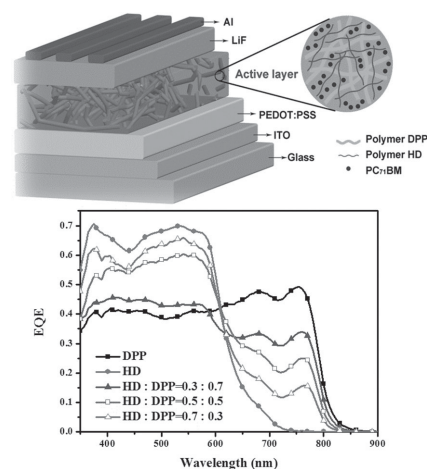


Performance Enhancement of Polymer Solar Cells by Using Two Polymer Donors with Complementary Absorption Spectra

Heng Lu, Xuejuan Zhang, Cuihong Li,* Hedi Wei, Qian Liu, Weiwei Li, Zhishan Bo*

Performance enhancement of polymer solar cells (PSCs) is achieved by expanding the absorption of the active layer of devices. To better match the spectrum of solar radiation, two polymers with different band gaps are used as the donor material to fabricate ternary polymer cells. Ternary blend PSCs exhibit an enhanced short-circuit current density and open-circuit voltage in comparison with the corresponding HD-PDPC-DTBT (HD)- and DT-PDPPTPT (DPP)-based binary polymer solar cells, respectively. Ternary PSCs show a power conversion efficiency (PCE) of 6.71%, surpassing the corresponding binary PSCs. This work demonstrates that the fabrication of ternary PSCs by using two polymers with complementary absorption is an effective way to improve the device performance.



1. Introduction

Bulk-heterojunction polymer solar cells (BHJ PSCs) have attracted a great deal of interest due to their unique properties of low-cost, light weight, and potential applications in large-area flexible devices.^[1] The bulk

heterojunction (BHJ) structure has been proven to be the most efficient structure for PSCs, which has a large donor/acceptor (D/A) interface to make the photogenerated excitons separate to form free charge carriers and the phase-separated interpenetrating network can effectively transport the free charge carriers to the collecting electrodes.^[2] High power conversion efficiency (PCE) polymer solar cells require that the polymers have a broad absorption to effectively absorb the solar radiation to produce high short-circuit current (J_{sc}).^[1,2] However, due to the intrinsic molecular excitonic nature of the primary excitation, conjugated polymers usually exhibited an absorption spectrum with a few 100 nm width.^[3] The main-chain donor-acceptor design can effectively extend the main absorption peak of conjugated polymers even to the near-infrared region via the internal charge transfer between the donor and acceptor units. However, this strategy will decrease the light absorption in the visible region and result in

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a low short-circuit current density (J_{sc}).^[1b,c,e] The influence of absorption width on the PCE of polymer solar cells was studied in a great detail by Minnaert and Burgelman,^[4] and their investigations have revealed that the broadening of the width of absorption band from 200 to 400 nm will result in an improvement of efficiency up to 35%. To more efficiently utilize the solar radiation, several major resolutions at different levels of maturity have been put forward. For instance, design and synthesis of conjugated terpolymers would expand the absorption spectrum and improve the light harvesting.^[5] However, random terpolymers are usually noncrystalline and exhibit lower PCE.^[6] Alternatively, tandem solar cells can provide much broader absorption than single junction cells.^[1f,7] In a tandem solar cell, two individual subcells with complementary absorption spectra are stacked together by a charge recombination layer.^[6] The open-circuit voltage (V_{oc}) of tandem solar cells equals the sum of two subcells while the short-circuit current is restricted to the lower one, resulting in an enhanced total PCE. Complexity and technical challenges are main drawbacks for tandem solar cells. To expand the absorption scope of the active layer, ternary blend systems with two donor materials and one fullerene acceptor have recently attracted more and more attention.^[3,8] And this strategy still maintains the simple processing conditions used for single junction devices. Various sensitizers, including dyes, polymers, small molecules and quantum dots, have been employed in a small quantity as the third component in the ternary system to expand the absorption range of active layers.^[3] For instance, Huang et al. recently demonstrated that the incorporation of 1.0% squaraine dye in P3HT:PC₆₀BM-based PSCs could give a 38% increase in PCE.^[9] The increase of PCE is believed due to the Förster resonance energy transfer from P3HT to squaraine dye, which helps to improve exciton migration over long distances. Yu et al. demonstrated that an improved PCE was achieved in PID2-doped PTB7:PC₇₁BM ternary PSCs.^[10] A low content of PID2 leads to fibrillar structures and smaller domain sizes, favoring light harvesting, charge dissociation, and transport. Unlike the aforementioned ternary PSCs, You et al. reported the parallel-like BHJ polymer solar cells, in which two or more polymer donors with different band gaps can be employed at any composition, regardless of their highest occupied molecular orbital (HOMO) or lowest unoccupied molecular orbital (LUMO) levels.^[11]

Here, we develop a ternary blend system that contains two conjugated

polymers HD-PDFC-DTBT (simply abbreviated as HD)^[12] and DT-PDPPTPT (simply abbreviated as DPP)^[13] with complementary absorption feature to expand the absorption profile of the active layer. More importantly, the DPP component can form fibrous nanostructures for hole transport.^[10] We investigated ternary PSCs with different HD/DPP ratios. The optimal device with an HD/DPP ratio of 0.5:0.5 exhibited higher PCE than the corresponding binary PSCs based on HD or DPP. Compared with the HD:PC₇₁BM-based binary PSCs, the increase of PCE for the ternary PSCs is mainly due to a significant enhancement of the photocurrent density (J_{sc}) and a small enhancement of the fill factor (FF). Compared with the DPP:PC₇₁BM-based PSCs, the enhancement of PCE for the binary system is mainly due to a marked enhancement of the open-circuit voltage (V_{oc}). The HD:DPP:PC₇₁BM ternary blend exhibited a marked enhancement of the space limited charge carrier (SCLC) mobility (μ) in comparison with the corresponding HD or DPP-based binary blend. External quantum efficiencies (EQEs) of the ternary and binary active layers were measured to investigate the photoresponse of different components.

2. Results and Discussion

The chemical structures of HD and DPP are shown in Figure 1a. Polymer HD consists of electron-rich 2,7-linked 3,6-difluoro-2-hexyldecylcarbazole unit, 2,5-linked thiophene spacer, and electron-deficient 5,6-bis(octyloxy)-benzothiadiazole unit, whereas polymer DPP consists of electron-rich 1,4-linked phenylene unit, 2,5-linked thiophene spacer, and electron-deficient 2,5-bis(2-hexyldecyl) diketopyrrolopyrrole unit. The absorption spectra of polymers HD and DPP are shown in Figure 1b and the absorption data are summarized in Table 1. The absorption spectra of these two polymers do not match the solar spectrum very well. The main absorption peak of HD is located in the orange region from 500 to 600 nm, whereas the

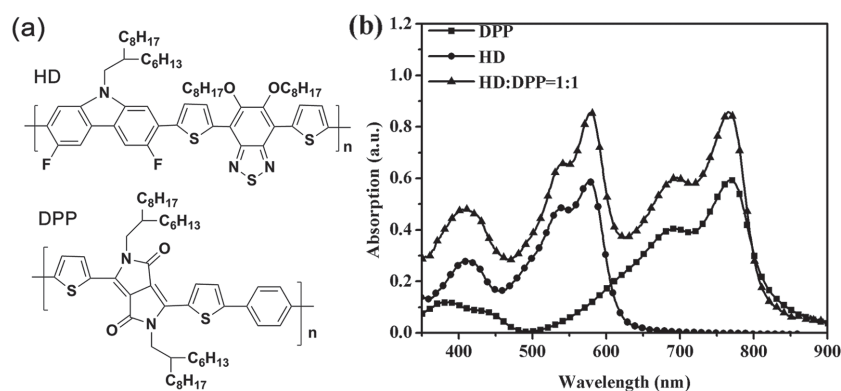


Figure 1. a) Chemical structures of HD and DPP; b) UV-vis absorption spectra of HD, DPP, and the blend of HD:DPP (1:1, by weight) films.

■ Table 1. Optical, electronic properties, and number-average molecular weights (M_n) of polymers and hole mobilities of solar cells.

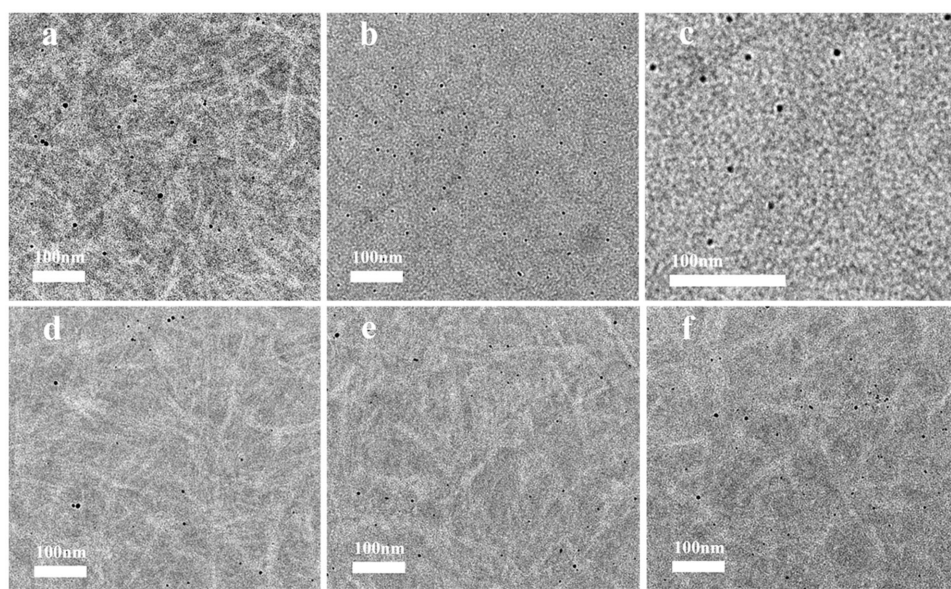
Donor	$\lambda_{\max}$ [nm]	$E_{g, \text{opt}}$ [eV] ^{b)}	HOMO/LUMO [eV]	M_n [kg mol ⁻¹]	μ_h [cm ² V ⁻¹ s ⁻¹] ^{c)}
HD	412 576	1.96	−5.57/−3.61	38	4.06×10^{-5}
DPP	684 767	1.53	−5.48/−3.95	72	8.90×10^{-5}
HD:DPP ^{a)}	412 765	—	—	—	2.70×10^{-4}

^{a)} 1:1 by weight; ^{b)} Energy levels calculated from the onsets of film absorption peaks according to equations $E_{g, \text{opt}} = 1240/\lambda_{\text{edge}}$ and $E_{\text{LUMO}} = E_{\text{HOMO}} + E_{g, \text{opt}}$; ^{c)} Measured by the SCLC method.^[14]

main absorption peak of DPP is located in the red region from 650 to 800 nm. It is worth noting that the absorption of HD and DPP is complementary. As shown in Figure 1b, the blend film of HD:DPP (1:1) shows a broad absorption ranging from 350 to 850 nm. HD can compensate for the weak absorption of DPP in the short-wavelength region from 350 to 600 nm.

HD exhibits good solubility in 1,2-dichlorobenzene (DCB) at elevated temperature, but poor solubility in chloroform (CF), whereas DPP exhibits good solubility in CF, but poor solubility in DCB. Therefore, we stirred the blend of HD and PC₇₁BM in a weight ratio of 1:2.5 in DCB at 60 °C for 2 h to guarantee to a sufficient dissolution. Meanwhile, a blend of DPP and PC₇₁BM in a weight ratio of 1:2.5 in CF (containing 6% DCB) was stirred at 60 °C for 2 h to ensure a sufficient dissolution to form a clear solution. After the two solutions were cooled to room temperature, they were mixed in different ratios and used for the fabrication of polymer solar cell devices. Since DCB is not a good solvent for DPP at room temperature, the formed nanofibrils by DPP in CF solutions could not

be destroyed after mixing with the HD solution in DCB. To investigate whether the nanofibrils still exist in the ternary blend films, bright field transmittance electron microscopy (TEM) experiments were carried out. As shown in Figure 2a, TEM images reveal that the formed nanofibrils in the DPP:PC₇₁BM (1:2.5) binary blends have a length of several hundred nanometers and a width of about 30 nm. TEM images of HD:PC₇₁BM (1:2.5) in Figure 2b showed a uniform morphology and the formed fine interpenetrated networks can clearly be seen in the magnified TEM image (Figure 2c). In the ternary blends films, TEM images also revealed that the nanofibrils were formed and the number of the nanofibrils decreased with the decreasing of DPP content. The packing of polymer chains as thin films is very crucial for the transportation of charge carriers. X-ray diffraction (XRD) analyses on polymer films on silicon substrates were performed and the diffraction patterns are shown in Figure S4 (Supporting Information). DPP films were spin-coated from a solution of CF:DCB (100:6), HD films were spin-coated from DCB solution, and HD:DPP (1:1) blend films



■ Figure 2. Bright-field TEM images of blend films. a) DPP; b) HD; c) the magnified image of HD; d) HD:DPP = 0.3:0.7; e) HD:DPP = 0.5:0.5; f) HD:DPP = 0.7:0.3 (all blend with PC₇₁BM by the weight ratio of 1:2.5).

Table 2. Photovoltaic parameters of solar cells.

HD:DPP:PC ₇₁ BM	PCE [%] ^{a)}	V _{oc} [V]	J _{sc} [mA cm ⁻²] ^{b)}	FF ^{c)}	d [nm]
1:0:2.5	5.72 (5.61)	0.97	8.82	0.67	95
0:1:2.5	6.34 (6.25)	0.79	11.49	0.69	110
0.3:0.7:2.5	6.24 (5.91)	0.85	10.46	0.70	96
0.5:0.5:2.5	6.71 (6.60)	0.87	11.02	0.70	103
0.7:0.3:2.5	6.53 (6.29)	0.90	10.51	0.69	98

^{a)}Value in brackets is the average over five nominally identical devices; ^{b)}J_{sc} was calculated by integrating the EQE spectrum with the AM1.5G spectrum; ^{c)}Fill factor.

were spin-coated from a solution of the blend in a solvent mixture of CF and DCB. All the concentrations are 10 mg mL⁻¹. DPP polymer exhibits one diffraction peak at a small angle of 4.86°, which reflects a distance of 18.15 Å between polymer backbones separated by alkyl side chains. The diffraction pattern of HD polymers shows three sharp peaks at the wide angel region, and the data are summarized in Table S2 (Supporting Information). The diffraction pattern of HD:DPP (1:1) blend films is similar to that of the HD polymer. The above results indicated that DPP is an amorphous polymer and HD is a semicrystalline polymer. As shown in Figure 2c, the blend films are composed of thick amorphous DPP nanofibers and the very thin semicrystalline HD nanofibers. Such a hierarchy structure of the active layer may be responsible for the improvement of the hole mobility (vide infra).

Photovoltaic properties of the ternary blends were investigated in a device configuration of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/active layer/LiF/Al (Figure S1, Supporting Information). The photovoltaic performance of ternary blend PSCs cells was optimized in different HD:DPP weight ratios and different polymer:PC₇₁BM ratios to afford the highest PCE. The ternary blend solutions used for the fabrication of PSCs via spin-coating were prepared by

mixing a solution of HD:PC₇₁BM in DCB and a solution of DPP:PC₇₁BM in CF. After optimization, a concentration of polymer:PC₇₁BM of 25 mg mL⁻¹, a weight ratio of polymer to PC₇₁BM of 1:2.5, and a thickness of the blend films of about 100 nm gave the best device performance. For the HD:PC₇₁BM (1:2.5)-based binary blend PSCs, a PCE of 5.82% was achieved with a V_{oc} of 0.97 V, an FF of 0.68, and a J_{sc} of 8.82 mA cm⁻². DPP:PC₇₁BM (1:2.5)-based PSCs gave a PCE of 6.34% with a V_{oc} of 0.79 V, an FF of 0.69, and a J_{sc} of 11.49 mA cm⁻². Ternary blend PSCs with HD:DPP:PC₇₁BM ratios of 0.7:0.3:2.5, 0.5:0.5:2.5, and 0.3:0.7:2.5 were fabricated and the device characters are summarized in Table 2. J–V characteristics of devices are shown in Figure 3a. As shown in Figure 3a, with the increase of the weight ratio of HD in the ternary blend, V_{oc} increased gradually from 0.79 to 0.97 V. For BHJ polymer solar cells, an empirical relation of $qV_{oc} \approx E_{CT} - 0.5$ eV (with q the elementary charge and E_{CT} the charge transfer state energy) has been used to estimate the V_{oc} under one sun illumination intensity at room temperature.^[15] The CT-state energy shifts with the composition of the blend, resulting in the changing of V_{oc}. For the ternary blend system, as shown in Figure S5a (Supporting Information), the V_{oc}, which exhibited a sublinear dependence on the composition of the two polymers, could be modeled as a parallel tandem

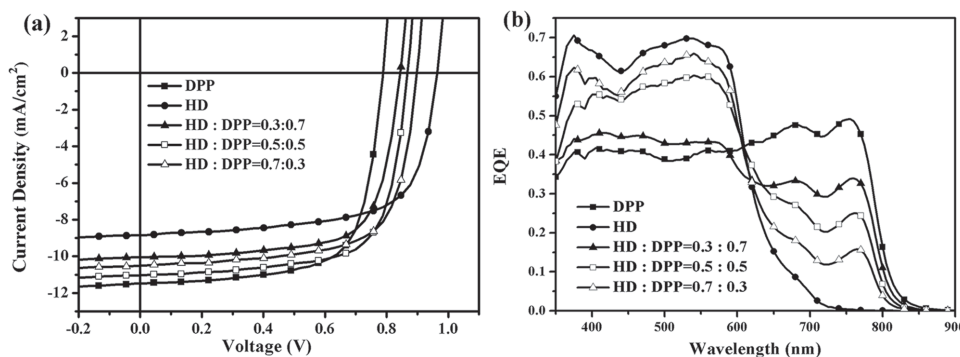


Figure 3. a) J–V characteristics of devices measured under the illumination of AM 1.5 G (100 mW cm⁻²) from a solar simulator; b) EQE curves of the binary and ternary PSCs.

device configuration.^[11,15] In the parallel tandem solar cells, there are two distinct CT states that both contribute to the absorption of light, depending on the composition of the two polymers.^[11,15] Compared with the DPP:PC₇₁BM (1:2.5)-based binary PSCs, J_{sc} of ternary PSCs with an HD:DPP:PC₇₁BM weight ratio of 0.3:0.7:2.5 decreased from 11.49 mA cm⁻² to 10.46 mA cm⁻². Increasing the HD content to an HD:DPP:PC₇₁BM weight ratio of 0.5:0.5:2.5, J_{sc} increased to 11.02 mA cm⁻². Further increasing the HD content to an HD:DPP:PC₇₁BM weight ratio of 0.3:0.7:2.5, J_{sc} unexpectedly decreased to 10.51 mA cm⁻². According to the equation $PCE = (J_{sc} \cdot V_{oc} \cdot FF) / P_{in}$, where P_{in} is the intensity of the incident light, we have to make a balance between pursuing high J_{sc} and V_{oc} . The highest PCE of 6.71% was achieved for ternary PSCs with an HD:DPP:PC₇₁BM weight ratio of 0.5:0.5:2.5. Compared with DPP:PC₇₁BM-based binary PSCs, the enhancement of PCE for the ternary PSCs is mainly due to the increase of V_{oc} , whereas compared with the HD:PC₇₁BM-based binary PSCs, the increase of PCE is mainly attributed to the increase of J_{sc} . High-efficiency PSCs require that the active layer have a balanced hole and electron mobility. Therefore, hole mobility of the binary and ternary blends was measured by the space charge limited current (SCLC) method and the results are shown in Table 1. The results clearly showed that the HD:DPP:PC₇₁BM (0.5:0.5:2.5)-based ternary blends have the highest hole mobility, benefiting the charge transportation. To verify the J_{sc} obtained by J - V measurement and to estimate the contribution of different polymers in the ternary blend system, EQE measurements were performed. EQE curves of binary and ternary PSCs are shown in Figure 3b. HD:PC₇₁BM (1:2.5)-based binary PSCs exhibited EQE values up to 70% in the range of 380–600 nm; whereas the EQE values of DPP:PC₇₁BM-based PSCs are around 40%–50% in the range of 350–800 nm. In the ternary PSCs, with the increasing of the content of DPP component, the EQE responses in the long wavelength region of 600–800 nm enhanced, and the EQE responses in the short-wavelength region of 350–700 nm decreased. It is reasonable to suppose that the DPP component is responsible for the EQE responses in the long wavelength region (600–800 nm) and the HD component contributes a large portion to the EQE response in the short-wavelength region (300–600 nm). The thickness of the active layer was around 100 nm for all devices. The EQE values at about 760 nm are almost linearly proportional to the content of DPP component (Figure S3, Supporting Information), indicating that the EQE response of DPP was not affected by the incorporation of the HD component. At the short-wavelength region, the EQE value of the HD:DPP:PC₇₁BM (0.5:0.5:2.5)-based ternary PSCs with a thickness of about 100 nm is roughly equal to the sum of contributions of HD and DPP-based binary PSCs with a thickness of about 50 nm. In

addition, the J_{sc} calculated by the integration of EQE curve is roughly equal to the J_{sc} obtained by J - V measurements, indicating that the obtained PCEs are believable.

3. Conclusions

In conclusion, HD:DPP:PC₇₁BM-based ternary polymer solar cells with enhanced photovoltaic performance were developed. The two polymers (HD and DPP) have complementary absorptions and the DPP component can form nanofibrils networks for the charge transportation. The ternary PSCs exhibited higher J_{sc} in comparison with the HD:PC₇₁BM-based binary PSCs and higher V_{oc} in comparison with the DPP:PC₇₁BM-based binary PSCs. V_{oc} of the ternary PSCs is related to the content of the two components. In an HD:DPP:PC₇₁BM weight ratio of 0.5:0.5:2.5, the devices exhibited a PCE of 6.71%, which surpass PCEs of the corresponding binary PSCs. This work demonstrates that the fabrication of ternary PSCs is an effective and easy way to achieve high photovoltaic performance.

4. Experimental Section

General Measurements and Characterization: UV–vis absorption spectra were obtained on a PerkinElmer UV–vis spectrometer model Lambda 750. The TEM (Tecnai F20, FEI) was operated at an accelerating voltage of 200 kV. J - V characteristics were recorded using a Keithley 2400 SMU under the irradiation of a 300 W solar simulator (San-Ei electric, XES-301S) as the white light source and the intensity was calibrated with a standard single-crystal Si photovoltaic cell. The thickness of the active layer was measured by a Dektak 6 M surface profilometer. XRD measurement was performed with Cu K α radiation using a 1.6 kW PANalytical Empyrean X-ray diffractometer.

Fabrication of PSCs: Photovoltaic performance was investigated in BHJ devices with a device configuration of ITO/PEDOT:PSS/active layer/LiF/Al for BHJ solar cells. ITO glasses with a conductivity of 20 Ω square⁻¹ were cleaned according to the standard procedures before use.^[1d] A thin layer of PEDOT:PSS (Clevios P VP AI 4083, purchase from H.C. Starck GmbH) was spin-coated on top of cleaned ITO substrate at 3000 rpm for 60 s and dried subsequently at 130 °C for 20 min on a hotplate before being transferred into a glove box. The binary PSCs HD-PDPC-DTBT:PC₇₁BM (99.5%, Solenne) and DT-PDPPTPT:PC₇₁BM in an optimal weight ratio of 1:2.5 spin coated on the top of ITO/PEDOT:PSS. Two polymers mixed with different ratios to fabricate the ternary PSCs. DPP:PC₇₁BM blend films were spin-coated from a solvent mixture of CF:DCB (100:6), HD:PC₇₁BM films were spin-coated from the DCB solution, and HD:DPP:PC₇₁BM films were spin-coated from a solvent mixture of CF and DCB. All the concentrations are 25 mg mL⁻¹. The solvents of DCB and CF were purchased from Aldrich. Finally, to complete the devices, the top electrode was thermally evaporated with a 0.6 nm LiF layer and followed by a 100 nm Al film at a base pressure below 10⁻⁶ torr. Five cells were fabricated on one substrate with an effective area

of 0.04 cm². The measurement of devices was conducted in air without encapsulation. The temperature while measuring the *J*–*V* curves was approximately 25 °C. The characteristics of the solar cells were optimized by testing approximately five cells.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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