Theory of magnetic small-angle neutron scattering of two-phase ferromagnets

Dirk Honecker and Andreas Michels*

Physics and Materials Science Research Unit, University of Luxembourg, 162A Avenue de la Faëncerie, L-1511 Luxembourg, Grand Duchy of Luxembourg

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Based on micromagnetic theory, we have derived analytical expressions for the magnetic small-angle neutron scattering (SANS) cross section of a two-phase particle-matrix-type ferromagnet. The approach—valid close to magnetic saturation—provides access to several features of the spin structure, such as perturbing magnetic anisotropy and magnetostatic fields. Depending on the applied magnetic field and on the magnitude of the magnetic anisotropy field relative to the magnitude of the jump in the longitudinal magnetization at the particle-matrix interface, we observe a variety of angular anisotropies in the magnetic SANS cross section. In particular, the model explains the “clover-leaf”-shaped angular anisotropy, which was previously observed for several nanostructured magnetic materials, and provides access to the magnetic interaction parameters, such as the average exchange-stiffness constant. It is also shown that the ratio of the magnetic SANS cross section depends on the Fourier coefficients of the magnetization, which are used in Sec. IV in order to compute the magnetic SANS cross section; here, we focus on the two most relevant scattering geometries where the externally applied magnetic field is perpendicular or parallel to the incident neutron beam. Section V discusses the results in real space in terms of the corresponding spin-spin correlation function. Finally, Sec. VI summarizes the main findings of this work.

I. INTRODUCTION

Magnetic small-angle neutron scattering (SANS) is one of the most important techniques for microstructure determination in magnetism and magnetic materials research. The essential advantages of the SANS method may be summarized by stating that (i) SANS provides nanometer-scale information (∼1–100 nm) from within the bulk of a sample and (ii) SANS experiments can be conducted under rather flexible conditions and different sample environments (temperature, electric and magnetic field, pressure, neutron polarization, time-resolved data acquisition, etc.). For instance, SANS has previously been used for investigating the anisotropy of the structure factor and the relaxation dynamics of magnetic fluids,1–3 the internal magnetic field, pressure, neutron polarization, time-resolved data acquisition, etc.). For instance, SANS has previously been used for investigating the anisotropy of the structure factor and the relaxation dynamics of magnetic fluids,1–3 the internal structure and morphology of magnetic nanoparticles and nanowire arrays,4–10 the vortex lattice of superconductors,11 domain sizes and magnetization reversal in exchange-bias systems,12 and the spin distribution of nanocrystalline13–28 and amorphous29–32 metals.

The scattering contrast in magnetic SANS arises from deviations of both the magnitude and orientation of the local magnetization vector field M(r) from its mean value. The phenomenological continuum theory of micromagnetics33–35 allows one to compute the magnetic microstructure of a material, i.e., the variation of the direction of M as a function of the position r and time. Since the elastic magnetic differential scattering cross section depends on the Fourier coefficients of the magnetization, it is, in principle, straightforward to use micromagnetic theory for predicting and modeling magnetic SANS.36–38 However, due to the nonlinearity of the governing equations—Brown’s equations of micromagnetics—it is rather difficult to obtain analytical closed-form expressions for the cross section and, indeed, such solutions are limited to the approach-to-saturation regime where the micromagnetic equations can be linearized.

Based on the pioneering work of Kronmüller et al. (Ref. 39), we have previously derived analytical expressions for the differential magnetic scattering cross section of ferromagnets with uniform values of the saturation magnetization and the exchange interaction, but with a highly nonuniform magnetocrystalline and/or magnetoelastic anisotropy.40,41 Examples for such materials are nanocrystalline or cold-worked metals which exhibit nanoscale variations in the direction and/or magnitude of the magnetic anisotropy, leading to a highly inhomogeneous spin structure along with a strongly field-dependent magnetic SANS signal. As summarized in Ref. 42, analysis of experimental SANS data on nanocrystalline cobalt and nickel and on cold-worked coarse-grained nickel yields information on the strength and spatial structure of the magnetic anisotropy field, the magnitude and field dependence of the magnetostatic field, the exchange constant, and the characteristic length of the spin misalignment.

It is the purpose of this paper to extend the micromagnetic scattering approach to materials having nonuniform saturation magnetization. Prominent examples are two-phase hard and soft magnetic nanocomposites, which are used, e.g., in electronics devices, transformers, and motors.43,44 Here, the jump in the magnitude of the magnetization at the particle-matrix interface gives rise to a magnetostatic stray field, which represents a dominating source of spin disorder.45

The paper is organized as follows: To start with, we summarize in Sec. II well-known results for the magnetic SANS cross section of a fully saturated two-phase particle-matrix-type ferromagnet. Section III describes the micromagnetic model and provides approximate expressions for the Fourier coefficients of the magnetization, which are used in Sec. IV in order to compute the magnetic SANS cross section; here, we focus on the two most relevant scattering geometries where the externally applied magnetic field is perpendicular or parallel to the incident neutron beam. Section V discusses the results in real space in terms of the corresponding spin-spin correlation function. Finally, Sec. VI summarizes the main findings of this work.

II. SATURATED STATE

It is our aim to derive an approximate expression for the magnetic microstructure and the ensuing magnetic SANS
cross section of a two-phase nanocomposite ferromagnet, i.e., a bulk material which consists of a distribution of ferromagnetic nanoparticles that are embedded in and magnetically exchange coupled to a ferromagnetic matrix; all particles are assumed to possess a saturation magnetization of $M_p$, whereas the saturation magnetization of the matrix phase is denoted by $M_m$. We remind the reader that for small-angle scattering, the discrete atomic structure of both the particle and matrix phases is generally of no relevance. Therefore, as far as magnetic SANS is concerned, the magnetization state may be represented by a continuous magnetization vector field $\mathbf{M}(r)$. Before discussing in detail in Sec. III the micromagnetic model of such a two-phase nanocomposite structure, we find it instructive to consider first the fully saturated netic model of such a two-phase nanocomposite structure, $R$ spherical particle (with radius $r$ and arrangement of the particles [compare Eq. (42) below].

At $V_p = 2\pi r^3$, where the longitudinal component

$$M_z = M_p u(r) + M_m (1 - u(r)), \quad (1)$$

where $u(r) = 1$ inside the particle phase and $u(r) = 0$ inside the matrix phase. By assuming that the function $M_z(r)$ can be expressed as a Fourier integral,

$$M_z(r) = \frac{1}{(2\pi)^{3/2}} \int \tilde{M}_z(q) \exp(iqr)d^3q, \quad (2)$$

$$\tilde{M}_z(q) = \frac{1}{(2\pi)^{3/2}} \int M_z(r) \exp(-iqr)d^3r, \quad (3)$$

it is straightforward to obtain the following limiting cases: At $q = 0$, one obtains the macroscopic saturation magnetization $M_s$ of the sample,

$$M_s = \langle M_z \rangle = V^{-1} \int M_z(r)d^3r = \frac{(2\pi)^{3/2}}{V} \tilde{M}_z(q = 0) = M_p p + M_m (1 - p), \quad (4)$$

where $V$ is the sample volume and $p$ equals the volume fraction of the particles. At $q \neq 0$, we obtain $\tilde{M}_z(q) = \frac{\Delta M}{(2\pi)^{3/2}} I_p(q), \quad (5)$

where $\Delta M = M_p - M_m$ denotes the jump in the magnetization magnitude at the particle-matrix interface and the function $I_p(q)$ contains information about the size distribution, shape, and arrangement of the particles [compare Eq. (42) below].

In the monodisperse and dilute limit, we find, for a single spherical particle (with radius $R$),

$$\tilde{M}_z(q) = \frac{\Delta M}{(2\pi)^{3/2}} 3 V_p j_1(qR) qR \quad (6)$$

where $V_p = \frac{4\pi}{3} R^3$ and $j_1(qR)$ denotes the spherical Bessel function of the first order. By using Eq. (6), we can immediately write down (in the monodisperse and dilute limit) the magnetic SANS cross section $d\Sigma_{SANS}/d\Omega$ for a collection of $N_p$ saturated particles in a saturated matrix. For the particular scattering geometry where the wave vector $k_0$ of the incident neutron beam is perpendicular to the applied magnetic field $H_0$, we obtain the well-known expression

$$d\Sigma_{SANS}(q) = \frac{N_p}{V} \Delta \rho_{mag}^2 V_p^2 F^2(qR) \sin^2 \theta, \quad (7)$$

where $\Delta \rho_{mag}^2 = b_H M^2$ represents the magnetic scattering-length density contrast, $F(qR) = 3 \frac{iqR}{\nu^2}$ is the form factor of the sphere, and $\theta$ denotes the angle between the scattering vector $q$ and $H_0$ [compare Eq. (22) below].

Deviations from the saturated state result in the emergence of transversal magnetization components. The associated so-called spin-misalignment scattering will be addressed in the following sections.

### III. MICROMAGNETIC MODEL

Analytical micromagnetic calculations of the type presented here have already been carried out by other authors (e.g., Refs. 46–48). We employ the micromagnetic approach for computing (in the high-field limit) the differential magnetic scattering cross section (see Sec. IV). In the following, we summarize the basic micromagnetic equations.

We are interested in the elastic spin-misalignment scattering which results from the static magnetic microstructure of a two-phase nanocomposite sample. Therefore, we start our analysis by writing down Brown's balance-of-torques equation,$^{31-35}$

$$\mathbf{M}(r) \times H_{eff}(r) = 0, \quad (8)$$

which expresses the fact that at static equilibrium, the torque on the magnetization vector field $\mathbf{M}(r)$ due to an effective magnetic field $H_{eff}(r)$ vanishes everywhere. In the micromagnetic model, we assume a uniform exchange interaction, but an explicitly wave-vector-dependent longitudinal magnetization (see below). The effective field,

$$H_{eff}(r) = H_0 + H_u(r) + H_p(r) + \frac{2A}{\mu_0 M_s^2} \nabla^2 \mathbf{M}(r), \quad (9)$$

is composed of a uniform applied magnetic field $H_0$, the magnetostatic field $H_u(r)$, the magnetic anisotropy field $H_p(r)$, and the exchange field [last term on the right-hand side of Eq. (9)]; $\mu_0 = 4\pi \times 10^{-7}$ Tm/A is the permeability of free space, and $V = e_x \partial / \partial x + e_y \partial / \partial y + e_z \partial / \partial z$, where $e_x$, $e_y$, and $e_z$ represent the unit vectors along the Cartesian laboratory axes. The parameter $A$ denotes the exchange-stiffness constant.

In the following, we assume the material to be nearly saturated along $H_0 \parallel \mathbf{e}_z$, i.e., we write

$$\mathbf{M}(r) = M_s(r)e_z + M_x(r)e_x + M_y(r)e_y, \quad (10)$$

with $M_z \ll M_x$ and $M_z \ll M_z$ (small-misalignment approximation). The local saturation magnetization is assumed to differ only slightly from its spatial average, i.e., $M_z(r) \equiv \langle M_z \rangle = M_s$. Note that the jump $\Delta M$ in the longitudinal magnetization enters the calculation in $q$ space via the expression for the Fourier coefficient of the magnetostatic field (see below). Furthermore, we assume that the anisotropy-energy density $\omega = \omega(r, \mathbf{M})$ depends only linearly on the components of the magnetization.$^{33}$ As a consequence, the resulting anisotropy
field, \( \mathbf{H}_p = -\mu_0^{-1}(\partial \mathbf{M} / \partial \mathbf{r}) \), is independent of \( \mathbf{M} \) and, therefore, also independent of the applied magnetic field, implying that near saturation \( \mathbf{H}_p = \mathbf{H}_p(\mathbf{r}) \). Due to the micromagnetic constraint \( \mathbf{M} = M_t \), an anisotropy-energy density of the form \( \omega = \omega(\mathbf{r}, M_x, M_y, M_z) \) may be reexpressed as \( \omega = \omega(\mathbf{r}, M_x, M_z) \), with the consequence that only two independent components of \( \mathbf{H}_p \) exist. In the approach-to-saturation regime, when \( \mathbf{M} \) is nearly aligned parallel to the external magnetic field \( \mathbf{H}_0 \), only those components of \( \mathbf{H}_p \) which are normal to \( \mathbf{H}_0 \) are physically effective in producing a torque on the magnetization.

Basic magnetostatics prescribes that \( \nabla \cdot (\mathbf{H}_0 + \mathbf{H}_p) = 0 \) and \( \nabla \times (\mathbf{H}_0 + \mathbf{H}_p) = 0 \). The magnetostatic field \( \mathbf{H}_d(\mathbf{r}) \) can be written as the sum of the surface demagnetizing field \( \mathbf{H}_d^s \) and of the magnetostatic field \( \mathbf{H}_d^b \) which is related to volume charges, i.e., \( \mathbf{H}_d = \mathbf{H}_d^s + \mathbf{H}_d^b \). In the high-field limit and for samples with an ellipsoidal shape with \( \mathbf{H}_0 \) directed along a principal axis of the ellipsoid, one may approximate the demagnetizing field due to the surface charges by the uniform field \( \mathbf{H}_d^s = -NM \mathbf{e}_z \), where \( N \) denotes the corresponding demagnetizing factor. In Fourier space (at \( \mathbf{q} \neq 0 \)), the above magnetostatic relations suggest the following expression for the Fourier coefficient \( h_y^s(\mathbf{q}) \) of \( \mathbf{H}_d^s(\mathbf{r}) \):\(^9\)

\[
h_y^s(\mathbf{q}) = - \frac{q[q \cdot \mathbf{M}(\mathbf{q})]}{q^2}.
\]

The terms in Eqs. (15) and (16) which contain the Fourier coefficients \( \mathbf{M}_s(\mathbf{q}) \) and \( \mathbf{M}_j(\mathbf{q}) \) model the influence of the two-phase microstructure on the magnetic SANS, is introduced into our theory only in the \( \mathbf{q} \) space via \( h_y^s(\mathbf{q}) = - \mathbf{q}[\mathbf{q} \cdot \mathbf{M}(\mathbf{q})]/q^2 \) [Eq. (11)]. This is an approximation, since in real space we have assumed that \( M_z \approx M_t = \text{const.} \), and hence \( \mathbf{M}_s(\mathbf{q}) \propto \delta(\mathbf{q}) \) would result, as is appropriate for a homogeneous single-phase material. However, by explicitly considering the \( \mathbf{q} \neq 0 \) Fourier coefficients of \( \mathbf{M}_s \) [Eq. (5)], it becomes possible to straightforwardly include the jump in the longitudinal magnetization at the particle-matrix interface, and one avoids the otherwise necessary calculation of convolution products.\(^{46-48}\)

For the following discussion of magnetic SANS, it is of interest to consider special projections of Eqs. (15) and (16) into the plane of the two-dimensional (2D) detector. The two scattering geometries which are of particular relevance to experiment have the external magnetic field \( \mathbf{H}_0 \) either perpendicular or parallel to the wave vector \( \mathbf{k}_0 \) of the incoming neutron beam. For \( \mathbf{k}_0 \parallel \mathbf{e}_i \), and \( \mathbf{H}_0 \parallel \mathbf{e}_i \), the scattering vector can be approximated as \( \mathbf{q} \approx (0,q_y,q_z) \), i.e., \( q_x \approx 0 \), and Eqs. (15) and (16) reduce to

\[
\mathbf{M}_s(\mathbf{q}) = M_s \frac{h_y(q)}{H_{\text{eff}}},
\]

\[
\mathbf{M}_j(\mathbf{q}) = M_j \frac{h_y(q) - \mathbf{M}_s(\mathbf{q}) q_y q_z}{H_{\text{eff}} + M_s q_y^2/q^2}.
\]
For \( \mathbf{k}_0 \parallel \mathbf{H}_0 \parallel \mathbf{e}_z \), \( q \equiv (q_x, q_y, 0) \), i.e., \( q_z \equiv 0 \), and the results for the Fourier coefficients simplify to

\[
\tilde{M}_s(q) = M_s \frac{h_x(H_{\text{eff}} + M_s q_x^2 / q^2) - h_y M_s q_y^2 / q^2}{H_{\text{eff}}(H_{\text{eff}} + M_s q_x^2 / q^2)},
\]

\[
\tilde{M}_s(q) = M_s \frac{h_y(H_{\text{eff}} + M_s q_y^2 / q^2) - h_x M_s q_x^2 / q^2}{H_{\text{eff}}(H_{\text{eff}} + M_s q_y^2 / q^2)}.
\]

Equations (20) and (21) are similar to Eqs. (2.15) in Ref. 42.

The high-field solutions for \( \tilde{M}_s(q) \) and \( \tilde{M}_m(q) \) can be seen as a sum of products of components of the anisotropy-field Fourier coefficient \( \mathbf{h}(q) \) and \( M_s(q) \) with micromagnetic functions which contain the effective magnetic field \( H_{\text{eff}} \) and terms that depend on the orientation of the wave vector (e.g., \( M_s q_x^2 / q^2 \)). The convolution theorem then implies that the magnetic microstructure in real space, \( \mathbf{M}(r) \), is tantamount to a complicated convolution product between the corresponding real-space functions. As a consequence, sharp features in the nuclear or anisotropy-field microstructure are washed out and smoothly varying magnetization profiles are at the origin of the related spin-misalignment scattering (compare, e.g., Figs. 2–4 in Ref. 51). Consistent with this notion is the observation of power-law exponents significantly larger than 4 (see Fig. 4 below) and the finding that the slope of the correlation function at the origin vanishes (see Fig. 5 below). 53

IV. SANS CROSS SECTION FOR UNPOLARIZED NEUTRONS

Although the following derivation of the magnetic SANS cross section is aimed to be self-contained, we refer the reader to Refs. 40 and 41 for further details.

A. \( \mathbf{k}_0 \perp \mathbf{H}_0 \)

For the transversal scattering geometry, one can express the total nuclear and magnetic SANS cross section \( d\Sigma /d\Omega \) at scattering vector \( q \) as

\[
d\Sigma /d\Omega(q) = \frac{8\pi^3}{V} b_H^2 \left[ \frac{|\tilde{N}|^2}{b_H^2} + |\tilde{M}_s|^2 \right] \cos^2 \theta + |\tilde{M}_m|^2 \sin^2 \theta - (\tilde{M}_s \tilde{M}_m^* + \tilde{M}_m^* \tilde{M}_s) \sin \theta \cos \theta.
\]

Here, \( V \) is the scattering volume, \( b_H = 2.9 \times 10^8 \text{ A}^{-1} \text{ m}^{-1} \), \( \tilde{N} \) is the nuclear scattering amplitude, and \( \theta \) denotes the angle between \( \mathbf{H}_0 \) and \( q \equiv q (0, \sin \theta, \cos \theta) \); \( c^* \) is a quantity that is complex conjugated to \( c \). The magnetic form factor in the expression for the magnetic scattering amplitude \( \langle \chi_{\mathbf{H}} \rangle \) was set to unity, which is permissible along the forward direction.

By inserting Eqs. (18) and (19) into Eq. (22), one can express \( d\Sigma /d\Omega \) as

\[
d\Sigma /d\Omega(q) = d\Sigma_{\text{res}}(q) + d\Sigma_{M}(q),
\]

where

\[
d\Sigma_{\text{res}}(q) = \frac{8\pi^3}{V} b_H^2 \left( \frac{|\tilde{N}|^2}{b_H^2} + |\tilde{M}_m|^2 \sin^2 \theta \right)
\]

represents the (nuclear and magnetic) residual SANS cross section, which may be measured at complete magnetic saturation. Subtraction of \( d\Sigma_{\text{res}} /d\Omega \) from a measurement of \( d\Sigma /d\Omega \) at a lower field yields the spin-misalignment SANS cross section \( d\Sigma_{M} /d\Omega \), which contains the scattering contributions due to the misaligned spins,

\[
d\Sigma_{M}(q) = S_H(q) R_H(q, \theta, H_{eff}) + S_M(q) R_M(q, \theta, H_{eff}).
\]

Following Weissmüller et al. (Ref. 40), we have here introduced the scattering function of the anisotropy field,

\[
S_H(q) = \frac{8\pi^3}{V} b_H^2 \tilde{h}^2(q),
\]

the scattering function of the longitudinal magnetization,

\[
S_M(q) = \frac{8\pi^3}{V} b_H^2 \tilde{M}_m^2(q),
\]

and the corresponding (dimensionless) micromagnetic response functions,

\[
R_H(q, \theta, H_{eff}) = \frac{p^2}{2} \left[ 1 + \frac{\cos^2 \theta}{(1 + p \sin^2 \theta)^2} \right],
\]

\[
R_M(q, \theta, H_{eff}) = \frac{p^2 \sin^2 \theta \cos^2 \theta}{(1 + p \sin^2 \theta)^2} + \frac{2p \sin^2 \theta \cos^2 \theta}{1 + p \sin^2 \theta},
\]

where \( p(q, H_{eff}) = M_s / H_{eff}(q, H_{eff}) \). We note that the term \( S_M \times R_M \) in Eq. (25) [and Eq. (31)] is not contained in the expression for the single-phase material case (homogeneous saturation magnetization), where

\[
d\Sigma_{M}(q) = S_H(q) R_H(q, \theta, H_{eff}).
\]

Near magnetic saturation, \( S_H \) and \( S_M \) are both approximately independent of the applied magnetic field and contain, respectively, information on the strength and the spatial structure of the magnetic anisotropy field, 43 as well as on the magnitude of the jump of the magnetization at the particle-matrix interface. The geometry of the microstructure is contained in \( S_H \) and \( S_M \). Both response functions depend on the magnitude and orientation of the scattering vector, the applied field, and the magnetic interaction parameters.

As mentioned previously, the effects of crystallographic texture or of other forms of anisotropy on \( d\Sigma_{M} /d\Omega \) enter the theory mainly through the magnetic anisotropy field \( \mathbf{h}(r) \). In deriving Eqs. (23)–(29), we have assumed that the corresponding anisotropy-field Fourier coefficients \( \mathbf{h}(q) = [h(q) \cos \beta, h(q) \sin \beta, 0] \) is isotropically distributed in the plane perpendicular to \( \mathbf{H}_0 \); in other words, the vector \( \mathbf{h}(q) \) takes on all orientations (i.e., angles \( \beta \)) with equal probability. This assumption allows us to average the response functions over \( \beta \), i.e., \( 1/(2\pi) \int_{0}^{2\pi} \ldots d\beta \), which results in Eqs. (28) and (29); note that interference terms \( \chi_{\mathbf{h}} \tilde{M}_s \) vanish in this averaging procedure (see also Sec. IV C below). The case of a texture in the orientation of the anisotropy field is treated in the appendix of Ref. 41.
Furthermore, by assuming that both Fourier coefficients $h^2$ and $\tilde{M}_z^2$ depend only on the magnitude q of the scattering vector $\mathbf{q}$, and not on its orientation $\theta$, one may average Eq. (25) [and Eq. (30)] with respect to the angle $\theta$, i.e., $1/(2\pi) \int_0^{2\pi} \cdots d\theta$, which results in

$$\frac{d\Sigma_M}{d\Omega}(\mathbf{q}) = S_H(q) R_H(q, H_i) \times S_M(q) R_M(q, H_i),$$

(31)

where

$$R_H(q, H_i) = \frac{p^2}{4} \left( 1 + \frac{1}{\sqrt{1 + p^2}} \right),$$

(32)

$$R_M(q, H_i) = \frac{\sqrt{1 + p^2} - 1}{2}. \quad (33)$$

Equation (31) may be compared to experimental data for $d\Sigma_M/d\Omega$ in order to obtain the functions $S_H$ and $S_M$ and a value for the average exchange-stiffness constant $A$. The micromagnetic equations for the spin-flip SANS cross section can be found in the Appendix.

B. $k_0 \parallel H_0$

For the longitudinal SANS geometry, $d\Sigma/d\Omega$ reads

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \sum_{j=1}^{N_p} \frac{8\pi^3}{V} b_H^2 \left[ |\mathbf{N}|^2 |\mathbf{b}_H|^2 + |\tilde{M}_x|^2 \sin^2 \theta + |\tilde{M}_z|^2 \cos^2 \theta \
+ |\tilde{M}_z|^2 - (\tilde{M}_x \tilde{M}_y + \tilde{M}_y \tilde{M}_x) \sin \theta \cos \theta \right]. \quad (34)$$

where $\mathbf{q} \equiv q (\cos \theta, \sin \theta, 0)$ and $\theta = \angle(\mathbf{q}, \mathbf{e}_z)$. Inserting Eqs. (20) and (21) into Eq. (34) and averaging over the orientations of the magnetic anisotropy field (angle $\beta$) results in

$$\frac{d\Sigma}{d\Omega}(\mathbf{q}) = \frac{d\Sigma_{\text{res}}}{d\Omega}(\mathbf{q}) + \frac{d\Sigma_{M}}{d\Omega}(\mathbf{q}),$$

(35)

where the residual SANS cross section equals

$$\frac{d\Sigma_{\text{res}}}{d\Omega}(\mathbf{q}) = \frac{8\pi^3}{V} b_H^2 \left[ |\mathbf{N}|^2 |\mathbf{b}_H|^2 + |\tilde{M}_x|^2 \right].$$

(36)

and the spin-misalignment SANS cross section is expressed as

$$\frac{d\Sigma_{M}}{d\Omega}(\mathbf{q}) = S_H(q) R_H(q, H_i),$$

(37)

with an isotropic (i.e., $\theta$-independent) response function,

$$R_H(q, H_i) = \frac{p^2}{2}. \quad (38)$$

We note that in the longitudinal SANS geometry, $d\Sigma_M/d\Omega$ is independent of $\mathbf{M}$, and equals the expression for the single-phase case [compare Eq. (33) in Ref. 40]. In other words, the two-phase nature of the underlying microstructure is (for $k_0 \parallel H_0$) only contained in the residual SANS, and not in $d\Sigma_M/d\Omega$.

C. Comment on the average of $d\Sigma_M/d\Omega$ over many statistically uncorrelated defects

The sample volume which is probed by the neutrons typically contains many defects (e.g., particles), with each one having a different orientation and/or magnitude of the magnetic anisotropy field. For ferromagnets with a nonuniform saturation magnetization, each particle or crystallite $j$ is additionally characterized by the form-factor function $M_{s,j}$. In order to discuss the statistical average over the spin-misalignment SANS cross sections of many defects, we ignore for the moment the $\Delta M$ fluctuations, i.e., we consider the case of a "homogeneous single-phase ferromagnet" with uniform values of $A$ and $M_s$. Here, the dominating source of spin disorder (in the approach-to-saturation regime) is related to spatially inhomogeneous magnetic anisotropy fields. We follow the arguments of Weissmüller[40,41] and assume that the total anisotropy-field Fourier coefficient of the sample, $h(q)$, can be expressed as the sum of the anisotropy-field amplitudes of the individual defects,

$$h(q) = \sum_j h_j(q). \quad (39)$$

If the $h_j$ of the individual defects are statistically uncorrelated (random anisotropy), then the expectation value of $|h(q)|^2$ reduces to the sum over the individual $|h_j|^2$,

$$|h(q)|^2 = \sum_j |h_j(q)|^2. \quad (40)$$

Since—within the linear micromagnetic approximation—$d\Sigma_M/d\Omega$ is proportional to $|h(q)|^2$ [compare Eq. (30)], it immediately follows that the above additivity also transfers to the total spin-misalignment SANS cross section of the sample. In other words,

$$\frac{d\Sigma_M}{d\Omega} = \sum_j \frac{d\Sigma_{M,j}}{d\Omega} = \sum_j S_{H,j} R_{H,j},$$

(41)

when homogeneous single-phase ferromagnets are considered.40,41 This is in contrast to nuclear SANS or small-angle x-ray scattering (SAXS), where the decomposition of the overall cross section into the sum over cross sections of individual particles is only permissible for small particle volume fractions. Including now the $\Delta M$ fluctuations in the discussion, an inspection of Eq. (19) shows that interference terms $x h_j \times \mathbf{M}_{s,j}$ appear in the spin-misalignment SANS cross section [compare Eq. (22)]. However, in deriving the final expression for $d\Sigma_M/d\Omega$ [Eq. (25)], these terms cancel when the averaging procedure over the (isotropic) orientations of the anisotropy field (angle $\beta$) are carried out, with the result that the scattering contributions due to inhomogeneous anisotropy fields, $S_H \times R_H$, and magnetostatic fluctuations, $S_M \times R_M$, are additive. Taking into account interparticle interference effects, the general expression for $|\bar{M}_z|^2$ (at $\mathbf{q} \neq 0$) may be expressed as

$$|\bar{M}_z(\mathbf{q})|^2 = \frac{(\Delta M)^2}{8\pi^3} \sum_{j=1}^{N_p} V_{p,j} F_j(q) \exp(-i \mathbf{q} \cdot \mathbf{r}_j)^2, \quad (42)$$

where $N_p$, $V_{p,j}$, $F_j$, and $\mathbf{r}_j$ denote, respectively, the number of particles, volume, form factor, and position vector of particle $j$ [compare Eq. (6)]. Equation (42) can be employed in Eqs. (25) or (31) in order to analyze experimental data.
FIG. 1. Micromagnetic response functions $R_H$ [Eq. (32), solid line] and $R_M$ [Eq. (33), dashed line] vs $p(q, H_i) = M_i/H_{eff}(q, H_i)$ (log-log scale).

D. Graphical representation of $d\Sigma_M/d\Omega$

In order to graphically display $d\Sigma_M/d\Omega$ [Eq. (25)], it is necessary to specify particular models for the Fourier coefficients $h(q)$ and $\tilde{M}_z(q)$. For $\tilde{M}_z(q)$, we use for simplicity the form factor of the sphere [Eq. (6)], which implies the neglect of interparticle interactions (dilute limit). We note, however, that the inclusion of the structure factor of the material, other particle shapes (form factors), or particle-size distribution is straightforward [compare Eq. (42)]. For $h(q)$, we employ the model introduced by Weissmüller et al. [Eq. (43) in Ref. 40], which considers a nanocrystalline material composed of spherical particles with a constant magnitude but random orientation of $\mathbf{H}_p$. The resulting expression for $h(q)$ is the form factor of the sphere, given by Eq. (6), where $\Delta M$ is replaced by the magnitude $H_p$ of the magnetic anisotropy field. For the particle radius, we assume a value of $R = 5$ nm for both $M_i(qR)$ and $h(qR)$. Since, then, $M_i(qR)$ and $h(qR)$ and, hence, $S_M$ and $S_H$ are equal except for the prefactors, the ratio $H_p/\Delta M$ determines the properties of $d\Sigma_M/d\Omega$. Unless otherwise stated, we have used the following materials parameters: $A = 2.5 \times 10^{-11}$ J/m, $\mu_0 M_s = 1.5$ T, and $\mu_0 \Delta M = 0.25$ T.

The spin-misalignment SANS cross section for $k_0 \perp H_0$ [Eq. (25)] contains scattering contributions due to the magnetic anisotropy field, $S_H \times R_H$, and due to jumps in the longitudinal magnetization, $S_M \times R_M$. In Fig. 1, we plot both response functions $R_H$ [Eq. (32)] and $R_M$ [Eq. (33)] as a function of the dimensionless parameter $p$. Assuming that $S_H$ and $S_M$ are of comparable magnitude, it is seen that at large applied fields or large momentum transfers (when $p \ll 1$), $d\Sigma_M/d\Omega$ is governed by the $S_M \times R_M$ contribution (see Fig. 2 below).

Figure 2 qualitatively displays the applied-field dependence of $d\Sigma_M/d\Omega$ for $k_0 \perp H_0$ [Eq. (25)] and for a ratio of $H_p/\Delta M = 1$ ($S_H = S_M$): the results are compared with the $d\Sigma_M/d\Omega$ of a single-phase material with a uniform saturation magnetization [Eq. (30)]. The $d\Sigma_M/d\Omega$ for the two-phase case [Figs. 2(a)–2(d)] reveal a strongly field-dependent angular anisotropy. At the largest fields, the pattern exhibits maxima roughly along the diagonals of the detector—the so-called clover-leaf anisotropy—previously observed in the Fe-based two-phase alloy Nanoperm (compare, e.g., Fig. 3 in Ref. 45). The position of the maxima in $d\Sigma_M/d\Omega$ depends on $q$ and

FIG. 2. (Color online) Upper row: Contour plots of $d\Sigma_M/d\Omega$ (in arbitrary units) for $k_0 \perp H_0$ [Eq. (25)]. $H_0$ is horizontal. $H_p/\Delta M = 1$ ($S_H = S_M$). $H_i$ values (in T): (a) 0.01, (b) 0.2, (c) 1.0, and (d) 10.0. Lower row: Field dependence of $d\Sigma_M/d\Omega$ for $k_0 \perp H_0$ for a homogeneous single-phase ferromagnet [Eq. (30)], where $\Delta M = 0$. Field values and materials parameters in (e)–(h) are the same as in (a)–(d). The yellow color corresponds to “high intensity” and the blue color corresponds to “low intensity.”
interaction and was first predicted by Weissmuller

\[ H(\mu, H_0) \text{ of arbitrary units} \] for \( k \) and the blue color corresponds to “low intensity.”

(b) 1.6, and (c) 8. The yellow color corresponds to “high intensity”

contribution to \( \mu \) into account, it is readily verified that the anisotropy-field

sharp spike in Figs. 2(a) and 2(e) is due to the magnetostatic

field-dominated scattering, with a characteristic clover-leaf-

/Delta_1 M

Figure 3 shows \( dS_M/d\Omega \) for \( k_0 \perp H_0 \) [Eq. (25)] at a fixed internal magnetic

field of \( \mu_0 H_1 = 2.0 \text{ T} \). \( H_0 \) is horizontal. Values of \( H_p/\Delta M \) are (a) 0.2, (b) 1.6, and (c) 8.

The yellow color corresponds to “high intensity” and the blue color corresponds to “low intensity.”

\( H_1 \) (see also Fig. 11 in Ref. 38). Such a type of clover-leaf

anisotropy cannot be reproduced by the \( dS_M/d\Omega \) for the single-phase case [Figs. 2(e)–2(h)]. Here, at large \( q \) and \( H_1 \), one observes an elongation of the spin-misalignment scattering along the field direction [due to the \( \cos^2 \theta \) term in Eq. (28)], with a “flying-saucer-type” pattern taking over at small \( q \) and \( H_1 \) [due to the \( \sin^2 \theta \) term in the denominator of Eq. (28)]. The sharp spike in Figs. 2(a) and 2(e) is due to the magnetostatic interaction and was first predicted by Weissmuller et al.*

Figure 3 shows \( dS_M/d\Omega \) for \( k_0 \perp H_0 \) [Eq. (25)] at a fixed internal magnetic field of \( \mu_0 H_1 = 2.0 \text{ T} \), but for different magnitudes of the magnetic anisotropy field \( H_1 \), relative to the jump \( \Delta M \) in the magnetization magnitude at the particle-matrix interface. One can clearly observe a transition from dipole-field-dominated scattering, with a characteristic clover-leaf-type pattern [Fig. 3(a)], to a more anisotropy-field-dominated \( \cos^2 \theta \)-type angular anisotropy of \( dS_M/d\Omega \) [Fig. 3(c)].

The following is in regard to the asymptotic power-law dependence of \( dS_M/d\Omega \). For particles with sharp interfaces, both \( h^2(q) \) and \( M^2_z(q) \) vary asymptotically as \( q^{-4} \), as does the function \( H_{\text{eff}}^2 \) [compare Eq. (17)]. Taking these dependencies into account, it is readily verified that the anisotropy-field contribution to \( dS_M/d\Omega \) varies as \( S_M \times R_M \propto q^{-8} \), whereas \( S_M \times R_M \propto q^{-6} \). Therefore, depending on the relative magnitude of both contributions to \( dS_M/d\Omega \), one observes different asymptotic power-law exponents (see Fig. 4). We note that other models for the anisotropy-field microstructure may result in different exponents.

\[ \text{Equation (44) embodies the convolution relationship between the anisotropy-field microstructure (through} \]

\[ l_c(H_1) = R + \frac{2A}{\mu_0 M_r H_1}. \]

Equation (44) embodies the convolution relationship between the anisotropy-field microstructure (through

\[ C(r) \propto \frac{1}{r} \int_0^\infty dS_M/d\Omega(q) \sin(qr)qdq. \]

Equation (43) has been solved numerically, and the results for \( C(r) \) at \( \mu_0 H_1 = 1.0 \text{ T} \) and for different ratios of \( H_p/\Delta M \) are shown in Fig. 5. The corresponding field dependence of the so-called correlation length \( l_c \) of the spin misalignment, which quantifies the size of inhomogeneously magnetized regions, can be seen in Fig. 6.

\[ C(r) \text{ (arb. units)} \]

\[ r \text{ (nm)} \]

\[ H_p/\Delta M = 4 \]

\[ H_p/\Delta M = 1 \]

\[ H_p/\Delta M = 0.2 \]

\[ \mu_0 H_1 = 1.0 \text{ T} \]

\[ C(r = l_c) = \exp(-1). \]

\[ \text{Dashed horizontal line: } C(r = l_c) = \exp(-1). \]
the field-independent parameter $R$ and micromagnetic functions [through the field-dependent exchange length $\sqrt{2 \lambda / (\mu_0 M_s H_s)}$]. Irrespective of the value of $H_p / \Delta M$, it is seen that at large fields, $l_C$ approaches the particle radius (dashed line in Fig. 6).

VI. SUMMARY AND CONCLUSIONS

Using the continuum theory of micromagnetics, we have derived in the approach-to-saturation regime analytical expressions for the magnetic small-angle neutron scattering cross section of a two-phase particle-matrix-type ferromagnet. For the particular scattering geometry where the applied magnetic field is perpendicular to the incoming neutron beam, the results for the spin-misalignment cross section $d \Sigma / d \Omega$ [Eq. (25)] exhibit a variety of angular anisotropies that are fundamentally different from the conventional $\sin^2 \theta$-type or $\cos^2 \theta$-type patterns. In particular, by explicitly taking into account the wave-vector dependence of the longitudinal magnetization, terms appear in $d \Sigma / d \Omega$ which give rise to maxima roughly along the diagonals of the detector (clover-leaf anisotropy), in agreement with experiment. Besides the value of the applied magnetic field, it is the ratio of the magnetic anisotropy field $H_p$ to the jump $\Delta M$ in the longitudinal magnetization at internal interfaces which determines the properties of $d \Sigma / d \Omega$, for instance, the asymptotic power-law exponent, the angular anisotropy, or the characteristic decay length of spin-misalignment fluctuations. Therefore, analysis of $d \Sigma / d \Omega$ will provide information on the strength and spatial structure of the magnetic anisotropy field, on $\Delta M$, and on the effective exchange-stiffness constant. Computation of the spin-spin correlation function corroborates the existence of long-range spin-misalignment fluctuations, with a characteristic field-dependent length that in the high-anisotropy-field limit ($H_p / \Delta M \gg 1$) can be described by Eq. (44). In contrast, perturbations in the spin structure that are due to magnetostatic fluctuations decay on a relatively short length scale.

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APPENDIX: MICROMAGNETIC EQUATIONS FOR THE SPIN-FLIP SANS CROSS SECTIONS

For polycrystalline bulk ferromagnets, the spin-flip SANS cross sections for the transversal ($\mathbf{k}_0 \perp \mathbf{H}_0$) and longitudinal ($\mathbf{k}_0 \parallel \mathbf{H}_0$) scattering geometry can, respectively, be written as25

$$
\frac{d \Sigma^{\pm \pm}}{d \Omega}(\mathbf{q}) = \frac{8 \pi^3}{V} b_H^2 \left[ |\vec{M}_z|^2 + |\vec{M}_y|^2 \sin^2 \theta \cos^2 \theta - (\vec{M}_y \vec{M}_z^* + \vec{M}_z \vec{M}_y^*) \sin \theta \cos^3 \theta \right],
$$

(A1)

$$
\frac{d \Sigma^{\pm \mp}}{d \Omega}(\mathbf{q}) = \frac{8 \pi^3}{V} b_H^2 \left[ |\vec{M}_z|^2 \sin^2 \theta + |\vec{M}_y|^2 \cos^2 \theta - (\vec{M}_y \vec{M}_z^* + \vec{M}_z \vec{M}_y^*) \sin \theta \cos \theta \right].
$$

(A2)

Since spiral and nuclear-spin incoherent scattering terms have been neglected in Eqs. (A1) and (A2), it follows that $d \Sigma^{\pm / \pm} / d \Omega = d \Sigma^{\pm / -} / d \Omega$. Furthermore, we note that $d \Sigma^{\pm / \pm} / d \Omega$ for the longitudinal geometry [Eq. (A2)] is identical to the corresponding spin-misalignment SANS cross section $d \Sigma / d \Omega$ for unpolarized neutrons [compare Eqs. (34)–(36)].

Subtracting (for $\mathbf{k}_0 \perp \mathbf{H}_0$) the scattering at saturation, $d \Sigma^{+ / -} / d \Omega = 8 \pi^3 V^{-1} b_H^2 |\vec{M}_z|^2 \sin^2 \theta \cos^2 \theta$, inserting Eqs. (18) and (19) into the remaining Eq. (A1), and averaging over the orientations of the anisotropy field results in

$$
\frac{d \Sigma^{z \mp}}{d \Omega}(\mathbf{q}) = S_H(\mathbf{q}) R_H^{+-}(q, \theta, H_i) + S_M(\mathbf{q}) R_M^{+-}(q, \theta, H_i),
$$

(A3)

where $S_H(\mathbf{q}) = 8 \pi^3 V^{-1} b_H^2 \vec{M}_z(\mathbf{q})$ and $S_M(\mathbf{q}) = 8 \pi^3 V^{-1} b_H^2 \vec{M}_z(\mathbf{q})$ remain unchanged, but

$$
R_H^{+-}(q, \theta, H_i) = \frac{p^2}{2} \left[ 1 + \frac{\cos^4 \theta}{(1 + p \sin^2 \theta)^2} \right],
$$

(A4)

$$
R_M^{+-}(q, \theta, H_i) = \frac{p^2 \sin^2 \theta \cos^6 \theta}{(1 + p \sin^2 \theta)^2} + \frac{2 p \sin^2 \theta \cos^4 \theta}{1 + p \sin^2 \theta}.
$$

(A5)
The azimuthal averages of the response functions read

\[
R^{±}_A(q, H_I) = \frac{2 + 2p^2 - (2 - p)\sqrt{1 + p}}{4}, \quad (A6)
\]

\[
R^{±}_B(q, H_I) = \frac{8(\sqrt{1 + p} - 1) - p[16 - 12\sqrt{1 + p} + p(9 - 4\sqrt{1 + p})]}{8p^2}. \quad (A7)
\]
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