Glass transition and crystallization of metallic films: a molecular dynamics study.

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We investigate the kinetics of crystallization of thin films of molten BCC metals. The study is made using the molecular dynamics method on the examples of molybdenum and tantalum. The temperatures studied lie below the calorimetric glass transition temperatures for the melts.

To analyze the dynamics of the phase transition, we used the distributions of the crystalline nuclei in the «size-asphericity parameter» coordinates. The statistics was gathered over a large number of MD trajectories corresponding to the same temperature and density. There are a few features showing the deviation of the crystallization dynamics from the mechanism assumed in the classical nucleation theory:

1. For thin (6 nm) molybdenum and tantalum films it is found that a significant fraction of MD trajectories have a longer lifetime with respect to crystallization. The analysis of the probability densities shows that those systems have characteristic long-lived crystalline clusters with the size of about 70 atoms.

2. We found two mechanisms of crystallization on the example of molybdenum films: some trajectories crystallize through the growth of one crystalline cluster by attachment of atoms from the melt, some — through coalescence of smaller clusters into larger ones.

3. The cooling rate used to obtain the supercooled melt state has a significant effect on the crystallization mechanism and the probability of occurrence of the long-lived clusters. In the systems obtained at high cooling rate, the probability of long-lived clusters is higher and coalescence is the dominant crystallization mechanisms.
In the systems obtained at lower cooling rate, the probability of long-lived clusters is lower, and the crystallization mechanism shifts to the attachment.

4. We have found on the example of tantalum films that increasing the film thickness from 6 nm to 10 nm leads to elimination of the long-lived MD trajectories.

In the conclusion, we have demonstrated the effects of the cooling conditions and the system size on the crystallization mechanisms in thin metallic films. In particular, there might be a fundamental limitation on the thickness of an amorphous metallic film not related to the possibility of fast cooling of the material that’s far from the film surface.