



# Magnetic aging in magnetic fluids: a static magnetic birefringence investigation

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## Abstract

The effect of magnetic aging on the static magnetic birefringence (SMB) data of a nickel ferrite-based ionic magnetic fluid was investigated. The traditional SMB model was improved to account for both the formation of agglomerates and the field dependence of the relative permeability associated to monomers and chain-like structures. Improvement of the SMB model allows determination of the sample polydispersity profile in excellent agreement with the transmission electron microscopy data. © 2001 Elsevier Science B.V. All rights reserved.

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Magnetic fluids (MFs) consist of magnetic nanoparticles dressed with a surface molecular layer and dispersed either in aqueous or non-aqueous media [1]. The strong response of concentrated MFs to an applied magnetic field ( $H$ ) has offered many options of technological applications [2]. In spite of the particular method used to produce nanosized particles it is well known from the transmission electron microscopy (TEM) data that the nanoparticle diameter ( $D$ ) distribution is adequately described by the lognormal probability function [3]. The static magnetic birefringence (SMB) measurements have been widely used to unfold the size polydispersity profile of an MF sample, although comparison between the data obtained directly from the TEM and indirectly from the SMB measurements very seldom fit one another. In this study, the analysis of the SMB data obtained from a MF sample is improved to account for both the contribution to the birefringence signal due to the formation of agglomerates and the field dependence of the relative permeability associated to monomers and chain-like structures. Special emphasis is given to the birefringence

signal contribution due to particle aggregates and the time evolution of them under the presence of a 100 G magnetic field (magnetic aging effect).

The ionic  $\text{NiFe}_2\text{O}_4$ -based MF sample used in this study was chemically synthesized by coprecipitating Ni(II) and Fe(III) ions in alkaline medium, following passivation and peptization of the oxide nanoparticles in acid medium, according to the Massart's method [4]. TEM was used to obtain the particle diameter polydispersity profile, which resulted in a mean particle diameter of 10.3 nm and a standard deviation of 0.38 (inset of Fig. 1). The room-temperature SMB data were obtained using the usual lock-in detection technique. The experimental setup consists of a chopped laser beam (632 nm) crossing perpendicularly the sample cell before illumination of the photodetector takes place. The sample cell consists of a double goniometer-like device that allows full angular rotation of both polarizer and analyzer and is mounted in the gap of an electromagnet in such a way that the laser beam and the external magnetic field are perpendicular to one another. The effective sample thickness is 1 mm and the axes of the polarizer and analyzer are set perpendicularly to each other during the SMB measurements. In order to evaluate the magnetic aging effect using the SMB measurements a sample containing about  $2.5 \times 10^{16}$  particle/cm<sup>3</sup> was analyzed before any previous magnetic treatment (ZFT sample). Afterwards,

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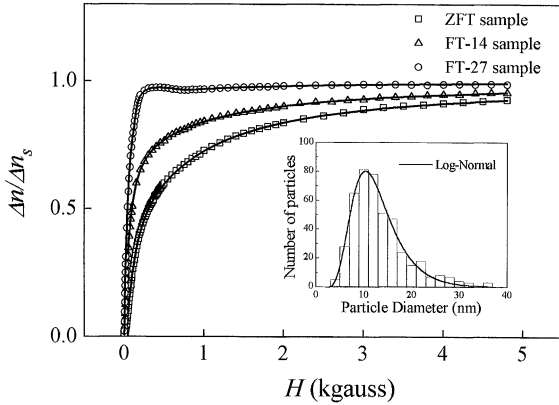


Fig. 1. Reduced SMB versus applied field for the ZFT, FT-14, and FT-27 samples containing  $2.5 \times 10^{16}$  particles/cm<sup>3</sup>. Symbols represent experimental data while solid lines represent the best fit according to the model described in this study.  $\Delta n_s$  is the saturation birefringence. The inset represents the particle size profile obtained from the TEM data.

the original sample (ZFT) was subjected to a field treatment of 100 G for 14 days (FT-14 sample) and 27 days (FT-27 sample), before performing the SMB measurements. Open symbols in Fig. 1 represent the normalized SMB data for the ZFT (square), FT-14 (triangle), and FT-27 (circle) MF samples, while the solid curves represent the best fit of the data according to the model described below.

The SMB model proposed by Xu and Ridler [5] is the starting point of the analysis carried out in the present study. The SMB signal ( $\Delta\bar{n}$ ) in MF systems needs to include the particle diameter function  $P(D)$  plus magnetization associated to the single-domain (SD) and multidomain (MD) magnetic structures, i.e.:

$$\Delta\bar{n}(H; \bar{D}_B, \sigma_B, \bar{H}_Q, \sigma_Q) = A \frac{\int_0^\infty \left[ \sum_Q C_Q \Delta n_Q(H, D) \right] D^3 P(D) dD}{\int_0^\infty D^3 P(D) dD} \quad (1)$$

with

$$\Delta n_Q(H, D) = \left[ 1 - \frac{3}{\xi_Q} \coth(\xi_Q) + \frac{3}{\xi_Q^2} \right].$$

The integral in Eq. (1) is carried out over the lognormal distribution function  $[P(D)]$  described through a mean particle diameter ( $\bar{D}_B$ ) and a standard deviation ( $\sigma_B$ ), while the summation takes into account the particle aggregation with  $C_Q$  (fraction of magnetic structure  $Q$ ) constrained by  $\sum_{Q=1}^Q C_Q = 1$ . Based on recent results [6,7] the SMB data were analyzed assuming the presence of the three following contributions: *single-domain* (SD)

Table 1

Some of the most relevant parameters obtained from the fit of the SMB data according to the model described in this study

	ZFT	FT-14	FT-27
$Q$ (MD)	1.21	1.20	8.13
$Q$ (AG)	2.87	5.97	6.25
$C_{SD}$	0.24	0.18	0.00
$C_{MD}$	0.60	0.63	0.63
$C_{AG}$	0.16	0.19	0.37
$\bar{D}_B$ (nm)	10.1	10.2	10.2
$\sigma_B$	0.28	0.36	0.35

*monomer, multidomain* (MD) *monomer* and *agglomerate* (AG).  $\xi_{SD} = (\pi/6)M_s D^3 H/kT$  accounts for SD monomer, while  $\xi_Q = Q \xi_{SD} [1 + \beta C_Q F_Q(H)]$  describes the contribution due to MD monomer ( $Q = 1$ ) or AG ( $Q > 1$ ).  $M_s$ ,  $k$ ,  $T$ , and  $\beta$  are the saturation magnetization, the Boltzmann constant, the absolute temperature, and a constant related to the Rayleigh's law, respectively. Notice that the relative permeability ( $\mu_{rel}$ ) is given by  $\mu_{rel} = 1 + \beta C_Q F_Q(H)$ , where  $F_Q(H)$  is the lognormal distribution function described by a mean field ( $\bar{H}_Q$ ) and a standard deviation ( $\sigma_Q$ ) [8]. In Eq. (1)  $A$  is a constant related to the refractive index of the suspending medium, the number of particles, and the average optical anisotropy factor of the monomer. Table 1 summarizes the main parameters obtained from the best fit of the experimental data. The parameter  $Q$  (MD) has assumed values close to the unit up to the 14th magnetic aging day, indicating the contribution of the MD monomer to the SMB. According to the parameters  $C_Q$  given in Table 1 the SD monomer is used to build AG's as the magnetic aging progresses. On the other hand, the MD monomer is used to build AG's at the 27th magnetic aging day, as indicated by the values of the parameter  $Q$  (MD).

In summary, the effect of magnetic aging on the SMB data of a 10.3 nm nickel ferrite-based ionic magnetic fluid was investigated. The SMB model was improved to account for the formation of agglomerates and the field dependence of the relative permeability associated to monomers and chain-like structures. The analysis of the SMB data provides excellent agreement with the TEM data, as far as the polydispersity profile of the sample is concerned.

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