Supplementary Information (SI) for Physical Chemistry Chemical Physics.
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Supporting Informations

Phonons Regulate Covalency: A New Way to Modulate Magnetism and Curie Temperature of CrI_3

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S1. Magnetic ground state of CrI_3 monolayer

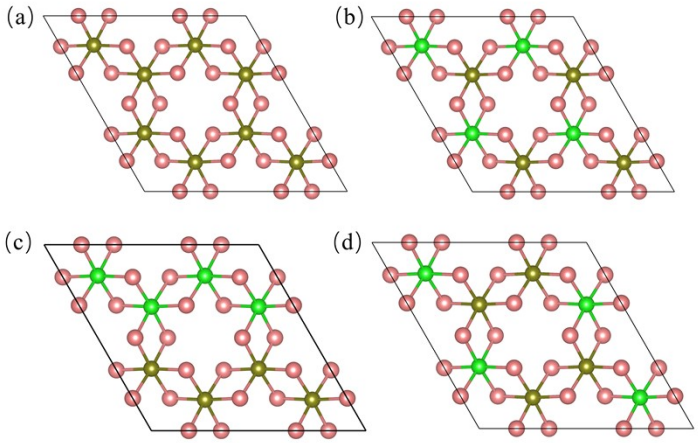

Figure S1. Four impossible magnetic phases for CrI_3 monolayer, including ferromagnetic (FM) (a), antiferromagnetic-Néel (AFM-N) (b), antiferromagnetic-stripy (AFM-S) (c), and antiferromagnetic-Zigzag (AFM-Z) (d). The dark gold and green balls represent the spin up and spin down magnetic lattices, respectively.

S2. Magnetic properties after imposing phonon modes

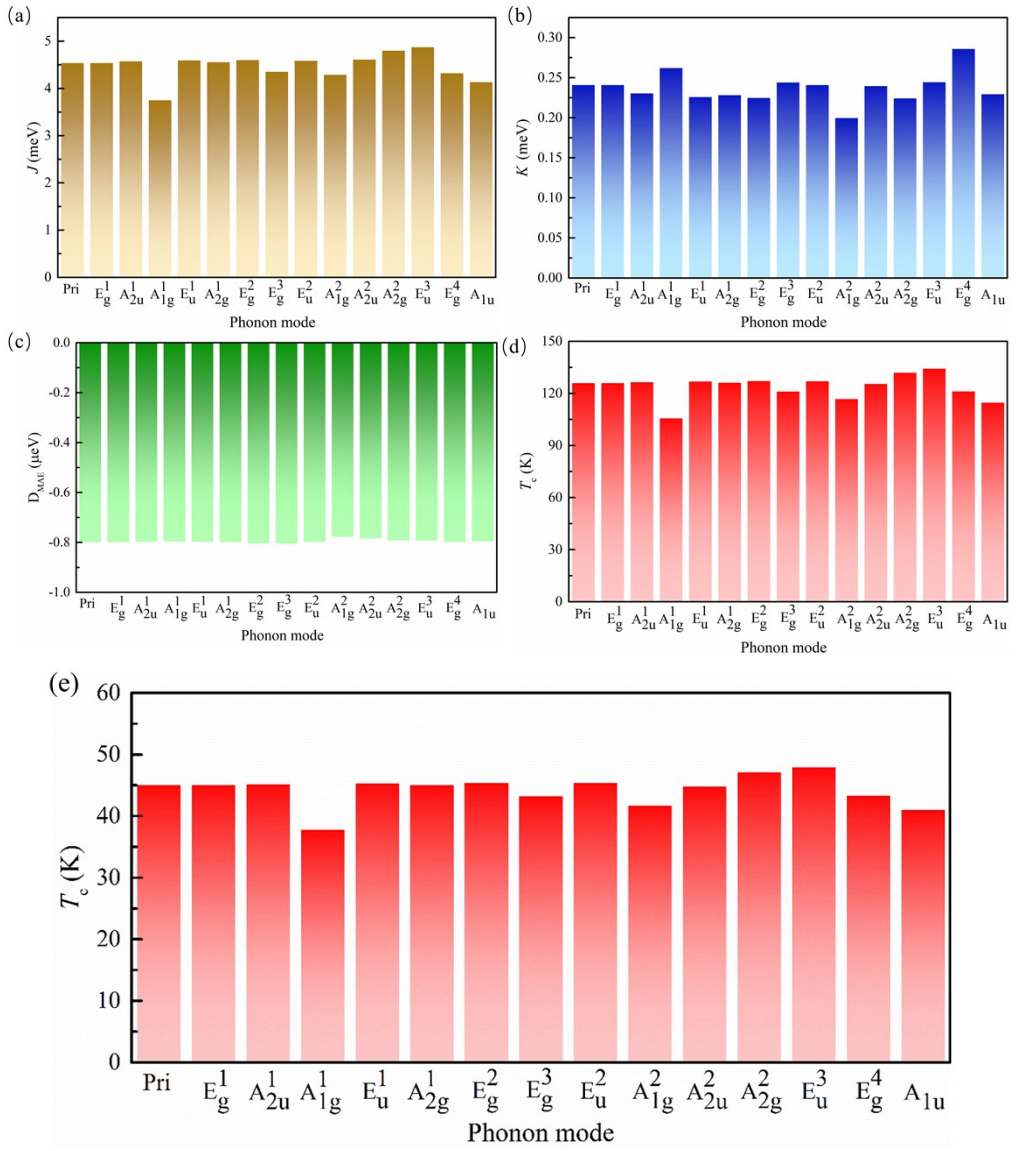

Figure S2. The coefficients of magnetic exchange coupling (a), magnetocrystalline anisotropy (b), the anisotropy of dipole-dipole coupling (c), the Curie temperature (d) after imposing phonon modes and after rescaling (e).

S3. Spin-wave spectrum before and after imposing phonon vibrations

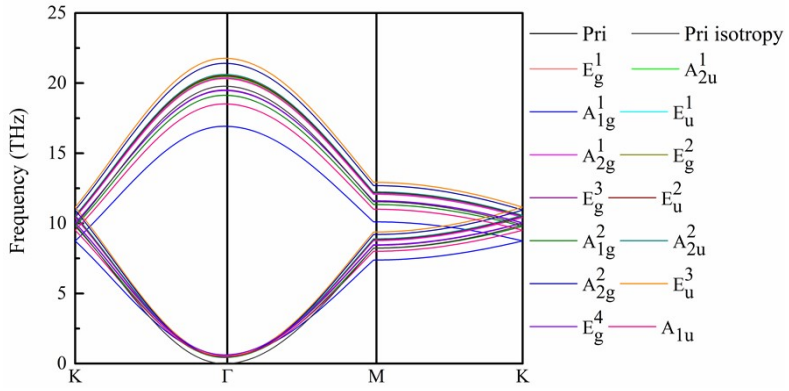

Figure S3. The spin-wave spectrum of CrI_3 monolayer with and without imposing phonon.

S4. Partial density of state of pristine CrI_3 monolayer

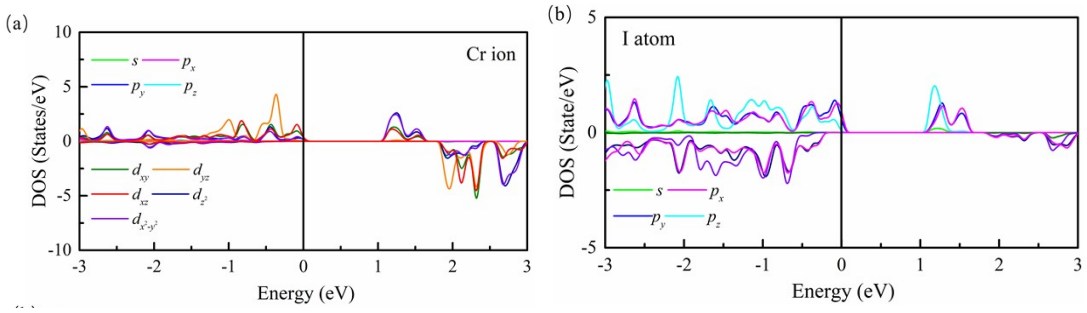

Figure S4. Partial density of state (PDOS) of Cr ion (a) and I (b) atom in pristine CrI_3 monolayer.