Supporting Information # 4: Magnetic properties of physical mixtures of the hard-magnetic hexaferrite and the soft-magnetic spinel ferrite

To show a clear difference between the hysteresis of the exchange-coupled composite nanoparticles and that of a physical mixture of the two magnetic phases, the core nanoparticles BaM_{100} (Figure S4(a)) were mechanically mixed with the spinel-ferrite maghemite nanoparticles S, which had a comparable volume to the spinel-ferrite layer at the hexaferrite cores of the composite nanoparticles (Figure S4(b)). To minimize the effect of inter-particle interactions, 10 wt.% of both nanoparticles were first mixed with diamagnetic BN and then the two types of the diluted nanoparticles were mixed together in different ratios.

First, the two magnetic phases were mixed in the same ratio, as determined in the composite. The phase composition was estimated from the Fe / Ba ratio measured by ICP-AS chemical analysis of the dissolved composite nanoparticles to be 61.5 wt.% $BaFe_{12}O_{19}$ / 38.5 wt.% Fe_2O_3. Figure S5 shows the hystereses for the core nanoparticles BaM_{100} (blue loop), the spinel-ferrite nanoparticles S (magenta loop) and the physical mixture of the two (black loop). The hysteresis of the mixture is close to the sum of the hystereses for the two components (red loop), calculated using the formula $M_{\mathrm{mix}}(H_i) = w_{HF}\, M_{HF}\,(H_i) + w_{Sp}\, M_{Sp}\,(H_i)$, where w_{HF} and w_{Sp} are the mass fractions of the BaM_{100} hexaferrite and the S spinel ferrite, respectively. The mixture S + BaM_{100} shows a characteristic constricted hysteresis loop as the two phases magnetize independently.

The shape of the hysteresis loop of the hard-magnetic phase is very sensitive to the addition of the non-coupled, soft-magnetic phase. Figure S6 shows experimental and calculated hysteresis loops for the S + BaM_{100} physical mixtures containing 10 wt.% and 2 wt.% of the spinel ferrite S.

If the coupling is absent, the shape of the loop for the mixture changed significantly even for the addition of just 2 wt.% of the soft-magnetic phase to the hard-magnetic phase.

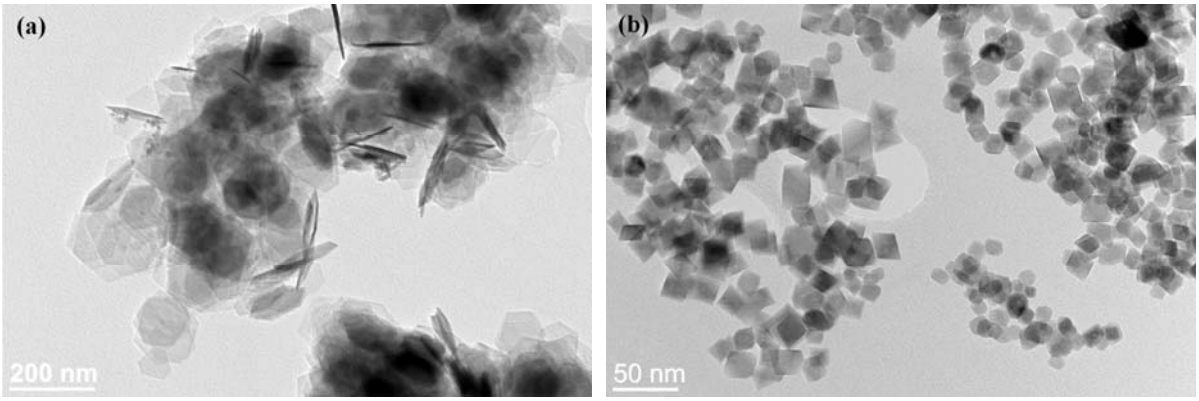

Figure S4: TEM images of the core nanoparticles BaM_{100} (a) and the spinel-ferrite nanoparticles S (b).

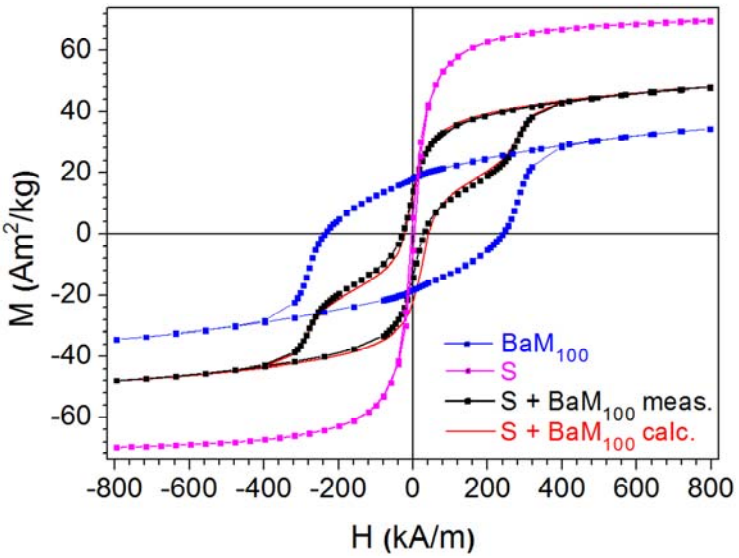

Figure S5: Measured hystereses for the core nanoparticles BaM_{100}, the spinel-ferrite nanoparticles S, and the physical mixture (S + BaM_{100} meas.), and the hysteresis loop calculated from the corresponding contributions of the two magnetic components in the mixture (S + BaM_{100} calc.).

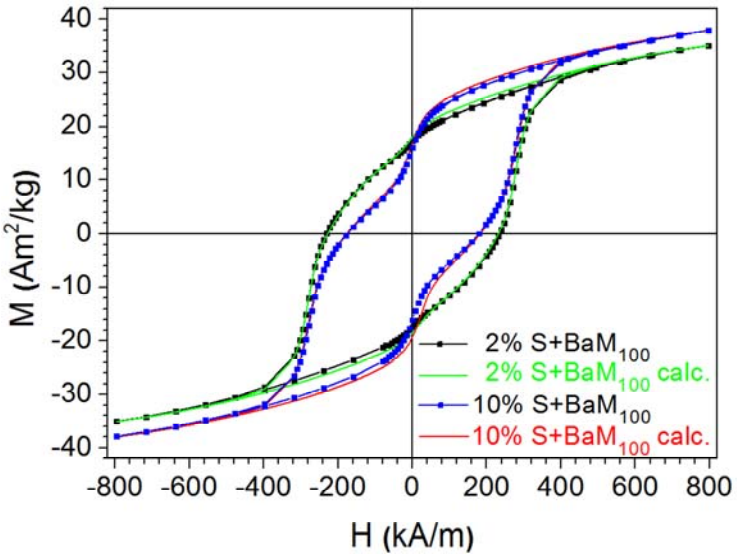

Figure S6: Measured and calculated hystereses for the S + BaM_{100} mixtures containing 10 wt.% and 2 wt.% of spinel-ferrite nanoparticles S.