Supplementary information to the paper: Transition from atomistic to macroscopic cratering

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ANIMATIONS

Animation 1 (au_101000_500eV.avi): Continuum-like illustration of the densification due to a Au_{101000} 500 eV/atom cluster impact event. The atoms are shown as small squares, which overlap completely in the bulk region, making the animation look like a continuum. The animation shows a 1000 Å wide and 1200 Å high cross section through the simulation cell. The cross section thickness is 4.08 Å (1 Au unit cell). The colors show the local density of each atom (estimated from the average distance to the nearest neighbors) relative to the bulk Au density. The red region which forms at 1-2 ps shows the high-pressure region which is consistent with the macroscopic cratering mechanism. At 2 - 5 ps a high-density pressure wave emanates from the crater. Between 10 and 50 ps long finger-like shapes form, and eventually break up due to the Rayleigh instability [1], leading to emission of drops of liquid matter similar to those formed when a stone is dropped on water.

Animation 2 (compression_animation.avi) shows a nanometer thick and 40 nm wide slice around the impact point of a 500 eV/atom Au_{315000} cluster. The color scale is the same as in Fig. 2 of the main article. The normal density regions are blue and compressed regions red. The animated event lasts 1.2 ps. After that the pressure will eventually release forming the displacement cascade. In this case the maximum density inside the compressed zone is more than two times the density of fcc Au. Notice that the compressed region is sinking while the cluster atoms are joining it, and the initial cavity is formed. This phenomenon is not seen in small cluster impacts. (Some sputtered clusters disappear from the scene, because they cross the borders of the visualization box.)

Animation 3 (3d_animation.avi) shows a 3d view of the crown induced by a 500 eV/atom Au_{750} cluster. The frame is 30 nm wide and the event lasts 50 ps. The colors show the distance from the viewer. Several phases are shown in the animation. Firstly, the cluster sinks into the substrate. Then fast recoil atoms are sputtered from the cavity. The rim of the cavity opens like a hatch and the cylindrically shaped crown is formed during the first 10 ps. After that, the fingers emerge and start to sputter clusters. In the last phase, the crown collapses slowly onto the surface. Notice that also some clusters land onto the surface. The clusters released in the later phases of the crown expansion have a rather slow velocity away from the surface. In 10 ns simulations, some of these clusters are seen to fragment and part of their material gain velocities towards the surface. This decreases the final sputtering yield [2, 3].

Animation 4 (surface_animation.avi) shows a 1 nm thick slice parallel with the surface seen from above, at the level of the initial surface. The frame is 40 nm wide and the event is a 500 eV/atom Au_{750} cluster impact on the Au(111) surface. The event lasts 50 ps. The AFM-like color scale shows the relative vertical positions of the atoms. In the first phase, the cluster sinks into the substrate and the cavity opens rising up the surface around the impact point. Already at this phase the crystal symmetry is seen in the rim. In the second phase between 10-30 ps, triangular symmetry is shown in the rim and three low surface areas are induced around the rim. This is an indication of stress in the lattice, although the crystal structure is not destroyed outside the crater region. In the last phase the stress is released, the rim is recrystallized and the final triangular shapes are formed inside the rim.

STOPPING OF SMALL CLUSTERS IN AU

The stopping of small atomic clusters in Au now rather well understood (Ref. [4] and references therein). At 500 eV/atom the cluster atoms loose their kinetic energy mostly in elastic collisions with the substrate atoms, which is called nuclear stopping. They loose some energy also in the interactions with the free electrons, but this does not have a significant effect. The energy distributed to the valence electrons by the electronic stopping is rapidly dissipated over the substrate [5], although in some cases it is claimed to induce minor effects in the substrate [6].

However, the stopping of a cluster can not be understood considering only binary collisions between cluster and substrate atoms. The interactions are complicated many-atom interactions [4]. The mesoscale effects, like craters and other surface modifications, can be analyzed with classical molecular dynamics, if the interaction model includes the possibility of many-atom interactions. In this study we have used the MD/MC-CEM poten-
...tial [7, 8], which takes account the many-atom nature of the interactions.

The collision process changes with the cluster size (number of atoms in the cluster). Fig. 3 shows an example of Au$_{103}$ impact on Au(111) at the same velocity as the large cluster impacts reported in the main article (500 eV/atom corresponding approximately to 22 km/s). The cluster pushes some substrate atoms away from the surface layers. A small cavity is formed. However, the cluster stops before a clear frontier of atoms is formed in the front of it. The cluster also transparently passes by some substrate atoms. As the impact energy increases, the cluster can travel considerably distances without small impact parameter collision with the cluster atoms. This channelling of an entire cluster is discussed in Ref. [4].

Fig. 3 shows an example of Au$_{103}$ impact at the same velocity. Now the cluster creates a cavity while penetrating into the substrate. Also a frontier of substrate atoms is formed in the front of the cluster. However, the cluster is not large enough to arrest the frontier atoms in a small volume, thus no compressed region is formed in this case. The analysis of compression, which is reported in Figs. 2 and 3 of the main article, show that the compressed region typical for macroscopic impact regime is formed at cluster sizes larger than 1000 atoms.

**CRATERS INDUCED BY LARGE CLUSTER IMPACTS**

Fig. 3 shows an example of a crater induced by a large cluster impact. In the case of Au, the crater rims re-crystallize at the end of cooling phase just after the main sputtering phase is over. However, there is still stress in the substrate because the substrate atoms form regions that are slightly displaced relative to each other, although the overall crystalline structure is preserved. Stress is present also because the crystalline orientation of the re-crystallized crater rim is different from the initial orientation in the substrate.

The volumes of the simulated craters are calculated assuming that they are ellipsoidal. In general, the radii of the simulated craters at 500 eV/atom are larger than their depths. When the impact energy/atom increases, the craters become deeper. The radii of the craters were measured on the level of the original Au(111) surface. The depths are the distances between the bottoms of the craters and the original surface level.

Love et al. [9] have measured depth-diameter ratio of craters formed in Al by nearly normal impacts of micrometeoroids and have found that the ratio is 0.56-0.60. In our simulations of Au impacts on Au(111) the ratio is 0.32-0.44, when the cluster has more than 750 atoms. In both cases, the impact velocity is approximately the same, so the difference of the crater shapes is due to different strength and melting properties of materials [10].

Perfectly spherical clusters were not used because one aim was to study the effect of cubic cluster shape on the symmetry of the crater. However, the simulations show that the orientation of the cubic clusters does not affect the crater shape, which is equally symmetric than the craters induced by spherical clusters.

**SCALING OF CRATER VOLUMES**

The crater growth in meteoroid impacts arrested either by the strength of the material, if the projectile is small enough, or by gravity, if the projectile is so large that gravitational forces dominate [11]. In this Letter we compare the results of atomistic simulations with the macroscopic scaling for the strength regime, because in the case of nanoparticles containing less than one million atoms, gravity is too weak to affect the results. In this regime, a crucial feature is the formation of a high pressure region called an isobaric core [12].

Scaling of planetary impact craters is discussed in K. A. Holsapple’s article. [13] In our simulations, both the impactor and the substrate have the same density, which simplifies the scaling laws considerably. Crater volumes scale with impactor velocity $U$ and mass $m$ according to the following scaling law

$$ V = K_1 \frac{m}{\rho} \left( \frac{\rho U^2}{\bar{Y}} \right)^{3\mu/2}, \quad (1) $$

where $\rho$ is the density of material and $\bar{Y}$ is a parameter that depends on the strength of the material. $K_1$ is a parameter. This scaling law is valid in the strength regime, which means that the impactor size is so small that the effects of gravity can be neglected. For strength regime impacts on hard rock, the impactor should have a diameter smaller than 32 meters. Eq. 1 is derived assuming that the energy is compressed into a point after the impactor has stopped in the substrate. This point source approximation is commonly used in the theory of macroscopic impacts. The simulations support this approximation, because the initial compressed region is small compared to the size of the crater.

Eq. 1 implies that $V$ depends linearly on $m$, if $U$ is constant. The empirical values of parameters for hard rock are $K_1 = 0.2$, $\bar{Y} = 18$ GPa, and $\mu = 0.55$. [13] Naturally, empirical parameters for Au are not available, because no macroscopic impact experiments are carried out for noble metal targets. (Parameters are available for rock, sand, water and soil.) If we use the density of Au $\rho = 19300$ kg/m$^3$ and Young’s modulus $Y = 78$ GPa instead of the modified strength parameter $\bar{Y}$ of hard rock, Eq. 1 gives

$$ V = 0.17 N \text{nm}^3, \quad (2)$$
where \( N \) is the number of atoms in the impacting cluster. For 22 km/s \( \text{Au}_{68,000} \) and \( \text{Au}_{161,000} \) clusters, the coefficients from the simulations are 0.24 and 0.22, respectively. From this result, we can not definitely conclude that the simulations and the empirical scaling law agree quantitatively because empirical scaling parameters are not available for \( \text{Au} \). However, we can say that with a reasonable choice of parameters, one gets an estimate of crater volume which is of about the same magnitude as the simulated volume. This result is not evident, because the macroscopic scaling is derived for impactors that are more than fifteen orders of magnitude larger in number of atoms than the clusters in this study, thus the empirical law is extrapolated downwards by more than fifteen orders of magnitude.

If the empirical strength parameter \( \bar{Y} = 18 \text{ GPa} \) for hard rock is used with the density of \( \text{Au} \), then \( V = 0.6 \text{N mm}^3 \). In other words, the craters would be approximately three times larger, if the material would be such that it has the same density as \( \text{Au} \) but the yield strength of hard rock. Thus, the macroscopic scaling is rather sensitive to the choice of material.

**CHANNELING**

Fast \( \text{Au} \) atoms channel easily in a perfect \( \text{Au} \) crystal \cite{4}. This occurs also in large cluster impacts, if the impact velocity is large enough (Fig. 4). Channelling means that an atom moves straight along a channel in the crystal structure without head-on collisions with substrate atoms.

In the early phases of the expansion of the displacement cascade, the fastest cluster or knock-on atoms move ahead of the main frontier of atoms along the channels due to channelling. They form subcascades or even satellite cascades, which are clearly visible in Fig. 4. Eventually the displacement cascade expands over the subcascades and the final form of the cavity is almost perfectly hemispherical. However, in the case of very small \((N < 20)\) and fast \((>100 \text{keV/atom})\) clusters the shape of the displacement cascade can be very irregular because of very large subcascades. This can be considered as a small size effect.

**FINGERS OF LIQUID MATERIAL**

Emergence of fingers is usually studied in the context of liquid drop impacts on solid or liquid surface \cite{14,15}. Fig. 5 shows how the fingers develop, when the corona grows. The first collisions between the cluster and substrate atoms occur at various angles because the cluster structure does not match symmetrically to the target lattice and because the atoms are initially not at their exact lattice positions due to the thermal motion. Because of this lack of symmetry and the presence of randomness in the simulation model, the atomic motion in the collision cascade and corona is not symmetric or uniform in all directions. Due to the attraction between the atoms, the motion of atoms moving near each other starts to correlate, and regions of faster and lower velocities appear. Eventually, the fingers appear and become more and more separated due to the surface tension. The reason to the cluster ejection out of the fingertips \cite{2} is minimization of surface tension, i.e. the Rayleigh instability \cite{1}.

**COHERENT DISPLACEMENTS**

Fig. 6 shows the crater during the main sputtering phase. The surface of the cavity is melted and the crystal structure is distorted near the cavity. Dislocation patterns are visible deeper in the structure. These patterns resemble dislocation loops observed on the surface during nanoindentation \cite{16,17}.

The strong pressure wave emanating from the cascade causes entire planes of atoms to slide coherently along the \{111\} planes of fcc metals \cite{18}. These coherent displacements are visible in the substrate near the crater (Fig. 7), on the surface outside the crater rim (Fig. 8) and inside the rim (Fig. 9). They also induce transient pile-up patterns around the crater. The permanent displacement pattern appears when the stress is released during the crater cooling phase. Coherent displacements are observed in large-scale hypervelocity simulations of \( \alpha \)-alumina substrates \cite{19,20}.

Because of these displacement and pile-up effects, successful cluster impacts on the \( \text{Au} \) surface not only change the surface topography but also modify the structure of the material inside the substrate. The simulated substrate was a perfect fcc lattice, but real substrates have defects which may initiate crack propagation. Impact induced cracks are shown for instance in micrometeoroid craters of the Hubble Space Telescope \cite{21}. Craters induced by asymmetric stress are observed on planetary surfaces \cite{22}.

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FIG. 1: An example of early phases of 500 eV/atom Au$_{13}$ cluster impact on Au(111). The frames show the process at 50 fs intervals.
FIG. 2: An example of early phases of 500 eV/atom Au$_{55}$ cluster impact on Au(111). The frames show the process at 50 fs intervals.
FIG. 3: An example of crater induced by a 500 eV/atom Au$_{161,000}$ cluster. The figure shows 100×100 nm wide and 2 Å thick slice at 210 ps. The crater rims are re-crystallized. The colours show the distance of the atoms from the viewer. Coloured areas indicate stress fields in the substrate.

FIG. 4: An example of high-energy impact induced by a 25 keV/atom Au$_{750}$ cluster. The figure shows 20×20 nm wide and 1 nm thick slice at 500 fs. Subcascades are clearly visible.
FIG. 5: Emergence of fingers in corona induced by a 107 keV/atom Au$_{13}$ impact. The corona is visualized as a 180° and 20 nm high panorama view seen from the centre of the cavity. Atoms in the red regions are moving faster upwards than atoms in the blue regions. Correlation of the motion of neighbour atoms begins already before 4 ps, which is seen as blue and red areas in the panorama. At 10 ps, the motion in the corona has become clearly inhomogeneous, and at 30 ps, only the tips of the fingers are moving upwards.
FIG. 6: An example of transient coherent dislocations induced by a 25 keV/atom Au$^{750}$ cluster. The figure shows $20 \times 20$ nm wide and 1 nm thick slice at 15 ps.

FIG. 7: An example of final coherent dislocations induced by a 50 keV/atom Au$_{55}$ cluster. The figure shows $20 \times 20$ nm wide and 1 nm thick slice at 250 ps.
FIG. 8: An example of crater rim induced by a 500 eV/atom Au$_{6,000}$ cluster. The figure shows 50×50 nm wide slice at 200 ps.

FIG. 9: The same event as in Fig. ?? but only a 1 nm slice of the surface is visualized.