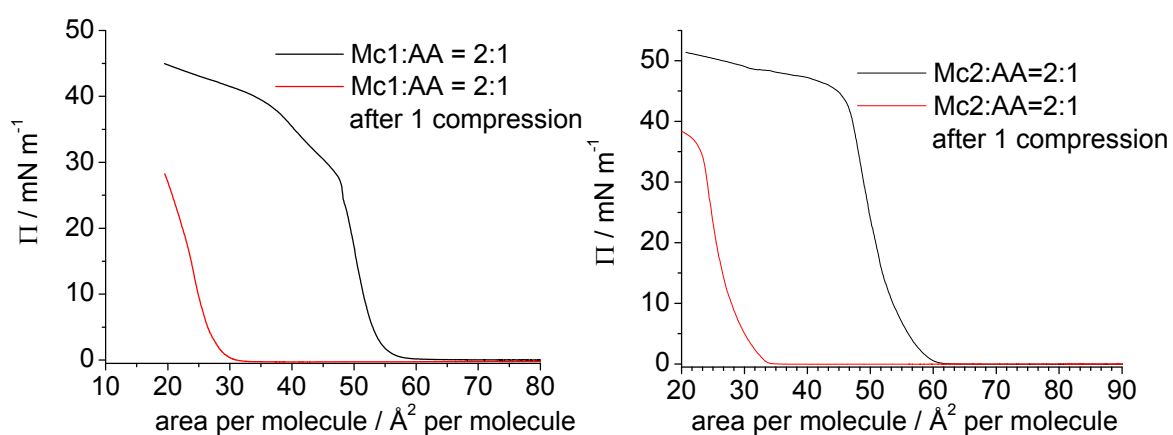


**Table SI1.**  $^1\text{H}$  NMR chemical shifts (ppm) for merocyanine MC1 in  $\text{CDCl}_3$ . See Scheme 1 for the atom numbering.

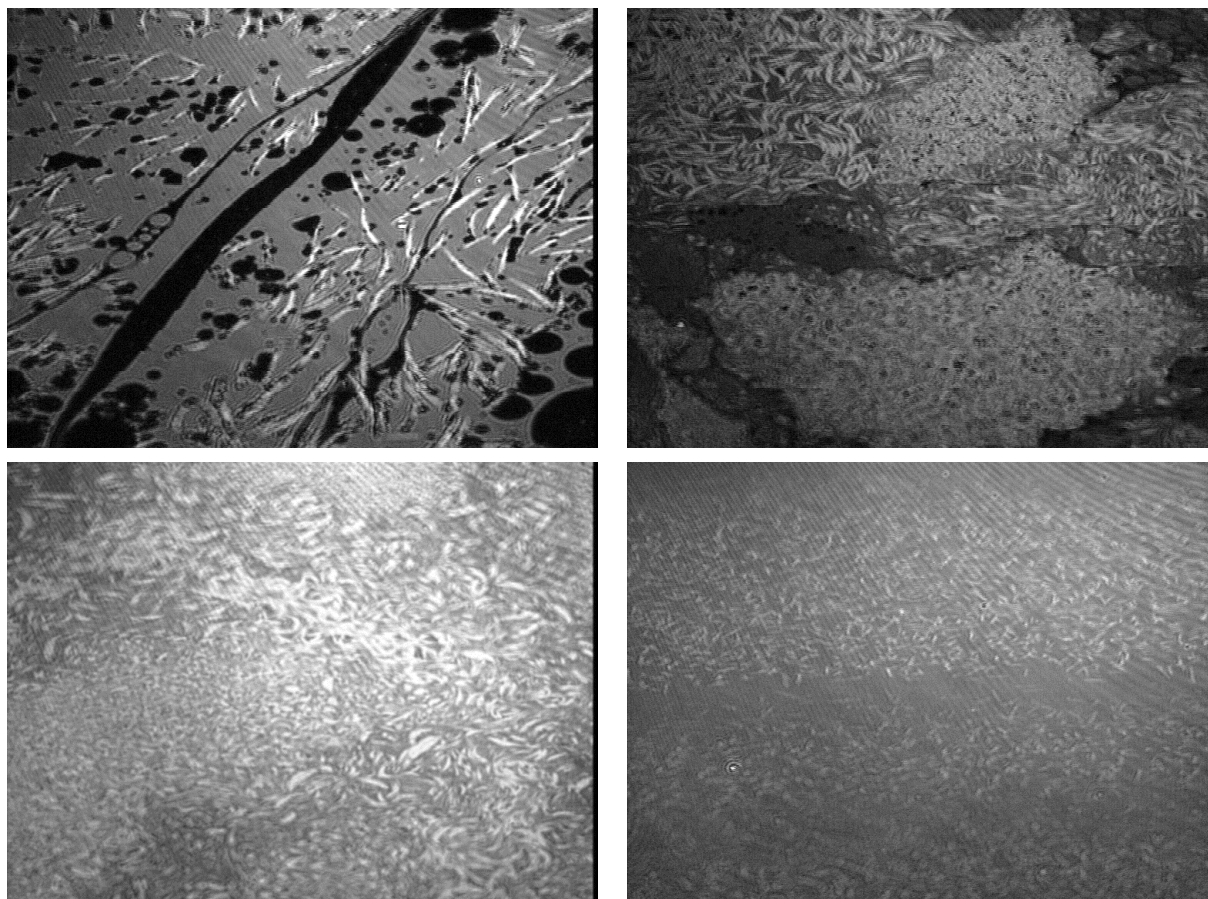
$\delta_1$	$\delta_2$	$\delta_3$	$\delta_4$	$\delta_5$	$\delta_6$	$\delta_7$	$\delta_8$	$\delta_9$	$\delta_{10}$
7.69	7.35	7.49	7.27	4.27	1.94	1.55	1.25-1.45	0.91	5.03

**Table SI2.** AFM roughness of LB films obtained from 1:2 MC(1/2):AA solutions ( $\pi = 20$  mN/m, transfer ratio = 100)

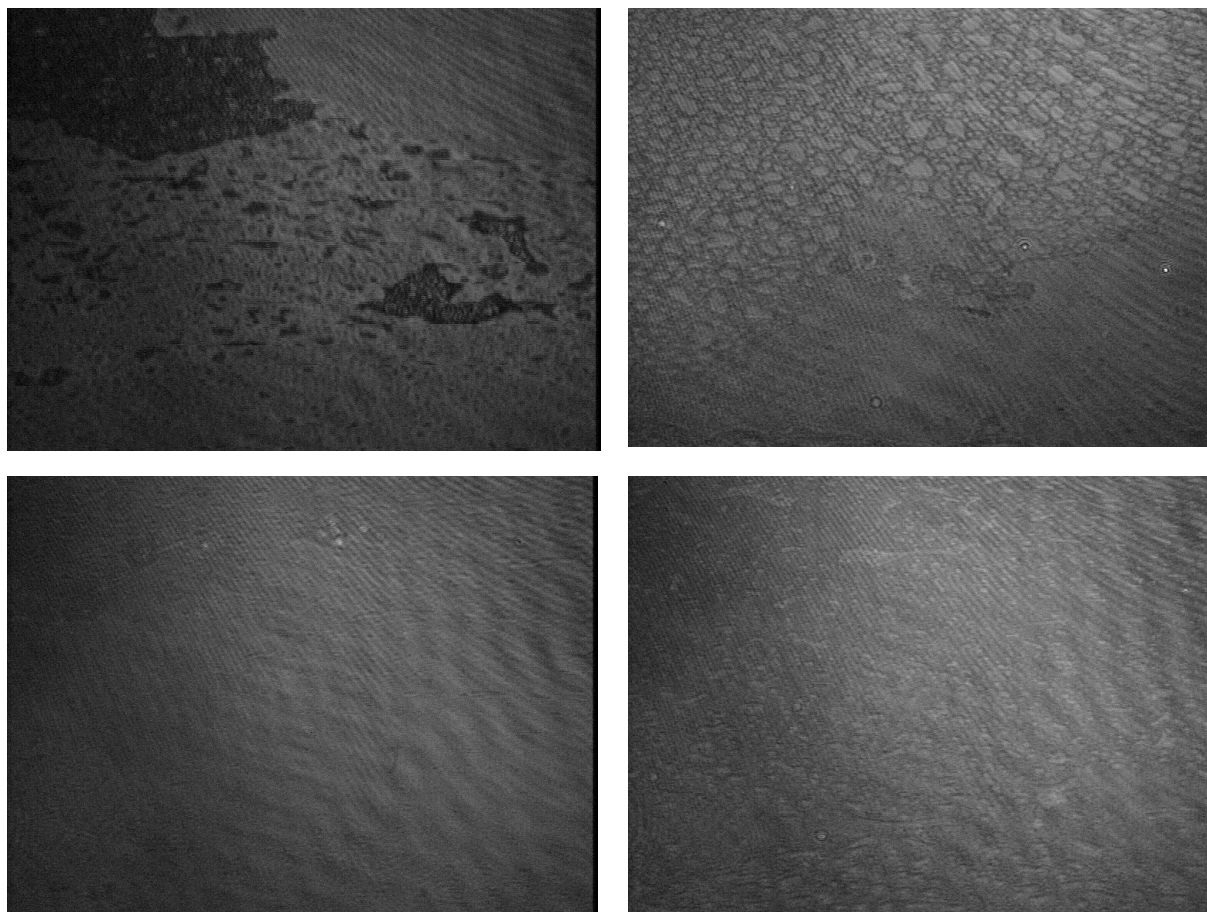
Layer Number	Roughness [ $\text{\AA}$ ]	
	MC1	MC2
3	35	20
9	40	25
29	60	45



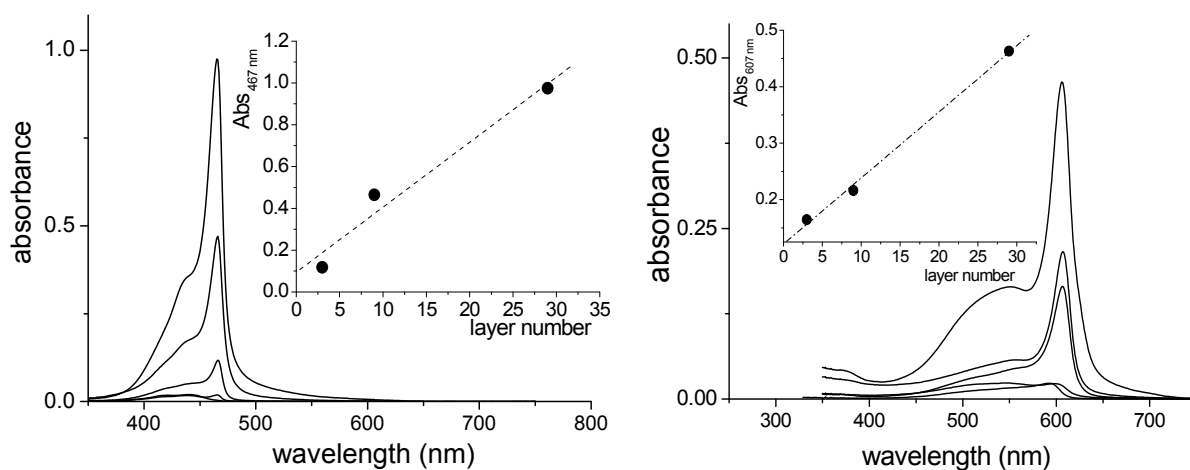
**Figure SI1.** Irreversibility of surface-pressure( $\pi$ )/area-per-molecule isotherms for 2:1 MC(1/2):AA mixtures.  $T = 20$  °C.



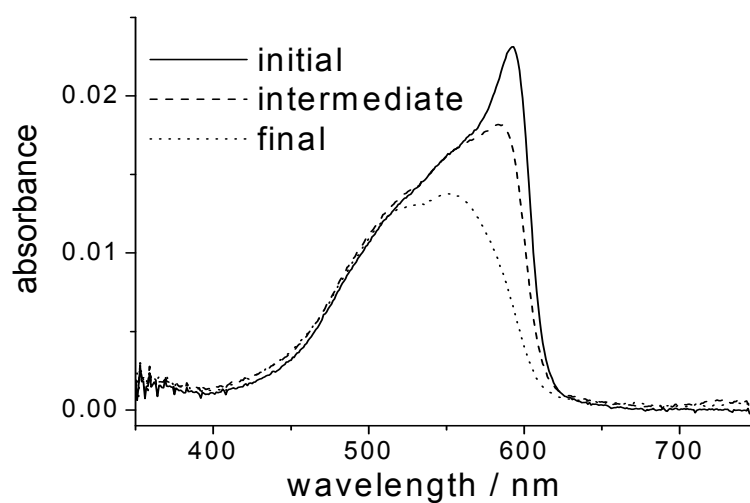
**Figure SI2.** Images of floating MC1 films taken at surface pressures 0.1, 3, 10 and 25 mN/m. The window width is 450  $\mu\text{m}$ .



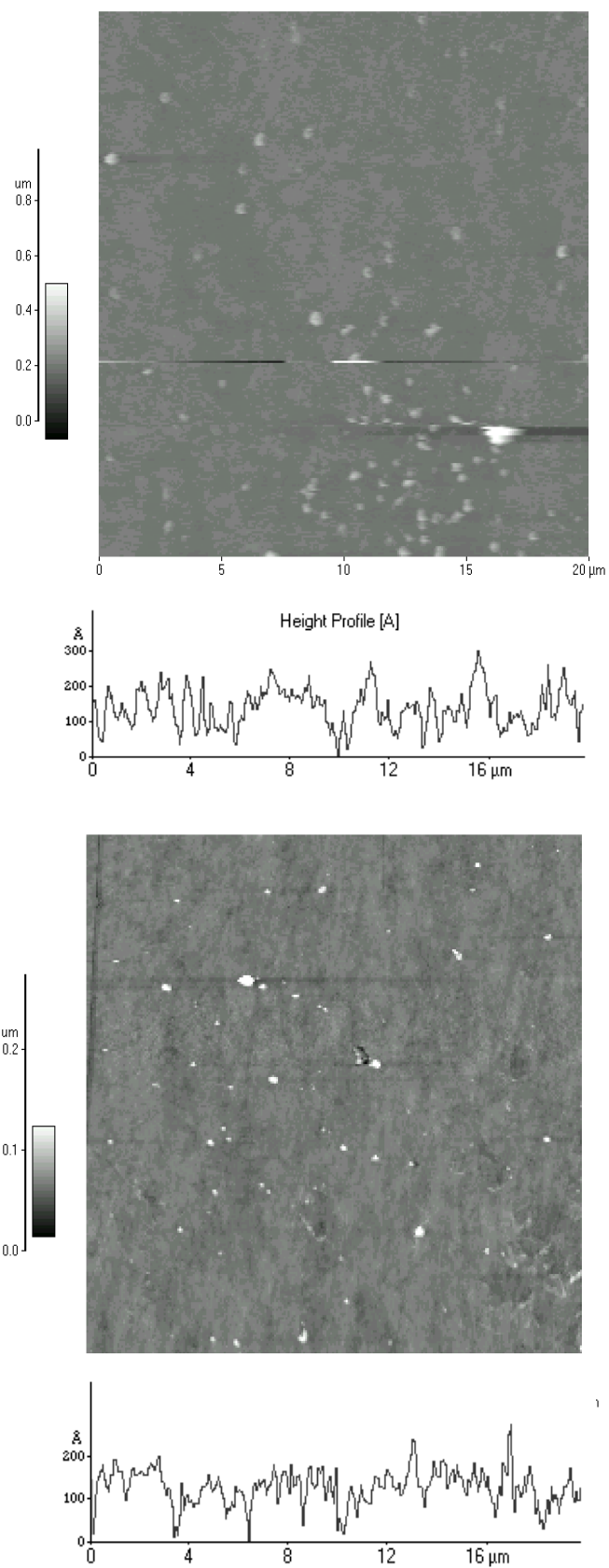
**Figure SI3.** Images of the MC1:AA 1: 2 mixture floating films taken at surface pressures 0, 10, 15 and 32 mN/m. The window width is 450  $\mu\text{m}$ .



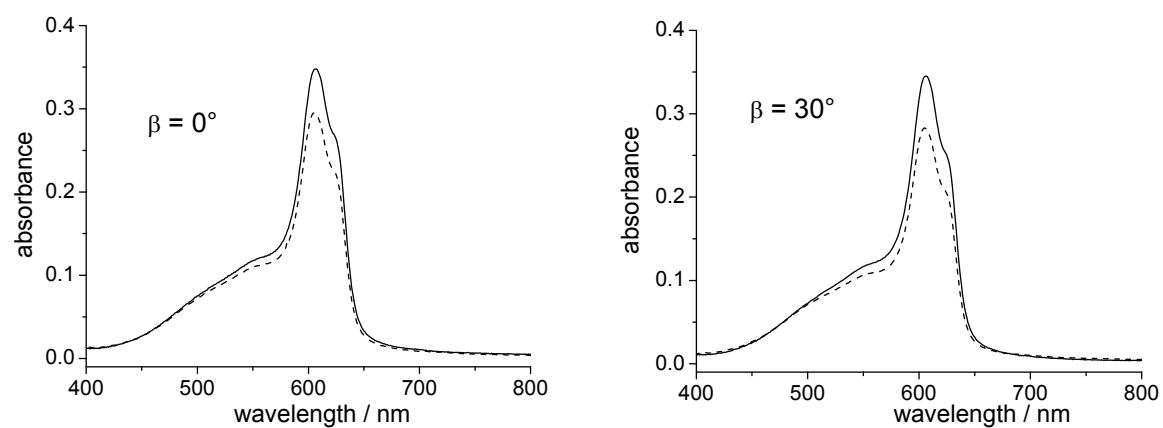
**Figure SI4.** UV-vis absorption spectra of LB films obtained with 1:2 MC:AA mole ratio: effect of the number of deposited layers. Left: MC1, right: MC2. Insets: maximum absorbances of the J bands vs. number of layers.



**Figure SI5.** Ambient-light induced photodegradation of an LB monolayer obtained from a 1:2 MC2:AA mixture.



**Figure SI6.** AFM images of 29-layer LB films obtained from 1:2 MC(1/2):AA solutions. MC1 on top.



**Figure SI7.** Linear dichroism of a 9-layer film deposited from a 1:2 MC2:AA mixture at 25 mN  $\text{m}^{-1}$ . Light polarization: vertical (solid), horizontal (dashed).