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PHYSICA B

Superparamagnetism versus exchange anisotropy in nanostructured Fe and Co: A SANS study at ambient and cryogenic temperatures

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Abstract

Small-angle neutron scattering (SANS) of nanostructured Fe and Co placed in intermediate external magnetic fields around 1 kOe shows a dominating intensity in the direction parallel to the external field. In the literature, such scattering is found from superparamagnetic clusters. In the present materials, the observed behavior is not compatible with such an interpretation because of the presence of perceptible exchange interactions coupling the nanometer sized ferromagnetic grains. The experimental results rather support the idea that ferromagnetic domains confine several grains and resist to some extent the external alignment forces and thereby induce a localized net magnetic component perpendicular to the external field.

Keywords: Small-angle neutron scattering; Nanoscale objects; Magnetic correlations; Superparamagnetism

1. Introduction

Nanostructured Fe and Co produced by inert-gas condensation [1] were found to form systems of consolidated nanoscale ferromagnetic clusters embedded in interfacial phases of ferri- or antiferromagnetic oxides [2]. We have used small-angle neutron scattering (SANS) to characterize the magnetic correlations on the scale of the grain size in these systems. Following the field dependence of the lines of equal intensity on the two-dimensional position-sensitive detector with a magnetic field applied in the plane of the sample, we observe at intermediate fields (0.8–3 kOe) a pronounced elliptical distortion in field direction, opposite to that expected for ferromagnetic alignment (parallel to the external field) of the grains. Similar observations in the literature were found from superparamagnetic spin systems [3–5]. An interpretation on this basis, applied to our materials, would ignore the distinct exchange interaction between the grains. In order to characterize the origin of the field-induced anisotropy in our materials, we performed a dedicated SANS experiment covering

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a broad range in temperature (1.7–300 K) and momentum transfer Q.

2. Experimental results

Fig. 1 shows the SANS intensity lines of nanostructured Fe and Co in an external magnetic field H of 0.8 kOe. The elliptical distortion in field direction is most pronounced for Fe, and is unchanged when cooling from room temperature to cryogenic temperatures. For Co, the elongation is obvious at 300 K, but has vanished after cooling to 1.7 K. Fig. 2 shows the Q dependence of the scattering parallel and perpendicular to the field direction for a nanostructured Fe sample. The scattering shows a crossover at around 0.15 nm^{-1}, where the elongation changes from parallel to the magnetic field for small Q to perpendicular to the field at larger Q values.

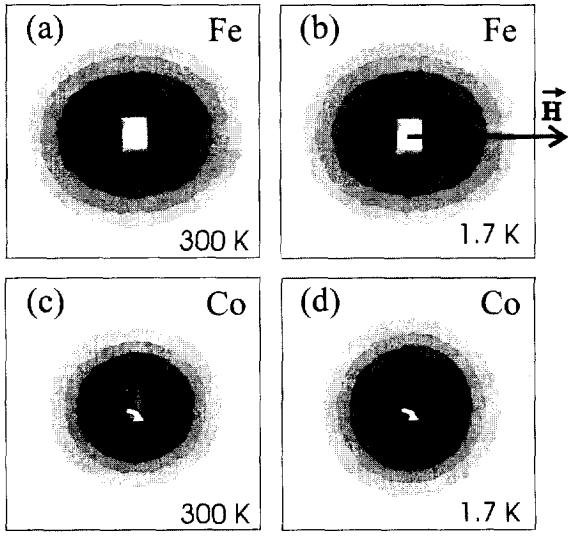

Fig. 1. Lines of equal SANS intensity on a two-dimensional position-sensitive detector, for nanostructured Fe (grain size $D = 20$ nm) and Co ($D = 10$ nm) in a horizontal magnetic field ($H = 0.8$ kOe), measured at different temperatures as indicated ($0.025 < Q < 0.1$ nm^{-1}; $Q = |\boldsymbol{Q}| = 4\pi \sin\theta/\lambda$ with $\theta =$ half the scattering angle and $\lambda =$ neutron wavelength).

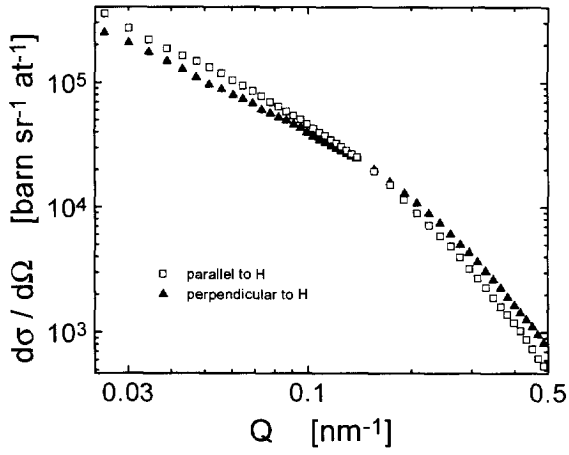

Fig. 2. SANS scattering cross section $d\sigma/d\Omega(Q)$ of nanostructured Fe, for directions parallel and perpendicular to the applied magnetic field of 1.5 kOe.

3. Discussion

These results are not compatible with an interpretation in terms of superparamagnetic clusters. Firstly, we would expect a marked temperature dependence of the degree of the elongation parallel to the field, with the tendency to be more pronounced at low temperatures. In contrast, for Fe, we observe no change between 300 and 1.7 K. For Co, we even observe a continuous relaxation of the distortion with decreasing temperature. Secondly, the elongation parallel to the field is restricted to the range of small Q (Fig. 2). The crossover at $Q = 0.15$ nm^{-1} represents a length scale of about 40 nm, significantly larger than the grain size itself. Hence, the spin component perpendicular to the field confines many grains to a common net magnetic alignment. Such an ensemble of coupled grains cannot be attributed to a loosely coupled superparamagnetic spin system. Rather it requires a perceptible exchange interaction across the interfaces. For the same samples, magnetization measurements show the effect of an exchange anisotropy between the grains and oxide interfacial phases by means of an asymmetric hysteresis loop after cooling the samples in an external magnetic field [2,6].

This exchange anisotropy is the basis for our interpretation of the spin canting out of the

external field direction. We find that several ferromagnetic grains are coupled among each other via exchange interaction. If such clusters of coupled grains are to some degree decoupled from the ferromagnetic surroundings owing to the presence of antiferromagnetic oxides, they may resist the external alignment forces at intermediate fields and thus induce a localized net magnetic component perpendicular to the field, consistent with the observed SANS intensity distortion. This interpretation is supported by the observation that the degree of elongation of the SANS intensity lines parallel to the field correlates with the magnitude of the bulk anisotropy constant. For a lower anisotropy constant, the ferromagnetic exchange length extends to larger distances and hence the probability for the formation of such clusters increases. Correspondingly, as can be seen from Fig. 1, a more pronounced horizontal elongation is found for Fe ($K_{Fe} = 4.7 \times 10^5$ erg/cm^3) than for Co ($K_{Co} = 5.2 \times 10^6$ erg/cm^3 at room temperature). K_{Fe} is practically independent of temperature for $T < 300$ K in accordance with an unchanged shape of the intensity lines at 1.7 K and 300 K (Fig. 1). K_{Co} increases, in turn, by more than 60% when the temperature is decreased from 300 to 1 K. In conjunction with this increase the horizontal elongation decreases (Fig. 1).

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