SUPPLEMENTARY INFORMATION

S1. Circularly polarized orbital transitions

In the main publication text we focus on only one circularly polarized point group (C_{3h}), thus here in Table 1 we present detailed information on which orbital transitions lead to circularly polarized triplet with $m_S = \pm 1$ for other point groups. For each point group the table includes, circularly polarized irreducible representation, symmetry of the orbital part of the triplet. The two last columns show pairs of orbitals between which transition result in CP triplet with $m_S = \pm 1$. They are separated into two categories: first one, transitions in which singlet and triplet with $m_S = 0$ are forbidden, the second category consist of transitions in which singlet and triplet with $m_S = 0$ are allowed, thus the triplet of interest can be hard to resolve. In practice, if in a molecule with a given symmetry the HOMO-LUMO pair (or HOMO-LUMO+1, HOMO-1-LUMO) is one of listed pairs, there is a great chance that triplet with $m_S = \pm 1$ associated with this transition is selectively circularly polarized.

POINT GROUP	CP irreducible representation	Orbital part symmetry of CP spin-polarized triplets	Orbital pairs leading to CP triplet with ms=±1 **without** other active states nearby	Orbital pairs leading to CP triplet with ms=±1 **with** other active states near
C_3	E	A, E		a - a, a - e, a - e
C_4	E	A,B	a - b	a - a, b - b, e - e
C_5	E_1	A, E_1	$a - e_2$	$a - a$, $e_1 - e_1$, $e_2 - e_2$, $e_1 - e_2$
C_6	E_1	A, E_2	$a - e_2$, b - e1	$a - a$, $b - b$, $e_1 - e_1$, $e_2 - e_2$
C_{3h}	E'	A'', E''	$a' - e''$, $a'' - e'$	$a' - a''$, $e' - e''$
C_{4h}	E_u	A_u, B_u, E_u	$a_g - b_u$, $a_u - b_g$	$a_g - a_u$, $b_g - b_u$, $a_g - e_u$, $b_g - e_u$, $a_u - e_g$, $b_u - e_g$, $e_g - e_u$
C_{5h}	E_1'	A'',E_2''	$a' - e_2''$, $a'' - e_2'$, $e_1' - e_2''$	$a' - a''$, $e_1' - e_1''$, $e_2' - e_2''$, $e_1' - e_2''$
C_{6h}	E_{1u}	A_u, E_{2u}	$a_g - e_{2u}$, $b_g - e_{1u}$, $e_{1g} - a_u$, $b_u - e_{2g}$	$a_g - a_u$, $b_u - b_g$, $e_{1g} - e_{1u}$, $e_{2g} - e_{2u}$, $e_{1g} - e_{2u}$
S_4	E	A, B	a - a, b - b	e - e, a - b
S_6	E_u	A_u, E_u		$a_g - a_u$, $e_g - a_u$, $e_g - e_u$, $a_g - e_u$
S_8	E_1	B, E_2	$a - e_2$, $b - e_2$, $e_1 - e_1$, $e_3 - e_3$	$a - b$, $e_1 - e_3$, $e_2 - e_2$

Table 1 Table presenting point groups supporting circularly polarized (CP) transitions and list of orbital transitions leading to possible spin-injection via CPL.

S2. C_{3h} Double group character and product tables

Table 2 and Table 3 present character table and cross product table respectively for C_{3h} double group.

		E	$\bar{E}$	S_3^2	$\bar{S}_3^2$	C_3	$\bar{C}_3$	σ_h	$\bar{\sigma}_h$	C_3^2	$\bar{C}_3^2$	S_3	$\bar{S}_3$	
Γ_1	A’	1	1	1	1	1	1	1	1	1	1	1	1	$x^2+y^2+z^2$
Γ_2	E’	1	1	$-\omega^2$	$-\omega^2$	ω^4	ω^4	1	1	$-\omega^2$	$-\omega^2$	ω^4	ω^4	i(x-iy)
Γ_3	E’	1	1	ω^4	ω^4	$-\omega^2$	$-\omega^2$	1	1	ω^4	ω^4	$-\omega^2$	$-\omega^2$	i(x+iy)
Γ_4	A"	1	1	-1	-1	1	1	-1	-1	1	1	-1	-1	z
Γ_5	E"	1	1	ω^2	ω^2	ω^4	ω^4	-1	-1	$-\omega^2$	$-\omega^2$	$-\omega^4$	$-\omega^4$	
Γ_6	E"	1	1	$-\omega^4$	$-\omega^4$	$-\omega^2$	$-\omega^2$	-1	-1	ω^4	ω^4	ω^2	ω^2	
Γ_7		1	-1	ω	$-\omega$	ω^2	ω^2	-i	i	$-\omega^4$	ω^4	$-\omega^5$	ω^5	Ψ(½,½)=’spin up’
Γ_8		1	-1	$-\ \omega^5$	ω^5	$-\omega^4$	ω^4	i	-i	ω^2	$-\omega^2$	ω	$-\omega$	Ψ(½,-½)=’spin down’
Γ_9		1	-1	$-\omega$	ω	ω^2	$-\omega^2$	i	-i	$-\omega^4$	ω^4	ω^5	$-\ \omega^5$	
Γ_{10}		1	-1	ω^5	$-\ \omega^5$	$-\omega^4$	ω^4	-i	i	ω^2	$-\omega^2$	$-\omega$	ω	
Γ_{11}		1	-1	-i	i	-1	1	-i	i	-1	1	i	-i	
Γ_{12}		1	-1	i	-i	-1	1	i	-i	-1	1	-i	i	

Table 2 Double point group C_{3h} character table. First column shows labelling according to Koster’s book [1] and the second one to the point group online base [15]. The last column contains functions which transform as according irreducible representations. ω is defined as $e^{i\pi/6}$.

Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	Γ_6	Γ_7	Γ_8	Γ_9	Γ_{10}	Γ_{11}	Γ_{12}	
Γ_1	Γ_2	Γ_3	Γ_4	Γ_5	Γ_6	Γ_7	Γ_8	Γ_9	Γ_{10}	Γ_{11}	Γ_{12}	Γ_1
	Γ_3	Γ_1	Γ_5	Γ_6	Γ_4	Γ_{11}	Γ_9	Γ_{12}	Γ_7	Γ_{10}	Γ_8	Γ_2
		Γ_2	Γ_6	Γ_4	Γ_5	Γ_{10}	Γ_{12}	Γ_8	Γ_{11}	Γ_7	Γ_9	Γ_3
			Γ_1	Γ_2	Γ_3	Γ_9	Γ_{10}	Γ_7	Γ_8	Γ_{12}	Γ_{11}	Γ_4
				Γ_3	Γ_1	Γ_{12}	Γ_7	Γ_{11}	Γ_9	Γ_8	Γ_{10}	Γ_5
					Γ_2	Γ_8	Γ_{11}	Γ_{10}	Γ_{12}	Γ_9	Γ_7	Γ_6
						Γ_5	Γ_1	Γ_2	Γ_4	Γ_6	Γ_3	Γ_7
							Γ_6	Γ_4	Γ_3	Γ_2	Γ_5	Γ_8
								Γ_5	Γ_1	Γ_3	Γ_6	Γ_9
									Γ_6	Γ_5	Γ_2	Γ_{10}
										Γ_4	Γ_1	Γ_{11}
											Γ_4	Γ_{12}

Table 3 Product table of double group C_{3h} from [1].

S3.Triethyls optimized geometries

Below, we present optimized geometries of Bet$_3$, AlEt$_3$, GaEt$_3$ and InEt$_3$. Structures were optimized using Gaussian with B3LYP functional and aug-cc-pvdz basis set (for Bet$_3$, AlEt$_3$ and GaEt$_3$) and for InEt$_3$ combination of aug-cc-pvdz and lanl2dz.

22
BEt3

B	0.000000	0.000000	0.000000
C	0.000000	1.580374	0.000000
C	1.338653	2.333412	0.000000
H	-0.610230	1.905136	0.864061
H	-0.610230	1.905136	-0.864061
H	1.190902	3.422869	0.000000
H	1.942190	2.084805	0.884800
H	1.942190	2.084805	-0.884800
C	1.368644	-0.790187	0.000000
C	1.351468	-2.326014	0.000000
H	1.955011	-0.424094	0.864061
H	1.955011	-0.424094	-0.864061
H	2.368841	-2.742787	0.000000
H	0.834399	-2.724389	0.884800
H	0.834399	-2.724389	-0.884800
C	-1.368644	-0.790187	0.000000
C	-2.690121	-0.007399	0.000000
H	-1.344782	-1.481043	0.864061
H	-1.344782	-1.481043	-0.864061
H	-3.559743	-0.680083	0.000000
H	-2.776589	0.639583	0.884800
H	-2.776589	0.639583	-0.884800

22
AlEt3

C	0.000000	1.985257	0.000000
C	1.366717	2.697814	0.000000
H	-0.594489	2.321289	0.869741
H	-0.594489	2.321289	-0.869741
H	1.264577	3.794085	0.000000
H	1.965361	2.428940	0.883023
H	1.965361	2.428940	-0.883023
C	1.719283	-0.992629	0.000000
C	1.653017	-2.532518	0.000000
H	2.307540	-0.645802	0.869741
H	2.307540	-0.645802	-0.869741
H	2.653486	-2.992199	0.000000
H	1.120843	-2.916522	0.883023
H	1.120843	-2.916522	-0.883023
C	-1.719283	-0.992629	0.000000
C	-3.019734	-0.165296	0.000000
H	-1.713051	-1.675487	0.869741
H	-1.713051	-1.675487	-0.869741

H	-3.918063	-0.801887	0.000000
H	-3.086204	0.487582	0.883023
H	-3.086204	0.487582	-0.883023
Al	0.000000	0.000000	0.000000

22

GaEt3

Ga	0.000000	0.000000	0.000000
C	0.000000	2.004613	0.000000
C	1.375837	2.689965	0.000000
C	1.736046	-1.002307	0.000000
C	1.641659	-2.536492	0.000000
C	-1.736046	-1.002307	0.000000
C	-3.017496	-0.153472	0.000000
H	-0.589519	2.333175	0.873175
H	-0.589519	2.333175	-0.873175
H	1.289104	3.787810	0.000000
H	1.968735	2.411656	0.883693
H	1.968735	2.411656	-0.883693
H	2.315348	-0.656049	0.873175
H	2.315348	-0.656049	-0.873175
H	2.635788	-3.010302	0.000000
H	1.104188	-2.910802	0.883693
H	1.104188	-2.910802	-0.883693
H	-1.725829	-1.677126	0.873175
H	-1.725829	-1.677126	-0.873175
H	-3.924892	-0.777508	0.000000
H	-3.072923	0.499147	0.883693
H	-3.072923	0.499147	-0.883693

22

InEt3

C	0.000000	2.176194	0.000000
C	1.378191	2.860921	0.000000
H	-0.587777	2.510756	0.872634
H	-0.587777	2.510756	-0.872634
H	1.297091	3.960251	0.000000
H	1.972875	2.582662	0.883099
H	1.972875	2.582662	-0.883099
C	1.884639	-1.088097	0.000000
C	1.788535	-2.624009	0.000000
H	2.468267	-0.746348	0.872634
H	2.468267	-0.746348	-0.872634
H	2.781133	-3.103440	0.000000
H	1.250213	-2.999891	0.883099
H	1.250213	-2.999891	-0.883099
C	-1.884639	-1.088097	0.000000
C	-3.166725	-0.236912	0.000000
H	-1.880490	-1.764408	0.872634
H	-1.880490	-1.764408	-0.872634

H	-4.078224	-0.856812	0.000000
H	-3.223088	0.417229	0.883099
H	-3.223088	0.417229	-0.883099
In	0.000000	0.000000	0.000000

S4.TDDFT calculations for BEt$_3$, AlEt$_3$, GaEt$_3$ and InEt$_3$

For each of BEt$_3$, AlEt$_3$, GaEt$_3$ and InEt$_3$ molecule we calculated excited states using Gaussian and Dirac without and with spin included. We used the same geometry for each molecule to eliminate variations in result obtained using Gaussian and Dirac due to changes in molecular geometry. In Dirac we used ‘Exact 2-Component one-electron Hamiltonian’ (X2C) and Gaunt interactions added to Hamiltonian, which includes 2-electron spin-same and spin-other-orbit corrections. Additionally for calculations without spin we included keyword ‘.SPINFREE’.

For InEt$_3$, structure optimization as well as TDDFT in Gaussian were performed using aug-cc-pvdz basis sets for C and H atoms and lan2ldz basis set for In atom. In Dirac, TDDFT for InEt$_3$ was done using dyall.ae2z basis set for C and H atoms and dyall.v3z for In atom. In all abovementioned calculations we used B3LYP functional.

Below, we present the sets of lowest excited states calculated for Bet$_3$, AlEt3, GaEt3 and InEt$_3$ obtained using three methods described above (Table 4-Table 7).

The symmetries of states calculated in Dirac with spin are given in a C$_{3h}$ subgroup: C$_s$, because C$_{3h}$ point group is not implemented in Dirac. In C$_s$ point group A’ refers to A’ and E’ of C$_{3h}$; A’’ refers to A’’ and E’’ of C$_{3h}$. Thus if there are two states A’ or A’’ (C$_s$ notation) very close in energy to each other we interpret it as doubly degenerate E’ or E’’ respectively in C$_{3h}$ notation. Otherwise it is A’ or A’’ (in C$_{3h}$ point groups).

BEt$_3$

Gaussian		Dirac X2C Gaunt spin free		Dirac X2C Gaunt			
Singlets	Triplets	Singlets	Triplets	States	Symmetry		Oscillator strength
					Calculated (C$_s$ PG)	Interpretation (in C$_{3h}$)	
	5.5183 (E’’)		5.5285 (E’’)	5.5274	2 A’	E’	0.00000002 0.00000002
				5.5285	A’’	E’’	-
				5.5285	A’’		-
				5.5296	A’	A’	-
				5.5296	A’	A’	-
5.9914 (E’’)		6.0044 (E’’)		6.0043	A’’	E’’	-
				6.0043	A’’		-
…		…		….			
6.5856 (E’) f=0.0022		6.5657 (E’) f=0.0020		6.4975	A’	E’	0.00000072

Table 4 Table with energies of the lowest excited states (T1 and S1) for BEt$_3$ calculated using different TDDFT implementations and levels of theory. All calculations were performed on the same molecular geometry – optimized in Gaussian using B3LYP functional and aug-cc-pvdz basis set. Oscillator strengths for presented excited states calculated in Gaussian and in Dirac without spin included are equal zero. Energies are given in eV. The last row shows the next optically allowed state with its oscillator strength f.

$AlEt_3$

Gaussian		Dirac X2C Gaunt spin free		Dirac X2C Gaunt			
Singlets	Triplets	Singlets	Triplets	States	Symmetry		Oscillator strength
					Calculated (C_s PG)	Interpretation (in C_{3h})	
	4.6506 (E’’)		4.6557 (E’’)	4.6527	2 A’	E’	0.00000069
							0.00000069
				4.6557	A’’	E’’	-
				4.6557	A’’		-
				4.6588	A’	A’	-
				4.6588	A’	A’	-
4.8989 (E’’)		4.9028 (E’’)		4.9029	A’’	E’’	-
				4.9030	A’’		-
…		…		….			
5.4294 (E’) f=0.0604		5.4008 (E’) f= 0.0292		5.1790	A’	E’	0.00001144

Table 5 Table with energies of the lowest excited states (T1 and S1) for $AlEt_3$ calculated using different TDDFT implementations and levels of theory. All calculations were performed on the same molecular geometry – optimized in Gaussian using B3LYP functional and aug-cc-pvdz basis set. Oscillator strengths for presented excited states calculated in Gaussian and in Dirac without spin included are equal zero. Energies are given in eV. The last row shows the next optically allowed state with its oscillator strength f.

$GaEt_3$

Gaussian		Dirac X2C Gaunt spin free		Dirac X2C Gaunt			
Singlets	Triplets	Singlets	Triplets	States	Symmetry		Oscillator strength
					Calculated (C_s PG)	Interpretation (in C_{3h})	
	4.3738 (E’’)		4.37623 (E’’)	4.35807	2 A’	E’	0.00002861
							0.00002862
				4.37493	A’’	E’’	-
				4.37500	A’’		-
				4.39484	A’	A’	-
				4.39530	A’	A’	-
4.6374 (E’’)		4.6385 (E’’)		4.63984	A’’	E’’	-
				4.63990	A’’		-
…		…		….			
5.4024 (E’) f=0.0576		5.3663 (E’) f=0.0568		5.1319	A’	E’	0.0001915

Table 6 Table with energies of the lowest excited states (T1 and S1) for $GaEt_3$ calculated using different TDDFT implementations and levels of theory. All calculations were performed on the same molecular geometry – optimized in Gaussian using B3LYP functional and aug-cc-pvdz basis set. Oscillator strengths for presented excited states calculated in Gaussian and in Dirac without spin included are equal zero. Energies are given in eV. The last row shows the next optically allowed state with its oscillator strength f.

$InEt_3$

Gaussian		Dirac X2C Gaunt spin free		Dirac X2C Gaunt			
Singlets	Triplets	Singlets	Triplets	States	Symmetry		Oscillator strength
					Calculated (C_s PG)	Interpretation (in C_{3h})	
	4.0799 (E’’)		3.8775 (E’’)	3.8307	2 A’	E’	0.00021963
							0.00021967
				3.8665	A’’	E’’	-
				3.8666	A’’		-
				3.9281	A’	A’	-
				3.9322	A’	A’	-
4.2901 (E’’)		4.0830 (E’’)		4.0946	A’’	E’’	-
				4.0946	A’’		-
…		…		….			
5.0340 (E’) f=0.1148		5.4058 (E’) f= 0.2272		5.1790	A’	E’	0.00111988

Table 7 Table with energies of the lowest excited states (T1 and S1) for $InEt_3$ calculated using different TDDFT implementations and levels of theory. All calculations were performed on the same molecular geometry – optimized in Gaussian using B3LYP functional and aug-cc-pvdz basis set. Oscillator strengths for presented excited states calculated in Gaussian and in Dirac without spin included are equal zero. Energies are given in eV. The last row shows the next optically allowed state with its oscillator strength f.

[1] H. S. G.F. Koster, J.O. Dimmock, R.G. Wheeler, (1963).