Supporting Information for

Two Dimensional Restriction-Induced Luminescence of Tetraphenyl Ethylene within the Layered Double Hydroxides Ultrathin Films and Its Fluorescence Resonance Energy Transfer

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Figure S1 (a) UV/vis absorption spectra of (PVK/LDH)$_n$ ($n = 3, 6, 9, 12$) UTFs. The inset shows linear relationship between the absorbance at 243, 296, and 345 nm and number of bilayers, $n$. (b) PL emission spectra for (PVK/LDH)$_n$ UTFs and (TPE@PVK/LDH)$_n$ UTFs, $n =3, 6, 9, 12$, for excitation at 293 nm.
Figure S2 The SEM and tapping-mode AFM images on top view of (TPE@PVK/LDH)$_n$ UTFs for $n = 10, 15$

Table S1. The roughness data for the UTF-$n = 5, 10, 15, 20$

<table>
<thead>
<tr>
<th>$N$</th>
<th>5</th>
<th>10</th>
<th>15</th>
<th>20</th>
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<tbody>
<tr>
<td>rms roughness (nm) $^{(a)}$</td>
<td>1.647</td>
<td>2.365</td>
<td>3.210</td>
<td>5.805</td>
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$^{(a)}$ The statistical rms roughness values were obtained by AFM.
**Figure S3.** The fluorescence decay curves of (a) (PVK/LDH)$_{30}$ UTF, (b) amorphous spin-coating film of TPE; (c) UTF-30 at 380 nm; (d) UTF-30 at 475 nm, respectively, in all of which the blank curves were plotted as the green ones.
Figure S4 The PL excitation and emission spectra of TPE powder (solid line), and PVK toluene solution (dotted line).

A spin-coating films of PVK@TPE (5wt%) without LDH nanoheats was fabricated, which can hardly reversibly respond to VOC atmosphere as shown in Figure S5.

Figure S5. (a) PL emission spectra of the (TPE@PVK) spin-coating film (TPE / PVK = 5 wt %) in the in the dry air (black curve), or in the THF(a); toluene(b); and acetone atmosphere (c) (blue curve), or again in the dry air (red curve) with 293 nm excitation.
In an air-tight quartz cell, the UTFs were exposed to various VOC vapors (acetone, toluene, chloroform, tetrahydrofuran (THF)) to undergo the VOC exposure cycles measurement. The UTFs were checked by fluorescence spectroscopy immediately, when exposing in the VOC vapor or in the dry air for 3 min each, and repeated for several times incessantly. That is, the blue fluorescence emission of TPE with 293 nm excitation can be quenched/recovered (off/on) by the adsorption/desorption of VOC molecules. The FRET behavior disappeared or recovered when exposing the UTFs-n to the common VOCs vapor or in the dry air.

**Figure S6.** (a) PL emission spectra of the (TPE@PVK/LDH)\textsubscript{20} UTF (TPE / PVK = 5 wt %) in the toluene vapor atmosphere (red curve) or in the dry air (black curve) with 293 nm excitation. (b) The reversible fluorescence response over five consecutive cycles.
Figure S7. (a) PL emission spectra of the (TPE@PVK/LDH)$_{20}$ UTF (TPE / PVK = 5 wt %) in the acetone atmosphere (red curve) or in the dry air (black curve) with 293 nm excitation. (b) The reversible fluorescence response over five consecutive cycles.