Supporting Information for

Protein Detection using Quadratic Fit Analysis Near Dirac Point of Graphene Field Effect Biosensors

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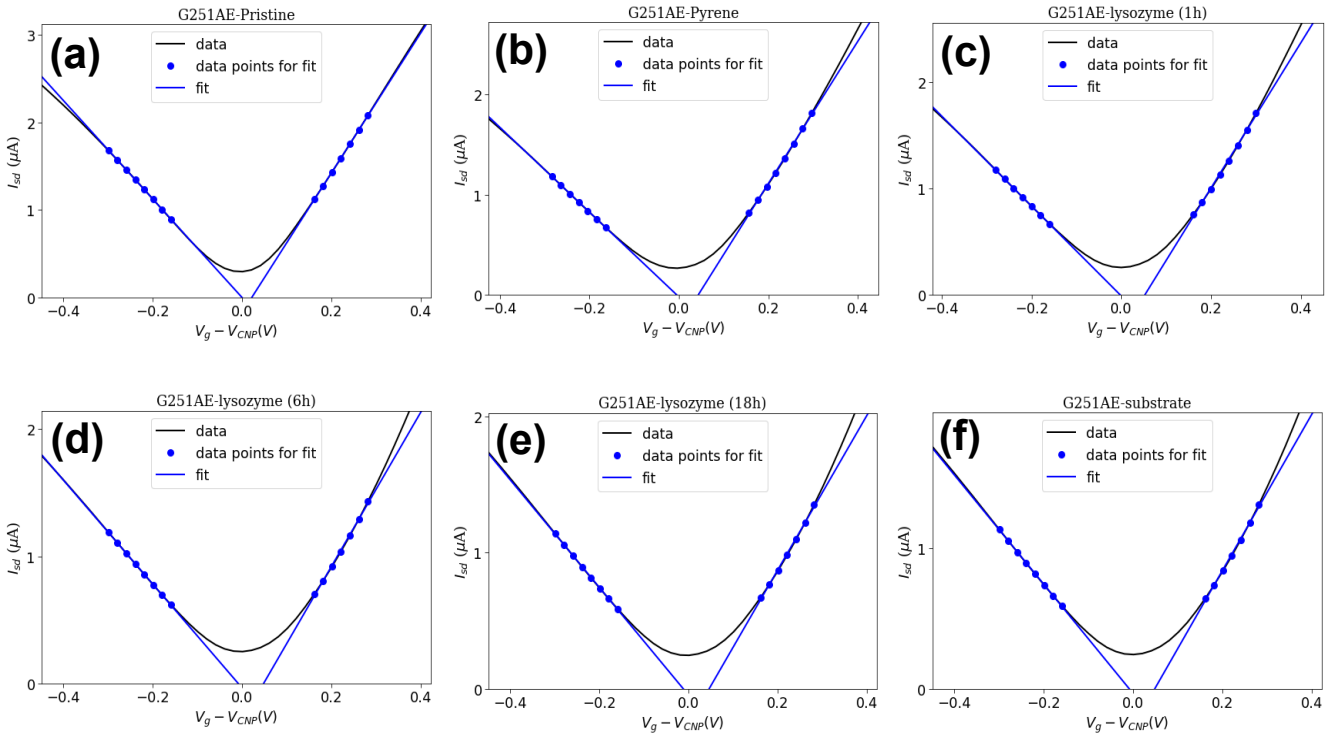

Figure S1. The linear fit model. $I(V_g)$ curves of (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, (e) 18 hr, and (f) introducing substrates, respectively, from Figure 2a in the main text.

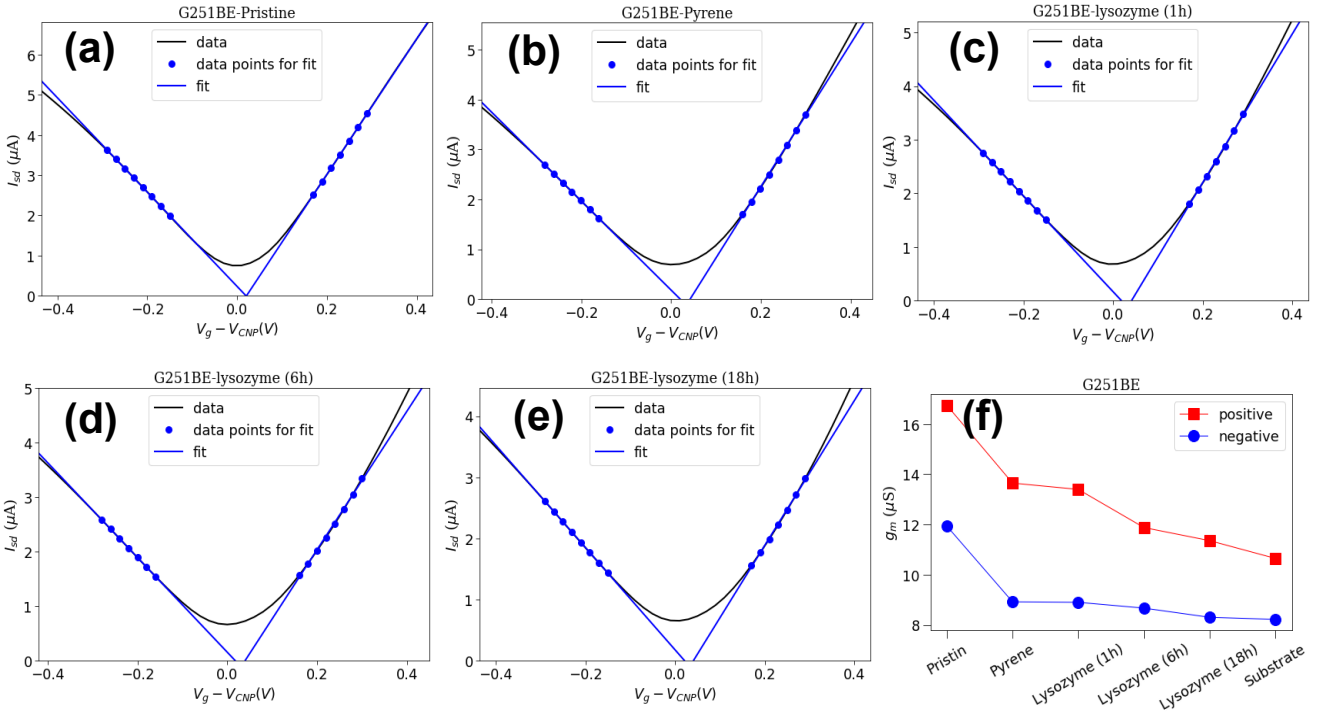

Figure S2. Additional example device with the linear fit model. $I(V_g)$ curves of (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, (e) 18 hr, respectively. (f) The trend of slope ($\pm g_m$) acquired from the fit.

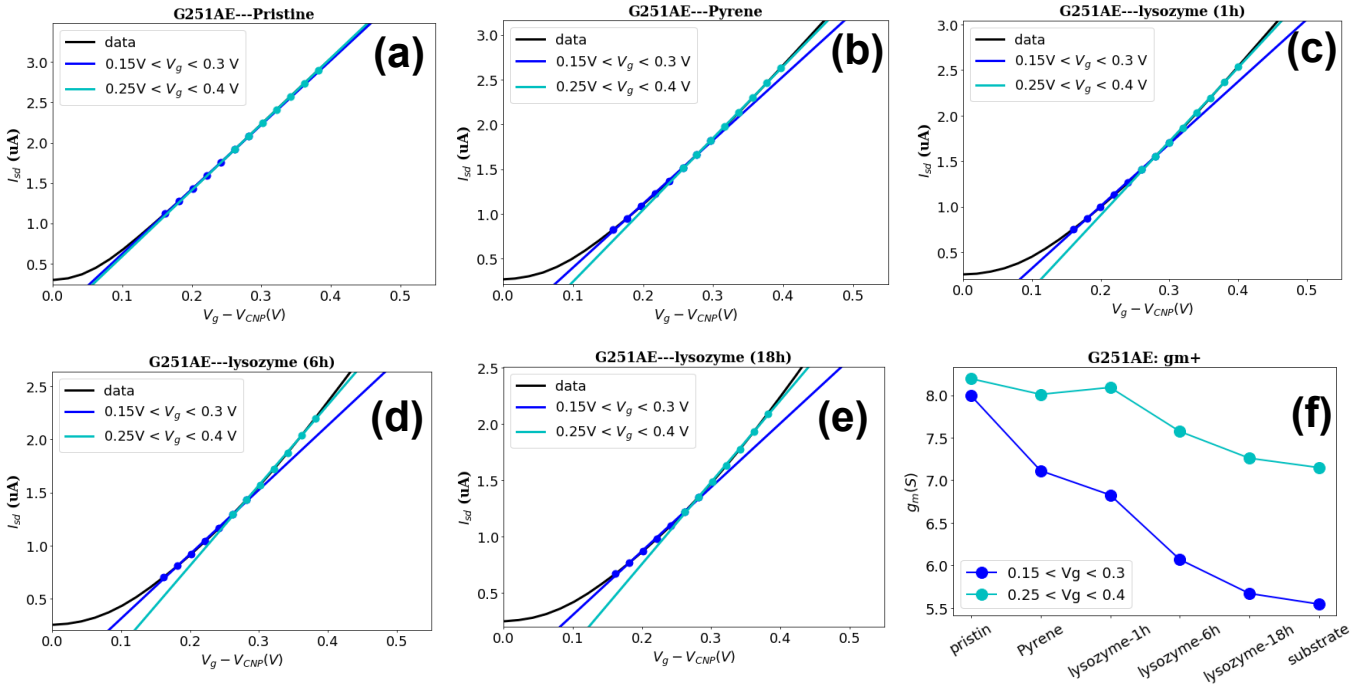

Figure S3. Comparison of the linear fit analysis obtained in two different gate voltage domains. The blue lines are the same fits shown in Figure 2 in the main text obtained at a gate voltage between 0.15 V and 0.3 V, and cyan lines are obtained from the interval of 0.25 V < V_g < 0.4 V. (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, (e) 18 hr. (f) The trends of slope (g_m) acquired from the fits at 0.15 V < V_g < 0.3 V and 0.25 V < V_g < 0.4 V. The pristine graphene in (a) shows linear $I(V_g)$ relation at V_g > 0.1 V but reveals a discrepancy between the data and the fit at V_g < 0.15 V and V_g > 0.3 V as a result of pyrene functionalization and protein conjugation processes (b) - (e).

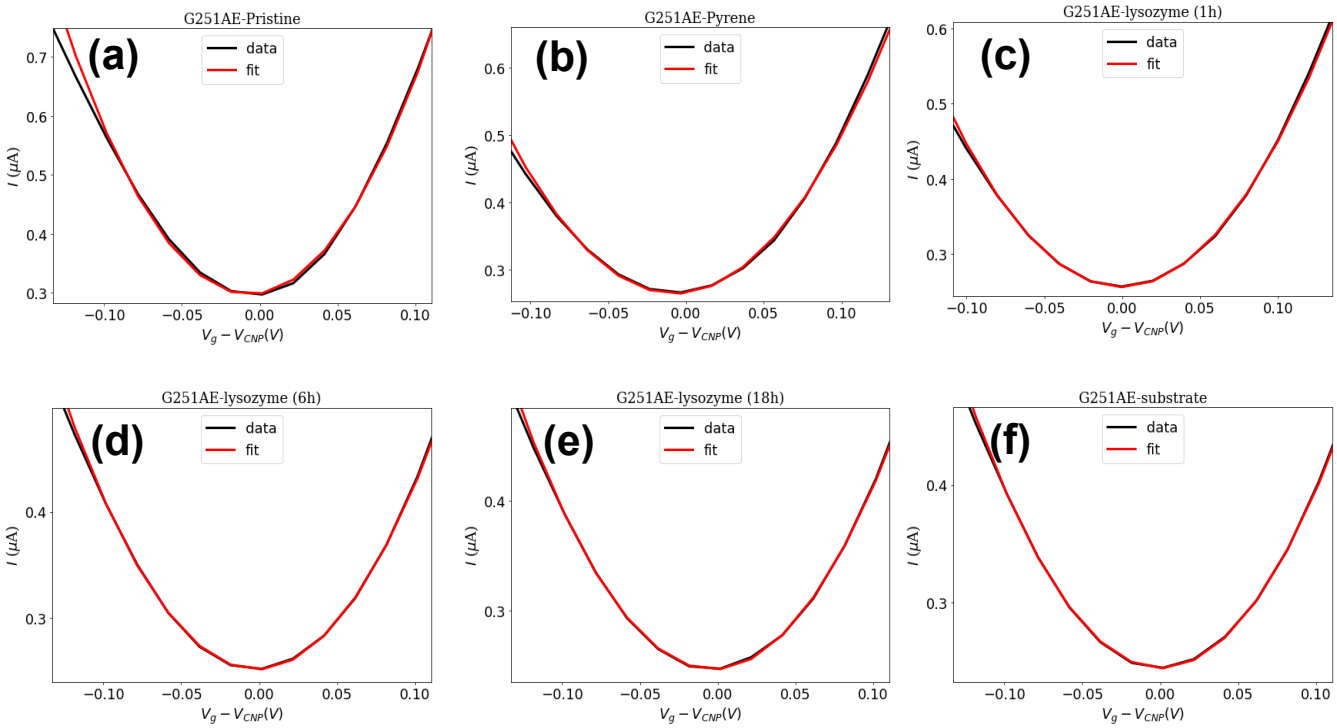

Figure S4. The quadratic fit model. $I(V_g)$ curves of (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, (e) 18 hr, and (f) introducing substrates, respectively, from Figure 2a in the main text.

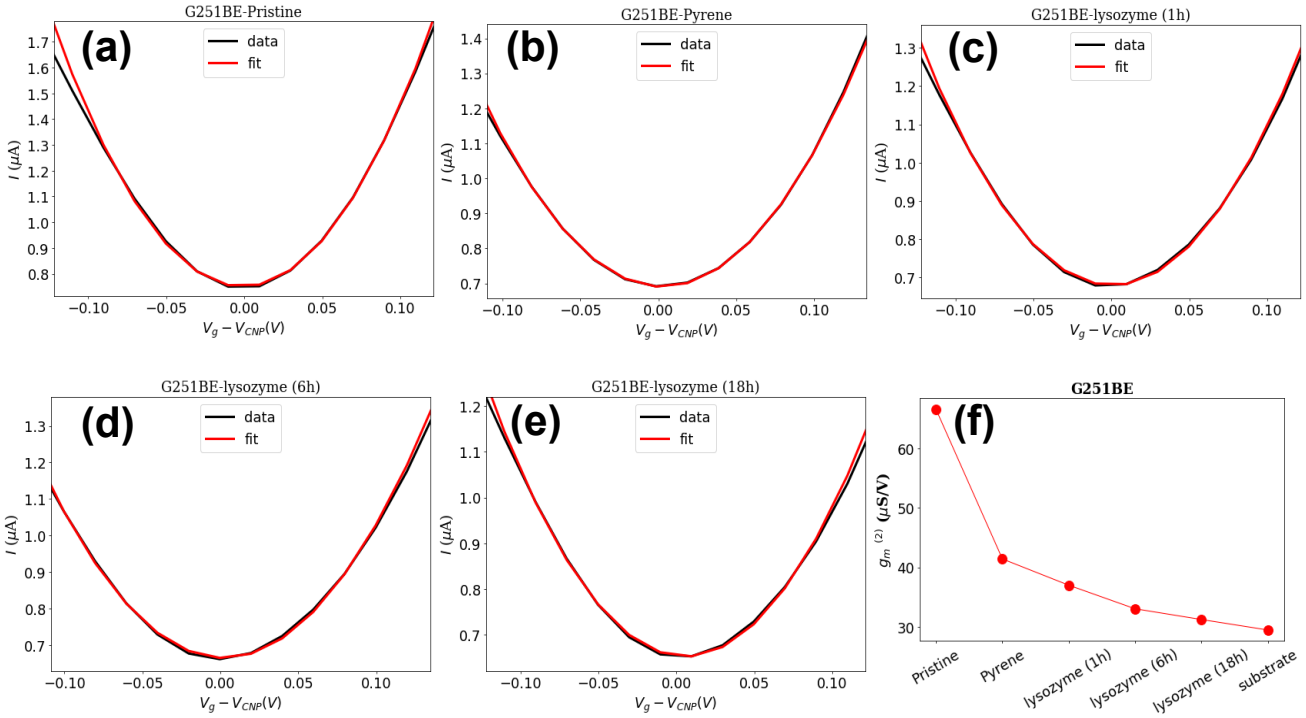

Figure S5. Additional example device with the quadratic fit model. $I(V_g)$ curves of (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, (e) 18 hr, respectively. (f) The fitting parameter ($g_m^{(2)}$) acquired from the fit.

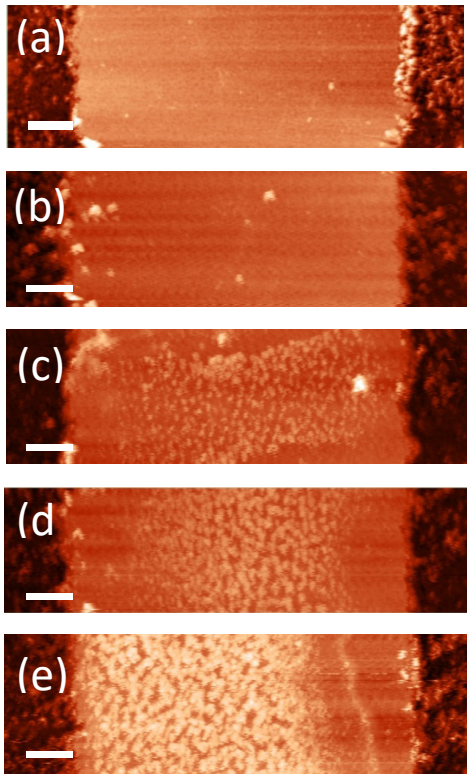

Figure S6. Additional examples of graphene devices imaged by AFM. (a) pristine graphene, (b) after linker functionalization, (c) bioconjugation with lysozyme for 1 hr, (d) 6 hr, and (e) 18 hr, respectively. All scale bare are 1 μm. The amount of attached proteins at each process is qualitatively distinguishable. The surface coverage of lysozymes on graphene estimated by ImageJ software in (c), (d), and (e) are 0.33, 0.49, and 0.73, respectively. However, the surface coverage is insufficient to determine the charge gating and the Coulomb interaction by each protein, since the orientation of protein through the pyrene linkers and interactions between proteins are not easily defined from the AFM images

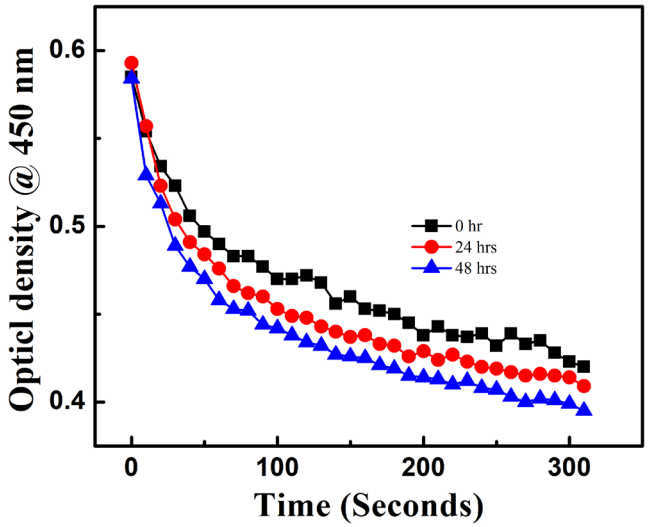

Figure S7. Long-term activity measurements of T4 lysozyme at room temperature using the standard assay kit (Micrococcus lysodeikticus, ATCC No. 4698, Sigma-Aldrich). All three curves acquired at varied times have almost identical slopes, indicating very consistent enzyme activity over 48-hour time period.

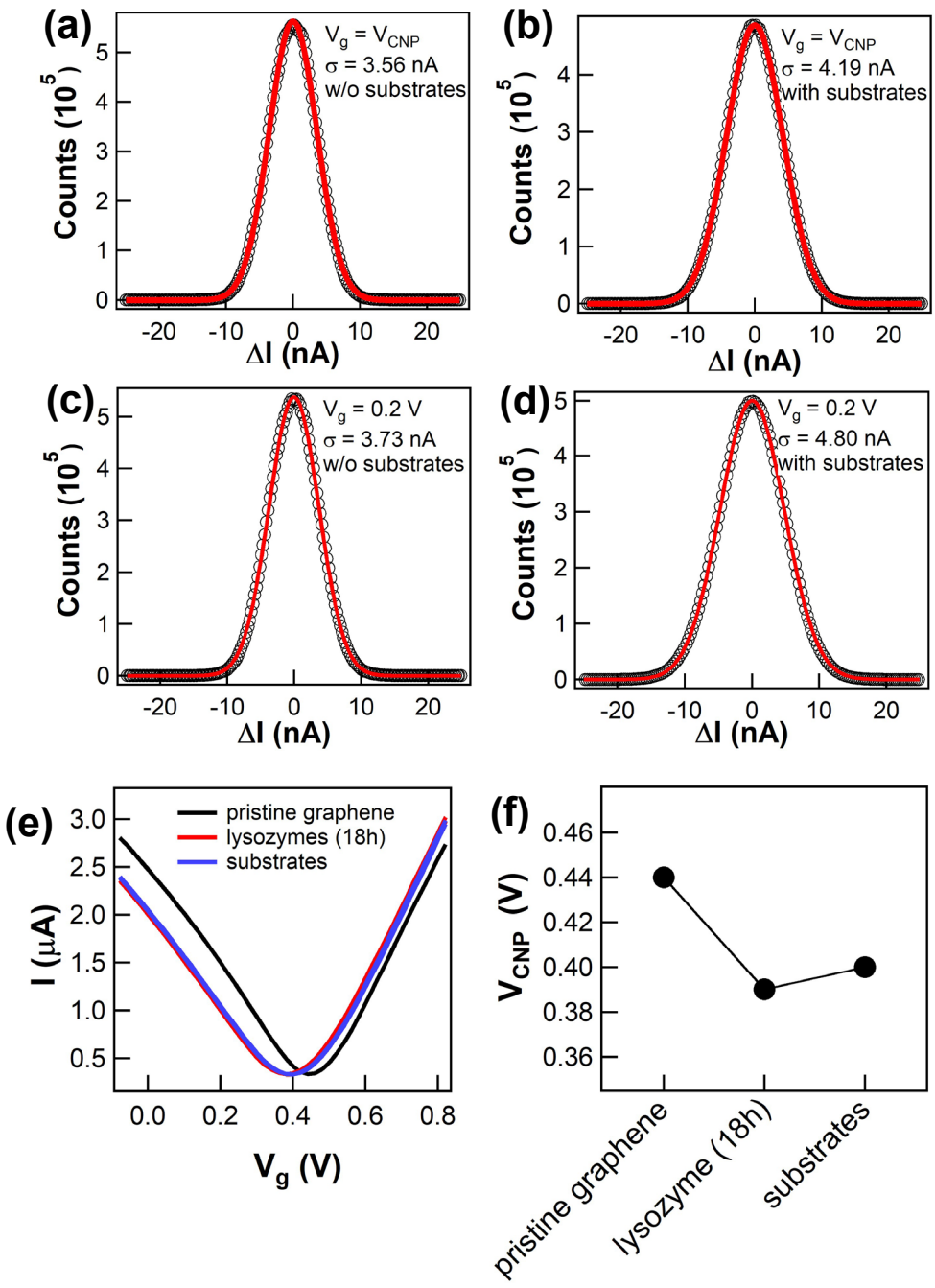

Figure S8. Control measurements performed with the graphene FETs without the pyrene linker functionalization. Histogram of $\Delta I(t)$ obtained at $V_g = V_{CNP}$ and $V_g = 0.2$ V in the absence of the substrates (a, c) and the presence of the substrates (b, d). The shift of $I(V_g)$ curve (e) and V_{CNP} value (f) corresponding to the lysozyme and substrate incubation.