Mechanisms of bubble growth and blistering on metals exposed to hydrogen

Alvaro Lopez Cazalilla, Flyura Djurabekova

Helsinki Institute of Physics and Department of Physics, University of Helsinki, Finland

Catarina Da Palma Serafim, Ana Teresa Perez Fontenla, Sergio Calatroni, Walter Wuensch

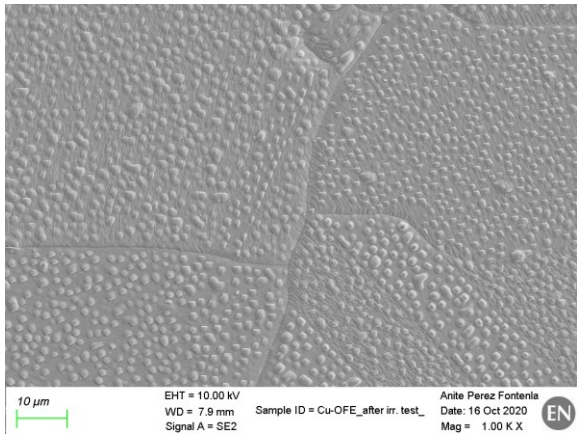
CERN Switzerland

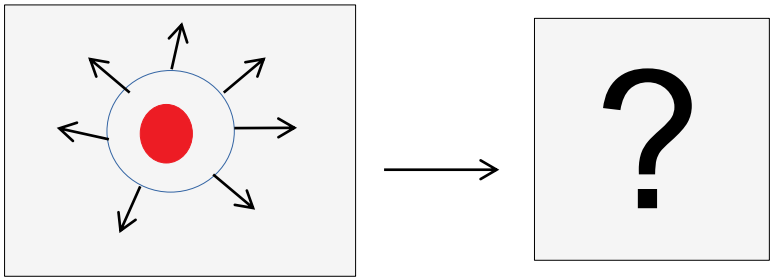
Outline

- Introduction & Motivation:
  - Dislocations, hydrogen and surface behavior.
- Effect on surfaces (MD). Influence of grain orientation at low fluences
- Cu-H machine-learned interatomic potential
- Conclusions

Motivation

- Cu is the first candidate for particle accelerators in the field of high energy physics
  - High thermal and electric conductivity as well as ductility properties make Cu superior to many other metals
  - It is widely used in accelerating structures such as CLIC and RFQ structures for proton linac injectors
- H tends to penetrate Cu and accumulates in some voids close to the surface, when relatively low energy H^- (~45 keV) is used in accelerator

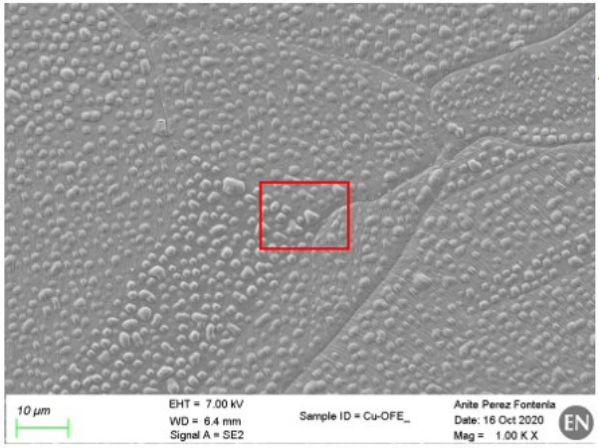


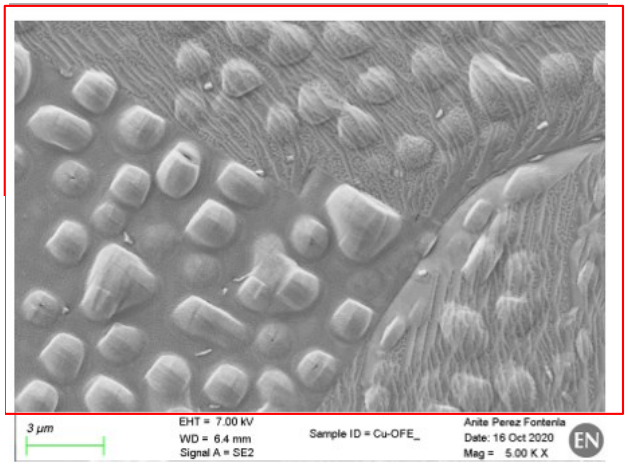


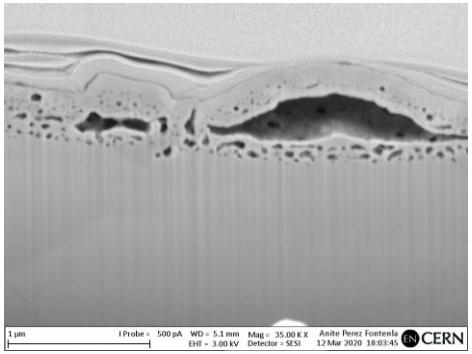
H gas bubble

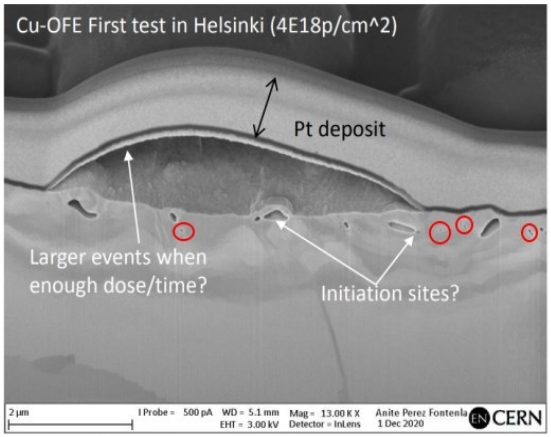
Blister at Cu surfaces

- Experimentally bubbles appear close to the surface seen in SEM images as protrusions or H blisters
- Also, protrusions that appear in the Cu surface are of different shapes



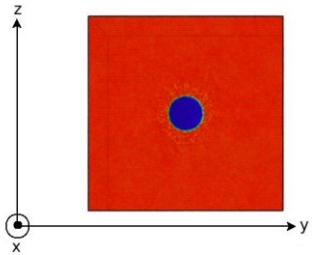


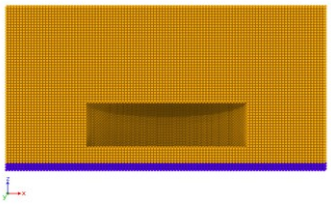


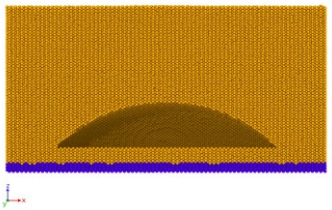


Simulation details

- We use molecular dynamics (MD) to study this effect. Mishin Embedded Atom Method (EAM) potential for Cu, EAM for H-H and purely repulsive H-Cu.
- MD is limited in the time and size of the cell, however, it provides a good description of the atomic interactions using interatomic potentials
- Problem of H accumulation and complete creation of defects is beyond the MD time span.
  - Solution for H accumulation: introducing directly high concentrations (high pressures) of H ($n_{H/Vac}$: number of H per vacancy in the void), we can speed up the process.



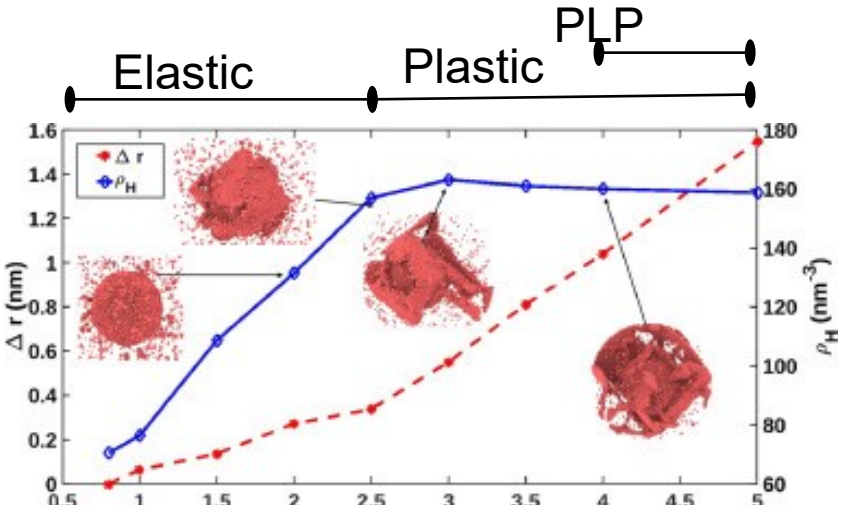




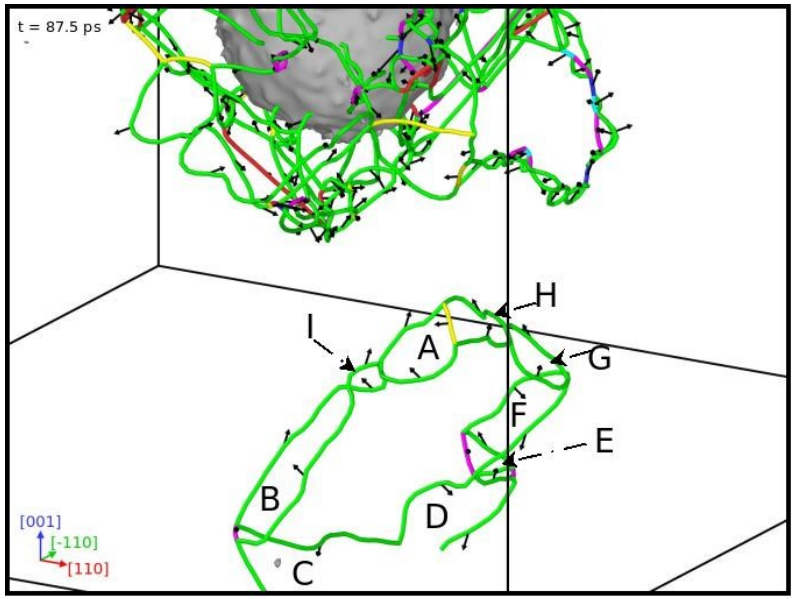
To enable large void simulations, we use different void shapes (sphere, hemisphere, disk) and surface orientations ({100}, {111}, {110}).

Bubble growth in Cu

- To model this effect, some computational approaches have been implemented before with different outcomes.
- The growth of tiny He bubble was followed in Cu, showing the dissociation of small prismatic loops [Jin et al., Sci. Rep. 11 (2021) 12839]
- We showed that even much larger bubbles grow via prismatic loop punching (PLP). In this case the network of shear loops is forming on adjacent {111} planes with Burgers vector either aligned or opposite to the gliding direction of the prismatic loop [A. Lopez, F. Djurabekova et al. Acta Materialia 225 (2022) 117554]

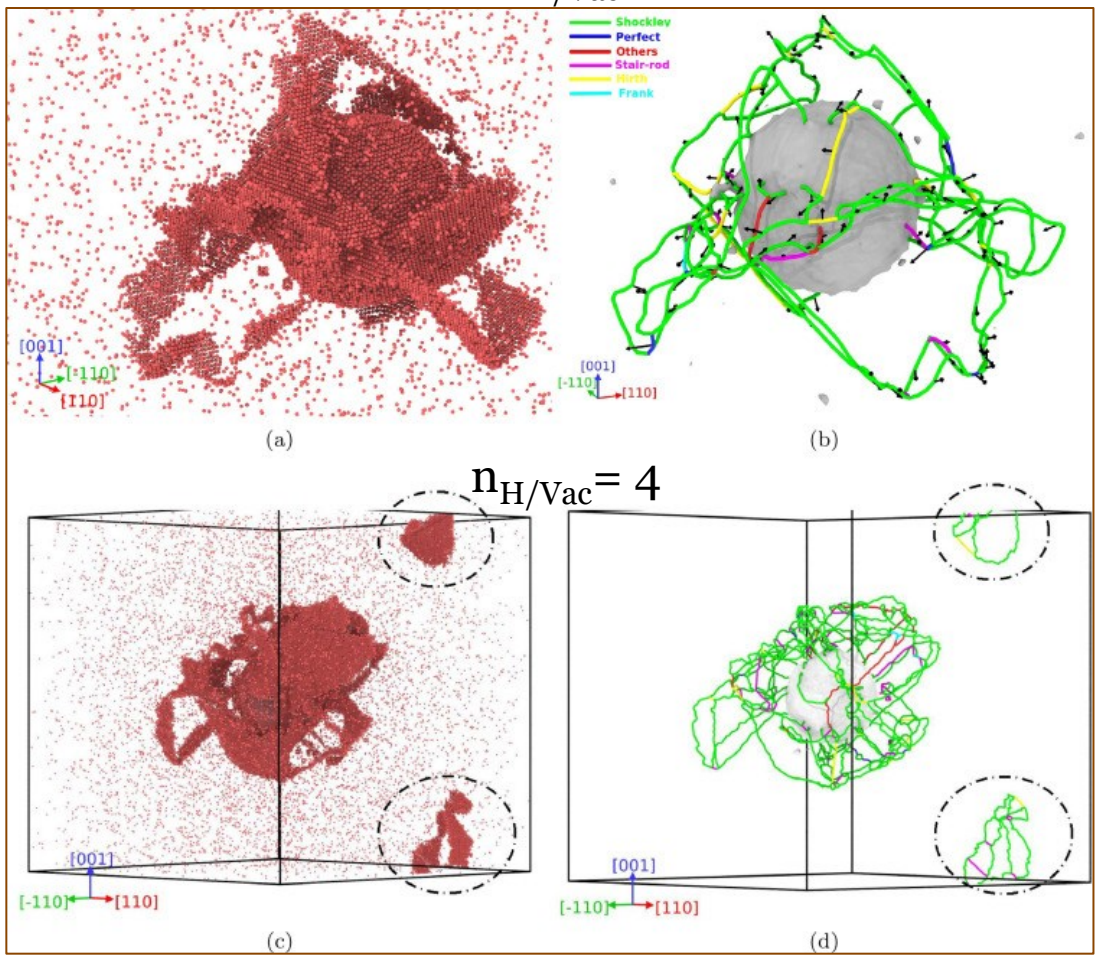


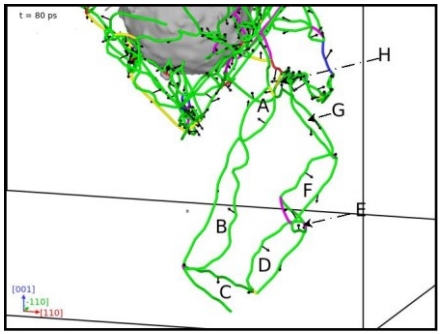
($n_{H/Vac}$: number of H per vacancy in the void)

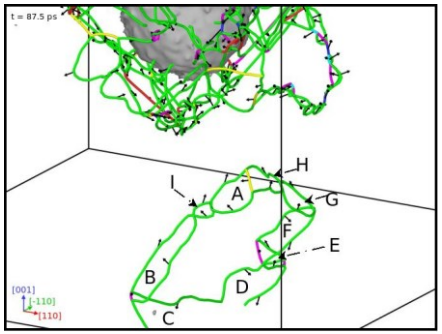


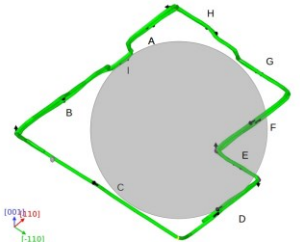
Prismatic loop of arbitrary face in FCC lattice

$n_{H/Vac} = 3$





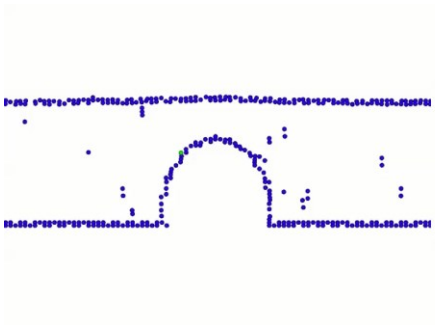




[A. Lopez, F.Djurabekova et al. Acta Materialia 225 (2022) 117554]

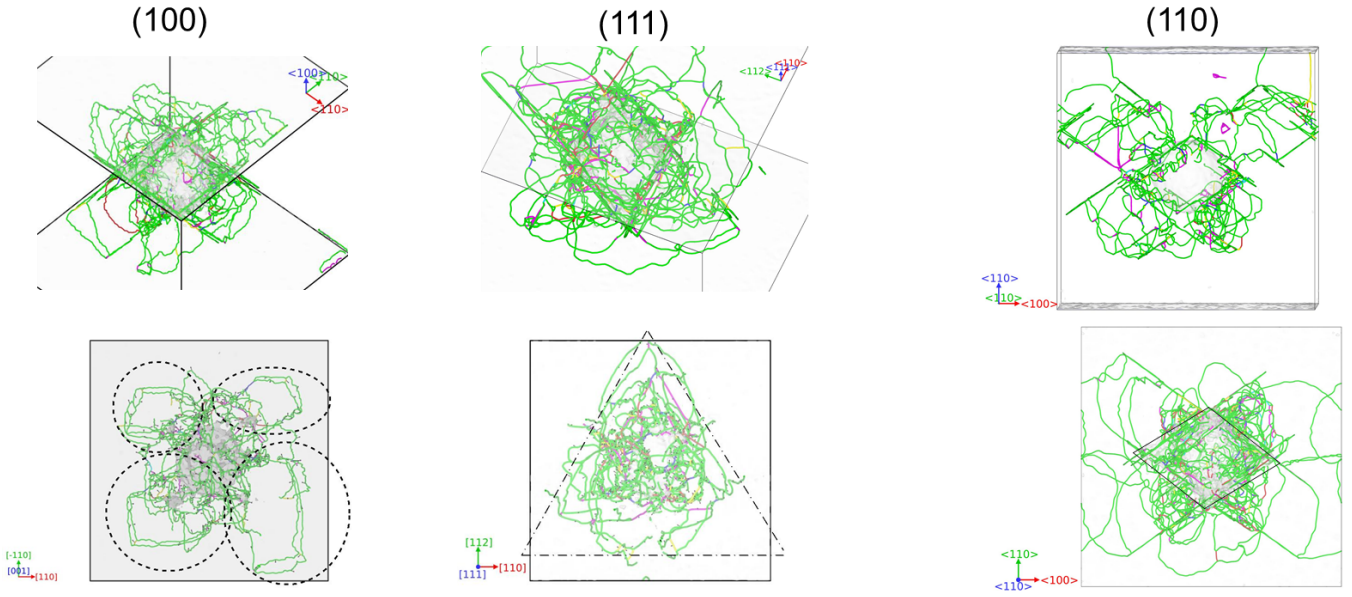
H bubble under the surface

- Now we place the void near surface
- Loop punching proceeds via very similar mechanism, but the dislocation loops are attracted by surface



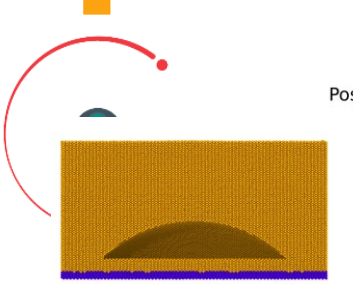
Surface protrusions with different crystallographic orietnations

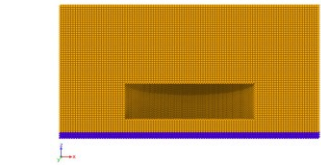
- In different directions we can see the formation of dislocation loops of a rhomboid shape in the <110> orientations.
- The dislocations travel in the slip planes, however the (100) surface attracts more dislocations than the (111), but less than the (110).
- The slip planes in FCC materials are the same, nevertheless, the interaction with the surface (or grain boundary) creates different features on surface



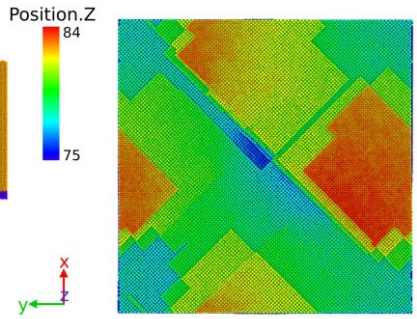
Green: Shockley partials. Blue: perfect partials. Red: "others" dislocations. Magenta: stair-rod. Yellow: Hirth. Cyan: Frank partial dislocations

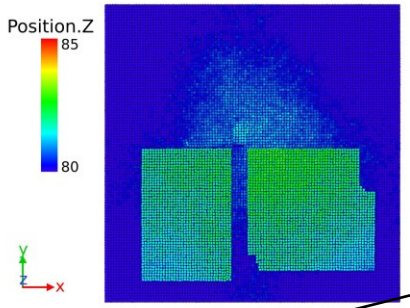
Hemispherical and disk bubbles ($n_{H/Vac}$ = 2)



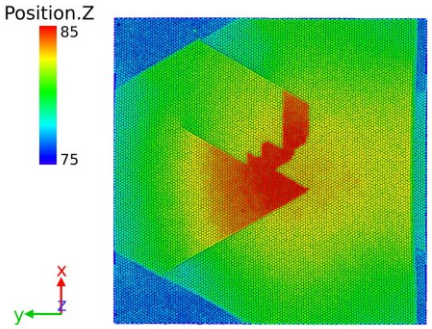


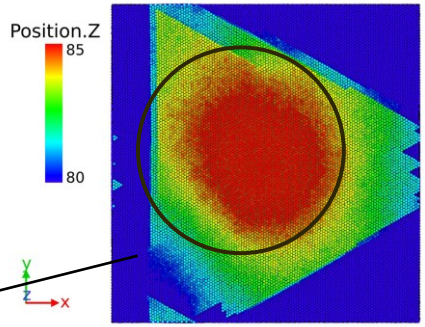
(100)



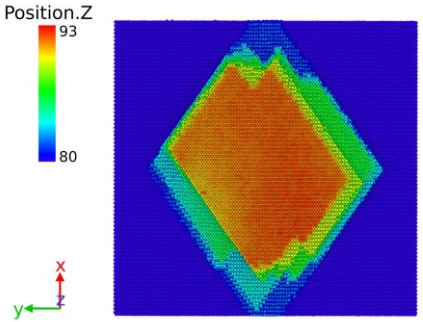


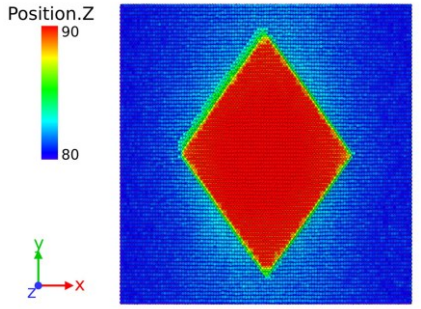
(111)



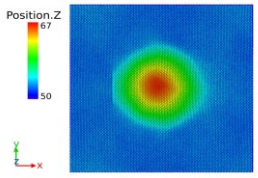


(110)



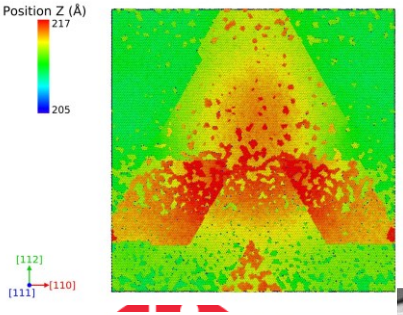


Circular shape at low H conc.

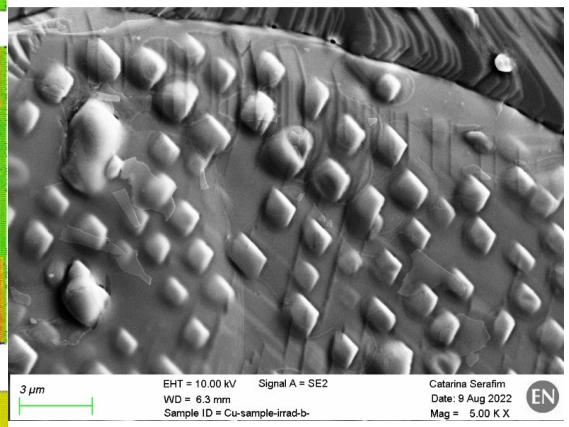


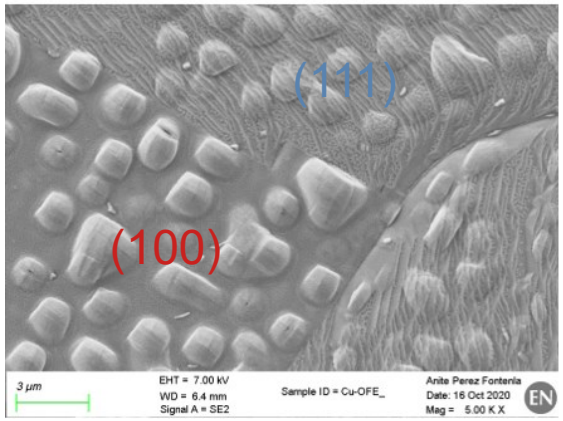
- Similar surface features as in the spherical void.
- However the disk-bubble generates clearer protrusions.

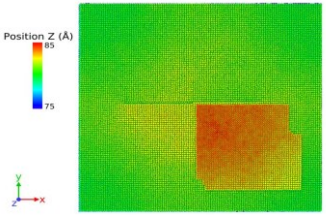
Comparison with experiments

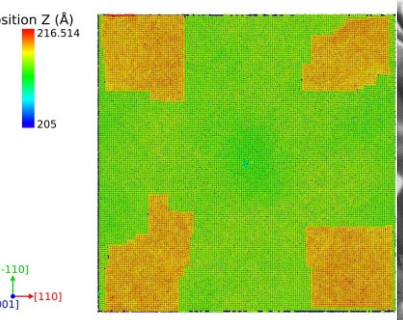


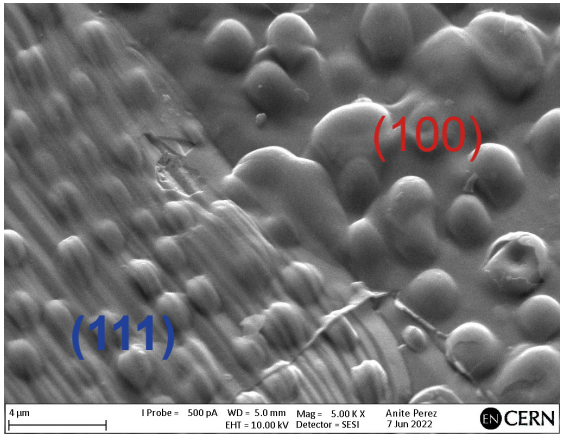
(110)

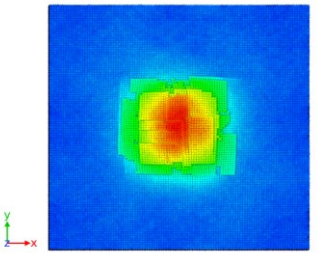


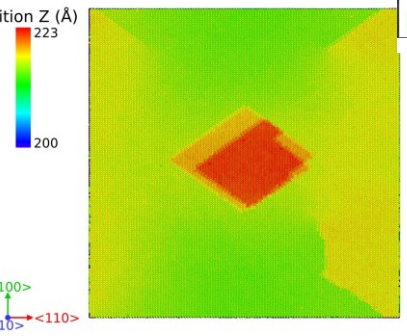


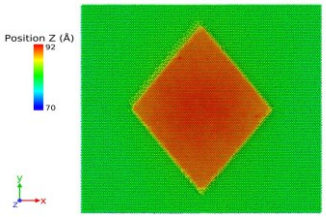


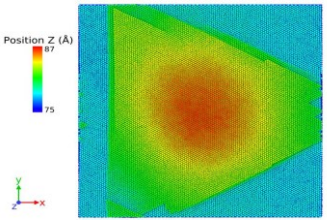


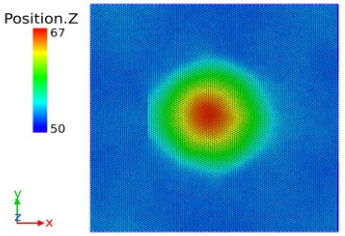






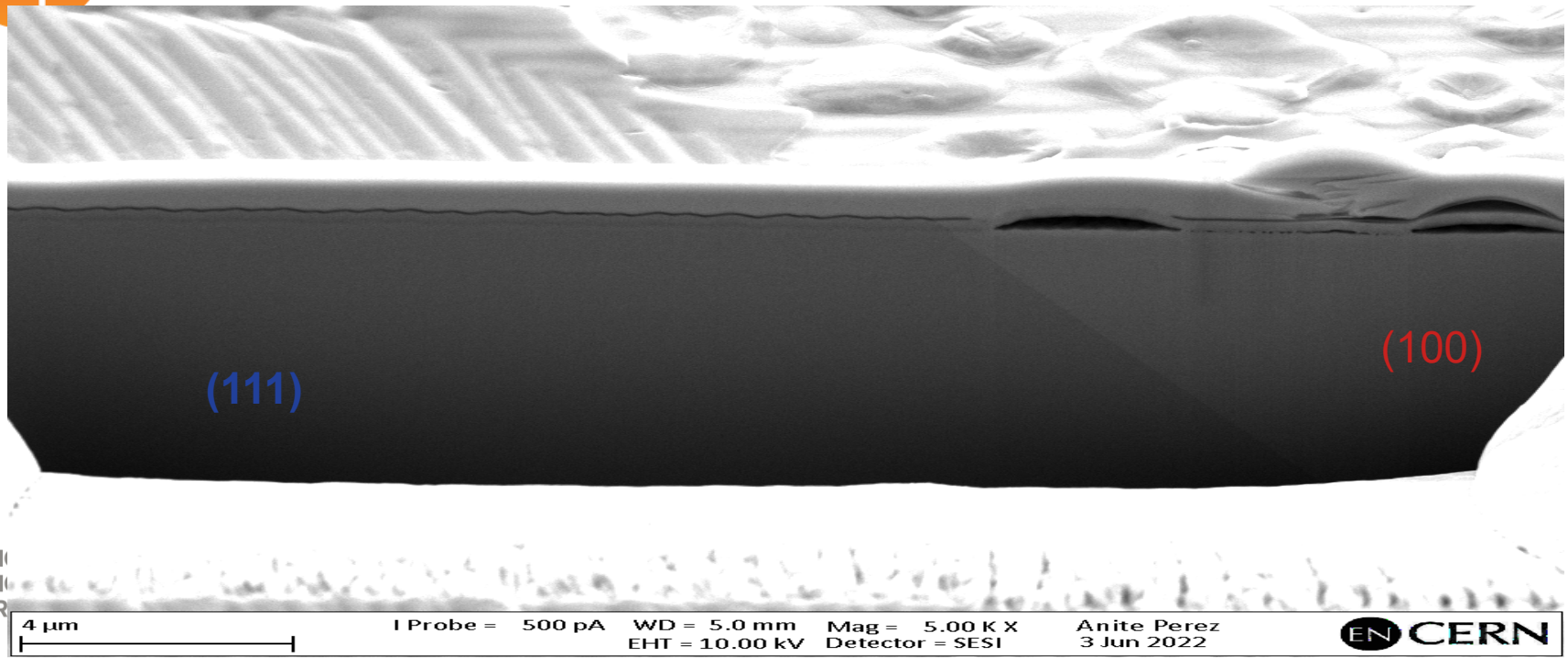




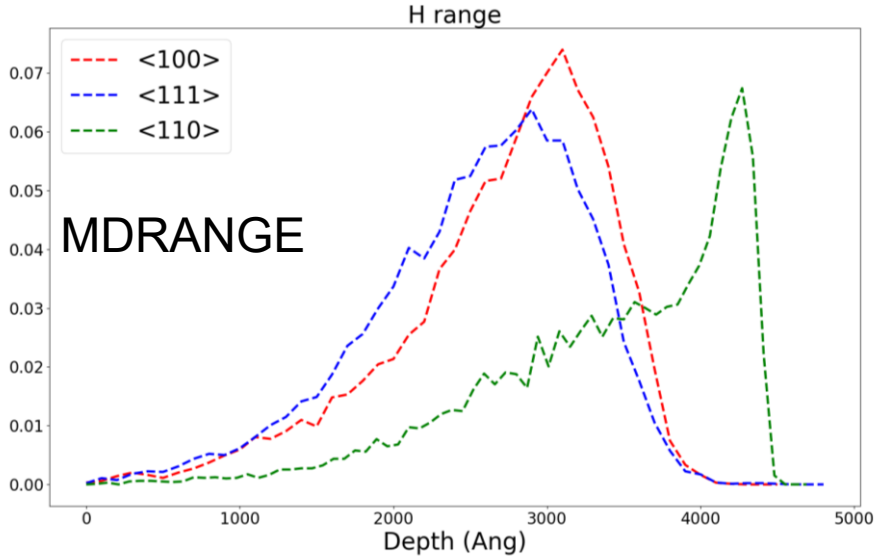


MDRANGE vs BCA

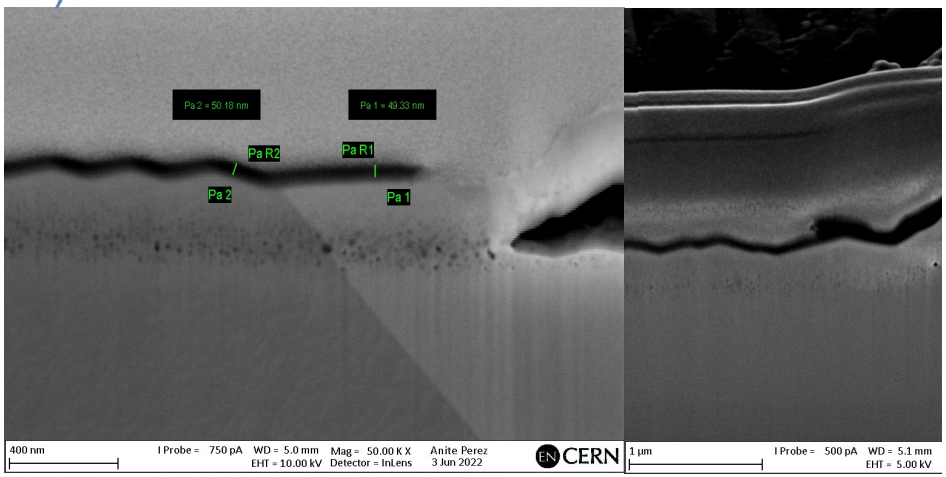
- The MD results do not provide information on why (110)/(111) are not the first surface yielding, being the glide direction/plane. We see (110) is the first yielding, contradicting the experiments.
- Since the initiation sites are formed at different depths depending on the grain orientation, we need to know how the penetration depths are for 45 keV H in Cu. For that we use MDRANGE, when typically Monte Carlo methods are commonly applied to this problem.
- MDRANGE* is an MD approach specifically designed for high-energy interactions and allows for consideration of different crystallographic orientations.

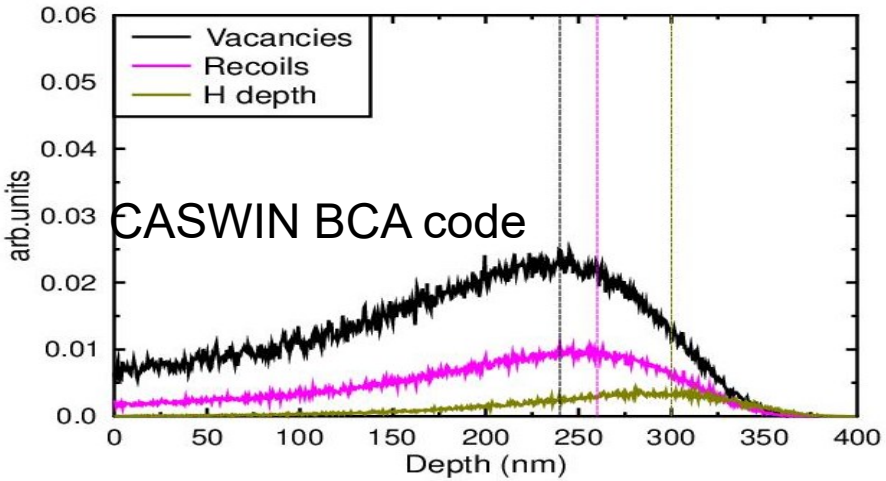


MDRANGE vs BCA



(111) (100) (110)





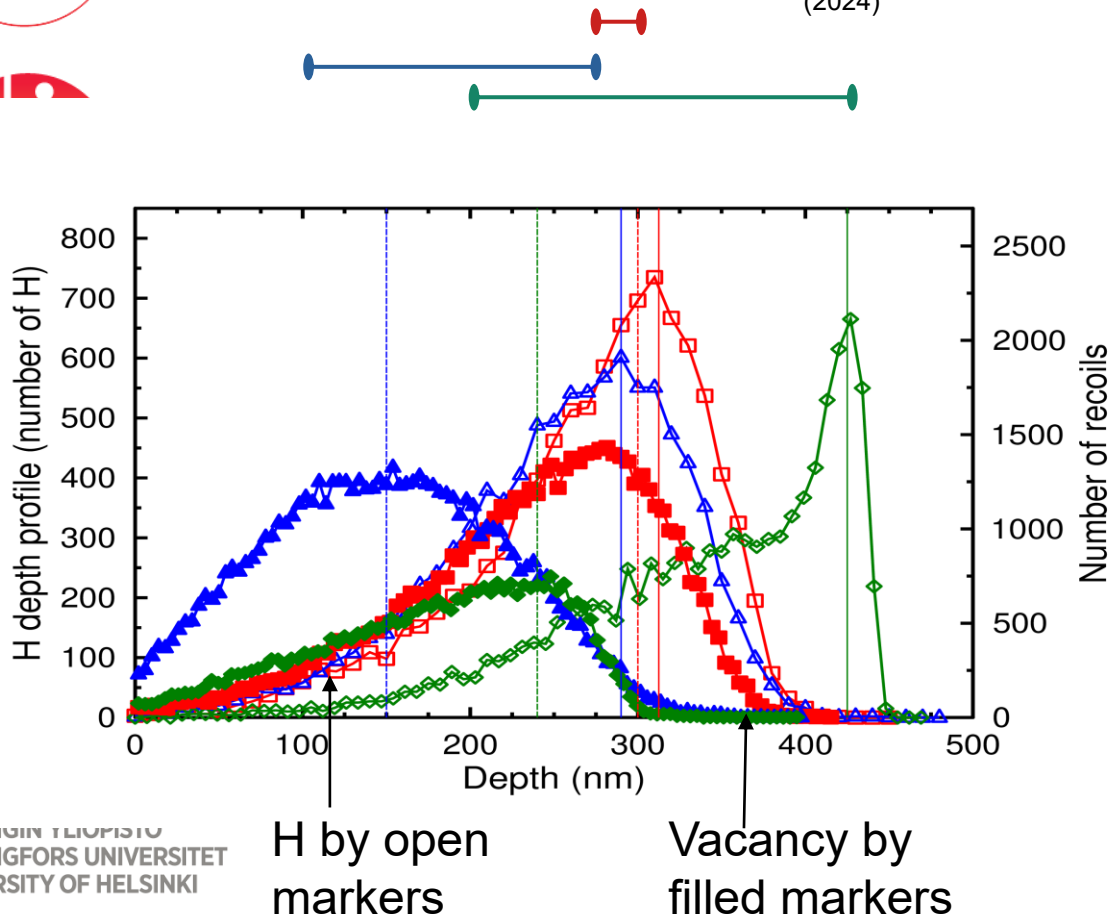
- The studied grains under low fluence show that bubbles are formed at different distances, which is consistent with MDRANGE results, observing a deeper formation in (110) grains.
- After a complete dataset of blisters/protrusions is obtained with MD, we understand the mechanisms of surface modification. However, we observe that under low fluences, only (100) grains experience blistering.

Recoils formation ~ vacancy formation

Comparison with experiments

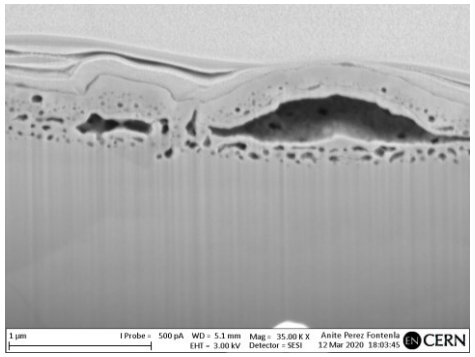
- We filter those recoils with kinetic energy lower than 40 eV (approximately the energy needed to create a vacancy), and we observe that for <100> the peaks are close, however the distances between them in <111> and <110> (specially the latter)

A. Lopez-Cazalilla, C. Serafim, F. Djurabekova, Acta Materialia, 266, 119699 (2024)



- The formation of vacancies occurs closer to the surface in <111> and <110>grains, but the hydrogen atoms need more time (fluence) to fill these gaps and develop larger blisters visible in the surface, while in <100> is immediate

Cu-H Machine-learned interatomic potential



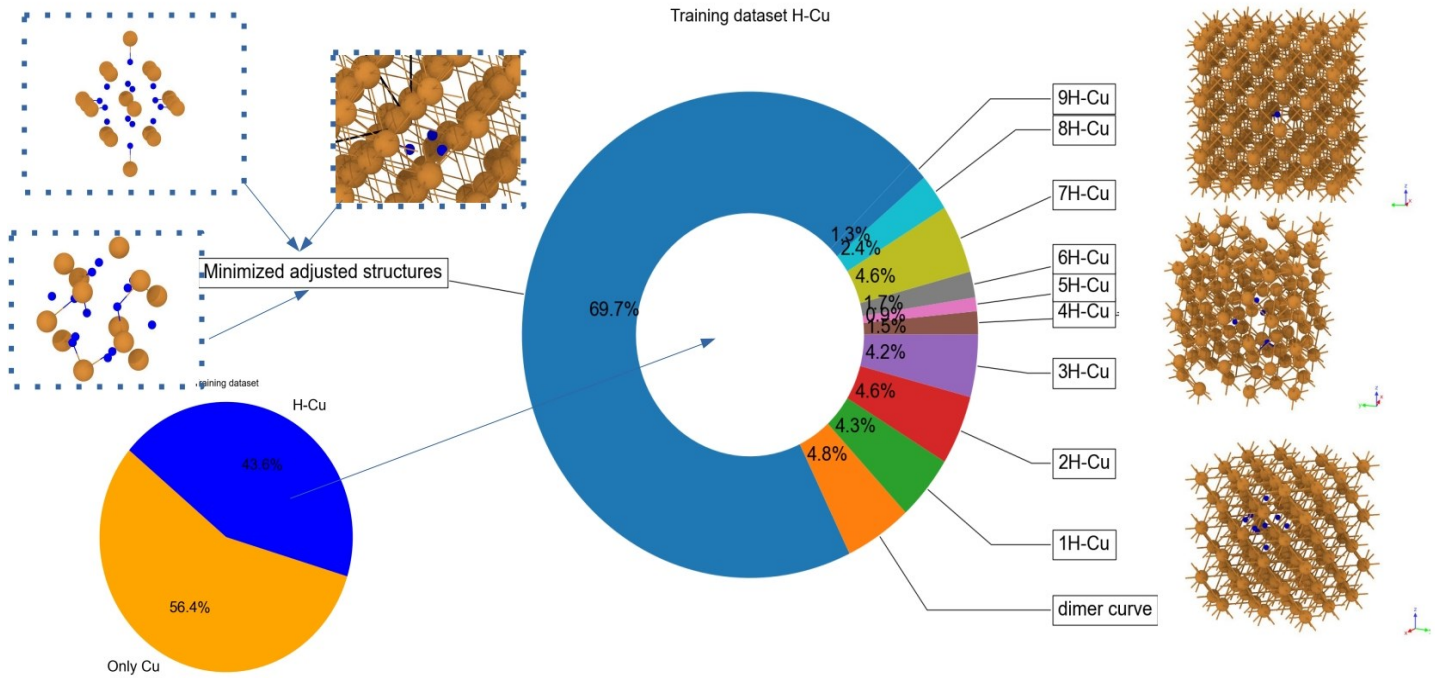
- The process of bubble coalescence and the interaction of H with dislocations formed during the growth of bubbles (intermediate stage) remain inaccessible by our current model. The application of purely repulsive interaction between H and Cu is limited.

- The development of a potential that can reproduce the processes that occur in the first stages of the bubble growth is needed.

- Only one interatomic potential (besides purely repulsive) is currently available for H-Cu interaction. However, the interaction between H and Cu is highly attractive, showing unrealistic results for bubble growth(3).

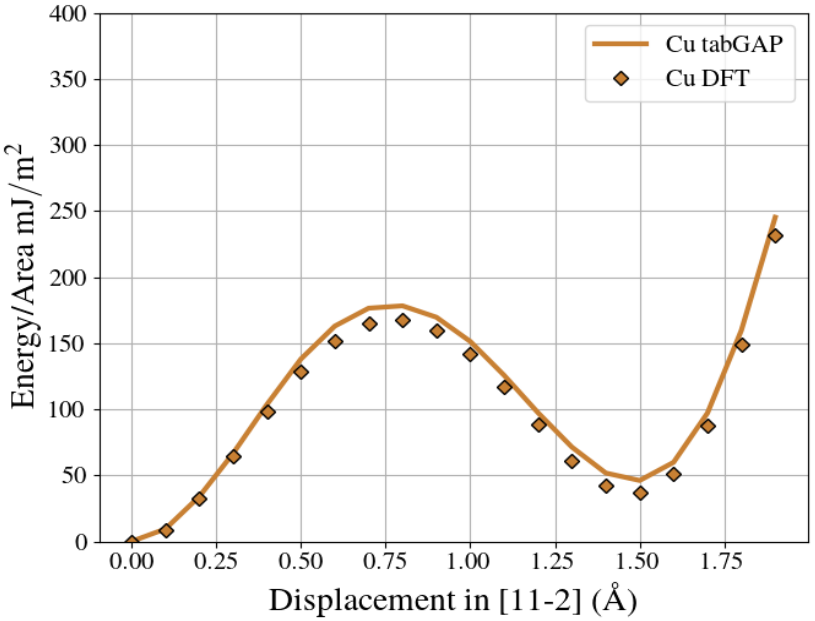
- We choose the machine learning approach to design a new potential with the inputs from density functional theory calculations

Cu-H MLIP. Training dataset

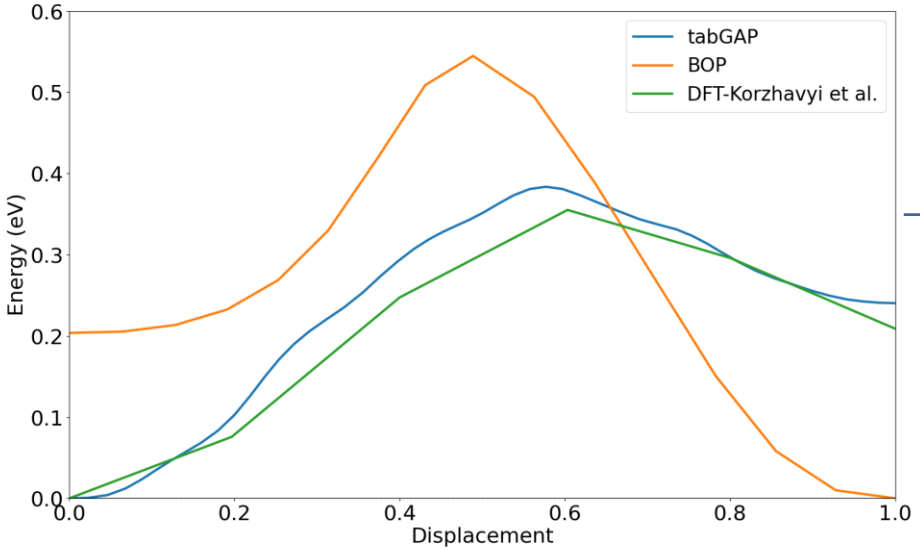


Cu-H MLIP. Performance tests

Courtesy of Aslak Fellman

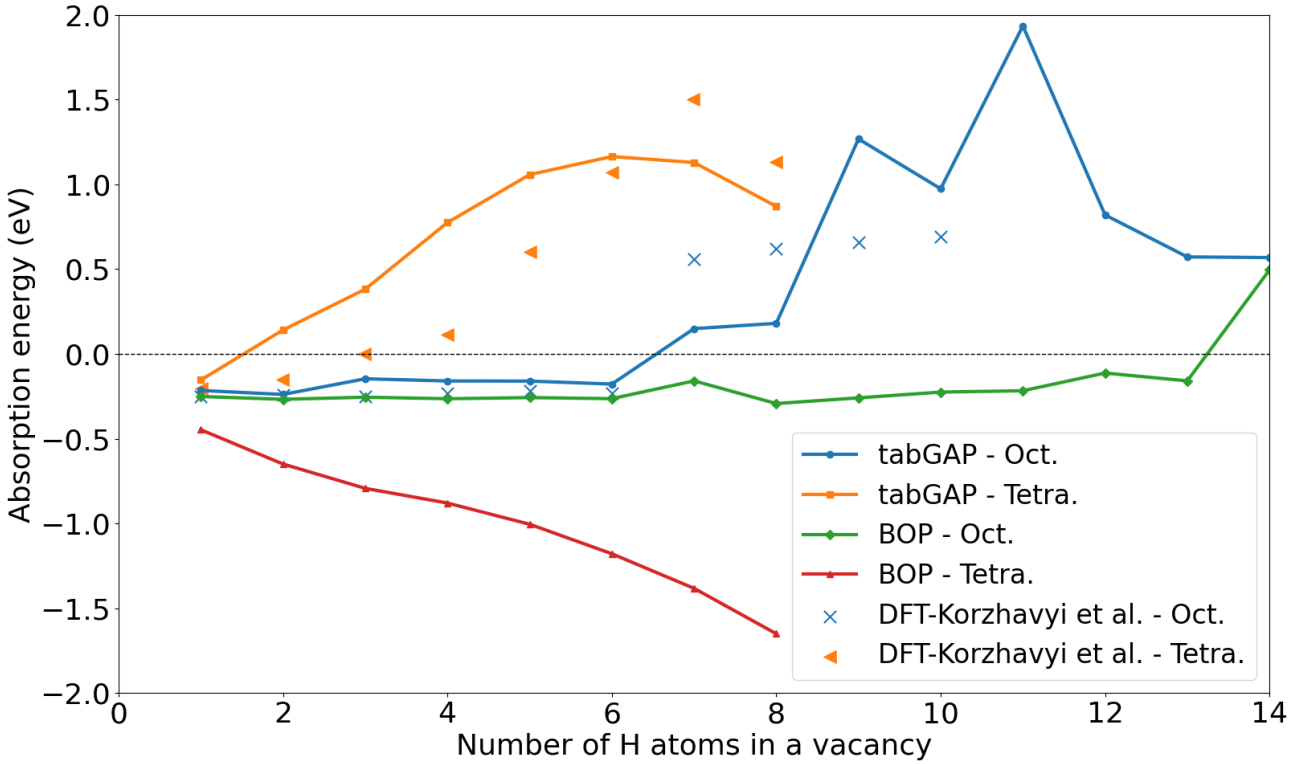


The insertion of the H dataset did not modify the mechanical properties of the copper part



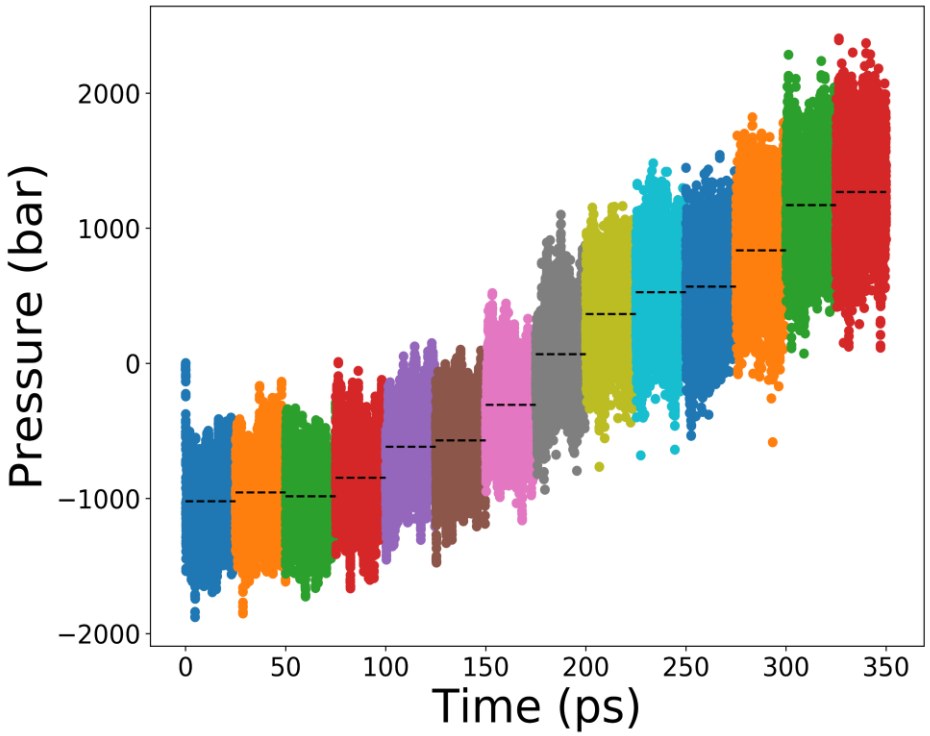
The barrier of a H atom moving from an octahedral to a tetrahedral position is predicted close to DFT precision. And better than the other available potential (BOP)

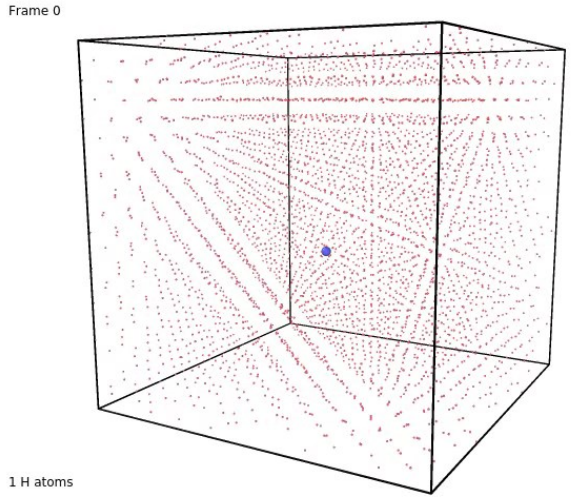
Cu-H MLIP. Performance tests



- Comparing with BOP, tabGAP predicts an absorption energy closer to DFT, with similar trend.
- DFT predicts that no more than 6 H can be accommodated in 1 vacancy

Cu-H MLIP. Sequential insertion of H





- This potential allows to follow processes such as the accumulation of H in small defects such as small vacancy clusters, and assists on what experimentally is not possible to follow.

Conclusions

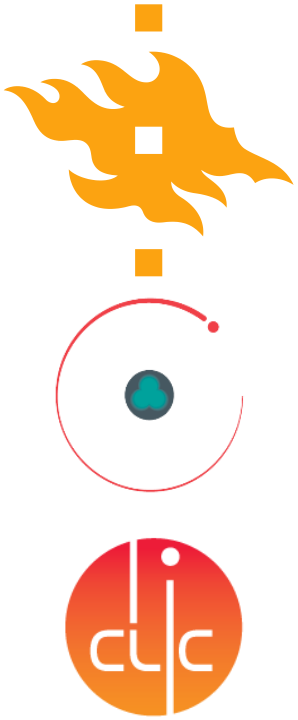
- In our simulations we clearly see the atomistic mechanisms of bubble and blister growth under hydrostatic internal H pressure. We identify the effect of surface orientation on the shape of appearing protrusion.
- The observed differences in the protrusion shapes with MD are consistent with those seen in experiment
- The difference between the formation of vacancies area and the penetration depth of H in (110) and (111) grains explain the later development of blisters, complementing the penetration depth as previous models predicted.
- The ML potential, with DFT precision, enables the exploration of mechanisms behind the blistering effect.

References

(1):

W. Pesch, T. Schober, and H. Wenzl Metall. Trans. A, vol. 11, p. 1821, 1980.
T. Schober, H. Conrads, and A. Schulz Fusion Technol., vol. 23, p. 227, 1993.
C. Ju and J. Rigsbee Mater. Sci. Eng., vol. 74, p. 47, 1985.
M. Iino Metall. Trans. A, vol. 9, p. 1581, 1978.
K. Ravi, V. Ramaswamy, and T. Namboodhiri Mater. Sci. Eng., vol. A129, p. 87, 1990.
J.-L. Lee and J.-Y. Lee J. Mater. Sci., vol. 22, p. 3939, 1987.
J.-Y. Lee and J.-L. Lee Phys. Chem. NF, vol. 146, p. 233, 1985.
J.-L. Lee and J.-Y. Lee Metall. Trans. A, vol. 20, p. 1793, 1989.
K. Kamada, A. Sagara, and H. Kinoshita, Difficulty of dislocation-punching from densely distributed high pressure bubbles, Radiation Effects, vol. 106:3, pp. 219-227, 1988.
K. Kamada,Hydrogen implantation effects in the subsurface layer of aluminum - bubble pressure and surface modifications, Journal of Nuclear Materials, vol. 169, pp. 141-150, 1989.

(2)Internal communication with CERN for Titanium not-blistering
(3) Zhou, X.W., Ward, D.K., Foster, M. et al. An analytical bond-order potential for the copper–hydrogen binary system. J Mater Sci 50, 2859–2875 (2015)
(4) J. Byggmästar, K. Nordlund, and F. Djurabekova, Simple machine-learned interatomic potentials for complex alloys, Phys. Rev. Materials 6, 083801 (2022)



Thank you for you attention!