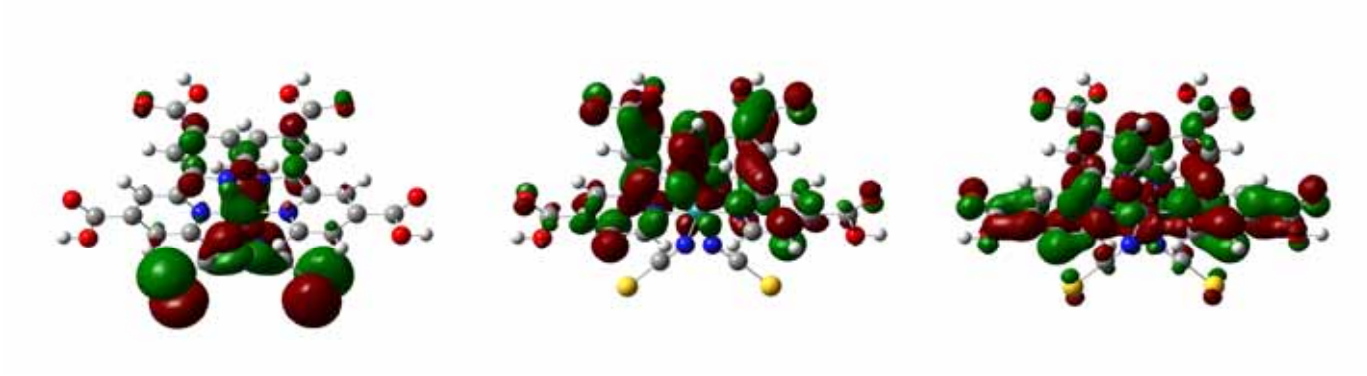


HOMO LUMO LUMO+1

Fig. 1S. Molecular orbitals of **N3** (dft/b3lyp/cep-4g, isodensity value = 0.02).

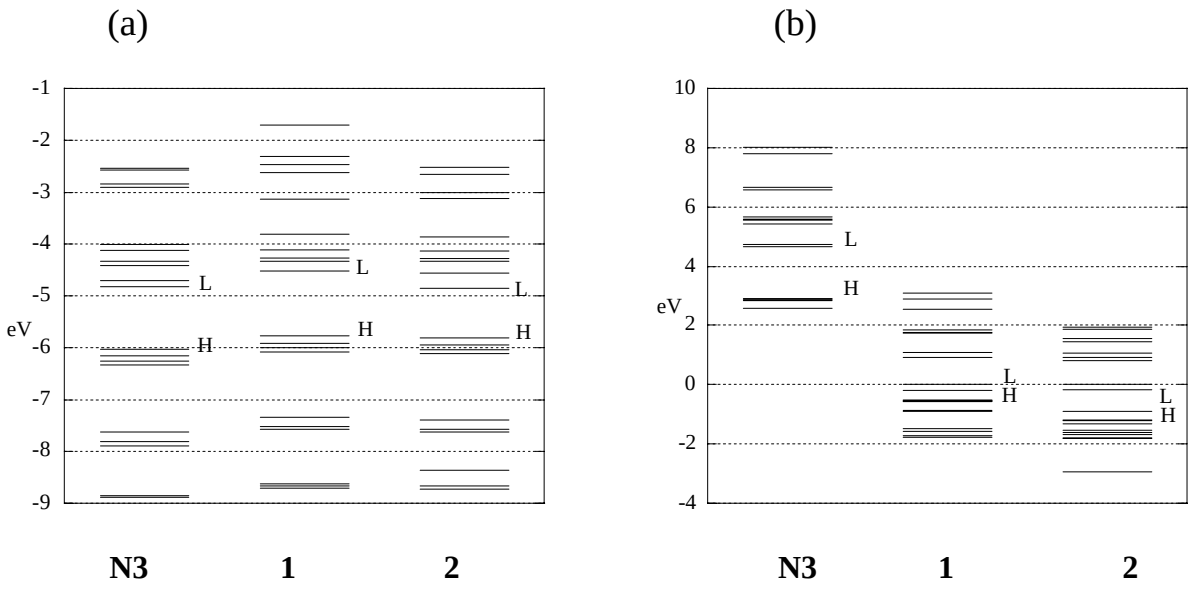
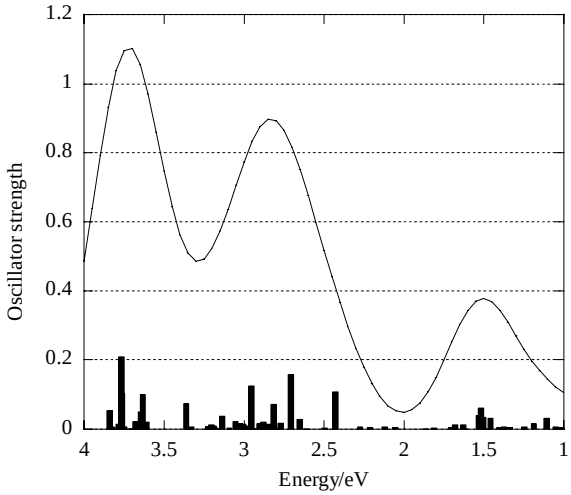
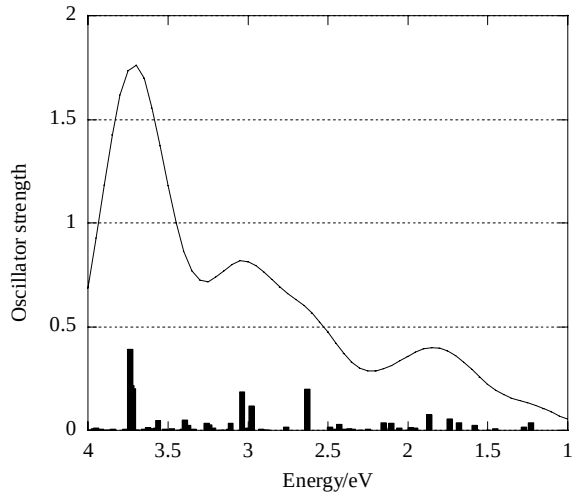


Fig. 2S. Energy levels (eV) of **N3**, **1,** and **2** (a) and deprotonated **N3**, **1,** and **2** in vacuo (b) Abbreviations: L, LUMO; H, HOMO.

(a) **N3** in vacuo



(b) **N3** in CH_3CN



(c) Deprotonated **N3** in vacuo

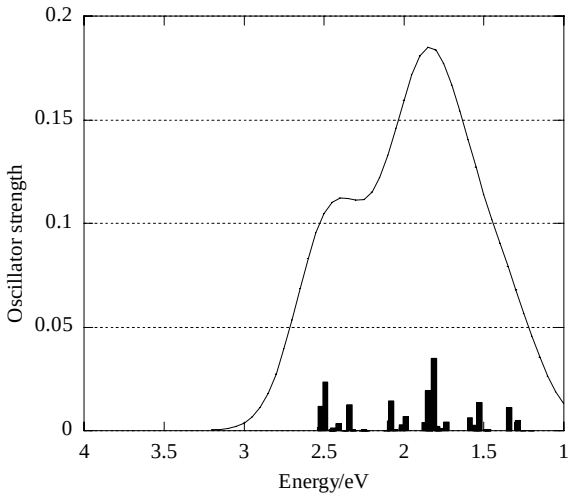
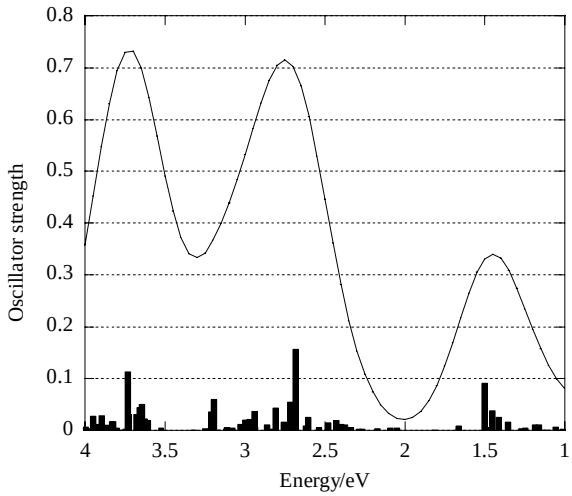
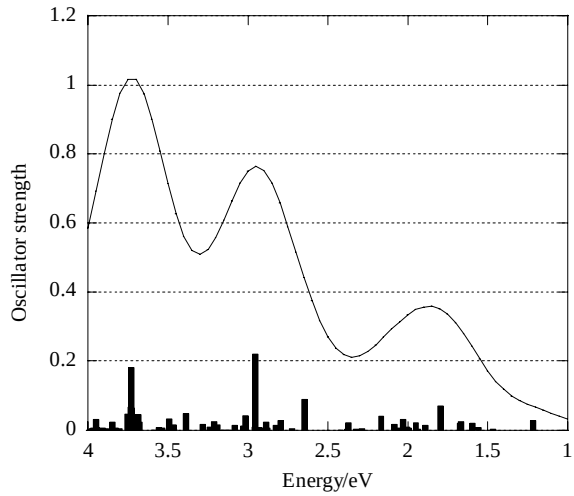


Fig. 3S. Calculated electronic spectra of **N3**: (a) **N3** in vacuo, (b) **N3** in CH_3CN, (c) deprotonated **N3** in vacuo. Vertical lines correspond to calculated excitation energies and oscillator strengths. The calculated spectra have been obtained by a gaussian convolution with σ=0.20 and 0.1eV for the transitions below and above 4.0 eV, respectively.

(a) **1** in vacuo



(b) **1** in CH_3CN



(c) Deprotonated **1** in vacuo

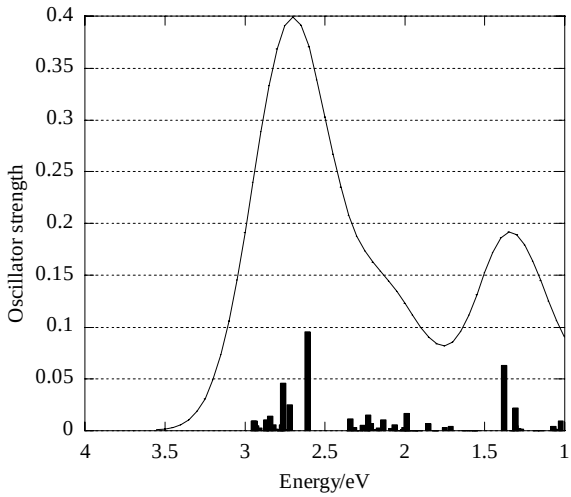
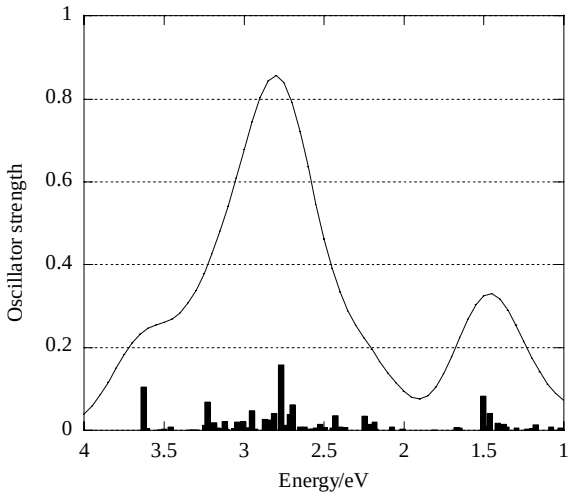
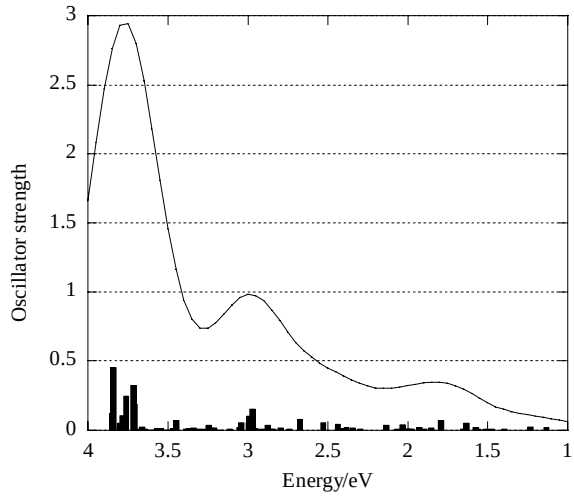


Fig. 4S. Calculated electronic spectra of **1**: (a) **1** in vacuo, (b) **1** in CH_3CN, (c) deprotonated **1** in vacuo. Vertical lines correspond to calculated excitation energies and oscillator strengths. The calculated spectra have been obtained by a gaussian convolution with σ=0.20 and 0.1eV for the transitions below and above 4.0 eV, respectively.

(a) **2** in vacuo



(b) **2** in CH_3CN



(c) Deprotonated **2** in vacuo

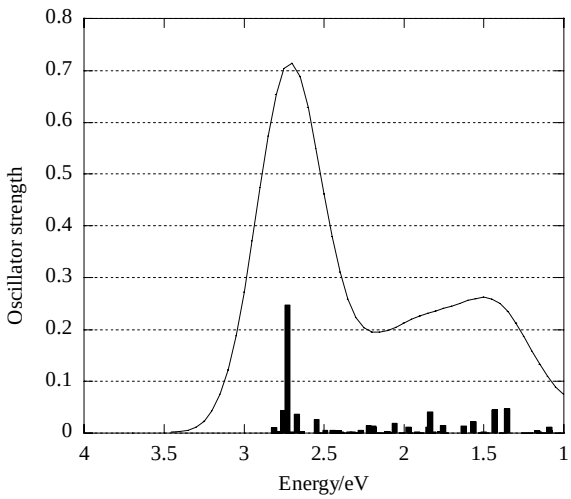


Fig. 5S. Calculated electronic spectra of **2**: (a) **2** in vacuo, (b) **2** in CH_3CN, (c) deprotonated **2** in vacuo. Vertical lines correspond to calculated excitation energies and oscillator strengths. The calculated spectra have been obtained by a gaussian convolution with σ=0.20 and 0.1eV for the transitions below and above 4.0 eV, respectively.