Supplementary Material

NMR Experiments:

In the HNCACB experiment $98 \times 32 \times 512$ complex points were acquired, with acquisition times of 9.6 ms, 17.8 ms, and 64.0 ms in t_1, t_2, and t_3 respectively, and 24 scans per increment, using a 1 s delay between scans, resulting in a total acquisition time of approximately 91 hours. The CBCA(CO)NH experiment was acquired with $60 \times 32 \times 512$ complex points, with acquisition times of 5.8 ms, 17.8 ms , and 64.0 ms in t_1, t_2, and t_3, respectively, and 32 scans per increment, using a 1 s delay between scans, resulting in a total acquisition time of approximately 74 hours. In the HNCO experiment $64 \times 32 \times 512$ complex points were acquired, with acquisition times of 21.2 ms, 17.8 ms, and 64.0 ms in t_1, t_2, and t_3, respectively, and 16 scans per increment, using a 1 s delay between scans, resulting in a total acquisition time of approximately 40 hours.

The H(CCO)NH-TOCSY experiment was acquired with $128 \times 32 \times 512$ complex points, with acquisition times of 16.0 ms, 17.8 ms , and 64.0 ms in t_1, t_2, and t_3, respectively, and 16 scans per increment, using a 1 s delay between scans, resulting in a total acquisition time of approximately 80 hours. The (H)C(CO)NH-TOCSY experiment was acquired with $96 \times 32 \times 512$ complex points, with acquisition times of 10.0 ms, 17.8 ms , and 64.0 ms in t_1, t_2, and t_3, respectively, and 24 scans per increment, using a 1 s delay between scans, resulting in a total acquisition time of approximately 89 hours. The carbon isotropic mixing time in both TOCSY experiments was 20.8 ms.

The (HB)CB(CGCD)HD experiment was acquired with 38×512 complex points, with acquisition times of 7.9 ms and 64.0 ms in t_1 and t_2, respectively, and 320 scans per increment, using a 0.9 s delay between scans, resulting in a total acquisition time of approximately 7 hours. The (HB)CB(CGCDCE)HE experiment was acquired with 76×512 complex points, with acquisition times of 7.9 ms and 64.0 ms in t_1 and t_2, respectively, and 320 scans per increment, using a 0.9 s delay between scans, resulting in a total acquisition time of approximately 14 hours.