Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Table S1: Total energies and edge energies for ZC_3NNR devices.

Device	Total Energy (eV)	Edge Energy (eV/Å)
9NC-2NN	-41540.878	-0.445
9NC-3NC	-45268.016	-0.571
9NC-4CC	-48985.410	-0.594
9NC-4NN	-48989.765	-0.640
9NC-5NC	-52720.729	-0.806
9NC-6CC	-56436.610	-0.814
9NC-6NN	-56445.061	-0.902
9NC-7NC	-60169.398	-0.999
9NC-8CC	-63885.993	-1.014
9NC-8NN	-63894.922	-1.108
9NC-3NC-H	-45374.809	-0.697
9NC-4NN-H	-49076.346	-0.720

Edge energies are related to the thermodynamically stable shape in exactly the same way that surface energies are used to predict the equilibrium shape of a three dimensional crystal through the Wulff construction[1-3]. Utilizing the method developed by Chee Kwan Gan on edge energy in hydrogenated and unhydrogenated graphene nanoribbons[2], the edge energy of C_3N model is calculated by the following formula,

$$E_{edge} = \frac{1}{2L}\left[E_{C_3N+H} - n_1 E_1 - \frac{n_H E_{H_2}}{2}\right]$$

,Where L is the length of ZC_3NNR device, E_{C_3N+H} is the total energy of the ZC_3NNR device which contains n_1 atomic combinations of C_3N, and E_1 is the energy per atomic combination of C_3N in the infinite, flat C_3N sheet with the lattice parameter that minimizes the energy. $E_{H_2}$is the energy of the H_2 molecule,

and n_H is the total number of H atoms.

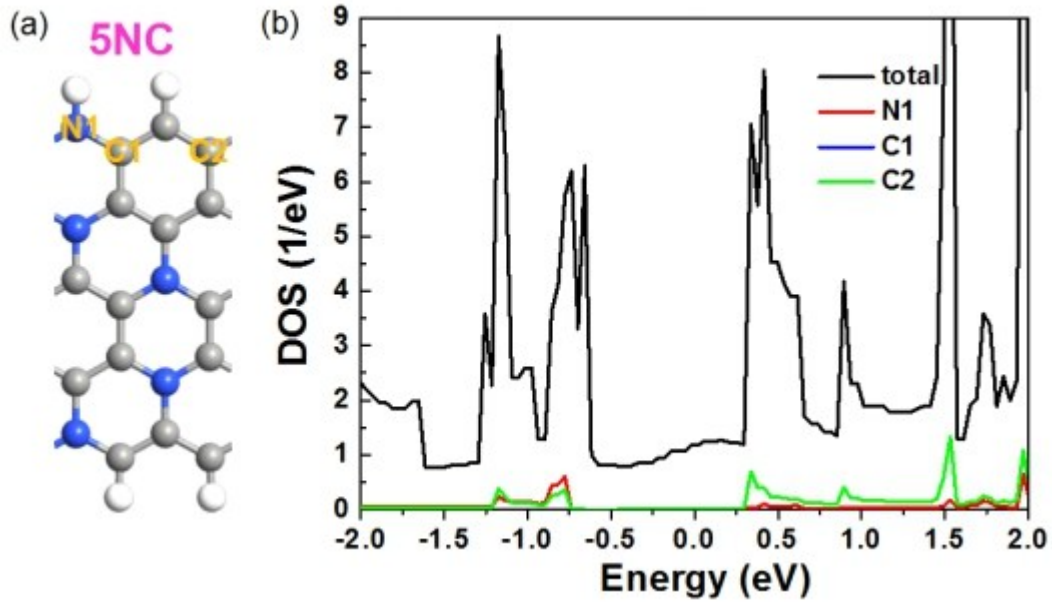
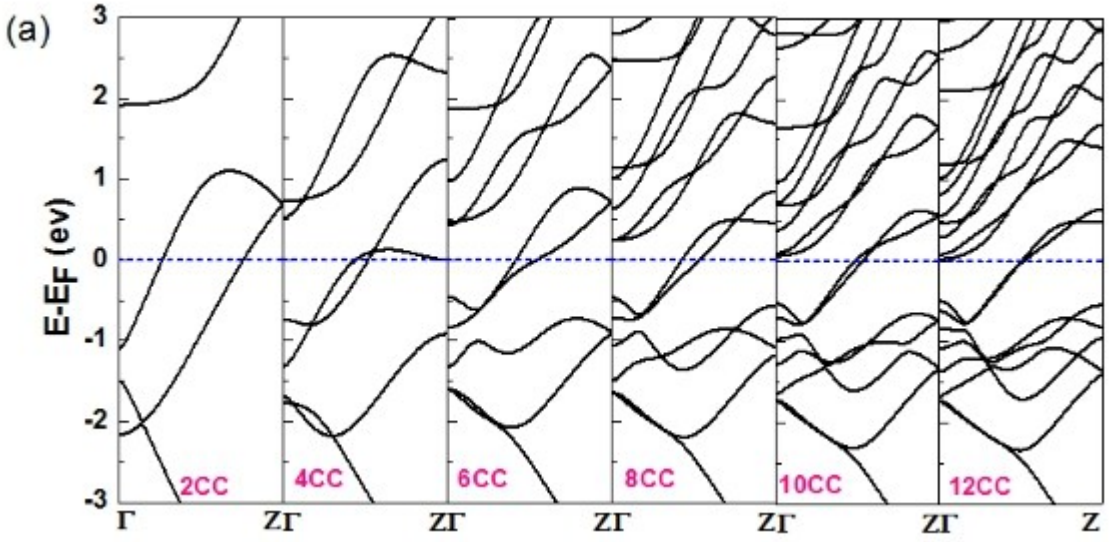


Fig. S1 (a) Schematic illustration of 5NC ZC_3NNR. The unit cell is omitted. Nitrogen, carbon and hydrogen atoms are shown by blue, gray and white balls, respectively. (b) The projected density of state (PDOS) as a function of energy for the 5NC ZC_3NNR.



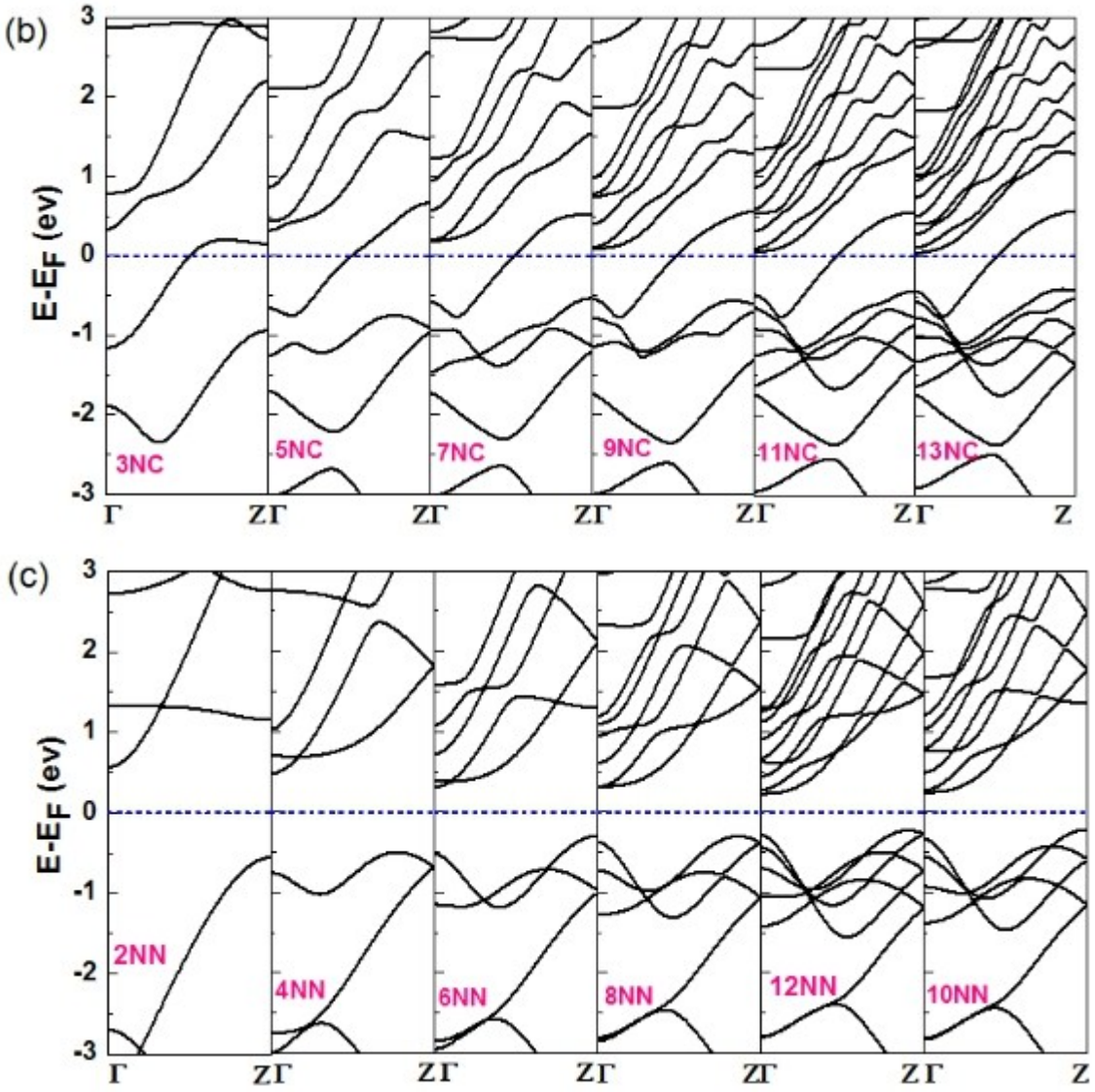


Fig. S2 Band structures around the Fermi level of (a) XCC; (b) XNC and (c) XNN ZC_3NNRs. X ranges from 2 to 13. The blue dotted line is the Fermi level.

Table S2: A summary of the structures and properties of ZC_3NNRs.

Edge morphology	ZC_3NNR	Property	9NC- heterojunction
All-carbon	4CC	Metal	Rectification
	6CC	Metal	Rectification、NDR
	8CC	Metal	NDR
One edge has N atoms	3NC	Metal	Rectification
	5NC	Metal	NDR
	7NC	Metal	NDR
Both edges have N atoms	2NN	Semiconductor	Rectifier diode
	4NN	Semiconductor	Rectifier diode
	6NN	Semiconductor	Rectifier diode
	8NN	Semiconductor	Rectifier diode

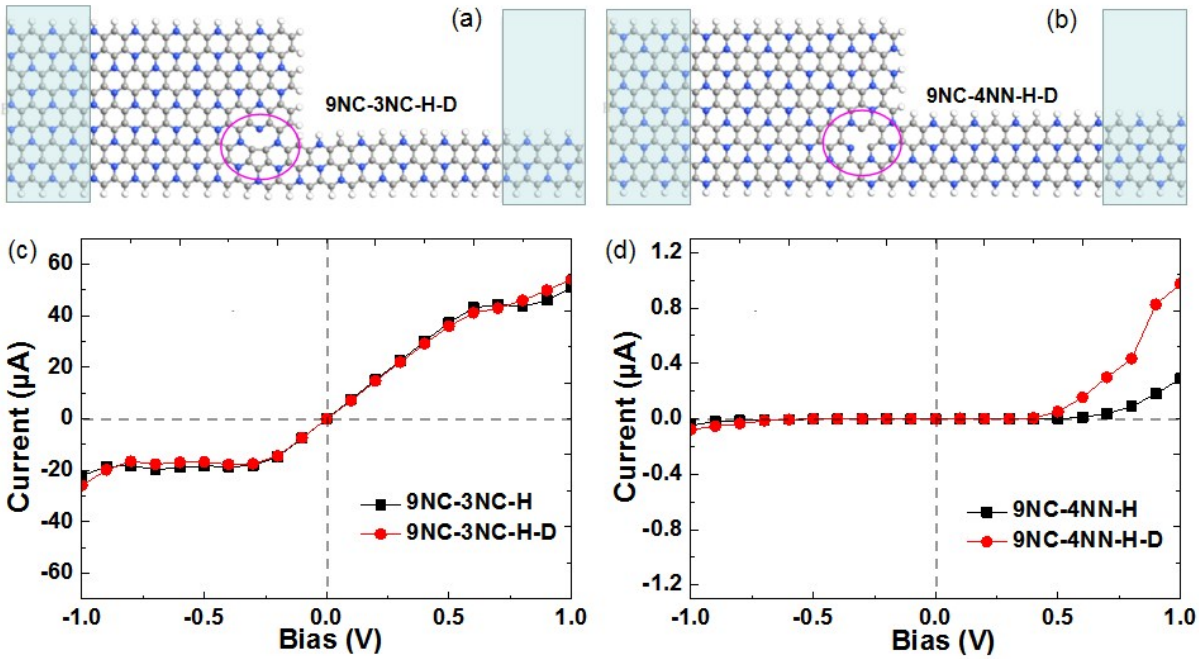


Fig. S3 Schematic illustrations of 9NC-3NC-H and 9NC-4NN-H devices with a point defect around the step, labeled as (a) 9NC-3NC-H-D and (b) 9NC-4NN-H-D, respectively. The I-V curves of (c) 9NC-3NC-H and 9NC-3NC-H-D devices; (d) 9NC-4NN-H and 9NC-4NN-H-D devices in the bias range from -1.0 to 1.0 V.

In Fig. S3 (a) and (b), both 9NC-3NC-H and 9NC-4NN-H devices have a point defect around the step, labeled as 9NC-3NC-H-D and 9NC-4NN-H-D, respectively. After optimization, a five-membered ring and a nine-membered ring are formed at the point vacancy of the 9NC-3NC-H-D, however the structural change at the point vacancy of 9NC-4NN-H-D is not obvious.

In Fig. S3(c), it can be seen that the presence of a point defect causes a very small change in the I-V curve of the 9NC-3NC-H device, indicating that the point defect here does not affect its electronic transport property. For the 9NC-4NN-H device, as shown in Fig. S3(d), the presence of a point defect causes an obvious increase in the forward-conducting current, while a little increase in the reverse current under high bias, so the forward-conducting and reverse-blocking rectifier diode behavior is still very significant. Therefore, such a point defect near the step does not change the overall electronic transport properties of the step-like ZC_3N devices.

Supplementary References

1. A. Pimpinelli and J. Villain, Physics of Crystal Growth Cambridge University Press, United Kingdom, 1998.

2. Gan C K, Srolovitz D J. First-principles study of graphene edge properties and flake shapes. Physical Review B, 2010, 81:125445.

3. Koskinen P, Malola S, Häkkinen H. Self-passivating edge reconstructions of graphene. Physical Review Letters, 2008, 101(11): 115502.