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ON THE RATIO BETWEEN THE MAXIMUM ABSORPTION FREQUENCY AND THE RESONANCE FREQUENCY OF COMPOSITE MAGNETIC FLUIDS*

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Abstract
The magnetic fluid samples investigated in this study were three kerosene-based magnetic fluids, stabilised with oleic acid, denoted as F1, F2 and F3. Sample F1 was a magnetic fluid with $Mn_{0.6}Fe_{0.4}Fe_2O_4$ particles, sample F2 was a magnetic fluid with $Ni_{0.4}Zn_{0.6}Fe_2O_4$ particles and sample F3 was a composite magnetic fluid obtained by mixing a part of sample F1 with a part of sample F2, in proportion of 1:1. Starting from the Landau-Lifshitz equation, it is demonstrated that in the case of a magnetic fluid, the measured resonance frequency, f_{res} is always different the corresponding maximum absorption frequency, f_{max} and the ratio, f_{max} / f_{res}, increases approaching unity with increase of polarizing field, H_{pol} . Measurements of the frequency dependent, complex magnetic susceptibility, $\chi(\omega) = \chi^{'}(\omega) - i\chi^{''}(\omega)$, in the GHz range, are used to investigate the effect which the mixing of two different magnetic fluids has on the dependence of the ratio f_{max} / f_{res} on an external strong polarizing magnetic field.

***Keywords*:** Magnetic fluid; Complex magnetic susceptibility; Ferrimagnetic resonance; Composite materials.

1. Introduction

Magnetic fluids are colloidal systems consisting of single-domain magnetic particles dispersed in a carrier liquid and are convenient model systems to explore fundamental properties of magnetic nanoparticle systems [1].

Taking into account the thermal fluctuations of the magnetic moment of the particles and neglecting interparticle interactions, Scaife has shown theoretically [2] that for a magnetic fluid the resonance frequency, f_{res}, and the frequency of maximum absorption, f_{max}, are always different except for the case of pure resonance.

Here we use an alternative theoretical approach to that of Scaife and we demonstrate that for a magnetic fluid the resonance frequency, f_{res}, is always different from the theoretical

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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.

In this paper, we have analyzed the effect which the mixing of two different magnetic fluids has on the dependence of the ratio f_{max} / f_{res} on an external strong polarizing magnetic field, H.

2. Theoretical considerations

In order to calculate the susceptibility of the magnetic fluid, the susceptibility of a 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 [3], [4]. In Ref. [4], starting from the equation of motion of the magnetization vector of the representative particle in the Landau-Lifshitz form, the complex magnetic susceptibility of a representative particle within the magnetic fluid is:

$$\chi = \frac{g^2\gamma^2\left(1+\alpha^2\right)}{\left(\omega_0^2-\omega^2\right)^2+\omega^2\Delta\omega^2}\left\{\left[\left(\omega_o^2-\omega^2\right)A+\omega^2\Delta\omega B\right]-i\omega\left[\Delta\omega A-\left(\omega_o^2-\omega^2\right)B\right]\right\} \quad (1)$$

where $\omega_0 = \frac{g\gamma\left(1+\alpha^2\right)^{1/2}}{M\sin\theta_0}\left(F_{\theta\theta}F_{\varphi\varphi}-F_{\varphi\theta}^2\right)^{1/2}$, is the theoretical resonance condition, $\Delta\omega = \frac{g\gamma\alpha}{M}\left(F_{\theta\theta}+\frac{F_{\varphi\varphi}}{\sin^2\theta_0}\right)$ is the frequency line width, $A = l^2F_{\theta\theta}+s^2\frac{F_{\varphi\varphi}}{\sin^2\theta_0}+2ls\frac{F_{\theta\varphi}}{\sin\theta_0}$, $B=\frac{\alpha M}{g\gamma\left(1+\alpha^2\right)}\left(l^2+s^2\right)$, $l=\sin\delta\sin\left(\varphi_0-\lambda\right)$, $s=\cos\theta_0\sin\delta\cos\left(\varphi_0-\lambda\right)-\cos\delta\sin\theta_0$ (δ and λ being the angular coordinates of the microwave magnetic field). Also, g is the spectroscopic splitting factor, γ is the gyromagnetic electronic ratio, (φ , θ) are the angular coordinates of the magnetization whilst $F_{\theta\theta}$, $F_{\varphi\varphi}$ and $F_{\theta\varphi}$ 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.

From Eq. (1), the frequency, f_{res}, at which $\chi'(\omega)$ becomes zero (i.e. the resonance frequency) is given by Eq. (2). In Eq. (2) the line width $\Delta\omega$ is a positive number and B is also a positive number. Because χ'' has to be positive for all frequencies, it follows from Eq. (1) that A is also a positive number (otherwise $\chi''< 0$ when $\omega<\omega_0$ in Eq. (1)). Consequently, Eq. (2) shows that the resonance frequency, f_{res} is always larger than f_0 . Since $\Delta\omega$ and B are directly proportional to the damping parameter, α, and as A is a positive number, Eq. (2)

shows that for a magnetic fluid f_0 and f_{res} are equal only for the case of pure resonance (i.e. $\alpha=0$).

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

In case of uniaxial anisotropy of particles, under the assumption of small anisotropy field and small interactions, the resonance condition and the line width for the particles become:

$$\omega_{0,p} \cong g\gamma \left(1 + \alpha_p^2\right)^{1/2} H \tag{3}$$

$$\Delta\omega_p \cong 2g\gamma\alpha_p H \tag{4}$$

Also, the derivatives of the free magnetic energy per unit volume of the particle become $F_{\theta\theta} = F_{\varphi\varphi} = M_S H$ and $F_{\theta\varphi}=0$. Introducing these expressions into (1), the frequency at which χ' becomes zero will be:

$$f_{res,p} = f_{0,p} \sqrt{\frac{1+\alpha_p^2}{1-\alpha_p^2}} \tag{5}$$

As is known, for the majority of the ferro-ferrimagnetic materials $\alpha_p \approx 10^{-2}$ [5, 6] therefore for a particle within the magnetic fluid in a strong polarizing field, one obtains for a particle, $f_{res,p} \cong f_{0,p}$. In the case of a strong polarizing field, the resonance condition (Eq.(3)) and the line width (Eq.(4)) 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, Eq.(5) can be written also for the magnetic fluid, following that for a magnetic fluid in a strong polarizing field. From the first derivative on ω of the imaginary part of the complex magnetic susceptibility, χ'', in ω_0 results in, $\frac{d\chi''}{d\omega}\left(\omega_{0,p}\right) = 0$. 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}. Hence, Eq. (5) for a magnetic fluid in strong polarizing field, can be written:

$$f_{res} = f_{\max} \sqrt{\frac{1+\alpha^2}{1-\alpha^2}} \tag{6}$$

where α, represent the damping parameter for the magnetic fluid.

3. Samples and experimental

The magnetic fluid samples investigated in this study were three kerosene-based magnetic fluids denoted as F1, F2 and F3. Sample F1 was a magnetic fluid with $Mn_{0.6}Fe_{0.4}Fe_2O_4$ particles dispersed in kerosene and stabilised with oleic acid [7], with a saturation magnetisation, $M_\infty = 5.53\ kA/m$. Sample F2 was a magnetic fluid with $Ni_{0.4}Zn_{0.6}Fe_2O_4$ particles dispersed in kerosene and stabilised with oleic acid, with a saturation magnetisation, $M_\infty = 5.04\ kA/m$. Sample F3 was a composite magnetic fluid obtained by mixing a part of sample F1 with a part of sample F2, in proportion of 1:1.

The complex magnetic susceptibility measurements, over the frequency range of *500 MHz* to *6 GHz*, were made using the short-circuited coaxial transmission line method [8], at room temperature. The short-circuited coaxial transmission line containing the magnetic fluid sample was placed between the pole faces of an electromagnet, the axis of the coaxial line being perpendicular to the field. The measurements were made at different values of the polarising field, H, over the range *0* and *125 kA/m*.

4. Results and discussions

The results of the frequency dependent, complex magnetic susceptibility, at different values of the polarising field, for sample F1 are presented in Fig.1. These measurements indicate the presence of a magnetic resonance phenomenon for the investigated samples by demonstrating a transition of the values of χ' from a positive value to a negative value, at the resonance frequency, f_{res}. As is shown in Fig. 1, in response to increasing the polarising field from *0* to approximately *125 kA/m*, f_{res} increases from *1.20 GHz* to *5.51 GHz* and f_{max} also increases from *0.88 GHz* to *4.94 GHz*. Similar behaviour is observed for the other two magnetic fluid samples, where f_{res} increases from *0.95 GHz* to *5.20 GHz* (for sample F2) and from *1.10 GHz* to *5.77 GHz* (for sample F3) ; f_{max} also increases from *0.39 GHz* to *4.97 GHz* (for sample F2) and from *0.80 GHz* to *5.47 GHz* (for sample F3), by increasing the polarising field over the above mentioned range.

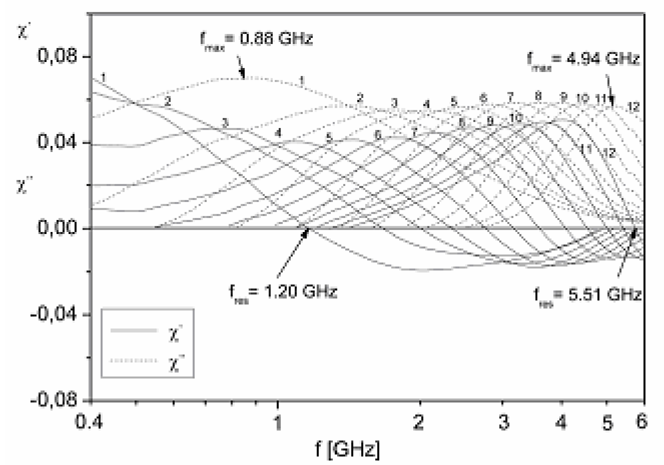

Fig.1. Plot of χ' and χ'' against frequency, for different values of polarising field, for the sample F1: $H_1= 0$, $H_2=15.23$ kA/m, $H_3=24.46$ kA/m, $H_4=35.28$ kA/m, $H_5=46.18$ kA/m, $H_6=57.07$ kA/m, $H_7=68.49$ kA/m, $H_8=79.33$ kA/m, $H_9=90.66$ kA/m, $H_{10}=102.40$ kA/m, $H_{11}=113.43$ kA/m, $H_{12}=124.47$ kA/m.

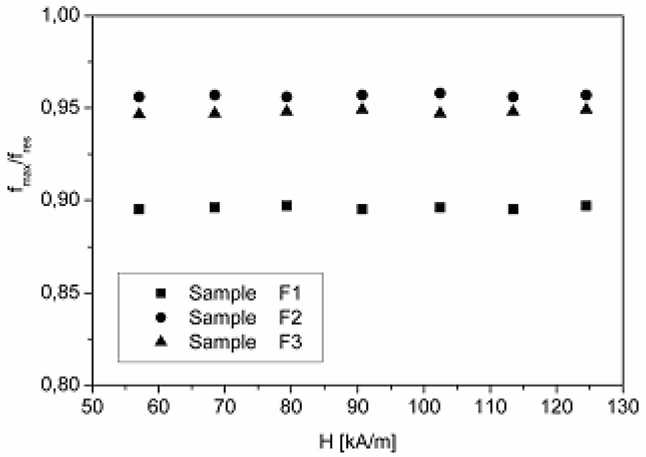

Fig.2. Plots of f_{max} / f_{res} against polarizing field, H, (H larger than $2H_A$) for the investigated samples.

In order to satisfy the condition of a strong polarizing field (i.e. H >> dipolar interaction field plus anisotropy field), we have determined the anisotropy field for the investigated samples from the linear fit of the experimental dependence of f_{res} on H [9]. The obtained values are: $H_A(F1) = 26.69$ kA/m, $H_A(F2) = 11.05$ kA/m, $H_A(F3) = 18.72$ kA/m.

From the complex magnetic susceptibility measurements, the ratio f_{max} / f_{res}, at different values of the polarizing field, H, was determined for each sample. The results are presented in Fig. 2 for values of H larger than $2H_A$, where the ratio f_{max} / f_{res} tends to a constant value, approaching unity in agreement with the theoretical considerations. As can be observed from Fig.2, the values of the ratio f_{max} / f_{res} of the composite sample (F3) are between the f_{max} / f_{res} values obtained for samples F1 and F2.

5. Conclusions

Measurements of the complex magnetic susceptibility, $\chi(\omega) = \chi'(\omega) - i\,\chi''(\omega)$, as a function of frequency, f, over the range *500MHz* to *6GHz*, and external polarising field, H, up to *1.25 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 the investigated samples.

From these dynamic measurements we investigate the effect which the mixing of two different magnetic fluids has on the dependence of the ratio f_{max} / f_{res} in an external strong polarizing magnetic field. The values of the ratio f_{max} / f_{res} of the composite sample (F3) are between the f_{max} / f_{res} values obtained for samples F1 and F2.

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