Experimental observation of novel long-period structures and phase transitions in fcc [111] tilt grain boundaries by atomic resolution STEM

Meiners, T.[1], Frolov, T.[2], Liebscher, C.H.[1] and Dehm, G.[1]

[1] Max-Planck-Institut für Eisenforschung GmbH, Germany, [2] Lawrence Livermore National Laboratory, United States

The mechanical properties of polycrystalline materials are strongly influenced by the grain boundaries (GB), since the degree of atomic order is lower than in the bulk crystals. With this, also other properties such as the solute concentration and diffusive properties differ from that of the bulk. Already 50 years ago it was realized that GBs can exist in more distinct structures, but the resolution of microscopic techniques was limited to provide clear experimental evidence. Just recently, the coexistence of GB phases at metallic GBs was predicted by atomistic simulations. These phase transitions, also called complexion transitions, strongly influence the diffusive properties and solute decoration of a boundary, and affect their kinetic and mechanical properties. Revealing such GB phase transitions at the atomic scale provides the required resolution and precision to directly detect complex atomic arrangements at interfaces. In our study, we investigate the structure of symmetric and asymmetric [111] tilt GBs in copper thin films by aberration-corrected STEM and directly correlate the experimental observations to molecular dynamic (MD) simulations.

High angle annular dark field (HAADF) images of two symmetric Σ 19b tilt boundaries (46.83°[111]/{2-53} (Fig. 1a) and 46.83°[111]/{1-87} (Fig. 1b)) reveal a complex GB structure. Both symmetric GBs show a regular pattern consisting of two alternating structural subunits, indicated by red and green dots, forming long-period structural units. Each of the two GBs contains a deformed rectangular unit with 8 atomic columns (green dots). In Fig. 1a, those are connected by a trapezoidal formation of 4 atomic columns while it is in Fig. 1b a curve of 5 atomic columns (red dots). Thus, a change in GB plane normal leads to a significant modification in the local atomic structure. Our MD simulations reproduce both GB phases and demonstrate a first order GB phase transition at finite temperature (see Fig. 1a and b). The asymmetric 46.83°[111]/{1-10}{9-2-7} tilt boundary consists of similar building blocks (green and red dots in Fig. 2) as its symmetric counterpart from Fig. 1b, but those are irregularly extended by an additional plane of atomic columns (yellow dots in structure #1 in Fig. 2). A close inspection of the asymmetric boundary reveals a sharp transition to a second structural motif. This shows a complex domino structure, where a full structural unit consists of two domino units with a size of 1x2 (see blue dots in structure #2). During the phase transition all geometric GB parameters (misorientation and GB planes) remain the same for both phases, indicating a congruent phase transition. To our knowledge, this is the first direct experimental observation of two coexisting GB phases in an asymmetric tilt boundary employing aberration-corrected STEM. The atomic ordering of the structural units and the transition will be discussed in detail and will also be compared with MD simulation, where a similar phase transition in the symmetric 46.83°[111]/{1-87} tilt boundary is established.

Figure 1

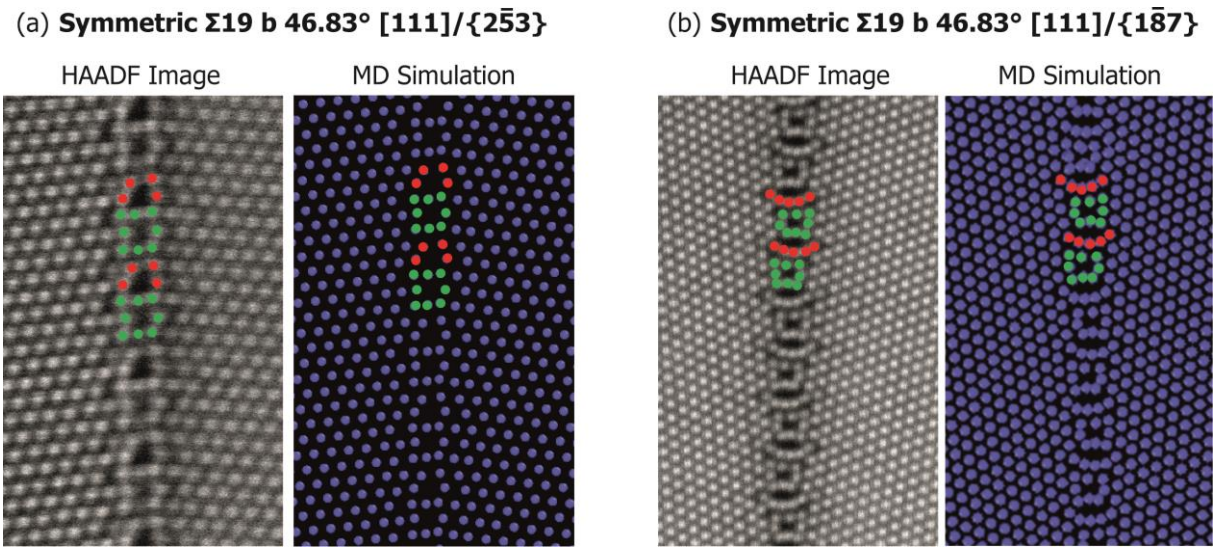
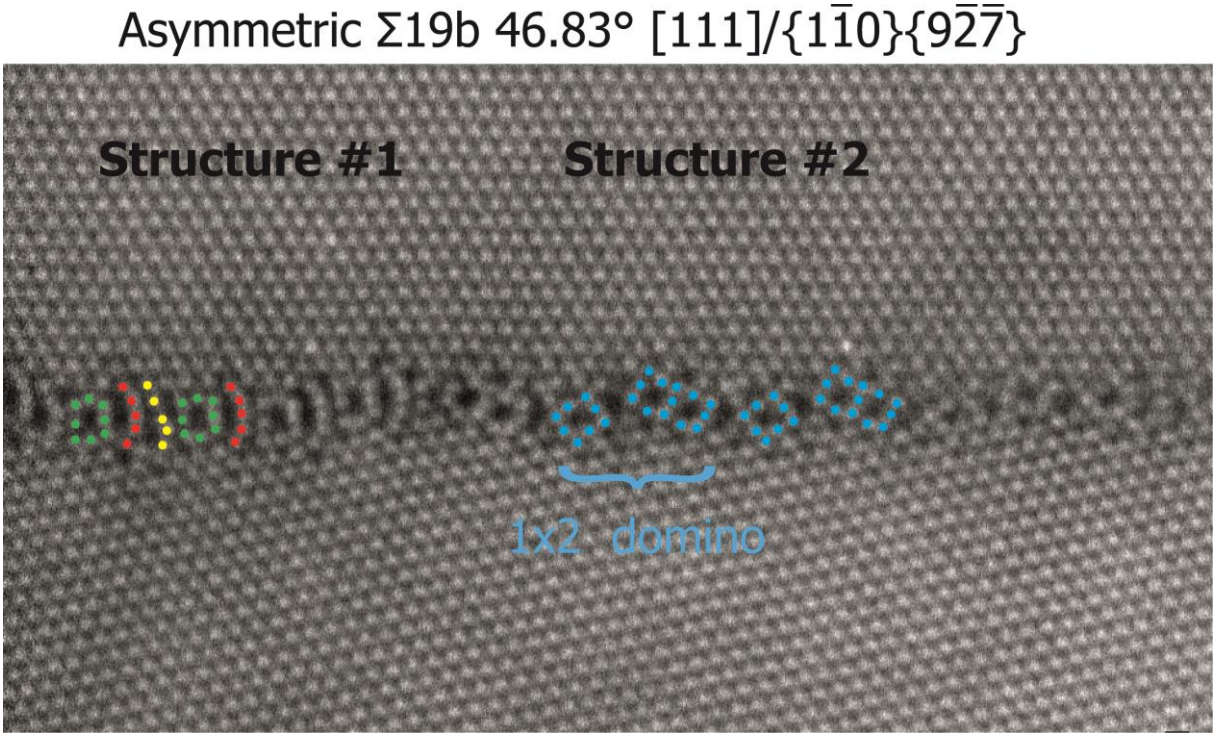

(a) Symmetric Σ19 b 46.83° [111]/{2$\bar{5}$3}
HAADF Image
MD Simulation
(b) Symmetric Σ19 b 46.83° [111]/{1$\bar{8}$7}
HAADF Image
MD Simulation

Figure 2


Asymmetric Σ19b 46.83° [111]/{1$\bar{1}$0}{9$\bar{2}\bar{7}$}
Structure #1
Structure #2
1x2 domino