Fast Field Cycling NMR Study in Ferrofluids and Ferronematic Materials

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Keywords: FC-NMR; nanostructures; complex fluids

Abstract: *The molecular dynamics of the carrying liquid in ferrofluid and ferronematic materials is investigated by fast field cycling NMR. Spin-lattice relaxation dispersion profiles in magnetic nanoparticles doped 5CB in different relative concentrations are measured as a function of temperature. The same study is carried out in aqueous ionic ferrofluid. From the comparison of the behaviours we conclude that due to the huge magnetic field gradients in the intergrain region (~10^4 T/m) the translational mobility of the liquid crystal molecules is restricted. That effect is not observed for water molecules in the ferrofluid material. Calculation of the magnetic forces for the water molecules of the ferrofluid in those regions gives energies slightly below k_BT. The same calculation with 5CB-molecules magnetic moment gives higher magnetic energies; in this case, thermal agitation is not enough to establish a freedom random- walk diffusion*

Ferrofluids are magnetic liquids consisting of magnetic particles (typically, $MnFe_2O_4$ or $CoFe_2O_4$) in colloidal solution with a carrier liquid. When the carrier liquid is a liquid crystal in its nematic phase, the material is known as ferronematic. It has been observed that the macroscopic orientational response of a liquid crystal to an external applied magnetic field is more effective when the liquid crystal contains magnetic particles, as the presence of magnetic grains affects the orientational viscosity of the liquid crystal.[1-3] The loss of that sort of *orientational viscosity* has been attributed to a coupling between magnetic particles and the Liquid Crystal Director,[4] but the physical nature of that coupling is still the object of research in many fields of basic science.

A question in ferrofluids research concerns the molecular dynamics of the carrying liquid, in particular whether molecular motions are affected by internal magnetic fields. Structural confinements of molecules provided by dispersed magnetic nanoparticles were recently studied with the aid of the line shape of the water protons NMR[5,6] and Raman spectroscopy.[7] It has been shown that under the presence of an external magnetic field, the magnetic grains arrange themselves in a structure that provides extremely high magnetic field gradients (namely, about 10^4 T/m).[2,5,6] In order to investigate this subject, we prepared magnetic nanoparticles doped 5CB in different relative concentrations. Proton NMR relaxometric spin-lattice dispersion was used to evaluate the diffusion processes.[8]

Measurements of T_1 vs. the Larmor frequency ν_L in the isotropic phase of the samples showed: *i)* at low Larmor frequencies T_1 is almost constant, as expected in an isotropic medium, and it increases for increasing ν_L's; *ii)* as a function of the

concentration of magnetic particles the crossover between these two behaviors goes through smaller ν_L's as the grains concentration increases; *iii)* At a given Larmor frequency the absolute values of T_1 decreases as particle concentration increases. In water-based ferrofluids FF, the crossover remains at the same Larmor frequency independently of particle concentration, showing no evidence of restriction in the translational diffusion. These different behaviors could be explained by considering the respective distinct magnetic susceptibilities of water and 5CB molecules. In doped 5CB, the restriction in molecular diffusion processes comes from magnetic interactions between the molecular magnetic moments and extremely high inter grains field gradients. The model we assume is based on magnetic traps present in the inter-nanograins space. The magnetic field gradients in these regions reaches extremely high values (~10^4 T/m), thus the translational mobility of LC molecules is restricted in that region. Calculation of the magnetic forces for the water molecules of FF material in these regions gives energies close to, but below, k_BT. The same calculation with e 5CB-molecules magnetic moment gives higher magnetic energies; in this case, thermal agitation is not enough to establish a freedom random walk diffusion.

Acknowledgements

The authors thank FAPESP, CNPq, CONICET and ANPCyT.

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