Supporting Information

for

$MoSe_2/C_{60}$ Heterojunction May Be Efficient for Photovoltaic Applications: Time-Domain Ab Initio Analysis of Interfacial Charge Separation and Recombination Dynamics

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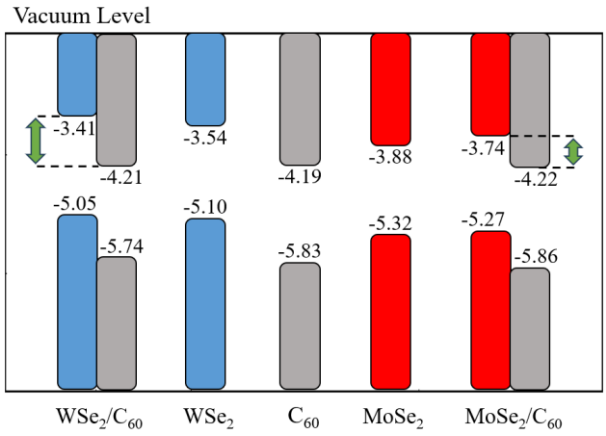

Figure S1. Band edge energies of the TMD/C_{60} heterostructures, the pristine $MoSe_2$ and WSe_2, and the pristine C_{60} calculated using PBE, with vacuum energy as reference level.

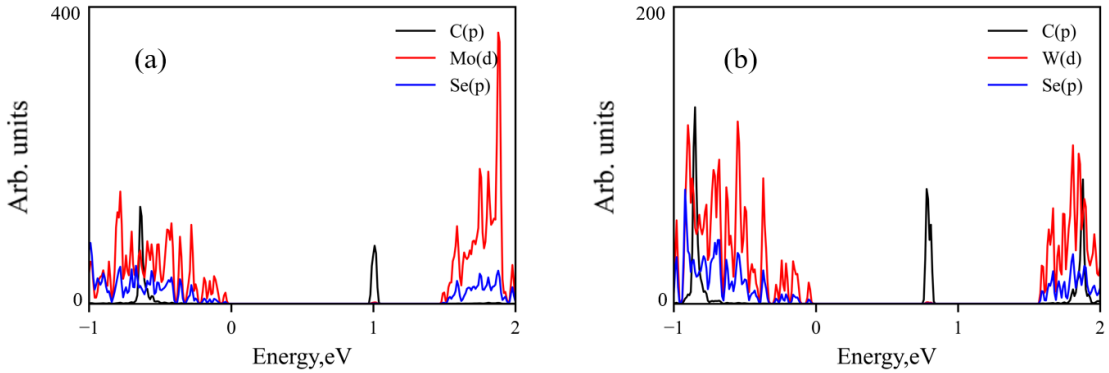

Figure S2. Atom and orbital resolved density of states for (a) $MoSe_2/C_{60}$ and (b) WSe_2/C_{60} calculated using PBE function with 4x4x1 kpoints.

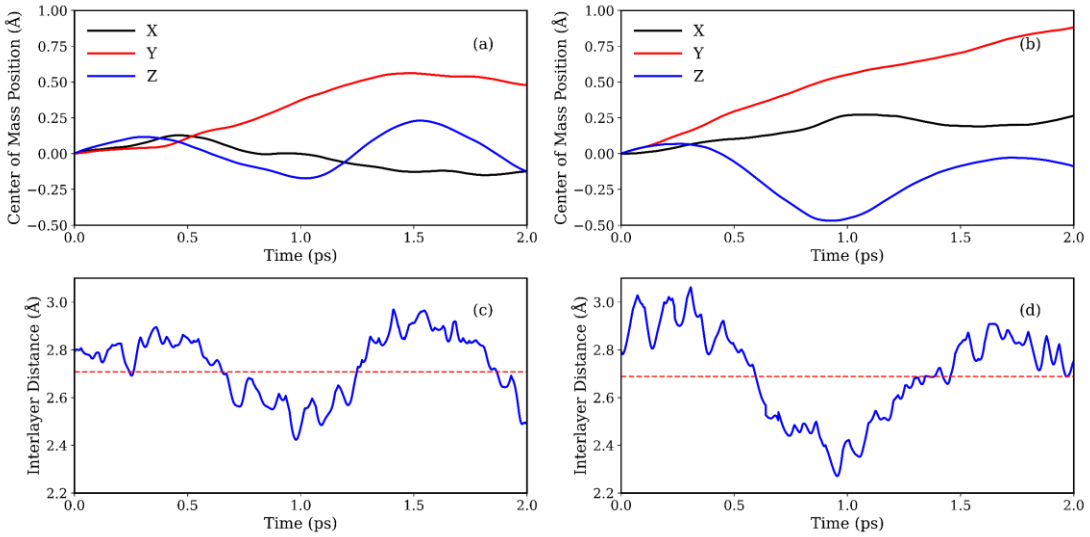

Figure S3. The center of mass position of C_{60} in a 2ps MD trajectory for (a) $MoSe_2/C_{60}$ and (b) WSe_2/C_{60} heterostructures. Time evolution of the interlayer distances for (a) $MoSe_2/C_{60}$ and (b) WSe_2/C_{60} heterostructures. Interlayer distance is defined as the distance between the bottom C atom of C_{60} and the top of Se atom of TMD.

Table S1. Computed donor-acceptor overlap (arb. unit) based on the optimized ground state structure at 0K.

	Electron transfer	Interface charge recombination	Intra-TMD recombination
$MoSe_2/C_{60}$	4.48×10^{-5}	3.02×10^{-5}	1.87×10^{-3}
WSe_2/C_{60}	4.41×10^{-5}	1.28×10^{-5}	1.78×10^{-3}

Table S2. Lowest-lying excitation energy computed by delta-SCF approach

	$MoSe_2/C_{60}$	WSe_2/C_{60}	$MoSe_2$	WSe_2	C_{60}
Excitation energy (eV)	1.45	1.39	1.45	1.56	1.64

Table S3. The calculated key orbital energies and band offset for electron transfer (TMD CBM/C_{60} LUMO) and charge recombination (C_{60} LUMO/TMD VBM) using PBE, PBE+SOC level of theory for $MoSe_2/C_{60}$ and WSe_2/C_{60} heterostructures. Unit in eV.

		PBE	PBE+SOC
$MoSe_2/C_{60}$	$MoSe_2$-CBM	1.487	1.378
	C_{60}-LUMO	1.002	0.902
	$MoSe_2$-VBM	-0.048	-0.05
	$MoSe_2$-CBM/C_{60}-LUMO	0.485	0.476
WSe_2/C_{60}	WSe_2-CBM	1.586	1.250
	C_{60}-LUMO	0.787	0.547
	WSe_2-VBM	-0.051	-0.052
	WSe_2-CBM/C_{60}-LUMO	0.799	0.703
$MoSe_2$	CBM	1.371	1.288
	VBM	-0.075	-0.047
	VBM/CBM	1.446	1.335
WSe_2	CBM	1.479	1.207
	VBM	-0.073	-0.053
	VBM/CBM	1.552	1.26