**Supporting Information for** 

## Single-step Preparation of Two-dimensionally Organized Gold Particles via Ionic Liquid/Metal Suputter Deposition

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Abbreviation	Molecular Structure	Name
HyEMI-BF4	+/ OH F-B-F F-B-F F	1-(2-hydroxyethyl)-3- methylimidazolium tetrafluoroborate
EMI-BF4	+ $N$ $P$	1-ethyl-3-methylimidazolium tetrafluoroborate
BMI-BF4	+ N N F- F-B-F F	1-butyl-3-methylimidazolium tetrafluoroborate
OMI-BF4	+ / F- F-B-F F-B-F F	1-octyl-3-methylimidazolium tetrafluoroborate
HyEMI-TFSA	$+ \bigvee_{N \searrow N} \bigvee_{OH} OH \qquad F \bigvee_{O' \stackrel{S \searrow O}{\sim} O' \stackrel{S \searrow O}{\sim} O' \stackrel{S \searrow O}{\sim} O'$	1-(2-hydroxyethyl)-3- methylimidazolium bis(trifluoromethanesulfonyl)amide
EMI-TFSA	$+ \bigwedge_{N \searrow N} N \xrightarrow{F \bigvee_{i=1}^{F} F_{i=1}^{-F} F_{i=1}^{-F}$	1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)amide
TMPA-TFSA	$\begin{array}{c} \begin{array}{c} & & \\ & & \\ & & \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} $	<i>N,N,N-</i> trimethyl- <i>N-</i> propylammonium bis(trifluoromethanesulfonyl)amide
Ch-TFSA	$ \begin{array}{c} \begin{array}{c} & & \\ & & \\ & & \\ & & \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array}{c} \\ \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \begin{array} \\ \end{array} \\ \end{array} \\ \end{array} \\ \end{array} \\$	2-hydroxyethyl- <i>N</i> , <i>N</i> , <i>N</i> - trimethylammonium bis(trifluoromethanesulfonyl)amide
SBMI-TFSA	$\xrightarrow{+} N \xrightarrow{OH} F \xrightarrow{F} F \xrightarrow{F} F \xrightarrow{F} F$	1-(4-sulfobutyl)-3- methylimidazolium bis(trifluoromethanesulfonyl)amide
EMI-EtSO4	$ \begin{array}{c} + \sqrt{1} \\ & & & & \\ & & & \\ & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & \\ & & & & \\ & & & & \\ & & & & \\ & & &$	1-ethyl-3-methylimidazolium ethylsulfate
EMI-Ac		1-ethyl-3-methylimidazolium acetate
BMI-SCN		1-butyl-3-methylimidazolium thiocyanate
BMI-lactate		1-butyl-3-methylimidazolium (L)-lactate
HyEA-formate		(2-hydroxyethyl)ammonium formate

## Table S1Molecular structures of RTILs used.



**Fig. S1** Extinction spectrum of Au nanoparticles uniformly dispersed in EMI-BF4. The particles were prepared by sputter deposition of Au on EMI-BF4 for 300 s with a discharge current of 10 mA under an Ar pressure of 20 Pa.



**Fig. S2** TEM images of Au monoparticle films prepared by sputter deposition of Au on HyEMI-BF4 for 30 s with a discharge current of 40 mA under an Ar pressure of 20 Pa. Panel b is a magnified image of panel a. The bars in pictures represent a length of 20 nm.



**Fig. S3** (a,b) TEM images of Au particles contained in multilayer particle films prepared by Au sputter deposition HyEMI-BF4 for 600 (a) and 1200 s (b). The bars in pictures represent a length of 20 nm. Samples for TEM measurements were prepared by pulverizing multilayer particle films with sonication. (c) Schematic illustration of the multilayer film formation of Au particles along with collapse of monoparticle layer by the crystal growth of individual particles on RTIL surface.



**Fig. S4** FT-IR spectrum of an Au monoparticle film immobilized on an Si substrate. The film was prepared by Au sputter deposition onto HyEMI-BF4 for 300 s.



**Fig. S5** Top view photographs of metal monoparticle layer films of Ag (a,b), Pd (c,d), and Pt (e,f) deposited on the HyEMI-BF4 surface (a,c,e) and corresponding TEM images of metal films transferred on TEM grids (b,d,f). The films of Ag and Pd were prepared by sputter deposition of corresponding metals with discharge current of 10 mA under an Ar pressure of 20 Pa, while Pt films were prepared by Pt sputtering with current of 40 mA at the same Ar pressure. The sputtering time for each case was 300 s.