Quasicrystalline superlattices (QC-SLs) generated from single-component colloidal building blocks have been predicted by computer simulations but were challenging to reproduce experimentally. We, for the first time, discovered that 10-fold QC-SLs could generate through a self-assembling process. We used truncated tetrahedral quantum dots with anisotropic patchiness as the building blocks and created the QC-SL upon evaporation of the solvent on a liquid-air interface. Transmission electron microscopy and tomography measurements allow structural reconstruction of the QC-SL from the nanoscale packing to the atomic-scale orientation alignments. Interestingly, the QC-SLs possessed a unique QC order, leading to the discovery of a new tiling concept, the “flexible polygon tiling rule.” The anisotropic shape and patchiness of the building blocks and the microscopic assembly environment were identified to be the keys for the single-component QC-SL formation. Our discovery may spur the creation of further complex superstructures through a kinetically-driven route.