RESONANCE ABSORPTION OF ULTRASONIC WAVES IN A MAGNETIC FLUID

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An external magnetic field applied to a magnetic fluid gives rise to the appearance of spherical and chain-like clusters.

The present work is based on an acoustic method permitting the determination of the presence and size of the clusters by resonance absorption of an ultrasonic wave in a magnetic fluid under the influence of a magnetic field.

Measurements by applying the pulse method of the variations of the ultrasonic absorption coefficient $\Delta\alpha$ for three frequencies in a selected magnetic fluid versus the external magnetic field strength have been reported.

1. Introduction

The magnetic fluid EMG-605 is a suspension consistsing of water as carrier and particles of magnetite $FeO \cdot Fe_2O_3$ about 20 nm in diameter. Thus, it is a colloid with a high degree of dispersion containing polydisperse particles of the ferrimagnetic that has an intrinsic magnetic moment.

The stability of the colloidal system of magnetic particles is ensured by an adsorption layer, the molecules of which have a polar group bound physically or chemically to the surface of the solid phase.

In the case of oleinic acid $CH_3(CH_2)_7 = CH(CH_2)_7COOH$ as the surface active substance, the thickness of the adsorption layer is determined by the length of the molecule of the acid which amounts to about 2 nm.

The externally applied magnetic field causes an ordering of the magnetic moments of the particles giving rise to a magnetization of the sample as a whole on a macroscopic scale. If the external magnetic field is weak, thermal motion counteracts the arraying of the magnetic moments into the direction of the field, whereas in a very strong field most of the particles become oriented and the magnetization of the sample attains saturation [4].

At equilibrium, the magnitude of the magnetization is a function of the volume concentration of the magnetite particles (their magnetic moments), the strength of the external magnetic field, and temperature.

For the majority of magnetic fluids, their state of magnetization is satisfactorily described by the classical Langevin law [6] magnetization of molecules of a paramagnetic gas.

In the case of magnetic fluids, the corresponding expression, is fulfilled on the assumption that dipolar magnetic or electric interactions between neighbouring particles are absent or that, at least, the energy of those interactions is less than kT, i.e. the energy of thermal oscillations.

Otherwise, the magnetic particles coagulate and the magnetic fluid is subject to restructurization. Spherical clusters, with a radius ranging from several tens of nanometers up to micrometers, arise as well as chain-like clusters accessible to microscopic observation.

The process of restructurization of the magnetic fluid requires some time depending on the evolution of the aggregates with increasing strength of the external magnetic field. The presence of hysteresis in the magnetization curve of the magnetic fluid (or hysteresis of the acoustic wave absorption coefficient) points to the formation of aggregates in the fluid that can continue to exist even after the removal of the magnetic field [2].

One of the methods enabling us to detect the presence of aggregates and to determine their size is based on resonance absorption of ultrasonic waves in magnetic fluids. The idea of this method and the analysis of experimental results is the subject of the present paper.

2. The behaviour of magnetic objects in a magnetic fluid to be subject to an external magnetic field

An elementary magnet having a magnetic moment m, when immersed in a magnetic field B, undergoes the action of a mechanical moment T reorienting it into the direction of the lines of the magnetic field. The moment T is in equilibrium with the moment resulting from the Newton's Second Law of rotational motion:

$$T = m \times B = mB \sin \phi = -I\frac{d^2\phi}{dt^2}, \tag{2.1}$$

whence we get:

$$\frac{d^2\phi}{dt^2} + \frac{mB}{I}\sin \phi = 0. \tag{2.2}$$

For $\phi < 0.1\,\mathrm{rad}$, we have approximately $\sin(\phi) \approx \phi$ which lead to the equation of on harmonic oscillator:

$$\frac{d^2\phi}{dt^2} + \omega_0^2\phi = 0, \tag{2.3}$$

where

$$\frac{\omega_0}{2\pi} = f_0 = \frac{1}{2\pi}\sqrt{\frac{mB}{I}}. \tag{2.4}$$

When an ultrasonic wave of frequency f_a propagates through a magnetic fluid containing *spherical magnetic particles* of magnetic moment $m = VM_s$ and a moment of

inertia $I = 2V\varrho_M r^2/5$, the energy of the wave is transferred first to the translational and then to the rotational degrees of freedom of the particles.

On the application of "magnetic field scanning" from zero up to a well defined value of the magnetic field induction and if the equality $f_a = f_0$, is fulfilled, resonance absorption of the energy of the ultrasonic wave by the rotational degrees of freedom occurs.

The expression for the frequency of low amplitude vibrations in rotational motion of spherical particles, when the shear viscosity of the medium is not taken into account, becomes now:

$$f_0 = \frac{1}{2\pi r}\sqrt{\frac{5M_sB}{2\varrho_M}}\,, \tag{2.5}$$

that allows to determine the radius

$$r = \frac{1}{2\pi f_0}\sqrt{\frac{5M_sB}{2\varrho_M}}\,, \tag{2.6}$$

of the magnetic particles of size corresponding to the frequency of the applied ultrasonic waves at which, for a given magnitude of B, resonance vibrations of the spherical particles occur as indicated by a maximum of the absorption coefficient of the ultrasonic wave.

For magnetite particles, for which $M_s = 477.7\,\mathrm{kA/m}$ and $\varrho_M = 5240\,\mathrm{kg/m^3}$, we obtain [3]:

$$r = \frac{2.4\,[\mathrm{m \cdot T^{-1/2} \cdot Hz}]\,\sqrt{B}}{f_0}\,. \tag{2.7}$$

If the magnetite particles form *spherical clusters*, the magnetization and density of the latters are smaller than those of a magnetite crystal [3].

On the assumption that the magnetite particles forming the cluster have a radius, r, of $10\,\mathrm{nm}$ and are packed hexagonally, and that the thickness of the layer of the surface active substance amounts to $\delta = 2\,\mathrm{nm}$, the maximum magnetization of such a cluster can be evaluated from the formula [3]

$$M_{cl} = \phi_h M_s \left(\frac{r}{r+\delta}\right)^3\,, \tag{2.8}$$

where $\phi_h = 0.74$ is the *volume concentration of magnetite particles* in the case of hexagonal packing of the latters within the cluster.

Hence, the maximum magnetization of the cluster is $M_{cl} = 204.6\,\mathrm{kA/m}$. Next, taking into consideration the mass of all the components of the cluster, we evaluated the density, ϱ_{cl}, of the cluster for a hexagonal packing, from the formula:

$$\varrho_{cl} = \phi_h(\varrho_M - \varrho_s)\left(\frac{r}{r+\delta}\right)^3 - \phi_h(\varrho_0 - \varrho_s) + \varrho_0\,, \tag{2.9}$$

with the following notation: $\varrho_0 = 988\,\mathrm{kg/m^3}$ — the density of water at $+50\,^\circ\mathrm{C}$, and $\varrho_s = 877.7\,\mathrm{kg/m^3}$ — that of the oleinic acid at $+50\,^\circ\mathrm{C}$. Thus, for our further calculations, we took $\varrho_{cl} = 2774.5\,\mathrm{kg/m^3}$.

When applying the expression (2.6) for the clusters, we have to use the values calculated from Eqs. (2.8) and (2.9) for their magnetization and density, respectively; these values obviously differ strongly from those of a single magnetite particle.

For comparison, Fig. 1 shows the free rotation frequencies of single magnetic particles and those of the clusters, formed by them, in an external magnetic field.
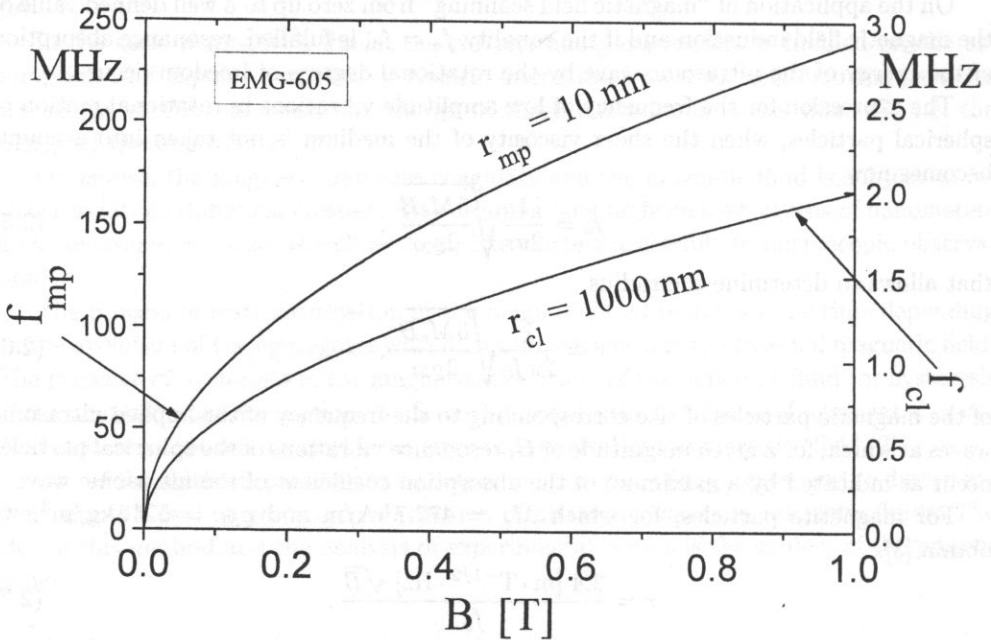

Fig. 1. Free rotational vibration frequencies for single magnetic particles f_{mp} ($r_{mp} = 10$ nm, $M_s = 477.7$ kA/m, $\varrho_M = 5240$ kg/m^3) and clusters f_{cl} ($r_{cl} = 10^{-6}$ m, $M_{cl} = 204.6$ kA/m, $\varrho_{cl} = 1774.5$ kg/m^3) versus the magnetic field induction (B).

The functions are of the form:

$$f_{mp} = 240 \, [\mathrm{M\,Hz \cdot T^{-1/2}}] \sqrt{B}, \qquad f_{cl} = 2.16 \, [\mathrm{M\,Hz \cdot T^{-1/2}}] \sqrt{B}. \tag{2.10}$$

2.1. Chain clusters in a magnetic field

In the case of chain-like clusters of length l [1, 5, 9] the magnetic moment is

$$m = V M_{cl} = \frac{4\pi r_{cl}^3 n}{3} M_{cl} = \frac{2\pi r_{cl}^2 l}{3} M_{cl}, \tag{2.11}$$

where $n = l/2r_{cl}$ is the number of clusters of radius r_{cl} forming the chain of length l. The moment of inertia of a chain of clusters calculated with respect to its centre is:

$$I = \int_{-l/2}^{l/2} \varrho_{cl} s x^2 \, dx = \frac{\varrho_{cl} s l^3}{12} = \frac{m l^2}{12}, \tag{2.12}$$

and since the mass of the chain and its length are $m = 2\pi r_{cl}^2 \varrho_{cl} l/3$ and $l = 2r_{cl}n$, respectively, we finally get

$$I = \frac{\pi \varrho_{cl} r_{cl}^2 l^3}{18} = \frac{4\pi \varrho_{cl} r_{cl}^5 n^3}{9}. \tag{2.13}$$

With regard to the above, the expression for the vibration eigen-frequency of the chains takes the form:

$$f_0 = \frac{1}{2\pi}\sqrt{\frac{mB}{I}} = \frac{1}{\pi l}\sqrt{\frac{3M_{cl}B}{\varrho_{cl}}}. \tag{2.14}$$

Formula (2.14) shows that chains of the same length have the same eigen-frequencies irrespective of the radius of the clusters forming them.

In the case of a chain of magnetic clusters of length $l = 10\,\mu m$ in a field $B = 0.5\,T$, the vibration eigen-frequency of the magnetic object as a whole is $f_0 = 372\,kHz$.

It should, however, be kept in mind that the above result was obtained on the assumption that no frictional forces are active between the magnetic particles and the carrier liquid.

For greater vibration amplitudes of spherical or chain-like clusters $\phi \gg 0.1\,rad$, the effect becomes *nonlinear* and the free vibration frequency becomes dependent on the vibration amplitude.

Restricting our calculations to the first two terms of the power expansion of the $\sin\phi$ function:

$$\sin\phi = \frac{\phi}{1!} + \frac{\phi^3}{3!} - \frac{\phi^7}{7!} + \dots , \tag{2.15}$$

Eq. (2.2) takes the form:

$$\frac{d^2\phi}{dt^2} + \frac{mB}{I}\left(\phi - \frac{\phi^3}{6}\right) = 0. \tag{2.16}$$

Inserting $\omega_0^2 = mB/I$, we finally arrive at the equation of motion of an anharmonic oscillator

$$\frac{d^2\phi}{dt^2} + \omega_0^2\phi - \frac{\omega_0^2}{6}\phi^3 = 0. \tag{2.17}$$

Equation (2.17) shows that the vibration frequency of the magnetic objects depends on the vibration amplitude ϕ_0:

$$f = f_0\left(1 - \frac{\phi_0^2}{16}\right). \tag{2.18}$$

2.2. Rotational oscillations of a spherical cluster in a viscous liquid

When considering the motion of a spherical cluster with a radius r_{cl} and a magnetic moment m in a field B, we have to take into account, in addition to the moment $m \times B$ due to the action of the field on the magnet and the moment of inertia of the sphere in rotational motion $I \times d\omega/dt$, the frictional moment originating from the friction between the sphere and the carrier liquid of viscosity η.

With regard to Fig. 2, the elementary moment dM of the force dF is:

$$dM = a \times dF = r_{cl}\sin\alpha\, dF, \tag{2.19}$$

whereas the elementary force dF acting tangentially to the element ds of the spherical surface is:

$$dF = \eta\frac{\nu(a)}{d}ds = \frac{\eta\omega a 2\pi a r_{cl}}{d}d\alpha = \frac{2\pi\omega\eta r_{cl}^3\sin^2\alpha}{d}d\alpha, \tag{2.20}$$
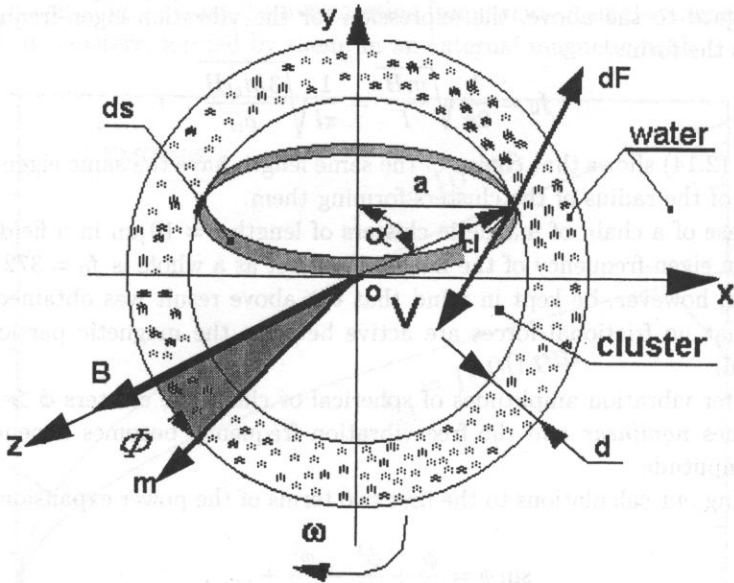
Fig. 2. Rotational vibrations of a spherical cluster in a viscous medium acted on by an external magnetic field.

where d is the thickness of the interface layer experiencing the gradient of the linear velocity $\nu(a)/d$ during the rotational motion of the sphere.

Consequently, the differential of the moment is given by the expression:

$$dM = \frac{2\pi\omega\eta r_{cl}^4 \sin^3 \alpha}{d} d\alpha. \qquad (2.21)$$

The total moment M is obtained by integration of (2.21):

$$M = \frac{2\pi\omega\eta r_{cl}^4}{d} \int\limits_{\alpha=0}^{\pi} \sin^3 \alpha \, d\alpha = \frac{2\pi^2\omega\eta r_{cl}^4}{3d}, \qquad (2.22)$$

and the equation of rotational motion takes the form:

$$I\frac{d^2\phi}{dt^2} + \frac{2\pi^2 r_{cl}^4 \eta}{3d}\frac{d\phi}{dt} + mB\sin\phi = 0. \qquad (2.23)$$

Since for a spherical cluster we have $m = V M_{cl}$, $I = 2V \varrho_{cl} r_{cl}^2/5$ and, moreover, at $\phi < 0.1\,\text{rad}$ and $\sin\phi \approx \phi$, we get:

$$\frac{d^2\phi}{dt^2} + \left(\frac{5\pi\eta}{4dr_{cl}\varrho_{cl}}\right)\frac{d\phi}{dt} + \left(\frac{5M_{cl}B}{2r_{cl}^2\varrho_{cl}}\right)\phi = 0. \qquad (2.24)$$

On insertion of

$$\omega_0^2 = \frac{5M_{cl}B}{2r_{cl}^2\varrho_{cl}}, \qquad \frac{1}{\tau} = \frac{5\pi\eta}{4dr_{cl}\varrho_{cl}}, \qquad (2.25)$$

we arrive at

$$\frac{d^2\phi}{dt^2} + \frac{1}{\tau}\frac{d\phi}{dt} + \omega_0^2\phi = 0. \tag{2.26}$$

We search for a solution for damped vibrations of the cluster in the form:

$$\phi = \phi_0 \exp^{-\beta t} \sin\omega t, \tag{2.27}$$

which will serve to determine β and ω.

The first and second derivatives of (2.27) are, respectively,

$$\frac{d\phi}{dt} = -\beta\phi_0 \exp^{-\beta t} \sin\omega t + \omega\phi_0 \exp^{-\beta t} \cos\omega t, \tag{2.28}$$

$$\frac{d^2\phi}{dt^2} = \beta^2\phi_0 \exp^{-\beta t} \sin\omega t - 2\omega\beta\phi_0 \exp^{-\beta t} \cos\omega t - \omega^2\phi_0 \exp^{-\beta t} \sin\omega t, \tag{2.29}$$

which, on insertion into (2.26) and after some algebra give

$$\left(\beta^2 - \omega^2 + \omega_0^2 - \frac{\beta}{\tau}\right)\phi_0 \exp^{-\beta t}\sin\omega t + \left(\frac{\omega}{\tau} - 2\omega\beta\right)\phi_0 \exp^{-\beta t}\cos\omega t = 0. \tag{2.30}$$

In (2.30) the coefficient of the second term consisting $\cos\omega t$ vanishes if

$$\beta = \frac{1}{2\tau} = \frac{5\pi\eta}{8dr_{cl}\varrho_{cl}}, \tag{2.31}$$

whereas that of the first one vanishes if

$$\omega^2 = \omega_0^2 + \beta^2 - \frac{\beta}{\tau} = \omega_0^2 - \left(\frac{1}{2\tau}\right)^2. \tag{2.32}$$

Thus,

$$\omega = \omega_0\sqrt{1 - \left(\frac{1}{2\omega_0\tau}\right)^2} \tag{2.33}$$

Inserting the above defined quantities into (2.33) we get the expression for the frequency of vibration of the spherical cluster in the damping medium:

$$f = \frac{1}{2\pi r_{cl}}\sqrt{\frac{5M_{cl}B}{2\varrho_{cl}} - \left(\frac{5\pi\eta}{8d\varrho_{cl}}\right)^2}. \tag{2.34}$$

Referring the above expression to the frequency f_0 for a non-viscous liquid, we obtain

$$\frac{f}{f_0} = \sqrt{1 - \frac{5\pi^2\eta^2}{32\varrho_{cl}Bd^2M_{cl}}}. \tag{2.35}$$

The determination of the correct thickness of the interface layer is a highly essential problem. Equating Eq. (2.22) to the equivalent expression derived from Ref. [8], we obtain:

$$M = \frac{2\pi^2\omega r_{cl}^4}{3d} = 8\pi\eta r_{cl}^3\omega, \tag{2.36}$$

that allows the evaluation of the interface layer

$$d = \frac{\pi r_{cl}}{12}.$$ (2.37)

Hence, the formulae (2.34) and (2.35) become,

$$f = \frac{1}{2\pi r_{cl}} \sqrt{\frac{5BM_{cl}}{2\varrho_{cl}} - \left(\frac{15\eta}{2r_{cl}\varrho_{cl}}\right)^2},$$ (2.38)

and

$$\frac{f}{f_0} = \sqrt{1 - \frac{22.5\eta^2}{BM_{cl}r_{cl}^2\varrho_{cl}}},$$ (2.39)

respectively. On fitting typical values for suspended in water clusters with a radius of $r_{cl} = 1\,\mu\text{m}$, for which $M_{cl} = 204.6\,\text{kA/m}$ and $\varrho_{cl} = 2774.5\,\text{kg/m}^3$, while the water shear viscosity is $\eta = 0.001\,\text{N}\cdot\text{s/m}^2$ and the magnetic field $B = 0.5\,\text{T}$, we get $f/f_0 = 0.96$.

Obviously, with increasing induction B we observe a increase in f/f_0 as shown in Fig. 3.
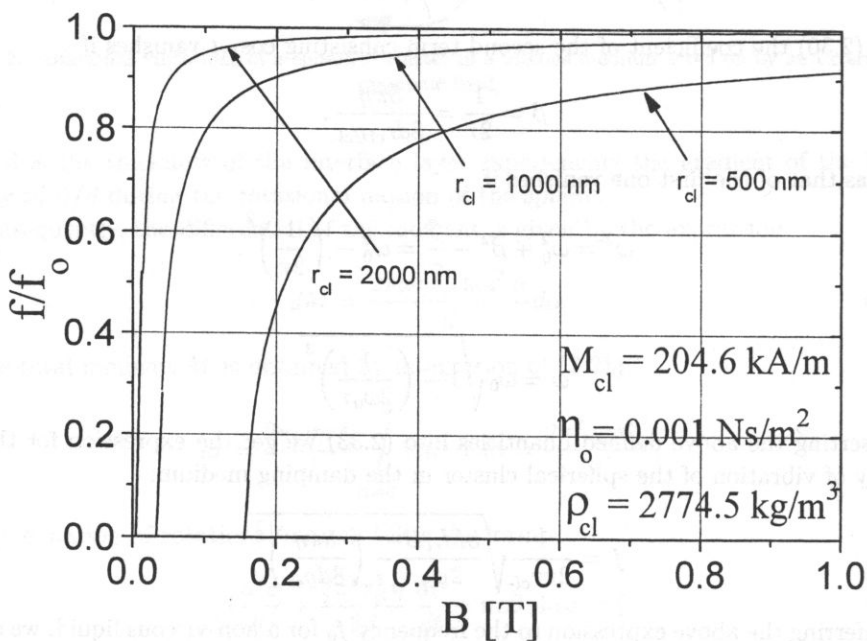
Fig. 3. f/f_0 versus B for spherical clusters with different radii in the magnetic fluid EMG-605.

Table 1 gives examples of B values for which the expression (2.39) vanishes for some selected radii of the clusters involving very strong damping of their rotation due to friction. Our evaluation was made for the following values: $M_{cl} = 204.6\,\text{kA/m}$, $\varrho_{cl} = 2774.5\,\text{kg/m}^3$, $\eta = 0.001\,\text{N}\cdot\text{s/m}^2$.

Hence one should expect an increase in the ultrasonic wave absorption in the range of small B values resulting from the friction due to shear viscosity.

Table 1. Magnetite cluster radii and the values of B for which a critical damping of rotation vibration of the clusters takes place.

r_{cl} [nm]	2000	1000	500
B [mT]	9.9	36.9	158.6

3. The method of determining the variations in ultrasonic wave absorption coefficient $\Delta\alpha$ in the magnetic fluid under investigation

The measuring set-up used for recording changes in the absorption of ultrasonic waves in the magnetic fluid caused by variations of the strength of the external magnetic field is shown in Fig. 4. Basing on the pulse method, it consists of subsets produced by "MATEC" and involves a "MODEL 2470B" block containing echo pulse delay circuits and channels with analog memories, one of which recording the amplitude value of the first echo.
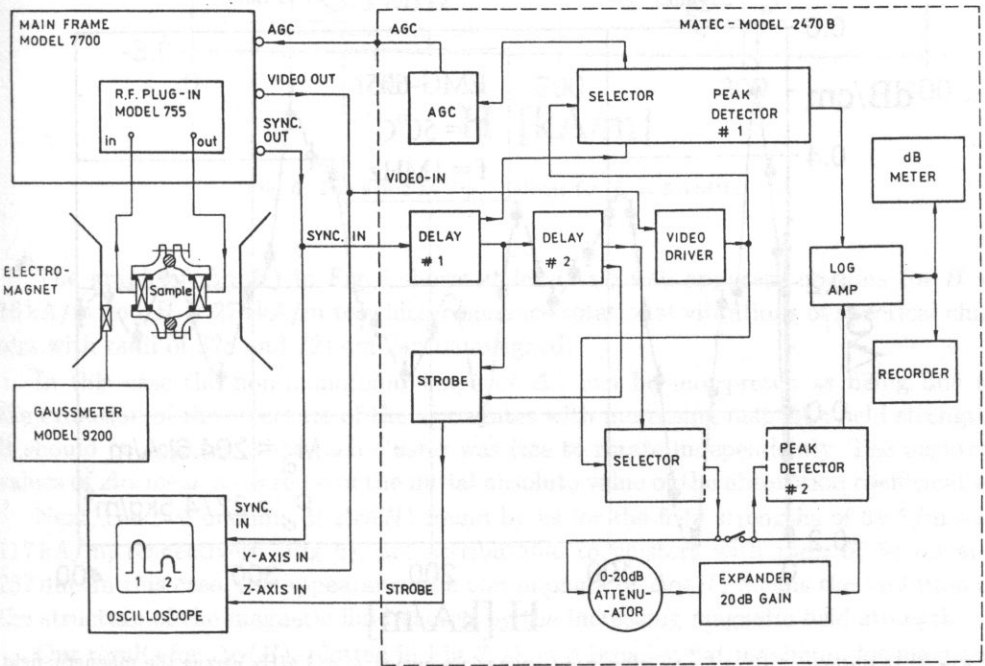
Fig. 4. Block diagram of the measuring stand for the determination of variations of $\Delta\alpha(H)$ for an ultrasonic wave in EMG-605.

In our measurements, the set-up operated according to the "single echo" principle and permitted to record variations in the absorption coefficient of ultrasonic waves $\Delta\alpha$. Hence the constant voltage signal was fed to a logarithmic amplifier giving in the output voltage a change proportional to that in the amplitude coefficient of the ultrasonic wave absorption in the magnetic fluid.

Additionally, we applied on the analog output of the "MODEL 2470B" block, a low transmission RC filter with constant delay time of about 5 s averaging the values of the voltage measured with a digital voltmeter.

The accuracy of our measurements, performed with the setup described above, amounted to about ±0.1 dB/cm in the range of 20 dB.

The magnetic field induction in the slit of the electromagnet was measured with a "BELL 9200" type teslometer to within 0.5 per cent. The temperature of the medium was stabilized to within ±0.01 K.

4. Experimental results and their analysis

The changes in absorption coefficient $\Delta\alpha$ in the magnetic fluid EMG-605 were measured for ultrasonic frequencies $f_a = 1$ MHz (Fig. 5); $f_a = 3.5$ MHz (Fig. 6); and $f_a = 6.7$ MHz (Fig. 7) versus increasing magnetic field strength at a temperature of +50°C.

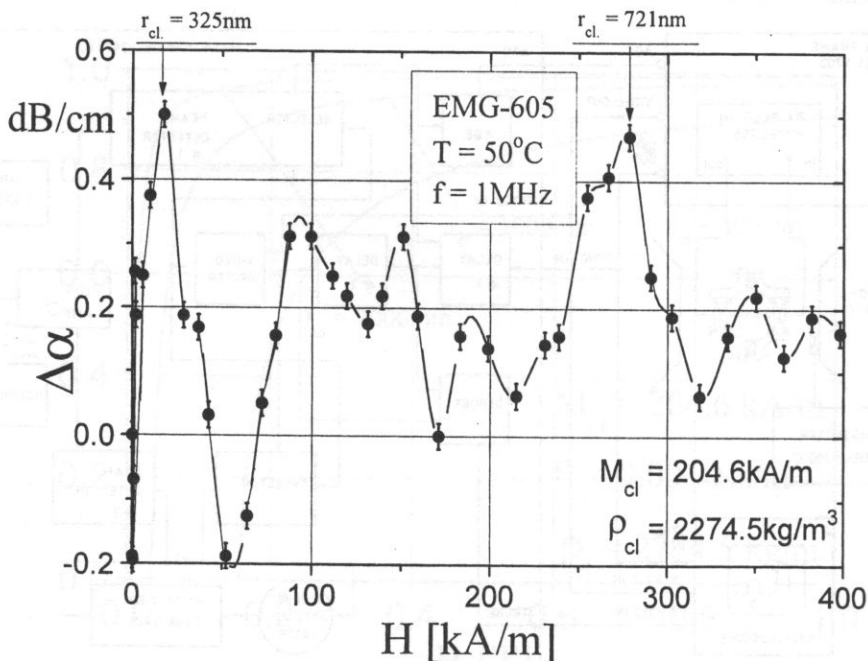

Fig. 5. Variations $\Delta\alpha(H)$ for an ultrasonic wave of frequency $f_a = 1$ MHz versus the magnetic field strength H.

The measuring cell, with ceramic transducers distant by 1.6 cm, had a volume of about 2 cm³ and was disposed so that the magnetic field vector and wave propagation vector overlapped.

The magnetic field was manually increased from zero, after previous demagnetization of the electromagnet core, up to the successive field strengths selected for the measurements of $\Delta\alpha$, after a lapse of 5 minutes in each case.
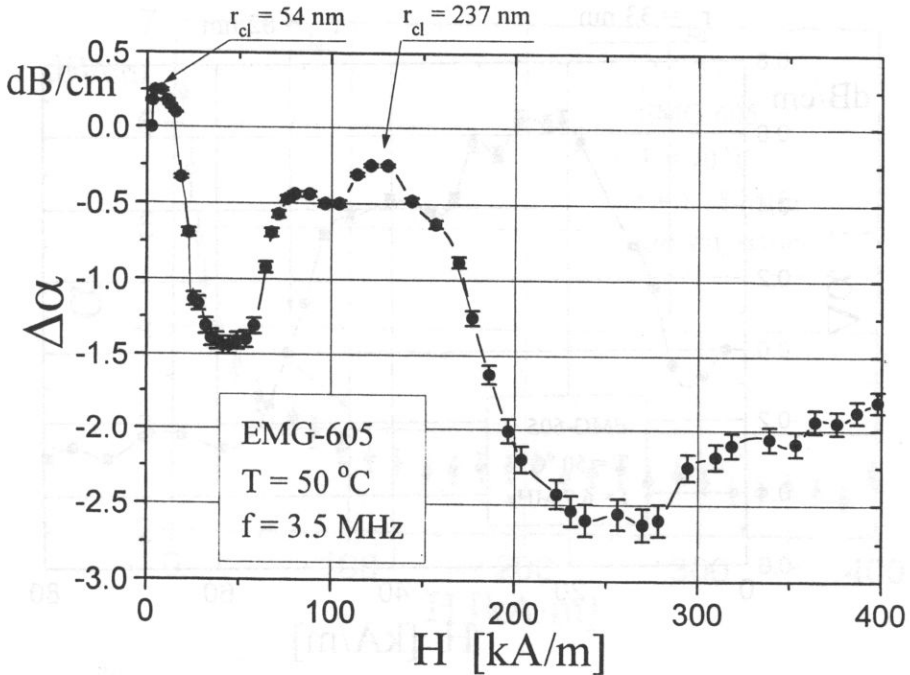
Fig. 6. The same as Fig. 5, albeit for $f_a = 3.5$ MHz.

The graph of $\Delta\alpha(H)$ in Fig. 5 shows at least two well apparent maxima for $H = 18$ kA/m and $H = 278$ kA/m to which resonance rotational vibrations of spherical clusters with radii of 325 and 721 nm can be assigned.

In this case the non-monotonic shape of $\Delta\alpha$ can be interpreted as being due to the evolution of the structure of the aggregates with increasing magnetic field strength. It should be apparent that each cluster was free to rotate independently. The negative values of $\Delta\alpha$ mean a decrease of the initial absolute value of the absorption coefficient α.

Next, the two maxima of $\Delta\alpha(H)$ found by us for the field strengths of 6 kA/m and 117 kA/m, respectively (Fig. 6), are attributable to clusters with radii of 54 nm and 237 nm. In this case, the appearance the two maxima of $\Delta\alpha(H)$ means the evolution of the structure of the magnetic fluid caused by the increasing magnetic field strength.

Our results for $\Delta\alpha(H)$, plotted in Fig. 7, show a broaden flat maximum for magnetic field strengths of $8.7 \div 52$ kA/m that points to the possible presence of spherical clusters with radii ranging from 33 to 82 nm in magnetic fluid.

It is noteworthy that the radius 54 nm, determined at $f_a = 3.5$ MHz from the first maximum of $\Delta\alpha(H)$ in Fig. 6, coincides with the range of the cluster radii in Fig. 7 determined from $\Delta\alpha(H)$ recordings.

Hence, the same cluster structures are responsible for the $\Delta\alpha(H)$ graphs in the two cases. The phenomenon points to a high degree of complexity of the restructurization processes in the magnetic fluid under the action of the external magnetic field [1, 5].
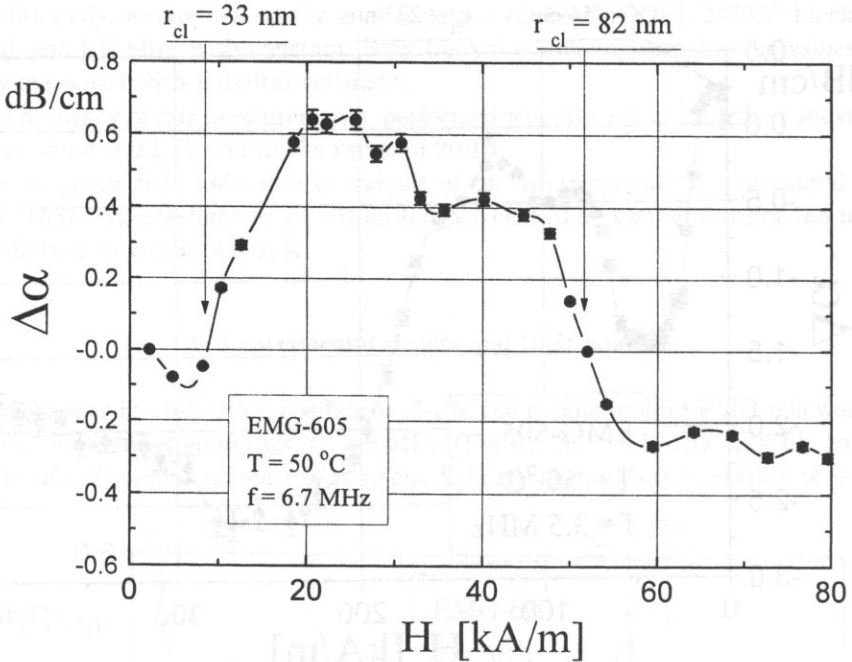
Fig. 7. The same as Fig. 5, albeit for $f_a = 6.7\,\text{MHz}$.

Moreover, in the magnetic field of changing strength, clusters can arise, which under energetically favourable conditions, can continue to exist even after the magnetic field is switched off. This becomes apparent as a hysteresis of the ultrasonic absorption coefficient: the values of α obtained with increasing magnetic strength do not coincide with those obtained in a decreasing field (see Figs. 8a and 8b). Dealing with the effect in question, we determined the absolute value of α in the magnetic fluid (with no magnetic hysteresis) at a slightly higher temperature (70°C) and at $f_a = 1.18\,\text{MHz}$ in fields ranging from 0 to 400 kA/m. In the region of very weak fields, the damping was very strong showing that only magnetic objects of a size of single magnetic particles took part in the process of resonance absorption.

The monotonic decrease of α with growing H (without any maxima of α) points to the absence of clusters in the virgin fluid. However, in a decreasing field (Fig. 8) certain irregularities appear in the behaviour of α pointing to the rise of clusters during the time involved. The following steep increase in the absorption coefficient when H approaches zero, proves that not all the magnetic particles were consumed in the cluster formation process.

Additionally, we carried out density measurements of the (Fig. 9) EMG-605 (with no magnetic prehistory) and water as function of the temperature in the range from 15 to 40°C, using a "DMA-38" microprocessor densitometer from Anton Paar that operates by the method proposed by KRATKY et al. [7].

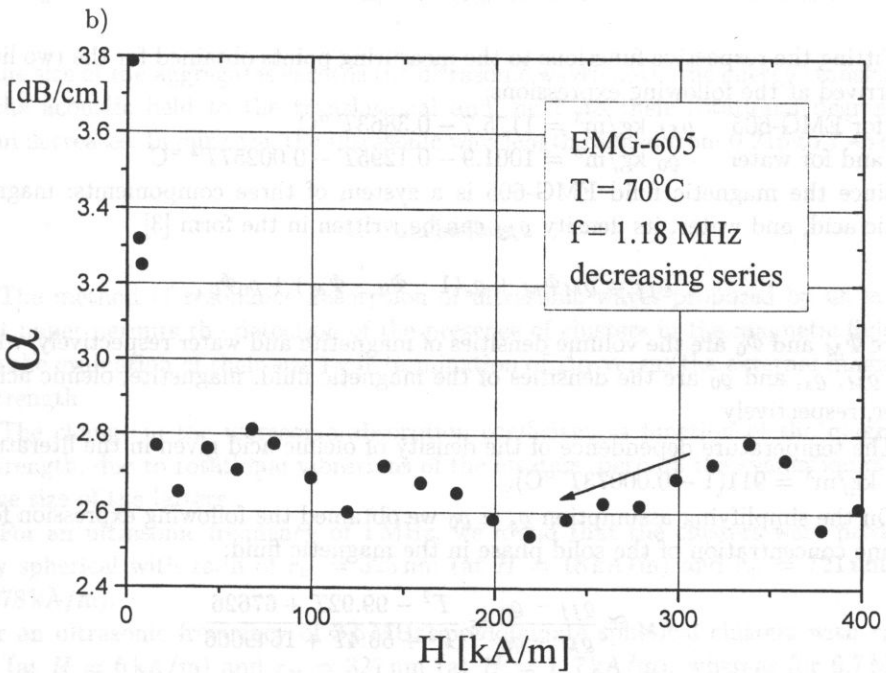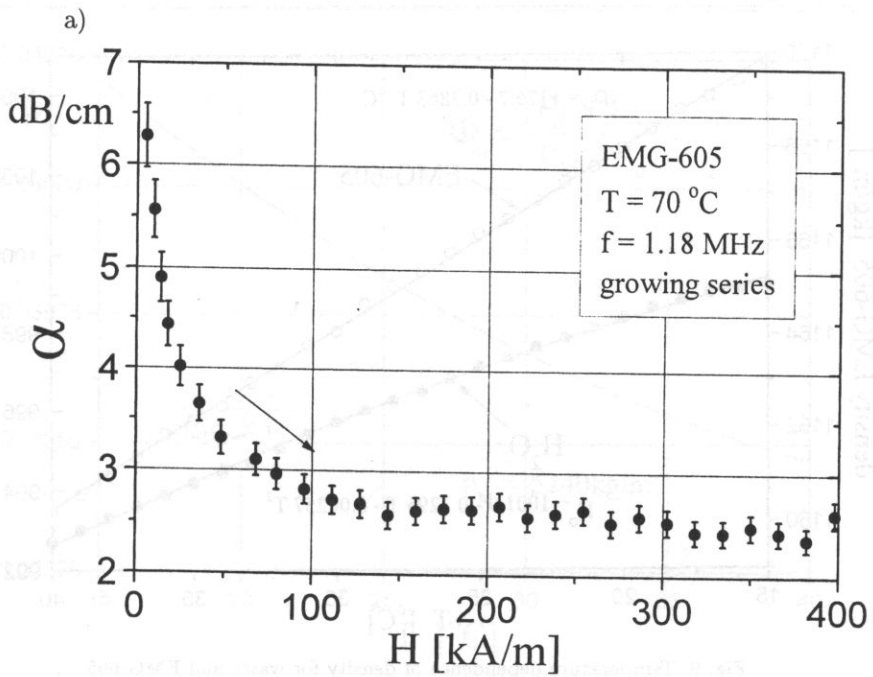The accuracy, as declared by the makers, amounted to $\pm 10^{-3}\,\text{g/cm}^3$ and $\pm 0.1\,°\text{C}$.

a)

b)

Fig. 8. a) Absolute value of the absorption coefficient $\alpha(H)$ of an ultrasonic wave ($f_a = 1.18$ MHz) in the magnetic fluid EMG-605 at $+70\,^\circ$C as function of the external, magnetic field at increasing field strength. b) Absolute value of the absorption coefficient $\alpha(H)$ of an ultrasonic wave ($f_a = 1.18$ MHz) in the magnetic fluid EMG-605 at $+70\,^\circ$C as a function of the external, magnetic field at decreasing field strength.
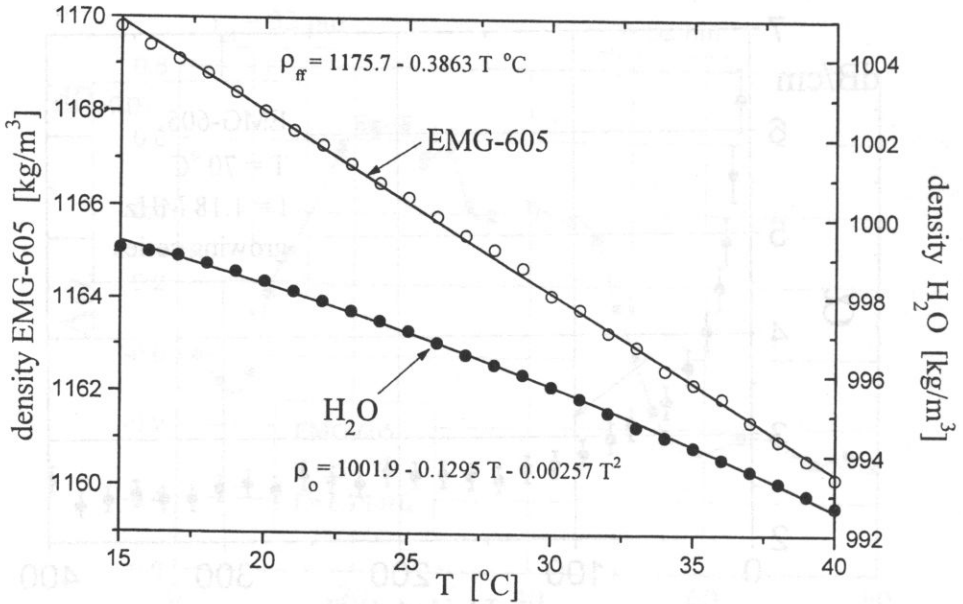
Fig. 9. Temperature dependence of density for water and EMG-605.

Fitting the respective functions to the measuring points obtained for the two liquids, we arrived at the following expressions:

for EMG-605 ϱ_{ff} kg/m^3 = 1175.7 − 0.3863T °C

and for water ϱ_0 kg/m^3 = 1001.9 − 0.1295T − 0.00257T^2 °C.

Since the magnetic fluid EMG-605 is a system of three compomemts: magnetite, oleinic acid, and water, its density ϱ_{ff} can be written in the form [3]

$$\varrho_{ff} = \varrho_M \Phi_M + \varrho_s(1 - \Phi_0 - \Phi_M) + \varrho_0 \Phi_0, \qquad (4.1)$$

where Φ_M and Φ_0 are the volume densities of magnetite and water respectively, whereas ϱ_{ff}, ϱ_M, ϱ_s, and ϱ_0 are the densities of the magnetic fluid, magnetite, oleinic acid and water, respectively.

The temperature dependence of the density of oleinic acid given in the literature [3] is ϱ_s kg/m^3 = 911(1 − 0.00073T °C).

On the simplifying assumption $\varrho_s \simeq \varrho_0$ we obtained the following expression for the volume concentration of the solid phase in the magnetic fluid:

$$\Phi_M \cong \frac{\varrho_{ff} - \varrho_0}{\varrho_M - \varrho_0} = \frac{T^2 - 99.92T + 67626}{T^2 + 50.4T + 1649066}. \qquad (4.2)$$

The expression (4.2) is plotted in Fig. 10 as a function of temperature. The aggregates of magnetic particles differ from the carrier liquid in their dynamic and thermodynamic properties. Thus, once the acoustic properties of the magnetic fluid are available, we are in a position to draw conclusions regarding the changes taking place therein.
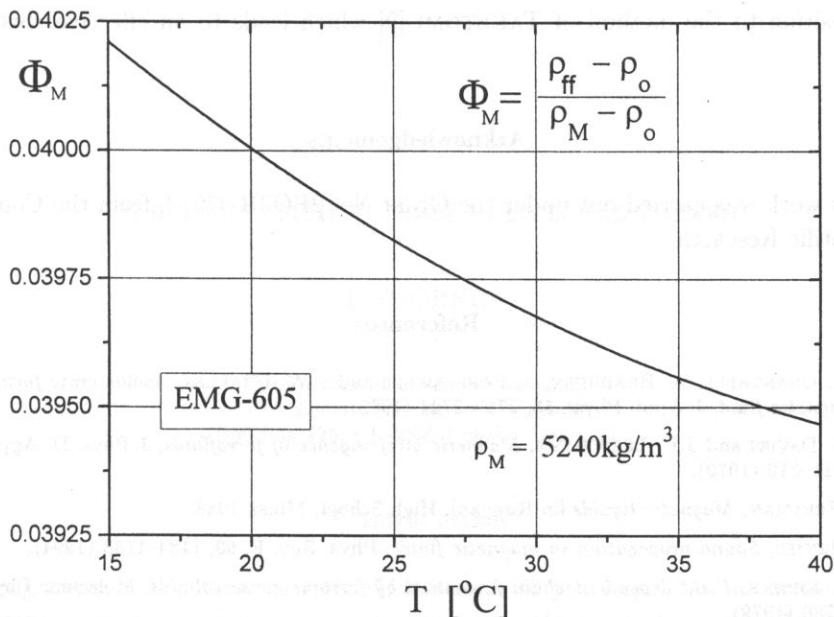
Fig. 10. Volume concentration of magnetite (Φ_M) in EMG-605 *versus* temperature.

If the size of the aggregates exceeds the ultrasonic wavelength, the energy transferred from the acoustic field to the translational and, next, to their rotational degrees of freedom decreases. In our case, the ultrasonic wavelength ranged from 0.216 to 1.45 mm.

5. Conclusions

1. The method of resonance absorption of ultrasonic waves proposed by us in the present paper permits the detection of the presence of clusters in the magnetic fluid as well as the evaluation of their size by determination of $\Delta\alpha$ versus the external magnetic field strength.

2. The change in the ultrasonic absorption coefficient as function of the magnetic field strength, due to rotational vibrations of the clusters, permits the evaluation of the effective size of the latters.

3. For an ultrasonic frequency of 1 MHz, we found that the clusters were predominantly spherical with radii of $r_{cl} = 325$ nm (at $H = 18$ kA/m) and $r_{cl} = 721$ nm (at $H = 278$ kA/m).

For an ultrasonic frequency of 3.5 MHz, predominate spherical clusters with $r_{cl} = 54$ nm (at $H = 6$ kA/m) and $r_{cl} = 327$ nm (at $H = 117$ kA/m), whereas for 6.7 MHz, spherical clusters with r_{cl} ranging from 33 nm to 82 nm were predominant (at $H = 8.7 - 52$ kA/m).

4. Our resonance method of ultrasonic wave absorption permits a simultaneous detection of clusters of different size from a single series of measurements of $\Delta\alpha = f(H)$,

in opposition to the method of TAKETOMI [9] which leads to an effective cluster size only.

Acknowledgements

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