Supporting Information

Calculation details:

According to thermal spike theory, the temperature distribution in cylindrical coordinates related to a bombarded spot is: [1-2]

$$T(F_d, r, t, T_S) = \frac{F_d}{4\pi\kappa t}\exp(-\rho C r^2 / 4\kappa t) + Ts \quad \text{(S1)}$$

where F_d is the energy deposited into the track per unit length, κ is the thermal conductivity of TiO_2 (11.7 W/m/k), ρ is the density of TiO_2 (3.8 g/cm^3), C is the heat capacity of TiO_2 (57.12 J/mol/K), t is the relaxation time, r is the related distance to the bombarded spot, and T_S is the substrate temperature.[3]

As a result, the viscosity on the substrate surface decreases due to thermal activation:

$$\eta_{thermal} = \eta_0 \exp(E_{flow} / kT) \quad \text{(S2)}$$

where η_0 is a material dependent prefactor, E_{flow} is the activation energy for flow, and k is Boltzmann's constant. We can use the first order estimates for η_0 and E_{flow}: $\eta_0 = 10^{-3}$ Pa·s and E_{flow} =1.0 eV. Subsequently, the shear stress can relax in the locally heated region at a rate of R= $\mu / \eta_{thermal}$ (where μ is shear modulus at a value of 90 GPa),[3] and the temperature is cooled back to the substrate temperature. The total amount of stress relaxation up to a time τ is:

$$\Omega(F_d, r, \tau, T_S) = 1 - \exp[\int_0^{\tau} -R(F_d, r, t, T_S)dt] \quad \text{(S3)}$$

The ion-induced viscosity η can be calculated by the cross section $\theta_{eff}(F_d, T_S)$ of full stress relaxation:

$$\eta_{rad} = \frac{\mu}{3B\theta_{eff}(F_d, T_S)} = \frac{\mu}{3B\int_0^{2\pi}\int_0^{\infty}\Omega(F_d, r, \infty, T_S) r dr d\theta} \quad \text{(S4)}$$

$$\eta = \frac{\eta_{rad}}{f} \quad \text{(S5)}$$

where B is dependent on Poisson's ratio v by: $B = 6(1-v)/(5-4v)$, f is the ion flux. Poisson's ratio v for TiO_2 is 0.28.[3]

As shown in Fig. S1, for 30 kV FIB sculpting, the total amount of stress relaxation at 1 ps overlaps with those of 10 ps and when $\tau \rightarrow \infty$, which means the stress has almost completely relaxed at a time scale of picosecond. Similarly, the time scale of stress relaxation for 2 kV-16 kV FIB sculpting is also picosecond. Monte Carlo simulation by SRIM shows F_d is 1.89 kV/nm, 1.6 kV/nm, 1.27 kV/nm, 1.06 kV/nm, and 0.71 kV/nm for 30 kV, 16 kV, 8 kV, 5 kV, and 2 kV Ga^+ beam sculpting, respectively.[4] Therefore, the temperature distribution at local thermal spikes under 2 kV-30 kV FIB sculpting can be calculation by equation (S1) as shown in Fig. 5.

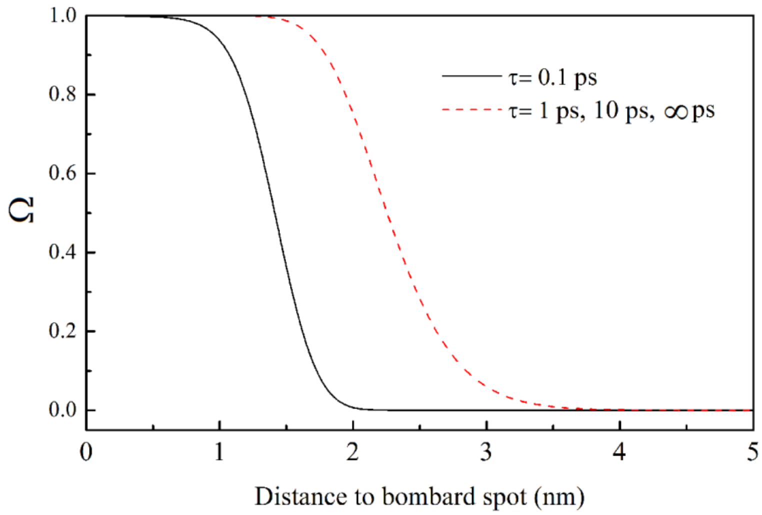

Fig. S1. Calculations of the total amount of stress relaxation at T_S= 300 K up to time τ: 0.1 ps, 1 ps, 10 ps, and ∞.

References:

1. M. L. Brongersma, E. Snoeks and A. Polman, *Appl. Phys. Lett.*, *1997*, ***71***, 1628.
2. H. Trinkaus, *J. Nuclear Mater.*, *1995*, ***223***, 196.
3. J. F. Shackelford and W. Alexander, *CRC Press*, *2001*.
4. J. F. Ziegler, J. P. Biersack and U. Littmark, *The Stopping and Range of Ions in Solids*. Pergamon: New York, 2008.