

## Dependence on size and shell fraction of exchange coupling and coercivity of hard/soft core/shell ferrite nanoparticles

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Symmetry breaking at the particle boundary of magnetic nanoparticles is a determining factor in their magnetic properties since it generally produces a structure with a magnetically ordered core and a shell with disordered spins. Ultrasmall particles are notorious for exhibiting a magnetization increase even at quite intense applied fields, behavior which can be interpreted as a non-saturation of the magnetic disordered shell even if the nanoparticle magnetization is reversible. This interface provides the appearance of unique interface effects such as the existence of an exchange coupling field, originating from magnetic interaction between core and shell spins lying at this interface [1, 2].

In this context we perform in the present work a study of the influence of the core/shell characteristics on the exchange coupling properties in hard/soft core/shell nanoparticles. For this purpose, we produce  $CoFe_2O_4@\gamma-Fe_2O_3$  nanoparticles with variable fractions of core and shell for a same size and with variable sizes and same shell fraction. These different characteristics have been obtained through modifications of the general scheme of hydrothermal synthesis of ferrite nanoparticles for ferrofluids [3]. Then, an extensive and systematic magnetic characterization of those particles was performed to investigate the saturation criterion associated to the closure of the major hysteresis loop [1]. Analyses of both closure and irreversible field, ZFC-FC magnetization measurements measured at different fields and forced minor loops procedure measured after either ZFC or FC processes are undertaken to test the anisotropy energy barrier. Once verified the saturation criteria, values of the exchange coupling field are extracted from FC hysteresis loops as a function of the cooling field. Our experimental results show that largest exchange bias field values are encountered when the nanoparticle size is smaller, and its surface shell fraction is larger. Moreover, we also show that larger surface layer leads to higher coercivity, suggesting a relationship between the intensity of the exchange coupling and the anisotropy field of the magnetic core.

To better understand the key mechanisms involved in the observed magnetic behavior, a mesoscopic scale approach and the Monte Carlo method has been employed and well reproduced the observed features. These calculations indicate that by increasing the core size, the shell thickness decreases and this results to the increase of the core anisotropy and the lowest values of the exchange bias field. In addition the small variations of the exchange coupling field observed are well accounted by the small difference in the anisotropy constants ratio between the cobalt ferrite core and the maghemite shell.

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<sup>[2]</sup> F. G. Silva, J. Depeyrot, Yu. L. Raikher, V. I. Stepanov, I. S. Poperechny, R. Aquino, G. Ballon, J. Geshev, E. Dubois, R. Perzynski, Sci. Rep. 11 (2021), 5474.

<sup>[3]</sup> J. A. Gomes, M. H. Sousa, F. A. Tourinho, R. Aquino, G. J. da Silva, J. Depeyrot, E. Dubois, R. Perzynski, J. Phys. Chem. C 112 (2008) 6220.