Figure S1. Radial distribution function of chlorides around the center-of-mass of the imid-10 micelle in the bulk aqueous phase ($\xi = 60 \text{ Å}$). A large peak at $\sim 16 \text{ Å}$ indicates that chlorides are predominantly present around the micelle of radius $\sim 16 \text{ Å}$. The RDF has been truncated at half of the box length.
Figure S2. Radial distribution function (RDF) between water-oxygen and chloride ions at various locations ($\xi = 10 \text{ Å}, 15 \text{ Å}, 35 \text{ Å}, 60 \text{ Å}$) of the imid-10 micelle from the metal surface. The peaks around 3 Å and 5 Å show that the chloride ions are highly solvated with water molecules. It has to be noted that the RDFs are deviating from 1 at large distances for the cases when the micelle is close to the surface. This is because when the micelle is close to the surface, the RDF histogram bins extend beyond the metal lattice where there are no water or chloride atoms.
Figure S3. Density profile of water as a function of distance from the gold surface (a) in the absence of a micelle, and (b) when the imid-10 micelle is located at different locations (ξ = 10 Å, 20 Å, 30 Å) from the metal surface. In the adsorbed state (ξ = 10 Å), the first peak in the density of water decreases because many water molecules are liberated from the metal surface.
Figure S4. Snapshots showing arrangement of constituent molecules in (a) the non-rigid, and (b) the rigid imid-10 micelles when the micelles are constrained at 17 Å (free energy peak for rigid micelle) from the metal surface. In the non-rigid micelle, the molecules are able to re-arrange themselves in order to interact more strongly with the metal surface, while this is not possible in the case of a rigid micelle. Adsorbed layers of water on the metal surface are also clearly visible.
Figure S5. Distribution of polar head groups of the molecules that comprise the rigid and non-rigid imid-10 micelles when the micelles are constrained at 17 Å from the metal surface. A large peak around 3 Å in the case of non-rigid micelle shows that some surfactant molecules now lie flat on the surface so that their head groups are in the vicinity of the metal surface. As opposed to this, the head groups in the rigid micelles are more uniformly distributed.
Figure S6. Radial distribution function of water-oxygen around the center-of-mass (COM) at various locations ($\xi = 10$ Å, 15 Å, 20 Å, 60 Å) of (a) the non-rigid imid-10 micelle, and (b) the rigid imid-10 micelle. It is observed that when the non-rigid micelle disintegrates at $\xi = 10$ Å, the water molecules come closer to the COM. Kindly note that for $\xi = 10$ Å, 15 Å and 20 Å, the RDF histogram bins extend beyond the metal lattice. Therefore, the RDF profiles have been properly normalized by multiplying the factor, $f = \frac{\text{Volume of a complete spherical RDF bin}}{\text{Volume of spherical cap of RDF bin outside the metal lattice}}$. 

\[ V_{\text{capped}} = \frac{1}{6} \pi r^2 (3h - r) \] 

\[ V_{\text{full}} = \frac{4}{3} \pi r^3 \] 

\[ f = \frac{V_{\text{capped}}}{V_{\text{full}}} \]
Figure S7. Radial distribution function of chlorides around the protonated nitrogen atoms at different locations ($\xi = 10 \, \text{Å}, 15 \, \text{Å}, 25 \, \text{Å}$) of the imid-10 micelle from the metal surface. When the micelle just adsorbs on to the surface ($\xi = 15 \, \text{Å}$) before disintegrating, the peaks in the RDF show a decrease which suggests that the counterions are unable to completely surround the micelle. After the disintegration of the micelle ($\xi = 10 \, \text{Å}$), the counterions are again observed close to the nitrogen atoms resulting in increase in the RDF peaks.
Figure S8. Angular orientation of a vector joining an atom at the center and an atom on the periphery of the rigid imid-10 micelle with respect to the vector normal to the metal surface. The large variations in the angle show that the micelle is sampling many different orientations. Only when the micelle is close to the metal surface, the orientations get restricted. The red point at 13 Å corresponds to the orientation when the micelle is at the metal surface.
**Movie S1.** Disintegration of the imid-10 micelle at the gold-water interface. The initial configuration is shown with the micelle at ~15 Å from the surface. The movie is captured for 160 ns. Disintegration of the micelle is seen by gradual loss of surfactant molecules in the proximity of the metal surface followed by their adsorption with their molecular axis lying parallel to the metal surface. Water molecules and chloride ions are not shown in the movie for clarity of disintegrating molecules.

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