Supplementary information for Insight into the electronic structure of the centrosymmetric skyrmion magnet $GdRu_2Si_2$

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A. Calculated bulk band structure

To reveal the band structure of $GdRu_2Si_2$ with helical AFM magnetic structure, we started from paramagnetic calculation where trivalent Gd potential is used in which strongly localized valence $4f$ electrons are treated as core states (Fig. S1a). The spectra of paramagnetic phase calculated along high symmetry directions of the bulk Brillouin zone (Fig. S1a, top) using the Heyd-Scuseria-Ernzerhof (HSE06) screened hybrid functional [1] and that obtained within bare GGA-PBE calculation are demonstrated in Figs. S1 a and b, respectively. As can be seen from comparison of these spectra the latter one does not reproduce well the more accurate HSE06 result, especially at the Fermi level. Applying DFT-1/2 corrections over GGA-PBE allows receiving more accurate description of the nonmagnetic spectrum in this energy range (Fig. S1d). Scalar-relativistic spectrum of AFM $GdRu_2Si_2$ was calculated with tetravalent Gd potential in which the $4f$ electrons are treated as valence states within the HSE06 approach (Fig. S1e). Fig. S1e also presents projections of weights of the Gd, Ru, and Si orbitals (left, middle, and right subpanels, respectively). As can be seen the Gd $4f$ localized band, located at ~ -7.5 eV, hybridizes with Si p_{xy} orbitals. Si s and p_z states contribute to the bottom of the main valence band, which is mainly formed by Ru d orbitals. However, the Gd d states also present near the Fermi level (E_F) where they have most pronounced weights in the vicinity of the M point. Thus we can conclude the Gd d are the orbitals which mediate magnetization of the bulk bands near the Fermi level via Gd $f-d$ hybridization. To mimic the HSE06 band structure the simplified GGA+U approach was applied (see details in the Method section). The GGA+U spectrum as obtained within PBE calculation reasonably well reproduces the position of the Gd $f-d$ band (Fig. S1f). However, like in the nonmagnetic case, the spectrum at E_F differs from that calculated within HSE06 approach as well as position of the deep Si p_{xy} states. The partial ionization of Si potential within DFT-1/2 method resolves both these problems (Fig. S1g).

B. Magnetic interaction

Exchange constants were calculated using the magnetic force theorem as it is implemented within the multiple scattering theory [2]. To describe localized Gd $4f$ electrons we used two different DFT functionals, a GGA+U

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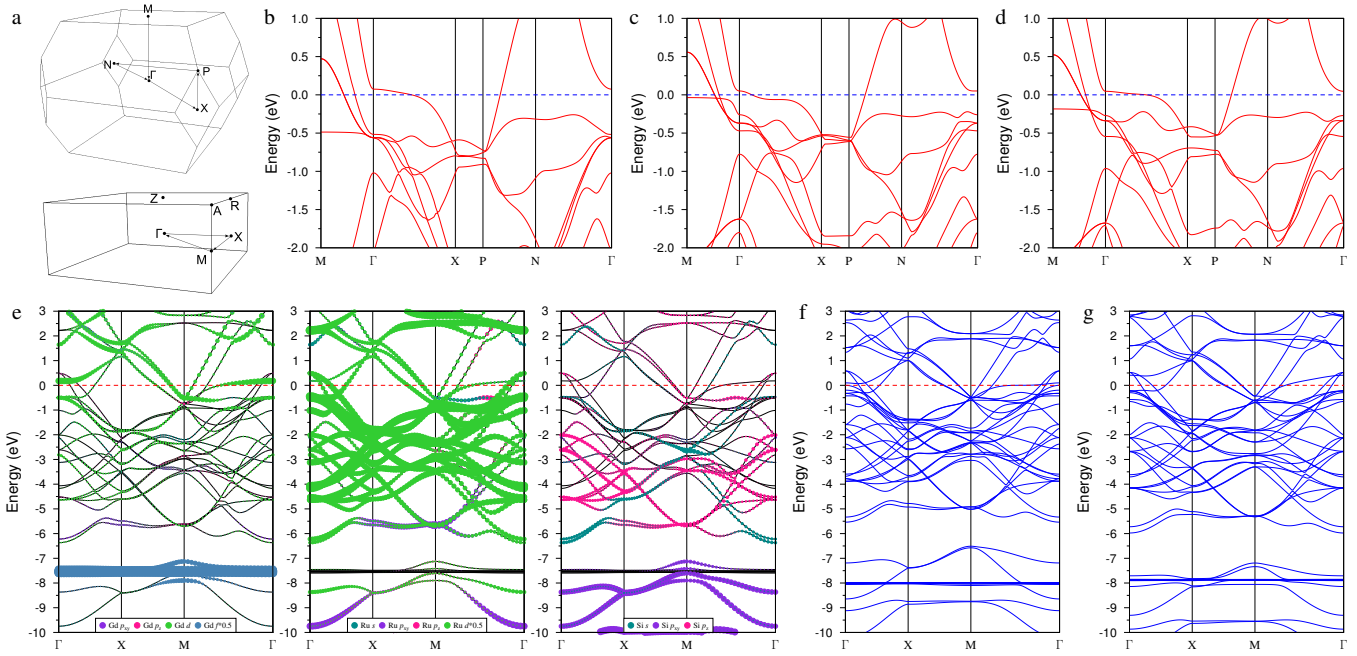

FIG. S1. (a) Bulk Brillouin zones for paramagnetic primitive (top) and for AFM tetragonal (bottom) cells. Arrows show the paths along which electronic spectra are calculated. (b) Bulk energy spectrum of paramagnetic $GdRu_2Si_2$ where Gd states are treated as core electrons as obtained with HSE06 hybrid functional. (c) The spectrum of nonmagnetic $GdRu_2Si_2$ calculated within bare GGA-PBE approach. (d) The same but within DFT-1/2 approach. (e) Scalar-relativistic spectrum of AFM $GdRu_2Si_2$ calculated within HSE06 approach with weights of the Gd (left), Ru (middle), and Si (right) orbitals. (f) Scalar-relativistic spectrum of AFM phase calculated within GGA-PBE with $U = 6.7$ eV and $J = 0.7$ eV. (g) The spectrum calculated within GGA-PBE with $U = 6.7$ eV and $J = 0.7$ eV and quaternary ionized Si p states within DFT-1/2 approach.

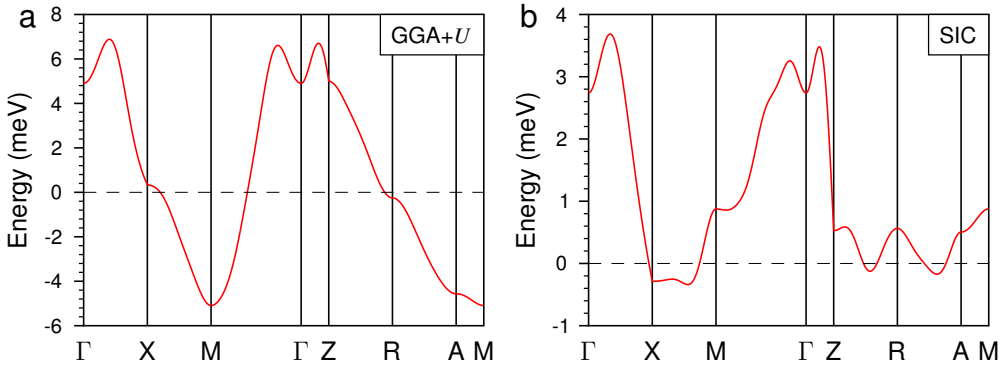

FIG. S2. $J(\vec{Q})$ calculated along the high symmetry directions of the BZ of the tetragonal cell. The global maximum of $J(\vec{Q})$ corresponds to the total energy minimum. Results are presented for a GGA+U (a) and self-interaction correction (SIC) (b) DFT functionals.

approach [3] and a self-interaction correction (SIC) method [4–6]. The results of our calculations are presented in Fig. S2. Both approaches provide a similar behaviour of $J(\vec{Q})$ and the $J(\vec{Q})$ maximum occurs at the same wave vector $Q \approx 0.2\ (2\pi/a)$ along $\Gamma - \mathrm{X}$ direction. However, the magnitude of J's calculated within the SIC method is significantly smaller than that obtained using the GGA+U approximation. The reason is that the SIC method overestimates localization of corrected orbitals. Therefore, in our manuscript we rely on the results obtained within the GGA+U approach.

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