

Supporting Information

Third-Order Nonlinear Optical Properties and Electroabsorption Spectra of an Organic Biradical, [Naphtho[2,1-*d*:6,5-*d'*]bis([1,2,3]dithiazole)]

Keigo Takauji,[†] Rie Suizu,[†] Kunio Awaga,[†] Hideo Kishida,[‡] and Arao Nakamura[‡]

[†]*Department of Chemistry and Research Center for Materials Science, Nagoya University,*

Furo-cho, Chikusa, Nagoya 464-8602 Japan

[‡]*Department of Applied Physics, Nagoya University, Furo-cho, Chikusa, Nagoya 464-8603 Japan*

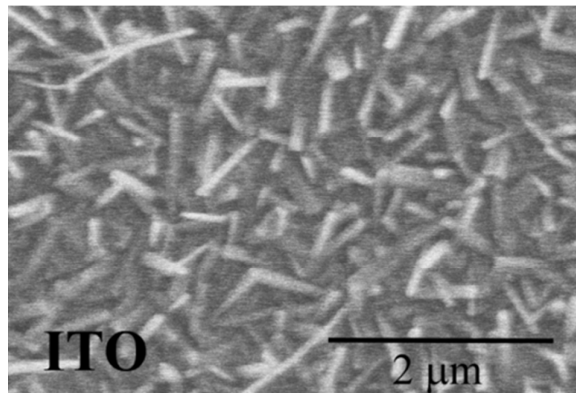


Figure S1. SEM image of the NT thin film.

SEM image of the NT thin film on an ITO substrate.

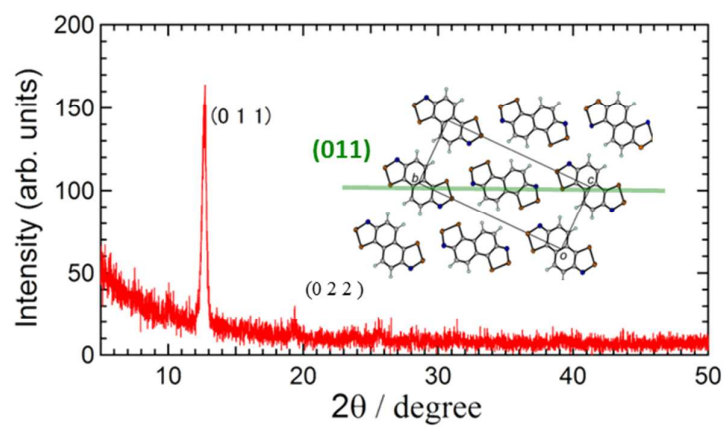


Figure S2. XRD measurement of the NT thin film.

(0 1 1) and (0 2 2) peaks vested in (0 t t) show that the NT molecules are oriented with the π electron plane perpendicular to the substrate.

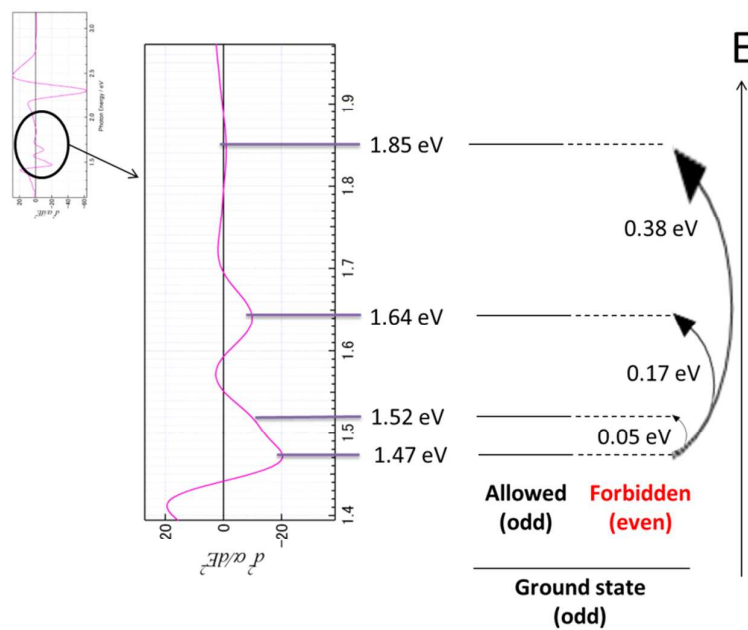


Figure S3. Electronic structure of NT.

Electronic structure of NT films under the assumption of degeneracy between the forbidden and allowed levels derived from the second differential of the absorption spectrum at 4 K.

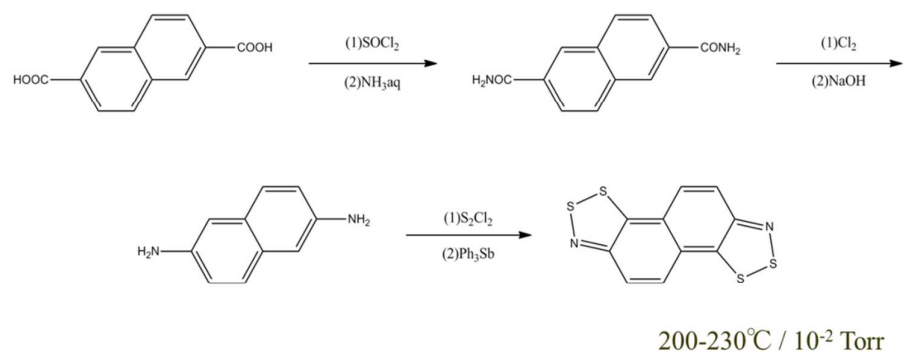


Figure S4. Synthesis.

Detail of the NT preparation method is described in Oakley's paper.²⁵ NT is deposited on the substrate after purification by sublimation (200-230 °C / 10^{-2} Torr).

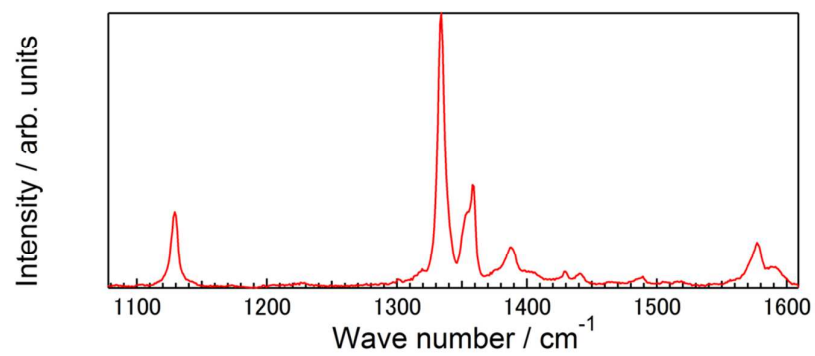


Figure S5. Raman spectrum of an NT film.

The wavelength of excitation laser is 633 nm.

energy (eV)		damping parameter (eV)		transition matrix element ($\text{\AA}$)	
$\hbar\omega_1$	1.47	$\hbar\gamma_1$	0.10	$\langle 0 x 1\rangle$	0.20
$\hbar\omega_2$	1.47	$\hbar\gamma_2$	0.050	$\langle 1 x 2\rangle$	0.90
$\hbar\omega_3$	1.64	$\hbar\gamma_3$	0.080	$\langle 0 x 3\rangle$	0.20
$\hbar\omega_4$	1.64	$\hbar\gamma_4$	0.060	$\langle 3 x 4\rangle$	0.60
$\hbar\omega_5$	1.85	$\hbar\gamma_5$	0.14	$\langle 0 x 5\rangle$	1.0
$\hbar\omega_6$	1.85	$\hbar\gamma_6$	0.10	$\langle 5 x 6\rangle$	0.20

Table S1. Parameters of the three-level-model used for the calculation of EA spectra.