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

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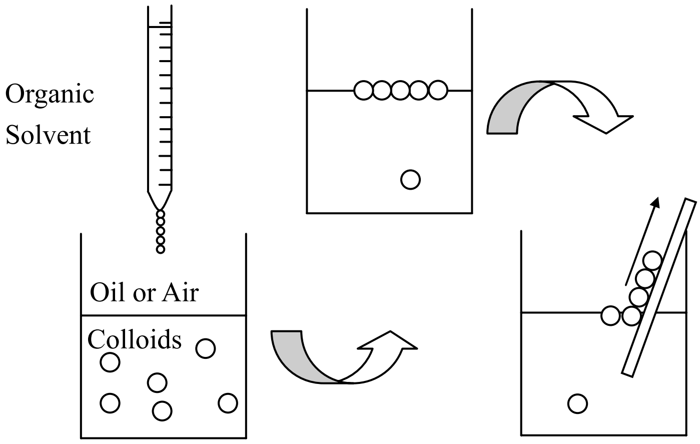
S1. Scheme for Preparation and Transfer of the Nanoparticle Films Formed at the Water-Air or Water-Oil Interface



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

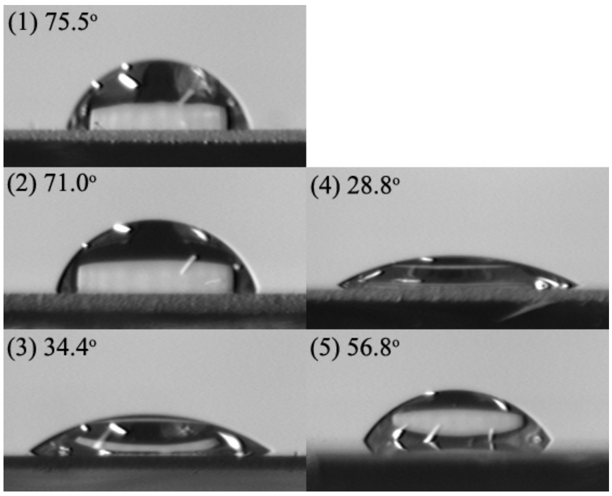


Figure S1. Photographs of 5 μL water drops on vacuum-evaporated Au films without (1) and with 28.6 volume % of methanol (2), ethanol (3), 2-propanol (4), and acetone (5), respectively.

S3. Correction of Zeta Potentials

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

$$\mu = \frac{\varepsilon_{\mathrm{r}}\varepsilon_0}{\eta}\zeta$$

where μ is electrophoretic mobility, η is medium viscosity, ε_r is medium dielectric constant, ε_0 is vacuum permittivity, and ζ is zeta potential.

Because the zeta potential analyzer contains only the parameters of pure media, a series of zeta potentials ζ_1 were first obtained by using the dielectric constant $\varepsilon_{r,1}$ and the viscosity η_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 ζ_2 of the colloidal particles dispersed in an organic solvent-water binary medium.

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

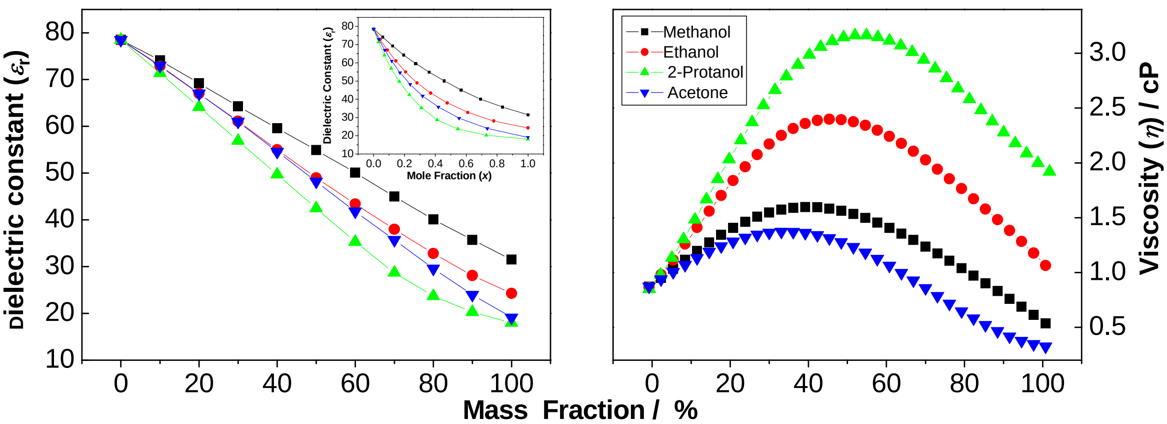


Figure S2. Dielectric constant (left panel) and viscosity (right panel) of organic solvent-water mixtures as a function of the mass fraction of the organic solvent at 25°C. The insert in the left panel shows the dielectric constant of the organic solvent-water mixtures as a function of the mole ratio 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.

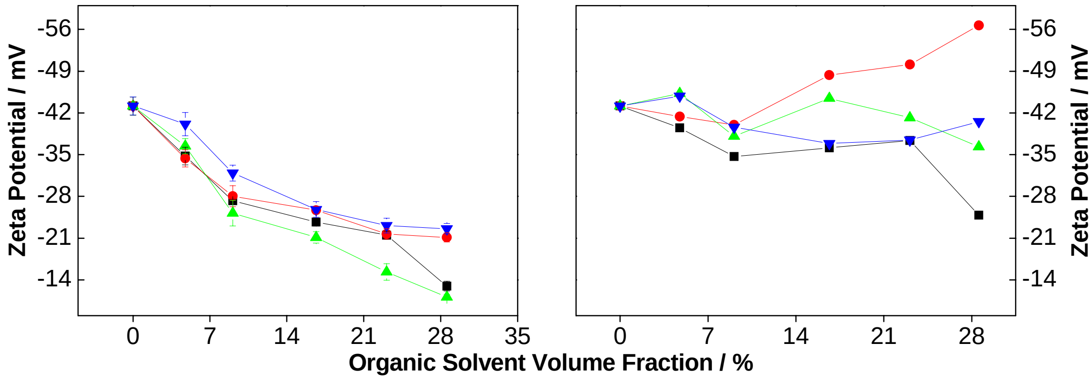


Figure S3. Left panel shows the ζ potentials of 40 nm gold colloids in organic solvent-water mixtures of methanol (■), ethanol (●), 2-propanol (▲), and acetone (▼). These ζ potentials were converted from the measured electrophoretic mobilities by using the dielectric constant and viscosity of pure water. 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.

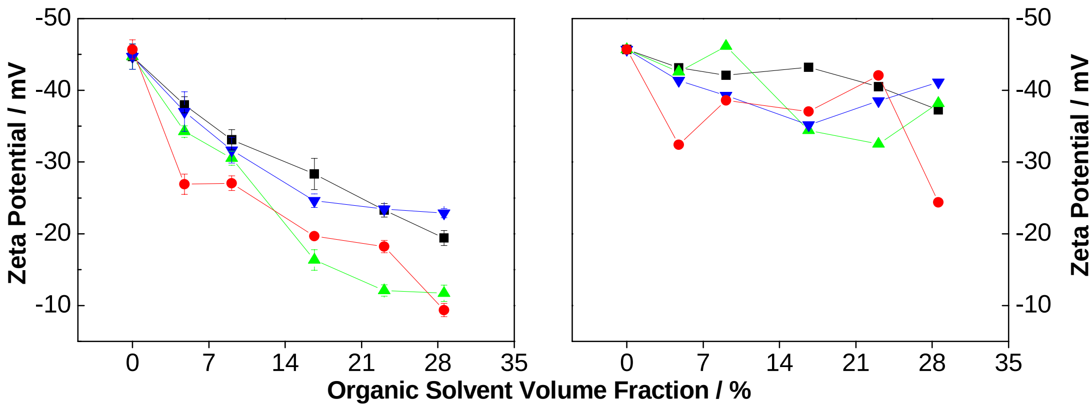


Figure S4. The left panel shows the ζ potentials of Ag colloids in organic solvent-water mixtures of methanol (■), ethanol (●), 2-propanol (▲), and acetone (▼), respectively. These ζ potentials were converted from the measured electrophoretic mobilities by using the dielectric constant and viscosity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