Supporting information

Strain-Controlled Single Cr Embedded Nitrogen-Doped Graphene Achieve Efficient Nitrogen Reduction

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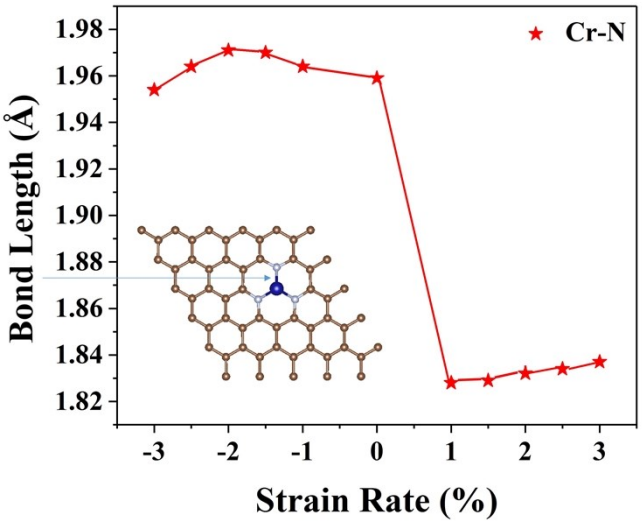

Figure S1. Bond length of Cr-N for CrN_3@Gaphene.

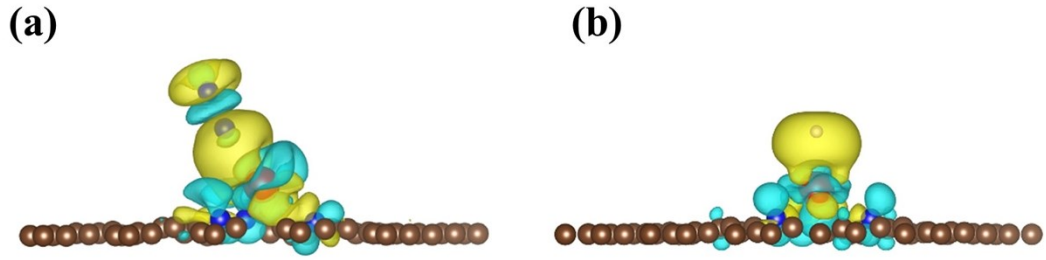

Figure S2. The charge density difference of H* **(a)** and N_2 **(b)** adsorbed in the CrN_3@Gaphene with lattice strain of 0%. The isovalue is set to 0.0014 e/Å^3 and 0.0012 e/Å^3 for H* and N2 adsorbed in the CrN_3@Gaphene with lattice strain of 0% respectively. The yellow/blue area in the figure indicates that the area get/lose electrons.

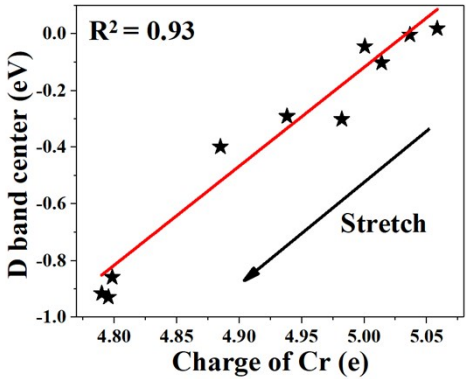

Figure S3. The linear relationship between the d-band center of CrN_3@Graphene and the charge of singe Cr atom.

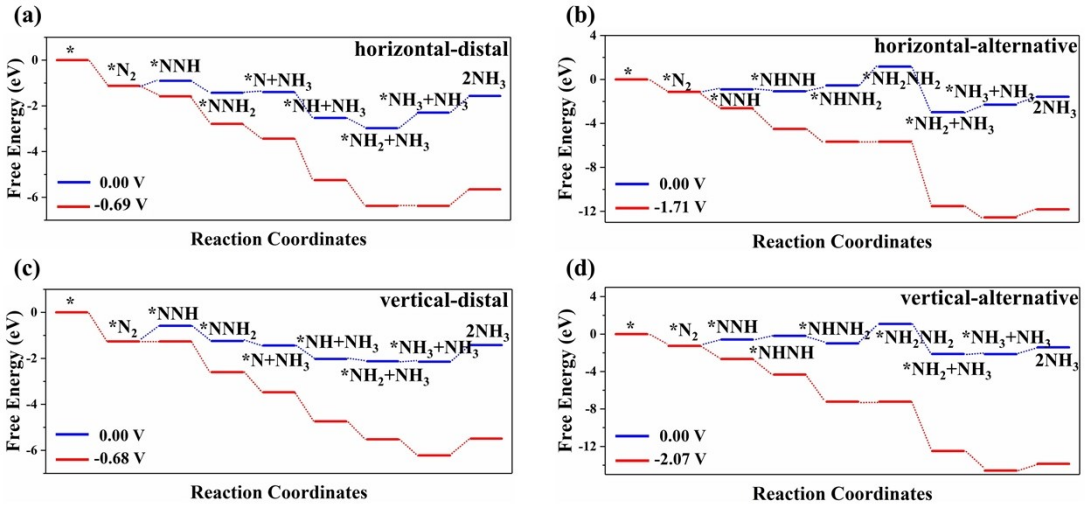

Figure S4. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of -3%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

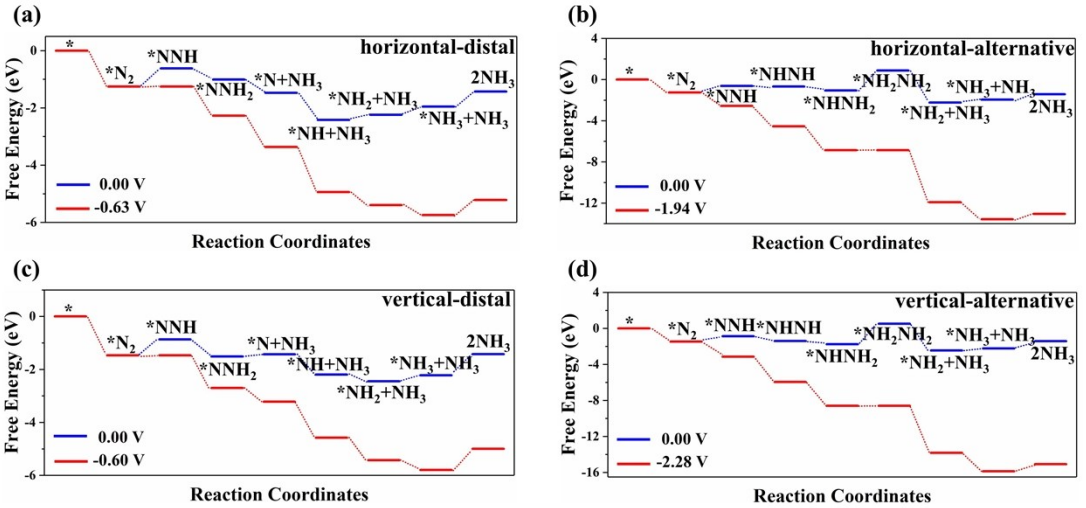

Figure S5. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of -2.5%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

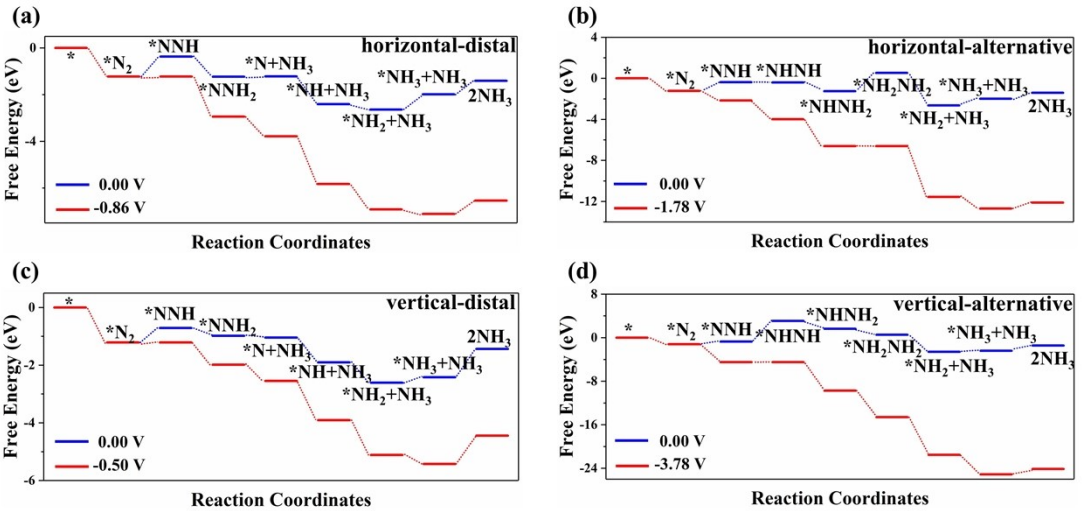

Figure S6. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of -2%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

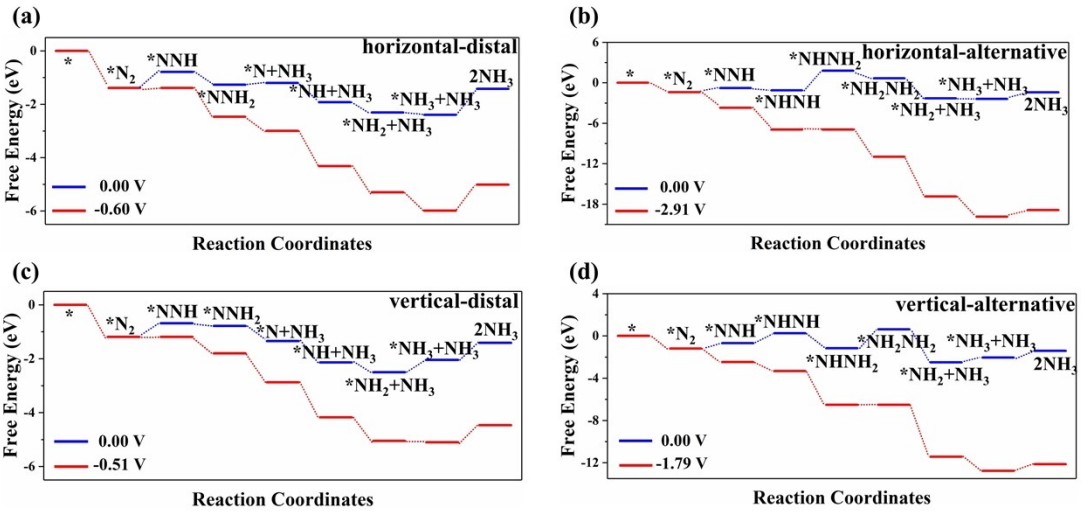

Figure S7. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of -1.5%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

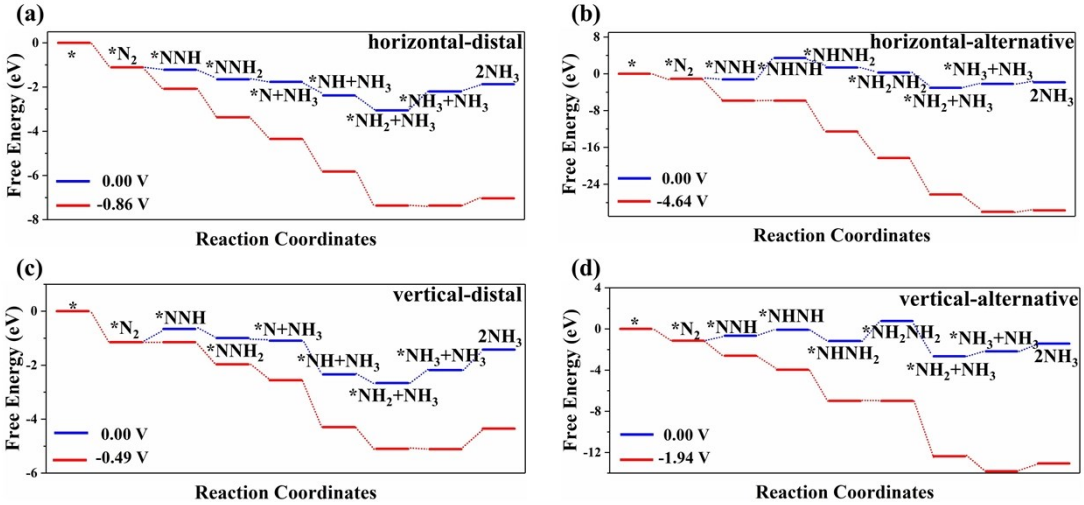

Figure S8. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of -1%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

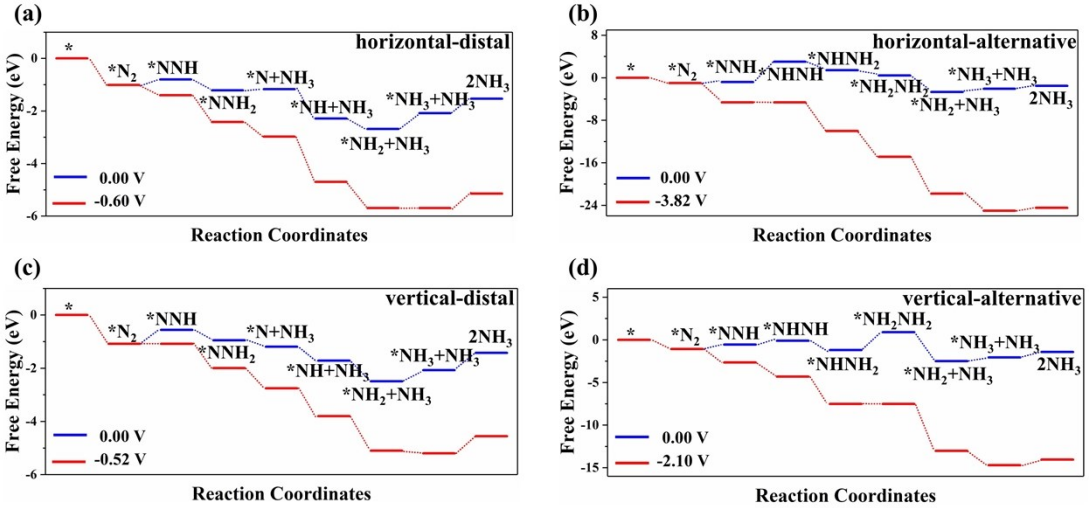

Figure S9. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of 0%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

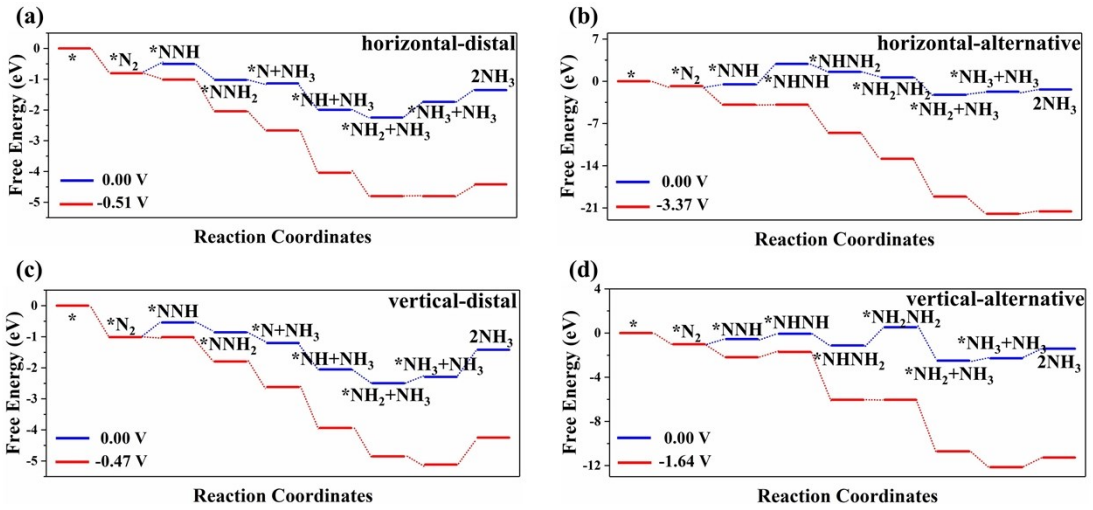

Figure S10. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of 1.25%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

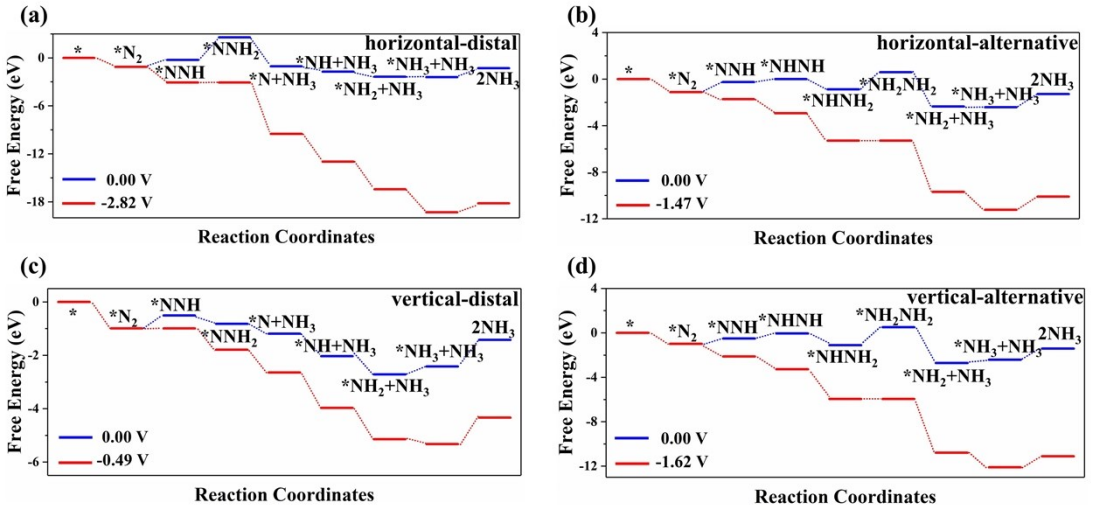

Figure S11. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of 1.75%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

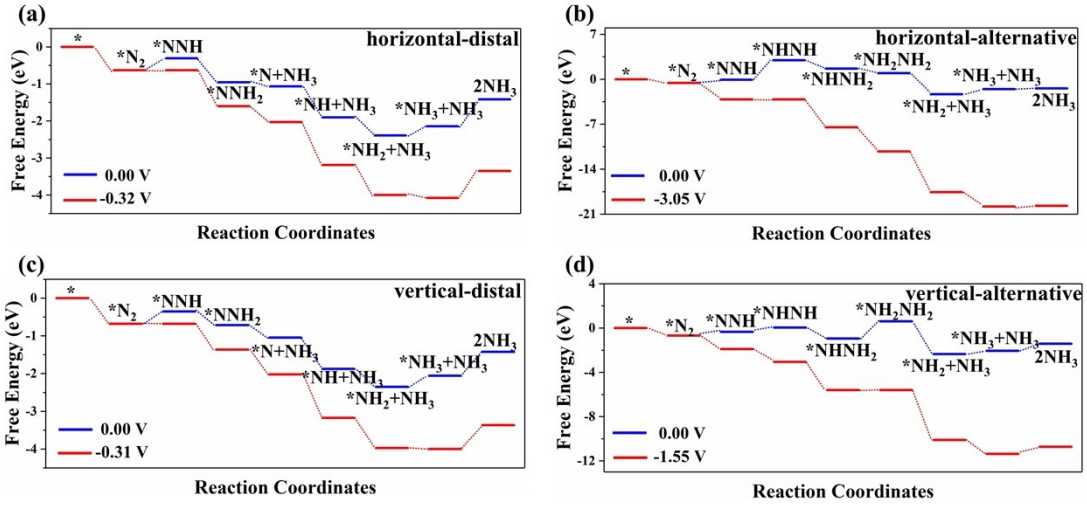

Figure S12. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of 2.5%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

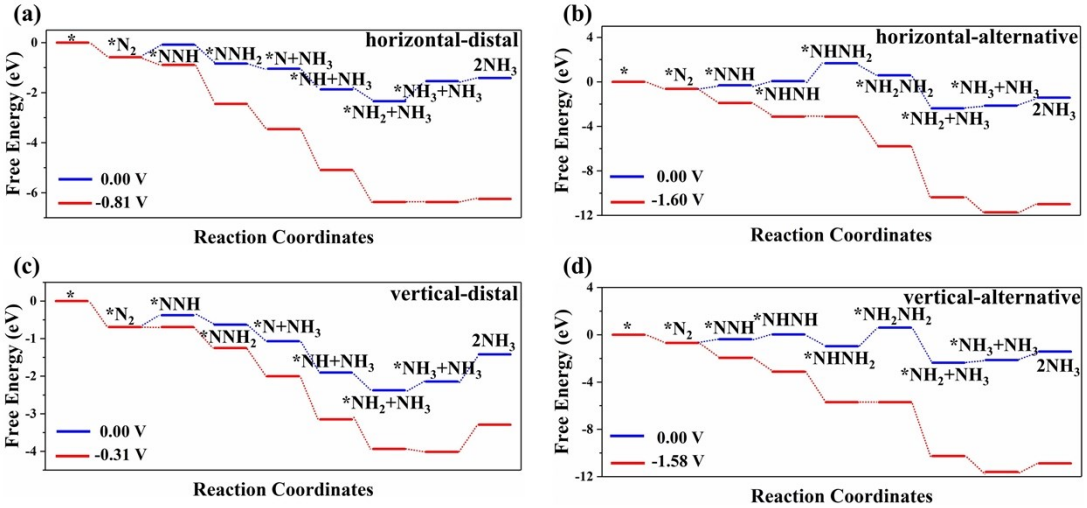

Figure S13. Free energy process for **(a).** horizontal-distal, **(b).** horizontal-alternative, **(c).** vertical-distal, **(d).** vertical-alternative reaction mechanism. The substrate is CrN_3@Gaphene with lattice strain of 3%. Structural evolution is shown in the upper right corner of each panel. Brown, blue, red, gray spheres represent carbon atoms, nitrogen atoms, chromium atoms, hydrogen atoms, respectively.

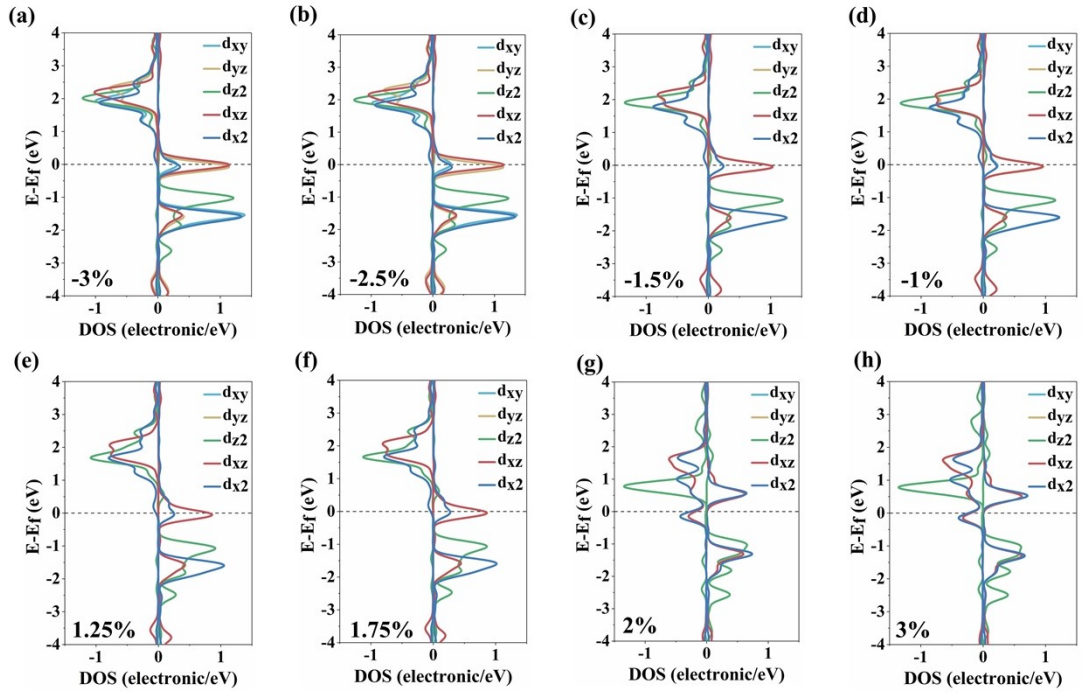

Figure S14. The density of states of single atom Cr for CrN_3@Gaphene with lattice strain of -3% **(a)**, -2.5% **(b)**, -1.5% **(c)**, -1% **(d)**, 1.25% **(e)**, 1.75% **(f)**, 2% **(g)**, 3% **(h)**.

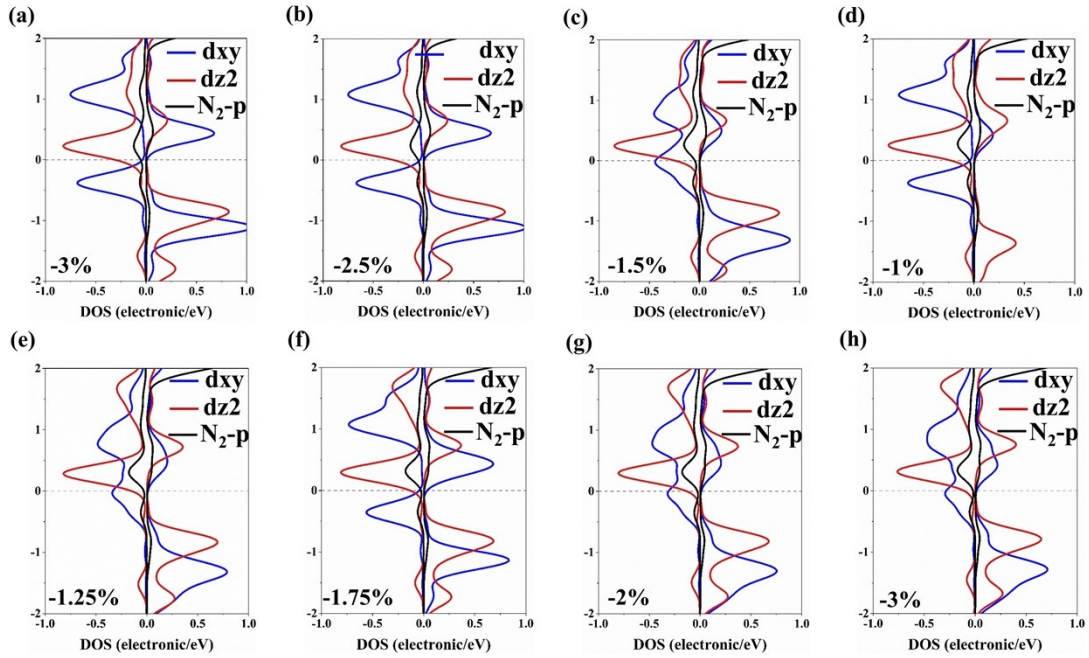

Figure S15. The partial density of states which divided by orbital components of N_2 and Cr for CrN_3@Graphene with lattice strain of -3% **(a)**, -2.5% **(b)**, -1.5% **(c)**, -1% **(d)**, 1.25% **(e)**, 1.75% **(f)**, 2% **(g)**, 3% **(h)**.The spin state of the electron filled in the right/left orbit is spin up/down.

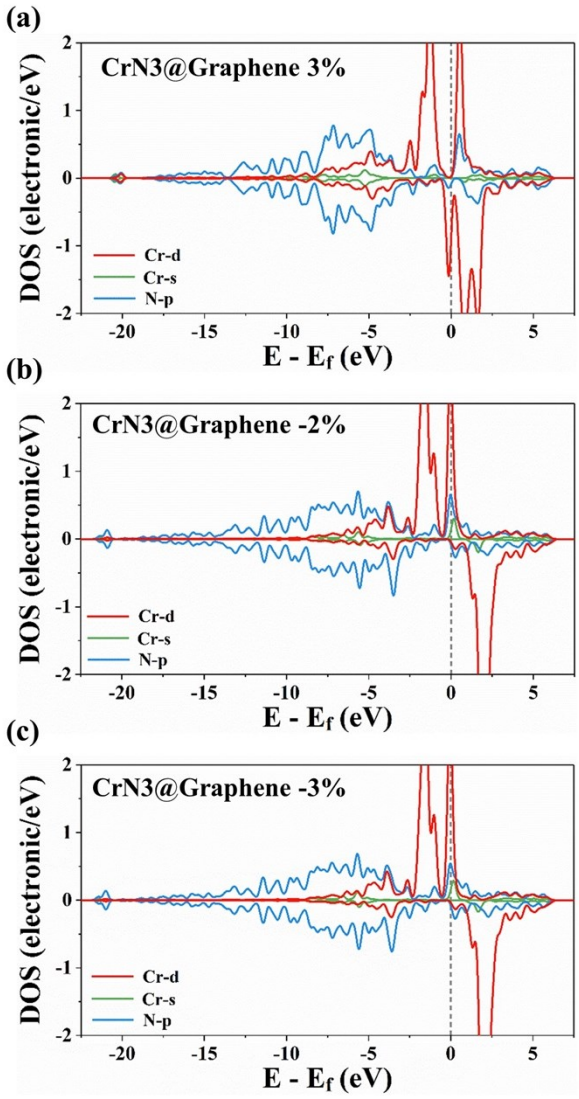

Figure S16. The partial density of states for CrN_3@Graphene with lattice strain of 3% **(a)**, -2% **(b)**, -3% **(c)**.

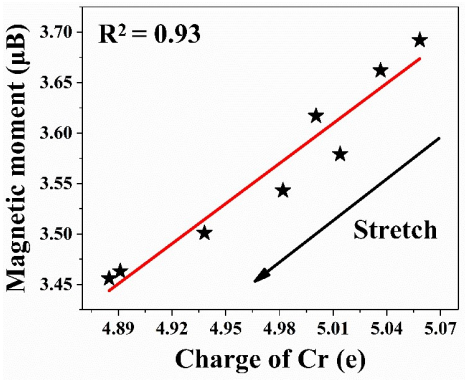

Figure. S17 The linear relationship between the magnetic moment of single Cr atom and the charge of single Cr atom.

Table S1. Bond length of N-N for nitrogen molecule which adsorbed on the single CrN_3@Gaphene with different lattice strain. The bond length of N-N for intrinsic nitrogen molecule is 1.11 Å.

Lattice Strain (%)	-3.0	-2.5	-2.0	-1.5	-1.0	0	1.25	1.75	2.0	2.5	3.0
Bond Length(Å)	1.15	1.15	1.15	1.15	1.15	1.15	1.15	1.15	1.15	1.15	1.15