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Exciplex spin-flip acceleration enables high-performance narrowband electroluminescence

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Exciplex-forming systems that harvest triplet excitons via a triplet-to-singlet spin flip (reverse intersystem crossing, RISC) enable thermally activated delayed fluorescence, providing a route to boost light emission in organic light-emitting diodes. Here, we report heavy-atom-incorporated exciplexes in which the triplet state is predominantly localized on the heavy-atom fragment, resulting in large spin-orbit coupling. Through positional isomer optimization, the RISC rate constant reaches $4.9 \times 10^6 \text{ s}^{-1}$, approximately an order of magnitude higher than in typical exciplexes. Organic light-emitting diodes based on the optimized exciplex host achieve a maximum external quantum efficiency (EQE) exceeding 40% and exhibit low efficiency roll-off (EQE > 33% at 1000 cd m^{-2}).

Introduction

The spin-triplet excitons are key determinants of efficiency and stability in organic optoelectronic devices.^{1–4} For organic emitters, their triplet excitons are primarily harnessed by two pathways: phosphorescence through radiative triplet decay,^{5–7} or thermally activated delayed fluorescence (TADF) via reverse intersystem crossing (RISC) from spin-forbidden triplets to spin-allowed singlets.^{8–10} Recent developments have shown that TADF can be realized in materials featuring twisted donor (D)–acceptor (A) configurations,^{11–15} or multiple-resonance (MR) nitrogen-boron fused polycyclic frameworks.^{16–20} In both cases, the electron densities of the highest occupied and lowest unoccupied molecular orbitals (HOMO and LUMO) can be spatially separated by precise molecular design, thereby facilitating spin-flip of triplet excitons for efficient utilization.^{21,22}

Another class of TADF materials, known as exciplexes, is formed in donor/acceptor (D/A) blends and offers greater design flexibility, attracting considerable industrial interest.^{23,24} Upon photo- or electrical excitation, D and A form an excited-state complex in which the HOMO resides mainly on D and the

LUMO on A, enabling through-space charge transfer and triplet-to-singlet spin flip.^{25,26} In such exciplex systems, however, the lowest singlet (S_1) and triplet (T_1) states typically have dominant charge-transfer (CT) character, resulting in weak spin-orbit coupling (SOC) and consequently slow RISC;^{27,28} reported exciplex TADF systems usually exhibit RISC rates (k_{RISC}) below 10^6 s^{-1} .^{29–32} Slow RISC prolongs triplet lifetimes and promotes bimolecular exciton annihilation events, leading to severe efficiency roll-off in exciplex-based OLEDs.³³

In this context, we are driven to accelerate the triplet-to-singlet spin flip in exciplex systems, achieving k_{RISC} of approximately $4.9 \times 10^6 \text{ s}^{-1}$, about an order of magnitude higher than those typically other reported exciplexes. This RISC enhancement enables excellent performance in exciplex-based OLEDs, particularly at high brightness, surpassing state-of-the-art TADF, phosphorescent, TADF-sensitized hyperfluorescent and phosphorescence-sensitized hyperfluorescent OLEDs based on exciplex-forming co-hosts. Theoretical calculations and photo-physical studies confirm that forming localized triplet states on heavy-atom-containing fragments, together with inducing additional orbital angular momentum through positional isomer optimization, drives the enhanced k_{RISC} , so as to reduce the triplet exciton concentration and minimize triplet-involved annihilations in the emission layer, and ultimately overcome the efficiency roll-off.

Results and discussion

Given that exciplex-forming co-hosts are based on bimolecular donor–acceptor systems,^{27,34} and recognizing the relatively high electronegativity of selenium,^{35–37} we strategically incorporated a planar five-membered selenium-containing heterocycle (dibenzo[*b,d*]selenophene) into the donor moiety. To minimize the

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formation of labile bonds,³⁸ we specifically compared *meta*- and *para*-conjugation linkages (*mSeCzCz* vs. *pSeCzCz*, Fig. 1a) for integrating selenium atom within the electron-donating framework. Density functional theory calculations (Fig. S1) indicated that *mSeCzCz* and *pSeCzCz* have similar dihedral angles between the dibenzo[*b,d*]selenophene and bicarbazole moieties (55.1° for *mSeCzCz* and 56.1° for *pSeCzCz*).

The selenium-containing donors (*mSeCzCz* and *pSeCzCz*) were synthesized *via* a multi-step procedure (Scheme S1). The synthetic route commenced with a lithium-halogen exchange reaction, followed by hydrolysis to yield aromatic boric acids. Chlorinated dibenzo[*b,d*]selenophenes, serving as key intermediates, were prepared by trimethylsilyl cyanide-catalyzed intramolecular cyclization. The final products were obtained through the Buchwald-Hartwig coupling reaction. The chemical structures were fully characterized by ¹H NMR, ¹³C NMR, and high-resolution mass spectrometry (Fig. S2–S11). Furthermore, the single-crystal structure of *mSeCzCz* (Fig. S12) revealed a twisted geometry between the peripheral dibenzo[*b,d*]selenophene group and the bicarbazole core. Notably, the crystal architecture displayed no significant π - π interactions. However, intermolecular C-H...Se interactions between the phenyl rings of bicarbazole and a neighboring molecule's selenium atom, along with multiple C-H... π interactions between neighboring dibenzo[*b,d*]selenophene and carbazole rings, were observed. Additionally, thermogravimetric analysis

(Fig. S13) demonstrated their excellent thermal stability, with high decomposition temperatures (5% weight loss) of 420 °C and 408 °C for *mSeCzCz* and *pSeCzCz*, respectively, making them suitable for forming uniform films by thermal evaporation.

To construct blue-light-emitting exciplex systems, *SiTrzCz2* (9,9'-(6-(3-(triphenylsilyl)phenyl)-1,3,5-triazine-2,4-diyl)bis(9*H*-carbazole)) with high triplet energy ($T_1 = 2.89$ eV) was selected as an electron-withdrawing component and blended with selenium-containing donors of *mSeCzCz* and *pSeCzCz*, respectively (Fig. 1a). The steady-state photophysical properties of *mSeCzCz*, *pSeCzCz*, *SiTrzCz2*, and their 1:1 molar blends (*mSeCzCz*:*SiTrzCz2* and *pSeCzCz*:*SiTrzCz2*, abbreviated as *mSe-EX* and *pSe-EX*) were investigated in solid films. As shown in Fig. 1b and c, both *mSeCzCz* and *pSeCzCz* exhibited absorption bands below 350 nm, closely matching the absorption of the reference molecule *SiCzCz* (9-(3-(triphenylsilyl)phenyl)-9*H*-3,9'-bicarbazole). The photoluminescence (PL) spectra of *mSeCzCz* and *pSeCzCz* films displayed emission maxima at 386 nm and 406 nm, respectively, with the latter showing a distinct red shift relative to *SiCzCz*. Interestingly, their 1:1 molar blends with *SiTrzCz2* (*mSe-EX* and *pSe-EX*) exhibited identical broad emission bands centered at 467 nm, slightly blue-shifted compared to the control exciplex (*SiCzCz*:*SiTrzCz2*, termed *Si-EX*). Phosphorescence spectra measured at 77 K (Fig. S14) revealed T_1 energies of 2.77 eV for *mSeCzCz* and 2.84 eV for *pSeCzCz*,

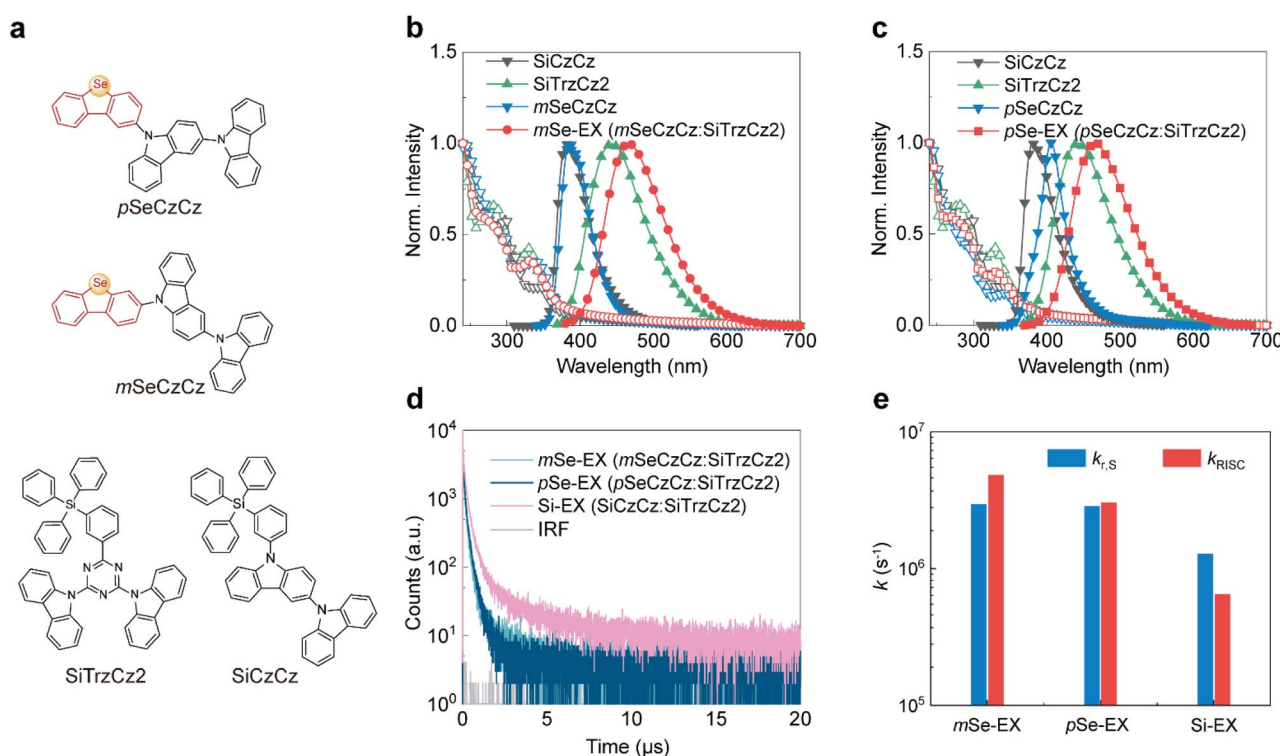


Fig. 1 (a) Chemical structures of the compounds. (b) Normalized UV-visible absorption spectra (open symbols) and PL spectra (filled symbols) of *SiCzCz*, *SiTrzCz2*, *mSeCzCz* and their exciplex blend *mSeCzCz*:*SiTrzCz2* (*mSe-EX*). (c) Normalized UV-visible absorption spectra (open symbols) and PL spectra (filled symbols) of *SiCzCz*, *SiTrzCz2*, *pSeCzCz* and their exciplex blend *pSeCzCz*:*SiTrzCz2* (*pSe-EX*). (d) Transient PL decay curves of *mSe-EX*, *pSe-EX* and the reference exciplex *SiCzCz*:*SiTrzCz2* (*Si-EX*) films under excitation at 375 nm. (e) Comparison of calculated rate constants for singlet radiative decay ($k_{r,s}$) and reverse intersystem crossing (k_{RISC}).



determined from the onsets of these spectra. The broad phosphorescence emission band around 500 nm arises from the cumulative contribution of multiple vibronic bands associated with LE-state emission. As anticipated, *mSe-EX* and *pSe-EX* films clearly exhibited prompt and delayed fluorescence components (Fig. 1d), confirming efficient RISC within these exciplexes. The corresponding prompt (τ_{PF}) and delayed (τ_{DF}) lifetimes of *mSe-EX* (58.5 ns/0.3 μs) and *pSe-EX* (72.0 ns/0.4 μs) were significantly shorter than those of *Si-EX* (186.8 ns/1.9 μs), despite similar PL quantum yields (31%, 29%, and 30% for *mSe-EX*, *pSe-EX*, and *Si-EX*, respectively). Key kinetic constants, including singlet radiative decay rate ($k_{\text{r,s}}$), singlet nonradiative decay rate ($k_{\text{nr,s}}$), intersystem crossing rate (k_{ISC}) and RISC rate (k_{RISC}), were extrapolated and are summarized in Fig. 1e and Table 1 (see SI for detailed calculation methods).^{39,40} Notably, *mSe-EX* and *pSe-EX* showed significantly enhanced $k_{\text{r,s}}$ values ($3.0 \times 10^6 \text{ s}^{-1}$ and $2.9 \times 10^6 \text{ s}^{-1}$, respectively) relative to *Si-EX* ($1.3 \times 10^6 \text{ s}^{-1}$). Selenium incorporation also accelerated singlet nonradiative contributions, including $k_{\text{nr,s}}$ ($5.4 \times 10^6 \text{ s}^{-1}$ for *mSe-EX*, $6.8 \times 10^6 \text{ s}^{-1}$ for *pSe-EX* and $2.9 \times 10^6 \text{ s}^{-1}$ for *Si-EX*) and k_{ISC} ($6.1 \times 10^6 \text{ s}^{-1}$ for *mSe-EX*, $3.3 \times 10^6 \text{ s}^{-1}$ for *pSe-EX* and $1.0 \times 10^6 \text{ s}^{-1}$ for *Si-EX*), resulting in similar PL quantum yields across these exciplex systems. More importantly, *mSe-EX* and *pSe-EX* demonstrated substantially increased k_{RISC} values ($4.9 \times 10^6 \text{ s}^{-1}$ and $3.1 \times 10^6 \text{ s}^{-1}$, respectively) compared to *Si-EX* ($6.9 \times 10^5 \text{ s}^{-1}$), indicating that selenium integration in exciplex systems promotes faster consumption of triplet excitons. This trend is consistent with other selenium-containing TADF emitters, and the k_{RISC} values of *mSe-EX* and *pSe-EX* surpass those of most previously reported selenium-containing TADF molecules (typically in the order of 10^5 – 10^6 s^{-1}).^{41–45}

To clarify the enhanced k_{RISC} in *mSe-EX* relative to *pSe-EX*, we performed time-dependent density functional theory calculations to analyse SOC matrix elements and natural transition orbital (NTO) distributions, with *Si-EX* as the reference. The SOC matrix elements for the $T_1 \rightarrow S_1$ transition (Fig. 2a) in *mSe-EX* (3.606 cm^{-1}) and *pSe-EX* (3.136 cm^{-1}) were markedly higher than that of *Si-EX* (0.265 cm^{-1}), corresponding to 13.6- and 11.8-fold enhancements, respectively (Fig. 2b). NTO analysis indicated charge-transfer characteristics in the S_1 states of all exciplexes (Fig. S15), while their T_1 states exhibited localized excitations (Fig. 2c). In *Si-EX*, the T_1 state primarily localized on the benzene unit of tetraphenylsilane and the *meta*-connected

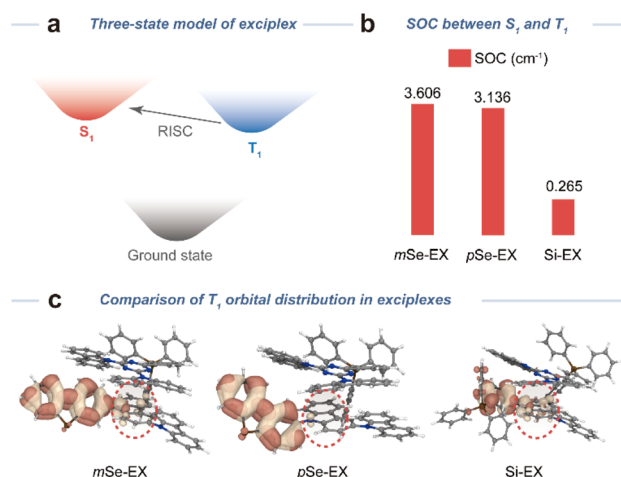


Fig. 2 (a) Schematic illustration of potential energy surfaces for the ground state (black), the lowest singlet state (S_1 , red), and the lowest triplet state (T_1 , blue) within the exciplex. (b) Comparison of calculated spin-orbit coupling (SOC) values between the S_1 and T_1 states for *mSe-EX*, *pSe-EX*, and *Si-EX* systems. (c) Hole-electron distributions of T_1 orbitals for *mSe-EX*, *pSe-EX*, and *Si-EX* at their optimized T_1 geometries.

carbazole groups. Conversely, the T_1 state in *pSe-EX* was mainly localized on the dibenzo[*b,d*]selenophene fragment, and the selenium incorporation enhanced the SOC between T_1 and S_1 states. Notably, for *mSe-EX*, the T_1 state extended from the dibenzo[*b,d*]selenophene fragment to the *meta*-connected carbazole group *via* two-lobed p-orbitals. The rotational overlap of adjacent p-orbitals generated additional orbital angular momentum, amplifying SOC and thereby enhancing k_{RISC} in *mSe-EX*.⁴⁶

To further investigate the exciplex hosts with fast k_{RISC} on MR-TADF emitters, we incorporated *v*-DABNA as the emitter into *mSe-EX*, *pSe-EX*, and *Si-EX* host matrices. PL spectra of the doped films (Fig. S16) showed a narrow emission peak at 470 nm with a FWHM of 17 nm, confirming efficient Förster resonance energy transfer (FRET) from the exciplex host to the MR-TADF emitter. Time-resolved emission spectra (Fig. 3a–c) revealed shorter delayed fluorescence lifetimes for *mSe-EX*:*v*-DABNA (2.8 μs) and *pSe-EX*:*v*-DABNA (3.0 μs) compared to *Si-EX*:*v*-DABNA (4.0 μs). Additionally, the delayed component

Table 1 Photophysical properties of *mSeCzCz*:*SiTrzCz2* (*mSe-EX*), *pSeCzCz*:*SiTrzCz2* (*pSe-EX*) and the reference exciplex *SiCzCz*:*SiTrzCz2* (*Si-EX*)

Film	λ_{PL}^a [nm]	Φ_{PL}^b [%]	τ_{p}^c [ns]	τ_{d}^d [μs]	$k_{\text{r,s}}^e$ [s^{-1}]	$k_{\text{nr,s}}^f$ [s^{-1}]	k_{ISC}^g [s^{-1}]	k_{RISC}^h [s^{-1}]
<i>mSe-EX</i>	467	31	58.5	0.3	3.0×10^6	5.4×10^6	6.1×10^6	4.9×10^6
<i>pSe-EX</i>	467	29	72.0	0.4	2.9×10^6	6.8×10^6	3.3×10^6	3.1×10^6
<i>Si-EX</i>	470	30	186.8	1.9	1.3×10^6	2.9×10^6	1.0×10^6	6.9×10^5

^a Peak wavelength of the photoluminescence spectra in the film. ^b Absolute photoluminescence quantum yield measured with an integration-sphere system under an argon atmosphere. ^c Lifetime of the prompt component determined from the transient photoluminescence decay curve at 298 K. ^d Lifetime of the delayed component determined from the transient photoluminescence decay curve at 298 K. ^e Rate constant of singlet radiative decay. ^f Rate constant of singlet nonradiative decay. ^g Rate constant of intersystem crossing. ^h Rate constant of reverse intersystem crossing.



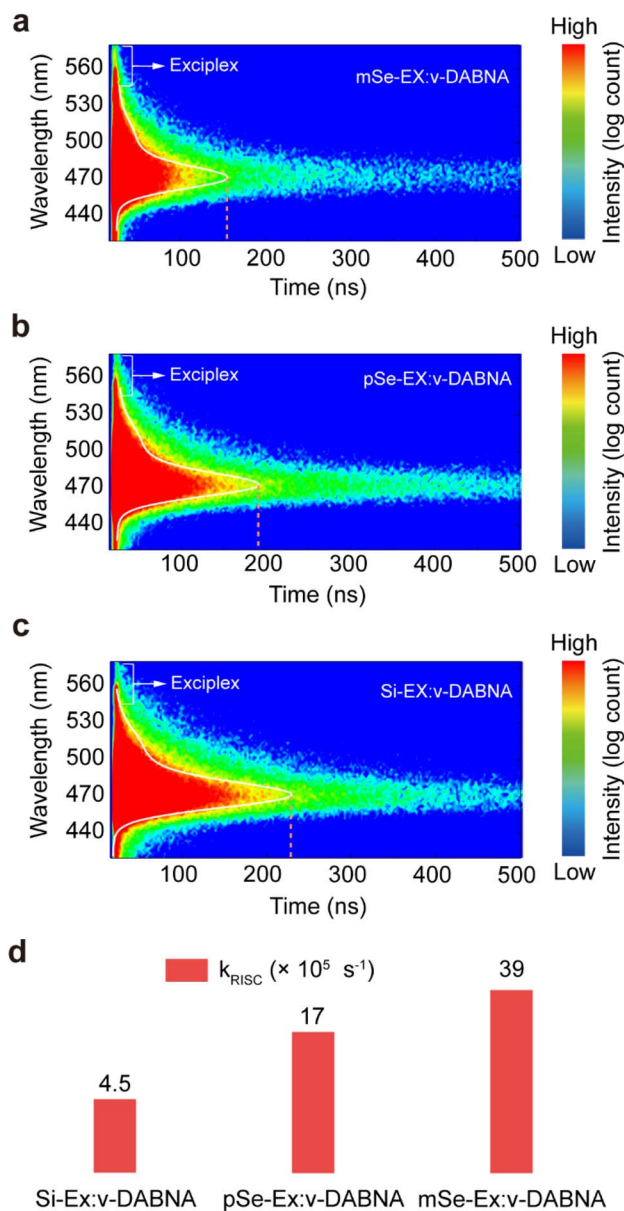


Fig. 3 (a)–(c) Time-dependent emission contour maps of (a) *mSe-EX:v-DABNA*, (b) *pSe-EX:v-DABNA*, and (c) *Si-EX:v-DABNA* films, recorded between 420 and 580 nm. (d) Comparison of calculated reverse intersystem crossing rate constants (k_{RISC}) of *mSe-EX:v-DABNA*, *pSe-EX:v-DABNA* and *Si-EX:v-DABNA* films.

contribution was highest for *mSe-EX:v-DABNA* (91%), followed by *pSe-EX:v-DABNA* (80%) and *Si-EX:v-DABNA* (45%). All doped films exhibited near-unity PL quantum yields of 99% (*mSe-EX:v-DABNA*), 97% (*pSe-EX:v-DABNA*), and 99% (*Si-EX:v-DABNA*). Consequently, the calculated k_{RISC} values (Fig. 3d) were progressively increased from $4.5 \times 10^5 \text{ s}^{-1}$ (*Si-EX:v-DABNA*) to $1.7 \times 10^6 \text{ s}^{-1}$ (*pSe-EX:v-DABNA*) and $3.9 \times 10^6 \text{ s}^{-1}$ (*mSe-EX:v-DABNA*). These results align closely with theoretical predictions, clearly demonstrating that exciplex hosts with fast k_{RISC} substantially accelerate the RISC process in MR-TADF emitters.

Encouraged by the near-unity PL quantum yield and accelerated RISC process of *v-DABNA* in the selenium-based

exciplex hosts, we fabricated corresponding OLED devices with the following architecture: indium tin oxide (50 nm)/1,4,5,8,9,11-hexaazatriphenylenehexacarbonitrile (HATCN, 5 nm)/4,4'-cyclohexylidenebis[*N,N*-bis(4-methylphenyl)benzenamine] (TAPC, 30 nm)/tris(4-carbazoyl-9-ylphenyl)amine (TCTA, 15 nm)/Se donor (15 nm)/Se donor:*SiTrzCz2:v-DABNA* (35 nm, 49.5 wt%:49.5 wt%:1 wt%)/*SiTrzCz2* (15 nm)/1-(4-(10-([1,1'-biphenyl]-4-yl)anthracen-9-yl)phenyl)-2-ethyl-1*H*-benzo[*d*]-imidazole (ANT-BIZ, 30 nm)/lithium quinolin-8-olate (Liq, 2 nm)/aluminium (Al, 100 nm). Specifically, for the emitting layers, *v-DABNA* was co-evaporated with the *mSe-EX* (*mSeCzCz:SiTrzCz2*) host for device A, and the *pSe-EX* (*pSeCzCz:SiTrzCz2*) host for device B. For comparison, a control device based on the *Si-EX* (*SiCzCz:SiTrzCz2*) host was also prepared. In this device architecture, HATCN, TAPC and TCTA served as hole-injection and hole-transport layers, while ANT-BIZ and Liq functioned as the electron-transport and electron-injection layers, respectively. Additionally, Se-containing donors (*mSeCzCz* or *pSeCzCz*) and *SiTrzCz2* acted as electron-blocking and hole-blocking layers, respectively. The device structure and corresponding energy level diagram are illustrated in Fig. 4a and b, and chemical structures of the organic materials are provided in Fig. S17. The electroluminescence properties are summarized in Table 2.

All fabricated devices exhibited nearly identical blue emission profiles (Fig. S18) peaking at 470 nm, with a narrow FWHM of 17 nm, closely matching the corresponding PL spectra obtained from thin films. This indicates complete FRET from the exciplex hosts to the *v-DABNA* emitter. According to the current density–voltage–luminance (J - V - L) curves (Fig. 4c), device A exhibited a turn-on voltage of approximately 3.2 V, an operating voltage of 4.2 V at a luminance of 1000 cd m^{-2} , and a maximum luminance of $36\,603 \text{ cd m}^{-2}$. In comparison, device B showed a higher turn-on voltage of approximately 3.6 V, an operating voltage of 5.0 V at 1000 cd m^{-2} , and a lower maximum luminance of $33\,204 \text{ cd m}^{-2}$. The EL spectra remained stable under high-brightness ($\sim 10\,000 \text{ cd m}^{-2}$) or high-voltage ($\sim 8 \text{ V}$) for all devices, indicating that the exciton recombination zone was essentially unchanged under these operating conditions (Fig. S19). Device A also achieved higher maximum current efficiency (39.7 cd A^{-1}) and power efficiency (39.0 lm W^{-1}) compared with device B, which displayed maximum current and power efficiencies of 31.1 cd A^{-1} and 27.1 lm W^{-1} , respectively (Fig. 4d).

As shown in Fig. 4e, device A achieved a maximum EQE of 40.9% (average: $40.45 \pm 0.29\%$, based on the measurements of 11 independent devices; Fig. S20), substantially exceeding those of device B (33.7%) and the control device (34.1%). This exceptional performance stemmed from the near-unity PL quantum yield of the emissive layer and favorable horizontal orientation of the transition dipole moment of *v-DABNA* doped in the *mSe-EX* host (transition dipole moment vector $S = -0.46$, Fig. S21).⁴⁷ Compared to the control device, devices A and B demonstrated significantly reduced efficiency roll-off, resulting from enhanced SOC values between the singlet and triplet manifolds provided by the *mSe-EX* and *pSe-EX*



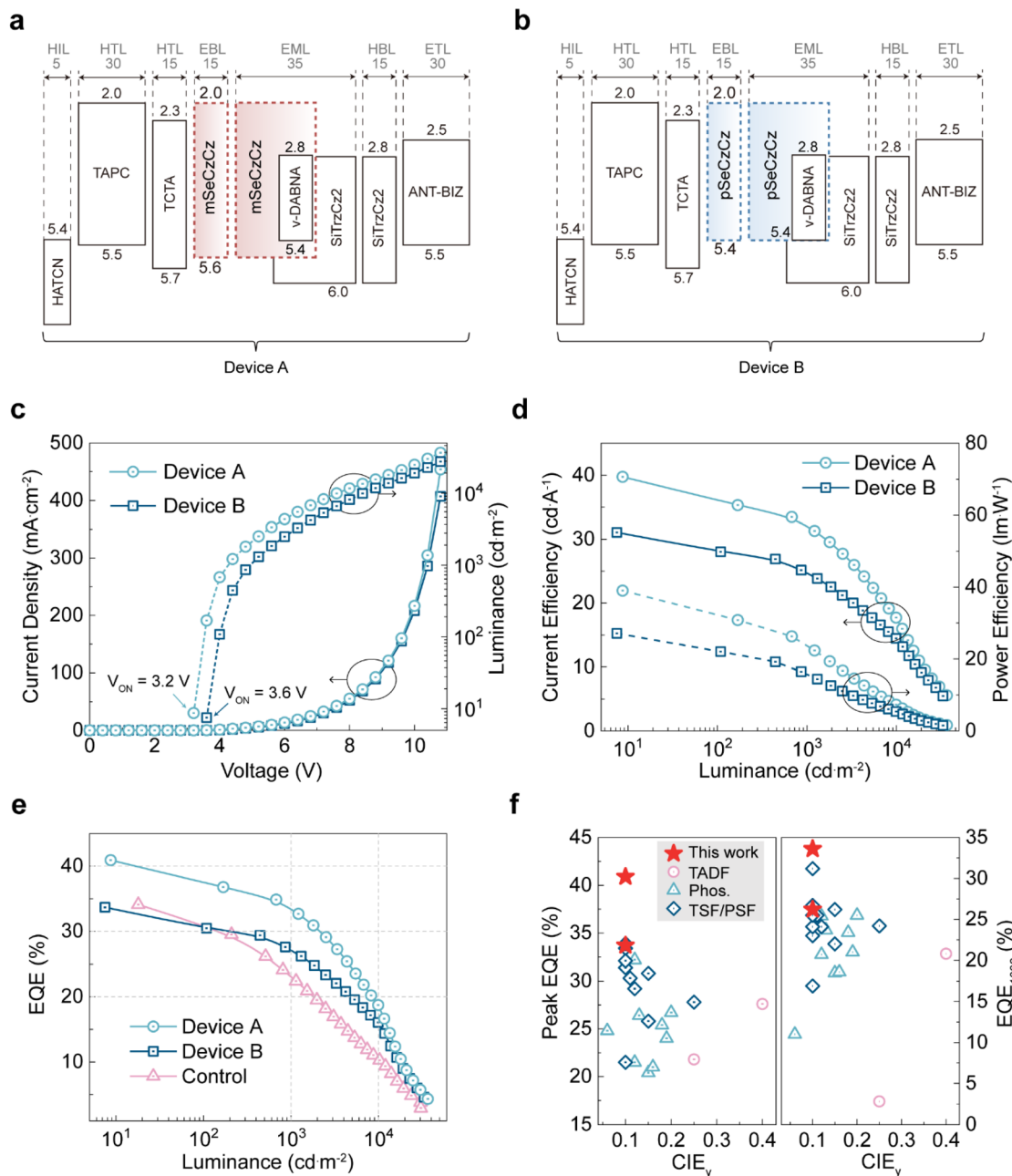


Fig. 4 (a) and (b) Device architecture and energy-level diagram of OLEDs based on (a) *mSeCzCz*:*SiTrzCz2*:*v-DABNA* (device A) and (b) *pSeCzCz*:*SiTrzCz2*:*v-DABNA* (device B). (c) Current density and luminance versus voltage (J - V - L) characteristics, and (d) current and power efficiency versus luminance curves for device A and device B. (e) External quantum efficiency (EQE) versus luminance plots of device A, device B and the control device. (f) Peak EQE and EQE at 1000 cd m^{-2} (EQE_{1000}) plotted against the Commission Internationale de l'Éclairage y -coordinate (CIE_y) for representative blue OLEDs employing exciplex-forming co-hosts. Definitions and references for these OLEDs are summarized in Table S1. TSF, TADF-sensitized fluorescence; PSF, phosphorescence-sensitized fluorescence.

hosts, which effectively suppresses triplet-involved annihilation processes at high brightness levels. Notably, owing to the stronger SOC provided by the *mSe-EX* host, device A exhibited considerably lower efficiency roll-off compared to recent blue OLEDs utilizing exciplex-forming co-host systems. Remarkably, device A maintained an EQE of 33.6% at a practical luminance of 1000 cd m^{-2} , surpassing those of state-of-the-art TADF, phosphorescent, TADF-sensitized

hyperfluorescent and phosphorescence-sensitized hyperfluorescent OLEDs based on exciplex-forming co-hosts (Fig. 4f and Table S1). These results clearly demonstrate that the exciplex hosts with fast RISC can effectively accelerate the RISC process in MR-TADF emitters, thereby substantially enhancing device performance under realistic display operating conditions.



Table 2 Summary performance of the devices

Device	λ_{PL}^a [nm]	L_{max}^b [cd m ⁻²]	EQE ^c [%]			CE ^d [cd A ⁻¹]		PE ^e [lm W ⁻¹]		FWHM ^f [nm]	CIE(x,y) ^g
			Max	1000 cd m ⁻²	Roll-off	Max	1000 cd m ⁻²	Max	1000 cd m ⁻²		
Device A	470	36 603	40.9	33.6	17.8	39.7	32.2	39.0	23.9	17	(0.13, 0.12)
Device B	470	33 204	33.7	26.2	22.3	31.1	23.9	27.1	14.4	17	(0.12, 0.12)
Control	470	30 511	34.1	22.4	34.3	35.8	23.1	40.2	16.5	17	(0.13, 0.12)

^a Electroluminescence peak wavelength recorded at 1000 cd m⁻². ^b Maximum luminance. ^c Maximum external quantum efficiency, and value at 1000 cd m⁻². ^d Maximum current efficiency, and value at 1000 cd m⁻². ^e Maximum power efficiency, and value at 1000 cd m⁻². ^f Full width at half-maximum. ^g CIE coordinates.

Conclusions

In summary, we have developed a heavy-atom-incorporated exciplex with k_{RISC} as far as $4.9 \times 10^6 \text{ s}^{-1}$, an order of magnitude higher than the typical values for exciplexes. The heavy atom participates directly in the triplet-state orbital distribution, resulting in strong spin-orbit coupling; combined with positional isomer optimization, this accelerates the spin-flip process. Notably, employing this exciplex with fast RISC as a host also enhances the k_{RISC} of the MR-TADF emitter to the 10^6 s^{-1} range. Devices employing the selenium-containing **mSe-EX** host exhibited exceptional electroluminescence performance, achieving a high EQE_{max} of 40.9% and minimal efficiency roll-off (EQE = 33.6% at 1000 cd m⁻²). We believe that the simple design and in-depth structure-property relationship established here would aid in the development of exciplex systems with efficient spin-flip transition and benefit the electroluminescence efficiency in narrowband OLEDs.

Author contributions

Z. Chen and C. Yang conceived and supervised the project. M. Wang synthesized and characterized the hosts, and performed photophysical measurements under the supervision of Z. Chen. Z. Chen and M. Huang fabricated the OLEDs and characterized the performance of the devices. Z. Chen performed theoretical calculations. D. Wang and Z.-H. Lu contributed to the UPS results. Z. Chen, M. Huang, C. Zhong, and C. Yang participated in discussions. All authors engaged in result analysis and provided feedback on the manuscript.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

The data supporting this article have been included as part of the SI. Supplementary information: all synthetic procedures, characterization data, spectroscopic data, supplementary figures and tables, and detailed information. See DOI: <https://doi.org/10.1039/d5sc06200d>.

CCDC 2408523 (**mSeCzCz**) contains the supplementary crystallographic data for this paper.⁴⁸

Acknowledgements

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