Electronic Supplementary Material (ESI) for Analyst.

SUPPORTING INFORMATION

Investigating non-specific binding to chemically engineered sensor surfaces using liposomes as models

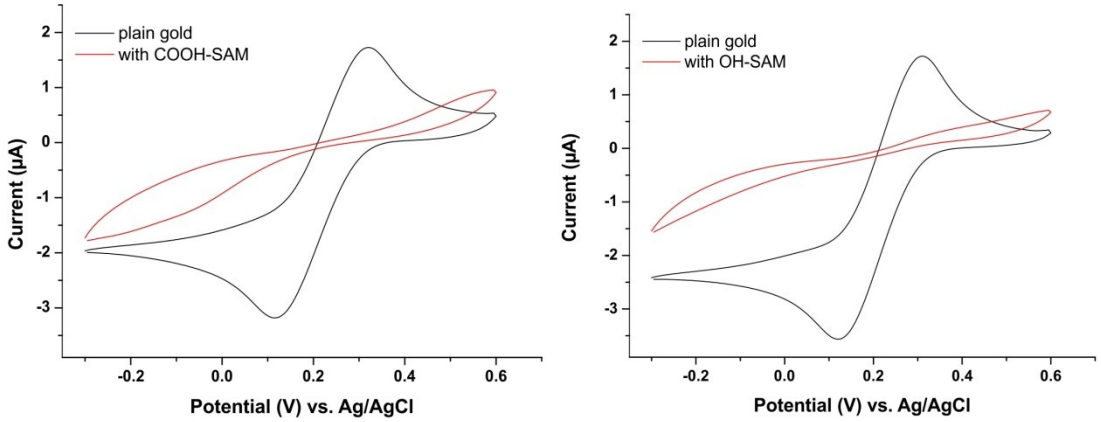
Christoph Fenzl[†], Christa Genslein[†], Celesztina Domonkos[‡], Katie A. Edwards[§], Thomas Hirsch[†], and Antje J. Baeumner[†,]*

[†]Institute of Analytical Chemistry, Chemo- and Biosensors, University of Regensburg, Universitaetsstr. 31, 93053 Regensburg, Germany

‡Research Centre for Natural Sciences, Hungarian Academy of Sciences, Magyar tudósok körújta 2., 1117, Budapest, Hungary.

§Department of Biological and Environmental Engineering, Cornell University, Riley-Robb Hall, 111 Wing Drive, Ithaca, NY 14853-5701

1) Cyclic voltammograms of self-assembled monolayer on Gold



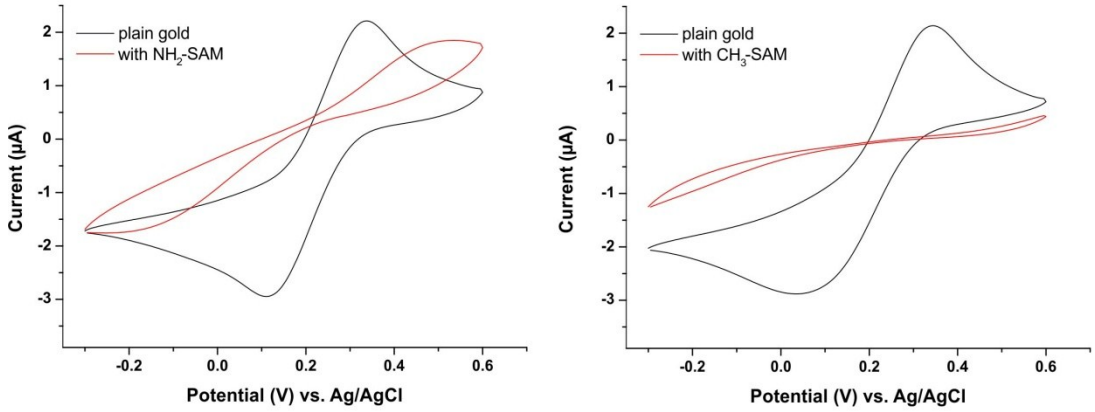


Figure S1. Cyclic voltammograms before and after the formation of self-assembled monolayers on gold electrodes. The solutions are 5 mM $Fe(CN)_6^{3-}$ in HEPES buffer. Scan rate 100 mV/s.

2) Fitting parameters of binding isotherms

Table S1. Parameters for the interaction of untagged liposomes with self-assembled monolayers on gold obtained by binding isotherms.

Fit model	Surface	$\Delta n_{D,max}$ [a] / RIU	K_L [b] / L·pmol^{-1}	K_D [c] / pmol·L^{-1}	h	R^2
simple	-COOH	0.0104±0.0013	0.00074±0.00012	1400±200		0.9991
	-OH	0.0094±0.0009	0.0032±0.0005	310±60		0.9924
	-CH_3	0.0013±0.0002	0.012±0.006	80±40		0.8501
	-NH_2	0.019±0.002	0.0061±0.0018	160±50		0.9730
	gold	0.0134±0.0010	0.00271±0.0003	370±40		0.9968
Fit model	**Surface**	$\Delta n_{D,max}$ [a] / RIU	K_L [b] / (L·pmol^{-1})^h	K_D [c] / (pmol·L^{-1})^h	h	R^2
extended	-COOH	0.014±0.010	0.0007±0.0003	1400±600	0.95±0.08	0.9989
	-OH	0.026±0.006	0.0036±0.0006	280±50	0.71±0.02	0.9998
	-CH_3	-	-	-	-	-
	-NH_2	0.0148±0.0008	0.0006±0.0005	2000±1400	1.62±0.19	0.9941
	gold	0.020±0.006	0.0035±0.0004	290±30	0.84±0.07	0.9985

[a]refractive index change at maximum surface loading, [b]Langmuir equilibrium constant, [c]Dissociation constant $K_D=1/K_L$

Table S2. Parameters for the interaction of liposomes with N-glutaryl-DPPE tag with self-assembled monolayers on gold obtained by simple and extended Langmuir fits.

Fit model	Surface	$\Delta n_{D,max}$[a] / RIU	K_L[b] / L·pmol^{-1}	K_D[c] / pmol·L^{-1}	h	R^2
simple	-COOH	0.0052±0.0019	0.0024±0.0012	400±200		0.9830
	-OH	0.0143±0.0009	0.0030±0.0003	330±30		0.9992
	-CH_3	0.0018±0.0005	0.02±0.013	50±30		0.7174
	-NH_2	0.018±0.002	0.014±0.004	70±20		0.9791
	gold	0.022±0.012	0.0010±0.0006	1000±600		0.9930
Fit model	**Surface**	$\Delta n_{D,max}$[a] / RIU	K_L[b] / (L·pmol^{-1})^h	K_D[c] / (pmol·L^{-1})^h	h	R^2
extended	-COOH	0.01±0.03	0.002±0.004	500±1000	0.9±0.3	0.9780
	-OH	0.020±0.005	0.0029±0.0004	340±50	0.91±0.04	0.9995
	-CH_3	-	-	-	-	-
	-NH_2	0.0142±0.0004	0.0028±0.0009	360±110	1.57±0.10	0.9983
	gold	0.011±0.009	0.0012±0.0005	800±300	1.1±0.3	0.9915

[a]refractive index change at maximum surface loading, [b]Langmuir equilibrium constant, [c]Dissociation constant $K_D=1/K_L$

3) SPR Simulation

SPR simulations were performed with the software Winspall (Freeware from MPIP Mainz, Germany). A 640 nm light source and a triangular prism (50 °) were chosen in best resemblance of the experimental setup. The following layer setup was used for approximation of a closest packed liposome monolayer:

Layer	Thickness / nm	*εX - real*	*εX - imaginary*
Glass	Infinite (starting layer)	2.59	0
Chromium[1]	3	-6.901	28.818
Gold[2]	50	-12.555	1.1464
Thiol	1.5	1.847	0
Lipid bilayer	4.8	2.25	0
300 mM NaCl	200	1.786	0
Lipid bilayer	4.8	2.25	0
Water	Infinite (ending layer)	1.778	0

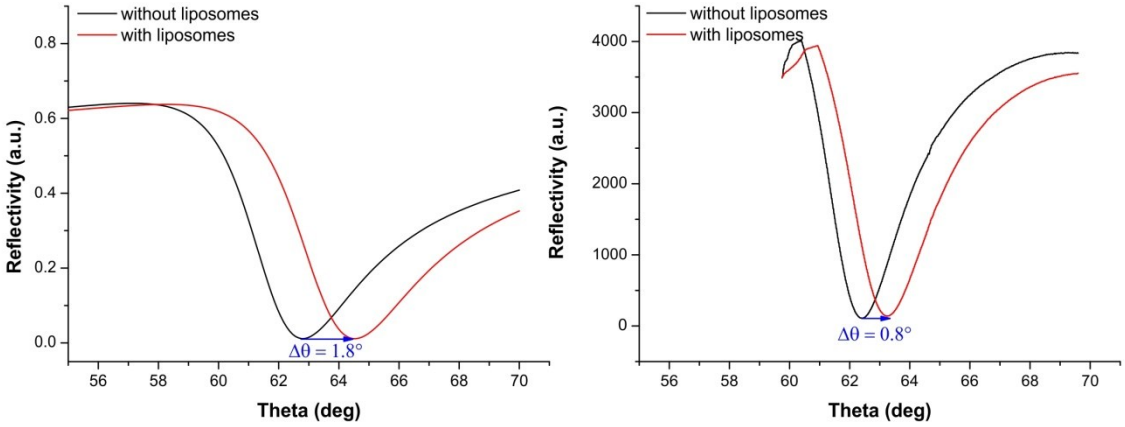


Figure S2. Change of the minimum of the SPR angle induced by anionic liposomes (300 mM NaCl encapsulant, without N-glutaryl-DPPE tag) on a $-NH_2$ SAM modified gold surface simulated for maximum surface loading (left) and obtained from experiment at 100 µM phospholipid concentration (right).

4) Van 't Hoff plot

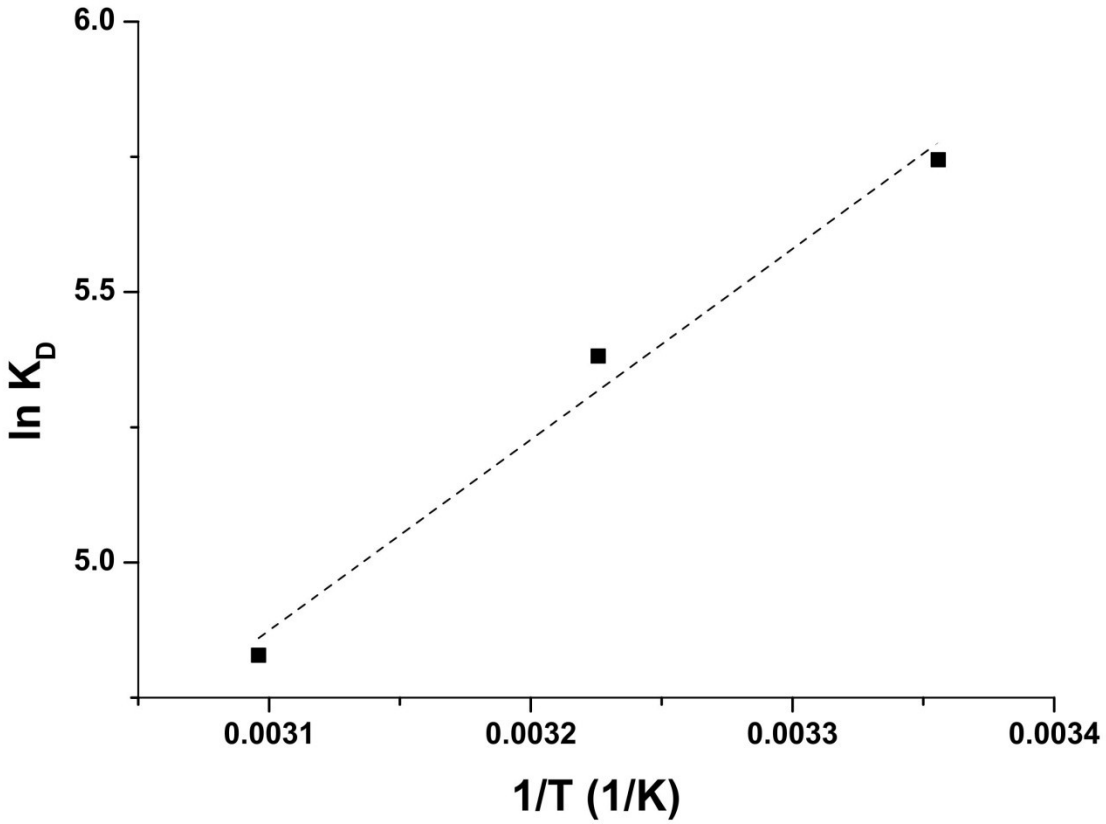


Figure S3. Van 't Hoff plot of the K_D values obtained from the Langmuir fits of the temperature dependent interactions of anionic liposomes (300 mM NaCl encapsulant, no tag) with -OH SAM modified gold surfaces at varying phospholipid concentrations at different temperatures.

References

(1) Rakic, A. D.; Djurišic, A. B.; Elazar, J. M.; Majewski, M. L. Optical Properties of Metallic Films for Vertical-Cavity Optoelectronic Devices. *Applied Optics* 1998, *37* (22), 5271–5283.

(2) Olmon, R. L.; Slovick, B.; Johnson, T. W.; Shelton, D.; Oh, S.-H.; Boreman, G. D.; Raschke, M. B. Optical Dielectric Function of Gold. *Phys. Rev. B* 2012, *86* (23), 235147.