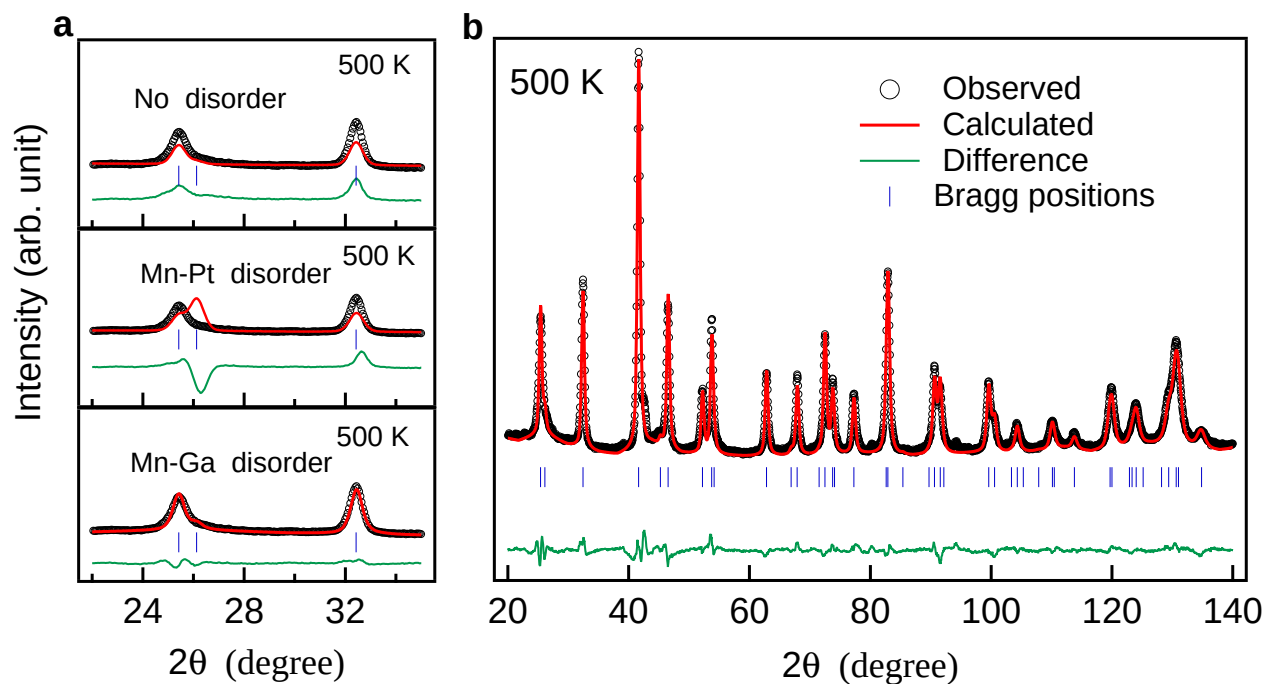
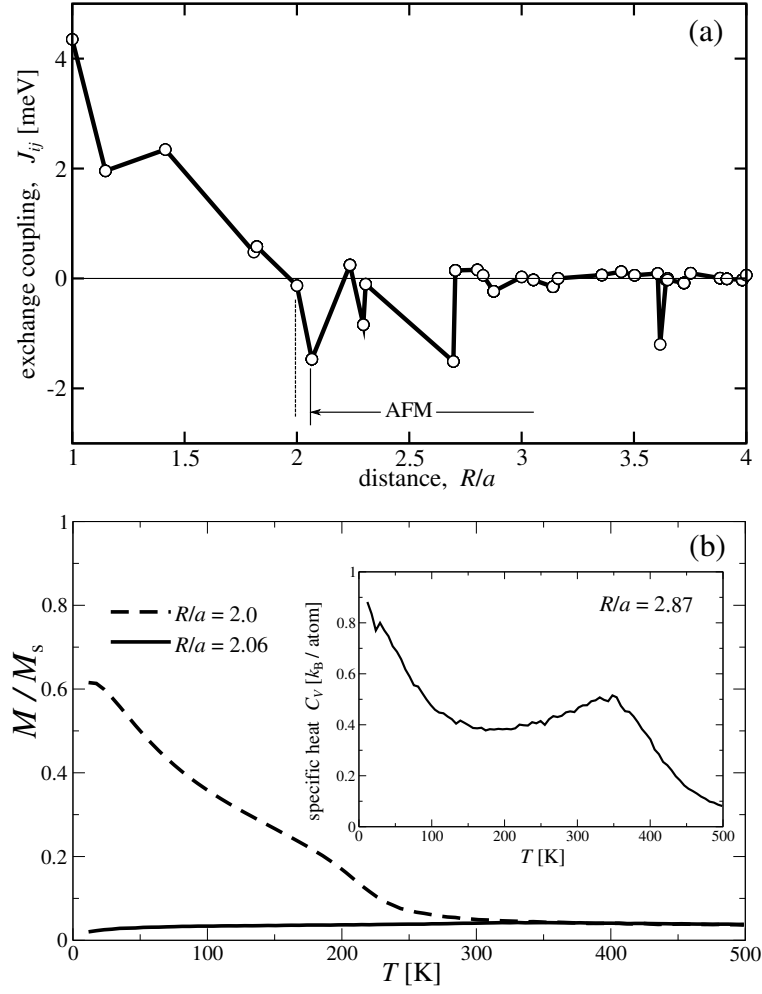




Supplementary Figure 1. Powder neutron diffraction on Pt_2MnGa at 500 K. The green curve shows the difference between observed (black) and calculated patterns (red). Vertical ticks indicate the nuclear Bragg peak positions. **a** The (002) and (110) Bragg peaks (black circles) have been fitted (red solid lines) by assuming (i) no disorder, (ii) Mn(2a)/Pt(4d) disorder, and (iii) Mn(2a)/Ga(2b) disorder. **b** Comparison of neutron diffraction patterns for the whole angular range.



Supplementary Figure 2. **a** Isotropic exchange coupling constants J_{ij} computed as functions of distance R (in the units of a) between i and j sites within $\text{Mn}(2a)$ sublattice. **b** Monte-Carlo simulated M/M_s temperature dependencies of the Heisenberg model parametrized by the computed J_{ij} . Solid line corresponds to the minimal cluster size needed to set the AFM order, dashed - by one shall smaller cluster. The inset shows the temperature dependency of the specific heat C_V (computed for the largest technically possible cluster size of $R/a = 2.87$) which indicates the position of T_N by the local maximum.

Supplementary Note 1

Crystal structure from neutron diffraction

Supplementary Figure 1 shows the observed and calculated neutron diffraction patterns at 500 K (paramagnetic phase) in the range of $20^\circ < 2\theta < 140^\circ$. All peaks are well indexed in Rietveld refinement assuming the space group $I4/mmm$ as in case of the X-ray diffraction. Refined lattice parameters are $a = b = 4.03 \text{ \AA}$, $c = 7.24 \text{ \AA}$. In a perfectly ordered unit cell, Mn, Ga and Pt atoms would occupy $2a (0, 0, 0)$, $2b (0, 0, 0.5)$ and $4d (0, 0.5, 0.25)$ Wyckoff positions, respectively. On the other hand, our Rietveld fit indicates possible presence of Mn(2a)-Ga(2b) disorder: by comparing several reasonable chemical configurations (focusing on (002) and (110) Bragg peaks), the perfect agreement is provided by assuming $\sim 33\%$ of Mn(2a)-Ga(2b) disorder (lower panel in Supplementary Figure 1 a).

Supplementary Note 2

Monte-Carlo simulations

Here we try to figure which mechanisms are responsible for setting such a long range ground-state modulation (by assuming a perfectly ordered structure). Since the relativistic effects do not affect the ground-state $\mathbf{q}$ vector substantially, in the following we calculate the isotropic exchange coupling constants J_{ij} using the real-space approach¹ implemented in the SPR-KKR Green's function method.² In Supplementary Figure 2 a they are plotted as functions of distance between the interacting sites i and j . Here, we drop all interactions involving Pt and Ga atoms as insignificant,

by leaving only those between Mn atoms. It follows, that all nearest Mn($2a$)-Mn($2a$) interactions are parallel ($J > 0$), whereas the antiparallel ones ($J < 0$) are encountered by starting from $R/a = 2$ (6-th shell within ($2a$)-sublattice). As it is shown by $M(T)/M_S$ curves (Supplementary Figure 2 b) obtained by the Monte-Carlo simulation (ALPS package³) of the classical Heisenberg model ($H = - \sum_{i>j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j$, where $\mathbf{e}_{i,j}$ are the unity vectors along the local magnetization directions on i and j sites), the AFM order sets in by including all interactions at least up to $R/a \approx 2.06$ (7-th shell); accounting of the higher shells does not affect the $M(T)$ behavior anymore. Such a superposition of the strong nearest parallel and the weaker long-range antiparallel exchange interactions typically allows for the long-range spin-spiral order. Its direction ($\mathbf{q} \parallel [001]$) follows from the symmetry reasons: the 7-th shell, critical for setting up the AFM order, contains 8 atoms at $\mathbf{R} = (\pm a, 0, \pm c)$ and $(0, \pm a, \pm c)$, situated above and below the ab -plane of the central atom. The corresponding Neel temperature can be estimated from the peak of the magnetic specific heat $C_V(T)$ computed for the largest cluster size ($T_N \approx 350$ K at $R/a \approx 2.87$, see the inset in Supplementary Figure 2 b). This again reasonably agrees with experimental $M(T)$ slope change.

Supplementary References

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