Time resolved PDF investigation of inhomogeneous melting in thin metal films

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We performed ultrafast time-resolved diffraction experiments at the X-ray Free-electron Laser (XFEL) facility in Pohang, Korea to study 300 nm polycrystalline thin films of gold evaporated onto silicon nitride windows, melted by a Ti-sapphire laser pulse [1]. A clear splitting of the (111) powder ring was found at certain fluences after a time delay of 20-100 ps. From the evolution of the X-ray diffraction lineshape, we established separate roles for the electron and phonon contributions in the melting dynamics, consistent with the prevailing 2-temperature model. We deduced that the laser energy is primarily taken up by the electrons, which becomes transmitted into the crystal lattice preferentially at the grain boundaries, converting to heat which diffuses into the grains and eventually melts them. The appearance of liquid was then tracked by pair-distribution function analysis and found to have a slight time dependence following melting [2]. We concluded that the melting process is highly heterogeneous, commencing at the domain boundaries [1]. The role of domain boundaries in the electrical and mechanical properties of crystals is known from electrical conductivity measurements and theoretical modelling. This model can be tested on other metals with focussed ultrafast lasers at high power levels which are sufficient to melt a thin film in a single shot.

[1] Tadesse A. Assefa, Yue Cao, Soham Banerjee, Sungwon Kim, Dongjin Kim, Sunam Kim, Jae Hyuk Lee, Sang-Youn Park, Intae Eom, Jaeku Park, Daewoog Nam, Sangsoo Kim, Sae Hwan Chun, Hyojung Hyun, Kyung Sook Kim, Pavol Juhas, Emil S. Bozin, Ming Lu, Changyong Song, Hyunjung Kim, Simon J. L. Billinge and Ian K. Robinson, Science Advances 6 eaax2445 (2020)

[2] Ian K. Robinson, Jack P. Griffiths, Robert Koch, Tadesse A. Assefa, Ana F. Suzana, Yue Cao, Sungwon Kim, Dongjin Kim, Heemin Lee, Sunam Kim, Jae Hyuk Lee, Sang-Youn Park, Intae Eom, Jaeku Park, Daewoog Nam, Sangsoo Kim, Sae Hwan Chun, Hyojung Hyun, Kyung sook Kim, Ming Lu, Changyong Song, Hyunjung Kim, Simon J. L. Billinge and Emil S. Bozin, IUCrJ 10 656–661 (2023)