Topical Review

Mathematical models for magnetic particle imaging

Tobias Kluth

Center for Industrial Mathematics, University of Bremen, Bibliothekstr. 5, 28357 Bremen, Germany

E-mail: tkluth@math.uni-bremen.de

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Abstract

Magnetic particle imaging (MPI) is a relatively new imaging modality. The nonlinear magnetization behavior of nanoparticles in an applied magnetic field is exploited to reconstruct an image of the concentration of nanoparticles. Finding a sufficiently accurate model to reflect the behavior of large numbers of particles for MPI remains an open problem. As such, reconstruction is still computed using a measured forward operator obtained in a time-consuming calibration process. The model commonly used to illustrate the imaging methodology and obtain first model-based reconstructions relies on substantial model simplifications. By neglecting particle–particle interactions, the forward operator can be expressed by a Fredholm integral operator of the first kind when describing the inverse problem. Here, we review previously proposed models derived from single-particle behavior in the MPI context and consider future research on linear and nonlinear problems beyond concentration reconstruction applications. This survey complements a recent topical review on MPI (Knopp et al 2017 Phys. Med. Biol. 62 R124).

Keywords: magnetic particle imaging, model review, particle dynamics

(Some figures may appear in colour only in the online journal)

1. Introduction

Magnetic particle imaging (MPI) is a relatively new imaging modality [15] that relies on the behavior of superparamagnetic iron oxide nanoparticles. The main goal of MPI is to reconstruct the spatially dependent concentration of particles. Measurements are obtained from multiple receive coils in which a voltage is induced by the particle’s nonlinear response to the applied dynamic magnetic field. These voltage measurements are then used for image reconstruction. A high temporal resolution and a potentially high spatial resolution make MPI suitable for several in vivo applications without any associated harmful radiation.
These applications also benefit from the short data acquisition times of MPI to enable fast imaging.

The number of potential medical MPI applications is continuously increasing, including those already suggested upon its introduction, such as vascular imaging and medical instrument tracking. The potential for imaging blood flow was demonstrated first in in vivo experiments using a healthy mouse [70]. The usability of a circulating tracer for long-term monitoring was recently investigated [24]. The high temporal resolution of MPI is advantageous for tracking medical instruments [21]. Recently, MPI was also shown to be suitable for tracking and guiding instruments for angioplasty [57]. Further promising applications of MPI include cancer detection [75] and cancer treatment by hyperthermia [48].

The relationship between particle concentration and measured voltage is modeled by a Fredholm integral equation of the first kind, based on the suppression of particle–particle interactions by a nonmagnetic coating. As such, determining the concentration is a linear inverse problem, in which the integral kernel is defined by the physical properties of the receive coils, the applied magnetic field, and the magnetization dynamics of the tracer material. MPI imaging methodologies differ in the applied magnetic field, which is generated by rapidly moving a field free point (FFP) [15] or a field free line (FFL) [67] along a given trajectory. The most well-known FFP trajectories in practice are the Lissajous and Cartesian trajectories, but other trajectories have also been investigated in simulation studies [29]. Cartesian sequences [8, 38, 76] enable modeling of the MPI signal by spatial convolution. In this case, deconvolution methods are commonly used to obtain image reconstructions, also known as x-space reconstruction [16, 17]. In contrast, when using a Lissajous trajectory, measurements can be obtained more quickly but the corresponding system matrix follows a complex structure. In the absence of suitable models for the tracer behavior in an applied field, the system matrix is usually measured [20, 37], whereby a ‘delta’ probe is moved through the entire field of view in a time-consuming measurement process. Unfortunately, the system matrix cannot be generalized to different scanner setups and tracer materials, so calibration must be performed whenever a new configuration is considered. Besides the long measurement times associated with calibration, this approach also has high memory requirements, particularly for three-dimensional imaging.

The problem of modeling MPI, with respect to finding the correct integral kernel/system function, remains an unsolved problem. Existing model-based reconstruction techniques incorporate particle behavior based on the theory of paramagnetism [16, 28, 31, 37, 46]. Methods based on ideal magnetic fields [16, 51] and on realistic magnetic fields [31] are promising but have not yet reached the necessary quality of measured system functions. One possible reason for this is their nonlinear dependence on the concentration which has been reported for large concentrations [42]. The range of concentrations over which nonlinear dependence emerges can be several times larger than typical experimental concentrations. As a result, this aspect has been neglected in MPI models thus far. When using measured system matrices, this issue might be relevant due to fact that the concentrations commonly chosen in the calibration process are as large as possible to guarantee optimal signal quality.

Another important aspect in modeling MPI that has aroused increasing interest is the magnetization dynamic of large numbers of particles, which is likely to become relevant to rapidly changing applied magnetic fields. To address this issue, simplified models given by ordinary differential equations have been integrated with superparamagnetic particle behavior [8], whereby the authors reported a fitted relaxation time constant of 2.9 $\mu$s for Resovist, a tracer often used in MPI, in an applied field of 22.9 kHz, which, when neglected, result in blurred reconstructions. The dynamic behavior of these particles is affected by Brownian and Néel mechanisms, which were initially suggested by Weizenecker et al [68] in the context of
MPI, as formulated in coupled stochastic differential equations. The first solutions using the Fokker–Planck equation for Brownian rotation were presented by Yoshida and Enpuku [71]; see also [53, 56, 69, 72, 74] for further reading. The behavior of Resovist was found to be mainly determined by Néel rotation, whereas Brownian rotation may influence the behavior in Resovist suspension, depending on the frequency of the applied field [43]. The Néel rotation mechanism is further influenced by the orientation of the particle’s easy axis, which significantly affects the magnetization behavior of a Resovist suspension [73]. The ongoing insufficiency of models for imaging, which neglect these particle dynamics, has motivated an increasing number of studies focusing on MPI excitation patterns in the context of Brownian and Néel rotations [9, 12, 19, 44, 58, 69]. An experimental validation of the Brownian rotation model using low frequencies in one-dimensional sinusoidal excitation patterns [44] confirmed its relevance. Furthermore, magnetization dynamics can substantially differ such that a distinction between different kinds of tracers is possible [50]. The use of these particle models is of particular interest, but they have not been applied extensively to the imaging problem. As yet, there have been only a limited number of works considering simplified models of the magnetic moment dynamics of individual particles in the concentration reconstruction [8]. The physical dynamics of single particle have not been included thus far in multidimensional MPI image reconstructions.

In MPI, reconstructing the concentration is a linear ill-posed inverse problem [30, 46], which is typically solved by applying Tikhonov regularization [36, 40, 52, 70]. In the literature, this problem is preferably solved by using the algebraic reconstruction technique [23, 35, 36, 70] combined with a nonnegativity constraint [70]. Tikhonov regularization has been applied to both kinds of system functions full-calibration-based (measured system matrix) and model-based. Recently, regularization techniques such as fused lasso regularization or other gradient-based methods have been applied to the full-calibration-based MPI problem [38, 61]. Directional total variation, which incorporates a priori information from another modality (e.g. MRI), has also been applied to the MPI problem in a first simulation study [1]. An investigation of the current state-of-the-art model with respect to operator uncertainty was conducted using the total-least-squares approach combined with standard Tikhonov regularization as well as a sparsity-promoting penalty term [28]. As yet, there have been only a small number of rigorous mathematical analyses of the parameter identification problems [22] in MPI. A first step toward a theoretical understanding of this problem is found in [46], in which the authors consider a similar problem by excluding the temporal nature of the MPI problem based on the simplified particle model using the Langevin function. Recently, the multidimensional imaging problem for different dynamic magnetic field patterns was analyzed in the context of inverse problems regarding their degree of ill-posedness [27].

However, due to the open modeling challenge, the use of mathematical models for MPI imaging remains limited but it is highly desirable from a number of perspectives. Potential applications stand to benefit from the high temporal resolution of MPI measurements, which require computationally efficient reconstruction methods to enable real-time imaging. For three-dimensional imaging, in particular, the development of fast model adapted algorithms is advantageous. The time- and memory requirements associated with system calibration may be reduced by the use of a sufficient model.

In this article, we review proposed MPI modeling approaches, which are based on physical models, to establish a basis for further investigations. This overview is complemented by numerical illustrations of model behaviors. In section 2, we formulate the general problem of MPI, considering the transition from the microscopic to the macroscopic scale as well as the measurement process. In section 3, we summarize various different models from a comprehensive mathematical perspective with respect to monodisperse and polydisperse tracers. We
consider particle behavior in equilibrium, as well as Brownian and Néel rotations. Additional information about these models and their relationships are provided in appendices A–C. To illustrate qualitative differences between the models, we present numerical simulations in section 4. We conclude with a discussion in section 5.

2. Preliminaries on magnetic particle imaging

To enable the formulation of the general problem in MPI, we first consider the interplay between magnetization and concentration on different scales. We then provide the technical definitions of the measurement process required for the formulation of the models. For a more detailed introduction to the principles of MPI the interested reader is referred to [32]. A recent review of MPI tracer development can be found in [2] and for a detailed chronological overview on the technical developments in MPI during the last decade we refer the reader to [34].

2.1. Definitions and notations

The inherent nature of the MPI problem is three-dimensional, which is why the relevant vector-valued functions remain three-dimensional even if the domain of the spatial variable is a subset of a $d$-dimensional affine subspace $E_d \subset \mathbb{R}^3$. Here, $E_d$ represents arbitrary lines ($d = 1$), planes ($d = 2$), or the whole $\mathbb{R}^3$ ($d = 3$). Let $\Omega \subset E_d$, $d = 1, 2, 3$, be a bounded domain in $E_d$, and let $S := S^2 = \{ x \in \mathbb{R}^3 | |x| = 1 \}$ be the unit sphere where $| \cdot |$ denotes the Euclidean norm of vectors. Gradient and divergence defined on the surface of $S$ are denoted by $\nabla_S$ and $\text{div}_S$. Furthermore, let $T > 0$ denote the maximal data acquisition time and $I := (0, T)$ the time interval during which the measurement process takes place. We denote the temporal partial derivative of a function $\vec{g} : I \rightarrow \mathbb{R}^3$, $k \in \mathbb{N}$, by $\dot{\vec{g}}$, i.e. $\dot{\vec{g}} = \frac{\partial}{\partial t} \vec{g}$. We label vector-valued functions with a superscript arrow, e.g. $\vec{g}$, and matrix-valued functions appear in bold type, e.g. $P : I \rightarrow \mathbb{R}^{3 \times 3}$. Note that vector-valued arguments of functions are not labeled with a superscript arrow.

2.2. MPI problem description

MPI modeling involves combinations of mathematical models at different scales. The magnetization dynamics of single particles with a ferromagnetic/ferrimagnetic core determine the behavior of an assembly of multiple particles in an immobilized ensemble or a ferrofluid. The behavior of a large number of particles in an external magnetic field then determines the characteristic of the measured signal in MPI, which is exploited to obtain a spatial image of the distribution of the tracer in the desired field of view; see figure 1 for a sketch of the scales. The main goal in MPI is to solve the problem on the macroscopic scale to obtain the spatially dependent particle concentration. To obtain suitable models for MPI, all scales must be taken into account. In total, we can identify three scales that are relevant in MPI, following the general level structure for ferromagnetic modeling presented in [39].

- **Microscopic scale (micromagnetic level; nanometer range):** on the microscopic scale, the behavior of single particles is described by micromagnetics [3]. In micromagnetics the quantum-physical description of matter is neglected. Ignoring the precise atomic structure, the particle’s core is described using continuum physics. The magnetization of the core is represented by a function with a continuous domain, e.g. as shown in figure 1(a), which approximates the behavior of a finite ensemble of magnetic spin moments at the atomic level. In the MPI context, typical core diameters range from 1–100 nm [32].
• **Mesoscopic scale (particle ensemble level):** on the mesoscopic scale, an ensemble of particles is considered. The transition from the microscopic to the mesoscopic scale is achieved by representing each individual particle by its magnetic moment, as shown in figure 1(b). This yields an ensemble of a finite number of magnetic moments in a reference volume. If the reference volume contains a sufficiently large number of particles, an approximation based on the mean magnetic moment of all the particles is desirable. When determining an approximate range of the particle ensemble level, typical concentrations during the calibration process in MPI are on the order $0.5 \text{ mol} \text{l}^{-1}$ with approximately $3 \times 10^8$ particles in $1 \mu\text{m}^3$.

• **Macroscopic scale (imaging level; millimeter to centimeter range):** on the macroscopic scale, the main goal is to obtain a spatial map of the concentration of the nanoparticles; see figure 1(c). The composition of the ensemble of particles can comprise nanoparticles with different properties, such as size or shape. A continuum approximation of the magnetization is used to describe the behavior of the tracer in the entire field of view that approximates the behavior of finite ensembles of the magnetic moments of the particles. On the imaging level, the spatial structure of the applied magnetic field is highly relevant to the spatial encoding of the tracer distribution in the measured temporal signal. A typical measurement device has a bore size with a diameter of approximately 12 cm. Figure 2 shows a scanner and the spatial structure of the applied field. The resolution is determined based on the interplay of the properties of the measurement device and the characteristics of the tracer material. In most studies, authors have reported resolutions of $\sim 1–5 \text{ mm}$, and resolutions in the sub-millimeter range remain an open challenge [34].

The main goal of modeling MPI is to find an appropriate model at the macroscopic scale. Approximate models might be derived by a transition from the micromagnetic level to the particle ensemble level. One possible general approach is as follows:

Let $\omega \subset \mathbb{R}^3$ be a bounded domain describing a single particle’s ferromagnetic core on the micromagnetic level (microscopic scale). Then, we can obtain the magnetization $\vec{M} : \omega \times (0, T) \to M_S S$, $M_S > 0$ being saturation magnetization, of the individual particles by the tools provided by the field of micromagnetics. The dynamic of the magnetization $\vec{M}$ is then given by the Landau–Lifshitz–Gilbert equation [14, 41], as follows:
\[ \frac{\partial}{\partial t} \vec{M} = \frac{\gamma}{1 + \alpha} \left( \vec{M} \times \vec{B}_{\text{eff}} + \frac{\alpha}{M_S} (\vec{M} \times \vec{B}_{\text{eff}}) \times \vec{M} \right), \]

(1)

\( \gamma, \alpha > 0 \), where the effective magnetic field \( \vec{B}_{\text{eff}} = -\nabla ME \) is given by the variational derivative of an energy functional \( E : \{\omega \rightarrow M_S S\} \rightarrow \mathbb{R} \), which comprises different energies like exchange, anisotropy, demagnetization, and Zeeman energy; we refer the reader to [39] for further details on micromagnetic modeling.

On the particle ensemble level (mesoscopic scale) a finite number of particles in a reference volume \( \Gamma \subset \mathbb{R}^3 \) is considered. For example, each of the \( N \in \mathbb{N} \) particles has a certain shape \( \omega_j \subset \mathbb{R}^3 \), with its centroid being at the origin. These particles are then located at positions \( (z_j)_{j=1}^N \subset \Gamma \). In general, all parameters that can substantially differ in individual particles are summarized in a sequence of tuples \( (P_j)_{j=1}^N \subset W^N \) for an entire ensemble where \( W \) defines the parameter space, e.g. for various locations and shapes we have \( W = \Gamma \times \{\omega_j\}_{j=1}^{N_{\omega}} \). The product space of parameters can usually be equipped with additional constraints such as, for example, pairwise disjoint domains \( \omega_{P_j} := z_j + \omega_{(j)} \), \( j = 1, \ldots, N \), of all the particles in an ensemble \( (i : \{1, \ldots, N\} \rightarrow \{1, \ldots, N_{\omega}\} \) being the particle-to-shape mapping). Given the magnetization \( \vec{M}_{(P_j)}_{j=1}^N : \bigcup_{j=1}^N \omega_{P_j} \times (0, T) \rightarrow M_S S \) for the ensemble of particles, the particles’ resulting magnetic moments \( \vec{m}_{P_j} : (0, T) \rightarrow \mathbb{R}^3 \) are as follows:

\[ \vec{m}_{P_j}(t) = \int_{\omega_{P_j}} \vec{M}_{(P_j)}_{j=1}^N (r, t) \, dr \]

(2)

for \( j = 1, \ldots, N \). By introducing the mean magnetic moment \( \bar{m}_{(P_j)}_{j=1}^N := \frac{1}{N} \sum_{j=1}^N \vec{m}_{P_j} \), we can exploit the following relation to estimate the number of particles

\[ \int_{\Gamma} \vec{M}(r, t) \, dr = \sum_{j=1}^N \vec{m}_{P_j}(t) = N \bar{m}_{(P_j)}_{j=1}^N (t) \]

(3)

where \( \bar{M} : \Gamma \times (0, T) \rightarrow \mathbb{R}^3 \) with \( \bar{M} = \vec{M}_{(P_j)}_{j=1}^N \) on \( \bigcup_{j=1}^N \omega_{P_j} \times (0, T) \) and zero elsewhere. The number of particles in an ensemble is expected to be large, so it is desirable to consider an
approximation instead of finding a set of parameter tuples that allows for the determination of the number.

Let \( \bar{m}_T : \{z_0\} \times (0, T) \rightarrow \mathbb{R}^3 \) be the expectation value over all possible ensemble compositions in terms of the tuples \((P^N_j)_{j=1}^N\), which reads as follows:

\[
m_T(z_0, t) = \sum_{N \in \mathbb{N}} p(N) \int_{\mathcal{W}^N} \rho_N((P^N_j)_{j=1}^N) \bar{m}_T(P^N_j) \, d(P^N_j),
\]

where \( \rho_N : \mathcal{W}^N \rightarrow \mathbb{R}^+ \cup \{0\} \) is a probability density function of the possible ensembles with length \( N \), \( p : \mathbb{N} \rightarrow [0, 1] \) is the probability of a number of particles contained in \( \Gamma \), and \( z_0 \in \mathbb{R}^3 \) is the centroid of \( \Gamma \). Thus, we obtain a spatial dependence on the position of the reference volume \( \Gamma \). Note that not all parameters in \( \mathcal{W} \) must be continuous variables that result in a combination of discrete sums and integrals in (4). The crucial step toward the macroscopic representation is the following approximation:

\[
N\bar{m}_T(P^N_j) \rightarrow \bar{N}m_T(z_0, t)
\]

for \( t \in (0, T) \), where \( \bar{N} \) indirectly determines the number of particles, since its semantic meaning strongly depends on the parameter space \( \mathcal{W} \) and the properties of \( \rho_N \).

At the imaging level (macroscopic scale) we consider a field of view given by the bounded domain \( \Omega \subset \mathbb{R}^3 \). Further, let \( Q \subset \mathbb{R}^3 \) be a volume with centroid as the origin. To obtain a concentration function \( c : \Omega \rightarrow \mathbb{R}^+ \cup \{0\} \) with \( \bar{N}(z) = \int_{\Omega} c(x) \, dx \), we can obtain the continuous formulation of the macroscopic problem using a moving average with respect to the volume \( Q \). The utility function \( \bar{f}_Q : \mathbb{R}^3 \times (0, T) \rightarrow \mathbb{R}^3 \) with

\[
\bar{f}_Q(z, t) = \int_{\Omega} \bar{M}(x, t) \chi_Q(z - x) \, dx
\]

defines a macroscopic moment of an ensemble of particles in \( Q \). Using the approximation on the particle ensemble in (5), we obtain the following:

\[
\bar{f}_Q(z, t) / |Q| \approx \bar{N}(z) / |Q| \bar{m}_T(z, t) = \int_{\Omega} c(x) \chi_Q(z - x) / |Q| \, dx \, \bar{m}_T(z, t).
\]

Choosing \( Q \) such that \( \chi_Q / |Q| \) is a Dirac sequence \( (Q) \rightarrow 0 \) yields \( \lim_{|Q| \rightarrow 0} \bar{f}_Q(z, t) / |Q| = \bar{M}(z, t) \) on the left-hand side and \( \lim_{|Q| \rightarrow 0} \bar{N}(z) / |Q| = c(z) \) on the right-hand side. This enables a continuous approximation \( \bar{M}(x, t) \approx c(x) \bar{m}_T(x, t) \) on the macroscopic scale.

In the remainder of this article, we consider the following special case motivated by experimental findings in the MPI context.

### 2.2.1 Transition in the MPI context

The following assumptions are motivated by experimental findings for MPI tracer. It was found that the main contribution to the MPI signal is expected to be caused by particles with diameters distributed around 26 nm potentially due to built aggregates of nanoparticles in Resovist suspension [11]. Furthermore, micromagnetic simulations, which take into account a single domain structure and the particle anisotropy, were used to predict the magnetization behavior of Resovist in sinusoidal excitation patterns [69, 74].

The authors also report an effective diameter larger than 25 nm [69] and a diameter distribution ranging from 15–40 nm [74]. Thus, the first assumption is that the particle diameter is in a range such that each individual particle has a single-domain structure on the microscopic level (see [47] for a brief introduction to size-dependent coercivity for nanoparticles). We then can represent the magnetic moment vectors on the mesoscopic scale as follows:
\[ \bar{m}_p(t) = V_C \bar{M}_{(P_i)}(z_j, t), \]  
where \( V_C \) is the volume of the \( j \)th particle’s core. As such, the dynamics from the micromagnetic level apply to the magnetic moment of the particle. We further assume that particle–particle interactions can be neglected and that the applied field is homogeneous in a reference volume \( \bar{z} + \Gamma \) (where the centroid of \( \Gamma \) is the origin). As a result the magnetization \( \bar{M}_p : \bar{z} + \omega_{(j)} \rightarrow M_S S \) of each particle can be determined independent of the ensemble and position.

\[ \bar{m}_p(t) = V_C \bar{M}_p(z, t) \]

for all \( j = 1, \ldots, N \). The parameters in \( \mathcal{V} \) are thus independent of the location of the particle within the reference volume. Let the remaining parameters of individual particles in an ensemble be i.i.d., which yields the following:

\[ N \bar{m}_{(P_i)}(t) \approx N \bar{m}_V(z, t) \]

as \( \lim_{N \to \infty} \bar{m}_{(P_i)} = \bar{m}_V(z, t) \). Subsequent considerations exploit the macroscopic scale approximation \( \bar{M}(x, t) \approx c(x) \bar{m}_V(x, t) \). The potential choice of \( \Gamma \) is always a tradeoff with respect to size. It must be sufficiently small for the spatial homogeneity assumption and sufficiently large for the particle mean approximation. Due to the large differences in magnitudes between the microscopic and macroscopic scales, this is not an issue in MPI. So, we omit \( \Gamma \) from the subsequent notation, and write \( \bar{m} \) instead of \( \bar{m}_V \).

### 2.3. Measurement process

In the following we define the complete problem, taking into account the measurement process. Then, we describe how the lower dimensional case is constructed. The signal \( v_k : I \rightarrow \mathbb{R} \), \( k = 1, \ldots, L \), which is obtained from the \( L \in \mathbb{N} \) receive coils, is given by the following:

\[ v_k(t) = -\int_I \int_{\Omega} \tilde{a}_k(t-\tau) c(x) \mu_0 \tilde{p}_k(x) \frac{\partial}{\partial \tau} \bar{m}(x, t) \, dx \, d\tau \]

\[ -\int_{\mathbb{R}^3} \tilde{a}_k(x) \frac{\partial}{\partial \tau} \tilde{B}_{app}(x, t) \, dx \, d\tau \]

where \( c : \Omega \rightarrow \mathbb{R}^+ \cup \{0\} \) is the concentration of the magnetic nanoparticles and \( s_k : \Omega \times I \rightarrow \mathbb{R} \), \( k = 1, \ldots, L \), denote the system functions that characterize the behavior of the nanoparticles. \( a_k : I \rightarrow \mathbb{R} \), \( k = 1, \ldots, L \), are the periodic kernel functions of the analog filter in the signal acquisition chain and \( \tilde{a}_k \) denotes its periodic continuation. \( \tilde{p}_k : \mathbb{R}^3 \rightarrow \mathbb{R}^3 \), \( k = 1, \ldots, L \), denotes the vector field which characterizes the sensitivity profile of the receive coils. The first integral in (11) is the analogously filtered voltage induced by the magnetization. We can obtain the dependence on the concentration from the macroscopic approximation \( \bar{M}(x, t) \approx c(x) \bar{m}(x, t) \). The second integral in (11) is the analogously filtered direct feedthrough of the field-generating coils. Since this value is several orders of magnitude larger than that of the particle signal, we must remove the feedthrough prior to digitization. Thus, in the remainder of this article we assume that the magnetic flux density \( \tilde{B}_{app} : \mathbb{R}^3 \times I \rightarrow \mathbb{R}^3 \) of the applied magnetic field and the kernel functions \( a_k \), \( k = 1, \ldots, L \), are chosen in a way such that \( v_{E,k} = 0 \) holds for all excitation signals \( v_{E,k} : I \rightarrow \mathbb{R} \), \( k = 1, \ldots, L \), as defined in (11).
Remark 2.1. Common choices for the analog filters $a_k$, $k = 1, \ldots, L$, are band-stop filters adapted to the frequencies of the sinusoidal excitations used in the subsequently described drive field. The assumption regarding the excitation signals $\nu_{E,k}$, $k = 1, \ldots, L$, is commonly made when the structure of the system functions is studied but not fulfilled in general MPI applications [63]. Efforts are made to remove the excitation signal or reduce its influence on the concentration reconstruction [28, 36, 63, 65].

The applied magnetic fields used in MPI can be characterized by a spatially dependent magnetic field $\vec{g} : \mathbb{R}^3 \rightarrow \mathbb{R}^3$ and a time-dependent homogeneous magnetic field $\vec{h} : I \rightarrow \mathbb{R}^3$. The applied magnetic field is then given by their superposition, i.e. $\vec{B}_{\text{app}}(x, t) = \vec{g}(x) + \vec{h}(t)$. The field $\vec{g}$, also known as the selection field, guarantees that a field-free region is generated; see figure 2(b). Ideally, $\vec{g}$ is assumed to be linear such that it can be represented by a matrix $G \in \mathbb{R}^{3 \times 3}$. Here we distinguish between two cases for the methodology, namely whether an FFP is generated ($\text{rank}(G) = 3$) or an FFL is used ($\text{rank}(G) = 2$). The field $\vec{h}$, also known as the drive field (see figure 2(b)), then moves the field-free region along a certain trajectory. The spatial homogeneity of the drive field is given in sufficiently small centered fields of view in the scanner.

Remark 2.2. In the literature it was also proposed in the FFL approach that the selection field be rotated over time such that the FFL is rotated. The authors also show a relation between the Radon transform and the FFL approach combined with the particle model based on the Langevin function (subsequently termed the equilibrium model). The selection field is then given by $\vec{g} : \mathbb{R}^3 \times I \rightarrow \mathbb{R}^3$ with $\vec{g}(x, t) = \mathbf{P}(t)^T G \mathbf{P}(t) x$ where $\mathbf{P} : I \rightarrow \mathbb{R}^{3 \times 3}$ is a rotation matrix for all $t \in I$.

Remark 2.3. In typical MPI applications, the drive field is defined by weighted sine or cosine functions with different frequencies in each component of $\vec{h}$. Other kinds of excitation signals have also been investigated via simulations [29].

We now can formulate the most general version of the MPI problem, which is to find the concentration $c$ that fulfills the following:

\[
\begin{align*}
\nu_k(t) & = - \int_\Omega \int_{\partial \Omega} c(x) \partial_x (t-t') s_k(x, t') \, \mathrm{d}x \, \mathrm{d}t' \\
\partial_s \delta c & = \mu_0 \vec{B}_0 \cdot \vec{m}
\end{align*}
\]  

(12) for $k = 1, \ldots, L$ and where $\vec{m} : \Omega \times I \rightarrow \mathbb{R}^3$ is the mean magnetic moment vector of the nanoparticles. The first equation in (12) models the analog filter process by a convolution with respect to $t'$. The spatial integration describes the induction of a voltage in the receive coil and is obtained via Faraday’s law of induction combined with the law of reciprocity to obtain the sensitivity profile of the coils [32]. The second equation comprises the sensitivity of the receive coil and the particle behavior in the applied magnetic field.

The $d$-dimensional case for $d < 3$ is constructed by assuming that the concentration is a $\delta$-distribution with respect to the orthogonal complement of the affine subspace $E_d$. Then the $d$-dimensional problem is constructed by assuming $c(x) = \bar{c}(x_1) \delta(x_2)$ where $x = x_1 + x_2$ with $x_1 \in E_d$, $x_2 \in E_d^\perp$, and $\bar{c} : \Omega \subset E_d \rightarrow \mathbb{R}^+ \cup \{0\}$. The affine linear parameterization of $\Omega \subset E_d$, i.e. the parameterization of shifted lines ($d = 1$) or planes ($d = 2$), then allows for the reformulation of the spatial integral in (12) as an integral over an integration domain $\Omega_d \subset \mathbb{R}^d$. Given a parameterization $\Psi : \Omega_d \rightarrow \Omega$ we can consider the problem with respect to $c_d : \Omega_d \rightarrow \mathbb{R}^+ \cup \{0\}$, $c_d(x) = \bar{c}(\Psi(x))$, and $\Omega_d = \Psi^{-1}(\Omega)$. All other spatially dependent functions are then treated analogously.

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Remark 2.4. The linear dependence on the concentration $c$ is based on the assumption that the particle–particle interactions can be neglected. There is increasing evidence that the demagnetization effects relying on these interactions can significantly influence the particle signal [42]. We do not consider the nonlinear dependence in this article, which remains to be explored in future work.

3. Models

Using the general problem description, we derive MPI models for different particle behaviors and tracer compositions. Subsequent considerations are based on the assumptions formulated in the special case in section 2.2. We assume single-domain particles, which is reasonable for ferromagnetic cores with sufficiently small diameters [7]. In this case, each particle has uniform magnetization for any applied magnetic field and is represented by its magnetic moment vector. In general, single-domain particles are not isotropic due to their shape, internal stress, and the internal structure [7]. In the following, we use the Stoner–Wohlfarth model [60] to reflect an uniaxial anisotropy with respect to an easy axis $n \in S$.

3.1. Monodisperse tracer

Monodisperse tracers are composed of a single type of nanoparticle only, all of which exhibit the same characteristic behavior. Here, we take into account two possible dynamical mechanisms, as reported in [53, 56, 68]. The particle can change the orientation of its magnetic moment vector by a rotation of the whole particle, which is known as Brownian rotation [6], and by an internal rotation, which is known as Néel rotation [4, 49]. First, however, we formulate a simplified model based on the Langevin function, termed the equilibrium model which is commonly used to illustrate this methodology. Then, we specify the general MPI problem with respect to the different dynamics.

3.1.1. Equilibrium model. One of the most extensively studied models in MPI is based on the Langevin function. As yet, it is the only model that has been studied with respect to the imaging problem. This model is motivated by the assumptions that the applied magnetic field is static and the particles are in equilibrium. The latter assumption also motivate the term equilibrium model which is used in the following. Under these assumptions, we assume that the mean magnetic moment vector of the nanoparticles immediately follows the magnetic field, i.e.:

$$\vec{m}(x, t) = m_0 L_\beta(|\vec{B}_{app}(x, t)|) \frac{\vec{B}_{app}(x, t)}{|\vec{B}_{app}(x, t)|}$$

(13)

where $L_\beta : \mathbb{R} \to \mathbb{R}$ is given in terms of the Langevin function by the following:

$$L_\beta(z) = \left(\coth(\beta z) - \frac{1}{\beta z}\right)$$

(14)

for $m_0, \beta > 0$. The Langevin function determines the length of the mean magnetic moment vector in equilibrium and can be derived from the Brownian and Néel rotations model, below, by assuming a static magnetic field. A more detailed description of the model’s relationship to Brownian rotation can be found in appendix B. The final problem with respect to the Langevin function is to obtain the concentration $c$ from the following system of equations:
Remark 3.1. \( m_0 \) and \( \beta \) are determined by the saturation magnetization \( M_s \) of the core material, the volume of the particle’s core \( V_c \), the temperature \( T_B \), and the Boltzmann constant \( k_B \). i.e. \( m_0 = M_s V_c \) and \( \beta = m_0/(k_B T_B) \). Assuming spherical particles, the influence of the particle diameter \( D \) is given by \( V_c = 1/6\pi D^3 \). Note that \( \beta \) also depends on \( D \) as it depends on \( m_0 \). For example, particles consisting of magnetite with a typical diameter of 30 nm (20 nm) at room temperature 293 K are characterized by \( \beta \approx 2.1/\mu_0 \times 10^{-3} \) (0.6/\mu_0 × 10^{-3}).

Remark 3.2. Under certain assumptions regarding \( B_{app} \), this problem can be formulated in terms of a spatial convolution, which is evaluated along the trajectory of the FFP with a temporally changing convolution kernel [28, 46]. A first theoretical investigation related to this model can be found in [46]. For further analyses of the problem we refer the reader to [27].

3.1.2. Brownian rotation. Given sufficiently large particle anisotropy, the magnetic moment vector is aligned with the easy axis. Thus, the magnetic moment vector changes its direction due to the rotation of the whole particle [6, 53] in a ferrofluid. By including the Brownian rotation and thermal noise, we obtain a Langevin equation. The dynamics of a particle’s magnetic moment vector \( \vec{m} \) : \( \Omega \times I \rightarrow m_0 S \) with \( |\vec{m}| = m_0 \) is given by the following:

\[
\frac{\partial}{\partial t} \vec{m}(x, t) = \frac{\nu}{m_0} \left( \vec{m}(x, t) \times \vec{B}_{app}(x, t) + \vec{D}(x, t) \right) \times \vec{m}(x, t)
\]

with physical parameters \( \nu, D > 0 \) and where thermal noise is taken into account by the white noise component \( \Gamma_x(t) \) with \( \langle \Gamma_x(t) \rangle = 0 \), \( \langle \Gamma_{x,t}(t_1) \Gamma_{x,t}(t_2) \rangle = \delta(t_1 - t_2) \), for all \( t, t_1, t_2 > 0, x \in \Omega \), and \( i, j = 1, 2, 3 \). \( \langle \cdot \rangle \) denotes the expected value of a random variable. In the case of zero noise the magnetic moment vector is directly damped into the direction of the magnetic field, as shown in figure 3(a). We use the Fokker–Planck equation to derive a differential equation for the probability density function \( f : S \times \Omega \times I \rightarrow \mathbb{R}^+ \cup \{0\} \) to obtain the mean magnetic moment vector. The problem then becomes as follows:

\[
\begin{align*}
\nu_k(t) &= -\int_I \int_{\Omega} c(x) \vec{a}_k(t-t') s_k(x, t') \, dx \, dt' \\
\frac{\partial}{\partial t} s_k(x, t) &= \mu_0 m_0 I_k \left( \frac{\partial}{\partial \vec{m}} \left( B_{app} \right) \right) + \sum_{k=1}^{L} I_k \left( \frac{\partial}{\partial \vec{m}} \left( B_{app} \right) \right)
\end{align*}
\]

for \( k = 1, \ldots, L \) and the identity matrix \( I_3 \in \mathbb{R}^{3×3} \).
\[
\begin{align*}
\alpha_k(t) &= -\int \int \int \sum_{k=1}^{L} \rho_k \frac{\partial}{\partial r^i} \delta c(x(t) \circ k(t) - r) \, dx \, dr' \\
\sum_{k=1}^{L} \rho_k \frac{\partial}{\partial r^i} \left( \mu_0 \frac{\partial m}{\partial \vec{r}} \right) &= 0 \\
\vec{m}(x,t) &= m_0 \int_S \rho f(m,x,t) \, dm \\
\frac{\partial f}{\partial x} &= -\nabla_x \left( \nu \left( \vec{B}_{app} - m \cdot \vec{B}_{app} \right) \right) - f - \frac{1}{\tau_B} \nabla_x f \\
\frac{\partial f}{\partial x} &= 0
\end{align*}
\]

where \( m_0, \nu, \tau_B > 0 \) and where \( f_0 : S \rightarrow \mathbb{R}_+ \cup \{0\} \) with \( \int_S f_0 \, dm = 1 \) is the initial distribution function. Note that divergence and gradients are defined with respect to the surface of the sphere. The first and second equations of (17) describe the general problem as stated in (12). The third equation defines the mean of the magnetic moment vector in terms of the probability density function \( f \). The fourth equation is the differential equation for the probability density function which takes into account the Brownian rotation. The derivation of the Fokker–Planck equation starting from the Langevin equation can be found in appendix A. The fifth equation is the initial condition. The normalization of the initial value together with the properties of the Fokker–Planck equation guarantee normalization of \( f \).

**Remark 3.3.** \( m_0, \nu, \vec{D}, \text{ and } \tau_B \) are determined by the saturation magnetization \( M_S \) of the core material, the volume of the core \( V_C \), the dynamic viscosity \( \eta \), the temperature \( T_B \), and the Boltzmann constant \( k_B \), i.e. \( m_0 = M_S V_C \), \( \nu = \frac{m_0}{k_B \eta} \), \( \tau = \frac{3k_B T_B}{4k_B T_B} \), and \( \frac{D}{|D|} = \sqrt{2|D|k_B T_B} \). \( \tau_B \) is the relaxation time constant. Assuming spherical particles, the particle diameter \( D \) influences \( V_C \) as well as \( V_B \). Thus \( m_0, \nu, \text{ and } \tau_B \) depend on the particle diameter. Table 1 gives an overview of common particle parameters.

**Remark 3.4.** Possible further applications in MPI may involve multi-color MPI [50], which also distinguishes between different kinds of tracers based on their dynamic behavior [64]. In this case the characteristic behavior encoded in multiple system matrices allows for the distinction. In the Brownian rotation model tracer behavior can differ due to changes in viscosity or temperature. Here, a distinction corresponds to the simultaneous reconstruction of \( c \) and a spatially dependent viscosity or temperature. As a result a nonlinear inverse problem must be solved.

### 3.13. Néel rotation

For spatially blocked particles, i.e. fixed \( n \in S \), Brownian rotation is suppressed. Each particle has a magnetic moment vector, which changes its direction due to a change in the internal electric states of the particle’s core \([4, 49]\). Starting at the microscopic scale and including the applied magnetic field, the particle’s anisotropy, and thermal noise yields a Langevin equation based on the Landau–Lifshitz–Gilbert equation for the particle’s magnetic moment vector \( \vec{m} : \Omega \times I \rightarrow m_0 S, |\vec{m}| = m_0 \), as given by the following:

\[
\frac{\partial \vec{m}(x,t)}{\partial t} = \vec{\gamma} \left( (\vec{B}_{eff}(\vec{m}(x,t)/m_0, x, t) + \vec{D} \vec{I}(t)) \times \vec{m}(x,t) \right) + \frac{\alpha}{m_0} \left( \vec{m}(x,t) \times (\vec{B}_{eff}(\vec{m}(x,t)/m_0, x, t) + \vec{D} \vec{I}(t)) \right) \times \vec{m}(x,t)
\]

with the physical parameters \( \vec{\gamma}, \alpha, \vec{D} > 0 \) and where \( \vec{I} \) is a white noise component with \( \langle I_x(t) \rangle = 0 \). \( \langle I_x(t_1) \vec{I}_y(t_2) \rangle = \delta_{x,t_1} \delta_{t_2} \), for all \( t, t_1, t_2 > 0, x \in \Omega \) and \( i, j = 1, 2, 3 \). In contrast to Brownian rotation, the magnetic moment vector moves on a precessional trajectory while it is damped in the direction of the magnetic field, as shown in figure 3(b). In contrast
to previous models, we consider an effective magnetic field \( \vec{B}_{\text{eff}} : S \times \Omega \times I \rightarrow \mathbb{R}^3 \), which consists not only of the applied magnetic field. Here, the uniaxial anisotropy of the particles is incorporated with a field \( \vec{B}_{\text{anis}} : S \rightarrow \mathbb{R}^3 \) with \( \vec{B}_{\text{anis}}(m) = 2K_{\text{anis}}V_C(m \cdot \hat{n})\hat{n} \) for a given easy axis \( \hat{n} \in S \) and anisotropy constant \( K_{\text{anis}} \in \mathbb{R} \). We obtain this field from the Stoner–Wohlfarth model for uniaxial anisotropic particles [60, 62]. Here, \( K_{\text{anis}} > 0 \) corresponds to the desired case of particles having an easy axis while \( K_{\text{anis}} < 0 \) describes an easy plane. The effective magnetic field is then given by the superposition of the applied and anisotropy fields, i.e. \( \vec{B}_{\text{eff}}(m, x, t) = \vec{B}_{\text{app}}(x, t) + \vec{B}_{\text{anis}}(m) \). We use the Fokker–Planck equation for the probability density function \( f : S \times \Omega \times I \rightarrow \mathbb{R}^+ \cup \{0\} \) to derive the mean magnetic moment vector function \( \vec{m} \). The problem then becomes as follows:

\[
\begin{align*}
\vec{v}_k(t) &= -\int_I \int_{S \times \Omega} c(x) \vec{a}_k(t, t') s_k(x, t') \, dx' \, dt' & k = 1, \ldots, L \\
\vec{s}_k &= \mu_0 \gamma \frac{\vec{m}_k}{\eta} & k = 1, \ldots, L \\
\vec{m}(x, t) &= n_0 \int_S mf(m, x, t) \, dm & \text{in } \Omega \times I \\
\frac{\partial f}{\partial t} &= -\text{div}_S \left( \vec{B}_{\text{eff}} \times m + \alpha(m \times \vec{B}_{\text{eff}}) \times m \right) f - \frac{1}{2\tau_0} \nabla_s f & \text{in } S \times \Omega \times I \\
f(\cdot, x, 0) &= f_0 & \text{in } \Omega
\end{align*}
\]

(19)
for $m_0$, $\tilde{\gamma}$, $\alpha$, $\tau_N > 0$ and where $f_0 : S \rightarrow \mathbb{R}^+ \cup \{0\}$ with $\int_S f_0 \, dm = 1$ is the initial distribution function. The first and second equations of (19) describe the general problem that was stated in (12). The third equation defines the mean of the magnetic moment vector with respect to the surface of the sphere $S$ together with the properties of the Fokker–Planck equation starting from the corresponding Langevin equation can be found in appendix C. The fifth equation is the initial condition. The normalization of the initial value of the Fokker–Planck equation guarantees normalization of $f$. Note that gradient and divergence operators must be considered with respect to the surface of the sphere $S$.

Remark 3.5. We determine $m_0$, $\tilde{\gamma}$, $Q$, and $\tau_N$ by the saturation magnetization $M_S$ of the core material, the volume of the core $V_c$, the gyromagnetic ratio $\gamma$, the damping parameter $\alpha$, the temperature $T_B$, and the Boltzmann constant $k_B$, i.e. $m_0 = M_S V_c$, $\tilde{\gamma} = \frac{\gamma}{T_B}$, $\tilde{Q} = \sqrt{\frac{2}{\gamma (1 + \alpha) |k_B|}}$, and relaxation time constant $\tau_N = \frac{m_0}{2 \pi |\gamma| k_B}$. Assuming spherical particles, the particle diameter $D$ influences $V_c$. Thus, $m_0$ and $\tau_N$ depend on the particle diameter. Note that $B_{anis}$ does not depend on the diameter as the volume of the core $V_c$ cancels out. Table 1 provides an overview of common particle parameters.

Remark 3.6. Temperature influences the behavior of nanoparticles. In the context of multi-color MPI, it was also suggested to determine the temperature while reconstructing the concentration [59]. The authors motivate the simultaneous reconstruction by real time monitoring in hyperthermia applications. The problem of finding a spatially dependent temperature $T_B$ requires solving a nonlinear problem. The simultaneous usage of different kinds of particles that differ in their physical properties may result in another nonlinear problem similar to the case of multi-color MPI with viscosity mapping. Structural differences might also be found in the relaxation time constants $\tau_N$ or the anisotropy parameters.

3.2. Polydisperse tracer

The investigations of particular polydisperse tracers also used in MPI have led to the introduction of a distribution function of the core diameter [37], which has mainly been motivated by related physical investigations [10, 11, 25, 26]. The tracer material is then modeled by a distribution of particles with different diameters $D > 0$. Assuming that the particle’s diameter distribution is given by the density function $\rho : \mathbb{R}^+ \rightarrow \mathbb{R}^+ \cup \{0\}$ with $\|\rho\|_{L(\mathbb{R}^+)} = 1$, we obtain the extended problems. Below, we present these models to highlight the parameter dependence on the particle’s core diameter.

Remark 3.7. The distribution function of the particle diameters in polydisperse tracers can be approximated by a log-normal distribution [10, 11, 25, 26]. For example, for Resovist a mean $\mu = \ln(13 \times 10^{-9})$ and a standard deviation of $\sigma = 0.37$, a log-normal distribution was reported for the diameter [11], i.e. $\rho(D) = \frac{1}{D_0 \sqrt{2 \pi \sigma^2}} e^{-(\ln(D) - \mu)^2/(2 \sigma)^2}$.

The equilibrium model in (15) can be extended to polydisperse tracers by adapting the function defining the length of the mean magnetic moment vector in (14). The extended problem is then given by the following:
\[
\begin{align*}
\{ v_k(t) &= -\int_I \int_\Omega c(x) \tilde{a}_k(t, t') s_k(x, t') \, dx \, dt' \\
\quad s_k &= \mu_0 B_k^2 \frac{\partial}{\partial t} \left( L_p(\tilde{B}_{app}) \frac{\partial}{\partial \tilde{B}_{app}} \right) 
\end{align*}
\] (20)

where \( L_p : \mathbb{R} \to \mathbb{R} \) is given in terms of the Langevin function by

\[
L_p(z) = \int_{\mathbb{R}^+} \rho(D)m_0(D)\mathcal{L}_\beta(D)(z) \, dD 
\] (21)

with \( m_0, \beta : \mathbb{R}^+ \to \mathbb{R}^+ \) describing the influence of the particle diameter on the volume of the core, respectively the magnetic moment. For further details on the physical parameters and their relationship to the particle diameter, we refer the reader to remark 3.1.

The Brownian rotation model stated in (17) can be extended for polydisperse tracers by extending the domain of the magnetic moment vector’s probability density function \( f : \mathbb{S} \times \Omega \times I \times \mathbb{R}^+ \to \mathbb{R}^+ \cup \{0\} \). The Brownian rotation model then becomes as follows:

\[
\begin{align*}
\{ v_k(t) &= -\int_I \int_\Omega c(x) \tilde{a}_k(t, t') s_k(x, t') \, dx \, dt' \\
\quad s_k &= \mu_0 B_k^2 \frac{\partial}{\partial t} \left( \rho(D)m_0(D) \int_\Omega mf(m, x, t, D) \, dm \, dD \right) \\
\quad \bar{m}(x, t) &= \int_{\mathbb{R}^+} \rho(D)m_0(D) \int_\Omega mf(m, x, t, D) \, dm \, dD \\
\quad \frac{\partial f}{\partial t} &= -\text{div} \left( \nu(D) \left( \tilde{B}_{app} - m(m \cdot \tilde{B}_{app}) \right) f - \frac{1}{\tau_0 m} \nabla f \right) \\
\quad f(\cdot, x, 0, D) &= f_0
\end{align*}
\] (22)

for \( m_0, \nu, \tau_0 : \mathbb{R}^+ \to \mathbb{R}^+ \) and where \( f_0 : \mathbb{S} \to \mathbb{R}^+ \cup \{0\} \) with \( \int_\Omega f_0 \, dm = 1 \). In contrast to the monodisperse model, the third equation of (22) comprises the mean of the magnetic moment vector and the mean of the particle diameter. The differential equation for the probability density function in the fourth and fifth equations are extended to take into account the dependence on the particle diameter \( D \). For further details on the physical parameters and their relation to the particle diameter we refer to remark 3.3.

The Néel rotation model is extended analogously by considering \( f : \mathbb{S} \times \Omega \times I \times \mathbb{R}^+ \to \mathbb{R}^+ \cup \{0\} \). It is thus given by the following:

\[
\begin{align*}
\{ v_k(t) &= -\int_I \int_\Omega c(x) \tilde{a}_k(t, t') s_k(x, t') \, dx \, dt' \\
\quad s_k &= \mu_0 B_k^2 \frac{\partial}{\partial t} \left( \rho(D)m_0(D) \int_\Omega mf(m, x, t, D) \, dm \, dD \right) \\
\quad \bar{m}(x, t) &= \int_{\mathbb{R}^+} \rho(D)m_0(D) \int_\Omega mf(m, x, t, D) \, dm \, dD \\
\quad \frac{\partial f}{\partial t} &= -\text{div} \left( \nu \left( \tilde{B}_{app} - m(m \cdot \tilde{B}_{app}) \right) f - \frac{1}{\tau_0 m} \nabla f \right) \\
\quad f(\cdot, x, 0, D) &= f_0
\end{align*}
\] (23)

for \( m_0, \tau_0 : \mathbb{R}^+ \to \mathbb{R}^+ \), \( \tau_0, \alpha > 0 \) and where \( f_0 : \mathbb{S} \to \mathbb{R}^+ \cup \{0\} \) with \( \int_\Omega f_0 \, dm = 1 \). Remark 3.5 provides further details on the physical parameters and their dependence on the diameter \( D \). The third equation in (23) comprises the mean of the magnetic moment vector and we extend the mean over the particle diameter and the fourth and fifth equations to take into account the dependence on the particle diameter.

**Remark 3.8.** The polydisperse models are equations parameterized by the diameter \( D \) that are similar to the monodisperse models based on the stochastic differential equations.
4. Numerical examples

To demonstrate the behavior of the models, we compute the temporal derivative of the mean magnetic moment vector \( \vec{m} \) in a highly symmetric special case where the probability density function is circular symmetric with respect to the \( e_3 \)-axis. On the \( e_3 \)-axis the third component of the mean magnetic moment vector is nonzero only. The numerical solution is based on the formulation of a system of ordinary differential equations obtained by approximating the probability density functions for the Brownian and Néel rotation by a finite number of Legendre polynomials. Further details about the numerical solution, which follows that in [9], can be found in appendix D. In the subsequent simulations, we used the first 50 Legendre polynomials and we assumed the initial distribution \( f_0 \) to be uniform.

The simulation setup is as follows. We assume an excitation in the direction of \( e_3 \) only and consider the case that \( \Omega \subset E_d = \{ qe_3 | q \in \mathbb{R} \} \). The drive field is given by \( \vec{h}(t) = A \sin(2\pi ft)e_3 \) where \( A > 0 \) is the excitation amplitude and \( f > 0 \) is the excitation frequency. We assume the election field to be linear, as defined by a diagonal matrix \( G \in \mathbb{R}^{3 \times 3} \).

For the simulations we use physical parameters typical in MPI applications. We assumed the nanoparticles to have a spherical magnetite core, and we chose the anisotropy constant \( K_{\text{anis}} \) as the bulk value for magnetite [13]. For non-spherical nanoparticles the effective anisotropy constant can be larger due to surface effects [18]. Table 1 provides an overview of the parameters. We computed the remaining parameters according to remarks 3.1, 3.3 and 3.5.

Figure 4 shows the simulation results of the monodisperse models for a diameter of 20 nm. The graphs of the equilibrium model show point symmetry to the midpoints between two zero crossings of the applied field (on the \( x \)-axis). The extremal points in the equilibrium model are close to the FFP in time. The relaxation time constants in the Brownian and Néel rotation models introduce a time lag between the applied field and the induced signal. The Brownian rotation model shows damped and skewed behavior and the Néel rotation model is shifted in the positive time direction, when compared to the equilibrium model. This shift is related to the particle anisotropy with the preferred \( e_3 \)-direction. A certain strength of the magnetic field is required before the magnetization changes. We can observe more structural differences at the \( z \)-positions next to the origin where one extremal point is more damped than the other. The more it is damped the less able is the applied magnetic field to counteract the anisotropy in the preferred direction of the easy axis.

Figure 5 shows the simulation results for the polydisperse models. The polydisperse equilibrium model is smaller in amplitude but its qualitative behavior remains similar to the monodisperse version with a diameter of 20 nm. The change in amplitude is a direct result of the diameter distribution shift toward particle diameters smaller than 20 nm. The relationship between the polydisperse Brownian rotation model and the equilibrium model is qualitatively similar to the relationship between their monodisperse versions. The polydisperse Néel rotation model changed its behavior toward a smaller amplitude and a shorter time delay.

5. Discussion

Above, we summarized several models relevant to MPI and showed simulation results for a special case to illustrate their characteristic behaviors. These different behaviors are the potential reasons why model-based reconstructions using the equilibrium model are not possible or not of the same quality as reconstructions using a measured linear operator. The dynamic effects are considered to be independent of each other as it is commonly assumed that one cause is dominant. However, combined models that consider simultaneous Brownian and Néel rotations
Figure 4. Simulated mean magnetic moment vector in e3-direction for different z-positions considering the monodisperse models (D = 20 nm) with parameters specified in table 1. Dotted vertical lines (red) highlight the point in time with zero applied magnetic field. T_R is the repetition time for one period of the applied field. The second and third periods are illustrated as no initial value f_0 was chosen to fulfill a periodicity constraint.
Figure 5. Simulated mean magnetic moment vector in $e_1$-direction for different $z$-positions considering the polydisperse models (log-normal particle diameter distribution) with parameters specified in table 1. Dotted vertical lines (red) highlight the point in time with zero applied magnetic field. $T_R$ is the repetition time for one period of the applied field. The second and third periods are illustrated as the initial value $f_0$ was not chosen to fulfill a periodicity constraint.
are desirable and were investigated by using their Langevin equations \[54\]. More recently
the coupled Fokker–Planck equation for both mechanisms was investigated and the system of
ordinary differential equations for the coefficients of spherical harmonics was formulated \[66\].

Imaging quality can suffer when neglecting these dynamics, as they influence the measured
temporal signal. The resulting loss of quality is due to spatial encoding in the time-
dependent signal. A certain delay, such as that in the Brownian and Néel rotations, may cause
spatially shifted reconstructions when taking into account Cartesian trajectories. The damping
and smoothing observed in the Brownian rotation model may result in an underestimated
concentration and a spatially blurred reconstruction. The ill-posed nature of the problem also
allows for only a certain degree of model errors. In the case of multidimensional trajectories,
such as Lissajous trajectories, the rotation of the applied magnetic field vector must be taken
into account. Due to the loss of circular symmetry, the probability density function must be
approximated for the whole surface of the sphere. The higher dimensionality also increases the
computation costs due to the increase in the number of equations in a corresponding system of
ordinary differential equations in the numerical computation. If the series expansion is truncated at \( N \) coefficients, the number of differential equations is of the order \( O(N) \) in the highly
symmetric case as illustrated in section 4. Considering a single dynamic mechanism with a
spherical harmonic approximation results in \( O(N^2) \) equations for an analogous truncation.
In the coupled case of Brownian and Néel rotations \[66\], it is of the order \( O(N^4) \). Therefore,
finding a direct and more efficient solution to compute the mean magnetic moment vector
is highly desirable, as it must be solved for all spatial positions and the whole measurement
time range. Given the numerical solutions for the multidimensional case with both Brownian
and Néel rotations, these models must still be physically validated for applied magnetic fields
in MPI. For this purpose, a recently proposed magnetic particle spectrometer (MPS) \[5\] can
provide the required measurements. The advantage of the proposed MPS is that it allows for
the application of a drive field with a three-dimensional excitation pattern and a constant offset
field simulating the selection field at a fixed position.

This review of mathematical models for MPI builds a basis for several future research
directions. The numerical treatment of all models in the multidimensional case requires fur-
ther analysis and the development of efficient algorithms. A first step in the theoretical direc-
tion was made by formulating a different problem setting based on the equilibrium model
\[46\]. However, by neglecting the temporal dependencies in the methodology, the equilibrium
model defined in this work is not directly covered. But recently, the MPI problem was investi-
gated analytically for different types of trajectories \[27\]. In addition to the linear inverse prob-
lem of reconstructing the concentration, several nonlinear inverse problems are presented by
applications and the particle behavior itself. For example, joint concentration reconstructions
combined with a spatial viscosity or temperature distribution have already been addressed in
the context of multi-color MPI \[50, 59\]. Prior to the consideration of combined problems or
a nonlinear concentration dependence \[42\], a series of analytical works is required regarding
the models presented above.

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Appendix A. Fokker–Planck equation for Brownian rotation

We derive (17) from the Langevin equation of the particle dynamics. As the spatial dependence in the probability density function \( f \) results solely from the spatially dependent applied magnetic field \( \vec{B}_{\text{app}} \), we consider the problem of determining \( f \) for one fixed \( x \in \Omega \), and also omit the spatial variable \( x \). The Langevin equation for Brownian rotation of a single particle with magnetic moment vector \( \vec{m} \) is as follows:

\[
\frac{\partial \vec{m}}{\partial t} = \frac{1}{6V_{\text{H}}\eta} \left( \vec{m} \times \vec{B}_{\text{app}} + \vec{D} \vec{\Gamma} \right) \times \vec{m},
\]

(A.1)

with \( \vec{D} > 0 \) determined below and where \( \vec{\Gamma} \) is a white noise component with \( \langle \vec{\Gamma}(t) \rangle = 0 \), \( \langle \vec{\Gamma}(t_1)\vec{\Gamma}(t_2) \rangle = \delta_{ij}(t_1 - t_2) \), for all \( t, t_1, t_2 > 0 \) and \( i, j = 1, 2, 3 \). \( \delta \) is the Kronecker delta and \( \delta \) is the Dirac delta distribution. Substituting \( \vec{m} = \vec{m}/m_0, m_0 = |\vec{m}| \) yields

\[
\frac{\partial \vec{m}}{\partial t} = \frac{1}{6V_{\text{H}}\eta} \left( m_0 \vec{m} \times \vec{B}_{\text{app}} + \vec{D} \vec{\Gamma} \right) \times \vec{m}
\]

(A.2)

where

\[
\Lambda(m) = \begin{pmatrix}
0 & -m_3 & m_2 \\
m_3 & 0 & -m_1 \\
-m_2 & m_1 & 0
\end{pmatrix}.
\]

(A.3)

The Fokker–Planck equation for the probability density function \( f : S \times (0, T) \to \mathbb{R}^+ \) thus becomes [55] as follows:

\[
\frac{\partial f(m, t)}{\partial t} = -\text{div}_S \left( \vec{a}(m, t)f(m, t) - \frac{1}{2}\vec{B}(m, t) \left( \text{div}_S (\vec{B}^j(m, t)f(m, t)) \right)_{j=1,2,3} \right)
\]

(A.4)

where \( \vec{a} : S \times (0, T) \to \mathbb{R}^3 \) and \( \vec{B} : S \times (0, T) \to \mathbb{R}^{3 \times 3} \) with \( \vec{B}^j \) being the \( j \)th column of \( \vec{B} \). Note that gradient and divergence operators are defined on the surface of the sphere; see, for example, [45] for a definition. From the Langevin equation it follows that:

\[
\vec{a}(m, t) = \frac{m_0}{6V_{\text{H}}\eta} \left( \vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m \right)
\]

(A.5)

and

\[
\vec{B}(m, t) = \frac{\vec{D}}{6V_{\text{H}}\eta} \Lambda(m)^T.
\]

(A.6)

Based on the fact that \( m \cdot \nabla f = 0 \) (as the surface gradient is tangent to the unit sphere), by using \( \Lambda(m)^T \Lambda(m) \nabla f = (m \times \nabla f) \times m = \nabla f \), we obtain the desired Fokker–Planck equation, as follows:

\[
\frac{\partial f(m, t)}{\partial t} = -\frac{1}{6V_{\text{H}}\eta} \text{div}_S \left( m_0 (\vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m)f(m, t) - \frac{\vec{D}^2}{12V_{\text{H}}\eta} \nabla f(m, t) \right).
\]

(A.7)

The diffusion coefficient \( \vec{D} \) is determined by considering the equilibrium case [56], i.e. let \( t_0 \in I \) be the point in time such that \( \frac{\partial}{\partial t} f(m, t_0) = 0 \). We further assume that
\(f_0 : S \rightarrow \mathbb{R}^+ \cup \{0\}\) with \(f_0 = f(\cdot, t_0)\) corresponds to the Boltzmann distribution for Brownian rotation in the equilibrium, i.e. \(f_0(m) = ke^{-\beta E(m, t_0)}\), where \(E\) is the total energy with \(\nabla_g E(\vec{m}, t_0) = -\vec{B}_{\text{app}}(t_0)\) and \(\beta = \frac{1}{k_B T_0}\). \(E(\vec{m}, t) = -\vec{B}_{\text{app}} \cdot \vec{m}\) and the definition of the surface gradient \(\nabla S f_0 = \nabla C f_0 - n(\nabla C f_0 \cdot n)\) (\(\nabla C\) gradient in Cartesian coordinates, \(n\) surface normal vector) yield the following:

\[
0 = \text{div}_S \left( m_0(\vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m) f_0 - \frac{\tilde{D}^2}{12 V h T} \frac{m_0}{k_B T} (\vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m) f_0 \right) = \left( m_0 - \frac{\tilde{D}^2}{12 V h T} \frac{m_0}{k_B T} \right) \text{div}_S \left( (\vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m) f_0 \right). \tag{A.8}
\]

From this it follows \(\tilde{D} = \sqrt{12 V h k_B T} \). Defining \(\tau_B = \frac{3 V h}{k_B T}\) yields

\[
\frac{\partial}{\partial t} f(m, t) = - \frac{1}{2 \tau_B} \text{div}_S \left( \frac{m_0}{k_B T} (\vec{B}_{\text{app}} - (m \cdot \vec{B}_{\text{app}})m) f(m, t) - \nabla S f(m, t) \right). \tag{A.9}
\]

**Appendix B. Derivation of Langevin function**

In the following, we give an example of how the equilibrium model, when using the Langevin function, is related to Brownian rotation. As we can see in appendix A, the Fokker–Planck equation is parametrized such that the probability density function of the magnetic moment vector in equilibrium at time \(t_0 \in I\) has the following form:

\[
f_0(m) = ke^{-\beta E(m, t_0)}. \tag{B.1}
\]

Considering the mean magnetic moment vector yields the following:

\[
\vec{m}(t_0) = m_0 \int_S m f_0(m) \, dm = m_0 k \int_S m e^{\beta m_0 (R^T \vec{m})(R^T \vec{B}_{\text{app}})} \, dm
\]

\[
= m_0 k R \int_S ye^{\beta m_0 b_0} \, dy
\]

\[
= m_0 k R \int_0^\pi \int_0^{2\pi} \sin(\theta) \sin(\phi) \left( \cos(\phi) \right) \left| \sin(\theta) \right| e^{\alpha \beta m_0 b_0} \, d\phi \, d\theta
\]

\[
= 2 \pi m_0 k R e \int_0^\pi \sin(\theta) e^{\alpha \beta m_0 b_0} \, d\theta
\]

\[
= -2 \pi m_0 k R e \int_{-1}^1 xe^{\alpha \beta m_0 b_0} \, dx
\]

\[
= -2 \pi m_0 k R e \left( \frac{1}{\beta m_0 b_0} e^{\alpha \beta m_0 b_0} \right)_{-1}^1 - \int_{-1}^1 \frac{1}{\beta m_0 b_0} e^{\alpha \beta m_0 b_0} \, dx
\]

\[
= -Re m_0 \left( e^{\alpha \beta m_0 b_0} + e^{-\alpha \beta m_0 b_0} - \frac{1}{\beta m_0 b_0} \right),
\]

where \(R \in \mathbb{R}^{3 \times 3}\) is a rotation matrix such that \(R^T \vec{B}_{\text{app}}(t_0) = e_3 b_0\), where \(b_0 = |\vec{B}_{\text{app}}(t_0)|\), and where \(k\) is obtained by similar transformations such that it is given by the following:
The length of the mean magnetic moment is thus given by $|\vec{m}(t_0)| = m_0 \mathcal{L}_{\vec{m}_0}(\vec{B}_{\text{app}}(t_0))$ with $\mathcal{L}_{\vec{m}}$ given by (14).

Appendix C. Fokker–Planck equation for Néel rotation

We derive (19) from the Langevin equation based on the Landau–Lifshitz–Gilbert equation. As the spatial dependence in the probability density function $f$ results solely and implicitly from the spatially dependent applied magnetic field $\vec{B}_{\text{app}}$ we consider the problem of determining $f$ for one fixed $x \in \Omega$, and also omit the spatial variable $x$. We obtain the Langevin equation for the Néel rotation of a single particle with the magnetic moment vector $\vec{m}$ from the Landau–Lifshitz–Gilbert equation, as follows:

$$\frac{\partial}{\partial t} \vec{m} = \tilde{\gamma} \left( (\vec{B}_{\text{eff}} + \vec{D}\vec{\Gamma}) \times \vec{m} + \frac{\alpha}{m_0} (\vec{m} \times (\vec{B}_{\text{eff}} + \vec{D}\vec{\Gamma})) \times \vec{m} \right),$$

with $\vec{D} > 0$ determined below, where $\tilde{\gamma} = \gamma/(1 + \alpha^2)$ and $\vec{\Gamma}$ is a white noise component with $\langle \Gamma_i(t) \rangle = 0$, $\langle \Gamma_i(t_1) \Gamma_j(t_2) \rangle = \delta_{ij}(t_1 - t_2)$, for all $t, t_1, t_2 > 0$ and $i, j = 1, 2, 3$. $\delta_{ij}$ is the Kronecker delta and $\delta$ is the Dirac delta distribution. Substituting $\vec{m} = \tilde{\gamma} \vec{m}/m_0$, $m_0 = |\vec{m}|$ yields the following:

$$\frac{\partial}{\partial t} \vec{m} = \tilde{\gamma} \left( \vec{B}_{\text{eff}} \times \vec{m} + \alpha m_0 \vec{m} \times (\vec{B}_{\text{eff}} + \vec{D}\vec{\Gamma}) \right) \times \vec{m} + \vec{D}(\vec{m})^T + \alpha \vec{m} \times (\vec{B}_{\text{eff}} + \vec{D}\vec{\Gamma}) \right) \times \vec{m}$$

$$\frac{\partial}{\partial t} \vec{m} = \tilde{\gamma} \left( \vec{B}_{\text{eff}} \times \vec{m} + \alpha m_0 \vec{m} \times (\vec{B}_{\text{eff}} + \vec{D}\vec{\Gamma}) \right) \times \vec{m}$$

where

$$\Lambda(m) = \begin{pmatrix} 0 & -m_3 & m_2 \\ m_3 & 0 & -m_1 \\ -m_2 & m_1 & 0 \end{pmatrix}$$

$$\Lambda(m)^T \Lambda(m) = -\begin{pmatrix} m_2^2 + m_3^2 & -m_1 m_2 & -m_1 m_3 \\ -m_1 m_2 & m_1^2 + m_3^2 & -m_2 m_3 \\ -m_1 m_3 & -m_2 m_3 & m_1^2 + m_2^2 \end{pmatrix}. \quad (C.4)$$

The Fokker–Planck equation for the probability density function $f : S \times (0, T) \to \mathbb{R}^+ \cup \{0\}$ thus becomes as follows [55]:

$$\frac{\partial}{\partial t} f(m, t) = -\text{div}_S \left( \vec{a}(m, t) f(m, t) - \frac{1}{2} \mathbf{B}(m, t) \left( \text{div}_S(\vec{B}^T(m, t) f(m, t)) \right) \right),$$

where $\vec{a} : S \times (0, T) \to \mathbb{R}^3$ and $\mathbf{B} : S \times (0, T) \to \mathbb{R}^{3 \times 3}$ with $\vec{B}^T$ being the $i$th column of $\mathbf{B}$. Note that gradient and divergence operators are defined on the surface of the sphere; see, for example, [45] for a definition. From the Langevin equation, it follows that:

$$\vec{a}(m, t) = \tilde{\gamma} \left( \vec{B}_{\text{eff}} \times m + \alpha m \times \vec{B}_{\text{eff}} \right) \times m$$

and
\[ \mathbf{B}(m, t) = \tilde{D} \tilde{\gamma} \left( \mathbf{A}(m)^T + \alpha \mathbf{A}(m)^T \mathbf{A}(m) \right). \]  

(C.7)

Based on the fact that \( m \cdot \nabla_s f = 0 \) (as the gradient is tangent to the unit sphere), by using \( \mathbf{A}(m)^T \mathbf{A}(m) \nabla_s f = (m \times \nabla_s f) \times m = \nabla_s f \), we obtain the desired Focker–Planck equation, as follows:

\[
\frac{\partial}{\partial t} f(m, t) = -\tilde{\gamma} \text{div}_S \left( \left( \tilde{\mathbf{B}}_{\text{eff}} \times m + \alpha (\tilde{\mathbf{B}}_{\text{eff}} - (m \cdot \tilde{\mathbf{B}}_{\text{eff}})m) \right) f(m, t) - \frac{\tilde{D}^2 \tilde{\gamma} (1 + \alpha^2)}{2} \nabla_s f(m, t) \right). 
\]

(C.8)

To determine the diffusion coefficient \( \tilde{D} \), we consider the equilibrium case \([56]\), i.e. let \( t_0 \in \mathbf{I} \) be a point in time such that \( \frac{\partial}{\partial t} f(m, t_0) = 0 \). We further assume that \( f_0 : S \rightarrow \mathbb{R}^+ \cup \{0\} \) with \( f_0(\cdot) = f(\cdot, t_0) \) corresponds to the Boltzmann distribution in the equilibrium, i.e. \( f_0 = k e^{-\beta E(m_m, t_0)} \), where \( E \) is the total energy with \( \nabla_{\tilde{m}} E(\tilde{m}, t_0) = -\tilde{\mathbf{B}}_{\text{eff}}(\tilde{m}/m_0, t_0) \) and \( \beta = \frac{1}{k T_B} \). We use \( \nabla_{\text{C}0} = \beta m_0 \tilde{\mathbf{B}}_{\text{eff}} f_0 \) and the definition of the surface gradient \( \nabla_s f_0 = \nabla_{\text{C}0} - n (\nabla_{\text{C}0} \cdot n) \) \( \nabla_{\text{C}} \) gradient in Cartesian coordinates, \( n \) surface normal vector) to obtain the following:

\[
0 = \text{div}_S \left( (\tilde{\mathbf{B}}_{\text{eff}} \times m + \alpha (\tilde{\mathbf{B}}_{\text{eff}} - (m \cdot \tilde{\mathbf{B}}_{\text{eff}})m) f_0 - \frac{\tilde{D}^2 \tilde{\gamma} (1 + \alpha^2)}{2} \nabla_s f_0 \right) \\
= \text{div}_S \left( (\tilde{\mathbf{B}}_{\text{eff}} \times m) f_0 \right) + \left( \alpha - \frac{\tilde{D}^2 \tilde{\gamma} (1 + \alpha^2) m_0}{2 k_B T_B} \right) \text{div}_S \left( (\tilde{\mathbf{B}}_{\text{eff}} - (m \cdot \tilde{\mathbf{B}}_{\text{eff}})m) f_0 \right) \\
= \left( \alpha - \frac{\tilde{D}^2 \tilde{\gamma} (1 + \alpha^2) m_0}{2 k_B T_B} \right) \text{div}_S \left( \tilde{\mathbf{B}}_{\text{eff}} - (m \cdot \tilde{\mathbf{B}}_{\text{eff}})m \right) f_0
\]

where the last equality holds as \( \text{div}_S \left( \tilde{\mathbf{B}}_{\text{eff}} \times m \right) = 0 \) for \( \tilde{\mathbf{B}}_{\text{eff}} = \tilde{\mathbf{B}}_{\text{app}} + \tilde{\mathbf{B}}_{\text{anis}} \). It follows \( \tilde{D} = \sqrt{2 \frac{\tilde{\gamma} \kappa_B T_B}{\gamma(1 + \alpha^2) m_0}} \). Defining \( \tau_N = \frac{m_0}{\tilde{D} k_B T_B} \) yields

\[
\frac{\partial}{\partial t} f(m, t) = -\text{div}_S \left( \tilde{\gamma} \left( \tilde{\mathbf{B}}_{\text{eff}} \times m + \alpha (\tilde{\mathbf{B}}_{\text{eff}} - (m \cdot \tilde{\mathbf{B}}_{\text{eff}})m) \right) f(m, t) - \frac{1}{2 \tau_N} \nabla_s f(m, t) \right).
\]

(C.10)

**Appendix D. Numerical solution for 1D excitation**

The following numerical solution follows that in [9]. We compute the mean magnetic moment vector \( \tilde{m} \) for an applied magnetic field with the form \( \tilde{B}_{\text{app}}(x, t) = b(x, t) e_3 \) with a given field \( b : \Omega \times \mathbf{I} \rightarrow \mathbb{R} \). We also assume that \( \Omega \subset \{ e_3 \} \). 

Under these assumptions, we can reformulate the Fokker–Planck equations for Brownian rotation in (17) and Néel rotation in (19) in terms of the angle \( \theta \) between \( m \) and \( e_3 \). Let \( x \in \Omega \) be fixed. By assuming \( z = \cos(\theta) \) the resulting ordinary differential equations for \( \tilde{f}_x : [-1, 1] \times \mathbf{I} \rightarrow \mathbb{R}^+ \cup \{0\} \) then become for Brownian rotation, as follows:

\[
2 \tau_B \frac{\partial}{\partial t} \tilde{f}_x(z, t) = \frac{\partial}{\partial z} \left( (1 - z^2) \left( \frac{\partial}{\partial z} \tilde{f}_x(z, t) - \beta b(x, t) \tilde{f}_x(z, t) \right) \right) 
\]

(D.1) 

with physical parameters \( \tau_B = 3 V_B \eta / (k_B T_B) \) and \( \beta = \frac{m_0}{k_B T_B} \). For Néel rotation, we obtain the following:
\[2\tau_n \frac{\partial}{\partial t} \tilde{f}(z,t) = \frac{\partial}{\partial z} \left( (1-z^2) \left( \frac{\partial}{\partial z} \tilde{f}(z,t) - \beta b(x,t) \tilde{f}(z,t) - \tilde{K} \tilde{f}(z,t) \right) \right) \]  

(D.2)

with physical parameters \( \tau_n = \frac{m_n}{2\tau_n^2 \mu_0} \) and \( \tilde{K} = \frac{2\mu_0 m_n}{M_0^2} \). Then, we obtain the mean magnetic moment vector component \( \bar{m}_3 \) by the following:

\[ \bar{m}_3(x,t) = m_0 \int_{-1}^{1} \tilde{f}_z(z,t) \, dz \varepsilon_3. \]  

(D.3)

We expand the probability density function \( \tilde{f} \) in Legendre polynomials \( \{P_n\}_{n \in \mathbb{N}} \) and \( a_n : I \to \mathbb{R}, n \in \mathbb{N} \), as follows:

\[ \tilde{f}(z,t) = \sum_{n=0}^{\infty} a_n(t) P_n(z). \]  

(D.4)

Using this relation and the constraint \( \int_{-1}^{1} \tilde{f}(z,t) \, dz = 1, \, t \in I \), we obtain the following system of differential equations for Brownian rotation

\[ \begin{cases} a_0(t) = \frac{1}{2} \\ \frac{2\tau_n}{m(n+1)} \frac{d}{dt} a_n(t) = -a_n(t) + \beta b(x,t) \left( \frac{a_{n-1}(t)}{2n-1} - \frac{a_{n+1}(t)}{2n+1} \right) \end{cases} \]  

for \( n \geq 1 \)  

(D.5)

and for Néel rotation

\[ \begin{cases} a_0(t) = \frac{1}{2} \\ \frac{2\tau_n}{m(n+1)} \frac{d}{dt} a_n(t) = -a_n(t) + \beta b(x,t) \left( \frac{a_{n-1}(t)}{2n-1} - \frac{a_{n+1}(t)}{2n+1} \right) \\ + \tilde{K} \left( \frac{(n-1)a_{n-3}(t)}{(2n-3)(2n-1)} + \frac{na_n(t)}{(2n-1)(2n+1)} \right) \\ - \frac{(n+1)a_{n+3}(t)}{(2n+1)(2n+3)} + \frac{(n+3)a_{n+1}(t)}{(2n+3)(2n+5)} \right) \right) \]  

for \( n \geq 1 \)  

(D.6)

by assuming \( a_{-1}(t) = 0 \). Assuming a uniform distribution as an initial value results in the initial values \( a_0(0) = 1/2 \) and \( a_n(0) = 0, \, n \geq 1 \), for both systems. Using the relation \( \int_{-1}^{1} \tilde{f}_z(z,t) \, dz = 2/3a_1(t) \), we can determine the mean magnetic moment vector. The time derivative of the mean magnetic moment vector is thus given by the respective differential equation for \( n = 1 \).

In the simulation we use an approximated probability density function for \( N \in \mathbb{N} \) given by the following:

\[ \tilde{f}(z,t) = \sum_{n=0}^{N} a_n(t) P_n(z). \]  

(D.7)

The applied magnetic field is given by the following:

\[ b(x,t) = A \sin(2\pi ft) + G_{3,3}x_3 \]  

(D.8)

where \( A, f > 0 \) and \( G_{3,3} \). Table 1 lists the physical parameters used in the simulation.

**ORCID iDs**

Tobias Kluth ⚫️ https://orcid.org/0000-0003-4814-142X
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