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Local structure of magnetic fluids: from a dilute gas to a colloidal glass

F. L. O. Paula [(1)]*, P. Coppola [(2)], J. Depeyrot [(1)] J. A. Gomes [(1)], R. Aquino [(3)] and F. A. Tourinho [(3)]

(1) Complex Fluids Group – Physics Institute – University of Brasilia – Brasília (DF) – Brazil
(2) Complex Fluids Group – Chemistry Institute – University of Brasilia – Brasília (DF) - Brazil
(3) Complex Fluids Group – Faculdade UnB Planaltina – University of Brasilia – Brasília (DF) – Brazil
*Corresponding author.

Abstract – We investigated here the colloidal stability and local order of concentrated ferrofluid (volume fraction of 9% to 30%) using Small Angle X-ray Scattering (SAXS).

Magnetic nanocolloids constitute a very attractive and promising class of nanomaterials as they may be confined, displaced, deformed and controlled by an external magnetic field. These unique and striking features make them suited for a large number of applications from engineering to biomedicals. In this work, we investigate by using Small Angle X-Ray Scattering (SAXS) measurements the colloidal stability and the local order (typically between 10 nm and 100 nm) of magnetic nanocolloids, based on manganese and cobalt ferrite nanocrystals. Our goal is to characterize quantitatively the balance of interparticles interactions and relate it to the colloidal state. We have previously used such experiments to investigate mixed system based on laponite discs and magnetic nanospheres. In particular, we have shown that the presence of interactions between magnetic nanoparticles, mediated by the presence of Laponite platelets, is associated to a progressive partial phase separation between spheres and discs [1].

Here, our ferrite nanoparticles prepared by a soft chemistry method are dispersed in acidic and neutral media, the attractive potential (van der Waals and magnetic dipolar) between particles being balanced by a repulsive electrostatic interaction. In order to elaborate concentrated samples of magnetic nanocolloids, we use the osmotic compression method which allows obtaining ferrofluids with volume fractions ϕ up to typically 30%. The dispersions are fluid for the lower concentration and present a vitreous transition fluid-solid when the concentration increases. SAXS experiments are realized at the Brazilian synchrotron (LNLS) and show that the 2D scattering pattern of all investigated dispersions is isotropic as depicted in Figure 1 for ϕ = 23.5 %. Figure 2 presents the behavior of the scattered intensity $I(q, \phi)$ with the volume fraction of magnetic particles The form factor associated to the particles can be determined by combining Guinier and Porod analyses of the intensity obtained for a dilute solution. Thus, it allows the extraction of the structure factor of the dispersions shown in Figure 3. It exhibits a correlation peak, whose intensity initially increases with increasing values of volume fraction and then, collapses for larger ones. This fluidlike structure is well characterized by an interaction parameter K_T proportional to the second virial coefficient, found here positive, expressing a repulsion of characteristic length κ^{-1}. The interactions balance is completely governed by the long range electrostatic repulsion and the dispersions behave as a hard-spheres system. The solid phase is always a colloidal glass with a glassy transition occurring always below 20 % due to the high size polidispersity.

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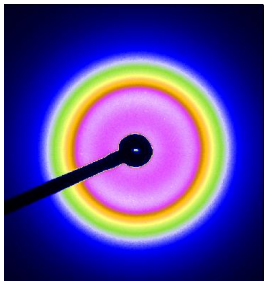

Figure 1: 2D scattering pattern

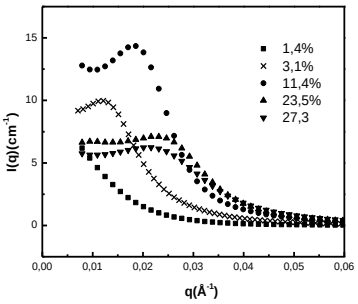

Figure 2: Scattering curves for samples of cobalt ferrite

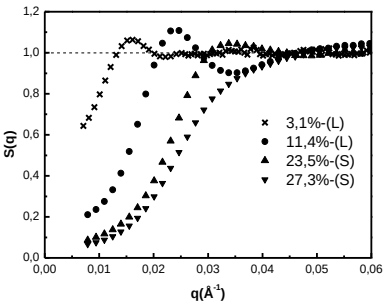

Figure 3: Structure factors (L) corresponds to samples in the liquid phase. (S) is the samples are presented in a solid phase.

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