Effect of packing fraction on ferromagnetic resonance in $NiFe_2O_4$ nanocomposites

Han Song, Sam Mulley, Nathan Coussens, Pallavi Dhagat,[a] Albrecht Jander, and Alexandre Yokochi
Oregon State University, Corvallis, Oregon 97331, USA

(Presented 2 November 2011; received 23 September 2011; accepted 19 December 2011; published online 13 March 2012)

Magnetic nanocomposites, composed of magnetic nanoparticles in an insulating matrix, can have properties not achievable in bulk, single-phase materials. This work reports on ferromagnetic resonance (FMR) measurements as a tool to investigate the internal magnetic field in nanocomposite samples of varying particle packing fractions. The ferromagnetic resonance frequency changes with particle packing fraction due to increased inter-particle interaction and demagnetizing field within the sample. Experimental results obtained on $NiFe_2O_4$ nanocomposite samples are compared to published theoretical models, and found to be consistent with the predicted trends. Extrapolation of the results to the limit of isolated particles indicates an average internal anisotropy field of 0.041 T for the $NiFe_2O_4$ nanoparticles. © *2012 American Institute of Physics*. [doi:10.1063/1.3679635]

I. INTRODUCTION

Magnetic composite materials, consisting of fine metallic magnetic particles in an insulating matrix, are promising candidates for high frequency applications, e.g., high frequency inductor cores. The composite, while possessing high resistivity to minimize eddy current losses, can simultaneously be tailored for high permeability and low coercivity by optimizing the magnetic properties and packing fraction of the constituent particles. Additionally, in contrast to traditional ferrite materials, magnetic composites can be compatible with standard semiconductor processing. In this effort, ferromagnetic resonance behavior, which limits the useful frequency range of such materials, is investigated as a function of particle packing fraction in nanocomposite samples. A phenomenological model of ferromagnetic resonance (FMR) behavior in composites was previously presented by Ramprasad *et al.*[1] Their results, reproduced with permission in Fig. 1, show that particle anisotropy and packing fraction influence the FMR frequency in a complex manner. These predicted trends are experimentally verified in this study with $NiFe_2O_4$ nanocomposite samples.

II. EXPERIMENTAL TECHNIQUE

Magnetic nanocomposite samples were prepared by mixing $NiFe_2O_4$ nanopowder (MTI Corporation, average diameter = 30 nm) with a non-magnetic binder (KBr) in a mortar. The powder mixture was then compressed into pellets (7 mm diameter, 1 mm thick) using a hand press, under zero magnetic field. A series of samples in the following packing fractions were made: 0.5%, 1.0%, 2.4%, 4.8%, 9.7%, 14.5%, 23.7%, 31.5%, and 39.2% (Packing fraction refers to the volume percent of magnetic nanoparticles in the KBr binder).

A standard lock-in detection experimental setup[2] was used to measure the ferromagnetic resonance response of the nanocomposite samples (Fig. 2(a)). The pellet samples were taped onto the center trace of a 50 Ω coplanar waveguide, illustrated in Fig. 2(b), and placed between the pole pieces of an electromagnet. The electromagnet was used to provide a *dc* magnetic field ranging from 0 T to 0.5 T. A small modulation field (3 mT, 100 Hz) was added to the *dc* field, to enable lock-in detection. To investigate the effects of sample shape and sample orientation, all samples were measured with the *dc* field directed both in-plane and perpendicular to the sample (out-of-plane), as shown in Figs. 2(c) and 2(d). Reproducibility of the results is verified by repeating the measurements on multiple samples for each packing fraction.

The waveguide is excited at a constant frequency, between 6 GHz and 10 GHz, using a microwave signal generator. The microwave signal is partially absorbed by the sample and the rest transmitted to a power detector. The power detector detects the oscillations in absorbed power, enabling the FMR response to be extracted by the lock-in amplifier at the modulation frequency. The measured signal is the field derivative of absorbed microwave power (dP/dH_{dc}) as a function of the *dc* magnetic field. Shown in Fig. 3 are example signal traces, obtained with a 23.7 vol% sample, for a range of microwave excitation frequencies. The zero crossing in the signal corresponds to the peak in absorption and thus, identifies the applied *dc* field, H_{app}, at which ferromagnetic resonance occurs for the given microwave excitation.

III. RESULTS AND DISCUSSION

The resonance frequency, f_{res}, for an ideal bulk magnetic sample without any anisotropy is

$$f_{res} = \frac{\gamma}{2\pi}\mu_0 H_{app}, \tag{1}$$

where γ is the gyromagnetic ratio (typically, $\gamma \approx 176$ GHz/T), μ_0 the permeability of free space, and H_{app} the applied *dc* field at which ferromagnetic resonance occurs. In the case of magnetic nanoparticles formed into nanocomposite samples of finite shape, Eq. (1) must be modified as follows to include an

[a]Corresponding author. Electronic mail: dhagat@eecs.oregonstate.edu.

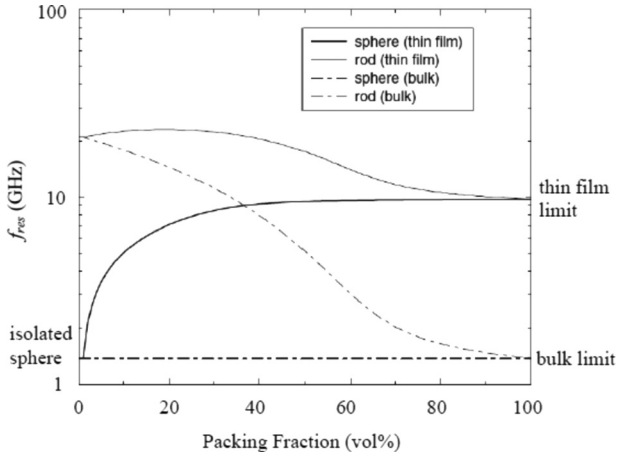

FIG. 1. FMR frequency, f_{res}, vs particle packing fraction for spherical and finite rod (aspect ratio of 2) particle composites. "Bulk limit" means demagnetizing field is negligible in any direction; "thin film limit" means demagnetizing field is significant only along the film normal. (Reprinted with permission from Ramprasad *et al*. (Ref. 1)).

effective field, H_{int}, arising from inter-particle dipole interactions, particle anisotropy, and the demagnetizing field within the sample:

$$f_{res} = \frac{\gamma}{2\pi}\mu_0(H_{app} + H_{int}). \quad (2)$$

H_{int} can be described by $H_{int} = H_{dem} + H_{anis} + H_{dip}$, where H_{dem} is the demagnetizing field within the sample; H_{anis} the particle anisotropy field; and H_{dip} the dipolar interaction field between particles.

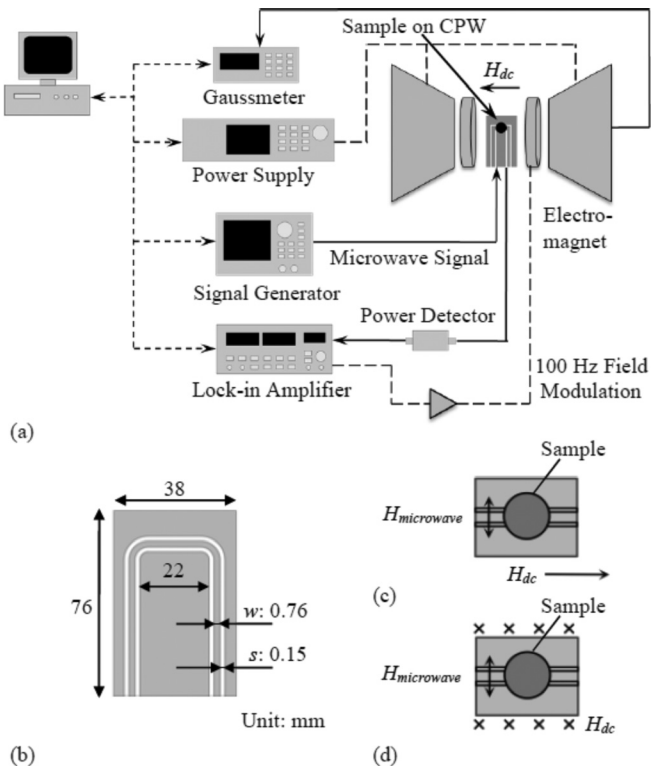

FIG. 2. (a) Experimental setup for FMR measurements. (b) Schematic of the coplanar waveguide. (c and d) In-plane and out-of-plane measurement configurations. $H_{microwave}$ is the microwave field. H_{dc} is the *dc* magnetic field.

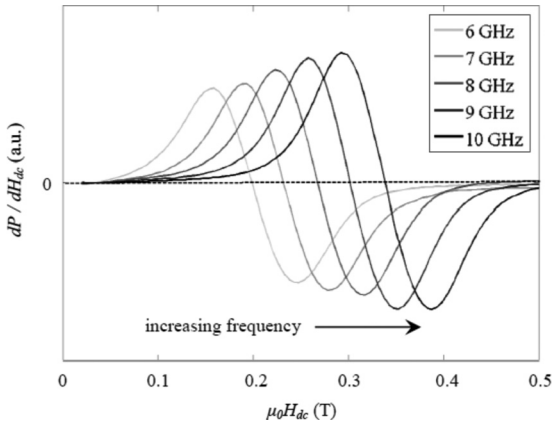

FIG. 3. FMR responses (out-of-plane measurement) of a 23.7 vol% $NiFe_2O_4$ nanocomposite sample at 6, 7, 8, 9, and 10 GHz, respectively.

The demagnetizing field, H_{dem}, depends on the particle packing fraction, sample shape and sample orientation. The field, H_{dip}, representing inter-particle dipole interactions is mainly packing fraction dependent. The anisotropy term, H_{anis}, is an effective field arising from particle shape anisotropy, crystalline anisotropy and orientation of the particle. In this study, the nanocomposite samples have no overall anisotropy since the particles are randomly oriented in the matrix. Thus H_{anis} is due only to the average internal anisotropy and shape of the nanoparticles themselves and will remain the same for different particle packing fractions and sample orientations.

Theoretically, in the isolated particle limit (that is, approaching a nanocomposite of zero volume percent packing fraction) the resonance behavior is affected only by the particle anisotropy. Both the sample demagnetizing field and inter-particle interactions become negligible. Therefore, assuming that the trend at very low packing is linear, the average isolated particle anisotropy field can be determined by extrapolation of the internal magnetic field, H_{int}, to zero vol%. Though not experimentally possible, for a nanocomposite approaching 100% packing fraction the resonance response should hypothetically reach the value expected for a fully dense, bulk sample of the given shape, and be determined predominantly by the sample demagnetizing field. Thus the resonance response, in this case, would follow Kittel's equation,[3]

$$f_{res} = \frac{\gamma\mu_0}{2\pi}\sqrt{(H_{app} + (N_x - N_z)M_s)(H_{app} + (N_y - N_z)M_s)}. \quad (3)$$

N_x, N_y, and N_z are demagnetizing factors in the x, y, and z directions with the *dc* field assumed to be in the z direction. M_s is the saturation magnetization of the magnetic material. Thus as the packing fraction of the nanocomposite is varied from close to zero to the maximum possible, the resonance response changes gradually from that of isolated magnetic particles to that of a magnetic bulk material.

Figure 4 shows the applied *dc* field at which ferromagnetic resonance is observed, $\mu_0 H_{app}$, as a function of the microwave excitation frequency, f_{res}. For in-plane measurements, $\mu_0 H_{app}$ is seen to decrease with increasing volume fraction. In contrast, for out-of-plane measurements, it

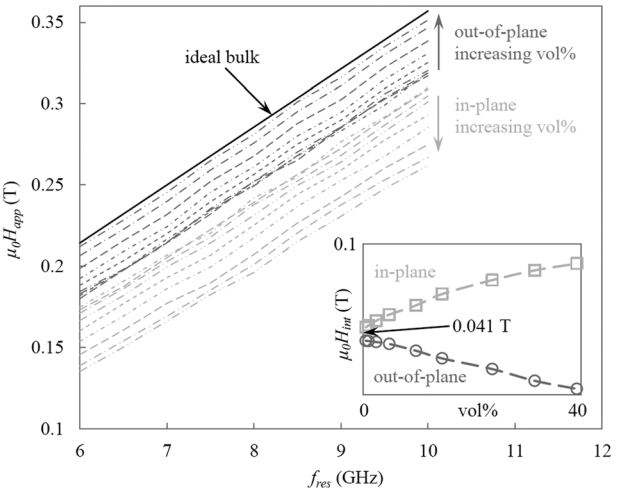

FIG. 4. Frequency dependence of the applied field at which FMR occurs in $NiFe_2O_4$ nanocomposite samples with different packing fractions. The "ideal bulk" line is given by Eq. (1). The inset shows the derived effective internal field, $\mu_0 H_{int}$, as a function of packing fraction and sample orientation.

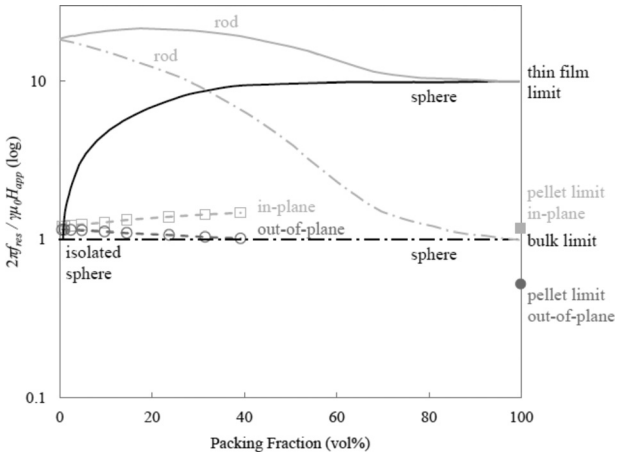

FIG. 5. Comparison between Ramprasad's model and experimentally measured FMR frequency normalized by $(\gamma\mu_0 H_{app}/2\pi)$ for different packing fractions.

increases with volume fraction. The standard deviation in the reported resonance field is less than 3 mT.

Also plotted in Fig. 4, is the response for an "ideal bulk" sample as described by Eq. (1). The vertical offset of the measured responses from the ideal trace is ascribed to the internal magnetic field, $\mu_0 H_{int}$, and obtained from linear fitting the measured data to Eq. (2). The inset in Fig. 4 shows the internal magnetic field as a function of the volume fraction. When extrapolated to zero vol%, the two curves for in-plane and out-of-plane measurements meet at a value of $\mu_0 H_{int} = 0.041$ T, which is identified as the average effective anisotropy field ($\mu_0 H_{anis}$) of the $NiFe_2O_4$ nanoparticles. Similar measurements can be performed for different magnetic nanoparticles, making this technique a tool for comparing particle anisotropy.

The measured data are compared to results from an effective medium model, presented by Ramprasad *et al.*[1] The model considers nanocomposites with varying packing fractions of sphere and rod shaped particles having identical properties. The rod-shaped particles possess relatively high shape anisotropy, while the shape anisotropy of the spherical particles is zero. The modeled and measured results are obtained with different applied fields; the data are therefore normalized to the respective ideal resonance frequency determined by Eq. (1) and re-plotted in Fig. 5 for comparison.

At the isolated particle limit, the resonance is influenced only by the average particle anisotropy as discussed earlier. For the $NiFe_2O_4$ samples, the resonance frequency in the isolated particle limit lies between that for the sphere-shaped and rod-shaped particles. Thus, not unexpectedly, the results suggest that the anisotropy of the nearly spherical $NiFe_2O_4$ nanoparticles, is slightly higher than that of the ideal sphere-shaped particles and considerably less than the rod-shaped particles.

Further, as the packing fraction increases, the in-plane and out-of-plane curves of the experimental samples diverge. This is due to the differing effects of demagnetizing field in the sample for the two cases. For in-plane measurements, the increasingly strong demagnetizing effects perpendicular to the applied field increase the resonance frequency. For out-of-plane measurements, the demagnetizing field tends to oppose the applied field and reduce the resonance frequency. As the packing fraction is increased to 100%, the demagnetizing field in the sample dictates the FMR behavior. Thus the modeled curves approach values corresponding to the shape of the assumed sample geometries, i.e., either an infinite thin film or a "bulk" sample of infinite extent in all dimensions. As for the experimental samples, the pellet-shaped geometry gives demagnetizing factors of 0.12 and 0.76 in-plane and out-of-plane, respectively. Using $\mu_0 M_s = 0.2$ T as provided by the manufacturer and $\mu_0 H_{app} = 0.3$ T in Eq. (3), the expected normalized resonance frequency at the 100% volume limit is 1.20 and 0.56 for the in-plane and out-of-plane measurements respectively (Fig. 5). Although packing fractions beyond 40% are not accessible experimentally, the trend in the curves for the in-plane and out-of-plane measurements is toward these limits.

In summary, though quantitatively different, the measured FMR responses are in agreement with Ramprasad's model in the sense of how demagnetizing field, dipole interactions and particle shape anisotropy influence the FMR behavior of magnetic nanocomposites.

IV. CONCLUSION

Ferromagnetic resonance responses of magnetic nanocomposites consisting of $NiFe_2O_4$ nanoparticles in a nonmagnetic binder were measured and analyzed. The results show that the packing fraction as well as the sample shape and orientation has a significant effect on the resonance behavior, due to the sometimes competing influences of demagnetizing field, inter-particle interactions and intra-particle anisotropy. The measured results are consistent with theoretical models published in literature.

[1]R. Ramprasad, P. Zurcher, M. Petras, M. Miller, and P. Renaud, Phys. Status Solidi B **233**, 31 (2002).
[2]S. S. Kalarickal, P. Krivosik, M. Wu, C. E. Patton, M. L. Schneider, P. Kabos, T. J. Silva, and J. P. Nibarger, J. Appl. Phys. **99**, 093909 (2006).
[3]C. Kittel, Phys. Rev. **73**, 155 (1948).