Range of Magnetic Correlations in Nanocrystalline Soft Magnets

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We have obtained the magnetic field dependence of static ferromagnetic correlations in nanocrystalline electrodeposited Co and Ni by means of the correlation function of the spin misalignment, determined from small-angle neutron scattering data. The approach yields a correlation length l_C, which is a measure for the spatial extent of inhomogeneities in the magnetization distribution. The correlation length depends strongly on the applied magnetic field with values ranging from 94 nm in nanocrystalline Co at low fields to about 15 nm at saturation. The results for l_C indicate that in Co the main source of nonuniformity in the spin system is the anisotropy field of each individual crystallite, whereas in nanocrystalline Ni the main sources of spin disorder originate from twin faults or from the defect cores of grain boundaries.

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The ferromagnetic correlation length is an important parameter in magnetism, since it is a measure for the average distance over which fluctuations of the magnetization are correlated [1]. Theory suggests that the excellent soft magnetic properties of nanocrystalline (NC) ferromagnets are due largely to the magnetic correlation length being larger than the "structural correlation length"—the average crystallite size D [2,3]. Since a given property of a material may depend crucially on the relation between that property's characteristic length scale and a relevant microstructural length scale, it would be of paramount importance to determine the volume-averaged magnetic correlation length in NC soft magnets. While standard techniques such as Lorentz, Kerr, or magnetic force microscopy probe/image the magnetic microstructure in the near vicinity of the surface of a specimen, magnetic small-angle neutron scattering (SANS) is able to resolve the spin arrangement within the bulk of magnetic materials and on the length scale of interest, between about 1 to 500 nm. It is the purpose of this Letter to provide a quantitative analysis of static ferromagnetic correlations in NC bulk ferromagnets based on the correlation function of the spin misalignment, which is straightforwardly obtained from the magnetic SANS cross section. As a case study, we determine the applied magnetic field dependence of the magnetic correlation length in NC electrodeposited Co and Ni, and we relate the values of the correlation length to characteristic microstructural length scales and to a prediction from micromagnetics theory. A preliminary study on NC Ni was published in a conference proceedings [4].

The correlation function $C(\mathbf{r})$ of the spin misalignment can be defined as follows [4,5]:

$$C(\mathbf{r}) = \frac{1}{V}\int \frac{\mathbf{M}_p(\mathbf{x})\mathbf{M}_p(\mathbf{x}+\mathbf{r})}{M_s^2}\,dV\,, \qquad (1)$$

where V denotes the scattering volume, M_s is the magnitude of the magnetization vector $\mathbf{M}(\mathbf{x})$, and the function $\mathbf{M}_p(\mathbf{x})$ represents the fluctuation of the local magnetization $\mathbf{M}(\mathbf{x})$ about its position-independent average $\langle\mathbf{M}\rangle$, i.e., $\mathbf{M}_p(\mathbf{x}) = \mathbf{M}(\mathbf{x}) - \langle\mathbf{M}\rangle$. For isotropic systems $C(r)$ can be related to an experimental magnetic SANS cross section $d\Sigma_{\mathrm{mag}}/d\Omega$ according to [4,5]

$$C(r) = \frac{a}{12\pi^2 b_{\mathrm{mag}}^2 \rho_a^2 r}\int_{q_{\min}}^{q_{\max}} \frac{d\Sigma_{\mathrm{mag}}}{d\Omega}\sin(qr)q\,dq\,, \qquad (2)$$

where b_{mag} and ρ_a denote, respectively, the atomic magnetic scattering length and the atomic density, and q is the modulus of the scattering vector $\mathbf{q}$. For the completely isotropic ferromagnet, at zero or vanishing applied magnetic fields, the parameter a in Eq. (2) takes on the value $a = 9$ and, similarly, $a = 8$ for the nearly saturated, texture-free ferromagnet, where variations of the magnetization are restricted to lie in a plane perpendicular to the applied field [4,5]. We are interested in evaluating a magnetic "correlation length" l_C, i.e., a characteristic dimension of regions in which the magnetic moments are coherently misaligned in the same direction relative to the direction of the mean magnetization $\langle\mathbf{M}\rangle$. There are various ways for defining and measuring l_C, for instance, in terms of the initial slope of $C(r)$. As a robust measure, which is independent of small oscillations of C at small r, we here identify l_C with the value of r for which C is decreased to $\exp^{-1}$ of its extrapolated value at $r = 0$. It is readily verified that this yields the correct correlation length when the correlation function decays exponentially, $C(r) = C(0)\exp(-r/l_C)$. It is emphasized that our definition does not require that $C(r)$ is an exponential; it is merely a convenient way to define a characteristic length which can be related to the magnetic microstructure and which can be computed model independently.

For unpolarized neutrons and in the limit of high applied magnetic fields the total SANS cross section $d\Sigma/d\Omega$ of a NC ferromagnet can be written as the sum

of a combined nuclear and magnetic residual scattering cross section $d\Sigma_{\rm res}/d\Omega$ and of a pure micromagnetic spin-misalignment scattering cross section $S_H R$ [6–8],

$$\frac{d\Sigma}{d\Omega}(q, H_i) = \frac{d\Sigma_{\rm res}}{d\Omega}(q) + S_H(q)R(q, H_i). \qquad (3)$$

The residual scattering cross section arises from regions within the sample of nonuniform nuclear density and/or composition (such as pores), and in the high-field limit $d\Sigma_{\rm res}/d\Omega$ is approximately independent of the internal magnetic field H_i. The pure magnetic scattering contribution is written as the product of the anisotropy-field scattering function $S_H(q)$ and of the micromagnetic response function for SANS, $R(q, H_i)$. The product $S_H R$ describes SANS from small static fluctuations of the magnetization perpendicular to the applied magnetic field. The approach via Eq. (3), which results from a combination of micromagnetics theory and neutron scattering formalism, has been successful in describing and analyzing SANS data on dense samples of soft magnetic electrodeposited NC Co and Ni near saturation [7–9]. In particular, quantitative information on the magnetic microstructure, the value of the exchange-stiffness constant, the strength and microstructure of the mean anisotropy field [7,8], and on the strength of the magnetostatic stray field could be obtained [9]. The above analysis was used here only to determine $d\Sigma_{\rm res}/d\Omega$, which was subtracted from the total scattering signal before further analysis.

The NC samples were synthesized by pulsed electrodeposition [10]. The materials parameters used in the data analysis along with sample characterization (and correlation functions of NC Ni) can be found in Refs. [4,8]. Here we report only experimental details that are relevant for the later discussion. The mean grain sizes of the NC samples were measured by x-ray diffraction; we obtained $D = 10$ nm for NC Co and $D = 49$ nm for NC Ni. The exchange-stiffness constants at room temperature, $A = (3.1 \pm 0.1) \times 10^{-11}$ J/m (NC Co) and $A = (7.6 \pm 0.3) \times 10^{-12}$ J/m (NC Ni), were determined by fitting the experimental SANS data to the micromagnetics theory [8]. The SANS experiments were conducted at room temperature at instrument NG 3 at National Institute of Standards and Technology Cold Neutron Research Facility using neutron wavelengths of $\lambda = 0.84$ nm (Co) and $\lambda = 0.60$ nm (Ni) with a respective wavelength spread $\Delta\lambda/\lambda = 0.10$. The applied magnetic field was provided by an (1.8 T) electromagnet and was applied normal to the incident neutron beam and in the plane of the sample. The SANS data were corrected for absorption, dark current, and background. The internal magnetic field H_i was estimated as $H_i = H_a - N_d\langle M\rangle$, where H_a denotes the applied magnetic field, $\langle M\rangle$ is the magnitude of the average magnetization at the particular H_a, and N_d is the demagnetizing factor, which was computed by approximating the sample shapes as ellipsoids.

Figure 1 depicts the azimuthal-average total scattering cross section $d\Sigma/d\Omega$ of NC Co as a function of the modulus q of the scattering vector at 295 K and at different magnetic fields H_i as indicated in the figure caption. The total scattering signal $d\Sigma/d\Omega$ is seen to decrease by more than 3 orders of magnitude as H_i is increased from 5 to 1800 mT, indicating a strong spin-misalignment scattering. From such data the residual scattering cross section $d\Sigma_{\rm res}/d\Omega$ can be extracted [7,8], and by subtracting $d\Sigma_{\rm res}/d\Omega$ from the total signal $d\Sigma/d\Omega$ the magnetic spin-misalignment scattering cross section $d\Sigma_{\rm mag}/d\Omega = S_H R$ is obtained [compare Eq. (3)] and can then be used in Eq. (2) in order to compute the correlation function $C(r, H_i)$. Because of the strong spin-misalignment scattering, this correction proved to be relevant only at the highest field.

Figure 2(a) shows the results for the correlation functions $C(r, H_i)$ of NC Co that were calculated using Eq. (2) with $a = 9$ at the lowest field and with $a = 8$ at the higher fields. Increasing the magnetic field H_i results in a suppression of the transversal magnetization fluctuations, which are described by $\mathbf{M}_p(\mathbf{x})$ and, hence, $C(r)$ decreases (at a given r) when the field is increased. Another way of monitoring the progressive alignment of magnetic moments is displayed in Fig. 2(b), where we plot the expectation value for the mean-square transversal magnetization component, $\langle|\mathbf{M}_p|^2\rangle_v/M_s^2$, as a function of H_i. Comparison with Eq. (1) shows that $\langle|\mathbf{M}_p|^2\rangle_v/M_s^2$ is given by $C(0)$, the extrapolated value of the correlation function at $r = 0$, where $\langle\ldots\rangle_v = V^{-1}\int(\ldots)dV$. At a given field, $\langle|\mathbf{M}_p|^2\rangle_v/M_s^2$ is a measure for the magnitude of the deviation of $\mathbf{M}$ relative to the mean magnetization $\langle\mathbf{M}\rangle$. As can be seen, $\langle|\mathbf{M}_p|^2\rangle_v/M_s^2$ depends strongly on the

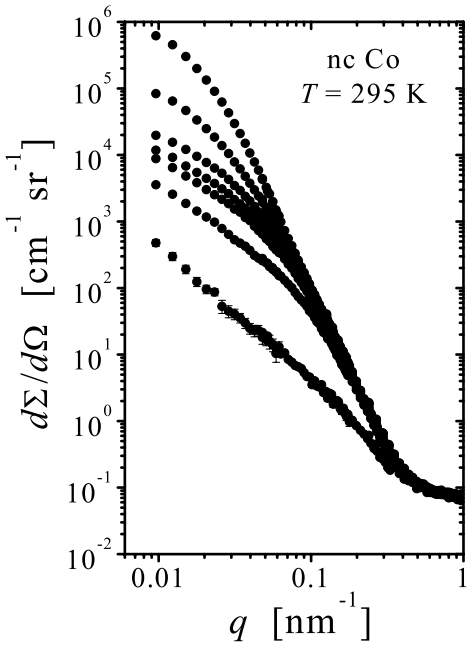

FIG. 1. (●) Macroscopic differential scattering cross section $d\Sigma/d\Omega$ of NC Co at different internal magnetic fields H_i. Values of the internal field $\mu_0 H_i$ (in mT) from top to bottom: 5, 25, 54, 80, 107, 243, 1800.

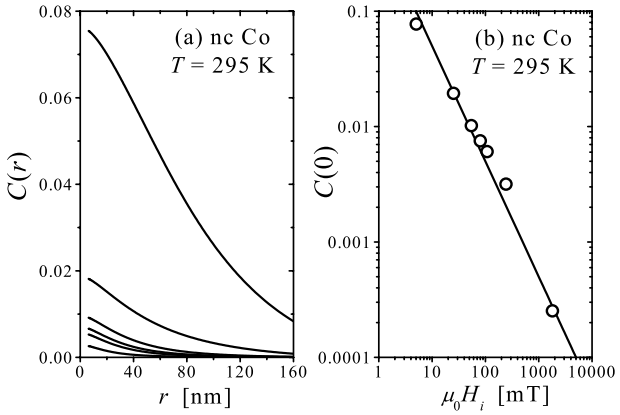

FIG. 2. (a) Experimental correlation functions $C(r, H_i)$. Values of $\mu_0 H_i$ (in mT) from top to bottom: 5, 25, 54, 80, 107, 243. (b) (○) Field dependence of the mean-square transversal magnetization component, $\langle |\mathbf{M}_p|^2 \rangle_v / M_s^2 = C(0)$. Solid line: $C(0) \propto H_i^{-1}$.

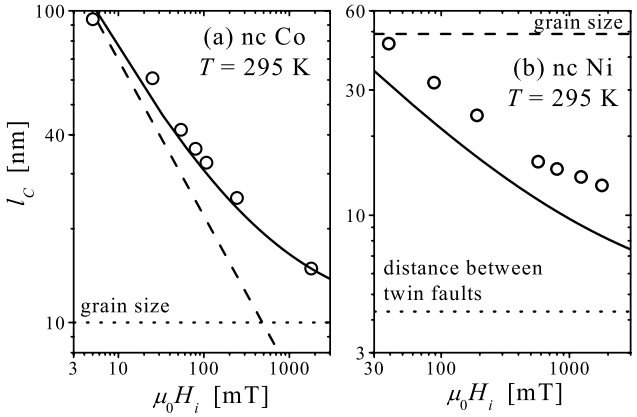

FIG. 3. (a) (○) Magnetic field dependence of the correlation length l_C in NC Co. Solid line: $l_C(H_i) = \mathcal{D} + l_H(H_i)$ with $\mathcal{D} = 10$ nm, $A = 3.1 \times 10^{-11}$ J/m, and $M_s = 1434$ kA/m. Dashed line: $l_C \propto H_i^{-1/2}$. Dotted line: experimental grain size. (b) (○) $l_C(H_i)$ in NC Ni. Solid line: $l_C = \mathcal{D} + l_H(H_i)$ with $\mathcal{D} = 4.3$ nm, $A = 7.6 \times 10^{-12}$ J/m, and $M_s = 522$ kA/m. Dotted line: average distance between twin faults. Dashed line: average grain size.

field [$C(0) \propto H_i^{-1}$] with values ranging from about 8% at the lowest H_i to 0.02% at the largest H_i. It is worth noting that at an applied field of about 100 mT, where NC Co is practically saturated [8] [$C(0) \cong 0.5\%$], the SANS cross section $d\Sigma/d\Omega$ still changes by more than 1 order of magnitude when H_i is further increased to the highest experimental field of 1.8 T (compare Fig. 1). This observation underlines the special importance of transversal magnetization fluctuations for the magnetic SANS of NC ferromagnets [7–9].

The correlation length l_C, determined as described above, can be seen in Fig. 3(a) as a function of H_i. The field variation of l_C confirms the conclusions that were already drawn by inspection of the $C(r, H_i)$, namely, a pronounced dependence on the applied field with values ranging from about 94 nm at the lowest field to about 15 nm at the highest field. The values for l_C are (at all fields) larger than those expected for uniformly magnetized grains of the experimental size 10 nm [dotted line in Fig. 3(a)].

Micromagnetics theory predicts that, near saturation, the magnetic microstructure is the convolution of the anisotropy-field microstructure with an exponential response function that decays with the characteristic length l_H given by [6,8,11,12]

$$l_H = \sqrt{\frac{2A}{\mu_0 M_s H_i}}. \tag{4}$$

Similarly to l_C, the quantity l_H is a measure for the length scale on which perturbations in the spin structure decay [6,8,11,12], or equivalently expressed, l_H describes how gradients in $\mathbf{M}(\mathbf{x})$, caused by a particular concentration of force, are transmitted by the exchange interaction into the lattice. For delta-function-like perturbations without any spatial extension the measured (average) value for l_C should be identical to l_H. However, since lattice defects such as dislocations, which in their vicinity give rise to gradients in the orientation of magnetic moments, have a finite spatial extension, the experimental value for l_C should at large applied fields reflect the typical size of the defect that causes the perturbation. For an ideal NC ferromagnet with atomically sharp grain boundaries, this length scale is related to the grain size D, which is the average distance over which the direction of the magnetic easy axes are uniform. Therefore, the field dependence of l_C cannot be reproduced by a pure $H_i^{-1/2}$ law over the entire range of fields, as expressed by $l_H(H_i)$ [compare dashed line in Fig. 3(a)]. Instead, it is suggested here to describe the data in Fig. 3(a) according to some function $l_C[\mathcal{D}, l_H(H_i)]$ that expresses the complicated convolution relationship between the nuclear (grain) microstructure and the magnetic microstructure predicted by micromagnetics theory, where $\mathcal{D}$ represents the "defect size." A simple choice is $l_C(H_i) = \mathcal{D} + l_H(H_i)$. By using the experimental values $\mathcal{D} = D = 10$ nm, $A = 3.1 \times 10^{-11}$ J/m, and $M_s = 1434$ kA/m [8], we obtain the solid line in Fig. 3(a) without any free parameters. This function provides a reasonable description of the field dependence of l_C at all fields, and it varies asymptotically as $H_i^{-1/2}$, in agreement with micromagnetics theory [6,8,11,12], which in turn implies that the small-misalignment approximation [for which Eq. (4) was derived] may be valid down to quite small fields. The observation in Fig. 3(a) that $l_C(H_i)$ deviates from a pure $H_i^{-1/2}$ behavior at large fields and approaches a constant value close to the experimental grain size suggests that in NC Co the dominating microstructural defect is the anisotropy field of each individual crystallite. In other words, the typical length scale over which the anisotropy field in our NC Co sample is uniform is of the order of D, and nonuniformities on a smaller scale, originating, e.g.,

from the defect cores of grain boundaries or stacking faults [8], play only a minor role.

By contrast, the field dependence of l_C in NC Ni [Fig. 3(b)] with an average crystallite size of $D = 49$ nm indicates the presence of inhomogeneities in the anisotropy field on a scale smaller than D [4]; l_C varies from about 45 nm at 39 mT to 13 nm at 1790 mT. The solid line in Fig. 3(b) represents the function $l_C(H_i) = \mathcal{D} + l_H(H_i)$, where we used the experimental values $A = 7.6 \times 10^{-12}$ J/m, $M_s = 522$ kA/m [8], and where we took the previously estimated average distance between twin faults, $\mathcal{D} = 4.3$ nm [8], to be the characteristic length scale of the nuclear microstructure. The above function $l_C(H_i)$ describes at least qualitatively the experimental data points of NC Ni, and the (almost constant) deviation is attributed to large variations in the characteristic length scales (grain size, distance between twin faults, etc.) that are associated with the individual microstructural anisotropy fields.

When the field is decreased, l_C increases to values of up to 94 nm [compare Fig. 3(a)] indicating the emergence of long-range magnetization fluctuations that are correlated over many grains. It would be of interest to follow the further evolution of l_C to very small values of the field or even to negative fields in order to investigate the behavior of l_C when domain nucleation sets in and magnetization reversal takes place. Rough estimates for the coercive field are of the order of 0.2 mT for both samples [8] and are much smaller than the fields that are accessible in the present experiment. In the available range of experimental fields, about 1800 mT down to several mT, the results in Fig. 3 suggest that the magnetic microstructure in NC Co and Ni is dominated by static nanometer-scale magnetization fluctuations, whose amplitude and wavelength increase continuously as the field is lowered. Eventually, the continuous increase of l_C is expected to break down when the coercive field is reached and a macroscopic domain structure with domain sizes of the order of several microns forms. In this situation, the dominating magnetic SANS contrast originates from the jump of the direction of the magnetization between neighboring domains, and such a discontinuity cannot be resolved anymore with conventional SANS.

Our results for l_C call for a comparison with the predictions of the random anisotropy model of Herzer [2], which relates the extraordinary soft magnetic properties of NC ferromagnets to the spatial extent of gradients in the magnetization distribution. Within Herzer's model the parameter $l_K = \sqrt{A/K}$ plays a crucial role, where K is an anisotropy constant. For grain sizes $D < l_K$ Herzer's correlation length L is predicted to increase as a function of D according to $L \propto D^{-3}$, leading finally to the well-known $H_c \propto D^6$ grain-size dependence of the coercivity H_c. However, since the grain sizes of the NC samples (Co: 10 nm; Ni: 49 nm) are approximately equal in magnitude to the respective value of l_K (Co: 8 nm; Ni: 40 nm), and since the above inequality should be considered as a rough estimate, a reliable comparison with Herzer's model is not possible at this point. By means of the presented formalism, a detailed study of the grain-size dependence of l_C is, however, straightforward.

In conclusion, we have presented the first analysis of the field dependence of static nanometer-scale magnetization fluctuations in nanocrystalline Co and Ni by means of the correlation function of the spin misalignment. The correlation length l_C in both samples depends strongly on the applied magnetic field with values ranging from some tens of nanometers (Co: 94 nm; Ni: 45 nm) at low fields to about 15 nm at saturation. The field dependencies of l_C follow a modified prediction from micromagnetics theory at all fields, suggesting that in NC Co the dominating source of spin nonuniformity is the anisotropy field of each individual crystallite, whereas in NC Ni inhomogeneities on a scale much smaller than the crystallite size, such as twin boundaries or disordered grain boundary regions, are the origin of disorder in the spin system. The possibility to measure the correlation length will enable one to scrutinize existing models for the magnetic properties of NC soft magnets [2,3].

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