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## Supporting information<sup>†</sup> for "Single protein sensing with asymmetric plasmonic hexamer via Fano resonance enhanced two-photon luminescence"



**Figure S1** (a) Charge distributions of two subradiant states of APH. The color scale is saturated. (b) The bird's eye view of the three dimensional electric intensity distributions  $|E|^2$  near the corresponding Fano resonant wavelengths at different excitation polarizations.



**Figure S2** (a) Evolutions of the extinction cross section when the protein is placed on several hot spots of the APH indicated in (b). (c) is the enlarge view for the features of the conventional sensing mechanism utilizing Fano resonance. (d) Dependence of the linear scattering cross section of the Fano resonance wavelengths on different locations of the protein above the APH.



**Figure S3** Evolutions of the normalized TPL intensity of the protein when the protein moves in the plane which is 6 nm above the surface of the APH at different directions (referring to different  $\theta$ ). It is normalized by the TPL of the protein without the APH under the same excitation condition. The normalized TPL is evaluated by using the Eq. 1 in the Method section. The inset indicates the excitation polarization.



**Figure S4** (a) Evolutions of the normalized electric field  $|E|^4$  (excitation wavelength at 920nm) and the electric field  $|E|^2$  (emission wavelength at  $\lambda$ =510nm) inside the protein when the protein moves towards the center of the APH with  $\theta$  = 90°, respectively. (b) Evolutions of the TPL intensity of the protein sphere from four representative directions towards the center of the APH. Solid lines are our approximated results (Eq. 2) in Fig. 2 (b) while the dotted lines stand for the results of Eq. 1.