**Supplementary Information**

**Effect of Phosphatidylserine and Cholesterol on Membrane-mediated Fibril Formation by the N-terminal Amyloidogenic Fragment of Apolipoprotein A-I**

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**This supplementary data consists of:**

Figures S1‒S6 and Table S1

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**Figure S1. Formation of amyloid-like structure of apoA-I 1‒83 bound to SUV.** Formation of amyloid-like structure was monitored by ThT fluorescence for apoA-I 1‒83 in the presence of SUV. PL/apoA-I weight ratio was 30. ○, in buffer; □, PC SUV; ▲, PC/Chol (2/1) SUV; ∇, PC/PS (7/3) SUV. Protein concentration was 0.05 mg/ml. *a. u.*, arbitrary units.

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**Figure S2. MALDI-TOF MS spectra of apoA-I 1-83/G26R before (A) and after (B) 120h incubation.** MALDI-TOF MS analysis was performed using a microflex instrument (Bruker Daltonics, Bremen, Germany) in the positive linear ion mode. Spectra were calibrated externally using a standard protein mixture (Protein calibration standard I and II, Bruker). The ion observed at m/z 9774.8‒9774.9 was estimated to be the protonated molecule (M + H)+: molecular mass of the recombinant apoA-I 1-83/G26R is calculated to be 9773.81.

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**Figure S3. Particle size distributions of PC (A) and PC/PS (7/3) (B) SUVs determined by dynamic light scattering measurements on a Zetasizer Nano ZS (Malvern).** The data were represented as volume-based distributions.

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**Figure S4. Far-UV CD spectra of apoA-I 1-83/G26R L22C (A and B) and S58C (C and D) variants bound to PC or PC/PS (7/3) SUVs.** Errors in the CD spectra of apoA-I 1-83/G26R bound to SUVs were shown as dotted lines.

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**Figure S5. Effect of salt concentration on fibril-forming property of apoA-I 1-83/G26R bound to SUV.** Formation of amyloid-like structure was monitored by ThT fluorescence for apoA-I 1‒83/G26R in the presence of SUV at NaCl concentration of 50 mM. PL/apoA-I weight ratio was 10. ○, PC SUV; ∇, PC/PS (7/3) SUV. Protein concentration was 0.05 mg/ml. *a. u.*, arbitrary units. The data at NaCl concentration of 150 mM (dotted line) are also shown for comparison.

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**Figure S6. Effects of PS on membrane-binding properties of apoA-I 8‒33 peptide.** (**A**) Comparison of α-helix contents of apoA-I 8‒33 and 8‒33/G26R peptides in buffer or bound to various SUVs. (**B**) Isothermal titration thermogram for binding of apoA-I 8‒33 peptide to PC/PS (7/3) SUV.

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| **Table S1. Thermodynamic parameters of binding of apoA-I 8‒33 peptide to SUVs at 25 °C**a | | | | | | | | |
|  |  |  |  |  | |  | |  |
| SUV | *K*d  (μg/ml) | *B*max  (amino acids/  mol PL) |  | ∆*G*b (kcal/mol) | | ∆*H* (kcal/mol) | | *T*∆*S*c (kcal/mol) |
| PC | 2.7 ± 0.9 | 0.05 ± 0.02 |  | −10.7 ± 0.2 | | −21.1 ± 1.6 | | −10.4 ± 1.8 |
| PC/PS (7/3) | 1.0 ± 0.2 | 0.16 ± 0.01 |  | −11.2 ± 0.1 | | −22.4 ± 0.1 | | −11.1 ± 0.1 |
| aThe data were from two or three independent experiments.  b Free energy was calculated according to ∆*G* = −*RT* ln 55.5(1/*K*d).  c The entropy of binding was calculated from ∆*G* = ∆*H* ‒ *T*∆*S* | | | | |  | |  | |
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