**ELECTRONIC SUPPLEMENTARY MATERIAL**

**Nonlinear Optical Activity of Imino-dyes with Furan, Thiophene or Thiazole Moieties as π-Conjugated Bridge. A Computational Investigation**

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**S2-5** Cartesian coordinates of the CAM-B3LYP/6-31G (d,p) optimised ground-states of dyes **1-4.**

**S6-9** Cartesian coordinates of the CIS/6-31G(d,p) optimised excited states of dyes **1-4.**

Dye **1** (CAM-B3LYP/6-31G (d,p) optimised ground-state )

C -2.99870800 -1.24350700 -0.51497800

C -3.91400600 -2.18329400 -0.90123000

C -5.19918700 -1.60642200 -0.70740200

C -4.95965400 -0.35961200 -0.22580700

H -3.68886200 -3.17080900 -1.27579400

H -6.17109800 -2.03589200 -0.89356800

S -6.08081900 0.94136500 0.18027800

O -5.73425700 2.13041500 -0.58757700

O -7.40952900 0.34446000 0.08564600

C -5.70572200 1.27381800 1.89408100

H -5.94805400 0.39487400 2.49045600

H -4.64986900 1.53195600 1.97588200

H -6.32716700 2.12079300 2.18777700

C -1.55009200 -1.28626700 -0.47831100

H -1.11651800 -2.22427800 -0.84595600

N -0.85769200 -0.30204500 -0.06373000

C 0.54136000 -0.38904300 -0.00089800

C 1.26234500 0.80306400 -0.09831800

C 1.26182700 -1.57225900 0.18676300

C 2.64366900 0.81674500 -0.06673600

H 0.70709000 1.72682600 -0.21957200

C 2.64444800 -1.56882000 0.24369200

H 0.73647100 -2.51121800 0.33050100

C 3.37599700 -0.37389600 0.10470900

H 3.15772300 1.76352700 -0.17220700

H 3.16240400 -2.50448800 0.41096400

C 5.50311900 -1.57973300 0.51022500

C 5.56416500 0.83076200 0.26484700

C 6.95562000 -1.10863900 0.54203000

H 5.34229200 -2.38266700 -0.21703400

H 5.18665100 -1.95832200 1.49404700

C 6.83541200 0.35425200 0.97432400

H 5.04002500 1.58222400 0.87113400

H 7.57027500 -1.70825600 1.21613200

H 7.39622700 -1.17836100 -0.45685100

H 6.70355700 0.41576100 2.05885300

H 7.69360700 0.97080400 0.70145000

N 4.75373000 -0.38112100 0.14968100

C 5.86957000 1.43828600 -1.10966100

H 6.46123800 0.72926100 -1.69538300

H 4.93707900 1.61233600 -1.66229600

O 6.64817600 2.61430100 -1.00571800

H 6.10002800 3.30682100 -0.61840300

O -3.63604500 -0.11832400 -0.10112500

Dye **2**  (CAM-B3LYP/6-31G (d,p) optimised ground-state)

C -2.66850400 -0.94440600 -0.50009300

C -3.57942200 -1.84764300 -0.98692100

C -4.91906700 -1.41379600 -0.83438700

C -4.98720000 -0.19081400 -0.23145000

S -3.43429400 0.46875300 0.14534600

H -3.29159900 -2.78671100 -1.44406100

H -5.79739900 -1.95681800 -1.15881500

S -6.49008300 0.66497900 0.12771500

O -6.22750800 2.09722600 0.02761400

O -7.54137600 0.02469600 -0.65734100

C -6.79665100 0.29277200 1.85079400

H -6.91956900 -0.78331900 1.96744600

H -5.96626200 0.66340300 2.45118700

H -7.71779500 0.81278900 2.11780900

C -1.22062100 -1.06881200 -0.50218300

H -0.82676100 -1.99271900 -0.94338600

N -0.48268200 -0.14437500 -0.03051900

C 0.91332600 -0.27568500 0.01036300

C 1.67054700 0.89750400 -0.00312100

C 1.59691800 -1.49179400 0.09946200

C 3.05184200 0.86604800 0.01690400

H 1.14461700 1.84503300 -0.04784700

C 2.97906500 -1.53537000 0.14292900

H 1.04314900 -2.42226200 0.17510700

C 3.74731900 -0.35642000 0.09036900

H 3.59450400 1.80196100 -0.02038000

H 3.46819200 -2.49684600 0.23218300

C 5.83641500 -1.65620400 0.38748100

C 5.97451800 0.76295600 0.31945000

C 7.30357700 -1.23518800 0.44393400

H 5.64637400 -2.39654300 -0.39687300

H 5.51230000 -2.09728400 1.34226600

C 7.23292100 0.19514400 0.98295900

H 5.47915500 1.48506000 0.98275800

H 7.90123000 -1.90197100 1.06838800

H 7.73757600 -1.24594900 -0.56020400

H 7.10781800 0.18073700 2.06989700

H 8.10952500 0.80211600 0.75012500

N 5.12394100 -0.41061900 0.12359500

C 6.29105300 1.45725800 -1.01055100

H 6.85500900 0.77311100 -1.65065100

H 5.36076700 1.70007500 -1.54053200

O 7.10861700 2.59687900 -0.83031900

H 6.58629300 3.27769000 -0.39019700

Dye **3** (CAM-B3LYP/6-31G (d,p) optimised ground-state)

N -4.82891000 0.48426100 0.02119500

C -3.64950800 -0.18190100 -0.10666200

C -3.44799200 -1.51822500 -0.40938900

S -2.13598400 0.65510900 0.14074000

C -2.07944700 -1.85196300 -0.45350900

H -4.24662300 -2.21886100 -0.60712700

C -1.23322100 -0.80593800 -0.18830500

H -1.71523600 -2.84633200 -0.68746100

C 0.19837500 -0.81222500 -0.17482100

H 0.66502800 -1.78120500 -0.40020300

C 2.28438300 0.17240500 0.03256700

C 3.01513800 0.87051000 1.00301900

C 2.97221700 -0.49686000 -0.98893200

C 4.39812900 0.86042000 0.98619900

H 2.46843600 1.41713000 1.76272700

C 4.35843200 -0.50392200 -1.01491300

H 2.41326100 -0.97839500 -1.78387900

C 5.06290200 0.16124300 -0.01861100

H 4.97080300 1.40940800 1.72544800

H 4.90058700 -0.99544800 -1.81517400

N 0.89047800 0.23008700 0.09959400

S 6.84059000 0.13958800 -0.04217700

O 7.27199300 -0.15732500 -1.40795200

O 7.31879900 1.33134000 0.65778300

C 7.26943400 -1.27632700 0.96906400

H 6.88311000 -1.12838500 1.97710500

H 6.85621500 -2.17805700 0.51814800

H 8.35906800 -1.32517000 0.98444100

C -4.93502400 1.94083500 -0.05869400

H -4.94991600 2.39147300 0.94225300

H -4.08108500 2.35810400 -0.59957800

C -6.25783700 2.15583800 -0.79127700

H -6.70676300 3.12716800 -0.57476800

H -6.09459400 2.09161800 -1.87106700

C -7.11035300 0.97937800 -0.31422200

H -7.54182200 1.21157100 0.66776900

H -7.93133200 0.74173000 -0.99289000

C -6.11722400 -0.18546600 -0.17673300

H -6.09289100 -0.78018200 -1.10069200

C -6.47225100 -1.11886400 0.98245100

H -6.49956400 -0.53507400 1.91396800

H -5.70936100 -1.88940900 1.10542200

O -7.68800600 -1.79590800 0.73956200

H -8.41120200 -1.16577000 0.83450600

Dye **4** (CAM-B3LYP/6-31G (d,p) optimised ground-state)

C -2.70137100 -1.07814900 0.01998700

C -4.83419400 -1.59066600 0.06389600

C -4.95104900 -0.23562500 -0.04272400

S -3.39548800 0.51219800 -0.12531400

H -5.67523700 -2.27086500 0.10690300

S -6.47829300 0.64449100 -0.12623600

O -6.28677300 1.79815700 -0.99824200

O -7.51733900 -0.34614600 -0.38756800

C -6.71345600 1.25627900 1.53794100

H -5.88305100 1.91084600 1.80164800

H -7.64811900 1.81891500 1.52730200

H -6.78536800 0.41193700 2.22251300

C -1.25674300 -1.26781800 0.03604300

H -0.93525100 -2.30939700 0.12719900

N -0.48717900 -0.25660500 -0.04021700

C 0.90790400 -0.36748700 -0.05554900

C 1.63357500 0.80310700 0.17873900

C 1.62781700 -1.54100800 -0.30973600

C 3.01439000 0.80892300 0.19833600

H 1.07841900 1.71812000 0.35434600

C 3.00870300 -1.54759600 -0.30255300

H 1.10759300 -2.46476800 -0.54058000

C 3.74392800 -0.37278200 -0.03823400

H 3.53052400 1.74164600 0.38498000

H 3.52767700 -2.47346800 -0.51438800

C 5.87489000 -1.63879800 -0.10741800

C 5.94309000 0.72867100 0.41838900

C 7.33038100 -1.19525100 0.03467100

H 5.69108100 -2.15684900 -1.05455400

H 5.58390100 -2.32200200 0.70400500

C 7.23145100 0.05918000 0.90534700

H 5.44657100 1.25964700 1.24173900

H 7.95790400 -1.97414600 0.47176000

H 7.74653200 -0.94919600 -0.94653200

H 7.13196400 -0.22043700 1.95848400

H 8.08498000 0.73271900 0.81185500

N 5.11770700 -0.39296500 -0.02825100

C 6.20708700 1.72437000 -0.71777400

H 6.76819500 1.22631600 -1.51335900

H 5.25750400 2.05930600 -1.15523900

O 7.00403700 2.81133800 -0.29089100

H 6.47724200 3.35981200 0.30210800

N -3.55629600 -2.06243800 0.10161900

Dye **1** (CIS/6-31G (d,p) optimised excited state)

C -3.11579900 -1.83872800 0.33561500

C -4.30174600 -2.06932600 0.97736000

C -5.07013000 -0.88204600 0.82873500

C -4.30561400 -0.04008500 0.11194200

H -4.58530500 -2.97050400 1.47840600

H -6.05550800 -0.67656800 1.19003500

S -4.62949400 1.57248500 -0.46185300

O -4.33634400 1.65841600 -1.86253100

O -5.93888800 1.90432900 0.02736900

C -3.44354200 2.58105700 0.38344200

H -3.62740600 2.53139300 1.44714500

H -2.44942400 2.23276000 0.14071000

H -3.58206000 3.59212100 0.02272500

C -1.95057400 -2.64580900 0.18812400

H -2.00702600 -3.64754200 0.58708100

N -0.94412400 -2.31841900 -0.61306000

C 0.19712000 -1.69223500 -0.28233000

C 1.13802500 -1.43513900 -1.30103500

C 0.52857200 -1.26919200 1.01974300

C 2.32175800 -0.80348000 -1.04361000

H 0.89549100 -1.75030100 -2.29873200

C 1.71420200 -0.63350700 1.28637600

H -0.16817300 -1.45071200 1.81613300

C 2.65181000 -0.37777000 0.26424900

H 3.00111100 -0.63549600 -1.85464100

H 1.92109500 -0.33141100 2.29348500

C 4.11192700 0.82798100 1.84974700

C 4.77985200 0.69998000 -0.48041100

C 5.47598200 1.49370300 1.67908300

H 4.12386500 0.06253400 2.61794900

H 3.35044400 1.55772300 2.11888100

C 5.50210000 1.86824300 0.19700000

H 4.25235200 1.04451800 -1.36496300

H 5.59541900 2.34729700 2.33519500

H 6.26912700 0.79045300 1.91022500

H 4.94351300 2.78538400 0.03647100

H 6.49877300 2.00712700 -0.20024400

N 3.82731000 0.25456800 0.53754600

C 5.74053000 -0.42439600 -0.88145600

H 6.34011600 -0.71428300 -0.02754400

H 5.18412800 -1.30402600 -1.19311600

O 6.63392100 -0.00731800 -1.87624500

H 6.19864900 0.02833600 -2.71193900

O -3.11574100 -0.60131600 -0.19115100

Dye **2** (CIS/6-31G (d,p) optimised excited state)

C -2.65924800 1.77531900 -0.46199600

C -3.80266900 1.96007600 -1.20622900

C -4.77997500 0.96984000 -0.96493200

C -4.38540500 0.04865400 -0.05159700

S -2.79773800 0.38049800 0.56837500

H -3.93210500 2.77847600 -1.88787300

H -5.74448800 0.93944200 -1.43449600

S -5.33894600 -1.31351800 0.46800100

O -5.02219100 -1.59612600 1.84037900

O -6.69646700 -1.06911200 0.06512500

C -4.74817300 -2.68464900 -0.49088800

H -4.91724600 -2.48567200 -1.53942100

H -3.69748100 -2.83782300 -0.28879600

H -5.31709200 -3.55016100 -0.17629900

C -1.48444000 2.58081900 -0.48199100

H -1.53093300 3.49012700 -1.06499400

N -0.45173700 2.39674300 0.33293300

C 0.66329200 1.69217000 0.08761000

C 1.64338300 1.61484000 1.10145000

C 0.93538000 1.01730700 -1.12097900

C 2.80364100 0.91700800 0.92799800

H 1.44823900 2.12458000 2.02644700

C 2.09743400 0.31417300 -1.30228700

H 0.20983700 1.05730900 -1.91116500

C 3.07213800 0.23691300 -0.28434700

H 3.51351900 0.89285000 1.72977500

H 2.25764600 -0.18251300 -2.23805300

C 4.44499600 -1.28672100 -1.65927500

C 5.21221300 -0.72418400 0.57463200

C 5.80384500 -1.93953600 -1.41311200

H 4.43865900 -0.68280000 -2.55998400

H 3.66122200 -2.03560900 -1.75450800

C 5.88640600 -2.01876500 0.11150500

H 4.71899200 -0.87635300 1.52994500

H 5.88101400 -2.90733300 -1.89336600

H 6.59826700 -1.31485500 -1.80762000

H 5.32054700 -2.87333400 0.46970800

H 6.89662900 -2.10110600 0.48967900

N 4.22183500 -0.46564500 -0.47199400

C 6.20570900 0.43502700 0.70688900

H 6.76932800 0.53965300 -0.21183600

H 5.67636900 1.37080800 0.86385300

O 7.13755500 0.19920100 1.72528200

H 6.74299400 0.34348200 2.56942300

Dye **3** (CIS/6-31G (d,p) optimised excited state)

N 4.70454700 0.52293600 0.05145600

C 3.60252400 -0.22013900 0.24519600

C 3.53588800 -1.58025500 0.57740500

S 2.00188400 0.47717700 0.07244500

C 2.24259500 -2.05934400 0.68846600

H 4.40673700 -2.18264900 0.74491200

C 1.25715200 -1.08275400 0.44847500

H 1.99724900 -3.07343200 0.94036600

C -0.12975400 -1.19141500 0.46826300

H -0.54905200 -2.15510900 0.71112100

C -2.19380700 -0.12205200 0.16535100

C -2.82532300 1.12403800 -0.09697600

C -3.03390500 -1.25441300 0.37153000

C -4.18634100 1.23809800 -0.15381100

H -2.19205900 1.98003700 -0.23809000

C -4.39919200 -1.13160600 0.31105700

H -2.61336100 -2.21867100 0.58215000

C -4.98589100 0.10430900 0.04242500

H -4.64891700 2.19173700 -0.32780800

H -5.02743700 -1.98527900 0.48610900

N -0.85148100 -0.11621300 0.20181900

S -6.73439200 0.23473600 -0.06677100

O -7.31976300 -0.87239700 0.64309700

O -7.11396300 1.58429300 0.25883800

C -7.11024600 -0.00170900 -1.78735200

H -6.62351900 0.76752700 -2.37027700

H -6.78269600 -0.98494900 -2.09434300

H -8.18553800 0.08119200 -1.88457800

C 4.70503200 1.98477400 0.07267100

H 4.58469400 2.38591600 -0.93173900

H 3.89278600 2.35705500 0.68279900

C 6.07798700 2.31532200 0.64814200

H 6.41488900 3.30858800 0.37620400

H 6.04281800 2.25510500 1.73110500

C 6.96405800 1.20712500 0.07941500

H 7.27281600 1.47159100 -0.92971300

H 7.85911600 1.03653700 0.66408600

C 6.05939200 -0.03677700 0.04078600

H 6.21393100 -0.64132600 0.93139500

C 6.31662000 -0.91369100 -1.18749700

H 6.18034600 -0.32033500 -2.08937200

H 5.61137500 -1.73112700 -1.22975000

O 7.59227300 -1.48572200 -1.12835300

H 8.24538400 -0.86652000 -1.40821600

Dye **4** (CIS/6-31G (d,p) optimised excited state)

C -2.64906100 -1.74121100 0.15323800

C -4.60097800 -1.22169900 -0.68438900

C -4.28484800 0.00433500 -0.20401400

S -2.72021300 -0.04500900 0.54979500

H -5.51185000 -1.43651100 -1.20834200

S -5.27683300 1.42761300 -0.32480900

O -4.41707200 2.57029300 -0.45823400

O -6.29151800 1.15663200 -1.30471500

C -6.08401900 1.56461300 1.24832100

H -5.34054200 1.66361300 2.02674200

H -6.69650400 2.45602000 1.20495200

H -6.70117800 0.69242900 1.40920900

C -1.51943800 -2.54943400 0.47173200

H -1.62702200 -3.60436700 0.26927500

N -0.47896700 -2.10427400 1.15643200

C 0.67259200 -1.59592300 0.69384900

C 1.64870700 -1.17846900 1.62456400

C 0.98024200 -1.44580900 -0.67550300

C 2.83912500 -0.64488400 1.22432400

H 1.42704600 -1.28993000 2.66956500

C 2.17314500 -0.91394700 -1.08479000

H 0.25799600 -1.75622100 -1.40666700

C 3.14532200 -0.49192300 -0.15049100

H 3.54258900 -0.33858800 1.97162800

H 2.36012100 -0.81816600 -2.13521800

C 4.70576500 0.08003900 -1.97625500

C 5.43897800 0.37755700 0.32055000

C 6.10625700 0.69037800 -1.96737400

H 4.00476300 0.67296600 -2.55286900

H 4.71738500 -0.92421900 -2.39465400

C 6.65954900 0.28908800 -0.59963700

H 5.51701300 -0.34977500 1.12292300

H 6.70953300 0.32983500 -2.79147600

H 6.04655100 1.76995400 -2.05452300

H 7.01985800 -0.73469000 -0.63059400

H 7.46715900 0.92042300 -0.25405400

N 4.32457100 0.03667700 -0.56584800

C 5.26824400 1.77376600 0.92914200

H 5.28024200 2.51896100 0.14364900

H 4.30709200 1.85443100 1.42907800

O 6.32259800 2.08533400 1.79658200

H 6.20513000 1.64976500 2.62444300

N -3.69449000 -2.20019300 -0.48365800