

Supplemental Figures

Figure 1S. Time evolution of the fluorescence emission spectra. (A) 15 μM dT_{12} with 90 μM Ag^+ and 90 μM BH_4^- in a pH = 10.5 buffer using $\lambda_{\text{ex}} = 340 \text{ nm}/\lambda_{\text{em}} = 540 \text{ nm}$. (B) 15 μM $\text{dT}_4\text{C}_4\text{T}_4$ with 90 μM Ag^+ and 90 μM BH_4^- in a pH = 10.5 buffer using $\lambda_{\text{ex}} = 360 \text{ nm}/\lambda_{\text{em}} = 475 \text{ nm}$.

Figure 2S. Dependence of the fluorescence intensity with the relative concentration of Ag^+ :oligonucleotide for dT_{12} (A) and $\text{dT}_4\text{C}_4\text{T}_4$ (B). The concentrations of the oligonucleotides were 15 μM , and the pH of the buffer was 10.5. One equivalent of BH_4^- was used for one equivalent of Ag^+ . For dT_{12} and $\text{dT}_4\text{C}_4\text{T}_4$, $\lambda_{\text{ex}} = 340 \text{ nm}/\lambda_{\text{em}} = 540 \text{ nm}$ and $\lambda_{\text{ex}} = 360 \text{ nm}/\lambda_{\text{em}} = 475 \text{ nm}$, respectively.

Figure 3S. Induced circular dichroism and absorption spectra for the electronic transitions of the cluster-oligonucleotide conjugates. The solid line (left axis) represents the circular dichroism spectrum, and the dashed line (right axis) represents the absorption spectrum. (A) 15 μM dT_{12} with 90 μM Ag^+ and 90 μM BH_4^- . (B) 15 μM $\text{dT}_4\text{C}_4\text{T}_4$ with 90 μM Ag^+ and 90 μM BH_4^- .

Figure 4S: Fluorescence intensity (circles) of the $\lambda_{\text{ex}} = 340 \text{ nm}/\lambda_{\text{em}} = 540 \text{ nm}$ fluorescence species and the absorbance (crosses) at 430 nm as a function of time for a sample with 6 $\text{Ag}^+:\text{dT}_{12}$ in a buffer with pH = 10.5. The data sets were fitted using single exponential functions. The rate is 0.004 /s for the fluorescence, and the rate is 0.005 /s for the absorbance.

Figure 5S. Absorption spectrum of 15 μM dT_{12} with 90 μM Ag^+ and 90 μM BH_4^- in a pH = 10.5 buffer. The dotted line represents the spectrum in a nitrogen saturated sample while the solid line represents the spectrum in an oxygen saturated sample.

Figure 6S. Fluorescence spectrum were collected using $\lambda_{\text{ex}} = 340 \text{ nm}$ for a sample with 15 μM dT_{12} with 90 μM Ag^+ and 90 μM BH_4^- in a pH = 10.5 buffer. The dotted line represents the spectrum in the air saturated sample while the solid line represents the spectrum in the O_2 saturated sample.

Figure 7S. Fluorescence spectra in solutions saturated with nitrogen (dashed line) and in same nitrogen-saturated solutions that were subsequently saturated with oxygen (solid line). The conditions were 15 μM $\text{dT}_4\text{C}_4\text{T}_4$ with 90 μM Ag^+ and 90 μM BH_4^- in a pH = 10.5 buffer.

Figure 8S: Fluorescence quantum yield measurements for the cluster- $\text{dT}_4\text{C}_4\text{T}_4$ conjugates. Using $\lambda_{\text{ex}} = 370 \text{ nm}$, the fluorescence intensity at 475 nm is plotted as a function of the absorbance at 370 nm. The sample has 6 $\text{Ag}^+:\text{dT}_4\text{C}_4\text{T}_4$ in a pH = 10.5 buffer. The slopes are $4.2 (\pm 0.1) \times 10^7 \text{ c/s}$ (intercept = 70000 +/- 14000) for quinine and $1.1 (\pm 0.1) \times 10^7 \text{ c/s}$ (intercept = 35000 +/- 140000) for the silver clusters.

Figure 9S. Fluorescence emission intensities as a function of the excitation power for a 10X diluted sample of 15 μM $\text{dT}_4\text{C}_4\text{T}_4$ with 90 μM Ag^+ and 90 μM BH_4^- in a $\text{pH} = 10.5$ buffer. The 30 μW power used for these studies is indicated by the vertical line.

Figure 10S. (A) A composite fluorescence spectrum of 0.5 μM $\text{dC}_4\text{T}_4\text{C}_4$ with 90 μM Ag^+ and 90 μM BH_4^- in water. The emission wavelengths are on the bottom axis and the excitation wavelengths are on the right axis. The spectra were acquired 16 hrs after adding BH_4^- . (B) Fluorescence spectra in solutions saturated with nitrogen (dashed line) and in same nitrogen-saturated solutions that were subsequently saturated with oxygen (solid line). (C) Fluorescence intensities of the $\lambda_{\text{ex}} = 340 \text{ nm}/\lambda_{\text{em}} = 495 \text{ nm}$ band as a function of pH .

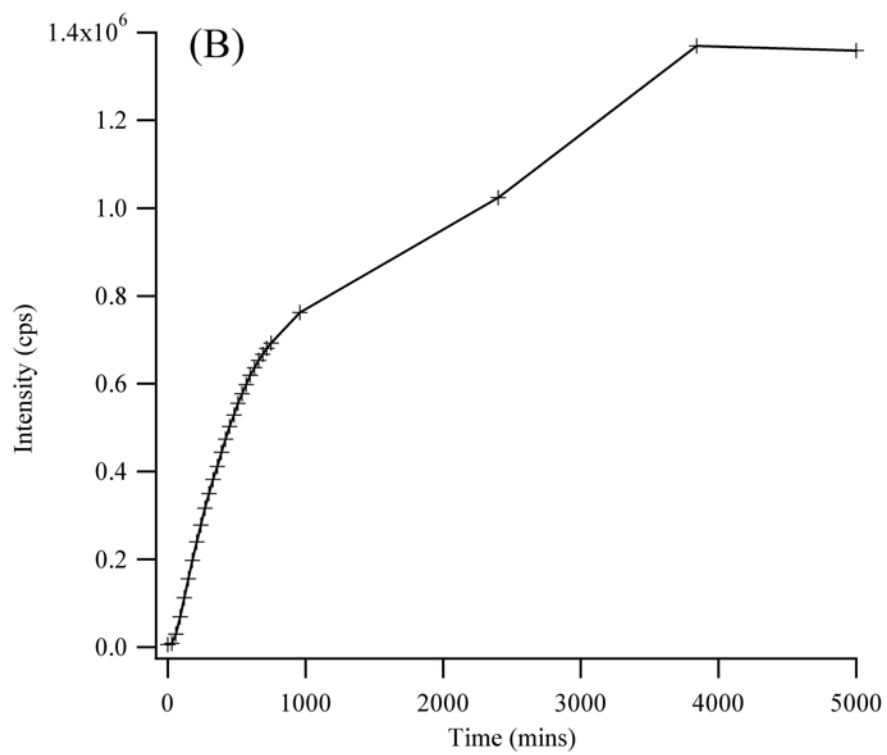
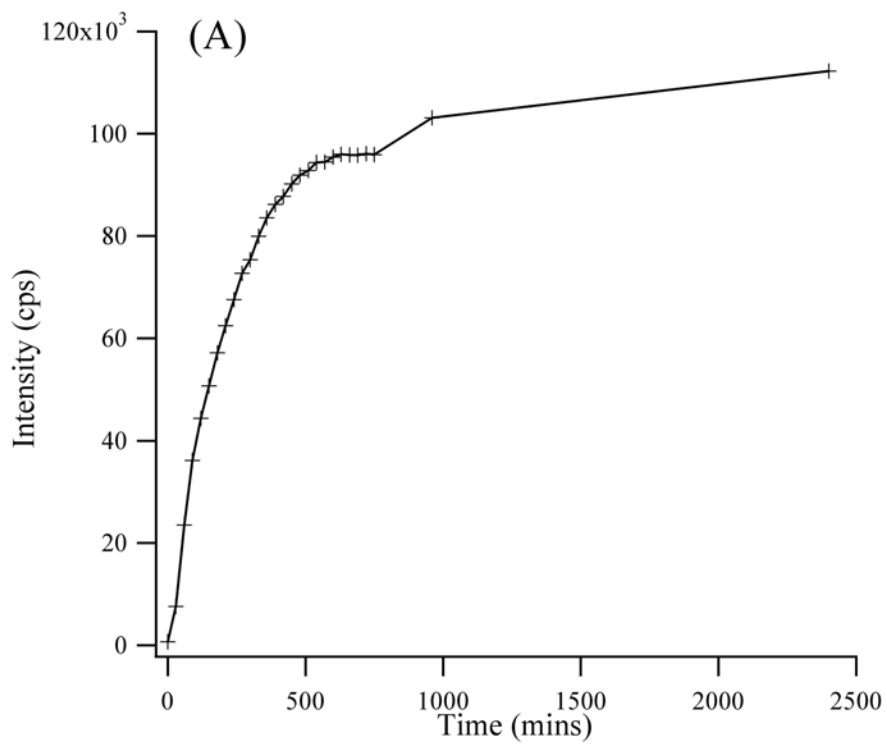


Figure 1S

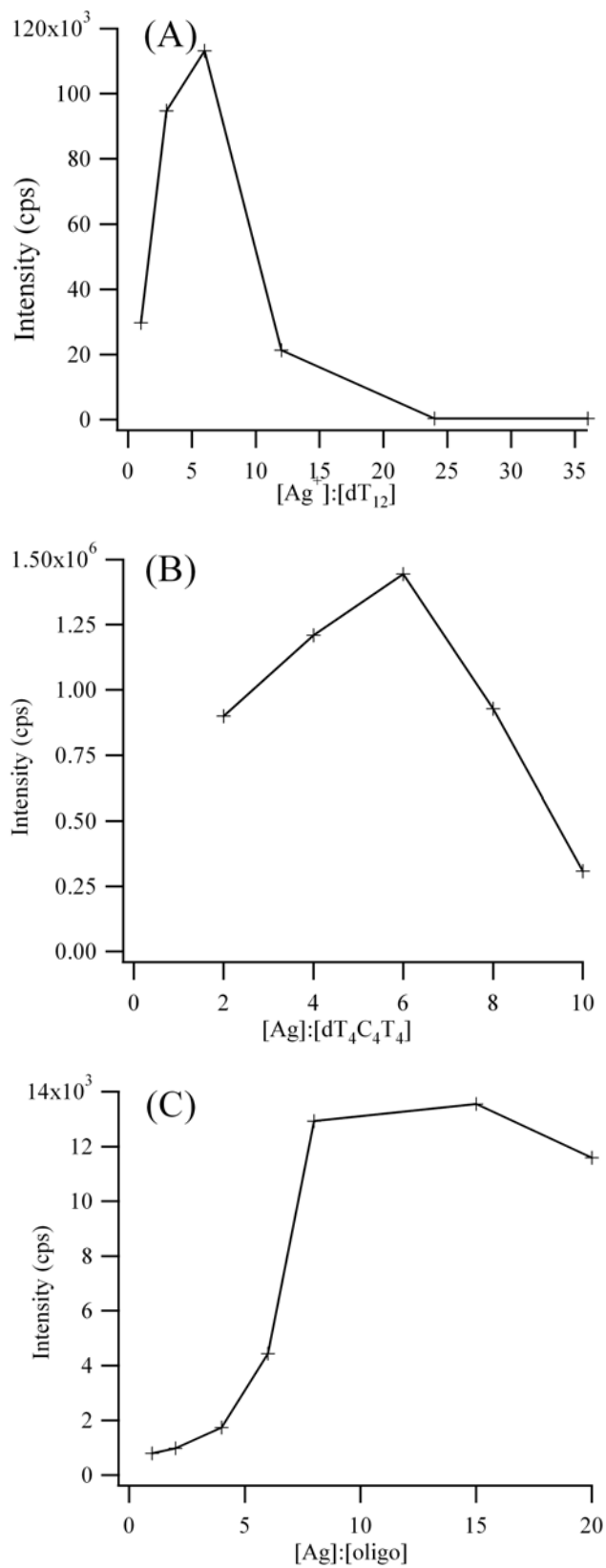


Figure 2S

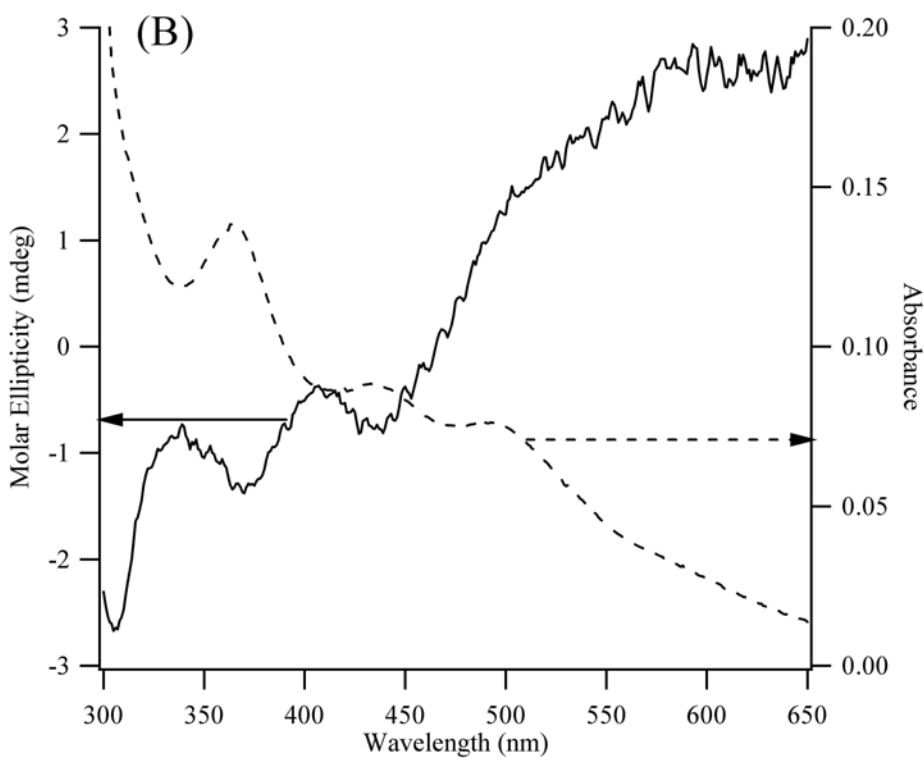
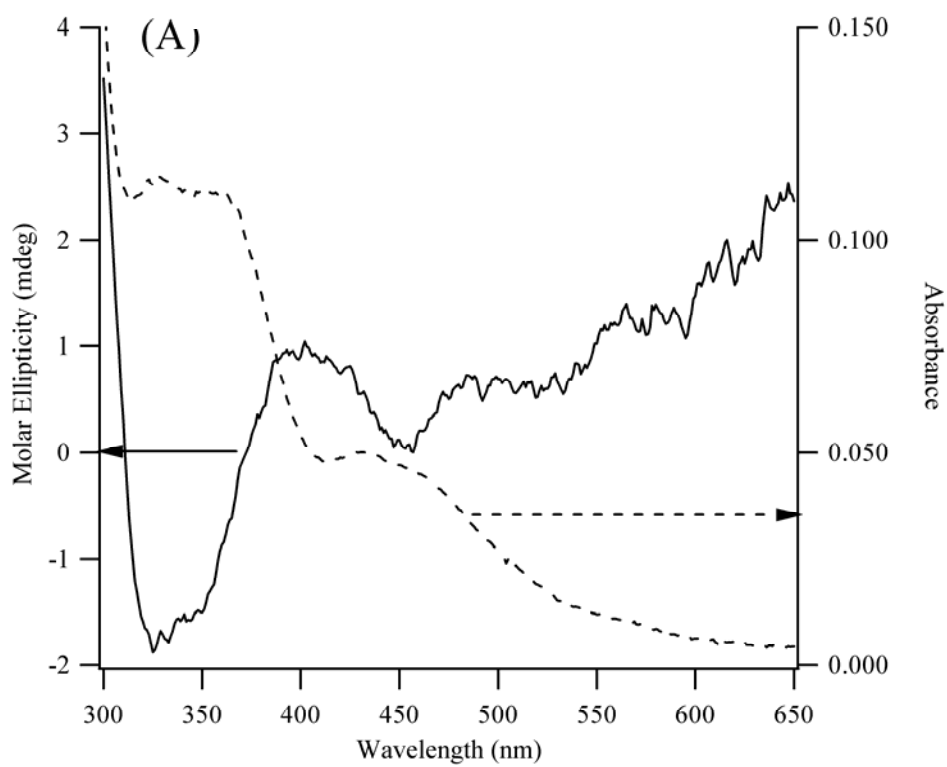


Figure 3S

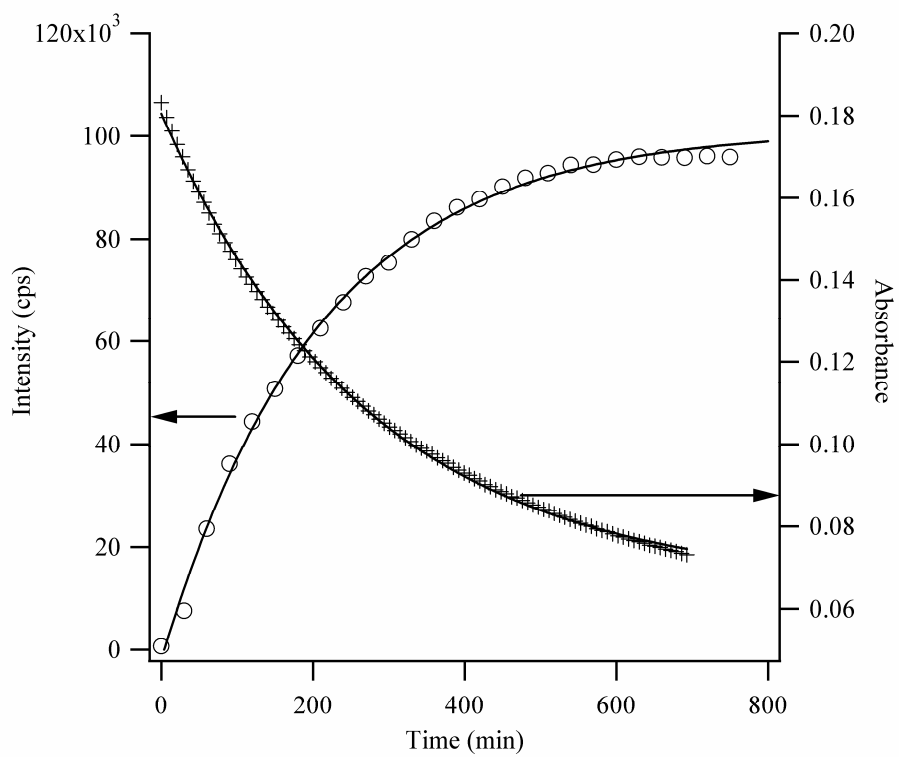


Figure 4S

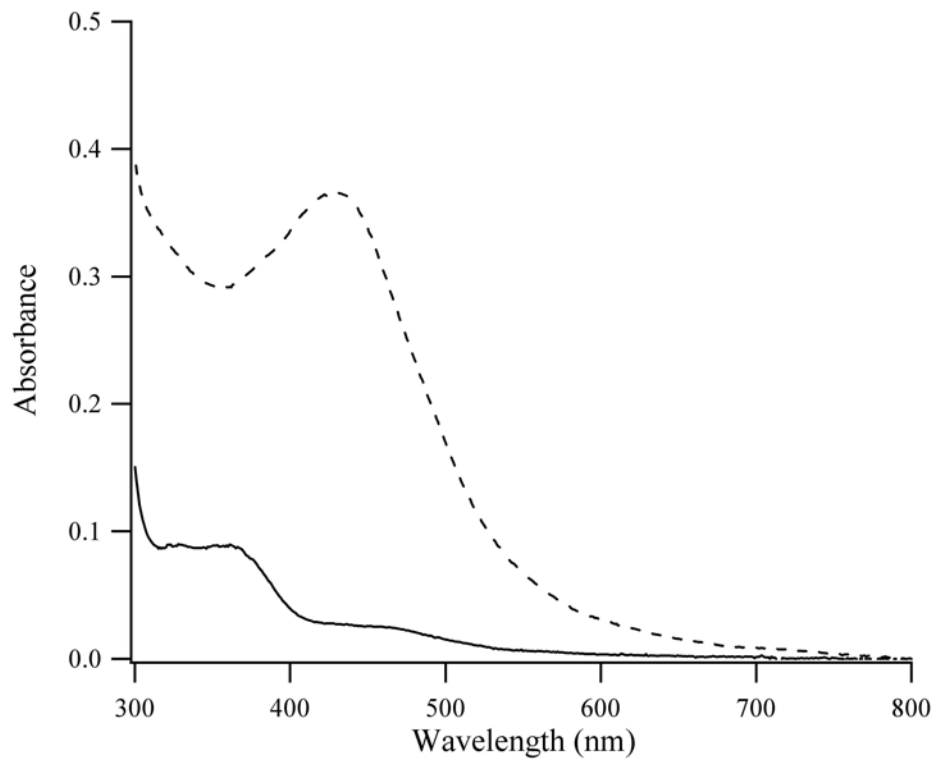


Figure 5S

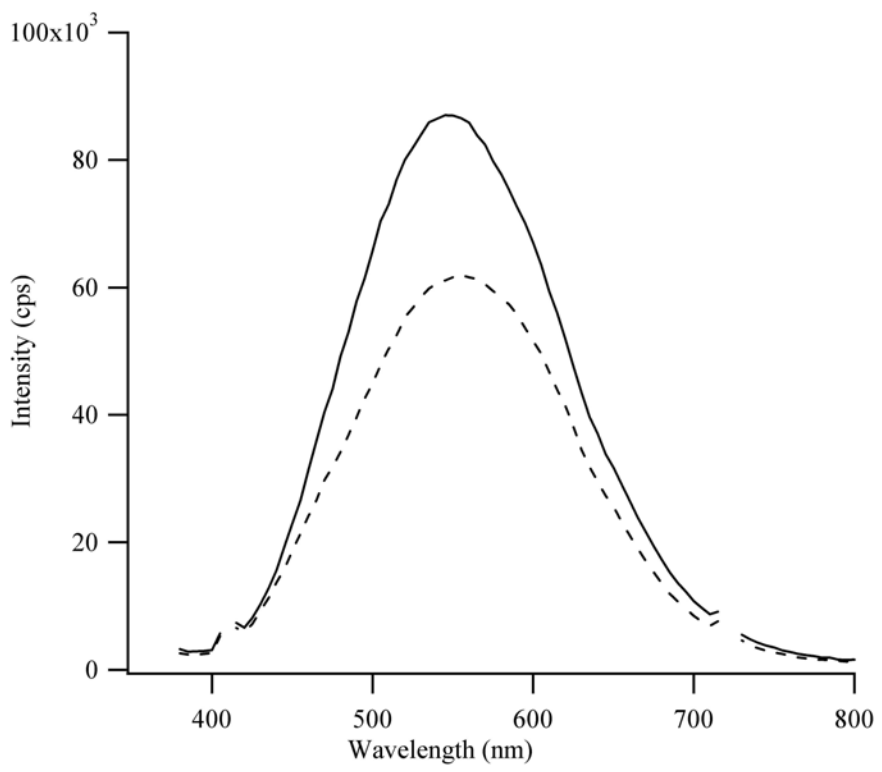


Figure 6S

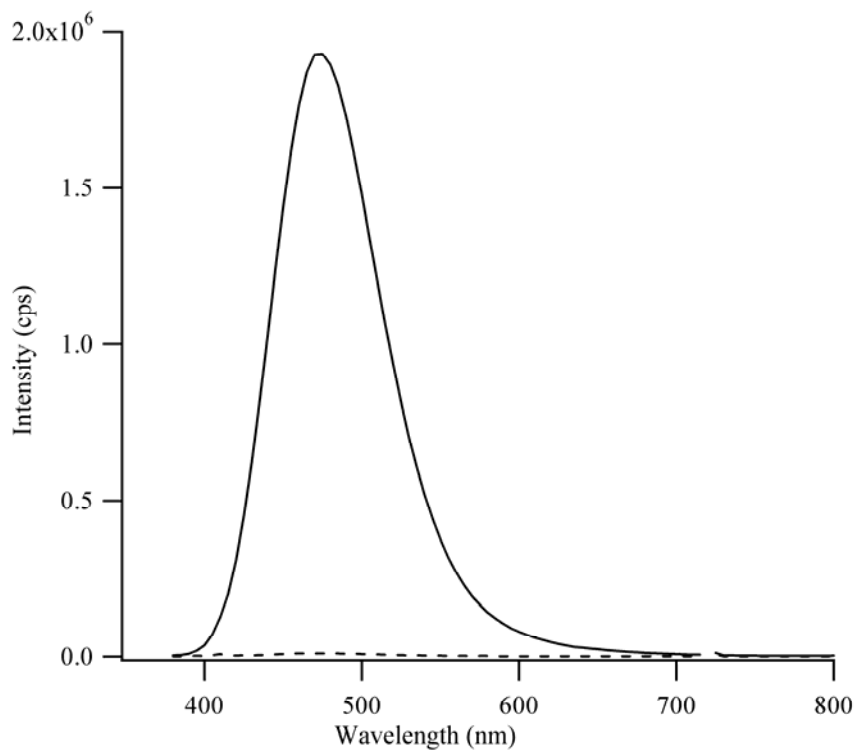


Figure 7S

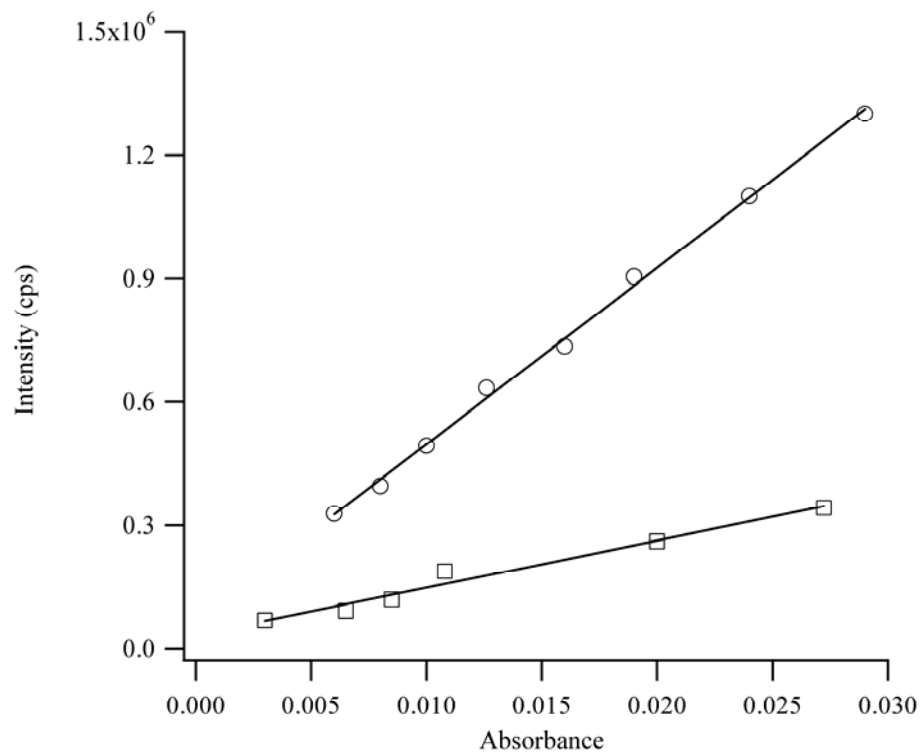


Figure 8S

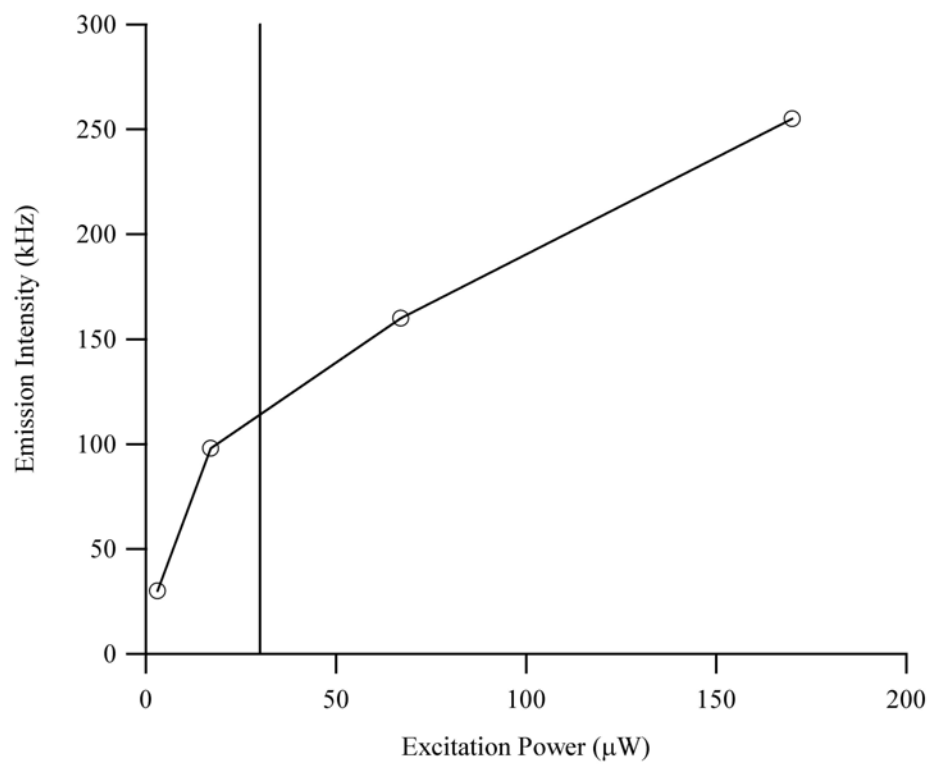


Figure 9S

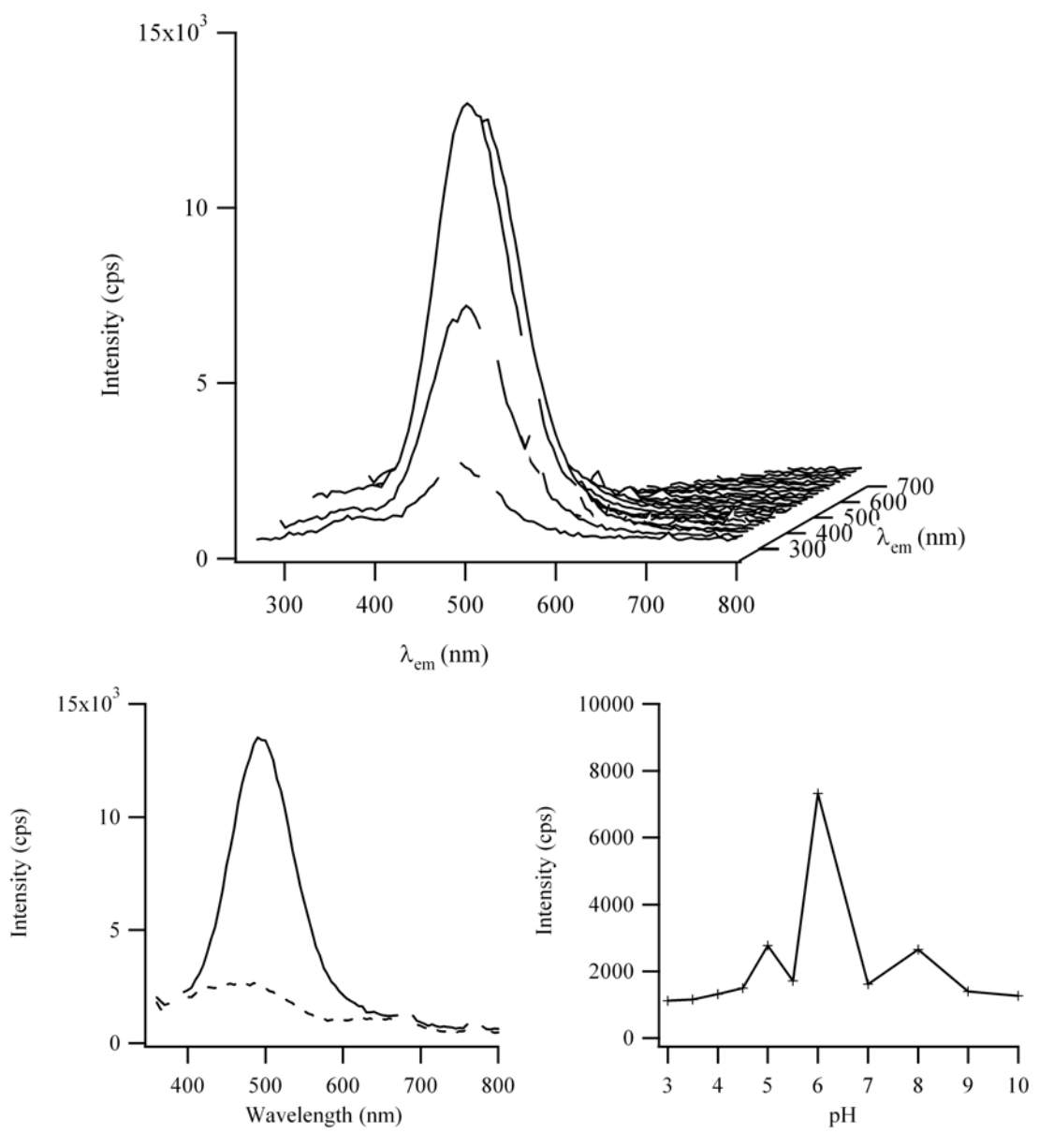


Figure 10S