

## Magnetic dimensional resonances in $\text{Fe}_3\text{O}_4$ spheres

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A series of new magnetic resonances were observed in magnetite ( $\text{Fe}_3\text{O}_4$ ) spheres in microwave magnetotransmission experiments, as follows. Microwaves (32–37 GHz) traveling in a cylindrical waveguide were incident on a small magnetite sphere suspended at the center of the guide. The waveguide passed axially through a superconducting solenoid, whose field could be varied from 0 to 6 T. The waves were circularly polarized in the plane perpendicular to the applied field. The experiments were performed at 5 K. The transmitted microwave power was measured as a function of the field. In addition to ferromagnetic resonances, new *size-dependent* resonances were observed, and were studied as a function of sphere diameter and microwave frequency. The behavior of the new resonances cannot be explained in terms of ordinary Walker modes. The dependence of these resonances on frequency and size is quite dramatic, showing a linear relation between the variables  $(B_0 - B_R)^{-1}$  and  $(\omega d)^{-2}$ , where  $B_0$  is the field at which the resonance occurs,  $B_R$  is the ferromagnetic resonance field for a given microwave frequency  $\omega$ , and  $d$  is the sphere diameter. The strength of the observed effect holds promise for application in nonreciprocal microwave devices.

### I. INTRODUCTION

Magnetite ( $\text{Fe}_3\text{O}_4$ ) belongs to the family of ferrites, which have a wide spectrum of applications. Studying the various properties of magnetite is therefore not only important in its own right, but bears on the understanding of the characteristics which ferrites have in common. Quite surprisingly, the magnetic properties of magnetite have not been studied in great detail. Magnetic resonance phenomena in this material have been investigated by Bickford<sup>1</sup> and others in the late 1950s and early 1960s. However, the early studies on magnetite in the areas of heat capacity and phase transitions have proved to be contradictory.<sup>2–7</sup> This is due almost entirely to the poor quality of the crystals used, and especially due to the lack of control of the stoichiometry of the samples. In this connection it is important to note that recently a new approach to crystal growth of  $\text{Fe}_3\text{O}_4$  via skull melting has yielded large single crystals of high purity.<sup>8</sup> Furthermore, reliable techniques for annealing  $\text{Fe}_3\text{O}_4$  crystals have been perfected, leading to a high degree of stoichiometry control.<sup>9,10</sup> The availability of samples of such improved quality motivated us to undertake a new series of experiments on magnetic resonance phenomena in this material.

We have studied magnetic resonances in  $\text{Fe}_3\text{O}_4$  spheres by microwave magnetotransmission experiments, to be described below. In this process, we have observed a new type of resonance that could not be described in terms of either the ferromagnetic resonance or magnetostatic (Walker) modes, which are the only magnetomicrowave resonance phenomena studied in ferrites so far. A typical transmission spectrum is shown in Fig. 1. We notice that, along with several weak, narrow resonances at

lower fields, the spectrum is dominated by a broad absorption line in the high-field region. It is the broad resonance which is new, and which we wish to concentrate on in the present paper. A striking feature observed in the case of the broad resonance is its strong dependence upon the sphere size and the frequency of the incident microwaves. In order to gain both a qualitative and a quantitative picture of this phenomenon, we have made a systematic study of the behavior of the resonance as a function of sphere size and microwave frequency. Based on earlier studies of dimensional resonances in anisotropic *magnetoplasma* spheres by Galeener and Furdyna,<sup>11</sup> by Galeener,<sup>12</sup> and by Dixon and Furdyna,<sup>13,14</sup> we have developed a simple phenomenological theory to describe

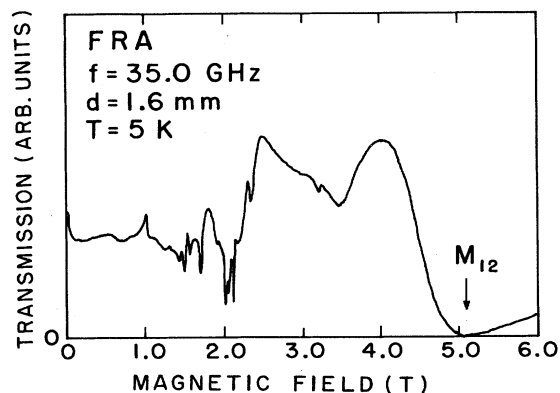


FIG. 1. A typical microwave transmission spectrum as a function of magnetic field for an  $\text{Fe}_3\text{O}_4$  sphere, 1.6 mm in diameter, at a microwave frequency of 35 GHz in ferromagnetic-resonance-active (FRA) configuration.

the observed dimension-related resonances in Fe<sub>3</sub>O<sub>4</sub>. This approach has been successful in describing the size and frequency dependences of the resonances. In addition, we were able to obtain by these means fairly good estimates for various material parameters.

In the next section, we briefly present the basic background needed for describing the interaction of microwaves with a magnetically anisotropic sphere. We then describe the experimental arrangement used in this investigation. Finally, we discuss the results which we have obtained, and relate them to the phenomenological formulation of dimensional resonances.

## II. THEORETICAL BACKGROUND

There are two principal resonances that have been observed in ferrites, namely, the ferromagnetic resonance and magnetostatic modes. Ferromagnetic resonance in ellipsoidal samples is given by the well-known Kittel condition,<sup>15</sup>

$$\omega = \gamma \mu_0 \{ [H_0 + (N_y - N_z)M_0][H_0 + (N_x - N_z)M_0] \}^{1/2}, \quad (1)$$

where  $\gamma$  is the gyromagnetic ratio,  $\mu_0$  is the permeability of vacuum,  $H_0$  is the external dc magnetic field,  $M_0$  is the magnetization, and  $N_x$ ,  $N_y$ , and  $N_z$  are the demagnetizing factors in the  $x$ ,  $y$ , and  $z$  directions, respectively (corresponding to the three principal axes of the ellipsoid). A sphere is a special and very important case, where

$$N_x = N_y = N_z = \frac{1}{3}. \quad (2)$$

Then Eq. (1) reduces to

$$\omega = \gamma \mu_0 H_0 \equiv \omega_0. \quad (3)$$

This is the ferromagnetic resonance condition for a sphere.

Magnetostatic modes are associated with modes of oscillation of an assembly of spins in which the spin phase varies throughout the sample, and are excited when the rf magnetic field is inhomogeneous at the sample. These modes have been observed by White and co-workers<sup>16,17</sup> and Dillon.<sup>18</sup> The conditions for excitation of these modes have been solved by Walker.<sup>19,20</sup> For spheres, the resonance conditions are as follows:

$$\begin{aligned} \text{set 1: } \omega &= \gamma \mu_0 \left[ H_i + M_0 \frac{m}{2m+1} \right], \\ \text{set 2: } \omega &= \gamma \mu_0 \left[ H_i + M_0 \frac{m}{2m+3} \right], \end{aligned} \quad (4)$$

where  $H_i$  is the internal dc magnetic field, and  $m$  are integers 1, 2, 3, . . . .

The two types of resonances described in Eqs. (1)–(4) depend on the shape, but neither of the resonances depends on the size of the sample. Small shifts with size have indeed been observed in the case of ferromagnetic resonance, the origin of such shifts lying in the propagation effects (i.e., the fact that the wavelength is finite within the sample medium), but so far this effect has been

in the nature of a correction to the main resonance condition.<sup>21,22</sup> The size dependence which we observe, on the other hand, is so large that it can no longer be dealt with as a perturbation to the ferromagnetic resonance, and so a new approach is necessary. In fact, we observe resonance harmonics, whereas in previous work only a small shift of the original ferromagnetic resonance was observed. We shall presently examine the observed behavior in terms of dimensional resonances in spheres, beginning with isotropic spheres.

### A. Dimensional resonances in an isotropic medium

Dimensional resonances will occur in a bounded specimen when a certain relationship exists between the wavelength inside the sample and the size of the sample, giving rise to internal multiple reflections which constructively interfere to give a high concentration of electromagnetic energy within the sample. This effect is analogous to the more familiar phenomenon of Fabry-Perot interference in plane parallel slabs and films involving multiple reflections.

Such effects can be rigorously formulated for a dielectric sphere. The solution to the problem of the interaction of a plane electromagnetic wave with a uniform isotropic dielectric sphere was first obtained by Mie.<sup>23,24</sup> The solution consists of expanding the incident, scattered, and interior waves in a well-defined infinite series of electric and magnetic vector waves, with appropriate weighting factors. For example, the weighting factors for the scattered waves (also known as scattering coefficients) are denoted by  ${}^e s_n$  and  ${}^m s_n$ , where the superscript indicates the electric ( $e$ ) or magnetic ( $m$ ) character of the wave, and  $n = 1, 2, 3, \dots$  gives the multipole order of the wave. Thus,  $n = 1$  corresponds to dipole waves,  $n = 2$  to quadrupole waves, and so on. The weighting factors for the three species of waves mentioned above are determined by matching the interior and exterior waves at the sphere surface by use of the isotropic boundary conditions for the rf electric ( $\mathbf{E}$ ) and magnetic ( $\mathbf{H}$ ) fields.

The extinction cross section,  $\Sigma_{\text{ext}}$ , is defined as the ratio of the sum of the absorbed and scattered power to the incident power, and can be expressed in terms of  ${}^e s_n$  and  ${}^m s_n$  as follows:<sup>11,12</sup>

$$\Sigma_{\text{ext}} = \frac{2\pi}{k_0^2} \sum_{n=1}^{\infty} (2n+1) [\text{Re}({}^e s_n) + \text{Re}({}^m s_n)], \quad (5)$$

where  $k_0$  is the wave number of the incident wave.

When the size of the sphere is small compared to the outside wavelength of the incident wave, the sphere is said to be in the *dipole limit*. This is expressed by the condition

$$k_0 a \ll 1, \quad (6)$$

where  $a$  is the radius of the sphere. In this limit, the quadrupole ( $n = 2$ ), octopole ( $n = 3$ ), and higher-order contributions can be neglected relative to the dipole ( $n = 1$ ) term. Also, for small spheres, we can neglect the

scattering cross section, so that the extinction and absorption cross sections are nearly equal.

Equation (6) does not imply that the radius of the sphere  $a$  is small compared to the *internal* wavelength. Indeed, for large values of the dielectric constant (and/or magnetic permeability), the inequality  $ka > 1$  may be satisfied simultaneously with Eq. (6), where  $k$  is the wave number *within* the medium from which the sphere is made. In that case, electric or magnetic dimensional resonances will occur whenever  $^e s_n$  or  $^m s_n$  diverge, giving rise to maxima in the absorbed power.

In general, the internal wave number  $k$  and the dielectric constant of the sphere medium  $\kappa$  are complex, and can be expressed in terms of their real and imaginary parts as follows:

$$\begin{aligned} k &= \alpha + i\beta, \\ \kappa &= \kappa' + i\kappa'', \end{aligned} \quad (7)$$

where  $\alpha$  and  $\beta$  are the phase and attenuation coefficients, respectively. When Eq. (6) holds, the expressions for  $^e s_n$  and  $^m s_n$  can then be expressed in terms of  $\kappa$  and  $k$ ,

$$\begin{aligned} ^e s_n &\approx \frac{-i}{2n+1} \frac{[(n+1)\kappa+n]j_n(z) - zj_{n-1}(z)}{n(\kappa-1)j_n(z) + zj_{n-1}(z)} \\ &\quad \times \left[ \frac{2^n n!}{(2n)!} \right]^2 (k_0 a)^{2n+1}, \\ ^m s_n &\approx \frac{-i}{2n+1} \frac{(2n+1)j_n(z) - zj_{n-1}(z)}{zj_{n-1}(z)} \\ &\quad \times \left[ \frac{2^n n!}{(2n)!} \right]^2 (k_0 a)^{2n+1}, \end{aligned} \quad (8)$$

where  $z = ka$  and  $j_n(z)$  is the spherical Bessel function of order  $n$ . It is easily shown that, owing to Eq. (6), the dominant contributions to Eq. (8) are from the  $n=1$  (i.e., dipole) terms.

In the limit of low losses,  $j_n(z)$  can be expanded as follows:

$$j_n(z) \approx j_n(x) + iyj'_n(x), \quad (9)$$

where  $x = \alpha a$ ,  $y = \beta a \ll x$ , and the prime on  $j_n(x)$  denotes a derivative with respect to the argument. From Eq. (8), the resonance condition for magnetic dipole resonances is then

$$xj_0(x) - y^2 j'_0(x) = 0, \quad (10a)$$

which for  $\beta \ll \alpha$  becomes simply

$$j_0(x) \approx 0. \quad (10b)$$

Similar expressions can be obtained for electric dipole resonances. For small losses and large  $\kappa'$ , it is observed that the conditions at which the electric and magnetic dimensional resonances occur are adequately given by<sup>11,12</sup>

$$\alpha a = E_m \quad (11)$$

for the electric dimensional resonances, and

$$\alpha a = M_m$$

for the magnetic dimensional resonances, where  $E_m$  and  $M_m$  correspond to the zeros of  $j_1(x)$  and  $j_0(x)$ , respectively. Since  $j_0(x)$  and  $j_1(x)$  each have a multiplicity of zeros, there is a series of solutions. These are identified by the index  $m$ . Physically, the different values of  $m$  may be approximately regarded as "harmonics," i.e., as the number of wavelengths "fitting" within the sphere diameter.

## B. Dimensional resonances on gyrotropic spheres

The interaction of a plane electromagnetic wave with a *gyrotropic* sphere is a complicated problem, with no known solutions in closed analytic form. The main difficulty arises from the fact that in a gyrotropic medium the dielectric constant (or the permeability) is a tensor instead of a scalar, and consequently Maxwell's equations are no longer separable in spherical coordinates. Ford and Werner solved the problem numerically for a gyrotropic dielectric sphere of arbitrary size, along the lines of the Mie theory.<sup>25</sup> The Mie solutions could then be retrieved from the general Ford-Werner formulation by replacing the dielectric tensor by a scalar.

Using the Ford-Werner formulation as a point of departure, Dixon and Furdyna developed a phenomenological approach which gives an approximate but rather satisfactory analytical description of the dimensional resonances in gyrotropic *plasma* spheres in the dipole limit. In the section below we shall extend their approach to the present gyromagnetic case.

The basic elements of the Dixon-Furdyna approach to the interaction of a plane electromagnetic wave with a spherical plasma in an external dc magnetic field are as follows. The dielectric tensor  $\vec{\kappa}$  for an infinite magnetized plasma from which the sphere is made has the form

$$\vec{\kappa} = \begin{pmatrix} \kappa_+ & 0 & 0 \\ 0 & \kappa_- & 0 \\ 0 & 0 & \kappa_{zz} \end{pmatrix}, \quad (12)$$

where  $\kappa_{\pm} = \kappa_{xx} \pm i\kappa_{xy}$  describes the dielectric response of the plasma to the two circular polarizations in the  $x$ - $y$  plane (i.e., in the plane transverse to the dc magnetic field  $\mathbf{B}_0$ ), and  $\kappa_{zz}$  describes the response of the plasma to  $\mathbf{E}_{\text{rf}} \parallel \mathbf{B}_0$ .

Dixon and Furdyna showed that when the sphere is excited by *one* of the independent modes (corresponding to the subscript  $+$ ,  $-$ , or  $zz$ ), Mie-type relations still hold. In particular, for excitation by one of the circular polarizations transverse to  $\mathbf{B}_0$ , the resonance condition is given by

$$\text{Re}(k_{\pm} a) = G_{1m}, \quad (13)$$

where  $k_+$  and  $k_-$  correspond to the wave numbers of the  $+$  and  $-$  circularly polarized excitations in the infinite plasma medium, respectively, and  $G_{1m}$  is a constant. In the specific case of a magnetized plasma,  $G_{1m}$  is equal to  $\sqrt{m(m-1/2)\pi}$  for the major magnetic dimensional resonances, and to  $\sqrt{m(m+1/2)\pi}$  for the major electric di-

mensional resonances. The empirical resonance conditions are therefore

$$\operatorname{Re}(k_{\pm}a) = \sqrt{m(m+1/2)}\pi$$

for the electric dimensional resonances, and (14)

$$\operatorname{Re}(k_{\pm}a) = \sqrt{m(m-1/2)}\pi$$

for the magnetic dimensional resonances. The results of Dixon and Furdyna are supported by exact numerical calculations employing the full Ford and Werner solution for the gyrotropic sphere.<sup>13</sup>

### C. Dimensional resonances in Fe<sub>3</sub>O<sub>4</sub>

The spheres used in our experiments were sufficiently small to be in the dipole limit ( $ka \ll 1$ ). Using the same qualitative approach as that of Dixon and Furdyna, we surmise that, in the case of an incident circularly polarized plane wave on a sphere made of a *magnetically* anisotropic medium, a series of dipole resonances will occur determined by the conditions

$$\begin{aligned} \operatorname{Re}(k_{+}a) &= G_{1m}, \\ \operatorname{Re}(k_{-}a) &= H_{1m}, \end{aligned} \quad (15)$$

where the + and - subscripts refer, respectively, to the ferromagnetic-resonance-active (FRA) and ferromagnetic-resonance-inactive (FRI) circular polarizations transverse to  $\mathbf{B}_0$ , the subscript 1 on  $G$  and  $H$  indicates dipole excitation, and  $m$  is an index indicating harmonics (i.e., how many standing wave antinodes are contained in the internal resonant field pattern).

Note that the empirical resonance conditions of Dixon and Furdyna given by Eq. (14)—i.e., the specific values of  $G_{1m}$ —emerged from the specific mathematical properties of the dielectric tensor  $\vec{\kappa}$  for a magnetized *plasma*. In extending this approach to the gyromagnetic case, we have assumed the *form* of Eq. (13), but we regard  $G_{1m}$  and  $H_{1m}$  in Eq. (15) as unknown constants (except that  $G_{12} > G_{11}$ , etc., as for the plasma case).

In the present case, the medium is described by an isotropic dielectric constant  $\kappa$  of the order of 15, and a gyrotropic magnetic permeability tensor  $\vec{\mu}$ , which can be written as

$$\vec{\mu} = \begin{pmatrix} \mu_{+} & 0 & 0 \\ 0 & \mu_{-} & 0 \\ 0 & 0 & \mu_0 \end{pmatrix}, \quad (16)$$

where  $\mu_{\pm} = \mu_0(1 + \chi_{\pm})$ , and  $\chi_{\pm}$  is the dynamic magnetic susceptibility corresponding to the two circularly polarized normal modes of propagation (FRA and FRI) for the medium from which the sphere is made. Equation (15) for the FRA mode can then be written as

$$\frac{\omega\sqrt{\kappa}}{c} \operatorname{Re}(1 + \chi_{+})^{1/2} = G_{1m}. \quad (17)$$

The expressions for the real ( $\chi'_{\pm}$ ) and imaginary ( $\chi''_{\pm}$ ) parts of  $\chi_{\pm}$  are given by the Bloch equations,

$$\begin{aligned} \chi'_{\pm} &= \frac{\omega_M(\omega_0 \mp \omega)T_2^2}{(\omega_0 \mp \omega)^2 T_2^2 + 1}, \\ \chi''_{\pm} &= \frac{\omega_M T_2^2}{(\omega_0 \mp \omega)^2 T_2^2 + 1}, \end{aligned} \quad (18)$$

where  $\omega_0 = \gamma\mu_0 H_0$ ,  $\omega_M = \gamma\mu_0 M_0$ ,  $T_2$  is the transverse relaxation time,  $H_0$  is the applied magnetic field,  $M_0$  is the saturation magnetization, and  $\gamma$  ( $=ge/2m$ ) is the gyromagnetic ratio.

For fields much higher than the ferromagnetic resonance field, i.e., for  $\omega_0 \gg \omega$ , and for  $(\omega_0 - \omega)T_2 \gg 1$ , inverting Eq. (17) yields

$$1 + \chi_{+} \approx 1 + \frac{\omega_M}{\omega_0 - \omega} = \frac{4G_{1m}^2 c^2}{\kappa\omega^2 d^2}, \quad (19)$$

where  $d = 2a$  is the sphere diameter. The above equation can be rewritten in terms of the fields. Writing  $\omega$ , the angular frequency of the microwaves, in the form

$$\omega = \gamma B_R, \quad (20)$$

where  $B_R$  is the ferromagnetic resonance field corresponding to  $\omega$ , and expressing  $\omega_0$  and  $\omega_M$  in terms of the applied magnetic field and saturation magnetization, respectively, we arrive at the resonance condition in the following form:

$$\frac{1}{B_0 - B_R} = \frac{4G_{1m}^2 c^2}{\kappa\mu_0 M_0} \frac{1}{\omega^2 d^2} - \frac{1}{\mu_0 M_0}. \quad (21)$$

The unknown parameters in our problem are  $M_0$ ,  $G_{1m}$ ,  $B_R$ , and  $\kappa$ , and the variable parameters are  $\omega$  and  $d$ . From the above equation we see immediately that (i) the magnetic field  $B_0$  corresponding to a dimensional resonance varies nonlinearly with  $\omega$  for a fixed  $d$  (and nonlinearly with  $d$  for a fixed  $\omega$ ), according to

$$B_0 = B_R + \mu_0 M_0 \left[ \frac{4G_{1m}^2 c^2}{\kappa\omega^2 d^2} - 1 \right]^{-1}, \quad (22)$$

(ii) a plot of  $(B_0 - B_R)^{-1}$  versus  $(\omega d)^{-2}$  should be a straight line, with a slope equal to  $4G_{1m}^2 c^2 / \kappa\mu_0 M_0$ , and an intercept equal to  $-(\mu_0 M_0)^{-1}$ . Equation (21) will be the point of departure for analyzing our experimental results.

## III. EXPERIMENT

### A. Microwave spectrometer

The microwave spectrometer employed in this study has been described previously.<sup>26</sup> The Fe<sub>3</sub>O<sub>4</sub> spheres were placed between two pieces of tape and suspended in a circular waveguide passing axially through the bore of a 60-kG superconducting solenoid. In all of our experiments, we used circularly polarized microwaves. The microwave power transmitted past the Fe<sub>3</sub>O<sub>4</sub> spheres was measured as a function of the dc magnetic field  $B_0$ . The experiments were carried out between 32 and 37 GHz.

### B. Sample preparation

Single crystals of  $\text{Fe}_3\text{O}_4$  of very high purity were grown at Purdue University by the skull melter process.<sup>8</sup> All the samples used in our experiments were stoichiometric. The spherical samples were prepared using an Enraf Nonius sphere grinder, as follows. Small irregular  $\text{Fe}_3\text{O}_4$  samples were placed inside the grinder, and air was blown tangentially through a number of channels. This swirled the pieces against the abrasive walls of the sample chamber, grinding them into spheres. The sphere diameter was measured by a traveling microscope. A single sphere was then placed between two pieces of tape, and placed inside the sample holder on the axis of the superconducting magnet, as described above.

## IV. RESULTS AND INTERPRETATION

For a given sphere size, magnetotransmission was studied at a series of microwave frequencies. We started with a sphere of diameter 1.6 mm and recorded the transmission spectra as a function of magnetic field for the following microwave frequencies: 32.4, 33.0, 33.5, 34.0, 34.4, 35.0, 35.5, and 36.0 GHz. At each frequency, the spectrum was obtained for both the FRA and FRI polarizations. The above procedure was repeated for three arbitrary orientations of the sphere, to check for possible anisotropy (crystallographic or geometrical). The sphere was then ground to a smaller size, and the above procedure repeated. This was done for a series of sizes. The successive diameters at which the spheres were studied were 1.6, 1.4, 1.2, 1.1, 1.0, 0.9, 0.8, and 0.7 mm. The temperature was maintained at 5 K for all the measurements.

As mentioned earlier, our aim was to gain an understanding of the dominant resonance shown in Fig. 1, i.e., its dependence on sphere diameter and on frequency, as well as to determine what material parameters could be extracted from such resonances. It was difficult to make a detailed analysis of the closely spaced resonances in the low-field region of the spectrum, since they did not show a systematic size or frequency dependence. Also, their positions depend upon the orientation of the sphere. In this paper we shall therefore focus our attention only on the broad resonances occurring at higher fields.

### A. Resonances in the FRA configuration

We have observed two-dimensional resonances in the FRA (or +) configuration. From our phenomenological model, this corresponds to

$$\text{Re}(k_+ a) = G_{1m}, \quad m = 1, 2,$$

where the index  $m$  indicates the resonance "harmonic," as explained earlier. The dimensional resonance corresponding to  $m=2$  (designated  $M_{12}$  in Figs. 2 and 3) was observed for sphere diameters equal to 1.6, 1.4, and 1.2 mm. As illustrated in Figs. 2 and 3, the resonance showed a rather dramatic size and frequency dependence. From Fig. 2 we observe that for a sphere of diameter 1.6 mm, the  $M_{12}$  resonance shifted from nearly 50 to 28 kG (almost a factor of 2) when the frequency was reduced

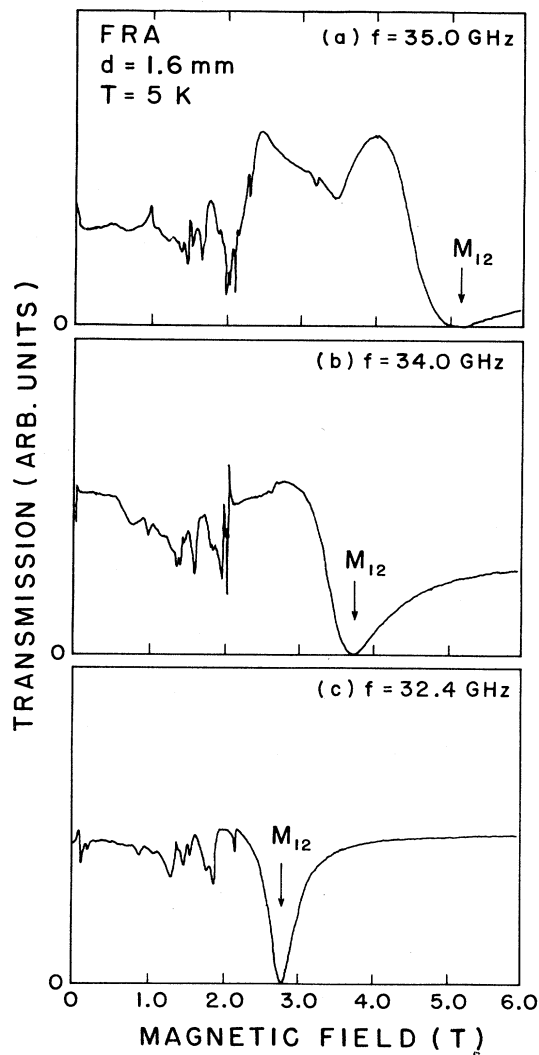


FIG. 2. Variation of the position of the  $M_{12}$  dimensional resonance with incident microwave frequency, for a fixed sphere diameter. Curve (a) is the transmitted power of circularly polarized microwaves for a 1.6-mm sphere at 35 GHz, curve (b) is for 34 GHz, and curve (c) corresponds to 32.4 GHz. All data are taken at 5 K.

from 35 to 32.4 GHz (less than 10%). Figure 3 shows the absorption spectra at a fixed frequency of 35 GHz for three spheres of diameters: 1.6, 1.4, and 1.2 mm. Once again we observe a very significant shift in the resonance field with only a small decrease in sphere size—the resonance shifted from 60 to 26 kG (nearly a factor of 2) when the sphere diameter was reduced by only 13%. When the sphere was further ground to 1.2 mm in diameter, the  $M_{12}$  resonance shifted to still lower fields, eventually becoming one of the sharp resonances in the low-field region as the microwave frequency was reduced, as shown in Fig. 4(a).

As the  $M_{12}$  resonance of the 1.2-mm sphere merged with the low-field lines when the frequency was de-

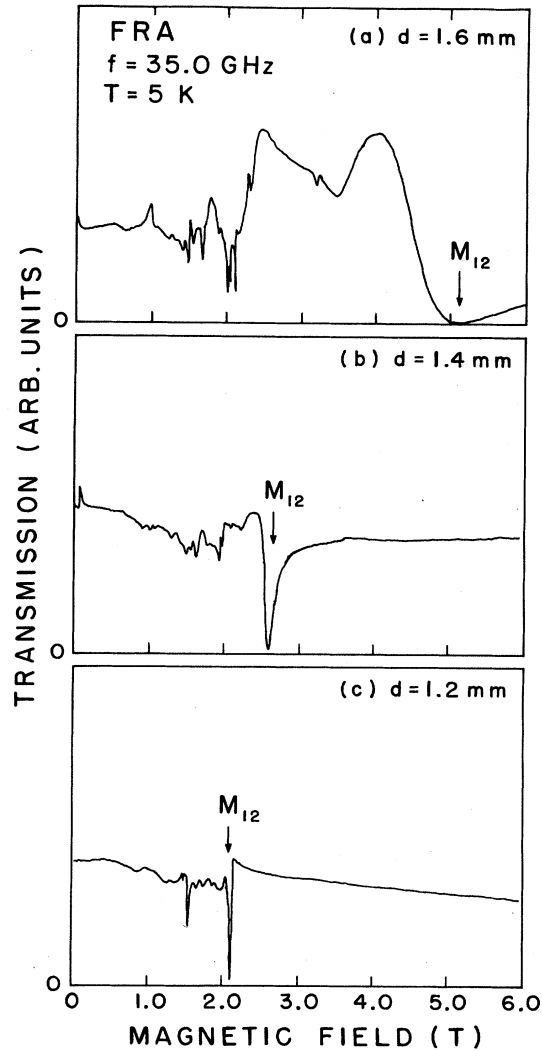


FIG. 3. Variation of the position of the  $M_{12}$  resonance with sphere size, at a fixed frequency of 35 GHz. Curve (a) is the transmitted power for circularly polarized microwaves for a sphere of 1.6 mm in diameter, curve (b) is for a diameter of 1.4 mm, and curve (c) corresponds to a diameter of 1.2 mm. All data are taken at 5 K.

creased, a *new* dimensional resonance  $M_{11}$  appeared, emerging near 60 kG for 33 GHz and moving to 45 kG at 32.4 GHz, as shown in Fig. 4(b). This resonance corresponds to  $\text{Re}(k+a) = G_{11}$ . We continued to study the behavior of the  $M_{11}$  resonance for sphere diameters equal to 1.1, 1.0, 0.9, 0.8, and 0.7 mm. The size and frequency dependences of the  $M_{11}$  resonance for the 1.1- and 1.0-mm spheres were qualitatively similar to those displayed by the  $M_{12}$  resonance. For smaller sizes the  $M_{11}$  resonance also merged with the sharp low-field lines. As would be expected, the intensity of the resonance decreased progressively with decreasing sphere size.

In order to check that the observed  $M_{11}$  resonance was a *magnetic-dipole-excited* dimensional resonance, we performed the following simple experiment. The 1.1-mm

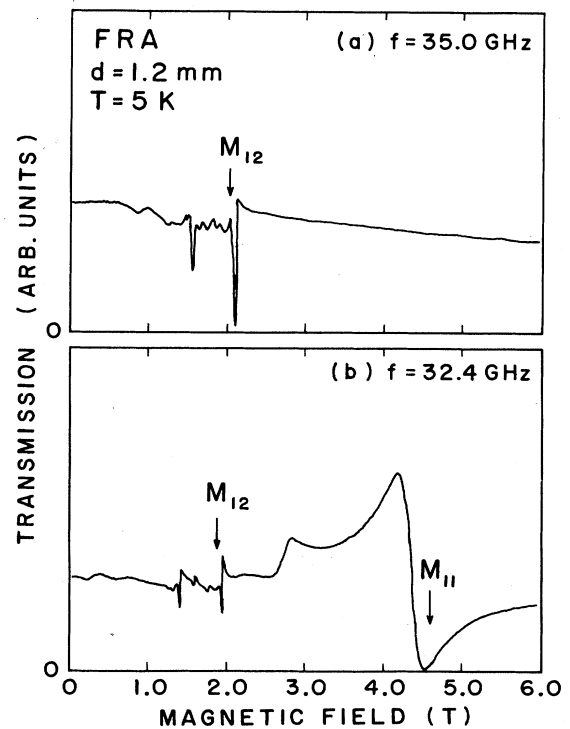


FIG. 4. Transmitted microwave power as a function of magnetic field for a 1.2-mm sphere at 35 and 32.4 GHz. Curve (a) shows the  $M_{12}$  resonance in the immediate vicinity of narrow lines in the low-field region. As  $M_{12}$  merges with these lines when the frequency is lowered, a new dimensional resonance,  $M_{11}$ , appears near 45 kG, as shown in curve (b).

sphere was mounted on top of a metal termination, and the reflected power was measured as a function of the dc magnetic field. The sphere was then raised by a distance equal to one-quarter of the microwave wavelength and the above procedure was repeated. In the first position, where the rf magnetic field was maximum and the rf electric field was zero, the  $M_{11}$  resonance occurred around 28 kG at 35 GHz, as expected. In the second position, where the rf electric field was a maximum, the intensity of the resonance diminished considerably, and no new resonances appeared, thereby confirming that the  $M_{11}$  resonance was a magnetic-dipole dimensional resonance.

We now test the phenomenological theory developed in Sec. II, Eq. (21), which predicts a linear relationship between  $(B_0 - B_R)^{-1}$  and  $(\omega d)^{-2}$  for  $M_{11}$  and  $M_{12}$ , the two straight lines having intercepts at  $-(\mu_0 M_0)^{-1}$ . In plotting the results, the position of the dimensional resonance  $B_0$  was taken to be the field at which the transmitted power was a minimum. The ferromagnetic resonance field  $B_R$  was defined through the relation  $\omega = \gamma B_R$ , with the gyromagnetic ratio  $\gamma$  treated as a parameter to be determined. In plotting  $(B_0 - B_R)^{-1}$  versus  $(\omega d)^{-2}$  for various values of  $\gamma$ , we found that fairly good linear fits could be obtained for  $\gamma$  between  $1.27 \times 10^7$  and  $1.70 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$  for both  $M_{11}$  and  $M_{12}$ , which correspond to  $g$  factors between 1.5 and 1.9. The best fit occurred for  $\gamma = 1.40 \pm 0.05$ , as shown in Fig. 5, corresponding to

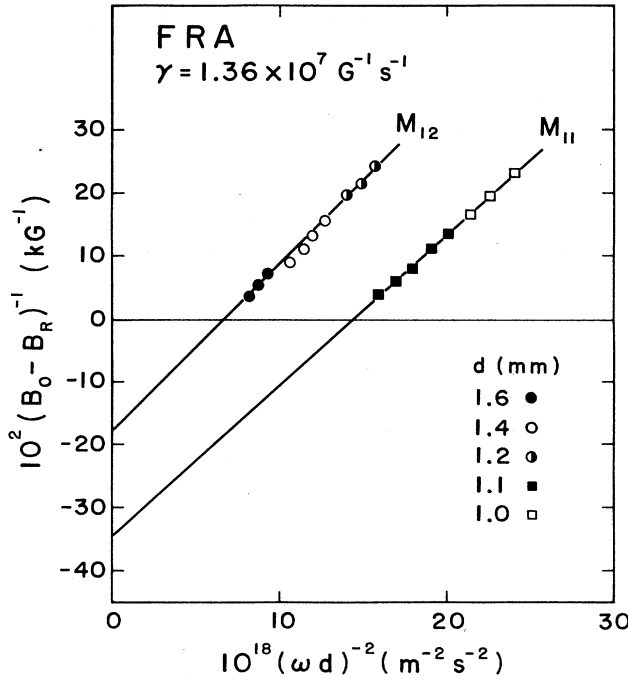


FIG. 5. A plot of  $(B_0 - B_R)^{-1}$  vs  $(\omega d)^2$  for  $\gamma = 1.36 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$ , for both the  $M_{11}$  and  $M_{12}$  dimensional resonances. Note that the plots are linear, but do not have a common intercept predicted by the phenomenological theory.

$g = 1.60 \pm 0.05$ .

The problem with this "best fit" is that, while it supports Eq. (21) for the  $M_{11}$  and  $M_{12}$  resonances taken separately, the two straight lines do not have a common intercept on the  $(B_0 - B_R)^{-1}$  axis, in contradiction with the phenomenological model. In fact, even if we allow the value of  $\gamma$  to range outside the limits given above, to where the plots deviate significantly from linearity, we are not able to find a value of  $\gamma$  for which the plots for both resonances extrapolate to a common intercept. If, instead, we allow the value of the parameter  $B_R$  to be different for  $M_{11}$  and  $M_{12}$ , two values of  $\gamma$  can be found for which the plots are linear, and which have the same intercept for both resonances, as illustrated in Fig. 6. These values of  $\gamma$  are approximately  $1.63 \times 10^7$  and  $1.36 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$  for  $M_{11}$  and  $M_{12}$ , respectively, corresponding to  $g$  factors of 1.85 and 1.54. Also, the value of  $M_0$  determined from the intercept is between  $4.3 \times 10^5$  and  $4.5 \times 10^5 \text{ A/m}$ , which is in reasonable agreement

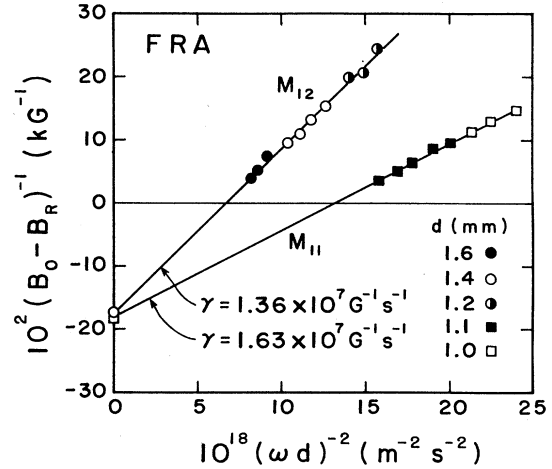


FIG. 6. A plot of  $(B_0 - B_R)^{-1}$  vs  $(\omega d)^2$  for  $\gamma = 1.63 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$  and  $1.36 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$  corresponding to the  $M_{11}$  and  $M_{12}$  dimensional resonances, respectively. Note that, to achieve a common intercept, different values of  $\gamma$  must be used for the two-dimensional resonances.

with the value  $5.06 \times 10^5 \text{ A/m}$  obtained for  $\text{Fe}_3\text{O}_4$  by dc magnetization measurements of Chikazumi and others.<sup>27</sup> The values of  $B_R$ , the intercepts  $M_0$ , and other parameters corresponding to the two dimensional resonances are tabulated in Table I.

We can thus summarize our results as follows. The fact that we get excellent linear fits for the  $(B_0 - B_R)^{-1}$  versus  $(\omega d)^2$  plots for each dimensional resonance separately implies that the phenomenological theory correctly identifies the mechanisms for the dramatic frequency and size dependences of the resonances. On the other hand, if we consider  $M_{11}$  and  $M_{12}$  together, the best linear fits do not intersect on the  $(B_0 - B_R)^{-1}$  axis, thereby resulting in different values of  $M_0$ . We can obtain a common intercept for  $M_{11}$  and  $M_{12}$  only by choosing different values of the ferromagnetic resonance field parameter  $B_R$  for the two resonances. We have thus shown that we cannot simultaneously describe all observed resonances using a single value of  $B_R$  and  $M_0$  in Eq. (21). While we do not understand this discrepancy with the model, it must be remembered that Eq. (21) represents an *ad hoc* phenomenological relation, and that the actual relationship connecting the resonance conditions with the Bloch equations is undoubtedly more complex. It is also possible that the assumption made implicitly in Eq. (17)—that the

TABLE I. Slopes and intercepts corresponding to the  $M_{11}$  and  $M_{12}$  FRA resonances for equal and different values of  $B_R$  (at 32.4 GHz).

Resonance	Ferromagnetic resonance field $B_R$ (kG)	Intercept ( $10^{-2} \text{ kG}^{-1}$ )	$10^{-17}$ slope	Saturation magnetization $M_0$ (A/m)	$G_{12}/G_{11}$
$M_{11}$	15.0	-34.15	2.38	2.33	~1.5
$M_{12}$	15.0	-17.54	2.63	4.53	
$M_{11}$	12.5	-18.25	1.39	4.36	~1.35
$M_{12}$	15.0	-17.54	2.63	4.53	

dielectric constant  $\kappa$  is real, isotropic, and frequency independent—may be too simplistic, thus leading to a qualitative departure from the exact predictions of Eq. (21).

Further experiments—perhaps on ferrites, which are better characterized and less complicated than  $\text{Fe}_3\text{O}_4$ —would be desirable in order to shed further light on this interesting question.

### B. Resonances in the FRI configuration

The features of the magnetotransmission spectrum in the ferromagnetic-resonance-inactive (or FRI) configuration resemble the FRA spectrum, consisting of a broad resonance at high fields and a series of closely spaced narrow resonances at lower fields. We have observed two magnetic dimensional resonances in this configuration, whose size and frequency dependences are qualitatively similar to the FRA case. Unlike the FRA configuration, no resonances were observed for spheres smaller than 1.1 mm in diameter. From our model, the two FRI dimensional resonances correspond to

$$\text{Re}(k_{-a}) = H_{1m}, \quad m = 1, 2,$$

where  $H_{11}$  and  $H_{12}$  are unknown constants. The model predicts a linear relationship between  $(B_0 + B_R)^{-1}$  and  $(\omega d)^{-2}$  for appropriate values of  $B_R$ , with a slope and intercept equal to  $4H_{1m}^2 c^2 / (\kappa \mu_0 M_0)$  and  $-(\mu_0 M_0)^{-1}$ , respectively. As before, we fit the data by using the gyromagnetic ratio  $\gamma$  as a fitting parameter. One such plot is shown in Fig. 7. Again, the two straight lines do not intersect on the  $(B_0 + B_R)^{-1}$  axis, in contradiction with the theory. Furthermore, the values of the intercepts (and therefore of the saturation magnetization) differ from those obtained from the FRA dimensional resonances by at least a factor of 2. Nevertheless, qualitatively the theory does appear to describe the observed size and frequency dependences. Beyond this, it is difficult to make any estimates for the ferromagnetic resonance field  $B_R$  or the saturation magnetization  $M_0$  from the limited FRI data available. However, the very existence of strong dimensional resonances in the FRI polarization should prove useful in providing additional data for a better understanding of the general problem presented in this paper, once a more rigorous model is formulated.

### V. CONCLUSION

We have extended the phenomenological approach used by Dixon and Furdyna in their investigation of dimensional resonances in gyrotropic plasma spheres to gyromagnetic  $\text{Fe}_3\text{O}_4$  spheres. It is clear that this approach is at least qualitatively successful in identifying

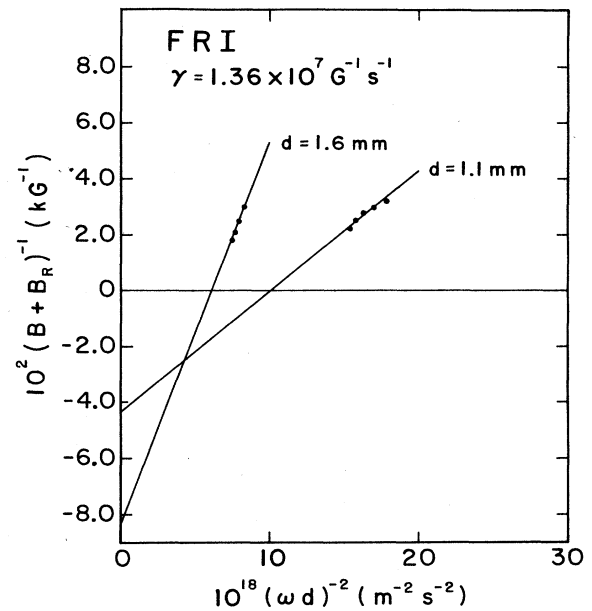


FIG. 7. A plot of  $(B_0 + B_R)^{-1}$  vs  $(\omega d)^{-2}$  for  $\gamma = 1.36 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$  for the two-dimensional resonances in the ferromagnetic-resonance-inactive (FRI) configuration, using the same value of  $\gamma$  ( $1.36 \times 10^7 \text{ G}^{-1} \text{ s}^{-1}$ ) as in Fig. 5.

the origin of the dimensional resonances in this system, and in describing the main features of their striking size and frequency dependences. As a result, we also get estimates for the ferromagnetic resonance field (and consequently the  $g$  factor) and the saturation magnetization. However, the fact that in fitting the dimensional resonances we require different ferromagnetic resonance field parameters  $B_R$  for different dimensional resonances is puzzling and not understood at the present stage.

As is clear from the data shown, the dimensional resonance effect observed in the small  $\text{Fe}_3\text{O}_4$  spheres is exceedingly strong. This strength of the interaction, along with the fact that the resonance condition can be *tuned* by the sphere size, holds promise for possible applications of the effect in microwave nonreciprocal devices, once this striking phenomenon is better understood quantitatively.

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