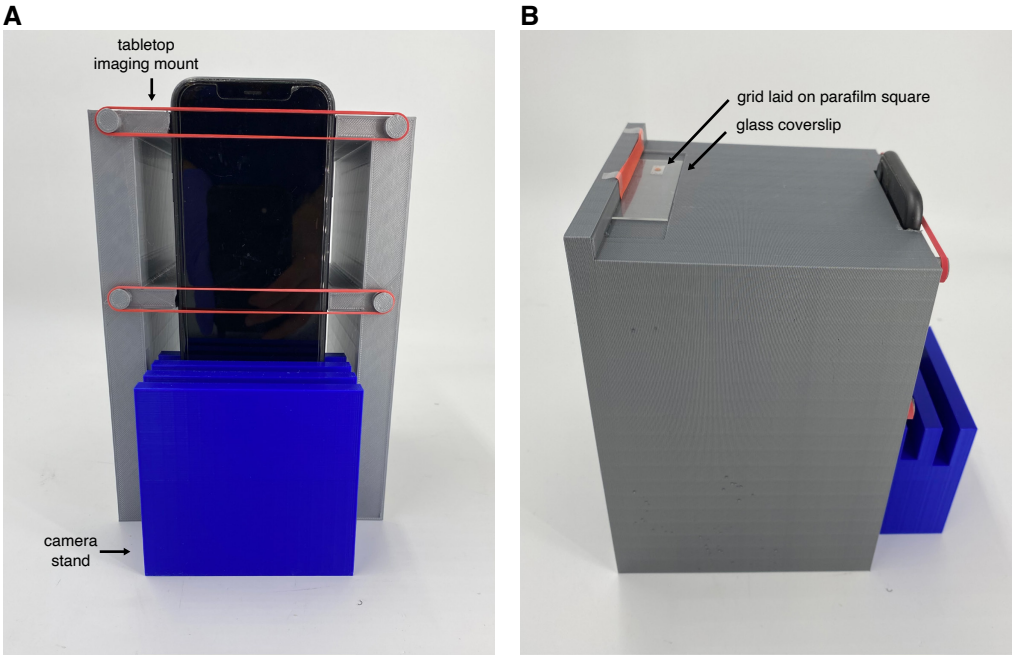
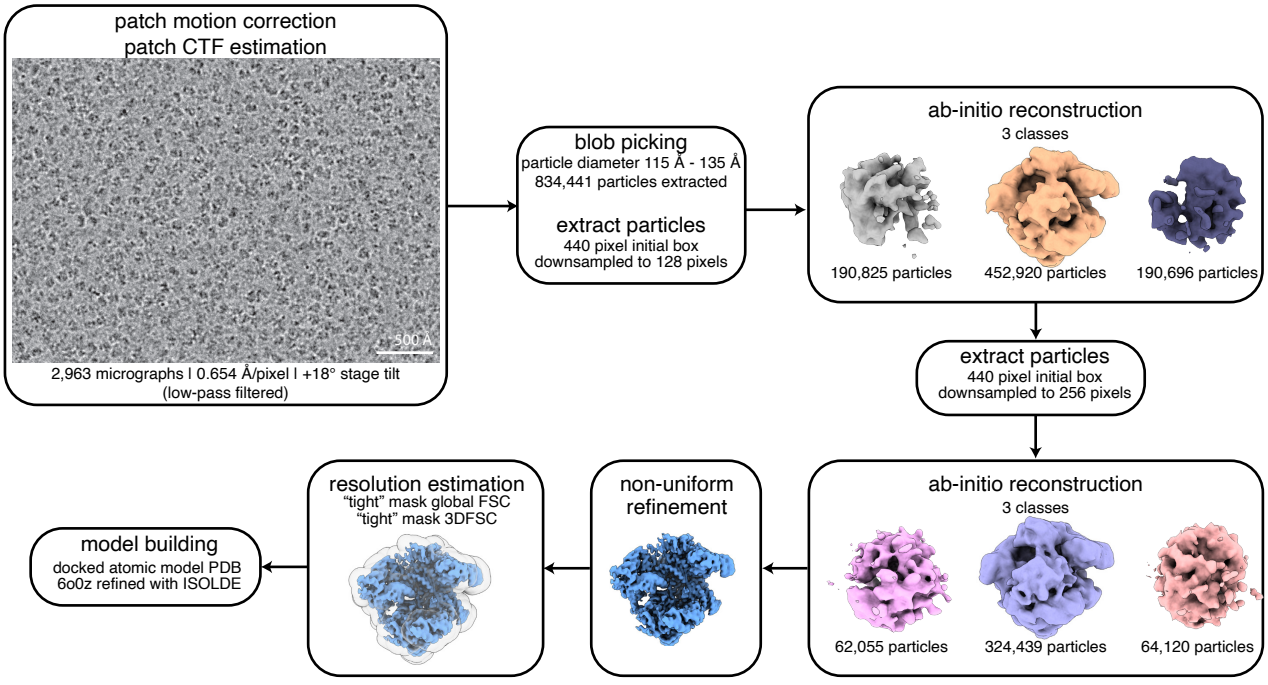


Supplementary Figure 1: Contact angle imaging stand. (A) 3D printed camera stand and tabletop imaging mount to secure camera in a position that aligns the camera in-plane with the coverslip. **(B)** Grid is placed on the coverslip on top of a 1 cm x 1 cm square piece of parafilm. Depicted imaging mount was 3D-printed using the provided .stl files, and uses an iPhone 11, but can be readily modified to accommodate other devices.



Supplementary Figure 2: Image processing workflow. CryoSPARC (v4.2.1) processing workflow for dCas9 complex. Job names, job details, and non-default parameters (italicized) are indicated.

SUPPLENTARY PROTOCOL

Protocol S1. Assessing grid hydrophilicity

S1.1 Prepare imaging setup.

- Locate a phone stand, tabletop imaging surface, phone with camera, glass coverslip, and parafilm.
  NOTE: Images shown in Figure 3 were obtained using a phone stand and tabletop imaging surface that were 3D printed using the provided .stl files, resulting in more easily reproducible and quantifiable contact angle measurements (**Figure S1A**).
- Lay a glass coverslip onto the flat surface.
- Cut a 1 cm by 1 cm square of parafilm and place it onto the glass coverslip.
- Place phone on the phone stand, orienting the phone such that the camera is in-plane with the glass coverslip. Secure phone in this position using rubber bands (**Figure S1B)**.

S1.2 Image grid.

- Take a sample photo to verify that the camera is aligned with the imaging surface.
- Directly after UV/ozone treatment of graphene-coated grids (see Protocol 2), place a single grid onto the square of parafilm on the glass coverslip.
  CRITICAL: Ensure the graphene side is facing upwards.
- Add a 2 µL water droplet onto the center of the grid surface with a pipette and immediately take a photo.

S1.3 Test treatment conditions.

- Repeat steps 1.5-1.6 after: *i*) desired intervals of UV/ozone treatment to determine a sufficient length of treatment; or *ii*) desired time intervals post UV/ozone treatment to measure how long after treatment the grid surface maintains its hydrophilic character.

S1.4 Quantify hydrophilicity.

- Calculate contact angles from photos by importing them into ImageJ[43] and using the Contact Angle plugin.

SUPPLEMENTARY PROTOCOL

Protocol S2. Single particle analysis of the dCas9 complex dataset.

NOTE: All image processing described in this protocol was performed using cryoSPARC version 4.2.1.

S2.1 Preprocess movies using the "Patch Motion Correction" and "Patch CTF Estimation" jobs.

S2.2 Perform particle picking using the "Blob picker" job, using a spherical blob ranging in diameter from 115 Å to 135 Å.

S2.3 Extract particles using the "Extract from micrographs" job, using normalized correlation coefficient (NCC) and power thresholds resulting in approximately 200-300 particles per micrograph.
NOTE: Appropriate thresholds and resulting particle count may vary, and users should inspect pick quality across a range of micrographs to identify suitable conditions.

S2.4 Perform multiclass initial reconstructions using the "Ab initio reconstruction" job, requiring three classes. Two of the three classes will likely contain non-Cas9 particles, including surface contaminants. Select the class resembling dCas9 for further processing. Additional rounds of multiclass "Ab initio reconstruction" or "Heterogeneous refinement" may be applied to further refine the particle stack.

S2.5 Perform 3D refinement using the "Non-uniform Refinement" job selecting default parameters.

S2.6 Estimate resolution of the reconstruction using the "Validation (FSC)", and "ThreeDFSC" jobs, employing the maps and mask from the final 3D refinement.