Uncovering the skyrmion bubble magnetic states in centrosymmetric kagome magnet Mn$_4$Ga$_2$Sn

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The successful realization of skyrmion based next generation spintronic devices greatly depends on the easy manipulation of underlying magnetic interactions in the skyrmion hosting materials. In this direction, although the mechanism of skyrmion formation in non-centrosymmetric magnets is comprehensively established, contradicting views on the stabilization process of skyrmion/bubble magnetic states in centrosymmetric magnets need further investigation. Here, we report the finding of a tunable skyrmion lattice up to room temperature in a new skyrmion hosting centrosymmetric kagome ferromagnet Mn$_4$Ga$_2$Sn. Using an extensive Lorentz transmission electron microscopy experiment at different temperatures, we demonstrate that both the Bloch-type skyrmions and type-II magnetic bubbles can be stabilized depending upon the nature of magnetic field excitation and strength of uniaxial magnetic anisotropy in the system. Most importantly, we illustrate a controlled switching between the topological skyrmions and the non-topological type-II magnetic bubbles by applying a small in-plane magnetic field. Our results categorically establish that the topological Bloch-type skyrmions are the energetically most stable magnetic objects in the centrosymmetric hexagonal magnets, whereas the type-II magnetic bubbles, instead of biskyrmions, are the excited magnetic states in the system. The present study is an important step towards the basic understanding of the skyrmion stabilization mechanism in centrosymmetric materials and paves the way for their future application in spintronics.

INTRODUCTION

Magnetic skyrmions are topologically protected nontrivial chiral spin configurations that can move in a small cut-off current by avoiding defects in their path$^4$, 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, such as cubic B20 compounds MnSi$^5$, Fe$_{1-x}$Co$_x$Si$^6$, CuxOSeO$_2$$^7$, $\beta_{mn}$ structured compound CoZnMn$^8$, and polar compound GaV$_3$S$_8$$^9$. In these materials, the competition between the Dzyaloshinskii–Moriya interaction (DMI) and the exchange interaction plays a significant role in stabilizing the skyrmion spin texture. Skyrmions have also been observed in multilayer thin films that in general host interfacial DMI$^{10,12}$. In recent times, skyrmion like whirling spin textures with various topological numbers have been found in certain centrosymmetric magnets that exhibit uniaxial magnetocrystalline anisotropy (UMA)$^{13,15,17,22,33}$. In these magnets, the dipolar energy prefers the in-plane arrangement of the spins, whereas the UMA favors the spins to align along the magnetic easy axis. The competition between these two energies gives rise to skyrmion like spin texture. In contrast to the chiral magnets, the skyrmions in centrosymmetric systems possess extra degrees of freedom in their helicity and vorticity$^4$. The helicity (vector chirality) that represents the rotational sense (clockwise or anti-clockwise) of the in-plane magnetization of the skyrmions is found to be tied with the DMI vector in case of the non-centrosymmetric magnets, whereas the absence of DMI in centrosymmetric systems is responsible for the realization of skyrmions with both helicities. On the other hand, the vorticity (winding number) of a skyrmion is related to the topological charge and can vary depending on the presence of Bloch lines (BLs) and their configuration$^{16,19}$. The high sensitivity of the BLs to the applied magnetic field can lead to the formation of skyrmionic bubbles with different topological numbers$^{17,19}$. The dipolar energy, which is one of the most important energy contributions for skyrmion formation in centrosymmetric magnets, can be easily manipulated by tuning the magnetization and thickness of the sample. This can enable us an easy control over the shape, size, chirality as well as the topological charge of the skyrmions in centrosymmetric materials.

In recent studies, skyrmionic bubbles with different internal spin configurations and topological charges have been experimentally found in centrosymmetric kagome ferromagnet Fe$_3$Si$_2$$^{17}$ and orthorhombic manganite La$_{1-x}$Sr$_2$MnO$_3$$^{18}$. Furthermore, the finding of magnetic biskyrmions, which represent an addition of two skyrmions with opposite helicity, have also been reported in several centrosymmetric magnets, e.g. tetragonal manganite La$_{2-x}$Sr$_{1+x}$Mn$_2$O$_7$$^{13}$, orthorhombic manganite La$_{1-x}$Sr$_2$MnO$_3$$^{19}$, hexagonal (Mn$_{1-x}$Ni$_x$)$_{65}$Ga$_{35}$ (x = 0.5)$^{15}$, MnPdGa$^{20}$, and NdCo$_5$$^{33}$. At the same time, few reports by Yao et al.$^{21}$ and Loudon et al.$^{22}$ have demonstrated that the conventional type-II bubbles with topological charge zero can exhibit similar Lorentz transmission electron microscopy (LTEM) magnetic contrast like the so-called biskyrmions when viewed from a certain angle with
FIG. 1: (Color online) Magnetic spin texture of (a) type-I bubble or skyrmionic bubble with counter-clockwise (CCW) helicity, (b) type-I bubble 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 LTEM. (i)-(k) The LTEM simulated images of the spin textures 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. (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.

respect to their axis. These conflicting reports about the skyrmion vorticities and/or topological numbers suggest that further study is required to understand the origin of the observed magnetic textures in centrosymmetric magnets. We have summarized all the possible magnetic textures found in the centrosymmetric magnets in Figure 1(a)-(n). The 3D spin texture, as well as 2D schematics of the in-plane magnetization distribution of different possible magnetic bubbles are shown in Figure 1(a)-(h). The LTEM simulated images of the spin textures are shown in Figure 1(i)-(k) and the corresponding experimental LTEM patterns are depicted in Figure 1(l)-(n). The skyrmionic bubbles (type-I bubbles) 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 Figure 1(k). The biskyrmion spin texture shown in Figure 1(d) needs further investigations, as contradicting reports are found on their experimental LTEM pattern. In this article, we have explored the presence of room temperature skyrmion lattice in a new centrosymmetric hexagonal kagome ferromagnet Mn$_4$Ga$_2$Sn. We have utilized the low temperature LTEM imaging technique to demonstrate a switching mechanism between the chiral skyrmions and the nonchiral type-II bubbles. In particular, we have experimentally demonstrated that the stable magnetic skyrmions can be converted into the metastable type-II bubbles with a finite in-plane magnetic field excitation, as schematically depicted in Figure 1(o).

RESULTS AND DISCUSSION

Mn$_4$Ga$_2$Sn crystallizes in Fe$_6$Ge$_4$-type hexagonal structure (space group P6$_3$/mmc) with alternative stacks of Mn-Sn and Mn-Ga-Sn atomic layers arranged along the c-axis as depicted schematically in Figure 2(a). The Mn atoms in the Mn-Sn layer form a kagome type of ordering with Sn sitting at the center of the hexagon, whereas the Mn in the Mn-Ga-Sn layer exhibits a breathing kagome lattice where Sn atoms sit alternatively at the center of the Mn triangles [Figure 2(b)]. For the present Mn$_4$Ga$_2$Sn sample, we have carried out room temperature powder x-ray diffraction (XRD) measurements (shown in supplementary Figure S1).
FIG. 2: (Color online) Structure and magnetic properties of Mn$_4$Ga$_2$Sn. (a) The crystallographic unit cell of Mn$_4$Ga$_2$Sn. (b) The top view (view from c-axis) of kagome lattice formed by Mn atoms in both Mn-Sn layer and Mn-Ga-Sn layers. (c) High resolution transmission electron microscopy (HRTEM) image taken on [001] oriented sample plate. The left inset shows the enlarged view of the atomic arrangement reconstructed by using the inverse fast Fourier transformation (iFFT) of the fast Fourier transformed HRTEM image. The right inset depicts the view of the atomic unit cell of Mn$_4$Ga$_2$Sn from the c-axis. (d) Room temperature selected area electron diffraction (SAED) pattern taken on the sample plate with [001] orientation. The inset shows the first derivative of the FC $M(T)$ curve $dM(T)/dT$. (e) Isothermal magnetization curves measured at different temperatures. The inset shows the first derivative of $M$ vs $H$ curves at selected temperatures. (g) ac susceptibility ($\chi'$) vs magnetic field ($\mu_0 H$) plots at different temperatures from 10 K to 300 K. The black arrows represent the anomalies corresponding to the magnetic transitions.

that confirm the single phase nature of our sample crystallizing in the hexagonal crystal structure with lattice parameter $a = b = 8.5527$ Å and $c = 5.3264$ Å, consistent with the earlier report [24]. To further verify the crystal structure, we have performed high-resolution transmission electron microscopy (HRTEM) imaging in our sample, as shown in Figure 2(c). The hexagonal unit cell formed by alternative Mn-Sn atoms is marked with a white box in the left inset of Figure 2(c) with the corresponding crystal structure shown in the right inset. Furthermore, the selected area electron diffraction (SAED) pattern shown in Figure 2(d) confirms the [001] orientation of our TEM lamella used for further study.

In order to map out the magnetic state of the present Mn$_4$Ga$_2$Sn sample, we have conducted temperature dependent dc magnetization measurements [$M(T)$] in zero field cooled (ZFC) and field cooled (FC) modes as shown in Figure 2(e). The ZFC and FC $M(T)$ curves measured in a field of 0.05 T almost trace on each other with a small hysteretic behavior for temperatures between 100 K and 300 K. As it can be found from the first derivative of the FC $M(T)$ curve with respect to temperature, shown in the inset of Figure 2(e), the sample exhibits a Curie temperature ($T_C$) of $\approx$ 320 K and an additional transition at about 85 K. This secondary transition at low temperature corresponds to a spin reorientation transition ($T_{SR}$), where the magnetic moments undergo a transition from easy-axis orientation at high temperatures to easy-plane alignment at the low temperatures [23, 24]. The effective anisotropy in the system continuously changes from negative (easy-plane) to positive value (easy-axis) with increasing temperatures. It is important to mention here that the mechanism of skyrmion formation in centrosymmetric magnets depends on the competing dipolar energy and easy axis magnetic anisotropy [15, 17, 19, 33].

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 [Figure 2(f)]. 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 $T > T_{SR}$ 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 Figure 2(d). A similar type of transition anomaly has also been found in many skyrmion hosting materials due to the first-order phase transition from the helical to skyrmion phase [9, 25, 26].

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 Figure 2(g). 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 [17, 27, 31]. 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 magnetic domain imaging using the LTEM technique. For this purpose, thin plates of Mn$_4$Ga$_2$Sn sample with [001] orientation are prepared using the focused ion beam (FIB). The required orientation of the TEM lamella is confirmed through the SAED pattern, as shown previously in Figure 2(d). The LTEM imaging in the present system is carried out from room temperature to cryogenic temperature ($T = 100$ K) using a liquid-N$_2$ TEM sample holder. Figure 3(a)-(d) shows over-focused LTEM images of the magnetic textures 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 [Figure 3(a)]. With increasing the fields, the stripe domains start to break into magnetic bubbles at a field of about 0.3 T [Figure 3(b)]. The sample completely transforms into the magnetic bubble lattice for a field of $\approx 0.35$ T, as shown in Figure 3(c). With further increasing the fields, the bubble domains start to disappear [Figure 3(d)], and finally a field polarized state is achieved at $\mu_0 H \approx 0.5$ T.

For a better understanding of the observed magnetic bubble domain state in the present sample, we have carried out LTEM imaging at different temperatures. The over-focused LTEM images taken at $T = 200$ K and 250 K for $\mu_0 H \approx 0.32$ T are depicted in Figure 3(e)-(f). The field evolution of the magnetic domain states for $T = 150$ K, $T = 200$ K, and $T = 250$ K are shown in the supplementary material (Figure S6, S7, S8). As it can be seen from Figure 3(e), a new type of magnetic contrast (shown inside the solid box) appears along with the magnetic bubble state 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 bubble states found at 200 K [Figure 3(f)]. Furthermore, the density of the newly formed magnetic textures greatly increases with increasing temperatures. To further access the nature of the observed LTEM contrasts over the temperatures, zoomed view of the magnetic texture at 100 K is shown in Figure 3(h). It looks like a circular pattern consisting of two incomplete circles of dark and bright contrast. A similar kind of LTEM intensity variation has also been found in other uniaxial anisotropy based centrosymmetric materials [14, 15, 17, 19, 22, 32, 33]. In some of the earlier reports, this magnetic texture is explained as a type-II bubble with topological number zero, as schematically shown in Figure 1(c) and (g) [13, 17, 21, 22]. Few other reports also claim that the given magnetic texture is composed of two magnetic skyrmions with opposite helicity, named as biskyrmion, resulting in a skyrmion number of two [schematically shown in the Figure 1(d) and (h)] [13, 15, 19, 32]. The zoomed view of the additional magnetic texture found at 200 K (enclosed in solid boxes) can be interpreted as a type-I magnetic bubble with spin rotation of a Bloch-skyrmion (Figure 1(m)). The ring like magnetic contrasts with reverse black and white arrangements depicted in Figure 1(h) and (m) correspond to the skyrmionic bubbles with opposite helicity with skyrmion number one.

It is very important to mention here that in order to avoid the bending contours during the LTEM measurements, all the images shown in Figure 3(a)-(f) are taken by tilting the sample at a certain angle from the zone-axis ($\alpha = 1^\circ$, $\beta = 3^\circ$, 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 always find the existence of type-II bubble or biskyrmion like magnetic contrast at 100 K. Interestingly, similar tilting condition at higher temperatures also gives rise to the finding of Bloch-skyrmion like magnetic contrast along with the magnetic textures at 100 K.

The existence of strong bending contours at low temperatures up to 200 K restrict us to perform the LTEM imaging completely 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 a nearly zero tilting condition where no in-plane magnetic field is present. Figure 3(g)-(l) show the over-focused LTEM
FIG. 3: (Color online) Over-focused LTEM images of magnetic textures evolved with magnetic fields and temperatures for Mn$_4$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-i) 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) and (i).

images of the magnetic textures 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 as shown in the supplementary Figure S9. 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 gives rise to the formation of a hexagonal skyrmion lattice consisting of both clockwise and counter-clockwise spin rotation [marked with a hexagon in Figure 3(g)]. An applied field of 0.4 T leads to the survival of a few scattered skyrmions with a reduced size [Figure 3(h)]. A field polarized state is achieved for fields greater than 0.4 T. Figure 3(i)-(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 Bloch-type skyrmions are the energetically favorable magnetic textures in the centrosymmetric magnets. The type-II magnetic bubble state is stabilized when the LTEM experiment is performed by a small tilting of the zone axis, thereby introducing some in-plane magnetic fields. For a better comprehension of the mechanism driving the nucleation of skyrmions/bubbles with in-plane magnetic fields, we have performed the LTEM measurements with controlled tilting conditions, as shown in Figure 4(b)-(e). The in-plane magnetic field is applied by tilting our sample around two orthogonal axes by angle α.
FIG. 4: (Color online) Evolution of magnetic domains with controlled tilting condition at an applied magnetic field \( H_{AP} \) of 0.2 T and temperature \( T = 250 \) K. (a) Schematic diagram of the sample orientation to describe the tilting geometry. (b)-(e) The over-focused LTEM images taken at different tilting angles. (f)-(i) The OOMMF simulated magnetic structures with different in-plane magnetic field directions along with a fixed out-of-plane magnetic field of 0.2 T. The insets of figure (f)-(i) represent the LTEM simulated images corresponding to the OOMMF images. The extended figures corresponding to (f)-(i) are shown in the supplementary material.

and \( \beta \), as shown schematically in Figure 4(a). 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 Figure 4(b), the over-focused LTEM image taken at \( T = 250 \) K in zero tilting condition and an applied out-of-plane magnetic field of 0.2 T shows a hexagonal lattice of skyrmions with both helicities. Introducing a 6\(^\circ\) \( \alpha \) tilting results in the formation of type-II bubbles as depicted in Figure 4(c). A close look at the individual bubble shown in the zoomed view of Figure 4(c) reveals that the deformation of the magnetic pattern exactly appears in the applied in-plane field direction (\( y \)-axis). Now, by reversing the tilting direction, i.e., -6\(^\circ\) \( \alpha \) tilting, the observed LTEM intensity contrast reverses its pattern as can be seen from Figure 4(d) and its zoomed view. This reversal of the LTEM intensity contrast 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\(^\circ\) \( \beta \) tilting, rotates the LTEM magnetic contrast by 90\(^\circ\) as shown in Figure 4(e) and its zoomed view. These findings establish that the in-plane field is the deciding factor for the BLs configuration in the internal magnetic structures of the observed bubble domains.

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 material. Figure 4(f) 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 Figure 4(f), corresponds to a Bloch-type skyrmion as observed experimentally. Now, we apply a small in-
plane magnetic field of 0.02 T along the ±y–directions in addition to the out-of-plane magnetic field of 0.2 T. The simulated spin textures resemble that of the type-II bubble as depicted in Figure 4(g) and (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 Figure 4(g) and (h) fully match with that of the experimental spin textures obtained for the α rotation given in Figure 4(c) and (d). When the in-plane magnetic field is changed to the x-direction, the elongation of the spin texture follows the field direction [Figure 4(i)]. As expected, the simulated LTEM pattern shown in the inset of Figure 4(i) replicates the experimental spin texture observed in Figure 4(c). 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 with topological number zero rather than biskyrmion with topological number two.

As it can be found from Figure 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 earlier that Mn₄Ga₂Sn undergoes from easy-axis anisotropy at high temperature to easy-plane anisotropy below T_{SR} [23, 24]. Since the magnetic anisotropy plays a major role in stabilizing skyrmions in centrosymmetric magnets, we have calculated the effective easy-axis anisotropy constant (K_{eff}) for Mn₄Ga₂Sn from T = 70 K to T = 300 K based on the law of approach to saturation [34]. As it can be found from Figure 5(a), the effective easy-axis anisotropy exhibits its lowest value near the T_{SR} before increasing for the higher temperatures. The K_{eff} attains a maxima at T ≈ 250 K and then decreases rapidly as the T_C of the sample falls near the room temperature. Such a change in the K_{eff} can be understood by considering the competition between the dipolar energy and magnetocrystalline anisotropy [35].

To examine the role of K_{eff} on the stabilization of isolated skyrmions, the experimentally observed number of isolated skyrmions (n_{SKX}) are also plotted as a function of temperature in Figure 5(a). Interestingly, a direct correlation between the n_{SKX} and K_{eff} can be seen over the temperatures. To further substantiate our findings, we have carried out micromagnetic simulation to find out the evolution of skyrmions with different
$K_{eff}$ in the presence of an in-plane magnetic field of 0.02 T (see the supplementary Figure [S14]). The simulated number of isolated skyrmions ($n_{SKX}_{\text{simulated}}$) as a function of $K_{eff}$ is shown in the inset of Figure [S1a]. Corroborating our experimental results, the number of simulated isolated skyrmions increases with increasing $K_{eff}$. All these results suggest that stronger the $K_{eff}$ higher the probability of stabilizing the skyrmion phase. Hence, the in-plane magnetic field can easily nucleate the type-II bubble magnetic state by destabilizing the skyrmion phase at lower $K_{eff}$.

Based on our LTEM results, we have constructed a phase diagram ($H$-$T$) as depicted in Figure [S1b]. We note that the present $H$-$T$ phase diagram is obtained for the case where we have to slightly tilt (nearly 4 degrees) the sample to get good quality images at all temperatures. With this sample tilting, the type-II bubbles are the energetically favorable magnetic states over the wide temperature range. Few numbers of isolated skyrmions along with type-II bubbles are observed nearby 250 K due to higher effective easy-axis anisotropy. As discussed earlier, the transformation between the skyrmions and type-II bubbles can be achieved by applying a non-zero in-plane magnetic field. Hence, it should be possible to realize the skyrmion phase instead of type-II bubbles at low temperatures. However, the bending contour effect at lower temperatures restricts us to perform LTEM experiment near the pole.

Again, the direct visualization of hexagonal skyrmion lattice near room temperature at nearly zero sample tilting condition suggests skyrmions as the stable magnetic state in this system. The conflicting reports about the stabilization of biskyrmion and type-II bubbles in centrosymmetric magnetic systems require a better understanding of the spin textures in these systems [15, 22]. Hence, the present findings of tunable spin textures in the centrosymmetric systems Mn$_4$Ga$_2$Sn is an important step in this direction. Our LTEM experiment with controlled tilting conditions combined with the micromagnetic simulations confirm that the type-II magnetic bubbles are the feasible magnetic excitations along with conventional skyrmions in the centrosymmetric magnets. Our results also suggest the possibility of a controllable transformation between the skyrmion and type-II bubble, which might play a pivot role in future applications. We have also uncovered a relationship between the magnetic anisotropy and the skyrmion nucleation probability, where the higher anisotropy prefers skyrmions. These skyrmions can be easily transformed to type-II bubbles by applying an in-plane magnetic field.

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 Mn$_4$Ga$_2$Sn. Our experiment along with micromagnetic simulation, conclusively establish that the Bloch-type skyrmions are the energetically most favorable magnetic states in the hexagonal crystal based centrosymmetric magnets when the external magnetic field is applied along the magnetic easy-axis. 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 as found in most of the centrosymmetric skyrmion hosting magnets. 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 centrosymmetric magnets. 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.

**METHODE**

**Sample preparation**

Polycrystalline ingots of Mn$_4$Ga$_2$Sn 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 CuK$_\alpha$ radiation.

**Magnetic 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).
**LTEM measurements**

For the LTEM measurements, thin sample platelets of Mn$_4$Ga$_2$Sn 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 [37]. Slab geometry of dimensions 1000 nm $\times$ 1000 nm $\times$ 100 nm were used for the simulation, with a rectangular mesh of 4 nm $\times$ 4 nm $\times$ 4 nm. The material parameters were chosen according to the experimental data of Mn$_4$Ga$_2$Sn. Exchange stiffness constant ($A$) was calculated using the formula, $A \approx K_B T_C / a$, where $K_B$ 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 at 250 K was calculated by using the formula $D = \pi \sqrt{A/K_u}$, where $D$ is the domain wall width observed in the LTEM experiment. The estimated value of magnetocrystalline anisotropy constant $K_u$ is about $\approx 1.02 \times 10^5$ J/m$^3$. 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 corresponding to 250 K to 100 K, respectively. We have used PYLorentz code to simulate the LTEM intensity contrasts corresponding to the OOMMF spin textures [35]. To get the 3D spin textures, as shown in figure 1. For the spirit code software is used [30].

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SUPPLEMENTARY INFORMATION

1. Sample characterization

1.1. X-ray diffraction (XRD)

Room temperature powder XRD pattern with Rietveld refinement for Mn$_4$Ga$_2$Sn is shown in Figure S1. The Rietveld refinement was performed using the space group P6$_3$/mmc. In this structure the Mn atoms occupy two Wyckoff positions 6g (0.5, 0, 0) and 6h (x, 2x, 0.25) (x = 0.1562), whereas, Sn atoms preferentially occupy the 2a (0, 0, 0) and 2c (0.6667, 0.3333, 0.75) Wyckoff positions. The Ga atoms fully occupy the 6h (x, 2x, 0.75) (x = 0.1989) atomic position. We have calculated the lattice parameters as a = b = 8.5527 Å and c = 5.3264 Å, which match well with the earlier reports [23, 24].

FIG. S1: Room temperature powder XRD pattern for Mn$_4$Ga$_2$Sn sample. Black open circles represent the experimental data and the red solid line represent the simulated XRD pattern obtained from Rietveld refinement. The green scattered lines represent the Bragg position and difference between the experimental and simulated data is shown with the blue solid line.
TABLE I: Comparison between the exact atomic percentage and EDX obtained data.

| Elements | Exact atomic percentage | Obtained atomic percentage from EDX |
|----------|-------------------------|-------------------------------------|
| Mn       | 57.14                   | 58.76 ± 2.35                        |
| Ga       | 28.57                   | 25.60 ± 1.27                        |
| Sn       | 14.29                   | 14.27 ± 0.71                        |

1.2. Scanning electron microscopy (SEM)

The compositional homogeneity of the Mn$_4$Ga$_2$Sn sample is studied by using SEM and energy dispersive X-ray spectroscopy (EDX). The SEM image of the sample is shown in Figure S2. The uniform contrast in the SEM image represents the single phase nature of our sample. The small black spots represent the small holes present in the sample. To determine the chemical composition of our sample we have performed EDX analysis at several places on our sample. The observed EDX data nearly matches with the exact atomic composition of Mn$_4$Ga$_2$Sn as shown in Table I.

![SEM image of Mn$_4$Ga$_2$Sn sample.](image)

FIG. S2: SEM image of Mn$_4$Ga$_2$Sn sample.

2. Lorentz transmission electron microscopy (LTEM)

![Images of FIB lamella of Mn$_4$Ga$_2$Sn sample.](image)

FIG. S3: Images of FIB lamella of Mn$_4$Ga$_2$Sn sample. (a) Top view and (b) side view of sample lamella.
For the LTEM measurements we have prepared 110 nm thin plate of Mn$_4$Ga$_2$Sn sample using Ga-based focused ion beam (FIB) technique. Figure S3 shows the FIB image of the sample lamella from top view and side view.

As our sample exhibits a Curie temperature of about 320 K, very poor LTEM magnetic contrast is found at room temperature as shown in Figure S4. To understand the magnetic texture with better contrast we have cooled down our sample in cryogenic temperature ($T = 100$ K) using liquid-N2 TEM sample holder. Figure S5 shows the magnetic ground state of the sample at 100 K.

Magnetic field dependent LTEM imaging were also performed at various temperatures from $T = 100$ K to $T = 300$ K. Variation of magnetic domains with change in magnetic field at temperatures of 150 K, 200 K and 250 K are shown in Figure S6, S7 and S8, respectively. Here, along with the type-II bubbles, we have observed a larger number of isolated skyrmions at a temperature about 250 K, as marked with white boxes. The field sweep measurement at 250 K with zero tilting condition is also shown in Figure S9. At zero tilting condition formation of skyrmionic bubble from stripe domain is observed.

3. Calculation for temperature dependent effective magnetocrystalline anisotropy

We have calculated effective uniaxial magnetocrystalline anisotropy ($K_{eff}$) of our polycrystalline sample Mn$_4$Ga$_2$Sn applying the law of approach to saturation [34]. The magnetization ($M$) near the saturation ($M/M_s \geq 0.9$), where $M_s$ is saturation magnetization, can be written as $M = M_s (1 - a_2/H^2)$ with $a_2 = 4K_{eff}^2/15M_s^2$ is the fitting parameter. From the straight line fitting between $M$ and $H^{-2}$ near the saturation, we have calculated $a_2$ to estimate the possible $K_{eff}$ values. One of the fittings is shown in Figure S10(a). The temperature variation of $a_2$, $M_s$ and $K_{eff}$ are plotted in Figure S10(b). The maximum effective uniaxial anisotropy is observed around 250 K. We have also estimated the out-of-plane easy axis anisotropy ($K_u$) of our sample using the domain wall width, $\delta = \pi \sqrt{A/K_u}$. In addition, the exchange stiffness constant ($A$) is calculated using the Curie temperature ($T_C$), $A \equiv K_B T_C/a$, where $K_B$ is Boltzmann constant and $a$ is the lattice parameter of the sample Mn$_4$Ga$_2$Sn. The estimated value of $A$ appears...
FIG. S6: Over-focused LTEM images of Mn$_4$Ga$_2$Sn sample lamella (with 4 degree sample tilting) at 150 K at different magnetic fields from $\mu_0H = 0$ T to $\mu_0H = 0.53$ T.

$5 \times 10^{-12}$ J/m. The average domain wall width of our sample is observed around 22 nm at 250 K. The estimated value of $K_u$ is $1.02 \times 10^5$ J/m$^3$. The order of the estimated value of $K_u$ is also nearly matches with the calculated values of Keff calculated using $M$ vs $H$ data of polycrystalline sample for 250 K, as shown in Figure S10(b). From the 100 K LTEM data we have found the increase in domain wall width of 29 nm and decrease in calculated $K_u$ value to $0.58 \times 10^5$ J/m$^3$. These findings suggest the decrease in anisotropy of our sample with decreasing temperature.

4. Micromagnetic simulation

We have carried out a detailed micromagnetic simulation to understand the mechanism of type-II bubble and skyrmion formation in our sample. The micromagnetic simulations is carried out using the Object Oriented Micromagnetic Framework (OOMMF) programme[37]. The equilibrium states are obtained by fully relaxing the random magnetic state. It is found that the uniaxial magnetocrystalline anisotropy has a fundamental role for the stabilization of stripe domain as magnetic ground state. For very low anisotropy the vortex ground state is observed rather than stripe domain state.

The magnetic field dependent micromagnetic simulations is carried out using the sample parameters corresponding to 250 K and considering the stripe domain as initial state at zero magnetic field (Figure S11). As can be seen in Figure S12, the stripe domains are transformed into magnetic skyrmions with both clockwise and counter-clockwise helicities with increase in magnetic field.

The micromagnetic simulations are also performed for various sample tilting conditions i.e, with application of additional in-plane magnetic field in different directions. We have observed almost all the skyrmions are transformed to type II bubble with application of additional in-plane magnetic field corresponding to the 6-degree sample tilting condition with respect to x-axis and y-axis, as shown in Figure S13.

To understand the effect of uniaxial magnetocrystalline anisotropy on the skyrmion to type-II bubble transformation with in-plane magnetic field, the spin textures are simulated with varying uniaxial anisotropy ($K_u$) as shown in Figure S14. The simulations are carried out with an out-of-plane magnetic field of 0.3 T and an in-plane magnetic field of 0.02 T. The number of isolated skyrmions per $\mu$m$^2$ area increases with increase in the uniaxial magnetocrystalline anisotropy.
FIG. S7: Over-focused LTEM images of Mn$_4$Ga$_2$Sn sample lamella (with 4 degree sample tilting) at 200 K at different magnetic fields from $\mu_0 H = 0$ T to $\mu_0 H = 0.5$ T.

FIG. S8: Over-focused LTEM images of Mn$_4$Ga$_2$Sn sample plate (with 4 degree sample tilting) at 250 K at different magnetic fields from $\mu_0 H = 0$ T to $\mu_0 H = 0.43$ T.
FIG. S9: Over-focused LTEM images of Mn$_4$Ga$_2$Sn sample plate (with nearly 0 degree sample tilting) at 250 K at different magnetic fields from $\mu_0 H = 0$ T to $\mu_0 H = 0.43$ T.

FIG. S10: Effective magnetocrystalline anisotropy calculation for Mn$_4$Ga$_2$Sn. (a) Straight line fitting between the magnetization and $1/H^2$ at 70 K. (b) The fitting parameter $a_2$, saturation magnetization ($M_S$), and calculated anisotropy with varying temperature.

FIG. S11: The simulated magnetic ground states for different uniaxial anisotropy constants, with the sample parameters corresponding to $T=250$ K, $A = 5 \times 10^{-12}$ J/m, $M_s = 5.35 \times 10^5$ A/m. The simulation is performed considering sample area $1 \mu m \times 1 \mu m$ and thickness 100 nm.
FIG. S12: The simulated magnetic structures at different out-of-plane magnetic fields, with the sample parameters corresponding to 250 K. The used parameters are, $A = 5 \times 10^{-12} \text{ J/m}$, $M_s = 5.35 \times 10^5 \text{ A/m}$, $K_{\perp}u = 1.0 \times 10^5 \text{ J/m}^3$. 
FIG. S13: The simulated magnetic structures with out-of-plane magnetic field ($\mu_0 H_{OP}$) of 0.2 T and in-plane magnetic field ($\mu_0 H_{IP}$) of 0.02 T at different directions (a)-(e). The simulations are performed with the sample parameters corresponding to 250 K. The used parameters are, $A = 5 \times 10^{-12}$ J/m, $M_s = 5.35 \times 10^5$ A/m, $K_{u\perp} = 1.0 \times 10^6$ J/m$^3$. All the equilibrium states are achieved after fully relaxing the random magnetic state at that particular conditions. The simulation is performed considering sample area $1 \mu$m $\times$ $1 \mu$m and thickness 100 nm. The LTEM simulations of the magnetic structure corresponding to the figure (a) – (e) in over-focused mode is performed using PYLorentz code (f) – (j).
FIG. S14: The simulated magnetic textures for different uniaxial anisotropy constant values with out of plane magnetic field of 0.3 T and in-plane magnetic field of 0.02 T. The used parameters are, $A = 5 \times 10^{-12}$ J/m, $M_s = 6.68 \times 10^5$ A/m (saturation magnetization corresponding to 100 K). The black arrows represent the in-plane magnetization components. The simulation performed considering sample area $1\mu m \times 1\mu m$ and thickness 100 nm. The skyrmion spin textures are marked with black dashed boxes.