

Supplementary Figure S1. The detailed procedure for TEM imaging of graphene torn edge. (a) TEM image of a graphene torn edge before the tear propagation. Once a tear is identified at low magnification, the electron beam is blocked to minimize the e-beam-induced damage. With a high magnification, we set a proper focus setting at a location far away from the identified tear. We guess its propagation direction with the shape of a graphene tear and start to image a sample approaching from the carbon support edge as shown. (b) TEM image of the same graphene torn edge after tear propagation with electron beam. (c) Diffraction pattern of the graphene membrane around the graphene edge. Microscopic edge directions can be assigned from the diffraction pattern.



Supplementary Figure S2. The calculated number of scattering events per second with transferred energy T shown as a function of T for single carbon atom. Two plot show p(T) with and without the thermal lattice vibrations, respectively. The same p(T) are shown as the semilog scale in the inset.

Supplementary Table 1. The analyzed data set to study graphene edges in armchair (AC) and zigzag (ZZ) edge configurations.

Edge type	# of analyzed images	Edge length	Pristine edge length	Otheredge length
Armchair (AC)	6	90.2 nm	82.1 nm (91 %)	8.1 nm (9 %)
Zigzag (ZZ)	4	43.6 nm	39.1 nm (90 %)	4.5 nm (10 %)

Supplementary Table 2. Detailed edge configuration (for location 1-16) of a zigzag edge analyzed in Figure 4. Pentagon-heptagon (5-7) reconstructed edge segments are shown in red, while hexagon (6) rings are shown in blue. The raw data can be found in the Supplementary Movie.



Supplementary Note 1

The cross section for Coulomb scattering between an electron and a carbon atom

An analytic approximation of the cross section for Coulomb scattering between an incident electron and a nucleus^{31,32} was employed to address the energy transfer rate to carbon atoms at the graphene edge during experimental imaging conditions. The scattering cross section for the events when energy *T* or higher is transferred can be written as

$$\sigma(T) = \frac{4Z^2 E_{\rm R}^2}{m_e^2 c^4} \left(\frac{T_{\rm max}}{T}\right) \pi a_0^2 \left(\frac{1-\beta^2}{\beta^4}\right) \left\{ 1 + 2\pi\alpha\beta \sqrt{\frac{T}{T_{\rm max}}} - \frac{T}{T_{\rm max}} \left[1 + 2\pi\alpha\beta + \left(\beta^2 + \pi\alpha\beta\right) \ln\left(\frac{T_{\rm max}}{T}\right) \right] \right\}$$
(S1)

where Z is the atomic number of the target atoms, $E_{\rm R}$ the Rydberg energy (13.6 eV), a_0 the Bohr radius of the hydrogen atom (5.3 × 10⁻¹¹ m), $\beta = v/c$ (electron velocity divided by the speed of light c), and $\alpha = Z/137$. $T_{\rm max}$ is the maximum transferred energy in the scattering event and can be written as

$$T_{\rm max} = \frac{2E(E + 2m_e c^2)}{Mc^2}$$
(S2)

under the assumption that the target atom mass M is much heavier that the electron mass m_e (E is the kinetic energy of the electron). At 80 kV TEM operation, T_{max} is 15.8 eV for carbon atoms. With the experimental imaging condition of electron beam intensity $j = 2 \times 10^6 \text{ e/s} \cdot \text{nm}^2$, we can calculate the number of scattering events per second as a function of the energy transfer T to single carbon atom.

$$p(T) = -\frac{\mathrm{d}\sigma(T)}{\mathrm{d}T}j \tag{S3}$$

Supplementary Note 2

The effect of thermal lattice vibrations on the cross section for Coulomb scattering

Recently, the thermal lattice vibrations were found to have an effect on the displacement cross section³³. When the target carbon atom is not at rest due to the vibrations of the lattice, the transferred energy T from incident electron can change. The modified maximum transferred energy can be written as

$$\widetilde{T}_{\max}(E_n) = \frac{(rc+t)^2}{2Mc^2}$$
(S4)

with $r = \frac{1}{c} \left(\sqrt{E(E + 2m_e c^2)} + \sqrt{2m_e c^2 E_n} \right)$ and $t = \sqrt{(E + E_n)(E + 2m_e c^2 + E_n)}$

where $E_n = Mv^2/2$ is the initial kinetic energy of the target atom³³.

The mean square velocity of a carbon atom in graphene can be calculated as

$$\overline{v^2} = \frac{9k_b}{8M}\theta_D + \frac{9k_bT}{M}\left(\frac{T}{\theta_D}\right)^3 \int_0^{\theta_D/T} \frac{x^3}{\exp(x) - 1} dx$$
(S5)

where k_b is the Boltzmann constant, *T* the temperature, and θ_D the Debye temperature (1287 K for graphene³³). At room temperature T = 293 K, we found that the average kinetic energy of the target atoms, $\overline{E_n} = M\overline{v^2}/2 = 0.069$ eV.

To estimate the average maximum transferred energy \tilde{T}_{max} , we put the average kinetic energy $\overline{E_n}$ into the Eq. (S4), which gives $\tilde{T}_{max}(\overline{E_n}) = 17.9 \,\text{eV}$. Therefore, $\tilde{\sigma}(T)$, Eq. (S1) with modified maximum transferred energy $\tilde{T}_{max}(\overline{E_n})$, gives the scattering cross section for the events when energy *T* or higher is transferred when we take thermal lattice vibrations into consideration. From this, the calculate the number of scattering events per second as a function of the energy transfer *T* to single carbon atom is written as

$$\tilde{p}(T) = -\frac{\mathrm{d}\tilde{\sigma}(T)}{\mathrm{d}T}\,j\tag{S6}$$

The supplementary Figure S2 shows the calculated p(T) and $\tilde{p}(T)$. We found that the effect of thermal lattice vibrations is not significant. The Figure 4c shows the p(T) in the energy range to 15.8 eV.

Supplementary Note 3

Other radiation effects on graphene from high-energy electron

We discuss different radiation effects on graphene from incident high-energy electrons. The important primary radiation effects^{32,39} includes

- 1) electronic excitation or ionization of individual atoms,
- 2) collective electronic excitations (plasmons)
- 3) generation of phonons, leading to heating of the targets,
- 4) displacement of atoms (including sputtering of atoms)

The first effect, electronic excitation and ionization of individual atoms, is quickly quenched due to the high density of delocalized electrons in metal and graphitic materials, including graphene. The energy will be quickly dissipated throughout the specimen, preventing a direct ionization-induced ionic movement. Phonon can be generated by electron scattering with carbon nuclei but it is mainly generated from the dissipation of plasmons into phonon modes^{32,39}. The energy transferred through processes from 1) to 3) together is related to internal thermal energy increase due to high-energy electron.

We can estimate the energy transfer rate to a specimen which is converted into internal thermal energy. If the beam-induced excitation of phonons is high enough, they will have an implication in the observed flipping rates between 57 and 66 zigzag edge configurations. Previously, for carbon samples, the average energy transfer value by one incident electron per unit mass thickness $(\Delta Q/\Delta x)$ to specimen heating was found to be ~ 3 eVcm²/µg³⁹. Temperature increase of the specimen can be written as

$$\Delta T = \frac{j\rho}{4e\kappa} \frac{\Delta Q}{\Delta x} R^2 \tag{S7}$$

where *j* is electron beam intensity, ρ is the mass density of graphene (2.2 g cm⁻³), κ is the thermal conductivity of graphene (~ 4000 W m⁻¹K⁻¹)⁴⁰, *R* is the distance between the center of electron beam to the location where the temperature stay constant (assumed to be 20 μ m)³⁹. From this, we obtain $\Delta T \sim 5$ K with our experiment parameters. We find that the temperature increase due to inelastic scattering is not significant in our experiment set-up. Therefore, our assumption in the main manuscript that the observed transformation between 5-7 and 6-6 ZZ edge configurations is mainly the consequence of the displacement effects (electron – carbon atom scattering) is reasonable.

Supplementary References

39. Reimer, L. & Kohl, H. Transmission electron microscopy: physics of image formation. (Springer, New York, U.S.A. 2008).

40. Balandin, A. A. *et al.*, Superior Thermal Conductivity of Single-Layer Graphene. *Nano Lett.* **8**, 902 (2008).