PHYSICAL REVIEW VOLUME 160, NUMBER 2 10 AUGUST 1967

Theory of Three-Magnon Ferromagnetic Relaxation Frequency for Low Temperatures and Small Wave Vectors

M. SPARKS

North American Aviation Science Center, Thousand Oaks, California

(Received 27 February 1967)

The three-magnon confluence relaxation frequency $1/\tau$ is calculated without making assumptions of the previous calculation of Sparks, Loudon, and Kittel. The results are such that the discrepancies between the experiments of Comstock, LeCraw, Nilsen, Remeika, Spencer, and Walker and the previous theory are removed. In particular, the exponentially small (rather than linear) dependence of $1/\tau$ on k_1 and T is explained by the new theory. A calculation of the bending down of $1/\tau$ below linearity in k_1 at large values of k_1 is also given.

All pertinent experimental results reported to date agree qualitatively with the new theoretical results; several experiments agree extremely well quantitatively. For example, LeCraw and Spencer's $1/\tau$'s are within 7% of the new theoretical values for all k_1 between 0.4×10^5 and 2.1×10^5 cm^{-1}. In other experiments, the observed values of $1/\tau$ are considerably larger than the theoretical values (an order of magnitude in the worst case); it appears that some other process is operative in these experiments. Measurements on ultrahigh-purity samples would be helpful in identifying this process.

I. INTRODUCTION

THE typical accuracy of ferromagnetic relaxation frequency calculations made previous to 1955 was a few orders of magnitude.[1] Recently there have been calculations for several processes which agree with experiment to within a few tens of percents.[2–4] The three-magnon confluence process should perhaps show the best agreement of any of these processes because the values of all of the parameters in the theoretical result are accurately known. Indeed, LeCraw and Spencer[5] found that the slope of the magnon line width ΔH_k versus wave number k_1 for very pure yttrium-iron garnet (YIG) with k_1 between 0.4 and 1.6×10^5 cm^{-1} at room temperature and a pump frequency ω_p of 8.2 Gc/sec agreed with the theoretical value calculated by Sparks, Loudon, and Kittel[4] to within 12%. Other results for YIG and europium and gadolinium iron garnets (EuIG and GdIG) reported by Comstock, Nilsen, Raymond, Remeika, and Walker,[6–9] at various temperatures and frequencies do not show such good agreement with theory.[4] In one case the disagreement is greater than an order of magnitude. Departures from the predicted[4] linearity in T at low values of T and from the predicted[4] linearity in k_1 at both high and at low values of k_1 have also been observed.

In the present paper the relaxation frequency for the three-magnon confluence process is calculated without making the following approximations which were previously made.[4,10] The high-temperature approximation is not made, and the bending down of ΔH_k from linearity at large k_1 values is calculated accurately. The first-order correction for the dipole–dipole demagnetization-energy term in the dispersion relation is calculated.

The new theoretical results agree to within 7% with LeCraw and Spencer's measurements[5] for all reported values of k_1 from 0.45 to 2.0×10^5 cm^{-1}, including the bending down from linearity starting at $k_1\cong1.6\times10^5$ cm^{-1}. The results also explain the observed departure from linearity in T and the drop below linearity in k_1 for small values of k_1. All pertinent experimental results reported to date agree qualitatively with the new theoretical results; however, several experiments at low temperature do not agree quantitatively with these results. It would be interesting to measure ΔH_k versus k_1 at various low temperatures for the ultrapure YIG samples of Spencer and Remeika[11] to determine if the discrepancy is associated with impurities in the sample.

2. RELAXATION FREQUENCY CALCULATION

Since the parallel-pumping experiment[12] measures the relaxation frequency of $\frac{1}{2}\pi$ magnons, i.e., magnons whose wave vector $\mathbf{k}_1$ is perpendicular to the applied field $\mathbf{H}_{\rm app}$, the calculation will be made explicitly for these magnons. The relaxation frequency $1/T$ is often

[1] E. Abrahams, *Advances in Electronics and Electron Physics* (Academic Press Inc., New York, 1954), Vol. 6.
[2] R. W. Teale and K. Tweedale, Phys. Letters **1**, 298 (1962).
[3] T. Kasuya and R. C. LeCraw, Phys. Rev. Letters **6**, 223 (1961).
[4] M. Sparks, R. Loudon, and C. Kittel, Phys. Rev. **122**, 791 (1961).
[5] R. C. LeCraw and E. G. Spencer, J. Phys. Soc. Japan. Suppl. **17**, 401 (1962).
[6] R. L. Comstock, Appl. Phys. Letters **6**, 29 (1965).
[7] W. G. Nilsen, R. L. Comstock, and L. R. Walker, Phys. Rev. **139**, A472 (1965).
[8] R. L. Comstock and W. G. Nilsen, Phys. Rev. **136**, A442 (1964).
[9] R. L. Comstock, J. J. Raymond, W. G. Nilsen, and J. P. Remeika, Appl. Phys. Letters **9**, 274 (1966).
[10] M. Sparks, *Ferromagnetic Relaxation Theory* (McGraw-Hill Book Company, Inc., New York, 1964).
[11] E. G. Spencer and J. P. Remeika, Proceedings of International Conference on Nonlinear Magnetism, Washington (1964) p. 12-1-1 (unpublished).
[12] F. R. Morgenthaler, J. Appl. Phys. **31**, 955 (1960), and doctoral dissertation, Massachusetts Institute of Technology, 1959, (unpublished); E. Schlömann, J. J. Green, and A. Milano, J. Appl. Phys. Suppl. **31**, 386S (1960).

reported as a line width ΔH_k defined as $1/\gamma T$, where $1/T$ is the *energy* relaxation frequency (related[13] to the magnon, or magnetization, relaxation frequency $1/\tau$ by $1/T=2/\tau$).

From Eqs. (5.41) and (5.34) of Ref. 10, the line width for three-magnon confluence process is

$$\Delta H_k = \frac{4\pi M\mu^2}{2\hbar}\left[\exp\left(\frac{\hbar\omega_1}{k_BT}\right)-1\right]F_1\int dk_2k_2^2\int_{-1}^{1}du\,\alpha$$
$$\times\exp\left(\frac{\hbar\omega_2}{k_BT}\right)n_2n_{1+2}\delta(\hbar\omega_{k_1+k_2}-\hbar\omega_{k_2}-\hbar\omega_1) \quad (1)$$

with

$$\alpha\equiv\frac{8}{2\pi}\int_0^{2\pi}d\phi_2\left|\frac{k_{2z}(k_{2x}-ik_{2y})}{k_2^2}\right|^2,$$

and

$$F_1\equiv\left[1+\left(\frac{\omega_m}{2\omega_1}\right)^2\right]^{1/2}-\frac{\omega_m}{6\omega_1},$$

where the n's are Bose-Einstein occupation factors, u is the cosine of the angle between $\mathbf{k}_2$ and $\mathbf{H}_{\text{app}}$, $\omega_m\equiv4\pi\gamma M$, and the subscript 1 refers to the relaxing magnon, 2 to the other incident magnon, and $3\equiv2+1$ to the output magnon. The correction factor[14] F_1, which does not appear in (5.41) of Ref. 10, arises from making the last Holstein–Primakoff transformation on the magnon creation and annihilation operations in the Hamiltonion.[15] This correction factor makes no more than a 5% correction when the approximations made in evaluating the integral are valid.

The approximations which will be made in evaluating the integral in (1) and in simplifying the results are listed for the convenience of the reader: (A) In the delta function the approximate dispersion relation

$$\hbar\omega=Dk^2+\hbar\omega_T-\tfrac{1}{2}\hbar\omega_m\cos^2\theta_k \quad (2)$$

is used for the 2 and 3 magnons, where ω_T defines the top of the magnon manifold at $k=0$. This is a good approximation when $Dk^2+\hbar\omega_T\gtrsim\hbar\omega_m\cos^2\theta_k$, as discussed in detail in Sec. 3.3 of Ref. 10. We shall see below that when the small k_1 approximation (B) $4Dk_1^2\ll\hbar\omega_1$ is satisfied, energy and momentum conservation make Dk_2^2 and Dk_3^2 large; consequently (A) is a good approximation when (B) is satisfied. When (B) is not satisfied, (A) is still a good approximation in the high-field limit: (C) $\omega_T\gtrsim\frac{3}{2}\hbar\omega_m$.

For example, for the worst case of $k_2=0$ and $\theta_{k_2}=\pi/2$ the approximate and exact dispersion relations differ by only 6% when $\omega_T=\frac{3}{2}\omega_m$. **Experiments should be carried out at the highest possible fields (i.e., highest frequencies), especially when (B) is not satisfied.**

[13] See p. 2 of Ref. 7.
[14] E. Schlömann, Phys. Rev. **121**, 1312 (1961).
[15] See Eq. (3.50) of Ref. 7.

(D) In the occupation number factors n in (1), the angle average of (2) is used, that is

$$\hbar\omega_2=Dk_2^2+\hbar\omega_{\text{av}}$$

with

$$\hbar\omega_{\text{av}}\equiv\hbar\omega_T-\tfrac{1}{6}\hbar\omega_m.$$

When (B) is satisfied, Dk_2^2 is large so that $\hbar\omega_{\text{av}}$ is a small correction term. But when Dk_1^2 is comparable to or larger than $\hbar\omega_T$, the error may become as large as $\sim\hbar\omega_m/3\hbar\omega_T$.

(E) In the delta function in (1), the approximation

$$2k_1k_2u_{12}\ll k_1^2+k_2^2$$

is made, where u_{12} is defined as the cosine of the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$. This is well satisfied for either $k_1\ll k_2$ or $k_1\gg k_2$. Even when $k_1\cong k_2$, the factor k_{2z}^2 in (3) below weights the small values of u_{12} since $\mathbf{k}_1$ is perpendicular to the internal field; thus this approximation is always rather well satisfied. Furthermore, this approximation effects only the value of a small correction term.

(F) The factor k_{2z}^2/k_2^2 is replaced by $\frac{1}{3}$ in a small correction term in the delta function.

(G) $\frac{1}{6}(\omega_m/\omega_T)[k_1^2/(k_1^2+k_{2mn}^2)]\ll1,$

and

$$\tfrac{1}{6}[\hbar\omega_mk_{2mn}^2/D(k_1^2+k_{2m}^2)^2]\ll1,$$

where k_{2mn} is defined in (7). Both of these inequalities are well satisfied if either (B) or (C) is satisfied.

(H) We set $\alpha=1$ in Eq. (1). This value of α is chosen to give the exact result in the high-T–low-k_1 limit, as discussed on p. 98 of Ref. 10. Consequently, in the region in which ΔH_k is linear in k_1, this approximation gives no error. Notice that this value 1 is very close to the value 1.067 obtained by averaging α over solid angles.

(I) k_2 and $k_3\ll k_{\text{BZ}}$, where k_{BZ} defines the edge of the Brillouin zone. More specifically, it is assumed that (2) holds for the 2 and 3 magnons even at large values of k_2 and k_3. This approximation is well satisfied everywhere except for extremely small values of k_1; then, according to (7), Dk_{2mn}^2 becomes large. At high temperatures this will make ΔH_k become small at a value of k_1 larger than it would if (I) were satisfied. For example, for YIG with $\hbar\omega_1=1690$ Oe (pump frequency$=9.53$ Gc/sec), $Dk_{2mn}^2=100$°K for the small value of $k_1=1.5\times10^4$ cm^{-1}.

(J) $k_BT\gg\hbar\omega_1,\quad k_BT\gg\hbar\omega_{2mn}.$

(K) $k_BT\gtrsim\frac{1}{2}\hbar\omega_{2mn}\cong k_BT_0.$

Consider now the delta function in (1). For a $\pi/2$ magnon $\mathbf{k}_1\cdot\hat{z}=0$, where $\hat{z}$ is the unit vector in the direction of the internal field. Using this result and approximation (E) we find

$$\cos^2\theta_{k_1+k_2}=[(\mathbf{k}_1+\mathbf{k}_2)\cdot\hat{z}/|\,\mathbf{k}_1+\mathbf{k}_2\,|]^2$$
$$=k_{2z}^2/(k_1^2+k_2^2+2k_1k_2u_{12})$$
$$\cong k_{2z}^2/(k_1^2+k_2^2)-[2k_1k_2k_{2z}^2u_{12}/(k_1^2+k_2^2)^2], \qquad (3)$$

where u_{12} is the cosine of the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$. With this equation and $\cos^2\theta_{k_2}=k_{2z}^2k_2^{-2}$, we find

$$\cos^2\theta_{k_2}-\cos^2\theta_{k_2+k_2}=\frac{k_{2z}^2}{k_2^2}\left(\frac{k_1^2}{k_1^2+k_2^2}+\frac{2k_1k_2^3u_{12}}{(k_1^2+k_2^2)^2}\right).$$

In this result we make the approximation (F) of replacing $(k_{2z}/k_2)^2$ by its solid-angle average value $\frac{1}{3}$, which gives

$$\delta(\hbar\omega_{k_1+k_2}-\hbar\omega_{k_2}-\hbar\omega_1)$$
$$=\delta\left\{\left[2Dk_1k_2+\tfrac{1}{6}\hbar\omega_m\frac{2Dk_1k_2^3}{D(k_1^2+k_2^2)^2}\right]u_{12}-\left[\hbar\omega_1-Dk_1^2-\tfrac{1}{6}\hbar\omega_m\frac{k_1^2}{k_1^2+k_2^2}\right]\right\}. \qquad (4)$$

The argument of the delta function has a zero at $u_{12}=u_0$, where

$$u_0\equiv\frac{\hbar\omega_T-\frac{1}{6}\hbar\omega_m k_1^2/(k_1^2+k_2^2)}{2Dk_1k_2+\frac{1}{6}\hbar\omega_m 2k_1k_2^3/(k_1^2+k_2^2)^2},$$

with $\hbar\omega_T\equiv\hbar\omega_1-Dk_1^2$. From this expression and $u_0<1$, we find

$$Dk_2^2>\frac{(\hbar\omega_T)^2}{4Dk_1^2}\left(1-\frac{1}{6}\frac{\omega_m}{\omega_T}\frac{k_1^2}{k_1^2+k_2^2}\right)^2\times\left(1+\frac{1}{6}\frac{\hbar\omega_m k_2^2}{D(k_1^2+k_2^2)^2}\right)^{-2}. \qquad (5)$$

Making approximation (G) that the two $\frac{1}{6}$ terms are small, i.e.,

$$Dk_{2mn}^2\cong(\hbar\omega_T)^2/4Dk_1^2, \qquad (6)$$

and iterating (5) once gives

$$Dk_2^2>\frac{\hbar\omega_T}{2\epsilon}\left[1-\frac{1}{3}\frac{\omega_m}{\omega_T}\frac{\epsilon^2(1+\epsilon^2)+2\epsilon}{(1+\epsilon^2)^2}\right]\equiv Dk_{2mn}^2, \qquad (7)$$

where

$$\epsilon\equiv 2Dk_1^2/\hbar\omega_T. \qquad (8)$$

Equation (4) may now be written as

$$\delta(\hbar\omega_{k_1+k_2}-\hbar\omega_{k_2}-\hbar\omega_1)$$
$$=\left[2Dk_1k_2\left(1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right)\right]^{-1}\delta(u_{12}-u_0). \qquad (9)$$

We now make the approximation (H) of replacing the angle factor α in (1) by 1. In evaluating the integral over $d\mathbf{k}_2$ the polar axis can be chosen along $\mathbf{k}_1$; thus $u_{12}=u\equiv\cos\theta_{k_2}$.

The u integral in (1) can now be evaluated as

$$\int_{-1}^{1}du\,\delta(u-u_0)=1 \qquad \text{for } u_0<1$$
$$=0 \qquad \text{for } u_0>0.$$

Using this expression and defining $z\equiv\hbar\omega_2/k_BT$ and $\hbar\omega_{2mn}\equiv Dk_{2mn}^2+\hbar\omega_i$, where $\hbar\omega_i$ defines the bottom of the magnon manifold, i.e.,

$$\hbar\omega_i=[(\hbar\omega_T)^2+\tfrac{1}{2}(\hbar\omega_m)^2]^{1/2}-\tfrac{1}{2}(\hbar\omega_m), \qquad (10)$$

reduces (1) to

$$\Delta H_k=4\pi M\mu^2k_BT\left\{8D^2k_1\left[1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right]\right\}^{-1}\times\left[\exp\left(\frac{\hbar\omega_1}{k_BT}\right)-1\right]$$
$$\times\int_{\hbar\omega_{2mn}/k_BT}^{\infty}dz\frac{\exp z}{(\exp z-1)[\exp(z+\hbar\omega_1/k_BT)-1]}. \qquad (11)$$

In writing the upper limit of the integral as ∞, we have made approximation (I) that the k_2^2 dispersion relation (2) is valid for all k_2 magnons.

The integral in (11) can be evaluated exactly by transforming to the variable $p\equiv[\exp(z)-1]$; the result is

$$\Delta H_k=4\pi M\mu^2k_BT\left\{8D^2k_1\left[1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right]\right\}^{-1}\times\ln\left[1+\frac{1-\exp(-\hbar\omega_1/k_BT)}{\exp(\hbar\omega_{2mn}/k_BT)-1}\right]. \qquad (12)$$

3. HIGH- AND LOW-TEMPERATURE LIMITS AND SMALL WAVE-VECTOR LIMIT

The behavior of ΔH_k as a function of T and as a function of k_1 can be seen by simplifying (12) in three limiting cases. In the high-temperature limit (J) of $\hbar\omega_1$ and $\hbar\omega_{2mn}$ both much less than k_BT, expanding the exponentials in (12) gives

$$(\Delta H_k)_{hiT}=4\pi M\mu^2k_BT\left\{8D^2k_1\left[1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right]\right\}^{-1}\times\ln\left\{1+\frac{\omega_1}{\omega_{2mn}}\left[1-\frac{\omega_1+\omega_{2mn}}{2k_BT}\right]\right\}. \qquad (13)$$

If the small k_1 approximation (B) of $4Dk_1^2\ll\hbar\omega_1$, i.e., $2\epsilon\ll1$, is also satisfied, the logarithm in (13) can be expanded to give

$$(\Delta H_k)_{\mathrm{high}T,\mathrm{low}k_1} = 4\pi M\mu^2 k_B k_1 \left\{2D\hbar\omega_1\left[1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right]\right\}^{-1} \times[1+\theta(\epsilon^2)](T-T_0), \quad (14)$$

where $k_BT_0 \equiv \hbar^2\omega_1^2/8Dk_1^2$, and the term $[1+\theta(\epsilon^2)]$ in the numerator indicates that the result is correct to first order in $\epsilon \cong 2Dk_1^2/\hbar\omega_1$. In the limiting case of ϵ small enough so that two terms in square brackets in (14) are negligible, but ϵ not too small (otherwise $T \gg T_0$ is not satisfied), (14) reduces to the previous result[4,10]

$$(\Delta H_k)_{SLK} = 4\pi M\mu^2 k_B T k_1/2D\hbar\omega_1. \quad (15)$$

When the small k_1 approximation B is satisfied, it is seen from (5) and $\omega_T \cong \omega_1$ that $Dk_{2mn}{}^2 \cong \hbar\omega_{2mn}$ is much

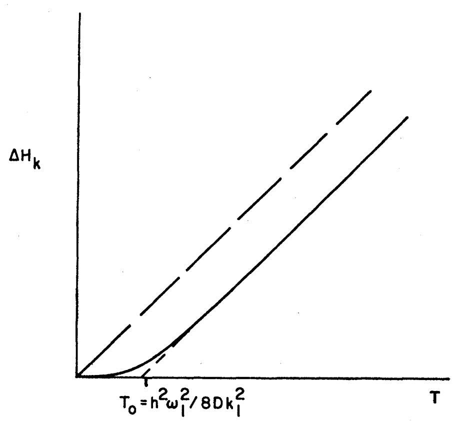

FIG. 1. Temperature dependence of ΔH_k for the case of $4Dk_1^2 \ll \hbar\omega_1$.

greater than $\hbar\omega_1$. In this case the fraction in the argument of the logarithm in (13) is $\ll 1$; so the zeroth-order expansion $\ln(1+\delta) \cong \delta$ is valid and (13) becomes

$$(\Delta H_k)_{\mathrm{lo}k_1} = 4\pi M\mu^2\hbar\omega_1 \left\{8D^2k_1\left[1+\frac{1}{12}\frac{\omega_m}{\omega_T}\frac{4\epsilon}{(1+\epsilon^2)^2}\right]\right\}^{-1} \times\left[\frac{1-\exp(-\hbar\omega_1/k_BT)}{\hbar\omega_1/k_BT}\right]\frac{1}{\exp(\hbar\omega_{2mn}/k_BT)-1}. \quad (16)$$

In the low-temperature limit (K) of $k_BT \lesssim \frac{1}{2}\hbar\omega_{2mn} \cong k_BT_0$, ΔH_k is exponentially small.

The temperature dependence of ΔH_k is illustrated in Fig. 1 for the case in which $4Dk_1^2 \ll \hbar\omega_1$ is satisfied. The region $T < T_0$ of exponentially small ΔH_k extends to larger temperatures (i.e., T_0 increases) as k_1 is decreased. For YIG with $k_1 = 10^5$ cm^{-1} and a pump frequency of 11.4 Gc/sec, i.e., a magnon frequency of 5.7 Gc/sec corresponding to $\hbar\omega_1 = 2035$ Oe, $T_0 = 1.5$°K, and for $k_1 = 0.25 \times 10^5$ cm^{-1}, $T_0 = 25$°K.

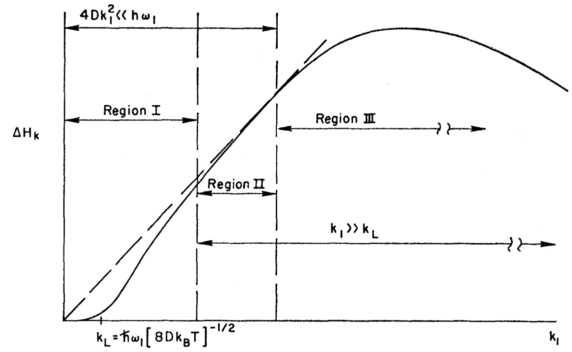

FIG. 2. Wave-vector dependence of ΔH_k. For $k_1 \gtrless 8k_L$ the curve is a plot of

$(2k_1/k_L)[\exp(2k_L^2/k_1^2)-1]^{-1} \cong (2k_1/k_L)[\exp(\hbar\omega_{2mn}/k_BT)-1]^{-1}$.

The k_1 dependence of ΔH_k is illustrated in Fig. 2. Region II of this figure is the region of ΔH_k linear in k_1. In the small k_1 region I, ΔH_k drops below the linear extrapolation, and ΔH_k becomes exponentially small at small k_1. In the large k_1 region III, ΔH_k drops below the linear extrapolation and for very large k_1, $\Delta H_k \sim \ln(k_1)/k_1$. The wave vector k_L above which ΔH_k becomes linear in k_1 is a function of temperature, and for sufficiently low temperatures there is no linear region. The temperature at which the linear region disappears is given roughly by $Dk_L^2 \equiv \hbar^2\omega_1^2/8k_BT \cong \frac{1}{8}\hbar\omega_1$, i.e., $k_BT \cong \hbar\omega_1$. Since $k_BT \gg \hbar\omega_1$ is usually well satisfied, there is usually a region of linear dependence of ΔH_k on k_1.

4. COMPARISON WITH EXPERIMENTS

LeCraw and Spencer[5] measured ΔH_k at room temperature and a pump frequency of 11.4 Gc/sec (magnon frequency of 5.7 Gc/sec) for values of k_1 between 0.4×10^5 and 2.1×10^5 cm^{-1}. Their experimental results (values from their Fig. 2 minus $\Delta H_{k\to 0}$ with k's increased by 5% to account for a later value[7] of D) are shown in Fig. 3. The predicted linear region

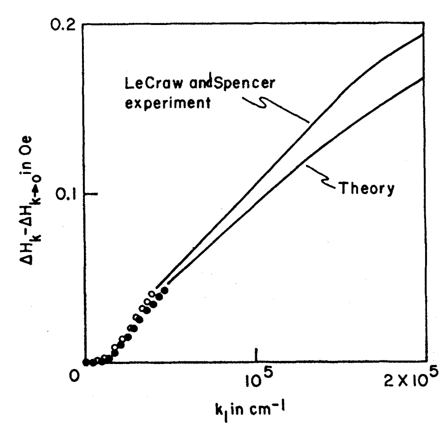

FIG. 3. Comparison of the results of LeCraw and Spencer (Ref. 5) with the theory.

of ΔH_k versus k_1 and the bending down at high values of k_1 are evident in this figure. In the region below 0.4×10^5 cm^{-1}, the predicted flattening was also observed,[16] as illustrated schematically by the open circles in the figure. Since the agreement between these experimental results and the theory is within a few percent, it is now feasible to make two additional small corrections to $(\Delta H_k)_{\text{high } T, \text{low } k_1}$ of (14). The bracketed term inside the braces in Eq. (13) is quite negligible since the high-temperature approximation (*K*) is well satisfied. Expanding the logarithm and using $\hbar\omega_{2mn}\equiv Dk_{2mn}^2+\hbar\omega_i$ [from above (10)] gives

$$\ln\left(1+\frac{\omega_1}{\omega_{2mn}}\right)\cong\frac{\hbar\omega_1}{Dk_{2m}^2}\left[1-\tfrac{1}{2}\,\frac{\hbar\omega_1+2\hbar\omega_i}{Dk_{2m}^2}\right]. \quad (17)$$

A simple calculation in which the angle approximation (*G*) is not made [Eq. (B.11) of Ref. 7 is used for α] shows that the numerical factor in the first correction factor in (16) should be $5/12=0.834$ $(\frac{1}{2})$, rather than $\frac{1}{2}$ as in (17). Thus the ln term in (13) is replaced by

$$\ln\left(1+\frac{\omega_1}{\omega_{2mn}}\right)\rightarrow\frac{4Dk_1^2}{\hbar\omega_1}F_2 \quad (18)$$

with

$$F_2\equiv1-0.834\left[1-\frac{\ln(1+\hbar\omega_1/\hbar\omega_{2mn})}{4Dk_1^2/\hbar\omega_1}\right], \quad (19)$$

which is correct to first order as ΔH_k first drops below linearity and retains the character of the logarithm. This expression (19) is not correct when ω_1/ω_{2mn} is large.

If we define

$$F_3\equiv\left[1+\frac{1}{12}\,\frac{\omega_m 4\epsilon}{\omega_T(1+\epsilon^2)^2}\right]^{-1} \quad (20)$$

then $(\Delta H_k)_{\text{high } T}$ is given by the expression

$$(\Delta H_k)_{\text{high } T}=(4\pi M\mu^2 k_B T k_1/2D\hbar\omega_1)F_1F_2F_3, \quad (21)$$

which is expected to be correct to within a few percent provided that the correction terms F_1, F_2, and F_3 do not differ greatly from 1.

The theoretical result (21) is shown in Fig. 3. The slope of the experimental curve in its linear region is 7% higher than the slope of the theoretical result (21) in its linear region (with $F_1=F_3=1$). At the maximum measured k_1, the theoretical curve is down by 12.5% from the linear curve, while the experimental curve is down by 10% from its linear extrapolation. The departure from linearity is a little more abrupt for the experimental curve than for the theoretical curve. The small k_1 portion of the theoretical curve is shown dotted because the flattening at the smallest values of $k_1\cong 0.15\times10^5$ cm^{-1} was exaggerated slightly since the Brillouin-zone approximation (I) is not well satisfied there.

[16] E. G. Spencer (private communication).

In Fig. 3, the following values were used: $D=4.5\times10^{-9}$ Oe cm$^2=0.834\times10^{-28}$ erg cm^2 from Nilsen, Comstock, and Walker[7]; $4\pi M=(0.726)(2470)=1792$ *G* from Geller *et al*[17]; and

$$\frac{4\pi M\mu^2 k_B T k_1}{2D\hbar\omega_1}=0.0579\,\frac{M(T)}{M(300)}\,\frac{T}{300}\,\frac{k_1}{10^5}\,\frac{2\pi\times10\text{ Gc/sec}}{\omega_1}\text{ Oe}. \quad (22)$$

Comstock[6] measured ΔH_k in YIG at 300°K and $\omega_1=4.76$ Gc/sec and saw the bending down at high k_1 and flattening at low k_1. But the value of $\Delta H_k=0.257$ Oe at $k_1=2\times10^5$ cm^{-1} is 22% higher than the theoretical value of 0.201 Oe. The agreement at lower temperatures was even worse. For example, at 100°K and $k_1=2\times10^5$ cm^{-1}, the experimental value of 0.170 Oe is 47% higher than the theoretical value of 0.090 Oe. Both Comstock[6] and Nilsen, Comstock, and Walker[7] found even greater disagreement with the theoretical results at lower temperature. For example, at 4.2°K and 8.2×10^5 cm^{-1} Nilsen and coworkers found $\Delta H_k=0.0232$ Oe. The value of $(\Delta H_k)_{SLK}$ in (15) is 5.3 mOe and the correct ΔH_k from (12) is considerably smaller; so the experimental result is well over five times larger than the theoretical value. Comstock[6] speculated that impurities gave rise to an additional contribution to ΔH_k. It would be interesting to repeat these experiments on ultrapure samples.[11]

Comstock and Nilsen[8] measured ΔH_k for lithium ferrite at room temperature for $\omega_p=34.67$ Gc/sec and $Dk_1^2=|H_c-H|=0$ to $50^2=2500$ Oe. It appears that there is another mechanism giving rise to ΔH_k in their sample because at $|H_c-H|^{1/2}=49$, (13) gives $\Delta H_k=0.08$ Oe, which is an order of magnitude smaller than the measured value of 0.5 Oe. Also, the theory indicates that ΔH_k should not be a linear function of k_1 for

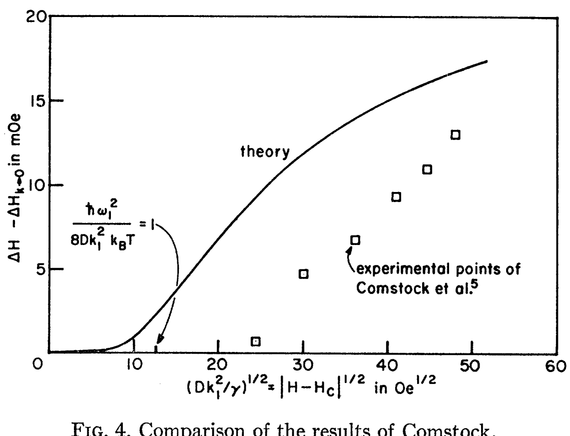

FIG. 4. Comparison of the results of Comstock, *et al.* (Ref. 9) with the theory.

[17] S. Geller, H. J. Williams, R. C. Sherwood, J. P. Remeika, and G. P. Espinosa, Phys. Rev. **131**, 1080 (1963).

$30 < |H - H_c|^{1/2} < 50$ Oe$^{1/2}$, but the experimental results were linear.

Comstock *et al.*[9] measured ΔH_k at 4.2°K as a function of k_1 for 6×10^5 cm$^{-1} < k_1 < 12\times10^5$ cm^{-1}. Figure 4 shows their experimental points and a plot of the theoretical result (12). Here the theoretical values are *greater than* the experimental ones. At least part of the discrepancy arises from making the angle approximation (H) of $\alpha=1$. It is easy to show that at very low temperatures retaining the angle factor α in (1) reduces ΔH_k well below the value obtained by setting $\alpha=1$.

5. SUMMARY

The three-magnon confluence relaxation frequency has been calculated without making assumptions which were previously made by Sparks, Loudon, and Kittel. The results are that the discrepancies between experiments and the previous theory of Sparks, Loudon, and Kittel are removed. In particular the exponentially small (rather than linear) dependence of $1/\tau$ on k_1 and T is explained by the new theory.

The most general result of the paper is the expression (12) for ΔH_k, which is valid when the approximate dispersion relation (2) is well satisfied and when Brillouin zone effects are negligible as in assumption (I). The result (12) can be simplified in several limiting cases such as high-temperature T and small wave vector k_1, as in (13), (14), (15), and (16). In particular, the Sparks, Loudon, and Kittel result is regained in the high-temperature, intermediate-k_1 limit.

ACKNOWLEDGMENTS

The author is grateful to R. L. Comstock, S. Geller, C. LeCraw, W. G. Nilsen, and E. G. Spencer for stimulating conversations. T. Wolfram made helpful suggestions on the manuscript.

PHYSICAL REVIEW VOLUME 160, NUMBER 2 10 AUGUST 1967

Volume Effect for Magnetic Transition Temperatures of Rare-Earth–Scandium Alloys*

E. O. Wollan
Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received 23 February 1967)

The alloys of the heavy rare earths with scandium have appreciably lower magnetic ordering temperatures than the metals themselves and their alloys with yttrium. The difference between these systems appears to be associated with the effect on the magnetic coupling of the smaller atomic volume of scandium. The Curie temperatures T_C of the Gd–Sc and the Gd–Y alloys are compared over the composition range for which they are ferromagnetic. The coupling energy per gadolinium atom T_C/x in the Gd–Y system is found to be essentially independent of composition whereas there is a linear decrease in this quantity for the Gd–Sc system. The rate of decrease of T_C/x with volume for the alloy system is compared with the corresponding decrease of T_C for pure gadolinium as determined in measurements at high pressures, and the results are found to be in good accord. The Néel-temperature data for the Tb–Sc system are also discussed.

The initial magnetic ordering properties of the heavy rare-earth metals and their alloys among themselves and with yttrium have been shown to be quite well represented by common curves in which the Néel temperature T_N, the Curie temperature T_C, and the initial turn angle ω_i are plotted against the spin variable $x=n(g-1)^2J(J+1)$. On the other hand, alloys with scandium and lanthanum which have the same type of outer electron configuration as yttrium do not show a simple correlation with the rare-earth metals or the yttrium alloys.

In attempting to resolve some of the characteristics of these systems on the basis of molecular-field considerations it became apparent that the crystal parameters associated with these systems have a pronounced effect on the magnetic coupling[1] which would not be anticipated on the basis of a simple free-electron Ruderman–Kittel–Kasuya–Yosida (RKKY) type of interaction.

It is interesting in this respect then to consider the rare-earth alloys with scandium because it has a much smaller atomic volume than yttrium and the rare earths. The effect of volume changes on the transition temperatures for the alloys can then also be compared with the corresponding changes brought about in the pure metals by the application of high pressures.

The simplest systems to consider in this respect would appear to be pure Gd and the Gd–Y [2] and the Gd–Sc [3] alloys. Since Gd is ferromagnetic, the changes in T_C should be a direct measure of the changes in the sum of the coupling constants without the effects of turn angle

* Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.
[1] E. O. Wollan, J. Appl. Phys. **38**, 1371 (1967).
[2] W. C. Thoburn, S. Legvold, and F. H. Spedding, Phys. Rev. **110**, 1298 (1958).
[3] H. E. Nigh, S. Legvold, F. H. Spedding, and B. J. Beaudry, J. Chem. Phys. **41**, 3799 (1964).