Transmission of a Lorentzian Spectral Line Through a Layer of Lorentzian Absorbers. Part I

JUERGEN HEBERLE

Department of Physics, Clark University, Worcester, Massachusetts, and Physik-Department, Technische Hochschule, München *

and SERGIO FRANCO

Department of Physics, Clark University, and Department of Computer Science, University of Illinois at Urbana

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This paper is concerned with the evaluation of the integral that represents the total transmitted intensity. We consider the general case in which the thin-absorber linewidth Γ_A is allowed to be different from the linewidth Γ_S of the source. The introduction of the parameter $\gamma = \Gamma_A/\Gamma_S$ has made it possible to expand the integral in an infinite series, each term of which is a rational algebraic expression. Our formulation avoids the tedium of numerical integration and lends itself readily to programming in FORTRAN. The application to Mössbauer spectroscopy is discussed. The results are valid in the case of a split source even if the spacing between the emission lines is less than their linewidth.

1. Introduction

We consider a source of radiation whose intensity is distributed about the mean energy E_S according to

$$I(E)\,\mathrm{d}E = \frac{A_0}{1+[(E-E_S)/\frac{1}{2}\Gamma_S]^2}\,\mathrm{d}E\,.$$

A beam of this radiation is incident upon a layer of particles, each of which has a cross section $\sigma(E)$ for removing radiation from the beam. The thickness of the layer is uniform and is specified by n, the number of particles per unit area normal to the direction of the beam. The cross section varies with energy according to

$$\sigma(E) = \frac{\sigma_{\max}}{1+[(E-E_A)/\frac{1}{2}\Gamma_A]^2}$$

where Γ_A is the *thin-absorber linewidth* (see Section 4). Then the total intensity transmitted by the layer is given by

$$P(\Delta E) = \int I(E)\,e^{-n\sigma(E)}\,\mathrm{d}E \qquad (1.1)$$

where $\Delta E = E_A - E_S$;

the integration is extended over the range of E within which the integrand is significantly greater than zero. The energy difference ΔE has been introduced to bring out the fact that the dependence of P upon the line centers E_S and E_A is such that only their difference is relevant.

The present paper is concerned with the evaluation of the integral in Eq. (1.1). It is our goal to put this expression into a form that is convenient for programming in the FORTRAN language.

As yet we have not specified the nature of the radiation or of the particles in the layer. This integral is of special interest in the spectroscopy based on the Mössbauer effect, where the radiation consists of recoilfree gamma rays and the absorbing particles are nuclei bound in crystal lattices. In this connection, the case $\Gamma_S = \Gamma_A$ has been treated by MARGULIES and EHRMAN[1], RUBY and HICKS[2], and by FRAUENFELDER et al.[3]. An approximative treatment of the general case, where $\Gamma_S \neq \Gamma_A$, has been given by O'CONNOR[4].

In the next section, we show how the integral can be expanded into an infinite series of finite algebraic expressions, whereas earlier formulas[2,3] involve infinite series, each term of which contains a Bessel function. In Section 3 we discuss properties of the individual terms. We explore special cases in Sec-

* On leave from the Department of Physics and Astronomy, State University of New York at Buffalo.

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[2] S. L. RUBY and J. M. HICKS, Rev. Sci. Instr. **33**, 27 [1962].

[3] H. FRAUENFELDER, D. E. NAGLE, R. D. TAYLOR, D. R. F. COCHRAN and W. M. VISSCHER, Phys. Rev. **126**, 1065 [1962].

[4] D. A. O'CONNOR, Nucl. Instr. Methods **21**, 318 [1963].

tion 4. The error incurred in terminating our series is estimated in Section 5. We show in Section 6 how these results can be applied in Mössbauer spectroscopy. A discussion follows in Section 7.

2. Expansion into an Infinite Series

Usually E_S and E_A are much greater than the widths Γ_S and Γ_A. This makes it possible to use infinite limits in the integral of Eq. (1.1). Furthermore, it is convenient to introduce the dimensionless variables

$$z = (E - E_A)/\tfrac{1}{2}\Gamma_S, \quad x = \Delta E/\tfrac{1}{2}\Gamma_S,$$
$$\gamma = \Gamma_A/\Gamma_S, \quad \text{and} \quad s = n\,\sigma_{\max}.$$

Then equation (1.1) becomes

$$P(\Delta E) = \tfrac{1}{2}A_0\Gamma_S\int_{-\infty}^{+\infty}\frac{\exp\{-s/[1+(z/\gamma)^2]\}}{1+(z+x)^2}\,dz. \quad (2.1)$$

Since in every subsequent integral the limits will be $-\infty$ and $+\infty$, it will not be necessary to indicate the limits explicitly. We evaluate $P(\infty)$ by changing the variable of integration to $y = z + x$. Thus

$$P(\infty) = \lim_{x\to\infty}\tfrac{1}{2}A_0\Gamma_S\int\frac{\exp\{-\gamma^2 s/[\gamma^2+(y-x)^2]\}}{1+y^2}\,dy$$
$$= \tfrac{1}{2}A_0\Gamma_S\int\frac{dy}{1+y^2} = \tfrac{1}{2}A_0\Gamma_S\pi.$$

Now we introduce the *transmission function* defined by

$$\mathrm{tran}(\gamma, s; x) = \frac{1}{\pi}\int\frac{\exp\{-\gamma^2 s/(z^2+\gamma^2)\}}{1+(z+x)^2}dz. \quad (2.2a)$$

Then Eq. (2.1) becomes

$$P(\Delta E) = \tfrac{1}{2}\pi A_0\Gamma_S\,\mathrm{tran}(\gamma, s; x), \quad (2.2b)$$

and our task is now to evaluate the transmission function.

After expanding the exponential in a Maclaurin series, we get

$$\mathrm{tran}(\gamma, s; x) = \sum_{m=0}^{\infty}\frac{(-s)^m}{m!}Q_m(\gamma, x) \quad (2.3a)$$

where

$$Q_m(\gamma, x) = \frac{1}{\pi}\int\left(\frac{\gamma^2}{z^2+\gamma^2}\right)^m\frac{dz}{1+(z+x)^2}. \quad (2.3b)$$

It is easy to see that $0 < Q_{m+1} < Q_m < 1$ for $m > 0$. Thus, for $m > 0$, the absolute value of each term in (2.3a) is less than the corresponding term in the Maclaurin series of e^s. It follows that our series (2.3a) is absolutely and uniformly convergent for all real values of s and x.

We use the method of residues to evaluate the integral for Q_1 and find that

$$Q_1 = \frac{\gamma(\gamma+1)}{x^2+(\gamma+1)^2} \quad \text{if} \quad \gamma > 0,$$
$$= \frac{\gamma(\gamma-1)}{x^2+(\gamma-1)^2} \quad \text{if} \quad \gamma < 0.$$

Since Γ_S and Γ_A are positive, we need to consider only the case $\gamma > 0$.

In principle, any Q_m can be evaluated by the method of residues. For large values of m, however, this approach is exceedingly cumbersome. Instead we differentiate (2.3b) with respect to γ and note that

$$\frac{\partial Q_m}{\partial\gamma} = \frac{2m}{\gamma}(Q_m - Q_{m+1}). \quad (2.4)$$

Since we have already integrated Q_1, it is possible to obtain all other Q_m from the recursive formula

$$Q_{m+1} = Q_m - \frac{\gamma}{2m}\frac{\partial Q_m}{\partial\gamma}. \quad (2.5)$$

By successive applications of (2.5), we have evaluated Q_2 to Q_6. We list these results together with Q_0 and Q_1, making use of the abbreviation $X = x^2 + (\gamma+1)^2$:

$$Q_0 = 1, \quad Q_1 = \frac{\gamma(\gamma+1)}{X},$$

$$Q_2 = \tfrac{1}{2}(\gamma/X) + (\gamma/X)^2(\gamma+1)^2,$$

$$Q_3 = \tfrac{3}{8}(\gamma/X) + \tfrac{3}{4}(\gamma/X)^2(\gamma+1) + (\gamma/X)^3(\gamma+1)^3,$$

$$Q_4 = \tfrac{5}{16}(\gamma/X) + \tfrac{1}{8}(\gamma/X)^2(4\gamma+5) + (\gamma/X)^3(\gamma+1)^2 + (\gamma/X)^4(\gamma+1)^4,$$

$$Q_5 = \tfrac{35}{128}(\gamma/X) + \tfrac{5}{64}(\gamma/X)^2(5\gamma+7) + \tfrac{5}{16}(\gamma/X)^3(\gamma+1)(2\gamma+3) + \tfrac{5}{4}(\gamma/X)^4(\gamma+1)^3 + (\gamma/X)^5(\gamma+1)^5,$$

and

$$Q_6 = \tfrac{63}{256}(\gamma/X) + \tfrac{21}{128}(\gamma/X)^2(2\gamma+3) + \tfrac{1}{32}(\gamma/X)^3(15\gamma^2+42\gamma+28) + \tfrac{3}{16}(\gamma/X)^4(\gamma+1)^2(4\gamma+7) + \tfrac{3}{2}(\gamma/X)^5(\gamma+1)^4 + (\gamma/X)^6(\gamma+1)^6.$$

It is seen that the Q_m have the general form

$$Q_m = \sum_{l=1}^{m}(\gamma/X)^l G_{lm}(\gamma), \quad (2.6)$$

where G_{lm} is a polynomial in γ. By applying the recursion relation (2.5) to Eq. (2.6), we obtain re-

cursion formulas for G_{lm},

$$G_{l,m+1} = \frac{1}{2m}\left[(2m-l)G_{lm} + 2(l-1)(\gamma+1)G_{l-1,m} - \gamma\frac{\mathrm{d}}{\mathrm{d}\gamma}G_{lm}\right] \quad \text{for} \quad 1<l<m+1, \tag{2.7a}$$

$$G_{1,m+1} = \frac{2m-1}{2m}G_{1,m} \tag{2.7b}$$

$$G_{m+1,m+1} = (\gamma+1)G_{mm}. \tag{2.7c}$$

Since on a digital computer it is much easier to perform algebraic operations than to carry out differentiations, we proceed to cast the recursion formulas (2.7) into a purely algebraic form. Toward this end we write

$$G_{lm} = \frac{1}{4^{m-l}}\sum_{k=1}^{l+1} a(k,l,m)\gamma^{k-1}. \tag{2.8}$$

By inserting (2.8) into (2.7) and equating the coefficients of like powers of γ, we are led to

$$a(k,1,m+1) = \frac{2(2m-k)}{m}a(k,1,m), \tag{2.9a}$$

$$a(1,l+1,m+1) = \frac{2(2m-l-1)}{m}a(1,l+1,m) + \frac{l}{m}a(1,l,m), \tag{2.9b}$$

$$a(k+1,l+1,m+1) = \frac{2(2m-k-l-1)}{m}a(k+1,l+1,m) + \frac{l}{m}[a(k,l,m)+a(k+1,l,m)], \tag{2.9c}$$

where it is understood that $a(k,l,m)=0$ if $k>l+1$.

From the above expression for Q_1, we see that

$$a(1,1,1) = a(2,1,1) = 1.$$

The recursion formulas (2.9) make it possible to compute all the other coefficients $a(k,l,m)$. Then by means of Eqs. (2.8), (2.6), (2.3a) and (2.2b), one can calculate $P(\Delta E)$ to any desired accuracy.

At this stage the problem has been essentially solved, except for the estimation of the error (see Section 5). It is useful, however, to point out various properties of our expansion and to consider special cases.

3. Some Properties of the Terms of the Series

By mathematical induction it is possible to obtain formulas for certain of the polynomials G_{lm} and for certain coefficients $a(k,l,m)$ which permit a direct computation of these quantities without having to evaluate similar quantities of lower index.

Thus by application of (2.9) it is possible to verify the following relations:

$a(k,l,m) \geqq 0$ for all values of the indices,

and

$a(k,l,m) = 0$ if $k = l+1$ and $l<m$.

The recursion formulas (2.7) for the G_{lm} permit one to verify the following equations:

$$G_{1,m+1} = \frac{1\cdot3\cdot5\ldots(2m-1)}{2\cdot4\cdot6\ldots(2m)} = \frac{(2m)!}{(2^m m!)^2} = (-1)^m P_{2m}(0) \quad \text{for} \quad m \geqq 1 \tag{3.1}$$

(where P_{2m} denotes a Legendre polynomial),

$$G_{2,m+1} = \frac{[2(m-1)]!}{m[2^{m-1}(m-1)!]^2}[2m-1+(m+1)\gamma] \quad \text{for} \quad m \geqq 2, \tag{3.2}$$

$$G_{mm} = (\gamma+1)^m,$$

$$G_{m-1,m} = \frac{m}{4}(\gamma+1)^{m-2},$$

$$G_{m-2,m} = \frac{m}{32}(\gamma+1)^{m-4}[4\gamma+(m+1)],$$

and

$$G_{m-3,m} = \frac{m}{64}(\gamma+1)^{m-6}\cdot[5\gamma^2+2(m+1)\gamma+\tfrac{1}{6}(m+1)(m+2)$$

If now we use (2.8) to extract from the above equations the values of $a(k,l,m)$, we find that these coefficients are integers. For example

$$a(1,m-3,m) = \tfrac{1}{6}m(m+1)(m+2),$$

which is an integer for all integral values of m. The question arises whether or not it is true that *all* the coefficients $a(k,l,m)$ are integers. We have not been able to prove the general case; however, by computation we have shown that $a(k,l,m)$ is an integer for $k \leqq l \leqq m \leqq 11$.

Since $a(k,l,m) \geqq 0$, it follows from (2.8) that G_{lm} is always positive. Furthermore, by inspecting Eq. (2.6) we see that for a fixed value of γ the function $Q_m(\gamma,x)$ reaches its greatest value at $x=0$; this will be important in Section 5 when we estimate the error.

4. Special Cases

The results of this section will be somewhat simpler if, instead of the transmission function, we

consider a related quantity, the *fractional absorption*, defined by

$$\varepsilon(\gamma, s; x) = 1 - \mathrm{tran}(\gamma, s; x).$$

Furthermore, in this section there occur series of the form

$$S_k(z) = k! \sum_{n=0}^{\infty} \frac{(2n)!\,(-1)^n}{(n!)^2\,(n+k)!} \left(\frac{z}{4}\right)^n.$$

It turns out that the $S_k(z)$ can be identified[5] with a special type of the confluent hypergeometric functions,

$$S_k(z) = \Phi(\tfrac{1}{2}, k+1; -z).$$

For the sake of brevity, however, we continue to use the notation $S_k(z)$. These hypergeometric functions are related to the modified Bessel functions of the first kind by the formula of HEBERLE[5]

$$\Phi(\tfrac{1}{2}, k+1; -z) = (k!)^2 e^{-z/2} \sum_{l=-k}^{k} \frac{I_l(z/2)}{(k-l)!\,(k+l)!}.$$

Thus, remembering that $I_{-l}(x) = I_{+l}(x)$, we obtain

$$S_0(z) = e^{-z/2} I_0(z/2), \tag{4.1a}$$

$$S_1(z) = e^{-z/2}[I_0(z/2) + I_1(z/2)], \tag{4.1b}$$

$$S_2(z) = \tfrac{1}{3} e^{-z/2}[3 I_0(z/2) + 4 I_1(z/2) + I_2(z/2)]. \tag{4.1c}$$

a. $s \ll 1$

For a very thin absorbing layer, such that $s \ll 1$, we can write

$$\varepsilon \approx \frac{\gamma(\gamma+1)}{x^2 + (\gamma+1)^2}\, s, \tag{4.2}$$

which represents a Lorentzian line shape with a width equal to $\Gamma_S + \Gamma_A$. We see that a thin absorber contributes an amount Γ_A to this width; hence our term *thin-absorber linewidth* for the quantity Γ_A (see Section 1).

b. $\gamma = 1$ and $x = 0$

Eq. (2.3b) is reduced to

$$Q_m(1, 0) = \frac{1}{\pi} \int \frac{dz}{(1+z^2)^{m+1}}. \tag{4.3}$$

A repeated integration by parts[5] leads to

$$Q_m(1, 0) = \frac{1 \cdot 3 \cdot 5 \ldots (2m-1)}{2 \cdot 4 \cdot 6 \ldots (2m)} = \frac{(2m)!}{(2^m m!)^2}, \tag{4.4}$$

and we have

$$\mathrm{tran}(1, s; 0) = \sum_{m=0}^{\infty} \frac{(2m)!\,(-1)^m}{(m!)^3} \left(\frac{s}{4}\right)^m = S_0(s).$$

By use of Eq. (4.1a) we obtain the well-known result[6,7,8]

$$\mathrm{tran}(1, s; 0) = e^{-s/2} I_0(s/2). \tag{4.5}$$

c. $\gamma/X \ll 1$

In order to consider this case, we write

$$\varepsilon = \sum_{n=1}^{\infty} (\gamma/X)^n b_n(\gamma, s) \tag{4.6}$$

where $$b_n(\gamma, s) = \sum_{m=n}^{\infty} \frac{(-s)^{m+1}}{m!} G_{lm}(\gamma).$$

Here the term in b_1 is the dominant one. We also work out the term in b_2 in order to provide a means of estimating the error incurred by approximating ε by the first term of the series in Eq. (4.6).

By use of (3.1) we write

$$b_1 = s \sum_{m=0}^{\infty} \frac{(-1)^m}{(m+1)!} G_{1,\,m+1}\, s^m = s\left[\gamma + 1 + \sum_{m=1}^{\infty} \frac{(2m)!(-1)^m}{(m!)^2\,(m+1)!} \left(\frac{s}{4}\right)^m\right] = s[\gamma + S_1(s)].$$

We proceed with b_2 in a similar way and make use of (3.2),

$$b_2 = s \sum_{m=1}^{\infty} \frac{(-1)^m}{(m+1)!} G_{2,\,m+1}\, s^m = s\left\{-\frac{(\gamma+1)^2}{2} s + \sum_{m=2}^{\infty} \frac{[2(m-1)]!\,(-1)^m\,[2m-1+(m+1)\gamma]}{[2^{m-1}(m-1)!]^2\,(m+1)!\,m}\, s^m\right\}.$$

[5] J. HEBERLE, to be published.

[6] H. A. BETHE, Rev. Mod. Phys. **9**, 69 [1937]; see equations (542) and (542a).

[7] P. P. CRAIG, J. G. DASH, A. D. MCGUIRE, D. E. NAGLE, and R. R. REISWIG, Phys. Rev. Letters **3**, 221 [1959].

[8] R. L. MÖSSBAUER and W. H. WIEDEMANN, Z. Phys. **159**, 33 [1960].

This expression becomes much simpler for the special case of $\gamma = 1$,

$$b_2(1,s) = -s^2\left\{2 + 3\sum_{m=2}^{\infty} \frac{[2(m-1)]!\,(-1)^{m-1}}{[(m-1)!]^2\,(m+1)!}\left(\frac{s}{4}\right)^{m-1}\right\}$$
$$= -s^2\left\{2 + 3\sum_{m=1}^{\infty} \frac{(2m)!\,(-1)^m}{(m!)^2\,(m+2)!}\left(\frac{s}{4}\right)^{m}\right\} = -\tfrac{1}{2}s^2\{1 + 3S_2(s)\}.$$

We resume our treatment of the general case,

$$b_2(\gamma,s) = s\left\{-\frac{\gamma^2 s}{2} + 4\sum_{m=1}^{\infty} \frac{(2m-1)!\,(-1)^m}{(m-1)!\,m!\,(m+1)!}\left(\frac{s}{4}\right)^m + 4\gamma\sum_{m=1}^{\infty} \frac{[2(m-1)]!\,(-1)^m}{(m-1)!\,(m!)^2}\left(\frac{s}{4}\right)^m\right\}$$
$$= -s\left\{\frac{\gamma^2 s}{2} + 2\left[1 - \sum_{m=0}^{\infty} \frac{(2m)!\,(-1)^m}{(m!)^2\,(m+1)!}\left(\frac{s}{4}\right)^m\right] + 4\gamma\sum_{m=0}^{\infty} \frac{(2m)!\,(-1)^m}{m!\,[(m+1)!]^2}\left(\frac{s}{4}\right)^{m+1}\right\}$$
$$= -s\left\{\frac{\gamma^2}{2}s + 2[1 - S_1(s)] + 2\gamma[S_0(s) + sS_1(s) - 1]\right\}.$$

Thus we obtain

$$\varepsilon = (\gamma/X)\,s[\gamma + S_1(s)] - (\gamma/X)^2\,\tfrac{1}{2}\,s[\gamma^2 s - 4\gamma + 4 + 4\gamma S_0(s) + 4(\gamma s - 1)\,S_1(s)] + O([\gamma/X]^3). \quad (4.7)$$

We want to point out that a Lorentzian line shape with width $\Gamma_S + \Gamma_A$ is approached in the two limiting cases $s \to 0$ and $\gamma \to 0$. For any values of s and γ, a Lorentzian is approached as $|x| \to \infty$.

d. $\gamma \gg 1$

Here we take γ so large that γ^{n-1} may be neglected in comparison with γ^n, but we make no assumptions about the magnitude of x relative to γ. Accordingly we have

$$Q_m \approx \left(\frac{\gamma}{x^2+\gamma^2}\right)^m \gamma^m = \left[\frac{1}{1+(x/\gamma)^2}\right]^m.$$

Substituting in (2.3a), we obtain

$$\mathrm{tran}(\gamma, s; x) \approx \sum_{m=0}^{\infty} \frac{1}{m!}\left[\frac{-s}{1+(x/\gamma)^2}\right]^m$$

and finally

$$\mathrm{tran}(\gamma, s; x) \approx \exp\left[\frac{-s}{1+(x/\gamma)^2}\right]. \quad (4.8)$$

5. Estimation of Error

We begin by writing Eq. (2.3a) in the form

$$\mathrm{tran}(\gamma, s; x) = \sum_{m=0}^{\nu-1} \frac{(-s)^m}{m!} Q_m + \sum_{m=\nu}^{\infty} \frac{(-s)^m}{m!} Q_m.$$

Here ν is to be chosen so that in the second summation the absolute values of the terms decrease with increasing m. This condition requires that

$$\frac{Q_{m+1}}{Q_m}\,\frac{s}{m+1} < 1 \quad \text{for} \quad m \geqq \nu. \quad (5.1)$$

In view of the fact (see Section 2) that $Q_{m+1} < Q_m$, a sufficient condition for (5.1) is

$$\nu \geqq s - 1. \quad (5.2)$$

Since we are concerned only with positive values of s, the inequality (5.2) ensures that the second summation is an alternating series with decreasing terms. Hence we can write

$$\mathrm{tran}(\gamma, s; x) = \sum_{m=0}^{\nu-1} \frac{(-s)^m}{m!} Q_m + \frac{1}{2}\,\frac{(-s)^\nu}{\nu!} Q_\nu \pm E_\nu, \qquad \nu \geqq s - 1,$$

where the error E_ν is given by

$$E_\nu = \tfrac{1}{2}(s^\nu/\nu!)\,Q_\nu(\gamma, x).$$

In order to be able to estimate E_ν in advance of a computation, we proceed to develop formulas for estimating Q_m directly, i.e. without the recursion relations. Since $Q_m > Q_{m+1}$, it follows from (2.4) that $\partial Q_m/\partial\gamma$ is always positive and that Q_m increases monotonely with γ. Thus

$$Q_m(\gamma, x) \leqq Q_m(1, x) \quad \text{for} \quad \gamma \leqq 1,$$
$$Q_m(\gamma, x) \geqq Q_m(1, x) \quad \text{for} \quad \gamma \geqq 1.$$

Another limit on Q_m is found by beginning with Eq. (2.3b)

$$Q_m(\gamma, x) = \frac{1}{\pi}\int \left(\frac{\gamma^2}{z^2+\gamma^2}\right)^m \frac{\mathrm{d}z}{1+(z+x)^2}$$
$$= \frac{\gamma^{2m}}{\pi}\int \left[\frac{1}{1+(\gamma^2-1)/(z^2+1)}\right]^m \cdot \left(\frac{1}{z^2+1}\right)^m \frac{\mathrm{d}z}{1+(z+x)^2}.$$

By considering the dependence on γ of the quantity in the brackets, we conclude that

$$Q_m(\gamma, x) \geqq \gamma^{2m} Q_m(1, x) \quad \text{for} \quad \gamma \leqq 1,$$
$$Q_m(\gamma, x) \leqq \gamma^{2m} Q_m(1, x) \quad \text{for} \quad \gamma \geqq 1.$$

In Section 3 we learned that the maximam value of Q_m occurs at $x = 0$. Therefore

$$Q_m(\gamma, x) \leqq Q_m(\gamma, 0) \begin{cases} \leqq Q_m(1, 0) & \text{for} \quad \gamma \leqq 1, \\ \leqq \gamma^{2m} Q_m(1, 0) & \text{for} \quad \gamma \geqq 1. \end{cases}$$

Using (4.4) we obtain

$$Q_m(\gamma, x) \begin{cases} \leqq \dfrac{(2m)!}{(2^m m!)^2} \text{ for} & \gamma \leqq 1, \\ \leqq \gamma^{m2} \dfrac{(2m)!}{(2^m m!)^2} \text{ for} & \gamma \geqq 1. \end{cases}$$

Auother approach is based on the Cauchy-Schwarz inequality[9]

$$\left|\int_a^b f^*(z)\, g(z)\, \mathrm{d}z\right|^2 \leqq \int_a^b f^* f\, \mathrm{d}z \int_a^b g^* g\, \mathrm{d}z,$$

which yields

$$(Q_m)^2 < \frac{1}{\pi^2} \int \frac{\mathrm{d}z}{[1 + (z + x)^2]^2} \int \left(\frac{\gamma^2}{z^2 + \gamma^2}\right)^{2m} \mathrm{d}z$$
$$= \frac{\gamma}{\pi^2} \int \frac{\mathrm{d}u}{(1 + u^2)^2} \int \frac{\mathrm{d}u}{(1 + u^2)^{2m}}.$$

These integrals have the form (4.3), and by use of (4.4) we obtain

$$Q_m < \frac{\sqrt{2\gamma[2(2m-1)]!}}{4^m (2m-1)!}.$$

We combine these results and obtain upper limits for the error

$$E_\nu \leqq \frac{1}{2} \frac{(2\nu)!}{(\nu!)^3} \left(\frac{s}{4}\right)^\nu = \frac{\Gamma(2\nu)}{\nu^2 [\Gamma(\nu)]^3} \left(\frac{s}{4}\right)^\nu < \frac{(e\, s/\nu)^\nu}{2\sqrt{2\pi}}, \quad \gamma \leqq 1 \tag{5.3a}$$

$$E_\nu \leqq \frac{\gamma^{2\nu}(2\nu)!}{2(\nu!)^3} \left(\frac{s}{4}\right)^\nu < \frac{\gamma^{2\nu}}{2\sqrt{2\pi}} \left(\frac{e\, s}{\nu}\right)^\nu, \quad \gamma \geqq 1 \tag{5.3b}$$

$$E_\nu < \frac{\sqrt{(\gamma/2)\,[2(2\nu-1)]!}}{\nu!\,(2\nu-1)!} \left(\frac{s}{4}\right)^\nu$$
$$= \sqrt{\frac{\gamma\, \Gamma(4\nu)}{2(4\nu-1)}} \frac{(s/4)^\nu}{\nu\, \Gamma(\nu)\, \Gamma(2\nu)} \tag{5.3c}$$
$$< \frac{1}{2\pi} \left(\frac{\pi}{2\nu}\right)^{1/4} \sqrt{\frac{\gamma}{4\nu - 1}} \left(\frac{e\, s}{\nu}\right)^\nu$$

where we have used Stirling's formula in the form

$$\Gamma(n) \approx \sqrt{2\pi/n}\ (n/e)^n \left(1 + \frac{1}{12\, n}\right)$$

to get rid of the factorials. We note that (5.3c) is valid for all values of γ and provides a closer estimate than either (5.3a) or (5.3b). The latter, however, are somewhat easier to evaluate.

[9] See, e.g., p. 424 in G. A. KORN and T. M. KORN, Mathematical Handbook for Scientists and Engineers, McGraw-Hill, New York 1961.

6. Application to Mössbauer Spectroscopy

The above considerations are applicable to Mössbauer experiments of the type in which the transmitted gamma rays are detected. In this section we are primarily concerned with relating the quantities defined above to quantities relevant to Mössbauer spectroscopy. At first we consider only unsplit spectra. One usually studies the transmission as a function of ΔE. This quantity is varied by varying either E_S or E_A via the Doppler effect. Thus one obtains

$$\Delta E = E_C + (v/c)\, E_\gamma$$

where E_C is the sum of the isomeric and thermal shifts and E_γ is either E_A or E_S (it does not matter which, since in practice they differ by less than one part in 10^8). It follows that

$$x = E_C/\tfrac{1}{2}\Gamma_S + (v/c)\, E_\gamma/\tfrac{1}{2}\Gamma_S.$$

We identify $I_S(E)$ with the intensity of recoilfree radiation from the source. Also we ignore, for the sake of simplicity of presentation, the effect of electronic absorption. Many authors, e.g. MARGULIES and EHRMAN[1], have described how to take into account the effects of background radiation and of electronic absorption. In some early papers self-absorption in the source was considered. Since that time one has learned how to prepare sources that are free of resonantly absorbing nuclei. Therefore self-absorption is no longer so important, and we do not consider it here.

In many experiments one encounters *environmental broadening*, which may occur in the absorber as well as in the source. Possible causes of environmental broadening have been listed by O'CONNOR[4]. For any one single nucleus in the absorber we have

$$\sigma(E) = \frac{f_A\, \sigma_{res}}{1 + [(E - E'_A)/\tfrac{1}{2}\Gamma]^2},$$

where

$$\sigma_{res} = \frac{\lambda^2}{2\pi} \frac{2J_{ex} + 1}{2J_{gd} + 1} \frac{1}{1 + \alpha}$$

and $\Gamma = \hbar/(2\pi\tau)$, in the customary notation; f_A is the Mössbauer fraction in the absorber. The value of E'_A may vary from nucleus to nucleus because of local variations of the isomer shift, and we can

describe this variation by means of a distribution function. Let us assume that this distribution is Lorentzian so that we have

$$\mathrm{d}n = \frac{n/\pi}{1+[(E'_A - E_A)/(\varkappa_A - 1)\,\frac{1}{2}\,\Gamma]^2}\,\frac{\mathrm{d}E'_A}{(\varkappa_A - 1)\,\Gamma/2}\,,$$

where $(\varkappa_A - 1)\Gamma$ is the width of the distribution. Then, on the average, each nucleus in the absorber has a cross section

$$\langle\sigma(E)\rangle = \frac{1}{n}\int \frac{f_A\,\sigma_{res}}{1+[(E-E'_A)/\frac{1}{2}\,\Gamma]^2} \cdot \frac{n/\pi}{1+[(E'_A - E_A)/(\varkappa_A-1)\,\frac{1}{2}\,\Gamma]^2}\,\frac{\mathrm{d}E'_A}{(\varkappa_A-1)\,\Gamma/2}$$
$$= \frac{f_A\,\sigma_{res}/\varkappa_A}{1+[(E-E_A)/\varkappa_A\,\frac{1}{2}\,\Gamma]^2}\,.$$

We see that the width of the average cross section is the sum of the width of the distribution of E'_A and of the natural width Γ. Thus we can write

$$\Gamma_A = \varkappa_A\,\Gamma \quad \text{and} \quad s = t/\varkappa_A\,,$$

where

$$t = n\,f_A\,\sigma_{res}\,.$$

Similarly, if the environmental broadening in the source is also produced by a Lorentzian distribution of the line centers, then the intensity distribution remains Lorentzian but is broadened by a factor $\varkappa_S$, so that

$$\Gamma_S = \varkappa_S\,\Gamma \quad \text{and} \quad \gamma = \varkappa_A/\varkappa_S\,.$$

These equations make it possible to apply the formulas of Section 2 to the case of an unsplit source and an unsplit absorber, even when they are broadened differently.

We proceed to consider the case of a split source and an unsplit absorber. Here the intensity of the radiation from the source is given by

$$I(E)\,\mathrm{d}E = \sum_{i=1}^{N} \frac{A_i}{1+[(E-E_i)/\frac{1}{2}\,\Gamma_i]^2}\,\mathrm{d}E \qquad (6.1)$$

where

$$\Gamma_i = \varkappa_i\,\Gamma.$$

We insert (6.1) into Eq. (1.1) and introduce the dimensionless variables

$$z = (E - E_A)/\tfrac{1}{2}\,\Gamma_i\,, \quad x_i = (E_A - E_i)/\tfrac{1}{2}\,\Gamma_i$$

and

$$\gamma_i = \Gamma_A/\Gamma_i\,.$$

Thus Eq. (1.1) becomes

$$P = \tfrac{1}{2}\sum_{i=1}^{N} A_i\,\Gamma_i \int \frac{\exp\{-\gamma_i^2\,s/(z^2+\gamma_i^2)\}}{1+(z+x_i)^2}\,\mathrm{d}z$$
$$= \frac{\pi}{2}\sum_{i=1}^{N} A_i\,\Gamma_i\,\mathrm{tran}\,(\gamma_i, s; x_i)\,,$$

which can be evaluated by the formulas of Section 2.

The methods presented here cannot be applied rigorously to the case of a split absorber. If the absorption lines are widely spaced so that the approximation

$$\int I(E)\exp\{-n\sum_j \sigma_j(E)\}\,\mathrm{d}E \approx \sum_j \int I(E)\,\exp\{-n\,\sigma_j(E)\}\,\mathrm{d}E$$

can be introduced, then it is possible to proceed in the manner described by MARGULIES et al.[10]. Here $\sigma_j(E)$ denotes the cross section of the j^{th} absorption line.

7. Discussion

Replacing the numerical integration of the integral in Eq. (2.1) by our series will result in a saving of computation time. This is important in problems involving repeated calculations of ε (e.g. in the Newton-Raphson method or in the analysis of experimental spectra by the method of least squares). Thus the computation[11] of numerical values of the line width (for $\gamma = 1$) would have benefitted from the present formulation. Another advantage of the series over numerical integration is that it is much easier to estimate the error of computation. A further benefit of the series is that it has opened the way to investigating other properties of the transmission function; the width formula of HEBERLE and FRANCO[12] was derived from this series by analytical methods.

In applying our results to Mössbauer spectra, the user is warned to consider to what extent his case satisfies the assumptions that we have made explicitly and implicitly. Is re-radiation into the detector negligible? Are the directions of all rays from the source to the detector nearly the same, so that the assumption of parallelism is warranted? Is the thickness of the absorber uniform? Another question is whether or not our description of environmental broadening corresponds to what occurs in nature. Other authors[10,13] have considered a Gaussian broadening. However, the fact that the use of a Lorentzian line shape in the fitting of experimental spectra has been successful so often, tends to support our description.

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[11] J. HEBERLE, Nucl. Instr. Methods **58**, 90 [1968].
[12] J. HEBERLE and S. FRANCO, Bull. Am. Phys. Soc. **13**, 61 [1968].
[13] S. MARGULIES, Z. Phys. **176**, 63 [1963].

A number of relevant questions remain unanswered by the present investigation. Is it possible to express $\mathrm{tran}(\gamma, s; 0)$ in terms of known functions for $\gamma \neq 1$? Is it possible to prove that all the $a(k,l,m)$ are indeed integers? We remark that $c(k,l,m) = (l/m)\, a(k,l,m)$ seem to be integers; perhaps it is easier to prove that the $c(k,l,m)$ are integers.

It has been reported[3,14] that for $\gamma = 1$ the line shape can be approximated by a Lorentzian in the range $0 \leqq s \leqq 10$. Is this also true for other values of γ? In view of Eq. (4.7), we may speculate that the Lorentzian line shape is an even better approximation for $\gamma < 1$ than for $\gamma = 1$.

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[14] P. STEINER, E. GERDAU, W. HAUTSCH, and D. STEENKEN, p. 364 in Hyperfine Interactions and Nuclear Radiations, edited by E. MATTHIAS and D. A. SHIRLEY, North-Holland Publishing Co., Amsterdam 1968.

Zur phänomenologischen Begründung erweiterter Casimir-Onsagerscher Reziprozitätsbeziehungen

W. MUSCHIK

II. Institut für Theoretische Physik der Technischen Universität Berlin

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Attempts are made to give phenomenological reasons for nonlinear Casimir-Onsager reciprocal relations. The fluxes can be defined as time derivations of state variables, or they can be explained by means of balance equations, because only their vectorial properties are used. At first, time reversal is replaced by an abstract parameter reversal from which involutoric transformations of forces and fluxes result. The connection between the parameter reversal of forces and fluxes allows to give reasons for relations which are equal to the Casimir-Onsager reciprocal relations apart from a sign. This sign is determined by experience. The connection between parameter and time reversal is discussed.

Als erster regte wohl DAVIES[1] an, die wohlbekannte statistische Begründung der **C**asimir-**O**nsagerschen **R**eziprozitäts-**B**eziehungen (CORB)[2–4] durch eine phänomenologische Begründung zu ergänzen. COLEMAN und TRUESDELL[5] und MACKE[6] haben unter gewissen einschränkenden Voraussetzungen wie der alleinigen Existenz von α-Kräften, der Abwesenheit ungerader Parameter sowie der Annahme, daß die Flüsse Zeitableitungen von Zustandsvariablen seien, mit phänomenologischen Hilfsmitteln eine Begründung für die Symmetrie der phänomenologischen Matrix gegeben.

Kürzlich wurde versucht[7], für nicht notwendig lineare, homogene phänomenologische Gleichungen unter Aufhebung der in [5] und [6] gemachten Einschränkungen eine phänomenologische Begründung der CORB zu geben.

Durch Zerlegung der phänomenologischen Gleichungen

$$\boldsymbol{i} = \mathfrak{L}(\boldsymbol{x}) \tag{1}$$

mit Hilfe der Taylor-Entwicklung in ihre symmetrischen und antisymmetrischen Anteile lassen sich dissipationslose und entropieerzeugende Flüsse

$$\boldsymbol{i}^{\mathrm{a}}(p) = \mathfrak{L}^{\mathrm{a}}_p[\boldsymbol{x}(p)], \quad \boldsymbol{i}^{\mathrm{s}}(p) = \mathfrak{L}^{\mathrm{s}}_p[\boldsymbol{x}(p)] \tag{2}$$

einführen. Dabei ist

$$p = \{p_1, p_2, \ldots, p_m\} \tag{3}$$

ein Satz von endlich vielen Parametern, die die folgenden Voraussetzungen erfüllen sollen:

A. Die Parameter sind umkehrbar.

B. Während der Prozesse, die in den betrachteten Systemen ablaufen, bleiben die Parameter konstant.

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