Supporting Information

Excellent Nonlinear Optical Effects in MoS_2/CrOCl Heterojunction

Xiaomin Wang[1,2], Xiguang Wang[1], Shi Qiu[1], Anbing Zhang[1], Hongsheng Liu[1*], Dawei Li[3], Chunyu Ma[1], Vladimir V. Uglov[4, 5], Konstantin Kozadaev[5], Junfeng Gao[1].

1. Key Laboratory of Materials Modification by Laser, Ion and Electron Beams (Dalian University of Technology), Ministry of Education, School of Physics, Dalian 116024, China.
2. School of Engineering and Technology, Baoshan University, Baoshan 678000, China.
3. School of Optoelectronic Engineering and Instrumentation Science, Dalian University of Technology, Dalian, Liaoning 116024, China.
4. Dalian University of Technology and Belarusian State University Joint Institute & Innovation Center, Dalian 116024, China.
5. Faculty of Physics, Belarusian State University, Nezalezhnastsi av.4, Minsk 220030, Belarus.

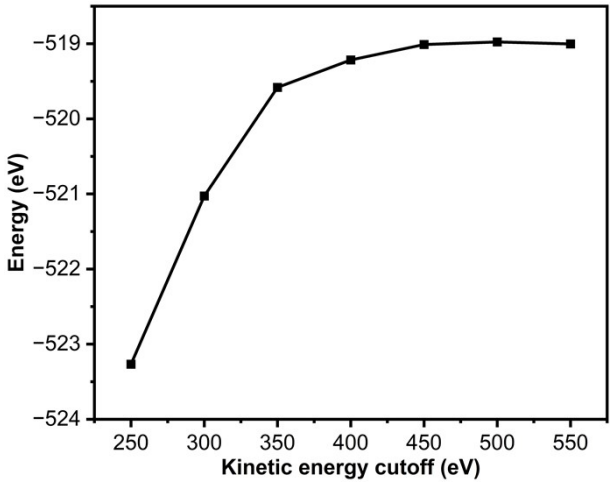

Figure S1. The total energy of the MoS_2/CrOCl heterojunction as a function of the kinetic energy cutoff for the planewave basis.

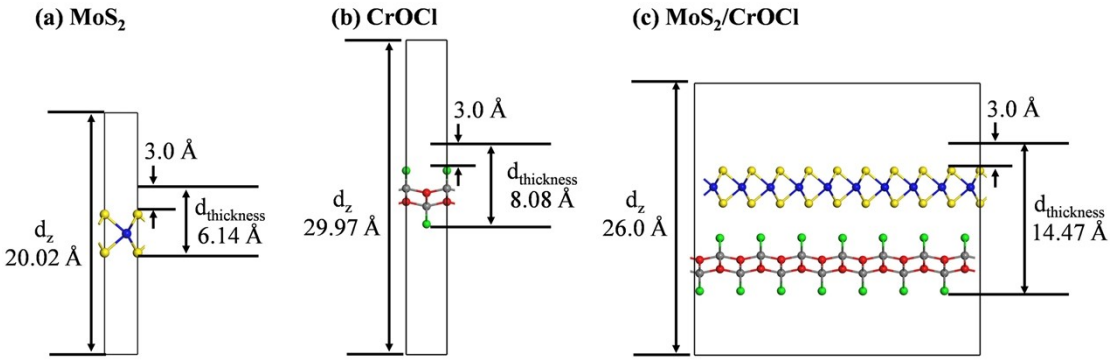

Figure S2. Details for the d_z and $d_{thickness}$ used in this study.

Table S1. Details about the lattice parameters of CrOCl supercell, MoS_2 supercell and MoS_2/CrOCl heterostructure. The strain in the CoOCl layer and MoS2 layer along a and b directions are also listed. Here, a and b are lattice constants along x and y directions. A positive value means a tensile strain and a negtive value means a compressive strain.

	CrOCl (7 × 1) supercell	MoS_2 (5 × 1) supercell	MoS_2/CrOCl heterostructure	strain in CrOCl	strain in MoS_2
a	27.288	27.407	27.347	0.22%	-0.22%
b	3.205	3.165	3.185	-0.62%	0.63%

The lattice constants of the MoS_2/CrOCl heterostructure were calculated as,

$$a_{heter} = (a_{CrOCl} + a_{MoS2})/2 ,$$

$$b_{heter} = (b_{CrOCl} + b_{MoS2})/2 ,$$

where, a_{heter} and b_{heter} are lattice constants of the MoS_2/CrOCl heterostructure. a_{CrOCl}, b_{CrOCl}, a_{MoS2} and b_{MoS2} are lattice constants of the CrOCl (7 × 1) supercell and MoS_2 (5 × 1) supercell.

The scanning of the potential energy surface of MoS_2/CrOCl heterostructures was performed by shifting the CrOCl layer along the x and y direction gradually (see Figure 1 in the main text). The grids are 56 × 7, i.e. a step of 0.488 Å along the x direction and a step of 0.455 Å along the y direction. For each configuration, all the atoms were fully relaxed including the interlayer distance.

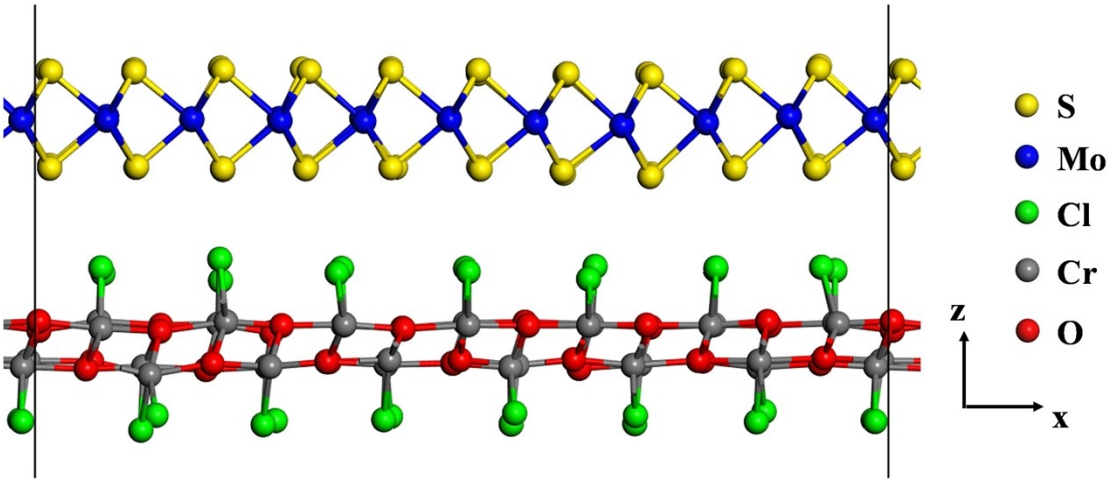

Figure S3. Snapshot structure of MoS_2/CrOCl heterostructure after 8 ps of AIMD simulations at 500K. The black rhombus represents the supercell used for AIMD.

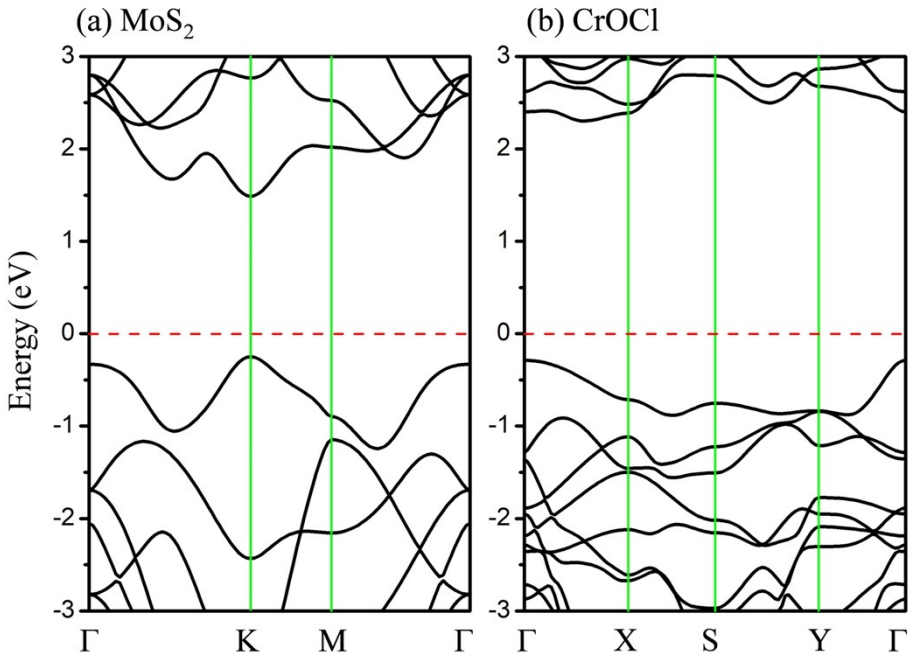

Figure S4. Band structure of (a) MoS_2 monolayer and (b) CrOCl monolayer.

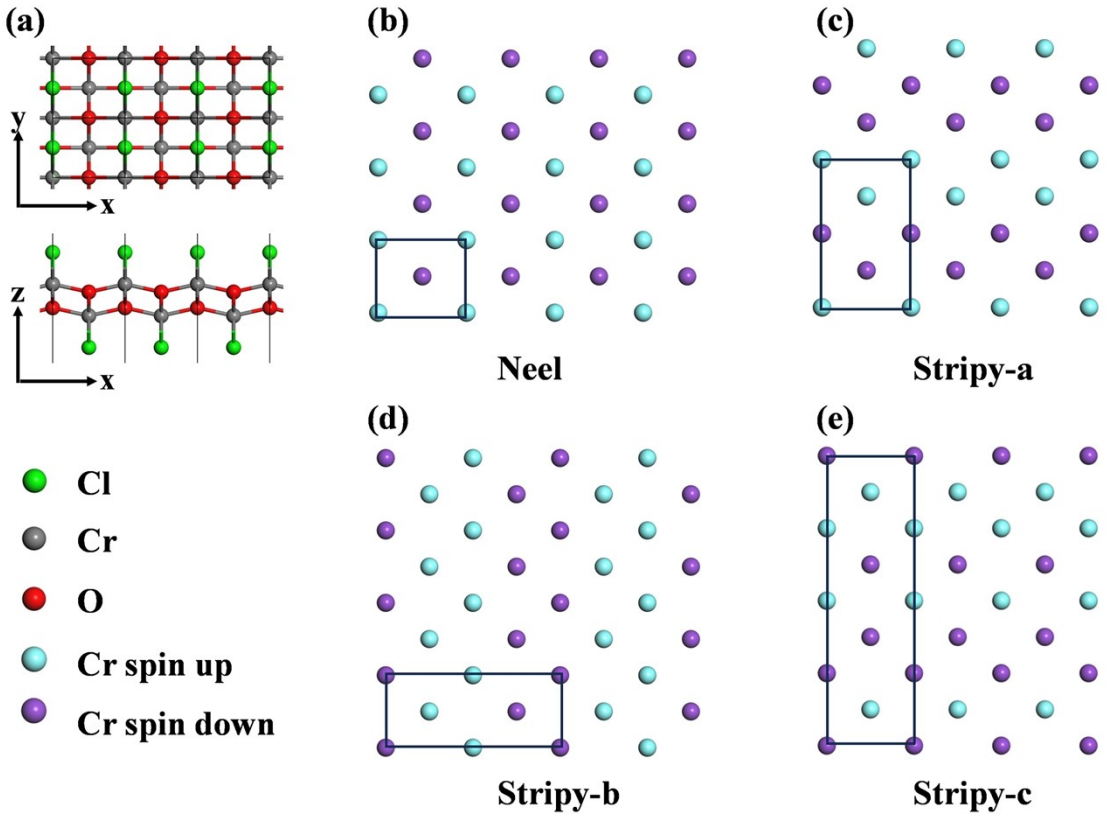

Figure S5. (a) Top and side views of monolayer CrOCl. (b-e) Top views of monolayer CrOCl with different antiferromagnetic configurations. Only Cr atoms are shown.

Table S2. Energy per CrOCl formula in monolayer CrOCl with non-magnetic (NM), ferromagnetic (FM) and antiferromagnetic (AFM-Neel, AFM-Stripy-a, AFM-Stripy-b, AFM-Stripy-c) states.

	NM	FM	AFM-Neel	AFM-Stripy-a	AFM-Stripy-b	AFM-Stripy-c
Energy (eV)	-18.522	-21.039	-21.068	-21.067	-21.065	-21.049

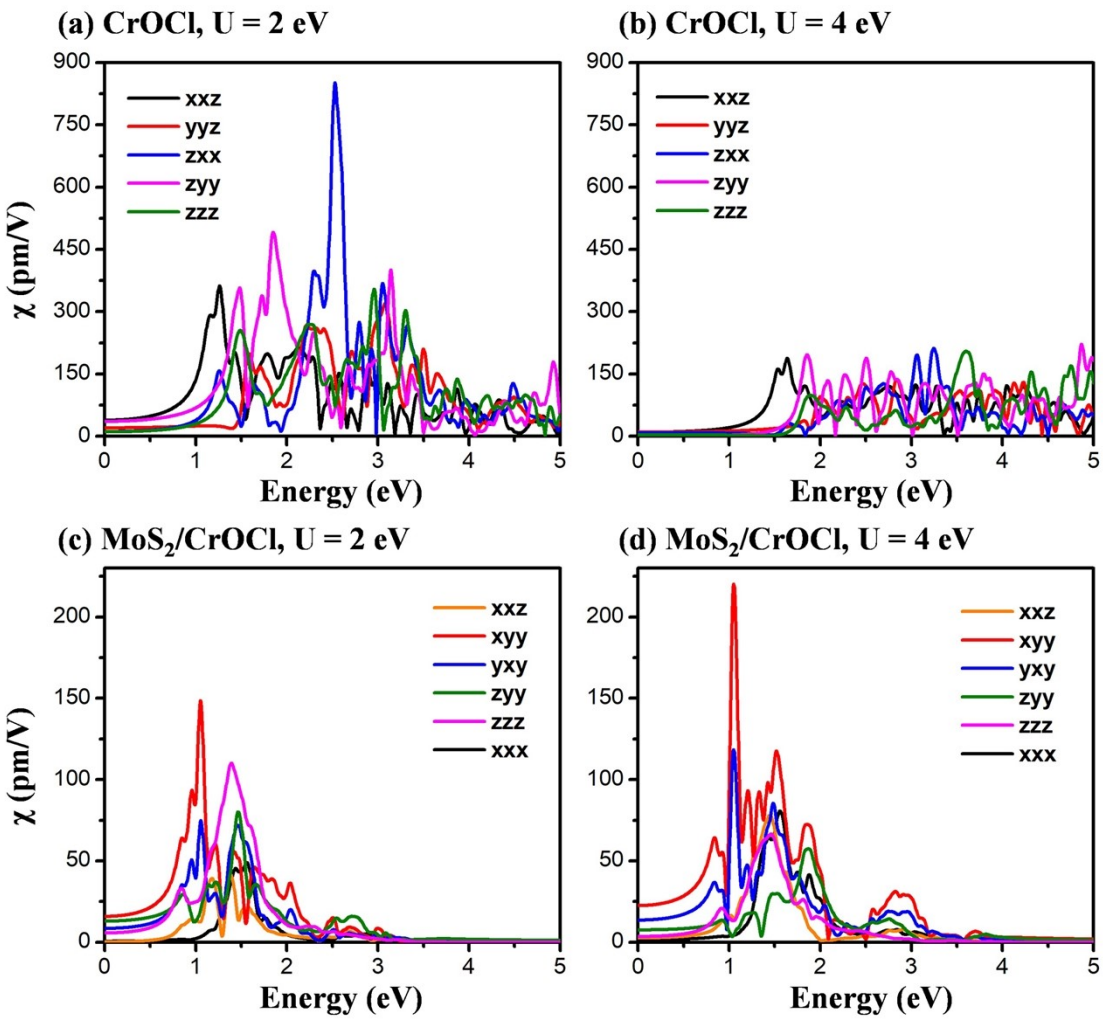

Figure S6. Non-zero second-order non-linear coefficients of (a-b) CrOCl and (c-d) MoS_2/CrOCl as functions of photon energy with different U values.

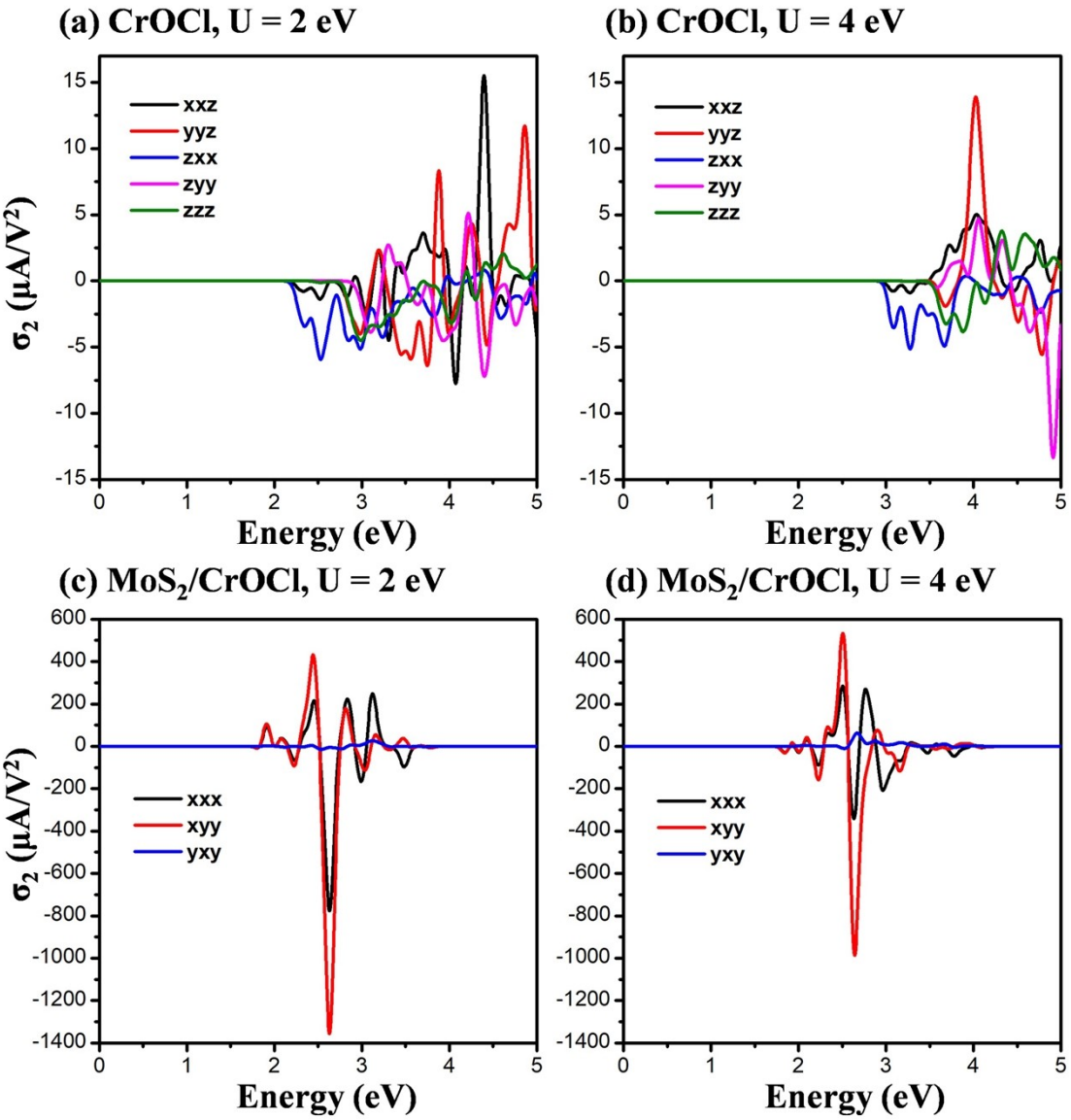

Figure S7. Shift photoconductivity versus photon energy of (a-b) CrOCl and (c-d) MoS_2/CrOCl with different U values.