Supporting Information

Two-dimensional ferromagnetic semiconductors of rare-earth Janus 2H-GdIBr monolayer with large valley polarization

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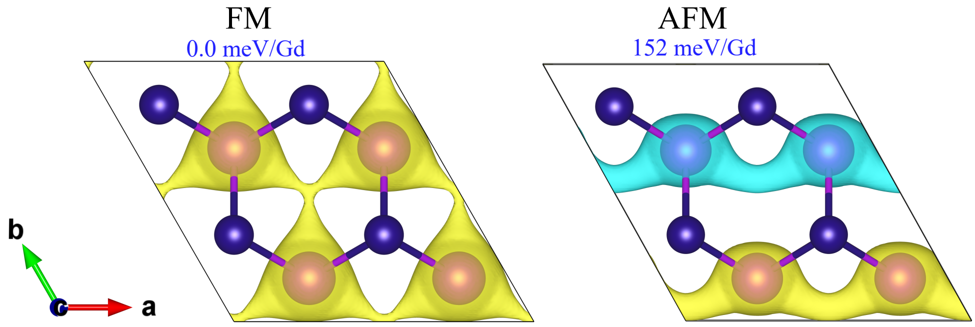

Figure S1. Spin density of 2×2×1 Janus 2H-GdIBr monolayer for different magnetic configurations. Yellow (blue) color refers to the spin up (down) charge density. The isosurface is 0.0094 e/Born^3.

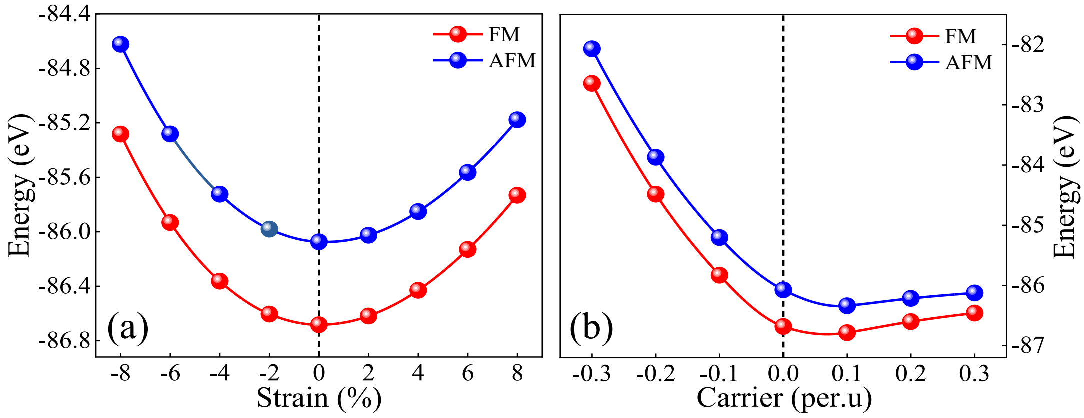

Figure S2. (a) Strain and (b) charge carrier dependence of the energy in FM and AFM states for the 2×2×1 Janus 2H-GdIBr monolayer.

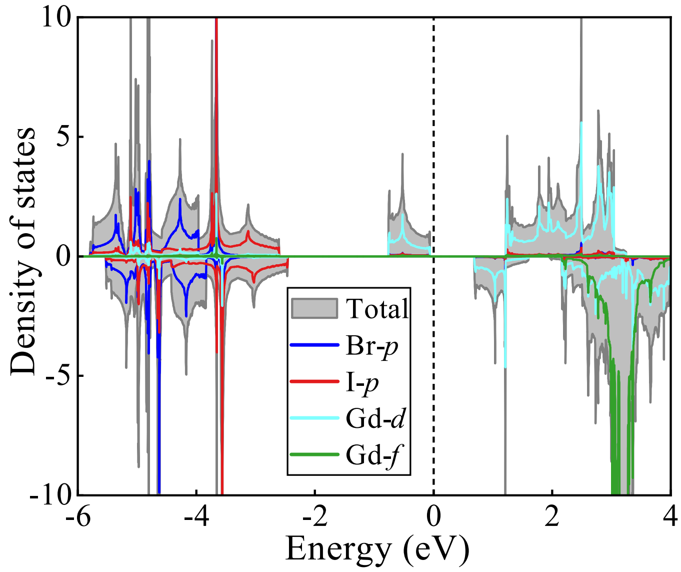

Figure S3. The spin-resolved PDOSs of Janus 2H-GdIBr monolayer.

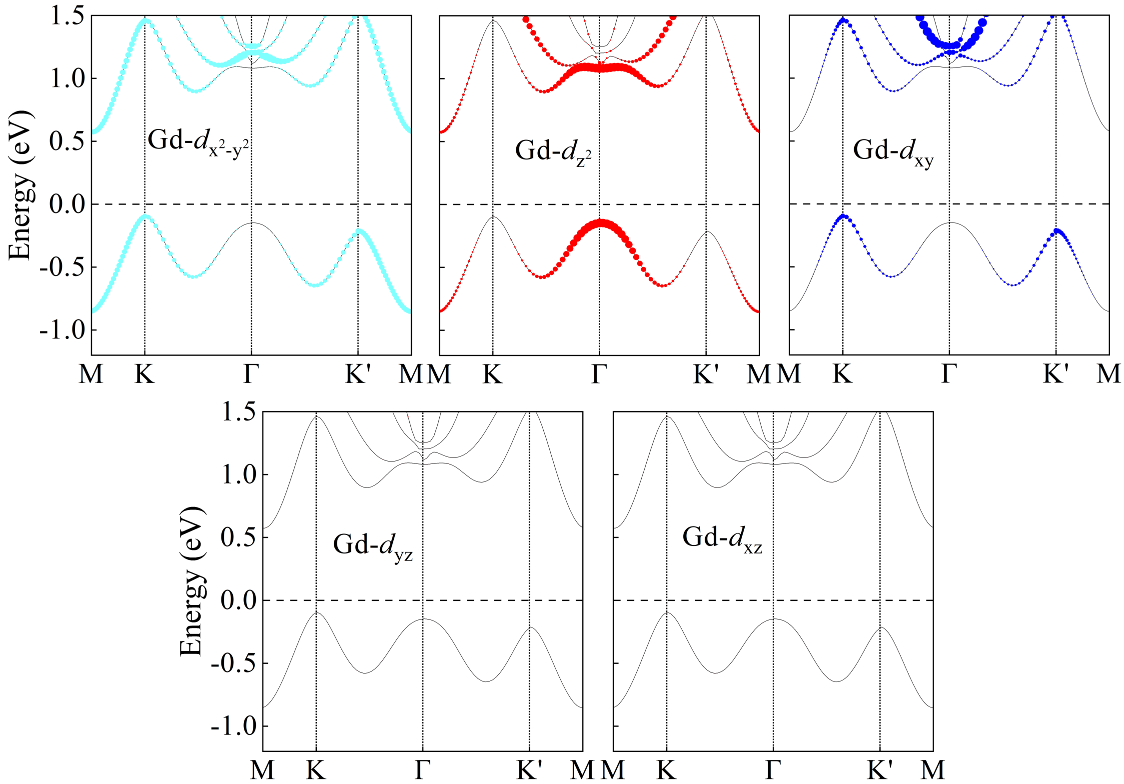

Figure S4. Projected band structure of Gd-*d* orbitals for the Janus 2H-GdIBr monolayer with SOC. The Fermi level is set to zero.

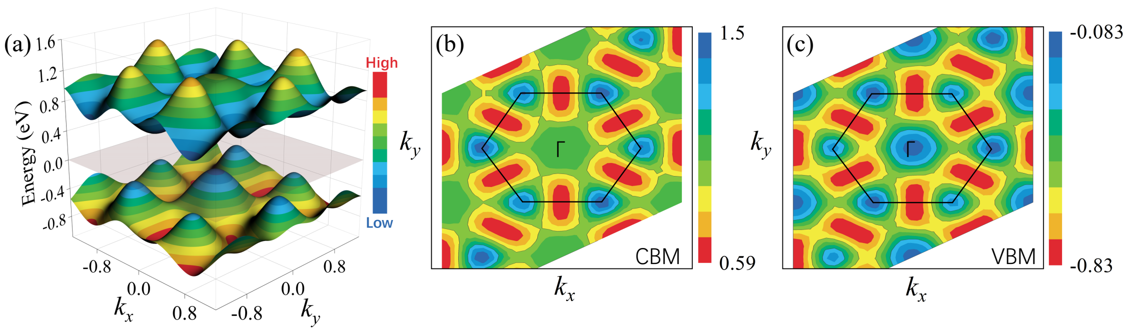

Figure S5. (a) 3D band structure as well as the 2D projected band structure of (b) CBM and (c) VBM at the k_xk_y-plane for the Janus 2H-GdIBr monolayer with considering SOC. The different colors in color bar show different iso-values. The blue and red color correspond to the smallest and largest energy values, respectively.

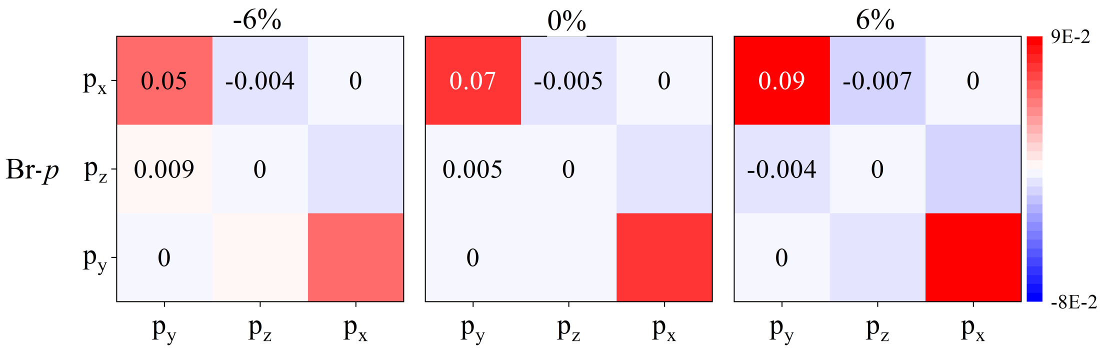

Figure S6. Br-p orbital resolved MAEs for the Janus 2H-GdIBr monolayer at the strains of –6%, 0% and 6%.

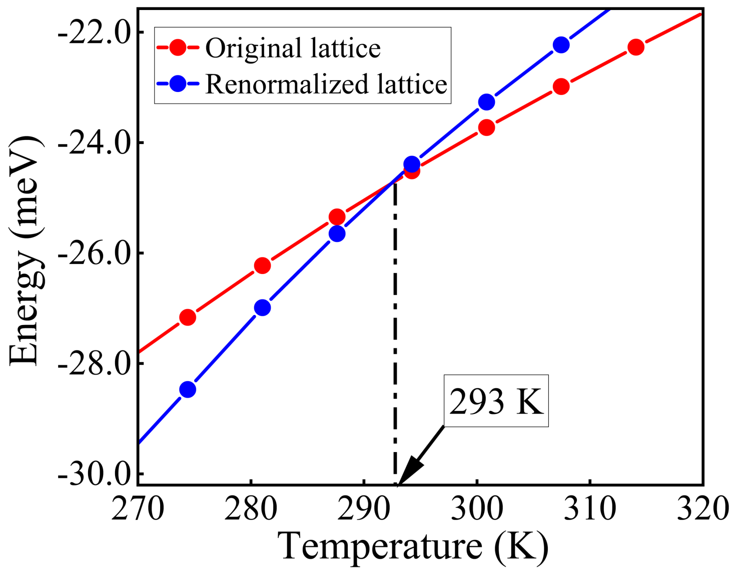

Figure S7. Nearly linear evolution of the internal energy per unit cell in the original 16×16 superlattice of spins (red) and the renormalized 32×32 superlattice of quasi-particles (blue). Lines are only to guide the eye.

The internal energies of the original spin model and the renormalized quasiparticle model are represented by the red and blue dots, respectively. The transition point of the renormalization group is the intersection of the two energies, and the corresponding horizontal coordinate value represents the T_c of the current system.

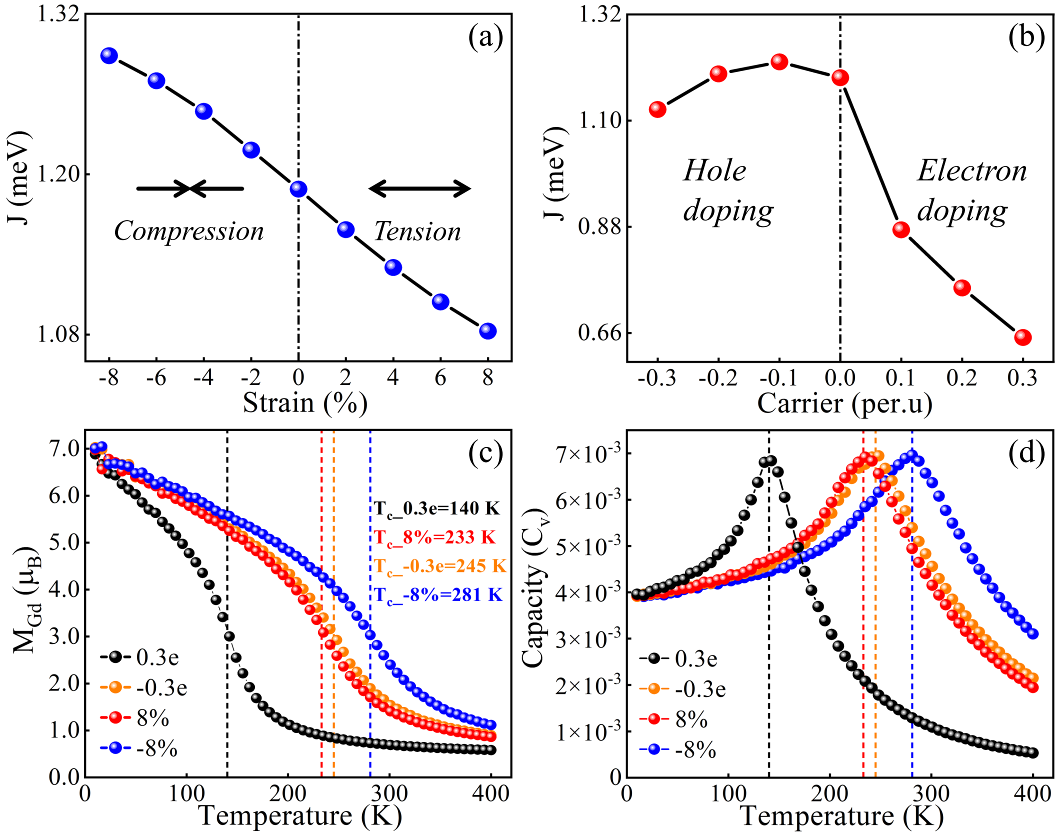

Figure S8. Dependences of exchange parameter *J* on (a) biaxial strain and (b) charge carrier doping for the Janus 2H-GdIBr monolayer. Dependences of (c) magnetic moment and (d) heat capacity on the temperature for the Janus 2H-GdIBr monolayer at the strains of ±8% and with carrier concentration of ±0.3e per.u..

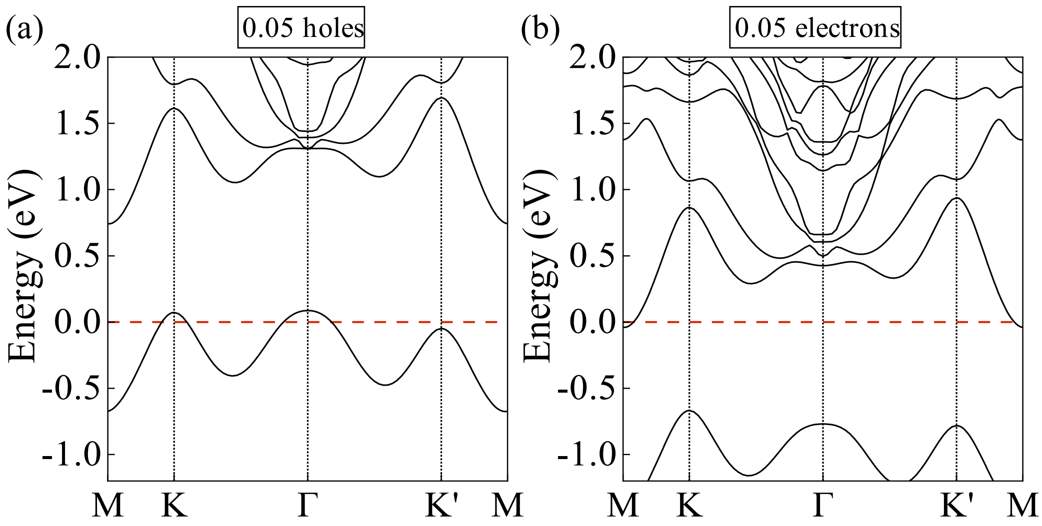

Figure S9. The band structures Janus 2H-GdIBr monolayer with (a) 0.05 holes and (b) 0.05 electrons doping. The Fermi level is set to zero.

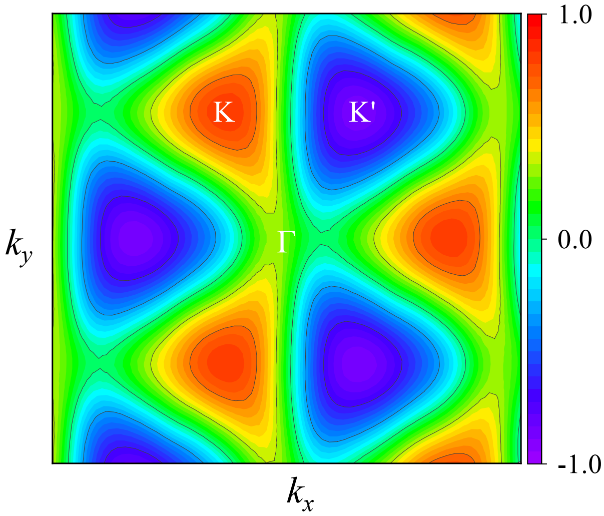

Figure S10. The circular polarization η(k) of the optical transition between the VBM and CBM in the first Brillouin zone.

In non-magnetic MoS_2 and magnetic 2H-VSeS, there is a valley-selective circular dichroism, which can selectively excite the valley carriers at K or K' with different circularly polarized light. The calculated degree of circular polarization $\eta^{V,C}(k)$, which is given by:

$$\eta^{V,C}(k)=\frac{\left|P_{+}^{V,C}(k)\right|^{2}-\left|P_{-}^{V,C}(k)\right|^{2}}{\left|P_{+}^{V,C}(k)\right|^{2}+\left|P_{-}^{V,C}(k)\right|^{2}}$$

The calculated interband transition matrix $P_{\pm}^{V,C}(k)=\left\langle\psi_{k}^{V}\right|\hat{P}_{\pm}\left|\psi_{k}^{C}\right\rangle$, where $\hat{P}_{\pm}=(\hat{P}_{x}\pm i\hat{P}_{y})/\sqrt{2}$. It is found that $\eta^{V,C}(k)$ is almost 1(–1) at and near K(K') point, which distinguishes the absorption of left and right circularly polarized (σ^{+} and σ^{-}) light between the valence and conduction bands. This suggests that left (σ^{+}) and right (σ^{-}) circularly polarized light excites the K and K' valleys.