission, and will henceforth be denoted by $(1-f)P_{\text{non res}}(\text{uniform})$. As $\mu_S t_S$ approaches zero, this quantity approaches $(1-f)$. The second term in (4), containing an integral over energy, represents the resonant contribution. Before discussing the general evaluation of the integral, we will first consider two special cases.

3.1 THIN SOURCE AND ABSORBER

When both source and absorber are thin in the sense that $T_S \ll 1$, $T_A \ll 1$, we can expand both exponentials in the second term of (4) and keep only lowest-order terms in effective thickness. Integrating over energy, we obtain

† Occasionally the target foil, which has been irradiated in a pile or in an accelerator beam to produce the desired radioisotope, may serve directly as a source. In such a case, the distribution of emitting atoms can be quite complex, and will not be treated here.

[8]) R. V. Pound and G. A. Rebka, Jr., Phys. Rev. Letters **3** (1959) 554

[9]) S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston and D. H. Vincent, Phys. Rev. Letters **4** (1960) 28

$$P(\mathscr{S}) = \left[(1-f)P(\text{uniform})_{\text{non-res}} + f\left(1-\frac{T_S}{4}\right)\right] - f\frac{T_A}{2}\frac{1}{1+(\mathscr{S}/\Gamma)^2} \tag{5}$$

The bracketed term in this equation represents the transmission when $\mathscr{S}$, the relative Doppler shift between source and absorber, is large. This asymptotic value is less than unity because of the self-absorption in the source. The second term represents the dip in the transmission due to resonance absorption in the external absorber. It is seen that the transmitted line, in the case of thin source and absorber, has a Breit-Wigner shape, but has an apparent width Γ_a which is twice that of either the emission or absorption spectrum. This broadening results from the overlap of the emission and absorption lines.

3 2. THIN SOURCE – TRANSMISSION AT ZERO DOPPLER SHIFT

When the effective source thickness T_S approaches zero (as, for example, in a source which has little resonance absorption because $a_S \to 0$), the transmission at zero Doppler shift can be found in terms of T_A, the effective absorber thickness. Expanding the term containing T_S in (4) and keeping only the lowest-order term leads to

$$P(0) \approx (1-f)P(\text{uniform})_{\text{non-res}} + f\,e^{-\frac{1}{2}T_A}\,J_0(\tfrac{1}{2}iT_A)\,, \qquad T_S \to 0\,, \tag{6}$$

where J_0 is the Bessel function of zero order. Because of the nature of resonance absorption, the resonant contribution to the transmission does not decrease exponentially with absorber thickness, but instead, shows a saturation behavior.

3 3 UNIFORM SOURCE – GENERAL CASE

Attempts to evaluate (4) analytically for arbitrary source and absorber thicknesses have been unsuccessful. Consequently, we have performed a numerical integration on the University of Illinois digital computer ILLIAC for values of T_S and T_A between zero and ten. It has been found empirically that over this range the transmitted line is, to a very good degree of approximation, a Breit-Wigner curve whose full width at half-height Γ_a, depends upon the value of T_S and T_A. The general variation of transmission with Doppler shift $\mathscr{S}$ is shown in fig. 2. The results of our numerical integration, in the form of the variation of Γ_a/Γ as a function of T_A with T_S as parameter, are shown in fig. 3. Note that as T_S and T_A both approach zero, Γ_a/Γ approaches the value two, since the conditions described in section 3.1 are applicable.

4. Gaussian Source Distribution

For the case of a Gaussian distribution of radioactive atoms, we will use

$$\rho(x) = (2N/\sqrt{\pi})\,e^{-x^2/t_S^2}\ \text{atoms/cm}, \qquad x \geqslant 0 \tag{7}$$

Here t_S represents a characteristic diffusion depth whose value depends upon the details of the source preparation. The above distribution is normalized so that Nt_S once again represents the total number of radioactive atoms.

Substitution of this Gaussian distribution into (1) leads to the following expression, which is normalized in the sense defined by (3):

$$P(\mathscr{S}) = (1-f)\,e^{(\frac{1}{2}\mu_S t_S)^2}\left[1-\Phi(\mu_S t_S/2)\right] + f\frac{\Gamma}{2\pi}\int_{-\infty}^{\infty}\frac{dE}{(E+\mathscr{S})^2+\Gamma^2/4}\exp\left(\frac{-T_A\Gamma^2/4}{E^2+\Gamma^2/4}\right)$$
$$\times\left[1-\Phi\left(\frac{T_S\Gamma^2/4}{2\left[(E+\mathscr{S})^2+\Gamma^2/4\right]}\right)\right]\exp\left(\frac{T_S\Gamma^2/4}{2\left[(E+\mathscr{S})^2+\Gamma^2/4\right]}\right)^2, \tag{8}$$

where Φ represents the error function,

$$\Phi(y) = (2/\sqrt{\pi})\int_0^y e^{-u^2}\,du\,.$$

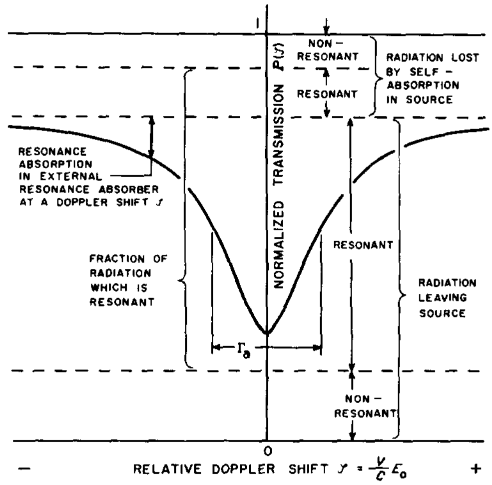

Fig 2 Normalized transmission of γ-radiation through a resonance absorber as a function of the relative Doppler shift between source and absorber The apparent full width at half-height of the transmitted line is denoted by Γ_a

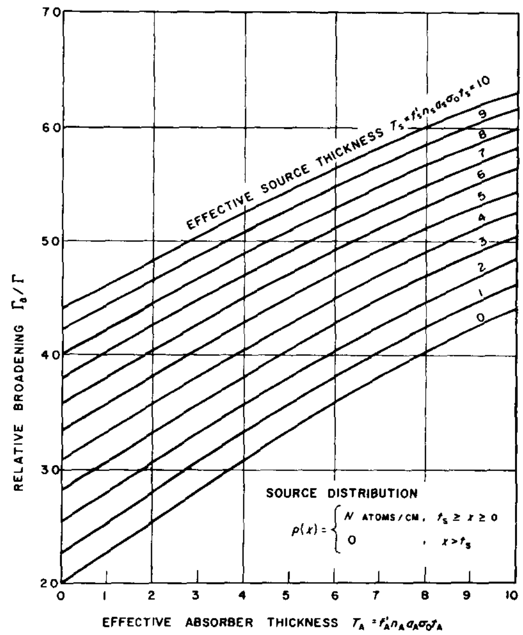

Fig 3

Fig. 3 Broadening of the transmitted line for a source having a uniform distribution of emitting atoms.

The first term in (8) corresponds to the transmission of the non-resonant fraction of the radiation, and will be denoted by $(1-f)P_{\text{non-res}}(\text{Gaussian})$. Again, as $\mu_S t_S$ approaches zero, the non-resonant contribution approaches $(1-f)$. The resonant contribution to the transmission is contained in the second term of (8). Before discussing the general evaluation of this equation, we will consider two special cases.

4 1. THIN SOURCE AND ABSORBER

If both source and absorber effective thicknesses satisfy the conditions $T_S \ll 1$, $T_A \ll 1$, we can expand the exponentials and the error function in the second term of (8), and keep only first-order terms in thickness. After integrating over energy, one obtains

$$P(\mathscr{S}) = \left[(1-f)P_{\text{non-res}}(\text{Gaussian}) + f\left(1 - \frac{T_S}{2\sqrt{\pi}}\right) \right] - f\frac{T_A}{2}\frac{1}{1+(\mathscr{S}/\Gamma)^2}. \tag{9}$$

In this equation the bracketed term represents the asymptotic transmission as $\mathscr{S} \to \infty$, and differs from unity because of self-absorption in the source. The second term results from resonance absorption in the external absorber. The transmitted line shape has the Breit-Wigner form, but is twice as wide as either the emission or the absorption line. Note that (9) differs from (5) only in that the Gaussian source results in more self-absorption than the uniform source. This conclusion follows from the choice of normalization for the Gaussian distribution and is subject to the thin source approximation used to derive both equations.

4 2. THIN SOURCE – TRANSMISSION AT ZERO DOPPLER SHIFT

Expansion of the appropriate terms to lowest order when $T_S \to 0$ allows (8) to be evaluated at the point $\mathscr{S} = 0$. Since both the Gaussian distribution of (7) and the uniform distribution described by (2) are normalized in the same way, it is not surprising that (6) once again follows for this special case, but with $P_{\text{non-res.}}(\text{uniform})$ replaced by $P_{\text{non-res.}}(\text{Gaussian})$.

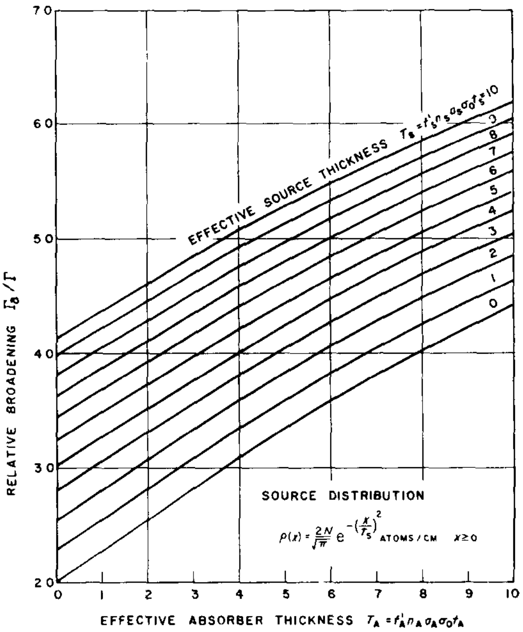

Fig 4 Broadening of the transmitted line for a source having a Gaussian distribution of emitting atoms

4 3 GAUSSIAN SOURCE – GENERAL CASE

Equation (8), representing the transmission from a source which has a Gaussian distribution of radioactive atoms, could not be evaluated analytically for arbitrary values of T_S and T_A. Again, we have performed a numerical integration on ILLIAC for the range $10 \geq (T_S, T_A) \geq 0$. As in the uniform case, it has been found empirically that the resulting transmitted lines differ but little from Breit-Wigner curves whose full widths at half-height vary with source and absorber thicknesses (see fig. 2). The calculated broadening of the transmitted line as a function of T_S and T_A is shown in fig. 4. As T_S and T_A approach zero, the conditions of section 4.1 apply, and Γ_a/Γ approaches the value two

5. Non-Resonant Absorbing Sources

As has been shown, the width of the transmission curve obtained from a resonantly absorbing source of finite thickness is always greater than twice the natural width of the transition. The way to obtain the narrowest lines with any given radioisotope is to use a source backing which has zero abundance of atoms that can absorb resonantly. In this way, only the external resonance absorber contributes to the line broadening. When this is the case, the ordinary electronic absorption, neglected so far for the resonant fraction of the radiation, must be considered. The transmission under these circumstances is given by

$$p(\mathscr{S}) = e^{-\mu_A t_A} \left\{ (1-f) \int_0^\infty dx\, \rho(x)\, e^{-\mu_S x} + f \frac{\Gamma}{2\pi} \int_{-\infty}^{\infty} dE \exp\left(\frac{-T_A \Gamma^2/4}{(E-E_0)^2 + \Gamma^2/4}\right) \times \int_0^\infty \frac{dx\, \rho(x)\, e^{-\mu_S x}}{(E-E_0+\mathscr{S})^2 + \Gamma^2/4} \right\} \tag{10}$$

The x-integral is the same in both the non-resonant and the resonant terms of this equation, and can be evaluated for the uniform distribution of (2) and the Gaussian distribution of (7). The transmission, normalized according to (3), is found to be

$$\left.\begin{aligned} P(\mathscr{S}) &= [(1-f) + fI(\mathscr{S})]\left[\frac{1 - e^{-\mu_S t_S}}{\mu_S t_S}\right] \\ &= [(1-f) + fI(\mathscr{S})]\, P_{\text{non-res}}(\text{uniform}) \end{aligned}\right\} \text{uniform source} \tag{11}$$

and

$$\left.\begin{aligned} P(\mathscr{S}) &= [(1-f) + fI(\mathscr{S})]\,[e^{(\frac{1}{2}\mu_S t_S)^2} (1 - \Phi(\mu_S t_S/2))] \\ &= [(1-f) + fI(\mathscr{S})]\, P_{\text{non-res.}}(\text{Gaussian}) \end{aligned}\right\} \text{Gaussian source} \tag{12}$$

where

$$I(\mathscr{S}) = \frac{\Gamma}{2\pi} \int_{-\infty}^{\infty} \frac{dE}{(E+\mathscr{S})^2 + \Gamma^2/4} \exp\left(\frac{-T_A \Gamma^2/4}{E^2 + \Gamma^2/4}\right) \tag{13}$$

In either case, the shape of the transmitted line is the same, only the amplitude being affected by the electronic absorption. This behavior follows from the fact that the ordinary mass attenuation coefficients are energy independent over the width of the emission and absorption lines. The shape of the transmitted line is determined by the energy integral of (13). Since this integral corresponds to the $T_S = 0$ case for either a uniform or a Gaussian source it has, in effect, already been considered. In the results presented below, it should be remembered that $I(\mathscr{S})$ must be substituted into either (11) or (12) to obtain the transmission, $P(\mathscr{S})$.

When the resonance absorber satisfies the condition $T_A \ll 1$, the integral of (13) reduces to

$$I(\mathscr{S}) = 1 - \frac{T_A}{2} \frac{1}{1 + (\mathscr{S}/\Gamma)^2}, \tag{14}$$

again representing a Breit-Wigner curve whose width at half-height is 2Γ.

For arbitrary values of T_A, the transmission also approximates a Breit-Wigner curve, but the apparent width Γ_a varies with the absorber thickness in the manner described by the $T_S = 0$ curve in either fig. 3 or fig. 4.

When there exists no Doppler shift between source and absorber, the energy integral of (13) can be evaluated

$$I(0) = e^{-\frac{1}{2}T_A} J_0(iT_A/2) . \tag{15}$$

In section 3.2 and 4.2, where very thin sources were considered, the evaluation of the energy integral led to the approximation given in (6). In the case of a non-resonantly absorbing source, no approximations are needed, and the result of (15) is exact. Since $I(\infty) = 1$, we can combine (15) with either (11) or (12) to get the useful result

$$\frac{P(\infty) - P(0)}{P(\infty)} = f\,[\,1 - e^{-\frac{1}{2}T_A} J_0(iT_A/2)\,] \tag{16}$$

Note added in proof Equation (16), firxt applied to the analysis of Mössbauer experiments by the Los Alamos group, is often being used when it is not applicable It must be remembered that (16) is

1) exact only for non-resonantly absorbing sources, it is a good approximation for sources where $T_S \ll 1$,

2) derived on the assumption that the emission and absorption lines overlap exactly at zero relative velocity between source and absorber If, as is often the case, there exists an energy shift between emission and absorption spectra, $P(0)$ must be replaced by $P(\mathscr{S}_0)$, where $\mathscr{S}_0$ is the Doppler shift required to produce coincidence,

3) valid for source and absorber half-widths equal to Γ, the natural width of the transition It is still correct if these widths are not the natural width, provided that they are equal $\Gamma_S = \Gamma_A$ In this case, however, the maximum absorption cross-section σ_0 must be multiplied by the factor Γ/Γ_S to keep the total absorption constant

We wish to emphasize that (16) can be used to extract f and f' from absorption measurements only if the conditions assumed in the derivation are at least approximately satisfied

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