C. J. LIN: I had considered the possibility of applying Dr. Schlömann's theory, but it is very hard to believe that definite spin wave modes could be excited in such small particles.

L.R. Bickford: I mean the earlier paper which predicted the asymmetric line shape for polycrystalline materials.

C.J. Lin: In our case, each grain of the sample consists of numerous single domain, single crystal particles and hence is quite different from Schlömann's case.

T. MIYADAI: Have you examined such a sample diluted in a non-magnetic substance?

C. J. LIN: We wish very much to do such kind of experiment but it is very hard to disperse the individual particles. We can disperese some of them for electron microscopic observation but most of them stick together.

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Ferrimagnetic Resonance of Iron-Oxide Micropowders*

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Ferrimagnetic resonance phenomena of single-domain γ-Fe_2O_3 powders with different particle shape distributions were studied at two frequencies: 24,116 Mc and 9,480 Mc. One of the powders had particles with close to spherical shape, which made it possible to estimate the g-factor, the first-order anisotropy constant, and the spin-spin relaxation time of γ-Fe_2O_3. The values obtained are: $g=1.97\pm0.02$, $K_1=-3.0\times10^5$ ergs/cm^3, $\tau_2=1.2\times10^{-10}$ sec. The resonance field of the acicular powders was lower than that of the spherical powder. The experimental data are in contradiction with theoretical results based on the Stoner-Wohlfarth model. The origin of this discrepancy is discussed.

Introduction

So far, few studies of the resonance phenomena of single-domain ferrimagnetic particles have been carried out. Brown, Hanton, and Morrish[1] investigated, theoretically and experimentally, the resonance absorption of suspensions of iron-oxide powders in zero applied steady magnetic field. Employing the Stoner-Wohlfarth model for a single-domain particle[2], they derived an expression for the absorption which was a simple function of the shape distribution of the particles in the powder, and their experimental results were in agreement with the theory. The present work is an investigation of the resonance phenomena of single-domain γ-Fe_2O_3 powders with different shape distributions when a steady magnetic field is applied. The results give some information on the properties of single-domain particles. Moreover, the fact that the particles of one of the powders were very close to spherical made it possible to estimate the g-factor, the first-order anisotropy constant, and the spin-spin relaxation time of γ-Fe_2O_3. An accurate determination of these quantities is at present impossible since no γ-Fe_2O_3 single crystal has as yet been grown.

Experiments

The experiments were carried out at room temperature and with two different frequencies: 24,116 Mc and 9,480 Mc. Three γ-Fe_2O_3

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powders, which differed from each other by particle shape distribution and particle size, were investigated. In powder 1, the particle shape was close to spherical. Electron-photomicrographs showed that the axial ratio was between 1.0 and 1.15 and that the particle size ranged from 0.03 to 0.2 micron, the average size being about 0.12 micron. Density measurements indicate that there are probably no cavities; there exists, however, the possibility of surface porosity. Powders 2 and 3 had acicular particle shape. Their approximate size and shape distributions were determined by means of an electron microscope previously, in connection with other experiments[3]. The particle length in these two powders ranged from 0.1 to 1.5 microns with a mean between 0.5 and 0.6 micron. In powder 2 the average axial ratio was 3.5 and in powder 3 it was 6.5. The powders were immobilized by suspension in Lucite or paraffin wax and from these suspensions, which had concentrations of 10%, 15%, and 19.4% by volume, spherical or disc samples were produced. For the X-band (9,480 Mc) measurements the diameters of the spherical samples were all in the vicinity of 1 mm, whereas for the K-band (24,116 Mc) experiments spheres with a diameter of about 0.5 mm were used.

Discussion of Results

Fig. 1 shows a typical set of absorption curves. These particular curves were obtained at 9,480 Mc. The line shape of the spherical powder is almost symmetrical; however, there is a slight asymmetry, the steeper branch of the curve being on the low-field side of resonance which indicates a negative first-order anisotropy constant[4]. The other two curves in Fig. 1 show that the resonance field decreases and the asymmetry of the line shape increases with increasing acicularity in such a way that the steeper branch of the curve remains on the low-field side. This is in contradiction with theoretical results based on the Stoner-Wohlfarth model, neglecting crystalline anisotropy and particle interaction, and assuming random orientation of the particles. This theory gives the imaginary part of the susceptibility as

$$\chi'' = \frac{\delta p}{4}\int_0^{2\pi} \Phi(\eta)d\eta \int_0^{\pi} \frac{[R(1+\cos^2\alpha)+(\cos 2\phi+\cos^2\alpha\cos^2\phi)]\sin\theta d\theta}{[R^2+R\eta(3\cos^2\phi-1)+\eta^2\cos^2\phi\cos 2\phi+\varepsilon]^2+\delta^2}, \qquad (1)$$

where

$$R=\frac{H_1}{M_s}\cos\alpha, \qquad \varepsilon=\frac{1-\omega^2\tau_2^2}{\tau_2^2\gamma^2M_s^2}, \qquad \text{and} \qquad \delta=\frac{2\omega}{\tau_2\gamma^2M_s^2}.$$

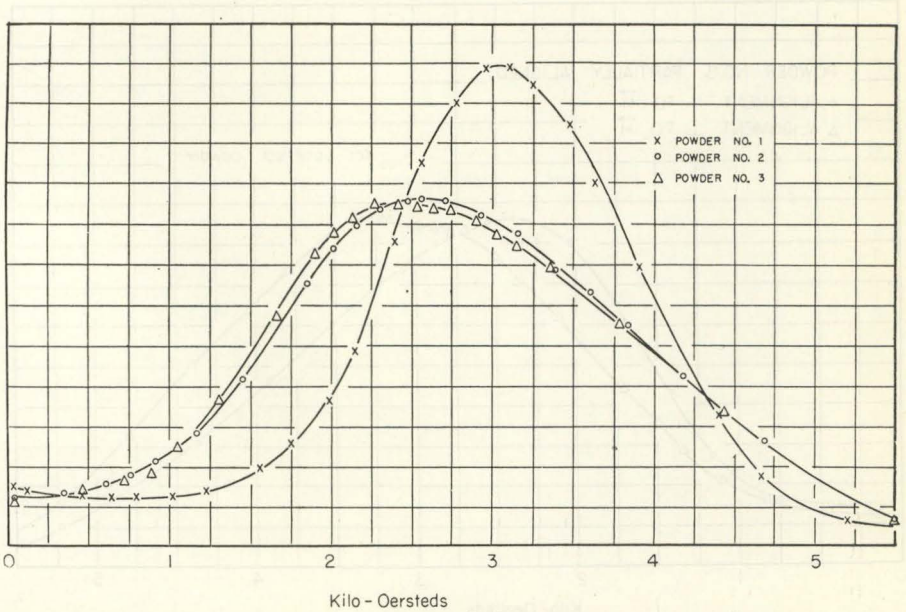

Fig. 1. Absorption curves of three γ-Fe_2O_3 powders with different shape distributions. Frequency = 9,480 Mc.

H_1 is the field seen by the particle, M_s the saturation magnetization, p the concentration by volume, and $\Phi(\eta)$ the shape distribution of the particles, where $\eta=D_b-D_a$, i.e., the difference between the transverse and the longitudinal demagnetizing factors. The damping is taken into account by the relaxation time τ_2. α is the angle between the steady field $\boldsymbol{H}_1$ and the equilibrium direction of magnetization of a certain particle, whereas θ is the angle between $\boldsymbol{H}_1$ and the easy axis of the particle. $\phi=\theta-\alpha$. The relation between these angles is given by the Stoner-Wohlfarth equation[2]

$$2H_1 \sin\alpha=M_s(D_b-D_a)\sin 2\phi\ . \qquad (2)$$

The integrations in equation (1) were carried out with the aid of a digital computer. The results thus obtained predicted a double-peak absorption curve, one peak being above and the other below the resonance field of the spherical powder. Hence, it appears that this model is not adequate for describing the behavior of the particles in question when a steady magnetic field is applied.

The results obtained with partially aligned powder 3, the most acicular powder, are shown in Fig. 2. When the alignment is along the field direction, the resonance field is lower than for the spherical powder, whereas for alignment perpendicular to the field direction, it is higher. This behavior is as predicted by theory. However, for perpendicular orientation, the resonance line is broadened and there is an indication of line structure. This phenomenon may be the origin of the discrepancy between our experimental results and the theory based on the Stoner-Wohlfarth model. It may be that an acicular particle ceases to be single-domain under the influence of a strong field perpendicular (or close to perpendicular) to its easy axis. A non-uniform magnetization may account for these phenomena, possibly in a manner analogous to the effect of domain structure on the ferromagnetic resonance in uniaxial crystals[5].

It was found that the resonance field of the spherical samples is virtually independent of the concentration of the suspension. This indicates that, at least for concentrations of the order of 10% to 20%, the field acting on the individual particle inside the sample is sufficiently well approximated by considering the particle located at the center of a spherical cavity in a continuous medium of magnetization pM_s, where p is the concentration. In the case of disc samples, a change of concentration shifted the resonance field by an amount in agreement with this model.

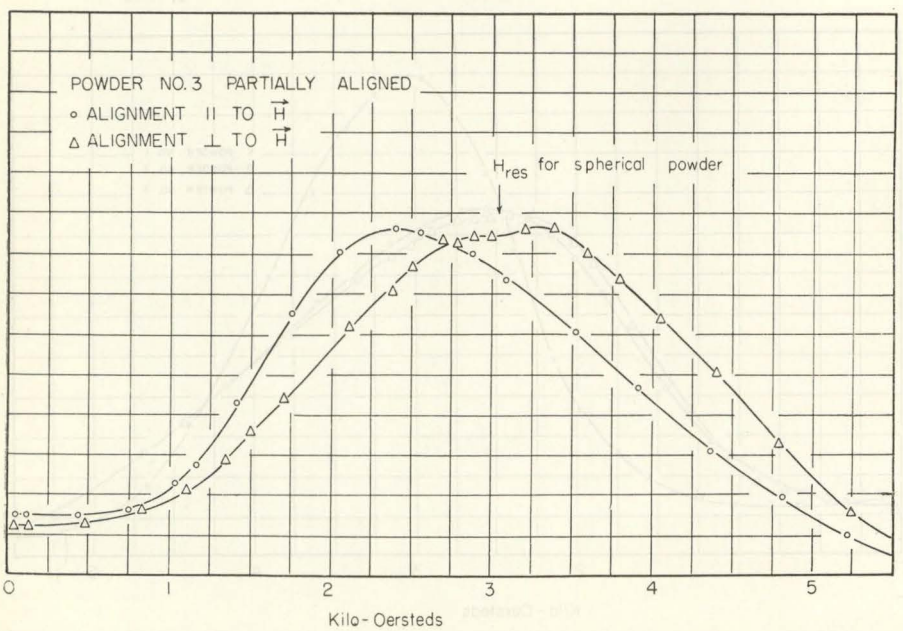

Fig. 2. Absorption curves of partially aligned acicular γ-Fe_2O_3 powder.

Table I. γ-Fe_2O_3 spherical powder

f Mc	H_{res} oe	ΔH oe	g	K_1 ergs/cm^3	τ_2 sec
9,480	3050	1750	1.97 ± 0.02	-3.0×10^5	1.2×10^{-10}
24,116	8360	1725			

The resonance field of an assembly of randomly oriented crystallites with cubic anisotropy is[4]

$$H_{res}=\frac{\omega}{\gamma}+\frac{K_1}{2M_s} \qquad (3)$$

and the line width is larger than that of a single crystal by $|K_1/M_s|$, where K_1 is the first-order anisotropy constant. (M_s=390 oe for γ-Fe_2O_3). Hence, by measuring the resonance field of the spherical powder at two different frequencies and using equation (3), the g-factor and K_1 were determined. By using K_1 and the measured line width, ΔH, of the spherical powder, the spin-spin relaxation time, τ_2, of γ-Fe_2O_3 was estimated. The results are given in Table I. A g-factor close to 2.00 is what one would expect, since the magnetic moments are those of the Fe^{3+} ions, which have a ^6S ground state.

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DISCUSSION

L. R. Bickford: Was the line shape the same at both frequencies?

A. H. Morrish: Much the same, except for the aligned sample with the field perpendicular to the alignment direction. The line structure observed at X-band was less certain at K-band.

E. P. Wohlfarth: The value of K_1 may presumably be affected by slight deviations from spherical shape and particle interactions.

A. H. Morrish: We agree, and no account has been taken of these factors.