## **Supplementary Materials**

PRO 1	HA	HIS 2	Н
HIS 2	HA	ASN 3	Н
ASN 3	HA	PRO 4	HD2
ASN 3	HA	PRO 4	HD3
ASN 3	HB2	PRO 4	HD3
PRO 4	HA	GLY 5	Н
GLY 5	Н	TYR 6	Н
TYR 6	HA	PRO 7	HD2
TYR 6	HA	PRO 7	HD3
PRO 7	HA	HIS 8	Н
HIS 8	HA	ASN 9	Н
HIS 8	HB2	ASN 9	Н
HIS 8	HB3	ASN 9	Н
ASN 9	HA	PRO 10	HD2
ASN 9	HA	PRO 10	HD3
ASN 9	HB2	PRO 10	HD2
ASN 9	HB3	PRO 10	HD2
PRO 10	HA	GLY 11	Н
GLY 11	Н	TYR 12	Н
PRO 1	HB2	PRO 1	HA
PRO 1	HB3	PRO 1	HA
HIS 2	HA	HIS 2	HB2
HIS 2	HD2	HIS 2	HE1
HIS 2	Н	HIS 2	HA
HIS 2	Н	HIS 2	HB2
HIS 2	Н	HIS 2	HB3
ASN 3	HA	ASN 3	HB2
ASN 3	HA	ASN 3	HB3
ASN 3	Н	ASN 3	HA
ASN 3	Н	ASN 3	HB2
ASN 3	Н	ASN 3	HB3
PRO 4	HB2	PRO 4	HD2
PRO 4	HB2	PRO 4	HD3
PRO 4	HB3	PRO 4	HA
PRO 4	HB3	PRO 4	HB2
PRO 4	HB3	PRO 4	HD2
PRO 4	HB3	PRO 4	HD3
PRO 4	HD2	PRO 4	HA
PRO 4	HD3	PRO 4	HD2
TYR 6	Н	TYR 6	HA
TYR 6	Н	TYR 6	HB2
TYR 6	Н	TYR 6	HB3

**Table S1.** Unambiguous inter-residues and intra-residues NOEs used as structural restraints for the ensemble calculation of 2-HexaPY peptide in  $H_2O/D_2O$  (90:10). NOEs were treated as described in the Experimental section.

PRO 1HAHIS 2HPRO 7HB3PRO 7HAPRO 7HB2PRO 7HAPRO 7HB3PRO 7HD3PRO 7HD2PRO 7HD3HIS 8HAHIS 8HB2HIS 8HAHIS 8HD2HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HB2HIS 8HB4HIS 8HB2HIS 8HB5HB4HIS 8HIS 8HB7HIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2ASN 9HAASN 9HB3ASN 9HAASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HB3PRO 10HB3PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2TYR 12HATYR 12HB3						
PRO 7HB2PRO 7HAPRO 7HB3PRO 7HD3PRO 7HD2PRO 7HD3HIS 8HAHIS 8HB2HIS 8HAHIS 8HD2HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HDHIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HB2QLY 11HGLY 11HA1GLY 11HGLY 11HA2	PRO 1	HA	HIS 2	Н		
PRO 7HB3PRO 7HD3PRO 7HD2PRO 7HD3HIS 8HAHIS 8HB2HIS 8HAHIS 8HD2HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HD2HIS 8HE1HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2ASN 9HAASN 9HB3ASN 9HAASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	PRO 7	HB3	PRO 7	HA		
PRO 7 HD2 PRO 7 HD3   HIS 8 HA HIS 8 HB2   HIS 8 HA HIS 8 HD2   HIS 8 HB2 HIS 8 HD2   HIS 8 HB2 HIS 8 HD2   HIS 8 HB3 HIS 8 HD2   HIS 8 HD2 HIS 8 HE1   HIS 8 HD2 HIS 8 HE2   HIS 8 H HIS 8 HB2   HIS 8 H HIS 8 HB2   HIS 8 H HIS 8 HB2   ASN 9 HA ASN 9 HB3   ASN 9 HA ASN 9 HB3   ASN 9 H ASN 9 HB3   PRO 10 HB2 PRO 10 HA   PRO 10 HB2 PRO 10 HA   PRO 10 HB3 PRO 10 HA   PRO 10 HB3 PRO	PRO 7	HB2	PRO 7	HA		
HIS 8HAHIS 8HB2HIS 8HAHIS 8HD2HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HD2HIS 8HE1HIS 8HDHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2ASN 9HAASN 9HB3ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	PRO 7	HB3	PRO 7	HD3		
HIS 8HAHIS 8HD2HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HDHIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	PRO 7	HD2	PRO 7	HD3		
HIS 8HB2HIS 8HD2HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HD2HIS 8HE1HIS 8HDHIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2HIS 8HHIS 8HB2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HA	HIS 8	HB2		
HIS 8HB3HIS 8HB2HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HD2HIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB3PRO 10HAPRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HA	HIS 8	HD2		
HIS 8HB3HIS 8HD2HIS 8HD2HIS 8HE1HIS 8HD2HIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HA3PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HB2	HIS 8	HD2		
HIS 8HD2HIS 8HE1HIS 8HHIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HA3PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HB3	HIS 8	HB2		
HIS 8HHIS 8HAHIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HAASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HA3PRO 10HB2PRO 10HA3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HD4PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HB3	HIS 8	HD2		
HIS 8HHIS 8HB2HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HAASN 9HASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3PRO 10HB2PRO 10HA3PRO 10HB2PRO 10HA3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HD4PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	HD2	HIS 8	HE1		
HIS 8HHIS 8HB3HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB3ASN 9HASN 9HB2ASN 9HASN 9HB3PRO 10HB2PRO 10HA3PRO 10HB2PRO 10HA3PRO 10HB3PRO 10HD3PRO 10HB4PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB4PRO 10HB4GLY 11HGLY 11HA1GLY 11HGLY 11HA2	HIS 8	Н	HIS 8	HA		
HIS 8HHIS 8HD2ASN 9HAASN 9HB2ASN 9HAASN 9HB3ASN 9HASN 9HAASN 9HASN 9HB2ASN 9HASN 9HB2ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HD3PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA2	HIS 8	Н	HIS 8	HB2		
ASN 9 HA ASN 9 HB2   ASN 9 HA ASN 9 HB3   ASN 9 H ASN 9 HA   ASN 9 H ASN 9 HA   ASN 9 H ASN 9 HB2   ASN 9 H ASN 9 HB2   ASN 9 H ASN 9 HB3   PRO 10 HB2 PRO 10 HA   PRO 10 HB2 PRO 10 HD3   PRO 10 HB3 PRO 10 HD2   PRO 10 HB3 PRO 10 HD3   PRO 10 HB3 PRO 10 HD2   PRO 10 HB3 PRO 10 HD3   PRO 10 HB3 PRO 10 HD3   PRO 10 HB3 PRO 10 HD3   PRO 10 HD2 PRO 10 HB2   GLY 11 H GLY 11 HA1   GLY 11 H GLY 11 HA2	HIS 8	Н	HIS 8	HB3		
ASN 9 HA ASN 9 HB3   ASN 9 H ASN 9 HA   ASN 9 H ASN 9 HB2   ASN 9 H ASN 9 HB3   PRO 10 HB2 PRO 10 HA   PRO 10 HB2 PRO 10 HA3   PRO 10 HB2 PRO 10 HD3   PRO 10 HB3 PRO 10 HD4   PRO 10 HB3 PRO 10 HD3   PRO 10 HB3 PRO 10 HD3   PRO 10 HB3 PRO 10 HD4   PRO 10 HB3 PRO 10 HD3   PRO 10 HD2 PRO 10 HB2   GLY 11 H GLY 11 HA2	HIS 8	Н	HIS 8	HD2		
ASN 9HASN 9HAASN 9HASN 9HB2ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HD3PRO 10HB3PRO 10HD4PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB4PRO 10HB4GLY 11HGLY 11HA1GLY 11HGLY 11HA2	ASN 9	HA	ASN 9	HB2		
ASN 9HASN 9HB2ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HD3PRO 10HB3PRO 10HAPRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB4PRO 10HD3PRO 10HD5PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	ASN 9	HA	ASN 9	HB3		
ASN 9HASN 9HB3PRO 10HB2PRO 10HAPRO 10HB2PRO 10HD3PRO 10HB3PRO 10HAPRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HD3PRO 10HB3PRO 10HB3PRO 10HB3PRO 10HB2GLY 11HGLY 11HA2	ASN 9	Н	ASN 9	HA		
PRO 10HB2PRO 10HAPRO 10HB2PRO 10HD3PRO 10HB3PRO 10HAPRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA2	ASN 9	Н	ASN 9	HB2		
PRO 10HB2PRO 10HD3PRO 10HB3PRO 10HAPRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	ASN 9	Н	ASN 9	HB3		
PRO 10HB3PRO 10HAPRO 10HB3PRO 10HD2PRO 10HB3PRO 10HD3PRO 10HD2PRO 10HB2GLY 11HGLY 11HA1GLY 11HGLY 11HA2	PRO 10	HB2	PRO 10	HA		
PRO 10   HB3   PRO 10   HD2     PRO 10   HB3   PRO 10   HD3     PRO 10   HD2   PRO 10   HB2     GLY 11   H   GLY 11   HA1     GLY 11   H   GLY 11   HA2	PRO 10	HB2	PRO 10	HD3		
PRO 10   HB3   PRO 10   HD3     PRO 10   HD2   PRO 10   HB2     GLY 11   H   GLY 11   HA1     GLY 11   H   GLY 11   HA2	PRO 10	HB3	PRO 10	HA		
PRO 10   HD2   PRO 10   HB2     GLY 11   H   GLY 11   HA1     GLY 11   H   GLY 11   HA2	PRO 10	HB3	PRO 10	HD2		
GLY 11   H   GLY 11   HA1     GLY 11   H   GLY 11   HA2	PRO 10	HB3	PRO 10	HD3		
GLY 11 H GLY 11 HA2	PRO 10	HD2	PRO 10	HB2		
	GLY 11	Н	GLY 11	HA1		
TYR 12 HA TYR 12 HB3	GLY 11	Н	GLY 11	HA2		
	TYR 12	HA	TYR 12	HB3		

Table S1. Cont.