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1 **The Physics of Interlayer Exciton Delocalization in Ruddlesden-**
2 **Popper Lead Halide Perovskites**

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1 **ABSTRACT**

2 Two-dimensional (2D) lead halide Ruddlesden-Popper perovskites (RPP) recently emerged as
3 a prospective material system for optoelectronic applications. Their self-assembled multi
4 quantum-well structure gives rise to the novel inter-well energy funnelling phenomenon, which
5 is of broad interests for photovoltaics, light-emission applications and in emerging technologies
6 (e.g., spintronics). Herein, we developed a realistic finite quantum-well superlattice model that
7 corroborates the hypothesis of exciton delocalization across different quantum-wells in RPP.
8 Such delocalization leads to a sub-50 fs coherent energy transfer between adjacent wells, with
9 the efficiency depending on the RPP phase matching and the organic large cation barrier
10 lengths. Our approach provides a coherent and comprehensive account for both steady-state
11 and transient dynamical experimental results in RPPs. Importantly, these findings pave the way
12 for a deeper understanding of the physics underpinning these systems crucial for establishing
13 materials design-rules to realize efficient RPP-based devices.

14

15 Keywords: Exciton, Ruddlesden-Popper perovskites, Quantum Well, Delocalization,
16 Funnelling

17

18 **INTRODUCTION**

19 Lead-halide Ruddlesden-Popper perovskites (RPPs) are the two-dimensional (2D)
20 analogues of the classical 3D APbX_3 perovskites (A = small organic cation, e.g., Cs^+ , CH_3NH_3^+
21 or $\text{NH}_2\text{CHNH}_2^+$; and X = is the halide anion, e.g., Cl, Br, or I). Owing to their robust
22 optoelectronic properties and better moisture resilience than their 3D counterparts¹⁻⁴, they
23 recently debuted as a viable alternative for efficient perovskite photovoltaics¹⁻⁴ and light-
24 emitting diodes (LED) applications⁴⁻⁶. The structure of RPP is formed by intercalating the 3D
25 perovskite crystal along its (001)-plane with organic large cations (or B-cation), with a generic

1 crystallographic formula of $B_2A_{n-1}Pb_nX_{3n+1}$, where n is the RPP phase index. This structure
2 creates a spontaneous self-assembled multiple-quantum-well (QW) system with alternating
3 layers of B cations [e.g., $C_6H_5C_2H_4NH_3^+$ or $C_4H_9NH_3^+$] as the barrier and inorganic $[PbX_6]^{4-}$
4 perovskite octahedra as the well (Fig. 1a). The well-thickness is quantized and increases
5 proportionally with the phase number n , while the barrier-thickness can be tuned by acting on
6 the chain length of the B-cation. Such modular structure gives rise to numerous fascinating and
7 tunable phenomena, such as quantum and dielectric confinement effects⁷⁻¹⁰, optical Stark
8 effect¹¹, inter-phase energy funnelling^{5, 12}, coherent phonon interactions¹³⁻¹⁴, *etc.* Our group
9 recently also validated that such ultrafast funnelling process also preserves the exciton spin
10 information¹⁵.

11 Using a standard solution-processed spin-coating method, thin films prepared with mixed
12 A- and B- cations form multitudes of RPP phases rather than a single pure phase. In such
13 systems, an ultrafast (sub-ps) energy funnelling from small- n towards large- n phases has been
14 reported^{12, 15-17}. Due to rapid exciton localization towards the lower bandgap emitters, highly
15 efficient light emission from RPPs has been achieved^{5, 12}. However, the detailed mechanism
16 behind this process is still nebulous. Albeit several interesting hypotheses presented, there is
17 still no consensus and the proposed mechanisms are unable to coherently account for all the
18 experimental data. Some of these proposed mechanisms include: quantum couplings¹²; inter-
19 well biexcitonic interactions¹⁶; sub-100 fs Forster energy transfer¹⁷; and long-lived charge-
20 transfer states in RPP¹⁸.

21 Apart from the nature of the dynamical processes, a coherent model should also be able to
22 explain and account for the effect of quantum and dielectric confinement on the static optical
23 properties of these materials. Hong *et al.*⁷ used an infinite quantum well model to reproduce
24 the bandgaps and exciton binding energies of the $n = 1$ and 2 lead-iodide RPP systems.
25 However, such a model could not be further generalized to other RPP phases. Blancon *et al.*¹⁰

1 proposed an empirical law to describe the 1s exciton binding energy of lead iodide RPP.
2 However, understanding on the link between electronic landscapes of different RPP phases and
3 their relations with dynamical processes are still lacking. Other attempts to model this material
4 system have been heavily reliant on density functional theory (DFT) calculations¹⁹⁻²⁰ or tight-
5 binding model²¹. Typical DFT calculations predict a flat dispersion along the stacking axis,
6 implying an impenetrable barrier of the organic layer. While such description has been
7 extremely useful in understanding the energetic landscape within the MQW systems²², it fails
8 to capture some important experimental observations., *e.g.*, subband effects²³⁻²⁴, electrical
9 current along the stacking axis²⁵, *etc.* Moreover, DFT modelling is only limited to the
10 descriptions of linear optical properties and could not be correlated to the experimental
11 dynamics. Hence, a comprehensive generic model that can account not only for the dynamics,
12 but also for its interplay with static effects, is urgently needed to advance our knowledge about
13 ultrafast funnelling in RPP systems.

14 Herein, we formulate a realistic modelling framework for the RPP systems using a simple
15 multi-finite-QW model. In doing this, we also challenge the belief about the failure of
16 superlattice approach in describing the RPP systems, arising from DFT results¹⁹⁻²⁰. In fact, the
17 superlattice approach adopted by our model could satisfactorily account for both steady-state
18 and dynamical photophysics observed in our experiments. Consequently, we can well describe
19 the exciton delocalization across multiple quantum-wells, which leads to coherent inter-well
20 exciton transfer and quantum energy transport within these systems. Furthermore, we propose
21 a rational approach to tune the transfer efficiency based on RPP phase matchings and barrier
22 lengths. This result sheds light on the physics underlying mixed-phased RPP and provides a
23 coherent framework that could explicate the observations reported previously in literature.
24 Importantly, by deepening the level of understanding for these systems, our work sets a way

1 for establishing material design-rules for developing efficient RPP-based optoelectronic
 2 devices.

3

4 **RESULTS AND DISCUSSION**

5 We consider the energy landscape of RPP superlattice along the stacking axis. The actual
 6 potential landscape could be complicated, due to various contributions from band-bending and
 7 image charge effect (Fig. 1b). Herein, we approximate the system to a periodic square well
 8 system (Fig. 1c) with periodic boundary conditions. For a 2D system in cylindrical coordinates
 9 (r, z) , the total wavefunction of an exciton $\Psi(z_e, z_h, r_e, r_h)$ is given by:

$$\Psi(z_e, z_h, r_{eh}) = \psi_e(z_e) \psi_h(z_h) \psi_x(z_e, z_h, r_{eh}). \quad (1)$$

10 Here, $\psi_{e,h}(z)$ is the electron and hole wavefunction; $\psi_x(r_{eh}, z_e, z_h)$ is the exciton envelope
 11 wavefunction; z_e and z_h is the z -coordinate of the electron and hole, respectively; and r_{eh} is the
 12 radial distance between electron and hole, where $r = 0$ is set at the electron and hole centre of
 13 mass and $z = 0$ is taken at the centre of one of the wells. The Hamiltonian for such an exciton
 14 system is given by:

$$H = H_e + H_h + H_C. \quad (2)$$

15 The last term represents the Coulombic interaction between the electron and hole – SI Section
 16 2. The first and second terms in Eq. 2 represents the Hamiltonian for electrons and holes, where:

$$H_{e,h} \equiv H_{e,h}(z) = -\frac{\hbar^2}{2m_{e,h}^*} \frac{d^2}{dz^2} + V_{e,h}(z). \quad (3)$$

17 The first two terms are the kinetic energy and the interaction with the potential landscape,
 18 respectively. Meanwhile $V_{e,h}(z) = V_0^{e,h}$ for the barrier regions and $V_{e,h}(z) = 0$ for the well
 19 regions. Note that in our current formulation, the contribution from exciton-phonon coupling
 20 is neglected. This matter is discussed in detail in SI Section 3.

21

1

2 Figure 1 | Ruddlesden-Popper perovskites (RPP) structure. (a) The crystal structures for different n phases of RPP.

3 (b) The actual energy landscape of RPP system along the stacking axis. (c) Our approximation of the actual energy

4 landscape into a periodic square quantum well. The 3D bandgap (E_G^{3D}), 2D bandgap (E_G^{2D}), and the effective5 electron and hole potential barriers ($V_0^{(e)}$ and $V_0^{(h)}$) are shown.

6

7 Applying 1st-order correction from perturbation theory to the exciton eigenenergy $E^{(1)}$ with8 periodic boundary condition: $\psi_{e,h}(z) = \psi_{e,h}(z + L_w + L_b)$, we have the wavefunction for the9 region $-(L_w + L_b)/2 \leq z \leq +(L_w + L_b)/2$:

$$\begin{aligned}
E^{(1)} &= E_G^{3D} + \left\langle \Psi^{(0)} \left| -\frac{\hbar^2}{2\mu} \nabla_r^2 + H_C + H_e + H_h \right| \Psi^{(0)} \right\rangle \\
&= E_G^{3D} + \left\langle \psi_e^{(0)} \left| H_e \right| \psi_e^{(0)} \right\rangle + \left\langle \psi_h^{(0)} \left| H_h \right| \psi_h^{(0)} \right\rangle + \left\langle \psi_x^{(0)} \left| -\frac{\hbar^2}{2\mu} \nabla_r^2 \right| \psi_x^{(0)} \right\rangle \\
&\quad + \left\langle \Psi^{(0)} \left| H_C \right| \Psi^{(0)} \right\rangle
\end{aligned} \tag{4}$$

$$\psi_{e,h}^{(0)}(z_{e,h}) = \begin{cases} A \frac{\cos(k_1 L_w/2)}{\cosh(k_2 L_b/2)} \cosh\left(k_2 \left[z_{e,h} + \frac{L_w + L_b}{2} \right]\right) & ; z < -L_w/2 \\ A \cos(k_1 z_{e,h}) & ; |z| \leq L_w/2 \\ A \frac{\cos(k_1 L_w/2)}{\cosh(k_2 L_b/2)} \cosh\left(k_2 \left[z_{e,h} - \frac{L_w + L_b}{2} \right]\right) & ; z > L_w/2 \end{cases} \tag{5}$$

$$\psi_x^{(0)}(z_e, z_h, r_{eh}) = \sqrt{\frac{2}{\pi a_B^2}} \exp\left\{-\frac{\sqrt{r_{eh}^2 + (z_e - z_h)^2}}{a_B}\right\}. \tag{6}$$

10 In eq. (4), E_G^{3D} represents the 3D bandgap of the perovskite; the second and third terms

11 represent the quantum confinement effect; the fourth term is the kinetic energy of the exciton;

12 and the last term represents the Coulombic interaction between the electron and the hole, *i.e.*,13 the exciton binding energy. Here, $\psi_{e,h}^{(0)}$ is the electron and hole wavefunctions; $\psi_x^{(0)}$ is the14 exciton envelope with the 2D 1s exciton as the trial wavefunction; $k_1 = \sqrt{2m^*E^{(0)}/\hbar^2}$ and15 $k_2 = \sqrt{2m^*(V_0 - E^{(0)})/\hbar^2}$ are the wavevectors; and a_B is the exciton Bohr radius. The

1 eigenenergies were solved numerically by using the Newton-Raphson method (SI Section 4).
 2 To solve the exciton wavefunction, we employed the variational method with a_B as the sole
 3 free parameter for minimizing the exciton energy (SI Section 5). Previously, a similar method
 4 was utilized to estimate the exciton binding energy from a single finite QW system²⁶. In this
 5 work, we further developed this approach for a multi-QW superlattice system employing
 6 periodic boundary conditions and successfully applied it for the RPP system.

7 As a first test of the model, we estimated the steady-state optical properties of the archetypal
 8 lead-iodide RPP $(C_6H_5C_2H_4NH_3)_2(CH_3NH_3)_{n-1}Pb_nI_{3n+1}$, henceforth called PEAMAPI. The
 9 absorption spectra of the mixed-phase RPP sample is shown in Fig. 2a-b. Excitonic absorption
 10 peaks for different RPP phases up to $n = 5$ are distinguishable. Beyond $n = 5$, the energy spacing
 11 between the adjacent phases is smaller than the linewidth broadening.

12

13

14 **Figure 2 | Exciton absorption and modelling of RPP.** (a, b) The absorption spectra of mixed-phase RPP thin
 15 film of $(C_6H_5C_2H_4NH_3)_2(CH_3NH_3)_{n-1}Pb_nI_{3n+1}$, (or PEAMAPI) at 295 K and 77 K, showing distinguishable
 16 excitonic peaks up to $n = 5$. The solid black line in (b) shows the reproduced absorption spectrum using the
 17 parameters obtained from the model. (c) The UV absorption spectrum of the pure $n = 1$ phase sample (or PEPI)
 18 at 295 K. (d) Modelling of the exciton resonance energy and the bandgap, (e) the exciton binding energy (E_b) vs
 19 experimental result from Ishihara et al. (1992)⁷, and (f) the exciton Bohr radius (a_B), as function of phase number
 20 n . The dotted line in (d) represents fitting by infinite quantum well model.

21

22 To construct our model, the electron and hole effective masses are set to be $0.15m_0$ and
 23 $0.23m_0$, respectively, (*i.e.*, exciton reduced mass of $0.09m_0$) where m_0 is the free electron
 24 mass²⁷⁻²⁸. The dielectric constant of the barrier layer (ϵ_b) is set to be 3.3^7 , while the dielectric
 25 constant for the inorganic layer (ϵ_w) are set to be $6.5^{7-8, 29}$. The L_w and L_b are set to be $n \times 0.63$
 26 nm and 1.0 nm, respectively⁷. Meanwhile, V_0^e , V_0^h , and E_g^{3D} were set as fitting parameters. The

1 model was then fitted to the series of exciton energies of different phase number n . The fitting
 2 result is shown in Fig. 2d, with the parameters displayed in Table 1. The derived exciton
 3 binding energy and Bohr radius are presented in Fig. 2e-f.

4

5 Table 1 | Fitting results for excitonic modelling of PEAMAPI. The 3 fitting parameters and their descriptions are
 6 displayed in the first 3 rows. Other parameters derived from the results are described in detail in the text.

Parameters	Descriptions	Results
V_0^e	Electron barrier height	0.84 ± 0.05 eV
V_0^h	Hole barrier height	0.55 ± 0.05 eV
E_g^{3D}	Bandgap of the 3D phase	1.81 ± 0.05 eV
E_B^{3D}	Exciton binding energy of the 3D phase	~ 10 meV

7

8 Here, the fitted 3D bandgap, which is larger than the reported value of $\text{CH}_3\text{NH}_3\text{PbI}_3$ of ~ 1.65
 9 eV is attributed to the distortion of 2D perovskite octahedral structure³⁰⁻³¹. Our result fully
 10 agrees with the experimental excitonic energies of the RPP sample, while the $1/L_w^2$ energy
 11 dependence from infinite QW model fails to account for the data (Fig. 2d). The accuracy of
 12 our model is upheld by the fact that these parameters could reproduce other independent
 13 experimental results. Firstly, the fitting was only performed over the exciton resonance energy.
 14 However, the exciton binding energy extracted from the fitting well reproduces (without
 15 fitting) the reported experimental results in the literature^{7, 27} of the low dimensions ($n = 1$ and
 16 2) and the 3D phase (extrapolated to ~ 10 meV). This is also seen when the model is extended
 17 to other RPP perovskites, which will be discussed later. Secondly, using these parameters and
 18 a simple 2D absorption model, we satisfactorily reconstructed the experimental absorption
 19 spectrum of the multi-phase PEAMAPI as shown in Fig. 2b. (with $R^2 = 0.996$). Further details
 20 may be found in SI Section 6.

1 It is also worth mentioning that our model is universally applicable to the general RPP
2 system and not limited to the current case of PEAMAPI. As proof-of-concept, we applied our
3 model to two other RPP systems, whose exciton resonances have been reported in literature:
4 $(\text{C}_4\text{H}_9\text{NH}_3)_2(\text{CH}_3\text{NH}_3)_{n-1}\text{Pb}_n\text{I}_{3n+1}$ (or BAMAPI)^{10, 32} and $(\text{C}_8\text{H}_{17}\text{NH}_3)_2(\text{CH}_3\text{NH}_3)_{n-1}\text{Pb}_n\text{I}_{3n+1}$ (or
5 OAMAPI)²⁵. While in previous reports it was noted that these bandgap values do not follow
6 the conventional quantum confinement model, a conclusive understanding of the underlying
7 physics has not been achieved. Herein, our model could satisfactorily reproduce these results
8 (SI Section 7) – thus validating its universality as a standard model for lead-halide RPP.

9 Fig. 3 shows the electron and hole probability densities of the lowest energy state as a
10 function of z for various n , where significant “leaking” of electron and hole eigenfunctions to
11 the organic barriers is observed. Such a ‘spill over’ implies static charge delocalization across
12 multiple RPP layers, which challenges the traditional fully localized photophysical picture of
13 the RPP systems. Previously, RPP were assumed to form an infinite QW system, where the
14 organic layer was thought to be insulating and hindering the charge transport^{3-4, 7, 33}. Such
15 assumption is supported by DFT calculations, where flat dispersion along the stacking axis was
16 observed¹⁹⁻²⁰. Per contra, our result shows that while the organic ligand indeed plays a
17 significant role in providing the quantum and dielectric confinement effects that enhance the
18 bandgaps and exciton binding energies, they are not the dominant impeding factor to the out-
19 of-plane inter-well charge transfer. Instead, other factors such as high exciton binding energy,
20 different phonon-scattering mechanism across the organic layers³⁴⁻³⁵, or inter-phase trap states
21 might be the major obstacles for the low out-of-plane mobility in RPP^{33, 36-37}. Another work by
22 Zhai *et al.*³⁸ also suggests a large exciton localization length of 10 nm in $n = 1$ RPP in the out-
23 of-plane direction, covering multiple consecutive QWs.

24 It is noteworthy to mention that in such system with strong inter-QW wavefunction overlap,
25 Kronig-Penney model predicts the formation of subband states, *i.e.*, non-flat dispersion along

1 the stacking axis. While such subband states have not been thoroughly investigated in the
 2 literature, their presence in RPP has been reported in other recent works²³⁻²⁴. A subband states-
 3 mediated transport mechanism is also necessary to explain our current-voltage (IV)
 4 measurement results along the stacking axis. The discussion on this matter is presented in detail
 5 in SI Section 8.

6
7

8 Figure 3 | RPP multiple quantum well (QW) superlattice eigenfunctions. The lowest eigen-solutions to the multi-
 9 QW superlattice potential landscape are shown for $n = 1$ to $n = 4$ (shown up to 3 QWs). The red and blue shading
 10 represents electron and hole probability densities, respectively. The wavefunction is normalized to one
 11 electron/hole per QW.

12

13 One important implication of this finding is the occurrence of electronic coherent energy
 14 transfers between different wells. To quantitatively understand this process, we model an
 15 isolated two-neighbouring RPP QWs system with phase numbers n_i and n_f , separated by barrier
 16 layer L_b . The eigenstate of such potential landscape is discussed in SI Section 9. The exciton
 17 delocalization across the QWs can be modelled by initially confining the wavefunction in one
 18 QW, before performing the time evolution operator on the wavefunction. The solution to the
 19 one-dimensional time-dependent Schrodinger equation $H\Psi = i\hbar(\partial\Psi/\partial t)$ is:

$$\Psi(x, t) = \sum_{\substack{m = \text{bound} \\ \text{states}}} c_m \psi_m(x) \exp\left[-i \frac{E_m}{\hbar} t\right]. \quad (7)$$

20 The c_m are complex constants, $\psi_m(x)$ are bound eigenfunctions, and $E_m < V_0$ are the bound
 21 eigenenergies of the systems. Fig. 4a shows the all bound eigenfunctions for the case of $n_i = 2$
 22 to $n_f = 3$ pair. Hard-wall boundary condition was set outside the region of interest, to limit the
 23 interactions to only between these two QWs. As a proof of concept without losing generality,
 24 we choose an ‘imperfect’ Gaussian (due to incomplete basis functions) as the initial boundary

1 condition. Such a function is sufficient to demonstrate the delocalized nature of the
 2 wavefunctions in two connected RPP QWs. The dynamics are almost unaffected by the choice
 3 of wavefunction for the localized initial boundary condition (SI Section 10).

4 The time-dependent probability density $P(z,t)$ of the solution is given by:

$$P(z, t) = \Psi^\dagger(z, t) \Psi(z, t) = \sum_{m,p} c_m^* c_p \psi_m^\dagger(z) \psi_p(z) \exp \left[-i \frac{(E_p - E_m)}{\hbar} t \right]. \quad (8)$$

5 Eq. (8) can be rearranged into:

$$P(z, t) = \sum_m |c_m|^2 |\psi_m(z)|^2 + \sum_{\substack{m,p \\ m \neq p}} c_m^* c_p \psi_m^\dagger(z) \psi_p(z) \exp \left[-i \frac{(E_p - E_m)}{\hbar} t \right] \exp \left[-\frac{t}{T_2} \right], \quad (9)$$

6 where the phenomenological decoherence factor with decoherence-time T_2 has been added to
 7 the cross/off-diagonal terms. Here, we set the decoherence time $T_2 = 20$ fs, which is the reported
 8 longest coherence for RPP system¹⁶. The resultant dynamics are oscillations of the weight of
 9 the probability density between the two QWs (See Fig. 4c, Supplementary Movie 1), where
 10 coherent oscillation of populations can be observed between the two QWs with frequency
 11 depending on the energy difference of involved eigenstates, *i.e.*, $\omega = \Delta E/\hbar$). See SI section
 12 11 and Supplementary Movie 2 for a GUI that simulates the coherent transfer for user-defined
 13 MQW parameters.

14

15

16 Figure 4 | Funneling model for RPP. **(a)** The bound eigenstates for electron and holes in connected $n_i = 2$ and $n_f =$
 17 3 pair. The probability density dynamics at different time are plotted in **(b)** Refer to Supplementary Movie 1 for
 18 the animated dynamics. Transfer from the initially localized population in the narrower quantum-well (QW) to
 19 the wider QW is clearly observed. **(c)** The integrated exciton population density at each QW region as function of
 20 time for $n_i = 2$ to $n_f = 3$ pair. Coherent oscillation of exciton populations between the two QWs with decoherence
 21 time $T_2 = 20$ fs are shown. **(d)** Transient absorption spectra of the mixed-phase PEAMAPI thin film, pumped

1 resonantly at $n = 1$, $n = 2$ and $n = 3$ exciton resonance with pump fluences 6.0, 9.9, and 29 $\mu\text{J}/\text{cm}^2$ respectively, at
 2 1 ps. (e) The deconvolved initial photoexcited exciton populations due to the pump at different phase n , and their
 3 corresponding populations at $t = 1$ ps. The total population is normalized to 1. (f) The change of exciton population
 4 the due to the ultrafast exciton funnelling (green square), as compared to the result from our model (magenta line).

5
 6
 7 The electron transfer efficiency P_e between the two QWs from n_i to n_f can be defined as the
 8 ratio of the final population at n_f versus the total population in both n_i and n_f :

$$P_e = \frac{\int_{n_f} P(z, \infty) dx}{\int_{n_i} P(z, \infty) dx + \int_{n_f} P(z, \infty) dx}. \quad (10)$$

9 The population which extends out into the barrier region is neglected. We subsequently
 10 estimated the electron transfer efficiencies between different sets of n_i and n_f for PEAMAPI.
 11 The results are presented in SI section 12.

12 To further validate our results, we compare the efficiencies obtained from our model to the
 13 experimental results from transient absorption (TA) spectroscopy, where the exciton
 14 population from each QW phases could be elucidated. Fig. 4d shows the transient absorption
 15 spectra of PEAMAPI thin film sample at 1 ps after pump excitation, photoexcited resonantly
 16 at $n = 1$, $n = 2$ and $n = 3$ excitonic peak. At this time delay, we expect all the coherent funnelling
 17 processes to have ended, and other excitonic recombination processes have yet to have taken
 18 place. Hence the observed population at each phase primarily originates from the exciton
 19 redistribution due to coherent funnelling processes. Fig. 4e shows a comparison between the
 20 initial and post-funnelling population distributions in each QW phase. The initial population
 21 distribution can be derived from the contribution of each phase to the linear absorption
 22 spectrum (see SI Section 6). A significant drop and increase in populations from the narrower
 23 and wider QW phases are observed, respectively. This signifies that such coherent funnelling

1 process contributes significantly to the dynamics in the system, and hence is crucial to be
2 studied.

3 It is noteworthy to mention that previous studies have reported charge transfer times in RPP
4 films in the order of several ps to 100s of ps³⁹⁻⁴¹. Such slower charge transfer dynamics are
5 different from this sub-ps coherent transfer process reported in this study. In fact, detailed
6 analysis of the TA kinetics of our RPP film could distinguish these two processes: the ultrafast
7 funnelling, which happens within the first 100 fs, and the these slower charge transfer,
8 attributed to hole back-transfer³⁹⁻⁴¹. The details of this analysis are presented in SI Section 13.

9 We proceed to quantify the population changes of each QW phase due to this funnelling
10 process. Estimating the change in population distribution of such a complex system is not
11 straightforward, as the funnelling could happen multiple times from one phase to another.
12 Hence, here we employ a statistical method to extend the result from our isolated 2 QWs model
13 for capturing the coherent funnelling dynamics of this complex real system, the details of which
14 are presented in SI section 14. The comparison between experimental result and our model is
15 shown in Fig. 4f. The model can satisfactorily reproduce the experimental data, especially for
16 $n = 1$ and $n = 2$ resonant excitations, while it slightly underestimates the contribution of
17 coherent funnelling in the case of $n = 3$ resonant excitation. This implies that such coherent
18 funnelling plays a more vital role for low n phases. For $n \gg 1$, other funnelling process, such
19 as Forster energy transfer¹⁷, have an increasing contribution as there is a decrease of inter-phase
20 energy gaps (and hence increasing inter-phase spectral overlaps).

21 Another process predicted by the exciton delocalization model is the coherent uphill/back
22 transfer; namely, an ultrafast transfer of excitation from thicker to thinner wells (SI Section
23 12). This coherent process is expected to be a transient phenomenon that disappears due to loss
24 of coherence within the ensemble system. To unravel this, we measured the transient absorption

1 dynamics of the PEAMAPI with average stoichiometric ratio of the precursor solution $\langle n \rangle =$
2 1.3. Such low $\langle n \rangle$ ratio results in a higher yield of lower n phases (Fig. 5a).

3 The sample was excited with pump energy 2.02 eV ($137 \mu\text{J}/\text{cm}^2$), resonant with the $n = 3$
4 RP phase (Fig. 5b). Such pump energy should not be able to excite $n = 1$ and 2 phases.
5 Interestingly, we observed a short-lived photobleaching signature at $n = 1$ and 2 resonant
6 energies (Fig. 5c-d) lasting about 100 fs. We considered the contribution from optical Stark
7 effect (OSE) and two-photon absorption (TPA)¹¹ to the signal. However, based on our
8 calculation (SI Section 15), OSE would contribute to less than 15% of the observed signal (Fig.
9 5c). We also exclude the contribution of TPA based on low pump fluence measurements (SI
10 Section 15). Thus, we could conclude that the observed PB peak originates from the state filling
11 of these two phases from coherent back-transfer process. Note that this process could also be
12 observed in our standard PEAMAPI sample (*i.e.*, $\langle n \rangle = 2$, datasets presented in Figure 4d),
13 although it is less obvious due to stronger negative photoinduced absorption background from
14 higher n phases. These coherent back-transfer signatures in our $\langle n \rangle = 2$ samples can also be
15 elucidated using global analysis of the TA spectra, as discussed in SI Section 13. This observed
16 ultrafast back-transfer process is an unambiguous experimental proof of exciton delocalization
17 in RPP system, as it would not occur in case of incoherent energy transfer (*e.g.* ultrafast Foster
18 energy transfer¹⁷).

19

20

21 Figure 5| Back transfer in PEAMAPI thin film. (a) Absorption spectrum of PEAMAPI thin film with average
22 stoichiometric ratio $\langle n \rangle = 1.3$. (b) Spectrum of the pump pulse used to the study back transfer, (c) TA spectrum
23 slice at 100 fs waiting time showing state filling of $n = 1$ and $n = 2$ exciton states. The red solid line indicates the
24 estimated contribution from optical Stark effect (OSE). (d) Normalized photobleaching kinetics probed at $n = 1$
25 (2.39 eV) and $n = 2$ (2.17 eV) resonance energies with pump excitation energy of 2.07 eV

26

1 CONCLUSION

2 In summary, we demonstrated a universal finite quantum well model that satisfactorily
3 describes both steady-state and dynamical optical properties in RPPs. Here, our model draws a
4 general connection between the electronic energetics of different RPP phases. In previous
5 studies, each phase requires an independent theoretical calculation (*i.e.*, density functional
6 theory) without a clear relation between the electronic structures of different phases. Our result
7 therefore offers a complementary picture of the electronic structures of RPP, which leads to a
8 more profound understanding of the system. Moreover, this model reveals the presence of
9 wavefunction delocalization across adjacent QWs, which penetrates the organic barrier layer.
10 This result challenges the prevailing notion where the organic layer was assumed to be a perfect
11 barrier. Our model also uncovers the presence of coherent electronic funnelling, which
12 successfully reproduces experimental data. This coherent funnelling is crucial especially for
13 low n phases.

14 Fundamental-wise, this discovery of exciton delocalization, and thus strong inter-QW
15 electronic coupling, provides a novel understanding of the physical processes behind these
16 inter-QW processes, which were previously ascribed to various independent weak-coupling
17 mechanisms. It pieces together the previous reports of various inter-QW interactions (*i.e.*, inter-
18 well charge transfer excitons¹⁸, inter-QW biexcitonic interactions¹⁶, interlayer exciton
19 diffusion⁴² *etc.*). Furthermore, such Dexter-like coherent energy transfer uncovered by our
20 model is expected to be spin-preserving. This could remarkably describe our previous
21 experimental observations of spin-preserved energy funneling in RPP¹⁵. Application-wise, our
22 result suggests that the insulating organic ligand is not the main obstacle for charge transport
23 applications in RPP. Hence, we delineate the direction for future optimization effort to be
24 directed at reducing the exciton binding energy or the inter-phase traps. Importantly, our work

1 uncovers the fundamental understanding of electronic properties in RPP, which is crucial for
2 setting-up a material design-rules for efficient RPP-based optoelectronics.

3 Beyond RPPs, our work also challenges the conventional understanding on the role of the
4 large organic molecules, e.g., in quantum dots (QDs), where they are often used as ligands⁴³⁻
5 ⁴⁴. The assumption that these insulating ligands form an “infinite” barrier is often taken for
6 granted and left unexamined – which if it is wrong, it will undoubtedly lead to a cascade of
7 misinterpretations of the underlying physics. Here, we show otherwise: tunnelling across these
8 ligands could still occur and lead to exciton delocalization across neighbouring systems (at
9 least in RPPs). Our work therefore serves as cautionary remark to re-examine this common
10 assumption, which could potentially hamper the development of exciton-based technology,
11 e.g., the emerging field of excitonics⁴⁵.

12

13 **ASSOCIATED CONTENT**

14 The data that support the findings of this study are available from the corresponding author
15 upon reasonable request. All codes used in the computation of results in this work and
16 designing of the GUI can be made freely available upon reasonable request. The Supporting
17 Information is available free of charge via the internet at <http://pubs.acs.org>

18

19 **AUTHOR’S CONTRIBUTIONS**

20 D.G., S.R., and T.C.S. conceived the idea for the manuscript. D.G. and S.R. designed and
21 performed the experiment. D.G., S.R., J.W.M.L., performed the modelling. D.G., S.R., M.R.,
22 N.M., and T.C.S. analysed the data. S.R. and Q.Z. synthesized the samples. Y.W. and S.Y.
23 prepared the devices and performed the IV measurement. Q.X. performed the DFT
24 calculations. T.C.S. led the project.

25

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