Lead-Halide Perovskites Meet Donor−Acceptor Charge-Transfer Complexes

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ABSTRACT: Low-dimensional lead halide hybrid perovskites are nowadays in the spotlight because of their improved stability and extensive chemical flexibility compared to their 3D perovskite counterparts, the current challenge being to design functionalized organic cations. Here, we report on the synthesis and full characterization of a perovskitoid hybrid (a perovskitoid) where the 1D lead iodide layout is patterned with a donor−acceptor charge transfer complex (CTC) between pyrene and tetracyanoquinodimethane, with a chemical formula of (C20H17NH3)PbI3·(C12H4N4). By combining multiple structural analysis and spectroscopic techniques with ab initio modeling, we show that the electronic, optical, and charge-transport properties of the hybrid materials are dominated by the organic CTC, with the inorganic backbone primarily acting as a template for the organization of the donor and acceptor molecules. Interestingly, time-resolved microwave conductivity (TRMC) measurements show an enhanced photocurrent generation in the 1D hybrid compared to the pure organic charge-transfer salt, likely associated with transient localization of the holes on the lead-iodide octahedra. This observation is in line with the close energy resonance between the valence crystal orbitals of the lead-iodide lattice and the frontier occupied molecular orbitals of pyrene predicted by the DFT calculations. Therefore, it paves the way toward the design of new hybrid low-dimensionality perovskites offering a synergic combination of organic and inorganic functionalities.

INTRODUCTION

Lead halide hybrid perovskites have received significant attention over the past decade as semiconductor materials, namely, for photovoltaic applications.1−3 These materials possess a peculiar crystalline structure with an ABX3 stoichiometry, where X is a halogen atom (I−, Br−, Cl−), B an inorganic cation (Pb2+, Sn2+, ...), and A an organic cation (CH3NH3+, NH2CHNH2+). The structure consists of a three-dimensional network of corner-shared BX6 octahedra with A cations located in the cavities of this framework.

While these 3D hybrid materials can be easily prepared from solutions of precursors and display impressive optoelectronic properties, e.g., with a certified power conversion efficiency of 24.2%,4 they suffer from degradation issues mainly associated with ionic diffusion in the inorganic network.5 This has driven the community to design low-dimensional hybrid perovskites that, in addition to higher stability, offer structural versatility, prompting their use as active components in a wide range of optoelectronic applications.6−13 Indeed, the constraint that the organic cation should obey a strict geometrical rule to fit in the...
cavities created by the inorganic octahedral network in the 3D case is alleviated when going to lower-dimensionality materials. As a result, by properly tuning the nature of the organic cation and/or the synthesis protocol, it is now possible to engineer lead halide organic−inorganic hybrids where the inorganic framework formed by the PbI$_6^{4−}$ octahedra spans the full dimensionality range from 3D to 2D, 1D, or 0D.

Layered perovskite materials containing alkylammonium cations have been scrutinized in the past. While the study of these provides useful insight into the effects of dielectric confinement and local lattice distortions, the range of tunability offered by inert organic cations is limited. An attractive strategy to convey additional functionalities to the resulting hybrid materials is to replace the inert cations by electroactive organic conjugated molecules. Notable work in this direction includes the work of Evans et al. and Maughan et al., who showed that synergic effects between the organic and inorganic components of hybrids can be achieved by making use of the process of charge transfer. In the case of the TTF containing hybrids of Evans et al., charge transfer between a neutral organic molecule (TTF) and a radical cation (TTF$^+$) in the hybrid occurs. For the tropyllium containing hybrid of Maughan et al., iodine-to-tropyllium charge transfer was suggested.

Organic charge-transfer complexes (CTCs) have distinct optical fingerprints and have been extensively studied over the last 50 years for their interesting properties, including ambipolar charge transport, metallicity, photoconductivity, and ferroelectricity. In this context, we have recently shown that organic donor−acceptor charge-transfer complexes can be combined with 2D layered hybrid perovskites. 2D perovskites with a nominal chemical formula (PyrC$_4$NH$_3$)$_2$Pbl$_4$, 2(TCNQ) or (PyrC$_4$NH$_3$)$_2$Pbl$_2$2(TCNB) with PyrC$_4$ being pyrene-C$_4$H$_8$ were obtained. The acceptors are incorporated into the organic layer through the formation of charge-transfer complexes with donors that are tethered to the inorganic framework via an alkylammonium chain. The acceptor molecule is therefore intercalated into the organic layer of the hybrid. In this regard, our work builds upon previous work on the intercalation of small molecules into the organic layer of 2D layered hybrids based on weak interactions, e.g., fluoroaryl−aryl interactions. We note that our approach differs from the works of Evans et al. and Maughan et al., since we introduce separate organic donor and acceptor molecules that self-assemble into charge-transfer complexes in the organic layer of the hybrid. Introducing separate donor and acceptor molecules imparts a high potential tunability to our new type of hybrids.

Unfortunately, we were at that time not able to resolve single-crystal structures of these 2D hybrids, preventing detailed structure−property relationships to be unraveled. Here, we report on our ongoing efforts toward the synthesis of hybrid materials, in which the organic component contributes to the charge transport, combining in a synergic way the complementary properties of their organic and inorganic components. More specifically, we synthesized a novel 1D hybrid composed of wires of face-sharing PbI$_6^{4−}$ octahedra separated by bilayers containing the CTCs, with a chemical formula of (C$_{20}$H$_{17}$NH$_3$)PbI$_3$·(C$_{12}$H$_4$N$_4$), which we rename as (PyrC$_4$NH$_3$)PbI$_3$·(TCNQ) (Figure 1a). A carbon chain of 4 carbon atoms ended by an ammonium group for the anchoring to the inorganic part is attached. This alkyl chain (which includes the positive charge of the cation via the ammonium...
group) is exclusively located on the pyrene molecule. The chains are running parallel to each other.

In addition to structural information, XPS valence band and TRMC measurements, combined with DFT computational investigations, provide detailed information about the electronic structure and the charge generation and transport properties of these novel hybrid materials. Altogether, our combined theory–experiment study conveys the message that (i) the inorganic lattice mostly acts as a template for the local organization of the organic conjugated molecules, and (ii) the close energy resonance between the occupied levels on the inorganic and organic parts improves photoinduced charge generation.

■ RESULTS AND DISCUSSION

Crystal Structure, Stability, and Film Morphology. The crystal structure of the mixed organic–inorganic hybrid has been elucidated using single-crystal X-ray diffraction (XRD) (Figure 1a) and was found to be monoclinic. The powder X-ray diffraction (PXRD) pattern of the bulk sample of crystals compared well with the simulated pattern from single-crystal XRD (Figure S1). The crystal structure of the pyrene-TCNQ organic CTC was taken from the literature, and a representation is shown in Figure 1b,c.41 The experimental PXRD pattern of our grown pyrene-TCNQ crystals matched well with the simulated pattern based on the crystal structure from literature (Figure S2). The 1D hybrid perovskitoid shows very good thermal stability in ambient atmosphere. The compound does not degrade into a new phase until a temperature of ∼190 °C, as assessed based on temperature-controlled XRD measurements (Figures S13 and S14). Films for TRMC were prepared via drop-casting followed by thermal annealing. The morphology of the resulting films was investigated using SEM measurements (Figure S4). As can be seen, no clear pinholes are present in the film. The absence of significant pinholes is the most important criterion in terms of morphology for the analyses carried out in the current study. The morphology of the films could potentially be optimized by using different processing techniques (e.g., gas quenching or antisolvent dripping), but this is outside of the scope of the current study.

Electronic Structure and Optical Properties. Optical absorption spectra measured by photothermal deflection spectroscopy for the 1D hybrid perovskitoid and the organic CTC crystals are reported in Figure 2 (see also absorption spectra in Figures S5 and S6). While the shapes of the two spectra are similar, pointing to the similar nature of the involved electronic excited states, a clear shift of the sharp absorption onset from ∼1.3 eV for the organic CTC to ∼1 eV for the hybrid is observed. Such a spectral displacement might originate from various (combined) contributions, namely, a different molecular packing of the CTC or direct full or partial involvement of the lead halide inorganic skeleton through hybridization with the organic chromophores.

We resort to DFT electronic structure calculations to address the origin of the observed differences in electronic structure and optical properties between the organic and the hybrid crystals. As a first step, an in-depth study was performed to analyze the sensitivity of the results against details of the modeling methodology. It has indeed been widely documented that the inclusion of spin–orbit coupling (SOC) effects and an accurate treatment of electronic exchange and correlation energy are key to a quantitative description of the electronic structure of lead halide perovskites.42,43 Details about the modeling approach are provided in the Supporting Information (Table S1, Figure S7). In a nutshell, after realizing that SOC does not influence the lowest unoccupied energy levels (localized on the TCNQ acceptors), we selected (unless otherwise specified) plane-wave DFT calculations in combination with a hybrid functional (PBE0), as implemented in the Quantum Espresso package suite.44 Using this methodology, we obtain an electronic bandgap of 1.93 eV for the 1D perovskitoid system, to be compared with 2.16 eV for the pure CTC crystal. While compared to the PDS data the theoretical values are expectedly blue-shifted, namely, by the exciton binding energy but also inherent errors associated with the choice of DFT functionals, the calculations do however correctly reproduce the observed spectral shift (∼0.23 eV calculated against ∼0.3 eV measured) between the two crystals.

Turning to the electronic structure energy diagrams displayed in Figure 3a, we find from the analysis of the partial density of states (DOS) that the band-edge levels in the 1D hybrid perovskitoid are fully confined on the organic part, namely, with the conduction band-edge (CBE) dominated by TCNQ empty orbitals and the valence band-edge (VBE) governed by pyrene occupied orbitals. It is important to stress, however, that the lead-iodide inorganic skeleton contributes with occupied crystalline orbitals that are located in close energy proximity (within ∼0.1 eV) to the VBE—in contrast, the corresponding PbI unoccupied crystalline orbitals are located at significantly higher energies than the CBE. In addition, inspection of the partial DOS reveals no or very limited hybridization between the organic molecular and inorganic crystalline orbitals, which would materialize through the presence of a partial contribution of lead and iodine atoms at the VBE (and CBE) energy. In view of the small energy difference, this absence of wave function mixing must originate from a very weak electronic coupling matrix element, likely resulting from the poor electronic contact between the inorganic scaffold and the conjugated chromophores. Looking retrospectively at the optical absorption data, we thus rule out hybridization as a mechanism for the observed red shift and rather invoke a reorganization of the donor and acceptor
molecules templated by the inorganic layout. This is consistent with the (slight) change in the ground-state charge transfer density when going from the organic CT salt (with an effective charge per TCNQ molecule calculated to be \( \sim 0.15 \) e\text{f} using a Bader charge analysis\( ^{45} \) reported in Supporting Information Table S2) to the lead halide CT hybrid (where the charge per TCNQ amounts to \( \sim 0.21 \) e\text{f}). We further note that the electronic band gap between the lead halide only crystalline orbitals is close to 4 eV at the level of theory considered. This is much higher than the corresponding bandgaps calculated at the same level of theory for 2D lead halide perovskites, highlighting the important role of dimensionality on the electronic structure of these materials.

Figure 3b displays both the predicted full (and partial) SOC-corrected DOS together with the simulated XPS spectrum, where relative intensities have been obtained through the density of states by the relative photoionization cross sections. While this approach should be considered with caution (as delocalized band states are obviously distinct from their contributing atomic orbitals), it explains the fact that occupied molecular orbitals arising from the organic components are hidden in the background noise of the XPS spectra because of their low ionization cross sections. With this caveat, we can proceed with the comparison of the theoretical results to the experimental valence band electronic structure, here probed using XPS (as a more bulk sensitive technique, information depth > 5 nm) instead of UPS (as a more surface sensitive technique, information depth about 1 nm). The measured and predicted valence band spectra are dominated by the iodine atoms at low binding energies, while the SOC-split 5d orbitals of lead contribute a twin peak at higher binding energies (below \(-10\) eV). It is interesting to see that a better match with experiment regarding the absolute energies of the lead orbitals is obtained considering the bare (unscaled) DOS, which could indicate partial hybridization or simply reflect uncertainties in the computed orbital energies. Unfortunately, the limited spectral resolution and the low cross sections of the sp2 carbon atoms from the pyrene molecules do not allow the theoretical finding that the VBE is mostly sourced by the organic molecules to be ascertained. As demonstrated below, this is indirectly supported by charge transport measurements.

**Charge Transport Properties.** Table 1 reports the calculated effective masses along high-symmetry paths for (i) the hybrid 1D organic–inorganic perovskitoid; (ii) the pure organic CT salt; and (iii) a hypothetical system built by removing all lead and iodine atoms from the crystal structure of the hybrid material and (to balance the charges) substituting the nitrogen atoms of the ammonium groups with carbons. This aims at mimicking a pure template effect where the inorganic lattice would participate not directly to the transport properties but eventually indirectly through a reorganization of the spatial rearrangement of the donor and acceptor molecules in comparison to the CT salt. As a matter of fact, in all relevant directions, the effective masses predicted by the DFT calculations are typical for organic molecular crystals with weak band dispersion. The differences observed between the hybrid and the organic crystal primarily stem from a template effect, as confirmed by comparing the relatively similar effective masses computed for the real hybrid and hypothetical structures. This template effect materializes mostly as differences in intermolecular distances along TCNQ and pyrene molecular rows in the \( b \) direction for the CTC crystal and the corresponding \( c \) direction for the 1D hybrid perovskitoids; see Figure 1c. More specifically, there is a reduced herringbone angle, or similarly a smaller longitudinal molecular translation, between successive donor (acceptor) molecules in the hybrid structure. While this is still far from being ideal from an intermolecular spatial overlap perspective, there is improved \( \pi-\pi \) stacking interaction between the organic molecules in the perovskitoid 1D systems compared to the pure CTC crystal.

### Table 1. Effective Masses of Electrons and Holes for the CTC Crystal and the 1D Hybrid (PyrC4NH3)PbI3(TCNQ) Crystals\(^{44} \)

<table>
<thead>
<tr>
<th>Direction</th>
<th>Organic CTC Pyrene-TCNQ</th>
<th>1D CTC Pyrene-TCNQ</th>
<th>1D Hybrid (PyrC4NH3)PbI3(TCNQ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( (001) )</td>
<td>( m_{\text{e}+} ) 10.42 ( m_{\text{h}+} ) 3.29</td>
<td>( m_{\text{e}+} ) 4.31</td>
<td>( m_{\text{h}+} ) 4.63</td>
</tr>
<tr>
<td>( (010) )</td>
<td>( m_{\text{e}+} ) 9.90 ( m_{\text{h}+} ) 2.11</td>
<td>( m_{\text{e}+} ) ( m_{\text{h}+} )</td>
<td>( m_{\text{e}+} ) ( m_{\text{h}+} )</td>
</tr>
<tr>
<td>( (001) )</td>
<td>( m_{\text{e}+} ) 6.66 ( m_{\text{h}+} ) 13.85</td>
<td>( m_{\text{e}+} ) 7.37 ( m_{\text{h}+} ) 2.35</td>
<td>( m_{\text{e}+} ) 4.33 ( m_{\text{h}+} ) 1.62</td>
</tr>
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</table>

\(^{44}\)We have also considered a hypothetical (1D CTC) obtained by removing the inorganic layout from 1D hybrid perovskitoids structure in order to highlight the template effect. Values in bold emphasize the reduced effective masses when going from the CTC to the 1D hybrid material.
which is driven by the inorganic layout. As a result, the electron effective masses along the high-mobility direction (c) are significantly smaller, especially for electrons, in the hybrid material with respect to the pure salt (for which b is the high-mobility direction), a difference that reflects the more substantial wave function overlap between the molecular orbitals in the presence of the inorganic template. Yet, the absolute values of the effective masses remain relatively large for both holes and electrons, irrespective of the direction; hence, we expect relatively poor charge transport mobility for both organic and hybrid crystals.

This is fully confirmed by time-resolved micro-conductivity (TRMC) measurements that probe the change in conductivity due to mobile charge carriers using high-frequency microwaves. The charge carriers can be generated either by a high energy electron pulse (pulse-radiolysis TRMC) or by a laser (photoconductivity TRMC). We have performed PR-TRMC measurements to monitor the temperature-dependent mobility of photogenerated free charge carriers in the two materials. The formation of free charge carriers is assured by the effective irradiation energy ($\sim 20$ eV) from the electron pulse. The results plotted in Figure 4a show modest ($10^{-3}$–$10^{-2}$ cm$^2$ V$^{-1}$ s$^{-1}$) and similar mobility values over a broad temperature range in the pure organic salt and in the 1D organic–inorganic hybrid crystals. In addition, similar recombination dynamics of the charge carriers for both materials are obtained; see Supporting Information (Figure S9 and S10).

To complement these studies, photoconductivity TRMC measurements have also been performed with laser excitation at different wavelengths (excitonic peak at $\sim 500$ nm and charge-transfer state at $\sim 600$ nm). In these measurements, the photoconductivity signal obtained is the product of the

![Figure 4](https://example.com/figure4.png)

**Figure 4.** (a) Charge carrier mobility as a function of temperature for the two investigated systems: 1D hybrid (PyrC$_n$NH$_3$)$_n$PbI$_3$ (TCNQ) and the CTC crystal pyrene-TCNQ. (b) Photoconductivity TRMC measurements of 1D hybrid (PyrC$_n$NH$_3$)$_n$PbI$_3$ (TCNQ) and the CTC crystal pyrene-TCNQ excited at the CT states ($\sim 600$ nm).
acid (57 wt % aqueous solution) were bought from Fisher Scientific. All chemicals were used without further purification. The dry dimethylformamide (DMF) that was used to make the precursor solutions and dry tetrahydrofuran (THF), which was used for the reactions, were obtained from our in-house solvent-purification system (MBRAUN SPS-800). All other solvents were purchased from Fisher Scientific.

The PyrC4NH3I salt was synthesized as detailed in a previous publication.10

**Single Crystal Growth.** The single crystals of (PyrC4NH3)2PbI4(TCNQ) were grown using an antigrowing vapor-assisted crystallization approach10 in which the components are dissolved together in a good solvent (gamma-butyrolactone; GBL) and dichloromethane (DCM) antigrowing slowly diffuses into the GBL solution. Specifically, PyrC4NH3I (0.108 mol/L), TCNQ (0.108 mol/L), and PbI2 (0.108 mol/L) were dissolved together in gamma-butyrolactone by stirring at 50 °C for 15 min. The precursor solution was filtered through a syringe filter (0.45 μm). The precursor solution (0.5 mL) was transferred to a small glass vial. The small vial (5 mL volume) was capped off with aluminum foil. A small hole was made in the aluminum foil. The small vial with the aluminum foil was put in a larger glass vial (20 mL volume). A small amount of dichloromethane (2 mL) was injected in the gap between the two flasks, and the larger flask was capped off with a plastic cap and parafilm. The vials were left undisturbed at room temperature. After 1 week black crystals suitable for single-crystal X-ray diffraction were harvested. These crystals were washed three times with dry dichloromethane and were subsequently dried under reduced pressure at room temperature.

The crystals of the organic CTC pyrene-TCNQ were grown by dissolving equimolar amounts of pyrene (0.025 mol/L) and TCNQ (0.025 mol/L) in tetrahydrofuran (THF) by stirring at 40 °C until full dissolution. The solution was allowed to cool down to room temperature and was filtered through a syringe filter (0.45 μm). Crystals were obtained by evaporation of the THF at room temperature.

**Thin Film Deposition.** Stoichiometric amounts of PyrC4NH3I (0.3 mol/L), TCNQ (0.3 mol/L), and PbI2 (0.3 mol/L) were dissolved in dry DMF by stirring at 50 °C for 30 min. The precursor solution was filtered through a syringe filter (0.45 μm) before use.

Quartz substrates were cleaned through consecutive sonication steps in a series of solvents (detergent water, deionized water, acetone, isopropanol) for 15 min each, followed by a UV–ozone treatment for 15 min. Films were obtained by drop-casting (~30 μL on a 2.5 cm × 2.5 cm substrate) on a quartz substrate. The films were annealed at 110 °C for 15 min. Films were placed in a polyether ether ketone filled glovebox. Photoconductivity TRMC measurements quantify the change in conductivity [micro-wave (8–9 GHz) power] upon pulsed excitation (repetition rate 10 Hz) due to free mobile charge carriers. The change of microwave power is related to the change in conductivity before and during the photoductive measurements, and the samples were kept in an inert nitrogen environment to prevent degradation by exposure to moisture.

**THEORETICAL METHODS**

**Electronic Properties.** Density functional theory calculations with periodic boundary conditions have been performed with the Quantum Espresso suite program10 for the electronic properties (and Bader analysis10) using a plane-wave/pseudopotential formalism. We adopted a norm-conserving pseudopotential, with a cutoff of 40 Ry for the expansion of the wave function and a correction for the van der Waals interactions (Grime DFT-D2 method). Different functionals (GGA and Hybrids) were applied: PBE, PBE0, and HSE. In the case of the PBE functional, we also performed the same calculations with and without a spin–orbit correction. We kept the same k-points mesh for all PBE calculations, 4 × 4 × 1 for the organic crystal (pyrene-TCNQ) and 4 × 1 × 4 for the 1D perovskitoid. When using hybrid functionals, the calculations were limited at the gamma point of the first Brillouin zone, because of the calculation costs.

**Charge Transport Properties.** Effective mass calculations have also been performed with Quantum Espresso at the PBE level. We instead resorted to the CRYSTAL suite program (CRYSTAL14, 50) for effective mass calculations using hybrid functionals, namely, the PBE0 functional, as these are not implemented in Quantum Espresso. For consistency between the two sets of results, we paid attention to keep the same cell parameters, the same number of atoms explicitly taken into account and the same k-points mesh for the single point calculations.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.chemmater.9b01289.

Crystallographic information file (CIF)

Experimental details of the single crystal XRD measurement, powder XRD, comparison between 2D and 1D hybrids containing CTCs, scanning electron microscopy, absorption spectra, theoretical methodological study, Bader analysis, partial density of states, pulse-radiolysis TRMC, photoconductivity TRMC, and thermal stability of the hybrid (PDF).
Author Contributions
The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes
The authors declare no competing financial interest.

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References
(7) Saida
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