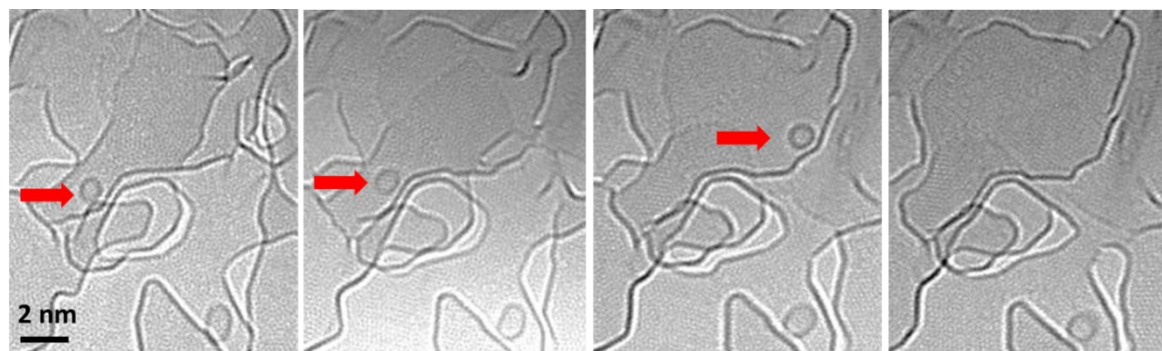
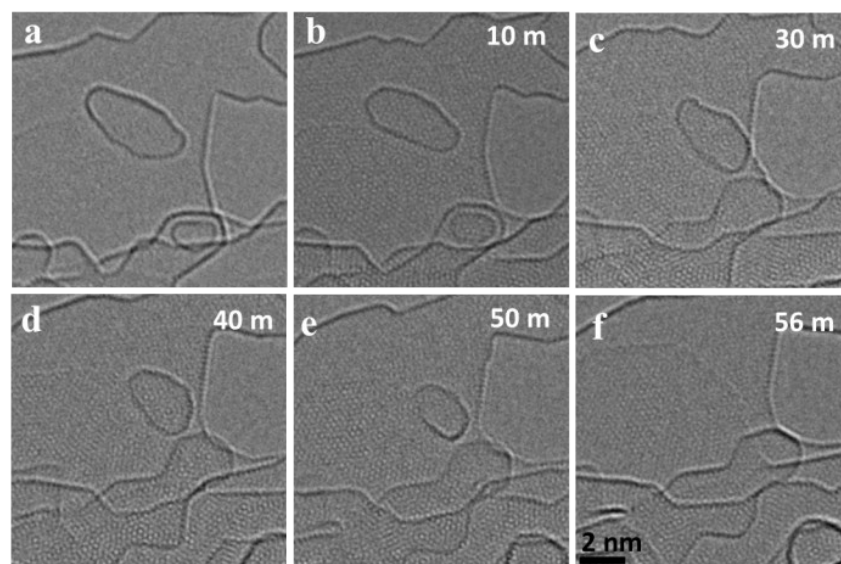


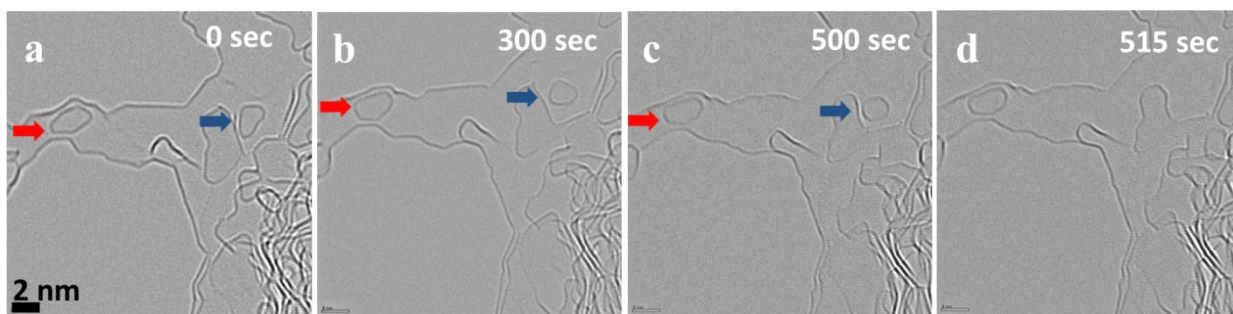
### Supplementary Information



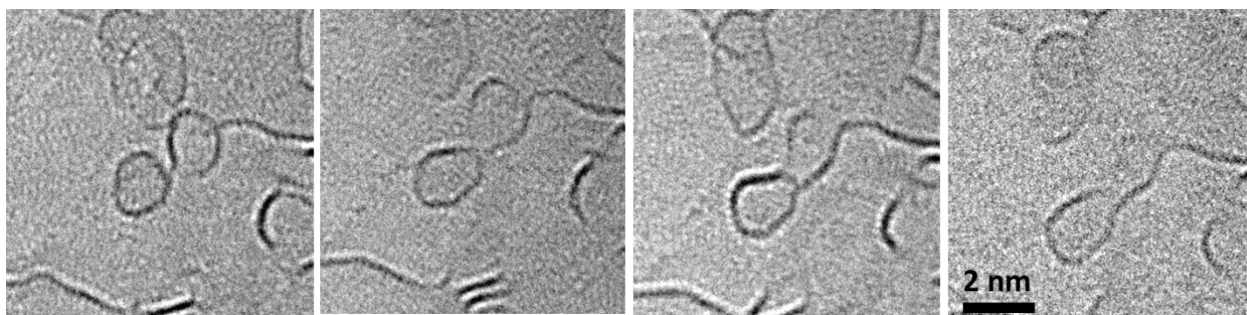
Supplementary figure 1: BF-TEM images showing migrating cage-like nanostructure during continuous imaging for 30 minutes at 1200 °C.



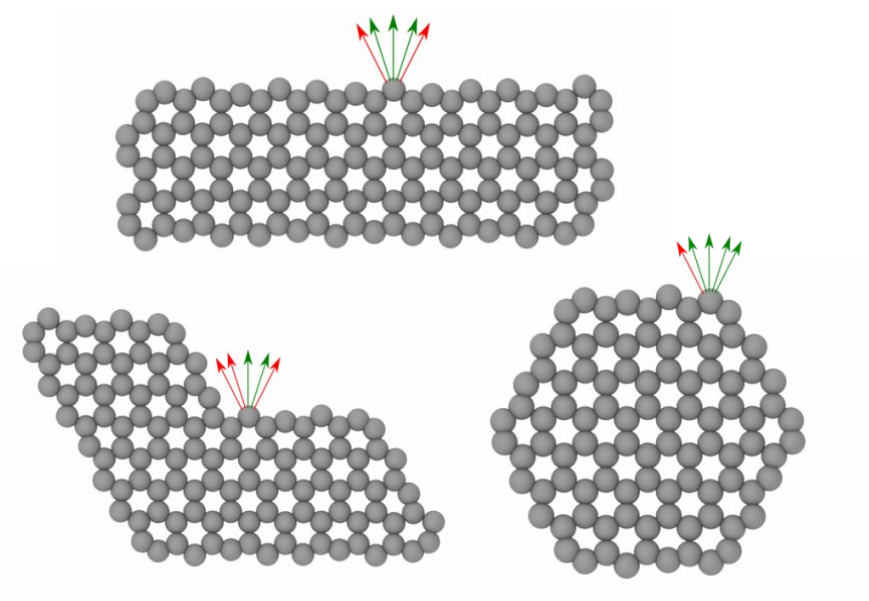
Supplementary figure 2:(a-f) shows the dynamics of a small flake heated over a period of 56 minutes at 1200 °C (supplementary video S6), where images have been taken at 1-minute intervals. The flake seems to be pinned to an edge. The flake is slowly moving towards an adjacent flake along an edge before it finally disappears from the frame. During migration, the flake shows a constant reduction in size while the adjacent flake is growing.



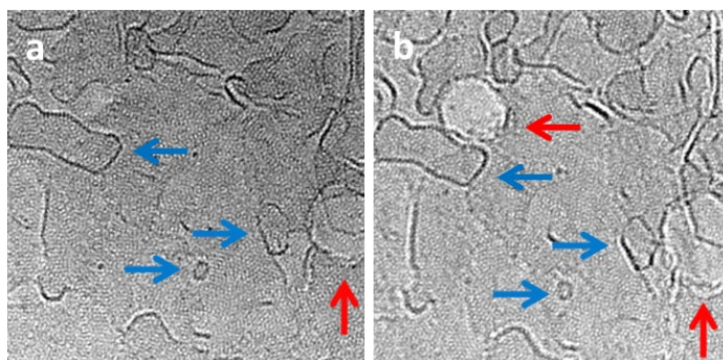
Supplementary figure 3: BF-TEM images showing the difference in behavior of two different flakes at the same temperature and illumination conditions: the flake marked by a red arrow is stable with minimum mass loss while the one indicated in blue is losing atoms quickly and migrating out of the field of view.



Supplementary figure 4: HRTEM images showing merging of two small, similar sized nanoflakes during heating at 900 °C



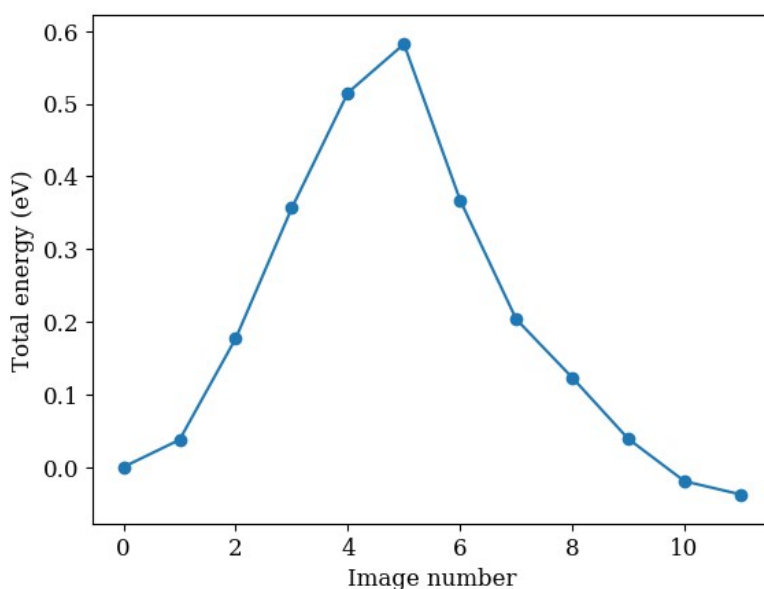
Supplementary figure 5: SMD simulations of knock-out events at different angles of momentum transfer (corresponding to a kinetic energy of 16 eV). The momentum is set on the first time step parallel to the flake plane in vertical direction and with deviations  $\pm 18^\circ$  and  $\pm 27^\circ$ . Green arrows: successful atom escape, red arrows: unsuccessful atom escape (only local rearrangement).



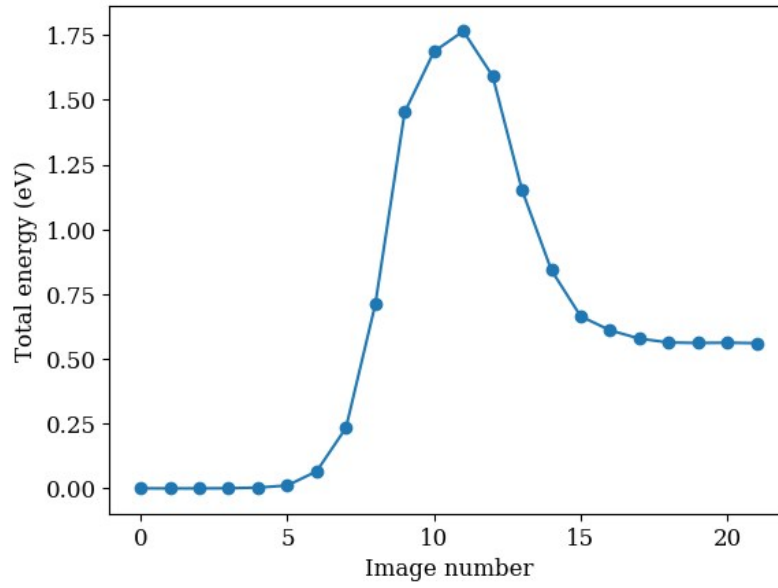
Supplementary figure 6: BF-TEM images at room temperature before (a) and after (b) continuous exposure for 10 minutes. The exposure resulted in an expansion of an already formed hole and the creation of a new hole (indicated by red arrows). The nanostructures (indicated by blue arrow) remained stable under the beam without any size/shape changes and migration, which shows the influence of temperature in the observed dynamics.

## Nudged Elastic Band (NEB) method

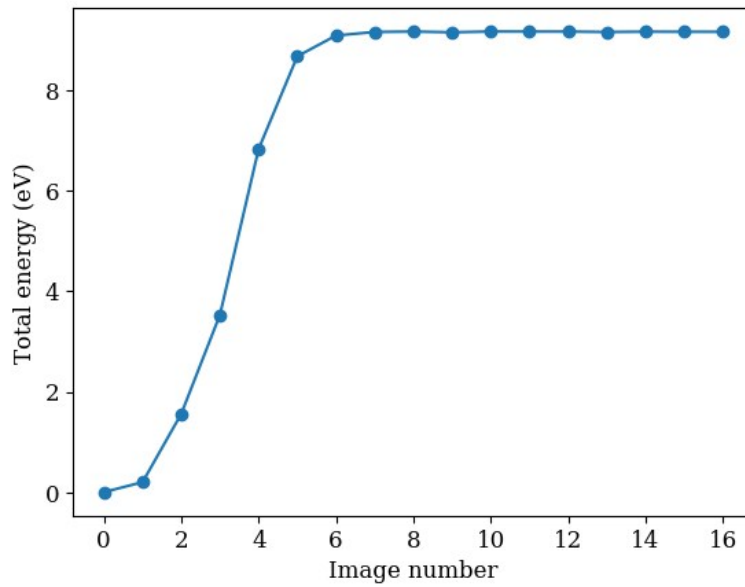
The NEB method is used to find the saddle point of the minimum energy path between given initial and final atomistic configurations. Therefore, intermediate states of the system (images) are coupled by harmonic springs, which ensure a uniform distribution of sampling points along the path. This coupled system is **then** minimized which results in the minimum energy path. The energy computation of each image can be based on analytical force fields as well as *ab initio* methods. Additionally, a climbing image, which doesn't feel any spring forces but tends towards the saddle point, can be used to improve the transition state. Detailed information about this method can be found in Henkelman *et al.*<sup>37</sup>.



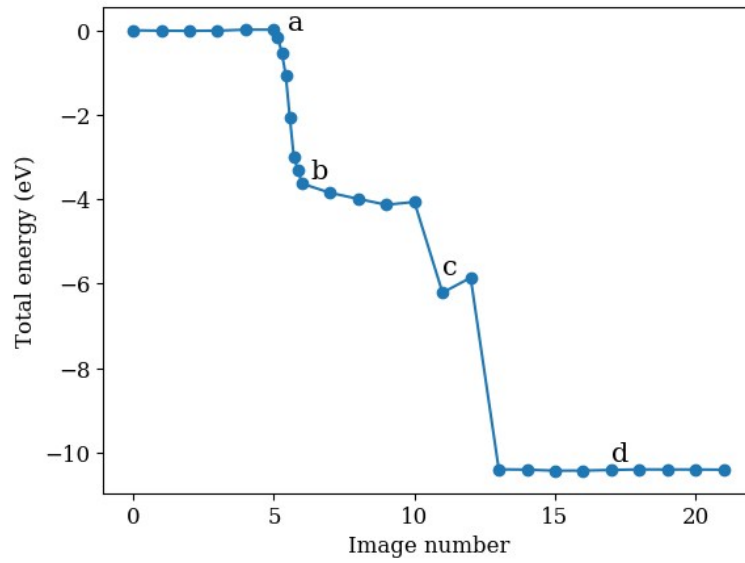
Supplementary figure 7: Energy profile of edge diffusion process (figure 5a,  $E_A = 0.58 \text{ eV}$ ). The energies are shifted by the value of the initial state.



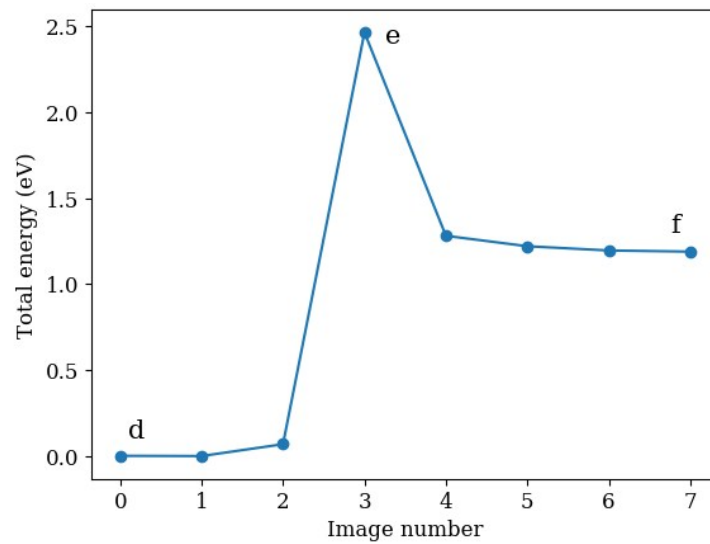
Supplementary figure 8: Energy profile of edge reconstruction process (figure 5b,  $E_A = 1.76 \text{ eV}$ ). The energies are shifted by the value of the initial state.



Supplementary figure 9: Energy profile of direct atom removal process (figure 5c,  $E_A = 9.17 \text{ eV}$ ). The energies are shifted by the value of the initial state.

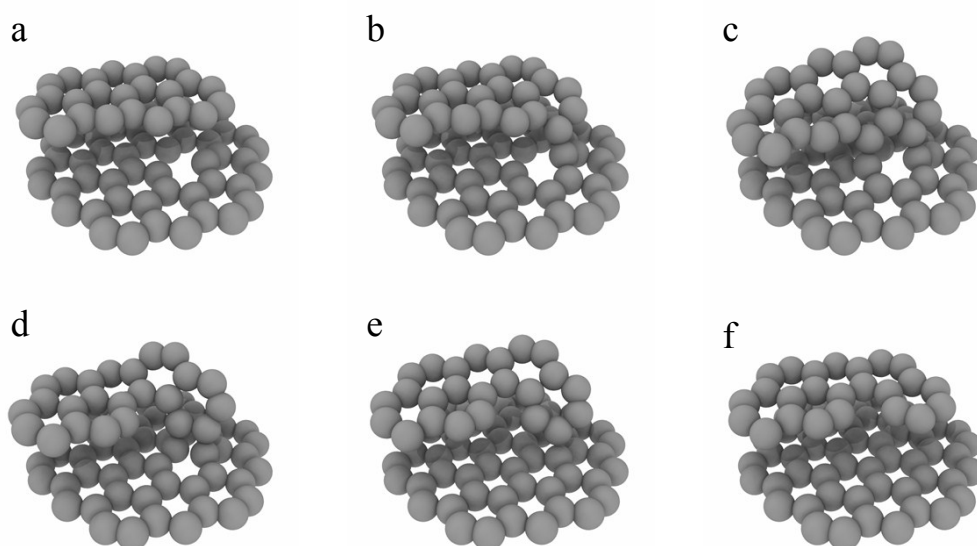


Supplementary figure 10: Energy profile of pinning process (figure 5d,  $E_A = 0.015 \text{ eV}$ ). The profile between image number 5 and 6 was recalculated with additional data points. The lowercase letters refer to the corresponding structures shown in supplementary figure 12. The energies are shifted by the value of the initial state.





Supplementary figure 11: Energy profile of depinning process with simultaneous atom removal (figure 5d,  $E_A = 2.47 \text{ eV}$ ). The lowercase letters refer to the corresponding structures shown in supplementary figure 12. The energies are shifted by the value of the initial state.



Supplementary figure 12: Intermediate states of a pinning (a - d) and depinning (d - f) process. The corresponding relative energies are given in supplementary figure 10 and 11. In the MD simulation of a similar pinning process (supplementary video S3) a stable intermediate pinned state can be observed, which indicates that the direct coupling of the reacting atoms to the substrate and the flake can enable a fast dissipation of the reaction energy, preventing an immediate depinning process. However, reactions with transient pinning states and instant depinning cannot be ruled out.