Microwave susceptibility and effective linewidth of iron-carbonyl powders

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The effective linewidth technique has been applied to study the microwave response and effective linewidth of iron carbonyl powders. The frequency and Q of a high-Q microwave cavity containing small powder samples was measured over a field range of 0–12 kOe at 10 GHz. The frequency data for high field (9–12 kOe) were used in conjunction with magnetization data and theory to calibrate the microwave system. The calibration parameters were then used in conjunction with the Q data to determine the negative imaginary part of the powder microwave permeability μ'' vs field. The μ'' results at high field were then fitted to theory to estimate the operational effective linewidth parameter for the powders. The procedure provided clear microwave evidence for (1) magnetic saturation of the powders in the expected field range of 7–8 kOe, (2) the operational validity of a model which treats the powder particles as noninteracting spheres, and (3) an off-resonance effective linewidth of about 500 Oe at 10 GHz.

The objective of this study was to perform off-resonance microwave permeability measurements and effective linewidth analyses on a series of iron-carbonyl powders. This technique has proved useful for the separation of resonance losses from off-resonance losses and the determination of intrinsic linewidth parameters under conditions for which direct FMR measurements are not possible.[1,2]

The effective linewidth technique is discussed at length in the references cited above. The basic idea is that cavity Q and frequency data can be used in conjunction with microwave perturbation theory and an analysis of the dynamic magnetic response to obtain the field dependence of the negative imaginary part of the microwave permeability for the material of interest. Based on various models for the microwave response, one can use the high-field permeability data to determine an operational "effective linewidth" for the material.

The complex microwave permeability μ for spherical, nonconducting particles magnetized to saturation by an applied static magnetic field H_0 may be written as

$$\mu - 1 = \omega_m(\omega_0 + i\eta_0)/[(\omega_0 + i\eta_0)^2 - \omega^2]. \quad (1)$$

The parameters $\omega_m = \gamma 4\pi M_s$ and $\omega_0 = \gamma H_0$ express the saturation induction $4\pi M_s$ and the static applied field H_0 in frequency units [γ is the (positive) gyromagnetic ratio, approximately 2.9 GHz/kOe for iron], ω is the microwave frequency, and η_0 denotes the relaxation rate. The "effective linewidth" for a material with the above response is given by $\Delta H_{\rm eff} = 2\eta_0/\gamma$. This $\Delta H_{\rm eff}$ parameter simply expresses the off-resonance relaxation rate in linewidth units for convenient comparison with the conventional half-power ferromagnetic resonance linewidths for saturated materials.

The microwave premeability is related to the microwave cavity Q and resonance frequency ω through perturbation theory.[2] With the usual definition of μ in terms of its positive real and negative imaginary parts, $\mu = \mu' - i\mu''$, the connections with Q and ω are given by

$$\mu' - 1 = -2K[(\omega - \omega_\infty)/\omega] \quad (2)$$

and

$$\mu'' = K(1/Q - 1/Q_\infty). \quad (3)$$

The ω_∞ and Q_∞ parameters correspond to the cavity resonance frequency and Q extrapolations at infinite field. From the response in Eq. (1), the infinite field frequency ω_∞ can be obtained from plots of ω vs a "$1/H$" parameter p given by

$$p = (\omega/\omega_0)/[1 - (\omega/\omega_0)^2]. \quad (4)$$

Such plots (essentially ω vs $1/H_0$) are linear at high field [as one would expect from Eqs. (1) and (2)] and extrapolate to ω_∞ at $p = 0$ (or $H_0 = \infty$). Similarly, Q_∞ is obtained from plots of Q vs a "$1/H^2$" parameter q given by

$$q = \frac{1}{2}\left(\frac{\gamma^2}{\omega_0^2}\right)\frac{[1 + (\omega/\omega_0)^2]}{[1 - (\omega/\omega_0)^2]^2}. \quad (5)$$

Such plots (essentially Q vs $1/H_0^2$) are linear at high field and extrapolate to Q_∞ at $q = 0$ (or $H_0 = \infty$). These extrapolation procedures were carried out for fields in the range 10–15 kOe.

The coefficient K in Eqs. (2) and (3) may be obtained empirically or directly from perturbation theory. The empirical approach was used here. This approach uses the fact that μ' at high field is relatively insensitive to the parameter η_0 and depends only on γ, $4\pi M_s$, H_0, and ω. Calculated values of μ' vs field were used in conjunction with measurements of the cavity frequency versus field and Eq. (2) to determine K for a given powder sample/cavity combination. This value of K was used to convert Q versus field data to values of μ'' versus field. The imaginary part of Eq. (1) was then fitted to the empirical μ'' vs H_0 results to determine η_0 and $\Delta H_{\rm eff}$. Values of K from perturbation theory were found to be almost a factor of 2 smaller than indicated by the empirical approach. These errors are attributed to problems in accurately estimating sample volumes, cavity loading effects for the metallic powder specimens, and modifications in μ due to particle interactions.

The basic experimental procedure was to load a small, loosely compacted powder sample in a high-Q TE011 cylindrical 9.9-GHz microwave cavity, use the technique of Refs. 1 and 2 to measure the cavity frequency and Q as a function of applied field, use the analysis described above to calibrate

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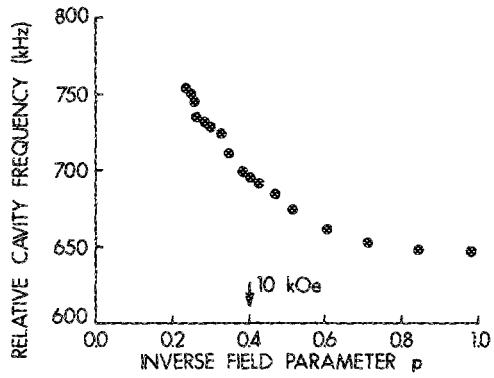

FIG. 1. Cavity frequency, relative to a reference frequency of 9.966 GHz, as a function of the "$1/H$" inverse field parameter p defined in the text.

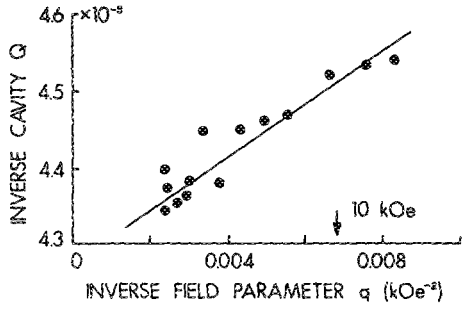

FIG. 2. Inverse cavity Q as a function of the "$1/H^2$" inverse field parameter q defined in the text.

the system response, and then determine μ'' versus field and the effective linewidth at high field.

Data were obtained on iron-carbonyl powder materials with 0.6–0.8 wt. % carbon, 0.6–0.8 wt. % nitrogen, nominal particle sizes of 1.5–2.5 μm, and nominal saturation inductions ($4\pi M_s$) of 17–19 kG. The $4\pi M_s$ values were obtained by standard vibrating sample magnetometry (VSM) techniques. Magnetic saturation required fields in excess of about 10 kOe, as expected for iron-based powders consisting of irregular but roughly spherical particles.[3] The measurements were made on 0.5–0.6-mg powder samples compacted and sealed in 1-mm-diam cups in small microwave dielectric sample holders.

Data on cavity frequency versus the dimensionless "$1/H$" parameter p are shown in Fig. 1. The frequency values on the vertical axis indicate the cavity frequency shift, in kHz, relative to 9.966 GHz. The 700-kHz point, for example, corresponds to a cavity frequency of 9.966 700 GHz. The measured cavity frequency increases as the "p" parameter decreases. There is a more-or-less linear region for $p < 0.4$ and a more gradual, somewhat rounded response for higher values of p. The $p = 0.4$ point corresponds to an applied static field H_0 of 10 kOe. As mentioned above, the VSM data showed that fields above about 10 kOe are necessary to magnetically saturate the powders, a necessary condition for the above microwave response equations to be applicable. The data for $p < 0.4$ were used, therefore, for the extrapolation to obtain ω_∞.

Figure 2 shows the corresponding results on inverse cavity Q vs the "$1/H^2$" parameter q. Here, the high field data region corresponding to magnetic saturation ($H_0 > 10$ kOe is for $q < 0.007$ kOe^{-2}). The changes in cavity Q at high field, however, are so small that the scatter in the data makes it difficult to separate the character of the $1/Q$ vs q response into saturated and unsaturated regions. Nevertheless, only the data for $q < 0.007$ kOe^{-2} were used for the $q = 0$ ($H_0 = \infty$) extrapolation to obtain Q_∞.

Figure 3 shows two theoretical curves and two sets of data points for the reduced real part of the permeability ($\mu' - 1$) vs static field H_0. The theoretical curves are based on Eq. (1) for two different values for ΔH_{eff}, 500 Oe [curve (a)] and 4000 Oe [curve (b)]. These curves show that ($\mu' - 1$) is relatively insensitive to the choice of the ΔH_{eff} parameter for fields above 10–12 kOe.

The open circle data points in Fig. 3 represent values of ($\mu' - 1$) which were obtained directly from perturbation theory, based on the theoretical empty-cavity unperturbed TE011 mode, a sample volume inferred from measured sample mass and density values, and the frequency data discussed above. It is evident from the figure that these points lie well *below* the theoretical curves. The discrepancy at the highest fields shown is almost a factor of 2. This discrepancy is attributed to cavity loading effects and the lack of a well-defined powder volume parameter for the metallic powders. The approach used here was to adjust empirically the perturbation theory coefficient K in Eq. (2) [and in Eq. (3)] to "force" a fit of the ($\mu' - 1$) data to the theoretical curves at very high field, $H_0 > 14$ kOe. This "empirical" K value was then used for the μ'' versus field and effective linewidth analysis (discussed below). The result of this procedure for ($\mu' - 1$) is shown by the *solid* circle data points in Fig. 3. These points track the theoretical curve for $\Delta H_{\text{eff}} = 500$ Oe rather well for fields above 9 kOe and fall below this curve at lower fields. This fall-off is attributed to the lack of magnetic saturation for fields below 10 kOe.

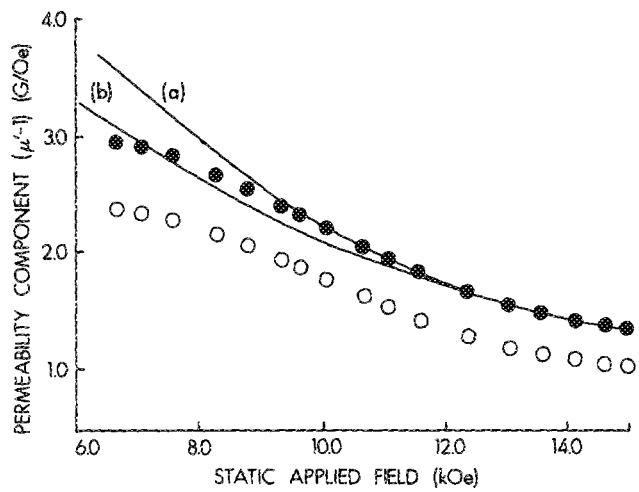

FIG. 3. Real part ($\mu' - 1$) of the reduced microwave permeability as a function of applied static field. The solid lines represent theoretical curves for $\Delta H_{\text{eff}} = 500$ Oe [curve (a)] and $\Delta H_{\text{eff}} = 4000$ Oe [curve (b)]. The open circles represent the experimental results based on the frequency data and cavity perturbation theory. The solid circles represent the experimental results adjusted to fit the theoretical curves at high field.

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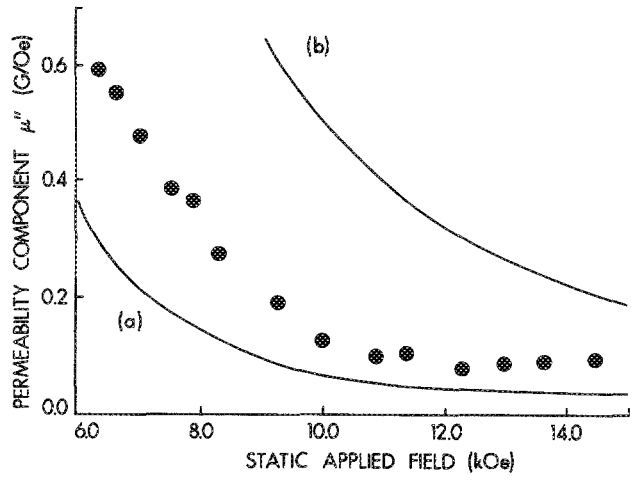

FIG. 4. Negative imaginary part μ'' of the reduced microwave permeability as a function of applied static field. The solid lines represent theoretical curves for $\Delta H_{\text{eff}} = 500$ Oe [curve (a)] and $\Delta H_{\text{eff}} = 4000$ Oe [curve (b)]. The solid circles represent the experimental results based on the empirical cavity calibration procedure discussed in the text.

Before considering the μ'' results, it is worthwhile to point out that the curves and fits in Fig. 3 for $(\mu' - 1)$ already give a rough indication of the operational ΔH_{eff} value for the powder. The theoretical curve for $\Delta H_{\text{eff}} = 500$ Oe tracks the solid data points down to somewhat lower fields than does the $\Delta H_{\text{eff}} = 4000$ Oe curve. This better tracking provides a preliminary indication that the operational powder effective linewidth is in the "hundreds of Oe" rather than the "thousands of Oe" range.

Figure 4 shows the corresponding theoretical curves and adjusted data points for μ'' vs H_0. The two curves are based on the same parameters as in Fig. 3, with ΔH_{eff} values of 500 Oe (lower curve) and 4000 Oe (upper curve). The data points (solid circles) were obtained from the Q vs H_0 data and the empirical K parameter from the $(\mu' - 1)$ fit discussed above. The theoretical μ'' vs H_0 curves for $\Delta H_{\text{eff}} = 500$ Oe and $\Delta H_{\text{eff}} = 4000$ Oe are quite different. As is evident from Eq. (1), the theoretical expression for μ'' contains η_0 (and hence ΔH_{eff}) as a multiplying factor. It is precisely this sensitivity of μ'' to ΔH_{eff} that makes it possible to use the μ'' versus field data to determine high-field effective linewidths. The data points in Fig. 4 lie slightly above the theoretical curve for $\Delta H_{\text{eff}} = 500$ Oe and track this curve fairly closely down to $H_0 = 10$ kOe. This high-field fit indicates an intrinsic X-band effective linewidth slightly higher than 500 Oe. The slight upturn in the data points at fields above 13 kOe is an artifact of the technique at high fields when μ'' becomes small.

For fields below 10 kOe, the data points move above the theoretical curve for $\Delta H_{\text{eff}} = 500$ Oe. As the powder becomes unsaturated, the microwave losses increase and the measured μ'' values become larger than expected on the basis of Eq. (1). It should be possible to use μ'' data for the entire range of fields below saturation, $0 < H_0 < 10$ kOe, to model the powder microwave properties for the partially magnetized and demagnetized states.

An alternative procedure to determine ΔH_{eff} should be mentioned. This procedure is based on the fact that the *slope* of the $1/Q$ vs q response at high field (Fig. 2) is proportional to η_0, and hence to ΔH_{eff} as well. The straight line in Fig. 2 corresponds to $\Delta H_{\text{eff}} = 500$ Oe, based on the empirical calibration parameter K discussed above.

The authors are grateful to Dr. I. B. Goldberg of the Rockwell International Science Center, Thousand Oaks, California, for supplying the samples, and to Dr. Gopalan Srinivasan for assistance with the measurements and analyses.

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