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Comment on “Magnetic Relaxation of Interacting Co Clusters: Crossover from Two- to Three-Dimensional Lattices”

In a recent Letter, Luis *et al.* [1] have studied superparamagnetic relaxation phenomena in samples with layers of Co nanoparticles in Al_2O_3 . It was shown that the blocking temperature, estimated from ac susceptibility measurements, increases when the number of layers of Co nanoparticles increases; i.e., the relaxation is slowed down because of dipole-dipole interactions. The results were explained by an increase of the single particle energy barriers due to dipole-dipole interactions.

The model used by Luis *et al.* [1] excludes collective dynamics of the particle moments and a transition to an ordered state at a finite temperature. In this Comment, we argue that their analysis is based on unrealistic assumptions, and we suggest an alternative interpretation of their data in terms of collective dynamics.

Luis *et al.* [1] wrote the expression for the relaxation time

$$\tau_{\pm} = \tau_0 \exp[(U_0 \pm E_{\text{dip}})/k_{\text{B}}T], \quad (1)$$

where U_0 is the anisotropy energy barrier in the absence of interactions, E_{dip} is the dipole interaction energy, and + and – correspond, respectively, to reversal against and towards the local dipolar field created by the neighboring particles. Inspired by the model by Dormann *et al.* [2], the authors assumed that the neighbors have time to relax to their equilibrium state, such that the central spin always has to relax against the dipolar field. This results in a larger effective energy barrier and thereby a longer relaxation time than for noninteracting spins. The assumption may be correct if the central spin interacts weakly with its neighbors and relaxes much slower than its neighbors. Thus, the model may be a fair approximation for the relaxation of the largest particles in a sample with a broad particle size distribution and weak interactions [3]. However, Luis *et al.* [1] considered an average particle in a sample with a narrow particle size distribution where all particles have similar relaxation times, and therefore the assumption is not fulfilled.

Several studies of dipole-interacting systems have shown that strong dipole interactions can result in a transition to an ordered state of otherwise superparamagnetic particles [4–8]. For a sample with a narrow particle size distribution and strong interactions, the transition to the ordered state may be a true second order phase transition accompanied by a critical divergence of the longest collective relaxation time [4–8]. For samples with weaker interactions or a wider size distribution, the dynamics may be a complex interplay between collective dynamics of some particles and other particles being thermally

blocked. In this case, a true phase transition may be absent although collective effects still play an important role [7,9]. It has been shown [3] that the correct order of magnitude of the temperature, T_{cr} , at which collective effects set in is

$$T_{cr} \sim \mu_0 \mu^2 / (4\pi r^3 k_{\text{B}}), \quad (2)$$

where μ is the magnetic moment of a particle of average size and r is the average interparticle distance. In the present study, the cobalt particles have a diameter of 2.9 nm and $r \approx 5$ nm. Inserting in Eq. (2) one finds that $T_{cr} \sim 30$ K. The authors report that the paramagnetic Curie temperature increases from $\theta = 5$ K for one cobalt layer to $\theta = 40$ K for 20 cobalt layers [1]. This increase by 35 K can only be due to interparticle interactions and, at least for ferromagnetic ordering, it would indicate an ordering temperature of ≈ 30 –40 K. This strongly suggests that collective phenomena play a significant role for the magnetic dynamics in the samples. We suggest that the experimentally observed interaction-induced changes of the relaxation time could be interpreted in terms of a transition from 2D to 3D collective dynamics.

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