Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

On the origin of very strong two-photon activity of squaraine dyes - A standard/damped response theory study

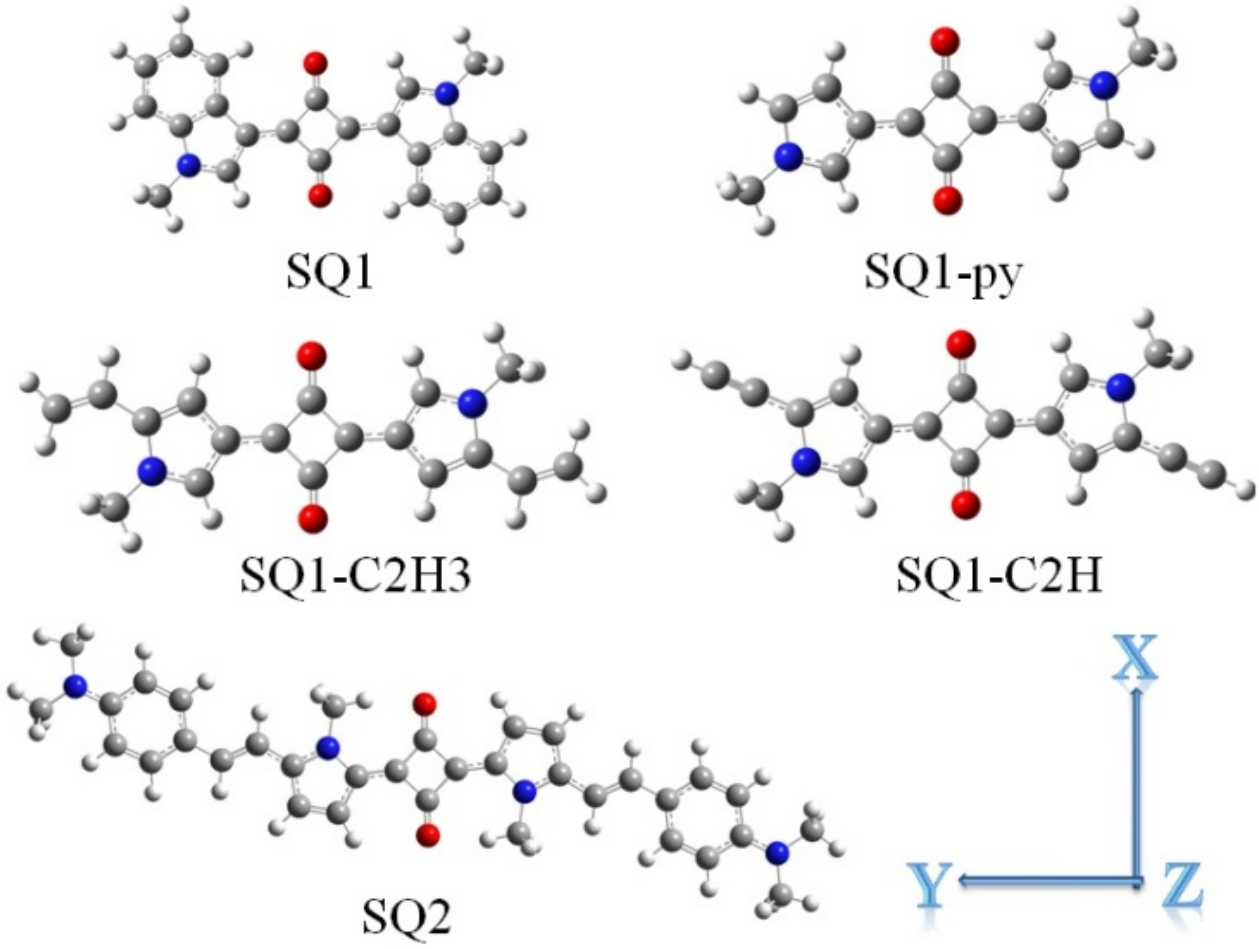
Md. Mehboob Alam,[a] Mausumi Chattopadhyaya[a], Swapan Chakrabarti*[a] and Antonio Rizzo*[b]

[a] Department of Chemistry, University of Calcutta, 92 A.P.C. Road, Kolkata-700 009, India. Fax: +91-33-23519755; Tel: +91-33-25950483; E-mail: swapanchem@yahoo.co.in

[b] Istituto per i Processi Chimico-Fisici del Consiglio Nazionale delle Ricerche (IPCF-CNR), Area della Ricerca, via G. Moruzzi 1, I-56124 Pisa, Italy. Fax: +39 050 315 2442; Tel: +39 050 315 2234; E-mail: rizzo@ ipcf.cnr.it

Contents

A) Cartesian Co-ordinates in gas phase



Gas phase optimized structures of the molecules considered in this work.

1) SQ1

O	-2.27476900	0.01295800	0.00000000
N	-1.04308300	4.47197000	0.00000000
C	-1.28232600	3.13928900	0.00000000
H	-2.28342200	2.73531800	0.00000000
C	-0.07487900	2.43103000	0.00000000
C	0.97907800	3.43116400	0.00000000
C	2.37628500	3.36856900	0.00000000
H	2.87290300	2.40483300	0.00000000
C	3.08944200	4.56171000	0.00000000
H	4.17304500	4.53462700	0.00000000
C	2.43615000	5.80515400	0.00000000
H	3.02347000	6.71620100	0.00000000
C	1.04808900	5.89061200	0.00000000

H	0.54810300	6.85192500	0.00000000
C	0.33891700	4.69097200	0.00000000
C	-0.00330000	1.03282800	0.00000000
C	-1.04808900	-0.00512900	0.00000000
C	-2.05036700	5.51866900	0.00000000
H	-1.95151100	6.14472800	0.89020800
H	-3.03812000	5.05991200	0.00000000
O	2.27476900	-0.01295800	0.00000000
N	1.04308300	-4.47197000	0.00000000
C	1.28232600	-3.13928900	0.00000000
H	2.28342200	-2.73531800	0.00000000
C	0.07487900	-2.43103000	0.00000000
C	-0.97907800	-3.43116400	0.00000000
C	-2.37628500	-3.36856900	0.00000000
H	-2.87290300	-2.40483300	0.00000000
C	-3.08944200	-4.56171000	0.00000000
H	-4.17304500	-4.53462700	0.00000000
C	-2.43615000	-5.80515400	0.00000000
H	-3.02347000	-6.71620100	0.00000000
C	-1.04808900	-5.89061200	0.00000000
H	-0.54810300	-6.85192500	0.00000000
C	-0.33891700	-4.69097200	0.00000000
C	0.00330000	-1.03282800	0.00000000
C	1.04808900	0.00512900	0.00000000
C	2.05036700	-5.51866900	0.00000000
H	1.95151100	-6.14472800	-0.89020800
H	3.03812000	-5.05991200	0.00000000
H	1.95151100	-6.14472800	0.89020800
H	-1.95151100	6.14472800	-0.89020800

2) SQ1-py

O	-2.24502900	0.37107300	0.00000000
N	-0.16261800	4.58205400	0.00000000
C	-0.70131000	3.34416800	0.00000000
H	-1.76550400	3.16961700	0.00000000
C	0.34505600	2.40852700	0.00000000
C	1.57154600	3.16319400	0.00000000
C	1.22797800	4.48309300	0.00000000
C	0.16261800	1.01776300	0.00000000
C	-1.03791800	0.16470000	0.00000000
C	-0.90375400	5.83437600	0.00000000
H	-0.66288500	6.42028100	0.88988600
H	-1.97038800	5.61579000	0.00000000
O	2.24502900	-0.37107300	0.00000000
N	0.16261800	-4.58205400	0.00000000
C	0.70131000	-3.34416800	0.00000000
H	1.76550400	-3.16961700	0.00000000
C	-0.34505600	-2.40852700	0.00000000
C	-1.57154600	-3.16319400	0.00000000
C	-1.22797800	-4.48309300	0.00000000
C	-0.16261800	-1.01776300	0.00000000

C	1.03791800	-0.16470000	0.00000000
C	0.90375400	-5.83437600	0.00000000
H	0.66288500	-6.42028100	-0.88988600
H	1.97038800	-5.61579000	0.00000000
H	0.66288500	-6.42028100	0.88988600
H	-0.66288500	6.42028100	-0.88988600
H	-1.83536700	-5.37387400	0.00000000
H	-2.56650800	-2.74847100	0.00000000
H	2.56650800	2.74847100	0.00000000
H	1.83536700	5.37387400	0.00000000

3) SQ1-C2H3

O	-2.27551500	0.01440000	0.00000000
N	-0.88299000	4.49906100	0.00000000
C	-1.21528500	3.19352000	0.00000000
H	-2.23808700	2.85145200	0.00000000
C	-0.03410000	2.43322000	0.00000000
C	1.05118400	3.36532900	0.00000000
C	0.52001600	4.63513800	0.00000000
C	0.00129700	1.03059500	0.00000000
C	-1.05118400	0.00026100	0.00000000
C	-1.84502000	5.59277200	0.00000000
H	-1.72527600	6.21403100	0.89001400
H	-2.84836300	5.17046900	0.00000000
O	2.27551500	-0.01440000	0.00000000
N	0.88299000	-4.49906100	0.00000000
C	1.21528500	-3.19352000	0.00000000
H	2.23808700	-2.85145200	0.00000000
C	0.03410000	-2.43322000	0.00000000
C	-1.05118400	-3.36532900	0.00000000
C	-0.52001600	-4.63513800	0.00000000
C	-0.00129700	-1.03059500	0.00000000
C	1.05118400	-0.00026100	0.00000000
C	1.84502000	-5.59277200	0.00000000
H	1.72527600	-6.21403100	-0.89001400
H	2.84836300	-5.17046900	0.00000000
H	1.72527600	-6.21403100	0.89001400
H	-1.72527600	6.21403100	-0.89001400
H	-2.09707600	-3.10435100	0.00000000
H	2.09707600	3.10435100	0.00000000
C	1.15190900	5.94318200	0.00000000
C	-1.15190900	-5.94318200	0.00000000
C	-2.46902500	-6.17698700	0.00000000
H	-3.19960400	-5.37622000	0.00000000
C	2.46902500	6.17698700	0.00000000
H	3.19960400	5.37622000	0.00000000
H	-2.85209900	-7.18947400	0.00000000
H	-0.48386600	-6.79950900	0.00000000
H	0.48386600	6.79950900	0.00000000
H	2.85209900	7.18947400	0.00000000

4) SQ1-C2H

O	2.27530300	-0.01802300	0.00000000
N	0.89297700	-4.49677300	0.00000000
C	1.21833000	-3.19202200	0.00000000
H	2.24007700	-2.84620800	0.00000000
C	0.03450800	-2.43289800	0.00000000
C	-1.05185500	-3.36473100	0.00000000
C	-0.50827500	-4.62932500	0.00000000
C	-0.00253500	-1.03005700	0.00000000
C	1.05185500	-0.00150200	0.00000000
C	1.83015300	-5.61202900	0.00000000
H	1.68265600	-6.22940300	0.88768600
H	2.84364200	-5.21489900	0.00000000
O	-2.27530300	0.01802300	0.00000000
N	-0.89297700	4.49677300	0.00000000
C	-1.21833000	3.19202200	0.00000000
H	-2.24007700	2.84620800	0.00000000
C	-0.03450800	2.43289800	0.00000000
C	1.05185500	3.36473100	0.00000000
C	0.50827500	4.62932500	0.00000000
C	0.00253500	1.03005700	0.00000000
C	-1.05185500	0.00150200	0.00000000
C	-1.83015300	5.61202900	0.00000000
H	-1.68265600	6.22940300	-0.88768600
H	-2.84364200	5.21489900	0.00000000
H	-1.68265600	6.22940300	0.88768600
H	1.68265600	-6.22940300	-0.88768600
H	2.10167000	3.12163100	0.00000000
H	-2.10167000	-3.12163100	0.00000000
C	-1.12929400	-5.89598400	0.00000000
C	1.12929400	5.89598400	0.00000000
C	1.65224900	6.98193800	0.00000000
H	2.12736900	7.93225000	0.00000000
C	-1.65224900	-6.98193800	0.00000000
H	-2.12736900	-7.93225000	0.00000000

5) SQ2

O	2.21969000	0.47972100	-0.02457600
N	0.09010000	3.50065700	-0.02305100
C	-0.67813600	2.33686900	-0.02284000
C	-2.03778300	2.73997600	-0.01911800
C	-2.07421900	4.11965700	-0.01673800
C	-1.01789100	7.04941000	-0.00555600
C	-0.24014000	5.93872300	-0.01913700
H	0.83783500	6.05197500	-0.03034000
C	-0.74007800	4.59290800	-0.01928500
C	-0.23080600	1.01561000	-0.02470200
C	1.01789100	0.22957500	-0.02483600
C	1.55006800	3.56242200	-0.02689800
H	1.90313400	4.08586500	-0.91898400

H	1.96019300	2.55332100	-0.02620900
O	-2.21969000	-0.47972100	-0.02457600
N	-0.09010000	-3.50065700	-0.02305100
C	0.67813600	-2.33686900	-0.02284000
C	2.03778300	-2.73997600	-0.01911800
C	2.07421900	-4.11965700	-0.01673800
C	1.01789100	-7.04941000	-0.00555600
C	0.24014000	-5.93872300	-0.01913700
H	-0.83783500	-6.05197500	-0.03034000
C	0.74007800	-4.59290800	-0.01928500
C	0.23080600	-1.01561000	-0.02470200
C	-1.01789100	-0.22957500	-0.02483600
C	-1.55006800	-3.56242200	-0.02689800
H	-1.90750700	-4.08883300	0.86163400
H	-1.96019300	-2.55332100	-0.02620900
H	-1.90313400	-4.08586500	-0.91898400
H	1.90750700	4.08883300	0.86163400
H	-2.86070900	2.04277200	-0.01850900
H	2.86070900	-2.04277200	-0.01850900
H	-2.95411900	4.74335600	-0.01458000
H	2.95411900	-4.74335600	-0.01458000
C	-0.58375900	8.43327100	-0.00607000
C	0.76477400	8.84014200	-0.02358600
C	-1.54482200	9.46034500	0.01758300
C	1.12838300	10.17275200	-0.02268900
H	1.55397300	8.09708400	-0.03522800
C	-1.20049600	10.80088100	0.02001700
H	-2.59686000	9.19366100	0.03879400
C	0.15432500	11.20199800	-0.00979300
H	2.18069900	10.42043300	-0.03114200
H	-1.98934400	11.53945900	0.04642000
C	0.58375900	-8.43327100	-0.00607000
C	1.54482200	-9.46034500	0.01758300
C	-0.76477400	-8.84014200	-0.02358600
C	1.20049600	-10.80088100	0.02001700
H	2.59686000	-9.19366100	0.03879400
C	-1.12838300	-10.17275200	-0.02268900
H	-1.55397300	-8.09708400	-0.03522800
C	-0.15432500	-11.20199800	-0.00979300
H	1.98934400	-11.53945900	0.04642000
H	-2.18069900	-10.42043300	-0.03114200
H	-2.09662000	6.91457100	0.00776100
H	2.09662000	-6.91457100	0.00776100
N	-0.51437500	-12.53308600	-0.03089400
N	0.51437500	12.53308600	-0.03089400
C	0.50169000	-13.56179900	0.11770600
H	1.01471100	-13.50908500	1.08798500
H	0.03255000	-14.54077400	0.03387700
H	1.25810400	-13.48866900	-0.67026500
C	-0.50169000	13.56179900	0.11770600
H	-1.01471100	13.50908500	1.08798500
H	-0.03255000	14.54077400	0.03387700

H	-1.25810400	13.48866900	-0.67026500
C	1.91385500	12.91166900	0.07693800
H	2.50560400	12.48138700	-0.73727700
H	1.99547100	13.99505100	0.00571000
H	2.36321800	12.59612200	1.02860300
C	-1.91385500	-12.91166900	0.07693800
H	-2.50560400	-12.48138700	-0.73727700
H	-1.99547100	-13.99505100	0.00571000
H	-2.36321800	-12.59612200	1.02860300

B) Cartesian Co-ordinates in CH_2Cl_2

1) SQ1

O	-2.27359600	0.01662300	0.00000000
N	-1.00683700	4.48707400	0.00000000
C	-1.25922300	3.16366600	0.00000000
H	-2.26838200	2.78270700	0.00000000
C	-0.05801300	2.43499900	0.00000000
C	1.00683700	3.42623900	0.00000000
C	2.40443800	3.35767100	0.00000000
H	2.89885000	2.39365200	0.00000000
C	3.12750800	4.54566100	0.00000000
H	4.21089900	4.51065400	0.00000000
C	2.48228900	5.79383100	0.00000000
H	3.07503400	6.70109800	0.00000000
C	1.09425300	5.88719100	0.00000000
H	0.59906500	6.85037800	0.00000000
C	0.37725200	4.69215100	0.00000000
C	-0.00007100	1.03693700	0.00000000
C	-1.04147000	-0.00178000	0.00000000
C	-2.00742100	5.54685600	0.00000000
H	-1.89667200	6.16905800	0.88994300
H	-2.99741000	5.09578600	0.00000000
O	2.27359600	-0.01662300	0.00000000
N	1.00683700	-4.48707400	0.00000000
C	1.25922300	-3.16366600	0.00000000
H	2.26838200	-2.78270700	0.00000000
C	0.05801300	-2.43499900	0.00000000
C	-1.00683700	-3.42623900	0.00000000
C	-2.40443800	-3.35767100	0.00000000
H	-2.89885000	-2.39365200	0.00000000
C	-3.12750800	-4.54566100	0.00000000
H	-4.21089900	-4.51065400	0.00000000
C	-2.48228900	-5.79383100	0.00000000
H	-3.07503400	-6.70109800	0.00000000
C	-1.09425300	-5.88719100	0.00000000
H	-0.59906500	-6.85037800	0.00000000
C	-0.37725200	-4.69215100	0.00000000
C	0.00007100	-1.03693700	0.00000000
C	1.04147000	0.00178000	0.00000000

C	2.00742100	-5.54685600	0.00000000
H	1.89667200	-6.16905800	-0.88994300
H	2.99741000	-5.09578600	0.00000000
H	1.89667200	-6.16905800	0.88994300
H	-1.89667200	6.16905800	-0.88994300

2) SQ1-py

O	-2.24959700	0.33339700	0.00000000
N	-0.14893400	4.59281000	0.00000000
C	-0.70194500	3.36689300	0.00000000
H	-1.76943400	3.21874500	0.00000000
C	0.33414400	2.41259800	0.00000000
C	1.56982500	3.15637800	0.00000000
C	1.24008300	4.47930100	0.00000000
C	0.14893400	1.02414600	0.00000000
C	-1.03209600	0.14929700	0.00000000
C	-0.87396800	5.86008700	0.00000000
H	-0.61763000	6.43690400	0.88985400
H	-1.94276200	5.65680800	0.00000000
O	2.24959700	-0.33339700	0.00000000
N	0.14893400	-4.59281000	0.00000000
C	0.70194500	-3.36689300	0.00000000
H	1.76943400	-3.21874500	0.00000000
C	-0.33414400	-2.41259800	0.00000000
C	-1.56982500	-3.15637800	0.00000000
C	-1.24008300	-4.47930100	0.00000000
C	-0.14893400	-1.02414600	0.00000000
C	1.03209600	-0.14929700	0.00000000
C	0.87396800	-5.86008700	0.00000000
H	0.61763000	-6.43690400	-0.88985400
H	1.94276200	-5.65680800	0.00000000
H	0.61763000	-6.43690400	0.88985400
H	-0.61763000	6.43690400	-0.88985400
H	-1.85589200	-5.36398000	0.00000000
H	-2.56405900	-2.74023500	0.00000000
H	2.56405900	2.74023500	0.00000000
H	1.85589200	5.36398000	0.00000000

3) SQ1-C2H3

O	-2.27429300	0.01378200	0.00000000
N	-0.81225000	4.52814400	0.00000000
C	-1.16943800	3.23265500	0.00000000
H	-2.20407400	2.92965200	0.00000000
C	-0.00929400	2.43591300	0.00000000
C	1.09476600	3.34733600	0.00000000
C	0.59496300	4.63073300	0.00000000
C	0.00324400	1.03442000	0.00000000
C	-1.04361200	0.00261400	0.00000000
C	-1.77600800	5.62565500	0.00000000
H	-1.65202700	6.24088800	0.89141700

H	-2.77741100	5.20041000	0.00000000
O	2.27429300	-0.01378200	0.00000000
N	0.81225000	-4.52814400	0.00000000
C	1.16943800	-3.23265500	0.00000000
H	2.20407400	-2.92965200	0.00000000
C	0.00929400	-2.43591300	0.00000000
C	-1.09476600	-3.34733600	0.00000000
C	-0.59496300	-4.63073300	0.00000000
C	-0.00324400	-1.03442000	0.00000000
C	1.04361200	-0.00261400	0.00000000
C	1.77600800	-5.62565500	0.00000000
H	1.65202700	-6.24088800	-0.89141700
H	2.77741100	-5.20041000	0.00000000
H	1.65202700	-6.24088800	0.89141700
H	-1.65202700	6.24088800	-0.89141700
H	-2.14017000	-3.08320900	0.00000000
H	2.14017000	3.08320900	0.00000000
C	-1.39241500	-5.84303200	0.00000000
C	-1.04361200	-7.13678400	0.00000000
H	-2.45520900	-5.61481600	0.00000000
H	-1.81585200	-7.89627400	0.00000000
H	-0.02417700	-7.49417700	0.00000000
C	1.39241500	5.84303200	0.00000000
C	1.04361200	7.13678400	0.00000000
H	2.45520900	5.61481600	0.00000000
H	1.81585200	7.89627400	0.00000000
H	0.02417700	7.49417700	0.00000000

4) SQ1-C2H

O	2.27424200	0.00229900	0.00000000
N	0.84630800	-4.51702600	0.00000000
C	1.19714100	-3.22367700	0.00000000
H	2.22902400	-2.91162100	0.00000000
C	0.02788300	-2.43525900	0.00000000
C	-1.07692000	-3.34751000	0.00000000
C	-0.55780800	-4.62115000	0.00000000
C	0.00404700	-1.03368300	0.00000000
C	1.04451000	0.00477500	0.00000000
C	1.76398100	-5.65335100	0.00000000
H	1.60200500	-6.26351500	0.88900900
H	2.78379800	-5.27554500	0.00000000
O	-2.27424200	-0.00229900	0.00000000
N	-0.84630800	4.51702600	0.00000000
C	-1.19714100	3.22367700	0.00000000
H	-2.22902400	2.91162100	0.00000000
C	-0.02788300	2.43525900	0.00000000
C	1.07692000	3.34751000	0.00000000
C	0.55780800	4.62115000	0.00000000
C	-0.00404700	1.03368300	0.00000000
C	-1.04451000	-0.00477500	0.00000000
C	-1.76398100	5.65335100	0.00000000

H	-1.60200500	6.26351500	-0.88900900
H	-2.78379800	5.27554500	0.00000000
H	-1.60200500	6.26351500	0.88900900
H	1.60200500	-6.26351500	-0.88900900
H	2.12336200	3.09102600	0.00000000
H	-2.12336200	-3.09102600	0.00000000
C	-1.20853200	-5.87312800	0.00000000
C	1.20853200	5.87312800	0.00000000
C	1.76398100	6.94310600	0.00000000
H	2.26045500	7.88414700	0.00000000
C	-1.76398100	-6.94310600	0.00000000
H	-2.26045500	-7.88414700	0.00000000

5) SQ2

O	2.22414600	0.45577600	-0.02558300
N	0.10918500	3.50930400	-0.02025700
C	-0.66120400	2.34559800	-0.02230700
C	-2.02082100	2.75670000	-0.01969800
C	-2.05429200	4.13540500	-0.01577400
C	-0.99673700	7.05772700	-0.00092300
C	-0.21278700	5.94744200	-0.01211200
H	0.86484700	6.06105200	-0.01732800
C	-0.71606500	4.60466300	-0.01596400
C	-0.22189900	1.02116400	-0.02482300
C	1.01400200	0.21879200	-0.02532700
C	1.56953000	3.57370100	-0.02180700
H	1.92293600	4.10066100	-0.91072600
H	1.97526800	2.56470900	-0.02442900
O	-2.22414600	-0.45577600	-0.02558300
N	-0.10918500	-3.50930400	-0.02025700
C	0.66120400	-2.34559800	-0.02230700
C	2.02082100	-2.75670000	-0.01969800
C	2.05429200	-4.13540500	-0.01577400
C	0.99673700	-7.05772700	-0.00092300
C	0.21278700	-5.94744200	-0.01211200
H	-0.86484700	-6.06105200	-0.01732800
C	0.71606500	-4.60466300	-0.01596400
C	0.22189900	-1.02116400	-0.02482300
C	-1.01400200	-0.21879200	-0.02532700
C	-1.56953000	-3.57370100	-0.02180700
H	-1.92498300	-4.09681900	0.86854700
H	-1.97526800	-2.56470900	-0.02442900
H	-1.92293600	-4.10066100	-0.91072600
H	1.92498300	4.09681900	0.86854700
H	-2.85142400	2.06928300	-0.02068100
H	2.85142400	-2.06928300	-0.02068100
H	-2.93270800	4.76117000	-0.01341200
H	2.93270800	-4.76117000	-0.01341200
C	-0.56826700	8.43966200	0.00441900
C	0.78181800	8.85119000	-0.00854400
C	-1.53629100	9.46409400	0.02491200

C	1.14040900	10.18383600	-0.00116300
H	1.57396500	8.11131600	-0.02484600
C	-1.19802300	10.80484000	0.03321300
H	-2.58712500	9.19221200	0.03510800
C	0.15956300	11.21224900	0.02036400
H	2.19111300	10.43765600	-0.01127100
H	-1.98846700	11.54189500	0.04931600
C	0.56826700	-8.43966200	0.00441900
C	1.53629100	-9.46409400	0.02491200
C	-0.78181800	-8.85119000	-0.00854400
C	1.19802300	-10.80484000	0.03321300
H	2.58712500	-9.19221200	0.03510800
C	-1.14040900	-10.18383600	-0.00116300
H	-1.57396500	-8.11131600	-0.02484600
C	-0.15956300	-11.21224900	0.02036400
H	1.98846700	-11.54189500	0.04931600
H	-2.19111300	-10.43765600	-0.01127100
H	-2.07522300	6.91995200	0.00627600
H	2.07522300	-6.91995200	0.00627600
N	-0.51281300	-12.53514200	0.02824000
N	0.51281300	12.53514200	0.02824000
C	0.51281300	-13.56916800	0.06460500
H	1.13798400	-13.48823900	0.96104100
H	0.03307100	-14.54518200	0.07537900
H	1.16734000	-13.52222300	-0.81311600
C	-0.51281300	13.56916800	0.06460500
H	-1.13798400	13.48823900	0.96104100
H	-0.03307100	14.54518200	0.07537900
H	-1.16734000	13.52222300	-0.81311600
C	1.91701100	12.92466500	0.01414400
H	2.43141600	12.55219400	-0.87864600
H	1.98365800	14.01024200	0.01217800
H	2.45106300	12.55308000	0.89610400
C	-1.91701100	-12.92466500	0.01414400
H	-2.43141600	-12.55219400	-0.87864600
H	-1.98365800	-14.01024200	0.01217800
H	-2.45106300	-12.55308000	0.89610400

C. Standard TPA results.

The two-photon absorption strength $\delta^{TP}(\omega_1,\omega_2)$ in the degenerate case $\left(\omega_1=\omega_2=\frac{\omega_{0f}}{2}\right)$ in the standard TPA calculations is given by the expression[1]

$$\delta^{TP}(\omega)\approx 8.35158\times10^{-4}\times\omega^2\sum_f g(2\omega,\omega_{0f},\Gamma)\cdot\delta^{TP}_{0f}(\omega_{0f}), \tag{1}$$

where, $\delta^{TP}_{0f}(\omega_{0f})$ is the orientation averaged two-photon probability (the quantity reported in the following tables)

$$\delta^{TP}_{0f}(\omega_{0f})=\sum_{\alpha\beta}\left(F\times S^{0f}_{\alpha\alpha}S^{*,0f}_{\beta\beta}+G\times S^{0f}_{\alpha\beta}S^{*,0f}_{\alpha\beta}+H\times S^{0f}_{\alpha\beta}S^{*,0f}_{\beta\alpha}\right), \tag{2}$$

$g(2\omega,\omega_{0f},\Gamma)$ is the line shape function, a Lorentzian centered in ω_{0f}, normalized to unity and with half width at half maximum (HWHM) Γ

$$g(\omega,\omega_{0f},\Gamma)=\frac{1}{\pi}\frac{\Gamma}{(\omega-\omega_{0f})^2+\Gamma^2}=\frac{1}{\pi}A(\omega,\omega_{0f},\Gamma) \tag{3}$$

and $S^{0f}_{\alpha\beta}$ is the two-photon transition matrix element which is a function of $\frac{\omega_{0f}}{2}$

$$S_{\alpha\beta}=\sum_i\frac{\mu^{0i}_\alpha\mu^{if}_\beta+\mu^{0i}_\beta\mu^{if}_\alpha}{\mathsf{h}\left(\omega_{0i}-\frac{\omega_{0f}}{2}\right)}=\sum_i\frac{\mu^{0i}_\alpha\mu^{if}_\beta+\mu^{0i}_\beta\mu^{if}_\alpha}{\Delta E^{(f)}_i}$$

F, *G* and *H* are scalars whose values are dependent on the polarization of the light. They are all equal to 2 for linearly polarized beam while, for a circularly polarized beam their value is -2, 3, 3, respectively. Equation (1) implies that the circular frequency ω, the line shape $g(2\omega,\omega_{0f},\Gamma)$ and the two-photon probability $\delta^{TP}_{0f}(\omega_{0f})$ are given in atomic units (1 a.u. of $\delta^{TP}_{0f}(\omega_{0f})$=

$e^4 \mathsf{h}^{-4} a_0^8 m_e^2 = 2.71847 \times 10^{-82}$ *SI* units), yielding TPA cross sections in Göppert-Mayer units (GM, $10^{-50} cm^4 \cdot \sec \cdot molec^{-1} \cdot photon^{-1}$).

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
4.108	301.8	7.260E+03	3.886	319.0	0.045897	**2.593**	478.1	-0.074513	4.038507	2.722	455.4	7.310E-02
4.300	288.3	9.890E+04	4.834	256.5	-0.336404	4.132	300.1	0.269105	0.250844	4.865	254.9	1.110E+01
4.529	273.8	4.540E+03	5.305	233.7	0.146029	4.319	287.0	-0.173312	1.215877	4.945	250.7	2.430E+04
5.110	242.6	**9.500E+07**	5.464	226.9	-0.009273	4.635	267.5	-0.940232	0.209224	5.433	228.2	3.060E+03
5.712	217.1	6.790E+05	5.537	223.9	-0.027168	5.085	243.8	-0.240717	1.091142	5.744	215.8	5.250E+01
5.820	213.0	6.860E+05	5.829	212.7	-0.098071	5.434	228.2	0.598826	0.516085	5.893	210.4	1.000E+01
5.930	209.1	1.530E+05	5.999	206.7	0.021229	5.563	222.9	0.510053	-1.405952	5.953	208.3	6.340E-01
5.962	208.0	4.520E+03	6.050	204.9	0.076595	5.796	213.9	-1.056345	0.394526	5.979	207.4	3.770E+02
6.040	205.3	3.290E+04	6.171	200.9	-0.092201	5.921	209.4	1.083460	-0.443363	6.057	204.7	1.890E+01
6.068	204.3	2.560E+05	6.260	198.1	-0.259979	6.006	206.4	-0.936984	1.014223	6.428	192.9	4.890E+01

Table C1. SQ1. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}), see Eq. (1) above, for the lowest ten electronic states of each symmetry. Standard TPA gas phase calculation. Here and in the following tables the states involved in the collection of data included in Table 2 of the manuscript are highlighted in bold.

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
4.236	292.7	2.230E+03	3.729	332.5	0.049905	**2.796**	**443.4**	-0.427107	-3.363390	2.602	476.6	1.050E-01
5.357	231.5	1.290E+06	5.056	245.2	0.187040	4.231	293.0	-0.427534	-0.495595	4.860	255.1	2.270E+00
5.419	228.8	**2.210E+07**	5.525	224.4	0.088817	4.923	251.9	0.184298	1.986122	5.185	239.1	2.080E+03
5.953	208.3	1.180E+04	5.620	220.6	-0.002690	5.475	226.5	0.172532	-0.525033	5.711	217.1	9.990E+03
6.142	201.8	8.680E+05	5.816	213.2	-0.248059	5.767	215.0	-1.361738	-0.014182	6.061	204.6	3.450E+02
6.291	197.1	3.690E+05	6.161	201.3	-0.392850	6.278	197.5	0.024456	0.246703	6.164	201.2	1.200E+02
6.412	193.3	1.940E+04	6.235	198.8	0.050542	6.415	193.3	-0.152897	-1.120721	6.306	196.6	8.700E+01
6.530	189.9	1.940E+01	6.397	193.8	-0.018110	6.500	190.8	0.407871	-1.250560	6.380	194.3	7.190E+01
6.556	189.1	6.030E+03	6.570	188.7	0.110315	6.526	190.0	0.384476	-0.626353	6.567	188.8	1.980E+01
6.728	184.3	3.500E+04	6.752	183.6	0.200136	6.886	180.0	0.156135	0.314217	6.815	181.9	6.550E+01

Table C2. SQ1-py. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA gas phase calculation.

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
3.763	329.5	2.170E+04	3.702	334.9	0.047872	**2.707**	457.9	-0.275942	3.985969	2.574	481.7	9.010E-02
4.728	262.3	1.760E+05	5.055	245.3	0.404464	3.796	326.6	0.248103	-0.524592	4.857	255.3	5.310E+00
5.325	232.8	1.170E+08	5.477	226.4	-0.172690	4.715	263.0	0.786246	1.809017	5.172	239.7	2.520E+04
5.515	224.8	**2.600E+08**	5.615	220.8	-0.015488	4.917	252.1	0.075635	-2.294135	5.592	221.7	2.340E+02
5.729	216.4	1.030E+07	5.651	219.4	0.097457	5.393	229.9	-0.212148	-2.384974	5.838	212.4	3.720E+02
6.000	206.6	1.870E+03	5.908	209.8	-0.027612	5.727	216.5	-1.211704	0.144926	5.915	209.6	4.230E+01
6.299	196.8	4.360E+05	5.968	207.8	0.002292	5.984	207.2	0.364527	-0.058846	5.936	208.9	9.270E+01
6.379	194.4	6.610E+03	6.031	205.6	-0.050045	6.378	194.4	0.033037	-0.269795	6.083	203.8	7.980E+01
6.414	193.3	1.460E+05	6.253	198.3	0.145324	6.585	188.3	0.105540	0.415170	6.310	196.5	5.080E+01
6.651	186.4	2.260E+01	6.357	195.0	0.015524	6.632	186.9	0.116144	0.093290	6.387	194.1	2.590E+01

Table C3. SQ1-C2H3. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA gas phase calculation.

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
3.916	316.6	1.840E+04	3.682	336.8	-0.047508	**2.712**	457.1	0.179852	-3.886471	2.558	484.7	8.340E-02
4.925	251.7	2.600E+05	5.170	239.8	-0.350080	3.930	315.5	-0.181914	0.715530	4.863	254.9	5.760E+00
5.368	231.0	**1.260E+09**	5.437	228.0	-0.032728	4.897	253.2	-0.683721	0.689790	5.331	232.6	1.090E+05
5.669	218.7	7.270E+06	5.622	220.6	-0.007688	4.948	250.6	-0.683361	-2.445937	5.445	227.7	1.080E+05
5.831	212.6	5.150E+06	5.664	218.9	0.140341	5.601	221.4	-0.526972	-1.979998	5.838	212.4	1.380E+02
6.052	204.9	5.170E+02	5.901	210.1	0.043029	5.771	214.8	-1.251063	0.378809	5.907	209.9	4.180E+00
6.353	195.2	1.010E+05	5.930	209.1	0.109208	6.204	199.8	-0.067784	0.097102	6.079	204.0	3.390E+01
6.382	194.3	2.270E+05	6.066	204.4	0.013753	6.448	192.3	0.043673	-0.339502	6.146	201.7	1.050E+02
6.611	187.6	1.170E+04	6.168	201.0	-0.008938	6.700	185.0	0.068843	0.319638	6.154	201.5	1.360E+02
6.745	183.8	3.620E+01	6.349	195.3	0.094621	6.745	183.8	-0.071377	-1.040932	6.325	196.0	1.330E+01

Table C4. SQ1-C2H. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA gas phase calculation.

A				*B*				
ΔE_{0f}	λ_{0f}	μ_z^{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	δ_{0f}^{TP}
eV	nm	a.u.	a.u.	eV	nm	a.u.	a.u.	a.u.
3.295	376.3	0.047107	2.800E+06	**2.002**	619.3	0.998949	7.693821	5.480E-02
3.435	361.0	0.026797	1.490E+06	2.928	423.4	-0.000971	-0.009828	1.840E+00
4.036	307.2	-0.039340	1.790E+05	3.570	347.3	-1.133537	-2.413757	3.550E+01
4.261	291.0	-0.011764	3.560E+05	3.837	323.1	0.696378	0.429895	6.050E+02
4.269	290.4	-0.065244	**1.010E+07**	4.257	291.2	0.787814	-0.295280	1.530E+01
4.301	288.3	0.361691		4.312	287.5	-0.189855	-0.862770	
4.477	276.9	0.255931		4.331	286.3	-0.083550	-0.530557	
4.631	267.7	-0.019204		4.548	272.6	-0.363326	0.190794	
4.642	267.1	0.069294		4.597	269.7	-0.033736	0.109378	
4.853	255.5	0.124533		4.619	268.4	0.008231	0.035376	

Table C5. SQ2. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f} , $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Two-photon transition strengths were computed only for the lowest five excited states. Standard TPA gas phase calculation.

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
4.051	306.1	1.560E+04	4.237	292.6	0.048562	2.488	498.4	-0.136267	-4.652937	3.057	405.6	1.070E-01
4.224	293.5	4.320E+05	4.888	253.7	-0.258685	4.071	304.6	-0.441390	-0.131524	4.849	255.7	7.390E+01
4.574	271.1	9.410E+04	5.595	221.6	0.304046	4.415	280.9	-0.158980	-0.847897	5.292	234.3	1.070E+04
5.120	242.2	**1.290E+07**	5.662	219.0	-0.002141	4.658	266.2	0.978850	0.151905	5.510	225.0	3.170E+03
5.771	214.8	2.200E+05	5.807	213.5	0.151770	5.075	244.3	-1.098908	-0.793731	5.985	207.2	4.220E+01
5.819	213.1	3.390E+05	5.980	207.3	0.093240	5.116	242.3	-0.585845	1.049407	6.134	202.1	3.060E+00
5.917	209.6	2.480E+05	6.200	200.0	-0.187885	5.578	222.3	-0.323249	-1.416771	6.189	200.3	1.710E+02
5.976	207.5	5.500E+04	6.213	199.6	-0.004277	5.854	211.8	-2.025153	-1.253592	6.232	198.9	1.390E+00
6.144	201.8	6.870E+04	6.342	195.5	0.061047	5.999	206.7	-0.600685	-0.113163	6.327	195.9	2.500E+00
6.198	200.0	4.120E+02	6.560	189.0	-0.245162	6.244	198.6	-0.183146	-0.794110	6.624	187.2	7.240E+01

Table D1. SQ1. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA solvent calculation (CH_2Cl_2).

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
4.053	305.9	7.650E+03	4.159	298.1	-0.051777	**2.748**	451.2	0.450649	-4.027084	3.023	410.1	1.400E-01
5.270	235.3	1.940E+04	5.147	240.9	0.237600	4.089	303.2	0.453091	-0.003321	4.869	254.6	3.170E+00
5.625	220.4	**2.910E+08**	5.734	216.2	0.000447	4.996	248.2	0.115823	-1.796087	5.637	220.0	2.760E+04
6.204	199.9	9.560E+02	5.947	208.5	-0.096124	5.283	234.7	1.103984	-0.336843	5.866	211.4	9.340E+02
6.253	198.3	6.380E+04	6.214	199.5	-0.345550	5.842	212.2	1.217793	0.632883	6.384	194.2	1.980E+02
6.379	194.4	5.390E+05	6.404	193.6	0.440212	6.512	190.4	-0.142650	-1.569413	6.468	191.7	1.820E+02
6.642	186.7	2.090E+04	6.451	192.2	0.069000	6.687	185.4	0.060962	0.663121	6.605	187.7	8.960E+01
6.838	181.3	5.210E+03	6.832	181.5	-0.011444	6.754	183.6	0.424662	0.856658	6.719	184.5	2.330E+01
6.958	178.2	1.140E+03	6.961	178.1	-0.304137	7.076	175.2	-0.434469	-0.132329	7.052	175.8	1.740E+02
7.063	175.5	1.900E+03	7.019	176.6	0.147865	7.139	173.7	-0.094580	-0.004589	7.118	174.2	2.000E+01

Table D2. SQ1-py. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA solvent calculation (CH_2Cl_2).

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
3.568	347.5	6.850E+04	4.116	301.2	-0.048470	**2.621**	473.1	-0.274919	4.678868	2.981	415.9	1.320E-01
4.824	257.0	1.500E+06	5.105	242.8	-0.371830	3.687	336.3	0.269407	0.177838	4.868	254.7	1.180E+01
5.249	236.2	**1.150E+13**	5.641	219.8	-0.293301	4.796	258.5	-0.782389	-2.402611	5.422	228.7	4.780E+04
5.560	223.0	4.220E+05	5.730	216.4	-0.001262	4.974	249.3	0.012248	-2.030466	5.741	216.0	1.390E+03
5.829	212.7	2.410E+06	5.937	208.8	-0.143583	5.290	234.4	-1.427946	-0.784815	6.094	203.5	3.800E+02
6.228	199.1	9.030E+02	6.181	200.6	0.251886	5.544	223.7	-0.435602	1.963724	6.117	202.7	1.220E+02
6.337	195.7	3.790E+05	6.296	196.9	-0.008971	6.226	199.1	-0.471042	0.133332	6.239	198.7	3.190E+01
6.436	192.6	7.290E+03	6.444	192.4	-0.203339	6.366	194.7	0.352858	0.203314	6.544	189.5	2.160E+01
6.493	190.9	1.300E+04	6.514	190.3	-0.024488	6.673	185.8	0.433574	0.147488	6.550	189.3	2.460E+01
6.545	189.4	1.410E+04	6.616	187.4	0.037774	6.708	184.8	0.042447	0.514789	6.700	185.1	5.830E+01

Table D3. SQ1-C2H3. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA solvent calculation (CH_2Cl_2).

A_g			A_u			B_u				B_g		
ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_z^{0f}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	ΔE_{0f}	λ_{0f}	δ_{0f}^{TP}
eV	nm	a.u.	eV	nm	a.u.	eV	nm	a.u.	a.u.	eV	nm	a.u.
3.766	329.2	5.660E+04	4.069	304.7	-0.047877	**2.641**	469.4	0.131190	-4.562366	2.941	421.5	1.250E-01
5.009	247.5	3.400E+06	5.233	236.9	0.339762	3.852	321.9	-0.210020	0.152940	4.876	254.3	1.100E+01
5.275	235.1	**8.920E+11**	5.545	223.6	0.030240	4.956	250.2	0.399866	-1.026784	5.542	223.7	6.950E+01
5.737	216.1	4.180E+05	5.727	216.5	-0.001933	5.001	247.9	0.822409	2.389894	5.629	220.3	1.020E+04
5.909	209.8	2.250E+06	5.778	214.6	-0.022036	5.379	230.5	-1.697109	-0.708532	5.778	214.6	9.150E+00
6.281	197.4	3.950E+02	5.885	210.7	-0.252627	5.729	216.4	0.219104	-1.773790	5.935	208.9	4.810E+02
6.432	192.8	2.440E+05	6.225	199.2	-0.073504	6.434	192.7	0.268630	0.133945	6.306	196.6	1.430E+02
6.697	185.1	4.010E+03	6.404	193.6	-0.181433	6.629	187.0	0.155849	-0.700348	6.412	193.4	2.060E+02
6.733	184.1	7.570E+02	6.427	192.9	-0.007929	6.771	183.1	0.055885	0.681899	6.435	192.7	1.670E+01
6.762	183.4	4.420E+03	6.518	190.2	-0.053876	6.910	179.4	0.523978	-0.014325	6.609	187.6	3.670E+01

Table D4. SQ1-C2H. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Standard TPA solvent calculation (CH_2Cl_2).

A				*B*				
ΔE_{0f}	λ_{0f}	μ_z^{0f}	δ_{0f}^{TP}	ΔE_{0f}	λ_{0f}	μ_x^{0f}	μ_y^{0f}	δ_{0f}^{TP}
eV	nm	a.u.	a.u.	eV	nm	a.u.	a.u.	a.u.
2.963	418.4	-0.028249	1.360E+07	**1.792**	691.9	-0.982347	-8.652518	1.890E+00
3.425	362.0	0.014485	**4.670E+07**	3.258	380.5	-0.000999	-0.003194	4.650E+01
4.241	292.4	0.001670	2.640E+04	3.404	364.2	1.234301	1.936956	4.550E+02
4.255	291.4	0.012931	6.060E+05	3.793	326.9	-0.949508	-1.138730	1.020E+02
4.301	288.3	0.307448	2.560E+01	4.241	292.3	-0.847292	-0.010902	4.490E-01
4.372	283.6	0.015916		4.298	288.5	0.137936	-1.102731	
4.483	276.6	-0.008856		4.433	279.7	-0.005746	0.004432	
4.528	273.8	0.447384		4.548	272.6	-0.471535	-0.616531	
4.722	262.6	-0.008115		4.595	269.8	0.284915	1.025026	
4.846	255.9	0.163919		4.670	265.5	-0.927875	0.515642	

Table D5. SQ2. Excitation energies (ΔE_{0f}), excitation wavelengths (λ_{0f}), transition moments (μ_α^{0f}, $\alpha = x, y, z$) and two-photon transition strengths (δ_{0f}^{TP}) for the lowest ten electronic states of each symmetry. Two-photon transition strengths were computed only for the lowest five excited states. Standard TPA solvent calculation (CH_2Cl_2).

	Gas phase								CH_2Cl_2 solvent							
	A_g					B_g			A_g					B_g		
	ΔE_{0f}	S_{xx}^{0f}	S_{yy}^{0f}	S_{zz}^{0f}	S_{xy}^{0f}	ΔE_{0f}	S_{xz}^{0f}	S_{yz}^{0f}	ΔE_{0f}	S_{xx}^{0f}	S_{yy}^{0f}	S_{zz}^{0f}	S_{xy}^{0f}	ΔE_{0f}	S_{xz}^{0f}	S_{yz}^{0f}
	eV	a.u.	a.u.	a.u.	a.u.	eV	a.u.	a.u.	eV	a.u.	a.u.	a.u.	a.u.	eV	a.u.	a.u.
SQ1	4.11	20.0	-193.5	-0.8	-26.3	2.72	-0.5	-0.1	4.05	28.9	-282.9	-0.4	42.9	3.06	0.6	-0.2
	4.30	12.5	674.7	-2.5	-161.3	4.86	-0.2	6.4	4.22	20.9	1429.2	-2.2	270.2	4.85	-0.6	-16.6
	4.53	1.6	-121.8	6.4	79.7	4.95	-17.4	-301.6	4.57	10.0	635.0	-5.5	221.7	5.29	29.6	198.0
	5.11	**24.0**	**-21798.3**	**-2.1**	**-427.8**	5.43	-14.5	106.1	**5.12**	**35.1**	**7974.7**	**1.6**	**758.7**	5.51	18.7	107.4
	5.71	23.7	-1849.7	19.3	-143.0	5.74	7.1	-12.1	5.77	-8.5	1021.3	2.5	212.1	5.99	-10.6	-6.7
	5.82	-2.3	1845.5	6.2	-125.4	5.89	4.3	-4.4	5.82	22.4	-1309.5	10.4	82.3	6.13	1.4	-3.1
	5.93	32.9	854.3	23.5	36.6	5.95	0.3	1.5	5.92	19.0	1105.7	3.3	-15.3	6.19	-17.9	18.0
	5.96	2.8	139.6	18.6	-23.6	5.98	6.3	37.1	5.98	-12.4	-509.7	-24.5	-34.4	6.23	1.7	-1.6
	6.04	-7.2	407.6	-3.0	26.2	6.06	-7.4	-4.0	6.14	58.1	-607.7	16.7	9.6	6.33	-1.2	2.8
	6.07	36.2	-1144.1	5.9	18.7	6.43	-7.0	-11.6	6.20	-10.4	37.3	-2.8	25.4	6.62	-12.6	10.6
SQ1-py	4.24	-2.3	97.7	0.1	36.4	2.6	-0.6	0.0	4.05	-4.7	189.2	0.0	-47.7	3.02	-0.7	0.1
	5.36	-29.7	2551.5	-6.3	60.2	4.86	0.4	2.9	5.27	-33.8	295.5	-2.2	108.3	4.87	0.3	-3.4
	5.42	**-10520.0**	**5.0**	**-1116.3**	**0.0**	5.19	-28.3	-83.6	**5.63**	**596.4**	**37536.7**	**-8.8**	**-4793.5**	5.64	-11.3	321.5
	5.95	14.3	221.4	0.9	76.1	5.71	15.4	193.0	6.20	-6.8	-44.9	-1.2	43.1	5.87	9.9	58.4
	6.14	-12.3	-2064.5	8.2	-229.6	6.06	5.5	-35.5	6.25	22.2	-576.1	15.8	21.2	6.38	-27.0	3.5
	6.29	27.7	1333.1	15.8	147.3	6.16	19.1	9.2	6.38	21.5	1620.0	14.5	-147.2	6.47	-7.2	25.2
	6.41	49.7	202.6	2.3	186.5	6.31	9.3	15.5	6.64	-26.9	-314.5	16.6	46.9	6.6	9.6	-15.6
	6.53	3.5	-9.2	-0.3	-3.9	6.38	1.0	-16.4	6.84	38.4	39.9	0.2	-128.3	6.72	1.6	-9.2
	6.56	16.6	166.0	-18.5	42.6	6.57	1.5	8.5	6.96	12.2	68.1	4.8	-6.2	7.05	13.7	-21.6
	6.73	-6.3	-415.3	2.4	-33.1	6.82	-13.9	-7.3	7.06	-1.7	96.8	-1.1	15.5	7.12	-2.2	8.4
SQ1-C2H3	3.76	8.3	-329.6	1.1	35.6	2.57	0.6	-0.1	3.57	12.4	-588.4	1.1	34.9	2.98	-0.7	0.0
	4.73	-16.7	930.4	0.3	138.3	4.86	-0.3	4.5	4.82	33.5	-2734.0	-1.2	-244.3	4.87	-0.4	6.6
	5.33	88.1	24058.3	-0.7	-1391.1	5.17	-35.6	305.4	**5.25**	**25779.7**	**7575823.0**	**2.5**	**-442374.2**	5.42	-12.4	423.4
	5.51	**-133.1**	**-35908.1**	**-1.1**	**2288.4**	5.59	20.0	21.9	5.56	44.4	-1458.2	-2.7	-129.8	5.74	27.5	66.9
	5.73	19.2	7134.1	-30.6	-564.6	5.84	-6.3	-36.8	5.83	10.5	-3476.0	29.1	249.4	6.09	-1.3	37.7
	6.00	-5.4	-80.0	-1.9	43.4	5.91	1.1	-12.5	6.23	-2.6	-52.8	-1.5	34.3	6.12	20.3	6.6
	6.30	-9.3	-1462.9	-24.8	49.5	5.94	-16.0	9.5	6.34	18.6	1360.4	24.4	-55.1	6.24	10.6	2.7
	6.38	-26.2	-42.7	-1.0	149.3	6.08	-6.8	-15.9	6.44	5.6	-180.4	0.1	58.4	6.54	-0.6	9.0
	6.41	-15.8	-838.1	-9.3	95.8	6.31	0.3	13.8	6.49	-9.7	-249.1	-7.9	4.4	6.55	-5.3	8.0
	6.65	8.6	0.9	1.3	4.1	6.39	2.1	-9.6	6.55	-12.0	-252.8	-21.9	-13.6	6.7	3.0	-14.5
SQ1-C2H	3.92	9.9	-305.0	0.8	26.2	2.56	0.6	0.0	3.77	-13.7	536.2	-0.9	-17.8	2.94	0.7	0.1
	4.93	-17.4	1118.0	-2.0	217.4	4.86	-0.2	4.6	5.01	28.2	-4097.9	1.1	-482.5	4.88	0.2	-6.4

	5.37	**131.9**	**79230.9**	**-0.4**	**-3166.9**	5.33	-44.6	636.8	**5.27**	**-2291.2**	**-2110255..1**	**-1.2**	**69515.5**	5.54	4.2	15.6
	5.67	16.4	-6032.0	1.8	160.8	5.45	-28.2	635.0	5.74	35.1	-1451.4	0.5	-107.9	5.63	-6.9	-195.8
	5.83	-10.3	5075.5	-21.5	-260.2	5.84	22.5	3.6	5.91	-19.3	3362.3	-20.5	-130.3	5.78	2.7	5.2
	6.05	3.9	26.4	2.2	-36.3	5.91	0.1	4.0	6.28	-0.6	-22.7	-1.2	32.7	5.93	-30.0	-30.0
	6.35	4.1	-693.7	-16.1	-111.5	6.08	0.4	-11.3	6.43	23.0	1087.7	26.7	-7.1	6.31	7.3	-21.9
	6.38	26.3	1038.5	21.0	-128.2	6.15	-13.8	-14.3	6.70	21.1	126.6	4.4	-32.0	6.41	-17.5	-21.6
	6.61	-3.7	243.3	-3.0	-13.4	6.15	4.4	-22.1	6.73	1.7	-51.0	0.4	30.7	6.43	-5.6	-5.6
	6.75	-5.3	-10.3	-0.7	-1.6	6.33	-7.0	0.9	6.76	5.4	-14.2	-4.1	-128.1	6.61	-11.1	-3.9

Table E1. Non vanishing two-photon transition tensor elements for **SQ1** and its derivatives. Standard TPA calculations. Again, the states involved in the collection of data included in Table 2 of the manuscript are highlighted in bold.

	Gas phase								CH_2Cl_2 solvent							
	A					*B*			*A*					*B*		
	ΔE_{0f}	S_{xx}^{0f}	S_{yy}^{0f}	S_{zz}^{0f}	S_{xy}^{0f}	ΔE_{0f}	S_{xz}^{0f}	S_{yz}^{0f}	ΔE_{0f}	S_{xx}^{0f}	S_{yy}^{0f}	S_{zz}^{0f}	S_{xy}^{0f}	ΔE_{0f}	S_{xz}^{0f}	S_{yz}^{0f}
	eV	a.u.	a.u.	a.u.	a.u.	eV	a.u.	a.u.	eV	a.u.	a.u.	a.u.	a.u.	eV	a.u.	a.u.
	3.30	117.2	3617.3	-0.3	674.6	2.00	0.0	-0.5	2.96	197.1	8053.2	-0.9	1302.8	1.79	-0.4	-2.6
	3.43	131.3	2564.6	-0.3	676.9	2.93	0.2	-2.6	**3.43**	**-150.6**	**15336.8**	**-0.4**	**198.8**	3.26	-0.9	-13.2
SQ2	4.04	-21.2	-924.1	0.0	-141.7	3.57	-1.8	-11.4	4.24	2.4	346.2	0.0	94.2	3.40	-5.4	-41.0
	4.26	-45.5	-1271.5	-0.8	-303.8	3.84	-6.7	-47.1	4.25	-11.6	-1732.1	1.4	-121.9	3.79	2.1	19.4
	4.27	**86.1**	**7005.6**	**-1.8**	**789.3**	4.26	-0.7	-7.5	4.30	0.4	10.9	0.5	1.5	4.24	-0.5	-1.2

Table E2. Non vanishing two-photon transition tensor elements for **SQ2**. Standard TPA calculations. Again, the states involved in the collection of data included in Table 2 of the manuscript are highlighted in bold.

F. Damped TPA results.

The two-photon absorption cross section $\delta^{TP}(\omega_1,\omega_2)$ in the degenerate case $\left(\omega_1=\omega_2=\frac{\omega_{0f}}{2}\right)$ is obtained from the data listed in the table below using the equation[2]

$$\delta^{\mathrm{TP}}(\omega)\approx 1.59501\times 10^{-2}\times\omega^2\times\sum_f \delta_{0f}^{DTP}(\omega,\omega_{0f},\Gamma) \tag{4}$$

where $\delta_{0f}^{DTP}(\omega,\omega_{0f},\Gamma)$, the two photon transition strength, listed in the following table and plotted in Figure 4 of the paper, is given by

$$\begin{aligned}\delta_{0f}^{DTP}(\omega,\omega_{0f},\Gamma) &= \frac{1}{30}\times\sum_{\alpha\beta}\left[F\times\delta_D^{\alpha\alpha\beta\beta}\left(\omega,\omega_{0f},\Gamma\right)+\left(G+H\right)\times\delta_D^{\alpha\alpha\beta\beta}\left(\omega,\omega_{0f},\Gamma\right)\right]= \\ &= \frac{1}{30}\times\sum_{\alpha\beta}\sum_f\left[F\times\left(\Phi_f^{\alpha\alpha\beta\beta}\left(\omega\right)+\Lambda_f^{\alpha\alpha\beta\beta}\left(\omega\right)\right)+\left(G+H\right)\times\left(\Phi_f^{\alpha\beta\alpha\beta}\left(\omega\right)+\Lambda_f^{\alpha\beta\alpha\beta}\left(\omega\right)\right)\times A\left(2\omega,\omega_{0f},\Gamma\right)\right]\end{aligned} \tag{5}$$

For further details and definitions see section C in this file and Ref. 2 in the paper. In the equations in this page, again ω and $\delta_{0f}^{DTP}\left(\omega_{0f}\right)$ are given in atomic units and the TPA spectra obtained are in GM units. In the table below, we list values of $\delta_{0f}^{DTP}(\omega_{TP},\omega_{0f},\Gamma)$. Γ =0.004 a.u., corresponding to a HWHM of 0.109 eV.

$h\omega_{TP}$	**SQ1**	**SQ1-py**	**SQ1-C2H3**	**SQ1-C2H**
0.816	1.083E+04	2.918E+03	1.024E+04	8.070E+03
0.952	1.555E+04	3.942E+03	1.465E+04	1.135E+04
1.088	2.281E+04	5.363E+03	2.153E+04	1.629E+04
1.225	3.458E+04	7.411E+03	3.308E+04	2.419E+04
1.361	5.495E+04	1.047E+04	5.453E+04	3.786E+04
1.497	9.322E+04	1.528E+04	1.013E+05	6.455E+04
1.633	1.743E+05	2.344E+04	2.377E+05	1.281E+05
1.769	3.809E+05	3.908E+04	9.913E+05	3.522E+05
1.905	1.123E+06	7.781E+04	4.909E+06	2.357E+06
2.041	6.475E+06	2.771E+05	1.075E+06	1.753E+06
2.177	2.283E+07	3.883E+05	1.646E+06	8.892E+05
2.313	1.313E+07	2.909E+05	1.556E+07	2.696E+06
2.449	5.810E+07	9.078E+05	2.304E+07	4.278E+07
2.585	1.081E+09	1.617E+07	1.107E+08	9.525E+07

Table F. Damped TPA calculations. Gas phase. ω_{TP} is the two-photon circular frequency. The two-photon transition strength $\delta_{0f}^{DTP}(\omega_{TP}, \omega_{0f}, \Gamma)$ is in a.u.

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