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Supporting Information

Transition from an Antiferromagnetic Ground State to Robust Ferrimagnetic Order with Curie Temperature above 420 K in Manganese-based Antiperovskite-type Structures

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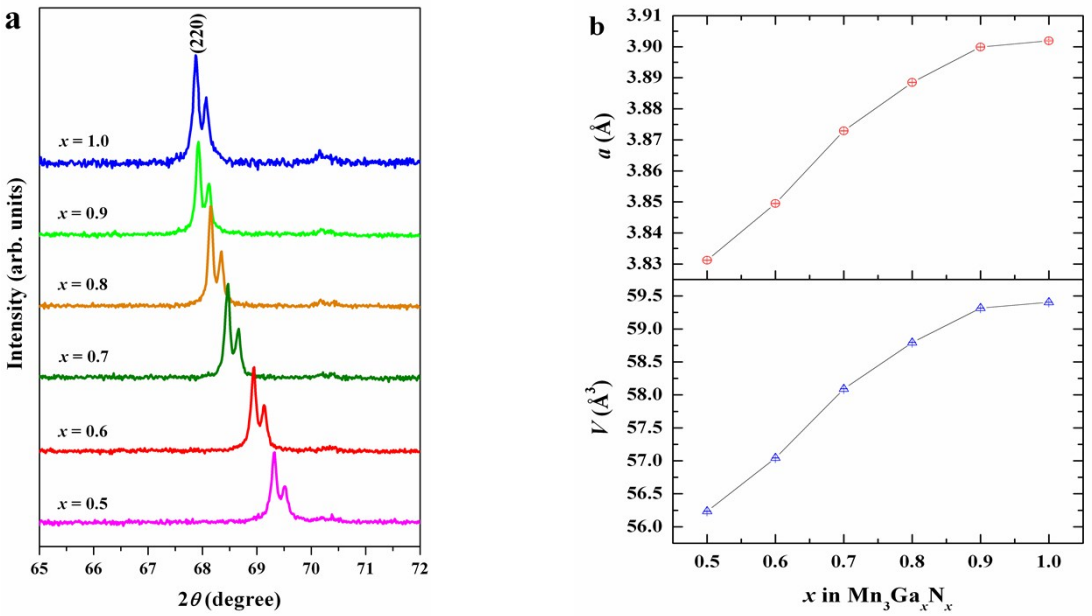

Fig. S1 (a) (220) peaks (λ = 1.5418 Å) and (b) lattice parameters and volumes of $Mn_3Ga_xN_x$ (x = 0.5−1.0) as a function of x from the laboratory powder X-ray diffraction (XRD) data.

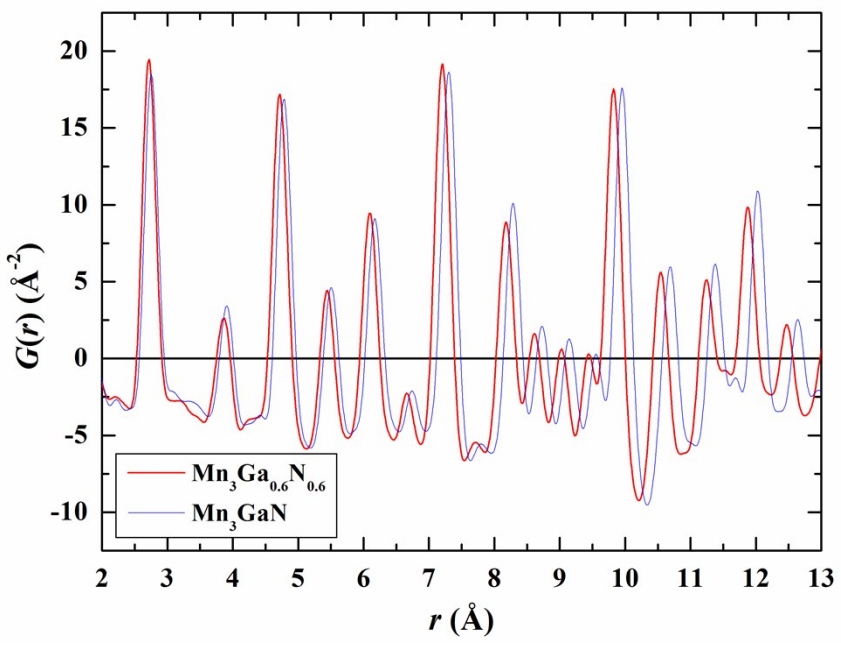

Fig. S2 Experimental pair distribution function (PDF) for $Mn_3Ga_{0.6}N_{0.6}$ and Mn_3GaN collected at room temperature.

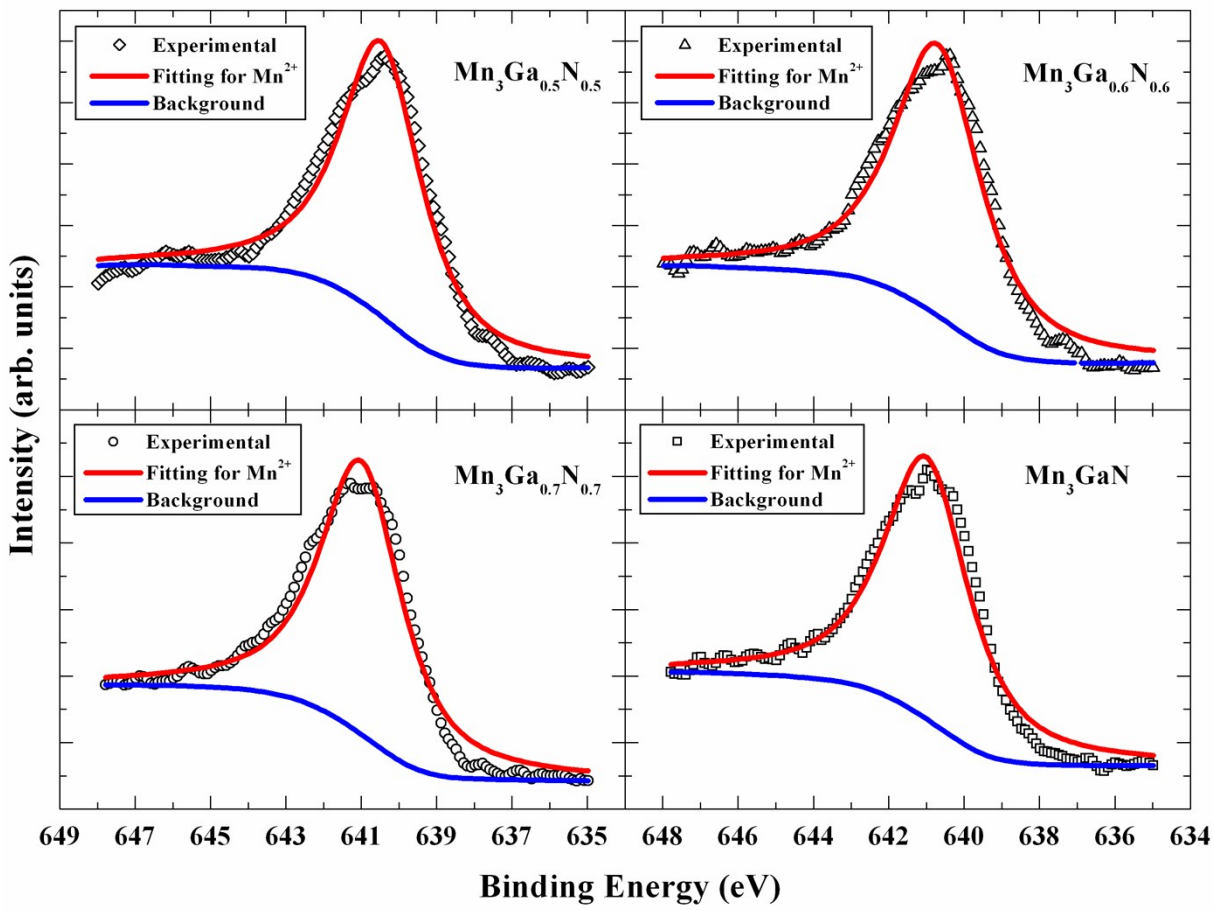

Fig. S3 X-ray photoelectron spectroscopy (XPS) spectra for $Mn_3Ga_xN_x$ (x = 0.5, 0.6, 0.7, and 1.0) at room temperature.

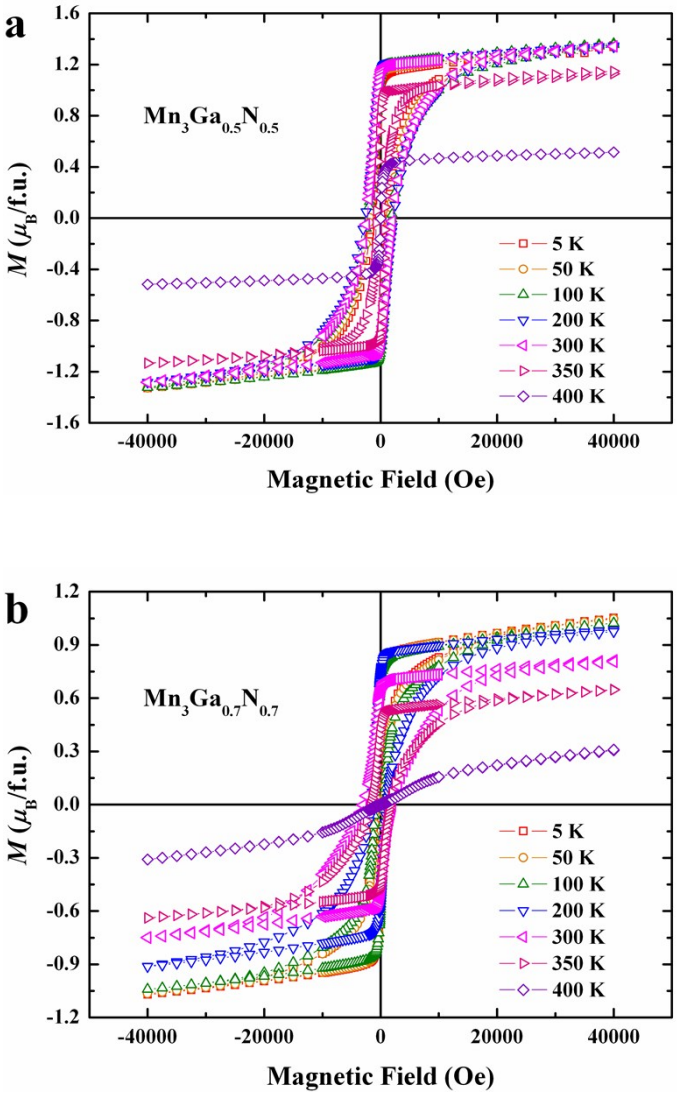

Fig. S4 Hysteresis loops for $Mn_3Ga_{0.5}N_{0.5}$ (a) and $Mn_3Ga_{0.7}N_{0.7}$ (b) measured in the range −40 kOe < H < 40 kOe at different temperatures. Here f.u. represents formula unit.

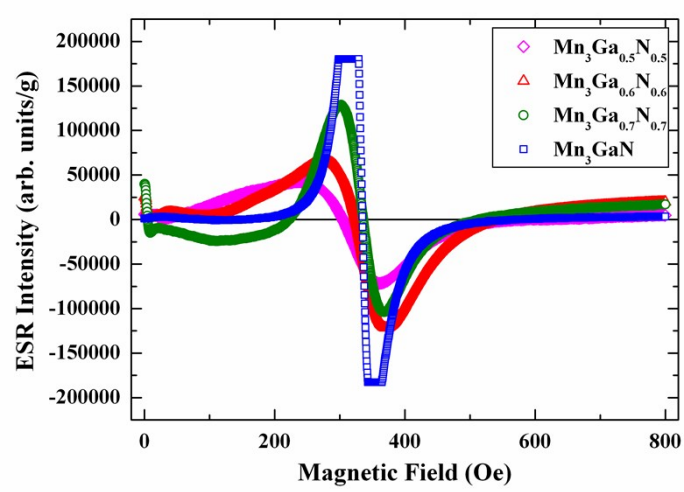

Fig. S5 Electron spin resonance (ESR) spectra for $Mn_3Ga_xN_x$ (x = 0.5, 0.6, 0.7, and 1.0) at room temperature.

Table S1 Refined lattice parameters and atomic coordinates of $Mn_3Ga_{0.6}N_{0.6}$ and Mn_3GaN from high-resolution synchrotron X-ray diffraction (SXRD) patterns at room temperature

Atom	**Site**	***x***	***y***	***z***	**U (Å^2)**	**Occ.**
$Mn_3Ga_{0.6}N_{0.6}$						
Space group: *Pm*-3*m*; a = 3.85762(2) Å; R_p = 3.68%, R_{wp} = 4.60%, χ^2 = 2.058.						
Mn	3*c*	0	0.5	0.5	0.0052(2)	1
Ga	1*a*	0	0	0	0.0096(5)	0.75(1)
Mn	1*a*	0	0	0	0.0096(5)	0.25(1)
N	1*b*	0.5	0.5	0.5	0.01	0.6
Mn_3GaN						
Space group: *Pm*-3*m*; a = 3.90471(5) Å; R_p = 3.38%, R_{wp} = 4.20%, χ^2 = 2.089.						
Mn	3*c*	0	0.5	0.5	0.0079(1)	1
Ga	1*a*	0	0	0	0.0086(2)	1
N	1*b*	0.5	0.5	0.5	0.014(1)	1

Table S2 Refined lattice parameters and atomic coordinates $Mn_3Ga_{0.6}N_{0.6}$ from neutron powder diffraction (NPD) patterns at 421 K and room temperature. The occupancies of 1*a* and 1*b* sites at room temperature are fixed as the values derived from the 421 K data.

Atom	**Site**	***x***	***y***	***z***	**U (Å²)**	**Occ.**
421 K						
Space group: *Pm*-3*m*; a = 3.85703(2) Å; R_p = 5.02%, R_{wp} = 5.51%, χ^2 = 0.963.						
Mn	3*c*	0	0.5	0.5	0.0119(7)	1
Ga	1*a*	0	0	0	0.0102(7)	0.712(5)
Mn	1*a*	0	0	0	0.0102(7)	0.288(5)
N	1*b*	0.5	0.5	0.5	0.0041(7)	0.614(9)
Room Temp.						
Space group: *Pm*-3*m*; a = 3.85612(5) Å; R_p = 6.76%, R_{wp} = 8.68%, χ^2 = 1.20.						
Mn	3*c*	0	0.5	0.5	0.0092(4)	1
Ga	1*a*	0	0	0	0.0084(4)	0.712
Ga	1*a*	0	0	0	0.0084(4)	0.288
N	1*b*	0.5	0.5	0.5	0.0078(4)	0.614

Table S3 Inductively coupled plasma-atomic emission spectrometry (ICP-AES) results for $Mn_3Ga_xN_x$

x	**1.0**	**0.9**	**0.8**	**0.7**	**0.6**	**0.5**
Mn	2.92	2.95	2.94	2.91	2.92	2.93
Ga	0.95	0.87	0.74	0.65	0.56	0.47
N	0.88	0.79	0.68	0.59	0.51	0.41
Fe	0.000003	0.000007	0.000005	0.000005	0.000004	0.000004
Co	0.000008	0.000006	0.000009	0.000003	0.000007	0.000009
Ni	0.000006	0.000002	0.000002	0.000006	0.000005	0.000003