

Table S1 Identified cross-linked peptides of sample 1 between ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn in LPC (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
1.43E-20	-0.67	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	23	T ²² <u>K</u> QGVAAEAAGK ³²	2268.2324
5.94E-14	0.65	32	Q ²⁴ GVAEAAGKTK ³⁴	10	G ⁹ LS <u>K</u> AK ¹²	1799.0151
8.04E-13	-0.04	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	23	T ²² <u>K</u> QGVAAEAAGK ³²	2376.2899
2.17E-12	-1.77	32	T ²² KQGVAAEAAG <u>K</u> TK ³⁴	23	T ²² <u>K</u> QGVAAEAAGK ³²	2484.3546
5.89E-12	0.46	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	12	T ²² <u>K</u> QGVAAEAAGK ³²	2733.4911
1.33E-11	0.22	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1897.0519
2.22E-11	-2.07	58	T ⁴⁴ KEGVVHGVATVAE <u>K</u> TK ⁶⁰	10	G ⁹ LS <u>K</u> AK ¹²	2493.4165
1.49E-10	-2.77	23	T ²² <u>K</u> QGVAAEAAGK ³²	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1884.0315
2.94E-10	-1.25	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	2389.3103
1.42E-08	-1.10	21	E ¹³ GVVAAA <u>E</u> TK ²³	10	G ⁹ LS <u>K</u> AK ¹²	1842.0461
2.07E-08	-0.67	58	E ⁴⁶ GVVHGVATVAE <u>K</u> TKEQVTNVGGAVVTGVTAVAQK ⁸⁰	12	G ⁷ LSK <u>A</u> KEGVVAAAEEK ²¹	5027.7505
3.43E-08	-2.37	43	T ³³ <u>K</u> EGVLYVGS <u>K</u> TK ⁴⁵	10	G ⁹ LS <u>K</u> AK ¹²	2149.2357
5.39E-07	2.62	43	T ³³ <u>K</u> EGVLYVGS <u>K</u> TK ⁴⁵	43	T ³³ <u>K</u> EGVLYVGS <u>K</u> TK ⁴⁵	2955.6531

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ¹⁴N-Ac- α -syn and ¹⁴N-Ac- α -syn, ¹⁴N-Ac- α -syn and ¹⁵N-Ac- α -syn, ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn, or ¹⁵N-Ac- α -syn and ¹⁵N-Ac- α -syn. Detailed data processing procedure could be referred to Methods session.

Table S2 Identified cross-linked peptides of sample 2 between ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn in LPC (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
2.56E-17	-0.42	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	2733.4911
3.92E-15	0.45	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	23	T ²² <u>K</u> QGVAAEAGK ³²	2268.2324
3.41E-14	0.97	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	23	T ²² <u>K</u> QGVAAEAGK ³²	2376.2899
2.82E-13	0.45	32	Q ²⁴ GVAEAAAG <u>K</u> TK ³⁴	10	G ⁷ LS <u>K</u> AK ¹⁰	1799.0151
6.88E-13	0.59	43	T ³³ KEGVLYVGS <u>K</u> TK ⁴⁵	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	2618.4529
4.51E-12	1.04	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1897.0519
2.59E-11	-2.90	23	T ²² <u>K</u> QGVAAEAGK ³²	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1884.0315
1.28E-10	0.36	32	Q ²⁴ GVAEAAAG <u>K</u> TK ³⁴	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1884.0315
1.07E-09	-1.42	21	A ¹¹ KEGVVAAA <u>E</u> KTK ²³	12	A ¹¹ <u>K</u> EGVVAAAEEK ²¹	1965.9903
2.81E-09	2.82	21	E ¹³ GVVAAA <u>E</u> KTK ²³	10	G ⁷ LS <u>K</u> AK ¹⁰	2510.3954
7.27E-08	0.30	43	T ³³ KEGVLYVGS <u>K</u> TK ⁴⁵	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1842.0461

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ¹⁴N-Ac- α -syn and ¹⁴N-Ac- α -syn, ¹⁴N-Ac- α -syn and ¹⁵N-Ac- α -syn, ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn, or ¹⁵N-Ac- α -syn and ¹⁵N-Ac- α -syn. Detailed data processing procedure could be referred to Methods session.

Table S3 Identified cross-linked peptides of sample 3 between ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn in LPC (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
3.49E-15	0.21	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	23	T ²² <u>K</u> QGVAAE <u>A</u> AGK ³²	2268.2324
1.32E-13	3.78	60	T ⁵⁹ <u>K</u> EQVTNVGGAVVTGVTAVAQK ⁸⁰	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	3473.8980
4.26E-13	2.34	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	23	T ²² <u>K</u> QGVAAE <u>A</u> AGK ³²	2376.2899
1.14E-12	0.67	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1897.0519
2.32E-12	1.24	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	23	T ²² <u>K</u> QGVAAE <u>A</u> AGK ³²	2720.4707
5.94E-12	1.20	96	T ⁸¹ VEGAGSIAAATGFV <u>K</u> K ⁹⁷	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	2815.5330
1.07E-10	0.52	43	T ³³ <u>K</u> EGVLYVGS <u>K</u> TK ⁴⁵	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	2618.4529
5.04E-09	0.91	21	A ¹¹ <u>K</u> EGVVAAA <u>E</u> KTK ²³	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	2126.1946
2.29E-08	1.40	96	T ⁹¹ VEGAGSIAAATGFV <u>K</u> K ⁹⁷	23	T ²² <u>K</u> QGVAAE <u>A</u> AGK ³²	2802.5126
1.52E-07	1.15	43	T ³³ <u>K</u> EGVLYVGS <u>K</u> TK ⁴⁵	10	G ⁷ LS <u>K</u> AK ¹⁰	2149.2357
2.52E-07	-3.62	34	T ³³ <u>K</u> EGVLYVGSK ⁴³	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	2389.3103
5.32E-07	-2.34	60	T ⁵⁹ <u>K</u> EQVTNVGGAVVTGVTAVAQK ⁸⁰	21	E ¹³ GVVAAA <u>E</u> KTK ²³	3395.8510
7.21E-07	0.58	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	2281.2528
7.33E-07	2.58	32	T ²² <u>K</u> QGVAAE <u>A</u> AG <u>K</u> TK ³⁴	23	T ²² <u>K</u> QGVAAE <u>A</u> AGK ³²	2484.3546
8.89E-07	0.99	21	E ¹³ GVVAAA <u>E</u> KTK ²³	10	G ⁷ LS <u>K</u> AK ¹⁰	1842.0461

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ¹⁴N-Ac- α -syn and ¹⁴N-Ac- α -syn, ¹⁴N-Ac- α -syn and ¹⁵N-Ac- α -syn, ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn, or ¹⁵N-Ac- α -syn and ¹⁵N-Ac- α -syn. Detailed data processing procedure could be referred to Methods session.

Table S4 Identified cross-linked peptides of sample 1 between ^{15}N -Ac- α -syn and ^{14}N -Ac- α -syn in DOPS (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
2.08E-16	0.45	32	Q ²⁴ GVAEAAG <u>K</u> TK ³⁴	10	G ⁷ LS <u>K</u> AK ¹⁰	1799.0151
2.67E-14	2.65	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	23	T ²² <u>K</u> QGVAEAAGK ³²	2720.4707
4.22E-14	-2.67	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	12	A ¹¹ <u>K</u> EGVVAAA EK ²¹	2733.4911
1.76E-12	-0.66	12	A ¹¹ <u>K</u> EGVVAAA EK ²¹	23	T ²² <u>K</u> QGVAEAAGK ³²	2268.2324
1.92E-09	-0.87	21	E ¹³ GVVAAA EK <u>T</u> K ²³	23	T ²² <u>K</u> QGVAEAAGK ³²	2298.2430
8.17E-09	1.29	21	A ¹¹ KEGVVAAA EK <u>T</u> K ²³	10	G ⁷ LS <u>K</u> AK ¹⁰	2041.1782

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ^{14}N -Ac- α -syn and ^{14}N -Ac- α -syn, ^{14}N -Ac- α -syn and ^{15}N -Ac- α -syn, ^{15}N -Ac- α -syn and ^{14}N -Ac- α -syn, or ^{15}N -Ac- α -syn and ^{15}N -Ac- α -syn. Detailed data processing procedure could be referred to Methods session.

Table S5 Identified cross-linked peptides of sample 2 between ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn in DOPS (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
3.01E-15	-0.73	43	T ³³ KEGVLYVGS <u>K</u> TK ⁴⁵	43	E ³⁵ GVLVYVGS <u>K</u> TK ⁴⁵	2726.5105
1.71E-11	2.06	96	T ⁸¹ VEGAGSIAAATGFV <u>K</u> K ⁹⁷	12	A ¹¹ <u>K</u> EGVVAAAEK ²¹	2815.5330
1.22E-10	-2.90	32	Q ²⁴ GVAEAAG <u>K</u> TK ³⁴	97	<u>K</u> ⁹⁷ DQLGK ¹⁰²	1884.0315
3.80E-08	-0.35	96	T ⁸¹ VEGAGSIAAATGFV <u>K</u> K ⁹⁷	10	G ⁷ LS <u>K</u> AK ¹⁰	2346.3157
4.64E-07	0.38	23	T ²² <u>K</u> QGVAEAAGK ³²	23	T ²² <u>K</u> QGVAEAAGK ³²	2255.2120

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ¹⁴N-Ac- α -syn and ¹⁴N-Ac- α -syn, ¹⁴N-Ac- α -syn and ¹⁵N-Ac- α -syn, ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn, or ¹⁵N-Ac- α -syn and ¹⁵N-Ac- α -syn. Detailed data processing procedure could be referred to Methods session.

Table S6 Identified cross-linked peptides of sample 3 between ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn in DOPS (protein: lipid= 1: 50, mol: mol).

Score	Δ (ppm)	Res of Pr 1*	Sequence of Pr 1*	Res of Pr2*	Sequence of Pr 2*	Mass (Da)
4.55E-14	0.36	32	Q ²⁴ GVAEAAG <u>K</u> TEGVLYVGSK ⁴³	21	A ¹¹ KEGVVAAA <u>E</u> KTK ²³	3429.8717
9.60E-14	0.25	32	Q ²⁴ GVAEAAG <u>K</u> TK ³⁴	10	G ⁷ LS <u>K</u> AK ¹⁰	1799.0151
1.05E-07	-1.97	45	T ⁴⁴ <u>K</u> EGVVHGVATVAEK ⁵⁸	43	E ³⁵ GVLYVGS <u>K</u> TK ⁴⁵	2841.5486
1.08E-07	1.16	23	T ²² <u>K</u> QGVAEAAGK ³²	32	Q ²⁴ GVAEAAG <u>K</u> TK ³⁴	2255.2120
2.30E-07	-1.49	96	T ⁸¹ VEGAGSIAAATGFV <u>K</u> K ⁹⁷	12	A ¹¹ <u>K</u> EGVVAAA <u>E</u> K ²¹	2815.5330
5.89E-07	3.15	6	M ¹ DVFM <u>K</u> GLSK ¹⁰	32	Q ²⁴ GVAEAAG <u>K</u> TK ³⁴	2351.2228

Δ : mass error for crosslink assignments; Res: crosslinked residue (sequence number) within corresponding proteins; Pr: protein; Underlined K in sequence is the cross-linked lysine.

*The crosslinked pair of residues were from two individual Ac- α -syn: ¹⁴N-Ac- α -syn and ¹⁴N-Ac- α -syn, ¹⁴N-Ac- α -syn and ¹⁵N-Ac- α -syn, ¹⁵N-Ac- α -syn and ¹⁴N-Ac- α -syn, or ¹⁵N-Ac- α -syn and ¹⁵N-Ac- α -syn. Detailed data processing procedure could be referred to Methods session.