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Three-dimensional neutron far-field tomography of a bulk skyrmion lattice

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Supplementary Materials

All experimental, phantom, and reconstructed datasets used in this study can be found in an online repository located here (https://doi.org/10.5281/zenodo.7115410) [1].

Forward Propagator

The scattering pattern from passing through a single MSFR volume $S_{Q,\theta}$ for some projection θ is computed by propagating a neutron wave function through the sample via the time-evolution operator $\mathcal{U}$

$$S_{Q,\theta} = \left| \left\langle Q \middle| \mathcal{U} \middle| K_\theta \right\rangle \right|^2 . \tag{1}$$

Which is estimated using a translation operator along the z-direction

$$\mathcal{J}_{\theta,dz} = e^{iK_z(\theta,Q)dz} \tag{2}$$

assuming elastic scattering

$$K_z(\theta, Q) = \sqrt{K^2 - (K\sin\theta - Q_x)^2 - Q_y^2}, \tag{3}$$

where K is the incoming neutron wave number, along with potential the potential V_z at layer z

$$\mathcal{U}_{\theta,z+dz} = \mathcal{J}_{\theta,dz}\left[1 - iV_z dz\, m_n/K_z\right], \tag{4}$$

where m_n is the neutron mass, and dz is the voxel height. The potential from the sample is computed from its resulting magnetic field by applying the characteristic $-\hat{Q}\times\hat{Q}\times$ operator in Fourier-space [2]

$$\boldsymbol{B}_s = -4\pi M_s \mathcal{F}^{-1}\left\{\hat{Q}\times\hat{Q}\times\mathcal{F}\left[\boldsymbol{m}\right]\right\} \tag{5}$$

where M_s is the saturated magnetization, with $4\pi M_s = 1900$ G estimated from DC susceptibility measurements taken at 310 K. The potential also includes the external field and operates on a neutron spinor via Pauli matrices

$$V = -\mu_n \boldsymbol{\sigma}\cdot(\boldsymbol{B}_s + \boldsymbol{H}_{\text{ext}}). \tag{6}$$

The unpolarized cross section

$$S_{Q,\theta} = \sum_{s,s'} \left|\langle Q, s'|\mathcal{U}|K_\theta, s\rangle\right|^2 \tag{7}$$

is then computed by summing over input s and selected s' spin states.

Assuming the longitudinal correlation length of the sample is smaller than the reconstruction height, the observed scattering pattern can be estimated by self-convolving the scattering pattern of a single MSFR N times

$$I_{Q\theta} = \sum_{Q'',Q'} Y_{QQ'} W^{\theta}_{Q'Q''} \left[(S*)^N I_0\right]_{Q''\theta}, \tag{8}$$

where N is the ratio of the sample dimension along the propagation direction and the corresponding size of the MSFR. For this sample $N = 860$. Because the forward propagator is a function of the projection of the momentum transfer in the $x-y$ plane Q_{xy}, the sparse matrix W shifts the scale of Q to be horizontal to the propagation axis. The sparse matrix Y smears out $Q = K\Omega_Q \simeq K_{\text{avg}}\Omega_Q(1-\delta\lambda/\lambda)$, where Ω_Q is the scattering angle, over the neutron's incoming wavelength distribution.

Seeding

The initial guess is formed by first estimating the three-dimensional vector amplitude of the MSFR in Fourier-space $|\widetilde{m}_0|^2$. This is accomplished by first performing a N^{th}-order deconvolution of the scattering signal with itself and the incoming beam profile, which solves for I_θ

$$I_{\text{meas}} = (I_\theta *)^N I_0 \tag{9}$$

with the I_θ slices of $|\widetilde{m}|$ for each measured projection unknown. This is accomplished with modified Richardson-Lucy algorithm, which is iterative

$$I_{\theta,i+1} = I_i\left[\left(\frac{I_{\text{meas}}}{\epsilon + (I_{\theta,i}*)^N I_0}\right) * (I_{\theta,i}*)^{N-1} I_0\right] \tag{10}$$

where ϵ is a small regulator. When N is large, this can be unstable, and we found it is often helpful to enact this algorithm in stages, with I_{meas} deconvolved from I_0, then performing the deconvolution in n stages with $N' = N^{1/n}$. It can also be helpful to track the error of the deconvolution and stop iterating, or change the regulator size when the error stops decreasing with iteration number.

After deconvolution is complete, the Bragg peaks were fit to a Lorentzian profile with respect to Q_z. The projection with the incoming beam aligned with the dominant direction of the skyrmion tubes $I_{\theta=0}$ was then taken as the $|\widetilde{m}|$-slice at zero Q_z and expanded along the $Q_0 = D/J$ sphere, but attenuated according to the fitted Lorentzian with respect to Q_z. The result is a smooth function with a Lorentzian envelope along Q_z that preserves the transverse correlation structure of $I_{\theta=0}$. All the $Q > 0$ structure is then scaled and a DC term is introduced to $\widetilde{m}_0$ to generate the expected net sample magnetization $\langle m \rangle = \widetilde{m}(Q = 0)$.

After forming an estimate of $|\widetilde{m}_0|$, a guess for m is generated with an alternating projections algorithm. The vector field of the MSFR m is iteratively amended according to its constraints in Fourier and real-space. The Fourier-space constraint is given by the magnitude estimated from the deconvolved SANS data $|\widetilde{m}_0|$

$$\widetilde{\boldsymbol{m}}_{i+1} = \widetilde{\boldsymbol{m}}_i \frac{|\widetilde{m}_0|}{|\widetilde{m}_i|} \tag{11}$$

For the first few iterations, the transverse components of $\widetilde{m}$ are also redefined according to the sign of the DM term and expected curl around m_z

$$\widetilde{m}_{xy} = \pm i Q_{yx} \widetilde{m}_z. \tag{12}$$

The real space constraint is $\boldsymbol{m}^2 = 1$. However, it can also be beneficial to let m relax through a few iterations of a free energy minimizer before reapplying Fourier-space constraints. The net result is a guess m that adheres reasonably-well to the measured SANS projections, while also having a low free energy.

Minimization

The chosen cost function was

$$f = \chi^2 + \beta \mathcal{F}, \tag{13}$$

where the χ^2 is the sum of weighted residuals for all the projections

$$\chi^2 = \sum_{Q,\theta} (I_{Q,\theta} - M_{Q,\theta})^2 w_{Q,\theta} \tag{14}$$

and the free energy includes symmetric and antisymmetric exchange terms and a Zeeman term

$$\mathcal{F} = -\frac{1}{2}\boldsymbol{m} \cdot \nabla^2 \boldsymbol{m} + Q_0\, \boldsymbol{m} \cdot (\nabla \times \boldsymbol{m}) - \boldsymbol{h} \cdot \boldsymbol{m} \tag{15}$$

where the reduced field $\boldsymbol{h} = \boldsymbol{H} Q_0^2/J$, and helical shell radius $Q_0 = D/J$ is taken from the SANS patterns. The χ^2 depends on the difference between the measured SANS patterns $M_{Q,\theta}$ and estimated SANS intensity for each projection $I_{Q,\theta}$ computed from the MSFR via Eqn 8. The residuals are weighted according to $w_{Q,\theta} = 1/\sigma^2_{Q,\theta}$, which is taken from the SANS reduction software [3], with the dominant uncertainty from Poisson counting statistics.

The Lagrange multiplier β acts like a Boltzmann factor. Since the $\chi^2 \sim \mathcal{O}(\sqrt{N_{Q,\theta}})$, where $N_{Q,\theta}$ is the total number of measured SANS pixels over all projections, and the two kinetic terms in $\mathcal{F} \sim \mathcal{O}(Q_0^2)$, a reasonable choice is $\beta = \sqrt{N_{Q,\theta}}/Q_0^2$. Adjusting the weight of the Zeeman term h will change the average magnetization $\langle m_z \rangle$. A reasonable value of h can be selected based on studying the behavior of the free energy term in isolation [4]. However, we found that larger values of h were needed when the χ^2 was introduced to achieve the same average magnetization, likely because the χ^2 acts like a kinetic term which further reinforces the helical shell size Q_0. We therefore chose to study the behavior of the reconstructions over a range of h, as is shown in Table 1.

The gradient of the free energy term was computed via finite difference methods, similar to OOMMF [5]. The derivative of the χ^2 necessitates taking the derivative of the forward operator, which results in computing the overlap of the forward-propagating wavefunction ψ_θ with a backward-propagating residual wavefunction χ_θ for each projection

$$\frac{\delta}{\delta \boldsymbol{m}} \chi^2 = i \mu_n M_s m_n dz \sum_\theta \frac{1}{K_z} \mathcal{F}^{-1} \left\{ \mathcal{F} \left\{ \mathrm{Im} \left[\chi_\theta^\dagger \boldsymbol{\sigma} \psi_\theta \right] \right\} \cdot \left(\mathbb{I} - \hat{Q}\hat{Q} \right) \right\}, \tag{16}$$

where $\mathcal{F}\{\cdots\}$ and $\mathcal{F}^{-1}\{\cdots\}$ indicate forward and reverse Fourier transforms, respectively. The wavefunctions are computed by a combination of forward and backward propagation operators

$$\begin{aligned} \psi_\theta &= \left\langle x \middle| \mathcal{U}_z \middle| K_\theta \right\rangle \\ \chi_\theta &= \sum_Q \left\langle x \middle| \mathcal{U}_{z,f}^{-1} \middle| Q \right\rangle \widetilde{\psi}_{f,Q} G_Q \\ G_Q &= 2 \sum_{Q',Q''} Y_{Q,Q'} W_{Q'',Q'} \left\{ I_0 * [(I - M) w] \right\}_{Q'',\theta} \end{aligned} \tag{17}$$

where tildes indicate Fourier-space representations; subscript f denotes the final state; and $\mathcal{U}_{z,f}^{-1}$ is the backward in time propagator, starting from the final MSFR layer.

Defect Densities

The emergent magnetic field is computed from the MSFR and its derivatives

$$b_i = \frac{1}{2}\epsilon_{ijk}\boldsymbol{m} \cdot (\partial_j \boldsymbol{m} \times \partial_k \boldsymbol{m}), \tag{18}$$

where ϵ_{ijk} is the fully antismmetric tensor; $\partial_i \equiv \partial/\partial x_i$; and repeated indices are summed over the three space coordinates. Summing z-component of this field over an area is identified as the skyrmion winding number in the enclosed area, and the emergent magnetic charge density is defined as the source term for the emergent magnetic field

$$4\pi\rho_{\mathrm{em}} = \nabla \cdot \boldsymbol{b}. \tag{19}$$

A peak-finding algorithm was used to identify local maxima of ρ_{em}^2 that survive a threshold cut after a Gaussian blurring. The total emergent charge of the defect was then estimated to be the sum of ρ_{em} over the neighboring ± 2 voxels in all directions. Further classification of branching (two skyrmion events) versus segmentation (single skyrmion events) was accomplished by summing $\partial_z m_z$ over the same neighborhood. The same or differing signs of the two summations indicate segmentation and branching defects, respectively.

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