Supplementary material for:

*Hydrophobic coating- and surface active solvent-mediated self-assembly of charged gold and silver nanoparticles at water-air and water-oil interfaces*

Lijun Xu,\textsuperscript{a} Guobin Han,\textsuperscript{b} Jiawen Hu,\textsuperscript{a} Yan He,\textsuperscript{a} Jiangao Pan,\textsuperscript{a} Yongjun Li,\textsuperscript{a} and Jiannan Xiang\textsuperscript{a}

\textsuperscript{a} State Key Laboratory for Chemo/Biosensing and Chemometrics, Biomedical Engineering Center, and College of Chemistry and Chemical Engineering, Hunan University, Changsha 410082, China

\textsuperscript{b} State Key Laboratory for Physical Chemistry of Solid Surfaces and Department of Chemistry, Xiamen University, Xiamen 361005, China

* Corresponding author: Dr. Jiawen Hu, Fax: +86-0731-8821740 E-mail: jwhu@hnu.cn

Dr. Yongjun Li, Fax: +86-0731-8821904 E-mail: liyje@hnu.cn
S1. Scheme for Preparation and Transfer of the Nanoparticle Films Formed at the Water-Air or Water-Oil Interface

![Scheme for Preparation and Transfer of the Nanoparticle Films Formed at the Water-Air or Water-Oil Interface](image)

S2. Contact Angle Measurements on Vacuum-Evaporated Au Films.

Figure S1. Photographs of 5 μL water drops on vacuum-evaporated Au films without (1) and with 28.6 volume % of methanol (2), ethanol (3), 2-propanol (4), and acetone (5), respectively.

S3. Correction of Zeta Potentials

Zeta potentials were calculated from the measured electrophoretic mobilities using the Smoluchowski equation

\[ \mu = \frac{\varepsilon \varepsilon_0 \zeta}{\eta} \]
where $\mu$ is electrophoretic mobility, $\eta$ is medium viscosity, $\varepsilon_r$ is medium dielectric constant, $\varepsilon_0$ is vacuum permittivity, and $\zeta$ is zeta potential.

Because the zeta potential analyzer contains only the parameters of pure media, a series of zeta potentials $\zeta_1$ were first obtained by using the dielectric constant $\varepsilon_{r,1}$ and the viscosity $\eta_1$ of pure water with increase of the proportion of organic solvent.

$$\zeta_1 = \mu_1 \frac{\eta_1}{\varepsilon_{r,1} \varepsilon_0}$$

Then, the zeta potentials were corrected for the change in the dielectric constant and the viscosity with addition of organic solvent to water to obtain the real zeta potentials $\zeta_2$ of the colloidal particles dispersed in an organic solvent-water binary medium.

$$\zeta_2 = \frac{\varepsilon_{r,1} \eta_2}{\varepsilon_{r,2} \eta_1} \zeta_1$$

Figure S2. Dielectric constant (left panel) and viscosity (right panel) of organic solvent-water mixtures as a function of the mass fraction of the organic solvent at 25°C. The insert in the left panel shows the dielectric constant of the organic solvent-water mixtures as a function of the mole ratio of the organic solvent at 25°C. Dielectric constant data used to fit the curves in the left panel were taken from *J. Am. Chem. Soc.*, 1932, 54, 4126 and viscosity data used to fit the curves in the right panel were measured using an Ubbelohde Viscometer.
Figure S3. Left panel shows the ζ potentials of 40 nm gold colloids in organic solvent-water mixtures of methanol (■), ethanol (●), 2-propanol (▲), and acetone (▼). These ζ potentials were converted from the measured electrophoretic mobilities by using the dielectric constant and viscosity of pure water. The corresponding corrected ζ potentials considering the change in dielectric constant and viscosity with addition of organic solvent to water are shown in the right panel.

Figure S4. The left panel shows the ζ potentials of Ag colloids in organic solvent-water mixtures of methanol (■), ethanol (●), 2-propanol (▲), and acetone (▼), respectively. These ζ potentials were converted from the measured electrophoretic mobilities by using the dielectric constant and viscosity of pure water. The corresponding corrected ζ potentials considering the change in dielectric constant and viscosity with addition of organic solvent to water are shown in the right panel.