Supporting Information

A Theoretical and Experimental Approach to the Compaction Process of DNA by Dioctadecyldimethylammonium Bromide/Zwitterionic Mixed Liposomes

Alberto Rodríguez-Pulido,[1] Alberto Martín-Molina,[2] César Rodríguez-Beas,[2] Oscar Llorca,[3] Emilio Aicart,[1] and Elena Junquera[1,*]

[1]Grupo de Química Coloidal y Supramolecular, Departamento de Química Física I, Facultad de Ciencias Químicas, Universidad Complutense de Madrid, 28040-Madrid, Spain

[2]Grupo de Física de Fluidos y Biocoloides, Departamento de Física Aplicada, Facultad de Ciencias, Universidad de Granada 18071- Granada, Spain

[3]Centro de Investigaciones Biológicas, CSIC, Ramiro de Maeztu 9, 28040-Madrid, Spain

Corresponding Author: E. Junquera

Tel: +34-91-394-4131

Fax: +34-91-394-4135

E-mail: junquera@quim.ucm.es

http://www.ucm.es/info/coloidal/index.html

*To whom correspondence should be addressed

TABLE S1: Values of electrophoretic mobility, μ_e, zeta potential, ζ, and surface density charge, σ_ζ, at different values of *L*/*D* mass ratios for DODAB/DOPE-DNA and DODAB/DLPC-DNA lipoplexes. DNA concentration was kept constant at 0.050 mg/mL

DODAB/DOPE-DNA

L/*D*	$10^2\ \mu_e$ ($m^2\ V^{-1}\ s^{-1}$)	ζ (mV)	$10^3\ \sigma_\zeta$ ($C\ m^{-1}$)
1.0	– 4.61	– 59	– 21
2.1	– 4.16	– 53	– 18
3.1	– 4.13	– 53	– 18
3.4	– 3.32	– 42	– 13
5.5	1.66	21	6
5.7	2.67	34	10
8.0	3.52	45	15
9.8	3.78	48	16
14.8	3.67	47	15
∞[a]	4.17	53	18

DODAB/DLPC-DNA

L/*D*	$10^2\ \mu_e$ ($m^2\ V^{-1}\ s^{-1}$)	ζ (mV)	$10^3\ \sigma_\zeta$ ($C\ m^{-1}$)
0.8	– 4.55	– 58	– 20
2.0	– 3.95	– 51	– 17
3.1	– 4.28	– 55	– 19
4.3	– 3.55	– 45	– 15
5.0	2.94	38	12
6.0	3.36	43	14
8.1	3.71	47	16
10.0	3.39	43	14
14.9	4.23	54	19
∞[a]	4.61	59	21

[a]Liposome in the absence of DNA.

Errors are estimated to be around 2 % in electrophoretic mobility, 3% in zeta potential and around 6% in surface charge density.

TABLE S2: Parameters of the deconvoluted gaussian bands of EtBr fluorescence emission spectra in the presence of DODAB/DOPE-DNA lipoplexes at different *L*/*D* ratios: wavelength, λ_i, width, W_i, and area, A_i, in terms of % contribution to the overall fluorescence emission area (within parenthesis). Medium: aqueous HEPES 40 mM, pH = 7.4. DODAB:DOPE ratio is 1:1; DNA:EtBr ratio is 6:1; [DNA] = 0.025 mg/mL

		2 Gaussians							1 Gaussian			
L/D	I_{588}	λ_1 (nm)	W_1	A_1 (%)	λ_2 (nm)	W_2	A_2 (%)	r^2	λ_1 (nm)	W_1	A_1	r^2
0	223	582	37	5670 (42)	603	50	7876 (58)	0.999	—	—	—	—
1.0	212	581	34	3494 (28)	599	51	8983 (72)	0.999	—	—	—	—
2.0	175	581	30	1712 (17)	596	51	8633 (83)	0.999	—	—	—	—
3.0	147	583	29	1138 (13)	594	52	7525 (87)	0.999	—	—	—	—
4.0	115	582	28	656 (10)	594	52	5714 (90)	0.999	—	—	—	—
5.0	27	—	—	—	—	—	—	—	592	52	1552	0.997
6.0	25	—	—	—	—	—	—	—	591	52	1265	0.991
7.0	21	—	—	—	—	—	—	—	591	55	1363	0.991
8.0	24	—	—	—	—	—	—	—	591	51	1389	0.992
∞ [a]	10	—	—	—	—	—	—	—	597	59	708	0.998
HEPES	12	—	—	—	—	—	—	—	597	57	839	0.984

[a] Liposome in the absence of DNA. [*L*] = 0.075 mg/mL

TABLE S3: Parameters of the deconvoluted gaussian bands of EtBr fluorescence emission spectra in the presence of DODAB/DLPC-DNA lipoplexes at different *L*/*D* ratios: wavelength, λ_i, width, W_i, and area, A_i, in terms of % contribution to the overall fluorescence emission area (within parenthesis). Medium: aqueous HEPES 40 mM, pH = 7.4. DODAB:DLPC ratio is 1:1; DNA:EtBr ratio is 6:1; [DNA] = 0.025 mg/mL

		2 Gaussians							1 Gaussian			
L/*D*	I_{588}	λ_1 (nm)	W_1	A_1 (%)	λ_2 (nm)	W_2	A_2 (%)	r^2	λ_1 (nm)	W_1	A_1	r^2
0	225	582	37	5713 (42)	603	50	7954 (58)	0.999	—	—	—	—
1.0	210	582	36	5016 (40)	602	50	7657 (60)	0.999	—	—	—	—
2.1	187	581	36	3787 (34)	601	51	7217 (66)	0.999	—	—	—	—
3.2	158	581	34	2266 (24)	598	52	7108 (76)	0.999	—	—	—	—
3.6	148	582	35	2239 (26)	598	52	6408 (74)	0.999	—	—	—	—
4.2	134	582	32	1561 (19)	596	52	6446 (81)	0.999	—	—	—	—
4.7	87	581	30	814 (16)	595	52	4317 (84)	0.999	—	—	—	—
5.2	24	—	—	—	—	—	—	—	593	51	1347	0.990
5.7	16	—	—	—	—	—	—	—	596	52	926	0.995
6.2	14	—	—	—	—	—	—	—	594	55	892	0.994
∞ [a]	9	—	—	—	—	—	—	—	598	57	692	0.998
HEPES	9	—	—	—	—	—	—	—	597	56	523	0.998

[a]Liposome in the absence of DNA. [*L*] = 0.180 mg/mL

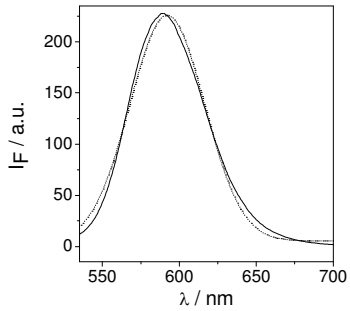
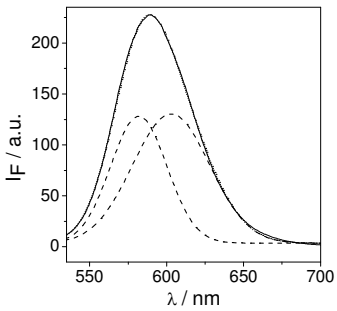
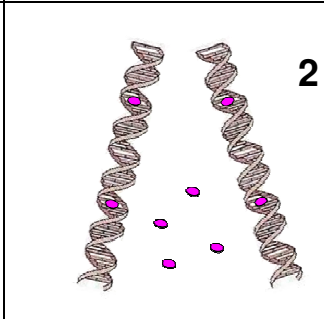
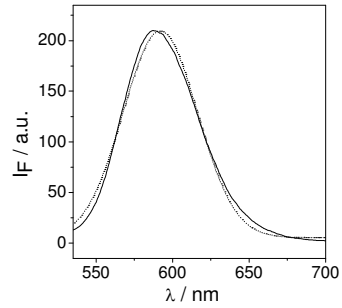
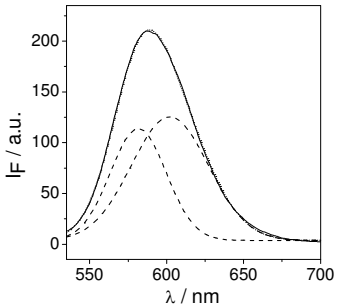
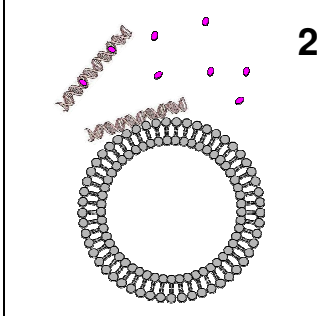
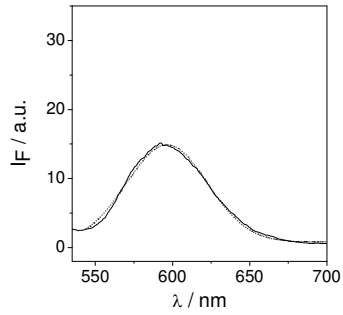
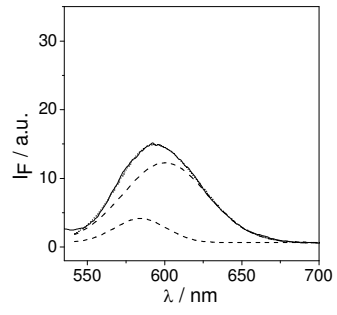
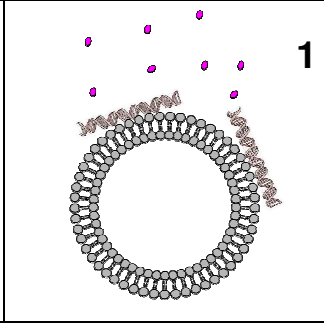
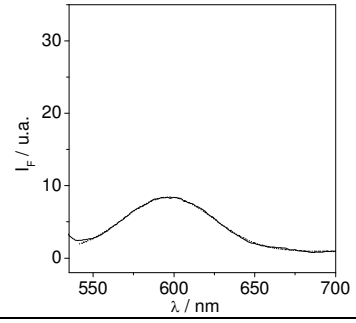
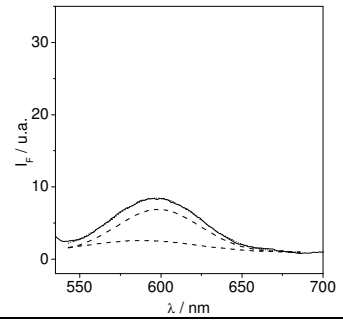
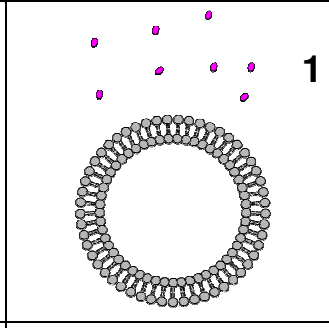
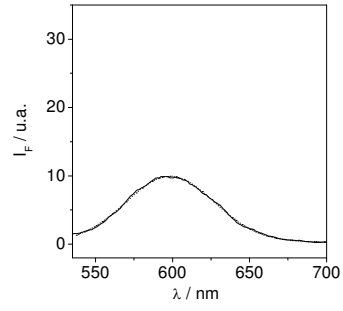
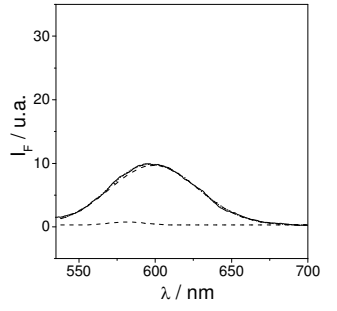
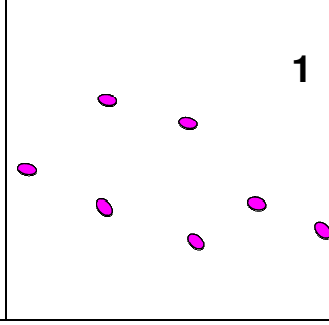
	1 Gaussian	*2 Gaussians*	*MICROENVIRONMENTS SCHEME*
$L/D = 0$			
$L/D = 1.0$			
$L/D = 5.7$			
$L/D = \infty$			
HEPES			

Figure S1. Emission fluorescence spectra of EtBr in the presence of DODAB/DLPC-DNA lipoplexes at a selection of *L*/*D* ratios, together with their deconvolutions into 1 or 2 gaussian components: *L*/*D* = 0, only DNA; *L*/*D* = 1.0, below $(L/D)_\phi$; *L*/*D* = 5.7, above $(L/D)_\phi$; *L*/*D* = ∞, only DODAB/DLPC liposomes; and HEPES, EtBr in the buffered medium, without liposomes and/or lipoplexes. Solid line: experimental spectra. Dash line: gaussian components. Dot line in 2 gaussian cases: total sum of gaussian components. Medium: aqueous HEPES 40 mM, pH = 7.4. DODAB/DLPC ratio is 1:1. A schematic diagram with the microenvironments where the probe is housed in included.

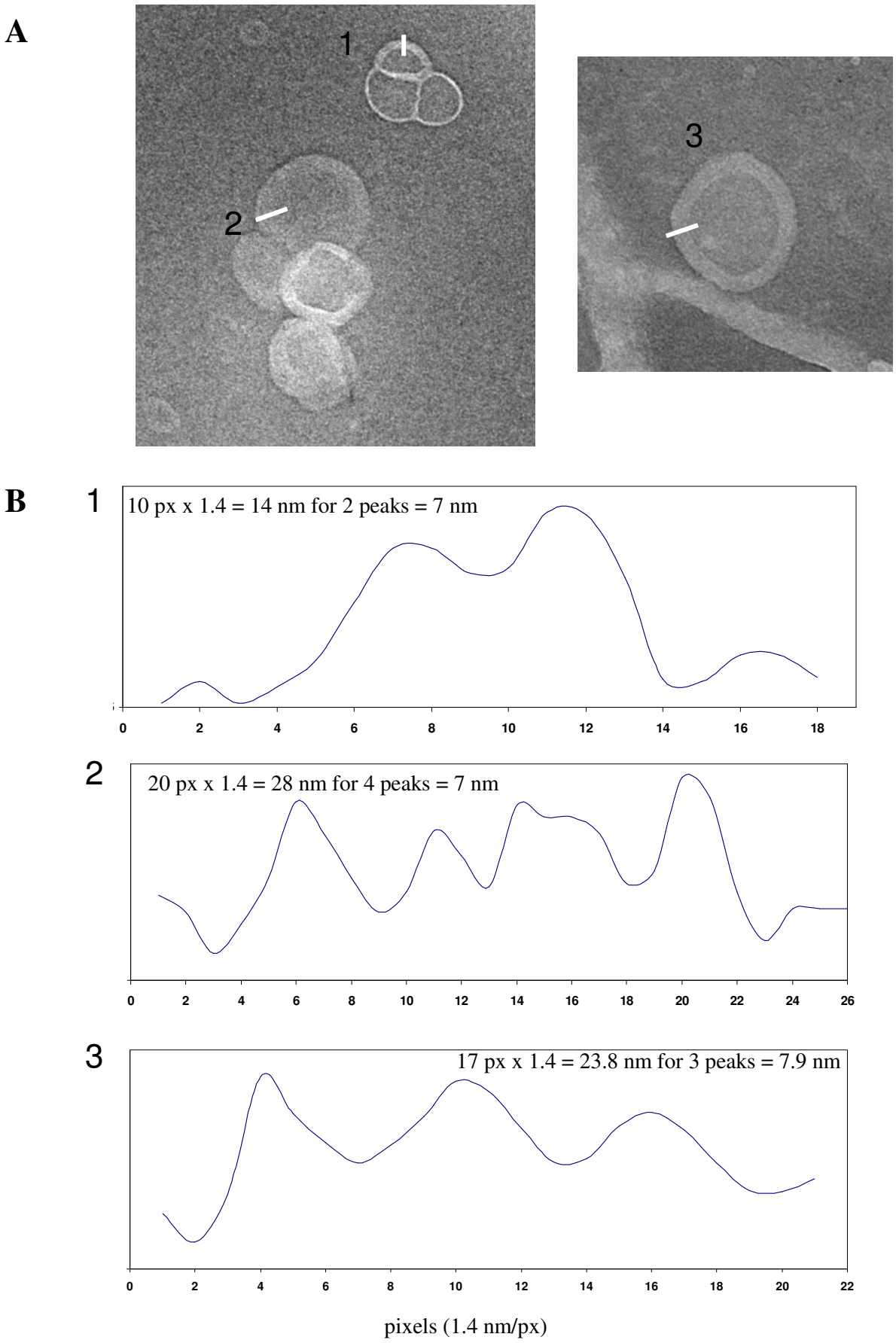


Figure S2. Details extracted from the original cryo-TEM micrographs of DODAB/DOPE-DNA lipoplexes, at $L/D >$ at $L/D > (L/D)_{\phi}$. (A) 2000x2000 pixel square images extracted from one micrograph, then low-pass filtered to remove high frequency noise, and finally scaled down to 500x500 and 1.4 nm/pixel. (B) Plots of the grey level vs. distance along a straight line across de 2D image shown in A. Averaged periodicity = 7.3 nm

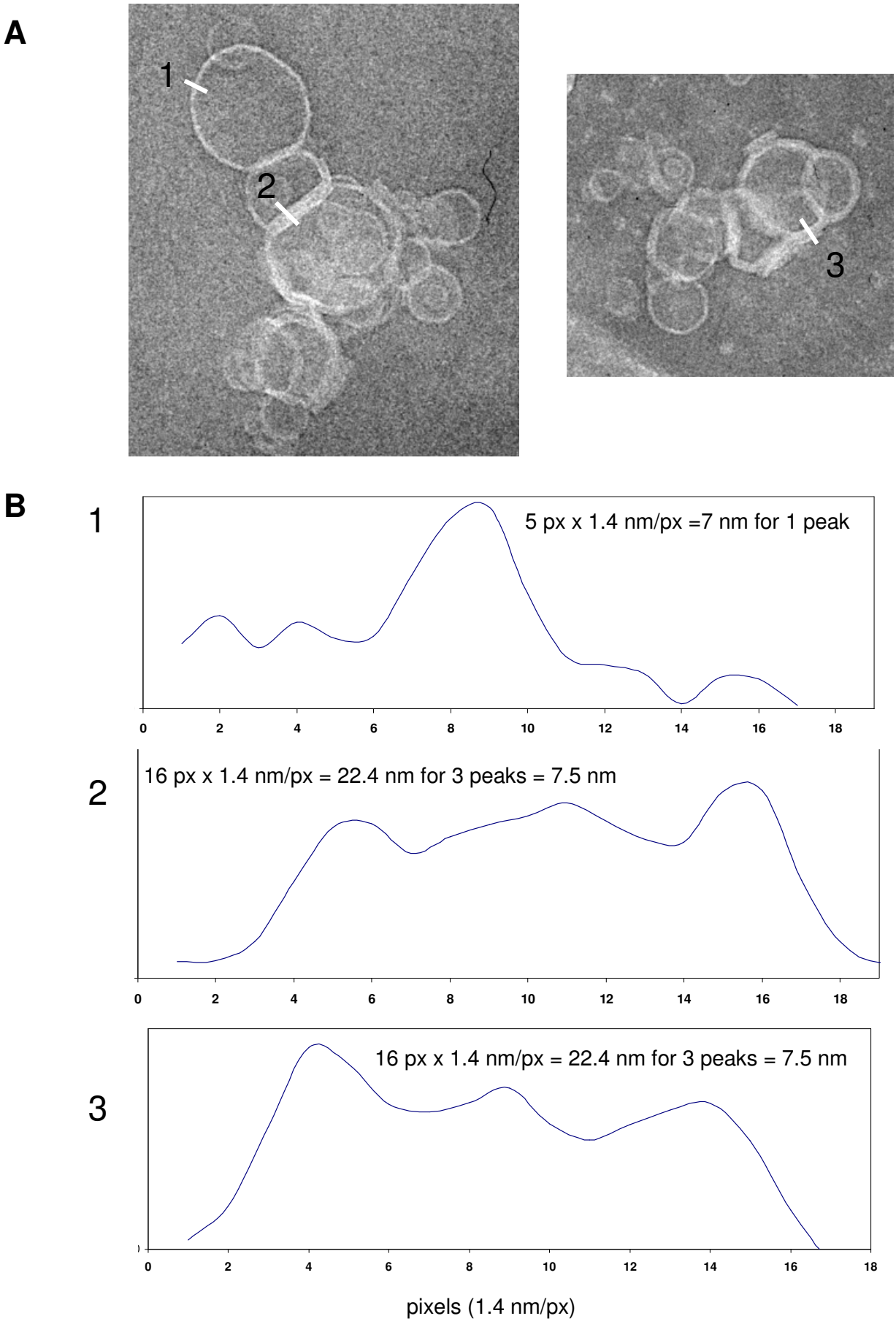


Figure S3. Details extracted from the original cryo-TEM micrographs of DODAB/DLPC-DNA lipoplexes, at L/D > at L/D > $(L/D)_{\phi}$. (A) 2000x2000 pixel square images extracted from one micrograph, then low-pass filtered to remove high frequency noise, and finally scaled down to 500x500 and 1.4 nm/pixel. (B) Plots of the grey level vs. distance along a straight line across de 2D image shown in A. Averaged periodicity = 7.3 nm