Experimental Section.

Synthesis of the gemini m-2-m $(AcAla_p)_2$ (m = 10-22, p = 3,4 and 5) Gemini-oligoalanines were synthesized according to reference [11] by adding gemini bromide to a suspension of the silver salt of the aceylated peptide in methanol (upon mixing the peptide acid and Ag_2CO_3 (1 eq.), followed by vigorous stirring under slight vacuum for 1 h). The mixture was stirred for 30 min at 40-50°C and filtered on Celite to give a colorless solution. After evaporation, the product was dissolved in chloroform/methanol (9/1, v/v) and precipitation with ether.

m-2-m $(AcAla_3)_2$ (example of m=18) ***RMN ^{1}H (400 MHz, CD_3OD, 25°C, δ ppm)*** **:** 4.38 (4H,q, ^{3}J=7.09 Hz); 4.17 (2H, q, ^{3}J=7.09 Hz); 3.94 (4H,s) 3.47 (4H, m); 3.24 (12H, s); 1.99 (6H, s); 1.85 (4H, m); 1.39 (18H, m) ; 1.31 (60H, m); 0.93 (6H, t, ^{3}J=6.95 Hz) ***NMR ^{13}C (100 MHz, CD_3OD, 25°C, δ ppm):*** 179.17; 174.94; 173.66; 173.14 ; 66.81; 56.97; 51.88; 51.83 ; 50.50; 50.45; 33.08; 30.81; 30.70; 30.63; 30.48 ; 30.36; 27.36; 23.74; 22.43; 19.19 ; 18.10 ; 17.95 ; 14.46

m-2-m $(AcAla_4)_2$ (example of m=18) ***RMN ^{1}H (400 MHz, CD_3OD, 25°C, δ ppm)*** **:** 4.34 (6H,m); 4.15 (2H, q, ^{3}J=7.09 Hz); 3.93 (4H,s) 3.44 (4H, m); 3.23 (12H, s); 1.99 (6H, s); 1.85 (4H, m); 1.38 (24H, m) ; 1.31 (60H, m); 0.92 (6H, t, ^{3}J=6.98 Hz) ***NMR ^{13}C (100 MHz, CD_3OD, 25°C, δ ppm):*** 179.36; 175.06; 174.58; 173.60 ; 173.36; 66.85; 56.98; 52.02 ; 51.85; 50.74 ; 50.52; 50.30; 33.08; 30.81; 30.76; 30.71; 30.63 ; 30.48; 30.36 ; 27.37; 23.75; 22.45; 19.24 ; 18.03 ; 17.99 ; 17.88 ; 14.45

m-2-m $(AcAla_5)_2$ (example m=18) ***RMN ^{1}H (400 MHz, CD_3OD, 25°C, δ ppm)*** **:** 4.34 (8H,m); 4.14 (2H, q, ^{3}J=7.03 Hz); 3.94 (4H,s) 3.46 (4H, m); 3.23 (12H, s); 1.99 (6H, s); 1.86 (4H, m); 1.38 (30H, m) ; 1.31 (60H, m); 0.92 (6H, t, ^{3}J=6.98 Hz) ***NMR ^{13}C (100 MHz, CD_3OD, 25°C, δ ppm):*** 178.86; 176.03; 174.56; 173.57; 173.47; 170.56; 66.89; 56.99; 51.98; 51.83; 50 ;88 ; 50.54 ; 50.39; 33.09; 30.81; 30.78; 30.71; 30.63 ; 30.49; 30.37 ; 27.37; 23.75; 22.44; 19.28 ; 18.12 ; 17.99 ; 17.81 ; 17.77 ; 14.45

Preparation of the samples for analysis after synthesis and prior to FTIR and CryoTEM

Samples were prepared as followed: the gemini m-2-m $(AcAla_p)_2$ (obtained as a white powder after synthesis) were dissolved in water by heating the surfactants above their Krafft temperature (determined by conductimetry, see the chart below) and cooling down at room temperature. Deionized water (Purelab Prima Elga, 18.2 MΩ.cm) was used to perform analysis by TEM, SAXS while D_2O (Eurisotop CEA Saclay, France) was used for FTIR studies (because of the strong absorption of water in the amide band domain). A control experiment by TEM showed no morphological change between D_2O and water. As the cation-anion interaction control the aggregation procedure, we preferred not to add any salt for the pH adjustment which may interfere with the gemini-peptide interaction. The pH of the system increased slightly with the increase in gemini-peptide concentration, and reached the equilibrium value of about 8.5 at 100 mM.

As the samples were cooled down, precipitation occurred for p=3, 4 ; m≥ 16 and for p=5, for all the investigated chain lengths. This precipitation was slow for p=3, 4 (few hours m≥ 18 to few days when m=16), whereas this phenomenon was much faster for p=5 (few minutes). The concentration used to perform all the analysis was 10 mM and 100 mM, except for p=5, only solutions of 10 mM were studied, since gemini m-2-m $(AcAla_5)_p$ could not be easily dissolved at 100mM.

m=	m-2-m $(AcAla_3)_2$	m-2-m $(AcAla_4)_2$	m-2-m $(AcAla_5)_2$
12	≤ 2°C	≤ 2°C	25°C
14	≤ 2°C	≤ 2°C	43 °C
16	≤ 2°C	≤ 2°C	55°C
18	≤ 2°C	≤ 2°C	76°C
20	15°C	23°C	96°C
22	30°C	44°C	≥ 100°C

Table S.1. Krafft temperatures (at 3 mM as usually defined) of gemini n-2-n $(AcAla_p)_2$ determined by conductimetry as a function of m and p. Note that at such a concentration Na+(AcAlap)2 are soluble for p=3and 4, and precipitate for p=5.

FT-IR Spectroscopy Measurements. The FT-IR spectra were recorded on a Bruker IFS 55 FT-IR spectrometer (Bruker, Karlsruhe, Germany) equipped with a detector (DKATGS with KBr window). To avoid the big absorption peak of H_2O, which hides the amide I peaks, samples were prepared in D_2O, at a concentration of 100 mM. Generally, 100 scans were added at a resolution of 4 cm^{-1}. The transmission measurements were performed with two ZnSe windows.

Transmission Electron Microscopy. A small drop of the gemini alanine samples in water was deposited on 400-mesh carbon-coated copper grid. After one minute, the excess liquid was blotted with filter paper. These air-dried specimens were then stained by applying a small drop of a 0.2% wt aqueous solution of uranylacetate and removing the excess solution on a filter paper. TEM images were recorded using a Philips EM 120 electron microscope operating at 120 kV.

Kinetics of chiral ribbons formation

Table S.1 : Time necessary for the ribbons observation, as a function of hydrophobic chain length *m* and alanine length *p*

	***m*-2-*m* ($AcAla_3$)$_2$**	***m*-2-*m* ($AcAla_4$)$_2$**	***m*-2-*m* ($AcAla_5$)$_2$**
m=10			~ 5 hours
m=12			~5 hours
m=16	~2 weeks	~ 2 days	
m=18	~2 weeks	~3 weeks	

For p = 3, twisted ribbons were observed with gemini 16-2-16 and 18-2-18, only after 2 weeks. For p = 4, this morphology was obtained much later (1 month) with 16-2-16. Multilayered tubules were obtained with 18-2-18 typically after 3 weeks. (In that particular case, twisted ribbons -which then transformed in tubular structures- were also observed). With p=5, only helical ribbons were formed after few hours, without showing intermediate twisted ribbons.

TEM images of AcAla3Na, AcAla4Na, and AcAla5Na aggregates.

At the same concentration as gemini Alap images shown above, AcAla3 is soluble and no aggregates were observed. AcAla4Na and AcAla5Na show precipitates with some elongated structures without any chirality.

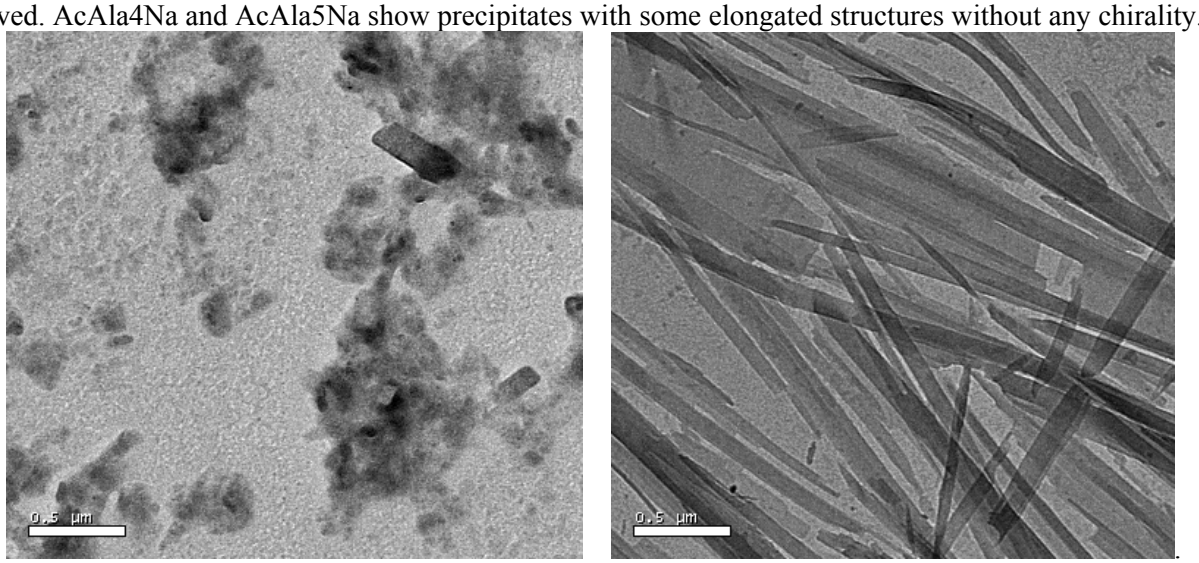

Fig. S2 TEM images of the aggregates of $AcAla_4$ (left) and $AcAla_5$(right)

Handedness of 10-2-10 $(AcAla5)_2$ and 12-2-12$(AcAla5)_2$

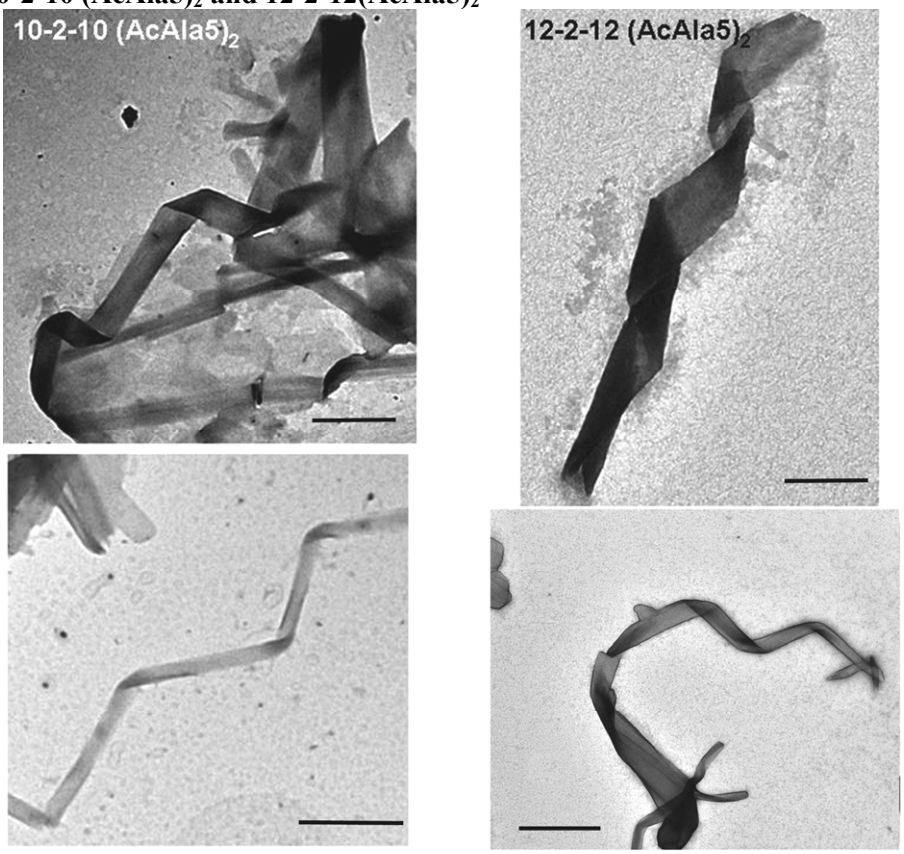

Fig. S3 TEM images of the aggregates of 10-2-10 $(AcAla5)_2$ and 12-2-12 $(AcAla5)_2$ showing all form right handed helices.

Infrared spectroscopy data : anti-symmetric / symmetric CH_2 stretching bands and amide I bands values as a function of time

The values of anti-symmetric / symmetric CH_2 stretching bands at around 2920 and 2850 cm^{-1} and amide I bands at around 1650 cm^{-1} are summarized for various chain lengths and alanine lengths. Wavenumbers shown in bold character represent the major peak, whereas those in italic labelled with w indicate weak peaks.

Table S.2: Values of anti-symmetric / symmetric CH_2 stretching bands and amide I bands after 1 day and 5 days respectively

	ν_{as}/ν_s CH_2 (cm^{-1}) **(1 day)**	ν_{as}/ν_s CH_2 (cm^{-1}) **(5 days)**	Amide I (cm^{-1}) **(1 day)**	Amide 1 (cm^{-1}) **(5 days)**
10-2-10 $(AcAla_3)_2$	2926.1 / 2855.3	2925.8 / 2855.1	1643.8	1643.1
12-2-12 $(AcAla_3)_2$	2926.2 / 2855.3	2926.4 / 2854.8	1643.1	1641.8
14-2-14 $(AcAla_3)_2$	2925.8 / 2855.1	2925.3 / 2854.8	1643.2	1641.6
16-2-16 $(AcAla_3)_2$	2924.6 / 2854.4	2924.6 / 2853.6	1643.5	1639.5
18-2-18 $(AcAla_3)_2$	2924.3 / 2852.7	2922.6 / 2852.4	1640.3	1637.6
20-2-20 $(AcAla_3)_2$	2923.1 / 2852.4	2922.1 / 2852.1	1641.3	1639.7
22-2-22 $(AcAla_3)_2$	2923.8 / 2851.1	2921.3 / 2851	1641.4	1639.8
10-2-10 $(AcAla_4)_2$	2926.2 / 2855.0	2925.9 / 2855.2	1644.8	1642.4
12-2-12 $(AcAla_4)_2$	2926.0 / 2855.1	2925.8 / 2854.9	1643.7	1641.1
14-2-14 $(AcAla_4)_2$	2925.8 / 2854.9	2925.6 / 2854.3	1643.2	1641.8

16-2-16 ($AcAla_4$)$_2$	2924.9 / 2853.9	2924.6 / 2852.5	1642.3	1639.0
18-2-18 ($AcAla_4$)$_2$	2924.1 / 2853.1	2920.9 / 2851.6	1641.4	*1685(w)*+**1630.2**
20-2-20 ($AcAla_4$)$_2$	2923.3 / 2852.8	2917.4 / 2849.7	1640.9	*1683(w)*+**1628.8**
22-2-22 ($AcAla_4$)$_2$	2923.2 / 2851.9	2916.4 / 2849	*1691*+ **1641**	*1683(w)*+**1629.1**
10-2-10 ($AcAla_5$)$_2$	2926.1 / 2855.1	2926.0 / 2855.4	*1685(w)*+**1633.5**	*1686(w)*+**1626**
12-2-12 ($AcAla_5$)$_2$	2925.3 / 2853.9	2924 / 2853.5	*1686(w)*+**1631.4**	*1685(w)*+**1624.3**
14-2-14 ($AcAla_5$)$_2$	2924.8 / 2853.2	2921.5 / 2851.1	*1685(w)*+**1630.7**	*1685(w)*+**1623.7**
16-2-16 ($AcAla_5$)$_2$	2924.1 / 2852.9	2918.2 / 2850.3	*1685(w)*+**1629.3**	*1685(w)*+**1623.1**
18-2-18 ($AcAla_5$)$_2$	2923.6 / 2852.7	2918.2 / 2850.1	*1686(w)*+**1626.1**	*1686(w)* +**1623.2**
20-2-20 ($AcAla_5$)$_2$	2923.1 / 2852.3	2818.6 / 2850.4	*1686(w)*+**1625.2**	*1686(w)* +**1623.1**
($AcAla_3$)Na				*1639*
($AcAla_4$)Na				*1670*(w) +**1622**
($AcAla_5$)Na				*1670*(w) +**1618**

The appearance of a peak at ~1685 cm^{-1}, in presence of a more intense peak situated between 1635-1620 cm^{-1} is typically attributed to the formation of anti-parallel β sheets. As can be seen in table S.2, this particular secondary structure appears only after few days with p=4, whereas anti-parallel β sheets are formed much faster with p=5. In parallel, the absence of the peak at 1670 cm^{-1} clearly demonstrate the absence of β turn. Therefore the antiparallel β sheet is due to intermolecular interaction rather than intramolecular organization. For comparison, the values of the amide I band of $AcAla_pNa$ are shown. In these cases, the strong peak at around 1620 cm^{-1} is accompanied by a weaker band at 1670 cm^{-1} indicating that the peptides form β turn when they are not coupled with gemini surfactants.

To illustrate the structural evolution with time with p=4, figure S.4 is also shown. The obtained spectra clearly show a shift of CH_2 stretching bands to lower wavenumbers, indicating a more crystalline organization. This slow organization is reciprocally associated with a progressive shift of the amide 1 band to lower wavenumbers and the observation of a peak at ~1685 cm^{-1}, characteristic of anti-parallel β sheets formation

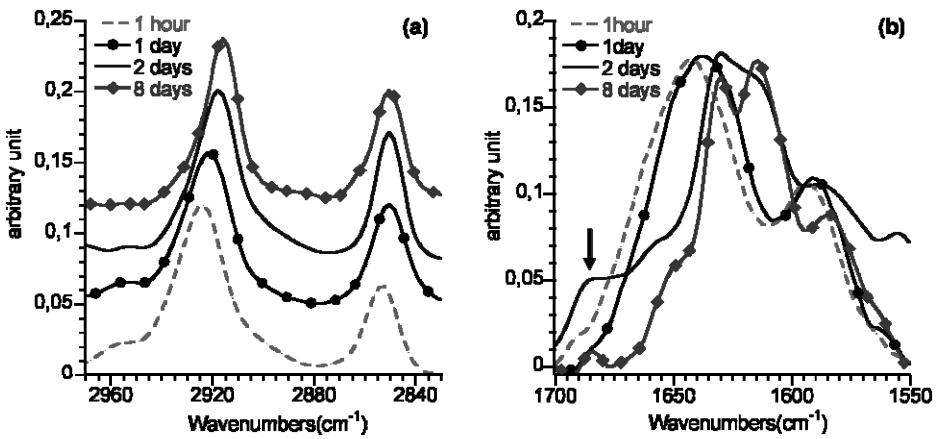

Fig. S4 Infrared spectra of 20-2-20($AcAla_4$)$_2$ at a concentration of 100 mM in D_2O. Temporal evolution of (a) the anti-symmetric / symmetric CH_2 stretching bands (b) the amide I bands. The arrow indicates the apparition of a peak at ~1685 cm^{-1}, characteristic anti-parallel β sheets formation.

CD spectra of gemini peptides compared to FT-IR spectra

Since all these ribbons are present as white gel-like precipitates, they strongly scatter light, and it turned out that CD measurements were difficult to perform for structural study. The concentration range that we were able to investigate even at 0.1 mm cell was below 10 mM (5 mM for Ala5) whereas the morphology study and IR study

were performed at 100 mM (10 mM for Ala5). And at such concentrations, some of those who formed ribbons at 100mM remained solution. e.g. m-2-m (AcAla 3 and 4)$_2$ with m = 16,18,20.

Some of the CD spectra are shown below for those solutions, principally showing random structure at solution state.

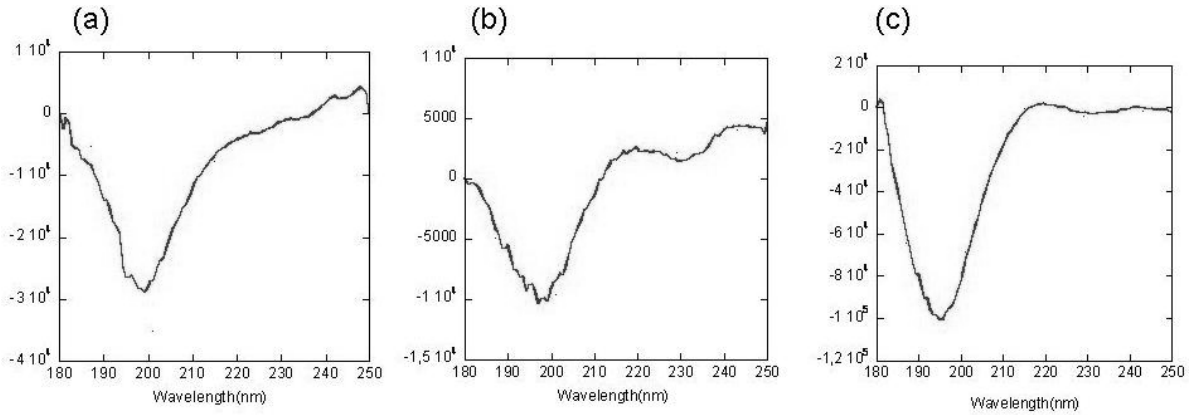

Fig. S5 CD spectra of (a) 18-2-18(AcAla$_3$)$_2$ 10 mM, (b) 16-2-16 (AcAla$_4$)$_2$ 10mM, and (c) 10-2-10 (AcAla$_4$)$_2$ 5 mM. The unit of y axis is deg. cm^2 / dmol

CD spectra of peptides

CD spectra of peptides AcAla$_p$Na are shown below. Concentration was fixed to 8 and 10 mM to avoid saturation for Ala5 which aggregates at this concentration. AcAla$_3$Na at both concentrations remain clear solution, and CD signal show (a) mainly random structure as revealed from the negative peak at around 195 nm. For the AcAla$_5$Na peptides, (c) the inversion of the signal at 195 nm was observed indicating β sheet structure although it does not exactly fit the expected spectra (this may be because these very short peptides may not produce the same spectra form expected from long peptides) as it is confirmed by FT-IR as β sheet structure (see the table above). For the AcAla$_4$Na peptides, interesting concentration dependent behaviour was observed: (b) at 8mM, mainly random structure was observed whereas at 10 mM, some β sheet structure started to form. This result was confirmed by FT-IR spectra at 100 mM which shows the presence of β sheet.

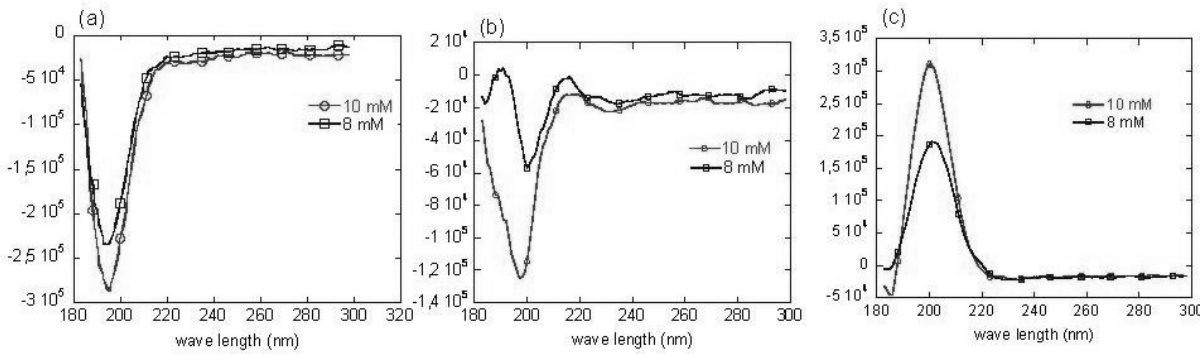

Fig. S6 CD spectra of AcAlap 8 and 10 mM. p = (a) 3, (b) 4 and (c) 5.