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Measurement of a Magnetic-Field Dependent Correlation Length in Nanocrystalline Ni Using Small-Angle Neutron Scattering

A. MICHELS[1]) (b), J. WEISSMÜLLER (a, b), U. ERB (c), and J. G. BARKER (d)

(a) Institut für Nanotechnologie, Forschungszentrum Karlsruhe, Karlsruhe, Germany

(b) Technische Physik, Universität des Saarlandes, Gebäude 43B, Postfach 151150, D-66041 Saarbrücken, Germany

(c) University of Toronto, Toronto, Canada

(d) NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg (MD), USA

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We have analyzed magnetic-field dependent small-angle neutron scattering data on nanocrystalline electrodeposited Ni by means of the correlation function of the spin misalignment. The approach yields a correlation length l_C of the spin misalignment that is a measure for the characteristic dimension of regions in which the magnetic moments are misaligned coherently into a common direction. We find that l_C varies strongly with the applied magnetic field H_i with values extending from about 50 nm (larger than the value for uniformly magnetized grains) at small H_i to about 10 nm (considerably smaller than the grain size) at large applied fields.

1. Introduction In nanocrystalline (nc) ferromagnets the direction and/or the strength of the magnetic anisotropy field changes randomly on a length scale of the order of the average grain size D which is typically about 10 nm. The static magnetic microstructure of *nanocrystalline* ferromagnets is therefore, compared to their *microcrystalline* counterparts, highly nonuniform on a nanometer scale, and this inhomogeneity results in a strong elastic magnetic neutron scattering signal at small scattering angles. This fact combined with the recent availability of high-purity, porosity-free electrodeposited nc soft magnets [1], where the ratio of interesting magnetic scattering to nuclear background is favorably high, render such samples attractive for studies by magnetic small-angle neutron scattering (SANS). Moreover, the technique of SANS is capable of resolving the magnetic microstructure in the *bulk* and on a length scale of a *few nanometers*, so that SANS may supplement surface sensitive methods such as Kerr-, magnetic force- or Lorentz microscopy. SANS studies do exist on soft magnetic [2–5] as well as on hard magnetic [6] single-component nc ferromagnets.

Here, we present a SANS study of the field dependence of the magnetic microstructure of nc Ni. Our previous analysis of SANS by soft magnets was based on a micromagnetics model which requires the material to be nearly saturated, and which supplies information on the magnetic interactions. Here, we employ a model-independent approach, which ignores the magnetic interactions, but which remains valid far from sa-

[1]) Corresponding author; Phone: +49-681-302-5174; Fax: +49-681-302-5222; e-mail: a.michels@mx.uni-saarland.de

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turation. We show how an appropriate, model-independent magnetic correlation length can be defined and measured, and we compare the results for the field dependence with theoretical predictions.

2. Experimental Details The nc Ni sample was prepared by pulsed electrodeposition [1] and consisted of a single 330 μm thick sheet with a purity of 99.99% and a mass density of (100.0 ± 0.2)% of the literature value of the coarse-grained material. An area-weighted average grain size, $D = 49$ nm, was determined by Warren-Averbach's analysis [7]. The SANS experiment was carried out at instrument NG 3 [8] at the National Institute of Standards and Technology using unpolarized neutrons with a wavelength $\lambda = 0.6$ nm and wavelength spread $\Delta\lambda/\lambda = 0.15$; the SANS data were corrected for absorption, background, dark current and converted to absolute units by means of a porous SiO_2 sample. The external magnetic field was provided by a 2 T electromagnet and was applied normal to the incident neutron wavevector and in the plane of the sheet. A magnetization isotherm at $T = 300$ K verified that a field of $\cong 0.1$ T is sufficient to approach saturation. For further experimental details see Refs. [5, 7].

3. The Correlation Function of the Spin Misalignment The correlation function $C(\mathbf{r})$ of the spin misalignment can be defined as follows [6]:

$$C(\mathbf{r}) = \frac{1}{V}\int \frac{\mathbf{M}_p(\mathbf{x})\,\mathbf{M}_p(\mathbf{x}+\mathbf{r})}{M_s^2}\,\mathrm{d}^3\mathbf{x}\,, \tag{1}$$

where V denotes the sample volume, M_s is the saturation magnetization and the function $\mathbf{M}_p(\mathbf{x})$ is the difference between the local magnetization $\mathbf{M}(\mathbf{x})$ and the mean magnetization $\langle\mathbf{M}\rangle$, which is directed along the applied magnetic field $\mathbf{H}_i$. For isotropic systems, $C(\mathbf{r})$ can be related to an experimental magnetic SANS cross-section $\mathrm{d}\Sigma_{mag}/\mathrm{d}\Omega$ [6],

$$C(r) = \frac{a}{12\pi^2 b_m^2 \varrho_a^2 r}\int_0^\infty \frac{\mathrm{d}\Sigma_{mag}}{\mathrm{d}\Omega}\,\sin(qr)\,q\,\mathrm{d}q\,, \tag{2}$$

where b_m and ϱ_a denote the atomic magnetic scattering length and the atomic density, respectively. For a completely isotropic ferromagnet, at zero or vanishing internal 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 the variation of the magnetization is confined to the plane perpendicular to the applied field [6] (the value $a = 8$ for the nearly saturated ferromagnet corrects the erroneous value $a = 32/\pi$ reported in Ref. [6]). We are interested in evaluating a magnetic "correlation length" l_C, that is, 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 $C(r)$ (compare Ref. [6]). As a robust measure, which is independent of small oscillations of C at small r, we shall here identify l_C with the value of r for which C is decreased to $1/e$ of the extrapolated value at $r = 0$. It is readily verified that this yields the correct correlation length when the correlation function decays exponentially, $C(r) = C(0)\exp(-r/l_C)$. For spherical particles with uniform magnetization, the correlation function is $C(r) = 1 - 3r/(2D) + r^3/(2D^3)$ [9], and the above definition yields $l_C \approx 0.45D$.

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 H_i the total SANS cross-section $d\Sigma/d\Omega$ of a nanocrystalline ferromagnet can be written as the sum of a combined nuclear and magnetic *residual* scattering cross-section $d\Sigma_{res}/d\Omega$ and of a pure *micromagnetic* scattering cross-section, $S_H \times R$ [10],

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

The residual scattering cross-section arises from regions within the sample of nonuniform nuclear density and/or composition (such as pores), and, in the limit of high applied fields, $d\Sigma_{res}/d\Omega$ is independent of 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*, $R(q, H_i)$; it describes the small-angle scattering from small static fluctuations of the magnetization perpendicular to the applied field H_i. The approach via Eq. (3), which results from a combination of micromagnetics theory and neutron scattering formalism [10], has been successful in describing SANS data on dense samples of soft magnetic electrodeposited nc Ni and Co near saturation [5]. In particular, quantitative information on the magnetic microstructure, the exchange-stiffness constant, and on the strength and microstructure of the mean anisotropy field could be obtained [5]. Here, we only used this approach to determine the residual scattering cross-section, which was subtracted from the data before further analysis. For the later discussion, it is worth noting that the micromagnetics model [10] predicts that perturbations in the spin structure decay with a characteristic length scale,

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

where A denotes the exchange-stiffness constant.

4. Experimental Results and Discussion Figure 1 shows the azimuthal-average total scattering cross-section $d\Sigma/d\Omega$ of the nc Ni sample as a function of the modulus q of the scattering vector at $T = 295$ K and at different magnetic fields H_i as indicated in the figure caption. The solid lines in the figure connect the best-fit values of $d\Sigma/d\Omega$ calculated according to Eq. (3). From such data, the residual scattering cross-section $d\Sigma_{res}/d\Omega$ can be obtained, and $d\Sigma_{res}/d\Omega$ is plotted in Fig. 1; for further details see Refs. [5, 7]. The total scattering signal $d\Sigma/d\Omega$ is seen to decrease by nearly three orders of magnitude as H_i is increased to 1.8 T, and $d\Sigma/d\Omega$ is everywhere above $d\Sigma_{res}/d\Omega$ indicating a strong magnetic scattering in nc Ni, even at the highest applied field. By subtracting $d\Sigma_{res}/d\Omega$ from the total signal $d\Sigma/d\Omega$, the magnetic scattering cross-section $d\Sigma_{mag}/d\Omega$ is obtained (compare Eq. (3)) and can then be used in Eq. (2) to compute the correlation function $C(r, H_i)$.

Figure 2a depicts the results for the correlation functions $C(r, H_i)$ that were calculated using Eq. (2) with $a = 8$. Increasing the magnetic field H_i results in a suppression of the transversal magnetic fluctuations, which are described by $\mathbf{M}_p(\mathbf{x})$, and, hence, $C(r)$ decreases (for given r) when the field is increased. Secondly, depending on the applied

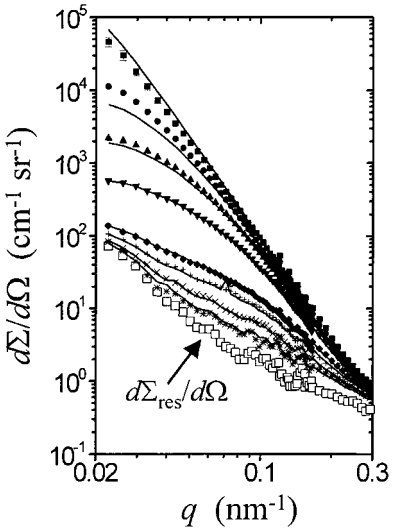

Fig. 1. Experimental (symbols) differential scattering cross-section $d\Sigma/d\Omega$ of nc Ni at $T = 295$ K versus modulus q of the scattering vector at different internal magnetic fields. Values of the internal field $\mu_0 H_i$ (in mT), from top to bottom: 0.5, 39, 88, 190, 570, 800, 1230, 1790. Fits to Eq. (3) (solid lines); the lines connect the fitted values of $d\Sigma/d\Omega$ at the discrete experimental q and H_i. Squares represent the residual scattering cross-section $d\Sigma_{res}/d\Omega$

magnetic field, the range of the ferromagnetic correlation extends to length scales that can be larger (at small H_i) or smaller (at large H_i) than the average grain size D of the nc Ni sample. The correlation length l_C determined as described above, is seen in Fig. 2b as a function of H_i. The variation of l_C confirms the conclusions that were already drawn by inspection of $C(r, H_i)$, namely a pronounced dependence on the applied field with values larger than expected for uniformly magnetized grains of the experimental size, $D = 49$ nm, at small H_i, and with values much smaller than D at large H_i.

It has been predicted 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 Eq. (4) [10]. This is supported by the field dependence of our experimental values for l_C , which vary asymptotically as $l_C \propto H_i^{-1/2}$ at small applied field (see Fig. 2b). However, the micromagnetics result would predict that the lower limit of l_C is given by the correlation length of the aniso-

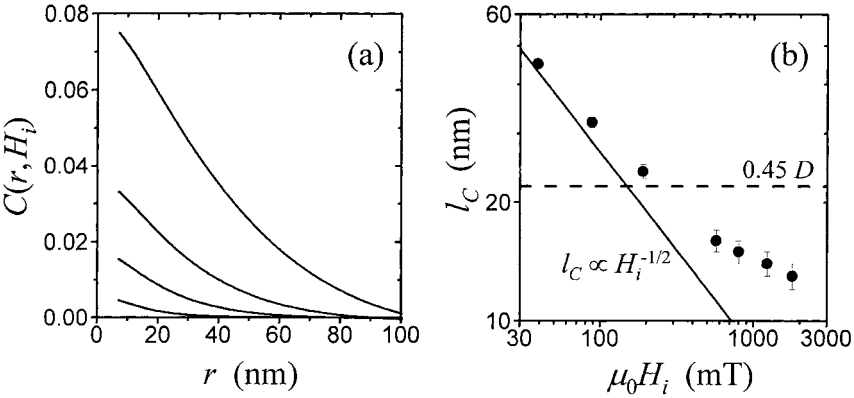

Fig. 2. a) Experimental correlation functions $C(r, H_i)$ of nc Ni at $T = 295$ K. The $C(r, H_i)$ have been computed using Eq. (2) with $a = 8$. Values of the internal magnetic field $\mu_0 H_i$ (in mT), from top to bottom: 39, 88, 190, 570. b) ● Correlation length l_C of nc Ni at $T = 295$ K versus internal magnetic field $\mu_0 H_i$. ——: $l_C \propto H_i^{-1/2}$; – – – $l_C = 0.45D$, with $D = 49$ nm being the experimental grain size

tropy field or, in other words, by the characteristic size of regions in which the magnetic easy axes are aligned in parallel. For grains with a uniform anisotropy this quantity is determined by the grain size D and by analogy to the correlation length of the spin misalignment it has the value $0.45D$, in our experiment about 22 nm. The experimental finding of $l_C < 0.45D$ at high fields implies that the anisotropy field $\mathbf{H}_p$ in the nc Ni sample is nonuniform on a scale smaller than the grain size. Possible origins of this internal nonuniformity of $\mathbf{H}_p$ within each grain are the large number of twin boundaries, or an interfacial anisotropy of grain boundaries, analogous to the magnetic anisotropy of free surfaces. The presence of extended inhomogeneities in $\mathbf{M}$ (at small H_i) on a length scale larger than D agrees qualitatively with predictions deduced from the "random anisotropy model" [11].

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References

[1] R. T. C. Choo, J. M. Toguri, A. M. El-Sherik, and U. Erb, J. Appl. Electrochem. **25**, 384 (1995).
[2] W. Wagner, A. Wiedenmann, W. Petry, A. Geibel, and H. Gleiter, J. Mater. Res. **6**, 2305 (1991).
[3] J. Weissmüller, R. D. McMichael, J. Barker, H. J. Brown, U. Erb, and R. D. Shull, Mater. Res. Soc. Symp. Proc. **457**, 231 (1997).
[4] J. Löffler, G. Kostorz, A. Wiedenmann, and W. Wagner, Physica B **241–243**, 603 (1997).
J. F. Löffler, H. B. Braun, and W. Wagner, Phys. Rev. Lett. **85**, 1990 (2000).
[5] A. Michels, J. Weissmüller, A. Wiedenmann, and J. G. Barker, J. Appl. Phys. **87**, 5953 (2000).
A. Michels, J. Weissmüller, A. Wiedenmann, J. S. Pedersen, and J. G. Barker, Philos. Mag. Lett. **80**, 785 (2000).
[6] J. Weissmüller, D. Michels, A. Michels, A. Wiedenmann, C. E. Krill, H. M. Sauer, and R. Birringer, Scripta Mater. **44**, 2357 (2001).
[7] J. Weissmüller, A. Michels, J. G. Barker, A. Wiedenmann, U. Erb, and R. D. Shull, Phys. Rev. B **63**, 214414 (2001).
[8] C. J. Glinka, J. G. Barker, B. Hammouda, S. Krueger, J. J. Moyer, and W. J. Orts, J. Appl. Cryst. **31**, 430 (1998).
[9] A. Guinier and G. Fournet, Small-Angle Scattering of X-Rays, John Wiley & Sons, New York 1955 (p. 15).
[10] J. Weissmüller, R. D. McMichael, A. Michels, and R. D. Shull, J. Res. NIST **104**, 261 (1999); http://www.nist.gov/jres.
[11] G. Herzer, IEEE Trans. Magn. **26**, 1397 (1990).