Energy Funneling in a Noninteger Two-Dimensional Perovskite

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ABSTRACT: Energy funneling is a phenomenon that has been exploited in optoelectronic devices based on low-dimensional materials to improve their performance. Here, we introduce a new class of two-octahedron-layer-thick (n = 2) 2D cesium lead bromide perovskite nanosheets; it consisted of a central n = 2 region surrounded by edge-lying n = 3 regions, as imaged by electron microscopy. Thicker noninteger 2D CsPbBr₃ nanostructures were obtained as well. These noninteger 2D perovskites formed a laterally coupled quantum well band alignment with virtually no strain at the interface and no dielectric barrier, across which unprecedented intramaterial funneling of the photoexcitation energy was observed from the thin to the thick regions using time-resolved absorption and photoluminescence spectroscopy.

KEYWORDS: perovskite, heterostructure, nanocrystal, photophysics, TEM

It is essential to build a fundamental view connecting the structure of semiconductors to their photophysical properties, which ultimately determine the performance of optoelectronic devices. Metal halide perovskites stand out because they tout numerous attractive features for optoelectronic applications, such as high absorption coefficients, easily tunable bandgaps, relative defect tolerance, and long carrier diffusion lengths. Following a decade of research, the photoluminescence quantum yield (PLQY) of lead halide perovskite quantum dots has reached unity, the efficiency of perovskite-based solar cells currently exceeds 33%, and perovskite-based photodetectors are competitive against those made from silicon.

Two-dimensional (2D) metal halide perovskites are of specific interest because they outcompete their bulk counterparts in terms of optical performance and environmental stability, which are among the most pervasive issues limiting the proliferation of perovskite materials into mainstream applications. However, the primary challenge with 2D perovskites is that confinement precludes the formation of long-lived free carriers through competition with strongly bound, radiative excitons. This ultimatum in confined semiconductors has motivated the exploration of perovskite dimensionality to achieve a material with an optimal optoelectronic performance and stability. The strategy of energy funneling has been developed to treat the adverse effects of confinement. The bandgap of 2D perovskites scales with material thickness due to confinement. In energy funneling, the photoexcitation energy is funneled downhill along a bandgap gradient formed by a distribution of packed 2D perovskites that increase in thickness, known as quasi-2D perovskites. The goal of energy funneling is to accumulate excited species on the thickest 2D perovskite, which is idealized by lower exciton binding energy. From the thickest 2D perovskite, the enhanced emission rate can then be exploited for light-emitting diode or lasing applications, or charge may be extracted more readily for solar cells. However, a fraction of the photoexcitation energy is lost with each successive transfer event in energy funneling depending on the dielectric constant of the ligand spacers surrounding the perovskite lattice. Such efficiency loss has motivated further research on energy transfer, ligand shell chemistry, and the development of alternative approaches to improve energy funneling. This includes superlattices with strong electronic coupling, compositional core/shell, core/crown, and ionic heterostructures, van der Waals heterostructures, mixed-dimensionality nanostructures, and dielectric doping.

In this regard, we propose a novel material characterized by thickness heterogeneity within a single 2D nanostructure,
which we term a “noninteger 2D semiconductor”. The absence of ligands between regions of different perovskite thicknesses in the noninteger system forms a downhill band alignment without dielectric barriers, which may circumvent the obstacles faced by quasi-2D perovskites in energy funneling. We report the occurrence of intramaterial energy funneling from thinner to thicker regions, as evidenced by time-resolved photoluminescence (PL) and femtosecond transient absorption (fsTA) spectroscopy. Based on atomic resolution transmission electron microscopy (TEM) imaging and structural modeling, the interface across which transfer took place was virtually strain-free, unlike that present in heterostructures of varying elemental composition. Furthermore, this work unveils confinement as a knob to tune the interfaces and properties of semiconductors local to a single nanoparticle and highlights the implications of thickness variability on the optoelectronic properties of semiconductors, portending immediate device applications.

It has been established that perovskites are amenable to postsynthetic modification due to their soft, ionic lattice. For instance, bandgap tunability in perovskites has been achieved via anion exchange, and their morphology can even change in the presence of polar solvent. In this work, noninteger 2D perovskites were formed by the introduction of polar ligands to colloidal cesium lead bromide nanosheets (Figure 1a), leveraging the dynamic ligand environment and lability of perovskite to induce desirable structural transformation. The product of this reaction was a previously undiscovered 2D nanostructure resembling the parent nanosheet but with thickened edges. 2D perovskites are identified by their thickness (phase number) in units of octahedra, $n = x$. Therefore, we term the nonuniform-thickness products of the structural transformation reaction “noninteger” because they cannot be fully described by just one integer phase number.

The starting colloidal nanosheets had an edge length of around 100 nm (Figure 1b and Figure S1). The cubic, two lead bromide octahedral layer thick ($n = 2$) CsPbBr$_3$ nanosheet was synthesized via our reported synthetic procedure with modifications. The 2D morphology enabled the observation of intense, room-temperature excitons, with an absorption peak at 428 nm and a PL emission peak at 436 nm (Figure S2), both consistent with literature results on the assignment of $n = 2$ CsPbBr$_3$ nanosheet thickness. The cubic phase was confirmed by TEM (Figure S3). The $n = 2$ nanosheets were
also monodisperse in thickness, as indicated by a single excitonic peak in the absorption and PL spectra. This high purity meant that optical measurements could be performed to unambiguously track the transformation of \( n = 2 \) nanosheets into noninteger products containing different thicknesses of lead bromide octahedra, with different excitonic energies, alongside TEM for structural identification, as follows.

Aliquots of the reaction mixture were removed and purified throughout the reaction, which allowed for the study of noninteger products \( \textit{ex situ} \) without further structural transformation. TEM imaging showed an increase in contrast growing at the edges of the nanosheets as the reaction progressed (Figure 1b−d). We note now that the nanosheet-loaded TEM grids were encapsulated with graphene to avoid electron beam damage (Figures S4 and S5), as ultrathin \( \text{CsPbBr}_3 \) perovskite is notoriously beam-sensitive.\(^{46,47} \)

The integrated TEM image line scan intensity increased discretely at the edges of the noninteger structures (Figure 1e−g). Accompanying spikes in the derivative were consistent across the different noninteger 2D \( \text{CsPbBr}_3 \) phase compositions. This contrast change corresponded to the formation of a thicker \( \text{CsPbBr}_3 \) phase relative to the starting \( n = 2 \) phase. To identify the phase of the thickened edges of the nanosheet, we turned to PL and absorption spectroscopy.

In coincidence with TEM, an absorption study was conducted that tracked the transformation of the pure \( n = 2 \) excitonic absorption peak at 428 nm into another excitonic absorption peak at 454 nm (Figure 1h−j). The respective PL peaks were located at 436 and 463 nm. This exciton was lower in energy because quantum confinement was alleviated as the \( n = 2 \) nanosheets grew thicker. The absorption intensities coincided with the observed edge-thickening as the \( n = 2 \) exciton diminished with the appearance of the 454 nm exciton. Furthermore, the appearance of another exciton at a different wavelength, rather than a continuous red shift, implied that the noninteger nanostructure was formed with discrete thickness resolution. This resolution was that of a single lead bromide octahedron layer, as the 454 nm exciton (463 nm PL) matched that of pure, as-synthesized \( n = 3 \) \( \text{CsPbBr}_3 \) nanosheets (Figure S6). Thus, we assign the growing, higher contrast edges of noninteger 2D \( \text{CsPbBr}_3 \) to the \( n = 3 \) phase and the lower contrast interior to the starting \( n = 2 \) phase, both present in a single nanostructure. To the authors’ knowledge, this is the first report of such a noninteger 2D semiconductor. While the \( n = 2:3 \) noninteger \( \text{CsPbBr}_3 \) perovskite is the model system of focus in the main text, further reaction yielded similarly interesting \( n = 3:4 \) \( \text{CsPbBr}_3 \) in the form of nanoframes and abnormal, sawtooth nanostructures. A collection of TEM images of all noninteger 2D \( \text{CsPbBr}_3 \) along with discussion of the structural transformation reaction can be found in Figures S7−S11 in the Supporting Information.

We now aim to substantiate our claim that this noninteger concept is an alternative approach to accomplish energy funneling. We hypothesize that the attached \( n = 2 \) and \( n = 3 \) exciton components of the pure \( n = 2:3 \) \( \text{CsPbBr}_3 \) can be modulated to form other noninteger \( \text{CsPbBr}_3 \) compositions with varying thickness resolutions. This concept is illustrated in Figure 2a. An energy funneling scheme is depicted in Figure 2b. The 2D PLE spectrum of \( n = 2:3 \) \( \text{CsPbBr}_3 \) with 40% \( n = 3 \) composition is shown in Figure 2c. The inset displays the normalized PL excitation spectrum viewed at the 463 nm emission wavelength. Figure 2d shows the PL lifetime map of \( n = 2:3 \) \( \text{CsPbBr}_3 \) with 40% \( n = 3 \) composition. Inset: the corresponding PL decay constants and relative contributions in parentheses (95% confidence fit).
phases of the noninteger perovskite form a double quantum well with in-plane type-I alignment (Figure 2a). This contrasts with quasi-2D perovskites in which the ligand barrier interrupts the type-I alignment (Figure 2b).

Above-bandgap photoexcitation produced the expected PL spectrum of the $n=2:3$ 2D perovskite, which consisted of exciton emission from the $n=2$ and $n=3$ phases as seen in the 2D photoluminescence-excitation spectrum (PLE, Figure 2c). However, direct photoexcitation of the $n=2$ exciton compared to above-bandgap photoexcitation caused greater intensity, or sensitized, emission from the $n=3$ exciton, as evidenced by the excitation spectrum viewed at the 463 nm emission wavelength (Figure 2c, inset) and in comparison to the $n=3$ nanosheet control (Figure S12). Furthermore, excitation between the optical bandgaps of the $n=2$ and $n=3$ excitons did not produce strong emission, and even direct excitation of the $n=3$ exciton at 454 nm produced relatively weak emission. Additionally, electronic structure calculations of a noninteger 2D perovskite showed that the highest occupied molecular orbitals of the $n=2$ phase extended across the $n=2:3$ interface (Figure S13). The PLE along with computations implied electronic coupling between the $n=2$ and $n=3$ phases and supported the occurrence of energy funneling. The same was true for higher order noninteger 2D CsPbBr$_3$ (Figure S14).

The transfer of photoexcitation energy in a donor–acceptor system is more authentically represented by a change in photodynamics. The PL decay of the $n=2$ and $n=3$ excitons of the pure $n=2$ and $n=3$ CsPbBr$_3$ nanosheets was triexponential, with respective lifetimes of 16 and 30 ns representing the time scale trapping/detrapping process, accompanied by lifetimes <7 ns (Figure S15). The corresponding PL lifetimes of the $n=2$ region to the $n=3$ region was expected because of the alleviation of quantum confinement with increasing CsPbBr$_3$ thickness as well as the aforementioned quenching of the $n=2$ exciton by the $n=3$ exciton. Two distinct regions were seen in the PL decay map of the 40% $n=3$ CsPbBr$_3$ (Figure 2d), which corresponded to the $n=2$ phase in the presence of the $n=3$ phase and vice versa. The increase in PL lifetime from the $n=2$ region to the $n=3$ region was expected because of the alleviation of quantum confinement with increasing CsPbBr$_3$ thickness as well as the aforementioned quenching of the $n=2$ exciton by the $n=3$ phase. This quenching behavior was observed by measuring the PL decay as a function of phase composition in the $n=2:3$ CsPbBr$_3$ complex (Figure 2e).
lifetime of the $n = 2$ exciton in the pure $n = 2$ nanosheet was quenched by a factor of 2.2 in the 40% $n = 3$ CsPbBr$_3$, which was direct evidence of photodynamic transfer from the $n = 2$ phase to the $n = 3$ phase. The relative contribution of this PL lifetime to the PL decay decreased from 61.1% in the $n = 2$ nanosheet to 12.8% in the 40% $n = 3$ CsPbBr$_3$. In coincidence, the PLQY decreased from 31.9% for the pure $n = 2$ nanosheet to 12.8% in the 40% $n = 3$ CsPbBr$_3$. Concomitantly, the relative contribution of the shortest decay constant increased with an increasing $n = 3$ phase composition. The intermediate PL lifetime, around $\sim 2$ ns, did not follow a trend as a function of phase composition. Based on the quenching of the PL lifetimes, high transfer efficiencies of 61% and 83% were calculated for the 10% $n = 3$ CsPbBr$_3$ and 10.4% for the 40% $n = 3$ CsPbBr$_3$. Analogous lifetime mapping and PL lifetime results were obtained for the higher order noninteger 2D CsPbBr$_3$ (Figures S16–S21).

To gain further insight into the photodynamic transfer, we turned to femtosecond transient absorption spectroscopy (fsTA). Unless stated otherwise, all measurements were of solution-state samples, which made it so that interparticle transfer phenomena were diffusion-limited. The ground state bleach (GSB) of the pure $n = 2$ exciton was located around 428 nm (Figure 3a), matching that of the steady-state absorption, with decay constants of $\tau_1 = 216$, $\tau_2 = 855$, and $\tau_3 = 1197$ ps. The $\tau_1$ and $\tau_3$ lifetimes of the GSB decay were consistent with the $\sim 0.3$ and $\sim 2$ ns radiative processes observed in the PL decay, respectively, while $\tau_2$ corresponded to a nonradiative lifetime, as it did not contribute to the PL decay. The fsTA data of the pure $n = 2$ nanosheet served as a standard for comparison against that of the $n = 2:3$ perovskites. It was useful to deconvolute the dynamics by comparing to the pure $n = 2$ nanosheet as a control because both the $n = 2$ and $n = 3$ phases were excited above bandgap. The noninteger 2D perovskites measured via fsTA were of the phase compositions depicted in Figure 1c,d: 10% and 40% $n = 3$ (Figure 3b,c). The GSB of the $n = 2$ exciton was around 428 nm, while that of the $n = 3$ exciton was around 454 nm, which matched the steady-state absorption. This can be seen more clearly in the corresponding fsTA spectral traces (Figure 3d and Figure S22). There was further evidence that the photoexcitation energy was being funneled from the $n = 2$ phase to the $n = 3$ phase, as the radiative $\tau_1$ and nonradiative $\tau_2$ of the $n = 2$ exciton were quenched significantly with increasing $n = 3$ presence in the nanostructure (Figure 3e). Similarly, the excited state absorption at 410 nm was quenched with increasing $n = 3$ presence (Figure 3f). These conclusions were also true for $n = 3:4$ CsPbBr$_3$ (Figures S23 and S24). The decay constants of all relevant GSB and excited state absorption (ESA) features can be found in Table S1. Furthermore, the rise of the $n = 3$ GSB was slower in the 40% $n = 3$ CsPbBr$_3$ than in the pure $n = 3$ nanosheet (Figure 3g). Kinetic modeling of the noninteger 2D perovskite successfully reproduced this behavior when a direct transfer channel was included between the conduction bands of the $n = 2$ and $n = 3$ phases (Figure 3h).

The mechanism by which the photoexcitation energy was transferred between the phases of the noninteger 2D perovskites will now be discussed. It is still a contended issue whether energy funneling in quasi-2D perovskites occurs by the energy or charge transfer mechanism and by what branching ratio.\footnote{35–49} Approaches to distinguish the two mechanisms in a quasi-2D perovskite have been made using their characteristic time scales: hundreds of femtoseconds for energy transfer and tens of picoseconds for charge transfer. Most reports have sided with the exciton energy transfer mechanism,\footnote{12–19,48} justifying by a high degree of spectral overlap and dipole alignment between the donor and acceptor in quasi-2D perovskite; meanwhile, free carrier transfer is suppressed by the interparticle ligand barrier.\footnote{45} However, ligand barrier is not present in noninteger 2D perovskite. We also measured the fsTA spectrum of a film containing a drop-cast mixture of pure $n = 2$ and $n = 3$ nanosheets (Figure 3e and Figure S25). Since interparticle phenomena were no longer...
diffusion-limited in the solid state, this film served as a control sample to mimic energy funneling in a quasi-2D perovskite. The n = 2 exciton decay constant of 0.01, 0.600, and 9.9 ps; the n = 3 exciton decay constant of the pure nano sheet was quenched down to a factor of ~18000. On the other hand, the dynamics observed for the noninteger 2D CsPbBr3 occurred over hundreds of picoseconds. This suggested that charge transfer, which is characteristic slower than energy transfer, could be a mechanism of energy funneling or that energy funneling uncharacteristically slow in n = 2:3 CsPbBr3. To suit particular applications, we note that the funneling efficiency and kinetics can be modified according to the overlap between the donor emission and acceptor absorption, which depends on the elemental composition and the thickness of the 2D perovskite. Ultimately, the disparity in the photodynamics between the quasi-2D CsPbBr3 imitation sample and noninteger 2D CsPbBr3 affirms that the implication of structural heterogeneity in semiconductors is intramaterial energy funneling.

Photoexcitation transfer has been identified at interfaces of semiconductors with lateral band alignments, such as ionic heterojunctions and conventional heterostructures in composition, including core−shell and core/crown nanostructures. However, ionic heterojunctions decompose via ion diffusion and lattice-mismatch strain at interfaces of conventional heterostructures can poison charge transfer and optical properties. In conventional heterostructures, the charge transfer state is typically a trap state that arises from strain at the interface of the constituent semiconductors, which has motivated the search for interfaces with an optimal lattice matching for device applications. Further investigation of the n = 2:3 interface was warranted given that the strain behavior in this kind of heterostructure with constant elemental composition has not been explored before nor the consequences on energy funneling. Atomic resolution TEM was used to probe the n = 2:3 CsPbBr3 lattice for a possible out-of-plane or in-plane structural interface. The pure n = 2 and n = 3 CsPbBr3 nanosheets had a cubic lattice with a Pb−Br bond distance of 3.1 ± 0.1 Å, respectively (Figures S3 and S6). The n = 2:3 CsPbBr3 also had a cubic lattice with a Pb−Br bond distance of 3.1 ± 0.1 Å. Therefore, a unique, out-of-plane structural interface was not discernible between the n = 2 and n = 3 phases (Figure 4a), confirming that n = 2 and n = 3 phases were attached coherently, with virtually no lattice strain between them. There were no Bragg peaks in the diffraction pattern local to the interface (Figure 4e). The selected diffraction pattern of the n = 2:3 CsPbBr3 lattice for the noninteger 2D perovskite and that of the selected diffraction pattern local to the interface (Figure 4e). The selected diffraction pattern of the n = 2:3 CsPbBr3 was a single cubic pattern, which confirmed that the growth of the third layer of octahedra on the n = 2 phase was perfectly epitaxial and disproved the existence of an in-plane interface. Structural modeling of the n = 2:3 CsPbBr3 did not reveal an in-plane or out-of-plane interface either and showed that the crystal structure remained cubic throughout (Figure 4f). The implied n = 2:3 interface across which energy funneling occurred was thus defined solely by confinement, which also makes this study among the first to demonstrate highly efficient transfer without the influence of strain in a heterostructure.

Energy funneling has been desired among 2D semiconductors to improve their performance in devices while maintaining the other advantageous properties gained from the reduced dimensionality. While efforts so far have been focused on solving this problem using quasi-2D perovskite, we have successfully demonstrated that energy funneling can also occur in the CsPbBr3-based noninteger 2D perovskite, which is the first of a new class of semiconductor. The structural transformation reaction demonstrated that a variety of noninteger 2D perovskites could be prepared with properties that depend on the noninteger character, such as the absorption and emission profile, PLQY, PL decay kinetics, and transfer efficiency. Unlike conventional heterostructures, the interface between phases of the noninteger 2D CsPbBr3 was structurally coherent and virtually strain-free, which may permit the high transfer efficiencies desired for device applications. The overall findings presented herein necessitate further investigation and review of semiconductors with thickness heterogeneity in the confined regime and expansion of the noninteger concept to other materials and devices.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.3c03058.

Information about the methods, additional discussion on graphene encapsulation and the structural transformation reaction, characterization of n = 2 and n = 3 CsPbBr3 nanosheets, detailed steady-state and time-resolved optical spectroscopy data of all noninteger structures, computational results, and raw atomic-resolution TEM data (PDF)

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A.M.O., M.G., and P.Y. conceived the idea and designed the research. D.W. performed the kinetic modeling and electronic structural calculations. D.W. and E.R. provided theoretical insight into the work. C.S. supervised and trained A.M.O. on the TEAM 0.5 microscope, and C.O. provided the scripts for processing images taken on the TEAM 0.5 microscope. J.J. and M.C.F. contributed intellectually through numerous discussions. T.M. trained A.M.O. on fsTA spectroscopy. P.Y. supervised the work. All authors contributed to writing the manuscript.

Notes

The authors declare no competing financial interest.

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