Supporting Information

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Donor-substituted Triaryl-1,3,5-Triazinanes-2,4,6-Triones: Octupolar NLO-phores with a Remarkable Transparency–Nonlinearity Trade-off

Including:

1. NLO Measurements p. S2

2. Crystal data and structure refinement for 4 p. S3

1. NLO Measurements

The first hyperpolarizabilities were determined by performing hyper-Raleigh scattering (HRS) experiments in CH_2Cl_2 solutions. This technique is based on the analysis of the incoherent light scattered second-harmonic signal from an isotropic medium. The scattered nonlinear intensity $I^{2\omega}$ can be expressed as $I^{2\omega} = C(N_S\langle\beta_S^2\rangle+N\langle\beta^2\rangle)(I^{\omega})^2$ for a solution containing N chromophores dissolved in N_S solvent molecules (concentrations expressed per milliliter of solution), where the C coefficient represents geometrical factors and experimental terms. The brackets refer to orientationally averaged β values. The C scaling factor is empirically deduced by a calibration experiment using a reference.

HRS measurements were conducted with a single-mode Nd:YAG laser emitting pulses of 10-MW peak power and 10-ns duration at 10-Hz repetition rate. The incident intensity I^{ω} is monitored by a half-wave plate and a Glan polarizer. A fraction of the incident beam is extracted from the main beam by a glass plate and sent onto a reference NPP frequency doubling calibrated powder. The fundamental beam is focused in a cell which contains the solution. The scattered harmonic signal is then collected at a right angle on a second photomultiplier after spectral selection through an interferential filter with 3-nm spectral resolution. For measurements performed in chloroform, the solvent was taken as reference, with a $\langle\beta_S^2\rangle^{1/2}$ value of 0.19 10^{-30} esu at 1.064 μm. For measurements effected in dichloromethane, ethyl violet was taken as an external reference standard with a $\langle\beta^2\rangle^{1/2}$ value of 170 10^{-30} esu at 1.907 μm. In both cases, *ca.* 10^{-3} M solutions were used. Note that the excitation wavelength was chosen so as not to allow two-photon excitation of any of the chromophores examined, *i.e.* at a much longer wavelength than twice the their absorption wavelength. The experimental accuracy is estimated to be ± 15%.

2. Crystal data and structure refinement for 4

Empirical formula	$C_{27}H_{15}N_3O_3$	
Formula weight	429.42	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system, space group	Trigonal, R 3 c	
Unit cell dimensions	a = 13.5264(13) Å	alpha = 90 deg.
	b = 13.526 Å	beta = 90 deg.
	c = 24.608(2) Å	gamma = 120 deg.
Volume	3899.2(5) $Å^3$	
Z, Calculated density	6, 1.097 Mg/m^3	
Absorption coefficient	0.073 mm^{-1}	
F(000)	1332	
Crystal size	0.25 × 0.23 × 0.15 mm	
Theta range for data collection	3.01 to 27.48 deg.	
Limiting indices	-16<=h<=17, -17<=k<=16, -25<=l<=31	
Reflections collected / unique	7587 / 1002 [R(int) = 0.0576]	
Completeness to theta = 27.48	99.8 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.989 and 0.975	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	1002 / 1 / 100	
Goodness-of-fit on F^2	1.162	
Final R indices [I>2sigma(I)]	R1 = 0.0693, wR2 = 0.1716	
R indices (all data)	R1 = 0.0720, wR2 = 0.1732	
Absolute structure parameter	-10(10)	
Largest diff. peak and hole	0.326 and -0.320 $e.Å^{-3}$	

Atomic coordinates (× 10^4) and equivalent isotropic displacement parameters ($Å^2$ × 10^3) for 4. U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
C(1)	6523(4)	1102(5)	5364(2)	36(1)
C(2)	5566(4)	931(4)	5309(2)	25(1)
C(3)	4414(3)	716(3)	5224(2)	25(1)
C(4)	4067(4)	852(4)	4725(2)	25(1)
C(5)	2953(3)	654(3)	4622(2)	22(1)
C(6)	3648(3)	358(4)	5659(2)	24(1)
C(7)	2538(4)	158(3)	5587(2)	24(1)
C(8)	2223(3)	322(3)	5072(2)	17(1)
N(9)	1079(3)	149(3)	4991(2)	19(1)
C(10)	966(3)	1123(3)	5012(2)	17(1)
O(10)	1777(2)	2072(2)	5053(1)	22(1)

Bond lengths [Å] and angles [deg] for 4.

C(1)-C(2)	1.203(6)
C(1)-H(1)	0.9500
C(2)-C(3)	1.450(6)
C(3)-C(4)	1.359(7)
C(3)-C(6)	1.398(6)
C(4)-C(5)	1.415(6)
C(4)-H(4)	0.9500
C(5)-C(8)	1.401(6)
C(5)-H(5)	0.9500
C(6)-C(7)	1.398(6)
C(6)-H(6)	0.9500
C(7)-C(8)	1.388(6)
C(7)-H(7)	0.9500
C(8)-N(9)	1.460(5)
N(9)-C(10)#1	1.396(5)
N(9)-C(10)	1.401(5)
C(10)-O(10)	1.206(5)
C(10)-N(9)#2	1.396(5)
C(2)-C(1)-H(1)	180.0
C(1)-C(2)-C(3)	178.2(6)
C(4)-C(3)-C(6)	119.5(4)
C(4)-C(3)-C(2)	120.8(4)
C(6)-C(3)-C(2)	119.7(4)
C(3)-C(4)-C(5)	123.0(4)
C(3)-C(4)-H(4)	118.5
C(5)-C(4)-H(4)	118.5
C(8)-C(5)-C(4)	115.5(4)
C(8)-C(5)-H(5)	122.2
C(4)-C(5)-H(5)	122.2

C(7)-C(6)-C(3)	120.5(4)
C(7)-C(6)-H(6)	119.8
C(3)-C(6)-H(6)	119.8
C(8)-C(7)-C(6)	118.1(4)
C(8)-C(7)-H(7)	120.9
C(6)-C(7)-H(7)	120.9
C(7)-C(8)-C(5)	123.3(4)
C(7)-C(8)-N(9)	119.0(4)
C(5)-C(8)-N(9)	117.6(4)
C(10)#1-N(9)-C(10)	123.8(4)
C(10)#1-N(9)-C(8)	118.0(3)
C(10)-N(9)-C(8)	116.8(3)
O(10)-C(10)-N(9)#2	121.7(3)
O(10)-C(10)-N(9)	122.3(4)
N(9)#2-C(10)-N(9)	115.9(4)

Symmetry transformations used to generate equivalent atoms:

#1 -x+y,-x,z   #2 -y,x-y,z

Anisotropic displacement parameters ($Å^2 \times 10^3$) for 4.

The anisotropic displacement factor exponent takes the form: $-2 pi^2 [h^2 a^{*2} U11 + ... + 2 h k a^* b^* U_{12}]$

	U11	U22	U33	U23	U13	U12
C(1)	17(2)	46(3)	48(3)	13(2)	2(2)	18(2)
C(2)	18(2)	30(2)	28(2)	8(2)	4(2)	12(2)
C(3)	16(2)	17(2)	44(3)	3(2)	0(2)	11(2)
C(4)	20(2)	23(2)	27(2)	1(2)	9(2)	7(2)
C(5)	20(2)	16(2)	28(2)	3(2)	5(2)	8(2)
C(6)	17(2)	25(2)	28(2)	6(2)	1(2)	10(2)
C(7)	18(2)	20(2)	36(2)	2(2)	6(2)	11(2)
C(8)	10(2)	8(2)	31(2)	-3(2)	-2(1)	4(1)
N(9)	14(2)	18(2)	27(2)	4(1)	2(1)	9(1)
C(10)	16(2)	12(2)	23(2)	-1(1)	-4(2)	8(2)
O(10)	17(1)	16(1)	31(2)	4(1)	3(1)	6(1)

Torsion angles [deg] for 4.

C(1)-C(2)-C(3)-C(4)	-26(16)
C(1)-C(2)-C(3)-C(6)	153(16)
C(6)-C(3)-C(4)-C(5)	0.4(6)
C(2)-C(3)-C(4)-C(5)	-180.0(4)
C(3)-C(4)-C(5)-C(8)	1.0(6)
C(4)-C(3)-C(6)-C(7)	-1.0(6)
C(2)-C(3)-C(6)-C(7)	179.3(4)
C(3)-C(6)-C(7)-C(8)	0.2(6)
C(6)-C(7)-C(8)-C(5)	1.4(6)
C(6)-C(7)-C(8)-N(9)	-178.3(4)
C(4)-C(5)-C(8)-C(7)	-1.9(6)
C(4)-C(5)-C(8)-N(9)	177.7(3)
C(7)-C(8)-N(9)-C(10)#1	-70.8(5)
C(5)-C(8)-N(9)-C(10)#1	109.6(4)
C(7)-C(8)-N(9)-C(10)	96.3(4)
C(5)-C(8)-N(9)-C(10)	-83.4(4)
C(10)#1-N(9)-C(10)-O(10)	171.7(3)
C(8)-N(9)-C(10)-O(10)	5.4(6)
C(10)#1-N(9)-C(10)-N(9)#2	-7.2(8)
C(8)-N(9)-C(10)-N(9)#2	-173.4(3)

Symmetry transformations used to generate equivalent atoms:

#1 -x+y,-x,z   #2 -y,x-y,z