Supporting Information for

Influence of Sensor Composition on Nanoparticle and Protein Interaction with Supported Lipid Bilayers

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Figure S1. Transmision electron microscopy images of MUA AuNPs.



Figure S2. UV-vis absorption spectroscopy of MUA AuNPs.

Characterization technique	Material	d _c (nm)ª	<i>d</i> _h (nm) ^{<i>b</i>}	ζ (mV) ^c
TEM ^d	MUA AuNPs ^e	7.5 ± 0.7	—	—
UV-vis ^f	MUA AuNPs ^g	—	8 ± 1	—
DLS^h or LDE^i	MUA AuNPs ^j	—	8.2 ± 0.4	-29.3 ± 5.6
	DOPC ^k	—	109.9 ± 0.5	-1.2 ± 1.2
	Cytochrome <i>c</i> [/]		—	-2.2 ± 0.4

Table S1. Characterization of nanoscale mate	erials
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^a*d*_c is nanoparticle core diameter. ^{*b*}*d*_h is nanoparticle hydrodynamic diameter. ^{*c*}ζ is nanomaterial zeta potential. ^{*d*}Transmision electron microscopy. ^{*e*}MUA AuNPs were suspended in water, drop cast onto a TEM grid, and dried. ^{*f*}Ultraviolet-visible absorption spectroscopy. ^{*g*}MUA AuNPs were measured in water. ^{*h*}Dynamic light scattering. ^{*i*}Laser doplar electrophoresis. ^{*j*}MUA AuNPs were measured at a concentration of 3 nM in 150 mM NaCl buffered to pH 7.4 with 10 mM HEPES. ^{*k*}DOPC vesicles were measured at a concentration of 0.125 mg·mL⁻¹ in 150 mM NaCl buffered to pH 7.4 with 10 mM HEPES.



Figure S3. Frequency and dissipation *vs* time plots for a) vesicle fusion bilayer formation on SiO₂, b) solvent assisted bilayer formation on SiO₂, and c) solvent assisted bilayer formation on Au. Vesicle fusion bilayers are formed in 150 mM NaCl buffered to pH 7.4 with 10 mM HEPES. Solvent-assisted bilayers section I and III are 150 mM NaCl buffered to pH 7.4 with 10 mM HEPES, section II is isopropanol. Dotted lines represent one standard deviation of four replicate measurements.



Figure S4. Au(100), Au(111) and SiO₂ SLBs. a) Au(100)-DOPC SLB before equilibration b) Au(111)-DOPC SLB before equilibration c) SiO₂-DOPC before equilibration d) Au(100)-DOPC SLB after equilibration e) Au(111)-DOPC SLB after equilibration e) SiO₂-DOPC after equilibration. *D*: 1 nm (effective distance between the DOPC and Au(100) or SiO₂ surfaces) *D*': 0.7 nm (effective distance between the DOPC and Au(111) surface). Water molecules, hydrogen atoms, Na⁺ and Cl⁻ ions are removed for clarity. C: cyan, O: red, N: blue, Au: yellow, Si: orange



Figure S5. The selected six protein orientations differing from each other by 90° rotations with respect to the SLB surface. Here the SLB is Au(100)-DOPC and only a part of the SLB is shown for each orientation. Water molecules, hydrogen atoms, Na⁺ and Cl⁻ ions are removed for clarity. C: cyan, O: red, N: blue, Au(100): yellow.

Formation method	Sensor composition	Δ <i>f</i> ₅ /5 (Hz)	$\Delta D_{5} (10^{-6})$
Vesicle fusion	SiO ₂	-24 ± 0.5	0.2 ± 0.1
Solvent assisted	SiO ₂	-24 ± 1	0.2 ± 0.1
	Au	-28 ± 2	0.3 ± 0.2

Table S2. Frequency and dissipation values for supported lipid bilayers^a

^aFrequency and dissipation values reported in 150 mM NaCl buffered to pH 7.4 with 10 mM HEPES.

Table S3. Final frequency and dissipation values for MUA AuNP^{*a*} and cytochrome c^{b} interaction with supported lipid bilayers

Formation method	Sensor composition	Nanoscale analyte	Δ <i>f₅</i> /5 (Hz)	ΔD ₅ (10 ⁻⁶)
Vesicle fusion	SiO ₂	MUA AuNP	-0.4 ± 0.5*	-0.04 ± 0.07*
		Cytochrome c	0.3 ± 0.5*	0.01 ± 0.08*
Solvent assisted	SiO ₂	MUA AuNP	0.1 ± 0.3*	-0.01 ± 0.06*
		Cytochrome c	-0.9 ± 1.1*	-0.03 ± 0.2*
	Au	MUA AuNP	-35 ± 0.3	3.2 ± 0.3
		Cytochrome c	-29 ± 4	3.0 ± 1.1
Bare sensor	SiO ₂	MUA AuNP	-0.1 ± 0.1*	0.07 ± 0.08*
		Cytochrome c	-14 ± 1	0.2 ± 0.07
	Au	MUA AuNP	-5.6 ± 0.8	0.05 ± 0.04
		Cytochrome c	-16 ± 2	0.2 ± 0.04

^aFrequency and dissipation values for MUA AuNP-bilayer interaction reported in 0.15 M NaCl buffered to pH 7.4 with 0.01 M HEPES. ^bFrequency and dissipation values for cytochrome *c*-bilayer interaction reported in 0.01 M NaCl buffered to pH 7.4 with 0.01 M HEPES. Error reported as the standard deviation of 4 replicate measurements. *Values are statistically indistinguishable from 0 (p < 0.05).

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Material	Hamaker constant (J) ^a
Water	3.70·10 ⁻²⁰
SiO2	6.30·10 ⁻²⁰
Au	4.00·10 ⁻¹⁹
DOPC	7.62·10 ⁻²⁰
Material-Media-Material	Calculated Hamaker constant (J) ^b
Material-Media-Material SiO2-Water-AuNP	Calculated Hamaker constant (J) ^{<i>b</i>} 2.58·10 ⁻²⁰
Material-Media-Material SiO2-Water-AuNP Au-Water-AuNP	Calculated Hamaker constant (J) ^b 2.58·10 ⁻²⁰ 1.94·10 ⁻¹⁹
Material-Media-Material SiO2-Water-AuNP Au-Water-AuNP SiO2-Water-DOPC	Calculated Hamaker constant (J) ^b 2.58·10 ⁻²⁰ 1.94·10 ⁻¹⁹ 4.91·10 ⁻²¹
Material-Media-Material SiO2-Water-AuNP Au-Water-AuNP SiO2-Water-DOPC Au-Water-DOPC	Calculated Hamaker constant (J) ^b 2.58·10 ⁻²⁰ 1.94·10 ⁻¹⁹ 4.91·10 ⁻²¹ 3.68·10 ⁻²⁰

Table S4. Hamaker constants and calculated Hamaker constants used for DLVO calculations^a

^{*a*}Hamaker constants taken from Israelachvili.^{1,2} ^{*b*}Hamaker constants calculated from literature Hamaker constants and equation 3.

Fable S5. Constants for van de	r Waals and electric	double layer calculations
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Constant	Variable	
Characteristic decay wavelength (nm)	λ	100ª
Permitivity of free space (C/Vm)	ϵ_0	8.85·10 ⁻¹²
Relative permitivity of medium	ε _r	78.5 ^b
Thickness of hydration layer (nm)	Z	0.3°
boltzman constant (J/K)	k _B	1.38·10 ⁻²³
temperature (K)	Т	298
Charge of an electron (C)	ei	1.60·10 ⁻¹⁹

^aDecay wavelength taken from Hahn and O'Melia.³ ^bPermittivity of water at pH 7.4 25 C. ^cThickness of hydration layer taken from Oleson and Sahai.⁴ amaker constants calculated from literature Hamaker constants and equation 3.

Calculations		
Material	Zeta potential (mV)	Surface potential (mV) ^a
SiO ₂ substrate		-42 ^b
Au substrate		0 <i>c</i>
DOPC	-1.2 ^d	-1.8
MUA AuNP	-29.3 ^d	-44.2

Table S6. Zeta potentials and surface potentials used for electric double layer calculations

^aSurface potential taken either from literature reference or calculated from zeta potential and equation 5. ^bZeta potential of SiO₂ substrte taken from Oleson and Sahai.⁴ ^cNeutral zeta potential of Au substrate taken from multiple reports of Au(111) being neutrally charged at neutral pH.^{5,6} ^dZeta potential of DOPC and MUA AuNPs taken from table S1.



Figure S6. The non-bonded interaction energies between interfacial surfaces of Au(100)-SLB, Au(111)-SLB and SiO₂-SLB. Electrostatic interactions (Elec), Van der Waals (vdW) interactions.



Figure S7. Averaged densities of the A, L, C, N sites of cytochrome *c* protein as a function of distance to the closest top surfaces for the SLB systems; a) Au(100)-SLB b) Au(111)-SLB c) SiO₂-SLB.



Figure S8. The hydrogen bonded interactions occurrence as a % of the total simulation time (100 ns) for the systems; a) Au(100)-SLB b) Au(111)-SLB c) SiO₂-SLB d) Au(100) e) Au(111) and f) SiO₂. The *x* axis gives the 105 residues available in the cytochrome c protein. The residues = ['G', 'D', 'V', 'E', 'K', 'G', 'K', 'K', 'I', 'F', 'V', 'Q', 'K', 'C', 'A', 'Q', 'C', 'H', 'T', 'V', 'E', 'K', 'G', 'G', 'K', 'H', 'K', 'T', 'G', 'P', 'N', 'L', 'H', 'G', 'L', 'F', 'G', 'R', 'K', 'T', 'G', 'Q', 'A', 'P', 'G', 'F', 'T', 'Y', 'T', 'D', 'A', 'N', 'K', 'N', 'K', 'G', 'I', 'T', 'W', 'K', 'E', 'E', 'T', 'L', 'M', 'E', 'Y', 'L', 'E', 'N', 'P', 'G', 'T', 'K', 'N', 'K', 'M', 'I', 'F', 'A', 'G', 'I', 'K', 'K', 'T', 'E', 'R', 'E', 'E', 'T', 'L', 'K', 'K', 'N', 'K', 'N', 'K', 'G', 'I', 'F', 'G', 'I', 'K', 'K', 'T', 'E', 'R', 'E', 'E', 'T', 'L', 'K', 'K', 'N', 'K', 'N', 'K', 'G', 'I', 'E', 'E', 'T', 'L', 'K', 'K', 'Y', 'I', 'P', 'G', 'T', 'K', 'M', 'I', 'F', 'A', 'G', 'I', 'K', 'K', 'K', 'T', 'E', 'R', 'E', 'D', 'L', 'A', 'Y', 'L', 'K', 'A', 'T', 'N', 'E', 'HEME'].



Figure S9. Radius of Gyration (*R*g)of cytochrome *c* when it is near the closest top surfaces for the systems; a) Au(100)-SLB b) Au(111)-SLB c) SiO₂-SLB d) Au(100) e) Au(111) and f) SiO₂.

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