## **Supporting Information**

## Flexibility and electrostatic interactions on the coupled bindingfolding mechanisms of Chz.core and H2A.z-H2B complex

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**Fig. S1** Chz.core Chemical shift difference ( $\Delta \omega_{RMS}$ , average value) between unbound and bound state at each residue (A) and the secondary chemical shift of  $C_{\alpha}$ . The calculations utilize structures corresponding to the experimental conditions. According to the experimental definition (Hansen et al., JMB, 2009), the chemical shift  $\omega$  is calculated by considering the unbound ensembles and the bound ensembles, separately. Therefore,  $\Delta \omega_{N(H)} = |\omega_{N(H)}^{bound} - \omega_{N(H)}^{unbound}|$ , where  $\omega_{N(H)}^{bound}$  and  $\omega_{N(H)}^{unbound}$  correspond to the chemical shifts of bound and unbound ensembles. Then the combined effect of H and N atoms are normalized according to BMRB database (http://www.bmrb.wisc.edu/).  $\Delta \omega_{RMS} = \sqrt{\frac{1}{2}\Delta \omega_{N}^2 / \Delta \omega_{STD,N}^2 + \Delta \omega_{H}^2 / \Delta \omega_{STD,H}^2}$ , where  $\Delta \omega_{STD,N}$ =3.8 ppm, and  $\Delta \omega_{STD,N}$ =0.6 ppm.  $\Delta \omega$  repersents the chemical shift difference between the unbound and the bound state. The secondary chemical shift  $\Delta \delta C_{\alpha} = \omega_{sim} - \omega_{randomcoil}$ , where  $\omega_{randomcoil}$  is the chemical shift values in the random coils, and  $\omega_{sim}$  represents chemical shift values of Chz.core  $C_{\alpha}$  atoms from the simulated ensembles at the bound and unbound states.



**Fig. S2** Chz.core backbone folding and side chain packing contact map at the encounter (A) and the loosely bound (B) state. The upper triangular map plots the side chain packing, while the lower region is for the backbone folding map.



Fig. S3 Native structure of Chz.core-H2A.z-H2B complex and the inter-chain interactions. Red, yellow and green represent N-terminus, motif and C-terminus of Chz.core.



**Fig. S4** Contact maps of inter-chain interactions at the transition state (A), the beginning of the flat binding stage (loosely binding)(B), and the native contacts in the model (C). (D) and (E) are the projections of contact map on Chz.core residues at transition state and the flat binding stage corresponding to the inter-chain contact maps (A) and (B).



**Fig. S5** Free energy landscape of Chz.core intrachain fraction of contacts Qf (A) and Qb between Chz.core and H2A.z-H2B (B) at different temperatures. The simulation temperature is in the unit of Boltzmann constant k



**Fig. S6** Distance map of Chz.core intra-chain interactions influenced by different salt concentrations in 0.01, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3 M.



**Fig. S7** Free energy landcapes as a function of Qb(A), Qf(B), and  $R_{com}(C)$  (center of mass distance between Chz.core and H2A.z-H2B). Results are collected at the transition temperature.



**Fig. S8** Probability evolution of forming "helix" during the binding process. A-F represent the different flexibilities (*c* from 0.1 to 5.0).



**Fig. S9** Probability evolution of forming "turn" during the binding process. A-F represent the different flexibilities (*c* from 0.1 to 5.0).



**Fig. S10** 3D free energy landscape as a function of 3 coordinates of Qb, Qf and Rg (A) sliced in the binding direction, and the 2D free energy landscape versus Qb and Rg (B).



**Fig. S11** The electrostatic interactions in the encounter complex with different relative positions between Chz.core and H2A.z-H2B.



Fig. S12 Structures evolution in binding process with different Qb at 0, 0.2, 0.4, 0.6, 0.8.