Supplemental Information

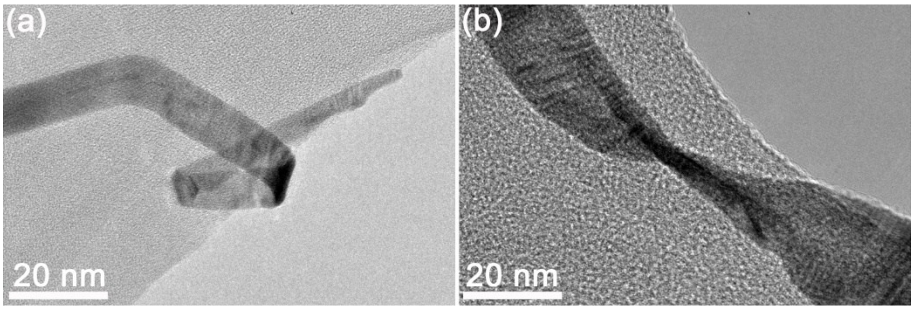

Figure S1 (a,b) Another two samples with folding parts which reveal the characteristics of 4H Au NRBs.

Table S1 Size evolution of the ultrathin 4H Au NRB (sample 1) under E-beam irradiation as a function of time (high magnification).

T (min)	w_p (nm)	w_t (nm)	w_m (nm)	w_e (nm)	w_s (nm)	φ_h	φ_l
0	10.9	10.9	10.9	10.9	0	0	0
10	10.5	9.4	9.9	9.95	0.55	0.05528	0.03362
20	10.5	8.1	9.5	9.3	1.2	0.12903	0.06724
30	10.4	7.3	9.0	8.85	1.55	0.17514	0.10085
40	10.3	6.7	8.6	8.5	1.8	0.21176	0.13447
50	10.3	5.7	8.3	8.0	2.3	0.2875	0.16810

Note: Black numbers are experimental values, and red numbers are calculated values.

The meanings and calculations of the alphabets used in the paper and SI:

T: Time,

t: Initial thickness,

t_p: Peak thickness,

t_t: Trough thickness,

w: Initial width,

w_p: Peak width,

w_t: Trough width,

w_a: Average width, $w_a = (w_p + w_t)/2$,

w_m: Mid width, width of the site in the mid of peak and trough, $w_m \approx w_a = (w_p + w_t)/2$,

w_e: Effective width, $w_e \approx w_m \approx w_a = (w_p + w_t)/2$,

w_s: Shrinking width, $w_s = (w_p - w_t)/2$,

φ: Reduction rate, $\varphi = \frac{w_s}{w_e} = \frac{w_e - w_t}{w_e} = \frac{(w_p + w_t)/2 - w_t}{(w_p + w_t)/2} = \frac{(w_p - w_t)/2}{(w_p + w_t)/2}$,

φ_h: Reduction rates at high irradiation intensity per unit area like sample 1 (high magnification as Figure 2),

φ_l: Reduction rates at low irradiation intensity per unit area like samples 2-4 (low magnification as Figure 4). It can be calculated using equation (S1). Note: for different magnification, the equation is also different. Equation (S1) is used for the magnification observing samples 2-4 in Figure 4.

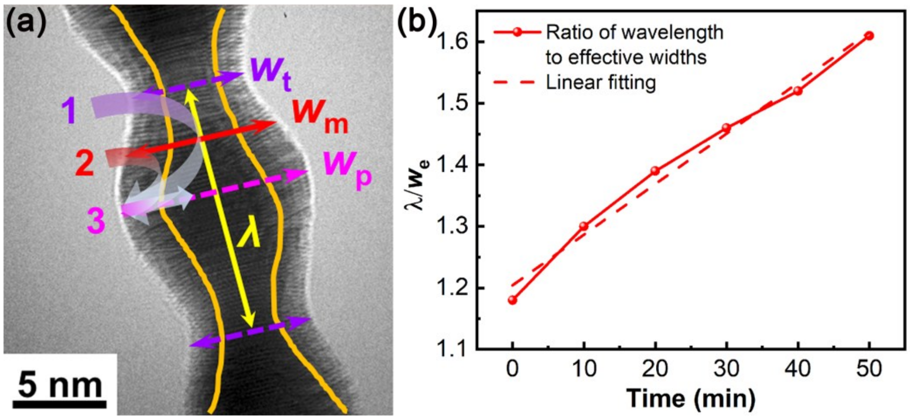

Figure S2 (a) Atom migration paths in the Rayleigh instability evolution processes of the ultrathin 4H Au NRB: at the trough sites, the atoms migrated to the inner radial direction, then to the longitudinal direction pointing to the peak sites, finally to the outer radial direction pointing to the peak sites (violet arrow 1, the density of the color represents the atom migration density; only one path was depicted, the other symmetric paths were omitted); at the peak sites, the atoms mainly migrated to the inner radial direction (pink arrow 3); at the mid sites, the atoms migrated like path 1, with a weak migration degree (red arrow 2); Notes: (i) the atoms also migrated to the width direction at the same time. The ripples left due to atom migrations between the edges and orange curves, and the wavelength of Rayleigh instability marked with yellow line from trough to trough; (ii) the TEM image was taken from Figure 2f. (b) Ratio of wavelength to effective widths as a function of time.

Table S2 Size evolutions of ultrathin 4H Au NRBs (sample 2-4) under E-beam irradiation as a function of time (low magnification).

Sample	0 min	20 min		30 min		40 min	
	w_m (nm)	w_p (nm)	w_t (nm)	w_p (nm)	w_t (nm)	w_p (nm)	w_t (nm)
2	11.8	11.4	9.9	11.2	8.95	11.0	8.0
3	9.6	10	8.8	10.2	8.4	10.4	8.0
4	14.5	13.9	11.7	13.6	10.3	13.3	8.9

Note: Black numbers are experimental values, and red numbers are calculated values according to linear relationship between the reduction rates and irradiation time.

Table S3 Size evolutions and reduction rates (φ_l) of ultrathin 4H Au NRBs (sample 2-4) under E-beam irradiation at 20 min (low magnification).

Sample	0 min	20 min		w_e (nm)	w_s (nm)	φ_l
	w_m (nm)	w_p (nm)	w_t (nm)			
2	11.8	11.4	9.9	10.65	0.75	0.07042
3	9.6	10	8.8	9.4	0.6	0.06383
4	14.5	13.9	11.7	12.8	1.1	0.08594

Note: the exponential equation between reduction rates (low magnification) and initial widths of the 4H Au NRBs:

$$\varphi_l = 6.44189 * 10^{-4} * \exp\left(\frac{w}{3.76321}\right) + 0.05557$$
, the fitting curve in Figure 4g. (S1)

Using equation (S1), the equivalent reduction rate (φ_l) of sample 1 with width of 10.9 nm at 20 min under low irradiation intensity per unit area can be obtained (0.067236). Then, according to the linear relationship between reduction rates and irradiation time, the equivalent reduction rates (φ_l) of sample 1 at other time can be decided by:

$$\varphi_l = \frac{0.067236}{20} * T = 0.0033618 * T$$
. (S2)

The equivalent reduction rate values are shown in Table S1. The relationship curve can be obtained as shown in Figure 4h (orange line).

s_l=0.0033618 is the slope of the reduction rate curve at low irradiation intensity per unit area. The slope of the actual reduction rate curve under high irradiation intensity per unit area can be obtained (s_h=0.00558) by linear fitting (blue dashed line in Figure 3d or Figure 4h). So the ratio (R=s_h/s_l=0.00558/0.0033618=1.66) of the two slopes indicates the enhancement of the high irradiation intensity on Rayleigh instability. The ratio will increase with the difference rise of the two irradiation intensities per unit area. Every irradiation intensity per unit area (magnification) has a specific slope reduction rate curve even for the same sample.