Direct observation of attractive skyrmions and skyrmion clusters in the cubic helimagnet Cu$_2$OSeO$_3$

J. C. Loudon, A. O. Leonov, A. N. Bogdanov, M. Ciomaga Hatnean, and G. Balakrishnan

1Department of Materials Science and Metallurgy, 27 Charles Babbage Road, Cambridge, CB3 0FS, United Kingdom
2Department of Chemistry, Faculty of Science, Hiroshima University Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8526, Japan
3Chiral Research Center, Hiroshima University, Higashi-Hiroshima, 739-8526, Japan
4IFW Dresden, Postfach 20016, D-01171 Dresden, Germany
5Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom

We report the discovery of attractive magnetic skyrmions and their clusters in noncentrosymmetric ferromagnets. These three-dimensional solitons have been predicted to exist in the cone phase of chiral ferromagnets [J. Phys: Condens. Matter 28, 35LT01 (2016)] and are fundamentally different from the more common repulsive axisymmetric skyrmions that occur in the magnetically saturated state. We present real-space images of these skyrmion clusters in thin (∼70 nm) single-crystal samples of Cu$_2$OSeO$_3$ taken using transmission electron microscopy and develop a phenomenological theory describing this type of skyrmion.

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I. INTRODUCTION

In magnetic compounds lacking inversion symmetry, the handedness of the underlying crystal structure induces an asymmetric exchange coupling called the Dzyaloshinskii-Moriya (DM) interaction [1] which stabilizes long-period spatial modulations of the magnetization with a fixed rotation sense [1,2]. There has been a renewed interest in chiral helimagnetism in the last few years since the discovery of two-dimensional localized modulations called chiral skyrmions [3–6]. Nonaxisymmetric skyrmions have been investigated theoretically in bulk [13] and confined chiral helimagnets [14], but no experimental observations of these states have been reported to date. Here we show that attractive skyrmions can occur in the cone phase of the cubic helimagnet Cu$_2$OSeO$_3$ by presenting transmission electron micrographs of skyrmion clusters and provide a theoretical description of these results.

II. THEORY

Chiral solitons and modulated phases are described mathematically by equations which minimize the energy functional for a chiral ferromagnet [1,6]:

$$w = A(\nabla M)^2 + K(M \cdot n)^2 - \mu_0 M \cdot H + w_D,$$

where $M$ is the magnetization $\mathbf{M} = M(\sin \theta \cos \psi; \sin \theta \sin \psi, \cos \theta)$ with $\theta$ being the polar angle and $\psi$ being the azimuthal angle between each magnetic moment and the applied magnetic field $\mathbf{H}$ which points in the $z$ direction. $A$ is the exchange stiffness constant, $K$ is the uniaxial anisotropy constant, and $\mathbf{n}$ is the unity vector along uniaxial anisotropy axis. The DM energy functionals $w_D$ are composed of Lifshitz invariants $L_{\alpha\beta}^{(k)} = M_i \partial M_j / \partial x_k - M_j \partial M_i / \partial x_k$ [13] where $x$ is the spatial coordinate.

Denoting the distance from the skyrmion axis as $\rho$, skyrmions in the cone phase approach the solutions for the cone phase [15,16] ($\psi, \psi_c$) at large distances from the axis [6]:

$$\theta_{\rho \rightarrow \infty} = \theta_c = \arccos (H / H_C), \psi_{\rho \rightarrow \infty} = \psi_c = 2\pi \zeta / L_D,$$

where $H_C = H_D(1 - K / K_0)$ is the magnetic field above which the saturated state forms and $H_D = D^2 M / (2A)$. The pitch of
the critical line of mechanical grinding and argon-ion polishing on the (110) face. Lorentz transmission electron microscopy (LTEM) was conducted using an FEI Tecnai F20 electron microscope and the sample cooled in situ using a liquid-helium cooled holder with a base temperature of 10 K. Skyrmions appear as black or white circles in the images produced using this technique. The images also show white linear features which are not magnetic but are parts of the specimen surface which had peeled off and rolled into tiny tubes. We have not encountered this with the preparation of other materials. (See the Supplemental Material for full experimental methods [17].)

IV. EXPERIMENTAL RESULTS

Figure 2 shows frames from one of several videos we acquired (see the Supplemental Material [17]) showing two skyrmion clusters (outlined in red) embedded in a host phase that produced no contrast in the image. At lower fields, the skyrmion lattice filled the whole sample. The smaller cluster contained seven skyrmions and the larger, 13. There does not appear to be a preferred number of skyrmions in a cluster and in other videos we observed a single skyrmion as well as clusters with 6, 18, and 21 skyrmions. The fact that the skyrmions form clusters demonstrates that the interaction between them must be attractive and later we show theoretically that skyrmions embedded in the cone phase can be attractive whereas skyrmions embedded in the saturated state are purely repulsive.

Figs. 3(a)–3(c) were taken under the same conditions from a different region of the sample and show a cluster of 30 skyrmions moving across the host phase and merging with the skyrmion lattice phase over 21 s. The boundaries of the cluster and the edge of the skyrmion lattice phase are delineated by red lines and by comparing panels (b) and (c), it can be seen that the phase boundary advances after merging and that the skyrmions in the cluster have spread evenly across the boundary.
in magnetic field will first create the cone phase and any coexistence should be between these two phases. Furthermore, coexisting phases like those we see here can only result from a first order transition whereas a transition from the skyrmion lattice to the saturated state should be a second order process which occurs via the gradual expansion of the period of the skyrmion lattice [4,20].

In all of the videos we recorded, the skyrmions were in constant motion as shown in Figs. 2(b)–2(d). Similar motion has been reported by Mochizuki et al. [22] who attribute this to the heating of the specimen by the electron beam. We suggest instead that it may be caused by the specimen charging under the electron beam as coating the sample in a thin layer of carbon to improve its electrical conductivity slowed the movement of the skyrmions. Such charging can occur if the sample is insulating, like Cu₂OSeO₃, or not well grounded. Mochizuki et al. calculated that a steady flow of electrons through the sample from the electron beam was three orders of magnitude too low to move skyrmions via the spin-torque effect, but it is possible that bursts of current caused by the specimen charging and discharging may be sufficient to cause this movement.

Areas with the cone and skyrmion lattice phases are separated by domain walls. The calculated contour plots of such a domain wall is presented in Fig. 3(e). The frontal parts of skyrmion cores in the wall have a similar structure as those in nonaxisymmetric skyrmions and play the role of nuclei for individual nonaxisymmetric skyrmions. The attractive interaction between such skyrmions [13] explains the formation of skyrmion clusters observed in our experiments.

\[ e(\rho) = (2\pi L_D)^{-1} \int_0^{L_D} dz \int_0^{2\pi} d\varphi w_z(\theta, \psi) \]  (3)

are plotted as functions \( \Delta e(\rho) = (e(\rho) - e_{\text{cone}}) \) for different values of the applied field (Fig. 4) where \( w_z(\theta, \psi) \) is the energy density [Eq. (1)] for an isolated nonaxisymmetric skyrmion and \( e_{\text{cone}} \) is energy density [Eq. (3)] calculated for the cone phase [Eq. (2)]. It should be noted that as the host cone phase and embedded nonaxisymmetric skyrmions are modulated along the film thickness \( L \), the skyrmion energy density [Eq. (3)] and Fig. 4(e)] and the interskyrmion coupling depend on the confinement ratio, \( v = L/L_D \). These subtle effects could be a subject of future theoretical and experimental investigations.

The characteristic lengths \( R_1, R_2, R_3 \) indicate several distinct regions in the radial energy density profiles \( \Delta e(\rho) \) [Figs. 4(c)–4(e)]. The functions \( R_i(h) \) are plotted in Fig. 4(d). For axisymmetric skyrmions [Fig. 4(c)], the energy densities
is enclosed by positive “shells” of the applied field. In the cone phase (a), (b) the skyrmion core extended shell density for axisymmetric skyrmions (interskyrmion potential. Negative asymptotics of the radial energy $e_{\text{skyrmion-skyrmion interaction}}$ (c). (e) The contour plot energy ($\Delta e$) have been also observed in MgB$_2$ and Sr$_2$RuO$_4$ [30–33]. The attractive skyrmions in the cone phase of noncentrosymmetric ferromagnets represent an alternative to solitons with the oscillatory interparticle potential investigated in Refs. [23–26,28,29].

In conclusion, we report direct observations of clusters of attractive skyrmions embedded in the cone phase of a noncentrosymmetric ferromagnet. The clusters were generated by the magnetic-field-induced fragmentation of the skyrmion lattice during the first-order transition to the cone phase. This investigation used Cu$_2$OSeO$_3$, but the same method could be used to investigate skyrmion clusters in the cone phases of other noncentrosymmetric ferromagnets.

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**REFERENCES**


