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Electronic Supplementary Information

Switching interlayer magnetic order in bilayer CrI_3 by stacking reversal

Xiangru Kong,[1,*] Hongkee Yoon,[2,*] Myung Joon Han,[2,†] and Liangbo Liang[1,†]

[1]*Center for Nanophase Materials Sciences, Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee 37831, USA*

[2]*Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea*

[*] These authors contributed equally to this work

[†] Corresponding authors: mj.han@kaist.ac.kr; liangl1@ornl.gov

Stacking	$R\bar{3}$	$R\bar{3}$-r	$C2/m$	$C2/m$-r	AA	AA-r
FM	3.44	4.01	3.49	3.48	3.60	4.10
AFM	3.46	4.00	3.48	3.49	3.59	4.06

Table S1. The averaged interlayer distance with the unit of Å in bilayer CrI_3 at different stacking patterns. The values are from the structures relaxed by Liechtenstein's DFT+U approach with $U = 3.9$ eV and $J = 1.1$ eV. The difference of the interlayer distance between the FM and AFM order for each stacking is small. The difference of the interlayer distance between $C2/m$ and $C2/m$-r is also small. However, it is not the case for stacking patterns of $R\bar{3}$ and AA: the interlayer distance of $R\bar{3}$-r is about 0.54~0.57 Å larger than that of $R\bar{3}$; the interlayer distance of AA-r is about 0.47~0.50 Å larger than that of AA. From the side views of Figure 1 in the main text, we can observe that in $R\bar{3}$-r and AA-r stackings, the I atom in the bottom sublayer of the top CrI_3 layer (I2b) sits almost directly above I1t, the I atom in the top sublayer of the bottom CrI_3 layer. Therefore, the orbitals from I2b and I1t atoms in these two stackings tend to repel each other, and push the top and bottom CrI_3 layers farther away, compared to their non-rotated stacking counterparts. This explains their larger interlayer distances (above 4.00 Å), and why their total energies are notably higher in comparison with the corresponding non-rotated stacking patterns $R\bar{3}$ and AA (Figure 2 in the main text). In contrast, for $C2/m$-r stacking, the rotation does not lead to I2b over I1t, and thus the interlayer distance barely changes compared to the non-rotated $C2/m$ stacking, which explains why $C2/m$-r and $C2/m$ share similar total energies.

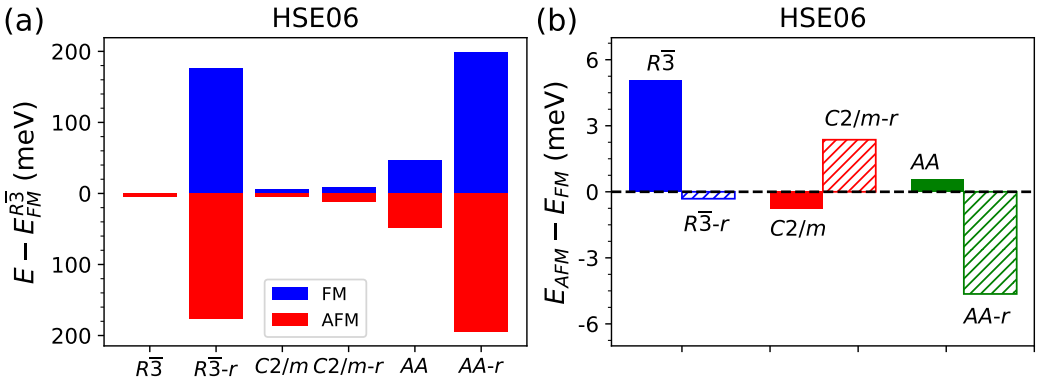

Figure S1. (a) The total energy of bilayer CrI_3 by hybrid functional (HSE06) calculations in all six stacking patterns ($R\bar{3}$, $R\bar{3}$-r, $C2/m$, $C2/m$-r, AA, and AA-r) with interlayer FM or AFM order. $R\bar{3}$ stacking with the FM order has the lowest energy and thus was used as the zero energy reference. (b) The energy differences between AFM and FM bilayer CrI_3 at different stacking orders computed by the HSE06 method. The HSE06 results without use of any U and J parameters are similar to those of Liechtenstein's DFT+U approach with $U = 3.9$ eV and $J = 1.1$ eV. This confirms that the 180° stacking rotation can switch the magnetic ground state for all three stacking orders.