Supplementary Information

Molecular Recognition and Supramolecular Self-Assembly of a Genetically Engineered Gold Binding Peptide on Au(111)

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1. Supplementary Figures

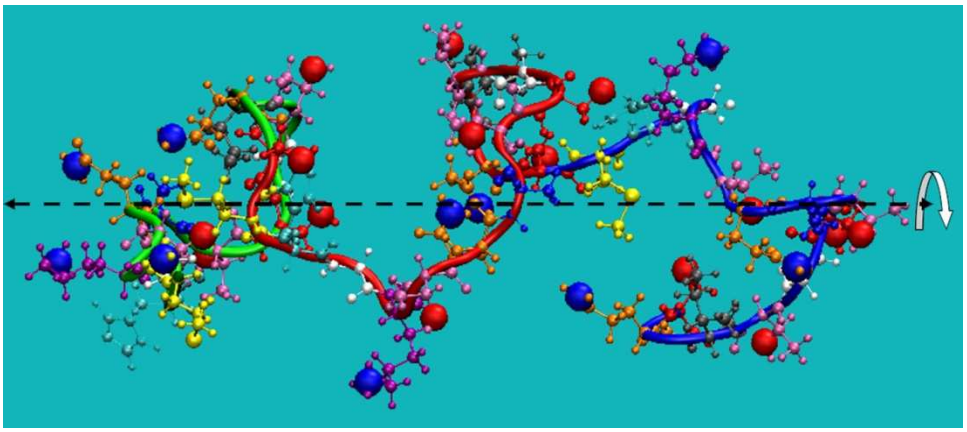

Figure S1. The possible binding sides on the NMR structure of 3rGBP$_1$. Distribution of hydrogen bonding donor/acceptor Lys, Thr, Gln, Ser sidechain and NH_3^+ main chain Nitrogen and Oxygen atoms which could hypothetically form non-bonding interactions with Au{111} surface. The dashed arrow represents the longest-axis of the peptide that has a one-fold symmetry. The blue and red spheres represent Nitrogen and Oxygen atoms, respectively, while the three different backbone colors stand for the repeating three units within the 3rGBP$_1$.

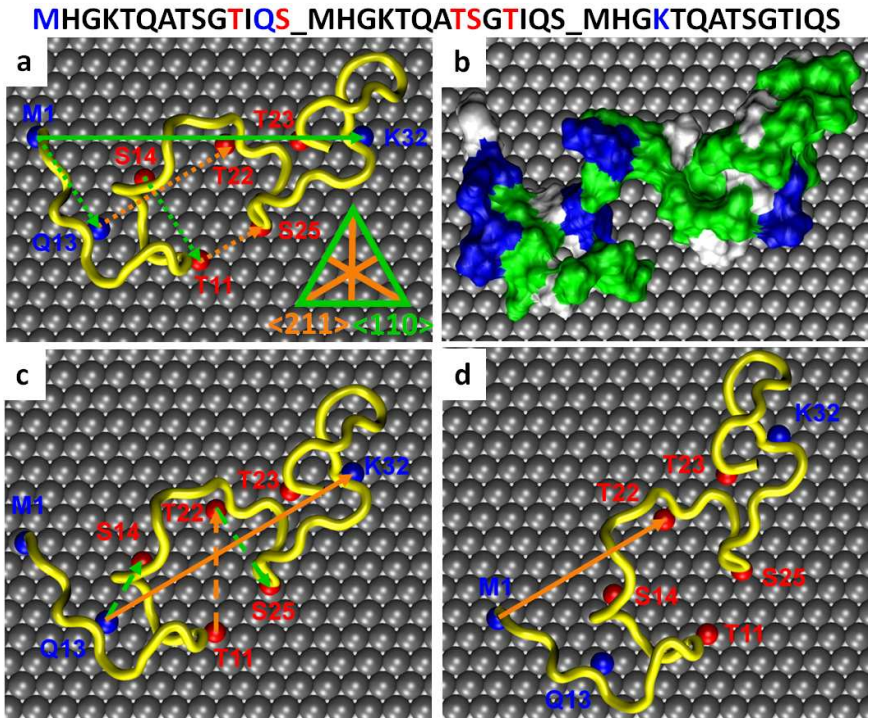

Figure S2. Top Three Correlating Orientations of 3rGBP$_1$ on the Au{111} surface lattice. The polypeptide backbone is represented in ribbon format (yellow), with the hetero-atoms of the docking surface residues (M1, T11, Q13, S14, T22, S23, T25, and K32) represented as space-filling atoms in either blue (N) or red (O). The crystallographic directions of the gold surface lattice for specific docking surface atoms are presented in green (<110>) or orange (<211>), depending on their correspondence with the appropriate Miller indices of the lattice. (a) The highest correlating orientation of 3rGBP$_1$ on gold surface lattice and space-filling representation in (b); (c)-(d) represent the second and third highest correlating orientations of 3rGBP$_1$ on Au{111}, respectively.

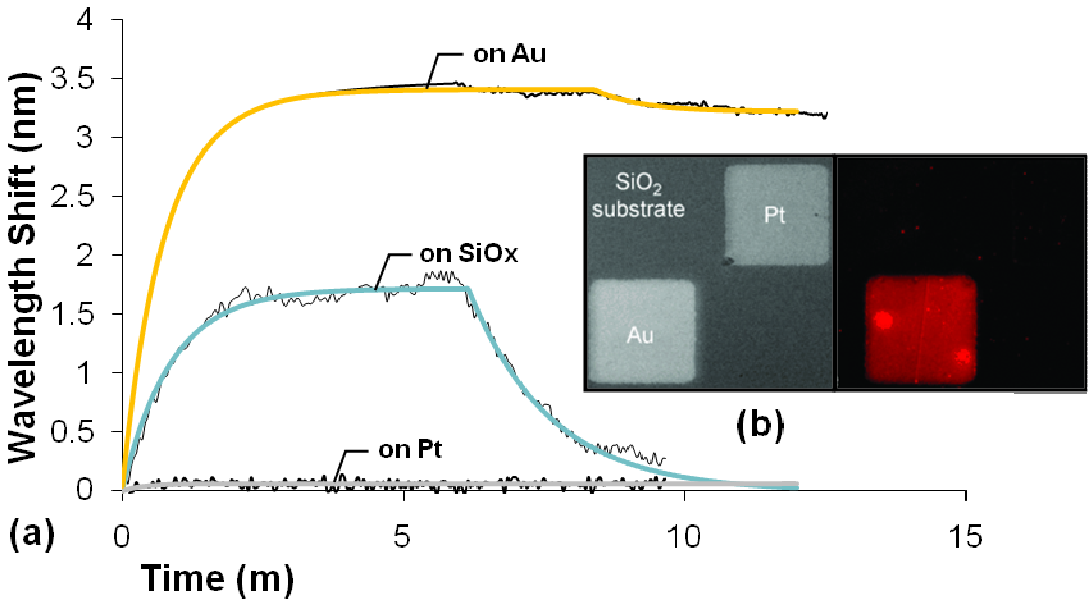

Figure S3. Material binding specificity of 3rGBP$_1$ on Au, silica and Pt substrates as determined by surface plasmon resonance spectroscopy (a) and fluorescence microscopy (b). (a) The Langmuir fitting of the SPR analyses of 3rGBP$_1$ on gold, platinum, and silica surfaces at 4 ug/mL concentration show specificity towards gold, but not towards SiO_x or Pt. Here the shift in the absorbance wavelength, due to adsorption on the Au surface, is plotted against time. The SPR measurements of 3rGBP$_1$ in (a) were made using an instrument (Kretschmann configuration) developed by the Radio Engineering Institute, Czech Republic. Lyophilized 3r-GBP1 was diluted in phosphate buffer (1:3 mixture of 10 mM KH_2PO_4, 10mM K_2HPO_4 and 100 mM KCl) with a 4ug/ml concentration. Peptide solutions were pumped over gold, platinum and silica substrate surfaces seperately and the adsorption of the peptide on these substrates individually was monitored in real-time. After the peptide-binding reached equilibrium on the surface, buffer (without peptide) was pumped over the substrate to observe its desorption from the surface. (b) The specificity of 3rGBP$_1$ is also demonstrated by fluorescence microscopy experiments. Here the biotinylated 3rGBP$_1$ is directed assembled onto the square Au micropatterns rather than either the substrate Si (covered with its native oxide) or Pt squares. To observe this, in (b), the CdSe quantum dots (QD, red) are surface functionalized by strepavidin

(SA). They are directed assembled on the Au surface because of the presence of biotinylated 3rGBP1 present at this site only, resulting in the red contrast, as observed by FM.

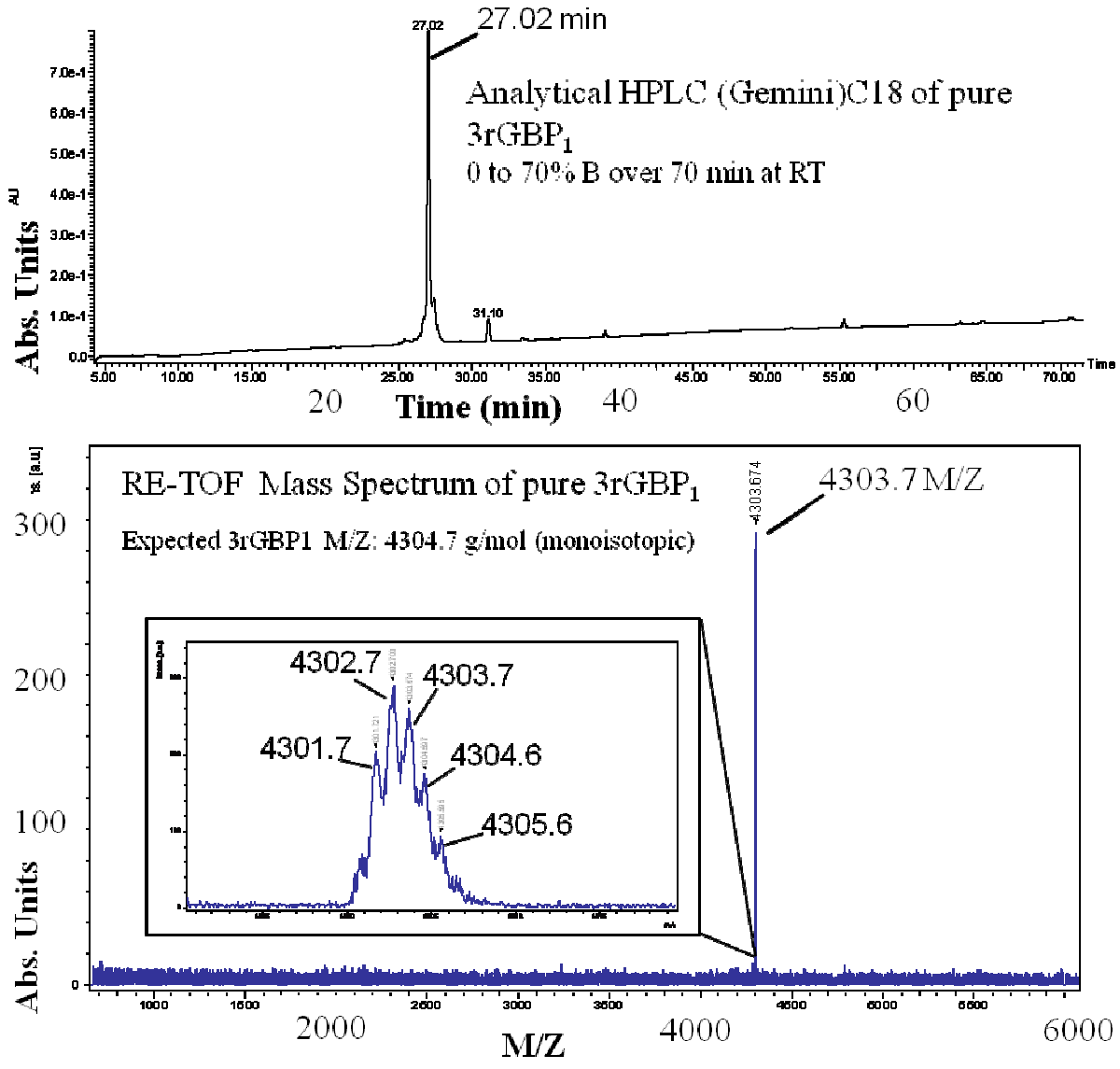

Figure S4. HPLC and RETOF-MS characterization of synthesized and purified 3rGBP$_1$.

2. Supplementary Tables

Table S1: The lateral distances between the points of the peptide where the hetero-atom interacts with the gold lattice; Hydrogen bonding donor/acceptor Lys, Thr, Gln, Ser sidechain and NH_3^+ main chain Nitrogen and Oxygen atoms.

(Å)	**M(1)**	**T(11)**	**Q(13)**	**S(14)**	**T(22)**	**S(23)**	**T(25)**	**K(32)**
M(1)	0	21.74	11.77	12.25	20.06	28.07	25.75	34.71
T(11)		0	11.32	10.52	12.72	16.77	7.58	21.85
Q(13)			0	7.28	16.31	23.58	17.45	29.89
S(14)				0	9.40	17.17	13.51	23.73
T(22)					0	8.04	9.58	14.69
S(23)						0	10.08	6.65
T(25)							0	14.38
K(32)								0

Table S2: The nearest-neighbor atomic distances on Au{111} surface.

Direction	**Distance (Å)**
<110>	2.884
<211>	4.995
<110>	5.767
<321>	7.630
<303>	8.651
<211>	9.990
<413>	10.397
<220>	11.535
<531>	12.570
<541>	13.215

Table S3: The spatial match between the 3rGBP$_1$ conformed architecture and the Au{111} surface lattice structure.

Direction	# of Alignments	Alignments
<110>	3	M(1)-T(22), M(1)-K(32), T(25)-K(32)
<211>	2	M(1)-T(22), Q(13)-K(32)
<110>	1	M(1)-K(32)
<321>	1	T(11)-T(25)
<303>	1	M(1)-K(32)
<211>	2	M(1)-T(22), Q(13)-K(32)
<413>	0	-
<220>	1	M(1)-K(32)
<531>	0	-
<541>	0	-