

COMPARISON OF THEORIES OF ANISOTROPY IN TRANSFORMER OIL-BASED MAGNETIC FLUIDS

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Abstract. *The external magnetic field in transformer oil-based magnetic fluids leads to the aggregation of magnetic nanoparticles and formation of clusters. These aggregations are the result of the interaction between the external magnetic field and the magnetic moments of the nanoparticles occurs. However, the temperature of magnetic fluids has also very important influence on the structural changes because the mechanism of thermal motion acts against the cluster creation. The acoustic spectroscopy was used to study the anisotropy of transformer oil-based magnetic fluids upon the effect of an external magnetic field and temperature. In present the anisotropy of the magnetic fluids can be described by two theories. Taketomi theory assumes the existence of spherical clusters. These clusters form long chains, aligned in a magnetic field direction. Shliomis in his theory supposed that only nanoparticles formed chains. A comparison of the experimental results with the predictions of the Taketomi theory allowed a determination of the cluster radius and the number density of the colloidal particles. The proportions of the acoustic wave energy used for excitation of the translational and rotational motion were determined.*

Keywords

Acoustic attenuation, anisotropy, clusters, magnetic fluid, rotation and translation motion.

1. Introduction

The transformer oil-based magnetic fluids are prepared by adding magnetic nanoparticles suspension to trans-

former oil. Nanoparticles with diameter (4–20 nm) in these fluids are generally monodomain and to prevent the interaction among them that may lead to their agglomeration and subsequent sedimentation they are coated by surfactants that produce entropic repulsion. These types of magnetic fluids have attracted remarkable physical properties that have recently found wide application in technology [1], [2]. They improve some of the oils insulating and thermal properties what can be an innovating example of development in power electronic technology. Their macroscopic magnetic properties are changed in the external magnetic field generated for example in high voltage transformer.

The transformer oil usually used both for high voltage insulation and power transformers cooling is subjected to extensive research to enhance its characteristics [1], [2]. The dielectric breakdown strength of transformer oil, however, is strongly influenced by the aggregation effects of magnetic particles and can induce electric breakdown [3], [4]. An externally applied magnetic field can cause a certain amount of colloidal particles to join into quasispherical formations and clusters as long as hundreds of nanometers or more [5], [6], [7], [8]. One of the useful tools to study improvements in the transformer oil-based ferrofluids, that is enable to describe the physical behavior of these liquids, and in particular, the magnetic field induced structure changes of magnetic nanoparticles is acoustic spectroscopy. The measurement of changes in the magnetic fluid structure is based on the measurements of changes in acoustic attenuation $\Delta\alpha$ of magnetic fluid under the influence of an external magnetic field and temperature. The structural changes (the process of clusters formation) in magnetic fluids induce also additional changes in acoustic attenuation, so that the interaction between the acoustic wave and the aggregated magnetic particles or clusters produced in the presence of exter-

nal magnetic field leads to the additional absorption of acoustic wave [8], [9], [10], [11], [12]. The dependence of the attenuation of acoustic wave on the angle φ between the direction of propagation and that of the magnetic field, known as the anisotropy of the attenuation of acoustic wave, provides also important information on the ferrofluid structure in a magnetic field. The comparison of the experimental results with the theoretical predictions [8], [9], [10], allows the distinguishing of two motions of the clusters of the ferrous colloidal particles in the fluid, the rotation and translation motions. The determination of the clusters radius, the density and other parameters of the colloidal particles is possible, too.

The ideal magnetic particles interact with the magnetic field, but not interact with each other. The degree of ideality is characterized by the coupling constant $\lambda = \mu^2 / (k_B T d_h^3)$, [9], where μ is the magnetic moment, $\mu = M_b V$, M_b is the bulk magnetization, V is the volume of the grain $V = \pi d^3 / 6$, k_B is Boltzman constant, T is temperature and d_h is hydrodynamic diameter of magnetic particle contains particle diameter and thinness of surfactant layer. The coupling constant determines the magnitude of dipolar attraction of neighboring grains. Large values of coupling constant λ mean agglomeration of particles in larger structures - clusters, chains.

In the present contribution we concern on two theories describes the arrangement of magnetic nanoparticles and their influence on the acoustic attenuation. In next the main properties of these theory's are described.

2. Theories

2.1. Model 1

Taketomi [10] in his model supposed that the ferrous colloidal nanoparticles ($\lambda > 5$) in the external magnetic field take chain-like formation due to their mutual magnetic interaction. Similar long thick chain-like clusters were presented in simulation work of Satoh [13]. These chain formations coalesce together into clusters with a sphere of radius a . These clusters line up like beads in the direction of external magnetic field (Fig. 1). According to Taketomi [10], this formation of clusters caused the acoustic attenuation α , which consists of two parts, the rotational motion of the clusters α_{rot} and their translational motion α_{tr} . The clusters, activated by the acoustic field, perform translational and rotational motions simultaneously. These irreversible processes dissipate the energy of the acoustic wave into heat. The rotating mechanism affecting the acoustic

attenuation is described as [10]:

$$\alpha_{rot} = \frac{2\pi^2 f}{\rho_0 c^3} \left(\frac{4}{3} \eta + \eta_V + 2\alpha_5 \cos^5 \varphi + \alpha_1 \cos^4 \varphi \right), \quad (1)$$

where ρ_0 is the density of the carrier fluid, c is the velocity of the acoustic wave propagating at a frequency f , η and η_V are the dynamic and volume viscosities of the ferrofluid and α_1 , α_5 are the Leslie coefficients appearing in the theory of liquid crystals [8]. The second and the third terms in Eq. 1 are non zero only in the presence of external magnetic field.

The calculation of acoustic attenuation caused by translational motion is described in this section. Taketomi assumed three types of forces in his theory. The first one is, a recovering force F_1 :

$$F_1 = -k \sin(\varphi) x. \quad (2)$$

This force acts on the cluster toward to the original point and it makes than periodic motion. It is the result of acoustic wave force. The second force is the frictional force F_2 . It attracts on the moving cluster, particles in viscose liquid. This force is described by the Stokes law and can be expressed as:

$$F_2 = -6\pi\eta_0 a (u - v_x), \quad (3)$$

where η_0 is the shear viscosity of the magnetic liquid, u is the velocity of cluster and $v_x = v_0 \exp[i(qx - \omega t)]$ is the velocity of fluid caused by acoustic wave.

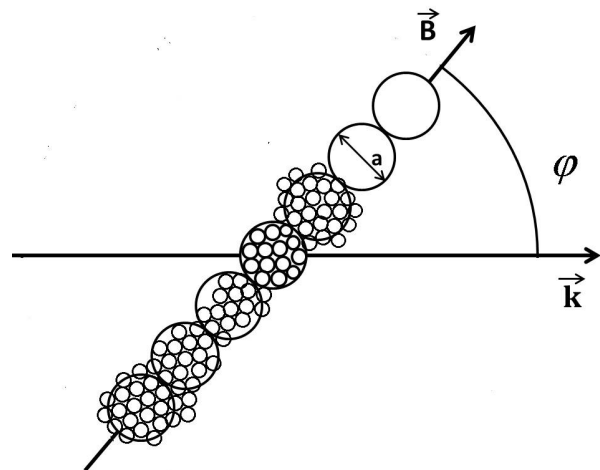


Fig. 1: Chain of clusters [10] with diameter a oriented in the direction of magnetic field \mathbf{B} , φ is the angle between the direction of magnetic field \mathbf{B} and wave vector \vec{k} .

From the acoustic theory of fluids [12] is derived the last force, the force of the pressure F_3 in form:

$$F_3 = \rho_0 V \omega v_x, \quad (4)$$

where ρ_0 is the density of the magnetic liquid, $V = (4\pi/3)a^3$ is the volume of the cluster and ω is a angular frequency of acoustic wave.

The calculation of the dissipative energy E_{dis} per unit volume of the magnetic field per unit time is expressed in work [10]. Finally, the acoustic attenuation caused by translational motion is calculated as dissipative energy divided by $\rho_0 c v_0^2$ and it can be written as:

$$\alpha_{tr} = \frac{3\pi\eta_0 a \omega^3 \rho_0 V N (6\pi\eta_0 a + \rho_0 V \omega)}{\bar{k} c^2} \left(\frac{\sin\varphi - \rho_m V \omega^2}{k} \right)^2 + \left(\frac{6\pi\eta_0 a \omega}{k} \right)^2. \tag{5}$$

The total sum of acoustic attenuation in the presence of external magnetic field is:

$$\alpha = \alpha_{rot} + \alpha_{tr}. \tag{6}$$

2.2. Model 2

In Shliomis, Mond, Morozov [9] model of the magnetic fluid considers mainly single particles (monomers) and dimers. The nanoparticles interact each separately ($\lambda < 5$) and do not create clusters. Although the number of dimers is rather small, nevertheless it is enough to provide the ultrasound absorption that is comparable with the viscous one. Of course, in the magnetic field higher oligomers can exist: trimers, tetramers, etc.

In this model is assumed only two forces that act on the dimers. The dimer represents a pair of nanoparticles with hydrodynamic diameter d_h . Nanoparticles are bound to each other due to the potential provided by magnetic dipole attraction and entropic and steric repulsion. So the first force is calculated as:

$$F_{1,j} = -\frac{\partial U}{\partial \vec{r}_j} = -\frac{\partial}{\partial \vec{r}_j} \left[\frac{1}{2} C (\vec{r} - \langle \vec{r} \rangle)^2 \right], \tag{7}$$

where C is the dimer's elastic constant; $j = 1, 2$; and $\vec{r} (\vec{r} = |\vec{r}_1(t) - \vec{r}_2(t)|)$ and $\langle \vec{r} \rangle$ are the instantaneous and the mean equilibrium distance between the nanoparticles, respectively. The second force, Stokes force is equivalent to force F_2 (Eq. 2) used in Taketomi theory. It is expressed as:

$$F_2 = -3\pi\eta_0 d_h (\vec{r}_j - \vec{v}), \tag{8}$$

where $\vec{v} = \vec{v}_0 \cos(\vec{k}\vec{r} - \omega t)$ is the fluid velocity.

Shliomis simplified the equations of motion for two particles ($j = 1, 2$) using the dimer's center of mass vector \vec{r}_c and the distance vector \vec{r} . Next, he used linearization the mean equilibrium distance: $\vec{r} = \langle \vec{r} \rangle + \varepsilon(t)$ plus another approximation. They were able

to determine the dissipative energy E_{dis} as Taketomi. In this model was also made the generalization for the effect of the N-particle oligomers with $N \geq 3$. The final relationship of the acoustic anisotropy attenuation is in the form:

$$\frac{\alpha(\varphi) - \alpha(\frac{\pi}{2})}{\alpha(0) - \alpha(\frac{\pi}{2})} = \frac{2(1 - S)\cos^2\varphi + 3\cos^3\varphi}{2 + S}, \tag{9}$$

where $\alpha(0), \alpha(\frac{\pi}{2})$ are real values from experiment. The degree of the anisotropy [9] is given by the function $S(\lambda\xi\theta) = \frac{1}{2} (3 \langle \cos^2\theta \rangle - 1)$ where θ is the angle between \vec{r} and the applied magnetic field \mathbf{B} .

3. Experimental Results

In these experiments we used same the experimental arrangement as in our previous works [14], [15]. The frequency of acoustic wave was 12,6 MHz. Transformer oil TECHNOL was used as the carrier liquid for the preparation of magnetic fluids for investigation by acoustic methods. The magnetic fluid (MF) used in experiments consisted of magnetite particles FeO, Fe₂O₃ with the mean diameter $a = 11,1$ nm, coated with oleic acid as a surfactant. The volume concentrations of magnetic particles were 1, 2 and 2,5 %. The basic properties of 2 % MF TECHNOL were following: the density 0,904 g/cm³ and saturation magnetization 9,1 mT. For this type of MF with hydrodynamic diameter $d_h=15$ nm is coupling constant $\lambda = 4,9$.

In our experiment we measured the change of acoustic attenuation as the function of external magnetic field (Fig. 2). From the obtained results it can be seen that with increasing magnetic field the acoustic attenuation also increases. When the magnetic field is swept at a constant rate the magnetic moment of the nanoparticle slowly move in the direction of this field. This interaction leads to the aggregation of nanoparticles to oligomers, chain and later to clusters [1], [5], [8]. The development of the attenuation of acoustic wave depends on the maximum of the external magnetic field and shows a hysteresis [2], [12], [15]. At higher maximum magnetic field the change of acoustic attenuation is more pronounced. This can be caused by more numbers and size of clusters. The process of agglomeration can continue also at degreasing magnetic field to certain saturated state. The value of acoustic attenuation does not return to the initial state immediately after the magnetic field has been removed. This effect can be caused by longer lifetime of clusters than time of decrease of the magnetic fluid.

The acoustic attenuation was investigated for various concentrations of MFs in the temperature range of 15–30 °C (Fig. 3). The obtained results indicate also the significant effect of temperature on the acoustic attenu-

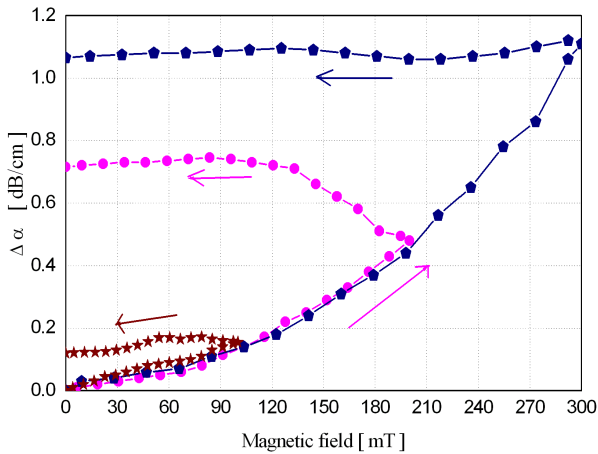


Fig. 2: Changes of the acoustic attenuation in external magnetic field for 2,5 % MF measured at 20 °C for different achieved maximum values of magnetic field (red - 100 mT, pink - 200 mT, blue - 300 mT). Magnetic field increased and/or decreased at a constant rate, 2,2 mT/min.

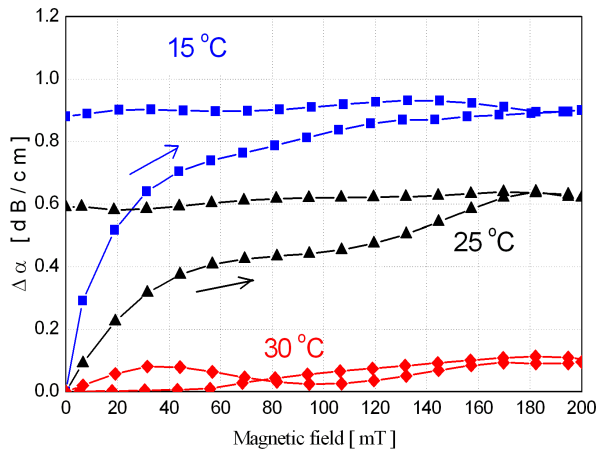


Fig. 3: The temperature influence of the attenuation changes on the external magnetic field for 1,0 % MF (blue 15 °C, black 25 °C, red 30 °C).

ation. The measurement of the acoustic attenuation at a lower temperature (15 °C) shows the largest changes originated from the process of the cluster creation and shows also higher hysteresis. At this temperature the creation of cluster is more effective because Brown thermal motion has not sufficient energy to destroy the clusters. At a higher temperature collision caused by thermal motion are more often and effective. These collisions can cause with higher probability decrease both numbers of clusters and their length. Above 30 °C the majority of the particles are not involved in the cluster structures. The influence of magnetic field on the acoustic attenuation happens small. The stability of such medium in a magnetic field is then better.

The results of the anisotropy of acoustic attenuation in the magnetic fluid of the value 200 mT are shown

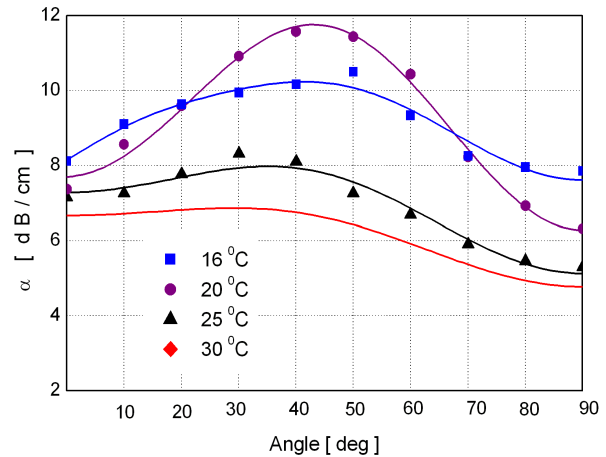


Fig. 4: Anisotropy measurement [15] of the acoustic attenuation at 200 mT external magnetic field in the 2 % MF and sum of the components $\alpha_{rot} + \alpha_{tr}$ of the Taketomi functions [10] (blue 16 °C, violet 20 °C, black 25 °C, red 30 °C).

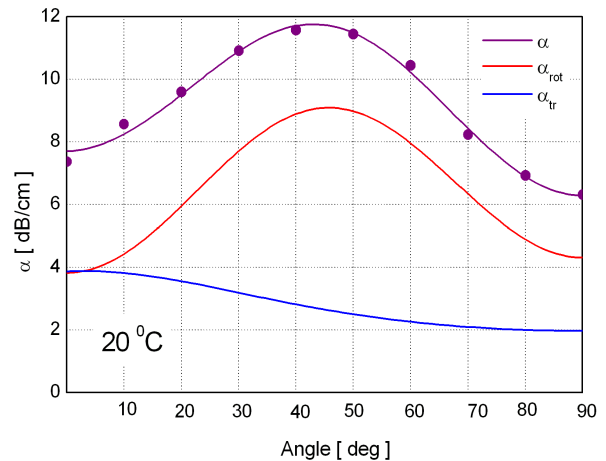


Fig. 5: Anisotropy measurement of the acoustic attenuation in external magnetic field 200 mT at the temperature 20 °C in the 2 % MF and the components $\alpha_{rot}, \alpha_{tr}$ from the Taketomi theory [12].

in Fig. 4. The measurements were made in dependence on the angle φ between the magnetic field \mathbf{B} and wave vector \vec{k} . Development of the anisotropy was investigated in the temperature range of 16–30 °C. The results indicate a significant effect of temperature on the anisotropy of acoustic attenuation in studied MF. The measurement at a lower temperature shows the largest changes of acoustic attenuation. These changes are caused by the arrangement of clusters in the direction of magnetic field and their change of orientation with respect to acoustic wave direction. The changes of acoustic attenuation are smaller at higher temperatures. We suppose that this effect is because that the clusters are smaller and chain of nanoparticles are shorter. The theoretical fit using the Taketomi functions (Eq. (1), (5)) to the experimental data are pre-

sented by solid lines also in Fig. 4. The part of acoustic wave energy used for stimulation of translational vibrations and rotational degrees of freedom were determined from these functions. Now it is possible also to estimate the percent contribution of acoustic attenuation from the individual kinds of cluster motion.

The representative anisotropy measurement of acoustic attenuation and its analysis by Taketomi model is illustrated in Fig. 5. In this figure are presented calculated values components for rotation (α_{rot}) and translation (α_{tr}). From Fig. 5 results that α_{rot} takes a maximum value about angle 55° , while $\alpha_{tr}(\varphi)$ decreases with to angle. These fits are presented also in normalized form in figures 6(a)—(c), for particular temperatures. By analyzing these results we can get more information about the influence of rotation and translation components to attenuation coefficient.

Tab. 1: Type values of parameters described 2 % magnetic fluid based on the TECHNOL obtained from the fit of the taketomi function to the experimental points.

	16 °C	20 °C	25 °C	30 °C
$\frac{4}{3}\eta_S + \eta_V$ [Nsm^{-2}]	0, 41	0, 36	0, 33	0, 28
α_5 [Nsm^{-2}]	0, 34	0, 83	0, 35	0, 23
α_1 [Nsm^{-2}]	-1, 0	-1, 7	-0, 54	-0, 30
k [Nm^{-1}]	22, 56	6, 90	1, 47	0, 49
r [nm]	300	138	71	37
$10^{-17}N$ [m^{-3}]	2, 35	12, 53	35, 63	250
$V \times N$ [10^{-3}]	3, 3	1, 72	0, 67	0, 66

Figure 6 shows normalized components of the acoustic wave rotation and translation absorption of the Taketomi functions (Eq. (1), (5)) for various temperatures (16 °C, 20 °C, 25 °C and 30 °C). As follows from the dependence $\alpha_{rot}(\varphi)$ in the range of the angle φ studied, the function takes a maximum value whose position depends on temperature (16–67 °C, 20–55 °C, 25–42 °C and 30–30 °C). On the other hand, the second component of the acoustic attenuation, $\alpha_{tr}(\varphi)$, decreases with to angle up to about 55° . Then its value slowly increases at higher temperatures.

From Fig. 6 it can be seen that at a lower temperature the translation part of acoustic absorption is dominant for small angles between wave vector \vec{k} and the magnetic field \mathbf{B} . At an angle $\sim 36^\circ$ these components are equal and the rotation component of the acoustic attenuation is dominant at higher angles. At temperature 20 °C components of absorption are same only in the parallel case of \vec{k} and \mathbf{B} . The rotation part of absorption is dominant for other angles and for higher temperatures. The reason for such behavior can be in the fact that at a higher temperature the magnetic

nanoparticles create chains rather than spherical clusters. The development of the anisotropy of acoustic attenuation of acoustic waves propagating in the magnetic fluid in the magnetic field was fitted by functions defined in Eqs. (1), (5) and (6). The parameters ($\frac{4}{3}\eta_S + \eta_V$), α_5 , α_1 , k , r and N from the fit are collected in Tab. 1. They describe the structure of a magnetic fluid subjected to an external magnetic field at various temperature.

Also from the values of a and N can be to determine the volume concentration of all clusters ($V \times N = 3, 3 \times 10^{-3}$ at the lowest temperature). This value implied which part (0, 3 %) of the magnetic grains (domains) are included in the clusters, while some other main part are free in the supporting fluid. The comparison of the volume of the magnetic nanoparticles (2 %) with the volume of the magnetic cluster of the magnetic fluid ($V \times N \approx 0, 33$ %, Tab 1.), indicates that only a small proportion (~ 15 %) of magnetic nanoparticles create the cluster structure [8].

4. Discussion

The observed results show the strong influence of the acoustic attenuation on the value of magnetic field in magnetic fluid based on transformer oil TECHNOL (Fig. 2). When the magnetic field is swept at a constant rate the magnetic moment of the nanoparticle slowly move in line of this field. This interaction leads to the aggregation of nanoparticles to chain-like formations and later to clusters with radius hundreds of nanometers (see Tab. 1). These clusters cause the increase of the acoustic attenuation with increasing external magnetic field. On the values of acoustic attenuation also influence the temperature as is shown in Fig. 3. With increasing temperature the speed of magnetic particles and their thermal energy (Brown motion) increase. Their collisions with clusters are more effective and can causes with higher probability decay of cluster. Decrease of the acoustic attenuation with temperature is caused also by a decrease of viscosity term (Eq. (1), Tab. 1). These effects agree with other works [5], [8].

The anisotropy of acoustic attenuation was investigated in the temperature range of 16–30 °C (Fig. 4). The obtained results indicate the significant effect of temperature on the acoustic attenuation. Measured developments of the anisotropy of the acoustic attenuation confirm our assumption, that nanoparticles are involved in the cluster structures. Clusters have the rotational and translational degrees of freedom, which are represented by two components of the acoustic attenuation α_{rot} and α_{tr} [8], [10]. At temperatures 16 and 20 °C are the largest changes coupled with the process of chains orientation in the direction of the field. The

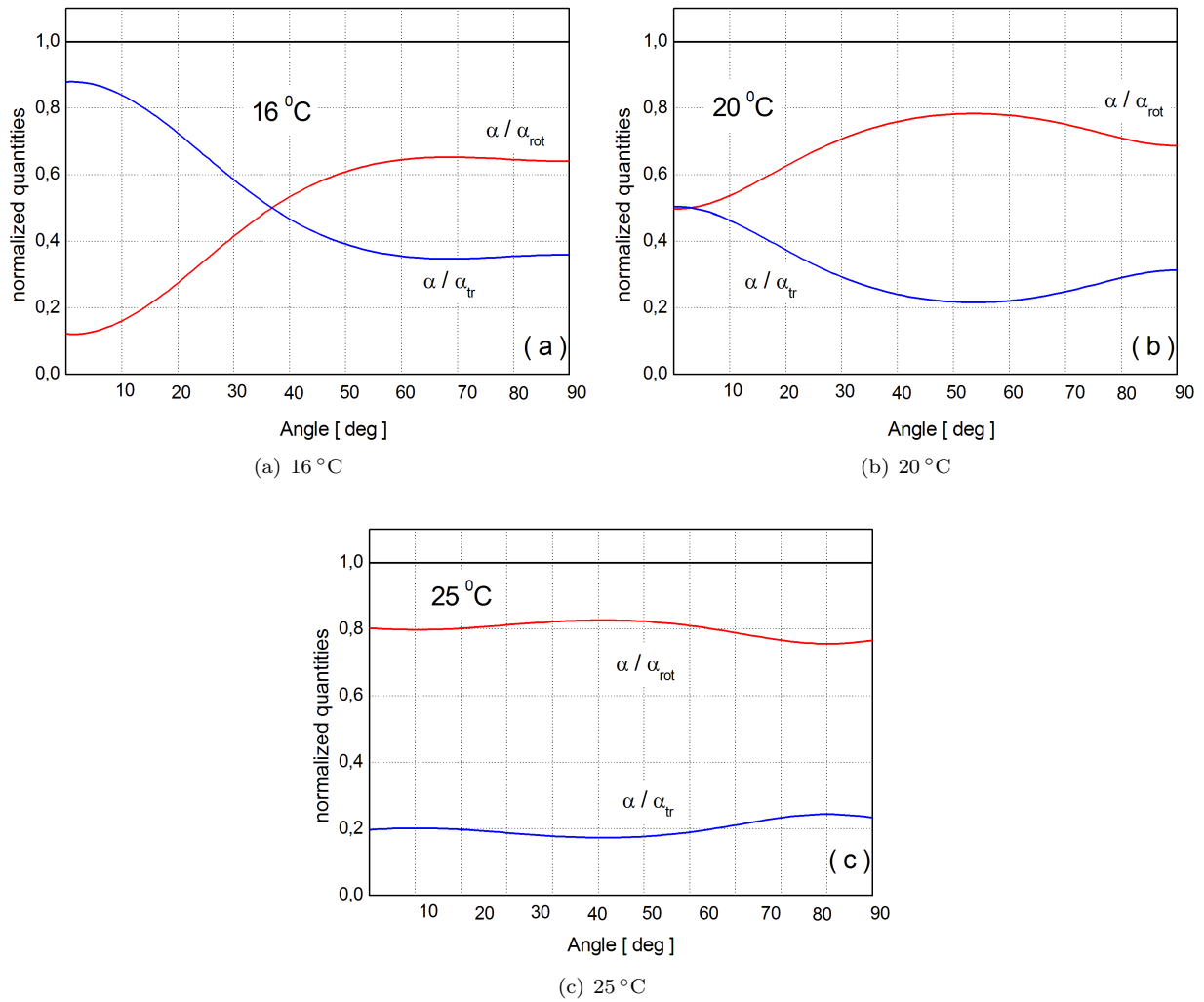


Fig. 6: The normalized components (α/α_{rot} and α/α_{tr}) of the Taketomi function in the 2 % MF based on TECHNOL at $B = 200$ mT calculated for (a) 16 °C, (b) 20 °C and (c) 25 °C.

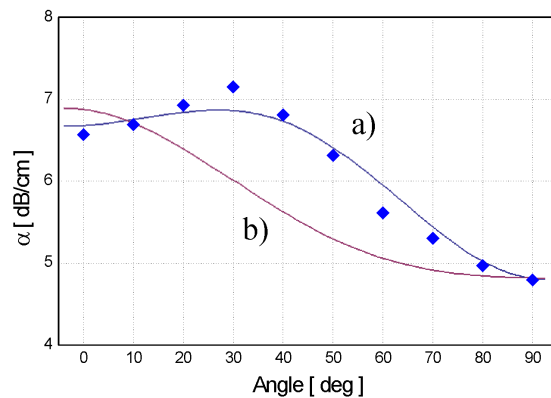


Fig. 7: Theoretical functions for the development of anisotropy acoustic attenuation ($B = 200$ mT, $T = 30$ °C) expressed as a) the sum of components $\alpha_{rot} + \alpha_{tr}$ (Eq. 5) from Taketomi theory and b) the Shliomis function (Eq. (9), $S = 0, 65$) compared with experimental results - blue.

translation part of cluster motion is comparable with their rotation part. Of course, there are also other effect influenced on the acoustic attenuation, like the radius of clusters and the length of their chains. At temperatures 25–30 °C the change of acoustic attenuation were less pronounced because rotation part was dominant (Fig. 6c).

Taketomi theory [10] was used (Eqs. (1), (5)) for the analysis of the angular dependence of the acoustic attenuation on the angle φ (Fig. 4). From this theory we were able to determine more information about existence, size and density of clusters. In the case of Shliomis theory [9] our results doesn't correspond with theoretical predictions. The absorption coefficient would be decrease with angle φ , but we had maxim at an angle around 400 for all measured temperatures (Fig. 4). Also from this theory could not be determined the characteristic parameters of the studied magnetic fluid.

Shliomis [9] in his theory predicts only translation and vibration motion of nanoparticles. From this and other assumption they derived function of translation acoustic attenuation defined by Eq. (9). In Fig. 7 are presented comparisons of the theoretical results of Taketomi (a) and Shliomis functions (b) of the acoustic attenuation at temperature 30 °C. As it can be seen from figure the curves are different. These differences are higher at lower temperatures. This is cases by ignoring the rotation motion of clusters in Shliomis theory, which is very important at big clusters or at higher coupling constant ($\lambda > 4$).

Temperature is very important parameter which has influence on the anisotropy of acoustic attenuation (Figs. 4, 5, 6) and also on the value of the clusters radius (Tab. 1). At a lower temperature (16 °C) the radius is much bigger than at a higher temperature. The calculated mean values change from 300 nm to 37 nm. In the first case the cluster consist of several tens of nanoparticles while at the radius 37 nm there is only about 5–9 magnetic nanoparticles. At a higher temperature the thermal motion increases resulting in a decrease of numbers of clusters and mainly their radius. The both smaller numbers of clusters and their shorter length induce the smaller influence on the acoustic attenuation. The influence of magnetic field on the acoustic attenuation happens small at above 30 °C [12], [14], and as follows from our results the majority of the particles are not involved in big cluster structures.

5. Conclusion

Two theories describing the anisotropy of acoustic attenuation that occurs in magnetic fluid were described

and compared. The acoustic spectroscopy was used to observe the influence of both magnetic field and temperature on the structures of TECHNOL transformer-oil based magnetic fluid. We measured and analyzed the anisotropy of the acoustic attenuation in the presence of external magnetic field of the value 200 mT. It was confirmed that the effect of external magnetic field on the creation of clusters of nanoparticles in magnetic fluids. The study of the anisotropy showed the important role translational and rotational motion of the clusters to the acoustic attenuation. Using Taketomi theory we are able to determine the radius of clusters, their density and viscous term as the function of temperature.

The transformer oil is usually used for high voltage insulation and power transformer cooling. The transformer oil-based magnetic fluids can improve some of the oils insulating and thermal properties. The dielectric breakdown strength of transformer oil is strongly influenced by the aggregation effects of magnetic particles and can induce electric breakdown.

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