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Generation of high-density biskyrmions by electric current

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Much interest has been focused on the manipulation of magnetic skyrmions, including the generation, annihilation, and motion behaviors, for potential applications in spintronics. Here, we experimentally demonstrate that a high-density Bloch-type biskyrmion lattice in MnNiGa can be generated by applying electric current. It is revealed that the density of biskyrmions can be remarkably increased by increasing the electric current, in contrast to the scattered biskyrmions induced by a magnetic field alone. Furthermore, the transition from the ferromagnetic state to the stripe domain structure can be terminated by the electric current, leading to the biskyrmions dominated residual domain pattern. These biskyrmions in such residual domain structure are extremely stable at zero magnetic and electric fields and can further evolve into the high-density biskyrmion lattice over a temperature range from 100 to 330 K. Our experimental findings open up a new pathway for the generation of skyrmion lattice by electric current manipulation.

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INTRODUCTION

Magnetic skyrmions are topologically stable particle-like spin textures. The manipulation of skymions by electric current, for example, the skyrmion dynamic behavior,[1–3] has been the focus of significant research. Due to their nanometer size and spin-transfer torque (STT) driven behavior using ultralow electric current,[2, 3] skyrmions are promising candidates for applications in the next-generation spintronic devices. Skyrmions with single chirality, caused by the intrinsic Dzyaloshinskii–Moriya interactions (DMIs), are usually studied in non-centrosymmetric chiral systems at low temperature.[4–6] Exploring new materials to overcome the temperature limitation is the everlasting pursuit. Room-temperature skyrmions have been discovered in centrosymmetric magnets where the magnetic dipolar interaction is dominant,[7, 8] in magnetic multilayers with enhanced interfacial DMI,[1, 9] and in the artificial skyrmion systems.[10, 11] Usually, the spin configuration in bulk magnets is typically the Bloch-type "spiral" skyrmion where the swirling direction is perpendicular to the radial direction, as opposed to the Néel-type "hedgehog" skyrmion where the swirling direction is in the radial plane, as commonly found in ultrathin magnetic films. When an electric current is applied to the skyrmion system, the coupling between the non-trivial spin configuration of skyrmion and the electric current usually gives rise to interesting responses to magnetic field and electric field, such as emergent electromagnetic induction, skyrmion Hall effects,[9, 12] angular momentum exchange via the STT,[2] and others. When an electric current passes through a ferromagnetic material or a magnetic layer, the spin-polarized current can change the angular momentum or orientation of magnetization, if the torque is strong enough.[13, 14] The resulting rotation[2, 3] and translation motion[7, 15–17] are among the main topics of researches on skyrmion manipulation.

Additionally, the nucleation and propagation of skyrmions enabled by electric current have been put forward by several theoretical proposals.[18–21] Recently, it has been demonstrated experimentally that the electric current does create the Néel-type skyrmions by cutting the stripe domains in the interfacially asymmetric magnetic multilayers.[9] The mechanism for the Néel-type skyrmion generation has been further studied by simulating the dynamics of chiral stripe domain structure. The interaction between topological charge and conduction electrons induces the spin Hall torque that generates the observed Néel-type skyrmions.[22] Based on the theoretical simulations, it has been proposed that the current-induced STT can facilitate the generation of the Bloch-type skyrmions.[22] However, thus far, no experimental study on this issue has been reported.

Recently, we have indeed observed the Bloch-type biskyrmions in a centrosymmetric $(Mn_{1-x}Ni_x)_{65}Ga_{35}$ ($x = 0.5$) (MnNiGa) alloy, which can survive over a wide temperature (T) range from 100 to 340 K.[8] Unfortunately, in this alloy, magnetic field can indeed induce some biskyrmions, but the density of biskyrmions is low and no well-ordered biskyrmion arrangement (lattice) is possible at room temperature. Along this line, any manipulation of these scattered biskyrmions into a high-density biskyrmion lattice is definitely appealing for skyrmion-dependent applications in high-density information storage. Here, we demonstrate that, in the MnNiGa alloy, such a lattice can be properly generated by applying an electric current with the assistance of a magnetic field. With increasing magnetic field, the magnetic domain structure experiences a transition from the stripe pattern to the biskyrmion lattice and further to the ferromagnetic state because of the corresponding energy landscape tailored by the magnetic field.[23] Due to the interaction of conduction electrons with the diversified spin configurations, it is the STT to trigger the generation and propagation of biskyrmions. The generation of high-density Bloch-

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type biskyrmion lattices by electric current control would provide key roadmap for establishment of theoretical models and prospects for spintronic devices.

RESULTS

Hereafter we focus on the biskyrmion evolution with the influences of electric current. We pass a *dc* electric current through the sample (the schematic configuration and the calculation shown in Supplementary Fig. S1) and simultaneously perform the Lorentz transmission electron microscopy (Lorentz TEM) observations in the thinner region. The linear *J*–*V* curve and the unchanged stripe domain pattern at the current density of 7.68×10^7 A/m^2 (Supplementary Fig. S2) indicate that the Joule heating effect plays minor role. The magnetic field dependence of the biskyrmion evolution at a fixed current density of 5.0×10^7 A/m^2 is shown in the Lorentz TEM images (Figs. 1a–d). The unchanged helical stripes in Fig. 1a indicate that the STT at the current density of 5.0×10^7 A/m^2 is not strong enough to have significant influence on the stripe domain structure. Given the fixed current density, the biskyrmions appear to be pinched off along the stripe lines with increasing magnetic field, generating the mixed states of biskyrmion lines and shrunk stripes at a magnetic field of 0.15 T (Fig. 1b) and 0.17 T (Fig. 1c), respectively. When the field exceeds ~0.19 T (Fig. 1d), a self-organized high-density hexagonal biskyrmion lattice appears in compensation of the stripe domain structure. Further increasing the magnetic field makes the biskyrmion size shrink and eventually a homogeneous ferromagnetic state appears above the saturation field.

It should be mentioned that the stripe domain pattern can always be recovered when magnetic field is reduced back to zero from the saturation field if there is no other external stimulus. Thus the same sample region is used to compare the different manipulation effects. We present in Fig. 1e, f, the magnetic field-dependent biskyrmion evolution without electric current. It is seen that these stripes transform into biskyrmions with increasing magnetic field. However, only several scattered biskyrmions are induced at a magnetic field of 0.24 T (Fig. 1f). This demonstrates that it is the electric current to drive a significant increase of the biskyrmion density in comparison with the scattered biskyrmions in the presence of a magnetic field but without electric current. While no obvious change of the stripe domain structure is observed at the same current density of 5.0×10^7 A/m^2 (Fig. 1a), the generation of high-density biskyrmions is significantly facilitated (Fig. 1d) when the system comes to the vicinity of stripe domains to biskyrmions transition, driven by a magnetic field. It also implies that the electric current should be applied before the magnetic domain transition occurs, in order to generate the high-density biskyrmions. If the electric current is applied behind the magnetic domain transition, the density cannot be enhanced, as supported by the data in Supplementary Fig. S3, where the creep motion is the only consequence of the conventional STT effects on the biskyrmions, evidenced by their displacements. We notice that the current density needed to break the stripe domains is fairly low.[22] This phenomenon indicates that the energy barrier at the vicinity of the magnetic domain transition is lowered by increasing magnetic field[23] and this might be the reason for the reduced current density. It also should be mentioned that the critical magnetic field to form the biskyrmions can be influenced by the sample thickness, which results in the sequence of biskyrmion generation and annihilation along the gradient thickness of the sample.

The relationship between the biskyrmion density and the pre-applied electric current density is systematically analyzed, based on the Lorentz TEM observations, as shown in Fig. 2. In the absence of electric current, only scattered biskyrmions induced by magnetic field can be identified (Fig. 2a). If a nonzero electric current passing through the sample first and then a vertical magnetic field are applied in sequence, the biskyrmion density can be greatly enhanced. The larger the pre-applied current is, the denser the biskyrmions will be (Fig. 2b–d). This tendency becomes apparent above the critical current density of 3.4×10^7 A/m^2, as seen in Fig. 2e where the *B*–*J* phase diagram over a broad region is evaluated from the biskyrmions contour mapping data using the Lorentz TEM. It can be concluded that the high-density biskyrmion lattice can be tuned out by a sufficiently large current injection. Moreover, the critical magnetic field to form the biskyrmions can be reduced if the electric current increases. This dependence can be understood if one considers that the current-induced STT can change the microscopic domain configuration so that the long-range stable stripe ground state is gradually replaced by a series of metastable biskyrmions states when the system goes through the magnetic domain transition. Here, the increased biskyrmions are generated from the same stripe domain region instead of accumulating skyrmions from other area driven by electric current.[9] This intriguing fact that the biskyrmions reorganize themselves into a remarkably dense distorted hexagonal lattice (Fig. 2d) due to the electric current excitation, provides the possibility for externally controlling the magnetic biskyrmion lattice required for information bits of high density and low-power consumption memory devices. It should be noted that a non-zero magnetic field is always required to stabilize the biskyrmion lattices generated by this electric current manipulation.

Much of previous research focused on the skyrmions evolution process by varying magnetic field.[24–26] It is known that the stripe domain structure instantly pops out from the homogeneous ferromagnetic state with decreasing magnetic field, without the intermediate skyrmion phase. In this sense, the magnetic field-free residual state should be the stripe domain structure. However, in our study, the magnetic field-free residual domain structure is a mixed state of biskyrmions and stripes. This state, evolved from the ferromagnetic state and with decreasing magnetic field, is observed only if an appropriate electric current is continuously applied to the sample. Figure 3 shows the Lorentz TEM images of several field-free residual domain structures obtained by several different roadmaps. The detailed processing procedures are schematically illustrated in the *insets* of Fig. 3 where the electric current and the magnetic field are imposed according to the assigned paths. The corresponding Lorentz TEM images are acquired at the stages marked by a *star*. In the absence of electric current, the regular stripe domain structure directly emerges with decreasing magnetic field from the homogeneous ferromagnetic state (driven by a high-magnetic field above the saturation threshold), as shown in Fig. 3a. The mixed states with coexisting biskyrmions and stripes are observed if the electric current is applied before turning-on the magnetic field. At a current density of ~6.4×10^7 A/m^2, one sees that the biskyrmions with elongated shape occupy major part of the region and the stripes occupy the minor part (Fig. 3c). Starting from this residual state in Fig. 3c, further increasing the magnetic field leads to the shrink of the irregular-shape biskyrmions, reaching a well-developed distorted biskyrmion lattice (Fig. 3d). The nucleation of the residual biskyrmions from the ferromagnetic state is different from the classic skyrmion formation where only magnetic field applied to the stripe-like domain structure is needed. For the present case, the energy barrier is relatively small[23] at the vicinity of the magnetic domain transition from the parallel spin configuration to the magnetic helical structure, and the generation of biskyrmions can be facilitated at an optimized electric current intervening condition. When the magnetic field decreases down to zero, the residual biskyrmion lattice is robust once it is generated. Even after removing both the magnetic field and the electric current, this lattice can exist over a wide temperature range from 100 to 330 K. Further increasing the magnetic field forces the biskyrmion size smaller (Supplementary Fig. S4).

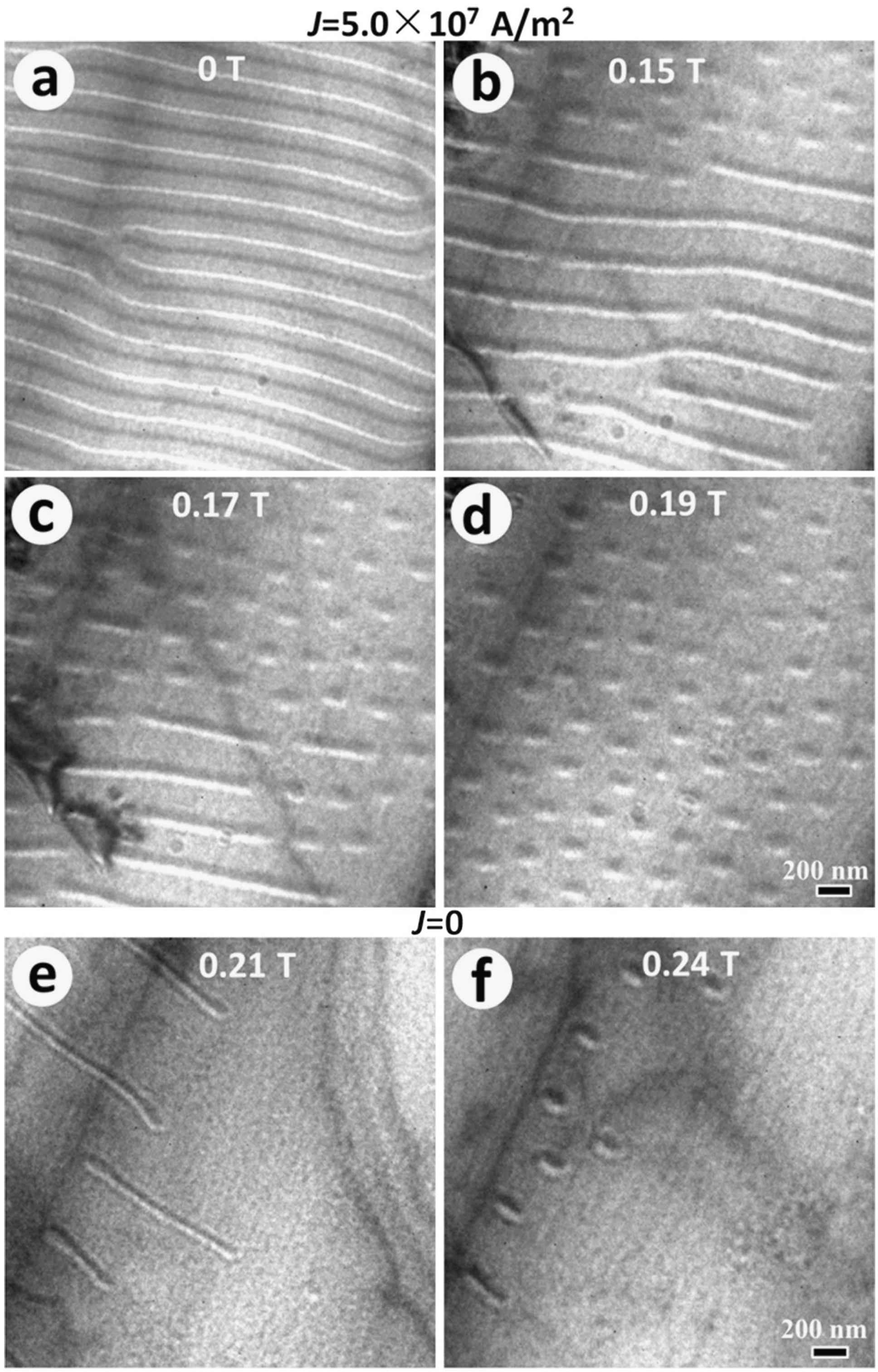

Fig. 1 High-density biskyrmions by electric current manipulation compared to the magnetic field-induced scattered biskyrmions. Lorentz TEM images of biskyrmions transition at a magnetic field of **a** 0 T, **b** 0.15 T, **c** 0.17 T, **d** 0.19 T while a fixed electric current of 5.6×10^7 A/m^2 is maintained. High-density biskyrmions pinched off along the stripe line, attributing to the spin transfer torque effects induced by electric current. The Lorentz TEM images showing the magnetic field dependence of biskyrmions at **e** 0.21 T and **f** 0.24 T, without electric current for comparison

It is noted that the typical size of the biskyrmions is ~90 nm, which approximately equals to the half periodicity λ of the helical stripe domain structure determined by the competing exchange interactions, and this value may vary slightly depending on different experimental conditions.[1] The corresponding lateral magnetizations for those outlined single biskyrmions (Fig. 3, *insets*) show similar magnetic configurations in spite of the different shapes. The spin configuration for the Bloch-type biskyrmion is composed of two skyrmions with opposite helicities (clockwise and counter-clockwise spin curl orientations with no

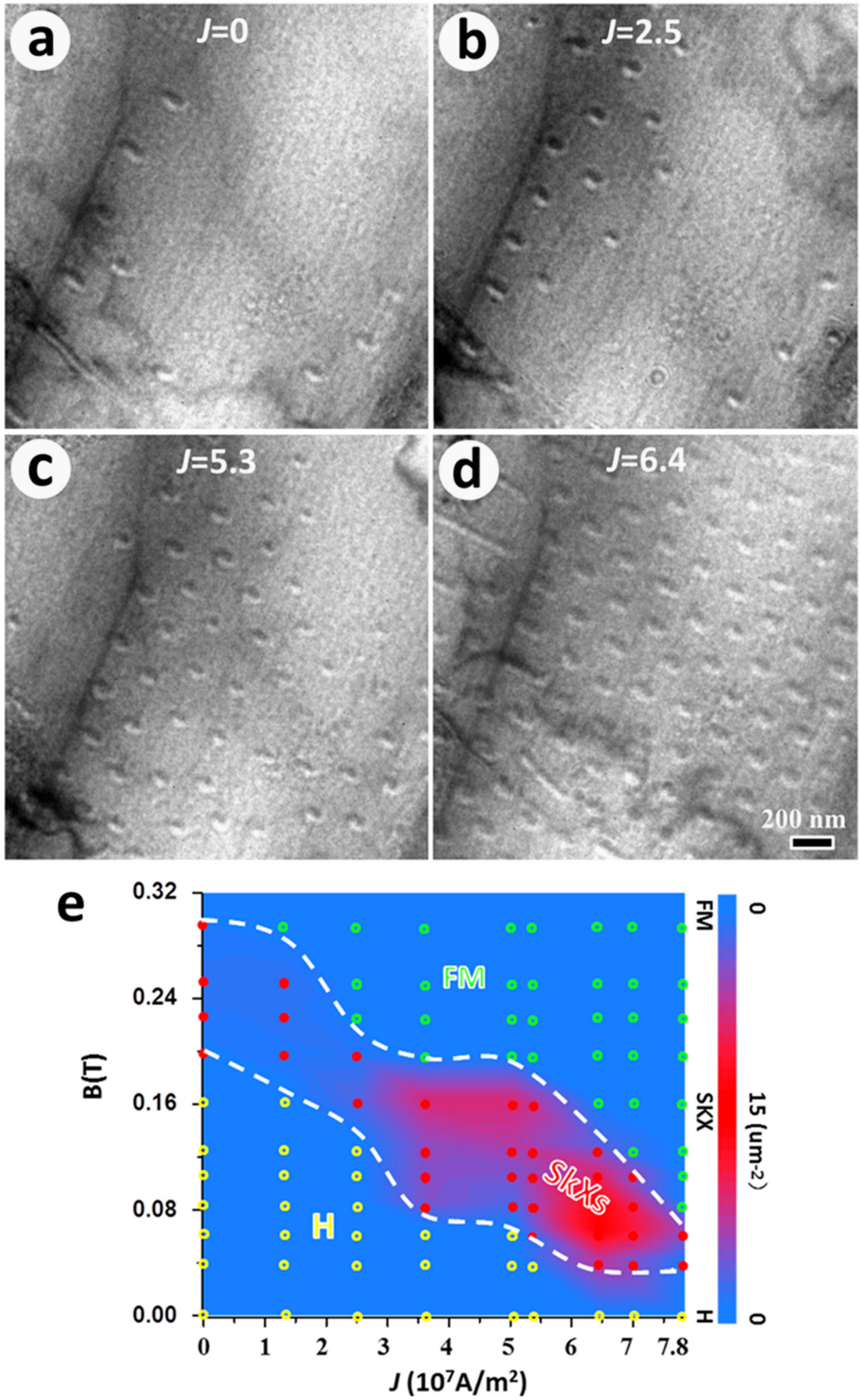

Fig. 2 The relationship between biskyrmion density and the pre-applied electric current density. Lorentz TEM images of biskyrmion distribution at different fixed current density of **a** 0, **b** 2.5×10^7 A/m^2, **c** 5.3×10^7 A/m^2, **d** 6.4×10^7 A/m^2. **e** The contour mapping of biskyrmions as a function of external magnetic field (*B*) and current density (*J*) based on in-situ Lorentz TEM observations. The *colorful dots* and *dashed lines* show the experimental points, from which the skyrmion density maps are extrapolated, and a guide to the eyes for the threshold and annihilation magnetic fields for the skyrmions evolution, respectively. The color scale indicates the skyrmion density per square micrometer. *H* helical stripe state, *SKXs* skyrmions phase, and *FM* ferromagnetic state

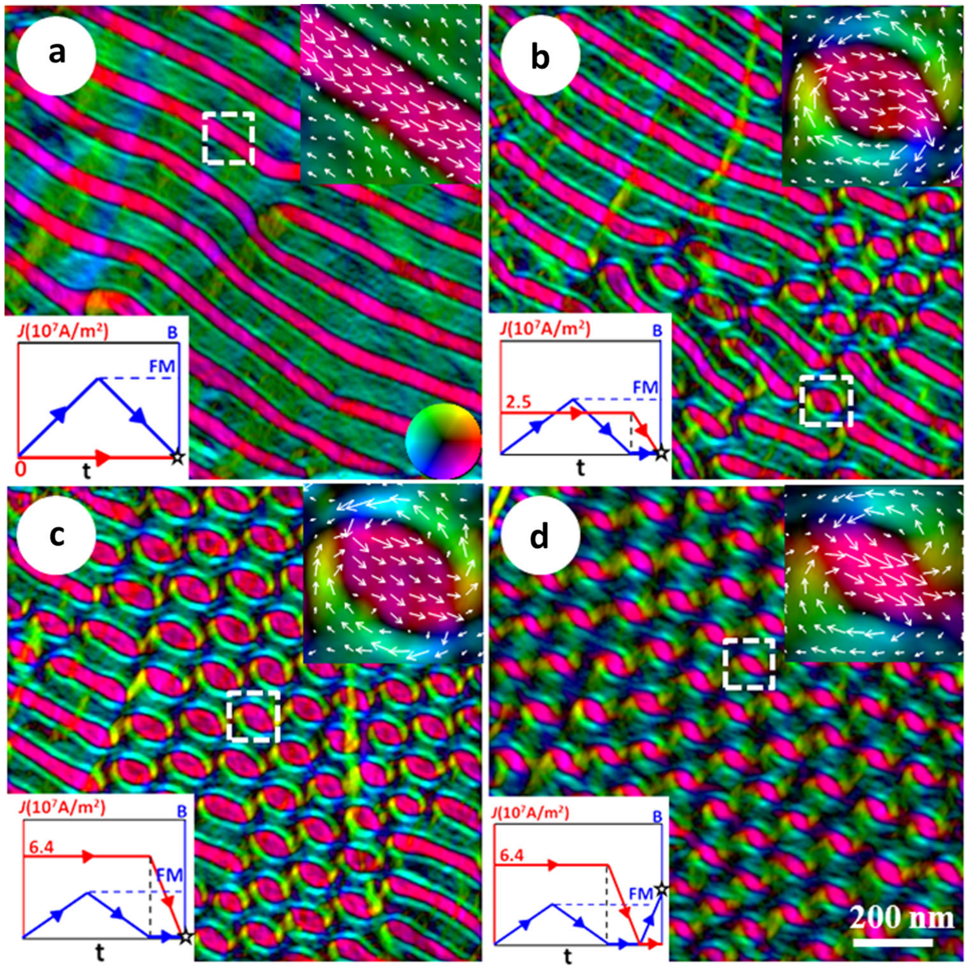

Fig. 3 Residual magnetic domain structures without and with interfering the transition from the ferromagnetic state to the stripe pattern by electric current. The Lorentz TEM images of the residual in-plane magnetization distribution formed at varied electric current density of **a** 0, **b** 2.5×10^7 A/m^2, and **c** 6.4×10^7 A/m^2, passing across the magnetic domain transition. The detailed experimental procedures are schematically shown in the *insets* with the current density and magnetic field applied accordingly. The *red axis* for applied current density, the *blue axis* for applied magnetic field. The Lorentz TEM images are taken at the experimental condition marked by a *star*. **d** The Lorentz TEM images of the biskyrmion lattice evolved from the residual magnetic domain structure shown in **c**, upon a magnetic field increasing up to 0.17 T. The corresponding in-plane magnetization maps for selected single biskyrmions are shown in the insets

net chirality) but the same central spin direction antiparallel to external magnetic field (at the core sites). This configuration is energetically stable due to the dipolar interaction in centrosymmetric magnets.[7] This current-induced biskyrmion formation is different from the traditional bubble domain dynamics where the bubble domains either collapse without moving or grow in the current direction,[27] which further confirms that the biskyrmions in MnNiGa have topologically stable properties. The realization of the high-density biskyrmions lattice by the present electric current control roadmap, together with the topological features such as nanometric localized spin configuration and fixed topological number, will be highly appreciated for practical applications.

Based on the corresponding Lorentz TEM imaging results, one may propose the schematic diagram in Fig. 4 where the evolution of high-density biskyrmions in response to electric current is summarized, in comparison with the evolution of conventional magnetic field-induced scattered biskyrmions. Four different evolution paths/experimental procedures with four colors are mapped out accordingly. The *red channel* represents the current-controlled procedure shown in Fig. 1a–d, where a fixed electric current is pre-applied before turning-on the magnetic field. The density of biskyrmions in the lattice is strikingly increased in contrast to the scattered biskyrmions induced directly by a magnetic field without electric current, as shown in Fig. 1e, f (*green channel*). The other current-controlled procedure is based on Fig. 3, and is shown here by the *pink channel*. The *green* and *purple channels* stand for two conventional procedures for comparison. As shown above, only scattered biskyrmions can be generated via the *green channel*, as shown in Fig. 1e, f. The current-driven biskyrmion formation at the vicinity of magnetic phase transition with mixed stripes and biskyrmions (*purple channel*) is based on the results shown in Supplementary Fig. S3. A comparison of all these procedures allows us to clearly see the advantages of the current-controlled procedures in manipulating the biskyrmion density, as proposed in this work, over the conventional procedures where only a magnetic field is used to induce the biskyrmions.

DISCUSSION

In summary, the generation of well-ordered Bloch-type biskyrmions with tunable density via electric current control has been clearly demonstrated in MnNiGa by the Lorentz TEM. The biskyrmion density can be strikingly increased by the proper electric current manipulation, in contrast to the case of scattered biskyrmions induced by a magnetic field. The commonly observed

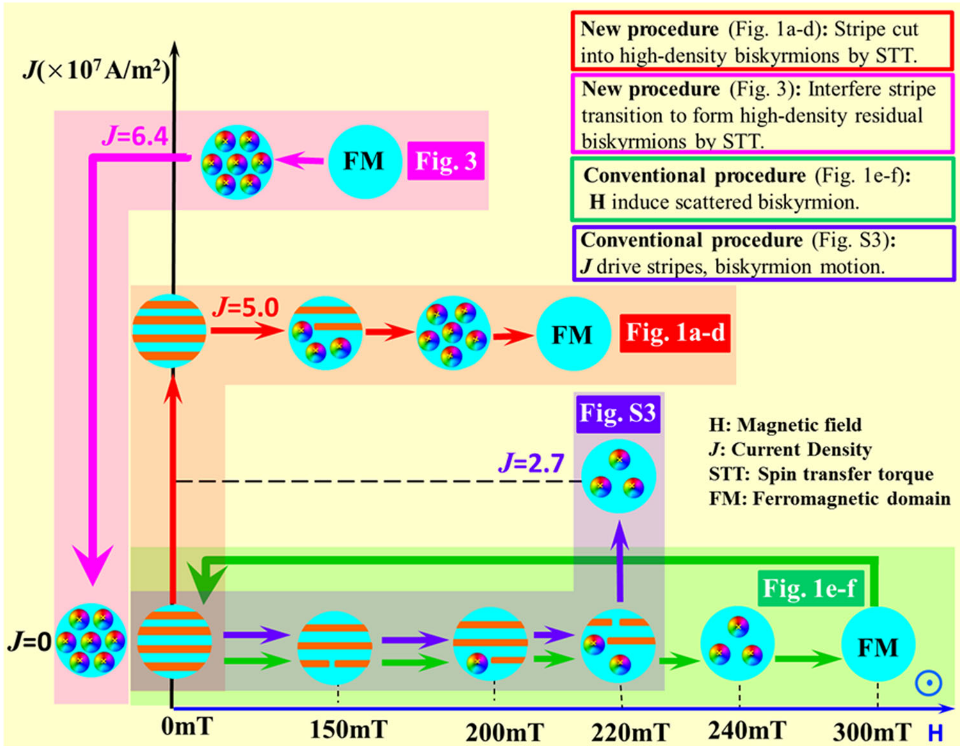

Fig. 4 Schematic diagram elaborating the effects of electric current in generating the high-density biskyrmions in MnNiGa alloy. Four different processing paths/experimental procedures are mapped out with four colors. The *red* channel stands for the procedure with high-density biskyrmions based on Fig. 1a–d, found in this work. The *pink* channel stands for the other procedure with high-density residual biskyrmion lattice based on Fig. 3, found too in this work. The *green* and *purple* channels stand for two conventional procedures with isolated biskyrmions based on Fig. 1e, f and Supplementary Fig. S3, respectively. The *arrows* indicate the sequence of applying external field. The direction of the magnetic field points out of the plane

transition from the ferromagnetic state to the stripe domain structure with decreasing magnetic field can be terminated by pre-applying an appropriate electric current, thus facilitating the generation of biskyrmions in compensation of the stripe domains. The residual biskyrmions are nonvolatile, and the high-density biskyrmion lattice that can survive over a wide temperature range from 100 to 330 K can be generated by proper current-controlled procedures. The reduced energy barrier at the vicinity of the magnetic phase transition and the STT effects both contribute to the high-density biskyrmions generation in these current-controlled procedures. The electric current is shown to be an effective option to control the density of magnetic biskyrmions. This easy and efficient tuning option together with the topological features over a wide temperature range make this material an excellent candidate as information carriers in the next-generation ultra-dense magnetic spintronic devices. Our experimental findings not only open up a new pathway for skyrmions generation, but also provide key data for further theoretical simulation to discover the nature of the interaction of the electric current with different spin configurations.

METHODS

The as-cast polycrystalline $(Mn_{1-x}Ni_x)_{65}Ga_{35}$ ($x = 0.5$) compound is fabricated by arc-melting method, as illustrated in details in ref. 8 and termed as MnNiGa in this paper. The thin plates for Lorentz TEM observations are cut from bulk samples and thinned by mechanical polishing and argon-ion milling. The averaged grain size is ~10 µm, large enough to be locally treated as single crystal. The MnNiGa TEM sample is trapezoidal-shaped with the electron-transparent area thickness around 40 nm and the thickness gradually increases away from the edge with the range of 40–100 nm. The magnetic domain contrast is observed using the Lorentz TEM (JEOL2100F) operated at 200 kV. The magnetic field applied normal to the thin plate is induced by the magnetic objective lens of the TEM. The current-controlled biskyrmion formation is observed using a double-tilt electrical TEM holder with two electrical conducting blocks at two sides of the TEM sample. The *dc* current is supplied by a source-measure unit instrument (Keithley 2601B). The approximately averaged current density of 2.8×10^7 A/m^2 corresponds to a current of 100 mA according to the cross-section area calculation, as shown in Supplementary Fig. S1.[3, 7] The magnetic structures could be examined directly in the electron microscope by three sets of images with under-, over-, and just (or zero) focal-lengths and then the high-resolution in-plane magnetization distribution map is obtained using the commercial QPt software based on the transport of the intensity equation.[28] The colors depict the magnitude and direction of the lateral magnetization, which can be deduced from the *arrows* and the *color wheel*.

Data availability

The data supporting the findings of this study are available from the corresponding authors on reasonable request.

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AUTHOR CONTRIBUTIONS

Y.Z. conceived and designed the experiments. L.C.P. and Y.Z. performed the experiments and analyzed the data. M.H. contributed to the electric current control set up. W.H.W. and B.D. synthesized the samples. H.F.T. contributed to the TEM set up. J.Q.L., B.G.S., S.G.W., J.W.C., G.H.W., and J.P.L. contributed to the results analysis. Y.Z., L.C.P., and M.J.K. wrote the manuscript. All authors discussed the results and commented on the manuscript.

ADDITIONAL INFORMATION

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