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Introduction

Organic solar cells (OSCs) have undergone a very encouraging development in past few decades,¹⁻⁴ however currently still suffering from inferior power conversion efficiencies (PCEs) with respect to their inorganic counterparts (like Si, GaAs, *etc.*).^{5,6} The greatest challenge of organic light-harvesting materials lies on their structural disorders or weak intermolecular interactions caused by the flexibility of large molecular skeletons. Unlike inorganic semiconductors that are formed by

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A rare case of iodinated non-fullerene acceptors for high-performance organic solar cells without posttreatments[†]

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Conventional halogenations such as fluorination, chlorination and bromination on non-fullerene acceptors (NFAs) generally contribute to a dramatic improvement in overall photovoltaic performance of organic solar cells (OSCs). Despite the potentially enhanced intermolecular interaction caused by iodine atoms with increased polarizability, iodinated NFAs are quite rare due to the high-reactive activity of iodine. Here, an iodinated acceptor (CH45) is constructed, rendering a first-class efficiency of 18.15% for OSCs without any extra post-treatments, attributed to the enlarged molecular polarizability and dielectric constant, and further reduced exciton binding energy of CH45. Moreover, the working mechanism of 1-chloronaphthalene additive to greatly improve fill factors of CH45-based OSCs is also revealed at single-crystal levels. By successfully establishing such a rarely iodinated but high-performance molecular platform, more delicately iodinated light-harvesting molecules with unexpected properties (such as significantly enhanced intermolecular non-covalent actions) will be inspired to further achieve high-efficiency OSCs without tedious post-treatments.

a spatial lattice with clear periodic structures through covalent bonds, organic semiconductors are only bound together by van der Waals forces, hydrogen bonds, π - π interactions, *etc.*, thus resulting in large molecular packing distances, small overlaps of p-orbital electron clouds, difficult exciton/electron delocalization and more crystal defects. All such disadvantages can lead to a small exciton radius (<1 nm), large binding energy $(E_{\rm b} >$ 0.3 eV; <0.01 eV for inorganic semiconductors),7,8 inferior exciton diffusion through Förster and Dexter energy transfer,9 hindered electron hopping10 and also too many non-radiative recombination centers in aggregates of organic lightharvesting materials. In order to address this issue, lots of post-treatment strategies (such as thermal annealing, solvent vapor annealing, etc.)11-13 have been applied in nearly all the high-efficiency OSCs,6,14,15 with the aim of achieving more compact/ordered molecular packings or better nanoscale film morphology. However, the widely applied post-treatments will make the morphology evolution in the active layer unpredictable and uncontrollable, more importantly, limiting the economic feasibility of OSCs especially for large-scale production.16,17 Therefore, developing light-harvesting material systems with already superior and thermodynamically stable nanoscale morphologies overlapped together once formed into blended films is essential, which is most likely achieved by significantly enhancing the intrinsic intermolecular packing strengths.

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Fig. 1 (a) Chemical structures of CH4 and CH45. (b) Regularity of atomic radius, crystallinity and polarizability. (c) Isotropic polarizabilities of CH4 and CH45, including phenazine central units with different halogenations. (d) Dielectric property (dielectric constant ε_r) of CH4 and CH45. (e) Derived E_b values based on the analysis of temperature-varying photoluminescence (PL) spectra.

The great structural complexity of organic molecules and intrinsically metastable micromorphology of blended films^{16,18-20} make the construction of OSCs with both high performance and free of post-treatments very challenging. Despite the lack of structure–activity relationships,²¹⁻²³ a relatively clear clue has been revealed that not only efficient π - π stackings but also sufficient noncovalent interactions, like halogen bonding *etc.*, should be considered, if much enhanced

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intermolecular interactions and thermodynamically stable morphologies are expected.24,25 It is known that F or Cl involved in secondary interactions (X···H, X···S, X··· π , etc.) can usually render more compact and ordered molecular packing of lightharvesting molecules.24,26 Especially, F or Cl, featuring small atomic radius and large electronegativity, have been extensively applied on end units of non-fullerene acceptors (NFAs), leading to an encouraging increase of PCE of OSCs.24,26,27 Note that the homomorphic iodine(I) atom possesses even more unique advantages than F or Cl, except for their similar abilities in tuning energy levels and absorption of NFAs: (I) the much enlarged atomic radius (F, 71 pm; Cl, 99 pm; Br, 114 pm; I, 140 pm) endows iodine with dramatically increased polarizability, which is conducive to increase the relative dielectric constant and reduce the exciton binding energy $(E_{\rm b})$ of NFAs;²⁸ (II) in light of the enlarged atomic radius but decreased electronegativity (F, 4.0; Cl, 3.0; Br, 2.8; I, 2.7) of iodine,²⁹ enhanced σ -hole interactions, halogen bonds and π /p-electron orbital overlap are expected due to the loose outmost electron cloud, consequently contributing strengthening of intermolecular to interactions;30-32 (III) iodide usually features enhanced crystallinity compared with fluoride and chloride, which could result in a better molecular crystalline ordering in theory.^{30,31,33} Given so many unique merits of iodine, the seldom employed iodization on NFAs and even polymeric donors really surprises us. Two potential factors as follows may account for that: (I) the conventional halogenation of NFAs occurs on end units, however, the large atomic radius of iodine may hinder the efficient intermolecular packings of NFAs through end units,18 especially considering that the π - π stacking distance between two neighboring and packed end units is only 0.32-0.36 nm;^{34,35} (II) the high reactive activity of iodine, especially in C-C and C-N cross-coupling reactions, makes it easy to remove during the construction of novel molecular platforms.³⁶ In light of the contrasting advantages and disadvantages of iodinated NFAs, high-performance OSCs free of post-treatments can be most promising if greatly maximizing the merits of iodine whilst circumventing its evident weaknesses.

The most distinctive advantage of our recently explored CHseries NFAs lies in the sufficient structural modification sites on the molecular skeleton of central units, which has afforded a blowout PCE growth of resulting OSCs.20,24,27 Particularly, a huge possibility for molecular packing optimization through quite a minor structural modification (like halogenation) on central units of CH-series NFAs has been manifested,21 demonstrating the powerful capacity in reducing $E_{\rm b}$, increasing relative dielectric constant, facilitating charge generation/ transport, etc.37 This unique property of CH-series NFAs is really exciting, because the hitherto inaccessible iodization of other NFAs such as ITIC, 38 F39 and Y-series molecules 40 could be easily achieved if introducing iodine on central units rather than end groups. In this way, the advantages of iodine could be fully exerted without hindering the crucially important intermolecular packings through end units simultaneously. Bearing these thoughts in mind, an A-D-A type NFA (CH45, Fig. 1a), featuring a rare but distinctive iodization on the phenazine central unit, has been designed and constructed. The delicate

iodization on the central unit increases molecular polarizability and dielectric constant, further reducing the exciton binding energy of CH45. Moreover, by utilizing a widely employed 1chloronaphthalene (CN) additive, molecular crystalline ordering and intermolecular interaction in CH45 films were further enhanced, thus contributing to a first-class efficiency of 18.15% for CH45-based OSCs without any extra posttreatments. More importantly, based on a single-crystal analysis, the working mechanism whereby CN greatly improves the FFs of OSCs was also revealed at molecular levels.

Results and discussion

Fig. 1a displays the chemical structures of CH45 and its counterpart CH4 (ref. 27) (see Scheme 1 and S1[†] for synthetic routes). Similar to that of CH4, a favorable A-D-A architecture can be verified (Fig S1[†]) for CH45, affording improved light harvesting, charge generation/transport and eventual PCEs.41 The decomposition temperature (~322 °C) of CH45 is near to that of other highly efficient NFAs (Fig. S2[†]),^{24,40,42} indicating the outstanding thermal stability can be maintained even when introducing highly reactive iodine atoms on the molecular skeleton. As we have mentioned above, iodine possesses a larger atomic radius and greater polarizability than hydrogen or its homomorphic elements (F, Cl, Br) (Fig. 1b), which may bring about increased molecular polarizability and relative dielectric constant (ε_r), further reducing the exciton binding energy (E_b) in theory.⁴³ As shown in Fig. 1c, the isotropic polarizability of the phenazine central unit with iodization displays a maximum value of >200 compared to phenazine central unit (146.3) and its halogenated derivatives (149.2, 179.6 and 185.3 for fluorination, chlorination and bromination, respectively). Note the easily polarized property of iodine is also maintained in CH45, indicated by a larger isotropic polarizability of 1388.5 than 1328.1 for CH4. Meanwhile, the dipole moment of CH45 is 2.01 Debye, much larger than that of 1.63 Debye for CH4 (Fig. S3[†]). As a result, CH45 neat film affords a larger ε_r of 2.26, comparing to that of 1.89 for CH4 (Fig. 1d) and this tendency can be also kept in blended films (2.69 for PM6:CH4 and 3.98 for PM6:CH45). Note that the increased ε_r of organic semiconductors is conducive to achieve efficient photodynamics and thus render an upgraded OSC with improved FF and J_{SC} .¹² It is well known that E_b of organic semiconductors is about 0.1-1 eV, larger than that of ~0.01 eV of inorganic semiconductors by several orders of magnitude. This should be one of reasons for the relatively larger energy loss and inferior PCEs of OSCs with respect to inorganic solar cells.28,44 After iodination on the phenazine unit, CH45 neat film possesses a significantly decreased experimental $E_{\rm b}$ (~61 meV) compared with that of ~110 meV for CH4 (Fig. 1e, S4 and S5[†]),⁴⁵ which agrees with the trend of theoretical calculations in the gas phase (1.66 and 1.61 eV for CH4 and CH45, respectively; Table S1^{\dagger}). The dramatically larger calculated E_b than that in the solid phase is plausible, given that E_b is greatly affected by solid-state polarizations and intermolecular electronic interactions of organic molecules.28,44 However, what truly excites us is that E_b reduces greatly with halogenation on the phenazine unit, making it possible to reach an $E_{\rm b}$ comparable to that of



inorganic semiconductors through delicate structural optimization.

CH4 and CH45 possess maximum absorption wavelengths of 752 and 742 nm, respectively, in dilute chloroform solutions (Fig. 2a and Table S2†). The blue-shift absorption of CH45 compared to that of CH4 should attributed to the substitution of iodine on the central unit, which decreases the electrondonating feature of central donor of NFAs. Moreover, an obvious bathochromic shift of absorption (66 nm for CH4 and 74 nm for CH45) can be observed from solutions to solid films with the maximum absorption peaks located at 818 nm for CH4 and 816 nm for CH45, suggesting strong intermolecular interactions, especially for CH45.⁴⁶ The experimental lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) are -3.84/-5.64 eV for CH4 and -3.85/-5.66 eV for CH45 (Fig. 2b and S6†), agreeing well with DFT calculated results (Fig. S7†). Note that the considerable LUMO/HOMO offset between polymeric donor PM6 (ref. 47) (Fig. 2b and S8†) and CH45 can enable more efficient charge transfer with a larger driving force in theory.



Fig. 2 (a) Normalized absorption spectra of PM6 (film), CH4 (solution), CH45 (solution), CH4 (film) and CH45 (film). (b) Energy level diagram of PM6, CH4 and CH45 derived from CVs. (c) *J*–*V* curves of OSCs based on PM6:CH4 and PM6:CH45 with/without CN. (d) EQE plots of OSCs based on PM6:CH4 and PM6:CH45 with/without CN.

Table 1 Summary of device parameters for optimized OSCs^a

Active layer	$V_{\rm OC}$ (V)	$J_{\rm SC}~({ m mA~cm^{-2}})$	Calc. $J_{\rm SC}^{b}$ (mA cm ⁻²)	FF (%)	PCE (%)
PM6:CH4	$0.908~(0.904\pm0.003)$	$24.67~(24.26\pm0.21)$	24.11	$66.4~(66.7\pm0.6)$	$14.87~(14.61\pm0.15)$
PM6:CH45 PM6:CH45 (CN)	$\begin{array}{c} 0.912 \; (0.908 \pm 0.003) \\ 0.910 \; (0.910 \pm 0.002) \end{array}$	$\begin{array}{c} 25.34 \ (25.47 \pm 0.17) \\ 25.57 \ (25.50 \pm 0.21) \end{array}$	24.52 24.97	$\begin{array}{c} 73.7~(72.8\pm0.4)\\ 78.0~(77.6\pm0.5)\end{array}$	$\begin{array}{c} 17.03 ~ (16.85 \pm 0.12) \\ 18.15 ~ (18.0 \pm 0.06) \end{array}$

^{*a*} Statistical (15 independent devices) and optimal results are listed in parentheses and outside of parentheses, respectively. ^{*b*} Current densities calculated from EQE plots.

In order to evaluate the photovoltaic performance of NFAs with iodination on the molecular backbone, OSCs with a conventional structure were fabricated by blending NFAs with a widely employed donor, PM6. A moderate PCE of 17.03% along with a $V_{\rm OC}$ of 0.912 V, $J_{\rm SC}$ of 25.34 mA cm⁻² and FF of 73.7% was afforded by as-cast OSCs based on CH45 (Table 1), displaying an overall improvement over CH4-based OSCs. When additive (CN) was introduced into the precursor of PM6:CH45. a much higher PCE of 18.15% was further reached with a comparable $V_{\rm OC}$ of 0.910 V, slightly higher $J_{\rm SC}$ of 25.57 mA cm⁻², but significantly improved FF of 78.0%. Meanwhile, similar but obvious improvements in PCE and FF can be also observed for CH4-based OSCs after the introduction of CN (Tables S3-S6[†]). The corresponding J-V curves and external quantum efficiency (EQE) spectra are presented in Fig. 2c and d, respectively. It is worth noting that the remarkable PCE of over 18% for CH45-based OSCs without extra post-treatments is more attractive for large-scale production of OSCs when comparing to those involving complicated film treatment methods, such as thermal annealing, solvent vapor annealing or vacuum treatments. The excellent photovoltaic performance for OSCs based on PM6:CH45 without any extra post-treatments should be attributed to the superior film morphology once the formation of blended films is realized, which might be caused by more iodine-induced enhanced intermolecular secondary interactions (discussed in detail below). As regards the PCE improvement of CH45 comparing to that of CH4, the main reason has been revealed by a systematic investigation in our previous work.20,24,27,37 Halogenation on the central unit of NFAs can help to construct a superior fibrillar network in blended films; contribute to more efficient photodynamics; and make a better trade-off among tangled J_{SC} and V_{OC} . However, what is more interesting is the significant improvement of photovoltaic performance of OSCs, especially for FFs, after utilizing CN as additive. Although this phenomenon is really common during optimization of OSCs,11 a systematic investigation, especially at molecular levels, is still lacking, thus making the working mechanism of CN quite unclear.

Bearing the above thoughts in mind, herein we just take CH45 for an example to meticulously analyze its molecular packing changes in single crystals grown in chloroform (CF) and CN (see the detailed method in ESI†), which roughly simulates the device-fabricating conditions with/without CN. The rectangle-shaped crystals of CH45 grown in CN were much larger than those in CF (Fig. S9†), suggesting that CN may contribute to enhanced intermolecular interaction and improved crystallinity. From an overall view of single crystals (Fig. 3a and b), the effective intermolecular packing networks can be formed for crystals grown in both CN and CF. Moreover, in spite of the strong π - π stacking through conjugated skeletons, an interesting molecular pair (Fig. S10[†]) was also formed through weak I-S/I-H secondary interactions. This may be ascribed to the enhanced σ -hole interactions caused by the increased polarizability of iodine, which is expected to enhance the intermolecular interactions of CH45 and form a superior 3D intermolecular packing network. As regards the π - π stacking behaviors, the different molecular packing modes may be observed in single crystals grown in different solvents. However, in such a specific iodide as that of CH45, the packing modes extracted from single crystals grown in CF and CN are almost the same, both of them exhibiting three distinctive packing modes with over 80 kJ mol⁻¹ of intermolecular potential (Fig. S11[†]). The most attractive molecular packing of mode 1 is actually a dual interaction between iodized central unit of one molecule and bridged thiophene of another molecule (dual E/ b), which exhibits the largest intermolecular potential of \sim 200 kJ mol⁻¹ among the three different packing modes. Mode 2 is molecular packing mainly involving an "end unit to end unit" ("E/E" mode) interaction, also playing a dominant role in other high-performance A-D-A type acceptors like ITIC,19,38 F series,³⁹ Y series^{42,48} and even many donor molecules.⁴⁹ Another interesting packing mode (mode 3) involves a central unit and end unit. Note that modes 1 and 3 exist in most CH-series acceptors due to the 2D conjugated central units but are quite rare in other NFAs.^{20,24,37} Most importantly, in spite of almost the same packing modes, the π - π stacking distances are slightly shortened for single crystals grown in CN compared to those in CF, especially for mode 1 and mode 3 (Fig. 3c), indicating that CN can effectively enhance the intermolecular interactions of NFAs.

The major X-ray diffraction peaks (Fig. S12[†]) generated from a single crystal can roughly match with grazing incidence wideangle X-ray scattering (GIWAXS) data (Fig. 4a and b and Table 2). The molecular π - π stacking distances are slightly shortened from 3.86 Å (3.93 Å) to 3.80 Å (3.87 Å) in neat (blended) film when employing CN as additive based on GIWAXS measurements, which agrees well with the single-crystal analysis. Moreover, CH45 neat films with CN afford a relatively larger crystal coherence length (CCL) (22.5 Å for (010) and 157.1 Å for (100)) than that for films without CN (20.5 Å for (010) and 137.9 Å for (100)), indicating that CN is beneficial to increase the molecular crystalline ordering.⁵⁰ In general, in such a specific iodide as that of CH45, the introduction of CN can not only shorten the π - π stacking distance to enhance intermolecular



Fig. 3 Top and side views of crystal structure including three main packing modes (a) for CH45 grown in CF and (b) for CH45 grown in CN. The X-ray crystallographic coordinates for structures of CH45 grown in CF and CN have been deposited at the Cambridge Crystallographic Data Centre (CCDC) with numbers 2269036 and 2269035, respectively. (c) Statistical distribution of π - π interlayer distances between acceptor molecular layers including three types of intermolecular packing modes for CH45 grown in CF and CN.

interactions of CH45, but also improve molecular crystalline ordering, thus leading to facilitated charge transport and much increased FFs in resulting OSCs.

Based on atomic force microscope (AFM) images displayed in Fig. 4c, clearly bundle-like nanofibers can be observed in PM6:CH45-based blended films, which are conducive to superior photodynamics and improved photovoltaic parameters in resulting OSCs.^{51,52} Furthermore, the root-mean-square roughness for blended films with CN is 1.05 nm, larger than that without CN (0.75 nm). This reflects the increased crystallinity of CH45 induced by CN, in good accordance with GIWAXS data. As shown in Fig. S13,[†] the electron/hole mobilities (μ_e/μ_h) afforded by space-charge-limited current (SCLC) measurements of PM6:CH45-based blended films with and without CN were 5.9 \times $10^{-4}/4.4$ \times 10^{-4} and 5.0 \times $10^{-4}/2.6$ \times 10^{-4} V^{-1} $s^{-1},$ corresponding to μ_e/μ_h ratios of 1.3 and 1.9, respectively. The slightly higher but much more balanced μ_e/μ_h ratios for blended films with CN are beneficial for reaching better FFs in resulting OSCs.12 In addition, CH45-based OSCs with CN display overall improved exciton dissociation efficiency (η_{diss}), charge collection efficiency (η_{coll}) and suppressed charge recombination

processes with respect to CH45 without CN (Fig. S14a-c[†]).^{53,54} The higher photoluminescence quenching yield (η_{PLO}) of 92% for PM6:CH45 blends with CN than that of 89% without CN (Fig. S14d[†]) is also consistent with the larger $\eta_{\rm diss}$ mentioned above. Interestingly, the η_{PLQ} of 32% for CH45 neat films with CN is an appropriate signal of CH45 as a bipolar molecule (Fig. S14d[†]), which has been demonstrated in Y-series molecules.48 Benefiting from the enhanced intermolecular stacking and molecular crystalline ordering, a low level of energy loss should be achieved for CH45-based OSCs. A detailed decomposition of energy losses^{55,56} is presented in Table S7† and the overall energy losses for CH45-based OSCs are around 0.52 eV, ranking among the smallest E_{loss} systems.^{20,57} As regards the most important non-radiative recombination energy losses (ΔE_3) , a small value of ~0.21 eV can be afforded, which is further confirmed by the relatively large EQE_{EL} (Fig. S16[†]) and small Urbach energies $(E_{\rm U})$ presented in Fig. S17⁺.⁵⁷

To sum up, the introduction of CN in the CH45 system can enhance the strength of intermolecular packings and molecular crystalline ordering, thus leading to a superior film morphology. The facilitated charge transfer/transport dynamics



-0.5 0.0 0.5 1.0 1.5 2.0

 $q_{xy}(\text{\AA}^{-1})$

PM6:CH45



 $q_{\rm xy}({\rm \AA}^{-1})$

PM6:CH45

200.0 nm

PM6:CH45



200.0 nm

Fig. 4 (a) 2D GIWAXS patterns of CH45 neat films (with/without CN) and PM6:CH45 blend films (with/without CN). (b) The corresponding inplane (IP) and out-of-plane (OOP) extracted line-cut profiles from Fig. 3a. (c) AFM height/phase images of optimized PM6:CH45 films; the corresponding root-mean-square roughness values are 1.05 and 0.76 nm with and without CN, respectively.

Table 2 Crystal parameters extracted from GIW	AXS
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Film	$q~(010,{ m \AA}^{-1})$	d-space ^{a} (010, Å)	$\operatorname{CCL}^{b}(010, \operatorname{\AA})$
CH45	1.63	3.86	20.5
CH45 (CN)	1.65	3.80	22.5
PM6:CH45	1.60	3.93	14.4
PM6:CH45 (CN)	1.63	3.87	22.7

^{*a*} Calculated from the equation: *d*-spacing = $2\pi/q$. ^{*b*} Obtained from the Scherrer equation: $CCL = 2\pi K/FWHM$, where FWHM is the full-width at half-maximum and *K* is a shape factor (K = 0.9 here).

in resulting blended films should be responsible for the excellent photovoltaic performances without any assistance of extra post-treatments.

The stability of OSCs is a prerequisite for their future applications. Also we have some concerns as to the stability of the iodinated CH45 itself and devices due to the labile C-I bond, which can lead to various reactive species such as free radicals, especially under conditions of light exposure. Therefore, we conducted a stability experiment to help dispel such concerns. Firstly, the iodinated CH45 itself exhibits excellent

photostability, indicated by the UV-visible spectra and nuclear magnetic resonance (NMR) spectra. There are no changes in the intensity and shape of absorption spectra after aging for 144 h under one sun illumination (Fig. S18[†]). Also, NMR spectra of CH45 do not show any difference before and after aging for over 144 h under one sun illumination (Fig. S19[†]), which indicates that the introduction of C-I bonds does not decrease the photostability of the acceptor itself. The excellent photostability of CH45 can meet well the chemical stability requirement as a light absorption material. Secondly, experiments were conducted for thermal stability and photostability of OSCs based on iodinated CH45, fluorinated counterpart CH6 (ref. 27) and counterpart CH4 (ref. 27) without halogenation in the central unit. As depicted in Fig. S20,† under continuous heating at 65 ° C in a nitrogen-filled glovebox, the optimized devices based on CH4, CH6 (ref. 27) and CH45 maintained 76.0%, 84.7% and 86.3% of the initial efficiencies after thermal aging for 289 h, respectively. Moreover, under continuous 1 sun illumination simulated by light-emitting diode arrays using maximum power point tracking mode, the PCEs of all devices slowly decreased without a dramatic burn-in degradation stage, and kept at around 90% of their original PCE after photo-aging for 288 h.

Phase

2.0

1.5

1.0

0.5

0.0

(c)

The overall slightly superior stability of CH45- than CH4- and CH6-based devices should be attributed to the enhanced weak noncovalent interactions induced by polarizable iodine of CH45.

Conclusion

To address the great challenge of structural disorders or weak intermolecular interactions confronted by organic lightharvesting molecules, delicate iodization on the phenazine central unit of CH45 was conducted with the aim of enhancing intermolecular noncovalent interactions. Due to the high polarizability of iodine, a relatively large dielectric constant and reduced exciton binding energy can be realized for CH45 films with respect to the CH4 counterpart without iodization. Combining iodinated CH45 with the widely employed additive CN, a first-class efficiency of 18.15% has been achieved for OSCs without any extra post-treatments. Moreover, the working mechanism of CN in improving FFs of CH45-based OSCs was revealed through both single-crystal and morphology analysis: (I) shortening of molecular π - π stacking distances to enhance intermolecular interactions at single-molecule level; (II) improving molecular crystalline ordering to construct superior film morphology at the level of aggregation. By successfully establishing such a seldom reported but high-performance iodinated molecular platform, our work will inspire further pioneered exploration of light-harvesting molecules (and their delicate iodination) for high-efficiency OSCs free of tedious post-treatments.

Author contributions

The synthetic works were carried out by H. C., and three acceptors were characterized by H. C., X. C. and P. W. X. C. carried out most of the device fabrication and measurements. T. H., H. C. and G. L. carried out the theoretical computation of three acceptors. F. H. and Z. M. performed the EQE_{EL} and sEQE experiments and analyzed the data. H. L., X. B. and W. F. performed the morphology characterization and analyzed the data. Y. G., B. K. and C. L. helped to analyze the data and revise the manuscript. Y. C., Z. Y., and X. W. supervised and directed this project; Y. C., H. C. and Z. Y. wrote the manuscript. All authors discussed the results and commented on the manuscript.

Conflicts of interest

The authors declare no conflict of interest.

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