Spin disorder:Key player in the hyperthermia *sweet spot*

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Every hour cancer kills four people on average [1], and given that not all cases are diagnosed, the clock actually ticks faster. Nanomedicine, which exploits the potentialities of nanoparticles (NPs) for diagnostic and therapeutic purposes, provides the framework to bring these numbers down. Magnetic hyperthermia (MHT) is one of the most promising therapeutical strategies, where the cancerous cells get deactivated by the heat released from NPs, excited by an external AC magnetic field. In the last decades, the race for achieving the most performant NP design complying with the biomedical safety limits has welcomed many participants. Tuning the size, shape, composition or assemblies [2] have been competing and joining forces to push the therapeutical limits of MHT, all within the boundaries imposed by biocompatibility, efficiency of synthesis methods and biomedical concerns. Very recently, a new player, spin disorder, has come to the pitch. There exist theoretical predictions indicating that inhomogeneous spin structures result in a higher heat release compared to their non-disordered counterparts [3]. This is in line with experimental data, where nanoflowers (NFs), a multi-core conglomerate of single-core grains, are shown as excellent candidates for MHT [4, 5].
In this work, we resolve the microscopic spin configuration of the NFs beyond the macrospin model configuration, connecting the actual microstructure with the macroscopic parameter of coercivity. Using Mumax3, we numerically approximated the nanoflower morphology as closely as possible by making a collection of grains within which the anisotropy direction is fixed, but random on the average (see inset of Fig. 1), and we vary the NF size, monitoring the coercive field Hc. As it can be seen in Fig. 1, beyond the single-domain limit (purple) and below the upper limit of a multidomain state (green), there is an intermediate region where the magnetization acquires a swirling configuration that can form a vortex (orange). The vortex core is pinned at the grain boundaries, triggering the rise of Hc. This vortex-like structure can be proven using polarized small-angle neutron scattering (SANS). We have numerically computed the spin-flip SANS cross section and extracted from it the pair-distance distribution function p(r), where we have observed a zero crossing for 20, 10 and 0 mT, a clear signature of the vortex. Our work showcases the critical role of spin disorder in the heat release of magnetic NPs, elucidating the actual microstructure configuration of the NFs and capturing the physics underlying the "sweet spot" for hyperthermia.

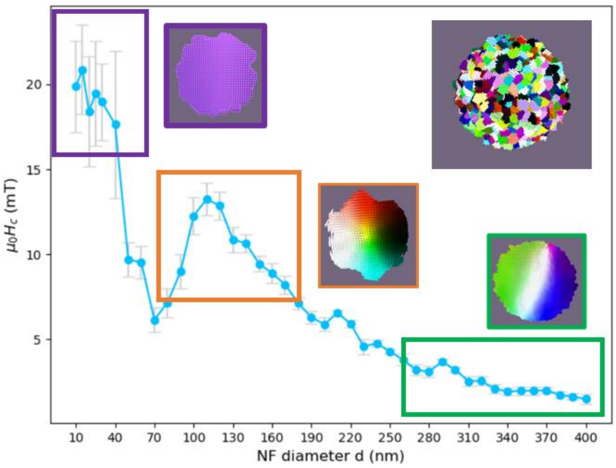

Figure 1. Coercivity vs. NF diameter: In purple, single-domain regime, in orange, the vortex configuration. In green, the multidomain state. The inset showcases a 3D simulation of NF100, where each color represents a different anisotropy direction.

References
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