Enhancing Long-Range Energy Transport in Supramolecular Architectures by Tailoring Coherence Properties

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ABSTRACT: Efficient long-range energy transport along supramolecular architectures of functional organic molecules is a key step in nature for converting sunlight into a useful form of energy. Understanding and manipulating these transport processes on a molecular and supramolecular scale is a long-standing goal. However, the realization of a well-defined system that allows for tuning morphology and electronic properties as well as for resolution of transport in space and time is challenging. Here we show how the excited-state energy landscape and thus the coherence characteristics of electronic excitations can be modified by the hierarchical level of H-type supramolecular architectures. We visualize, at room temperature, long-range incoherent transport of delocalized singlet excitons on pico- to nanosecond time scales in single supramolecular nanofibers and bundles of nanofibers. Increasing the degree of coherence, i.e., exciton delocalization, via supramolecular architectures enhances exciton diffusivities up to 1 order of magnitude. In particular, we find that single supramolecular nanofibers exhibit the highest diffusivities reported for H-aggregates so far.

INTRODUCTION

Supramolecular chemistry provides intriguing opportunities to create nano- to mesoscale assemblies with unprecedented optical and electronic functionalities owing to cooperative interactions between the constituent building blocks.1–7 A key functionality for potential applications is, for example, efficient long-range excitation energy transport.8–14 In general, energy transport in organic materials is governed by the delicate interplay between electronic Coulomb coupling between densely packed molecules and electronic and structural disorder. On the one hand, electronic coupling leads to the formation of delocalized exciton states; that is, electronic excitations are coherently shared by many molecules, which we refer to as (quantum) coherent transport. On the other hand, electronic and structural disorder leads to a localization of excitons on small domains of supramolecular assemblies.15,16 If disorder dominates, long-range transport cannot be realized, because incoherent Förster-type hopping of strongly localized excitons limits transport to some tens of nanometers.17 In contrast, reducing disorder increases exciton delocalization, and thus the degree of coherence.15,18–20 Such combined incoherent–coherent transport, i.e., incoherent hops of delocalized excitons,2,21–26 with a strong contribution of coherence, indeed allowed achieving distances beyond 1 μm.12,23 However, a full understanding and control of long-range energy transport is still highly complex, because in supramolecular nanostructures the electronic coupling and disorder are typically of the same order of magnitude. This so-called intermediate regime renders disentangling the different contributions to energy transport difficult, on both the theoretical and experimental side.16 Since transport efficiencies and distances are predicted to be largest in this regime,2,4,5,7–9 a unique picture is desirable for the design of novel excitonic materials and devices.16

A straightforward approach to modify electronic coupling and disorder makes use of the self-assembly of defined nanostructures based on the same building blocks. In this context, so-called pathway complexity can be exploited to form thermodynamically and kinetically stable supramolecular aggregates with different structural order2,4,7,24 and thus with significantly altered photophysical and energy transport properties. In contrast, supramolecular aggregates with different hierarchical levels11,29–31 feature the same structural arrangement of the building blocks, i.e., the same electronic coupling, with only subtle variations in the local electronic
environment. Such structures are therefore ideal candidates to reveal the interplay between the electronic coupling and disorder.

Here we present stable and robust supramolecular architectures based on a carbonyl-bridged triarylamine trisamide (CBT, compound 1, see ref 32) with different hierarchical levels depending on the solvent, i.e., single supramolecular fibers and bundles of supramolecular fibers (Figure 1). The molecular design of compound 1 results in columnar structures with a well-defined, cofacial H-type arrangement of the CBT cores that is driven by directed hydrogen bonding between amide groups. Excitons in these supramolecular architectures possess different degrees of coherence (delocalization), tuned by bundling-induced electronic disorder. We are thus able to resolve the competition between coherence and disorder and to demonstrate its impact on long-range, pico- to nanosecond, incoherent transport of charge carriers on the level of single nanostructures at room temperature. In contrast to previous work on nanotubular assemblies,11,30 we obtain an intercolumnar spacing of 3.2 nm. This distance is substantially smaller than the calculated diameter of about 4.4 nm for the extended compound 1.32 In bundles the peripheries of adjacent nanofibers therefore strongly interact. Based on these data, we estimate that one bundle consists of approximately 2000 nanofibers.

■ RESULTS AND DISCUSSION

Controlled Self-Assembly of Single Nanofibers and Bundles of Nanofibers. Using two selected solvents, we are able to self-assemble compound 1 into distinct supramolecular morphologies (see Supporting Information section 1). In n-dodecane, compound 1 forms single supramolecular nanofibers of several micrometers in length (Figures 2A and S1) and uniform heights of about 2 nm (Figure 2B), as shown by atomic force microscopy (AFM), which is consistent with our previous work on single CBT-nanofibers.23,32 Transmission electron microscopy (TEM, Figure 2C) reveals single supramolecular nanofibers and nanofibers partially located next to each other due to sample preparation. The selected area electron diffraction (SAED) pattern features a signal corresponding to a distance of 0.33 nm (Figure 2D) caused by cofacial stacking of CBT cores along the supramolecular nanofibers.23,32

Self-assembly of compound 1 in anisole results in bundles of supramolecular nanofibers with widths and heights on the order of 100 nm, as shown by AFM and TEM measurements (Figure 2E,F,G and Figure S1). The SAED pattern yields the characteristic cofacial stacking distance of 0.33 nm between CBT cores (Figure 2H). Additionally, SAED at smaller angles (Figure S2) reveals defined signals corresponding to a distance of 2.8 nm. Assuming a columnar hexagonal packing of the nanofibers,32 we obtain an intercolumnar spacing of 3.2 nm. This distance is substantially smaller than the calculated diameter of about 4.4 nm for the extended compound 1.32 In bundles the peripheries of adjacent nanofibers therefore strongly interact. Based on these data, we estimate that one bundle consists of approximately 2000 nanofibers.

Optical Properties of Nanofibers and Bundles. UV–vis and photoluminescence (PL) spectra of dissolved compound 1 in THF (Figure 3A) exhibit the characteristic vibronic progression of an aromatic molecule (see Supporting Information section 4, Table S1). The maxima at 460 nm and 490 nm, respectively, are attributed to the electronic (0–0) π−π* transition of the CBT core.33,34

The absorption spectra of supramolecular nanofibers and bundles of nanofibers are shown in Figure 3B and C, respectively (see also Figure S3). Both feature a substantially reduced intensity of the highest-wavelength (lowest-energy) peak around 520 nm (labeled A1) compared to the spectrum of dissolved compound 1. This spectral change is characteristic for delocalized absorbing excitons in H-aggregates, formed by substantial electronic Coulomb coupling between adjacent CBT cores.35 Circular-dichroism spectra of dispersions of both supramolecular morphologies feature nearly identical signatures (Figure S4) indicating no significant difference in the structural arrangement of the CBT cores in supramolecular nanofibers and bundles of nanofibers.

After absorption rapid, subpicosecond relaxation within the exciton bands takes place toward lower-energy, relaxed exciton states,22 from where emission occurs (see Supporting Information section 4, Table S1).
The intercolumnar distance of 3.2 nm within bundles prevents delocalization of electronic excitations between columns. A bundle is thus modeled as an arrangement of independent nanofibers. Electronic (energy) disorder is included by taking the CBT cores’ transition energy offsets from a Gaussian distribution with a width $\sigma$. Moreover, we include a correlation length $l_0$ that accounts for differences in the spatial distribution of disorder in the transition energies (Figure S5).

The simulations (Figure 3B,C, black lines) agree very well with the experimental data in the relevant spectral region. The absorption spectra of both architectures are well described by a common set of parameters, i.e., by an electronic coupling of $g_0 = 735 \text{ cm}^{-1}$ (91 meV) and an electronic disorder of $\sigma = 1036 \text{ cm}^{-1}$ (134 meV, see Figure S9 and Supporting Information section S), which places both morphologies in the intermediate coupling regime.

The differences between the PL spectra of nanofibers and bundles can only be modeled using different correlation lengths. The absence of the 0–0 PL peak in the spectrum of bundles requires a vanishing singlet excitation correlation length ($l_0 \geq 10$ CBT cores) due to the distinct spatial transition energy landscapes along one column (single nanofiber and within a bundle, respectively) compared to the rough excited-state energy landscape along nanofibers in bundles (Figure 3F). The different excited-state energy landscapes along one column (single nanofiber and within a bundle, respectively) due to the distinct spatial transition energy correlations is visualized in Figure 3E,F. We note that these realizations are directly taken from the numerical simulations.

**Direct Visualization of Exciton Transport.** Our optical spectroscopy data demonstrate that we are able to tune the
coherence characteristics of the relaxed excitons along the H-type columns by altering the hierarchical level of our architectures. These relaxed excitons are responsible for incoherent long-range transport, since they perform many hopping steps within their substantial excited-state lifetime in our H-aggregates. Importantly, the hopping rates of delocalized excitons have to be described by a generalized Förster theory, in which optically dark exciton states contribute to the hopping rates.36−38 These systems are thus ideal to resolve the interplay between morphology, correlated electronic disorder, and coherence (delocalization) in the long-range incoherent transport of excitons along individual, spatially isolated nanostructures on pico- to nanosecond time scales. Figure 4A and B display representative wide-field PL images of isolated nanostructures, both with lengths of several micrometers, in agreement with the AFM data (Figure 2A,E). The single nanofiber shows a small PL signal (Figure 4A), which demonstrates the weakly optically allowed nature of the emitting excitons and thus the high degree of coherence within the nanofiber. The signal from the bundle of nanofibers is significantly stronger mainly owing to the large number of columns within the bundle (Figure 4B) and to a lesser extent due to the higher PL quantum efficiency of one column in a bundle (see Table S2).

Having located isolated nanostructures, we switched the microscope to confocal illumination and centered each nanostructure in the diffraction-limited focus of a pulsed laser (red dashed circles, Figure 4A,B). Combining detection-beam scanning with time-correlated single-photon counting39 (see Supporting Information sections 1 and 6), we measured
PL decay curves while scanning the detection position along the long axis of the nanostructures (dashed arrows in Figure 4A,B). Figure 4C and D show the resulting PL intensity distributions, I(x, t), as a function of the distance x relative to the center of the excitation spot and time t after laser excitation. Normalization of the spatial intensity distributions at each point in time (Figure S13) reveals the broadening of the PL signal along the nanostructures’ long axes on (sub-) nanosecond time scales. Hence, the initial singlet exciton population, created by the diffusion-limited excitation pulse, is transported away from the excitation spot prior to (radiative) decay. This energy transport is significantly more pronounced for the single nanofiber compared to the bundle of nanofibers (Figure 4C,D, white contour lines). We attribute this difference to the distinct excited-state energy landscapes (Figure 3E,F). We rule out artifacts due to saturation and technical issues, since we operate under very low excitation fluences, and we have performed an independent control experiment on a system that does not show long-range energy transport (see Supporting Information, Materials and Methods, and Figure S12).

To quantitatively describe the time-dependent broadening of the spatial intensity distributions, we calculated the second moments \( \mu_2(t) \) at time \( t \) as a measure for their widths (Figure S13). We evaluated changes of \( \mu_2(t) \) with respect to the second moment (width) of the initial distribution \( \mu_2(0) \):

\[
\Delta \mu_2(t) = \mu_2(t) - \mu_2(0)
\]

The \( \Delta \mu_2(t) \) curves retrieved from the data in Figure 4C,D are shown as thick dashed lines in Figure 4E,F. At short times (\( t \leq 1 \) ns), we find similar slopes for both curves. However, the \( \Delta \mu_2(t) \) values for the single nanofiber are larger by more than 1 order of magnitude compared to those for the bundle. This observation reflects the faster and more pronounced broadening of the initial exciton population in the single nanofiber due to more efficient energy transport. For longer times (\( t \geq 2.5 \) ns) the broadening slows down and a plateau is reached for both architectures. We confirmed the same trend for in total 56 single nanofibers and 32 bundles, illustrated with thin solid green and blue lines in Figure 4E,F.

The second moments \( \Delta \mu_2(t) \) follow to a good approximation a power law for \( t < 1 \) ns. We can thus fit the transport dynamics with a diffusion model \( D(t) = \frac{1}{2} A t^{\alpha-1} \) (see Supporting Information section 7):

\[
\Delta \mu_2(t) = A t^\alpha
\]

Here \( \alpha \) is the diffusion exponent and \( A \) is the exciton hopping coefficient, which is related to the time-dependent diffusivity \( D(t) \). Figure 4G shows the exponents for nanofibers (green bars) and bundles (blue hatched bars) for all curves in Figure 4E,F. We find a broad distribution with \( 0 < \alpha < 1 \) (for \( t < 1 \) ns) due to the intrinsic electronic disorder in deposited supramolecular nanostructures. For single nanofibers the mean exponent is \( \alpha_{\text{Fiber}} = 0.78 \pm 0.04 \), and a significant fraction exhibits \( \alpha \approx 1 \), which indicates normal diffusion visualized in Figure 4E,F with black solid lines. In contrast, the mean exponent for bundles is smaller with \( \alpha_{\text{Bundle}} = 0.37 \pm 0.47 \), which is characteristic for strongly subdiffusive transport due to the disordered energy landscape.\(^{22} \) Notably, for bundles the highest occurrence of exponents is at \( \alpha \approx 0 \). This behavior is expected for a system with strong local electronic perturbations (Figure 3F), in which trapping hinders exciton transport.\(^{24} \) For the single nanofibers the average exciton hopping coefficients \( A \) as a function of the exponent \( \alpha \) are larger (Figure 4H). This
translates into higher diffusivities $D(t)$ for single nanofibers. For example, for the nanofiber shown in Figure 4C we find $D_{\text{fibre}}(t = 1 \text{ ns}) = 1.03 \text{ cm}^2/\text{s}$, which is the largest value reported for an H-aggregate and is more than 1 order of magnitude larger than the diffusivity for the bundle shown in Figure 4D with $D_{\text{bundle}}(t = 1 \text{ ns}) = 0.05 \text{ cm}^2/\text{s}$ (see also Figure S14).

**CONCLUSION**

Our H-type supramolecular architectures with different hierarchical levels represent a versatile system to understand the subtle interplay between electronic coupling, disorder, and coherence for efficient long-range, incoherent transport of delocalized singlet excitons. We have demonstrated remarkable hindrances due to trapping in local energy minima.22 The uncorrelated transition energies in bundles of nanofibers can be explained by very subtle local electronic perturbations due to interacting peripheries.30 Alternatively, disorder on a local scale between columns may arise from a geometric frustration in a hexagonal packing due to compensation of macrodipoles.42 Both effects can destroy shared electronic environments. Our observations are a manifestation of coherence-enhanced diffusivities of excitons45,18,43 and highlight the critical role of spatially correlated transition energies of the supramolecular building blocks for long-range energy transport.44 The present data therefore add a new dimension to the spectroscopic properties as well as in the energy transport characteristics of single supramolecular nanofibers and bundles of nanofibers. The transition energies of adjacent CBT cores in single supramolecular nanofibers are spatially correlated, resulting in smooth excited-state energy landscapes. The concomitant high degree of coherence (exciton delocalization) facilitates long-range incoherent energy transport. In contrast, in bundles of nanofibers spatial correlations in the transition energies are found to be absent. This gives rise to a disordered excited-state energy landscape with strongly localized excitons. Hence, exciton transport is hindered by trapping in local energy minima.25 The uncorrelated transition energies in bundles of nanofibers can be explained by very subtle local electronic perturbations due to interacting peripheries.30 Alternatively, disorder on a local scale between columns may arise from a geometric frustration in a hexagonal packing due to compensation of macrodipoles.42 Both effects can destroy shared electronic environments. Our observations are a manifestation of coherence-enhanced diffusivities of excitons45,18,43 and highlight the critical role of spatially correlated transition energies of the supramolecular building blocks for long-range energy transport.44 The present data therefore add a new dimension to the development of a detailed theoretical understanding of energy transport in columnar H-type supramolecular nanostructures45 as well as for the design of novel, optimized nanophotonic applications.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/jacs.0c01392.

Materials and methods; additional characterization of supramolecular architectures by AFM, electron diffraction, and optical spectroscopy; numerical simulations of optical spectra and exciton diffusion model (PDF)

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