DIPOLAR INTERACTIONS IN Fe-Fe OXIDE COLLOIDAL NANOPARTICLES: SIZE AND CONCENTRATION EFFECTS

José M. Vargas^{1,2}, Leandro M. Socolovsky², Marcelo Knobel², Daniela Zanchet¹ ¹Laboratório Nacional de Luz Síncrotron, 13084-971 Campinas (SP), Brazil. ²Laboratório de Materiais e Baixas Temperaturas – IFGW - Universidade Estadual de Campinas, CP 6165, 13083-970 Campinas (SP) Brazil.

E-mail: <u>zanchet@lnls.br</u>

Studies of magnetic nanoparticle systems have attracted much interest in the last few years owing to their fundamental interest and technological applications. The correlation of parameters such as morphology and crystalline structure of the particles with the resulting magnetic properties has been thoroughly investigated, but many open questions remain to be answered. In particular, the intrinsic complexity of nanostructured materials makes a reliable correlation between experimental data and proposed theories difficult. In this aspect, the use of "model" samples, with well characterized morphological and structural data, is an important step towards the improvement of theories that describe the real phenomena that take place in these systems. One of the crucial topics is the role played by magnetic interactions among the magnetic entities. This subject has been extensively studied from both experimental and theoretical approaches [1,2] but until now there is not a clear picture of how the dipole-dipole interactions could affect the macroscopic magnetic response of the system.

In this work, we have produced size-controlled Fe-Fe oxide samples by colloidal methods [3,4], given a further step in the study of dipolar interactions in nanosystems. The dependence of magnetic properties with particle size (6 - 12 nm) has been evaluated in powder samples (after solvent evaporation) by measuring Zero Field Cooled (*ZFC*) and Field-Cooled (*FC*) magnetization curves and magnetization loops (*M*(*H*); *H* is the applied magnetic field) [5]. On the other hand, the evolution of dipolar interaction with inter-particle distances has been evaluated by embedding the particles in a matrix at different concentrations [6]. Different models have been applied, such as the so-called superferromagnetic model, proposed by Hansen & Mørup [7], and the T* model recently proposed by Allia et al. [8], and compared to independent morphological characterization. The very good agreement between morphological analysis and two independent magnetic measurements for each case have allowed us to quantify some relevant physical parameters in this system, which will be here presented.

Briefly, Figure 1 shows a typical Transmission Electron Microscopy (TEM) image of colloidal Fe nanoparticles, revealing spherical-like particles with narrow size distribution (diameter = 7 ± 1 nm). The corresponding Small Angle X Ray Scattering (SAXS) data is presented in the inset corroborating the sample homogeneity. From the structural point of view, High Resolution TEM and X Ray Diffraction have shown that these particles are amorphous as-synthesized and that an amorphous/polycrystalline oxide is formed with time [4].

The ZFC - FC curves for powder samples with mean particle size of 6.7, 8.2 and 11.4 nm are shown in Figure 2. It can be clearly seen that the irreversibility temperature, T_{irr} , defined as the highest blocking temperature, T_B , increases as a function of mean particle size (T_{irr} is also defined as the joint of ZFC and FC curves). The quantitative analysis has been performed in the frame of the superferromagnetic model, by introducing a correction to the theory of superparamagnetism due to dipolar interactions. Relevant physical parameters, such as anisotropy constant (K), mean magnetic moment per particle (\overline{n}) and interacting volume (V_{app}) have been evaluated as a function of particle size (Table I), in very good agreement with TEM characterization [5].

The dependence of dipolar interaction with inter-particle distance has been evaluated by diluting similar fractions of a given sample in increasing mass of paraffin (up to 99.9% of paraffin). The results have clearly shown an increase of dipolar interaction strength with the decrease of interparticle distance ($\sim 140 - 10$ nm), which have been evaluated by the T* model [7]. This is the first

experimental validation of the T* model using a well-behaved system composed by true isolated spherical nanoparticles with a small size distribution, which will be discussed in detail [7,8].

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Figures:





Fig.1 : TEM image of Fe nanoparticles. Inset: SAXS data.

Fig. 2: ZFC – FC magnetization curves as a function of particle size (H = 20 Oe).

Table I. Parameters obtained as a function of particle size. d_{TEM} = mean diameter obtained by TEM; K = anisotropy constant; σ = normalized standard deviation; \overline{n} = mean number of Bohr magnetons per particle; V_{app} = interacting volume.

$d_{TEM}(nm)$	$K(10^4 \text{ erg/cm}^3)$	$\sigma(\Delta d \ / \ \overline{d} \)$	\overline{n}	$V_{app}(nm^3)$
6.7	7.9	0.20	2589	2141
8.2	8.6	0.25	4059	4134
11.4	8	0.20	9293	9837