Table S1. MALDI-TOF mass-spectrometry analysis of **LB1**-peptides and their PNA705 conjugates synthesized using SELPEPCON

Peptide	Peptide mass found (*m*/*z*)[a]	Peptide mass calculated (*m*/*z*)	Conjugate[b] mass found (*m*/*z*)[a]	Conjugate mass calculated (*m*/*z*)
LB1_1	2460	2464	7711	7714
LB1_2	2461	2464	7716	7714
LB1_3	2462	2464	7716	7714
LB1_4	2153	2154	7406	7405
LB1_5	2478	2479	7732	7729
LB1_6	2479	2480	7735	7730
LB1_7	2536	2537	7793	7787
LB1_8	2447	2448	7703	7698
LB1_9	2350	2352	7599	7602
LB1_10	2384	2388	7634	7638
LB1_11	2186	2187	7435	7437
LB1_12	2354	2356	7606	7606
LB1_13	2469	2471	7723	7721
LB1_14	2470	2471	7723	7721
LB1_15	2470	2471	7722	7721
LB1_16	2469	2471	7721	7721

[a] Main signal as found by MALDI-TOF mass- spectrometry.

[b] Mass after conjugation with N_3-PNA(705)-S-S-biotin and workup providing the **LB1**-PNA705-SH conjugate containing a triazole linker between the peptide and the PNA as shown in **Schemes 1 - 3**.

Table S2a. Sequences and MALDI-TOF mass-spectrometry analysis of **LB2** CPP-library containing a *N*-terminal alkyne linker and conjugation to N_3-PNA705-S-S-biotin.

Peptide	Sequence[a]	Peptide mass (*m/z*)[b]		Conjugate mass (*m/z*)[c]	
		Found	Calculated	Found	Calculated
LB2_1	GWTLNSAGYLLGPHIDNHRSFHDKYGLA	3223	3220	8472	8470
LB2_2	GALFLGFLGAAGSTMGAWSQPKKKRKV	2891	2886	8156*	8137
LB2_3	GWTLNSAGYLLGKINLKALAALAKKIL	2918	2921	-	8171
LB2_4	GALFLFWLGAAGSTMGAWSQPKKKRKV	3012	3016	8262	8266
LB2_5	MGLGLHLLVLAAALQGAWSQPKKKRKV	2990	2994	8241	8244
LB2_6	RRRRRRRQIKIWFQNRRMKWKKGG	3383	3377	8650**	8627
LB2_7	RXRRXRRXRIKILFQNRRMKWKK	3251	3245	8519**	8495
LB2_8	RXRRBRRXRILFQYRXRBRXRB	2989	2989	8249	8240
LB2_9	RXRRBRRXRYQFLIRXRBRXRB	2990	2989	8249	8240
LB2_10	MVTVLFRRLRIRRACGPPRVRV	2819	2820	8094**[d]	7982
LB2_11	AGYLLGKINLKALAALAKKIL	2262	2262	7518	7512
LB2_12	KETWWETWWTETSQPKKKRKV	2925	2927	8176	8178
LB2_13	KETWFETWFTEWSQPKKKRKV	2847	2849	8099	8100
LB2_14	VTVLFRRLRIRRCSGPPRVR	2603	2605	7855**[d]	7767
LB2_15	GLWRALWRLLRSLWRLLWRA	-	2701	-	7951
LB2_16	CQSWQQGWCSLGPIHAHLDR	2576	2578	-	7652
LB2_17	HGLASTLTRWAHYNALIRAF	2375	2378	7624	7628
LB2_18	HGLASTLTRWAHYNALIRAF	2378	2378	7625	7628
LB2_19	AGYLLGKINLKALAALAKKI	2146	2149	7400	7399
LB2_20	GYLLGKINLKALAALAKKIL	2189	2191	7444	7441
LB2_21	VTVLFRRLRIRRASGPPRVR	2484	2485	7739	7735
LB2_22	MPGEPRRANVMAHKLEPASL	2282	2284	7538	7534
LB2_23	PQRDTVGGRTTPPSWGPAKA	2158	2158	7414	7409
LB2_24	AGYLLGKLKALAALAKKIL	2034	2020	7288	7270
LB2_25	KMTRAQRRAAARRNRWTAR	2432	2436	7680	7686
LB2_26	RGGRLSYSRRRFSTSTGR	2178	2179	7424	7430
LB2_27	KLALKLALKALKAALKLA	1955	1957	7210	7207
LB2_28	RQIKILFQNRRMKWKKGG	2381	2367	7635	7617
LB2_29	VRLPPPVRLPPPVRLPPP	2074	2077	7327	7327
LB2_30	LGISYGRKKRRQRRRPPQ	2330	2332	7583	7582
LB2_31	LLIILRRRIRKQAHAHSK	2287	2289	7542	7539
LB2_32	IAWVKAFIRKLRKGPLG	2032	2033	7287	7283

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus.

[b] Main signal as found by MALDI-TOF mass- spectrometry. Note that cysteines contain StBu protection group.

[c] Mass after conjugation with N_3-PNA(705)-S-S-biotin and workup providing the **LB2**-PNA705-SH conjugate containing a triazole linker between the peptide and the PNA as shown in **Schemes 1** - **3**. Main signal contains (*) oxidized methionine, (**) sodium

[d] The mass spectrum contains a statistical mixture of peaks corresponding to StBu adducts corresponding to the amount of cysteines; the major observed signal is shown

Table S2b. Sequences and MALDI-TOF mass-spectrometry analysis of **LB2** CPP-library containing a *N*-terminal alkyne linker and conjugation to N_3-PNA705-S-S-biotin.

Peptide	Sequence[a]	Peptide mass (*m/z*)[b]		Conjugate mass (*m/z*)[c]	
		Found	Expected	Found	Expected
LB2_33	RBRRXRILFQYRXRRBR	2379	2380	7635	7630
LB2_34	RXRRXRILFQYRXRRXR	2464	2464	7720	7714
LB2_35	RQIKIWFQNRRMKWKK	2341	2326	7597**	7576
LB2_36	FFLIPKGRRRRRRRRR	2305	2306	7560	7556
LB2_37	AAKKAAKKAAKKAAKK	1690	1691	6943	6941
LB2_38	RQPKIWFPNRRKPWKK	2242	2245	7494	7495
LB2_39	SDLWEMMMVSLASQY	-	1870	-	7120
LB2_40	GEAHIPTSEMREKGW	1820	1807	7070**	7057
LB2_41	KWFETWFTEWPKKRK	2174	2177	7427	7427
LB2_42	RKKLWTPPKAKKWK	1873	1874	7127	7125
LB2_43	RXRRXRRXRRXRXB	1983	1983	7238	7233
LB2_44	GKKAFKEAEKGFKK	1675	1675	6930	6925
LB2_45	GHKALKLAAKLLHH	1616	1616	6872	6866
LB2_46	GKKALKLAAKLLKK	1589	1589	6846	6839
LB2_47	RXRRBRRXRRBRXB	1899	1899	7158	7149
LB2_48	RRRRNTRRNRRRVR	2086	2087	7334	7338
LB2_49	TRRQRTRRARRNR	1862	1862	7110	7112
LB2_50	KMDCRWRWKCCKK	2115	2115	7366[d]	7101
LB2_51	INLKKLAKLKKIL	1602	1602	6860	6852
LB2_52	GRKKRRQRRRPPQ	1797	1798	7055	7048
LB2_53	GPFHFYQFLFPPV	1674	1675	6933	6925
LB2_54	GSPWGLQHHPPRT	1548	1549	6805	6799
LB2_55	PIRRRKKLRRLK	1670	1699	6951	6949
LB2_56	RRQRRTSKLMKR	1695	1695	6947	6945
LB2_57	WWWWRRRRRRRR	2092	2091	7343	7342
LB2_58	RRRRWWWWRRRR	2092	2091	7343	7342
LB2_59	RWRRWRRWRRWR	2092	2091	7342	7342
LB2_60	ALSSSPSKHCG	1239	1240	6489**[d]	6402
LB2_61	YARAAARQARA	1283	1283	6537	6534
LB2_62	RRRRRRRRFF	1798	1797	7053	7047
LB2_63	RRRRFFRRRR	1798	1797	7055	7047
LB2_64	ARWRWKAAKK	1379	1380	6636	6630

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus.

[b] Main signal as found by MALDI-TOF mass- spectrometry. Note that cysteines contain StBu protection group.

[c] Mass after conjugation with N_3-PNA(705)-S-S-biotin and workup providing the **LB2**-PNA705-SH conjugate containing a triazole linker between the peptide and the PNA as shown in **Schemes 1** - **3**. Main signal contains (*) oxidized methionine, (**) sodium

[d] The mass spectrum contains a statistical mixture of peaks corresponding to StBu adducts corresponding to the amount of cysteines; the major observed signal is shown

Table S2c. Sequences and MALDI-TOF mass-spectrometry analysis of **LB2** CPP-library containing an *N*-terminal alkyne linker and conjugation to N_3-PNA705-S-S-biotin.

Peptide	Sequence[a]	Peptide mass (*m/z*)[b]		Conjugate mass (*m/z*)[c]	
		Found	Expected	Found	Expected
LB2_65	TQIENLKEKG	1238	1238	6495	6489
LB2_66	KFFKFFKFFK	1492	1493	6750	6743
LB2_67	SRWRWKSSKK	1426	1428	6684	6678
LB2_68	CRWRWKCCKK	1740	1740	6991[d]	6726
LB2_69	RRLSYSRRRF	1475	1476	6726	6726
LB2_70	RRRRRRRRR	1500	1503	6759	6753
LB2_71	RWRWKXXKK	1391	1393	6647	6643
LB2_72	RRRRRRFF	1327	1329	6582	6579
LB2_73	GRKKRRQR	-	1163	-	6414
LB2_74	NFKFGLSS	978	978	6231	6228
LB2_75	KKKKKKKK	-	1122	-	6373
LB2_76	PKKKRKV	962	962	-	6212
LB2_77	FFLIPKG	900	900	6154	6150
LB2_78	FLFLFL	-	878	-	6128

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus.

[b] Main signal as found by MALDI-TOF mass- spectrometry. Note that cysteines contain StBu protection group.

[c] Mass after conjugation with N_3-PNA(705)-S-S-biotin and workup providing the **LB2**-PNA705-SH conjugate containing a triazole linker between the peptide and the PNA as shown in **Schemes 1** - **3**. Main signal contains (*) oxidized methionine, (**) sodium

[d] The mass spectrum contains a statistical mixture of peaks corresponding to StBu adducts corresponding to the amount of cysteines; the major observed signal is shown

Table S3a. Yields of parallel peptide and PNA705-conjugate synthesis of **LB2**-CPP-Library.

Peptide[a]	Length	Number of Arg's	Net charge	Successful peptide synthesis[b]	Yield peptide (μmol)[c]	Successful conjugation[d]	Yield conjugate (nmol)[e]
LB2_1	28	1	+4	Y	4.4	Y*	12.1
LB2_2	27	1	+5	Y	3.2	Y*	14.5
LB2_3	27	0	+4	Y	2.3	N	-
LB2_4	27	1	+5	Y	2.9	Y*	16.7
LB2_5	27	1	+5	Y	3.1	Y*	19.4
LB2_6	24	9	+13	Y	4.8	Y	18.7
LB2_7	23	8	+12	Y	3.0	Y	20.1
LB2_8	22	10	+10	Y	5.0	Y	18.5
LB2_9	22	10	+10	Y	4.9	Y	27.9
LB2_10	22	7	+7	Y	4.7	Yx	16.1
LB2_11	21	0	+4	Y	6.1	Y*	13.8
LB2_12	21	1	+3	Y	2.1	Y	18.6
LB2_13	21	1	+3	Y	2.3	Y	16.7
LB2_14	20	7	+7	Y	4.7	Yx	21.2
LB2_15	20	5	+5	N	-	N	-
LB2_16	20	1	+2	Y	3.7	N	-
LB2_17	20	2	+2	Y	4.3	Y	19.6
LB2_18	20	2	+2	Y	3.1	Y	20.3
LB2_19	20	0	+4	Y	3.6	Y	21.7
LB2_20	20	0	+4	Y	3.6	Y	17.2
LB2_21	20	7	+7	Y	3.5	Y	21.8
LB2_22	20	2	+1	Y	3.9	Y	21.7
LB2_23	20	2	+4	Y	4.3	Y	20.1
LB2_24	19	0	+3	Y	2.6	Y*	13.6
LB2_25	19	7	+8	Y	4.2	Y	22.4
LB2_26	18	6	+6	Y*	3.9	Y	19.3
LB2_27	18	0	+5	Y	3.2	Y	20.1
LB2_28	18	3	+7	Y	4.0	Y	21.6
LB2_29	18	3	+3	Y	4.9	Y	20.7
LB2_30	18	6	+8	Y	2.9	Y	19.8
LB2_31	18	4	+6	Y	3.3	Y*	20.5
LB2_32	17	2	+5	Y	3.5	Y	21.2

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus. See **Table S2** for sequences. Length = number of amino acids

[b] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant impurities.

[c] Crude yield from a 5 μmol synthesis after Oasis HLB cartridge workup. Based on crude weight and corrected for TFA salts.

[d] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant amount of impurities (In most cases low conversion to the conjugate resulting in significant amount of unconjugated PNA Conjugates indicated with an "x" contain cysteines that are modified with *t*BuS-groups.

[e] From a 30 nmol reaction based on N_3-PNA705-S-S-biotin, determined by UV absorption at 260 nm.

Table S3b. Yields of parallel peptide and PNA705-conjugate synthesis of **LB2**-CPP-Library.

Peptide[a]	Length	Number of Arg's	Net charge	Successful peptide synthesis[b]	Yield peptide (μmol)[c]	Successful conjugation[d]	Yield conjugate (nmol)[e]
LB2_33	17	8	+8	Y	3.5	Y	22.1
LB2_34	17	8	+8	Y	3.7	Y	19.9
LB2_35	16	3	+7	Y	3.0	Y	22.7
LB2_36	16	9	+10	Y	1.0	Y	20.1
LB2_37	16	0	+8	Y*	1.8	Y	18.4
LB2_38	16	9	+7	Y	2.5	Y	21.4
LB2_39	15	0	0	N	-	N	-
LB2_40	15	1	-1	Y*	3.1	Y	19.7
LB2_41	15	1	+3	Y	1.9	Y	21.1
LB2_42	14	1	+7	Y	2.0	Y	21.5
LB2_43	14	8	+8	Y	3.9	Y	20.7
LB2_44	14	0	+4	Y	2.6	Y	21.5
LB2_45	14	0	+3	Y	2.3	Y	21.0
LB2_46	14	0	+6	Y	2.0	Y	19.2
LB2_47	14	8	+8	Y	3.4	Y	18.5
LB2_48	14	10	+10	Y	3.6	Y	13.9
LB2_49	13	8	+8	Y	3.5	Y	7.5
LB2_50	13	2	+7	Y	2.4	Yx	18.0
LB2_51	13	0	+5	Y*	2.3	Y	22.4
LB2_52	13	6	+8	Y	2.7	Y	16.7
LB2_53	13	0	0	Y	0.7	Y	21.0
LB2_54	13	1	+1	Y	2.9	Y	23.1
LB2_55	12	5	+8	Y	4.0	Y	17.0
LB2_56	12	5	+7	Y	4.3	Y	12.8
LB2_57	12	8	+8	Y	2.3	Y	15.0
LB2_58	12	8	+8	Y	4.5	Y	21.3
LB2_59	12	8	+8	Y	2.7	Y	17.9
LB2_60	11	0	+1	Y	6.7	Y	20.4
LB2_61	11	3	+3	Y	3.2	Y	21.5
LB2_62	11	9	+9	Y	2.2	Y	21.8
LB2_63	11	9	+9	Y	2.7	Y	24.3
LB2_64	10	2	+5	Y	2.7	Y	22.7

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus. See **Table S2** for sequences.

[b] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant impurities.

[c] Crude yield from a 5 μmol synthesis after Oasis HLB cartridge workup. Based on crude weight and corrected for TFA salts.

[d] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant amount of impurities (In most cases low conversion to the conjugate resulting in significant amount of unconjugated PNA Conjugates indicated with an "x" contain cysteines that are modified with *t*BuS-groups.

[e] From a 30 nmol reaction based on N_3-PNA705-S-S-biotin, determined by UV absorption at 260 nm.

Table S3c. Yields of parallel peptide and PNA705-conjugate synthesis of **LB2**-CPP-Library.

Peptide[a]	Length	Number of Arg's	Net charge	Successful peptide synthesis[b]	Yield peptide (μmol)[c]	Successful conjugation[d]	Yield conjugate (nmol)[e]
LB2_65	10	0	0	Y	4.7	Y	20.5
LB2_66	10	0	+4	Y	3.7	Y	15.7
LB2_67	10	2	+5	Y	2.8	Y	23.6
LB2_68	10	2	+5	Y	3.5	Yx	25.7
LB2_69	10	5	+5	Y	5.5	Y	24.2
LB2_70	9	9	+9	Y	1.9	Y	17.2
LB2_71	9	2	+5	Y	3.2	Y	17.9
LB2_72	8	6	+6	Y	2.8	Y	17.2
LB2_73	8	4	+6	N	-	N	-
LB2_74	8	0	+1	Y	3.4	Y	17.5
LB2_75	8	0	+8	N	-	N	-
LB2_76	7	1	+5	Y	3.3	Y*	8.5
LB2_77	7	0	+1	Y	2.6	Y	18.0
LB2_78	6	0	0	N	-	N	-

[a] All peptides contain a *C*-terminal amide and a 4-pentynoic acid group coupled to the *N*-terminus. See **Table S2** for sequences.

[b] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant impurities.

[c] Crude yield from a 5 μmol synthesis after Oasis HLB cartridge workup. Based on crude weight and corrected for TFA salts.

[d] Main product was identified by MALDI-TOF mass-spectroscopy, Y = Yes, N = No, * = significant amount of impurities (In most cases low conversion to the conjugate resulting in significant amount of unconjugated PNA Conjugates indicated with an "x" contain cysteines that are modified with *t*BuS-groups.

[e] From a 30 nmol reaction based on N_3-PNA705-S-S-biotin, determined by UV absorption at 260 nm.

Table S4. Library of **LB1** peptides containing a C-terminal alkyne, peptide and conjugate synthesis yield.

Peptide	Sequence (*N* to *C* – term)[a]	Net charge	Number of Arg's	Yield peptide (μmol, (%))[b]	Yield conjugate (nmol, (%))[c]
LB1_17	RXRRXRILFQYRXRRXRZ	+8	8	3.3 (67)	22.3 (74)
LB1_18	RXRRXRILFQYRXRRXRGGZ	+8	8	3.3 (66)	24.4 (82)

[a] All peptides contain a *C*-terminal amide and a free amine on *N*-terminus. Z = Bpg (L-bishomopropargylglycine).

[b] Crude yield from a 5 μmol synthesis after Oasis HLB cartridge workup. Based on crude weight and corrected for TFA salts.

[c] Yield of the **LB1**-PNA705 conjugate obtained via SELPEPCON (**Schemes 1** - **3**) from a 30 nmol scale synthesis based on N_3-PNA705-S-S-biotin, determined by UV absorption at 260 nm.

Table S5. MALDI-TOF mass-spectrometry analysis of C-terminal alkyne containing **LB1**-peptides and their PNA705 conjugates synthesized using SELPEPCON.

Peptide	Peptide mass found (*m*/*z*)[a]	Peptide mass calculated (*m*/*z*)	Conjugate[b] mass found (*m*/*z*)[a]	Conjugate mass calculated (*m*/*z*)
LB1_17	2503	2507	7747	7757
LB1_18	2617	2621	7864	7871

[a] Main signal as found by MALDI-TOF mass- spectrometry.

[b] Mass after conjugation with azido-PNA(705)-S-S-biotin and workup providing the **LB1**-PNA705-SH conjugate containing a triazole linker between the peptide and the PNA as shown in **Schemes 1 - 3**.

Examples of MALDI mass-spectra of parallel-synthesized LB1- and LB2-peptides obtained by SELPEPCON

Below, in **Figures S1** - **S12**, examples are given of MALDI-TOF mass-spectra of **LB1**- and **LB2**-peptides obtained using the SELPEPCON parallel synthesis procedure. **Figures S11** - **S12** show peptides that showed several peaks belonging to impurities.

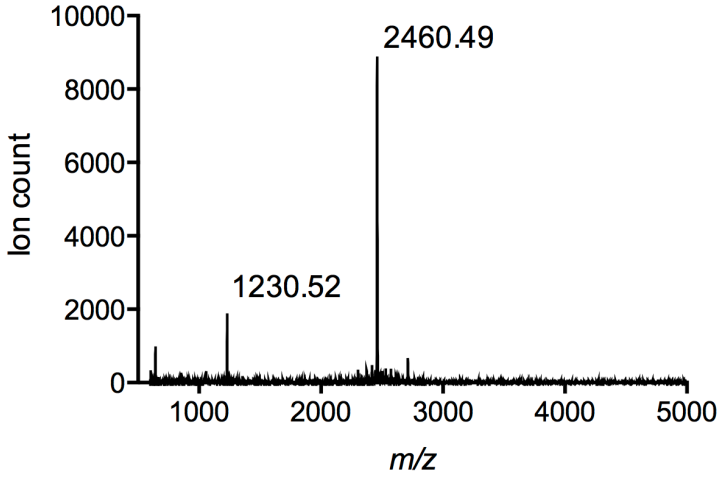

Figure S1. MALDI-TOF mass-spectrum of **LB1_1**

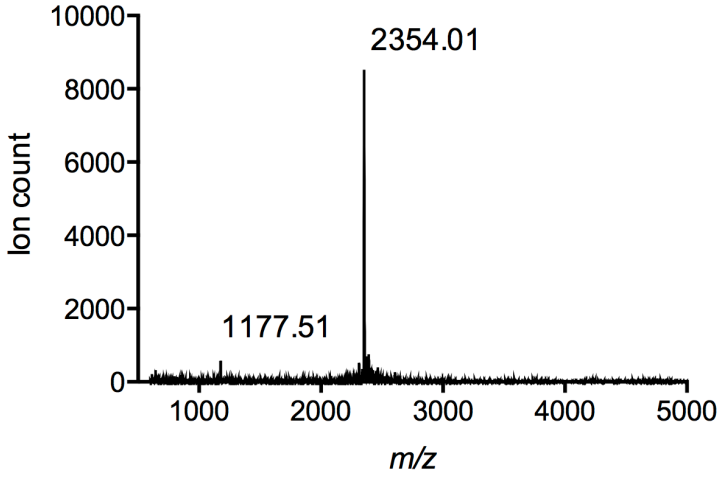

Figure S2. MALDI-TOF mass-spectrum of **LB1_12**

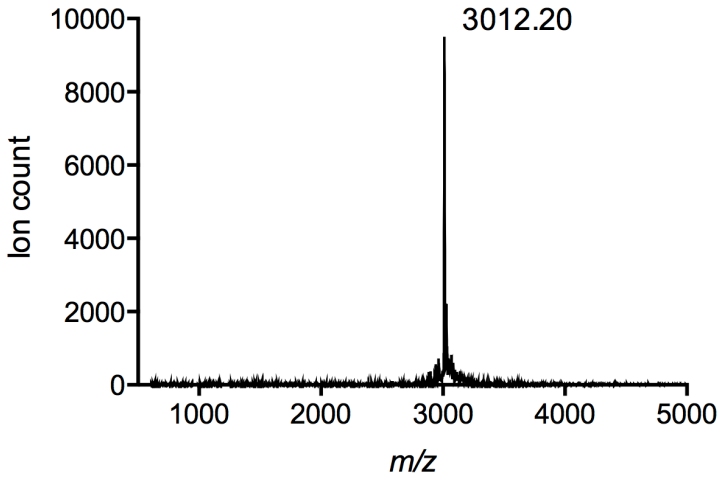

Figure S3. MALDI-TOF mass-spectrum of **LB2_3**

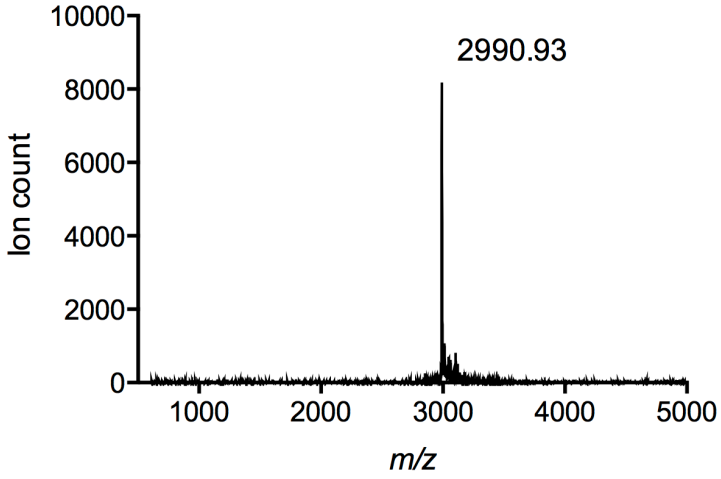

Figure S4. MALDI-TOF mass-spectrum of **LB2_5**

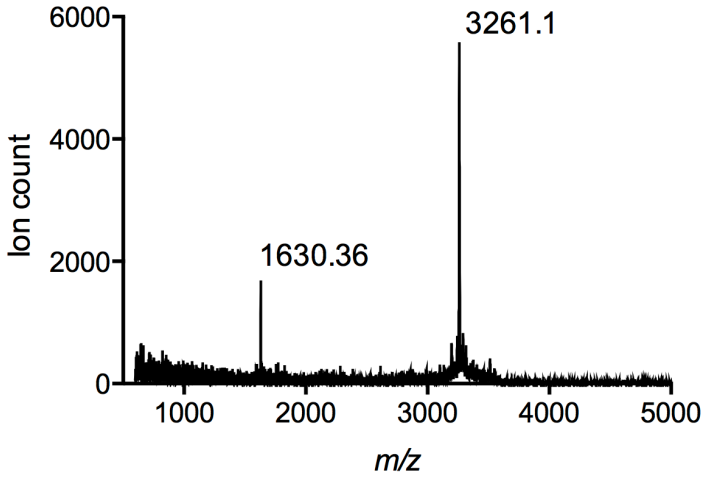

Figure S5. MALDI-TOF mass-spectrum of **LB2_7**

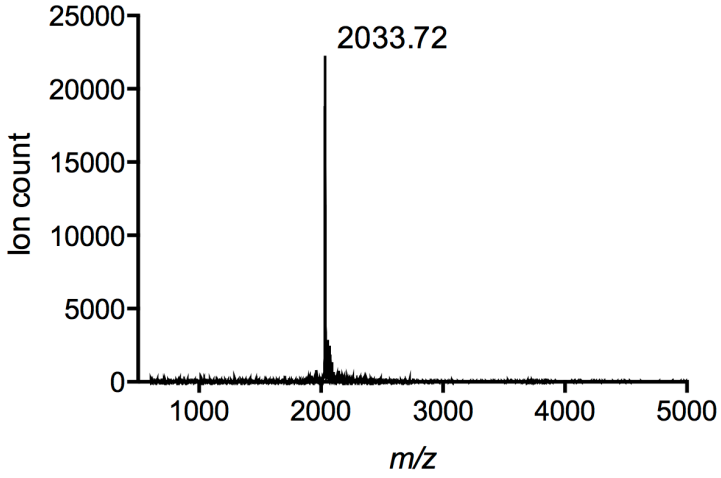

Figure S6. MALDI-TOF mass-spectrum of **LB2_24**

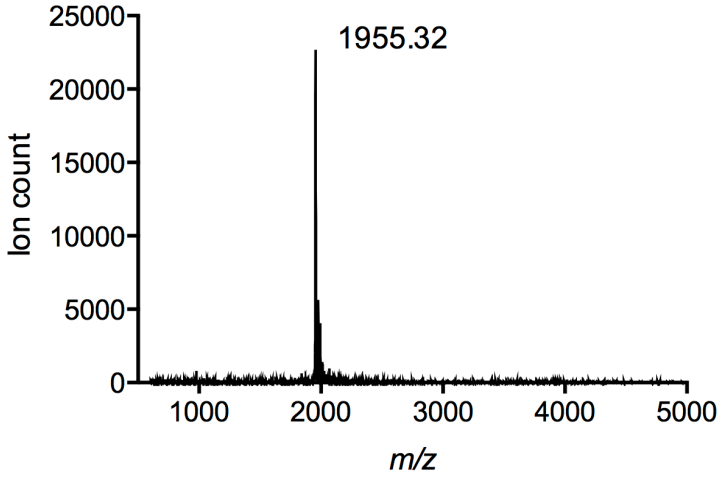

Figure S7. MALDI-TOF mass-spectrum of **LB2_27**

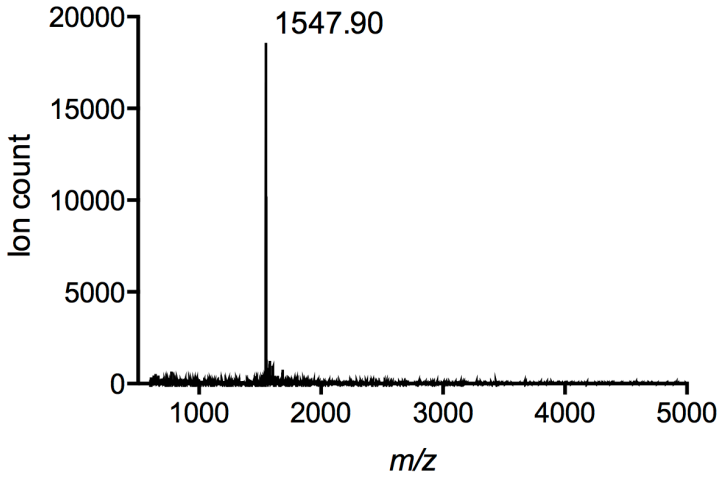

Figure S8. MALDI-TOF mass-spectrum of **LB2_54**

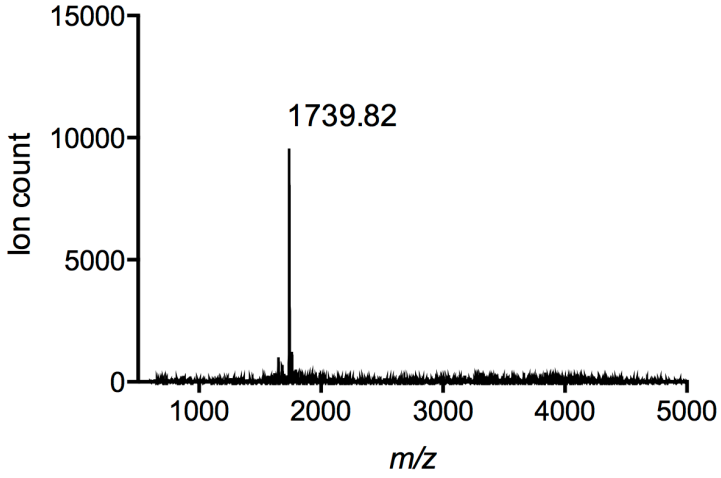

Figure S9. MALDI-TOF mass-spectrum of **LB2_68**

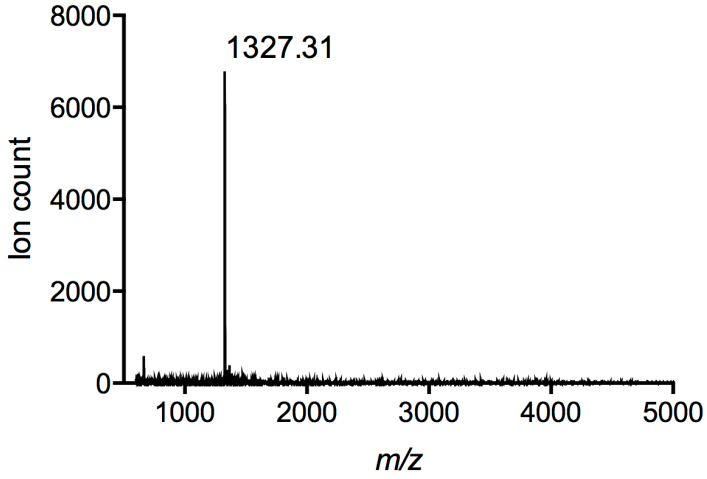

Figure S10. MALDI-TOF mass-spectrum of **LB2_72**

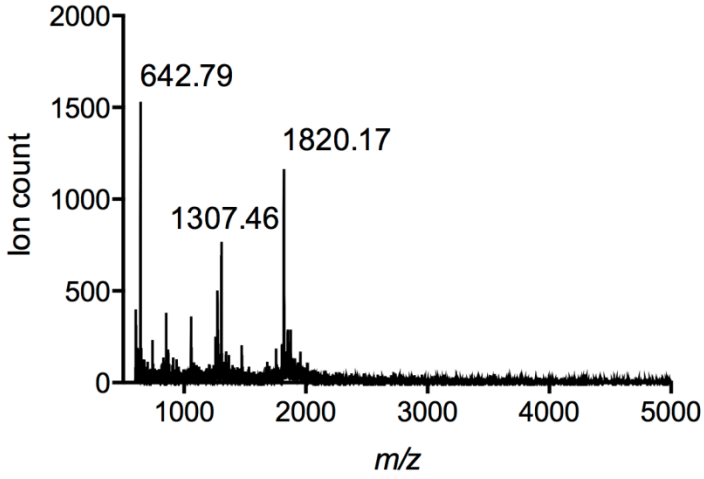

Figure S11. MALDI-TOF mass-spectrum of **LB2_40**

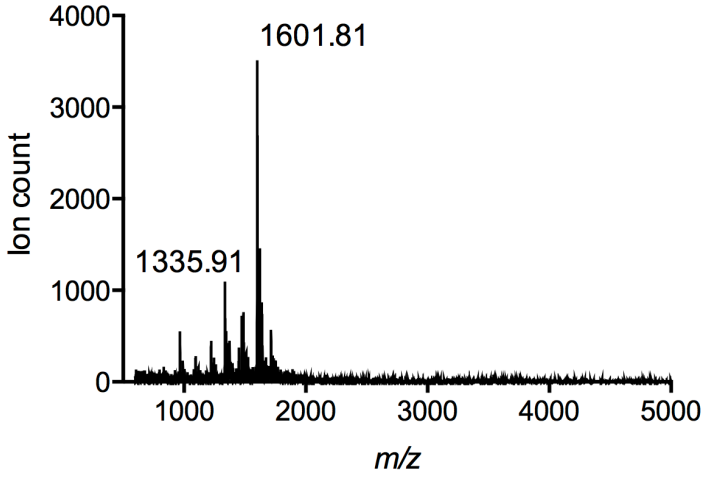

Figure S12. MALDI-TOF mass-spectrum of **LB2_51**

Example HPLC graphs of parallel-synthesized peptides obtained by SELPEPCON

Below, in **Figures S13** and **S14**, two example HPLC graphs of peptides obtained by SELPEPCON are shown. These are recorded on a Phenomenex analytical C18 Jupiter column (250 x 4.6 mm, 5 micron) using the following gradient (A: 0.1% TFA, B: 90% acetonitrile, 0.1% TFA) 0-2 min 15% B 2-20 min 15%-30% B 20-25 min 30%-90% B.

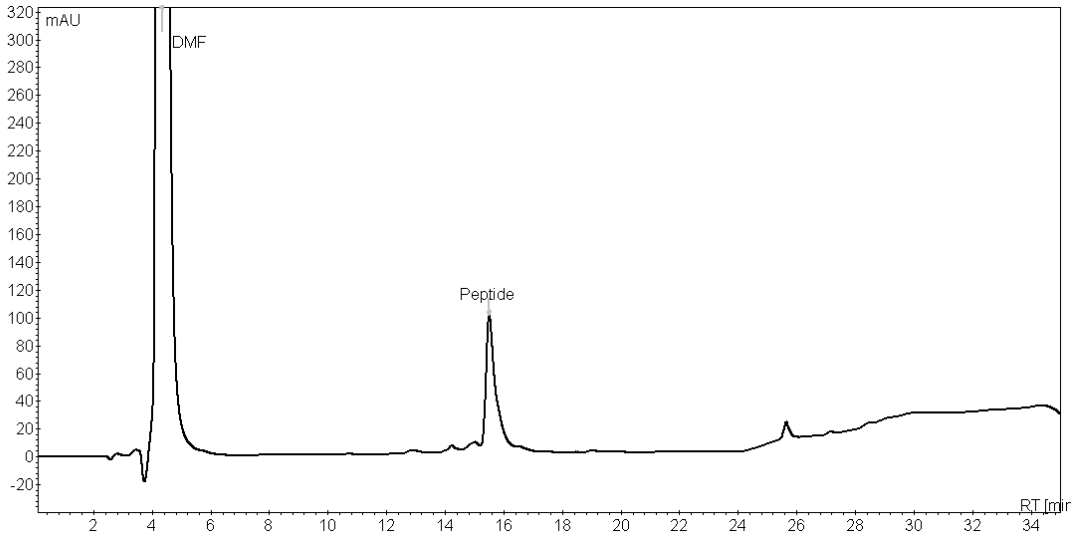

Figure S13. HPLC-graph of the **LB1_5** obtained by SELPEPCON.

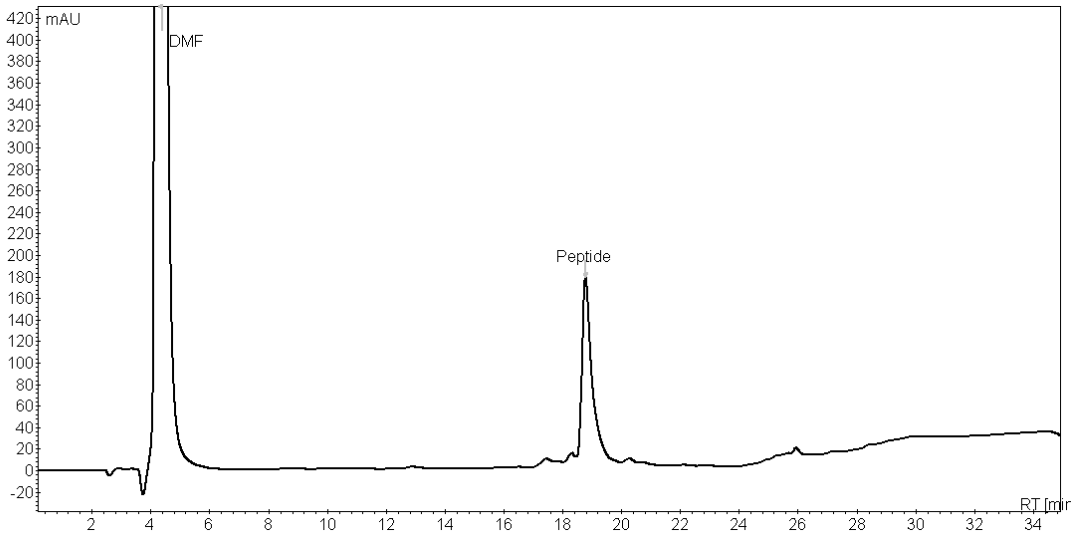

Figure S14. HPLC-graph of the **LB1_10** obtained by SELPEPCON.

Examples of MALDI mass-spectra of LB1- and LB2–PNA705 conjugates obtained by SELPEPCON

Below, in **Figures S15** - **S27,** examples are given of MALDI-TOF mass-spectra of conjugates obtained using the SELPEPCON procedure. **Figures S15** - **S26** show MALDI-TOF mass-spectra of PNA705 conjugates from libraries **LB1** and **LB2** that were tested in the splicing redirection assay. As an example **Figure S27** shows a MALDI-TOF mass-spectrum of a conjugate for which the conversion of the conjugation reaction was low and was thus not tested. **Figure S25** shows an example of a conjugate containing a cysteine in the CPP-sequence for which the mixture of conjugate containing different amounts of StBu groups was found by MALDI-spectroscopy.

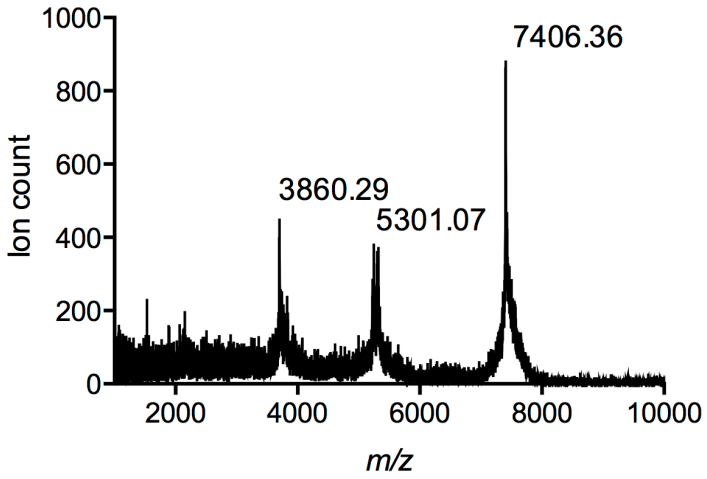

Figure S15. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB1_4**

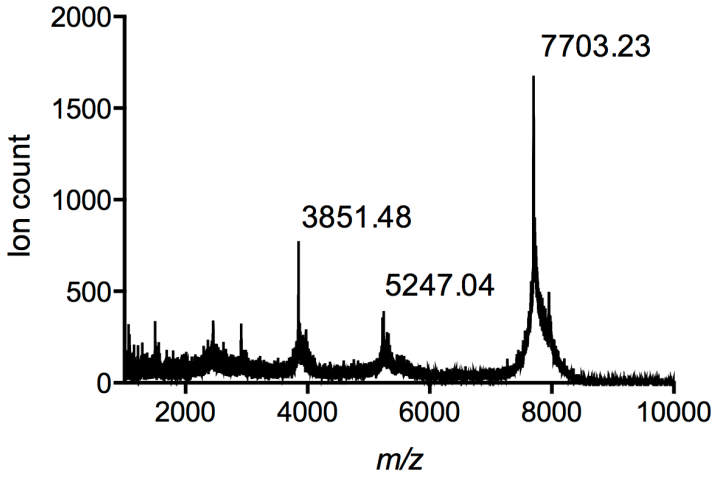

Figure S16. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB1_8**

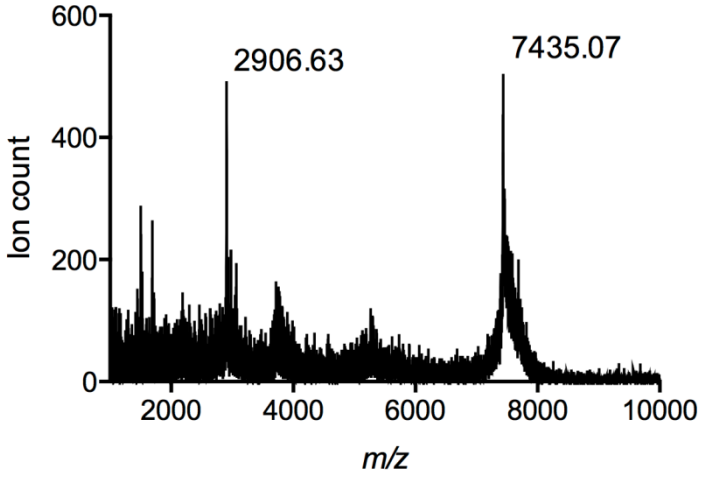

Figure S17. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB1_11**

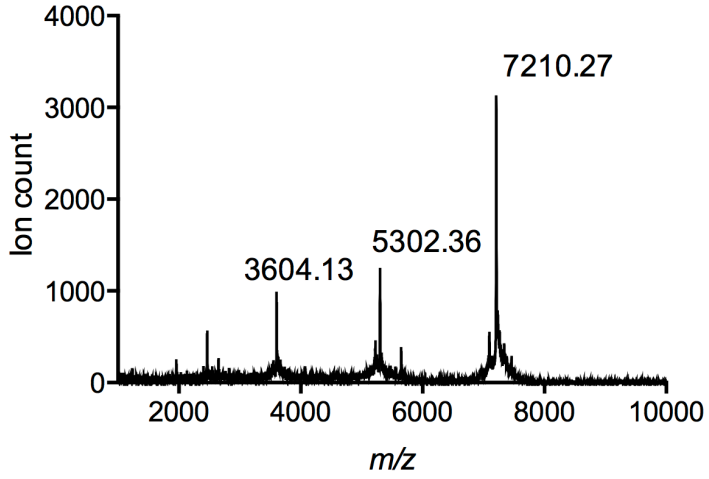

Figure S18. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_27**

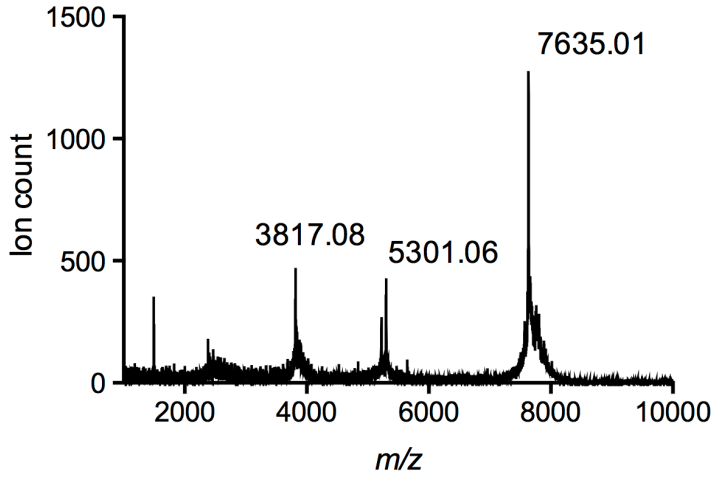

Figure S19. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_28**

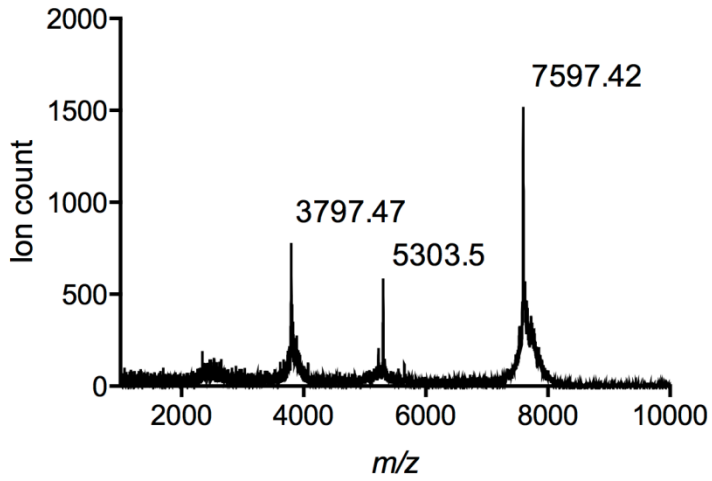

Figure S20. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_35**

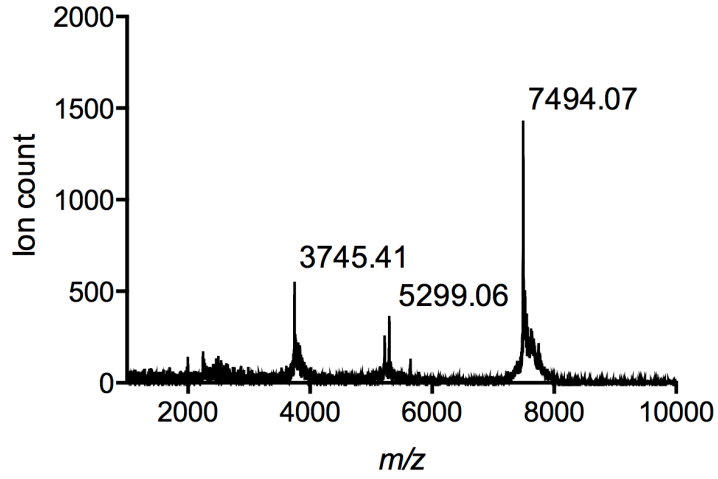

Figure S21. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_38**

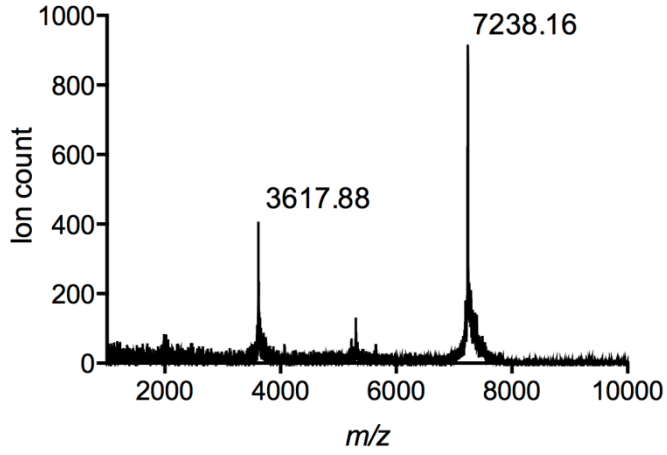

Figure S22. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_43**

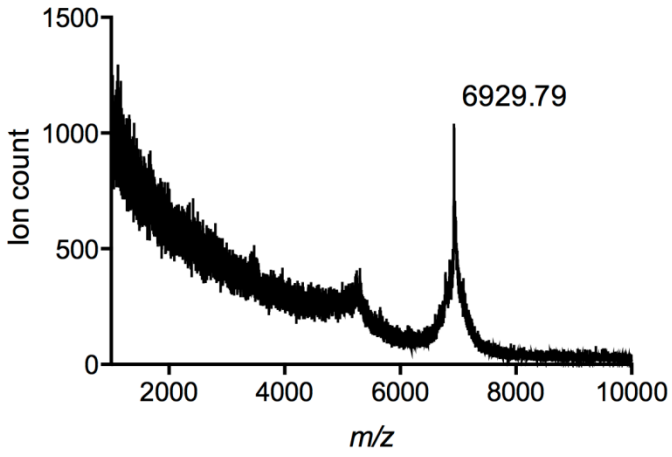

Figure S23. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_44**

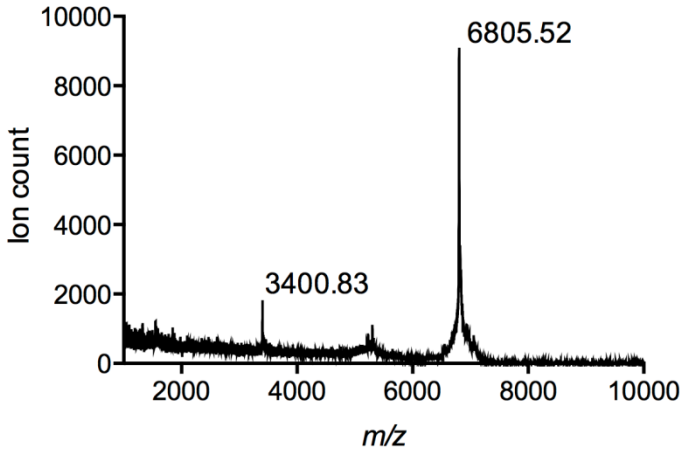

Figure S24. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_54**

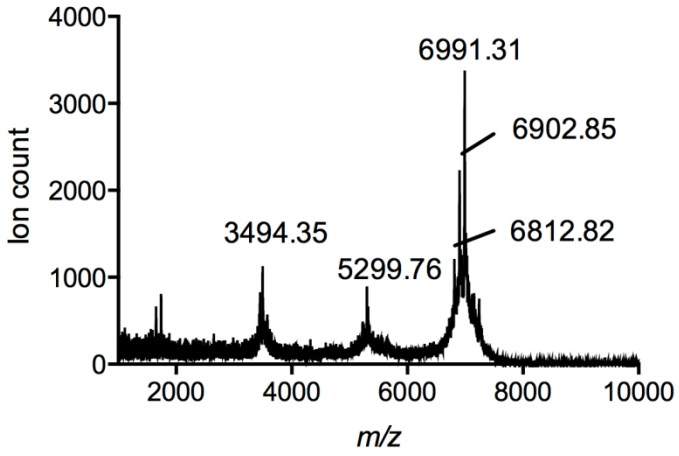

Figure S25. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_68**

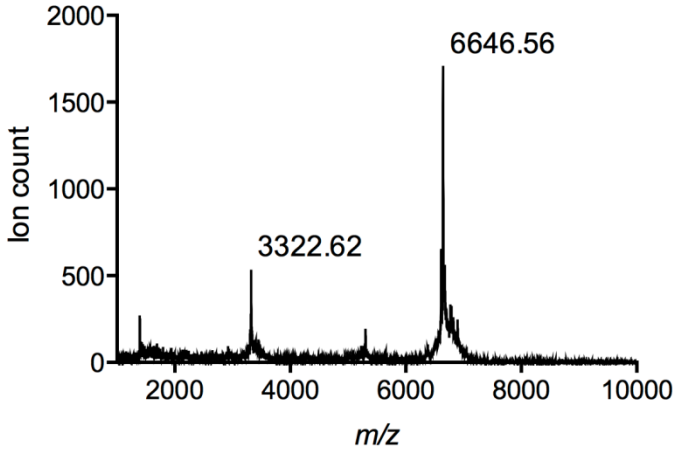

Figure S26. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_71**

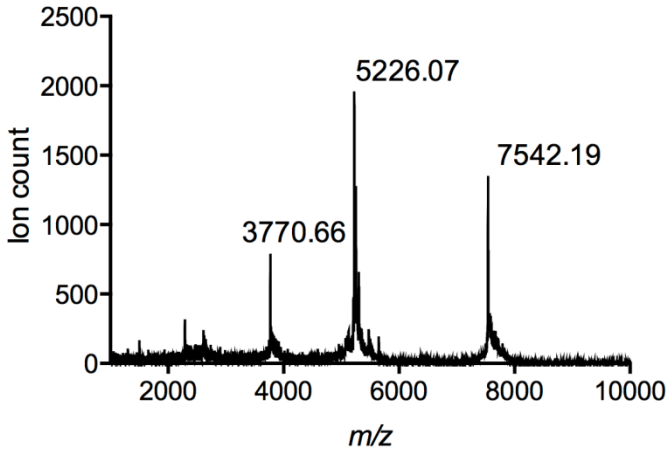

Figure S27. MALDI-TOF mass-spectrum of the PNA705 conjugate of **LB2_31**

Example HPLC graphs of CPP–PNA705 conjugates obtained by SELPEPCON

Below in **Figures S28** and **S29**, two example HPLC graphs of **LB2**–PNA705 conjugates are shown. These are recorded on a Phenomenex analytical C18 Jupiter column (250 x 4.6 mm, 5 micron) using the following gradient (A: 0.1% TFA, B: 90% acetonitrile, 0.1% TFA) 0-2 min 10% B 2-20 min 10%-30% B 20-30 min 30%-50% B 30-35 min 50%-90% B. Unconjugated PNA impurities (±10%) can be observed which are likely the results of a small amount of PNA-impurties without an azide functional group. In addition, impurities are observed that share a mass close to that expected of the conjugate. These can be the result of either impurities in the peptide or degradation of the conjugate.

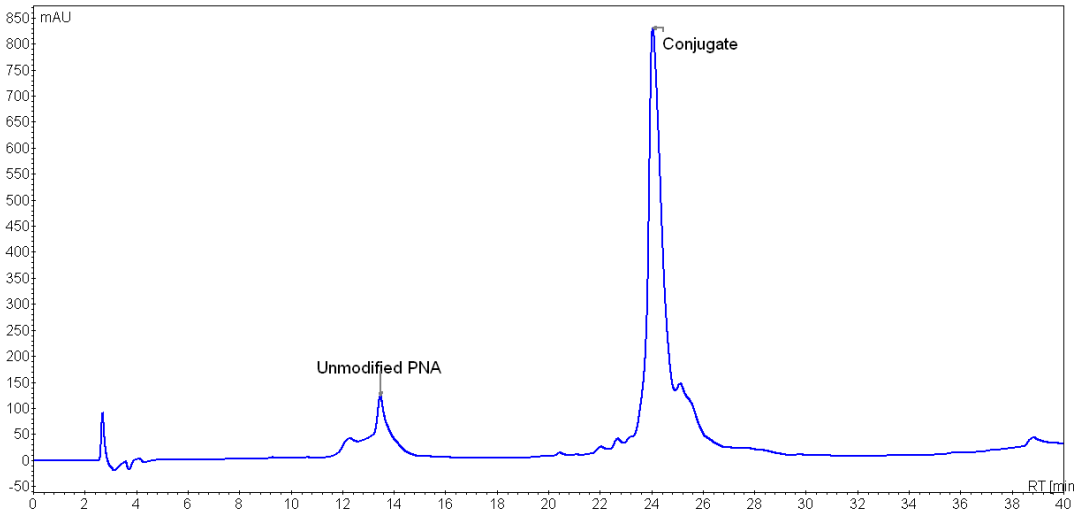

Figure S28. HPLC-graph of the **LB2_21**–PNA705 conjugate obtained by SELPEPCON.

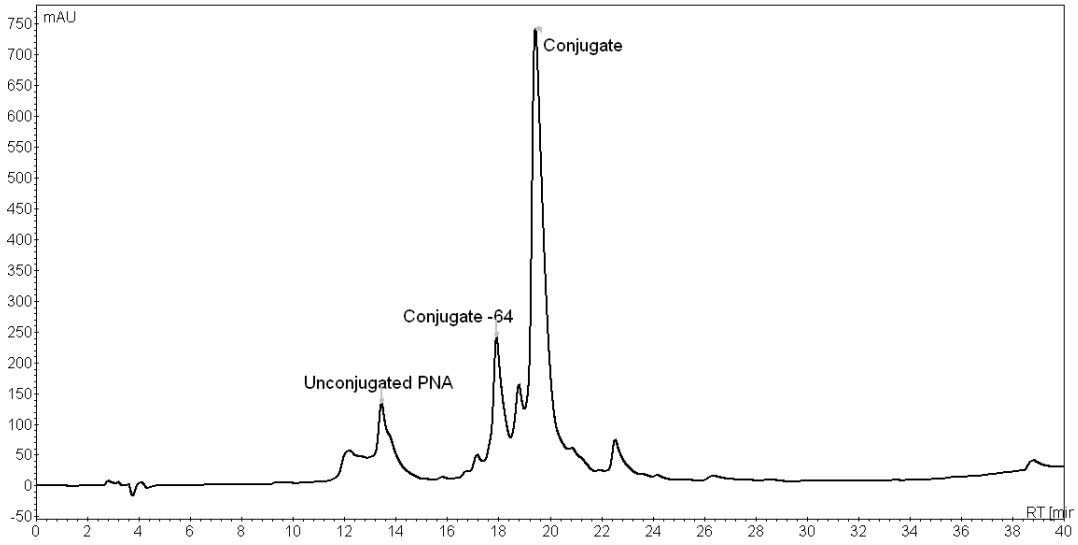

Figure S29. HPLC-graph of the **LB2_22**–PNA705 conjugate obtained by SELPEPCON.

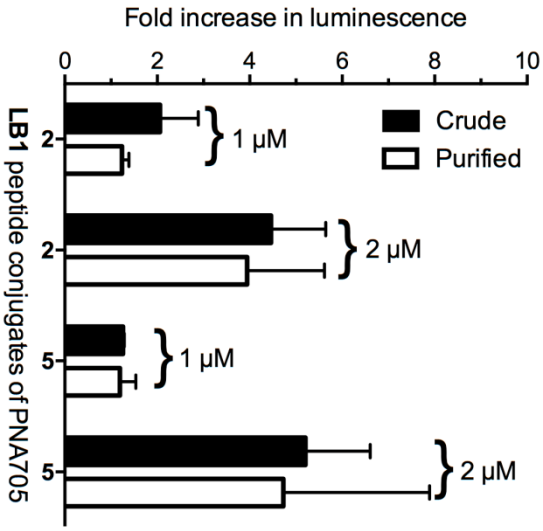

Figure S30. Fold increase in luminescence caused by PNA705 conjugates of **LB1**-peptides obtained by SELPEPCON (Crude) and RP-HPLC purified (Purified) compared to a buffer blank induced by the conversion of Beetle luciferin into oxyluciferin by expressed luciferase via splicing redirection in HeLa pLuc705 cells at 1 and 2 μM conjugate concentration.

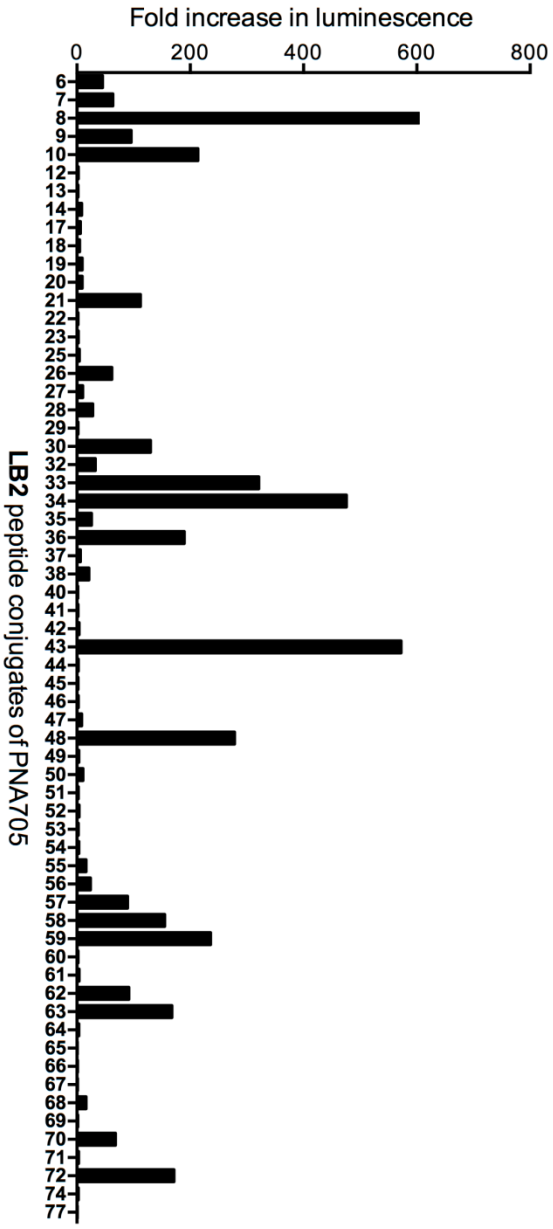

Figure S31. Fold increase in luminescence caused by PNA705 conjugates of **LB2**-peptides compared to a buffer blank induced by the conversion of Beetle luciferin into oxyluciferin by expressed luciferase via splicing redirection in HeLa pLuc705 cells in a single point screening assay at 5 μM conjugate concentration.