

Supplementary information

Predicting Intersystem Crossing Rate Constants of Alkoxy-Radical Pairs with Structure-Based Descriptors and Machine Learning

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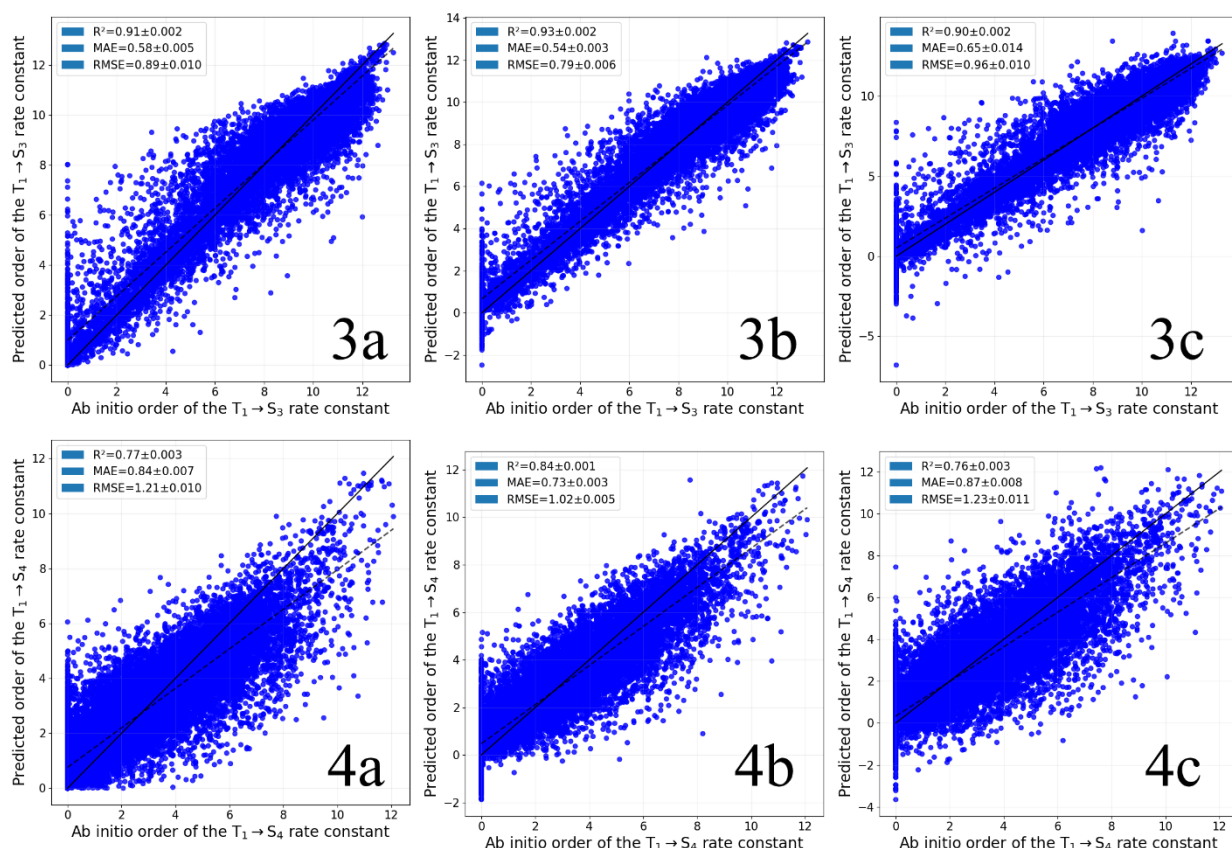


Figure S1. *Ab initio* versus predicted orders of the $T_1 \rightarrow S_3$ and $T_1 \rightarrow S_4$ rate constants, denoted as **3** and **4**, respectively. The predicted values, obtained using Random Forest, CatBoost, and Neural Network models, are labeled as a, b, and c, respectively. The solid black line represents $y = x$, while the dashed line corresponds to the linear regression fit. The legend displays the mean and standard deviation of R^2 , RMSE, and MAE, averaged over five runs.

Table S1. Root mean squared error (RMSE), mean absolute error (MAE), and coefficient of determination (R^2) obtained during hyperparameter tuning of Random Forest for predicting the order of $T_1 \rightarrow S_n$ ISC rate constants. The table presents only parameter sets where MAE differs by at most 0.01 from the best-performing model. The hyperparameters include max depth, number of estimators. The best-performing model is highlighted in **bold**, along with the **chosen model**. The parameters were optimized using Grid Search and Repeated K-Fold cross-validation.

Max. depth	Number of estimators	RMSE	MAE	R^2
30	1000	0.64±0.0014	0.43±0.0007	0.81±0.0013
30	1250	0.64±0.0013	0.43±0.0006	0.81±0.0013
30	1500	0.64±0.0013	0.43±0.0006	0.81±0.0013
30	1500	0.64±0.0013	0.43±0.0006	0.81±0.0013

Table S2. Root mean squared error (RMSE), mean absolute error (MAE), and coefficient of determination (R^2) obtained during hyperparameter tuning of feed-forward neural network for predicting the order of $T_1 \rightarrow S_n$ ISC rate constants. The table presents only parameter sets where MAE differs by at most 0.01 from the best-performing model. The hyperparameters include batch size, epochs, learning rate, number of hidden layers and number of neurons in each hidden layer. The best-performing model is highlighted in **bold**, along with the **chosen model**. The parameters were optimized using Grid Search and Repeated K-Fold cross-validation.

Batch size	Epochs	Learning rate	Number of hidden layers	Number of neurons in hidden layers	RMSE	MAE	R ²
16	50	0.001	2	200	0.68±0.002	0.47±0.002	0.79±0.002
16	50	0.001	3	200	0.68±0.001	0.47±0.001	0.78±0.002
16	50	0.001	4	100	0.68±0.005	0.47±0.003	0.78±0.004
16	50	0.001	4	150	0.68±0.004	0.47±0.003	0.78±0.003
16	50	0.001	4	200	0.68±0.002	0.47±0.002	0.79±0.002
16	100	0.001	3	100	0.69±0.003	0.48±0.003	0.78±0.003
16	100	0.001	3	150	0.69±0.003	0.47±0.001	0.78±0.002
16	100	0.001	3	200	0.69±0.002	0.47±0.002	0.78±0.002
16	100	0.001	4	50	0.69±0.006	0.47±0.003	0.78±0.004
16	100	0.001	4	100	0.69±0.001	0.47±0.001	0.78±0.001
16	100	0.001	4	150	0.69±0.002	0.47±0.002	0.78±0.001
16	100	0.001	4	200	0.69±0.004	0.47±0.003	0.78±0.003
32	50	0.001	4	150	0.68±0.005	0.47±0.004	0.78±0.004
32	50	0.001	4	200	0.68±0.004	0.47±0.003	0.79±0.003
32	100	0.001	4	100	0.69±0.001	0.47±0.001	0.78±0.001
32	100	0.001	4	150	0.70±0.003	0.47±0.002	0.78±0.003
32	100	0.001	4	200	0.70±0.002	0.47±0.002	0.78±0.002
64	50	0.001	4	200	0.69±0.003	0.47±0.003	0.78±0.001
16	50	0.001	4	200	0.68±0.002	0.47±0.002	0.79±0.002

Table S3. Root mean squared error (RMSE), mean absolute error (MAE), and coefficient of determination (R²) obtained during hyperparameter tuning of CatBoost for predicting the order of T₁→S_n ISC rate constants. The table presents only parameter sets where MAE differs by at most 0.01 from the best-performing model. The hyperparameters include depth, number of estimators, and learning rate. The best-performing model is highlighted in **bold**, along with the **chosen model**. The parameters were optimized using Grid Search and Repeated K-Fold cross-validation.

Depth	Number of estimators	Learning rate	RMSE	MAE	R ²
8	1250	0.1	0.58±0.0009	0.40±0.0005	0.85±0.0010
8	1500	0.1	0.58±0.0010	0.39±0.0005	0.85±0.0010
9	1500	0.05	0.58±0.0013	0.40±0.0007	0.85±0.0012
9	1000	0.1	0.58±0.0013	0.40±0.0008	0.84±0.0013
9	1250	0.1	0.58±0.0014	0.39±0.0009	0.85±0.0013
9	1500	0.1	0.57±0.0014	0.39±0.0009	0.85±0.0013
10	1250	0.05	0.58±0.0011	0.40±0.0007	0.84±0.0011
10	1500	0.05	0.58±0.0010	0.39±0.0007	0.85±0.0011
10	1000	0.1	0.58±0.0014	0.39±0.0006	0.85±0.0012
10	1250	0.1	0.58±0.0014	0.39±0.0006	0.85±0.0012
10	1500	0.1	0.57±0.0014	0.39±0.0007	0.85±0.0013
10	1500	0.1	0.57±0.0014	0.39±0.0007	0.85±0.0013

Table S4. Root mean squared error (RMSE), coefficient of determination (R²), mean absolute error (MAE), and the MAE for each of the 10 considered dimers, comparing ab initio and predicted orders of the ISC rate constants (T₁→S₁, T₁→S₂, T₁→S₃, T₁→S₄, T₁→S_n) using Random Forest regressor. The metrics were obtained using the optimal set of custom descriptors.

	T ₁ →S ₁	T ₁ →S ₂	T ₁ →S ₃	T ₁ →S ₄	T ₁ →S _n
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MSE	0.99±0.010	0.47±0.015	0.80±0.017	1.46±0.023	0.38±0.009
RMSE	0.99±0.005	0.69±0.011	0.89±0.010	1.21±0.010	0.62±0.007
MAE	0.76±0.004	0.46±0.004	0.58±0.005	0.84±0.007	0.42±0.004
R ²	0.89±0.002	0.83±0.007	0.91±0.002	0.77±0.003	0.83±0.006
MAE(OAce-OAce)	0.77±0.013	0.47±0.004	0.54±0.012	1.02±0.014	0.42±0.006
MAE(OAce-OEt)	0.73±0.008	0.36±0.008	0.53±0.011	0.77±0.014	0.32±0.006
MAE(OAce-HOBuO)	0.81±0.013	0.56±0.005	0.72±0.020	1.04±0.017	0.52±0.005
MAE(OAce-MeO)	0.77±0.010	0.42±0.008	0.49±0.013	0.82±0.022	0.38±0.009
MAE(OEt-OEt)	0.56±0.014	0.30±0.007	0.38±0.013	0.59±0.007	0.23±0.007
MAE(OEt-HOBuO)	0.76±0.008	0.55±0.009	0.56±0.011	0.74±0.019	0.53±0.007
MAE(OEt-MeO)	0.68±0.010	0.27±0.007	0.34±0.008	0.51±0.007	0.23±0.007
MAE(HOBuO-HOBuO)	0.82±0.008	0.70±0.017	0.85±0.011	1.03±0.018	0.64±0.017
MAE(HOBuO-MeO)	0.80±0.006	0.43±0.006	0.69±0.013	0.97±0.013	0.39±0.003
MAE(MeO-MeO)	0.73±0.017	0.25±0.006	0.32±0.005	0.67±0.013	0.20±0.005

Table S5. Root mean squared error (RMSE), coefficient of determination (R²), mean absolute error (MAE), and the MAE for each of the 10 considered dimers, comparing ab initio and predicted orders of the ISC rate constants (T₁→S₁, T₁→S₂, T₁→S₃, T₁→S₄, T₁→S_n) using CatBoost regressor. The metrics were obtained using the optimal set of custom descriptors.

	T ₁ →S ₁	T ₁ →S ₂	T ₁ →S ₃	T ₁ →S ₄	T ₁ →S _n
MSE	0.89±0.008	0.38±0.010	0.63±0.009	1.04±0.010	0.31±0.005
RMSE	0.95±0.004	0.62±0.008	0.79±0.006	1.02±0.005	0.56±0.004
MAE	0.72±0.003	0.42±0.003	0.54±0.003	0.73±0.003	0.38±0.002
R ²	0.90±0.001	0.87±0.005	0.93±0.002	0.84±0.001	0.86±0.004
MAE(OAce-OAce)	0.75±0.018	0.44±0.006	0.52±0.011	0.82±0.014	0.38±0.007
MAE(OAce-OEt)	0.69±0.010	0.34±0.009	0.52±0.015	0.69±0.014	0.31±0.009
MAE(OAce-HOBuO)	0.77±0.007	0.51±0.007	0.65±0.008	0.88±0.011	0.47±0.008
MAE(OAce-MeO)	0.74±0.010	0.40±0.009	0.47±0.014	0.72±0.022	0.36±0.007
MAE(OEt-OEt)	0.55±0.012	0.29±0.004	0.36±0.015	0.52±0.008	0.23±0.009
MAE(OEt-HOBuO)	0.71±0.010	0.49±0.005	0.53±0.011	0.65±0.010	0.46±0.008
MAE(OEt-MeO)	0.66±0.014	0.27±0.006	0.35±0.007	0.50±0.006	0.22±0.006
MAE(HOBuO-HOBuO)	0.77±0.012	0.62±0.015	0.74±0.016	0.86±0.015	0.56±0.010
MAE(HOBuO-MeO)	0.75±0.004	0.39±0.005	0.61±0.005	0.82±0.006	0.36±0.005
MAE(MeO-MeO)	0.71±0.016	0.24±0.006	0.30±0.006	0.61±0.008	0.19±0.004

Table S6. Root mean squared error (RMSE), coefficient of determination (R²), mean absolute error (MAE), and the MAE for each of the 10 considered dimers, comparing ab initio and predicted orders of the ISC rate constants (T₁→S₁, T₁→S₂, T₁→S₃, T₁→S₄, T₁→S_n) using a feed-forward neural network. The metrics were obtained using the optimal set of custom descriptors.

	T ₁ →S ₁	T ₁ →S ₂	T ₁ →S ₃	T ₁ →S ₄	T ₁ →S _n
MSE	1.11±0.013	0.52±0.018	0.91±0.019	1.52±0.028	0.42±0.005
RMSE	1.05±0.006	0.72±0.013	0.96±0.010	1.23±0.011	0.65±0.004
MAE	0.80±0.002	0.49±0.007	0.65±0.014	0.87±0.008	0.45±0.003
R ²	0.88±0.002	0.82±0.009	0.90±0.002	0.76±0.003	0.81±0.004
MAE(OAce-OAce)	0.83±0.016	0.48±0.014	0.58±0.013	0.96±0.015	0.44±0.017
MAE(OAce-OEt)	0.73±0.013	0.39±0.023	0.56±0.021	0.77±0.011	0.35±0.007
MAE(OAce-HOBuO)	0.87±0.014	0.60±0.008	0.81±0.022	1.07±0.008	0.55±0.012
MAE(OAce-MeO)	0.80±0.003	0.45±0.012	0.58±0.022	0.85±0.018	0.41±0.008
MAE(OEt-OEt)	0.62±0.019	0.34±0.008	0.46±0.046	0.61±0.014	0.27±0.020
MAE(OEt-HOBuO)	0.80±0.011	0.56±0.012	0.62±0.030	0.77±0.028	0.54±0.016
MAE(OEt-MeO)	0.72±0.028	0.33±0.012	0.42±0.012	0.56±0.019	0.26±0.004
MAE(HOBuO-HOBuO)	0.91±0.021	0.77±0.017	0.95±0.022	1.07±0.023	0.70±0.012

MAE(HOBuO-MeO)	0.83±0.003	0.44±0.018	0.76±0.031	1.00±0.022	0.40±0.007
MAE(MeO-MeO)	0.76±0.015	0.29±0.015	0.36±0.033	0.69±0.026	0.23±0.010

Table S4. Mean absolute error (MAE), root mean squared error (RMSE), and coefficient of determination (R^2) for predicting the order of the $T_1 \rightarrow S_n$ ISC rate constant, obtained using a CatBoost model with descriptors ranked via recursive feature elimination (RFE). Descriptors are listed in order of decreasing importance, and the metrics were computed using the descriptor set that includes the descriptor in each row and all descriptors ranked higher.

	MAE	RMSE	R^2
$\angle(A1,C1,Ou1)$	0.984	1.373	0.132
$\angle(B2,C2,Ou2)$	0.817	1.140	0.402
$\angle(B1,C1,Ou1)$	0.722	1.015	0.526
C1-Ou1	0.669	0.951	0.583
B1-C1	0.627	0.898	0.628
$\angle(A2,C2,Ou2)$	0.578	0.817	0.693
C2-Ou2	0.556	0.787	0.715
$\angle(A1,C1,B1)$	0.524	0.747	0.743
$\angle(A2,C2,B2)$	0.500	0.716	0.764
Ou1-H21	0.474	0.680	0.787
$\angle(B1,C1,H11)$	0.464	0.667	0.795
$\angle(B2,C2,H12)$	0.455	0.654	0.803
B2-C2	0.449	0.645	0.808
C1-C2	0.437	0.624	0.820
min(O-O)	0.424	0.610	0.829
A1-C1	0.421	0.605	0.831
$\angle(Ou1,C1,H11)$	0.416	0.601	0.833
Ou1-Op1	0.410	0.593	0.838
Ou2-Op2	0.398	0.576	0.847
$\angle(Ou2,C2,H12)$	0.396	0.573	0.849
$\angle(A2,C2,H12)$	0.395	0.574	0.848
H12-Ou2	0.394	0.571	0.850
$\angle(A1,C1,H11)$	0.394	0.571	0.849
Ou2-H22	0.391	0.566	0.852
Ou1-Ou2	0.388	0.562	0.855
H11-Ou1	0.388	0.561	0.855
C2-Ou1	0.385	0.558	0.857
C1-Ou2	0.382	0.555	0.858
A2-C2	0.382	0.554	0.859
$\angle(C1,B1,Op1,H21)$	0.378	0.551	0.860
H11-C1	0.378	0.551	0.860
Op1-H21	0.378	0.551	0.860
h,bonds	0.375	0.547	0.862
$\angle(C2,B2,Op2,H22)$	0.374	0.545	0.863
H12-C2	0.373	0.545	0.863
Op2-H22	0.373	0.544	0.864
min(O-O)	0.373	0.544	0.863
Ou2-Op1	0.372	0.543	0.864
Ou1-Op2	0.372	0.544	0.864
min(H-H)	0.373	0.545	0.863
$\angle(C1,Ou1,C2,Ou2)$	0.374	0.545	0.863
A1-Ou2	0.374	0.545	0.863
C1-B2	0.373	0.545	0.863
B1-Ou2	0.373	0.544	0.863

Ou2-H21	0.372	0.544	0.864
Ou1-A2	0.373	0.545	0.863
B1-C2	0.373	0.545	0.863
$\angle(C1,A1,A2,C2)$	0.373	0.545	0.863
Ou1-B2	0.374	0.546	0.863
$\angle(C1,B1,B2,C2)$	0.374	0.546	0.863
B1-B2	0.374	0.546	0.863
unpaired hbonds	0.374	0.546	0.862
$\angle(Ou1,C1,C2,Ou2)$	0.375	0.547	0.862
A1-C2	0.375	0.547	0.862
$\angle(B1,C1,B2,C2)$	0.375	0.547	0.862
C1-A2	0.375	0.548	0.862
Ou1-H22	0.375	0.548	0.862
A1-A2	0.375	0.548	0.862
Op1-H22	0.375	0.548	0.861

Table S5. Mean absolute error (MAE), root mean squared error (RMSE), and coefficient of determination (R^2) for predicting the order of the $T_1 \rightarrow S_1$ ISC rate constant, obtained using a CatBoost model with descriptors ranked via recursive feature elimination (RFE). Descriptors are listed in order of decreasing importance, and the metrics were computed using the descriptor set that includes the descriptor in each row and all descriptors ranked higher.

	MAE	RMSE	R^2
C1-C2	1.397	1.749	0.663
min_dist	1.176	1.491	0.755
Ou1-Ou2	1.033	1.341	0.802
min(O-O)	0.991	1.289	0.817
C1-Ou2	0.929	1.212	0.838
Ou1-C2	0.905	1.182	0.846
B1-Ou2	0.878	1.146	0.855
Ou1-B2	0.860	1.122	0.861
min(O-O)	0.838	1.097	0.867
$\angle(B2,C2,Ou2)$	0.808	1.059	0.876
B1-C1	0.790	1.037	0.882
C2-Ou2	0.783	1.028	0.883
$\angle(A1,C1,Ou1)$	0.776	1.019	0.885
Ou1-H21	0.767	1.008	0.888
min(H-H)	0.763	1.002	0.889
B2-C2	0.760	1.000	0.890
Ou1-A2	0.756	0.993	0.891
A1-Ou2	0.749	0.984	0.893
unpaired hbonds	0.749	0.985	0.893
$\angle(A2,C2,Ou2)$	0.744	0.978	0.894
A2-C2	0.741	0.974	0.895
A1-C1	0.741	0.973	0.896
B1-C2	0.739	0.971	0.896
$\angle(B1,C1,Ou1)$	0.735	0.968	0.897
$\angle(A1,C1,B1)$	0.735	0.965	0.897
A1-C2	0.734	0.965	0.897
Ou2-H21	0.732	0.963	0.898
C1-Ou1	0.731	0.963	0.898
Ou1-Op2	0.732	0.964	0.898
$\angle(C1,Ou1,C2,Ou2)$	0.731	0.962	0.898
$\angle(Ou2,C2,H12)$	0.732	0.964	0.898
Op1-H21	0.732	0.963	0.898

$\angle(\text{A2,C2,H12})$	0.732	0.963	0.898
C1-B2	0.732	0.963	0.898
Ou2-Op1	0.729	0.960	0.899
C1-A2	0.730	0.962	0.898
$\angle(\text{A1,C1,H11})$	0.733	0.963	0.898
$\angle(\text{Ou1,C1,H11})$	0.732	0.962	0.898
Ou1-Op1	0.730	0.962	0.898
Ou1-H22	0.731	0.962	0.898
H11-C1	0.732	0.963	0.898
h,bonds	0.731	0.962	0.898
H12-C2	0.731	0.962	0.898
H11-Ou1	0.731	0.962	0.898
B1-B2	0.733	0.964	0.898
$\angle(\text{B2,C2,H12})$	0.732	0.963	0.898
$\angle(\text{C1,B1,B2,C2})$	0.734	0.965	0.897
$\angle(\text{B1,C1,H11})$	0.733	0.964	0.898
H12-Ou2	0.735	0.965	0.897
A1-A2	0.733	0.964	0.898
Ou2-Op2	0.733	0.964	0.898
$\angle(\text{B1,C1,B2,C2})$	0.735	0.967	0.897
$\angle(\text{C1,B1,Op1,H21})$	0.735	0.967	0.897
$\angle(\text{Ou1,C1,C2,Ou2})$	0.735	0.967	0.897
$\angle(\text{C1,A1,A2,C2})$	0.737	0.969	0.897
Op1-H22	0.737	0.970	0.896
Ou2-H22	0.736	0.968	0.897
Op2-H22	0.738	0.969	0.896
$\angle(\text{C2,B2,Op2,H22})$	0.735	0.967	0.897

Table S6. Mean absolute error (MAE), root mean squared error (RMSE), and coefficient of determination (R^2) for predicting the order of the $T_1 \rightarrow S_2$ ISC rate constant, obtained using a CatBoost model with descriptors ranked via recursive feature elimination (RFE). Descriptors are listed in order of decreasing importance, and the metrics were computed using the descriptor set that includes the descriptor in each row and all descriptors ranked higher.

	MAE	RMSE	R^2
$\angle(\text{B1,C1,Ou1})$	1.114	1.673	0.008
$\angle(\text{B2,C2,Ou2})$	1.101	1.653	0.031
$\angle(\text{A1,C1,Ou1})$	1.078	1.623	0.066
$\angle(\text{A1,C1,B1})$	1.074	1.614	0.077
C2-Ou2	1.067	1.606	0.086
Ou1-Op1	1.063	1.602	0.091
$\angle(\text{A2,C2,H12})$	1.056	1.590	0.103
C1-Ou1	1.042	1.569	0.127
$\angle(\text{Ou2,C2,H12})$	1.030	1.555	0.142
Ou1-H21	0.859	1.304	0.397
$\angle(\text{Ou1,C1,H11})$	0.784	1.187	0.500
$\angle(\text{A1,C1,H11})$	0.739	1.116	0.559
min(O-O)	0.670	0.975	0.663
$\angle(\text{A2,C2,Ou2})$	0.639	0.932	0.692
A2-C2	0.640	0.935	0.690
min dist	0.619	0.904	0.711
A1-C1	0.620	0.903	0.711
Ou2-Op2	0.619	0.901	0.712
$\angle(\text{C1,B1,Op1,H21})$	0.617	0.898	0.714

B2-C2	0.576	0.827	0.758
Ou1-Ou2	0.564	0.812	0.766
Ou1-B2	0.548	0.792	0.778
H11-C1	0.548	0.791	0.778
B1-C1	0.511	0.740	0.806
H12-Ou2	0.490	0.714	0.819
A1-Ou2	0.491	0.716	0.818
$\angle(\text{B1,C1,H11})$	0.492	0.715	0.819
H11-Ou1	0.486	0.708	0.822
Op1-H21	0.485	0.704	0.824
Ou1-C2	0.485	0.707	0.823
Op2-H22	0.479	0.697	0.828
Ou1-Op2	0.476	0.693	0.830
Ou2-H22	0.475	0.692	0.830
h,bonds	0.475	0.691	0.830
B1-Ou2	0.473	0.690	0.831
$\angle(\text{B2,C2,H12})$	0.474	0.690	0.831
min(O-O)	0.470	0.685	0.833
C1-C2	0.467	0.684	0.834
H12-C2	0.460	0.673	0.840
$\angle(\text{C1,Ou1,C2,Ou2})$	0.460	0.673	0.839
B1-B2	0.459	0.671	0.841
Ou1-A2	0.459	0.671	0.840
Ou2-Op1	0.458	0.669	0.841
min(H-H)	0.457	0.668	0.842
A1-C2	0.457	0.668	0.842
C1-Ou2	0.453	0.661	0.845
C1-B2	0.453	0.660	0.846
B1-C2	0.450	0.660	0.846
C1-A2	0.449	0.657	0.847
$\angle(\text{Ou1,C1,C2,Ou2})$	0.450	0.658	0.846
$\angle(\text{C1,A1,A2,C2})$	0.443	0.646	0.852
$\angle(\text{B1,C1,B2,C2})$	0.442	0.646	0.852
$\angle(\text{C2,B2,Op2,H22})$	0.439	0.644	0.853
A1-A2	0.440	0.646	0.852
$\angle(\text{C1,B1,B2,C2})$	0.441	0.645	0.852
Ou2-H21	0.441	0.646	0.852
Ou1-H22	0.438	0.640	0.855
unpaired hbonds	0.438	0.640	0.855
Op1-H22	0.436	0.638	0.856

Table S7. Mean absolute error (MAE), root mean squared error (RMSE), and coefficient of determination (R^2) for predicting the order of the $T_1 \rightarrow S_3$ ISC rate constant, obtained using a CatBoost model with descriptors ranked via recursive feature elimination (RFE). Descriptors are listed in order of decreasing importance, and the metrics were computed using the descriptor set that includes the descriptor in each row and all descriptors ranked higher.

	MAE	RMSE	R^2
$\angle(\text{A1,C1,Ou1})$	1.944	2.617	0.240
$\angle(\text{B2,C2,Ou2})$	1.546	2.208	0.459
$\angle(\text{A2,C2,Ou2})$	1.278	1.880	0.608
$\angle(\text{B1,C1,Ou1})$	1.170	1.733	0.667
C2-Ou2	1.049	1.573	0.725
$\angle(\text{Ou1,C1,H11})$	0.962	1.444	0.768
B1-C1	0.893	1.343	0.800

C1-Ou1	0.856	1.281	0.818
$\angle(A2,C2,H12)$	0.825	1.234	0.831
min_dist	0.775	1.161	0.850
Ou2-Op2	0.732	1.103	0.865
min(O-O)	0.689	1.035	0.881
Ou1-H21	0.644	0.971	0.895
A1-C1	0.635	0.951	0.900
$\angle(A1,C1,B1)$	0.623	0.934	0.903
H12-Ou2	0.615	0.922	0.906
Ou1-Op1	0.604	0.906	0.909
h,bonds	0.603	0.904	0.909
B2-C2	0.598	0.897	0.911
$\angle(Ou2,C2,H12)$	0.595	0.896	0.911
$\angle(A1,C1,H11)$	0.593	0.890	0.912
Op2-H22	0.588	0.884	0.913
Ou2-H22	0.583	0.876	0.915
$\angle(B1,C1,H11)$	0.577	0.866	0.917
Ou1-Ou2	0.575	0.860	0.918
C1-C2	0.571	0.852	0.919
$\angle(B2,C2,H12)$	0.566	0.842	0.921
Op1-H21	0.566	0.842	0.921
$\angle(C1,B1,Op1,H21)$	0.559	0.833	0.923
A2-C2	0.560	0.836	0.922
H11-C1	0.559	0.831	0.923
min(O-O)	0.559	0.831	0.923
Ou1-Op2	0.557	0.827	0.924
H11-Ou1	0.557	0.829	0.924
$\angle(C1,Ou1,C2,Ou2)$	0.557	0.826	0.924
C1-Ou2	0.557	0.828	0.924
min(H-H)	0.555	0.822	0.925
Ou1-B2	0.556	0.825	0.924
unpaired_hbonds	0.557	0.828	0.924
H12-C2	0.556	0.825	0.924
B1-Ou2	0.554	0.824	0.925
Ou1-C2	0.555	0.823	0.925
A1-Ou2	0.554	0.823	0.925
Ou2-Op1	0.554	0.823	0.925
$\angle(C2,B2,Op2,H22)$	0.551	0.819	0.925
C1-A2	0.551	0.818	0.926
Ou1-A2	0.552	0.820	0.925
Ou2-H21	0.552	0.820	0.925
B1-C2	0.550	0.818	0.926
C1-B2	0.551	0.820	0.925
A1-C2	0.552	0.818	0.926
B1-B2	0.553	0.820	0.925
$\angle(B1,C1,B2,C2)$	0.553	0.822	0.925
Ou1-H22	0.554	0.822	0.925
$\angle(Ou1,C1,C2,Ou2)$	0.556	0.827	0.924
A1-A2	0.556	0.824	0.925
$\angle(C1,A1,A2,C2)$	0.558	0.827	0.924
$\angle(C1,B1,B2,C2)$	0.560	0.829	0.924
Op2-H21	0.558	0.827	0.924

Table S8. Mean absolute error (MAE), root mean squared error (RMSE), and coefficient of determination (R^2) for predicting the order of the $T_1 \rightarrow S_4$ ISC rate constant, obtained using a CatBoost model with

descriptors ranked via recursive feature elimination (RFE). Descriptors are listed in order of decreasing importance, and the metrics were computed using the descriptor set that includes the descriptor in each row and all descriptors ranked higher.

	MAE	RMSE	R ²
B1-C1	1.915	2.342	0.134
$\angle$ (B2,C2,Ou2)	1.627	2.123	0.289
$\angle$ (A2,C2,Ou2)	1.461	1.959	0.394
B2-C2	1.351	1.832	0.470
min_dist	1.250	1.719	0.533
$\angle$ (B1,C1,Ou1)	1.179	1.637	0.577
$\angle$ (A1,C1,Ou1)	1.083	1.521	0.635
$\angle$ (A1,C1,B1)	1.034	1.451	0.667
$\angle$ (Ou1,C1,H11)	1.007	1.416	0.683
Ou1-Ou2	0.952	1.334	0.719
min(O-O)	0.936	1.309	0.729
$\angle$ (Ou2,C2,H12)	0.909	1.272	0.744
$\angle$ (B2,C2,H12)	0.885	1.244	0.756
min(H-H)	0.879	1.234	0.759
$\angle$ (A2,C2,H12)	0.864	1.215	0.767
C1-C2	0.855	1.201	0.772
A1-C1	0.845	1.185	0.778
Ou2-H22	0.820	1.153	0.790
Ou1-Op1	0.793	1.117	0.803
$\angle$ (B1,C1,H11)	0.785	1.102	0.808
Ou2-Op2	0.777	1.092	0.812
Ou1-H21	0.762	1.073	0.818
Op2-H22	0.763	1.073	0.818
C2-Ou2	0.762	1.070	0.819
C1-Ou1	0.762	1.070	0.819
H12-Ou2	0.757	1.063	0.821
A2-C2	0.760	1.067	0.820
$\angle$ (A1,C1,H11)	0.757	1.064	0.821
$\angle$ (C1,B1,Op1,H21)	0.751	1.055	0.824
Ou1-C2	0.749	1.051	0.826
h,bonds	0.748	1.050	0.826
C1-Ou2	0.745	1.045	0.827
$\angle$ (C2,B2,Op2,H22)	0.742	1.042	0.829
H11-Ou1	0.741	1.040	0.829
Op1-H21	0.741	1.039	0.829
min(O-O)	0.743	1.042	0.829
Ou1-Op2	0.744	1.041	0.829
unpaired hbonds	0.739	1.038	0.830
H11-C1	0.743	1.041	0.829
H12-C2	0.744	1.042	0.829
Ou2-H21	0.743	1.040	0.829
C1-B2	0.741	1.038	0.830
Ou2-Op1	0.741	1.038	0.830
B1-C2	0.741	1.037	0.830
$\angle$ (C1,Ou1,C2,Ou2)	0.741	1.037	0.830
C1-A2	0.743	1.038	0.830
B1-Ou2	0.743	1.038	0.830
Ou1-B2	0.742	1.037	0.830
A1-Ou2	0.742	1.038	0.830
$\angle$ (Ou1,C1,C2,Ou2)	0.744	1.040	0.829

A1-C2	0.746	1.042	0.828
B1-B2	0.747	1.041	0.829
Ou1-H22	0.743	1.039	0.830
Ou1-A2	0.745	1.039	0.829
A1-A2	0.746	1.043	0.828
$\angle(B1,C1,B2,C2)$	0.747	1.043	0.828
$\angle(C1,B1,B2,C2)$	0.749	1.044	0.828
$\angle(C1,A1,A2,C2)$	0.752	1.048	0.827
Op2-H21	0.751	1.047	0.827