Electronic Supplementary Information for

Atomistic modelling of electron beam induced structural transformations in deposited metal clusters

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Content:

1) Summary of typical STEM experimental parameters used in the present study

Table S1. Typical experimental parameters for STEM measurements of Au_{309} clusters deposited on a carbon substrate. These parameters have been provided by the Swansea Nanomaterials Lab[1].

Parameter	Value
Deposition energy (eV/atom)	0.06
Electron energy (keV)	300
Beam current (pA)	25
Beam width (pm)	50
Field of view 1D (nm)	18.08
View Area (nm^2)	326.89
Pixel size (px/nm)	20.143
Total number of pixels	1.326×10^5
Pixel dwell time (μs/px)	20
Acquisition time (s)	2.65

[1] https://swanseanano.uk/

2) Complementary analysis of the local atomistic environment in the Au_{309} cluster using CNA and PTM methods

The validity of the results (see Fig. 2b) obtained in this study by the Common Neighbour Analysis (CNA) method [S1,S2] was checked by performing a similar analysis using the polyhedral template matching (PTM) method [S3] available in the OVITO software [S4]. The PTM method also allows the identification of lattice structures according to the topology of the local atomic environment and, in a general case, provides a higher reliability than CNA in the presence of strong thermal fluctuations and strains (see Ref. [S3] for details). In brief, this method identifies the local crystalline order by matching the local atomic environment to the templates of the structural types (in particular, fcc, hcp and bcc) determined by the PTM algorithm. For each atom, PTM maps the local environment to each possible structure. Then, a root-mean square deviation (RMSD), a measure of the spatial deviation from the ideal structure template, is calculated, and the structure type with the lowest RMSD value is assigned to the atom.

Table S2 shows the comparison of the fractions of atoms of the Au_{309} cluster belonging to fcc and hcp lattices, as determined by the interval CNA (i-CNA) [S2] and PTM methods. The results obtained for the final cluster structure obtained after deposition on the graphite substrate and relaxation for 1 ns are averaged over five independent MD simulations, with standard errors indicated. The very close values of the fcc and hcp fractions obtained by the two methods highlight the main result of this analysis, namely the reorganisation of the cluster core into a Dh-like structure after the deposition.

Table S2. Fractions of atoms of the Au_{309} cluster belonging to fcc and hcp lattices, as determined by the i-CNA [S2] and PTM [S3] methods using the OVITO software [S1,S4]. "Initial structure" refers to the structure of the free Au_{309}-Ih cluster before deposition. "Final structure" refers to the structure after the deposition on the graphite substrate and relaxation for 1 ns. Values obtained for the final structure are averaged over five independent MD simulations, with standard errors indicated.

	Initial structure			**Final structure**		
	fcc	hcp	unidentified	fcc	hcp	unidentified
i-CNA	0.06	0.27	0.67	0.13 ± 0.04	0.12 ± 0.04	0.75 ± 0.04
PTM	0.07	0.29	0.64	0.14 ± 0.04	0.14 ± 0.04	0.73 ± 0.05

3) Snapshots of the relaxed structure of the Au_{309} cluster after irradiation and its comparison with a cluster structure after deposition

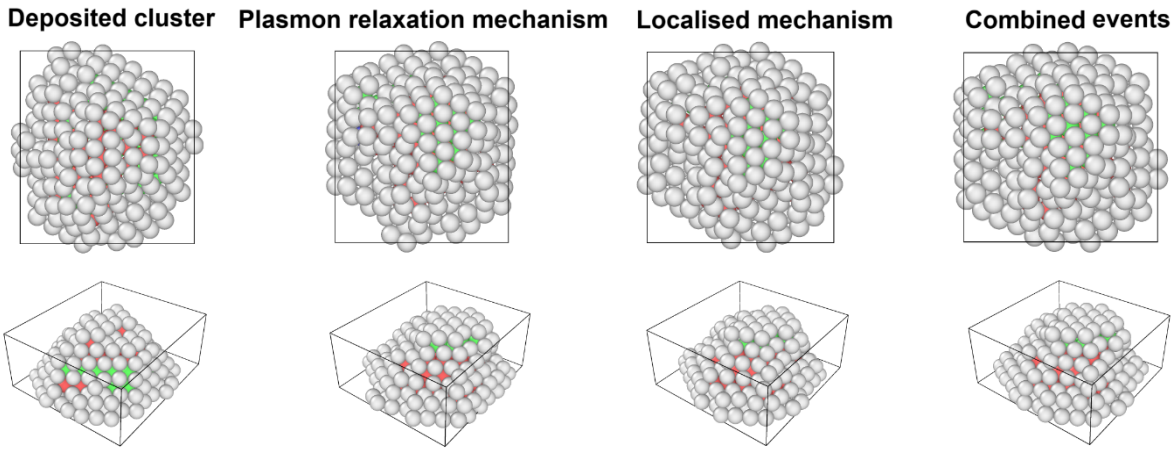

Figure S1. Snapshots of the Au_{309}-Ih cluster after deposition on a few-layer graphite substrate (1st column) and at the end of each post-irradiation energy relaxation simulation (2nd, 3rd and 4th columns). The top row shows the top view of the cluster, the bottom row shows the perspective view. The cluster is on the graphite substrate, which is not shown for clarity. Green and red spheres show atoms assigned to the fcc and hcp crystal lattices by the i-CNA method of the OVITO software [S1,S4]. The corresponding fractions of atoms in the fcc and hcp local atomistic environment are shown in Fig. 6 in the main text. The snapshots were rendered with OVITO.

References

[S1] A. Stukowski, Structure identification methods for atomistic simulations of crystalline materials, *Modell. Simul. Mater. Sci. Eng*. **20** (2012) 045021.

[S2] P. M. Larsen, Revisiting the common neighbour analysis and the centrosymmetry parameter, arXiv:2003.08879 [physics.comp-ph].

[S3] P. M. Larsen, S. Schmidt and J. Schiøtz, Robust structural identification via polyhedral template matching, *Modelling Simul. Mater. Sci. Eng*. **24** (2016) 055007.

[S4] A. Stukowski, Visualization and analysis of atomistic simulation data with OVITO–the Open Visualization Tool, *Modelling Simul. Mater. Sci. Eng*. **18** (2010) 015012.