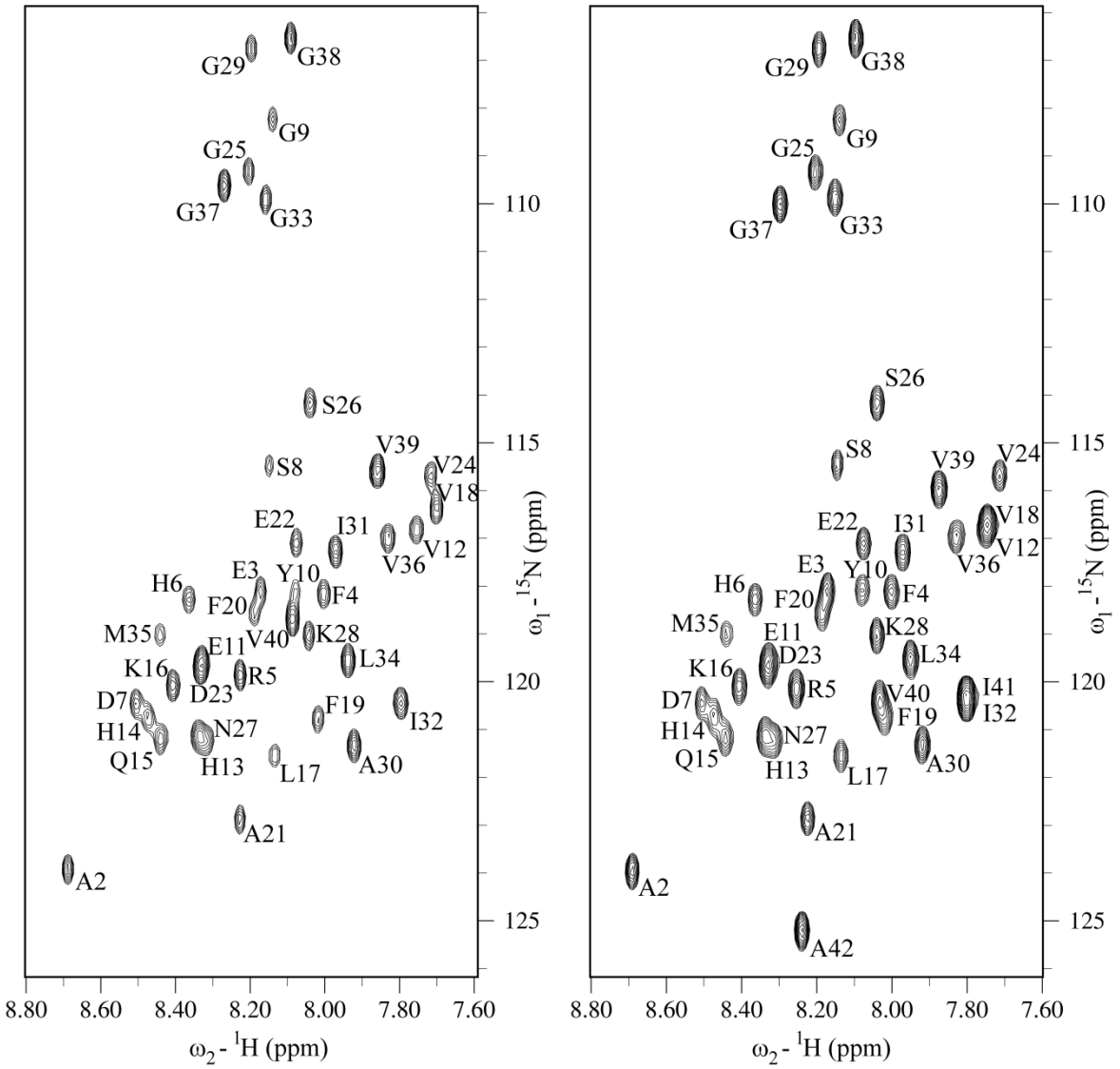
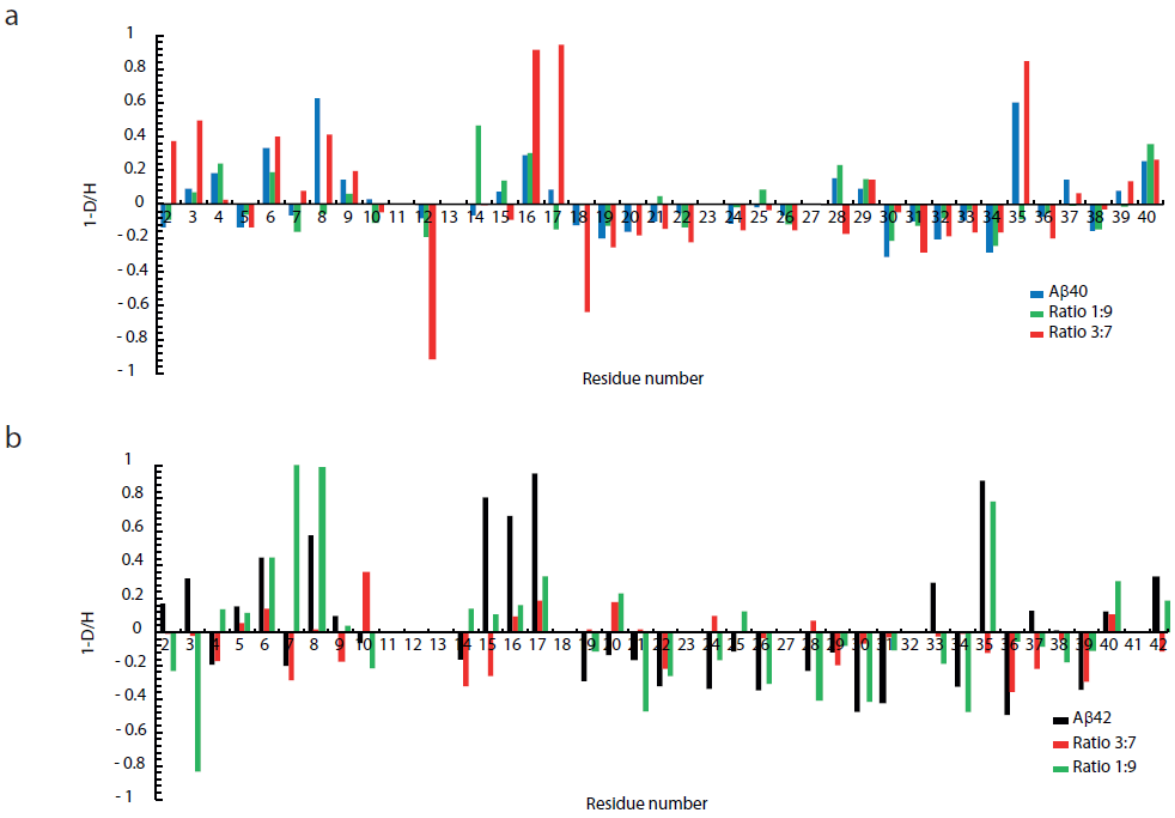
SUPPLEMENTAL DATA



Supplemental Fig. S1. HSQC spectra obtained after redissolving Aβ fibrils in perdeuterated dimethylsulfoxide containing 0.01% deuterated trifluoroacetic acid. *A.* HSQC spectrum of ^{15}N-labeled monomeric $A\beta_{40}$ obtained from resolubilized fully protonated amyloid fibrils. The cross-peaks of residues 11/23 and 13/27 overlap in the $A\beta_{40}$ spectrum. *B.* HSQC spectrum of ^{15}N-labeled monomeric $A\beta_{42}$ obtained from fully protonated fibrils. For $A\beta_{42}$, there are (partial) overlaps in the cross-peaks of residues 11/23, 12/18, 13/27, 19/40 and 32/41. The sequence-specific assignment of the peptide backbone amides is indicated next to each cross-peak. The presence of the unlabeled Aβ alloform in the mixtures does not influence the spectra as expected for monomeric peptides, since the cross-peaks of labeled peptides in the 1:9 and 3:7 ratios display identical chemical shifts. For the HDX interpretation, the intensities of the peaks are determined by the height of the spectral signals.



Supplemental Fig. S2. Atomic resolution HDX using NMR reveals that small differences at atomic level may be present between fibrillar Aβ ratios. The protection factor of each residue is derived from comparing the corresponding peak intensities observed under fully protonated conditions to the signal intensities after exchange with D_2O. The results are shown as bar plots. In these plots no data is available for residues 11, 12, 13, 18, 19, 23, 27, 32, 40 and 41 for $A\beta_{42}$, and 11, 13, 23 and 27 for $A\beta_{40}$ due to (partial) overlapping peaks (and therefore ambiguous N-H cross-peak assignments). A) signals for the ^{15}N-labeled $A\beta_{40}$ alloform in the ratios; B) signals for the ^{15}N-labeled $A\beta_{42}$ alloform.

Supplementary Table 1. Aβ oligomer population patterns as derived from mass spectrometry analysis. Three different incubation times were analyzed for each ratio and the detected molecular assemblies (kDa) are reported in the tables.

# Aβ	Pure $Aβ_{40}$			Ratio 1:9			Ratio 3:7			Pure $Aβ_{42}$		
	1h	**3h**	**6h**	**1h**	**3h**	**6h**	**1h**	**3h**	**6h**	**1h**	**3h**	**6h**
1	4.36	4.32	4.35	4.43	4.44	4.46	4.46	4.47	4.46	4.45	4.45	4.46
2	8.73	8.75	8.72	8.90	8.94	8.91	8.96	8.92	8.95	8.94	8.92	8.98
3	13.07	13.06	13.08	13.28	13.53	13.57	13.57	13.58	13.56	13.37	13.39	13.55
4	17.42	17.43	17.41	17.72	18.08	18.10	18.10	18.09	18.08	17.79	17.77	17.70
5	21.83	21.83	21.84		22.68	22.67	22.67	22.67	22.67	22.24	22.22	22.26
6	26.28	26.29	26.29		27.13	27.12	27.12	27.12	27.13	26.69	26.67	26.65
7	30.57	30.56	30.58		31.58			31.17	31.17	31.21	31.22	31.20
8	34.91	34.91	34.92		36.02			35.57	35.57	35.99	35.98	35.95
9	39.29	39.30	39.30					40.02	40.05	40.45	40.49	40.45
10	43.69	43.68	43.70					44.49		44.92	44.89	44.88
11	47.91	47.92	47.92					48.99		49.36	49.31	49.29
12	52.23	52.24	52.24					53.61		53.73	53.76	
13	56.64	56.57	56.66					58.13		58.20	58.18	
14		60.91	61.01					62.72			62.63	
15		65.25	65.30					66.70			65.32	
16		69.65	69.61									
17		73.92	73.98									
18			78.33									
19			82.67									
20			87.03									
21			91.39									
22			95.77									
23			99.97									
24			104.32									
25			108.67									
Higher Mass Range												
		156	186			172		156			156	187
			348			355		315			253	
			714			515		471				
			852					626				