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The influence of particle concentration and polarizing field on the resonant behaviour of magnetic fluids

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Abstract

Starting from the Landau–Lifshitz equation, with resonant frequency $f_0 = \omega_0/2\pi$, it is demonstrated that, in the case of a magnetic fluid, the measured resonant frequency, f_{res} is always different from f_0 , except for the case of pure resonance (i.e. zero damping parameter of Landau–Lifshitz equation) where $f_{res} = f_0$. It is also shown that f_{res} and the corresponding maximum absorption frequency, f_{max} , are different, thus supporting the deductions of Scaife, who arrived at this conclusion using an alternative theoretical approach.

Furthermore, based on complex magnetic susceptibility measurements, over the frequency range 100 MHz–6 GHz, the dependence of the ratio f_{max}/f_{res} on an external polarizing magnetic field, H_{pol} , over the approximate range 0 and 1.3 kOe and on particle concentration has been examined for different magnetic fluid samples. It is demonstrated how the ratio f_{max}/f_{res} tends to unity both by (i) increasing the polarizing field and (ii) decreasing the particle concentration of the samples.

1. Introduction

Magnetic fluids are colloidal systems that consist of single-domain ferro-ferrimagnetic particles dispersed in a carrier liquid and coated with a surfactant to prevent agglomeration [1]. The magnetic properties of magnetic fluids have been studied intensively because they have multiple applications, ranging from instrumentation to medicine. Also, magnetic fluids seem to be an advantageous model system for the simulation and study of disordered systems, as their structure and particle concentration can be controlled easily, depending on the production process, temperature or presence of external fields [2].

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Taking into account the thermal fluctuations of the magnetic moment of the particles and neglecting interparticle interactions, Scaife has shown theoretically [3] that, for a magnetic fluid, the resonant frequency, f_{res} , and the frequency of maximum absorption, f_{max} , are always different except for the case of pure resonance (i.e. zero damping parameter of the Landau–Lifshitz equation [4]) where $f_{res} = f_{max}$.

Here we use an alternative theoretical approach to that of Scaife and demonstrate that, for a magnetic fluid, the resonant frequency, f_{res} , is always different to the theoretical resonant frequency, f_0 , except for the case of pure resonance where $f_{res} = f_0$. It is also shown that f_{res} and the frequency of maximum absorption, f_{max} , are different.

For magnetic fluids, we have analysed the effect that a polarizing field has on the ratio of f_{max}/f_{res} at constant particle concentration. Furthermore, by changing the particle concentration, n , of the magnetic fluid we have been able to observe and report on the effect that interparticle interaction has on the ratio of f_{max}/f_{res} . For this purpose, room-temperature measurements of the complex magnetic susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, were obtained for a number of magnetic fluid samples with different values of particle concentration over the frequency and polarizing field ranges of 100 MHz–6 GHz and 0–1.3 kOe, respectively.

2. Theoretical considerations

The usual approach for the theoretical description of the magnetic resonance of systems consisting of single-domain particles (such as magnetic fluids) is based on the analysis of the free magnetic energy per unit volume of a representative particle [5, 6]. In order to calculate the susceptibility of the magnetic fluid, the susceptibility of the representative particle has to be averaged over all particle sizes, over all orientations of the anisotropy axes and over all orientations of the dipolar field [5, 6].

In [6], starting from the equation of motion of the magnetization vector of the representative particle in the Landau–Lifshitz form, the resonance condition and the line width are calculated. As the Landau–Lifshitz equation can be written for every magnetic system, starting from the equation of motion of the magnetization vector, \vec{M} , of the magnetic fluid (equation (1)),

$$\frac{d\vec{M}}{dt} = -g\gamma(\vec{M} \times \vec{H}) - \alpha \frac{g\gamma}{M} [\vec{M} \times (\vec{M} \times \vec{H})] \quad (1)$$

the resonance condition (equation (2)) and the line width (equation (3)) of the magnetic fluid will have the same form as in [6]:

$$\omega_0 = \frac{g\gamma(1 + \alpha^2)^{1/2}}{M \sin \theta_0} (F_{\theta\theta} F_{\varphi\varphi} - F_{\varphi\theta}^2)^{1/2} \quad (2)$$

$$\Delta\omega = \frac{g\gamma\alpha}{M} \left(F_{\theta\theta} + \frac{F_{\varphi\varphi}}{\sin^2 \theta_0} \right). \quad (3)$$

In equations (1)–(3), \vec{H} is the effective magnetic field, g is the spectroscopic splitting factor, γ is the gyromagnetic electronic ratio, and α is the damping parameter of the magnetic fluid. Also, φ and θ are the angular coordinates of the magnetization, while $F_{\theta\theta}$, $F_{\varphi\varphi}$ and $F_{\varphi\theta}$ are the second derivatives of the free energy per unit volume of the magnetic fluid at the equilibrium position of its magnetization (φ_0, θ_0) , where F has a minimum.

The real and imaginary components of the frequency-dependent complex magnetic susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of magnetic fluids are shown in appendix A to have the following form:

$$\chi' = \frac{A}{(\omega_0^2 - \omega^2)^2 + \omega^2 D^2} [(\omega_0^2 - \omega^2)B + \omega^2 DC] \quad (4)$$

$$\chi'' = \frac{A\omega}{(\omega_0^2 - \omega^2)^2 + \omega^2 D^2} [DB - (\omega_0^2 - \omega^2)C]. \tag{5}$$

From equation (4), the frequency, f_{res} , at which $\chi'(\omega)$ becomes zero (i.e. the resonant frequency) is found to be:

$$f_{res} = f_0 \left(\frac{B}{B - DC} \right)^{1/2}. \tag{6}$$

In equation (6) D is a positive number (because it is the line width, $\Delta\omega$) and C is also a positive number (see appendix A). Because χ'' has to be positive for all frequencies, it follows from equation (5) that B is also a positive number (otherwise $\chi'' < 0$ when $\omega < \omega_0$ in equation (5)). Consequently, equation (6) shows that the resonant frequency, f_{res} , is always (in the real cases) larger than f_0 . Since D and C are directly proportional to the damping parameter, α , and B is a positive number, equation (6) shows that, for a magnetic fluid, f_0 and f_{res} are equal only for the case of pure resonance (i.e. $\alpha = 0$).

It is shown in appendix A that

$$\frac{d\chi''}{d\omega}(\omega_{res}) = \frac{A(BCD^2 + C^2D\omega_0^2 - B^2D - 2BC\omega_0^2)(CD - B)}{D^2(BCD - B^2 - C^2\omega_0^2)\omega_0^2}. \tag{7}$$

It follows that, if f_{res} is equal to the maximum absorption frequency, f_{max} , then the derivative (7) must be zero. Therefore, from the associated equation of relation (7), it is found that the frequency corresponding to the resonance condition at which the derivative (7) is zero is

$$\omega_0 = \frac{\pm 1}{C(DC - 2B)} \sqrt{-C(DC - 2B)DB(DC - B)}. \tag{8}$$

From equation (6) it follows that $B - DC > 0$ (otherwise f_{res} will be an imaginary number). As B, C and D are positive numbers and $B > DC$, from equation (8) ω_0 is found to be an imaginary number. This result is obviously absurd, and therefore one deduces that the derivative, $d\chi''/d(\omega)$, calculated at $\omega = \omega_{res}$ cannot be zero. Consequently, we can assert that f_{res} is not equal to f_{max} . This result concurs with that reported by Scaife [3], who used an alternative theoretical approach.

In the following section we present an experimental analysis concerning:

- (i) the polarizing field effect on the ratio f_{max}/f_{res} , at constant particle concentration, and
- (ii) the effect of interparticle interaction on the ratio f_{max}/f_{res} in zero polarizing field.

3. Samples and experimental details

The magnetic fluid samples used in this study were magnetic fluids with magnetite particles dispersed in kerosene and stabilized with oleic acid. The colloidal particles of magnetite were obtained by chemical co-precipitation of Fe^{2+} and Fe^{3+} ions in aqueous solution with an excess of NH_3 . The stabilization of magnetite particles was done by hydrofobization with technical oleic acid in the absence of a dispersion medium [7]. Finally, the magnetic fluid was filtered in a magnetic field gradient in the presence of magnetic wool, consisting of steel wires of approximately 17 μm in diameter, which was placed in the filter vessel [8]. The resulting sample was denoted sample A. From the initial magnetic fluid (sample A), four other samples—namely A1, A2, A3 and A4—were obtained by successive dilution of each sample with kerosene, with a dilution ratio of 2:3.

The saturation magnetization of the initial magnetic fluid was $M_\infty(A) = 46.3$ magnetic moment cm^{-3} (i.e. 46.3 kA m^{-1}), measured using the ballistic method. Particle concentration

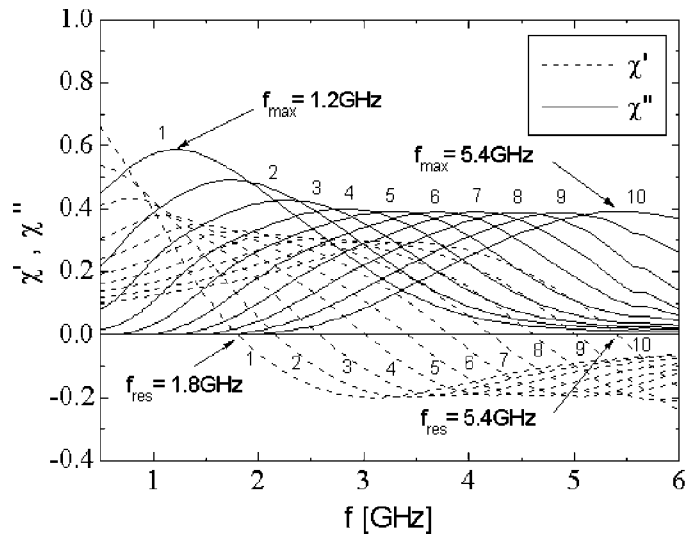


Figure 1. A plot of χ' and χ'' (normalized to the value of χ' at 0.1 GHz corresponding to zero polarizing field) against frequency, for ten values of polarizing field, for the initial sample (sample A): $H_1 = 0$, $H_2 = 191.4$ Oe, $H_3 = 307.5$ Oe, $H_4 = 443.3$ Oe, $H_5 = 580.3$ Oe, $H_6 = 717.2$ Oe, $H_7 = 860.8$ Oe, $H_8 = 997$ Oe, $H_9 = 1.14$ kOe, $H_{10} = 1.29$ kOe.

within the initial magnetic fluid, n , and mean particle magnetic diameter, D_m , were determined using the Chantrell method [9], resulting in $n(A) = 3 \times 10^{17} \text{ cm}^{-3}$ and $D_m = 8.5$ nm.

The complex magnetic susceptibility measurements, over the frequency range 100 MHz–6 GHz, were made using the short-circuited coaxial transmission line method [10], at room temperature. The short-circuited coaxial transmission line that contained the magnetic fluid sample was placed between the pole faces of an electromagnet, with the axis of the coaxial line perpendicular to the field. Automatically swept measurements over the entire frequency range of the input impedance of the line containing the sample were measured using a Hewlett Packard network analyser (HP 8753 C) and, from these measurements, the components $\chi'(\omega)$ and $\chi''(\omega)$ of the complex magnetic susceptibility of the samples were determined.

4. Results and discussions

The results of the frequency-dependent complex magnetic susceptibility, at different values of the polarizing field, for sample A are presented in figure 1. Magnetic resonance is observed for each value of polarizing field, indicated by the transition of the real part of the complex magnetic susceptibility from a positive to a negative value. As is shown in figure 1, f_{res} increases from 1.8 to 5.4 GHz in response to an increase in the polarizing field from 0 to 1.29 kOe.

The frequency of maximum absorption at resonance, f_{max} , also increases from 1.2 to 5.4 GHz by increasing the polarizing field from 0 to 1.29 kOe, as we can observe from figure 1.

The dependence of the ratio of f_{max}/f_{res} as a function of the external magnetic field, H_{pol} , is plotted in figure 2 for sample A. As can be observed, the ratio f_{max}/f_{res} is different from unity—a result that is in agreement with our theoretical considerations as well as with those of Scaife [3]. An increase in polarizing field leads to an increase in the ratio of f_{max}/f_{res} , with the ratio approaching unity in the case of the strongest polarizing field (see figure 2).

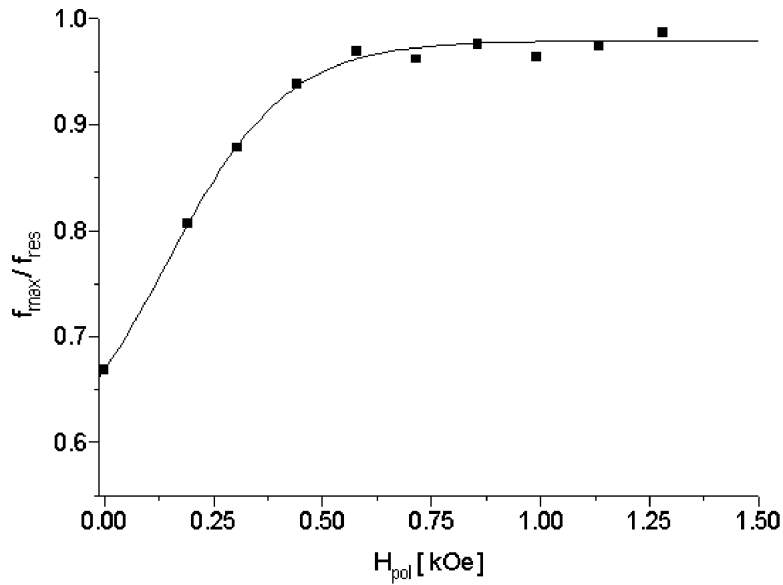


Figure 2. The dependence of the ratio of the maximum absorption frequency, f_{max} , and the resonant frequency, f_{res} , on an external magnetic field, H_{pol} , for sample A. The full curve is an eye-guiding line having the equation $r(x) = a+b/(1+\exp(-(x-c)/d))$, with $a = 0.57$, $b = 0.39$, $c = 153.74$ and $d = 127.93$.

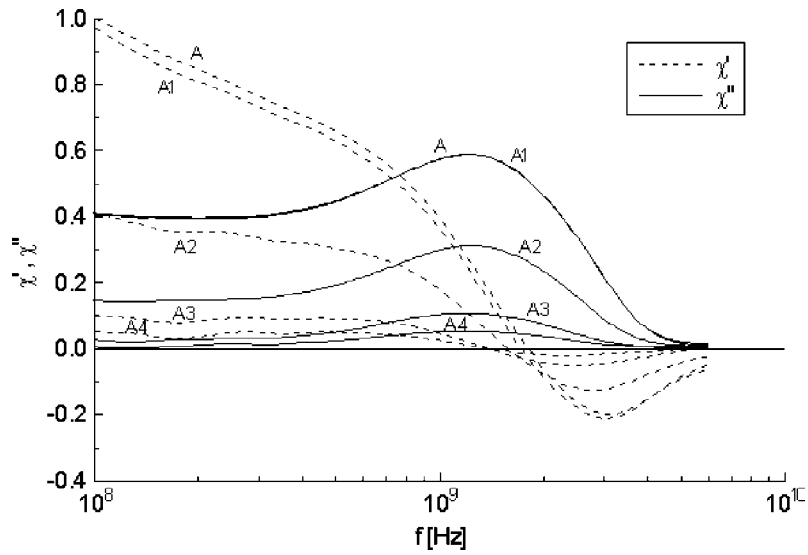


Figure 3. A plot of χ' and χ'' (normalized to the value of χ' at 0.1 GHz) against frequency, at zero polarizing field, for samples A, A1, A2, A3 and A4.

The effect of the interparticle interactions on the ratio f_{max}/f_{res} was analysed from the dependence of the ratio f_{max}/f_{res} on particle concentration, n , in zero polarizing field at room temperature. The particle concentration within the magnetic fluid sample was changed by successive dilution with kerosene, with a dilution ratio 2:3, starting from sample A. Figure 3 shows the results obtained for the frequency-dependent complex magnetic susceptibilities, χ'

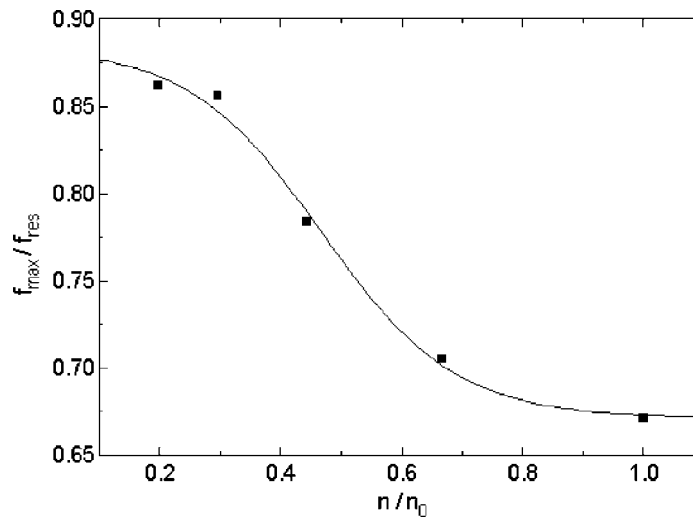


Figure 4. The plot of the ratio f_{max}/f_{res} against normalized particle concentration, n/n_0 , at zero polarizing field. n_0 is the initial particle concentration (of sample A) and n is the particle concentration of the diluted samples. The full curve is an eye-guiding line having the equation $r(x) = a + b/(1 + \exp(-(x - c)/d))$, with $a = 0.67$, $b = 0.21$, $c = 0.46$ and $d = -0.11$.

and χ'' , for all dilutions of sample A. All data from the plot are normalized to the value of χ' of sample A at 0.1 GHz.

As can be seen from figure 3, the resonant frequency, f_{res} , decreases with dilution from 1.81 GHz for the most concentrated sample (sample A) to 1.41 GHz for the most diluted sample (sample A4). We also note that the magnitude of χ' and χ'' decreases with dilution, as expected, and that f_{max} is approximately constant, at 1.2 GHz.

The plot of the ratio f_{max}/f_{res} as a function of particle concentration at zero polarizing field is presented in figure 4. It can be observed that the ratio f_{max}/f_{res} increases with decreasing particle concentration.

Any explanation of the dependence of the ratio f_{max}/f_{res} on an external magnetic field and on particle concentration must start from the structure of the magnetic fluids.

A magnetic fluid consists of single-domain particles, which obey a dimensional distribution and are dispersed in a carrier liquid. In zero magnetic field, the orientation of the uniaxial anisotropy axes of the particles is uniform while, with increasing biasing field, the anisotropy axes tend to align close to the direction of the magnetic field [11]. Also, an external magnetic field can induce particle agglomerations, resulting in a change in the local interparticle interaction field [12].

The resonance condition of a particle within the magnetic fluid depends on the particle axis orientation, on particle size [11], and on local interparticle interactions [12]. The measured resonance line comprises a great number of single particle lines, which have centres shifted relative to each other and which have different line widths. Consequently, for the case of a small polarizing field and high particle concentration (see figures 2 and 4), the superposition of the single particle resonance lines is possibly responsible for the large difference between f_{res} and f_{max} .

In a small polarizing field, the distribution of the anisotropy axes of the particles in a magnetic fluid is approximately uniform [11]. Also, we would expect a large spread in the values and in the orientations of the dipolar magnetic field acting on a particle due to all

others particles in the sample. This will result in a large spread in the values of the theoretical resonant frequency of a particle within the magnetic fluid, $f_{0,p}$, and in the values of line width of the particles (see equations (B.3) and (B.4) in appendix B). Therefore, the values of χ' and χ'' corresponding to the individual particles within the magnetic fluid will also have a large spread. This will probably lead to a large difference between f_{res} and f_{max} of the magnetic fluid. In the case where the polarizing field is large (relative to the interparticle interaction field and to the anisotropy field), the contribution of both the dipolar and anisotropy terms to the expressions for resonance and line width can be neglected. Consequently, the resonance condition (equation (B.5)) and the line width (equation (B.6)) will be the same for all particles. As a result, the resonance condition and the line width of one particle within the magnetic fluid will be the same as those of the magnetic fluid. In this case, $f_{res} \cong f_{max} = f_0$ (see appendix B). This result is in agreement with our experimental findings (see figure 2).

For the case of zero polarizing field and small interactions (by comparison with the anisotropy field), there exists a large spread in the values of frequency corresponding to the resonance condition of a particle, $f_{0,p}$, and in the values of line width of a particle within the magnetic fluid (see equations (B.13) and (B.14) in appendix B). Consequently, this will result in a large spread in the values of χ' and χ'' corresponding to the individual particles of the magnetic fluid and manifest itself in the form of a large difference between f_{res} and f_{max} . For the diluted magnetic fluids, the dipolar magnetic field acting on the particle due to all others particles in the sample can be neglected. Therefore the resonance condition (see appendix B) will be the same for all particles. In this case, the resonance condition and the line width of one particle will be the same as those of the magnetic fluid, and one obtains $f_{res} \cong f_{max} = f_0$ (see appendix B)—a result that is in agreement with our experimental findings (see figure 4).

5. Conclusions

Measurements of the complex magnetic susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, as a function of frequency, f , over the range 100 MHz–6 GHz and the external polarising field, H_{pol} , up to 1.3 kOe, are obtained by means of the coaxial transmission line technique. Ferromagnetic resonance, indicated by the $\chi'(\omega)$ component going from a positive to a negative quantity at a frequency f_{res} , is observed in the case of five magnetic fluid samples, namely samples A, A1, A2, A3 and A4. In the case of sample A, the variation in the polarizing field results in f_{res} covering the frequency range 1.8–5.5 GHz. The corresponding $\chi''(\omega)$ profiles have f_{max} values covering the frequency range 1.8–5.4 GHz.

From these dynamic measurements we investigate the factors which influence the ratio between the maximum absorption frequency, f_{max} , and the resonance frequency, f_{res} . These experimental results show that the ratio f_{max}/f_{res} is always less than unity, thereby confirming the deductions of Scaife.

It is further shown how the ratio f_{max}/f_{res} increases, approaching unity, with (i) an increase in polarizing field, H_{pol} , and (ii) a decrease in interparticle interactions; the interparticle interactions being reduced by means of changing the particle concentration of the magnetic fluid samples.

It is also demonstrated that, for the case of an unpolarized magnetic fluid sample, the measured resonant frequency, f_{res} , and the resonant frequency of the Landau–Lifshitz equation, f_0 , are always different, with $f_{res} > f_0$. The exception is the case of pure resonance (i.e. null damping parameter of the Landau–Lifshitz equation). Again, the influence that the polarizing field and/or interparticle interactions have on f_{res} is investigated theoretically and it is proven that, under conditions of strong polarizing field and/or small interparticle interactions, $f_{res} \cong f_0$.

Appendix A. The behaviour of f_0 , f_{max} and f_{res} in magnetic fluids in the general case

The susceptibility of the magnetic fluid χ will have the following form, as in [6]:

$$\chi = \frac{\gamma^2 g^2 (1 + \alpha^2)}{\omega_0^2 - \omega^2 + i\omega \Delta\omega} \left[l^2 \left(F_{\theta\theta} + \frac{i\omega\alpha M}{\gamma g(1 + \alpha^2)} \right) + s^2 \left(\frac{F_{\varphi\varphi}}{\sin^2 \theta_0} + \frac{i\omega\alpha M}{g\gamma(1 + \alpha^2)} \right) + 2ls \frac{F_{\theta\varphi}}{\sin \theta_0} \right]. \quad (\text{A.1})$$

The parameters l and s are:

$$l = \sin \delta \sin(\varphi_0 - \lambda) \quad (\text{A.2})$$

$$s = \cos \theta_0 \sin \delta \cos(\varphi_0 - \lambda) - \cos \delta \sin \theta_0. \quad (\text{A.3})$$

In the above equations δ and λ are the angular coordinates of the microwave magnetic field. To separate the real and imaginary parts of the complex magnetic susceptibility, the expression of the complex susceptibility is multiplied by $\omega_0^2 - \omega^2 - i\omega\Delta\omega$, resulting in:

$$\chi = \frac{g^2 \gamma^2 (1 + \alpha^2) (\omega_0^2 - \omega^2 - i\omega\Delta\omega)}{(\omega_0^2 - \omega^2)^2 + \omega^2 (\Delta\omega)^2} \left[l^2 \left(F_{\theta\theta} + \frac{i\omega\alpha M}{\gamma g(1 + \alpha^2)} \right) + s^2 \left(\frac{F_{\varphi\varphi}}{\sin^2 \theta_0} + \frac{i\omega\alpha M}{g\gamma(1 + \alpha^2)} \right) + 2ls \frac{F_{\theta\varphi}}{\sin \theta_0} \right]. \quad (\text{A.4})$$

Expanding expression (A.4), one obtains

$$\chi = \frac{g^2 \gamma^2 (1 + \alpha^2)}{(\omega_0^2 - \omega^2)^2 + \omega^2 (\Delta\omega)^2} \left[(\omega_0^2 - \omega^2) l^2 F_{\theta\theta} + (\omega_0^2 - \omega^2) l^2 \frac{i\omega\alpha M}{g\gamma(1 + \alpha^2)} + (\omega_0^2 - \omega^2) s^2 \frac{F_{\varphi\varphi}}{\sin^2 \theta_0} + (\omega_0^2 - \omega^2) s^2 \frac{i\omega\alpha M}{g\gamma(1 + \alpha^2)} + 2ls (\omega_0^2 - \omega^2) \frac{F_{\theta\varphi}}{\sin \theta_0} - i\omega \Delta\omega l^2 F_{\theta\theta} + \Delta\omega l^2 \frac{\omega^2 \alpha M}{g\gamma(1 + \alpha^2)} - i\omega \Delta\omega s^2 \frac{F_{\varphi\varphi}}{\sin^2 \theta_0} + \Delta\omega s^2 \frac{\omega^2 \alpha M}{g\gamma(1 + \alpha^2)} - 2i\omega \Delta\omega ls \frac{F_{\theta\varphi}}{\sin \theta_0} \right]. \quad (\text{A.5})$$

Separating equation (A.5) into its real and imaginary components, one obtains

$$\chi' = \frac{g^2 \gamma^2 (1 + \alpha^2)}{(\omega_0^2 - \omega^2)^2 + \omega^2 (\Delta\omega)^2} \left[(\omega_0^2 - \omega^2) \left(l^2 F_{\theta\theta} + \frac{s^2 F_{\varphi\varphi}}{\sin^2 \theta_0} + \frac{2ls F_{\theta\varphi}}{\sin \theta_0} \right) + \frac{\omega^2 \Delta\omega \alpha M}{g\gamma(1 + \alpha^2)} (l^2 + s^2) \right] \quad (\text{A.6})$$

$$\chi'' = \frac{g^2 \gamma^2 (1 + \alpha^2)}{(\omega_0^2 - \omega^2)^2 + \omega^2 (\Delta\omega)^2} \left[\omega \Delta\omega \left(l^2 F_{\theta\theta} + \frac{s^2 F_{\varphi\varphi}}{\sin^2 \theta_0} + 2ls \frac{F_{\theta\varphi}}{\sin \theta_0} \right) - \frac{\omega \alpha M (\omega_0^2 - \omega^2)}{g\gamma(1 + \alpha^2)} (l^2 + s^2) \right]. \quad (\text{A.7})$$

To simplify matters, the following notation is used:

$$A = g^2 \gamma^2 (1 + \alpha^2) \quad (\text{A.8})$$

$$B = l^2 F_{\theta\theta} + s^2 \frac{F_{\varphi\varphi}}{\sin^2 \theta_0} + 2ls \frac{F_{\theta\varphi}}{\sin \theta_0} \quad (\text{A.9})$$

$$C = \frac{\alpha M}{g\gamma(1 + \alpha^2)} (l^2 + s^2) \quad (\text{A.10})$$

and

$$D = \Delta\omega \tag{A.11}$$

resulting in $\chi'(\omega)$ and $\chi''(\omega)$ being expressed in the form

$$\chi' = \frac{A}{(\omega_0^2 - \omega^2)^2 + \omega^2 D^2} [(\omega_0^2 - \omega^2)B + \omega^2 DC] \tag{A.12}$$

$$\chi'' = \frac{A\omega}{(\omega_0^2 - \omega^2)^2 + \omega^2 D^2} [DB - (\omega_0^2 - \omega^2)C]. \tag{A.13}$$

From (A.12) one obtains

$$f_{res} = f_0 \left(\frac{B}{B - DC} \right)^{1/2}. \tag{A.14}$$

The first derivative of χ'' with respect to ω is

$$\frac{d\chi''}{d\omega} = \frac{A}{[(\omega_0^2 - \omega^2)^2 + \omega^2 D^2]^2} \{ [DB + 2\omega^2 C - (\omega_0^2 - \omega^2)C][(\omega_0^2 - \omega^2)^2 + \omega^2 D^2] - [\omega DB - (\omega_0^2 - \omega^2)\omega C][2\omega D^2 - 4\omega(\omega_0^2 - \omega^2)] \} \tag{A.15}$$

and

$$\frac{d\chi''}{d\omega}(\omega_{res}) = \frac{A(BCD^2 + C^2 D\omega_0^2 - B^2 D - 2BC\omega_0^2)(CD - B)}{D^2(BCD - B^2 - C^2\omega_0^2)\omega_0^2}. \tag{A.16}$$

Expression (A.16) is zero when

$$BCD^2 + C^2 D\omega_0^2 - B^2 D - 2BC\omega_0^2 = 0 \tag{A.17}$$

resulting in

$$\omega_0 = \frac{\pm 1}{C(DC - 2B)} \sqrt{-C(DC - 2B)DB(DC - B)}. \tag{A.18}$$

Appendix B. The behaviour of f_0 , f_{max} and f_{res} in magnetic fluids: the case of strong polarizing field and small particle concentration

Assuming that the particle has uniaxial anisotropy, the free magnetic energy per unit volume of the particle within the magnetic fluid is

$$F = -M_S H_{pol}(\vec{e} \cdot \vec{e}_H) - M_S H_D(\vec{e} \cdot \vec{e}_D) - K(\vec{e} \cdot \vec{e}_A)^2. \tag{B.1}$$

In equation (B.1), e , e_H , e_D and e_A are the unit vectors that define the directions of the magnetization, M_S , the static polarizing magnetic field, H_{pol} , the dipolar magnetic field, H_D , and the anisotropy axis, respectively.

Under the assumption of small anisotropy field, $2K/(M_S H_{pol}) \ll 1$, and small interactions, $H_D/H_{pol} \ll 1$, the magnetization of the representative particle at the equilibrium position is approximately parallel to the polarizing field. Also, assuming that the static magnetic field is parallel to the y axis, after calculation of the free energy derivatives of the particle, $F_{\theta\theta}$, $F_{\varphi\varphi}$ and $F_{\varphi\theta}$, equation (2) written for the particle becomes

$$\omega_{0,p} = g\gamma(1 + \alpha_p^2)^{1/2} \left\{ \left[H_{pol} + H_D \sin \theta_D \sin \varphi_D - \frac{2K}{M_S}(\cos^2 \theta_A - \sin^2 \theta_A \sin^2 \varphi_A) \right] \times \left[H_{pol} + H_D \sin \theta_D \sin \varphi_D - \frac{2K}{M_S} \sin^2 \theta_A \cos(2\varphi_A) \right] - \frac{4K^2}{M_S^2} \sin^2 \theta_A \cos^2 \theta_A \cos^2 \varphi_A \right\}^{1/2}. \tag{B.2}$$

In the equation (B.2), (θ_D, φ_D) and (θ_A, φ_A) are, respectively, the angular co-ordinates of the dipolar magnetic field and of the unit vector, which define the direction of the anisotropy axis. Also, in equation (B.2), α_p is the damping parameter of the particle. For the case of small anisotropy field and small interactions, the terms $(H_D/H_{pol})^2$, $2KH_D/(M_S H_{pol}^2)$ and $(2K/H_{pol}M_S)^2$ can be neglected and, using the approximation $(1+x)^{1/2} \cong 1+x/2$ (for $x < 0.5$), equation (B.2) becomes

$$\omega_{0,p} = g\gamma(1+\alpha_p^2)^{1/2} \left[H_{pol} + H_D \sin \theta_D \sin \varphi_D + \frac{K}{M_S} (2 \sin^2 \theta_A \sin^2 \varphi_A - \cos^2 \theta_A - \cos^2 \varphi_A \sin^2 \theta_A) \right]. \quad (\text{B.3})$$

Using the same approximations, the line width of the particle is

$$\Delta\omega_p = 2g\gamma\alpha_p \left[H_{pol} + H_D \sin \theta_D \sin \varphi_D + \frac{K}{M_S} (2 \sin^2 \theta_A \sin^2 \varphi_A - \cos^2 \theta_A - \cos^2 \varphi_A \sin^2 \theta_A) \right]. \quad (\text{B.4})$$

For the case in which the dipolar term and the anisotropy term can be neglected, equations (B.3) and (B.4) become

$$\omega_{0,p} \cong g\gamma(1+\alpha_p^2)^{1/2} H_{pol} \quad (\text{B.5})$$

$$\Delta\omega_p \cong 2g\gamma\alpha_p H_{pol}. \quad (\text{B.6})$$

Also, the derivatives of the free magnetic energy per unit volume of the particle become $F_{\theta\theta} = F_{\varphi\varphi} = M_S H_{pol}$ and $F_{\theta\varphi} = 0$. Introducing these expressions into (A.6), the frequency at which χ' becomes zero will be

$$f_{res,p} = f_{0,p} \sqrt{\frac{1+\alpha_p^2}{1-\alpha_p^2}}. \quad (\text{B.7})$$

As is known, for the majority of the ferro-ferrimagnetic materials $\alpha_p \approx 10^{-2}$ [13, 14] therefore, for a particle within the magnetic fluid in a strong polarizing field ($2K/(M_S H_{pol}) \ll 1$ and $H_D/H_{pol} \ll 1$), one obtains for a particle, $f_{res,p} \cong f_{0,p}$. In the case of a strong polarizing field, the resonance condition (equation (B.5)) and the line width (equation (B.6)) will be the same for all particles. Consequently, the resonance condition and the line width of one particle within the magnetic fluid will be the same as those of the magnetic fluid. In this case, equation (B.7) can also be written for the magnetic fluid, following that for a magnetic fluid in a strong polarizing field, $f_{res} \cong f_0$.

From equation (A.15), the first derivative with respect to ω of the imaginary part of the complex magnetic susceptibility, χ'' , in ω_0 results in

$$\frac{d\chi''}{d\omega}(\omega_{0,p}) = \frac{A}{\omega_{0,p}^2 D^2} [2\omega_{0,p}^2 C - DB]. \quad (\text{B.8})$$

For the case of a strong polarizing field, the constant B defined in equation (A.9) becomes

$$B = M_S H_{pol} (l^2 + s^2). \quad (\text{B.9})$$

Inserting (A.10), (A.11) and (B.9) into (B.8), one obtains:

$$\frac{d\chi''}{d\omega}(\omega_{0,p}) = \frac{A(l^2 + s^2)M_S}{\omega_{0,p}^2 D^2} \left[2\omega_{0,p}^2 \frac{\alpha_p}{g\gamma(1+\alpha_p^2)} - \Delta\omega_p H_{pol} \right]. \quad (\text{B.10})$$

Using equation (B.5) and (B.6) in (B.10), results in:

$$\frac{d\chi''}{d\omega}(\omega_{0,p}) = 0. \tag{B.11}$$

This means that in the case of strong polarizing field, for each particle within the magnetic fluid, the frequency corresponding to the resonance condition, $f_{0,p}$, is equal to the maximum absorption frequency at resonance, $f_{max,p}$. Also, for the magnetic fluid, the frequency corresponding to the resonance condition, f_0 , is equal to the maximum absorption frequency at resonance, f_{max} .

In zero polarizing fields, the density of the free magnetic energy of the particle is:

$$F = -M_S H_D (\vec{e} \cdot \vec{e}_D) - K (\vec{e} \cdot \vec{e}_A)^2. \tag{B.12}$$

Under the assumption of small interactions by comparison with the anisotropy field, the magnetization of the particle at the equilibrium position is approximately parallel to the anisotropy axis. Therefore, from equations (2) and (3) written for the magnetic particle, using equation (B.12) in the calculation of $F_{\theta\theta}$, $F_{\varphi\varphi}$, and $F_{\theta\varphi}$ one obtains:

$$\omega_{0,p} = g\gamma(1 + \alpha_p^2)^{1/2} \left(\frac{2K}{M_S} + H_D \sin \theta_D \sin \varphi_D \right) \tag{B.13}$$

and

$$\Delta\omega_p = 2g\gamma\alpha_p \left(\frac{2K}{M_S} + H_D \sin \theta_D \sin \varphi_D \right). \tag{B.14}$$

In the case of a diluted magnetic fluid, $F_{\theta\theta} = 2K$, $F_{\varphi\varphi} = 2K \sin^2 \theta_A$ and $F_{\theta\varphi} = 0$. Equations (B.13) and (B.14) become:

$$\omega_{0,p} = g\gamma(1 + \alpha_p^2)^{1/2} \frac{2K}{M_S} \tag{B.15}$$

$$\Delta\omega_p = 4g\gamma\alpha_p \frac{K}{M_S}. \tag{B.16}$$

Substituting the expressions for the derivatives of the density of free magnetic energy into equation (A.6), it becomes equal to equation (B.7). As a result of small particle concentration, the resonance condition (equation (B.15)) and the line width (equation (B.16)) will be the same for all particles. Consequently, the resonance condition and the line width of one particle within the magnetic fluid will be the same as those of the magnetic fluid. In this case, equation (B.7) can be written also for the magnetic fluid, following that for a diluted magnetic fluid $f_{res} \cong f_0$.

In diluted magnetic fluids, the constant B defined in equation (A.9) becomes:

$$B = 2K(l^2 + s^2). \tag{B.17}$$

Inserting (A.10), (A.11) and (B.17) in (B.8), one obtains:

$$\frac{d\chi''}{d\omega}(\omega_{0,p}) = \frac{A(l^2 + s^2)}{\omega_{0,p}^2 D^2} \left[2\omega_{0,p}^2 \frac{\alpha_p M_S}{g\gamma(1 + \alpha_p^2)} - 2K \Delta\omega_p \right]. \tag{B.18}$$

Using equation (B.15) and (B.16) in (B.18), one obtains:

$$\frac{d\chi''}{d\omega}(\omega_{0,p}) = 0. \tag{B.19}$$

This shows that in the case of a diluted magnetic fluid, the frequency corresponding to the resonance condition of a particle within the magnetic fluid, $f_{0,p}$, is equal to the maximum absorption frequency of the particle, $f_{max,p}$. Furthermore, for the magnetic fluid having small particle concentration, the frequency corresponding to the resonance condition, f_0 , is equal to the maximum absorption frequency at resonance, f_{max} .

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