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

Supplemental materials for "**Intrinsic Ferromagnetism with High Curie Temperature in Two Dimensional $XCrY_3$ (X = Al, Ga, In; Y = S, Se, Te) Monolayers.**"

In this Supplemental Material we provide (1) the phonon spectra and the MD simulation results for all $X\mathrm{Cr}Y_3$ (X=Al, Ga, In; Y=S, Se, Te) monolayers, (2) the electronic band structures and PDOSs of all $X\mathrm{Cr}Y_3$ monolayers, (3) the top and side view of deformation charge density distribution of the $\mathrm{AlCrS_3}$ monolayer, (4) the orbital-projected contribution to MAE from the SOC interaction, (5) Magnetic moment and specific heat capacity as a function of temperature by Monte Carlo simulations, and (6) the total energies per cell of monolayer $X\mathrm{Cr}Y_3$ with different magnetic configurations at the GGA+ U level.

Figures S1 and S2 shows that the studied $X\mathrm{Cr}Y_3$ monolayers are kinetically anddynamically stable.

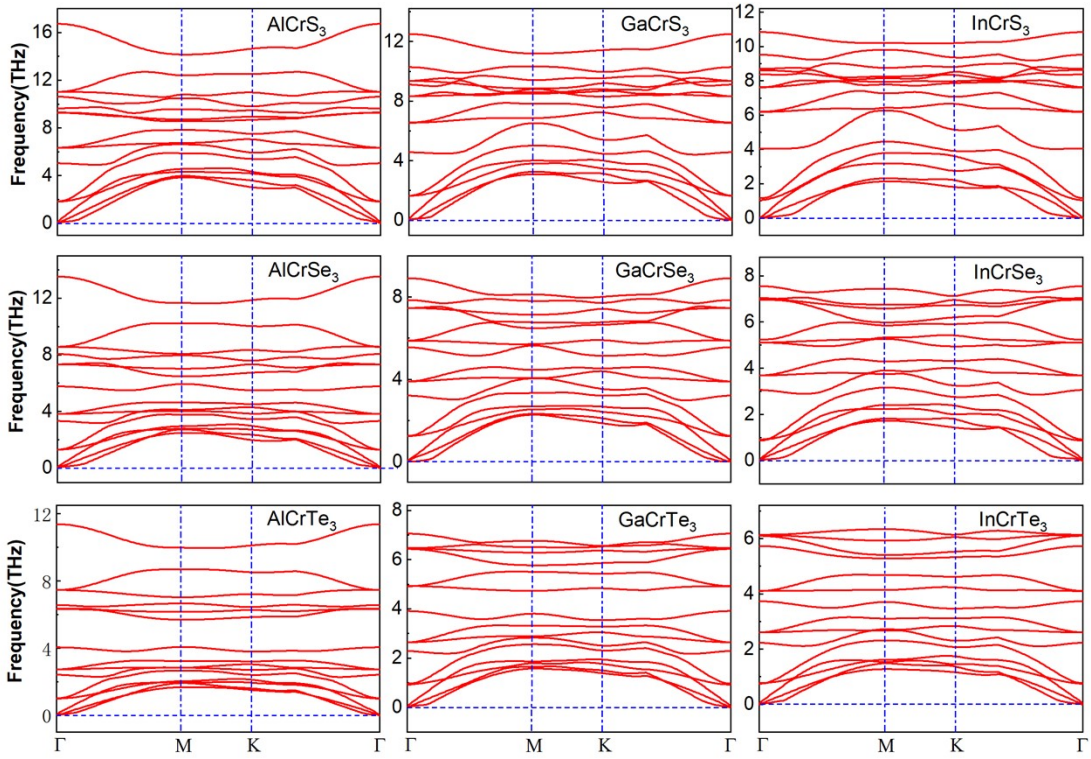

Figure S1. The phonon spectra of all $X\mathrm{Cr}Y_3$ (X=Al, Ga, In; Y=S, Se, Te) monolayers.

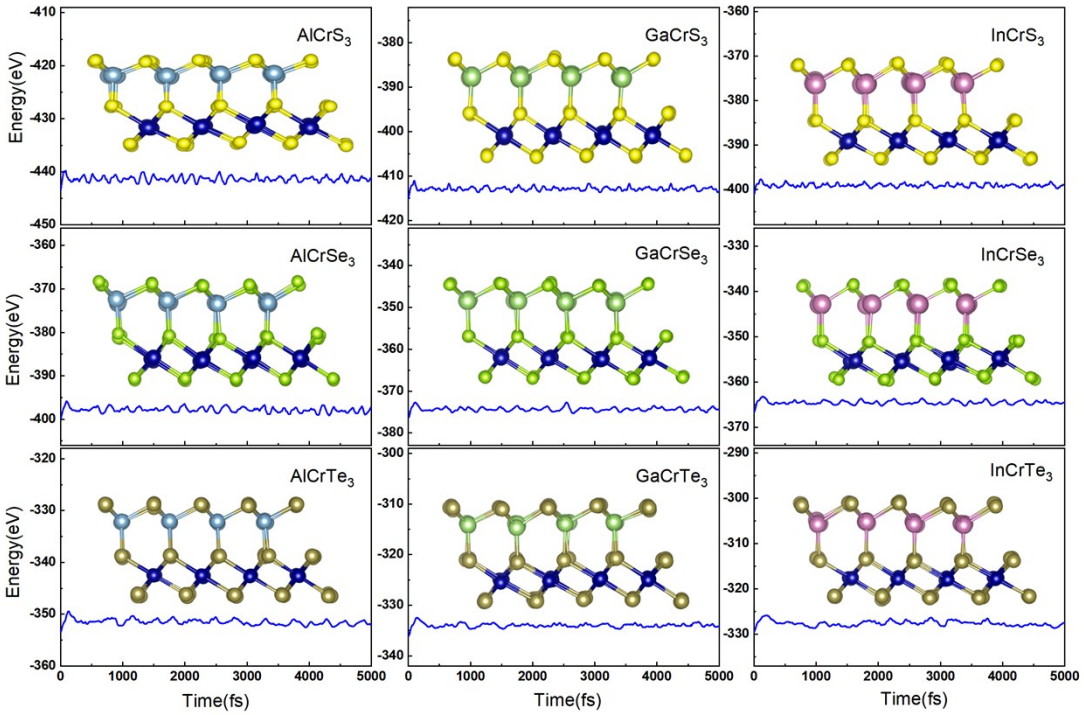

Figure S2. Results of molecular dynamicssimulations of all $XCrY_3$ (X=Al, Ga, In; Y=S, Se, Te) monolayers , as well as the final structures after being heated at 300K for 5ps inserted.

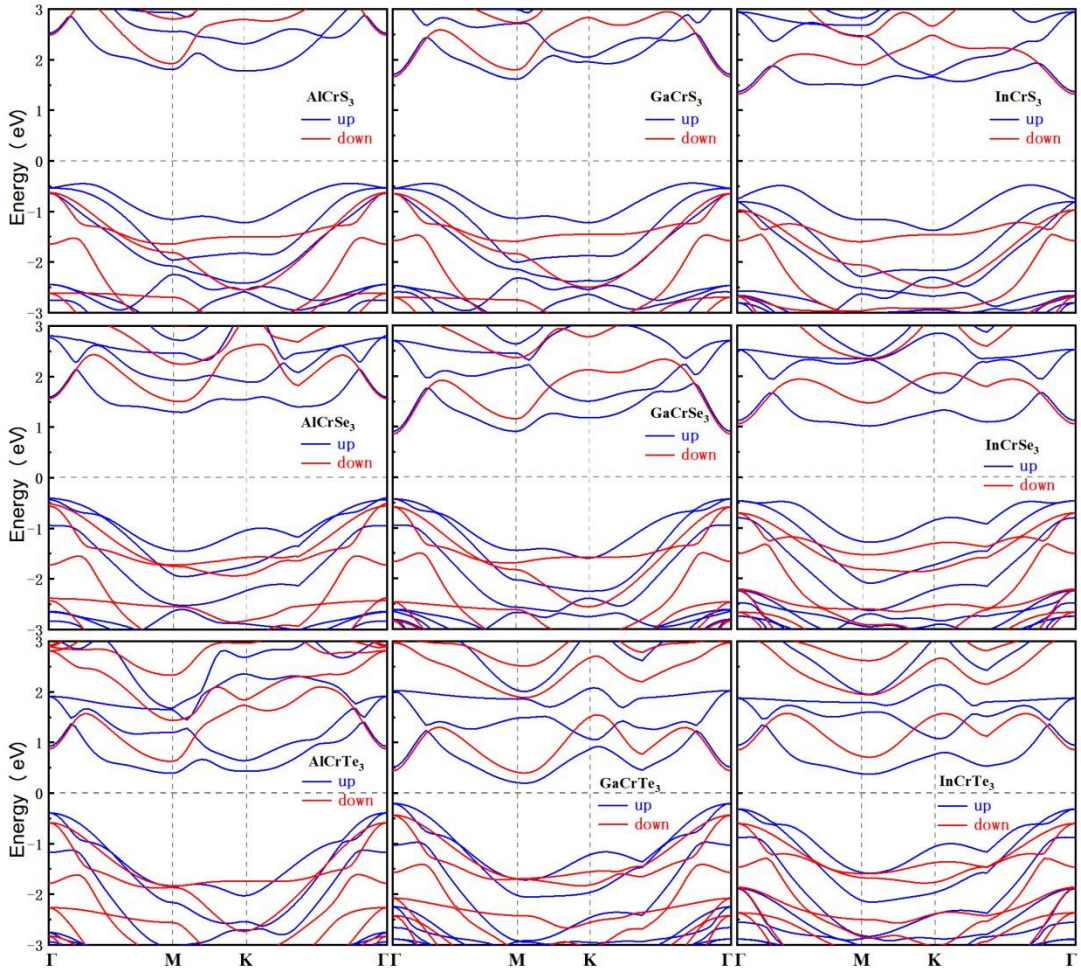

Figure S3. Band structures of all $X\mathrm{Cr}Y_3$ (X=Al, Ga, In; Y=S, Se, Te) monolayers under HSE06 calculation methods. Blue and red lines represent up and down spin, respectively.

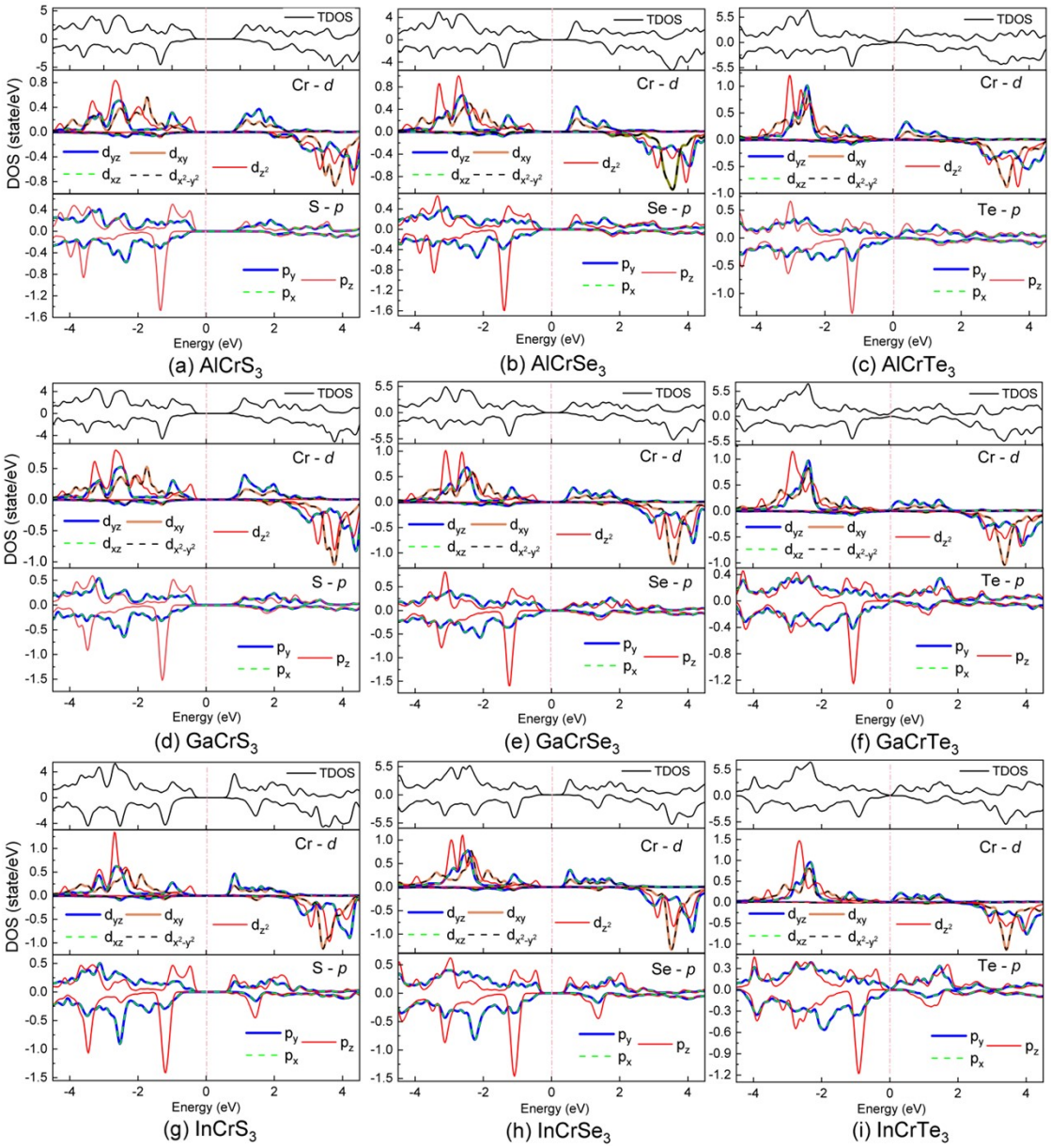

Figure S4. Total DOS and corresponding PDOS ofmonolayer $AlCrS_3$, $AlCrSe_3$, $AlCrTe_3$, $GaCrS_3$, $GaCrSe_3$, $GaCrTe_3$, $InCrS_3$, $InCrSe_3$, $InCrTe_3$, respectively. The vertical dashed line represents the Fermi level which is set as zero.

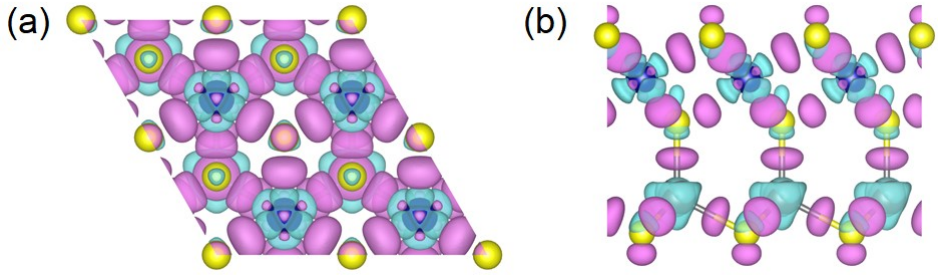

Figure S5. The (a) top and (b) side view of deformation charge density distribution of the $AlCrS_3$ monolayer. The value of the isosurface is 0.003 e/bohr^3. The silver, blue and yellow balls represent Al, Cr and S atoms, respectively.

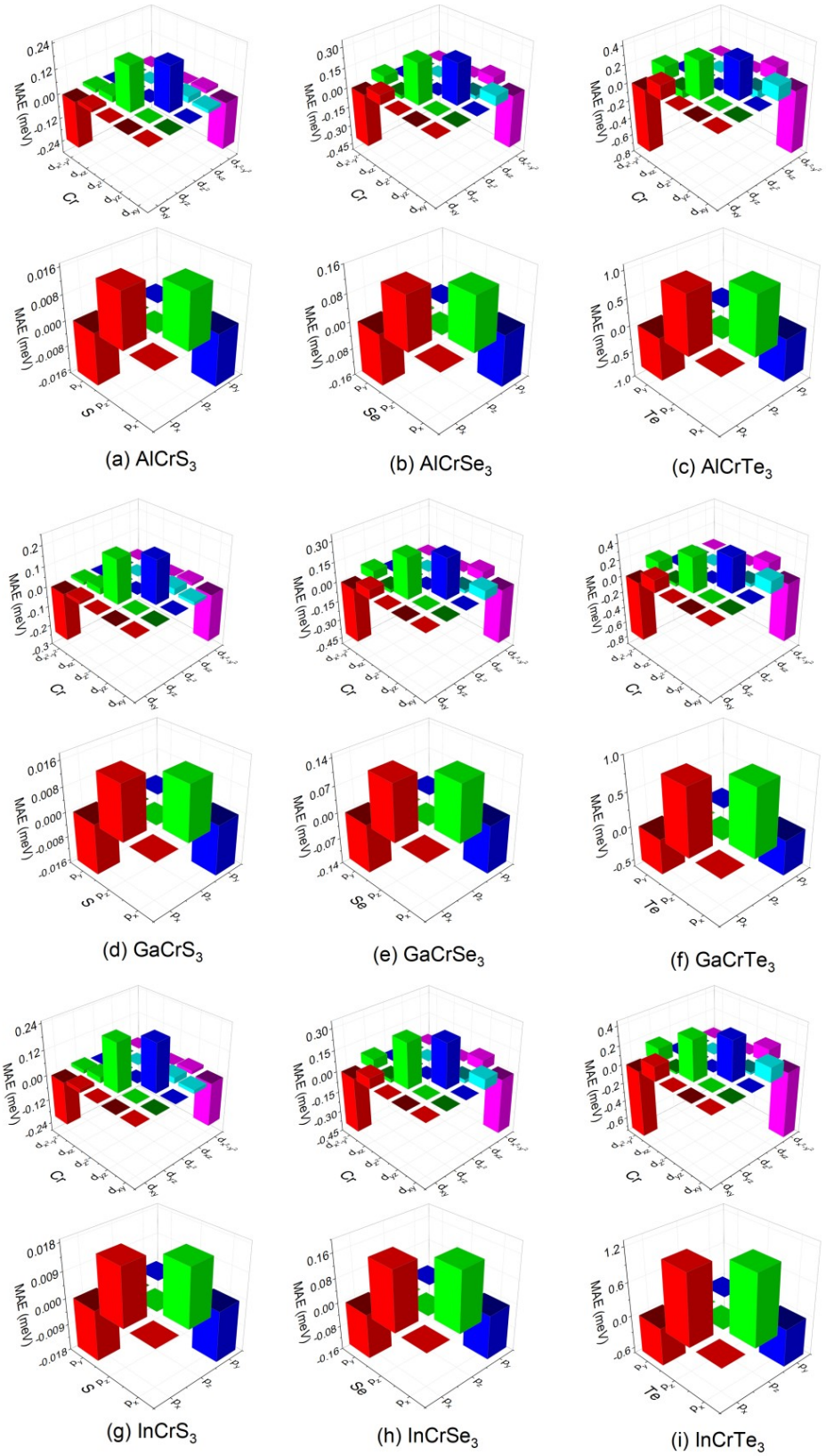

Figure S6. (a)-(i) are the orbital-projected contribution to MAE from the SOC interaction for$AlCrS_3$, $AlCrSe_3$, $AlCrTe_3$, $GaCrS_3$, $GaCrSe_3$, $GaCrTe_3$, $InCrS_3$, $InCrSe_3$, $InCrTe_3$, respectively.

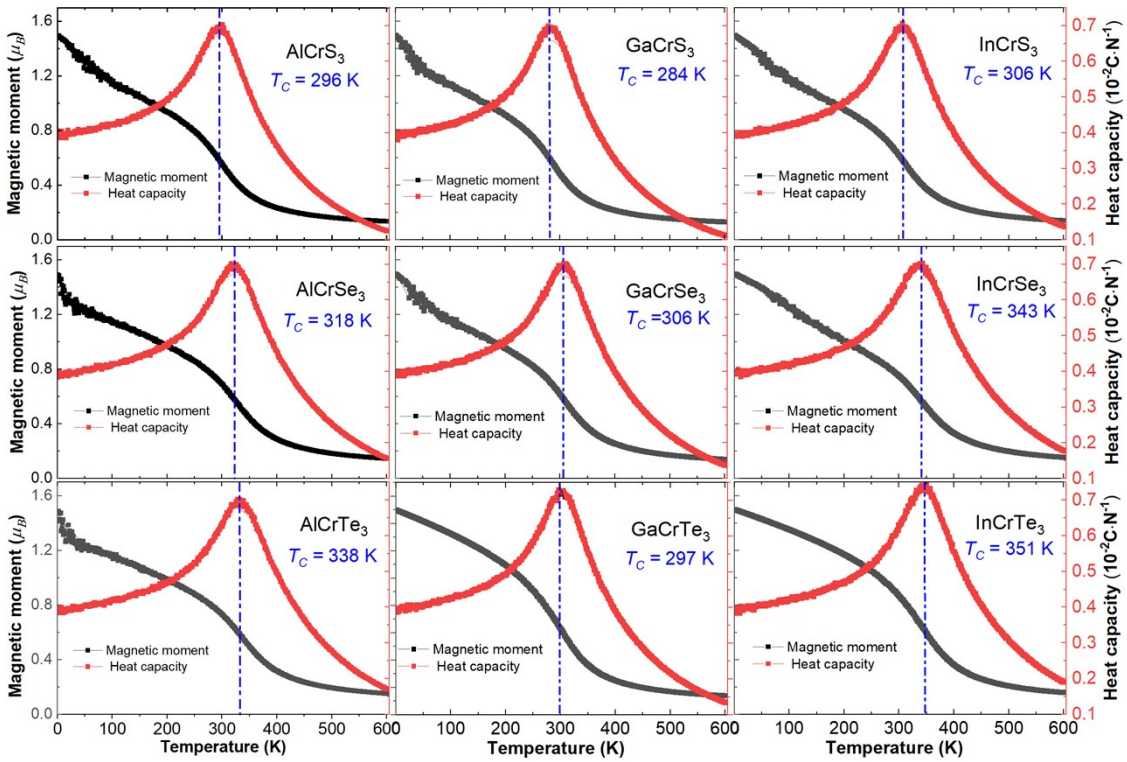

Figure S7. Magnetic moment and specific heat capacity as a function of temperature by Monte Carlo simulations.

Table S1. The total energies per cellof monolayer $XCrY_3$with different magnetic configurations at the GGA+ U level, including FM, zigzag AFM (z-AFM), stripe AFM (s-AFM), and 120° noncollinear AFM (n-AFM).

$XCrY_3$	E_{FM}/eV	$E_{z\text{-}AFM}$/eV	$E_{s\text{-}AFM}$/eV	$E_{n\text{-}AFM}$/eV
$AlCrS_3$	-27.72	-27.62	-27.61	-27.60
$AlCrSe_3$	-24.99	-24.87	-24.86	-24.85
$AlCrTe_3$	-22.09	-22.05	-22.02	-22.03
$GaCrS_3$	-25.94	-25.85	-25.84	-25.83
$GaCrSe_3$	-23.53	-23.47	-23.44	-23.43
$GaCrTe_3$	-21.01	-20.99	-20.97	-20.98
$InCrS_3$	-25.07	-24.90	-24.90	-24.89
$InCrSe_3$	-22.92	-22.78	-22.77	-22.76
$InCrTe_3$	-20.59	-20.52	-20.50	-20.50