TABLE I. Spin-orbit coupling parameters and energy separations of pure spin states.

	$\frac{1}{2}\zeta$ (ev)	λ	$2G_I$ (ev)	$2G_e$ (ev)
In^+	0.15	0.88	2.3	0.72
Tl^+	0.51	0.87	2.1	−0.21

TABLE II. Calculated oscillator strength ratios, without and with crystalline interactions, and observed ratios.

	$R_{\text{free ion}}$	R_{crystal}	R_{observed}
In^+	259	30	25
Tl^+	28	2	5

In Table II are shown the experimental and calculated oscillator strength ratios. The calculated values are tabulated both neglecting and including crystalline interaction. The improved agreement, due to inclusion of the crystalline interactions, in the calculated ratios with experiment is evident from Table II. The close agreement of the calculated and experimental results for KCl:In is obviously fortuitous. The reduction in oscillator strength ratios of the crystal states compared to the free-ion states arises from the decrease in energy separation of the unperturbed $^3P^0$ and $^1P^0$ states by crystalline interactions. The higher oscillator strength ratios calculated and observed for In^+ obviously arise from the smaller spin-orbit coupling in this ion.

CONCLUSIONS

Crystalline interactions have been shown to have a marked influence on the oscillator strength ratios for transitions from the ^1S to the "$^1P_1{}^0$" and "$^3P_1{}^0$" crystal states of KCl:Tl and KCl:In. The inclusion of these interactions by utilizing experimental transition energies, before applying spin-orbit interaction as a perturbation, permits the successful calculation of the oscillator strengths for these materials. This analysis supports the identification of these transitions but also indicates that the free-ion notation is somewhat less appropriate since the pure spin states are more strongly mixed in the crystal than in the free ions.

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Narrowing Effect of Dipole Forces on Inhomogeneously Broadened Lines

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Under the proper circumstances dipole forces can decrease an inhomogeneous broadening of a magnetic resonance line. The necessary condition is that the magnetization of the material be very much greater than the inhomogeneity, and is therefore most easily observed in ferromagnetic materials. Experiments are described in which this narrowing effect was observed in manganese ferrite and yttrium iron garnet. The theory of this effect is discussed for the two cases in which the spatial period of the inhomogeneity is large compared to atomic distances and either (I) short compared to sample size or (II) comparable to sample size. Applications to line broadening in paramagnets and polycrystalline ferrites, and to the low-temperature peak in single crystals of yttrium iron garnet and manganese ferrites, are discussed.

I. INTRODUCTION

IN electron and nuclear paramagnetic resonance, the effect of dc field inhomogeneities on the resonance line width is frequently met with in practice and is quite familiar. One observes a broadened line whose width is roughly equal to the absolute value of the field inhomogeneity, since at constant frequency, different parts of the sample come into resonance at different values of the mean external dc magnetic field.

We wish to point out in this paper that under certain circumstances the magnetic dipole forces can decrease inhomogeneous broadening of the resonance line. In effect, even though different parts of the sample see different dc magnetic fields, the long-range dipole forces can couple different regions together so that they assume a common resonant frequency. The necessary condition for this to happen is that the magnetization of the sample be considerably greater than the field inhomogeneity. This condition is virtually impossible to satisfy in nuclear resonance, can be satisfied in particular cases in electron paramagnetic resonance, but almost invariably holds in ferromagnetic resonance. The experimental observation of this narrowing effect is therefore easiest in ferromagnetic materials and experiments demonstrating the effect are described in Sec. III.

We shall consider, in this paper, field inhomogeneities of two sorts. First, there is case I in which the field fluctuates from point to point with a spatial period large compared to the distance between spins but short compared to the dimensions of the sample. A fine-grained polycrystalline ferrite is an example of this case since the direction of the magnetic anisotropy will vary in passing from one crystallite to the next. The second

case II, arises when the spatial period of the field inhomogeneity is of the order of the sample size. This case corresponds to the familiar case of inhomogeneity of the external field, and has also been the easiest to realize experimentally by distortion of the ferromagnetic samples from ellipsoidal shape. The theory presented in Sec. II will discuss the two cases in order.

An earlier presentation of line broadening in ferromagnetic resonance treated the effect of field inhomogeneities with a spatial period comparable to the distance between spins, arising from the disorder of the magnetic lattice. In this case, the exchange field is the effective agent in narrowing the resonance line. In cases I and II above, the spatial period of the fluctuation of the field is so large that the torques produced by the exchange field are negligible. The exchange coupling cannot tie together the motion of distant regions of the sample, whereas the long-range dipole forces can.

II. THEORY

The theory of the dipole narrowing of inhomogeneously broadened resonance lines in ferromagnetic salts can be developed in a way very similar to the theory of ferromagnetic resonance line width given by Clogston, Suhl, Walker, and Anderson.[1] According to this reference the mode spectrum of a ferromagnetic ellipsoid placed in a uniform magnetic field is given by

$$\omega_\kappa=\gamma[(H-N_zM+H_ea^2\kappa^2)\times(H-N_zM+H_ea^2\kappa^2+4\pi M\sin^2\theta)]^{\frac{1}{2}}, \quad (1)$$

where ω_κ is the frequency of a spin wave of wave number κ, a is the distance between spins, γ equals $-g\beta/\hbar$, H is the applied external field, N_z is the longitudinal demagnetizing factor, M is the magnetization of the sample, H_e is an exchange field, and $\cos\theta_\kappa=\kappa_z/\kappa$. The sample is supposed to be symmetric about the axis. The frequency of the uniform-precession, or infinite-wavelength mode $\kappa=0$, is given by the Kittel relation,

$$\omega_0=\gamma[H-(N_z-N_t)M], \quad (2)$$

where N_t is the transverse demagnetizing factor.

The spectrum given by Eqs. (1) and (2) is shown in Fig. 1. The frequencies ω_2 and ω_1 are given by

$$\omega_2=\gamma[(H-N_zM)(H-N_zM+4\pi M)]^{\frac{1}{2}}, \quad (3)$$

$$\omega_1=\gamma(H-N_zM). \quad (4)$$

The important features of this spectrum, including the degeneracy of ω_0 with a manifold of spin-wave modes, have been discussed in reference 1. In Fig. 1, the range of κ values has been divided into three regions by lines drawn at points κ_1 and κ_2. The line at κ_2 marks the point at which exchange first becomes important and is given roughly by $\kappa_2=(1/a)(4\pi M/H_e)^{\frac{1}{2}}\sim10^6$ cm^{-1}. Below the line at κ_1, the spin wavelength becomes comparable to the dimensions of the specimen and spin waves are no longer a good representation of the modes of oscillation.

As derived in reference 1, Eq. (1) applies strictly only to magnetic crystals whose magnetization M is close to the saturation value M_0. Actually the dispersion relation (1) can be derived classically on the basis of the Landau-Lifshitz, or Bloch equations. If restricted to a range of κ values for which $\kappa<(1/a)(4\pi M/H_e)^{\frac{1}{2}}$ so that the term in H_e may be neglected, Eq. (1) applies to any insulating crystal with a magnetization M per unit volume, including paramagnets.

In reference 1, a perturbation was assumed imposed upon the spin system that gave a connection between the spin wave $\kappa=0$ and a manifold of degenerate spin waves of higher κ value. The specialized assumptions made about this perturbation rendered it independent of the magnitude of κ. The line width resulting from this perturbation is given in Eq. (29) of reference 1 as

$$\Delta H=\frac{3}{20}\left(\frac{H_P^2(4\pi M)^{\frac{1}{2}}}{H_e^{\frac{3}{2}}}\right)I\left(n_t,\frac{H}{4\pi M}\right), \quad (5)$$

where I is a shape factor, and H_p^2 is the mean square value of the perturbation through the specimen expressed in oersteds.

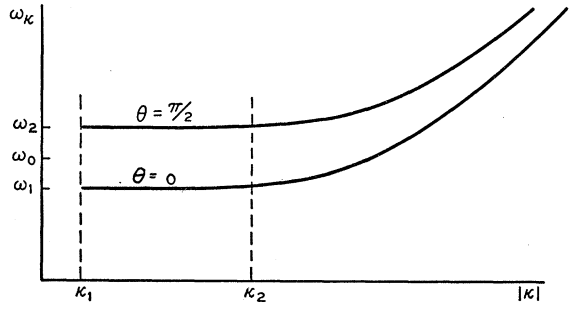

FIG. 1. The mode spectrum of a magnetized ferromagnetic ellipsoid of finite extent. The high κ values correspond to the region where exchange is important. The region below κ_2, comprises the modes for which the exchange is unimportant and whose frequencies are determined by dipole forces alone.

[1] Clogston, Suhl, Walker, and Anderson, J. Phys. Chem. Solids 1, 129 (1956).

Case I

We now wish to examine the result of confining the perturbation to the range of κ values between κ_1 and κ_2. Using methods entirely similar to reference 1, it is found that

$$\Delta H=\left(\frac{18\pi}{10}\right)\frac{H_p^2}{4\pi M}J, \quad (6)$$

where J is a shape factor given by

$$J=\frac{1}{6}\left(\frac{\omega_0}{\omega_1}\right)\left(\frac{\omega_1\omega_M}{\omega_2^2-\omega_0^2}\right)^{\frac{1}{2}}\left[1+5\left(\frac{\omega_0^2+\omega_1^2}{2\omega_0\omega_1}\right)\right], \quad (7)$$

with $\omega_M=\gamma 4\pi M$. [The quantities ω_0, ω_1 and ω_2 depend upon the shape through the demagnetizing factors appearing in Eqs. (2), (3), (4).] The exchange field no longer plays a part in this expression. The mean square perturbation H_p^2 is narrowed by dipole forces alone to give a line width of order $H_p^2/4\pi M$.

There are two points of comparison between Eqs. (5) and (6) that are of interest. Expression (5) was developed to deal with a rapid spin-to-spin variation of the dipolar or pseudodipolar forces in a ferromagnet and shows the very great narrowing brought about in such a case by the exchange field. Equation (6) shows, by way of contrast, that a more slowly varying perturbation will be narrowed only by dipole forces, and therefore to a much smaller degree.

The second point of interest lies in the shape factor found in both Eqs. (5) and (6). For the first case this is given by the quantity I shown in graphical form in reference 1. It can be seen there that I decreases very fast with transverse demagnetizing factor, approaching zero for an infinitely thin disk specimen. A similar graph giving J as a function of $n_t=N_t/4\pi$ and $H/4\pi M$ is shown in Fig. 2. We see here, again, a general tendency for the line width to be smaller the more nearly the specimen approaches the shape of a flat disk. The line width, however, no longer approaches zero for a flat disk, but approaches a constant value corresponding to $J=1$. However, the disk must not be allowed to become infinitely thin, as it will be remembered that a condition for validity of the calculation is for the perturbation to be of finer grain than the thickness of the specimen.

With increasing values of n_t the line width becomes very large approaching infinity for a value of n_t given by $\omega_0=\omega_2$. For values of $H/4\pi M$ less than unity, this condition obtains for two values of n_t. For values of n_t outside the vertical asymptotes, ω_0 is greater than ω_2, so that the uniform mode is no longer degenerate with any spin waves of $\kappa<\kappa_2$. Therefore the line width arising from a perturbation confined to κ values less than κ_2 will be zero in this case.

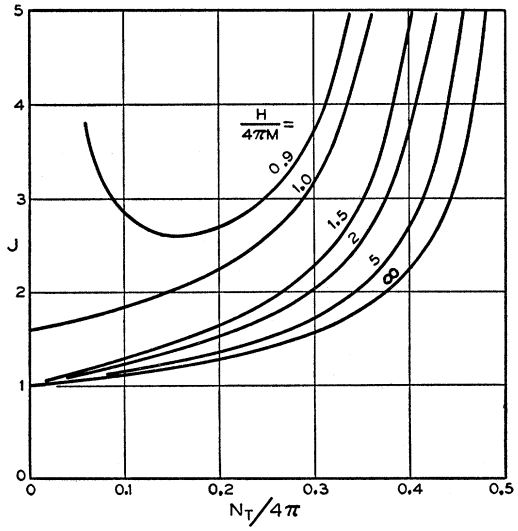

FIG. 2. Specimen shape effect for dipole narrowing of line, broadened by inhomogeneity, with spatial period between κ_1 and κ_2.

Case II

Let us now consider the effect upon the resonance line width if the system is subject to a perturbation whose characteristic length places it in the region below κ_1. In this region the oscillations of the system must be discussed in terms of a set of normal modes described by Walker[2] and Mercereau and Feynman[3] and observed by White and Solt[4] and Dillon.[5]. The frequencies of these modes are dispersed by the dipole fields into a spectrum extending between the limits ω_1 and ω_2.

We may now, because of our assumption that the spatial period of the perturbation is comparable to the sample size, confine our attention to those members of the set of modes which have only a small number of nodal surfaces within the specimen. Generally speaking, small perturbations will make a connection between the uniform mode and the set of normal modes that will lead to an additional absorption of power in regions away from the resonant frequency. Depending upon the particular case, this may take on the character of satellite absorption lines or of a non-Lorentzian broadening of the fundamental peak or both. The larger the magnetization, the greater the dispersion of these magnetostatic modes and the more these effects of the perturbation will be decreased. In effect, the line is narrowed by the dipole forces as before.

There will, in the present case be a shape effect as before. In Fig. 4 of reference 2 it will be observed that there is a pronounced crowding of the modes into the region about ω_1 for a thin disk and into the region about ω_2 for a thin rod. This effect is peculiar to the long wavelength modes as distinguished from the spin wave modes. In contrast to Case I above, the resonance line may then be expected to be broadened more in these extreme cases than in the case of a spherical specimen. If the spread of the modes becomes smaller than the size of the perturbation, both expressed in oersteds, there will be no narrowing effect at all. Thus, an infinitely thin disk or rod can be expected to exhibit a line width comparable to the magnitude of the inhomogeneity. This would also be the case for a sphere whose magnetization is so small that the spread between ω_1 and ω_2 is smaller than the perturbation, as would be normally encountered with a paramagnetic specimen.

[2] L. R. Walker, Phys. Rev. **105**, 390 (1957).
[3] J. E. Mercereau and R. P. Feynman, Phys. Rev. **104**, 63 (1956).
[4] R. L. White and D. N. Solt, Jr., Phys. Rev. **104**, 56 (1956).
[5] J. F. Dillon, Jr., Bull. Am. Phys. Soc. Ser. II, **1**, 125 (1956).

III. EXPERIMENTAL

Field inhomogeneities as large as several hundred oersteds were desired over the region of the specimen. As a typical size of the samples used in ferromagnetic resonance is 0.010 in., it would be exceedingly difficult to obtain such a field gradient with an external field. Instead, the desired field gradient was obtained from an internal field by using nonellipsoidal-shaped samples. Consequently, the spatial period of the inhomogeneity was of the order of the sample size and makes the present experiments a realization of case II as discussed above.

As an illustration, consider the hemisphere shown in Fig. 3, placed in a uniform external magnetic field. If the external field is sufficiently large so that the magnetization can be assumed to be uniform throughout the sample, then one may calculate by simple magnetostatics the demagnetizing fields at different points in the specimen. For example, at point A, $H_{\text{demag}}=-\frac{2}{3}\pi M$ while at point B, $H_{\text{demag}}=-\frac{5}{6}\pi\sqrt{2}M$. Thus, for $MnFe_2O_4$, for which $4\pi M_s$ at room temperature is approximately 4500 gauss, there is an effective inhomogeneity across the sample of the order of 570 gauss. Similarly, one finds for a hemisphere of yttrium iron garnet ($4\pi M_s=2200$) an effective inhomogeneity of the order of 280 gauss.

Flats parallel to the 110 plane were ground on the sphere and the resonance line width alternately observed with the dc field parallel to the three principal crystalline directions. The flats ranged from a slight perturbation to where nothing but a polar cap of the sphere was left. The results were that the different line widths in the three principal crystalline directions remained essentially unchanged. This is portrayed in Figs. 4 and 5 for the 100 direction in both materials. The resonance shifts to lower fields, the more the sphere is truncated, i.e., as it approaches a disk geometry. In addition, higher eigenmodes of magnetic resonance of the sample, analogous to those of ellipsoids, are excited.

The experimental results described above indicate that, although a field inhomogeneity of hundreds of oersteds exists across the specimen, distant regions of the sample do not oscillate at different frequencies but are coupled together by long-range dipole forces and oscillate in some eigenmode of magnetic resonance of the truncated sphere.

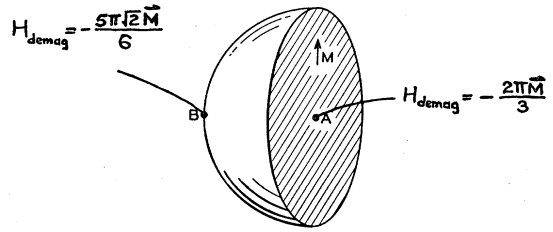

FIG. 3. Local demagnetizing fields in hemisphere with uniform magnetization.

These results can be discussed in terms of the treatment of Case II in Sec. II. In the present case the perturbing field varies so slowly through the truncated sphere that only a few modes of low symmetry are involved. The major effect of the perturbation H_p is therefore initially to lead to absorption of power at several discrete field values away from resonance, without perceptible broadening of the main line [Figs. 4(b) and 5(b)]. It is not surprising indeed that this state of affairs persists even when the specimen has been cut to a hemispherical shape. Since the relevant modes are dispersed in a range $4\pi M$ of field and may be considered to have roughly equal matrix components of H_p with the main mode of oscillation, the fractional excitation of each mode will be of order $H_p/4\pi M$. If the magnetization is several thousand oersteds, a variation of H_p over the hemisphere of several hundred oersteds will maintain $H_p/4\pi M$ in the range of small perturbations.

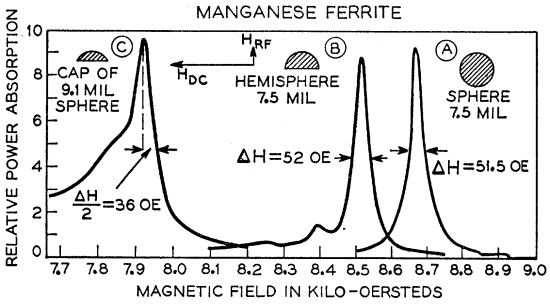

FIG. 4. Resonant absorption at 24 000 Mc/sec in manganese ferrite with the dc magnetic field in the [100] direction. As the sphere is ground down, the resonant peak shifts to lower field values and higher modes are excited. The main resonance does not broaden, however, over a large range of such perturbation.

As the magnitude of the inhomogeneity is increased (by grinding the flat still further), more of the modes close to the uniform precession are excited, resulting in a broadened line, first indications of which may be seen in the low-field side of Figs. 4(c) and 5(c). Of course, we are now passing out of the regime of small perturbations and it becomes more correct to speak directly in terms of the eigenmodes of magnetic resonance of the truncated sphere and their excitation by the driving microwave field.

An even more drastic inhomogeneity was introduced by using an irregular chip of single-crystal yttrium iron garnet whose largest dimension was 0.010 in. Many more modes are now excited resulting in a considerable broadening of the absorption spectrum, with many of these modes still distinctly resolved, however. (See Fig. 6.)

IV. OTHER APPLICATIONS OF THE THEORY

There are several other situations in which an application can be made of the theory of dipole narrowing.

(a) Polycrystalline Ferrimagnetic Materials

It has rather generally been assumed in the past that a polycrystalline specimen of some ferrimagnetic material, such as manganese ferrite or yttrium iron garnet, will show a resonance line broadened by the root-mean-square variation of the anisotropy field. It is now apparent that, if the crystallites are small compared to the size of the specimen, the line will be narrowed by dipole fields in accordance with Eq. (6). If the specimen is extremely dense so that the perturbation comes mainly from the crystalline anisotropy fields, rather than from the shape anisotropy of the crystallites, the narrowing can be very marked. Suppose that H_p is of order 50 oersteds and $4\pi M$ approximately 2000 oersteds. The line should then be narrowed by about a factor of four to about 12 oersteds.

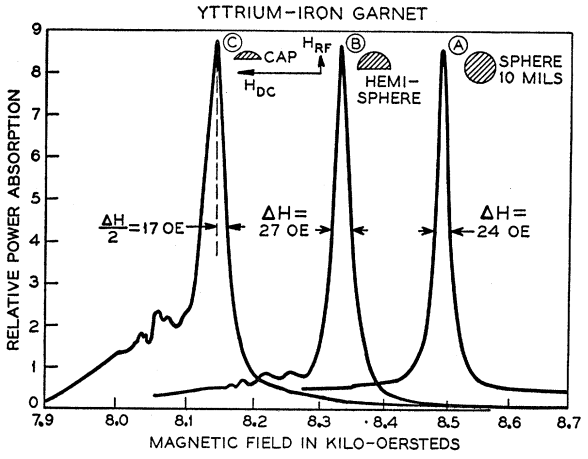

FIG. 5. Absorption in yttrium iron garnet [$Y_3Fe_2(FeO_4)_3$] with the dc field in the [100] direction.

(b) Low-Temperature Peak in Single-Crystal Ferrimagnets

The resonance line width of a single crystal of manganese ferrite or yttrium iron garnet still presents many puzzles, although the behavior may in part be accounted for by the theory of reference 1. One feature common to these two materials is a sharp rise in line width as temperature is decreased, followed by an abrupt decrease at still lower temperatures. No explanation has yet been made of this notable effect. The increase in line width is in both cases accompanied by a steep rise in crystalline anisotropy. The following explanation may be conjectured. Since the anisotropy is changing very fast with temperature, it is possible that minor differences from point to point in the crystal

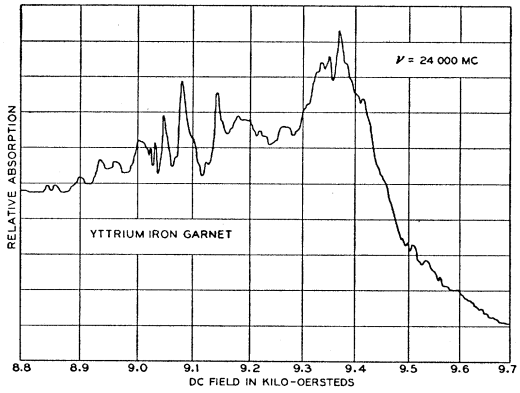

FIG. 6. Resonance in an irregular chip of yttrium iron garnet whose largest dimension is 0.010 in. The strong demagnetizing field inhomogeneities in such a chip give rise to a mode spectrum to which the uniform driving field can easily couple.

cause a substantial dispersion of the anisotropy field. In a particular yttrium iron garnet crystal reported by Dillon,[6] the line width rises to a value of 400 oersteds, which would correspond to a root-mean-square fluctuation of the anisotropy of about 300 oersteds. Since the anisotropy field rises from 100 to 3000 oersteds in this temperature range such a dispersion in value may be possible.

(c) Dipolar Narrowing in Paramagnets

Since the necessary condition for observation of dipolar narrowing of an inhomogeneously broadened line is that $4\pi M_s > H_p$ (H_p is the rms magnitude of the inhomogeneity), one should be able to observe the effect of dipolar narrowing in certain paramagnets at sufficiently low temperatures. For example, in the free radical $\alpha\alpha$-diphenyl β-picryl hydrazil at a temperature of 4.2°K in a field of 20 000 gauss (operating at 56 kMc/sec) the magnetization, M, becomes approximately 7.0 gauss and so inhomogeneities below this value should be narrowed. Furthermore, recalling that the separation between the magnetostatic modes in the region below κ_0 in Fig. 1 is of the order M, one should be able to excite and resolve these higher modes of magnetic resonance in this material as the line width is only one oersted.

ACKNOWLEDGMENTS

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[6] J. F. Dillon, Jr., Bull. Am. Phys. Soc. Ser. II, 2, 22 (1957).