

## Primitive Theory of Ferrimagnetic Resonance Frequencies in Rare-Earth Iron Garnets

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The effective  $g$  factor for ferrimagnetic resonance frequencies in rare-earth iron garnets is calculated by direct inspection of eigenvalues rather than study of the equations of motion. The rare-earth ions are treated as captive in the exchange field from the iron, but subject to decomposition of their energy levels by crystalline fields and/or spin-orbit interaction. With a crystalline field, the problem is tractable in a simple way only if these decompositions are large or small compared to those which could be produced by the exchange field acting alone. Anisotropy, actually very important at low temperatures, is neglected except insofar as it can be represented by an anisotropy field. The concept of "fictitious spin" is useful, and the spectroscopic

splitting factors turn out to be more relevant than the true gyromagnetic ratios. For europium garnet, our theory becomes essentially that of Wolf. It is shown that Kittel's formula  $g_{\text{eff}} = 2(M_{\text{Fe}} + M_{\text{RE}})/M_{\text{Fe}}$  has approximate validity if most of the magnetic moment of the rare earth arises from nondiagonal matrix elements joining ionic energy levels with separations large compared with the Zeeman energy in the exchange field. The fact that in certain cases the experimental results are represented fairly well by Kittel's formula is hence not necessarily to be construed as evidence that the rare-earth ion is highly damped by spin-lattice interaction as in his original model.

IN the early days of ferromagnetic resonance, there was considerable confusion between the spectroscopic splitting factor and the true gyromagnetic ratio. Some of this confusion seems to still persist in the ferrimagnetic garnets, as we hope the present paper will show.

There are two general methods commonly employed for calculating ferrimagnetic resonance frequencies. One is the use of classical equations of motion; the other employs spin wave or harmonic oscillator approximations applied to the secular problem of the coupling between two large angular momentum vectors. The two methods are essentially equivalent.<sup>1</sup> In our opinion, the second is a little clearer in exhibiting the physical nature of the approximations.

In this paper we present still a third method which is more primitive, but in our opinion more intuitive and less likely to be misleading. We shall assume that the specimen is spherically cut so that there are no demagnetization corrections. Also, we will forget about anisotropy, except, of course, insofar as it can be handled phenomenologically by the usual artifice of an anisotropy field. We will develop our theory explicitly for the garnets, but it is readily adapted to any situation in which the magnetization of one sublattice is a "captive" one created by the exchange field from the other.

If the crystal exists in a state of spontaneous magnetization, which is not substantially changed by the applied field, it will have  $2n+1$  equally spaced eigenvalues in an applied field, where  $2n+1$  is the number of possible quantized orientations of the magnetization of the specimen. The quantity  $n$  may be called the fictitious spin and is, of course, an enormous number.

<sup>1</sup> For references and some discussion of this point, see J. Van Kranendonk and J. H. Van Vleck, *Revs. Modern Phys.* **30**, 18 (1958). The harmonic oscillator approximation, incidentally, is more or less similar to that employed by S. Golden and J. K. Bragg in the mathematically related chemists' problem of approximating high eigenvalues of the asymmetrical top [J. Chem. Phys. **17**, 439 (1949)].

We may define quantities  $n_{\text{Fe}}$ ,  $n_{\text{RE}}$  similarly for the iron and rare-earth sublattice, and we may take  $n = n_{\text{Fe}} \pm n_{\text{RE}}$  inasmuch as in the garnets, the moment of the captive rare-earth lattice is created by the exchange field largely from the iron, and so the moment of the rare-earth lattice is practically parallel or antiparallel to that of the iron. Assumptions fully as restrictive as those we have just made, and probably essentially equivalent, are introduced in the two standard procedures described above when it is assumed in them that the moduli of the magnetic moment of the two sublattices are each constants of the motion. The state of lowest energy is that where the moment is parallel to the applied field, so that

$$E = -H(M_{\text{Fe}} + M_{\text{RE}}).$$

The effective  $g$  factor in ferrimagnetic resonance, when multiplied by  $\beta H$ , is the spacing of two adjacent eigenvalues and is, hence,

$$g_{\text{eff}} = (M_{\text{Fe}} + M_{\text{RE}})/[(n_{\text{Fe}} \pm n_{\text{RE}})\beta], \quad (1)$$

where  $\beta$  is the Bohr magneton.

As the ferric ions are effectively in  $S$  states, we may take  $n_{\text{Fe}} = M_{\text{Fe}}/2\beta$ . To make our expression (1) acquire a form superficially like that usually employed in the literature, we define a ratio  $G_{\text{RE}}$  by

$$G_{\text{RE}} = M_{\text{RE}}/[\pm(n_{\text{RE}})\beta]. \quad (2)$$

Then

$$g_{\text{eff}} = (M_{\text{Fe}} + M_{\text{RE}})/(\frac{1}{2}M_{\text{Fe}} + G_{\text{RE}}M_{\text{RE}}). \quad (3)$$

We have taken  $M_{\text{Fe}} > 0$ , and then  $M_{\text{RE}}$  is negative for most of the rare-earth garnets; but up to samarium, and also for samarium at low temperatures,  $M_{\text{RE}}$  has the same sign as  $M_{\text{Fe}}$ .

Our problem is now to evaluate  $\pm n_{\text{RE}}$ , and our essential point is that this is a matter of numerology rather than of calculating true angular momentum. We assume that in the absence of the exchange and applied fields, an individual rare-earth ion has a number of energy levels  $E_f$  which are separated from each

other because of the crystalline field and/or spin-orbit interaction. Each of these levels is in general  $(2S_{\Gamma}+1)$ -fold degenerate, and we speak of  $S_{\Gamma}$  as the fictitious spin of the level. For spin-orbit splitting in the absence of a crystalline field,  $S_{\Gamma}$  is, of course, the same as the inner quantum number  $J$ . We use capital letters throughout for spins and gyromagnetic ratios or spectroscopic splitting factors which have only a pseudo-meaning. For a cubic field  $S_{\Gamma}$  can be  $\frac{1}{2}$  or  $\frac{3}{2}$  for an odd ion, and 0 or 1 for an even one.<sup>2</sup> The important thing is that inside each of these degenerate manifolds, the transformation properties of the angular momentum and magnetic moment matrices are, except for a proportionality factor, the same as those for an ion with an honest spin of  $S_{\Gamma}$ . Consequently, we can write

$$\pm n_{\text{RE}} = -A \sum_{\Gamma} \sum_{M=-S_{\Gamma}}^{S_{\Gamma}} M e^{-E_{\Gamma M}/kT}. \quad (4)$$

Here  $E_{\Gamma M}$  is one of the substates into which  $E_{\Gamma}$  is decomposed by the exchange field  $H_{\text{ex}}$  emanating from the iron sublattice, and the proportionality constant  $A$  is the number of rare-earth ions divided by the partition function of an ion. The negative sign in (4) is necessary, as the ferrimagnetic lattice has a negative gyromagnetic ratio because of the negative sign of electronic charge, and so the spin of the ferric sublattice in the direction of  $M_{\text{Fe}}$  is negative if  $M_{\text{Fe}} > 0$ . The question of sign choice in (2) is eliminated when (4) is used, since the right side of (4) is negative when the minus sign applies. To compute  $M_{\text{RE}}$  we introduce for mathematical convenience a small applied field  $H_0$  parallel to the exchange field. We then can use the relation

$$M_{\text{RE}} = -A \sum_{\Gamma} \sum_M (-\partial E/\partial H_0) e^{-E_{\Gamma M}/kT}. \quad (5)$$

After the derivative is taken,  $H_0$  may be set equal to zero, as any applied field is negligible compared with the exchange one. We now assume that  $\beta H_{\text{ex}}$  is small compared with the splitting of the energy levels  $E_{\Gamma}$  produced by the crystalline field or spin-orbit interaction. Then the energy levels can be expressed as a

<sup>2</sup> It should, however, be noted that the method of fictitious spin is rigorous for the case  $S = \frac{3}{2}$  only if the first-order Zeeman energy is linear in the magnetic quantum number  $M$ , as assumed in our Eq. (6). Actually, this need not be the case in a cubic field, and instead it is either necessary to include a term in  $M^3$  or alternatively to assume that  $g$  depends on the magnetic quantum number  $M$  [Cf. A. Abragam and M. L. Pryce, *Proc. Phys. Soc. (London)* **63**, 409 (1950), and especially B. Bleaney, *ibid.* **73**, 939 and **74**, 493 (1959)]. The cubic term gives anisotropy in the Zeeman effect, though not the susceptibility; and in the framework of our isotropic theory, which of necessity neglects the cubic term in  $M$ , the best approximation is probably to use the single  $g$  value which yields the proper susceptibility or, in other words, proper second moment. It should also be mentioned that for even ions in a cubic field there can be doubly degenerate levels (Bethe's type  $\Gamma_3$ ); since they are void of any magnetic moment, it is immaterial whether they are formally treated as having a fictitious spin  $S = \frac{1}{2}$  or as two coincident levels with  $S = 0$ .

series

$$E_{\Gamma M} = E_{\Gamma} + (G_{\Gamma} H_0 + \tilde{G}_{\Gamma} H_{\text{ex}}) \beta M + \frac{1}{2} [\alpha_{\Gamma M} H_0^2 + 2\tilde{\alpha}_{\Gamma M} H_0 H_{\text{ex}} + \alpha'_{\Gamma M} H_{\text{ex}}^2]. \quad (6)$$

Note particularly that the quantity  $\tilde{G}_{\Gamma}$ , which may be termed the exchange spectroscopic splitting factor, is not the same as the conventional splitting factor  $G_{\Gamma}$ , since the exchange field acts only on the spin moment, whereas a true magnetic field acts on the total moment. (The relation between the two  $G$ 's is  $\tilde{G}_{\Gamma} = 2(g_J - 1)G_{\Gamma}/g_J$ , where  $g_J$  is the Landé factor of the multiplet component  $J$  of the free ion from which  $\Gamma$  is derived.) We will keep only the terms of the first order in the development of (4) and (5) in the exchange field. With this approximation, we find from (4) and (5), evaluating  $\sum M^2$  in the usual way, that (2) becomes

$$G_{\text{RE}} = \frac{\sum_{\Gamma} (2S_{\Gamma}+1) [S_{\Gamma}(S_{\Gamma}+1) G_{\Gamma} \tilde{G}_{\Gamma} + 3kT \tilde{\alpha}_{\Gamma} \beta^{-2}] e^{-E_{\Gamma}/kT}}{\sum_{\Gamma} \tilde{G}_{\Gamma} (2S_{\Gamma}+1) S_{\Gamma}(S_{\Gamma}+1) e^{-E_{\Gamma}/kT}}, \quad (7)$$

where

$$\tilde{\alpha}_{\Gamma} = \sum_M \tilde{\alpha}_{\Gamma M} / (2S_{\Gamma}+1).$$

It is to be cautioned that formula (7), or even the expression (4) for the enumeration of states and our whole concept of fictitious spin for the RE sublattice, should not be used unless  $\beta H_{\text{ex}}$  is small compared to the intervals  $E_{\Gamma} - E_{\Gamma'}$  separating the various states  $E_{\Gamma}$ . Fortunately, this condition is usually met in the rare-earth garnets, especially at higher temperatures, where the exchange fields are smaller. It is clear that (7) cannot be employed for arbitrary values of  $\beta H_{\text{ex}}$ , for (7) does not in general reduce in the limit  $E_{\Gamma} - E_{\Gamma'} = 0$  to the value  $g_J$  for the free ion. (This difficulty does not arise, however, if the splittings are caused solely by spin-orbit interactions.) If the crystalline field is small compared to the exchange fields, it is, of course, a better approximation to treat the ion as free. In the intermediate case, there appears to be no simple formula.

Another difficulty is that, actually, in the garnets the field is not cubic locally, although macroscopically the symmetry is cubic. We can, however, regard our model as the first approximation and treat the local deviations from cubic symmetry as a perturbation which we ignore or, rather, relegate to the catch-all of anisotropy corrections. An alternative procedure, in case the deviations from cubic symmetry are quite pronounced, is to treat all levels as at most two-fold degenerate ( $S_{\Gamma} = \frac{1}{2}$ ) and use for each Kramers doublet an isotropic  $G$  equal to the mean of that for the three principal local directions.

Equations (3) and (7) are to be compared with the Tsuya-Wangsness<sup>3</sup> formula:

$$g_{\text{eff}} = (M_{\text{Fe}} + M_{\text{RE}}) / (\frac{1}{2} M_{\text{Fe}} + g^{-1} M_{\text{RE}}), \quad (8)$$

<sup>3</sup> N. Tsuya, *Progr. Theoret. Phys. (Kyoto)* **7**, 263 (1952); R. K. Wangsness, *Phys. Rev.* **91**, 1085 (1953).

and the formula of Kittel<sup>4</sup>:

$$g_{\text{eff}} = (M_{\text{Fe}} + M_{\text{RE}}) / \frac{1}{2} M_{\text{Fe}}. \quad (9)$$

The quantity  $g$  in (8) is the gyromagnetic ratio (in units of  $-e/2mc$ ) of the rare-earth lattice, or equally well, its Landé  $g$  factor, as Tsuya and Wangsness did not consider crystalline fields or more than one multiplet component. Comparing (8) and (3), we see that our formula is similar to that of Tsuya or Wangsness except that it involves  $G_{\text{RE}}$ , which we may call a fictitious gyromagnetic ratio, instead of a real one.

The relation of our formula to the Tsuya-Wangsness and Kittel ones, and its meaning, will be clearer if we consider some specific examples:

(i) Gadolinium iron garnet. Here the crystal field can be considered as negligible compared to the exchange field, and one has  $G_{\text{RE}} = g = 2$ . Then (3) reduces to the Tsuya-Wangsness formula, which is known to work quite well in gadolinium garnet. This is not surprising, as this is the case where the ions are least influenced by the crystalline field.

(ii) Europium iron garnet. Analysis of the susceptibility shows that it is a pretty good approximation in europium to treat the ions as free, the reason being that the two lowest levels,  $J=0$  and  $J=1$ , are undecomposed by a cubic field. The decomposition of the levels is then to be considered as due to spin-orbit interaction, and it is well-known that Eu and Sm are the two rare earths for which it is necessary to include more than one multiplet component. The case of Sm we will discuss elsewhere.

If we neglect the crystalline field, the quantity  $G_{\text{RE}}$  is the same as the gyromagnetic ratio  $\tilde{g}$  in an exchange field. In Eu the formula for  $\tilde{g}$  is

$$\tilde{g} = \frac{48 + (9X - 3)e^{-X} + (45X - 5)e^{-3X} + \dots}{X(6e^{-X} + 30e^{-3X} + \dots)}, \quad (10)$$

where

$$X = (E_{J=1} - E_{J=0}) / kT = 480/T.$$

This is not quite the same as the expression given in my book<sup>5</sup> for the gyromagnetic ratio of Eu but is obtained by adaptation to the case that the field acts only on the spin, as discussed by Wolf and Van Vleck.<sup>6</sup> Since the denominator of (10) vanishes except insofar as the upper multiplet components are inhabited,  $1/\tilde{g}$  is quite small ( $\sim 0.04$  at  $300^\circ$ ). As a first approximation, one can take  $1/\tilde{g} = 0$ , and then one has the same formula as that used by Wolf<sup>7</sup> to explain the results of Miyadai<sup>8</sup> on ferrimagnetic resonance in the europium garnet. Thus, the formula which Wolf<sup>7</sup> employed is, as he

observed, the same as that of Kittel, but derived by a completely different mechanism. Wolf noted that in europium the state  $J=0$  does not contribute to the angular momentum. As long as one is dealing with free ions not subject to crystalline fields, angular momentum is a measure of the number of states. Thus, though couched in different physical language, the physical picture which Wolf uses is basically similar to our own. By means of (10), we can correct for the effect of upper states. Unfortunately, this gives a slight correction in the wrong direction, the discrepancies with experiment becoming respectively about 4%, 12%, rather than 2%, 10% at  $T = 300^\circ, 500^\circ$ . These annoying corrections can be avoided if it is assumed that the level  $J=1$  is split by the noncubic portion of the field by an amount large compared to the Zeeman energy in the exchange field.

(iii) Ytterbium iron garnet. Besides Gd and Eu, this is the only garnet for which the crystalline field theory of the susceptibility seems pretty well under control. The lowest level is a Kramers doublet whose  $G_{\text{R}}$  value is moderately isotropic and has approximately the value  $G_{\text{R}} = 24/7$  appropriate to a cubic field.<sup>9</sup> The next level is about  $580 \text{ cm}^{-1}$  up, so that its Boltzmann factor can be neglected, and (7) becomes

$$G_{\text{RE}} = (24/7)[1 + (T/310)]. \quad (11)$$

The factor  $1 + (T/310)$  results from including the  $\tilde{\alpha}_{\text{R}}$  part of (7), or in other words, the temperature-independent portion of the susceptibility, which is known in YbG both theoretically and experimentally.<sup>10</sup> For a free Yb ion, the Landé  $g$  factor or gyromagnetic ratio is

$$g = 8/7, \quad (12)$$

and this is the value of  $g$  to be used in the Tsuya-Wangsness formula taken literally or naively. Comparison of (11) and (12) illustrates the danger of confusing spectroscopic splitting or fictitious gyromagnetic factors with true gyromagnetic ratios. As YbIG has a compensation point  $M_{\text{RE}} = -M_{\text{Fe}}$  near  $T=0$ , there is a temperature at which (8) has a zero denominator if (12) is used, but this catastrophe does not take place with (11) in (3).

If one interprets the resonance data of Rodrigue, Meyer, and Jones<sup>11</sup> by a formula of type (3), then the value of  $G_{\text{RE}}$  which is needed is about 4.0 at room temperatures and above; while at lower temperatures, the empirical value of  $G_{\text{RE}}$  so obtained drops gradually to about 1.0 at  $T=0$ . The experimental error in so determining  $G_{\text{RE}}$  is considerable; Rodrigue states

<sup>9</sup> Y. Ayant and J. Thomas, *Compt. rend.* **248**, 387 (1959); D. Boakes, G. Garton, M. J. M. Leask, and W. P. Wolf, *Proc. Phys. Soc. (London)* **74**, 663 (1959); J. W. Carson and R. L. White, *J. Appl. Phys.* **31**, 53S (1960).

<sup>10</sup> M. Ball, G. Garton, M. J. Leask, and W. P. Wolf, *Proceedings of the Seventh International Conference on Low-Temperature Physics* (University of Toronto Press, Toronto, Canada, 1960).

<sup>11</sup> G. P. Rodrigue, H. Meyer, and R. V. Jones, *J. Appl. Phys.* **31**, 376S (1960).

<sup>4</sup> C. Kittel, *Phys. Rev.* **115**, 1587 (1959).

<sup>5</sup> J. H. Van Vleck, *The Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1932), p. 256.

<sup>6</sup> W. P. Wolf and J. H. Van Vleck, *Phys. Rev.* **118**, 1492 (1960).

<sup>7</sup> W. P. Wolf, *J. Phys. Soc. Japan* **15**, 2104 (1960).

<sup>8</sup> T. Miyadai, *J. Phys. Soc. Japan* **15**, 2205 (1960); Y. Shichijo, T. Miyadai, and H. Takata, *ibid.* **15**, 530, 1534 (1960).

(unpublished thesis) that it may be as much as 50%. Even so, the fit with (11) is not good, though at least better than with (12) or with the value  $G_{\text{RE}} = \infty$  demanded by (9). The reason for the discrepancy is not clear—whether it is caused by difficulty with anisotropy corrections, failure to correct for the repercussions of the exchange field on the spin of the iron lattice, or what.

(iv) Kittel's formula. For certain garnets at high temperatures, Kittel<sup>4</sup> has proposed the formula (9) which is identical with the limit  $1/G_{\text{RE}} = 0$  of our expression (3). His model is, however, quite different from our own, as he has the rare-earth ion heavily damped by spin-lattice interaction, whereas we have no damping and simply utilize the static crystalline field (and in Sm and Eu, the natural multiplet splittings). We believe our model is the more plausible. Our rederivation of essentially his result as a limiting case by another mechanism under certain conditions we regard as the main contribution of the present paper.

Our reluctance to interpret (9) as mainly the result of a damping or spin-lattice process is chiefly because this interpretation would imply that the spin-lattice interaction distorts the Zeeman pattern as much as does the crystalline field, and this does not seem very reasonable. Also, we have some doubts as to whether the Landau-Lifshitz damping term used by Kittel is, in general, an adequate representation of the necessarily complicated spin-lattice coupling effects.

In a certain sense, however, the difference between our theory and Kittel's is a semantic one, and it is easy to see why we get somewhat similar formulas. The important thing to obtain his formula is that the contribution of the rare earths to the number of eigenvalues of the specimen be suppressed. At the same time, however, the rare earths must make a substantial contribution to the magnetization of the crystal, as otherwise one has the trivial case  $g_{\text{eff}} = 2$ . Under such circumstances the magnetic moment of the rare-earth sublattice may be said to be semiquenched. One can immediately understand why (9) usually applies, if at all, only at high temperatures, for a necessary condition for semiquenching is that the exchange field be small compared to the crystalline field, be it static or oscillatory, and this condition is more likely to be met at high temperatures. We prefer to think of semiquenching not as damping of angular momentum, but rather as relegation of the rare earths' magnetic contribution to being via second- rather than first-order Zeeman effect. As already mentioned, this difference is to a certain extent semantic. There does remain, however, the question as to whether the semiquenching is a static or dynamic effect; i.e., is caused primarily by the ordinary crystalline field or by the modulations of this field by lattice vibrations. No doubt both effects are present, and in this sense Kittel's theory and ours are complementary. However, we wonder whether it would be possible to have most of the semiquenching caused by

spin-lattice interaction without introducing excessive real quenching at high temperatures; i.e., suppression of the rare earths' magnetic susceptibility when the temperature is raised and spin-lattice interaction becomes stronger.

To obtain exactly the Kittel formula with our model, we would have to have the degeneracy of the inhabited rare-earth states lifted completely with a decomposition large compared to  $\beta H_{\text{ex}}$ . This could conceivably happen for even ions in sufficiently asymmetrical fields. However, we doubt if this is the actual situation. In odd ions there is always the Kramers degeneracy, so that, with our theory, the Kittel formula would never apply accurately [barring very fortuitous cancellation of the various terms in the denominator of (7), for as we shall see later,  $\tilde{G}_T$  can have different signs for different states]. In our opinion, the explanation is rather that a large part, but not all, of the susceptibility often arises from the nondiagonal part of the magnetic moment which does not contribute to the number of states. Furthermore, oftentimes the spectroscopic splitting factors  $G_T$  involved in (7) are quite large. We thus expect that normally Kittel's formula is but an approximation, and so it is experimentally. All told, on our model one might expect Kittel's formula to apply somewhat better to even than to odd ions. Actually, of the two ions for which Kittel's formula holds best—Ho and Er—one is even and one is odd. Possibly this is evidence for the dynamic rather than static interpretation, for with overpowering spin-lattice coupling, though Kramers' theorem retains its validity, there are so many energy levels that it ceases to be of interest and the even-odd distinction disappears. Another argument in favor of the dynamic view can be made from considerations of linebreadths, since the damping model nicely explains the anomalously large linebreadth over a certain temperature interval when small amounts of certain rare earths are added to YIG. It seems clear that the anomaly occurs in the transition region where the exchange field becomes large enough to overcome semiquenching. Such a situation can arise with either the static or oscillatory crystal fields, but whether the dependence on temperature is critical enough with the static model is uncertain.

We should not forget to mention the possibility that sometimes the denominator of our expression (3) can be larger than in Kittel's formula (9), either because  $G_{\text{RE}} < 0$ ;  $M_{\text{RE}} < 0$ , or  $G_{\text{RE}} > 0$ ;  $M_{\text{RE}} > 0$ . The expression (7) for  $G_{\text{RE}}$  can, indeed, in principle, on occasion be negative, since some states carry negative  $G_T$ . An example of a negative  $G_T$  is provided by the  $\Gamma_4$  triplet of a  $J=3$  level in a cubic field, for which  $G_T = -\frac{3}{2}g_J$ .<sup>12</sup> We doubt, however, that negative values of  $G_{\text{RE}}$  are very common. The situation  $M_{\text{RE}} > 0$ , however, arises in the first part of the rare-earth sequence, since here the exchange field works in the opposite direction from an

<sup>12</sup> A. Abragam and M. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951); J. H. Van Vleck, Physica **26**, 544 (1960).

applied field, and, in consequence, the coupling between the rare-earth and iron sublattices, though basically of antiferromagnetic sign, makes the moment of these sublattices parallel rather than antiparallel.<sup>13</sup>

In closing, we should emphasize that the present paper makes no pretense of including anisotropy, and it can be regarded rather as an attempt to see how far one can push the theory with a purely isotropic model. Of course, the effects of anisotropy are particularly

important at low temperatures. The theory usually does not appear to work too well at low temperatures if the correction for anisotropy is made in the usual way by introducing an anisotropy field. The question of how far it is warranted to include the anisotropic part of the crystalline potential simply through this artifice is a subject into which we do not want to enter here.

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<sup>13</sup> V. Jaccarino, B. T. Matthias, M. Peter, H. Suhl, and J. H. Wernick, *Phys. Rev. Letters* **5**, 251 (1960); G. Goldring, M. Schieber, and Z. Vager, *J. Appl. Phys.* **31**, 2057 (1960); W. P. Wolf, *ibid.* **32**, 742 (1961).

## Debye-Waller Factor in Mössbauer Interference Experiments

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A simple calculation is presented of the effects of lattice dynamics on interference between Mössbauer processes and corresponding atomic processes, i.e., between Mössbauer and Rayleigh scattering, or between internal conversion of Mössbauer radiation and the photoelectric effect. When the energy of the emitted  $\gamma$  ray or electron is not measured, it is necessary to sum over all possible final states of the lattice. The interference contribution is found to be attenuated by the same "Debye-Waller" factor as the ordinary Mössbauer contribution, depending only upon the momentum of the incident  $\gamma$  ray. If the energy of the emitted  $\gamma$  ray is measured (e.g., by a Bragg scattering experiment), the atomic contribution is attenuated by the usual x-ray Debye-Waller factor, depending upon the momentum transfer, the Mössbauer contribution by the square of the usual Mössbauer factor, and the interference term by the geometric mean of the atomic and Mössbauer factors.

IT is now generally known<sup>1-3</sup> that the effects of lattice dynamics in Mössbauer experiments are expressed very simply in terms of the fraction  $f$  of gamma rays emitted from the source without energy loss due to recoil. This is the Debye-Waller factor<sup>4</sup>

$$f = |\langle i | \exp(-i\mathbf{k} \cdot \mathbf{X}_L) | i \rangle|^2, \quad (1)$$

where  $|i\rangle$  is the initial state of the lattice,  $\hbar\mathbf{k}$  is the momentum of the gamma ray, and  $\mathbf{X}_L$  is the coordinate of the nucleus emitting the gamma ray. Interest has recently been expressed in interference between atomic effects and the Mössbauer effect; e.g., between Rayleigh and Mössbauer scattering,<sup>5</sup> or between atomic photoelectric absorption and Mössbauer absorption followed by emission of a conversion electron.<sup>6</sup> The purpose of this note is to point out that the effect of

the lattice dynamics on the interference term is given by the *same factor*  $f$  which appears in the direct Mössbauer term.

Let us consider the scattering of a gamma ray of momentum  $\hbar\mathbf{k}_1$  into a state of momentum  $\hbar\mathbf{k}_2$  by an atom whose motion in the lattice is described by the coordinate  $\mathbf{X}_L$ . Let  $M$  be the probability amplitude for the process due to the Mössbauer effect, and let  $A$  be the amplitude for Rayleigh scattering by the atomic electrons. Then the scattering cross section will be given by

$$\sigma \propto \{|A|^2 + |M|^2 + 2C\text{Re}(A^*M)\}, \quad (2)$$

where  $C$  is a factor expressing the degree of coherence of the two elementary processes. This factor  $C$  is independent of the lattice and is not considered further here.

We wish to investigate the effect of the lattice dynamics upon Eq. (2). From ordinary Mössbauer and Rayleigh scattering we know that the direct Mössbauer term  $|M|^2$  is proportional to  $f$ , and that the direct Rayleigh term  $|A|^2$  is independent of the lattice dynamics. The dependence of the interference term upon the lattice is not evident, *a priori*.

The coherence properties of the final lattice states

<sup>1</sup> *Proceedings of Illinois Conference on the Mössbauer Effect*, edited by H. Frauenfelder and H. Lustig (University of Illinois Urbana, Illinois, 1960).

<sup>2</sup> H. J. Lipkin, *Ann. Phys.* **9**, 332 (1960).

<sup>3</sup> C. Tzara and R. Barloutaud, *Phys. Rev. Letters* **4**, 405 (1960).

<sup>4</sup> I. Waller, *Ann. Physik* **79**, 261 (1926); W. Marshall and J. P. Schiffer, Atomic Energy Research Establishment Report, 1959 (unpublished).

<sup>5</sup> P. J. Black and P. B. Moon, *Nature* **188**, 481 (1960).

<sup>6</sup> L. J. Tassie (to be published). See also reference 1, p. 25.