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Resolving the Complex Spin Structure in Fe-Based Soft Magnetic Nanocrystalline Material by Magnetic Small-Angle Neutron Scattering

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Over the last decades, the development of novel Fe-based nanocrystalline soft magnetic materials raised considerable interest owing to their great potential for technological applications. The most well-known examples are FINEMET- [1], VITROPERM- [2], and NANOPERM-type [3] soft magnetic alloys, which find widespread application as magnetic cores in high-frequency power transformers or in interface transformers in the ISDN-telecommunication network. Typically, these materials are synthesized by the ultra-rapid annealing of melt spun amorphous precursor material, which results in a nanocrystalline microstructure consisting of Fe-based nanoparticles that are embedded in an amorphous magnetic matrix.

The magnetic softness of such materials is due to the so-called exchange-averaging effect of the local magnetocrystalline anisotropy and has been successfully modeled within the framework of the random anisotropy model [4–7]. More specifically, this model predicts that the variation of the macroscopic coercivity H_c of soft magnetic nanocrystalline materials scales as:

$$H_c \sim (D/L_0)^n$$

where $n = 3$ or $n = 6$ depending on the nature of the magnetic anisotropy, D is the average particle size, and $L_0 \sim \sqrt{A_{ex} / K_1}$ is the exchange correlation length, with A_{ex} the exchange-stiffness constant and K_1 the magnetocrystalline anisotropy. Based on the above expression, it appears obvious that an improvement of the magnetic softness comes about by either refining the grain microstructure (smaller D) and/or by maximizing (minimizing) A_{ex} (K_1).

While D can be routinely measured using X-ray diffraction and electron microscopy, the exchange and anisotropy constants are more difficult to obtain, in particular on polycrystalline bulk materials. Regarding the measurement of the exchange constant, most of the research activities in this direction are focused on the overall macroscopic characterization, e.g., via hysteresis-loop measurements (coercivity, saturation magnetization, and permeability) and magnetic-anisotropy determination (crystalline, shape, or stress related) rather than on the quantitative determination of A_{ex}. One reason for this might be related to the fact that many of the conventional techniques for measuring A_{ex} (e.g., magnetooptical methods, Brillouin light scattering, spin-wave resonance, or inelastic neutron scattering) require thin-film or single-crystal samples.

Recent progress regarding the theoretical understanding of magnetic small-angle neutron scattering (SANS) allows one to quantitatively analyze the magnetic interactions in bulk ferromagnets [8, 9]. Magnetic SANS provides volume-averaged information about the spatial variation of the magnitude and orientation of the magnetization vector field on a length scale of about 1–300 nm. This technique was previously successfully employed to study e.g., the mesoscale magnetic microstructure of Invar and Heusler alloys [10–12], or to disclose the role played by the crystal defects on the magnetic microstructure of nanocrystalline materials prepared by the high-pressure torsion process [13, 14].

Our recent study [15] unravels the magnetic softness in $(Fe_{0.7}Ni_{0.3})_{86}B_{14}$ alloy via unpolarized magnetic SANS experiments. This particular alloy is a promising HiB-NANOPERM-type soft magnetic nanocrystal-

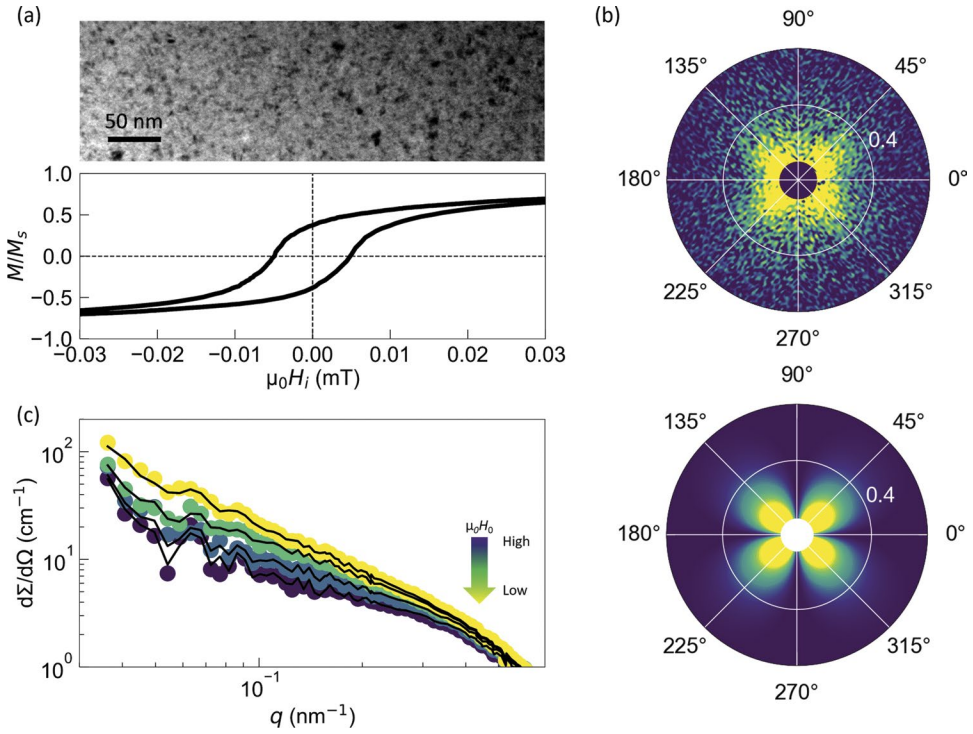

Figure 1. (a) Top panel: Bright-field electron microscopy image of HiB-NANOPERM highlighting its nanocrystalline microstructure. Bottom panel: Normalized magnetization curve revealing a coercivity of $\mu_0 H_c \cong 4.9$ μT. (b) Top panel: Experimental purely magnetic SANS cross section $d\Sigma_{mag}/d\Omega$. Bottom panel: Corresponding theoretical $d\Sigma_{mag}/d\Omega$ based on the micromagnetic SANS theory. (c) Magnetic-field dependence of the azimuthally-averaged total (nuclear + magnetic) SANS cross section $d\Sigma/d\Omega$ of $(Fe_{0.7}Ni_{0.3})_{86}B_{14}$ alloy. Solid lines: Prediction by the micromagnetic theory.

line material [16], which exhibits an ultrafine-grained microstructure with an average particle size as small as 4 nm and an extremely small coercive field of ~ 4.9 μT (see Figure 1a). For comparison, the magnitude of the earth's magnetic field at its surface varies between about 25 to 65 μT [17], i.e., 5 to 13 times larger. Our neutron data analysis (summarized in Figure 1b,c) provides access to the magnetic interaction parameters such as the exchange-stiffness constant and the strength and spatial structure of the magnetic anisotropy and magnetostatic fields. As discussed before, these parameters play a crucial role towards the optimization of the magnetic softness of Fe-based nanocrystalline materials. The analysis of the field-dependent SANS data suggests the presence of strong spin misalignment on a mesoscopic length scale. The clover-leaf-type angular anisotropy patterns observed in the magnetic SANS signal (Figure 1b) can be well reproduced by our micromagnetic SANS theory. The origin of these patterns is related to the jump of the magnetization at the interfaces between the particles and the amorphous magnetic matrix. Analysis of the azimuthally-averaged total (nuclear and magnetic) SANS cross section (Figure 1c) yields an exchange constant of $A_{ex} = (10 \pm 1) \times 10^{-12}$ J/m, a value that is 2–3 times larger than those reported previously for similar alloys [18]. The large value of A_{ex} together with the small grain size and low anisotropy is believed to be responsible for the extreme softness of this alloy.

The study highlights the strength of the unpolarized magnetic SANS technique to characterize magnetic materials on the mesoscopic length scale. The structural and magnetic results provide valuable information on the $(Fe_{0.7}Ni_{0.3})_{86}B_{14}$ ribbons, and further confirm the strong potential of Fe-B-based HiB-NANOPERM-type alloys as soft magnetic nanocrystalline materials. In the context of the random anisotropy model, we provided further support for the notion that the magnetic softness in these systems is due to the combined action of the small particle size and an increased exchange constant resulting in an enhanced exchange correlation length.

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