Supplemental Material for: Polarization-sensitive photoluminescence from aligned carbon chains terminated by gold clusters

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S1. PHOTOLUMINESCENCE DEPENDENCE ON ROTATION ANGLE FOR SAMPLES SYNTHESIZED WITHOUT GOLD

To investigate the dependence of the photoluminescence of a gold-free, non-aligned carbynes sample on the polarization of the optical excitation, we rotated the polarization plane of the linearly-polarized incident light ray, which was normal to the sample plane, using a half-wavelength plate. It can be seen from Fig. S1 that the colloidal sample synthesized without gold nanoparticles exhibits no discernible dependence of photoluminescence on the polarization angle. It should also be noted that the integral photoluminescence intensity in this experiment is over an order of magnitude less than that observed with samples synthesized in the presence of gold (see Fig. 3 of the main text) for the same pumping power and concentration of carbon atoms.

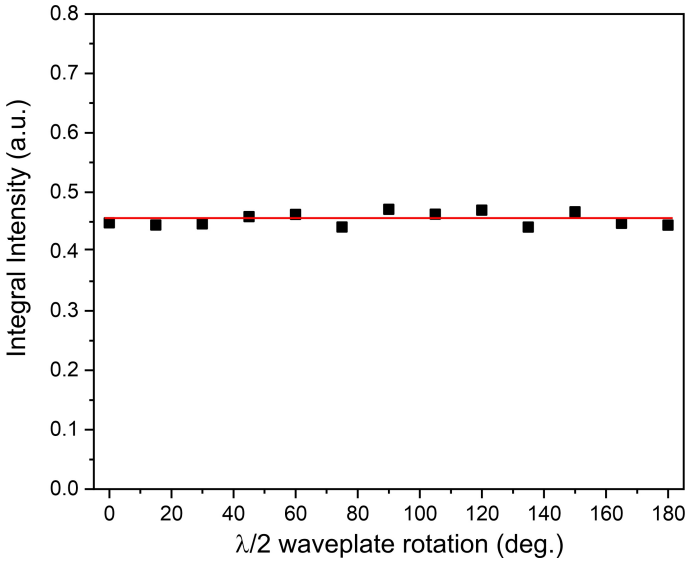

FIG. S1. Photoluminescence dependence on the rotation angle of the half-wavelength plate for the colloidal sample synthesized without gold nanoparticles. The laser excitation wavelength used was 375 nm, at a power of 15 mW.

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S2. ESTIMATE FOR CHAIN LENGTH

In this section, we provide a simple estimate for the inter-level spacing in a finite carbyne chain. We then use this estimate to determine the minimum chain length for the blue shift to be observed when the excitation of the film is at the gold plasmon frequency. Utilizing the nearest-neighbor tight-binding model, the energy spectrum of an infinite carbyne chain is expressed by the following equation:

$$E = \pm\sqrt{(\alpha-\beta)^2 + 4\alpha\beta\cos^2\left(\frac{kc}{2}\right)}, \tag{S1}$$

where α and β are the transfer integrals associated with the short (triple) and long (single) covalent bonds, respectively, k is the the wave vector along the chain, and c is the lattice constant. From Eq. (S1), the difference in energy between the top of the conduction band and the bottom is $2\,|\beta|$. A finite chain, composed of N atoms, with a configuration given in Ref. 6 of the main text, possesses N discrete energy levels. Two of these levels (edge states) are within the band gap of the infinite chain, and $N/2-1$ are in the conduction band. Therefore, the typical level spacing in the conduction band is approximately $2|\beta|/(N/2-1) \approx 4|\beta|/N$. Transitions between states of the same parity are forbidden, see Fig. S2. Therefore, there must be a minimum of three carbyne energy levels within the excitation photon energy for the blue shift to be observed, leading to the condition that $12|\beta|/N < h\nu$.

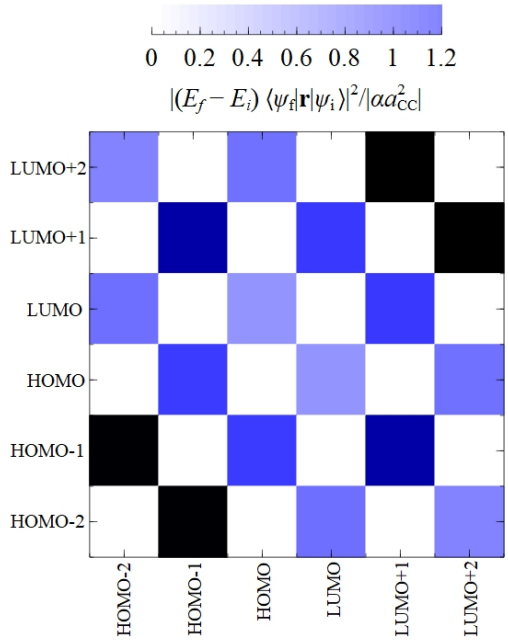

FIG. S2. The absolute value of the oscillator strength, normalized by a dimensionless constant, for inter-level transitions in a chain composed of $N = 40$ atoms. The tight-binding parameters are $\alpha = -4.657$ eV and $\beta = -3.548$ eV, with on-site energies of 0.1 eV for the end atoms.