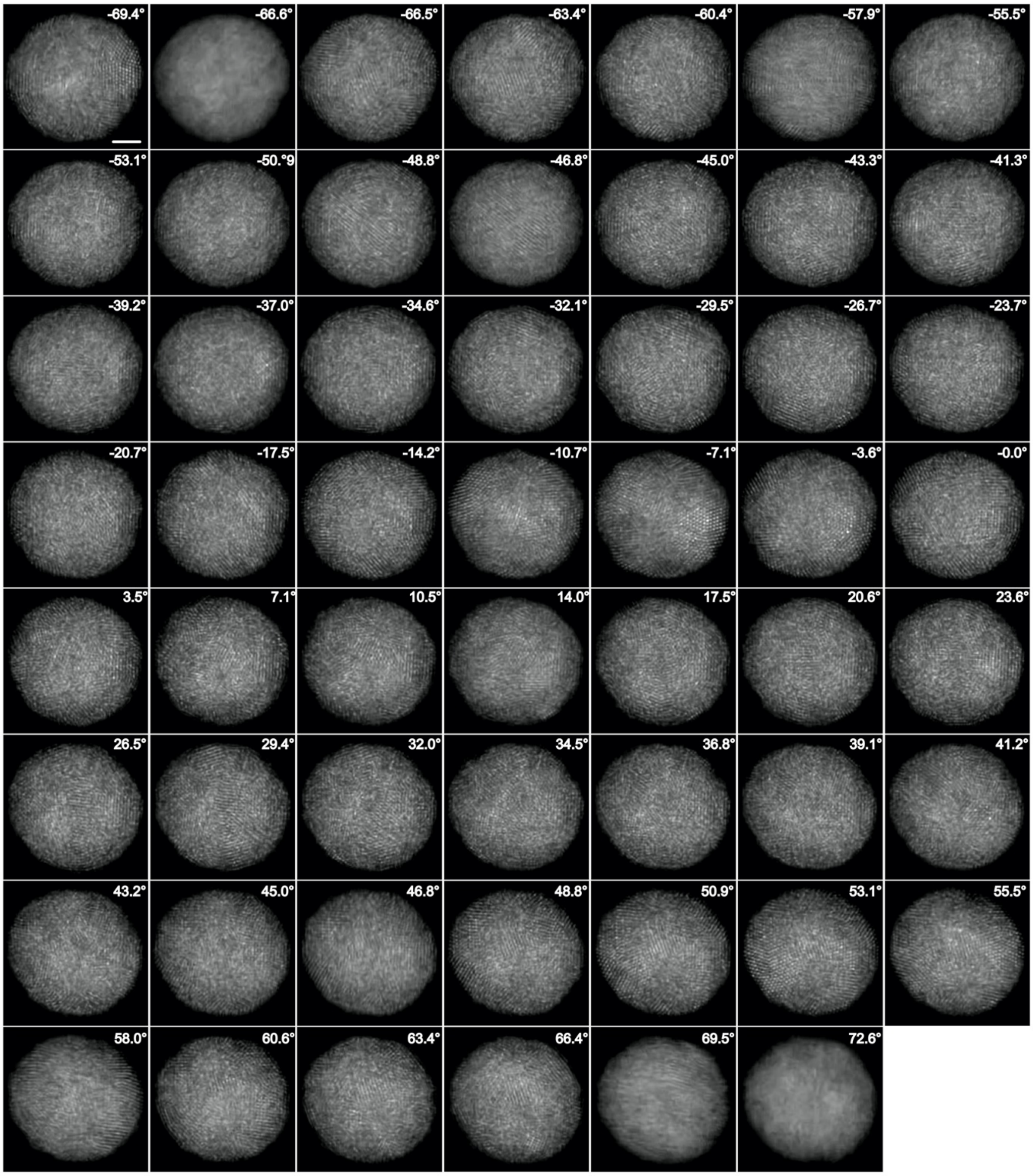
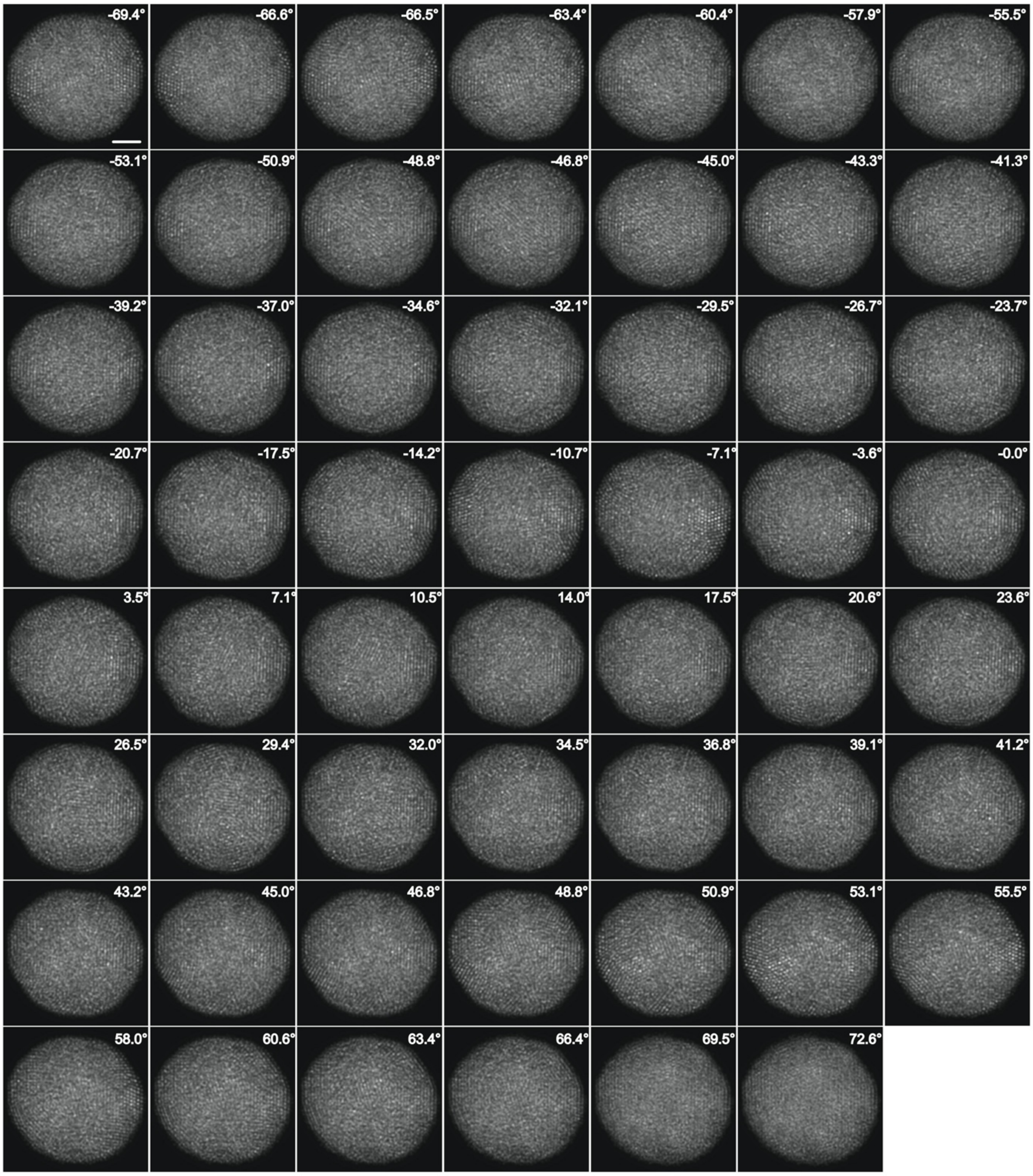
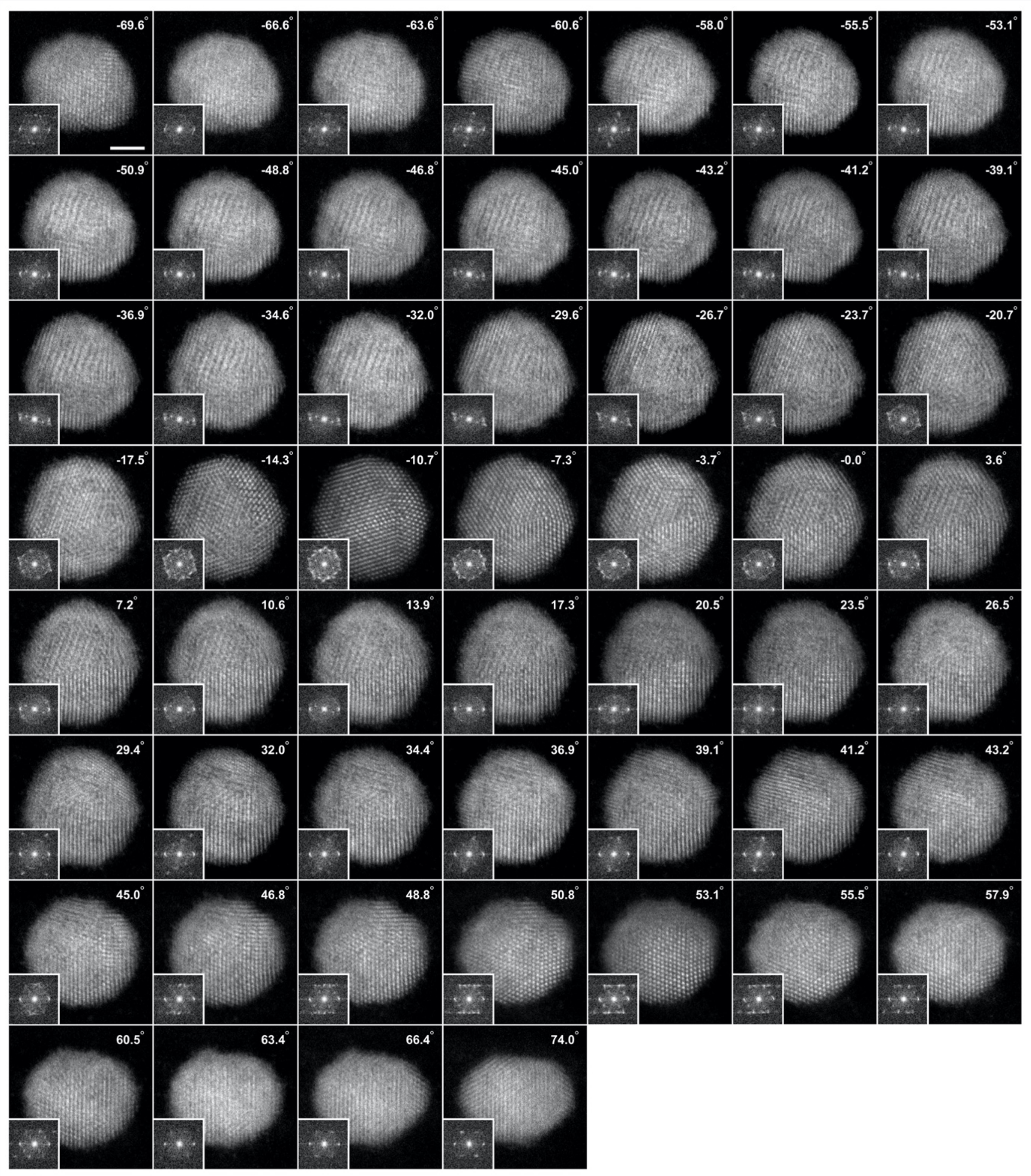
Supplementary Figures



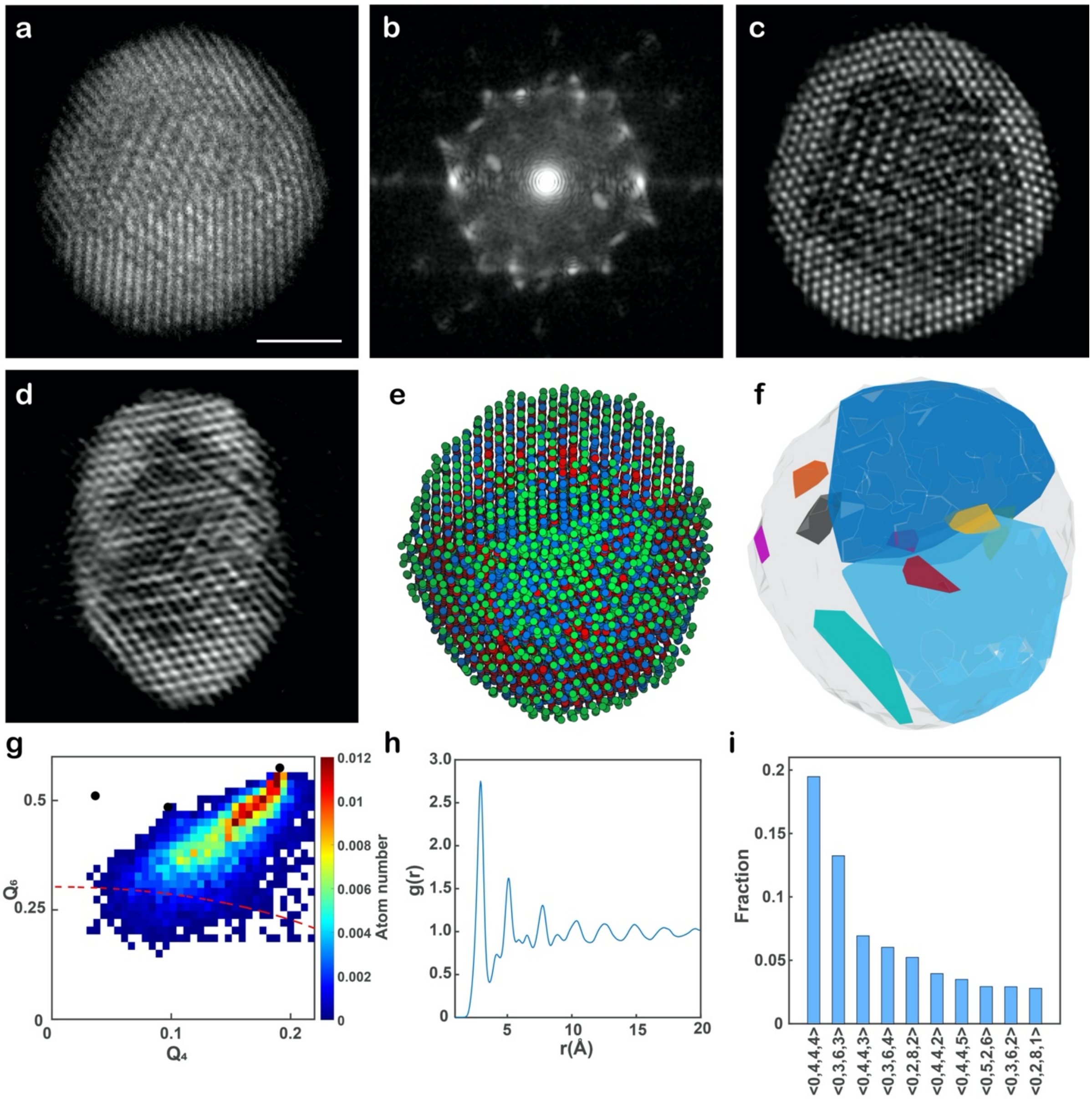
Supplementary Fig. 1 | Experimental tomographic tilt series of the multi-component metallic glass nanoparticle (particle 1) after denoising. 55 ADF-STEM images of the nanoparticle with a tilt range from -69.4° to +72.6°. Scale bar, 2 nm.



Supplementary Fig. 2 | 55 multislice STEM images of the multi-component metallic glass nanoparticle (particle 1). The multislice simulation was calculated from the experimental 3D atomic model (Methods). To account for the source size and incoherent effects, each multislice image was convolved with a Gaussian function. Scale bar, 2 nm.



Supplementary Fig. 3 | 53 raw experimental images of a polycrystalline nanoparticle (particle 5). The insets show the diffraction patterns calculated from the experimental images, where the Bragg peaks are visible. Scale bar, 2 nm.



Supplementary Fig. 4 | Determination of the 3D atomic structure of a polycrystalline nanoparticle (particle 5). a, A representative raw experimental image at 0°. Scale bar, 2 nm. **b**, Average diffraction pattern obtained from 53 experimental images (Supplementary Fig. 3), in which the Bragg peaks are visible. **c, d**, Two representative 2.4-Å-thick internal slices of the 3D reconstruction in the *xy* and *xz* planes, respectively. **e**, Experimental 3D atomic model of the polycrystalline nanoparticle, where green, blue and red balls represent type 1, 2 and 3 atoms, respectively. **f**, Distribution of ten crystalline grains (in different colors) inside the nanoparticle. **g**, The local BOO parameters of all the atoms in the polycrystalline nanoparticle. Based on the criterion of the normalized BOO parameter ≥ 0.5 (the dashed red curve), there are 94.25% crystalline atoms. **h**, The RDF of all the atoms in nanoparticle, where peaks corresponding to the different crystal planes are visible. **i**, Ten most abundant Voronoi polyhedra in the polycrystalline nanoparticle.