Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Tunable Electrical Properties and Multiple-phases of Ferromagnetic GdS_2, $GdSe_2$ and Janus GdSSe Monolayers

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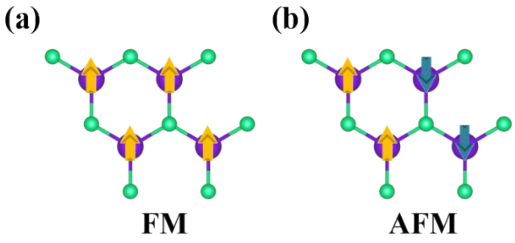

Figure S1. (a) Ferromagnetic (FM) and (b) antiferromagnetic (AFM) configurations of GdS_2, $GdSe_2$ and Janus GdSSe monolayers.

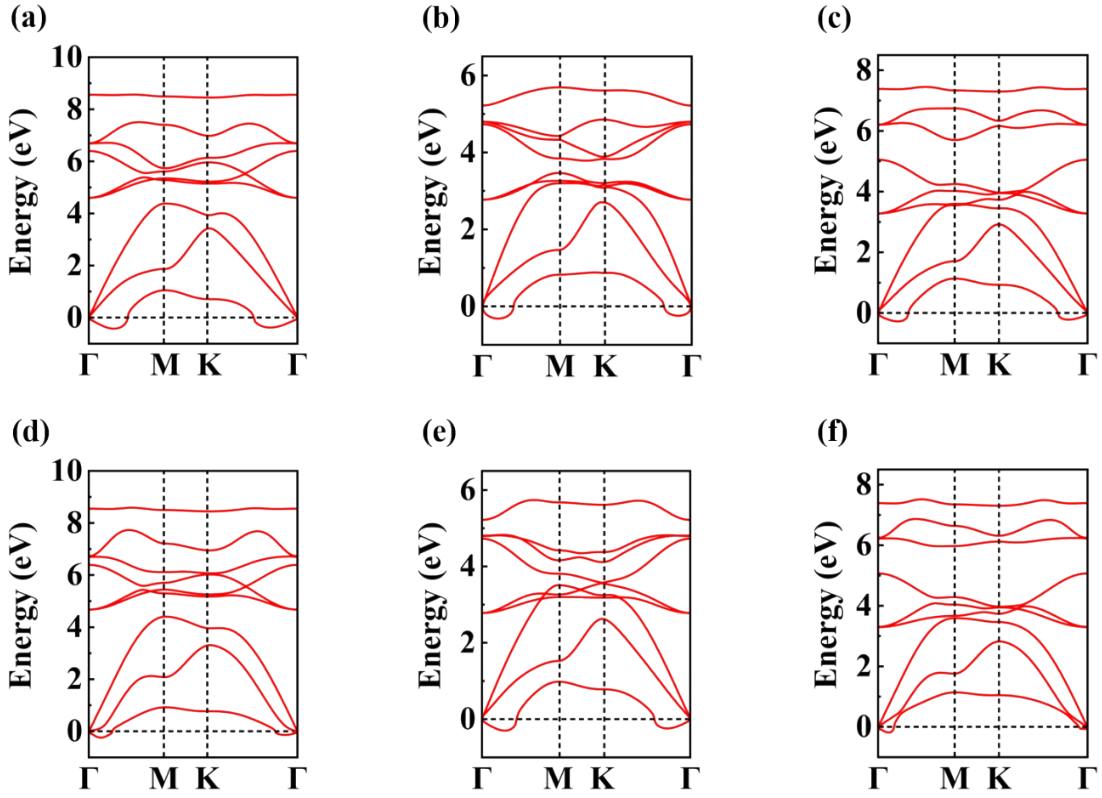

Figure S2. Calculated phonon spectrums of (a,d) GdS_2, (b,e) $GdSe_2$ and (c,f) Janus GdSSe monolayers by adopting (a-c) 2×2×1 and (d-f) 3×3×1 supercells.

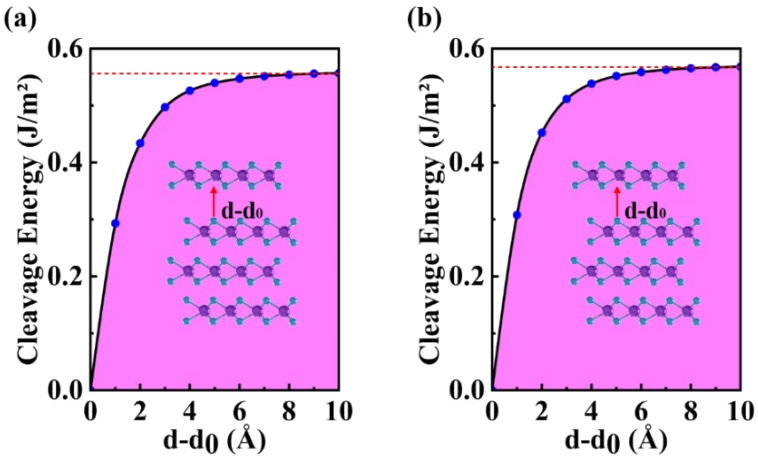

Figure S3. Calculated exfoliation energy of (a) GdS_2 and (b) $GdSe_2$ monolayers.

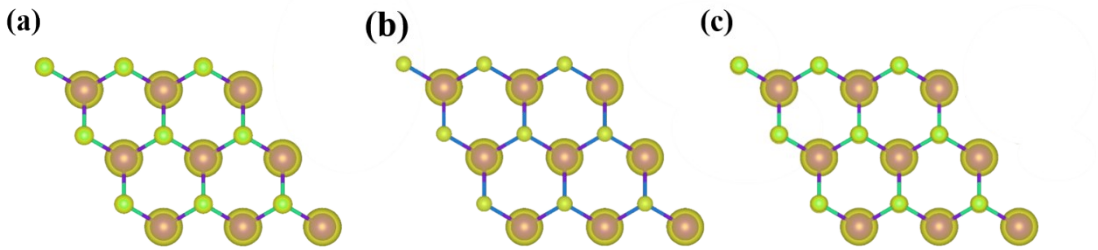

Figure S4. Spin-resolved charge density of (a) GdS_2, (b) $GdSe_2$ and (c) Janus GdSSe monolayers in ferromagnetic (FM) configuration. The yellow color represents the spin-up charge. The isosurface value is set as 0.003 e $Å^{-3}$.

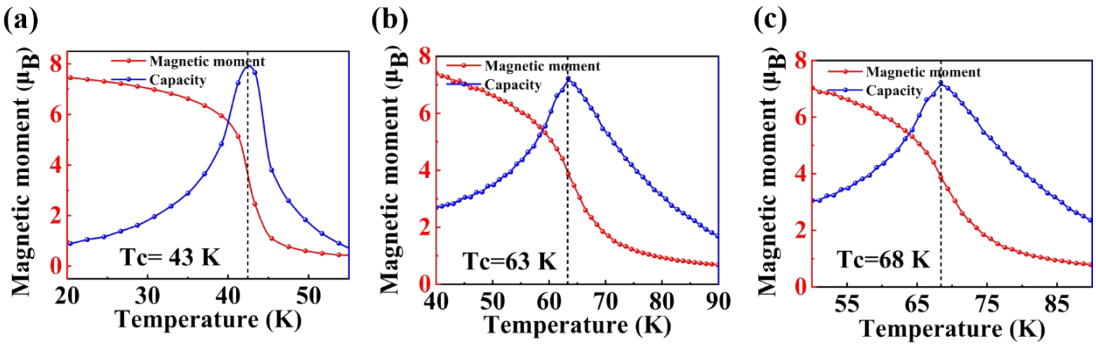

Figure S5. The variation of magnetic moment of Gd atoms and the specific heat as a function of temperature for (a) GdS_2, (b) $GdSe_2$ and (c) Janus GdSSe monolayers based on the Heisenberg model.

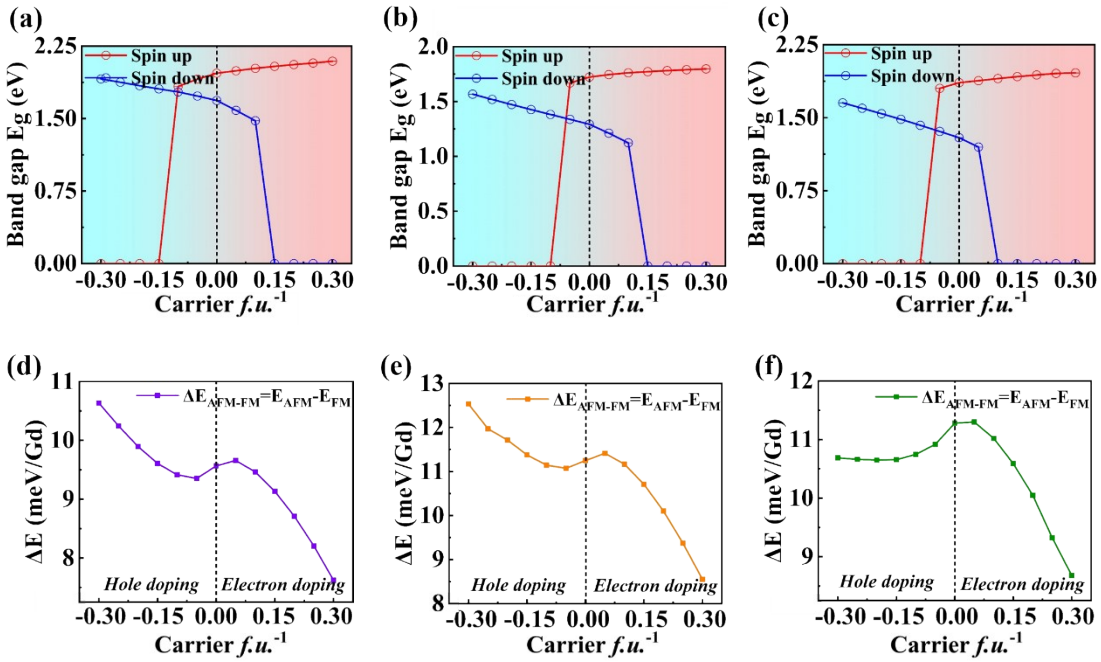

Figure S6. The band gaps of spin-up and spin-down channels of (a) GdS_2, (b) $GdSe_2$ and (c) Janus GdSSe monolayers under carrier doping from -0.3 to 0.3e *f.u.*$^{-1}$, and the exchange energy ($\Delta E_{AFM-FM} = E_{AFM} - E_{FM}$) of (d) GdS_2, (e) $GdSe_2$ and (f) Janus GdSSe monolayers. Positive/negative value means electron/hole doping.

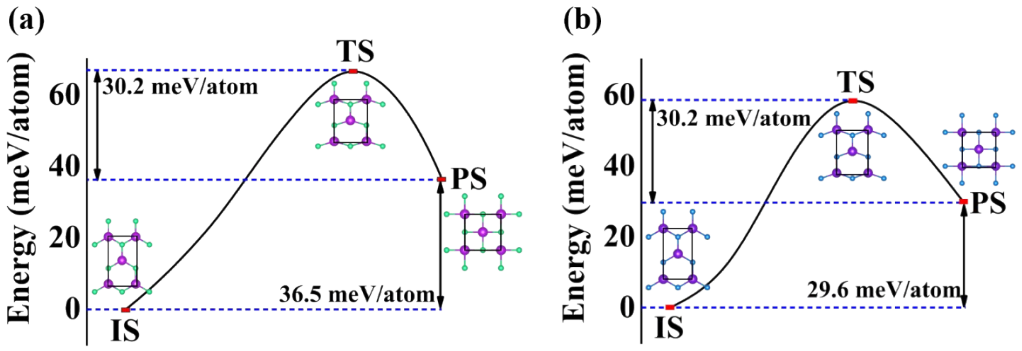

Figure S7. Minimum energy path for the structural phase transitions between initial states (IS) and parastatic states (PS) of (a) GdS_2 and (b) $GdSe_2$ monolayers calculated by SSNEB method.

Table S1. The structural parameters (Å) of the initial state (IS) and paraelastic state (PS), energy barriers (meV/atom) and reversible ferroelastic strain during ferroelastic transformation of GdS_2 and $GdSe_2$ and GdSSe monolayers.

	Initial state		Paraelastic state		Energy barrier	Strain
	a_{IS}	b_{IS}	a_{PS}	b_{PS}		
GdS_2	7.15	4.13	5.13	5.13	66.2	73.2%
$GdSe_2$	7.40	4.27	5.33	5.33	60.8	73.2%
GdSSe	7.27	4.20	5.29	5.29	62.3	73.2%

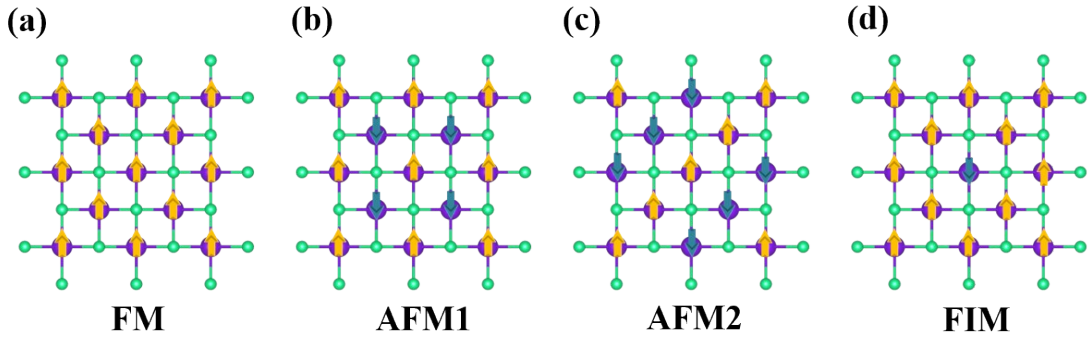

Figure S8. (a) Ferromagnetic (FM), two antiferromagnetic (AFM) of (b) AFM1 and (c) AFM2, and (d) ferrimagnetic (FIM) configurations for the square lattices of GdS_2 and $GdSe_2$ and GdSSe monolayers.

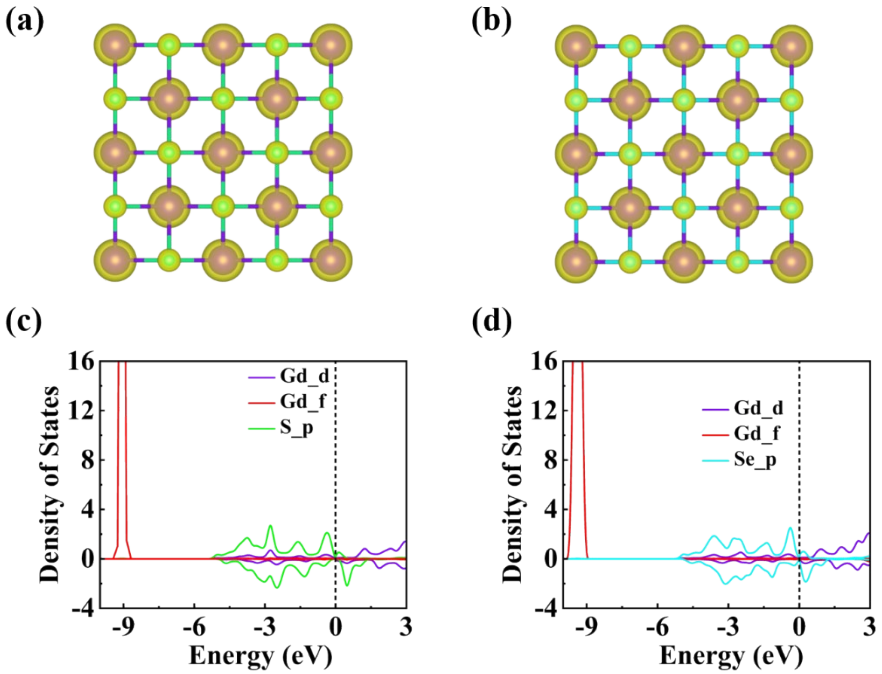

Figure S9. Spin-resolved charge density of the square lattices of the (a) GdS_2, (b) $GdSe_2$ monolayers in ferromagnetic (FM) configuration. The yellow color represents the spin-up charge. The isosurface value is set as 0.003 e $Å^{-3}$. Orbital-projected density of states of the square lattices of the (a) GdS_2, (b) $GdSe_2$ monolayers.

Table S2. The energies for the ferromagnetic (FM), antiferromagnetic (AFMI and AFM2) and ferrimagnetic (FIM) configurations of the square lattices of the GdS_2 (P-GdS_2), $GdSe_2$ (P-$GdSe_2$) and GdSSe (P-GdSSe) monolayers. Magnetic anisotropic energies (MAE) of the P-GdS_2, P-$GdSe_2$ and P-GdSSe monolayers calculated by the DFT+U (U_{eff} = 8 eV) method with considering spin orbit coupling (SOC). Negative value represents the easy magnetization axis along the in-plane direction.

	FM	AFM1	AFM2	FIM	MAE
		(meV/Gd atom)			(μeV/Gd atom)
P-GdS_2	0	111.4	109.2	5.1	-46.5
P-$GdSe_2$	0	53.3	53.3	11.2	-340.5

Results of PBE+U method

Considering with the 4f orbitals of the Gd atoms, we adopted the PBE+U method. The related Hubbard on-site Coulomb parameter U of 9.2 eV and exchange interaction parameter J of 1.2 eV were chosen for U_{eff} (8 eV), which is defined as U_{eff} = U-J. Previous results have shown that the calculated lattice constant (3.99 Å) of $GdGe_2$ monolayer at the U_{eff} value of 8 eV matches the experimental observation (4.01 Å) [1]; the rocksalt structure of bulk GdN calculated at the similar U_{eff} value match the experimental observation[2]; similarly U_{eff} value successfully reproduce the magnetic moment of Gd atom in bulk GdI_2.[3]

POSCAR files of the hexagonal honeycomb lattice of GdS_2, $GdSe_2$ and Janus GdSSe monolayers

GdS_2-POSCAR

1.0

4.1292424202 0.0000000000 0.0000000000

-2.0646636004 3.5760219793 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd S

1 2

Direct

0.666660482 0.333331327 0.499968290

0.333329931 0.666662053 0.570505571

0.333329513 0.666666587 0.429506111

$GdSe_2$-POSCAR

1.0

4.2736334801 0.0000000000 0.0000000000

-2.1368600116 3.7010738531 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd Se

1 2

Direct

0.666660522 0.333331253 0.499965620

0.333329957 0.666662313 0.576242542

0.333329536 0.666666436 0.423771811

GdSSe-POSCAR

1.0

4.1989555359 0.0000000000 0.0000000000

-2.0996223742 3.6363808907 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd S Se

1 1 1

Direct

0.666656841 0.333332286 0.504613113

0.333332132 0.666660704 0.570961428

0.333330959 0.666666933 0.424415493

POSCAR files of the square lattice of GdS_2, $GdSe_2$ and Janus GdSSe monolayers

GdS_2-POSCAR

1.0

5.1326365471 0.0000000000 0.0000000000

0.0000000000 5.1326365471 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd S

2 4

Direct

0.000000000 0.000000000 0.492970984

0.500000000 0.500000000 0.492970984

0.000000000 0.500000000 0.435988444

0.500000000 0.000000000 0.435988444

0.000000000 0.500000000 0.549960488

0.500000000 0.000000000 0.549960488

$GdSe_2$-POSCAR

1.0

5.3323807716 0.0000000000 0.0000000000

0.0000000000 5.3323807716 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd Se

2 4

Direct

0.000000000 0.000000000 0.492969844

0.500000000 0.500000000 0.492969844

0.000000000 0.500000000 0.431552639

0.500000000 0.000000000 0.431552639

0.000000000 0.500000000 0.554397515

0.500000000 0.000000000 0.554397515

GdSSe-POSCAR

1.0

5.2285308838 0.0000000000 0.0000000000

0.0000000000 5.2285308838 0.0000000000

0.0000000000 0.0000000000 20.0000000000

Gd Se S

2 2 2

Direct

0.000000000 0.000000000 0.497041480

0.500000000 0.500000000 0.497041480

0.000000000 0.500000000 0.431907849

0.500000000 0.000000000 0.431907849

0.000000000 0.500000000 0.549970629

0.500000000 0.000000000 0.549970629

The Used software and input files of Monte Carlo simulations

Monte Carlo method[4] based on the standard Metropolis-Hasting algorithm [5]has been widely adopted to estimate the Curie temperature of FM materials. The software what we use for Monte Carlo simulation is MCSOLVER.[6] In practice, the software divides the continue pare of Hamiltonian into twenty bins, each site thus can have at most twenty possible magnetic moments, and each site has its own energy according to the Hamiltonian. In one Monte Carlo step, the software first chose one site then decide which magnetic moment it would take, stochastically. Based on the energy variation, the software would accept or reject the operation with appropriate possibility and then repeat this operation for the next site until the magnetic moment of all sites in the supercell were taken. There is 32×32×1 supercell in our simulations, using > 10^8 sweeps (> 10^{14} Monte Carlo steps) to get converged results. Below are the input files for our Monte Carlo simulations of GdS_2, $GdSe_2$ and Janus GdSSe monolayers with hexagonal honeycomb lattices.

The input files of GdS_2 monolayer

This is mcsolver's save file, version: 2.3

Lattice:

1.000000000 0.000000000 0.000000000

-0.500000000 0.866000000 0.000000000

0.000000000 0.000000000 1.000000000

Supercell used in MC simulations:

32 32 1

Orbitals in cell:

1

Positions, initial spin states and onsite-anisotropy of every orbital:

orb 0: type 0 spin 4.000000000 pos [0.666670000 0.333330000 0.500000000] Dx 0.000000000 Dy 0.000000000 Dz 1.553000000 h 0.000000000

Bonds:

4

id, source, target, overLat, exchange matrix elements of each bond:

bond 0: Jx -1.726700000 Jy -1.726700000 Jz -1.726700000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [1 0 0]

bond 1: Jx -1.726700000 Jy -1.726700000 Jz -1.726700000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [1 1 0]

bond 2: Jx -1.726700000 Jy -1.726700000 Jz -1.726700000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 1 0]

bond 3: Jx -1.726700000 Jy -1.726700000 Jz -1.726700000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 0 0]

Temperature scanning region:

Tmin 10.000000000 Tmax 100.000000000 nT 50

Field scanning region:

Hmin 0.000000000 Hmax 0.100000000 nH 1

Dipole long-range coupling:

alpha 0.000000

Measurement:

measure the correlation function between orb0 and orb0 over [0 0 0]

Supergroup

OrbGroup:1

Supergroup

group0 orb0-orb0

Distribution output frame: 0

Sweeps for thermalization and statistics, and relaxiation step for each sweep:

80000 640000 0

XAxis type:

T

Model type:

Heisenberg

Algorithm:

Metropolis

Ncores:

3

The input files of $GdSe_2$ monolayer

This is mcsolver's save file, version: 2.3

Lattice:

1.000000000 0.000000000 0.000000000

-0.500000000 0.866000000 0.000000000

0.000000000 0.000000000 1.000000000

Supercell used in MC simulations:

32 32 1

Orbitals in cell:

1

Positions, initial spin states and onsite-anisotropy of every orbital:

orb 0: type 0 spin 4.000000000 pos [0.666670000 0.333330000 0.500000000] Dx 0.000000000 Dy 0.000000000 Dz 12.500000000 h 0.000000000

Bonds:

4

id, source, target, overLat, exchange matrix elements of each bond:

bond 0: Jx -2.028000000 Jy -2.028000000 Jz -2.028000000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 0 0]

bond 1: Jx -2.028000000 Jy -2.028000000 Jz -2.028000000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [1 0 0]

bond 2: Jx -2.028000000 Jy -2.028000000 Jz -2.028000000 Jxy 0.000000000 Jxz

0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 1 0]

bond 3: Jx -2.028000000 Jy -2.028000000 Jz -2.028000000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 0 0]

Temperature scanning region:

Tmin 10.000000000 Tmax 100.000000000 nT 50

Field scanning region:

Hmin 0.000000000 Hmax 0.100000000 nH 1

Dipole long-range coupling:

alpha 0.000000

Measurement:

measure the correlation function between orb0 and orb0 over [0 0 0]

Supergroup

OrbGroup:1

Supergroup

group0 orb0-orb0

Distribution output frame: 0

Sweeps for thermalization and statistics, and relaxiation step for each sweep:

80000 640000 0

XAxis type:

T

Model type:

Heisenberg

Algorithm:

Metropolis

Ncores:

3

The input files of Janus GdSSe monolayer

This is mcsolver's save file, version: 2.3

Lattice:

1.000000000 0.000000000 0.000000000

-0.500000000 0.866000000 0.000000000

0.000000000 0.000000000 1.000000000

Supercell used in MC simulations:

32 32 1

Orbitals in cell:

1

Positions, initial spin states and onsite-anisotropy of every orbital:

orb 0: type 0 spin 4.000000000 pos [0.666670000 0.333330000 0.500000000] Dx 0.000000000 Dy 0.000000000 Dz 7.470000000 h 0.000000000

Bonds:

4

id, source, target, overLat, exchange matrix elements of each bond:

bond 0: Jx -2.028000000 Jy -2.028000000 Jz -2.028000000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 0 0]

bond 1: Jx -2.039620000 Jy -2.039620000 Jz -2.039620000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [0 1 0]

bond 2: Jx -2.039620000 Jy -2.039620000 Jz -2.039620000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [1 1 0]

bond 3: Jx -2.039620000 Jy -2.039620000 Jz -2.039620000 Jxy 0.000000000 Jxz 0.000000000 Jyz 0.000000000 Jyx 0.000000000 Jzx 0.000000000 Jzy 0.000000000 orb 0 to orb 0 over [1 0 0]

Temperature scanning region:

Tmin 10.000000000 Tmax 100.000000000 nT 50

Field scanning region:

Hmin 0.000000000 Hmax 0.100000000 nH 1

Dipole long-range coupling:

alpha 0.000000

Measurement:

measure the correlation function between orb0 and orb0 over [0 0 0]

Supergroup

OrbGroup:1

Supergroup

group0 orb0-orb0

Distribution output frame: 0

Sweeps for thermalization and statistics, and relaxiation step for each sweep:

80000 640000 0

XAxis type:

T

Model type:

Heisenberg

Algorithm:

Metropolis

Ncores:

3

References

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