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ULTRASONIC ANISOTROPY IN MAGNETIC LIQUIDS

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Experimental results for magnetic field influence on the ultrasonic anisotropy in magnetic fluids are reported. The measurements have been carried out for the frequency 3.37 MHz, at the temperature 20°C and magnetic field of strength $B = 100$ mT, 200 mT and 500 mT. The results were analyzed in terms of Gotoh/Chung theory and Taketomi theory. The former was found to exhibit a lack of inner consistency while the latter gives reliable values of the physical parameters obtained by best-fit procedure.

1. Introduction

Magnetic liquids are homogeneous colloidal suspensions of ferromagnetic particles in a carrier liquid, such as water, mineral oil, or an organic compound [1, 2]. Typically, the diameter of the magnetic particles or grains in the suspension ranges from 50 to 100 Å. They are coated with a surface-active dispersive medium to avoid coagulation. However, even if atrraction between the magnetic particles is weak some coagulation dependent on the temperature, the external magnetic field, as well as their concentration cannot be avoided.

An external magnetic field acting on a magnetic liquid gives rise to a volume magnetic moment: simultaneously, the magnetization of the liquid undergoes a variation achieving saturation at a well defined value of magnetic field induction. The physical mechanism underlying the macroscopic magnetization of a magnetic liquid in an external magnetic field is related with the fact that the particles possess a rotational degree of freedom. Under the influence of the external magnetic field, by way of the electromagnetic interaction, a spatial moment appears in the liquid and orients the particle magnetic dipoles along the field direction.

Moreover, the magnetic particles in the magnetic liquid were observed to form chain-like structures due to their mutual magnetic interaction under an external magnetic field. The clusters arising under these conditions can contain more than 10^6 particles and attain a size of several micrometers [3, 4].

One of the most interesting features of magnetic liquids is their anisotropic behaviour: their physical properties exhibit directional effects of the magnetic field. One of the methods to study this anisotropy is by measuring the ultrasonic velocity and attenuation as a function of the magnetic field angle θ.

The anisotropy of sound velocity and attenuation in a magnetic fluid under an external magnetic field was studied by several authors both theoretically and experimentally [5-8]. However, there are still many problems that require elucidation. In this and subsequent articles we will report the results of our experimental studies of the ultrasonic properties of magnetic liquids and a comparison of the experimental findings with existing theories.

2. Theoretical background

The linear hydrodynamical theory of a magnetic liquid in a magnetic field is due to PARSONS [5], who dealt with magnetic fluids as nematic liquid crystals. Thus, he introduced a director n, i.e. a unit vector parallel to the local magnetization vector. Neglecting the spatial derivative terms of order higher than the first, he obtained the following expressions for the velocity and absorption coefficient of the ultrasonic wave in a magnetic liquid:

$$\frac{c-c_0}{c_0}=\frac{\omega\gamma_2^2}{8\rho c_0^2\gamma_1}\,\frac{\omega\tau[1-(\omega/\omega_c)^2]}{[1-(\omega/\omega_c)^2]^2+(\omega\tau)^2}\sin^2 2\theta, \tag{1}$$

$$\alpha=\frac{\omega^2\gamma_2}{8\rho c^3}\,\frac{[1-(\omega/\omega_c)^2]^2}{[1-(\omega/\omega_c)^2]^2+(\omega\tau)^2}\sin^2 2\theta, \tag{2}$$

where ρ is the magnetic liquid density, ω is the angular frequency of the sound wave, c is the sound velocity and ω_c is given by

$$\omega_c=\sqrt{m_0 H/I}, \tag{3}$$

with I – the moment of inertia density of the colloidal particles, m_0 – the average of the magnetization throughout the fluids, and τ is defined as

$$\tau=\gamma_1/(m_0 H). \tag{4}$$

The constants γ_1 and γ_2 can be expressed in terms of Leslie's coefficient appearing in the theory of liquid crystals [9]. Parson's theory states that the ultrasonic velocity and absorption coefficient are endowed with anisotropy dependent on the angle θ between the propagation direction of the wave and the direction of the magnetic field **H**. According to Parsons, this anisotropy is proportional to $\sin^2 2\theta$.

Experiment, however, failed to confirm Parsons' theory [6, 7]. Accordingly, GOTOH and CHUNG [7], on the basis of the work of TARAPOV [10] proceeded to derive a set of magnetohydrodynamical equations for magnetic liquids in an external DC magnetic field. Linearizing the equations, they obtained expressions for the velocity and attenuation of the sound wave. In a first approximation, their theory leads to the following expressions for the attenuation and velocity of the ultrasonic wave:

$$c = c_0\sqrt{\frac{1+a_1x}{1+a_2x}}, \tag{5}$$

$$\alpha = \alpha_0\sqrt{\frac{1+a_2x}{1+a_1x}}\,\frac{1+a_3x+a_4x^2}{(1+a_1x)^2}, \tag{6}$$

where $x = \sin^2\theta$. The coefficients a_1, a_2, a_3 and a_4 are in general highly complicated functions of the squared magnetic field strength $\mathbf{H}^2$ and various material constants characterizing the magnetic liquid. Their determination requires that the functions of state $p(\rho, s)$, $T(\rho, s)$ and $M(\rho, T. H)$ shall be available. Nonetheless, the Eqs. (5) and (6) permit a qualitative prediction of the behaviour of the ultrasonic velocity and attenuation as functions of the angle θ between the direction of **H** and the propagation direction of the wave. Analysis of the derivative

$$\frac{dc}{d\theta} = c_0\sin\theta\cos\theta(1+a_1x)^{-1/2}(1+a_2x)^{-3/2}(a_1-a_2) \tag{7}$$

shows that in the interval $0 \leqslant O \leqslant 90^0$ the $dc/d\theta$ undergoes no change in sign and vanishes at the boundaries. Thus, the velocity changes monotonically with θ and has extreme values at both ends of the above interval, whereas the answer to the question of whether we deal with a decrease or an increase of the velocity depends on the sign of the expression $a_1 - a_2$. Similarly, the analysis of the derivative

$$\frac{d\alpha}{d\theta} = \alpha_0\sin\theta\cos\theta(1+a_1x)^{-1/2}(1+a_4x)^{-7/2}(A_0+A_1x+A_2x^2), \tag{8}$$

where

$$A_0 = a_1+2a_2-5a_4,\quad A_2 = 3a_1a_2-4a_1a_4-3a_2a_4+4a_3,\quad A_2 = 5a_1a_3-2a_1a_2a_4-a_3a_4$$

shows that depending on the value of the coefficients a_j's, attenuation has two, one or no extreme points in the interval $0 \leqslant O \leqslant 90^0$. Obviously, the linear hydrodynamical theory of Parsons and the magnetohydrodynamical theory of Gotoh and Chung lead to completely different predictions concerning the anisotropy of the ultrasonic wave propagation velocity in magnetic liquids.

The equations of motion adopted by GOTOH and CHUNG [7] do not take into account internal freedoms of the fluid. However, magneto-optical experiments [3, 4] suggest that in the presence of an external magnetic field the magnetic particles form chain-likes clusters which remain in the sample even after removal of the field. With this in mind TAKETOMI [8]) developed a theory which attributes the anisotropy of the sound absorption coefficient to the two types of motion of the clusters: rational and translational. Using a special liquid crystal theory [11] he obtained the following expression for the sound attenuation arising from rotation of the magnetic particles:

$$\alpha_{\rm rot}=\frac{\omega^2}{2\rho c^3}\left(\frac{4}{3}\eta_s+\eta_b+2\alpha_5\cos^2\theta+\alpha_1\cos^4\theta\right), \tag{9}$$

where η_s, η_b are the shear and bulk viscosities respectively, and α_1, α_5 are Leslie's coefficients [9]. It can be noted that neglecting the third and fourth terms on the righthand side of Eq. [9] one obtains the sound absorption coefficient of ordinary fluids. In order to evaluate the translational motion contribution to the coefficient of absorption of the ultrasonic wave in a magnetic fluid Taketomi assumed the model of a vibrating sphere in viscous fluid. Calculating the dissipative energy per unit volume of the magnetic fluid, he obtained the following equation for the additive sound attenuation due to translational motion of the clusters:

$$\alpha_{\rm tra}=\frac{3\pi\eta_0 a\omega^3 VN(6\pi\eta_0+\rho_0 V\omega)/(k^2c)}{(\sin\theta-\rho_m V\omega^2/k^2)+(6\pi\eta_0 a\omega/k^2)}, \tag{10}$$

where k is the force constant, ρ_0, η_0 are the density and shear viscosity of the solvent, N is the number density of the clusters and ρ_m, V, a are the density, volume and radius of the cluster respectively. Qualitatively, the sum of Eqs. (9) and (10) gives predictions similar to the theory of Gotoh and Chung (two extrema in the interval $0 \leqslant \theta \leqslant 90^0$) but is more useful since it enables the assessment of some physical parameters characterizing magnetic fluids such as the number density and radious of the clusters.

3. Experimental technique

Measurements of absorption and velocity were carried out using the Matec pulse-echo technique (Fig. 1). The radio-frequency gated amplifier model 755 and gating modulator model 7700 were used to drive the piezoceramic transducer. The ultrasonic pulse, on traversal of the sample, was detected by the receiver transducer and amplified in a wide-band amplifier. The resulting pulse-echo train was observed on a CRT display. The velocity of the ultrasonic wave can be evaluated from the expression

$$c=2lf, \tag{11}$$

where l is the distance between the transducers and f is the inverse of the double round trip time in the sample.

The model 2460B Automatic Attenuation Recorder measures the logarithmic difference (in dB) between two selected echos, say A and B, if the time gates correctly cover the main portion of the echoes. When A and B are two consecutive echoes the log(A/B) output is proportional to the absorption coefficient of the sample. The variations of log(A/B), e.g. as a function of the angle θ, give direct changes in the absorption coefficient. The accuracy of our determination of the velocity was of the order of 0.05% whereas changes in absorption coefficient were measured with error less than 1%. The absolute values of the absorption coefficient were accurate within $\pm$ 5%.

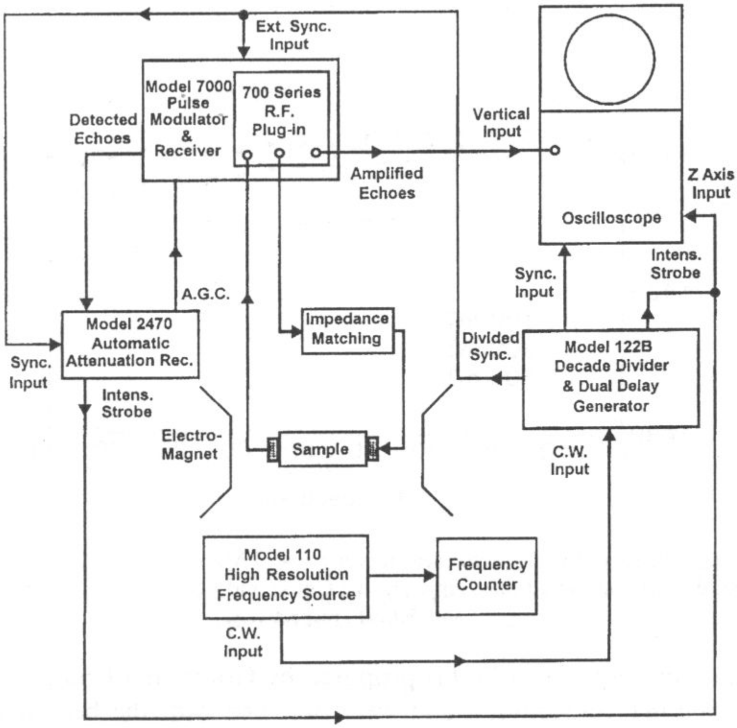

Fig. 1. Block diagram of the experimental setup.

The ultrasonic cell used in our measurements was made of brass. For the angular dependence experiment the magnetic field was rotated by five degrees each time while the measuring cell remained stationary in the gap between the electromagnetic pole pieces. The magnetic field induction was measured to within 0.5% with a F.W. Bell Gaussmeter model 9200.

4. Results and analysis

Our measurements were carried out in a magnetic liquid denoted as EMG-605 (produced by Ferrofluidics Inc.) consisting of magnetite particles Fe_3O_4 suspended in water. The values of saturation magnetization, initial susceptibility, volume concentration and viscosity were 20 mT, 0.5, 3.5% and $< 0.5\ N \cdot m^{-2}$ (at 25°C), respectively. The particle distribution was a normal distribution with a maximum at 100 Å The measurements were carried out in 20°C at 4.37 MHz and in three magnetic fields B, namely 100 mT, 200 mT and 500 mT.

The angular dependence of ultrasonic velocity for the three values of B is shown in Fig. 2. Without any numerical analysis it is obvious that the experimental data do not vary in the manner predicted by Parson's theory, i.e as $\sin^2 2\theta$. The solid, dashed

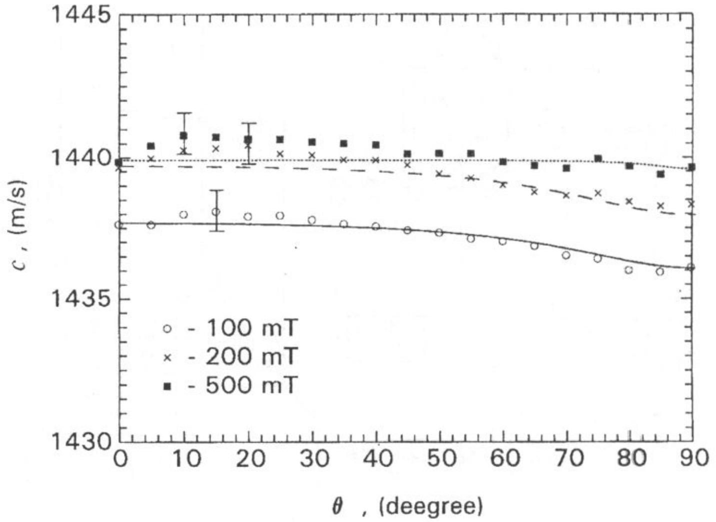

Fig. 2. Angular dependence of the ultrasonic velocity for $f = 4.37$ MHz in three magnetic field strengths. The solid, dashed and dotted lines represent the Gotoh and Chung theory for 100 mT, 200 mT and 500mT, respectively.

and dotted lines correspond to Eq.(5) proposed by Gotoh and Chung for 100 mT, 200 mT and 500 mT respectively, and were obtained with the best-fit procedure. Agreement between experiment and theory is fair, except for higher values of B for which the experimental points lie systematically above the theoretical curves. However, the data distribution does not decrease monotonically as predicted by Eq.(5) but exhibit two small extrema on both ends of the interval $0 \leqslant \theta \leqslant 90^0$.

Figure 3 shows the results for attenuatiuon, $\Delta\alpha = \alpha - \alpha_0(\theta = 0)$, *versus* θ for three values of B: 100 mT, 200 mT and 500 mT. Here, also, the experimental data do not match Parson's experssion given by Eq.(2). The lines drawn in Fig. 2 were obtained by fitting the attenuation data to Eq.(6) derived by Gotoh and Chung. Qualitatively, agreement between the experimental data and the theoretical lines is quite good though one aspect of this fitting requires clarification. Below we give the expressions used to draw the theoretical lines in Figs. 1 and 2:

$$\alpha = 28.8\sqrt{\frac{1+2.20x}{1+0.46x}\,\frac{1+0.17x+0.23x^2}{(1+0.46x)^2}}, \qquad \text{for 100 mT}$$

$$c = 1437.68\sqrt{\frac{1-0.82441x}{1-0.82399x}},$$

$$\alpha = 29.3\sqrt{\frac{1+3.19x}{1+1.63x}\,\frac{1+2.57x+1.53x^2}{(1+1.63x)^2}}, \qquad \text{for 200 mT}$$

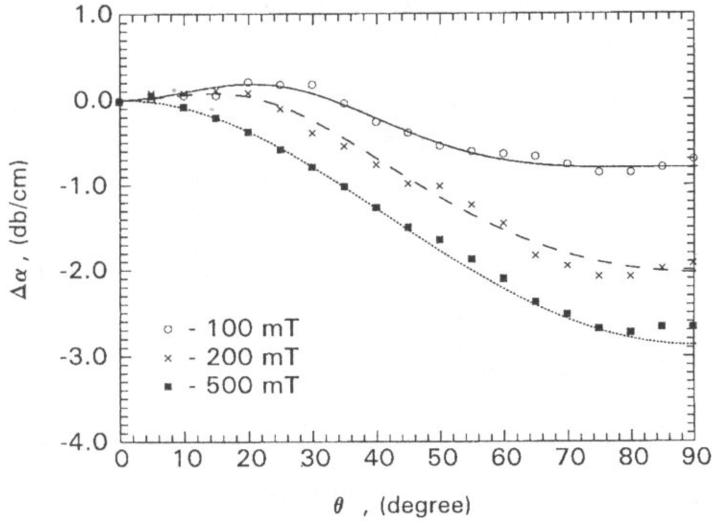

Fig. 3. Angluar dependence of ultrasonic attenuation $\Delta\alpha$ for $f = 4.37$ MHz in three magnetic field strengths. The solid, dashed and dotted lines represent the Gotoh and Chung theory for 100mT, 200 mT and 500 mT, respectively.

$$c = 1439.69\sqrt{\frac{1-0.82508x}{1-0.82465x}},$$

$$\alpha = 29.9\sqrt{\frac{1+0.35x}{1+0.29x}}\,\frac{1+0.44x+0.03x^2}{(1+0.29x)^2}, \qquad \text{for 5200 mT}$$

$$c = 1439.90\sqrt{\frac{1-0.97819x}{1-0.97818x}},$$

The coefficients a_1 and a_2 which appear both in Eqs.(5) and (6) should have similar (if not the same) values for given magnetic fields since the velocity and attenuation were measured under the same thermodynamical conditions and at the same frequency of the ultrasonic wave. This is not the case. In fact, it is impossible to get satisfactory agreement between experiment and Gotoh/Chung theory when the coefficients a_1 and a_2 obtained from fitting the velocity data to Eq.(5) are inserted into Eq.(6) for attenuation and the fitting procedure is restricted only to the coefficients a_3 and a_4. This indicates that the Gotoh/Chung theory lacks inner consistency. One possible origin of this situation seems to reside in the approximation used to solve the eigenvalue equation. However, the usefulness of the Gotoh/Chung theory is very limited since it is rather difficult to deduce any valuable information concerning the magnetic liquid on the basis of the parameters obtained through the fitting process.

Figure 4 shows the results of the analysis of the angular dependency of the absorption coefficient in terms of Taketomi's theory. The solid, dashed and dotted

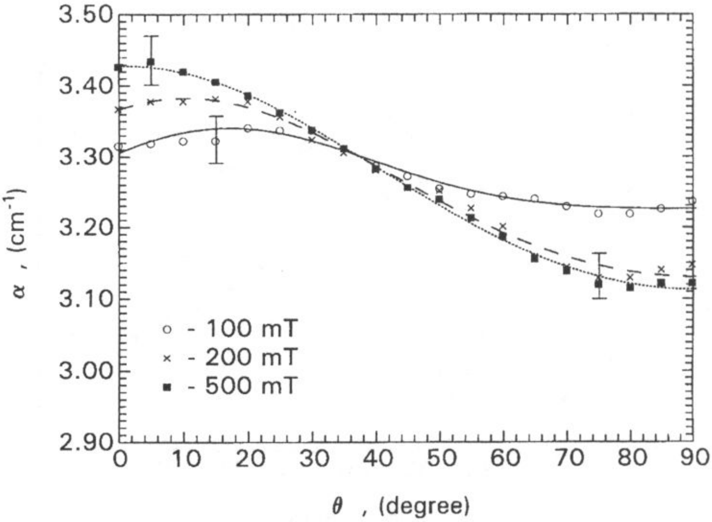

Fig. 4. Angular dependence of the absorption coefficient α for $f =$ MHz in three magnetic field strengths. The solid, dashed and dotted lines represent Taketomi theory for 100 mT, 200 mT and 500 mT, respectively.

lines were obtained by fitting the experimental results to the sum of Eqs.(9) and (10). The fitting parameters values, $\eta_s+4/3\eta_b$, α_5, α_1, a, k, N are listed in Table 1. Here, we take the values $\eta_0 = 0.1003\ \mathrm{N\cdot s\cdot m^{-2}}$, $\rho_0 = 996\ \mathrm{km\cdot m^{-3}}$ for water, and $\rho_m = 5240\ \mathrm{kg\cdot m^{-3}}$ for magnetite.

The values of $\eta_s+4/3\eta_b$ listed in Table 1 show that viscosity is about 4 – 5 cP and increases slightly with the magnetic field. The viscosity values obtained are close to those reported by the Ferrofluidics Corp.

Table 1

B mT	$\eta_s+4/3\eta_b$ $\mathrm{N\cdot m^{-2}\cdot s^{-1}}$	α_5 $\mathrm{N\cdot m^{-2}\cdot s^{-1}}$	α_1 $\mathrm{N\cdot m^{-2}\cdot s^{-1}}$	$a\times10^6$ m	k $\mathrm{N\cdot m^{-1}}$	$N\times10^{-16}$ $\mathrm{m^{-3}}$
100	0.41 ± 0.02	0.07 ± 0.02	0.13 ± 0.01	1.678 ± 0.002	4.27 ± 0.05	56 ± 4
200	0.42 ± 0.06	0.13 ± 0.03	0.08 ± 0.01	1.673 ± 0.009	2.87 ± 0.09	56 ± 4
500	0.48 ± 0.03	0.10 ± 0.01	0.05 ± 0.01	1.684 ± 0.009	0.14 ± 0.02	57 ± 3

Table 1 shows that the cluster radius and number density of the clusters do not change (within error) with respect to the magnetic field. We speculate that in the external magnetic field $B = 100$ mT most of the magnetic particles are bounded within the clusters so the further increase of the magnetic field neither changes the number density of the cluster nor its radius. If we assume the magnetic particles to be spheres of radius 100 Å then the number density of the magnetic particles is obtained to be $2.6\times10^{24}\ \mathrm{m^{-3}}$.

The force constant k depends strongly on the magnetic field, i.e. k decreases with the external field. The same effect was observed by TAKETOMI [11] who proposed the explanation of this behaviour.

The Taketomi's theory enables the assessment of some useful parameters characterizing magnetic liquids. However, it would be desirable to expand the Taketomi model to include an expression for the ultrasonic velocity.

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