SUPPLEMENTAL MATERIAL

Figure S1 shows the calculated spectral response for a mono-metallic c-BNA system comprising an Au bowtie and Au cap. While the NIR and VIS peaks follow the same spectral behavior as the bi-metallic case, the overall enhancement of electric and magnetic fields is lower.

![Fig. S1 Spectral response as a function of cap thickness for an all-Au mono-metallic c-BNA system.](image1)

Figure S2 shows the calculated spectral response for a mono-metallic c-BNA system comprising an Ag bowtie and Ag cap. Electric and magnetic field enhancement values are larger for the all-Ag c-BNAs compared to bi-metallic and mono-metallic Au c-BNAs.

![Fig. S2. Spectral responses as a function of cap thickness for an all-Ag mono-metallic c-BNA system.](image2)
Electromagnetic absorption and plasmon damping is related to the imaginary part of the dielectric function. Figure S3 shows the loss ratio for Au and Ag, defined as:

\[
\text{Loss ratio} = \frac{\text{Im}\{\varepsilon_{\text{Ag}}\}}{\text{Im}\{\varepsilon_{\text{Au}}\}}.
\]

Clearly, losses associated with an Ag cap in the c-BNAs are lower than the corresponding mono-metallic system. This leads to overall higher enhancement values in the bi-metallic system compared to the mono-metallic case.

Figure S4 shows the full set of calculated charge densities as a function of cap thickness for the NIR resonance. Evidently, increasing cap thickness increases the spatial separation of charges, leading to the blue shift in the electric and magnetic plasmon resonances. Further, the elementary charge density associated with the bowtie is unperturbed by the cap thickness.
Fig. S4 Calculated charge density distributions for the NIR resonance as a function of cap thickness.

Figure S5 shows the spectral response of bi-metallic c-BNAs ($l' = 425$ nm, $t = 20$ nm, $g = 20$ nm) as a function of increasing cap length ($l$) along the tip-to-tip axis. A blue shift is evident in the NIR mode with increasing $l$ that is due to reduced Coulomb screening between opposing charges, in accordance with the proposed hybridization model.

Fig. S5 Spectral response of bi-metallic c-BNAs as a function of increasing cap length $l$. 
Fig. S6. Spectral sensitivity of mono-metallic Au c-BNAs. Grey squares represent data for the VIS resonances whereas red squares represent NIR results. Linear fits are included as black lines.

Fig. S7. Absorption cross sections of bi-metallic and mono-metallic c-BNAs ($\Gamma = 425$ nm, $t = 20$ nm, $g = 20$ nm).