

Description of Additional Supplementary Files

Supplementary Movie 1. HS-AFM videos of three representative CaMKII β homooligomers on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Image size, 360 $\times$ 98 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 68 or 69 nm²; frame rate, 3.3 fps.

Supplementary Movie 2. HS-AFM videos of three representative CaMKII β homooligomers with 50 μ M bosutinib on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Image size, 240 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 64 nm²; frame rate, 3.3 fps.

Supplementary Movie 3. HS-AFM videos of three representative CaMKII α / β 3:1 heterooligomers on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Green arrowheads indicate the kinase domains of CaMKII β subunits. Image size, 240 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 64 nm²; frame rate, 3.3 fps.

Supplementary Movie 4. HS-AFM videos of three representative CaMKII α /GFP-CaMKII β 3:1 heterooligomers on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Green arrowheads and arrows indicate the kinase domains of CaMKII β subunits and GFP, respectively. Image size, 120 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 64 nm² or 100 $\times$ 80 nm²; frame rate, 3.3 fps.

Supplementary Movie 5. HS-AFM videos of three representative CaMKII β homooligomers treated with Ca²⁺/CaM (1 mM Ca²⁺ and 800 nM CaM) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. White dotted circles indicate Ca²⁺/CaM bound to the kinase domains, and blue dotted squares indicate kinase domain complexes. Image size, 360 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 82 or 80 nm²; frame rate, 3.3 fps.

Supplementary Movie 6. HS-AFM videos of three representative CaMKII α / β 3:1 heterooligomers treated with Ca²⁺/CaM (1 mM Ca²⁺ and 800 nM CaM) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Green arrowheads indicate the kinase domains of CaMKII β subunits. White dotted circles indicate Ca²⁺/CaM bound to the kinase domains, and blue dotted squares indicate kinase domain complexes. Image size, 240 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 64 nm²; frame rate, 3.3 fps.

Supplementary Movie 7. HS-AFM videos of three representative CaMKII β homooligomers treated with Ca²⁺/CaM and ATP (1 mM Ca²⁺, 800 nM CaM, and 1 mM ATP) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. White dotted circles indicate Ca²⁺/CaM bound to the kinase domains, and blue dotted squares indicate kinase domain complexes. Image size, 360 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 77, 72 or 76 nm²; frame rate, 3.3 fps.

Supplementary Movie 8. HS-AFM videos of three representative CaMKII α / β 3:1 heterooligomers treated with Ca²⁺/CaM and ATP (1 mM Ca²⁺, 800 nM CaM, and 1 mM ATP) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Green arrowheads indicate the kinase domains of CaMKII β subunits. White dotted circles indicate Ca²⁺/CaM bound to the kinase domains, and blue dotted squares indicate kinase domain complexes. Image size, 240 $\times$ 96 pixels² (acquired in HS-AFM observations); scan area, 80 $\times$ 64 or 100 $\times$ 80 nm²; frame rate, 3.3 fps.

Supplementary Movie 9. HS-AFM videos of three representative CaMKII β homooligomers treated with EGTA (2 mM) and ATP (1 mM) after Ca²⁺/CaM/ATP stimulation (1 mM Ca²⁺, 800 nM CaM, and 1 mM ATP) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Blue dotted squares indicate kinase domain complexes. Image size, 360 × 96 or 80 × 73 pixels² (acquired in HS-AFM observations); scan area, 80 × 74 or 73 nm²; frame rate, 3.3 fps.

Supplementary Movie 10. HS-AFM videos of three representative CaMKII α / β 3:1 heterooligomers treated with EGTA (2 mM) and ATP (1 mM) after Ca²⁺/CaM/ATP stimulation (1 mM Ca²⁺, 800 nM CaM, and 1 mM ATP) on a P[5]A+-modified mica surface. The trajectories of the kinase domains are shown on the right. Green arrowheads indicate the kinase domains of CaMKII β subunits. Blue dotted squares indicate kinase domain complexes. Image size, 240 × 96 or 120 × 96 pixels² (acquired in HS-AFM observations); scan area, 80 × 64 nm²; frame rate, 3.3 fps.

Supplementary Movie 11. HS-AFM videos of CaMKII α _{T286A} and CaMKII β _{T287A} homooligomers treated with Ca²⁺/CaM (1 mM Ca²⁺ and 800 nM CaM) on a P[5]A+-modified mica surface. An enlarged view of the white dotted box is presented on the right. White dotted circles indicate Ca²⁺/CaM bound to the kinase domains, and white dots indicate the center of the kinase domains. Frame rate, 3.3 fps.