Supplementary Note 1: Strain effect on edge states of zigzag GNR by first-principles calculations

In the main text, we study the strain effect on the electronic property of a zigzag graphene nanoribbon (GNR) with $n = 8$ C chains using self-consistent charge density-functional tight-binding (SCC-DFTB) method\(^1\). The results summarized in Fig. 2 of the manuscript show that electronic bands of edge states shift downwards (upwards) if the GNR is in extension (compression).

To confirm it, we also carry out a first-principles calculation on the same GNR using the Vienna ab initio simulation package (VASP)\(^2\) within the density functional theory (DFT), where the projector augmented plane wave (PAW) method\(^3,4\) is adopted, and the generalized gradient approximation (GGA) in the framework of Perdew-Burke-Ernzerhof (PBE)\(^5\) was adopted for the exchange-correlation interaction. The positions of the atoms are obtained by structural optimization until the forces on each atom are smaller than 0.01 V/Å, and a vacuum layer of 20 Å is used to avoid possible effects of image supercells. A plane wave basis is set with a kinetic-energy cutoff of 500 eV. For this quasi-one dimensional GNR, the Brillouin zone is sampled with 100 \(k\) points. The fully relaxed GNR has a translational periodicity of $|T_0| = 2.47$ Å, close to that ($|T_0| = 2.46$ Å) obtained by SCC-DFTB. The results for the GNR with axial strains of $\pm 5\%$ are shown in Supplementary Figure 1, which displays similar behaviors as revealed by SCC-DFTB.

Supplementary Note 2: Shift of energy levels of edge states with axial strain

In the main text, we show that for a zigzag GNR with $n = 8$ C chains the energy levels of edge states shift downwards (upwards) with axial compression (tension) using SCC-DFTB, see also Supplementary Figure 2(a). To get more insight to this shift, we consider different strains within the range of $-5\% < \varepsilon < 5\%$. Supplementary Figure 2(b) shows the shift of the valance band of edge states at $k = \pi/T$, which display a linear dependence on strain $\varepsilon$. 
Supplementary Note 3: Half-metallicity obtained through external electric field

Son et al. suggested that a half-metallic state can be induced by applying an external electric field transversely across zigzag GNRs and demonstrated this idea using first-principles calculation. Here, we carry out similar calculation on a zigzag GNR with \( n = 8 \) C chains using SCC-DFTB. Our calculation shows that spin-splitting in edge states are induced upon the presence of external electric field, Supplementary Figure 3(b). Furthermore, band gap of the spin-up states reaches a complete closure as the electric field strength reaches \( E_{\text{ext}} = 0.11 \ \text{V/Å} \), indicating the emergence of half-metallic state, Supplementary Figure 3(c). These results are consistent with those first-principles prediction of Son et al., thus demonstrate once more the validity of SCC-DFTB in dealing with the spin-polarization in zigzag GNRs. It is also interesting to point out that the half-metallic gap obtained with in-plane bending compares well with that obtained through external electric field, Supplementary Figure 3(d).
Supplementary Figure 1 | Electronic band structures of a zigzag GNR with $n = 8$ C chains under different conditions: under axial tension $\varepsilon = 5\%$ (left), stress-free $\varepsilon = 0$ (middle), and under axial compression $\varepsilon = -5\%$ (right).
Supplementary Figure 2 | (a) Electronic band structures of a zigzag GNR with \( n=8 \) C chains under different conditions: under axial tension \( \varepsilon = 5\% \) (left), stress-free \( \varepsilon = 0 \) (middle), and under axial compression \( \varepsilon = -5\% \) (right). (b) The shift of the energy level of the valance band of edge states at \( k = \pi/T \) with strain \( \varepsilon \). The dashed line represents a simple linear fitting.
Supplementary Figure 3 | Electronic band structures of a zigzag GNR with n=8 C chains under external electric fields ($E_{ext}$): (a) $E_{ext} = 0.00$, (b) $E_{ext} = 0.05$ V/Å, and (c) $E_{ext} = 0.11$ V/Å. The red dashed lines (green solid lines) denote energy bands with spin-up (spin-down) orientation. (d) Comparison of half-metallic (HM) gaps obtained through external electric field and in-plane bending, respectively.
Supplementary References