Supplemental materials : Tunable Magnetic Coupling of Monolayer CrI_2 by Strain Engineering

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February 3, 2025

1 Linear response method on determining U value

The GGA+U accounts for describing the strongly correlated system. The standard method of determining U is to take U as a semi-empirical value by comparing it with the experimental result (e.g., energy gap). However, the reported energy gap from the experiment is not consistent with each other. Hence, we use the linear response method proposed by Cococcioni and Gironcoli. In this method, the U can be determined as the difference between the screened and bare second derivative of the energy to localized state occupation n^I at site I. This can be written as

$$U = \frac{\partial^2 E\left[\{n^I\}\right]}{\partial (n^I)^2} - \frac{\partial^2 E_0\left[\{n^I\}\right]}{\partial (n^I)^2}$$

Here,

$$E\left\{n_I^l\right\} = \min_{\alpha_I}\left\{E\left\{\alpha_I\right\} - \sum_I \alpha_I n_I^l\right\}$$

$$\frac{\partial E\left[\{n^I\}\right]}{\partial n^I} = -\alpha_I\left(\{n^I\}\right)$$

$$\frac{\partial^2 E\left[\{n^I\}\right]}{\partial (n^I)^2} = -\frac{\partial \alpha_I\left(\{n^I\}\right)}{\partial n^I}$$

We can get occupation-dependent energy functional by applying localized potential shifts to the d levels of the Cr atoms to excite charge fluctuation on their orbitals and solving the Kohn–Sham equations self-consistently. From above, we then have

$$U = \left(\frac{\partial \alpha_I}{\partial n^I_{\mathrm{NSCF}}}\right) - \left(\frac{\partial \alpha_I}{\partial n^I_{\mathrm{SCF}}}\right) = \chi_0^{-1} - \chi^{-1}$$

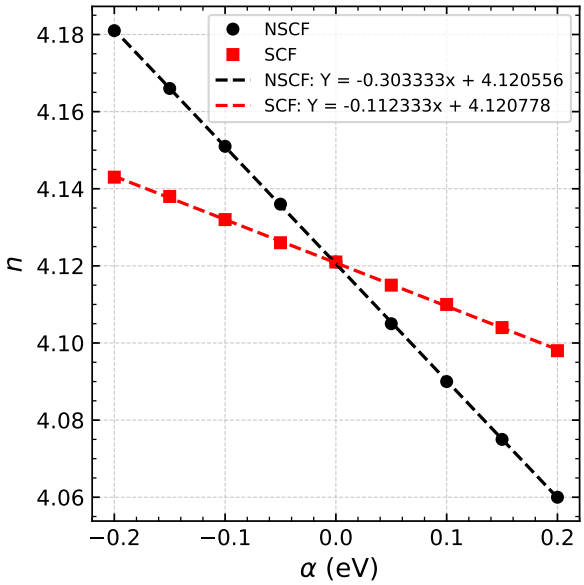

Figure S1: Linear response d orbital electron of Cr atom under spherical potential perturbation.

2 Lattice-constant dependence on U value.

we optimize the lattice constant using several different U values. As shown in the table, a higher U value induces a larger lattice constant due to higher electron-electron interaction.

Table S1: Lattice-constant dependence on U value.

U	$U = 0$	$U = 3$	$U = 5.6$	$U = 7$
a (Å)	3.9177	3.9730	4.0240	4.0524
b (Å)	7.4857	7.5478	7.5508	7.5433

3 Angle of Cr-I-Cr

The modulation of Cr-I-Cr angle of Θ_1 and Θ_2 under external strain is small. The angle is nearly 90°, which supports the FM superexchange interaction in the considered strain regime.

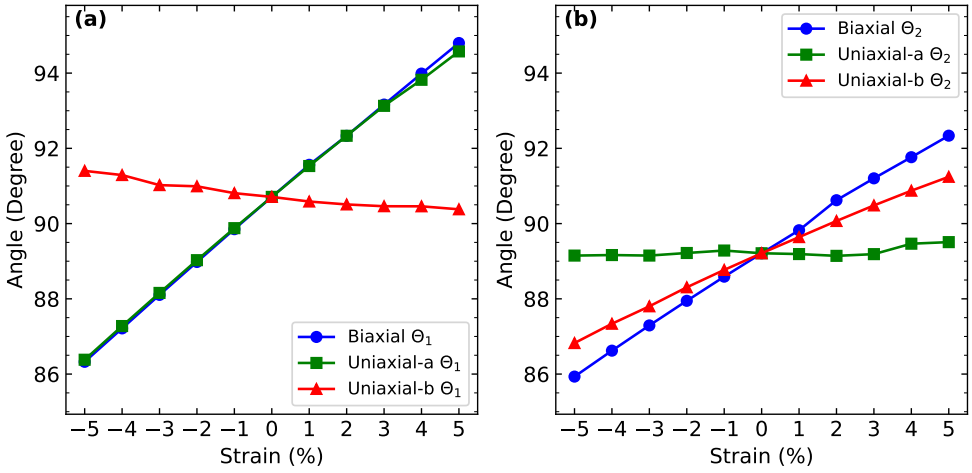

Figure S2: Angle of Cr-I-Cr for the (a) Θ_1 and (b) Θ_2.

4 The FM superexchange mechanism for Cr1-I-CrI' and Cr1'-I-Cr2.

As explained in the main text, the superexchange applied symmetrically when considering the Cr1'-I bond interaction. The −ICOHP data shows that Ip_y interact strongly with the Cr1' $d_{x^2-y^2}$ orbitals.

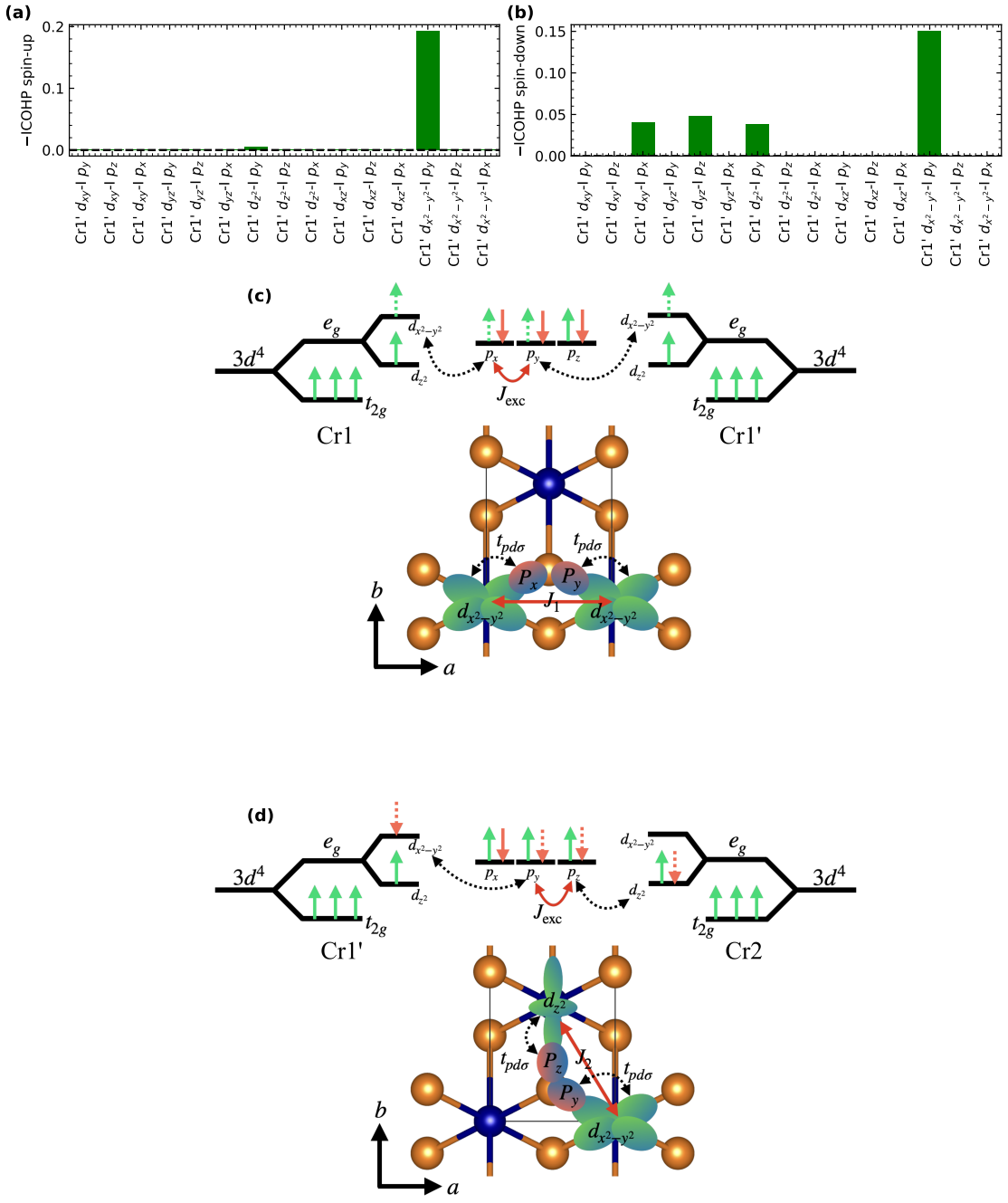

Figure S3: The orbital resolved −ICOHP for the Cr1'-I (a) spin-up component and (b) spin-down component. The mechanism of FM superexchange of (c) Cr1-I-Cr1' and (d) Cr1'-I-Cr2.