Heterogeneous Magnetism in (Zn,Co)O

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Since the theoretical suggestion by *ab initio* computations that (Zn,Co)O can be intrinsically ferromagnetic, and the subsequent experimental observation of high-temperature ferromagnetism, this compound has reached the status of a model system for a broad class of dilute magnetic oxides and dilute magnetic semiconductors, in which a robust ferromagnetism is observed despite a minute concentration of magnetic impurities. However, despite the apparent agreement between experiment and theory for many of such materials systems, it was rather soon realized that the origin of the abundant high-temperature ferromagnetism is far from being understood [1]. Indeed, over the recent years it has become more and more obvious that the understanding of these ferromagnets requires the use of advance nanocharacterizations tools in order to assess how magnetic impurities are actually incorporated and distributed depending on the growth conditions and co-doping.

We investigate (Zn,Co)O films grown on silicon substrates by Atomic Layer Deposition using organic zinc and cobalt precursors [2]. The samples with different thickness and cobalt content ranging from a fraction of a percent to about 40% are thoroughly characterized (XRD, XPS, XMCD, TEM, SIMS, EDX [3]) and investigated by SQUID magnetometry [4] to reveal their magnetic characteristics which are then combined with the microscopic details of the Co distribution and the crystallographic structure of these layers. We find that purely paramagnetic (Zn,Co)O layers in the full range of the cobalt content can be prepared. Their magnetic response can be consistently described by the group theory model relevant to the Co ion in the wurtzite host with antiferromagnetic Co-Co superexchange and magnetic anisotropy with respect to the wurtzite *c*-axis adequately taken into account. However, under slightly modified growth conditions a clear above-room-temperature ferromagnetism appears. Interestingly, it comes in two guises. Firstly, as very thoroughly investigated in [3], it is related to a kind of metallic Co mesh formed at the (Zn,Co)O – substrate (Si or Al_2O_3) interface. Secondly, despite the Co accumulation on the interface, other samples show a pronounced superparamagnetic response with substantially weaker magnetic anisotropy. The magnitude of this effect scales rather to the volume of the sample than to its surface and both findings together indicate a presence of Co-rich ferromagnetic nanoclusters dispersed throughout the whole volume of the layer.

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