Supplementary Information for

Magnetically Textured Powders — An Alternative to Single-Crystal and Powder X-ray Diffraction Methods

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1. Magnetic energy

The magnetic energy of a particle induced by the interaction of an induced magnetic dipole and applied magnetic field is expressed by

$$E = \frac{-V}{2\mu_0} {}^{t}\mathbf{B}(\boldsymbol{\chi}\mathbf{B}) , \qquad \text{(S1)}$$

where μ_0 is the magnetic permeability of free space, V is the volume of the particle, $\boldsymbol{\chi}$ is the magnetic susceptibility tenor, $\mathbf{B}$ is the magnetic flux density (simply called the magnetic field), and the suffix t indicates the transpose. Here, $\boldsymbol{\chi}$ and $\mathbf{B}$ are assumed to be expressed in the laboratory coordinate system. The tensor $\boldsymbol{\chi}$ is derived from the tensor $\boldsymbol{\chi}'$ that is expressed in its principal-axis system as follows:

$$\boldsymbol{\chi} = {}^{t}\mathbf{A}\boldsymbol{\chi}'\mathbf{A} . \qquad \text{(S2)}$$

We use the Euler angles ϕ, θ, and ψ (Fig. S1) defined by the *zyx* (yaw, pitch, and roll) convention. Then, the transformation matrix **A** is given by (1)

$$\mathbf{A} = \begin{pmatrix} \cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta \\ \sin\psi\sin\theta\cos\phi - \cos\psi\sin\phi & \sin\psi\sin\theta\sin\phi + \cos\psi\cos\phi & \cos\theta\sin\psi \\ \cos\psi\sin\theta\cos\phi + \sin\psi\sin\phi & \cos\psi\sin\theta\sin\phi - \sin\psi\cos\phi & \cos\theta\cos\psi \end{pmatrix} . \qquad \text{(S3)}$$

The tensor $\boldsymbol{\chi}'$ is expressed by

$$\boldsymbol{\chi}' = \begin{pmatrix} \chi_1 & 0 & 0 \\ 0 & \chi_2 & 0 \\ 0 & 0 & \chi_3 \end{pmatrix} , \qquad \text{(S4)}$$

where χ_1, χ_2, and χ_3 are the principal values, where we define $\chi_1 > \chi_2 > \chi_3$ for biaxial crystals.

In the case where a static magnetic field **B** is applied to a biaxial crystal whose susceptibility tensor is defined by eq. (S4) in the *x*-direction, we insert $\mathbf{B} = (B, 0, 0)$ into eq. (S1) to obtain

$$E_{\text{stat}} = \left(\frac{VB^2}{2\mu_0}\right)\left\{(\chi_1 - \chi_2)\phi^2 + (\chi_1 - \chi_3)\theta^2\right\}, \qquad \text{(S5)}$$

where the isotropic and higher terms are discarded. In the case where a magnetic field rotating in the *xy*-plane at angular velocity ω is applied to a biaxial crystal, we insert $\mathbf{B} = B(\cos\omega t, \sin\omega t, 0)$ into eq. (S1) to obtain

$$E_{\text{rot}} = \left(\frac{VB^2}{4\mu_0}\right)\left\{(\chi_2 - \chi_3)\psi^2 + (\chi_1 - \chi_3)\theta^2\right\}, \qquad \text{(S6)}$$

where the time-average over one cycle,

$$\frac{-V}{2\mu_0}\frac{\omega}{2\pi}\int_0^{2\pi/\omega} {}^t\mathbf{B}(\chi\mathbf{B})dt \qquad \text{(S7)}$$

was made under the assumption of the "rapid rotation regime" (RRR) condition, which will be described in the next section. The isotropic and higher terms are discarded. If we take an AMMF, $\mathbf{B} = B(\cos\omega t, q\sin\omega t, 0)$ with $0 < q < 1$, and apply the same procedure, we obtain

$$E_{\text{AMMF}} = (2\mu_0)^{-1}B^2V\{\frac{q^2}{2}(\chi_2 - \chi_3)\psi^2 + \frac{1}{2}(\chi_1 - \chi_3)\theta^2 + \frac{(1-q^2)}{2}(\chi_1 - \chi_2)\phi^2\}, \qquad \text{(S8)}$$

which is equal to the result described in the main text.

2. Orientation kinetics

The equation of motion of a biaxial microcrystal under a dynamic magnetic field $\mathbf{B}(t)$ is expressed by the balance of the magnetic torque **N** and hydrodynamic torque **M** as follows:

$$\mathbf{N} + \mathbf{M} = 0 \qquad \text{(S9)}$$

with

$$\mathbf{N} = V\boldsymbol{\chi}\mathbf{B}\times\mathbf{B}/\mu_0 \text{ and } \mathbf{M} = -\eta\,\mathbf{L}\boldsymbol{\Omega}. \qquad \text{(S10)}$$

Here, **L** is the hydrodynamic tensor, η is the viscosity of the medium, and $\boldsymbol{\Omega}$ is the angular velocity vector. The tensor $\mathbf{L}$ is derived by the transformation of the tensor $\mathbf{L}'$ that is expressed in its principal-axis system as follows:

$$\mathbf{L} = {}^{t}\mathbf{A}\,\mathbf{L}'\mathbf{A}, \qquad \text{(S11)}$$

where the transformation matrix $\mathbf{A}$ is given by eq. (S3) and $\mathbf{L}'$ is expressed by

$$\mathbf{L}' = \begin{pmatrix} L_1 & 0 & 0 \\ 0 & L_2 & 0 \\ 0 & 0 & L_3 \end{pmatrix}. \qquad \text{(S12)}$$

If the particle is prolate or oblate ellipsoid, the three components of **L** are functions of their aspect ratio, and if the particle is a sphere, $L_1 = L_2 = L_3 = 8\pi a^3$, where a is the radius of the sphere (2,3). The vector $\boldsymbol{\Omega}$ is expressed by (1)

$$\boldsymbol{\Omega} = \begin{pmatrix} \dot{\psi}\cos\theta\cos\phi - \dot{\theta}\sin\phi \\ \dot{\psi}\cos\theta\sin\phi + \dot{\theta}\cos\phi \\ \dot{\phi} - \dot{\psi}\sin\theta \end{pmatrix}, \qquad \text{(S13)}$$

where a dot indicates the derivative with respect to time. Inserting eq. (S2) and (S10)–(S13) into eq. (S9), we obtain simultaneous differential equations for ϕ, θ, and ψ with respect to time.

These differential equations were solved numerically using Mathematica, where a sphere was assumed and the following parameters were used:

$\mathbf{B} = B(\cos\zeta, \sin\zeta, 0)$ with $\zeta = \omega t - 0.4\sin(2\omega t)$ and $B = 0.5$ T; $\chi_1 = -9.81\times10^{-6}$,

$\chi_2 = -10.11\times10^{-6}$, and $\chi_3 = -10.41\times10^{-6}$; η =1 Pa s;

initial conditions, $\phi(0) = \theta(0) = \psi(0) = \pi/4$; τ =50.3 s according to eq. (6).

Simulation 1 (SRR): ω = 0.0040 rad/s and time duration was 2000 s. $\omega\tau = 0.2$ rad.
Simulation 2 (ARR): ω = 0.020 rad/s and time duration was 1000 s. $\omega\tau = 1$ rad.
Simulation 3 (RRR): ω = 0.20 rad/s and time duration was 1000 s. $\omega\tau = 10$ rad.

The numerical results for three Euler angles ϕ, θ, and ψ are plotted as a function of time in Fig. S1. According to the $\omega\tau$ values, Simulation 1, 2, and 3 correspond to the "synchronous rotation regime" (SRR), "asynchronous rotation regime" (ARR), and RRR, respectively. The movies corresponding to SRR, ARR, and RRR are attached and designated as SRR_02rad, ARR_1rad, and RRR_10rad, respectively. We find that the alignment is fixed with respected to the laboratory coordinates only when the RRR condition is satisfied.

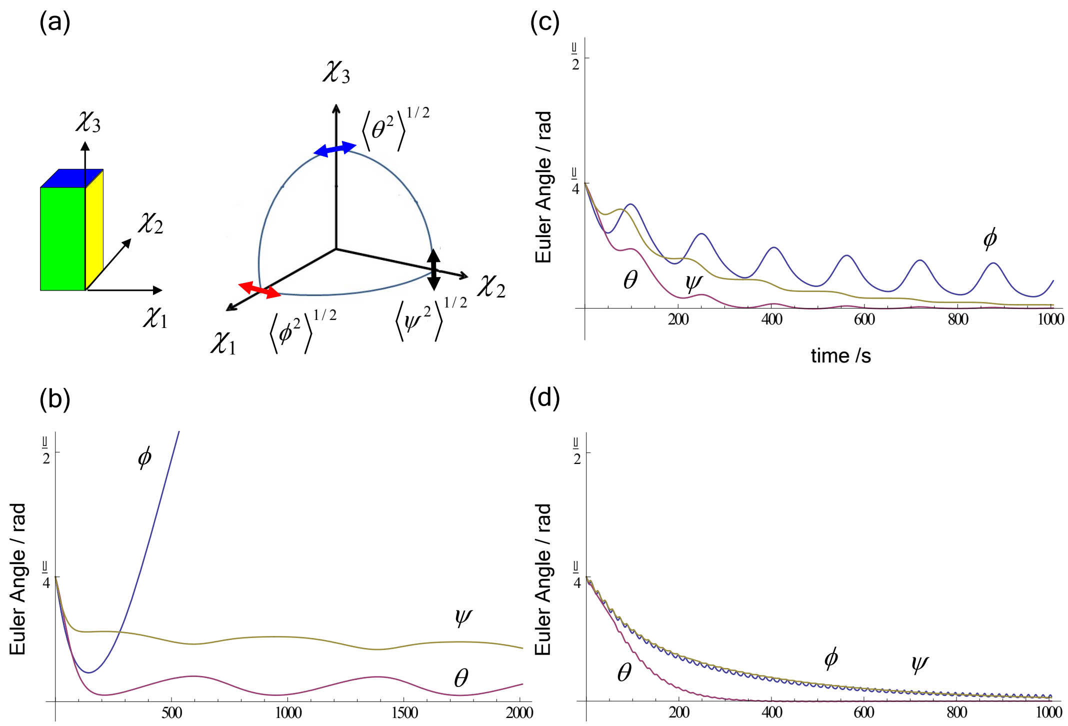

Fig. S1 (a) The definition of magnetic axes used in the movies and the definition of the fluctuations of the Euler angles ϕ, θ, and ψ in relation to the magnetic axes. The results for the temporal changes of Euler angles, solved numerically by Mathematica, are shown for the (b) SRR,

(c) ARR, and (d) RRR, which correspond to movie SRR_02rad, ARR_1rad, and RRR_10rad, respectively. For the SRR, the angle ϕ completely follows the magnetic rotation; for the ARR, the angle ϕ oscillates and the average does not reach zero; for the RRR, all the angles reach zero (orientation is fixed with respect to the laboratory coordinates), although a slight oscillation of ϕ still occurs.

Captions for movies

SRR_02rad shows the motion under the SRR condition, in which the χ_1 axis follows the rotation of the magnetic field, and thus the alignment cannot be fixed.

ARR_1rad shows the motion under the ARR condition, in which the χ_3 axis aligns in the direction of the z-axis, but the χ_1 axis rotates asynchronously with the rotating magnetic field, resulting in oscillations.

RRR_10rad: under the RRR condition, biaxial (3D) alignment is achieved.

References

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(3) C. Tsuboi, S. Tsukui, F. Kimura, T. Kimura, K. Hasegawa, S. Baba and N. Mizuno, *J. Appl. Cryst*., 2016, **49**, 2100–2105.