Surface and interfaces effects in two and three phases magnetic nanoparticles with onion-like architecture

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Nowadays, magnetic nanoparticles are widely used in many applications going from high density data storage and high performance magnets, to biomedical application as MRI or magnetic hyperthermia. The magnetic anisotropy is one of the main properties that determines its range of application, while in the first case compounds with enhanced effective magnetic anisotropy and thermal stability of the magnetic moment are required; biomedical applications need NPs with low anisotropy and superparamagnetic behavior at room temperature. The onion-like architecture opens a wide range of new possibilities to control the magnetic response of the systems allowing to combine in a single nanoparticle several components, with controlled size and high quality interfaces. In this talk I will present the strategies followed to design and fabricate magnetic NPs with core/shell and core/shell/shell architecture in order to tune the magnetic anisotropy and the magnetic coupling between the different phases. In ~10 nm Fe₃O₄/Co_{1-x}Zn_xFe₂O₄ core/shell exchange coupled nanoparticles, the effective magnetic anisotropy (K_{eff}) can be tune by the content of Co²⁺ in the shell. In this way the magnetic anisotropy can be changed an order of magnitude from 6.6×10^5 erg/cm³ for x=0, to 0.7×10^5 erg/cm³ for x=1. This result has important implications in the inversion mechanism of the magnetization providing, in example, a tool to tune the Brown or Néel relaxation mechanism and also allowing the optimization of the heating efficiency in magnetic hyperthermia experiments. On the other hand, by introducing a diamagnetic insulator between the ferrimagnetic phases the magnetic coupling can be modulated. In this case core/shell/shell nanoparticles were synthesized formed by a ~22 nm Fe₃O₄ ferrimagnetic core, encapsulated by a MgO shell of ~1nm thickness, and a second CoFe₂O₄ ferrimagnetic outer shell of ~2.5 nm thickness. The complexity of the system demands a detailed and local structural and compositional analysis which was performed by TEM and EELS. The growth of the third layer results in an enhancement of the coercivity field and the presence of exchange bias field at low temperature. This behaviour is discussed in terms of the strong coupling of the ferrimagnetic phases even in presence of the MgO separator, and the freezing of the surface spins below the freezing temperature T_F=32 K that pinned the magnetic moment of the outer ferrimagnetic shell. Finally, we will discuss the advances and the challenges still pending to achieve a fine-tune of the magnetic response in these complex nanostructures.