Tunable room temperature magnetic skyrmions in centrosymmetric kagome magnet Mn$_4$Ga$_2$Sn

Dola Chakrabartty$^1$, Sk Jamaluddin$^1$, Subhendu K. Manna$^1$ & Ajaya K. Nayak$^{1,2}$

The successful realization of skyrmion-based spintronic devices depends on the easy manipulation of underlying magnetic interactions in the skyrmion-hosting materials. Although the mechanism of skyrmion formation in non-centrosymmetric magnets is comprehensively established, the stabilization process of different skyrmion-like magnetic textures in centrosymmetric magnets needs further investigation. Here, we utilize Lorentz transmission electron microscopy study to report the finding of a tunable skyrmion lattice up to room temperature in a centrosymmetric kagome ferromagnet Mn$_4$Ga$_2$Sn. We demonstrate that a controlled switching between the topological skyrmions and non-topological type-II magnetic bubbles can be realized at the optimal magnetic anisotropy. We find that the topological skyrmions are the energetically most stable magnetic objects in the centrosymmetric hexagonal magnets, whereas application of in-plane magnetic field stabilizes type-II magnetic bubbles as an excited state. The present study is a significant step towards understanding of the skyrmion stabilization mechanism in centrosymmetric materials for their future applications.

$^1$School of Physical Sciences, National Institute of Science Education and Research, HBNI, Jatni, 752050 Bhubaneswar, India. $^2$Center for Interdisciplinary Sciences (CIS), National Institute of Science Education and Research, HBNI, Jatni, 752050 Bhubaneswar, India. *Email: ajaya@niser.ac.in
Magnetic skyrmions are topologically protected nontrivial chiral spin configurations that can move in a small cut-off current by avoiding defects in their path\(^1\)–\(^3\), and hence are considered as excellent candidates for the future high density based racetrack memory devices\(^4\). Skyrmions have been mostly observed in the non-centrosymmetric chiral magnets\(^5\)–\(^9\) and multilayer thin films\(^10\)–\(^12\), where the competition between the Dzyaloshinskii-Moriya interaction (DMI) and the exchange interaction plays a significant role in stabilizing the underlying spin textures. In recent times, skyrmion-like swirling spin textures with various topological numbers are found in certain centrosymmetric magnets with uniaxial magnetocrystalline anisotropy (UMA)\(^13\)–\(^19\). In these magnets, skyrmion-like spin textures can be obtained as a result of competing dipolar energy and the magnetic anisotropy. As the dipolar energy is one of the most important energy contributions towards the formation of skyrmions in centrosymmetric magnets, an easy control over the shape, size, chirality as well as the topological charge of the skyrmions can be achieved by tuning the magnetization and thickness of the sample.

Along with the observation of skyrmionic bubbles, the finding of magnetic biskyrmions, which represent an addition of two skyrmions with opposite helicity, have also been reported in several centrosymmetric magnets\(^14\)–\(^19\). A summary of different spin textures reported in the centrosymmetric magnets is depicted in Fig. 1. The 3D spin texture, as well as 2D schematics of the in-plane magnetization distribution of different possible magnetic bubbles are shown in Fig. 1a–h. The Lorentz transmission electron microscopy (LTEM) simulated images of the spin textures are shown in Fig. 1i–k and the corresponding experimental LTEM patterns are depicted in Fig. 1l–n. The skyrmions with opposite helicity exhibit reverse black and white ring arrangement, whereas the type-II bubbles display alternative white and black half circles, as shown in Fig. 1k. It has been shown that the conventional type-II bubbles (Fig. 1c) with topological charge zero can exhibit similar LTEM magnetic contrast like the biskyrmions (Fig. 1d) when viewed from a certain angle with respect to their axis\(^20\),\(^21\). Although there exists few recent reports highlighting the skyrmion to bubble transformation, we have carried out a systematic study on the aforementioned phenomenon in a new skyrmion hosting hexagonal kagome ferromagnet Mn\(_4\)Ga\(_2\)Sn with skyrmion size of about 100 nm. We have mainly utilized the low temperature LTEM imaging technique to demonstrate a switching mechanism between the chiral skyrmions and the nonchiral type-II bubbles by systematically tilting the sample in different directions to provide a controlled in-plane magnetic field. In particular, we have shown that the stable magnetic skyrmions can be converted into the metastable type-II bubbles or vice versa by applying an in-plane magnetic field, as schematically depicted in Fig. 1o.

Results and discussion

Mn\(_4\)Ga\(_2\)Sn crystallizes in Fe\(_{0.5}\)Ge\(_{0.5}\)-type hexagonal structure (space group P\(_6_3\)/mmc) with alternative stacks of Mn-Sn and Mn-Ga-Sn atomic layers arranged along the c-axis. A detailed analysis of the structural information is given in the “Supplementary Note 1”. We have performed high-resolution transmission electron microscopy (HRTEM) imaging on our thin plate sample as used for the LTEM study (Fig. 2a). The hexagonal unit cell formed by alternative Mn-Sn atoms is marked with a white box in the inset of Fig. 2a along with the corresponding crystal structure. Furthermore, the selected area electron diffraction (SAED) pattern shown in Fig. 2b confirms the [001] orientation of our TEM lamella. The sample exhibits a Curie temperature \(T_C\) of \(\sim 320\) K and an additional spin reorientation transition \(T_{SR}\) at about 85 K (see “Supplementary Note 2”). Field dependent isothermal magnetization, \(M(H)\), measurements are carried out at different temperatures to further access the magnetic state of our polycrystalline Mn\(_4\)Ga\(_2\)Sn sample (Fig. 2c). A large saturation magnetization of about 8.58 \(\mu_B\)/f.u. is found at 10 K. A close look at the low field regime of the \(M(H)\) curves measured at

![Fig. 1 Different types of skyrmion-like spin textures in centrosymmetric magnets.](https://example.com/fig1.png)

**Fig. 1 Different types of skyrmion-like spin textures in centrosymmetric magnets.** Magnetic spin texture of (a) skyrmion with counter-clockwise (CCW) helicity, (b) skyrmion with clockwise (CW) helicity, (c) type-II bubble, and (d) biskyrmion. (e–h) Schematics of the arrangement of in-plane magnetization components for the spin textures corresponding to (a–d), respectively. The black solid arrows in (e–h) represent the possible direction of electron deflection in Lorentz transmission electron microscopy (LTEM). (i–k) The LTEM simulated images corresponding to the schematics in (e–g). (l–n) Experimental LTEM images of the simulated LTEM pattern in (e–g). The dotted black arrows from (h) to (k) and (n) point that the biskyrmion spin structure can also give rise to the same kind of LTEM contrast to that of type-II bubble. Scale bars shown in the images (i–n) correspond 100 nm. (o) Schematic diagram showing that both skyrmion and type-II bubble can exist in a material with skyrmion being the lowest energy state. Application of a small in-plane magnetic field can destabilize the skyrmion and nucleate type-II bubble as the system lacks any particular chirality.
$T > T_{SR}$ reveals the presence of kink kind of features that signify the existence of field induced magnetic phase transition in the system. This transition like characteristic can be clearly seen from the first-derivative of the $M(H)$ curves plotted in the inset of Fig. 2c. A similar type of transition anomaly has also been found in different skyrmion hosting materials\textsuperscript{9,22,23}. For further verification of the observed transitions in the $M(H)$ measurements, we have carried out magnetic field dependent ac susceptibility measurements $\chi'(H)$ at various temperatures from $T = 10$ K to $T = 300$ K as depicted in Fig. 2d. The $\chi'(H)$ data taken at 10 K exhibit a typical ferromagnetic like feature, whereas the measurement at 100 K shows an additional hump-like anomaly that persists for the $\chi'(H)$ data measured at 200 K and 300 K. It is important to mention here that the presence of transition like anomaly in the $\chi'(H)$ data has been extensively used as a tool to indirectly probe the skyrmion phase in several skyrmion hosting materials\textsuperscript{16,24–28}. Hence, the magnetic phase transitions found in the isothermal magnetization as well as in the ac susceptibility measurements suggest the existence of a possible skyrmion phase above $T_{SR}$ in the present system.

Motivated by the signature of field induced magnetic phase transitions in $M(H)$ and $\chi'(H)$ data, we have performed an extensive real space LTEM imaging study at different temperatures. Figure 3a–d shows over-focused LTEM images recorded at 100 K by increasing the magnetic field from 0 T to 0.4 T. The presence of spontaneous stripe domains with an average period of about 100 nm are found at zero magnetic field (Fig. 3a). With increasing the fields, the stripe domains start to break into magnetic bubble like textures at a field of about 0.3 T (Fig. 3b) and a lattice state is achieved at $\approx 0.35$ T (Fig. 3c). These magnetic textures start to disappear around 0.4 T (Fig. 3d) and a field polarized state is obtained at $\mu_0 H = 0.5$ T. The over-focused LTEM images taken at $T = 200$ K and 250 K for $\mu_0 H = 0.32$ T are depicted in Fig. 3e, f. The field evolution of the magnetic states for these temperatures are shown in the “Supplementary Note 3”. As it can be seen from Fig. 3e, a new type of magnetic contrast (shown inside the solid box) appears along with the magnetic textures found at 100 K. By further increasing the temperature to 250 K, the emergence of additional magnetic contrast (shown in the dotted boxes) can be noticed together with the magnetic states found at 200 K (Fig. 3f). Furthermore, the density of the newly formed magnetic textures greatly increases with increasing temperatures. A close look at the additional magnetic textures (shown inside the boxes) formed at 200 K and 250 K reveals the typical LTEM contrast of skyrmions as previously shown in Fig. 1l, m. Similarly, the magnetic patterns found at 100 K correspond to that of a type-II bubble (as shown in Fig. 1n).

It is important to understand the stabilization mechanism that governs the formation of different magnetic states in our system. In order to avoid bending contours during the LTEM experiment in the present case, all the images shown in Fig. 3a–f are taken by tilting the sample at a certain angle from the zone-axis ($\alpha = 10^\circ$, $\beta = 30^\circ$, where $\alpha$ and $\beta$ represent the sample tilting along x-axis and y-axis, respectively, which lie in the $ab$ plane of the sample). As a result, a small in-plane magnetic field always acts along the basal plane of the sample in addition to the out-of-plane magnetic field. In this tilting condition, we find the existence of type-II bubble-like magnetic contrast at 100 K. Interestingly, similar tilting condition at higher temperatures also gives rise to the finding of skyrmions with opposite helicity along with type-II bubbles. The existence of strong bending contours at low temperatures up to 200 K restrict us to perform the LTEM imaging along the zone axis. However, the bending contour effect improves considerably at 250 K. To examine whether a non-zero in-plane magnetic field...
affects the nucleation of the observed magnetic textures, we have recorded LTEM images at nearly zero tilting condition that makes sure the absence of any in-plane magnetic field in the sample. Fig. 3g–l show the over-focused LTEM images recorded by increasing and decreasing the out-of-plane magnetic fields at 250 K. Stripe domains with an average periodicity of 80 nm are observed as magnetic ground state (Supplementary Fig. 11). With increasing the magnetic fields, the stripe domains first start to shrink before skyrmion bubbles appear at a field of 0.15 T. Further increasing the field to 0.3 T helps in the formation of a hexagonal skyrmion lattice consisting of both CW and CCW spin rotation (marked with a hexagon in Fig. 3g). An applied field of 0.4 T leads to the survival of a few scattered skyrmions with a reduced size (Fig. 3h). Figure 3i–l show the evolution of skyrmion lattice with decreasing the magnetic fields starting from the saturated state. As it can be seen, a hexagonal skyrmion lattice can be nucleated upon decreasing the field to 0.25 T. Further decreasing the field to 0.1 T enables a mixed skyrmion and stripe domain textures, which remain as a remanent magnetic state at zero magnetic field.

Our LTEM imaging carried out with nearly zero tilting angle categorically demonstrates that the skyrmions are the energetically favorable magnetic textures in the centrosymmetric magnets. The type-II magnetic bubbles are stabilized when a small in-plane magnetic field is applied to the sample. For a better comprehension of the mechanism driving the nucleation of skyrmions/bubbles with in-plane magnetic fields, we have performed the LTEM imaging with controlled tilting conditions, as shown in Fig. 4. The in-plane magnetic field is applied by tilting our sample around two orthogonal axes by angle $\alpha$ and $\beta$, as shown schematically in Fig. 4a. When the sample is tilted by $\alpha$ and $\beta$, it produces an in-plane magnetic field $H_{IP}$ along the $y$ and $x$ directions, respectively. As it can be seen from Fig. 4b, the over-focused LTEM image taken at $T=250$ K in zero tilting condition and applied out-of-plane magnetic field of 0.2 T shows a hexagonal lattice of skyrmions with both helicities. Introducing a 6° $\alpha$ tilting results in the formation of type-II bubbles as depicted in Fig. 4c. A close look at the individual bubble shown in the zoomed view of Fig. 4c reveals that the deformation of the magnetic pattern exactly appears in the applied in-plane field direction ($y$–axis). By making the tilting angle to zero, we can stabilize the skyrmion state as before (see “Supplementary Note 3”). Now, by reversing the tilting direction, i.e., -6° $\alpha$ tilting, the observed LTEM intensity contrast reverses its pattern as can be seen from Fig. 4d and its zoomed view. This reversal of the LTEM intensity represents a flipping of the in-plane magnetization direction of the underlying spin texture. Further introducing a rotation of the in-plane magnetic field, i.e., applying a 6° $\beta$ tilting, rotates the LTEM magnetic contrast by 90° as shown in Fig. 4e and its zoomed view. These findings establish that the in-plane field is the deciding factor for the internal magnetic structures of the observed magnetic textures.

To further support our experimental results, we have performed micromagnetic simulations using Object Oriented Micromagnetic Framework (OOMMF) by applying in-plane magnetic fields in different directions. The details of the simulation can be found in the “Supplementary Note 4”. Figure 4f shows one of the simulated spin textures obtained by applying 0.2 T out-of-plane magnetic field. The ring type pattern obtained from the LTEM simulation, shown in the inset of Fig. 4f, corresponds to a Bloch-type skyrmion as observed experimentally. Now, we apply a small in-plane magnetic field of 0.02 T along the $\pm y$ – directions in addition to the out-of-plane magnetic field.

---

**Fig. 3** Over-focused Lorentz transmission electron microscopy (LTEM) images of magnetic textures evolved with magnetic fields and temperatures for Mn$_2$Ga$_2$Sn. a–d LTEM images of the magnetic states recorded with increasing external magnetic field from 0 T to 0.4 T at 100 K. e, f LTEM images of the magnetic domain evolution at magnetic field 0.32 T for temperatures 200 K and 250 K. The solid and dotted boxes represent skyrmionic textures with two different helicities. All the images in (a–f) are taken at nearly 4 degrees of sample tilting condition. g–l The field evolution of magnetic skyrmions at 250 K taken with increasing and decreasing the out-of-plane magnetic field at nearly zero tilting condition. The hexagonal skyrmion lattices are marked with the dotted hexagon in (g, i). The scale bar shown in the images corresponds to 500 nm. $H$ corresponds to the magnetic field and $\mu_0$ is the permeability of vacuum.
of 0.2 T. The simulated spin textures resemble that of the type-II bubble as depicted in Fig. 4g, h. It can be clearly seen that the observed spin textures exhibit an elongation along the applied in-plane field direction by breaking the clockwise/counter-clockwise spin rotation of the Bloch-type skyrmion. The LTEM simulated patterns shown in the insets of Fig. 4g, h fully match with that of the experimental spin textures obtained for the α rotation given in Fig. 4c, d. When the in-plane magnetic field is changed to the x-direction, the elongation of the spin in the field direction follows the field direction (Fig. 4i). As expected, the simulated LTEM pattern shown in the inset of Fig. 4i replicates the experimental spin texture observed in Fig. 4e. Hence, our experimental results along with the micromagnetic simulations, categorically establish that the magnetic textures obtained by application of small in-plane magnetic fields are indeed type-II bubbles.

As it can be found from Fig. 3, for a given in-plane magnetic field, the probability of skyrmion nucleation is higher at the higher temperatures, whereas type-II bubbles get easily nucleated at lower temperatures. This observation suggests that the effect of the in-plane magnetic field is more prominent at lower temperatures. It has been reported that Mn₄Ga₂Sn undergoes a phase transition from the high temperature easy-axis to easy-plane anisotropy at the Tₕₕₜₜₖₕ = 29,30. Since the magnetic anisotropy plays a major role in stabilizing skyrmions in centrosymmetric magnets, we have calculated the effective uniaxial anisotropy constant (Kₑₑₑₑₙₙₑₑₑₑ) for Mn₄Ga₂Sn from T = 70 K to T = 300 K based on the law of approach to saturation31 (see “Supplementary Note 5”). As it can be found from Fig. 5, the effective anisotropy exhibits its lowest value near the Tₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚ₢
Stabilization of different kinds of topological spin textures in the centrosymmetric materials have drawn a great attention because of their high degrees of freedom pertaining to use as multiple bits in data storage devices. Recently, nanometric skyrmions of size about 1–2 nm size are observed in a few centrosymmetric systems, such as triangular lattice magnet Gd₃PdSi₃₁,₂₃, breathing kagome magnet Gd₃Ru₄Al₁₂,₃₂ and tetragonal GdRu₂Si₂₃. The mechanism of skyrmion formation in these systems is attributed to the presence of geometrical frustration and four-spin exchange interaction. In contrast, the skyrmions stabilized by competing dipolar and uniaxial magnetic anisotropy on other centrosymmetric systems are comparatively larger in size. However, skyrmions stabilized by geometrical frustration and higher order magnetic interactions are found to exist at very low temperatures in comparison to the room temperature skyrmions in other centrosymmetric skyrmion hosting materials. Even though there are few reports of room temperature skyrmions in the centrosymmetric magnets, the size of the skyrmions in these materials happens to be greater than 200 nm³.¹³,₁⁶ In contrast, the present system hosts near room temperature skyrmions with size of about 100 nm. Besides the LTEM observations, the topological Hall effect (THE) measurements have also frequently been used as a signature for the skyrmion phase existence.⁵,₂₃ In general, the THE is inversely related to the size of the skyrmions. For the present skyrmions with size of 100 nm, the topological Hall resistivity should be in the order of nano-ohm-cm. The observation of THE in micro-ohm-cm order in the present system signifies that the THE mostly originates from the non-coplanar spin structure of the sample [see "Supplementary Note 6"].

Recently, few reports show the transformation between topologically nontrivial skyrmions and trivial type-II bubbles in the centrosymmetric system.⁴,³⁴,³⁵ The present study makes a distinct effort to understand the underlying mechanism of skyrmion and bubble formation in centrosymmetric magnets. Similar kind of topological transition, e.g. antiskyrmion to non-topological bubble transformation, have also been experimentally observed in case of D₃d symmetry systems,⁵⁶ where the spin helix only stabilizes along the [100] or [010] direction. The anisotropic DM interaction (Dₓ = −Dᵧ) in these systems dictates the helicity reversal of the antiskyrmions. Hence, the in-plane magnetic anisotropy in the D₃d skyrmion hosting system is only determined by the DM interaction. In contrast, there is no preferential in-plane anisotropy in case of the centrosymmetric materials. The transformation between the topological skyrmions and the non-topological magnetic bubbles occurs due to nearly degenerate energies or small energy barrier between these spin textures. With the application of in-plane magnetic field, the Zeeman energy gain initiates the topological transformation easily. Hence, in the case of centrosymmetric system application of in-plane magnetic field in any direction gives rise to two Bloch lines with same in plane configuration. This results in the transformation of chiral skyrmion into achiral type-II bubble. When the in-plane magnetic field is removed the Bloch lines again disappear, enabling the transformation of achiral type II bubble to skyrmion. The reversal of spin textures with the applied in-plane magnetic field

![Fig. 5 Dependency of skyrmion density on effective magnetic anisotropy.](image)

Temperature variation of effective magnetic anisotropy constant (Kₐₐ₈) and number of skyrmions (nSKX) stabilized in tilted magnetic field in each 4 μm² area of the sample plate. The error bars in the Kₐ₈ are calculated from the standard deviation of the fitting parameters. The isolated skyrmion numbers up to 250 K are calculated for a field of 0.32 T, whereas at 280 K, the number is taken for the field 0.2 T. The inset shows the number of simulated skyrmions (nSKXsimulated) in constant tilted magnetic field by varying the magnetic anisotropy (detailed simulation is given in the supplementary note 5).

![Fig. 6 Phase diagrams.](image)

(a) H – T phase diagram constructed using the magnetization data of polycrystalline Mn₄Ga₂Sn sample. The critical fields for constructing the phase diagram are taken from the point of anomaly in the M(H) measurements (filled circles) and ac susceptibility data (filled squares). (b) H – T phase diagram for Mn₄Ga₂Sn, as deduced from the temperature and field dependent Lorentz transmission electron microscopy (LTEM) measurements of the sample at nearly 4 degree tilting condition. The color contrasts represent the average numbers of skyrmions (nSKX) and type-II bubbles in each μm² area. The asterisk symbols correspond to the point (H and T) where images are recorded. H corresponds to the magnetic field and μ₀ is the permeability of vacuum. SKX and FP correspond to skyrmion and field polarized, respectively.
should be universal for all centrosymmetric skyrnion hosting materials where skyrmions stabilized by competing dipolar and uniaxial magnetic anisotropy. However, the magnitude of in-plane magnetic field may differ from system to system depending on the strength of the magnetic anisotropy in the system. Here, we show that the magnetic anisotropy of the system decides the magnitude of energy barrier for the transformation between the topological skyrmions and the non-topological magnetic bubbles. We systematically vary the in-plane magnetic field in different directions, both in our LTEM measurements and micromagnetic simulations, to demonstrate the mechanism of skyrmion ↔ bubble transformation. All our measurements categorically establish a direct correlation among the probability of skyrmion formation, in-plane magnetic field and magnetic anisotropy of the system.

**Conclusion**

In conclusion, we have carried out an extensive LTEM measurement over a wide temperature range to uncover the possible spin textures in the centrosymmetric kagome magnet Mn₄Ga₂Sn. In the case of LTEM imaging, it is extremely difficult to record magnetic contrast at the crystal zone axis due to the presence of bending contours. Hence, the application of an in-plane magnetic field by a small tilting of the sample to avoid the bending contour can transform the skyrmionic state to the type-II bubble magnetic state. We have also discussed the correlation between the skyrmion stabilization and change in the uniaxial anisotropy that plays an important role in skyrmion formation in these systems. Although skyrmions have been observed and well studied in many non-centrosymmetric systems, the skyrmion hosting centrosymmetric magnets having higher degrees of freedom to manipulate their magnetic states deserve a thorough study for the realization of skyrmion based memory devices.

**Method**

**Sample preparation and structural characterization.** Polycrystalline ingots of Mn₄Ga₂Sn were prepared using high pure Mn, Ga, Sn metals in the argon atmosphere by using an arc melting furnace. For further homogeneity, the ingots were sealed in a quartz tube in the argon atmosphere and annealed at 823 K for ten days. The compositional homogeneity was verified using field emission scanning electron microscopy (FESEM) and energy-dispersive x-ray spectroscopy (EDX). The crystal structure of the sample was verified by powder x-ray diffraction (XRD) measurements performed using Rigaku smartlab diffractometer with CuKα radiation.

**Micromagnetic measurements.** The DC magnetic measurements of the sample were performed by utilizing Superconducting Quantum Interference Device Vibrating Sample Magnetometer (SQUID-VSM, Quantum design). The ac-susceptibility measurements were carried out by using the measurement ACMS option in Physical Properties measurement System (PPMS, Quantum design).

**Lorentz transmission electron microscopy (LTEM) study.** For the LTEM measurements, thin sample platelets of Mn₄Ga₂Sn with [001] orientation were cut from the polycrystalline sample using a Ga-based focused ion beam (FIB). The magnetic domains were studied using JEOL TEM (JEM-F200) in the Lorentz-TEM mode. A double tilting liquid nitrogen holder (GATAN-636) was used to study the temperature evolution of magnetic domains.

**Micromagnetic simulations.** Micromagnetic simulations were carried out using Object Oriented Micromagnetic Framework (OOMMF) code. Slab geometry of dimensions 1000 nm × 1000 nm × 100 nm were used for the simulation, with a rectangular mesh of 4 nm × 4 nm × 4 nm. The material parameters were chosen according to the experimental data of Mn₄Ga₂Sn. Exchange stiffness constant (A) was calculated using the formula $A = K_p T_C / a$, where $K_p$ is the Boltzmann constant, $T_C$ is the Curie temperature and $a$ is the lattice constant of the sample. The estimated value of the exchange constant $A$ is about $5 \times 10^{-12}$ J/m. The out-of-plane easy axis magnetocrystalline anisotropy ($K_u$) at 250 K was calculated by using the formula $D = \pi A / K_u$, where $D$ is the domain wall width observed in the LTEM experiment. The estimated value of magnetocrystalline anisotropy constant $K_u$ is about $1.02 \times 10^5$ J/m³. The equilibrium states were obtained by fully relaxing the randomly distributed magnetization. The simulations for the magnetic field dependent domain evolution for 250 K and 100 K were conducted at zero temperature with the experimental parameters are consistent with the experimental results.

**Data availability**

All data used to obtain the conclusions in this paper are presented in the paper and/or the Supplementary Materials. Other data may be requested from the authors. Please direct all inquiries to A.K.N. (ajaya@niser.ac.in).

**References**

1. Jonietz, F. et al. Spin transfer torques in MnSi at ultralow current densities. *Science* **330**, 1648–1651 (2010).
2. Schulz, T. et al. Emergent electrodynamics of skyrmions in a chiral magnet. *Nat. Phys.* **8**, 301–304 (2012).
3. Yu, X. et al. Skyrmion flow near room temperature in an ultralow current density. *Nat. Commun.* **3**, 988 (2012).
4. Nagaosa, N. & Tokura, Y. Topological properties and dynamics of magnetic skyrmions. *Nat. Nanotech* **8**, 899–911 (2013).
5. Mühlbauer, S. et al. Skyrmion lattice in a chiral magnet. *Science* **323**, 915–919 (2009).
6. Yu, X. et al. Real-space observation of a two-dimensional skyrmion crystal. *Nature* **465**, 901–904 (2010).
7. Adams, T. et al. Long-wavelength helimagnetic order and skyrmion lattice phase in Cu₃OSeO₃. *Phys. Rev. Lett.* **108**, 237204 (2012).
8. Tokunaga, Y. et al. A new class of chiral materials hosting magnetic skyrmions beyond room temperature. *Nat. Commun.* **6**, 7638 (2015).
9. Kézsmárki, I. et al. Néel-type skyrmion lattice with confined orientation in the polar magnetic semiconductor GaV₃S₄. *Nat. Mater.* **14**, 1116–1122 (2015).
10. Jiang, W. et al. Blowing magnetic skyrmion bubbles. *Science* **349**, 283–286 (2015).
11. Soumyanarayanan, A. et al. Tunable room-temperature magnetic skyrmions in Ir/FeCo/Pt multilayers. *Nat. Mater.* **16**, 898–904 (2017).
12. Kwon, H. Y. et al. High-density Néel-type magnetic skyrmion phase stabilized at high temperature. *NPJ Quantum Mater.* **2**, 18009 (2017).
13. Yu, X. et al. Magnetic stripes and skyrmions with helicity reversals. *Proc. Natl Acad. Sci.* **109**, 8856–8860 (2012).
14. Yu, X. Z. et al. Biskyrmion states and their current-driven motion in a layered manganese. *Nat. Commun.* **5**, 3198 (2014).
15. Wang, W. et al. A centrosymmetric hexagonal magnet with superstable biskyrmion magnetic nanodisks in a wide temperature range of 100–340 K. *Adv. Mater.* **28**, 6887–6893 (2016).
16. Hou, Z. et al. Observation of various and spontaneous magnetic skyrmionic bubbles at room temperature in a frustrated kagome magnet with uniaxial magnetic anisotropy. *Adv. Mater.* **29**, 1701144 (2017).
17. Yu, X. et al. Variation of topology in magnetic bubbles in a colossal magnetoresistive manganite. *Adv. Mater.* **29**, 1603958 (2017).
18. Xiao, X. et al. Low-field formation of room-temperature biskyrmions in centrosymmetric MnPdGa magnet. *Appl. Phys. Lett.* **114**, 142404 (2019).
19. Zuo, S. et al. Spontaneous topological magnetic transitions in NiCo5 rare-earth magnets. *Adv. Mater.* **33**, 2103751 (2021).
20. Yao, Y. et al. Magnetic hard nanobubble: a possible magnetization structure behind the bi-skyrmion. *Appl. Phys. Lett.* **114**, 102404 (2019).
21. Louden, J. C. et al. Do images of biskyrmions show Type-II Bubbles? *Adv. Mater.* **31**, 1806598 (2019).
22. Bauer, A. & Pfeiffer, C. Magnetic phase diagram of MnSn inferred from magnetization and ac susceptibility. *Phys. Rev. B* **85**, 214418 (2012).
23. Kurumaji, T. et al. Skyrmion lattice with a giant topological Hall effect in a frustrated triangular-lattice magnet. *Science* **365**, 914–918 (2019).
24. Thiesse, C. et al. Field dependence of the magnetic quantum phase transition in MnSn. *J. Phys. Condens. Matter* **9**, 6677 (1997).
25. Seki, S. et al. Observation of skyrmions in a multiferroic material. *Science* **336**, 198–201 (2012).
26. Nayak, A. K. et al. Magnetic antiskyrmions above room temperature in tetragonal Heusler materials. Nature 548, 561–566 (2017).

27. Jamaluddin, S. et al. Robust antiskyrmion phase in bulk tetragonal Mn-Pd-Sn Heusler system probed by magnetic entropy change and AC-susceptibility measurements. P. Adv. Funct. Mater. 29, 1901776 (2019).

28. Sen, S. et al. Observation of the topological Hall effect and signature of room-temperature antiskyrmions in Mn-Ni-Ga D2d Heusler magnets. Phys. Rev. B. 99, 134404 (2019).

29. Ihou-Mouko, H. Ph.D. thesis, University Henri Poincaré - Nancy I, Nancy, France, November, 2006.

30. Eichenberger, L. et al. Magnetic and magnetocaloric properties of Mn4−xFexGa2Sn. J. Appl. Phys. 116, 103909 (2014).

31. Andreev, S. V. et al. Law of approach to saturation in highly anisotropic ferromagnets Application to NdFeB melt-spun ribbons. J. Alloy. Compd. 260, 196 (1997).

32. Hirschberger, M. et al. Skyrmion phase and competing magnetic orders on a breathing kagomé lattice. Nat. Commun. 10, 5831 (2019).

33. Khanh, N. D. et al. Nanometric square skyrmion lattice in a centrosymmetric tetragonal magnet. Nat. Nanotechnol. 15, 444–449 (2020).

34. Wu, Y. et al. A strategy for the design of magnetic memories in bubble-hosting magnets. Appl. Phys. Lett. 118, 122406 (2021).

35. Wei, W. et al. Current-controlled topological magnetic transformations in a nanostructured Kagome Magnet. Adv. Mater. 33, 2101610 (2021).

36. eng, L. et al. Controlled transformation of skyrmions and antiskyrmions in a non-centrosymmetric magnet. Nat. Nanotechnol. 15, 181–186 (2020).

37. Donahue, M. J. & Porter, D. G. OOMMF Users Guide, Version 1.0, Interagency Report NISTIR, 2009.

38. McCray, A. R. et al. Understanding complex magnetic spin textures with simulation-assisted Lorentz transmission electron microscopy. Phys. Rev. Appl. 15, 044025 (2021).

Author contributions
A.K.N. conceived the original idea of the present work and supervised the project. D.C., S.K.M. synthesized the materials. S.J. prepared the TEM lamella and carried out related microstructural study. D.C. performed the LTEM, magnetic, and structural study and micromagnetic simulation. A.K.N. and D.C. wrote the manuscript.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s42005-022-00971-7.

Correspondence and requests for materials should be addressed to Ajaya K. Nayak.

Peer review information Communications Physics thanks the anonymous reviewers for their contribution to the peer review of this work. Peer reviewer reports are available.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Acknowledgements
AKN acknowledges the support from Department of Atomic Energy (DAE), the Department of Science and Technology (DST)-Ramanujan research grant (No. SB/S2/RJN-081/2016) and Nanomission research grant [SR/NM/NS-1036/2017(G)] of the Government of India.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2022