Mapping magnetic signals of individual magnetite grains to their internal magnetic configurations using micromagnetic models

David Cortés-Ortuño¹, Karl Fabian², and Lennart Vincent de Groot¹

¹Utrecht University
²Norwegian University of Science and Technology (NTNU)

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Abstract

Micromagnetic Tomography (MMT) is a technique that combines X-ray micro computed tomography and scanning magnetometry data to obtain information about the magnetic potential of individual grains embedded in a sample. Recovering magnetic signals of individual grains in natural and synthetic samples provides a new pathway to study the remanent magnetization that carries information about the ancient geomagnetic field and is the basis of all paleomagnetic studies. MMT infers the magnetic potential of individual grains by numerical inversion of surface magnetic measurements using spherical harmonic expansions. The magnetic potential of individual particles in principle is uniquely determined by MMT, not only by the dipole approximation, but also more complex, higher order, multipole moments. Here we show that such complex magnetic information together with particle shape and mineralogy severely constrains the internal magnetization structure of an individual grain. To this end we apply a three dimensional micromagnetic model to predict the multipole signal from magnetization states of different local energy minima. We show that for certain grains it is even possible to uniquely infer the magnetic configuration from the inverted magnetic multipole moments. This result is crucial to discriminate single-domain particles from grains in more complex configurations such as multi-domain or vortex states. As a consequence, our investigation proves that by MMT it is feasible to select statistical ensembles of magnetic grains based on their magnetization states, which opens new possibilities to identify and characterize stable paleomagnetic recorders in natural samples.
Supporting Information for "Mapping magnetic signals of individual magnetite grains to their internal magnetic configurations using micromagnetic models"

David Cortés-Ortuño¹, Karl Fabian², and Lennart V. De Groot¹

¹Paleomagnetic laboratory Fort Hoofddijk, Department of Earth Sciences, Utrecht University, Budapestlaan 17, 3584 CD Utrecht, The Netherlands.
²Norwegian University of Science and Technology (NTNU), S. P. Andersens veg 15a, 7031 Trondheim, Norway

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S1 Magnetic signal of a single cuboid

In the manuscript the magnetic signal from a grain simulated with the micromagnetic code was calculated by computing the dipole field of every magnetization vector in the grain model. This allowed to compute the total magnetic flux produced by the grain at a surface above it. To analyze the accuracy of this approximation a test model is defined using a cuboidal particle. The stray field of this grain can be calculated analytically and therefore the magnetic field it produces at a surface, which can be compared directly with the simulated grain.

S1.1 Physical model

For a uniformly magnetized cuboid grain (located at the origin), the demagnetizing field can be computed from the potential function that is defined over the boundary surfaces (faces) \( \partial \Omega \) of the cuboid as

\[
\phi(r) = \frac{1}{4\pi} \int_{\partial \Omega} \frac{\mathbf{M} \cdot d\mathbf{S}'}{|\mathbf{r} - \mathbf{r}'|}.
\] (1)

In equation 1, \( d\mathbf{S}' = \hat{n} \cdot dS' \) is the surface element, with \( \hat{n} \) as the unit vector normal to the faces, \( \mathbf{M} \) is the cuboid magnetization, \( \mathbf{r} \) is the location of the reference point and the vectors \( \mathbf{r}' \) point to the locations of the magnetic sources, \( i.e. \) the cuboid faces infinitesimal elements. This model is illustrated in Fig. S1, where position vectors are defined with respect to an arbitrary origin \( \mathcal{O} \). The magnetic field is defined as

\[
\mathbf{B}(\mathbf{r}) = -\mu_0 \nabla \phi(\mathbf{r}).
\] (2)
SUPP. FIG. S1: Model system for the calculation of the stray field of a uniformly magnetized cuboid. Position vectors are defined with respect to an arbitrary origin $\mathcal{O}$. The scan grid is defined by a rectangular grid of sensor points in the $xy$-plane. The cuboid center is specified by the vector $\xi$ such that the positions of infinitesimal cuboid face elements $r'$ are defined as $r' = \xi + r''$, with $r''$ as the location of the magnetic sources with respect to the cuboid center reference system.

For the case studied in the manuscript a single scan sensor is approximated as a physical point. Therefore, by defining $\gamma_B = \mu_0 / (4\pi)$, the field component normal to a sensor grid point defined in the $xy$-plane is calculated as
\[ B_z(r) = -\mu_0 \frac{\partial}{\partial z} \left( \frac{1}{4\pi} \int_{\partial \Omega} \frac{\mathbf{M} \cdot d\mathbf{S}'}{|\mathbf{r} - \mathbf{r}'|} \right) \] (3)

\[ = -\gamma_B \int_{\partial \Omega} \mathbf{M} \cdot d\mathbf{S}' \frac{\partial}{\partial z} \left( \frac{1}{|\mathbf{r} - \mathbf{r}'|} \right) \] (4)

\[ = \gamma_B \int_{\partial \Omega} \mathbf{M} \cdot d\mathbf{S}' \frac{\partial}{\partial z'} \left( \frac{1}{|\mathbf{r} - \mathbf{r}'|} \right) \] (5)

In equation 5 the derivative variable was changed, which is more convenient for two of the cuboid faces. The integral limits can be simplified if the integration variable is changed to the coordinate system of the cuboid center \( \mathbf{\xi} \) by noticing that \( \mathbf{r}' = \mathbf{\xi} + \mathbf{r}'' \), as shown in Fig. S1. In this case \( d\mathbf{S}' = d\mathbf{S}'' \) and \( \frac{\partial f}{\partial z'} = \frac{\partial f}{\partial z''} \) for an arbitrary function \( f \), hence the field can be expressed as

\[ B_z(r) = \gamma_B \int_{\partial \Omega} \mathbf{M} \cdot d\mathbf{S}'' \frac{\partial}{\partial z''} \left( \frac{1}{|\mathbf{r} - (\mathbf{\xi} + \mathbf{r}'')|} \right) \] (6)

\[ = \gamma_B \int_{\partial \Omega} \mathbf{M} \cdot d\mathbf{S}'' \frac{\partial}{\partial z''} \left( \frac{1}{|\mathbf{X} - \mathbf{r}''|} \right) \] (7)

with \( \mathbf{X} = \mathbf{r} - \mathbf{\xi} \). Alternatively, the field can also be written as

\[ B_z(r) = -\gamma_B \frac{\partial}{\partial z} \int_{\partial \Omega} \mathbf{M} \cdot d\mathbf{S}'' \left( \frac{1}{|\mathbf{X} - \mathbf{r}''|} \right) \] (8)

To compute the integrals we refer to (Hubert & Schäfer 1998, p. 122) where integrations are carried out based on the source function \( F_{000}(r) \) such that
Using equation 7 it is now possible to compute the field contribution of the cuboid face in the $yz$-plane, where $x'' = a$ and $\hat{n} = +\hat{x}$, as
Similarly, for the opposite face with normal in the $-\hat{x}$ direction the resulting field is

\[ B_{z}^{[-\hat{x}]} (\mathbf{r}) = -\gamma_{B} (M_{x}) F_{010} (\mathbf{r} - [\xi + r'']) \]

and for the faces in the $xz$-planes (normals in the $\pm \hat{y}$ directions) the field expressions are

\[ B_{z}^{[\pm \hat{y}]} (\mathbf{r}) = -\gamma_{B} (\pm M_{y}) F_{100} (\mathbf{r} - [\xi + r'']) \]

Finally, for the faces in the $xy$-planes (normals in the $\pm \hat{z}$ directions), the field is

\[ B_{z}^{[\pm \hat{z}]} (\mathbf{r}) = \gamma_{B} (M_{z}) \int_{y'' = -b}^{y'' = +b} \int_{z'' = -c}^{z'' = +c} dy'' dz'' \left( \frac{1}{|\mathbf{X} - \mathbf{r''}|} \right) \]

...
SUPP. FIG. S2: Comparison of the stray field of the uniformly magnetized cuboid model computed from both an analytical formulation and a micromagnetic simulation. The cuboid is defined as a cube of 50 nm edge size with its center at $z = -35$ nm. The stray field is calculated at a height of $z = 500$ nm.

Computed using equation 8 to obtain

$$B_z^{[\pm \xi]}(r) = -\gamma_B (\pm M_z) \frac{\partial}{\partial z} \left[ \int_{x''=-a}^{x''=+a} \int_{y''=-b}^{y''=+b} dx'' dy'' \left( \frac{1}{|X - r''|} \right) \right]_{z''=\pm c}$$

$$= -\gamma_B (\pm M_z) \frac{\partial}{\partial z} \left[ (-1)^2 F_{110} (r - [\xi + r'']) \right]_{z''=\pm c, y''=\pm b, x''=\pm c}$$

$$= -\gamma_B (\pm M_z) F_{11-1} (r - [\xi + r''])$$

$$\left. \right|_{z''=\pm c, y''=\pm b, x''=\pm a}$$
SUPP. TABLE S1: Results of multipole inversions of different order applied to the test cuboid system.

**S1.2 Cuboid test model**

The theoretical results obtained for the stray field of a cuboid particle are compared to the results obtained from a micromagnetic simulation using the finite element code MERRILL (Ó Conbhui et al. 2018). A cuboid of dimensions $50 \text{ nm} \times 50 \text{ nm} \times 50 \text{ nm}$ is defined with its center at $z = -35 \text{ nm}$. In addition, the particle’s magnetization is specified with a magnitude of $|\mathbf{M}| = 0.48 \text{ MA/m}$ and orientation $\mathbf{M}/|\mathbf{M}| = 0.577 \times (1, 1, -1)$. The $z$-component of the stray field for both the theoretical and simulation models are calculated on a scan grid of size $1.5 \mu\text{m} \times 1.5 \mu\text{m}$ at a height position of $z = 0.5 \mu\text{m}$. A comparison of the stray field results is shown in Fig. S2. The relative error of the simulation with respect to the theoretical solution in this case is
estimated as \( B_{\text{err}} = \| B_{z}^{\text{sim}} - B_{z}^{\text{theory}} \|_F / \| B_{z}^{\text{theory}} \|_F = 2.7183 \times 10^{-5} \). This small value confirms that the stray field approximation of the micromagnetic simulation, where the magnetization vectors at finite element nodes are treated as point dipoles, is in excellent agreement with the analytical formulation. Therefore the micromagnetic simulation can be used as a model to analyze the stray field signal of more complex magnetic structures.

Numerical inversions of the micromagnetic model of the test cuboid system were performed using multipole expansions up to the octupole order. The results of these inversions are summarized in Table S1 where the small relative error of the residuals indicate that the inverted stray field is approximated with high accuracy, which is supported by the correct magnitudes of the inverted magnetization and the inverted dipole moments. In particular, a dipole order expansion is sufficient to reproduce the right values of the cuboid magnetization.
SUPP. FIG. S3: Visualization of the initial states used for the simulations of a magnetite sphere using the MERRILL code. The selected uniform states are used to obtain magnetic states oriented in the [101] direction. Arrows depict the normalized magnetization vectors which are colored by their $z$-component. The titles indicate the type of initial state used in the simulations. For the RANDOM configuration, the Randomize All Moments command was used. For the UNIFORM configuration, the Uniform Magnetization 1.00 0.00 1.00 command together with Randomize Magnetization 10 were used. For the UNIFORM RANDOM state the random component was specified with a value of 35 degrees. In the manuscript, the uniform states are specified in 26 different directions in the Cartesian plane.
S3  Surface reconstruction

The generation of the finite element mesh for the grain 4 of Area 2 defined in the synthetic sample of (de Groot et al. 2018b) requires to reconstruct a surface that can be partitioned into tetrahedral units. This surface can be generated from an unstructured cloud of points which, in this case, is obtained from the the microCT data. Specifically, in (de Groot et al. 2018b) the voxel representation of the grains from the microCT data is transformed into a cuboid decomposition of the grains where voxels are grouped into cuboids of maximal available size that fit within the grain geometry with an efficient final representation of the grain shape. For the reconstruction of the grain surface, a cloud of points is specified by taking both the cuboid vertices and the center of every cuboid face, in order to produce more points for the reconstruction algorithm. This point data is publicly available in (de Groot et al. 2018a) for the synthetic sample of (de Groot et al. 2018b). In the case of grain 4, the cuboid decomposition together with the cuboid vertices and cuboid face centers are shown in Fig. S4. For this model the grain size was downscaled to 2% of the original grain size.

Optimized algorithms using unstructured set of points are implemented in the Computational Geometry Algorithms Library (CGAL) (The CGAL Project 2021). For the model used in the manuscript, the Poisson Surface Reconstruction (PSR) (Alliez et al. 2021) was applied, which requires a calculation of the normal vectors at every point. The calculation of the normals was achieved using the CGAL: :jet_estimate_normals function and consequently the normals were oriented with the CGAL: :mst_orient_normals function. To generate a surface with the PSR algo-
SUPP. FIG. S4: Surface reconstruction and finite element mesh for the magnetic grain 4 in the synthetic sample of (de Groot et al. 2018b). The decomposition of the grain into cuboids (by grouping voxels from microCT data) is shown by transparent structures with orange edges and was obtained from the grain data in (de Groot et al. 2018a). Cuboid vertices and face centers are depicted as small spheres. The surface was constructed via the Poisson Surface Reconstruction algorithm and the volume was partitioned into tetrahedral units.

The resulting surface is depicted in Fig. S4, which shows that the algorithm approximates grain geometry efficiently. An improved surface reconstruction can be achieved in the future by using the voxel representation of the grains from the microCT data.
directly.

The final step to obtain a finite element mesh for the micromagnetic simulations is to partition the volume defined by the reconstructed surface into tetrahedrons with edge lengths smaller than the exchange length of the material. As specified in the main manuscript, the CGAL library contains tetrahedralization algorithms. In particular, for the grain 4 model the Tetrahedral Isotropic Remeshing method (Tournois et al. 2021) was applied, which is based on the Multi-Material Adaptive Volume Remesher algorithm. The tetrahedralization of the grain 4 surface was achieved by firstly, generating a three dimensional volume mesh using the CGAL::make_mesh_3 function and consequently refining this mesh with the remeshing function CGAL::tetrahedral_isotropic_remeshing. This last method produced more efficient results (more consistent edge length sizes) than refining the mesh with the CGAL::refine_mesh_3 function, and was implemented in one of the latest CGAL releases with version number 5.1.

The CGAL code generates the mesh in .mesh format while the MERRILL code accepts both PATRAN .neu and Tecplot .tec mesh formats. Therefore, to convert the mesh for the grain model the meshio (Schlömer 2021) code is used with modifications from the authors of this work.

S3.1 Energy minimization and mesh statistics

Finding metastable magnetic configurations in a mesh with substantial number of nodes is a computationally cost calculation. In particular when the system is initialized with randomly oriented magnetization vectors because the algorithm requires
SUPP. FIG. S5: Statistical data of the finite element mesh for the magnetite grain model from a synthetic sample at room temperature. (a) Cumulative distribution of the edge lengths of the mesh tetrahedra for both a coarsely and finely discretized mesh. Vertical lines indicate the 50th and 99th percentiles. (b) Mesh quality of tetrahedra, or cells, using the normalized shape ratio parameter (Field 1991) (see discussion in text). Vertical lines indicate the 1st and 50th percentiles.

to find LEM states in a complex energy landscape by following different paths that minimize the micromagnetic energy. To speed up the process of energy minimization for the grain model of the synthetic sample, the strategy used in the paper is:

- Define a coarsely discretized mesh, *i.e.* using tetrahedra with edge lengths
larger than the material’s exchange length $\ell_{ex}$.

- Start the minimization and find a candidate for a LEM.

- Define the \textit{physically correct} mesh using tetrahedron’s edge lengths smaller than $\ell_{ex}$.

- Interpolate the magnetization field of the LEM found with the coarse mesh into the magnetization field of the refined mesh. This feature is implemented in the MERRILL code.

- Minimize the energy of the refined mesh using a Conjugate Gradient method in Cartesian coordinates and find a LEM.

Statistics of the FEM tetrahedra, or cells, for the mesh of the grain model of the synthetic sample (see Fig. S4) are detailed in Fig.S5. Plot (a) shows the cumulative distribution of edge lengths for both the coarse and fine meshes. In the case of the fine mesh, the 50th and 99th percentiles of the edge lengths are located at 5.90 nm and 8.12 nm, respectively, which are smaller than $\ell_{exch} = 9.59$ nm of magnetite at room temperature. For the coarse mesh the 99th percentile is located at 20.37 nm. Furthermore, the number of tetrahedron edges for the fine mesh is an order of magnitude larger than that of the modeled coarse mesh and its distribution is narrower than that of the coarse mesh. Plot (b) depicts a quantification of the quality of the meshes by means of a normalized shape ratio (Field 1991). For every tetrahedron cell, which is a 3-simplex, this parameter is defined as the ratio of the radius of the sphere inscribed in the cell (inradius) divided by the radius of the sphere enclosing
the cell (circumradius), multiplied by the circumradius to inradius ratio of the regular \( k \)-simplex, or simply the cell dimension, which in this case is 3. Meshes with normalized shape ratios closer to 1 translates into a good mesh quality, since the cells are close to regular tetrahedra. Tetrahedrons with ratios closer to zero are poorly defined by having edges with large variations in size or are flat in shape, which can affect the convergence of the algorithm (Ó Conbhuí et al. 2018). For the fine mesh the 1st and 50th percentile are 0.58 and 0.87, respectively. For the coarse mesh the 1st percentile is located at 0.51. These mesh statistics were computed using the Dolfin (Logg & Wells 2010) finite element code.
SUPP. FIG. S6: Magnetic configurations obtained after energy minimization of the grain 4 of Area 2 of the synthetic sample of (de Groot et al. 2018b), starting from six different states where magnetization vectors are randomly oriented. In the main manuscript, configurations 1, 5 and 6 were chosen to perform numerical inversions of the stray field produced by them.
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