Size-enhanced Spin Moment in NiO Nanoparticles

Nanomagnetism promises much, given the scope of proposed applications and associated phenomena. These include the targeted delivery of drugs to tumors, magnetic storage with nanomagnetic particles, improved battery lifetimes and also quantum computing. For much of this exploitation, there is a pressing need to study directly the microscopic spin-polarized electronic properties.

In the experiment reported here, spin-polarized Compton scattering was used on the **BL08W** beamline to study NiO nanoparticles. Compton scattering measures a one-dimensional projection of the electron momentum density in the sample. In the spin-polarized technique, as used here, one can obtain the momentum density distribution of just those electrons that contribute to the net *spin* magnetic moment (the orbital moment is not measured). The Compton profile is obtained from the spectrum of inelastically scattered high energy photons with the use of the known scattering cross-section. The sample is maintained in a magnetic field, which is reversed regularly so that profiles can be obtained for both field directions. Use of circularly polarized photons results in a difference between these profiles, and upon subtraction, the charge scattering cancels out, but the spin scattering does not. A schematic of the experimental set-up is shown in Fig. 1. The total area under the spin-polarized Compton profile gives the total *spin* moment.

In this experiment, nanoparticles of NiO were investigated. Three samples were synthesized by heat treatment of NiOH powder [1,2], and characterized using powder X-ray diffraction and SQUID magnetometry. The estimated mean particle diameters of the samples under study were 6±1 nm, 8±1 nm and 13±1 nm. Bulk NiO is an antiferromagnet, with T_N = 522K. In bulk NiO, there is a 17% orbital contribution to the total magnetic moment [3]. NiO nanoparticles, however, exhibit ferromagnetism, with moments of around 0.05 μB per formula unit in the smallest particles. The contribution of the orbital moment in the nanoparticles is not known. In Fig. 2 the hysteresis loops for the three different particle size are plotted. The magnetic moment is observed to increase with decreasing particle size. It is thought that the ferromagnetic component is confined to the near-surface region, so that the spin moment increases with smaller particle sizes because the surface to volume ratio is larger.

In Fig. 3 we plot the magnetic Compton profile for the smallest NiO nanoparticle sample measured in a magnetic field of 2.5 T and a temperature of 10 K. The data are compared to a spin-polarized Compton profile of bulk ferromagnetic nickel. At large momenta (p_z > 2.5 a.u., say) the profile of bulk nickel is dominated by the 3*d* spin moment [4]. The fact that the two profiles are identical in this region shows that the 6±1nm nanoparticles also exhibit this 3*d*-like moment. However, at low momenta the two profiles are distinctly different. This is likely to be due to the insulating nature of NiO and its different electronic band structure. For a fuller discussion electronic

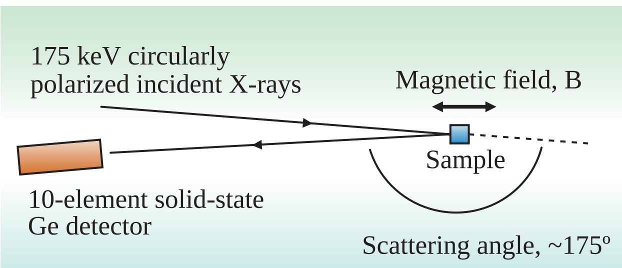

Fig. 1. A schematic of the experimental configuration used to measure spin-polarized Compton profiles on BL08W.

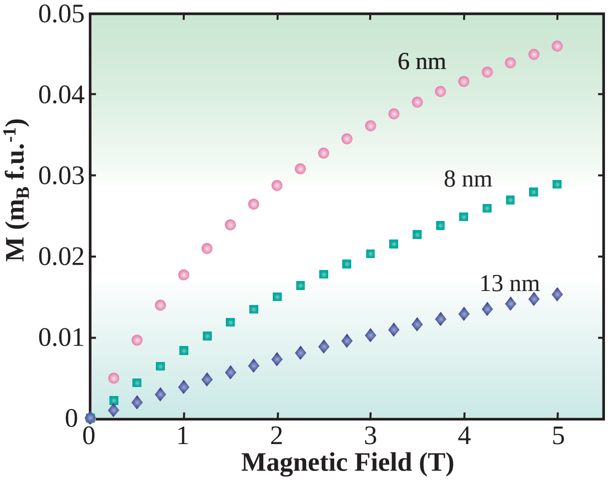

Fig. 2. The total moment, measured with SQUID magnetometry, as a function of applied magnetic field for the three particle sizes under study measured at 10 K.

structure calculations will need to be performed and compared to the data.

The spin magnetic moment was found for our samples by integrating the magnetic Compton profiles. The 8±1 nm and 13±1 nm NiO samples have a spin moment that are comparable to the total moment. However, the smallest of the samples has a spin moment that appears to be significantly larger than the total magnetic moment. This is the most surprising result; it suggests that there is an appreciable orbital moment, and that there is an *antiparallel* arrangement of the spin and orbital moments.

In summary we have used magnetic Compton scattering to study the effect of reducing the diameter of NiO nanoparticles on the spin moment. The spin moment increases as the nanoparticle diameter decreases. It appears that an enhanced orbital moment is exhibited by the smallest particles, and further studies are planned to investigate this.

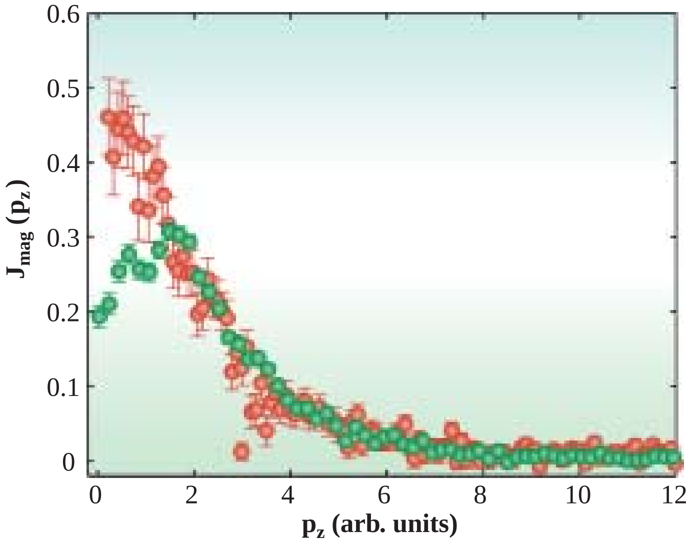

Fig. 3. Magnetic Compton profile of NiO nanoparticles with a mean particle size of 6±1 nm (red circles) and of bulk nickel (green circles).

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