Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information for

Two dimensional Cr_2GaX_4 (X = S, Se, Te)： Half-Metallic Intrinsic Room Temperature Ferromagnets with Large Magnetic Anisotropy

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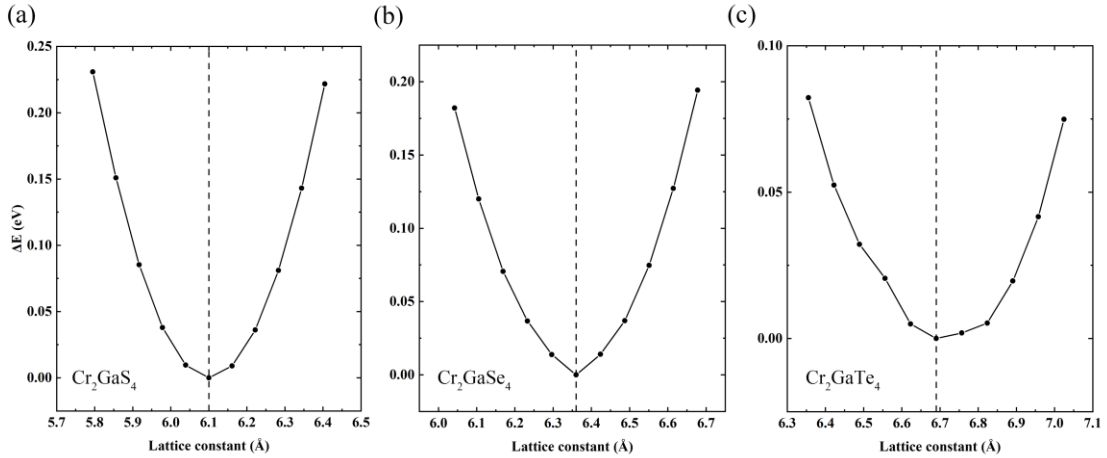

Figure S1: Total energy as a function of lattice constants in the ferromagnetic configurations of (a) Cr_2GaS_4, (b) Cr_2GaSe_4, and (c) Cr_2GaTe_4 monolayers.

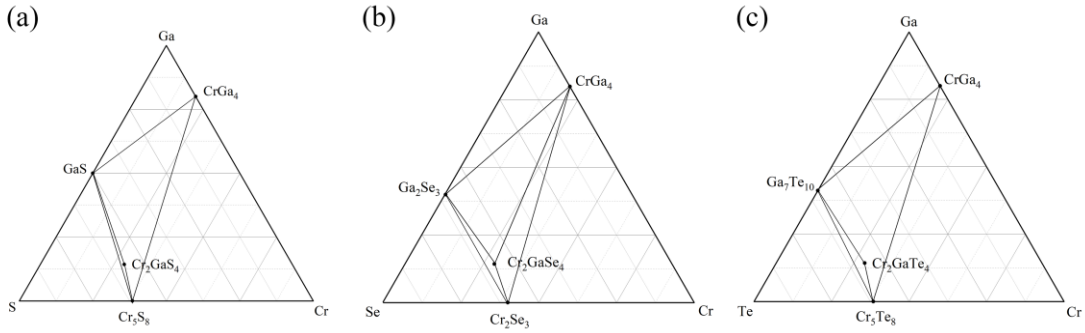

Figure S2: The phase diagram of a ternary (a) Cr-Ga-S, (b) Cr-Ga-Se, and (c) Cr-Ga-Te system.

Table S1: The optimized lattice constants (L, Å), the Cr-Ga bond lengths (d_1, Å), the Cr-X bond lengths (d_2, Å), the Cr-Ga-Cr bond angles (θ_1), the Cr-X-Cr bond angles (θ_2), and the stiffness constants (C, N/m).

	L	d_1	d_2	θ_1	θ_2	C_{11}	C_{12}	C_{66}
Cr_2GaS_4	6.10	3.05	2.40	90	128.26	13.39	-0.328	6.608
Cr_2GaSe_4	6.36	3.19	2.53	90	125.63	6.509	0.772	5.931
Cr_2GaTe_4	6.69	3.35	2.73	90	120.24	3.761	-1.35	6.115

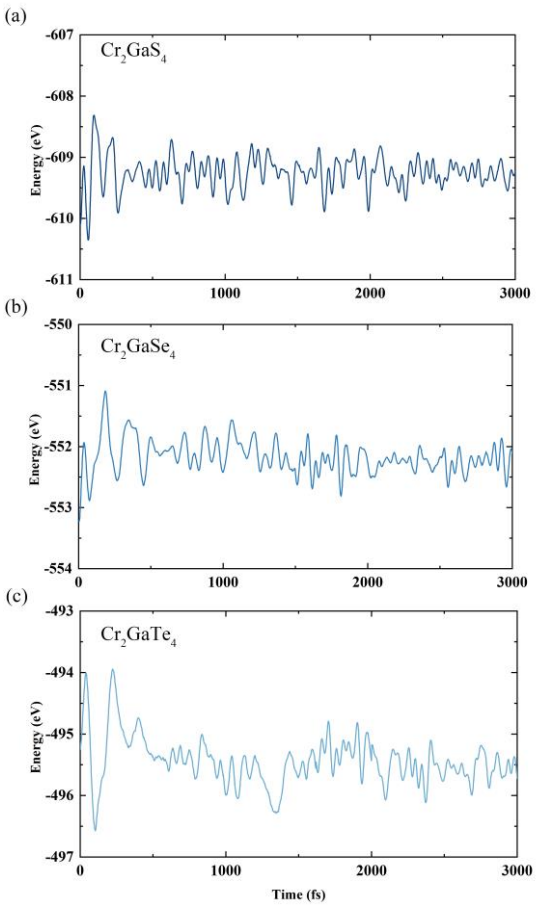

Figure S3: The evolution of total energy of the Cr_2GaX_4 (X = S, Se, Te) monolayers as a function of time at 300 K.

Table S2: Test calculation results of the band gaps (eV) on the spin-down channel and MAE (meV/Cr) of Cr_2GaX_4 (X = S, Se, Te) monolayers whether the van der Waals corrections are adopted with the density functional dispersion correction (DFT-D3).

Cr_2GaS_4	band gap	MAE
without vdW	3.60	0.28
with vdW	3.69	0.29

Cr_2GaSe_4	band gap	MAE
without vdW	3.21	0.55
with vdW	3.20	0.58

Cr_2GaTe_4	band gap	MAE
without vdW	2.37	3.45
with vdW	2.35	3.47

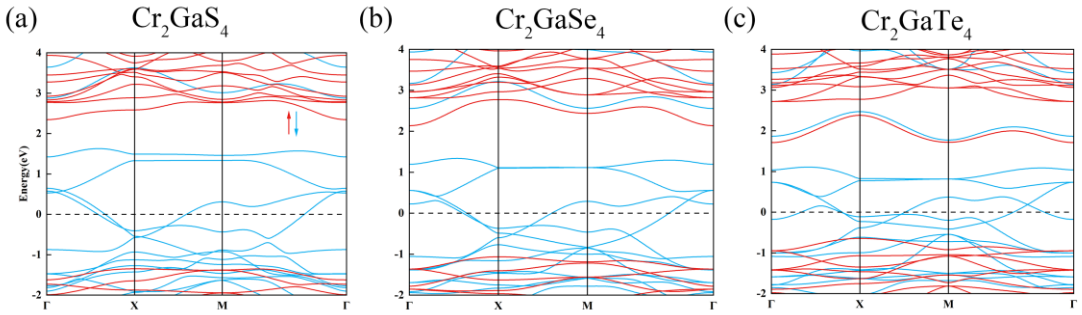

Figure S4: The electronic band structures of the Cr_2GaX_4 (X = S, Se, Te) monolayers for which the van der Waals corrections are adopted with DFT-D3.

Table S3: Test calculation results of the band gaps (eV) on the spin-down channel and MAE (meV/Cr) of Cr_2GaX_4 (X = S, Se, Te) monolayers with the different values of Hubbard *U*.

Cr_2GaS_4	band gap	MAE
U = 3.0 eV	3.60	0.28
U = 3.5 eV	3.96	0.28
U = 4.0 eV	4.01	0.27

Cr_2GaSe_4	band gap	MAE
U = 3.0 eV	3.21	0.55
U = 3.5 eV	3.25	0.52
U = 4.0 eV	3.28	0.54

Cr_2GaTe_4	band gap	MAE
U = 3.0 eV	2.37	3.45
U = 3.5 eV	2.38	3.42
U = 4.0 eV	2.38	3.28

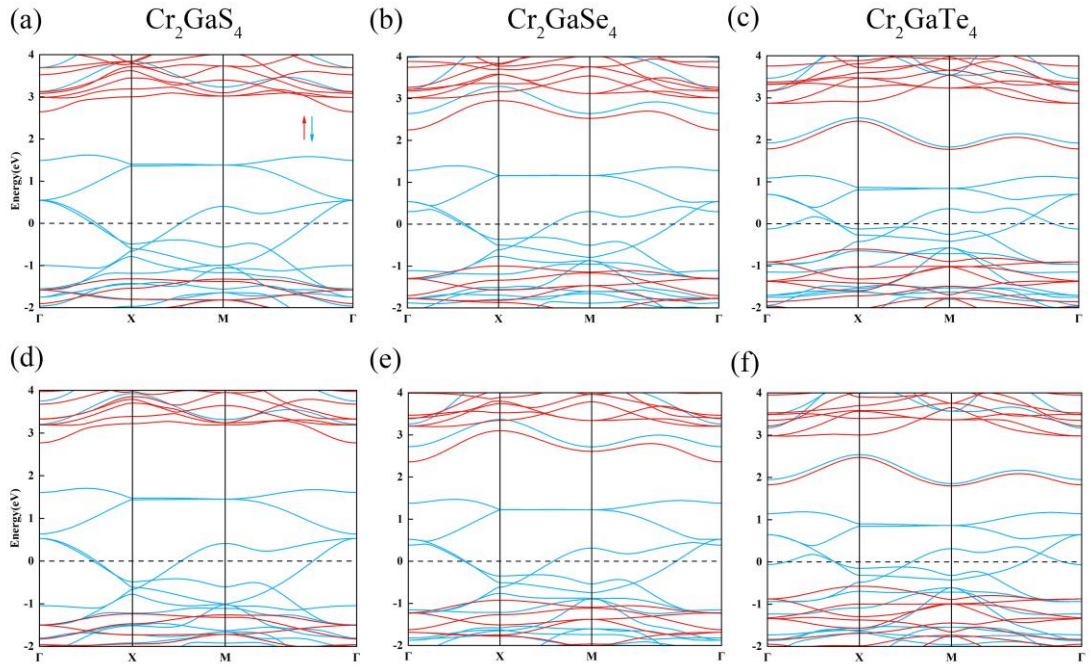

Figure S5: The electronic band structures of the Cr_2GaX_4 (X = S, Se, Te) monolayers, which the values of Hubbard *U* are set to (a-c) 3.5 and (d-f) 4.0 eV.

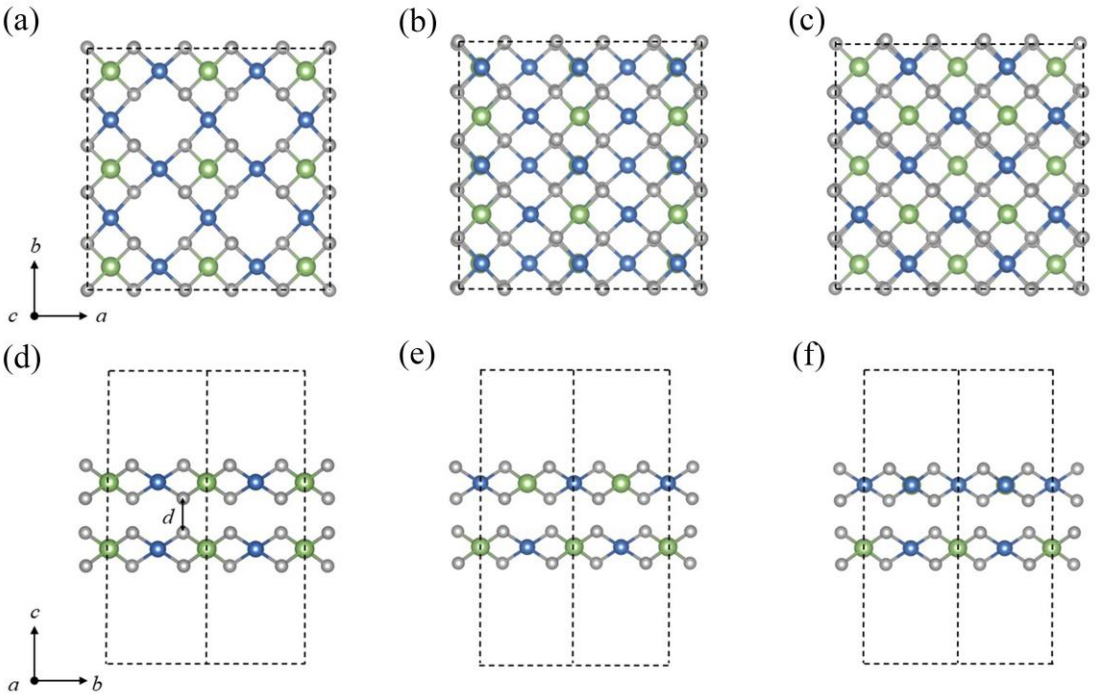

Figure S6: The Cr_2GaX_4 (X = S, Se, Te) bilayers with AA, AB-I, and AB-II stacking. (a-c) Top and (d-f) side views of the optimized structures.

Table S4: The optimized lattice constants (L, Å) and layer distances (d, Å) of the Cr_2GaX_4 (X = S, Se, Te) bilayers.

Cr_2GaS_4	L	d
AA	6.08	3.63
AB-I	6.08	3.58
AB-II	6.08	3.66

Cr_2GaSe_4	L	d
AA	6.33	3.74
AB-I	6.33	3.66
AB-II	6.34	3.76

Cr_2GaTe_4	L	d
AA	6.72	3.99
AB-I	6.69	3.82
AB-II	6.69	3.90

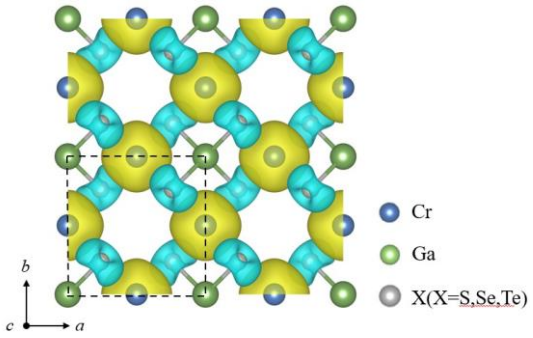

Figure S7: Top view of spin charge density for Cr_2GaX_4 (X = S, Se, Te) with iso-surface values of 0.002 e $bohr^{-3}$. The yellow and cyan colors represent spin-up and spin-down charge accumulation, respectively.

Table S5: The charge transfer (Q, e) by the Bader charge analysis. The negative and positive represent the gain and loss of electrons, respectively.

	Q_{Cr}	Q_{Ga}	Q_X
Cr_2GaS_4	1.41	0.90	-1.02
Cr_2GaSe_4	1.11	1.31	-0.77
Cr_2GaTe_4	0.87	0.58	-0.58

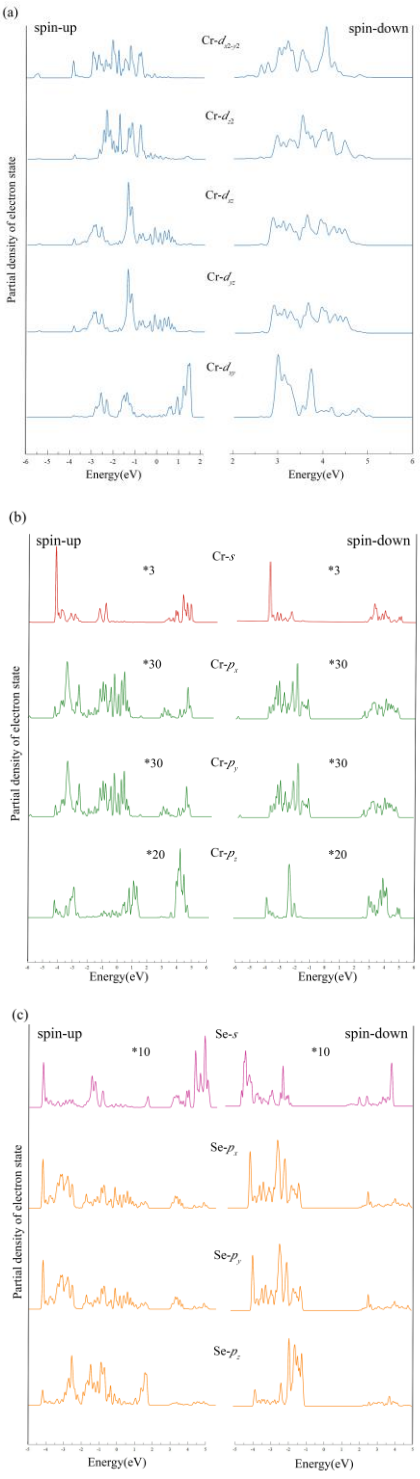

Figure S8: Partial density of electron state (PDOS) of Cr and Se atoms in Cr_2GaSe_4 monolayer. Spin-up and spin-down channels are from left to right. The smaller PDOS are multiplied by appropriate times to gain clear discrimination.