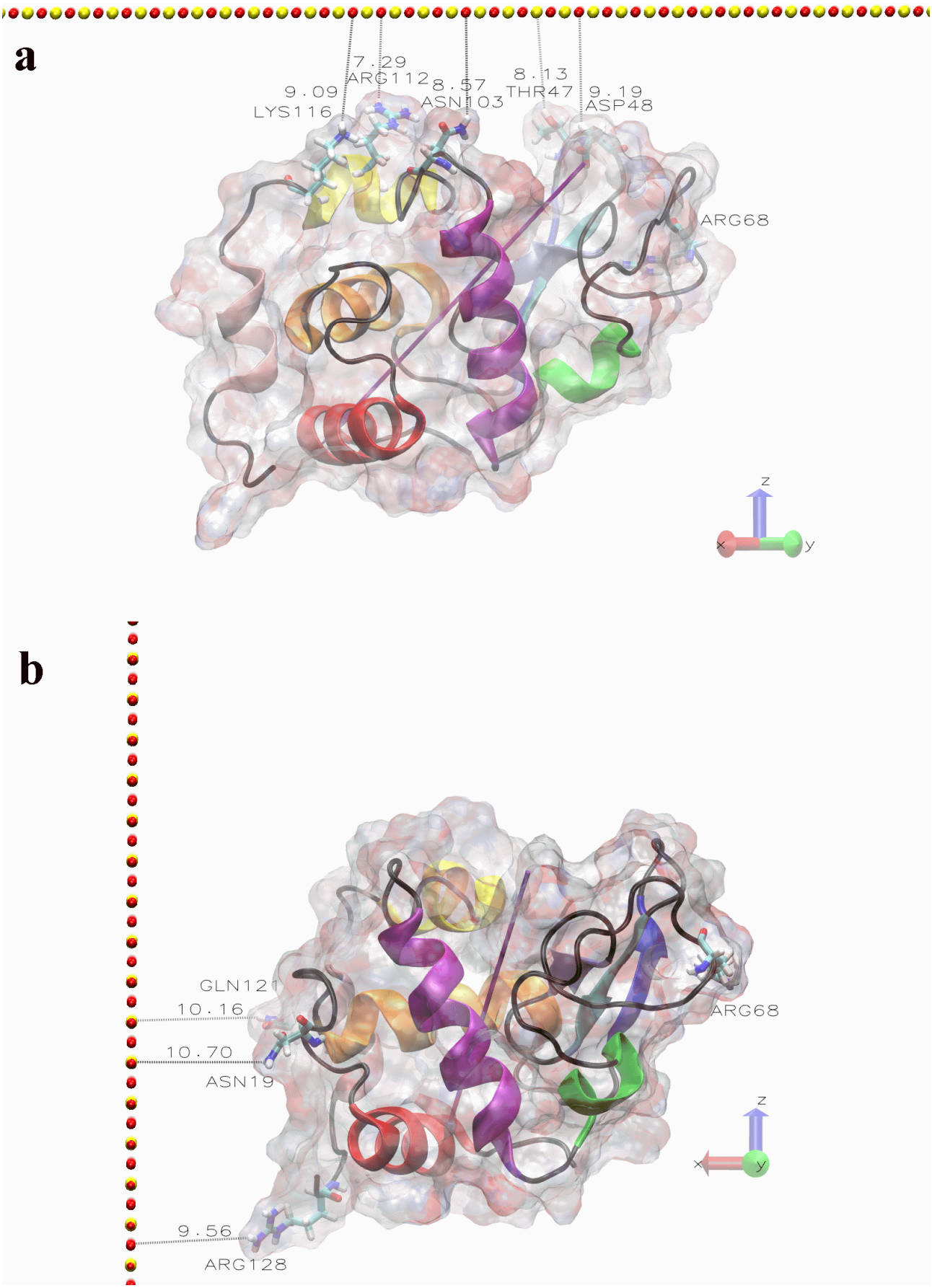
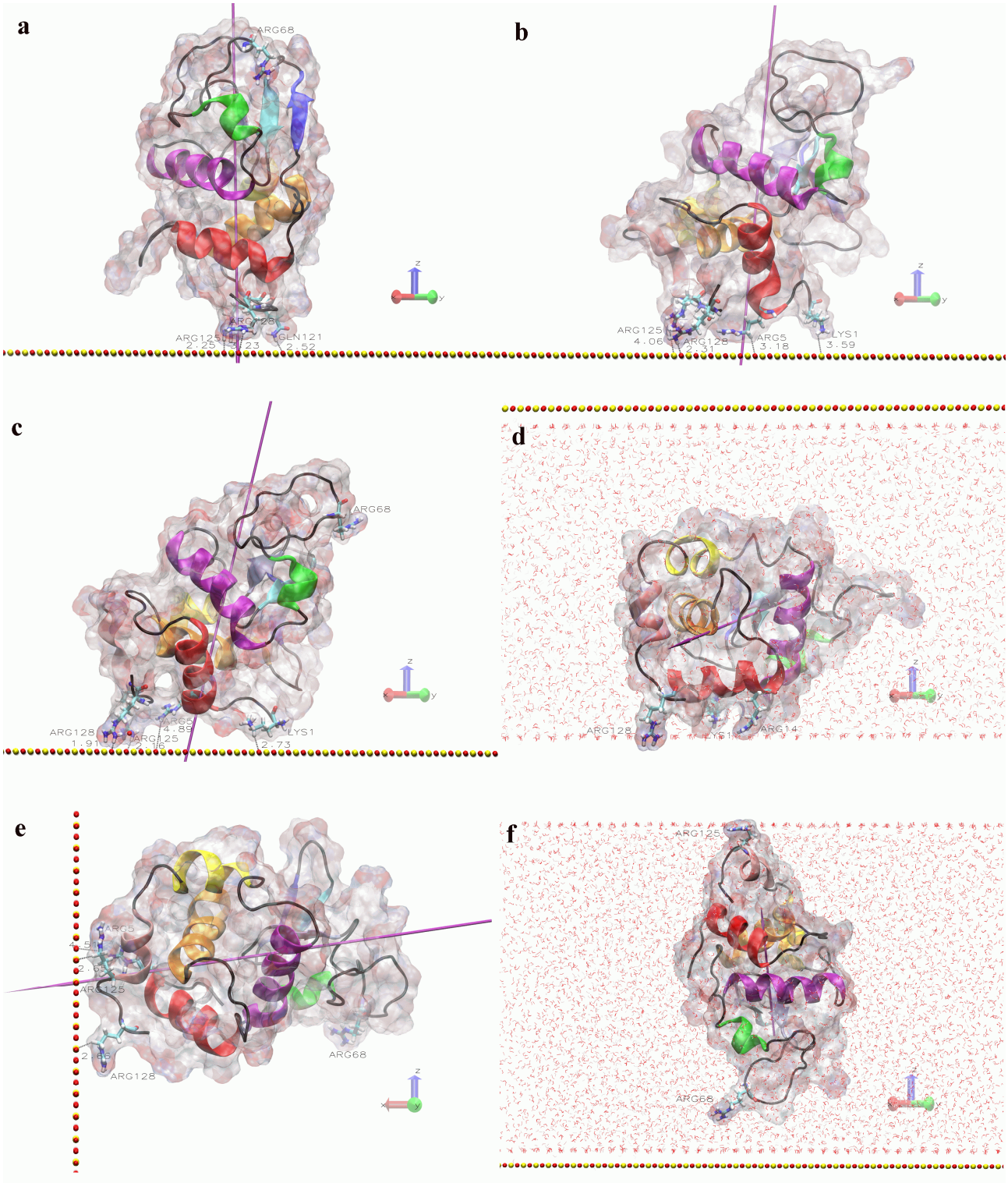
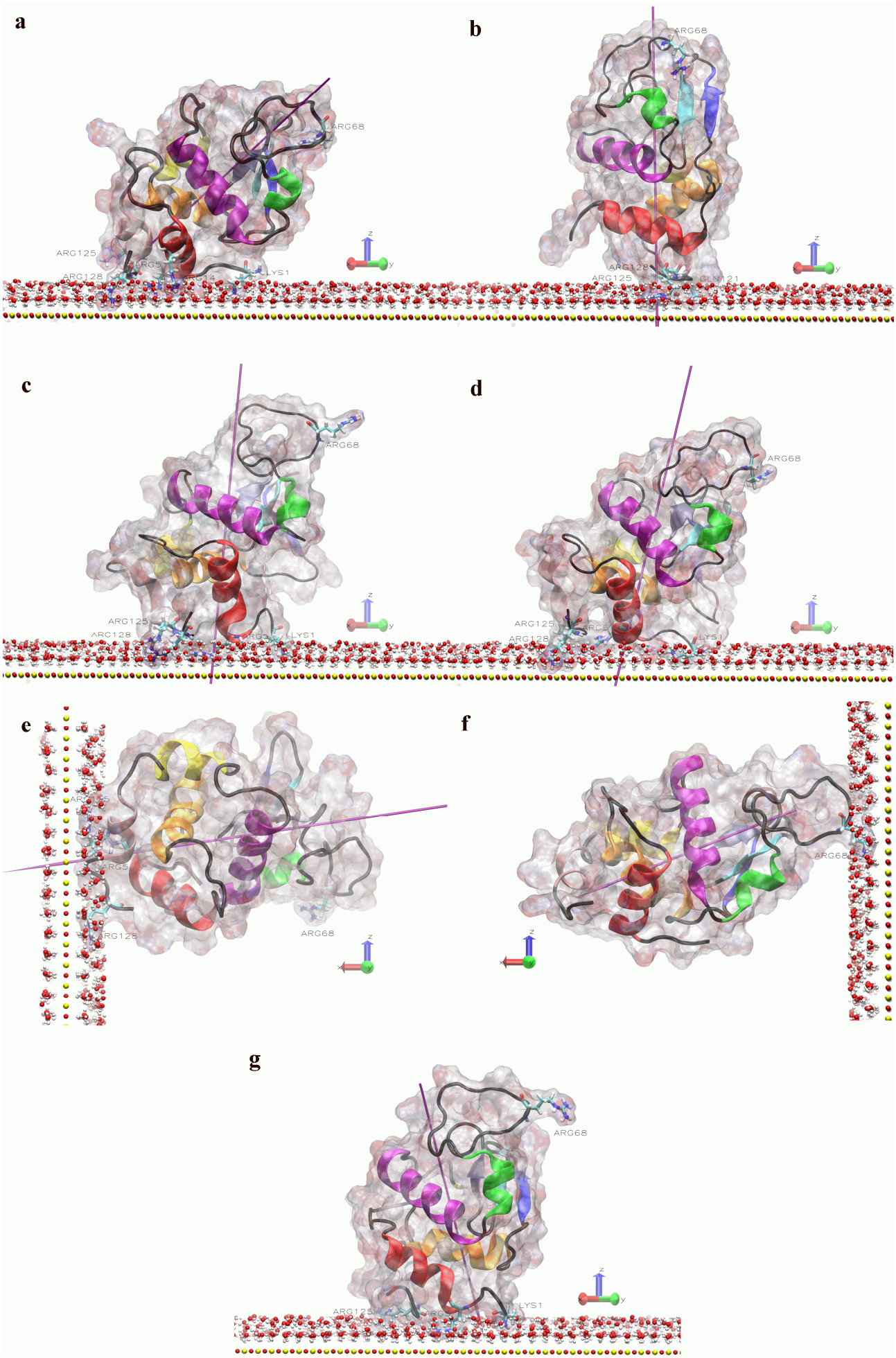

a
7.29
ARG112
9.09
LYS116
8.57
ASN103
8.13
THR47
9.19
ASP48
ARG68
z
x
y
b
GLN121
10.16
10.70
ASN19
ARG68
9.56
ARG128
z
x
y

Supplementary Figure 1. Protein and surface orientations at the initial stages of the trajectories. a, Orientation O2. b, Orientation O3. The shortest protein – surface distances are shown and important residues are labelled. The surface atoms are shown as red (oxygen atoms) and yellow (silicon) spheres. The HEWL surface is shown as a ghost surface coloured by charge, with protein secondary structure elements indicated as a cartoon and coloured as follow: red – helix A, orange – helix B, purple – helix C, yellow – helix D, pink – helix 3_{10} from domain α, green – helix 3_{10} from domain β, blue – sheet β1, cyan – sheet β2, grey – sheet β3, black – other structures including loops. Important residues are shown as licorice and coloured by element (hydrogen – white, carbon – cyan, nitrogen – blue, oxygen – red). The magenta needle indicates the protein dipole moment.



Supplementary Figure 2. Final protein – surface orientations. a, O1_002M_1. **b**, O1_002M_2. **c,** O1_002M_3. **d,** O2. **e,** O3. **f,** O1_R128G. The colouring method is given in Fig. S1. If the protein adsorbed to the image of the surface, the water molecules are shown as red lines.


a
ARG68
ARG125
ARG5
ARG128
LYS1
b
ARG68
ARG128
ARG125
LYS1
c
ARG68
ARG125
ARG128
LYS1
d
ARG68
ARG125
ARG128
LYS1
e
ARG125
ARG5
ARG128
ARG68
f
ARG68
g
ARG68
ARG125
LYS1

Supplementary Figure 3. Penetration through the water layers in the final structures. a, O1_002M. **b,** O1_002M_1. **c,** O1_002M_2. **d,** O1_002M_3. **e,** O3. **f,** O4. **g,** O1_90ns_R128G. Two surface water layers are shown by CPKs (distance from the surface smaller than 6 Å), residues penetrating the water layers are shown by licorice as is Arg68. Further details about the colouring method are given in Fig. S1.

Supplementary Movies:

O1.avi shows the 90ns adsorption simulation in orientation O1 discussed in the main text.

O4.avi shows the 90ns adsorption simulation in orientation O4.

O1_90ns_R128G.avi shows the 90 ns simulation starting the mutation at the end of the O1_002M simulation.

Methods

The simulations follow those of reference 5, extending the timescale from 20 ns to 90 ns for orientations O1, O2 and O3. Entirely new trajectories have also been performed for this paper, including 3 alternative realisations of O1, a new orientation O4, and two mutation simulations.

The starting protein structure was the X-ray structure of HEWL (1iee.pdb) solved by Sauter *et al*[6] with all four disulphide bridges kept. Trajectories were performed using NAMD 2.6 with the CHARMM27 force-field. Nine simulation systems were prepared. Two of them contained the protein placed in a periodic rectangular water box, with the ionic strength 0.5 M (trajectory I_05M) and 0.02M (trajectory I_002M), respectively, which are used to test the stability of the structure. Where it is not explicitly specified in the text, data and discussions relate to the 0.02M ionic strength simulations. The contrasting ionic strength was achieved in the protein neutralisation procedure by adding NaCl salt in two different concentrations. Seven further native systems were

composed from the protein and the surface in four different orientations (O1 to O4) with respect to each other. O1 denotes orientation 1 in which the surface was located as shown in Fig. 1a, in the *x,y* plane, close to the N,C-terminal protein face and approximately parallel to this part of protein surface. Translation of the surface in the *z* direction put its initial position close to the opposite protein side yielding orientation O2. In orientations O3 and O4 the surface was lying in the *y,z* plane. In O3, the surface was located close to Arg128, whereas translation in the *x* direction put the surface close to the opposite protein end yielding O4. The surface interactions presented here were performed with 0.02M. Due to its importance, we performed four separate 90 ns calculations in O1, denoted as O1_002M and three other trajectories O1_002M_1 to O1_002M_3.

We have also performed two mutation trajectories. From the starting configuration in O1, we have changed Arg128 to Gly, denoting this trajectory as O1_R128G. In addition, we mutated Arg128 to Gly at the end of the O1_02M_0 90 ns simulation, denoting this case as O1_90ns_R128G. Both these structures were subject to 90 ns dynamics.

The 86.4 x 92.8 $Å^2$ mica surface model was built from a square array of silicon and oxygen atoms located 1.6 Å away from each other with charges +1.11 e and -0.66 e, respectively. SiO_2 surface atoms were fixed in all stages of the MD simulations. The resultant surface charge density of σ = -0.0217 e/$Å^2$ was almost equal to that of mica at pH=7 (σ = -0.021 e/$Å^2$)[11]. The Lennard – Jones parameters for Si and O atoms were interpolated basing on chemical structure and similarity to already parametrized chemical moieties. For instance, similar to the phosphate atom bonded to four oxygen atoms (DMPA residue), epsilon and $1/2R_{min}$ of the Si atom were -0.585 kcal/mol and 2.15 Å, respectively. The parameters for oxygen atoms were -0.152 kcal/mol and 1.77 Å respectively, as in the existing ON2b atom. We use a single charged SiO_2 plane surface to simulate the mica surface.

The production MD simulations, preceded by water equilibration, minimisation, heating to 300 K and equilibration, were pursued for 90 ns at 300 K in the NVT ensemble. The integration step was 2fs, the SHAKE algorithm and PBC were used. The cut-off distance for both van der

Waals and Coulomb interactions was 12 Å. For ionizable residues the most probable charge states at pH 7 were chosen. The accuracy of the chosen protocol was checked by running test calculations with both cut-offs multiplied by 2 and also by using the SPME method. As we have already described, the results are not much affected by these changes[5]. No additional restrictions on momentum were used to allow for unconstrained protein – surface interactions and movement.