Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting Information

Trap-Mediated Photogating in Hybrid Organic–Inorganic Heterojunction Phototransistors for Photo-Memory and Photodetection

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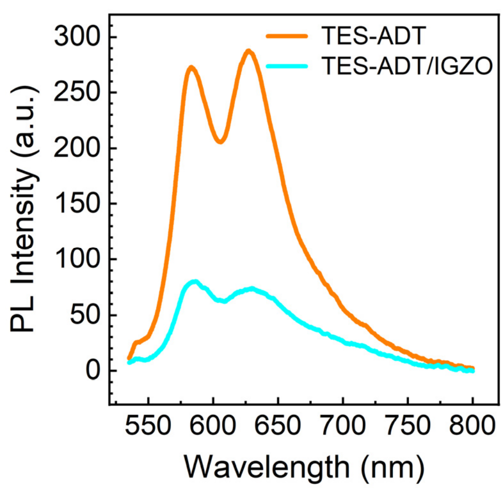

Figure S1. Steady-state photoluminescence (PL) spectra of pristine TES-ADT and TES-ADT/IGZO heterojunction films. The pronounced PL quenching in the heterojunction indicates the presence of a nonradiative decay pathway due to interfacial charge transfer.

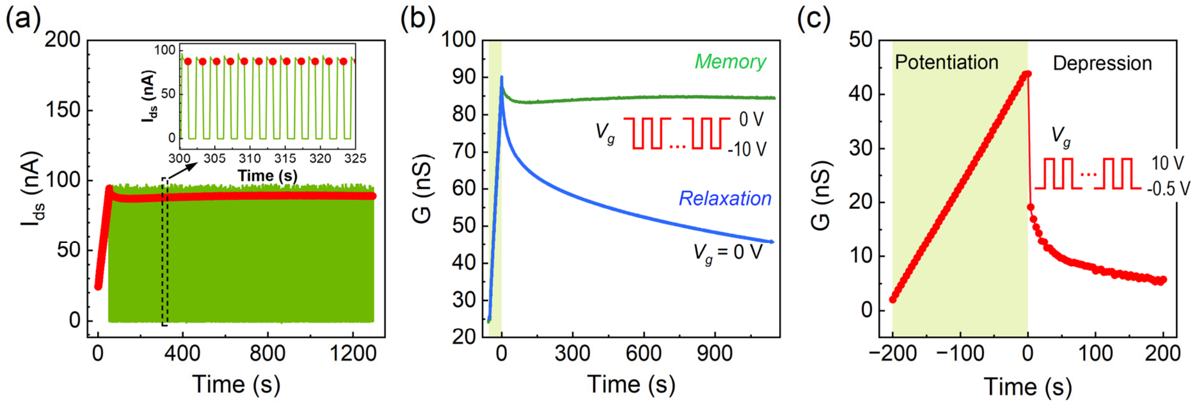

Figure S2. (a) Raw (green) and sampled (red dots) channel conductance traces of the TIPS-Tc/IGZO phototransistor operated in memory mode. Negative gate pulses alternating between −10 and 0 V (1 s each) were applied, with conductance sampled once per cycle at $V_g = 0$ V during the neutral interval. (b) Sampled channel conductance evolution under negative periodic gate pulsing (memory mode), and constant $V_g = 0$ V (relaxation mode). (c) Sampled conductance under positive gate pulsing (–0.5 V to +10 V) corresponding to the forgetting mode. All gate pulses use a 1 s width and 2 s period, and conductance is sampled during the low-bias interval (0 V or -0.5 V).

S2. Derivation of the Biexponential Trap-Dynamics Model

S2.1. Population Dynamics and Continuity Equations

Photogenerated holes in the organic layer exist as either free or trapped charges. The total hole density is

$$p_0(t) = p(t) + T(t) + M(t).$$

where $p(t)$ is the free-hole density, $T(t)$ and $M(t)$ are the shallow and deep trapped-hole populations, respectively. The temporal evolution of the free hole population follows:

$$\frac{dp(t)}{dt} = g(t) - \frac{p(t)}{\tau_R} - \gamma_T p(t)[T_0 - T(t)] - \gamma_M p(t)[M_0 - M(t)] + \beta_T T(t) + \beta_M M(t) \qquad (S1)$$

where $g(t)$ is the volumetric photogeneration rate, τ_R is the free carrier recombination lifetime, $\gamma_{T,M}$ and $\beta_{T,M}$ are the trapping and detrapping rate coefficients, and T_0, M_0 the total available shallow and deep trap densities, respectively. The trapped hole population follows its own continuity relations:

$$\frac{dT(t)}{dt} = \gamma_T p(t)\big(T_0 - T(t)\big) - \beta_T T(t), \qquad \frac{dM(t)}{dt} = \gamma_M p(t)\big(M_0 - M(t)\big) - \beta_M M(t) \qquad (S2)$$

S2.2. Light Pulse Excitation and Quasi-Steady Approximation

For a single optical pulse of duration t_p, the generation rate is given by

$$g(t) = \begin{cases} g_0, & 0 \leq t \leq t_p \\ 0, & t > t_p \end{cases}. \qquad (S3)$$

The photocarrier generation rate in organic is further described by $g(t) = \alpha P_0(t)(1-R)e^{-\alpha z}/h\upsilon$ and $\alpha = 4\pi\kappa/\lambda_{ex}$ is the absorption coefficient at the excitation wavelength λ_{ex}, κ is the imaginary part of the complex refractive index, R is the reflectivity, $P_0(t)$ is the incident optical power density, and $h\upsilon = hc/\lambda_{ex}$ is the photon energy. For sufficiently thin organic layers such that $\alpha z \ll 1$, the generation rate is reduced to $g(t) \approx \alpha P_0(t)(1-R)/h\upsilon$.

When recombination dominates over trapping or detrapping

$$\frac{1}{\tau_R} \gg \gamma_T T_0, \beta_T, \gamma_M M_0, \beta_M, \qquad (S4)$$

the dynamic of $p(t)$ decouple from those of $T(t)$ and $M(t)$, and the continuity equation for free holes reduces to

$$\frac{dp(t)}{dt} = g(t) - \frac{p(t)}{\tau_R}$$

which yields

$$p(t) = \begin{cases} g_0\tau_R\left(1 - e^{-t/\tau_R}\right), & 0 \leq t \leq t_p \\ g_0\tau_R\left(1 - e^{-t_p/\tau_R}\right)e^{-(t-t_p)/\tau_R}, & t > t_p \end{cases}.$$

Due to the fast recombination dynamics, the free carrier concentration rapidly reaches a steady-state value of $p(t) \approx g_0\tau_R$ during illumination and vanishes shortly thereafter. Therefore, over the timescales relevant to trapping and detrapping, the free hole concentration can be approximated by

$$p(t) \approx \begin{cases} g_0\tau_R, & 0 \leq t \leq t_p \\ 0, & t > t_p \end{cases}. \qquad (S5)$$

Substituting this steady-state approximation of the free hole concentration into Eq. (S2) gives

$$\frac{dT(t)}{dt} = \gamma_T g_0 \tau_R\big(T_0 - T(t)\big) - \beta_T T(t), \qquad \frac{dM(t)}{dt} = \gamma_M g_0 \tau_R\big(M_0 - M(t)\big) - \beta_M M(t). \qquad (S6)$$

Here, the product $\gamma_T g_0 \tau_R$ and $\gamma_M g_0 \tau_R$ represents the effective shallow and deep trap filling rate per trap (in units of s^{-1}) respectively under constant optical generation. We define this trap-filling rate as

$$k_T = \gamma_T g_0 \tau_R, \qquad k_M = \gamma_M g_0 \tau_R. \qquad (S7)$$

S2.3. Analytical Solution for a Single Pulse

Illumination phase ($0 \le t \le t_p$):

With this substitution and assuming an initial shallow trapped hole concentration $T(0) = T_i$, and deep trapped hole concentration $M(0) = M_i$ the solution for $T(t)$ and $M(t)$ is given by

$$T(t) = T_{ss} + (T_i - T_{ss})e^{-(k_T+\beta_T)t}, \qquad M(t) = M_{ss} + (M_i - M_{ss})e^{-(k_M+\beta_M)t}. \qquad (S8)$$

where the steady-state occupancies of shallow and deep traps are

$$T_{ss} = \frac{k_T T_0}{k_T + \beta_T}, \qquad M_{ss} = \frac{k_M M_0}{k_M + \beta_M}. \qquad (S9)$$

The equations reflect the equilibrium balance between the trapping and detrapping processes. In the early time limit, where $(k_T + \beta_T)t \ll 1$, $(k_M + \beta_M)t \ll 1$, the solutions can be approximated by their linear expansions:

$$T(t) \approx T_i + [k_T(T_0 - T_i) - \beta_T T_i]t, \qquad M(t) \approx M_i + [k_M(M_0 - M_i) - \beta_M M_i]t.$$

At $t = t_p$:

$$T(t_p) = T_{ss} + (T_i - T_{ss})e^{-(k_T+\beta_T)t_p}, \qquad M(t_p) = M_{ss} + (M_i - M_{ss})e^{-(k_M+\beta_M)t_p}. \qquad (S10)$$

Dark phase ($t > t_p$):

Since $p(t) = 0$, the trapped hole concentrations decay exponentially:

$$T(t) = T(t_p)e^{-\beta_T(t-t_p)}, \qquad M(t) = M(t_p)e^{-\beta_M(t-t_p)}. \qquad (S11)$$

Thus, the total trapped hole concentration becomes:

$$Q(t) = \begin{cases} T_{ss} + M_{ss} + (T_i - T_{ss})e^{-(k_T+\beta_T)t} + (M_i - M_{ss})e^{-(k_M+\beta_M)t_p}, & 0 \le t \le t_p \\ T(t_p)\, e^{-\beta_T(t-t_p)} + M(t_p)e^{-\beta_M(t-t_p)}, & t > t_p \end{cases}, \qquad (S12)$$

S2.4. Periodic Excitation and Recurrence Relation

Under periodic excitation with period P and light-on duration t_{on}, the hole concentration at the n-th pulse period is:

$$p(t) = \begin{cases} g_0\tau_R, & nP \le t < nP + t_{on} \\ 0, & nP + t_{on} \le t < (n+1)P \end{cases}.$$

Based on Eq (S8), the trapped hole concentrations after the illumination sub-internal of the n-th pulse cycle ($t = nP + t_{on}$) follows:

$$T(nP + t_{on}) = T_{ss} + (T_n - T_{ss})e^{-(k_T+\beta_T)t_{on}}, \quad (13a)$$

$$M(nP + t_{on}) = M_{ss} + (M_n - M_{ss})e^{-(k_M+\beta_M)t_{on}}. \quad (13b)$$

The trapped-hole concentrations during the illumination and dark sub-intervals of the n-th pulse cycle can be expressed as follows.

$$T_{n+1}(t) = \begin{cases} T_{ss} + (T_n - T_{ss})e^{-(k_T+\beta_T)(t-nP)}, & 0 \le t - nP < t_{on} \\ T(nP + t_{on})\, e^{-\beta_T(t-nP-t_{on})}, & t_{on} \le t - nP < P \end{cases}, \quad (S14a)$$

$$M_{n+1}(t) = \begin{cases} M_{ss} + (M_n - M_{ss})e^{-(k_M+\beta_M)(t-nP)}, & 0 \le t - nP < t_{on} \\ M(nP + t_{on})\, e^{-\beta_M(t-nP-t_{on})}, & t_{on} \le t - nP < P \end{cases}, \quad (S14b)$$

Defining the light- and dark-phase retention factors.

$$\eta_{on,i} = e^{-(k_i+\beta_i)t_{on}}, \qquad \eta_{on,i} = e^{-\beta_i(P-t_{on})}, \qquad i \in \{T, M\} \quad (S15)$$

and combining Eq. (S13) and the dark sub-interval solutions in Eq. (S14) yields the following recurrence relations:

$$T_{n+1} = T_n\, \eta_{on,T}\, \eta_{\text{off},T} + T_{ss}\big(1 - \eta_{on,T}\big)\eta_{\text{off},T}, \quad (S16a)$$

$$M_{n+1} = M_n\, \eta_{on,M}\, \eta_{\text{off},M} + M_{ss}\big(1 - \eta_{on,M}\big)\eta_{\text{off},M}. \quad (S16b)$$

The total trapped hole concentration Q(t) after the (n+1)-th pulse cycle is:

$$Q_{n+1} = T_{n+1} + M_{n+1}, \quad (S17)$$

The factors $\eta_{on,(T,M)}$ quantify how the previously trapped-hole populations influence the state after illumination, while $\eta_{\text{off},(T,M)}$ describe the degree of charge retention during the dark period governed by detrapping.

Through the photogating mechanism, the trapped charge $Q(t)$ modulates the IGZO channel potential and thereby the drain current:

$$I_{ds}(t) = f[Q(t)]$$

allowing quantitative extraction of parameters $k_T,\ k_M, \beta_T, \beta_M, T_0, M_0$ by fitting the measured $I_{ds}(t)$ transients to Eqs. (S12) and (S16)–(S17).

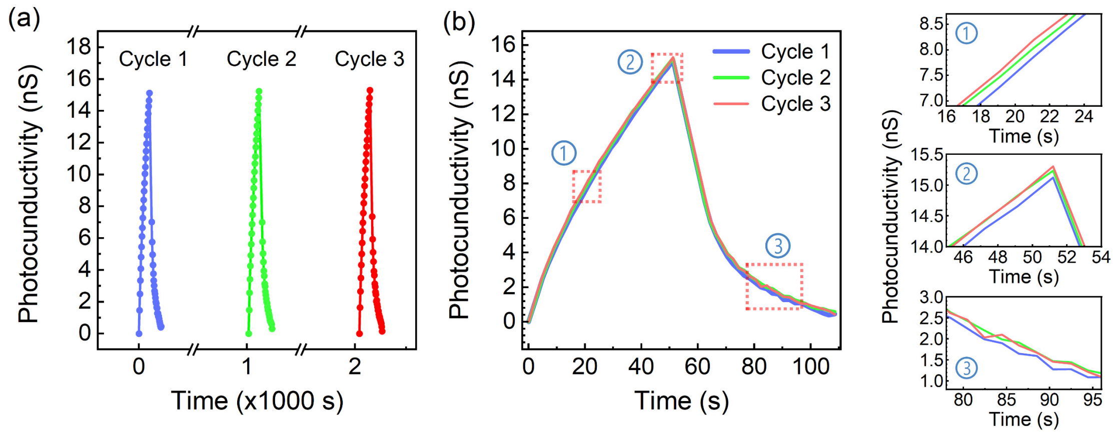

Figure S3. Reversibility and repeatability of photoconductance modulation under repeated potentiation–depression cycles. (a) Channel photoconductance of the TIPS-Tc/IGZO phototransistor plotted on a consecutive time scale over three successive optical potentiation–electrical depression (LTP–LTD) cycles. Each cycle consists of optical potentiation followed by electrical depression under pulsed positive gate bias (+10 V). (b) The same cycles overlapped for direct comparison. Three zoom-in panels highlight the onset of optical potentiation, the peak conductance state, and the voltage-induced conductance suppression during the forgetting process. The nearly identical responses in these regions across cycles demonstrate highly repeatable modulation dynamics. The peak conductance after each optical potentiation step remains unchanged, confirming that conductance suppression under pulsed positive gate bias (+10 V) is fully reversible and the observed "forgetting" behavior originates from reversible suppression of persistent photogating via redistribution and detrapping of photogenerated holes in the organic layer, rather than irreversible degradation of the IGZO channel due to bias stress.