Amplified Two-Photon Brightness in Organic Multicomponent Nanoparticles

Elisa Campioli,[a] Domna Maria Nikolaidou,[a] Vincent Hugues,[b] Marco Campanini,[c] Lucia Nasi,[c] Mireille Blanchard-Desce,*[b] and Francesca Terenziani*[a]

[a] *Dipartimento di Chimica, Università di Parma, Parco Area delle Scienze 17/a, 43124 Parma, Italy.*
[b] *Univ. Bordeaux, Institut des Sciences Moléculaires (CNRS UMR 5255), 351 Cours de la Libération, 33405 Talence, France..*
[c] *CNR-IMEM, Parco Area delle Scienze 37/a, 43124 Parma, Italy.*

Supporting Information

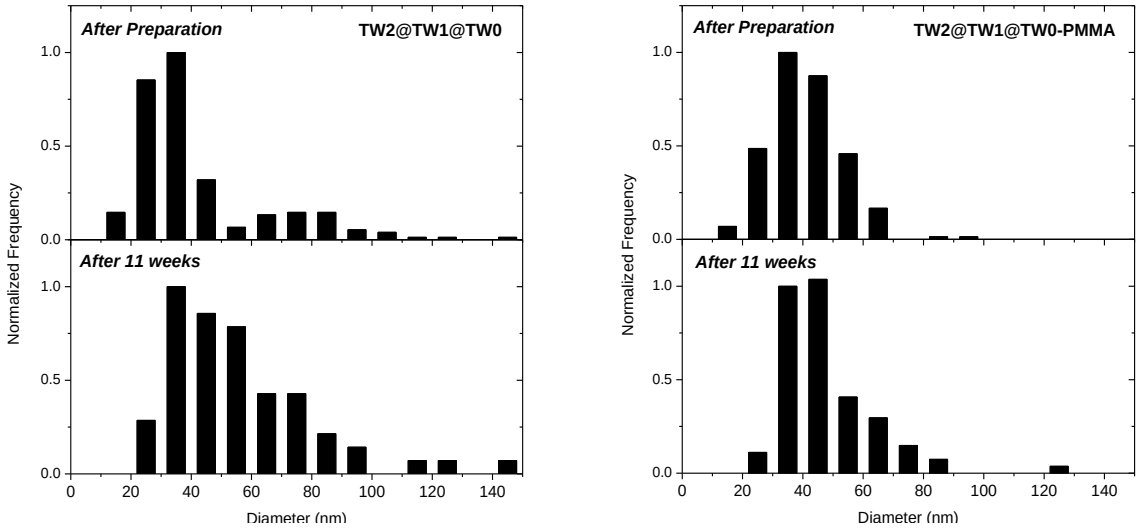

Fig. S1. Distribution analysis by TEM (performed over about 100 nanoparticles of each type) of undoped (left) and PMMA-doped (right) core@shell@shell nanoparticles. Top panels: nanoparticles deposited from freshly-prepared suspensions; Bottom panels: nanoparticles deposited from 11-weeks-aged suspensions.

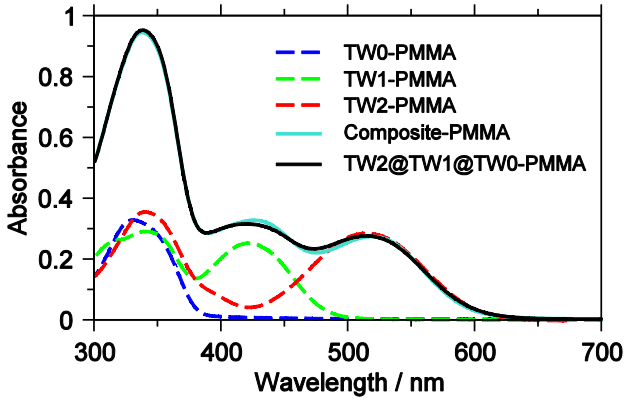

Fig. S2. Absorption spectra of the PMMA-doped nanoparticles suspensions.

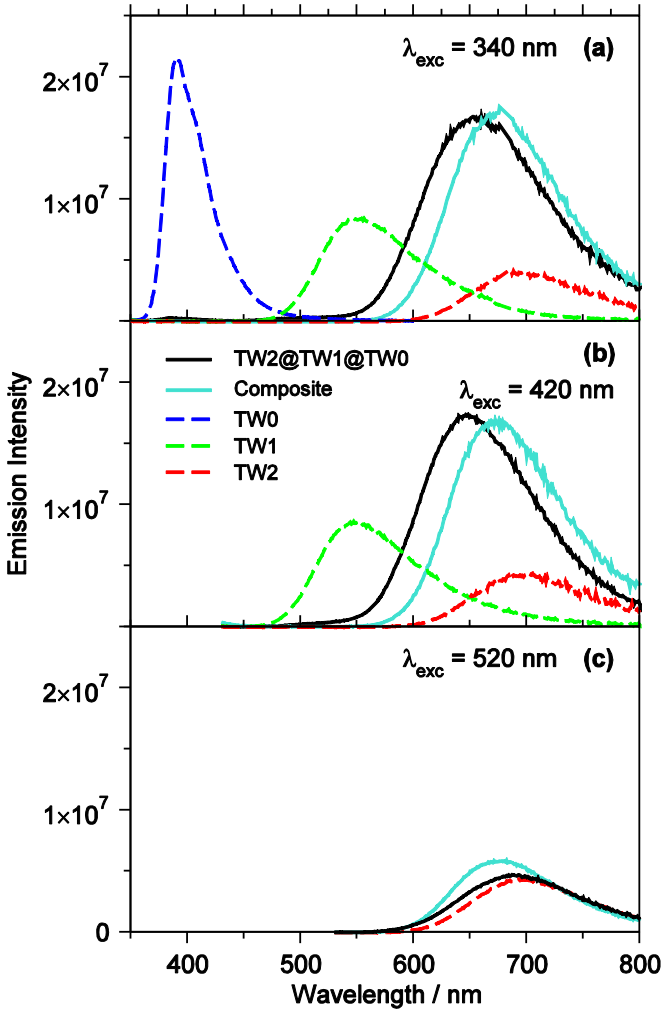

Fig. S3. Fluorescence emission spectra of the freshly-prepared PMMA-doped nanoparticles suspensions for different excitation wavelengths.

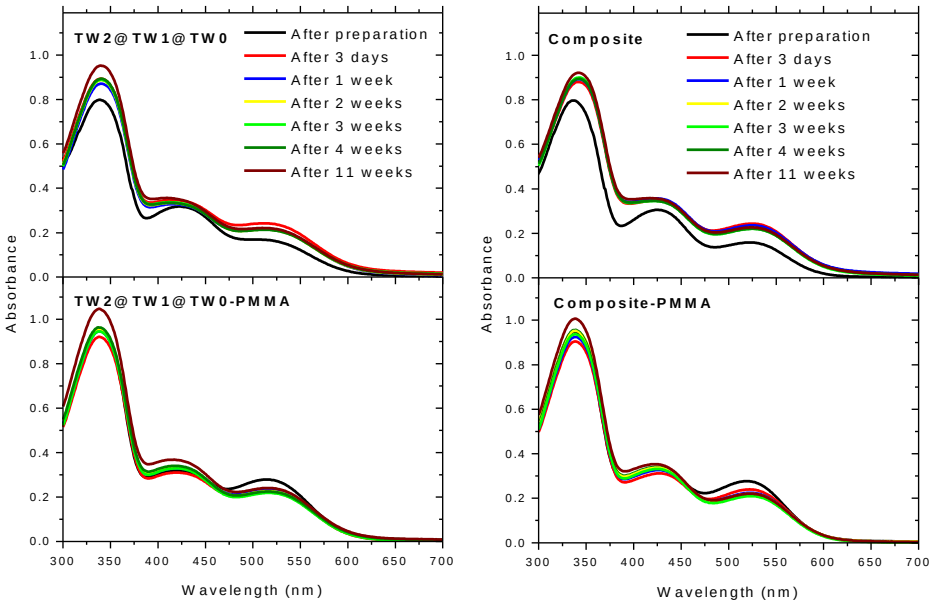

Fig. S4. Left: temporal evolution, of the absorption spectrum of undoped (top) and PMMA-doped (bottom) **TW2@TW1@TW0** nanoparticles suspension. Right: temporal evolution of the absorption spectrum of undoped (top) and PMMA-doped (bottom) composite nanoparticles suspension.

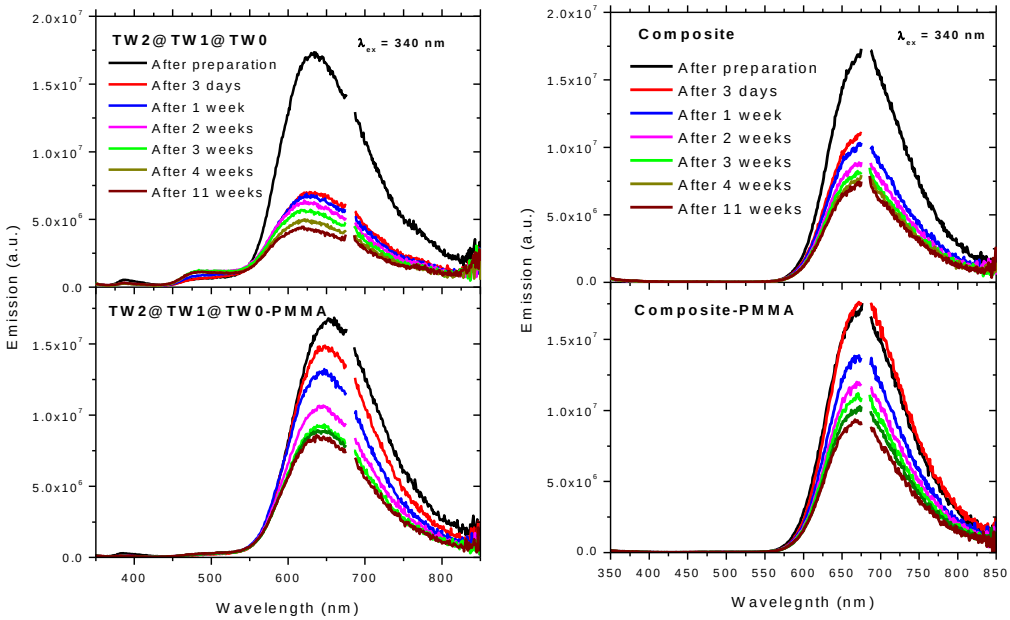

Fig. S5. Left: temporal evolution of the fluorescence spectrum of undoped (top) and PMMA-doped (bottom) **TW2@TW1@TW0** nanoparticles. Right: temporal evolution of the fluorescence spectrum of undoped (top) and PMMA-doped (bottom) composite nanoparticles suspension.

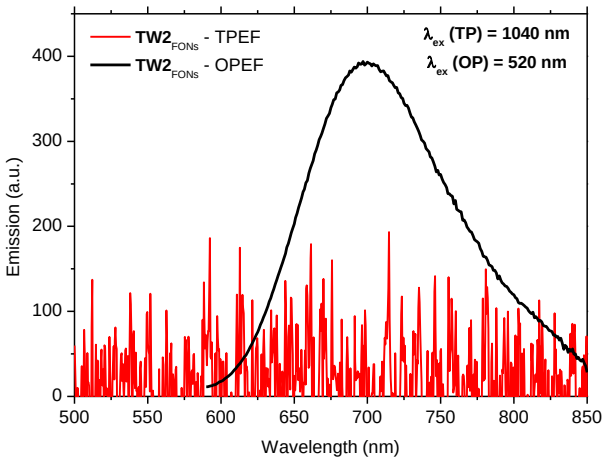

Figure S6. Comparison between two-photon excitation fluorescence (TPEF) spectrum and one-photon excitation fluorescence (OPEF) spectrum of **TW2** FONs. No significant TPEF signal could be detected.

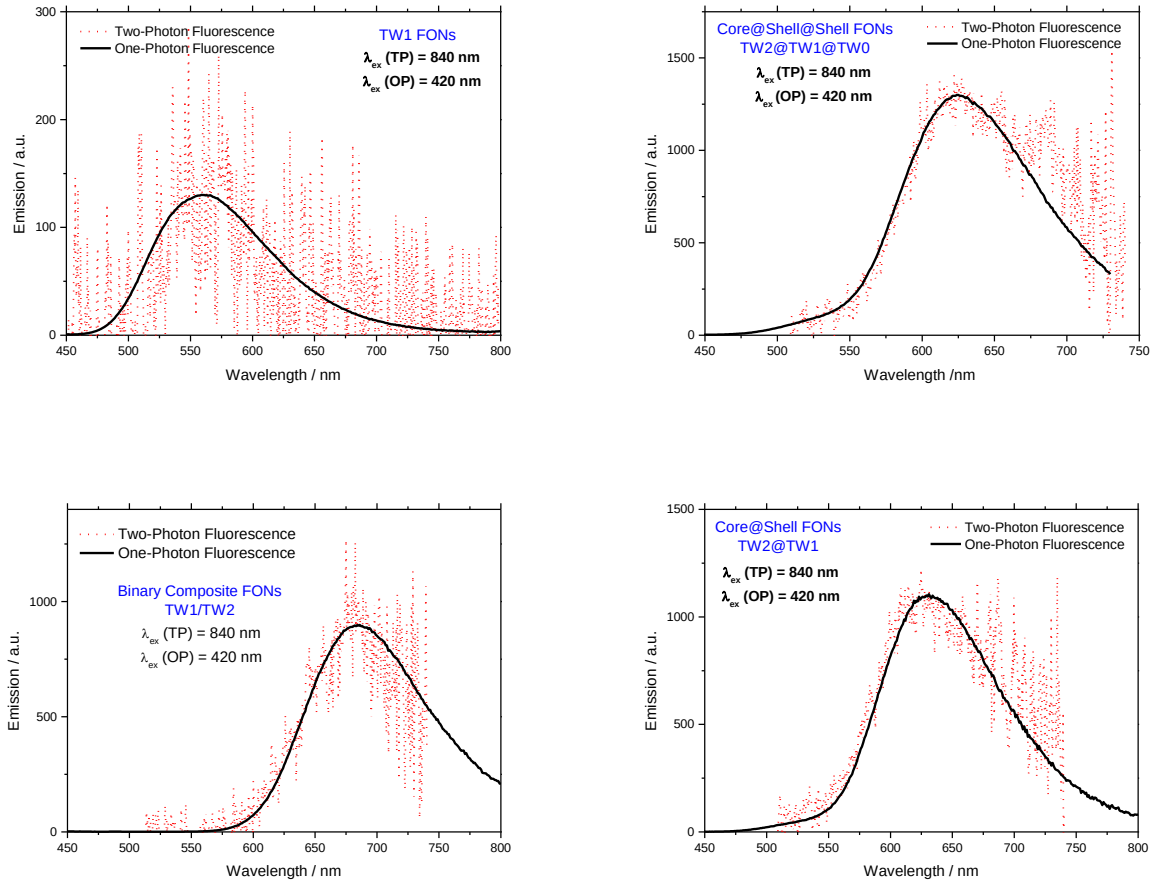

Figure S7. Comparison between the two-photon excited fluorescence (TPEF) spectrum and the one-photon excited fluorescence spectrum of the nanoparticle suspensions displaying a sizable TPEF signal.

Table S1. Fluorescence lifetimes of the freshly-prepared nanoparticles suspensions and, in parentheses, the relative contributions to the total decay. Excitation wavelength = 340 nm; variable detection wavelength, as specified in the table.

	λ_{em} = 400 nm			λ_{em} = 525 nm			λ_{em} = 620 nm		
	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)
TW0	0.48 (0.98)	2.26 (0.02)	-	-	-	-	-	-	-
TW1	-	-	-	0.40 (0.40)	1.48 (0.43)	3.93 (0.17)	-	-	-
TW2	-	-	-	-	-	-	0.73 (0.23)	2.64 (0.54)	5.83 (0.23)
Composite	ND	ND	ND	ND	ND	ND	2.07 (0.19)	6.66 (0.65)	22.94 (0.16)
TW2@TW1@TW0	< 0.4	-	-	0.35 (0.64)	1.48 (0.30)	6.71 (0.06)	2.60 (0.26)	7.51 (0.60)	25.80 (0.14)
TW0-PMMA	0.28 (0.28)	0.62 (0.71)	3.59 (0.01)	-	-	-	-	-	-
TW1-PMMA	-	-	-	0.32 (0.38)	1.29 (0.46)	3.70 (0.16)	-	-	-
TW2-PMMA	-	-	-	-	-	-	0.48 (0.23)	2.92 (0.49)	6.97 (0.28)
Composite-PMMA	ND	ND	ND	ND	ND	ND	0.54 (0.18)	3.34 (0.47)	8.25 (0.35)
TW2@TW1@TW0-PMMA	ND	ND	ND	0.34 (0.82)	1.60 (0.12)	6.66 (0.06)	1.05 (0.14)	4.62 (0.61)	13.35 (0.25)

ND: not detectable: the fluorescence is too weak to be detected.

Table S2. Fluorescence lifetimes of the freshly-prepared nanoparticles suspensions and, in parentheses, the relative contributions to the total decay. Excitation wavelength = 405 nm; variable detection wavelength, as specified in the table.

	λ_{em} = 525 nm			λ_{em} = 620 nm		
	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)
TW0	-	-	-	-	-	-
TW1	0.40 (0.38)	1.41 (0.43)	3.79 (0.19)	-	-	-
TW2	-	-	-	0.47 (0.25)	1.90 (0.60)	5.68 (0.15)
Composite	ND	ND	ND	0.82 (0.17)	4.65 (0.60)	13.12 (0.23)
TW2@TW1@TW0	<0.4 (0.52)	1.07 (0.39)	2.82 (0.09)	1.98 (0.19)	6.30 (0.70)	20.96 (0.11)
TW0-PMMA	-	-	-	-	-	-
TW1-PMMA	0.39 (0.28)	1.57 (0.50)	4.15 (0.22)	-	-	-
TW2-PMMA	-	-	-	0.29 (0.36)	1.70 (0.44)	5.06 (0.20)
Composite-PMMA	ND	ND	ND	0.48 (0.19)	3.15 (0.47)	8.85 (0.34)
TW2@TW1@TW0-PMMA	0.17 (0.82)	0.88 (0.14)	4.14 (0.04)	1.14 (0.18)	5.06 (0.62)	14.91 (0.20)

ND: not detectable: the fluorescence is too weak to be detected.