## ADVANCED MATERIALS

## **Supporting Information**

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A Centrosymmetric Hexagonal Magnet with Superstable Biskyrmion Magnetic Nanodomains in a Wide Temperature Range of 100–340 K

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## A centrosymmetric hexagonal magnet with superstable biskyrmion magnetic nanodomains in a wide temperature range of 100K to 340K

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Figure S1. The concentration, x, dependence of (a) lattice constants and (b) magnetic-ordering temperature,  $T_{c}$ .





Figure S2. Typical under-focused Lorentz TEM images show the magnetic stripes for  $(Mn_{1-x}Ni_x)_{65}Ga_{35}$  alloys with (a) x=0.2; (b) x=0.3; (c) x=0.4; (d) x=0.45.





Figure S3. M(H) curves measured for MnNiGa thin plate with the field direction perpendicular to the sample plane.





Figure S4.  $\frac{\rho_{xy}}{H}$  vs  $\frac{\rho_{xx}^2 M}{H}$  curves for varous temperatures. As in the high field rigion above Hc, the hall resistivity obeys  $\frac{\rho_{xy}}{H} = R_0 + S_A \frac{\rho_{xx}^2 M}{H}$ , we obtain the intercept as  $R_0$  and the slope as  $S_A$  by a linear fit from ~1.5T to 3T, as indicated by the red line. Inset shows the obtained  $R_0$ and  $S_A$  values.



Figure S5. The  $\rho_{xy}$ -*H* curve measured at different temperatures, calculated  $R_0H + R_SM$  curve (red line) used the above fitted  $R_0$  and  $S_A$  values ( $R_S = S_A \rho_{xx}^2$ ), and derived  $\rho_{xy}^T$  curve (blue line) from the analysis. At  $H > H_c$  (the critical field marked by arrows), the value of  $R_0H + R_SM$  is in accord with the experimental data.





Figure S6. Temperature dependence of Lorentz TEM images. Obvious biskyrmion domains are formed at (a) 167K with a perpendicular magnetic field of 0.57T. (b) At 190K with a magnetic field of 0.54T. (c) At 215K with a magnetic field of 0.52T. (d) At 230K with a magnetic field of 0.5T. The density of the biskyrmion domains shows a rough maximum at T=215K.



Figure S7. Part of the band structure and density of states (DOS) of  $(Mn_{1-x}Ni_x)_{65}Ga_{35}$  with x=0.5: (a) majority spin, the green lines are used for emphasis, (b) density of states, (c) minority spin. In the DOS, the numbers of the majority and minority electrons differ at the Fermi level, leading to a spin-polarized electronic structure of a 33% bulk spin polarization.



Sample	Temperature (K)	Field for biskyrmion formation	Field for biskyrmion state	Field for FM state
#1	338K	<b>0.04T</b>	<b>0.11T</b>	<b>0.20T</b>
#1	<b>297K</b>	<b>0.20T</b>	<b>0.40T</b>	<b>0.45</b> T
#2	<b>297K</b>	<b>0.20T</b>	<b>0.33T</b>	<b>0.37T</b>
#1	230K	0.25	<b>0.48T</b>	<b>0.58T</b>
#1	215K	<b>0.27T</b>	0.53T	<b>0.62T</b>
#2	<b>167K</b>	<b>0.34T</b>	<b>0.57T</b>	<b>0.64T</b>
#1	100K	<b>0.37T</b>	0.58T	0.65T

Table S1. *In-situ* Lorentz TEM data carried out at different temperatures to confirm the applied magnetic fields for biskyrmion formation, complete biskyrmion state, and FM state, respectively. All the data were used to outline the stability of the biskyrmions for MnNiGa thin plate in Figure 5.