Electronic Supplementary Information for

# Triplet Exciton Dissociation and Electron Extraction in Graphene-Templated Pentacene Observed with Ultrafast Spectroscopy

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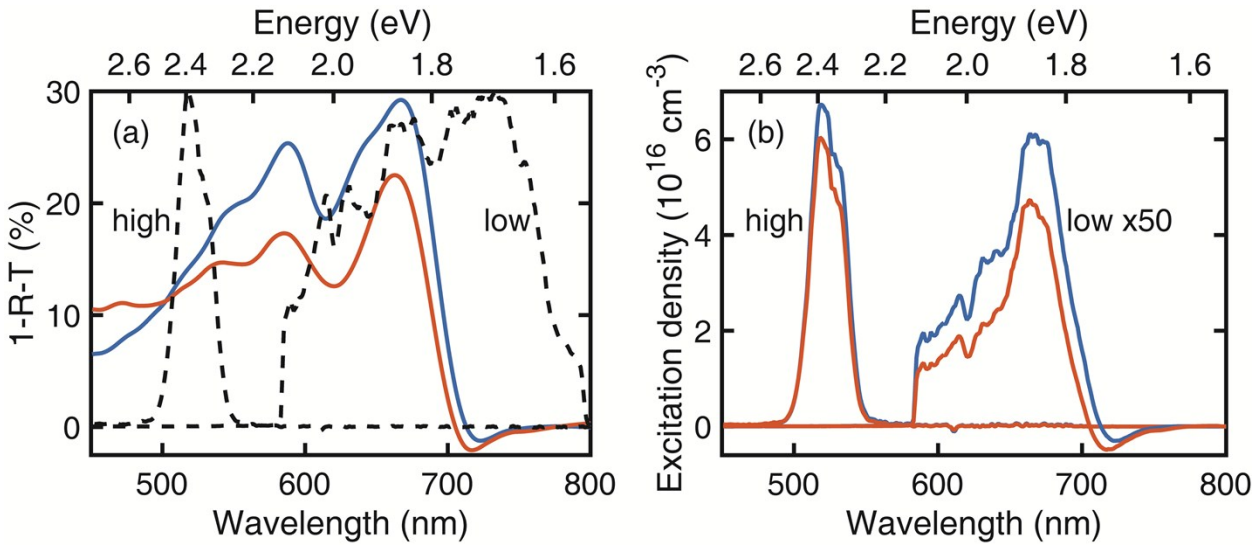


**Fig. S1** (a) Percent absorption for pentacene on glass (blue) and graphene (red) with pump spectra for the high and low fluence measurements (dashed black lines). (b) Excitation density vs. wavelength for the high (50 μJ/cm$^2$) and low (2 μJ/cm$^2$) fluence measurements accounting for pump fluence, percent absorption, and sample thickness (100 nm). The low fluence curves are scaled by a factor of 50 for visualization. The excitation densities reported in the text are the integrated areas of these curves.

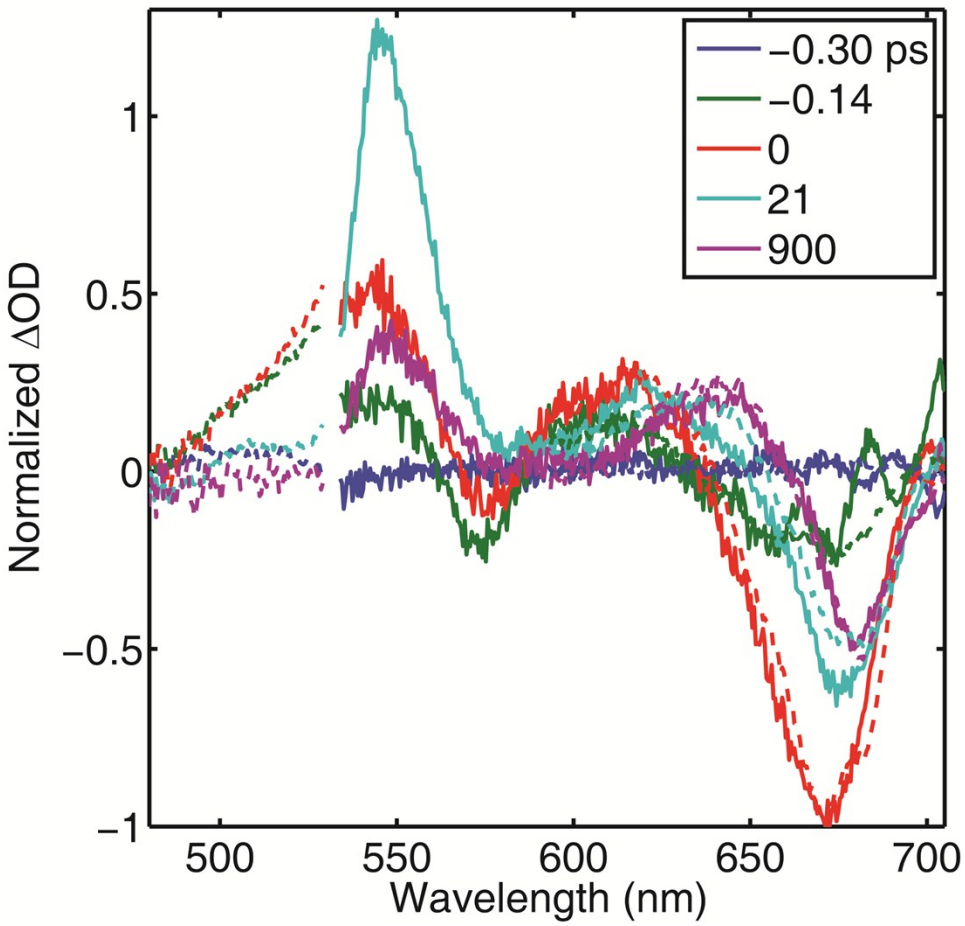


**Fig. S2** Transient absorption spectra for pentacene on graphene with excitation at 505 nm (solid lines) and 565 nm (dashed lines). The fluence was 35 μJ/cm$^2$ for both wavelengths.

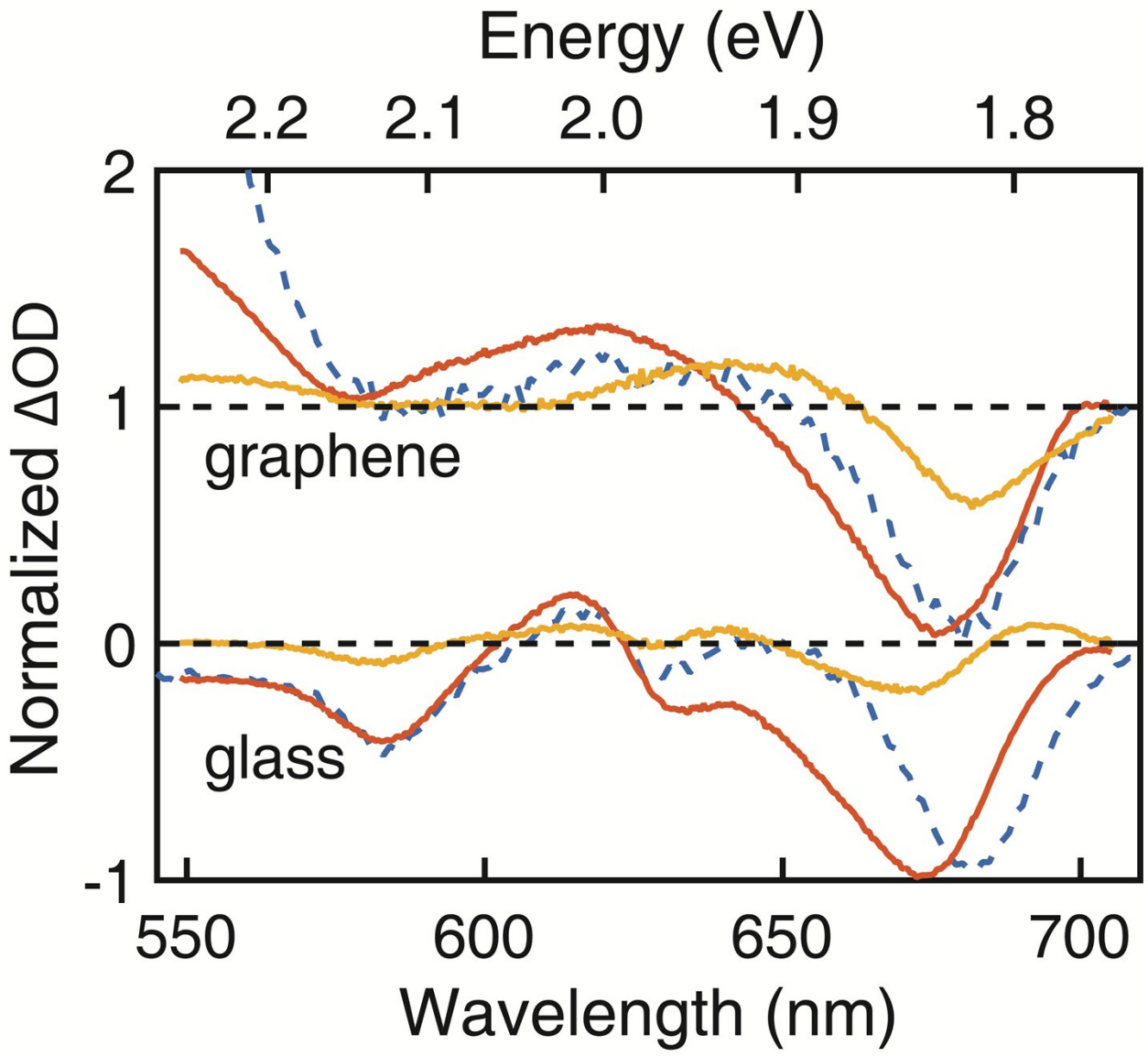


**Fig. S3** Transient absorption spectra for pentacene on glass (bottom) and graphene (top) at low fluence, T = 3.3 ps (blue dashed lines); high fluence, T = 0 ps (red solid lines); and high fluence, T = 900 ps (orange solid lines).

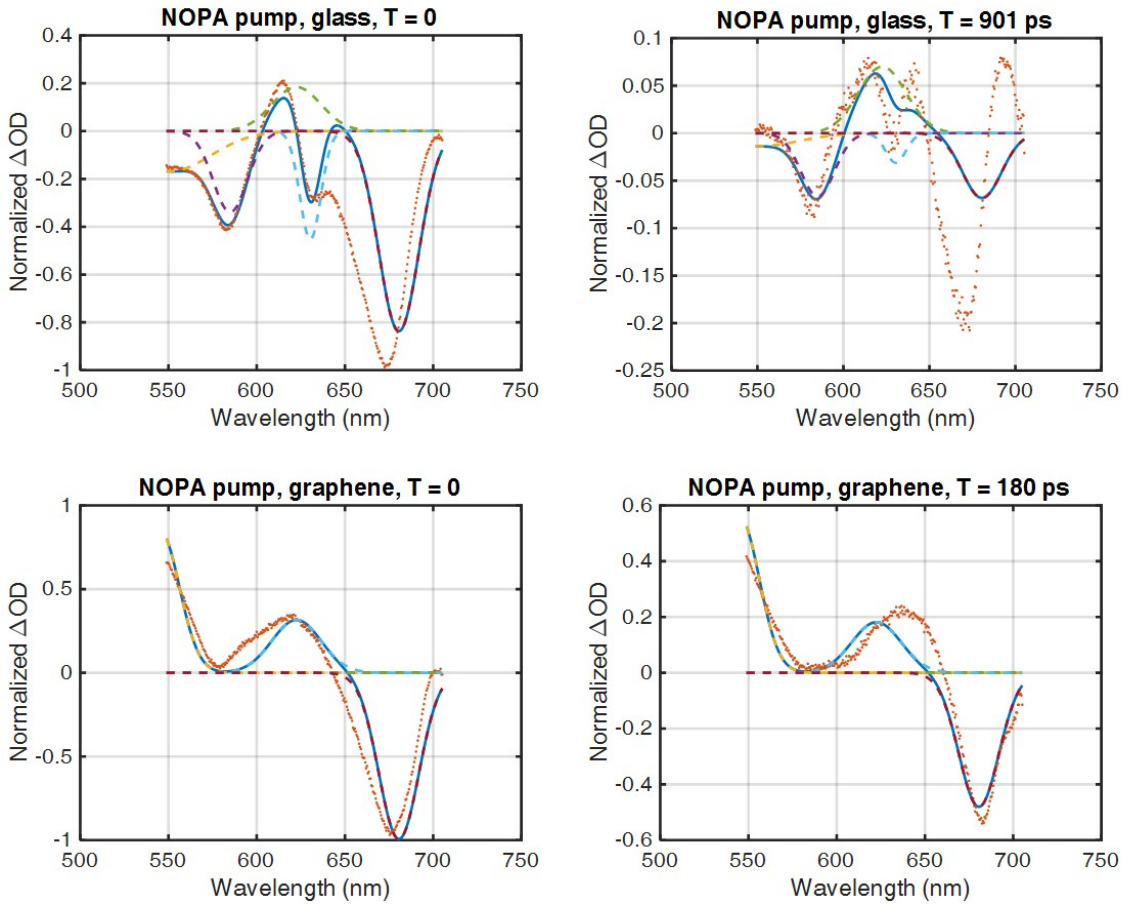


**Fig. S4** Experimental transient absorption spectra (red dots), fits (solid blue lines), and component Gaussians (dashed lines) using only the Gaussians obtained from fits to the low fluence data (see Fig. 4 and Table 1 in the main text). These fits clearly miss the additional spectral features observed at high fluence.

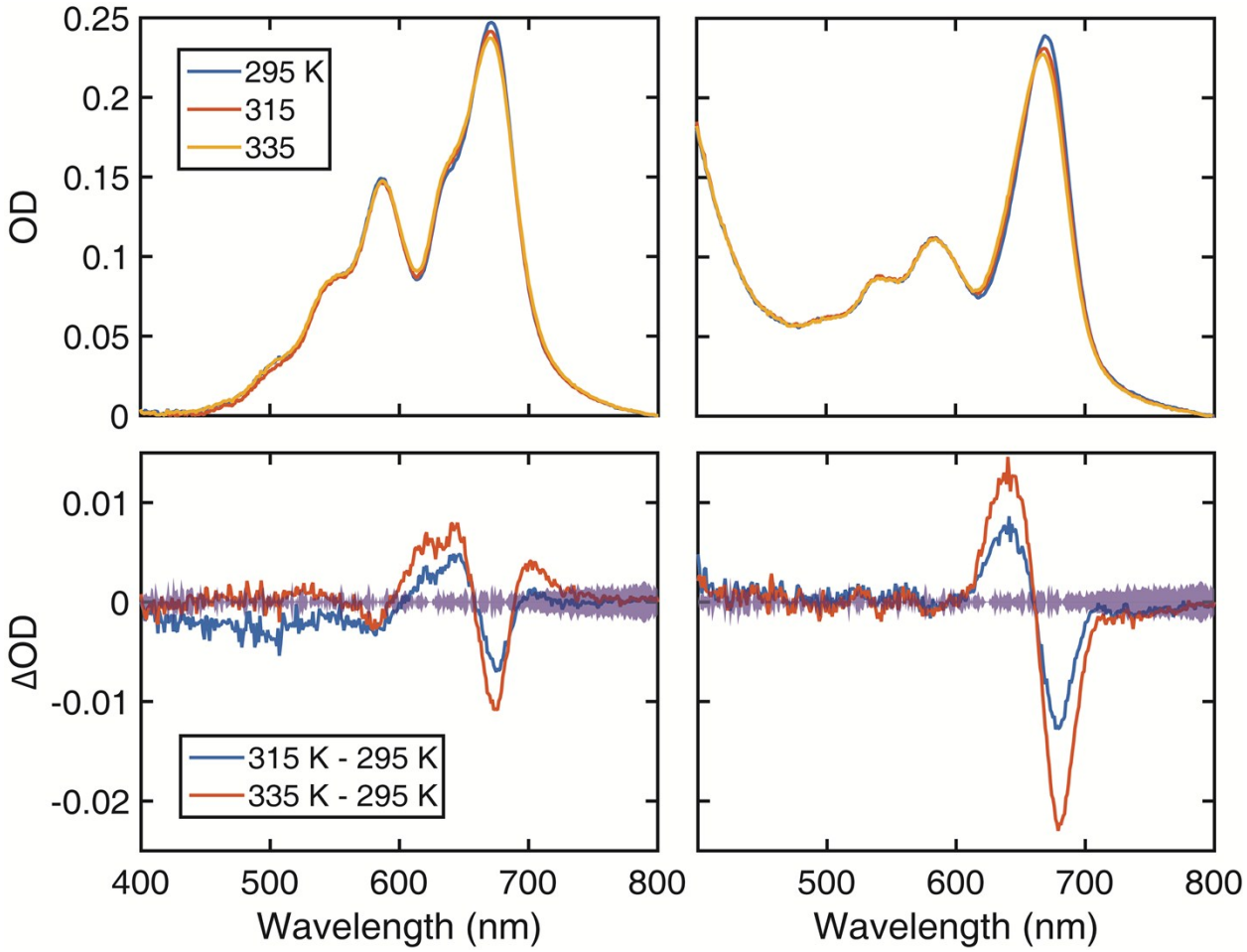


**Fig. S5** Temperature-dependent absorption spectra of pentacene on (top left) glass and (top right) graphene for 295 (blue), 315 (red), and 335 K (orange). We assumed that the absorbance at 800 nm should be zero, so the absorbance value at this wavelength was subtracted as an offset across the entire spectrum to obtain the data shown. Difference spectra between 315 and 295 K (blue) and 335 and 295 K (red) for pentacene on (bottom left) glass and (bottom right) graphene. The noise level due to the drift in the lamp intensity over the course of the measurement (a few hours) is shown in purple. We utilize the difference spectra between 335 and 295 K in our fits shown in the main text.

**Temperature-dependent absorption method**

Absorption of pentacene on glass/graphene was measured as a function of temperature using an optical cryostat (Janis ST-100) equipped with a sample heater. Samples were placed in the cryostat and evacuated to a pressure of $10^{-5}$ Torr. A beam of monochromatic (Horiba microHR) light was chopped and sent through a beam splitter. One beam was sent through the sample and collected by a calibrated silicon photodiode. The other beam was monitored by a second calibrated silicon photodiode to track changes in the light source intensity over the course of the experiment. The sample heater was used to heat the sample to 315 and 335 K. The sample was allowed to equilibrate for 15 minutes at each temperature. The absorption spectrum of the sample was measured by stepping through the wavelength range (400-800 nm) in 1 nm increments.

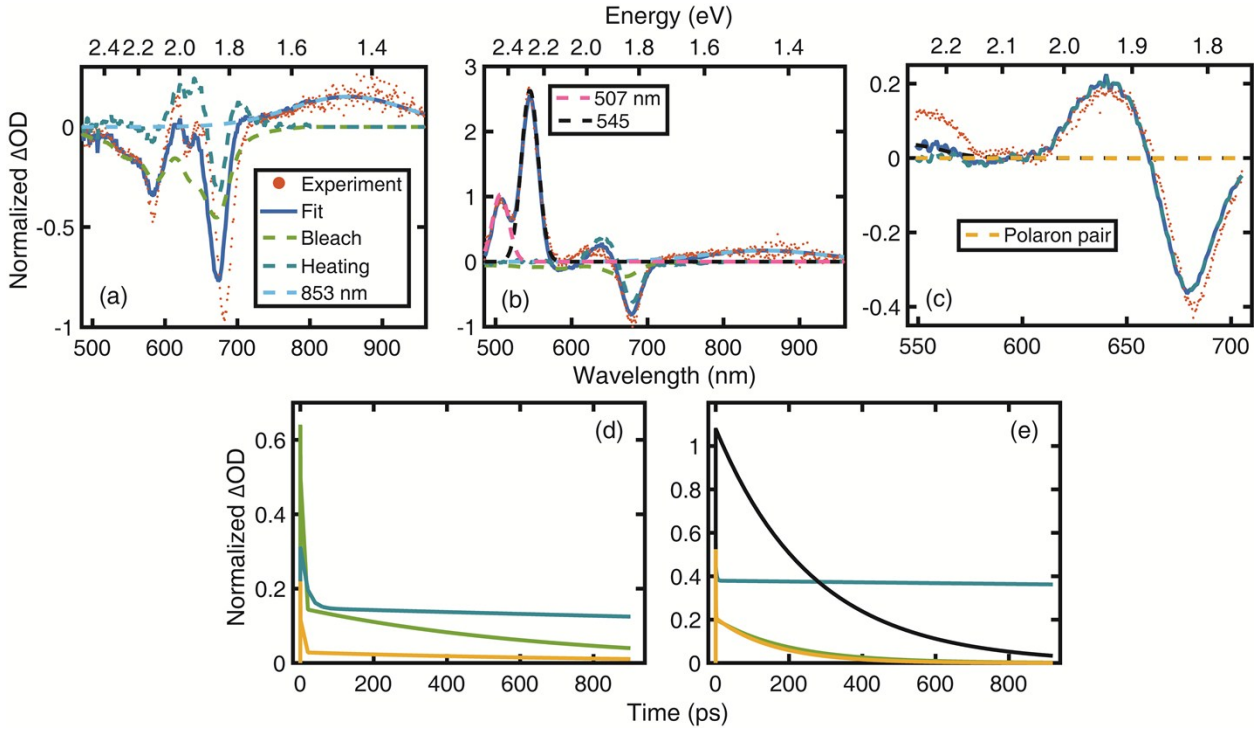

**Fig. S6** Transient absorption spectra (red dots) and corresponding fits (blue solid lines) with select spectral components (dashed lines). Pentacene on (a) glass and (b) graphene at low fluence (T = 3.3 ps) fit with bleaching (green), transient heating (teal), 853 nm Gaussian (light blue), and triplet (507 nm – pink, 545 nm – black; graphene only) components. (c) High fluence transient absorption spectrum on graphene (T = 900 ps) fit with bleaching, transient heating, polaron pair (orange), and triplet components. High fluence component kinetics from transient heating fits for pentacene on (d) glass and (e) graphene. The colors of the traces in (d,e) match the colors of the associated spectral components in (a-c).