

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

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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 transition¹. 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.

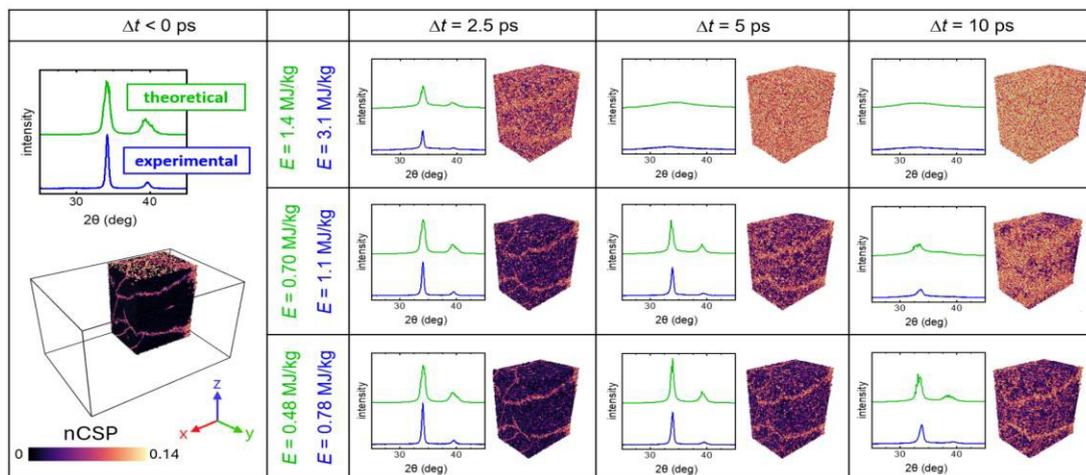


Figure 1. Structural evolution of the laser-irradiated polycrystalline Pd film, as revealed by molecular dynamics simulations. Experimental (blue) and theoretical (green) X-ray diffraction patterns are compared for different energy densities and pump-probe delay times, capturing various stages of the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

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References

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