Bridging experiment and simulation: X-ray free-electron laser probing of optically induced melting of Palladium

WE_O4

J. Antonowicz[1]*, A. Olczak[1], K. Sokolowski-Tinten[2], P. Zalden[3] and R. Sobierajski[4]

[1]*Faculty of Physics, Warsaw University of Technology, 00-662 Warsaw, Poland*
[2]*Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany*
[3]*European XFEL, 22869 Schenefeld, Germany*
[4]*Institute of Physics Polish Academy of Sciences, 02-668 Warsaw, Poland*

*e-mail: jerzy.antonowicz@pw.edu.pl

Due to its extremely short timescale, the non-equilibrium melting of metals is exceptionally difficult to probe experimentally. As a result, our understanding of melting mechanisms relies primarily on theoretical predictions. This work investigates the ultrafast melting of thin polycrystalline Pd films using optical laser pump – X-ray free-electron laser probe experiments combined with molecular dynamics simulations. By acquiring X-ray diffraction snapshots with sub-picosecond resolution, we capture the atomic structure of the sample during its transition from the crystalline to the liquid state.

Bridging the timescales of experiments and simulations enables us to formulate a realistic and consistent microscopic picture of the crystal-liquid transition[1]. We demonstrate that the melting process accelerates progressively with increasing absorbed energy density. This acceleration results from a gradual shift in the solid-to-liquid transition mechanism, evolving from heterogeneous melting—initiated within structurally disordered grain boundaries—to homogeneous melting, which occurs catastrophically within the crystal volume on a picosecond timescale, close to that of electron-phonon coupling in Pd.

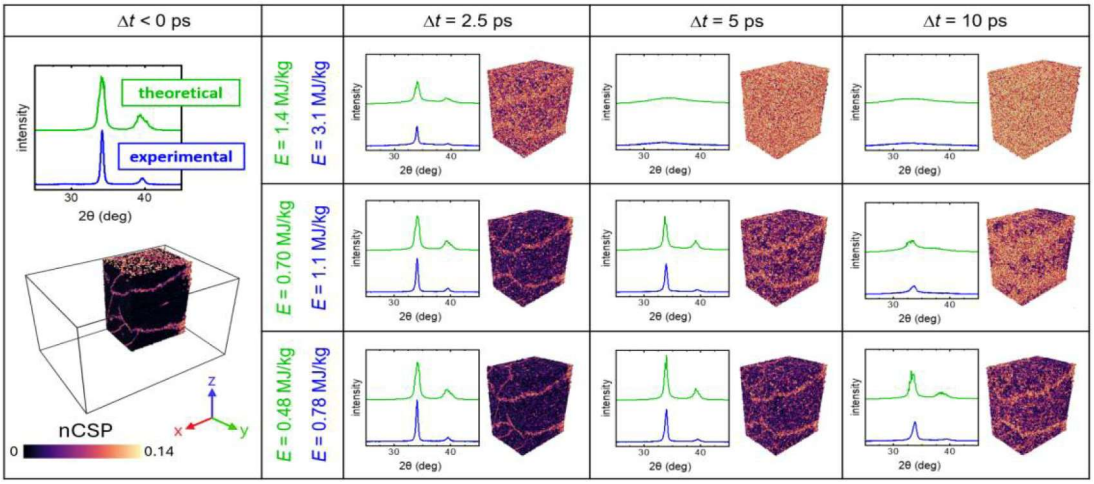

Figure 1. Structural evolution of the laser-irradiated polycrystalline Pd film, as revealed by molecular dynamics simulations. Experimental (blue) and theoretical (green) X-ray diffraction patterns are compared for different energy densities and pump-probe delay times, capturing various stages of the melting transition. Atoms are color-coded based on their deviation from local centrosymmetry.

Acknowledgements: This work was supported by the National Science Centre, Poland - grant agreement No 2017/27/B/ST3/02860 and No 2021/43/B/ST5/02480. The access to the European XFEL was supported by a grant of the Polish Ministry of Science and Higher Education - decision no. 2022/WK/13.

References

1. J. Antonowicz et al., Acta Mater. 276 (2024) 120043.