



## Temperature dependence of the FMR absorption lines in viscoelastic magnetic materials

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### ABSTRACT

The magnetic properties of the viscoelastic materials filled with magnetic nanoparticles strongly depend on the viscosity of these materials. The time–temperature changes of the viscosity can affect the orientation ordering of the anisotropy axes of the magnetic nanoparticles after an external magnetic field is applied. In consequence, the absorption lines obtained in the ferromagnetic resonance experiment (FMR) will possess an additional temperature dependence through the viscosity of the materials under consideration. The particular case of the temperature dependence of the FMR signal detected from the  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> (maghemite) magnetic nanoparticles dispersed at low concentration (0.1 wt.%) in a poly(ether-ester) multiblock copolymer (PEN-block-PTMO) matrix has been investigated. A strong increase of the resonance line amplitude is observed with increasing temperature. We note also two qualitatively different temperature dependences of the magnetic resonance field at low and high temperatures. A simplified theoretical model of a single cluster consisting of *N* magnetic nanoparticles in an elastic polymer matrix is introduced to explain these temperature dependences of the absorption lines. The model is based on the stochastic version of the Lifshitz–Landau equation for nanoparticle magnetization.

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### 1. Introduction

Recent investigations on the magnetic nanoparticles dispersed in non-magnetic matrix show that the main peak corresponding to the uniform resonance mode in the FMR spectra can be accompanied by a series of other peaks which originate both from a spin-wave exchange model [1,2] and dipolar inter-particle interactions as well as they can be coupled with the magneto-elastic phenomena. The latter case has been discussed in [3] concerning the FMR experiment with the  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> ferrimagnetic nanoparticles embedded in a multiblock poly(ether-ester) copolymer non-magnetic matrix. Their results suggest that some additional peaks in low temperatures originate from the orientational anisotropy of frozen polymer blocks. This suggestion has been confirmed theoretically in one of the last papers [4] where a simple stochastic model for non-interacting magnetic nanoparticles in a non-magnetic polymer matrix has been introduced. The orientational dependence of the FMR spectra has been found earlier by Owens [5] for a colloidal suspension of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles which have been solidified in a constant magnetic field (dc magnetic field). Similar observa-

tion has been found theoretically in a recent paper by Sukhov et al. [6]. In their paper there is a very interesting discussion on the shape of the ferromagnetic resonance spectra for the ensemble of the randomly distributed magnetic anisotropy axes as well as the discussion of the dependence of these spectra on temperature. They used a stochastic model which shares many features common with the experimental results, like the broadening of the FMR signal for the randomly distributed magnetic anisotropy axes as compared to the magnetic nanoparticles which all have the same orientation of the magnetic anisotropy. The model [6] is restricted to the case when the orientation of each anisotropy axis is frozen during computer simulation. The experiments with the magnetic nanoparticles dispersed in a block copolymer matrix [3,7,8] suggest that the assumption of the frozen anisotropy axes can be too restrictive for this type of material to explain the specific temperature dependence of the resonance amplitude. The example of this behaviour is presented very well in Fig. 3 in paper [7] where the absorption lines derivatives,  $d\chi''/dH_{dc}$ , have been plotted for magnetic agglomerates dispersed in a block copolymer. The symbols  $\chi''$  and  $H_{dc}$  represent the “out-of-phase” component of the complex ac susceptibility

$$\chi = \chi' - i\chi'', \quad (1)$$

and the external dc magnetic field experienced by the magnetic nanoparticles, respectively.

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In the text below we discuss the results of measuring the temperature dependence of FMR spectra of 0.1%  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> in PEN-block-PTMO matrix. A simplified theoretical model of a single cluster consisting of N magnetic nanoparticles in an elastic polymer matrix is introduced to explain this temperature dependence of the absorption lines.

## 2. Experimental

The ethylene 2,6-naphthalene dicarboxylate ester and tetramethylene oxide multiblock copolymer containing maghemite magnetic nanoparticles (0.1 wt.%) were synthesized by melt polycondensation of dimethylene naphthalate (DMN), ethylene glycol (EG) and poly(tetramethylene glycol) (PTMEG) in the presence of tetrabutyl orthotitanate as catalysts with phenolic antioxidants IRGANOX 1010 (Ciba Geigy) as stabilizers. Maghemite in the ethylene glycol was grounded and stirred for 5 min at 20,000 rpm and next the dispersion was sonicated for 5 min (Sonoplus Homogenisator HD 2200) and the procedure was repeated six times. All substrates were introduced into the reactor, where the two-stage process of PEN-block-PTMO synthesis proceeded with continuous mixing. The transesterification process of DMN with PG was carried out under atmospheric pressure in the temperature range of 150–190 °C. The progress of the reaction was monitored by the amount of distilled methanol in reaction relative to the 90% theoretical amount. The transesterification of di(2-hydroxyethylene) naphthalate with PTMEG and the polycondensation were performed. The process of polycondensation proceeded at 200–260 °C under a pressure of 0.1 hPa and was carried out until the desired torque value was achieved. The polymer composite filled the reactor by compressed nitrogen in the form of a filament. Scanning electron microscopy (SEM) (digital scanning electron microscopy Zeiss LEO) was used for characterization of the sample. Ferromagnetic resonance measurements were performed on powder samples sealed in quartz tubes using a Bruker E 500 X-band spectrometer ( $f=9.45$  GHz) with 100 kHz field modulation and an Oxford helium flow cryostat for temperature dependent measurements (3.5–300 K).

## 3. Experimental results

Fig. 1 presents the SEM picture of 0.1%  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> in PEN-block-PTMO matrix. The SEM shows almost homogenous system of magnetic nanoparticles in the agglomerate states with sizes below 100 nm. Five different parts of the same bulk sample have produced almost the same FMR spectra at room temperatures what suggest that the agglomerates are homogeneously dispersed in the matrix. The temperature dependence of the FMR spectra of 0.1%  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> in PEN-block-PTMO matrix is shown in Fig. 2. The spectrum is dominated by an intense slightly asymmetric single resonance line at high region of temperatures. For the clarity of presentation a few low-temperature spectra is also shown in Fig. 3.

The peculiar feature of the synthesized PEN-block-PTMO copolymer is that the dispersed magnetic nanoparticles form clusters. In the clusters the interparticle dipole–dipole magnetic interaction becomes important as well as the interaction of the magnetic nanoparticles with a non-magnetic matrix. This makes

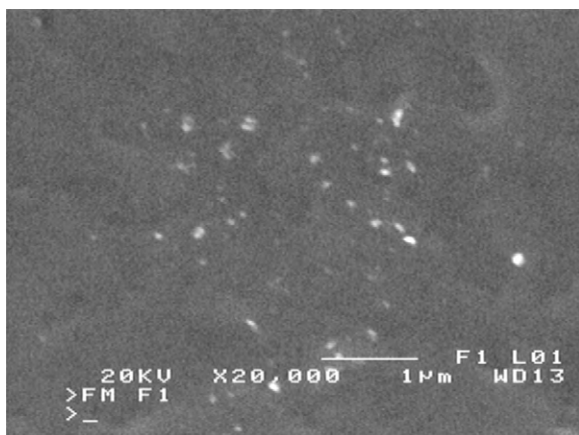


Fig. 1. SEM picture of magnetic nanoparticles 0.1%  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> in PEN-block-PTMO matrix.

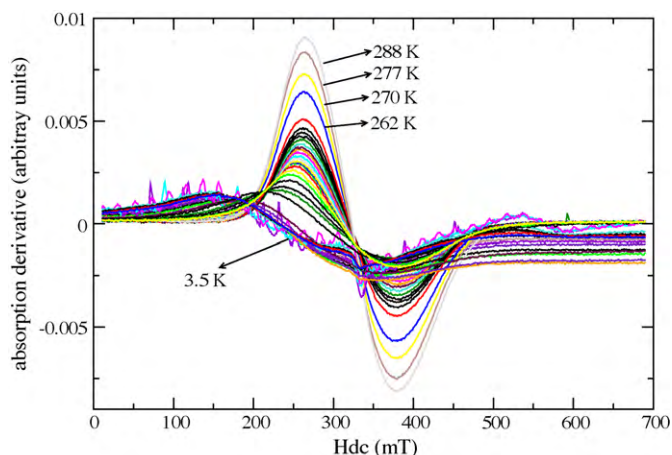


Fig. 2. The temperature dependence of FMR spectra (the derivative of the out-of-phase susceptibility,  $d\chi''/dH_{dc}$ ) of magnetic nanoparticles 0.1%  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> in PEN-block-PTMO matrix in the wide range of temperatures 3.5–288 K.

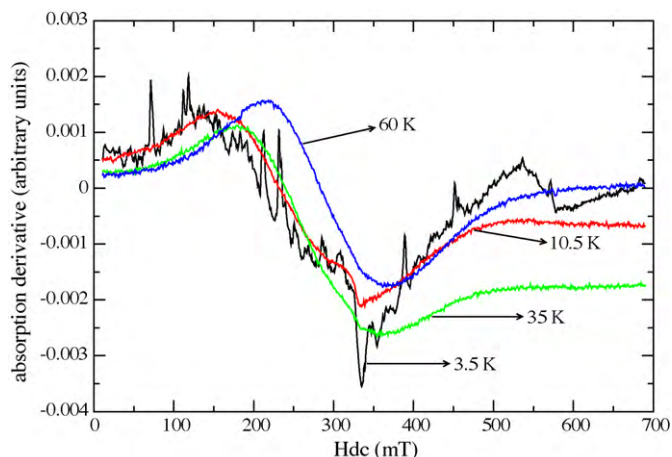


Fig. 3. A few absorption lines derivatives chosen from Fig. 2 in the range of low temperatures.

the magnetic relaxation to be a complex process. The question arises whether the completely different FMR spectra presented in Fig. 2 in the range of low temperatures and high temperatures could suggest a phase transition connected with  $\alpha$ ,  $\beta$  or  $\gamma$  relaxation processes. These relaxation processes could give a smooth transition between two asymptotically regions with high viscosity and small viscosity of the polymer matrix. These two ranges of temperatures are evident in the plot of the resonance field  $H_r$  in Fig. 4.

It is necessary to take into account that temperature dependence of polymer viscosity can substantially influence the dynamics of magnetic nanograins in low and high temperatures. In low temperature region we have solid-like non-magnetic matrix where magnetic relaxation takes place only through the process of magnetization relaxation (Neél relaxation). It is not the case of high temperatures where magnetic relaxation takes place both through the magnetization relaxation and rotation of the whole nanoparticle in a non-magnetic surrounding (Brownian relaxation). These two processes are typical for ferrofluids and they have different relaxation time scale.

## 4. Theory

We show that the particular temperature dependence of the FMR spectra observed in Figs. 2 and 4 as well as in the exper-

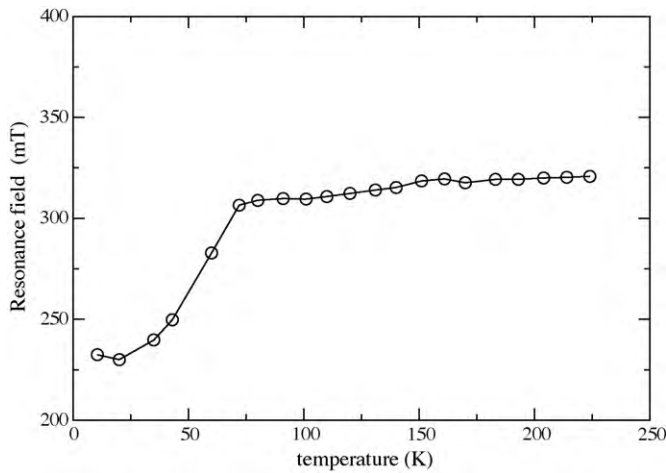


Fig. 4. Dependence of the resonance field  $H_r$  on temperature for magnetic nanoparticles 0.1%  $\gamma$ - $\text{Fe}_2\text{O}_3$  in PEN-block-PTMO matrix.

iments [3,7,8] can be explained by different elastic response of the polymer matrix to the mechanical stress produced by the rotating nanograins in an applied external dc magnetic field and the effect of the magnetic dipole–dipole interactions between the magnetic nanoparticles on the process of their orientation. To this aim we extended a simplified theoretical model constructed for non-interacting magnetic nanoparticles [4] to the case when agglomerates of magnetic nanograins can appear in a polymer matrix. In the model, the magnetic anisotropy axes can undergo rotational oscillations in an elastic medium but the distances between the magnetic nanoparticles stay frozen during computer simulation. Two assumptions have been introduced. The first one is assuming an empirical model for the viscosity parameter  $\nu$ , the Arrhenius law,

$$\nu(T) = \nu_0 e^{E/kT} \quad (2)$$

of the magnetic nanograins within the polymer matrix where  $E$  is the activation energy. The Williams–Landel–Ferry model is usually used to describe polymer melts relating to glass transition but it appears that assuming the simple Arrhenius law is sufficient for our model. The second assumption is introducing the Bloch law approximation

$$M_s(T) = M_0(0) \left( 1 - \left( \frac{T}{T_0} \right)^\alpha \right) \quad (3)$$

for the magnetization of the magnetic nanoparticles where  $T$  is temperature,  $\alpha = 1/3$  and  $T_0$  is some constant. It has been shown in paper by Zhigao and Youwei [9] that for the pure cluster, the Bloch law in Eq. (3) is well satisfied at low temperatures  $T < 0.5T_c$ . Another value of  $\alpha$  can be also found in publications on magnetic materials.

The model under consideration represents a cluster consisting of  $N$  magnetic nanograins embedded randomly into a non-magnetic polymer matrix. Each of the magnetic nanograins  $i = 1, 2, \dots, N$  has magnetization

$$\vec{M}_i = M_s V \vec{s}_i, \quad (4)$$

where  $M_s$  represents saturation magnetization and  $V$  is the nanoparticles' volume  $V$  which are the same for each nanograin,  $\vec{s}_i$  represents a unit vector in the direction of  $\vec{M}_i$ .

In the FMR experiments two external magnetic fields are switched on, a dc magnetic field  $\vec{H}_{dc}$  and  $\vec{H}_{ac}$  which denotes an alternating magnetic field. We have chosen the dc field to be in the

$z$ -direction and the ac field to be in the  $x$ -direction [2],

$$H_x = H_{ac} = H_{ac}^0 \cos(\omega t), \quad (5)$$

where  $\eta = f = 9.37$  GHz and  $\omega = 2\pi f$ .

In the cluster, the dipole–dipole inter-particle interactions are included. Thus, each nanograin  $i$  experiences an effective magnetic field  $\vec{H}_{eff,i}$  of the form [10]:

$$\vec{H}_{eff,i} = \vec{H}_{dc} + \vec{H}_{ac} + \frac{H_a}{|\vec{M}_i|} (\vec{M}_i \cdot \vec{n}_i) \vec{n}_i - \frac{M_s V}{4\pi} \sum_{j=1, j \neq i}^N \left( \frac{\vec{s}_j}{r_{ij}^3} - 3 \frac{(\vec{s}_j \cdot \vec{r}_{ji}) \vec{r}_{ji}}{r_{ji}^5} \right). \quad (6)$$

It is assumed that magnetic nanoparticles are immersed in a non-magnetic polymer matrix in such way that they become additionally coated with a polymer layer representing a rigid sphere of diameter  $R$  and the layer is pinned to the regions of the ends of the easy axis of the magnetic nanoparticles. The sphere makes rotational oscillations in a flexible non-magnetic matrix with a spring constant  $K_{el}$ . The total energy of a magnetic nanoparticle  $i$  with an uniaxial anisotropy as its dominating magnetic anisotropy [11,12] is given by the following form:

$$E_i = -K_a V \sin^2(\psi_i) - \mu_0 M_s V H_{eff,i} \cos(\phi_i) \quad (7)$$

where  $K_a$  is anisotropy constant,  $\psi_i$  is the angle between the particle magnetization  $\vec{M}_i$  and its magnetic anisotropy axis,  $\mu_0$  is constant of permeability, and  $\phi_i$  is the angle between  $\vec{M}_i$  and  $\vec{H}_{dc}$ . The symbol  $\vec{n}_i$  in Eq. (6) is a unit vector along the magnetic anisotropy direction of the nanograin  $i$  with the components

$$n_{x,i} = \sin(\phi_i) \cos(\theta_i), \quad (8)$$

$$n_{y,i} = \sin(\phi_i) \sin(\theta_i), \quad (9)$$

$$n_{z,i} = \cos(\phi_i). \quad (10)$$

and  $\phi_i$  and  $\theta_i$  are the angles vector  $\vec{n}_i$  makes with the  $z$ -axis and  $x$ -axis, respectively. The symbol  $H_a$  in Eq. (6) represents the magnetic anisotropy field which is defined as

$$H_a = \frac{2K_a}{\mu_0 M_s}. \quad (11)$$

The magnetization dynamics is described with the help of the stochastic version of the Landau–Lifshitz equation [13,14] for nanoparticle magnetization:

$$\frac{d\vec{M}_i}{dt} = -\gamma \vec{M}_i \times [\vec{H}_{eff,i} + \vec{B}_i] - \alpha \frac{\gamma}{M_s V} \vec{M}_i \times (\vec{M}_i \times [\vec{H}_{eff,i} + \vec{B}_i]), \quad (12)$$

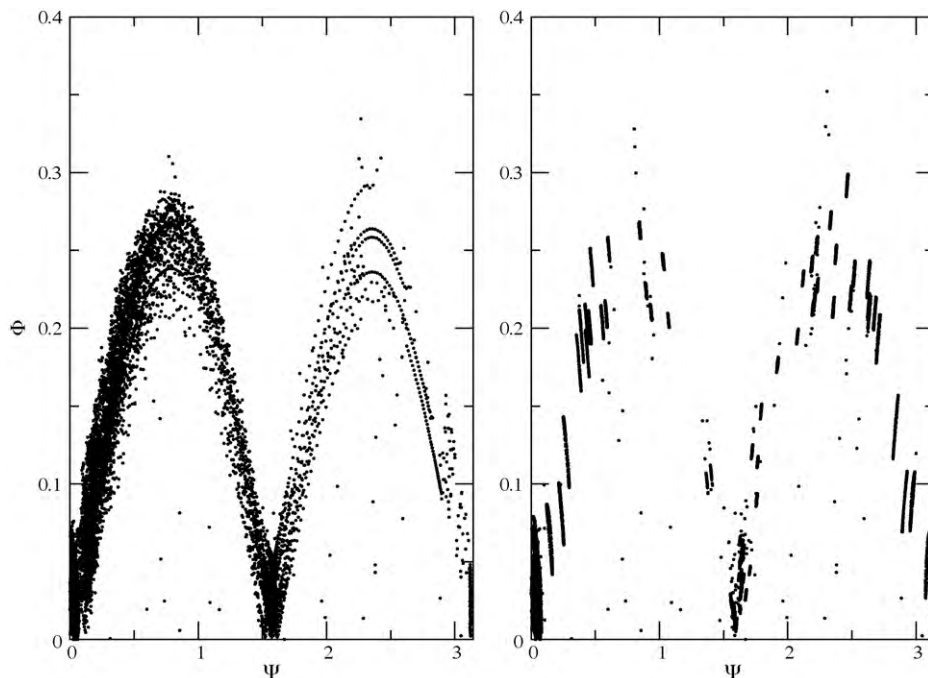
where  $i = 1, 2, \dots, N$ , and the constants  $\gamma$  and  $\alpha$  denote the gyro-magnetic ratio and the damping constant, respectively. As in the paper by Jönsson [11] and Usadel [15] we assume white-noise fluctuations which represent  $\vec{B}_i$ . Then the thermal averages of  $\vec{B}_i = (B_{x,i}, B_{y,i}, B_{z,i})$  fulfill the relations:

$$\langle B_{q,i}(t) \rangle = 0, \quad q = x, y, z, \quad (13)$$

$$\langle B_{q,i}(t) B_{p,i}(t') \rangle = \frac{2\alpha k_B T}{m\gamma} \delta_{q,p} \delta(t-t'), \quad p = x, y, z. \quad (14)$$

The rotational dynamics of the magnetic anisotropy axes is considered in the diffusion limit where the inertial effects of the rotating sphere with a radius  $R/2$  can be neglected. Then, the dynamics are described with the help of two Langevin equations for the angles  $\phi_i$  and  $\theta_i$  and they take the following form introduced in [4]:

$$\frac{d\phi_i}{dt} = -\frac{2}{R\xi} |K_a V \sin(2\psi_i)| \sin(\phi_i - \phi'_i) - \frac{K_{el}}{\xi} \sin(\phi_i - \phi_{0,i}) + \frac{1}{\xi} \lambda_{\phi_i}, \quad (15)$$

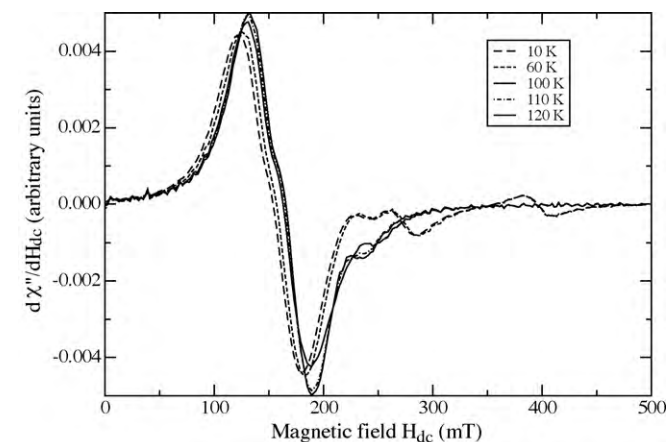


**Fig. 5.** Histogram of the values of the angles  $\psi$  (the angle the grain magnetization makes with the anisotropy axis) and  $\phi$  (the angle the grain magnetization makes with the dc magnetic field direction) of the nanoparticle's magnetization after long time computer simulation for an ensemble of  $N = 100$  magnetic nanoparticles with initially randomly distributed magnetic anisotropy axis.

$$\frac{d\theta_i}{dt} = -\frac{2}{R\xi} |K_a V \sin(2\psi_i)| \sin(\theta_i - \theta'_i) - \frac{K_{el}}{\xi} \sin(\theta_i - \theta_{0,i}) + \frac{1}{\xi} \lambda_{\theta_i} \quad (16)$$

where  $\xi$  represents the friction of the  $i$ th nanoparticle in the elastic non-magnetic polymer matrix and  $\lambda_{\psi_i}$  and  $\lambda_{\theta_i}$  represent the white-noise driving torque [16,17] for  $i$ th nanoparticle. They are stochastic equations where the thermal rotational fluctuations of the  $i$ th magnetic nanoparticle are characterized by temperature  $T$  and  $\lambda_{\psi_i}$  and  $\lambda_{\theta_i}$  fulfill the relations:

$$\langle \lambda_{\psi_i}(t) \rangle = 0 \quad (17)$$



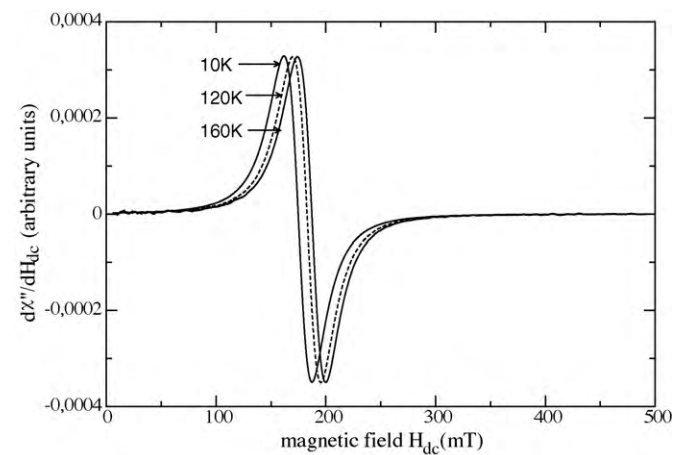
**Fig. 6.** Absorption lines derivatives,  $d\chi''/dH_{dc}$  (the derivative of the out-of-phase susceptibility) resulting from theoretical model under consideration for a cluster consisting of  $N = 30$  magnetic nanograins. Simulation data are averaged over 4 bins and each bin is equal to 2 mT. Some parameters of the computer simulation: demagnetizing factor in shape anisotropy  $D = 0.15$  ( $K_a = \mu_0(1 - 3D)M_s^2/4$ ),  $\alpha = 0.066$ ,  $R = 2$  nm,  $M_s = 450$  kA/m. The pre-exponential factor  $\nu_0 = 0.35$  Pa s in the expression for the viscosity  $\nu(T)$  (Eq. (2)) and the activation energy  $E = 0.1$  eV.

$$\langle \lambda_{\psi_i}(t) \lambda_{\psi_j}(t') \rangle = 2k_B T \xi \delta(t - t'), \quad (18)$$

where  $q = \psi_i, \theta_i$ . The angles  $\psi'$  and  $\theta'$  represent the angles which the magnetization  $\vec{M}$  makes with z-axis and x-axis, and the angles  $\psi_0$  and  $\theta_0$  are the initial angles of the easy axis after the magnetic nanoparticle has been built into polymer matrix. The numerical scheme applied to the stochastic equations is the Euler–Maruyama method.

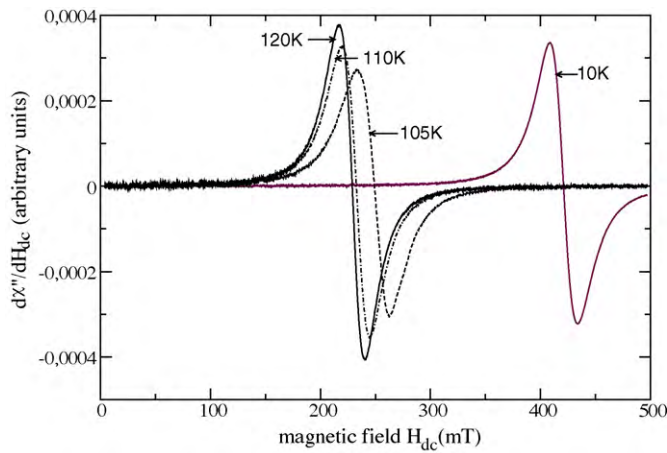
In the model under consideration the components of the complex ac susceptibility (Eq. (1)) have been calculated by performing the Fourier transform on the time averaged x-component of the magnetization, i.e.,

$$\chi = \frac{1}{\tau H_{ac}^0} \int_0^\tau dt M_x(t) e^{-i\omega t}, \quad (19)$$



**Fig. 7.** Absorption lines derivatives,  $d\chi''/dH_{dc}$  in theoretical model for a single ( $N = 1$ ) magnetic nanograin in the case when initially its magnetic anisotropy axis is parallel to the direction of the external dc magnetic field. The plotted curves correspond to temperatures  $T = 10, 120,$  and  $160$  K, respectively. Simulation data are averaged over 100 bins and each bin is equal to 0.05 mT. The parameters are the same as in Fig. 6.





**Fig. 8.** Absorption lines derivatives,  $d\chi''/dH_{dc}$  in theoretical model for a single ( $N=1$ ) magnetic nanograin in the case when initially its magnetic anisotropy axis is perpendicular to the direction of the external dc magnetic field. The plotted curves correspond to temperatures  $T=10, 105, 110,$  and  $120$  K, respectively. The parameters are the same as in Fig. 7.

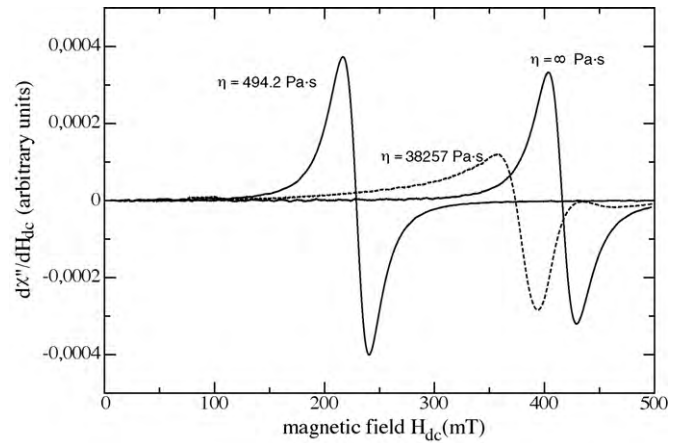
where  $\tau = 1/f$ . The absorption spectra coinciding with the ones presented in Fig. 3 in [7] can be obtained in the case when the viscosity  $\nu$  of the polymer becomes temperature dependent. In particular,  $\nu$  can take the form from Eq. (2). It is related to friction parameter  $\xi$  as follows:

$$\xi = 8\pi\nu(T)r^3, \quad (20)$$

where  $r=R/2$  denotes the radius of polymer sphere representing magnetic nanoparticle and its polymer coating. The additional assumption in Eq. (3) makes possible to introduce dependence of the saturation magnetization on temperature.

## 5. Discussion of the results

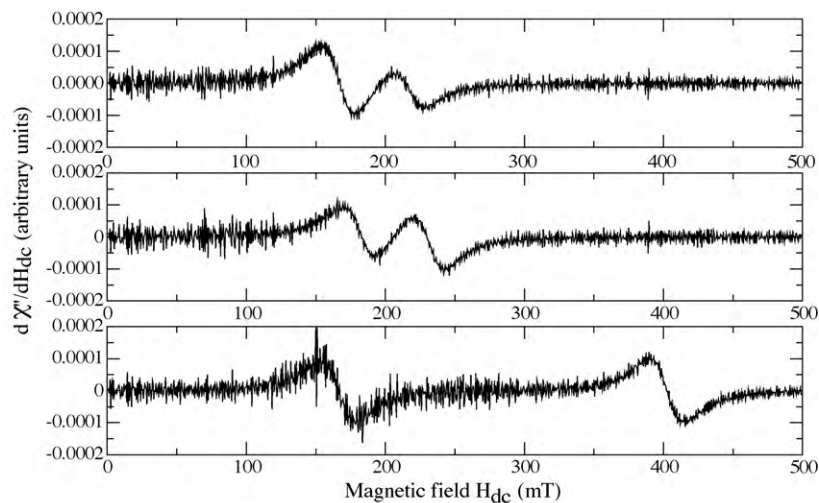
In the model under consideration, the orientation of the nanoparticle's magnetization  $\vec{M}_i$  ( $i=1, 2, \dots, N$ ) with respect to magnetic easy axis and external dc magnetic field is determined by the angles  $\psi_i$  and  $\phi_i$ , respectively. The larger the value of the anisotropy constant  $K_a$  the larger is the coupling between the nanoparticle's magnetization and the anisotropy field  $\vec{H}_a$ . In the case of the large



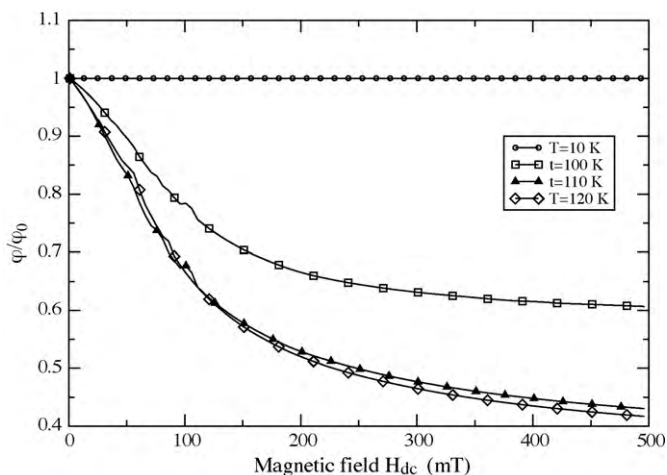
**Fig. 9.** The absorption lines derivatives,  $d\chi''/dH_{dc}$ , which are calculated for the single magnetic nanoparticle from Fig. 8 in the case when the viscosity parameter  $\nu$  does not depend on temperature. The plotted curves correspond to the same value of temperature,  $T=120$  K, and three different values of the viscosity,  $\infty, 38,257,$  and  $494.2$  Pa·s.

value of  $K_a$  and the small value of the viscosity parameter  $\nu$  of the non-magnetic matrix, e.g. in liquids, the movement of the nanoparticle's magnetization becomes strongly correlated with the rotational movement of the whole nanoparticle. In the opposite case of the large value of  $\nu$  the magnetic nanoparticles stay frozen into their surrounding and a Brownian-type rotation of their magnetic moment in an uniaxial anisotropy potential dominates the thermalization process. In the latter case the FMR spectra are characterized by multimodal absorption lines as in Fig. 3. A distinction between the case when the magnetic nanoparticles are dispersed in an elastic medium with small viscosity and solid medium (large viscosity) has been presented in Fig. 5 where a histogram of the values  $\psi$  and  $\phi$  collected from the computer simulations is plotted. The simulations were performed on a random cluster consisting of  $N=100$  magnetic nanoparticles.

The experimental FMR spectra in Fig. 2 show a particular temperature dependence such that the resonance amplitude is increasing with temperature. Our theoretical model shares qualitatively the same property of the FMR spectra. This can be observed in Fig. 6, where the absorption lines derivatives,  $d\chi''/dH_{dc}$ , have

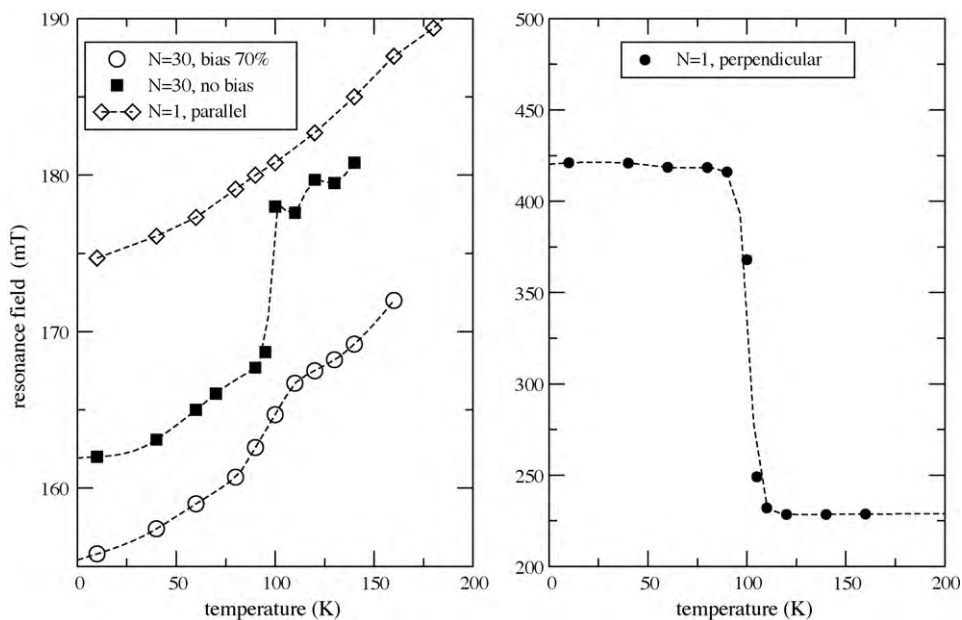


**Fig. 10.** Absorption lines derivatives,  $d\chi''/dH_{dc}$  in the case of two magnetic nanoparticles at temperature  $T=110$  K with two different initial orientations of the magnetic anisotropy axes (parallel and perpendicular to dc magnetic field). Upper panel: the dipole-dipole magnetic interaction is excluded. Middle panel: the dipole-dipole magnetic interactions is present. Bottom panel: the dipole-dipole magnetic interactions is present but the viscosity has a large value and it does not depend on temperature. The remaining parameters as in Fig. 6.



**Fig. 11.** Dependence of the average angle,  $\varphi = \frac{1}{N} \sum_{i=1}^N \varphi_i$ , the magnetic anisotropy axis makes with the direction of the  $H_{dc}$  on the strength  $H_{dc}$  for different temperatures. The symbol  $\varphi_0$  represents an initial angle (Eq. (15)). The data represent the same FMR experiment as in Fig. 6.

been plotted for the particular case of a magnetic agglomerate consisting of  $N=30$  magnetic nanograins. In the model under consideration, the temperature dependent viscosity  $\nu(T)$  has the most important effect on the increase of the resonance amplitude with increasing temperature. It is assumed that the viscosity obeys the Arrhenius law introduced in Eq. (2). In the majority of the computer simulations under consideration we have chosen the pre-exponential factor  $\nu_0 = 0.35$  Pa s and the activation energy  $E = 0.1$  eV. This choice ensures that the values of the viscosity  $\nu(T)$  at high temperatures appear to be close to the experimentally measured viscosity of the thermoplastic polymers (like PEN composites) [18,19]. In this case, the viscosity at  $T = 290^\circ\text{C}$  as a function of shear rate is changing from a few Pa s to the values of the order of  $10^3$  Pa s.



**Fig. 12.** Dependence of the resonance field  $H_r$  on temperature in theoretical model under consideration. The clusters consisting of  $N=1$  and  $N=30$  magnetic nanoparticles are considered. Two cases of the single magnetic nanoparticle are presented, the one with the magnetic anisotropy axis parallel to the dc magnetic field and the one with the perpendicular magnetic anisotropy axis. The agglomerates consisting of  $N=30$  magnetic nanoparticles represent two cases: when all magnetic nanoparticles are randomly oriented and when 70% of them is aligned with the dc magnetic field.

The effect of the temperature dependence of the viscosity on the absorption lines as well as the influence of the magnetic dipole–dipole interaction on them is evident already in the case of a single magnetic nanograin (Figs. 7–9).

In Fig. 7 the derivatives of the absorption lines are shown in the case of the magnetic anisotropy axis initially parallel to the external dc magnetic field. As it is expected the plots corresponding to the higher temperatures are shifted into the right-hand side direction of the figure with respect to the lines representing the lower temperatures. On the other hand, in the case of the magnetic anisotropy axis initially perpendicular to the external dc magnetic field (Fig. 8) the value of the resonance field decreases in a wide range of temperatures. It starts to increase only at some higher temperatures. The results of Figs. 7 and 8 can be compared with the results presented in Fig. 9 where the magnetic anisotropy axis is initially perpendicular to the external dc magnetic field and the viscosity  $\nu$  does not depend on temperature. In Fig. 10 the case of two magnetic nanoparticles is considered with the magnetic anisotropy axis being parallel and perpendicular to the dc magnetic field, respectively. Note that the dipole–dipole magnetic interaction strongly influences the value of the resonance magnetic field. All these factors: the temperature dependent viscosity, magnetic dipole–dipole interaction and orientation of the magnetic anisotropy axes with respect to the dc magnetic field contribute to the increase of the resonance amplitude presented in Fig. 6.

The value of the viscosity parameter  $\nu(T)$  has an effect on the elastic–magnetic coupling presented in Fig. 11. In the figure the angle  $\varphi$  the anisotropy axis makes with the dc magnetic field versus the value of the dc magnetic field is plotted and  $\varphi$  is averaged over all angles  $\varphi_i$  ( $i = 1, 2, \dots, N$ ) of the magnetic nanoparticles in the cluster. It is evident from Fig. 11 that in elastic medium the stronger the magnetic dc field the better is the aligning the anisotropy axis with the direction of the magnetic field.

The results of the computer simulation in Fig. 12 of the resonance field  $H_r$  suggest two separate temperature ranges, the ‘low temperature’ and ‘high temperature’ range when a large agglomerate of the magnetic nanoparticles is created. The observed qualitatively different dependence of  $H_r$  on temperature  $T$  for

the single nanoparticle with different orientation of the magnetic anisotropy axes (parallel and perpendicular to dc magnetic field) suggests that in a large cluster of magnetic nanoparticles a random orientation of the magnetic anisotropy axes is the reason for different 'low temperature' and 'high temperature' behaviour of  $H_r$ . This can be the explanation of the experimental results in Fig. 4.

## 6. Conclusions

We have shown that temperature dependence of the viscosity in the block copolymers can affect the absorption lines in FMR experiments. Namely, in the temperature range where the viscosity parameter drops down the magnetic nanoparticles are not frozen into random directions any more but they have possibility to become oriented to dc magnetic field. This process of orientation of the magnetic nanoparticles can be the major mechanism for the unusual increase of the resonance amplitude with increasing temperature in a poly(ether-ester) multiblock copolymer (PEN-block-PTMO) matrix. In the theoretical model under consideration the distances between the magnetic nanoparticles are kept constant and therefore the model cannot describe properly the whole range of temperatures. This kind of experimental and theoretical study could be important for characterization of different materials with the very low or ultra-low concentration of magnetic nanoparticles.

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