On Room-temperature Ferromagnetism of Diluted Magnetic Semiconductors: Magnetic Crossover of NiO Nanocrystals

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Spintronics based on diluted magnetic semiconductors (DMSs) provides a technology that transforms reading and writing information for many uses by spin rather than by electron charge [1,2]. The practical applications of spintronics devices are, however, strongly challenged by low reproducibility of room-temperature ferromagnetism of less than 10% for typical DMSs [3]. For example, although ZnO doped with transition metals (e.g., Ni$^{2+}$ [4,5], Co$^{2+}$ [6,7], and Fe$^{2+}$ [8]) is viewed as one of the promising DMSs because of its room temperature ferromagnetism, Ueda et al.[3] did not observe ferromagnetic behavior in Ni$^{2+}$ doped ZnO, which is in apparent contradiction to the theoretical prediction for 3d-transition–metal-doped ZnO [9]. The low reproducibility and controversial observations raise concerns over the origins of room-temperature ferromagnetism in DMSs.

The ferromagnetism observed in some materials was explained by researchers in terms of the carrier-induced ferromagnetism, double exchange mechanism, or micro metallic clusters, while the magnetic contributions from the secondary phases of component transition metal oxide nanocrystals were always excluded as a likely source under the consideration that (1) the component oxides (e.g., NiO, CoO) were generally antiferromagnetic and that (2) most of the nanoscale counterparts would only exhibit ferromagnetic-like behavior at very low temperatures [10] such as at 4.2K, due to the low Curie temperature (e.g., Tc<5K for 3.1 nm NiO) [11]. These explanations lack evidence and should be critically examined, because the simple addition of transition metal ions to a reaction system does not necessarily result in incorporation of dopants into the ZnO lattice.

As a consequence, experimental verification of room-temperature magnetic properties of the transition metal oxide nanocrystals as a function of their physical dimension can be expected to have many implications in developing an understanding of the origins of room-temperature ferromagnetism in DMS nanostructures.

In this work, we initiated a systematic experimental study of particle size dependence of room-temperature magnetic properties of NiO nanocrystals. NiO was chosen as the target material because it has lattice and magnetic structural similarities to that of transition metal oxides such as CoO, FeO, and MnO that could be possible component oxides in DMSs of doped ZnO. We found that with a size reduction of NiO nanocrystals, a maximum coercivity force of 933 Oe was obtained, which is strikingly much larger than that of 100-126 Oe reported in literature for Ni$^{2+}$:ZnO nanocrystalline thin films and aggregates.

![Fig. 1 Size dependence of the room temperature coercivity for NiO nanocrystals. Reference data [12,13] were also given for comparison.](image)