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Extremely fast Au–Ag alloy–dealloy associated reversible plasmonic modifications in SiO₂ films†

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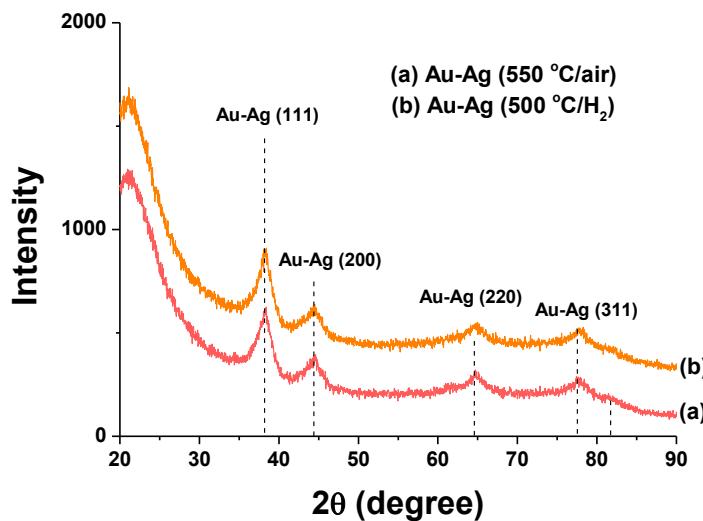


Fig S1. GIXRD patterns of the final heat-treated Au–Ag alloy nanoparticles incorporated SiO₂ films: (a) 550 °C (air), (b) 500 °C (10% H₂–90% Ar). The vertical lines with relative intensities are the positions of fcc Au/Ag reflections. The patterns were recorded at room temperature (22 °C).

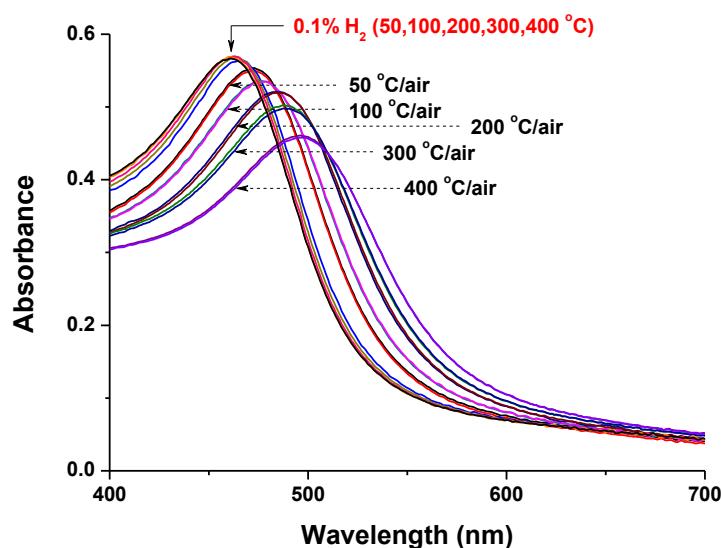


Fig S2. Optical spectra of the Au–Ag alloy film recorded in air–0.1% H₂–air cycles at the temperature range of 50–400 °C. Recording conditions of the spectra are indicated in the body of the figure.

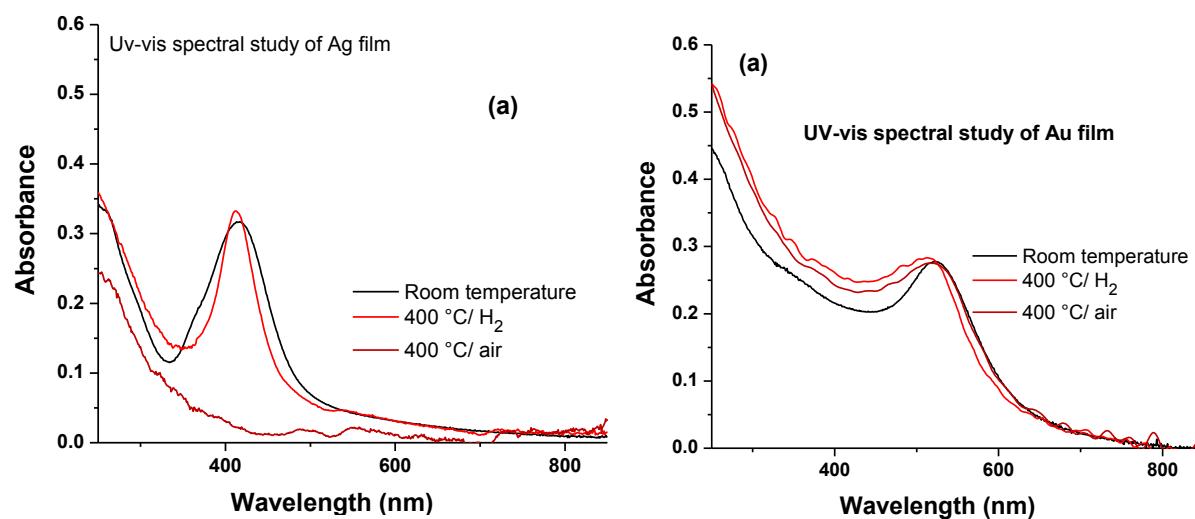


Fig S3. Optical spectra of the pure Ag (a) and Au (b) doped films in air and reducing (10% H₂–90%) atmospheres at 400 °C.

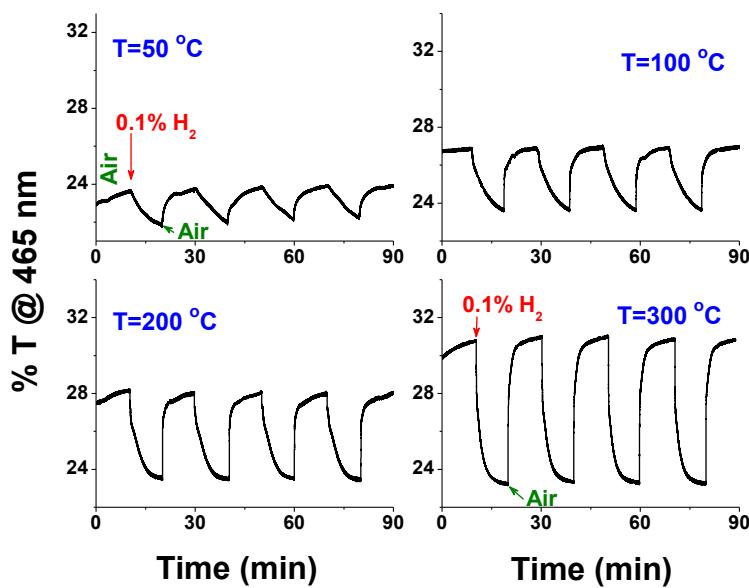


Fig. S4 Dynamic change of SPR transmittance of the Au–Ag (1:1) film when exposed to air–0.1% H₂–air cycles. The test was performed at 50, 100, 200 and 300 °C as marked in the figure and optical transmittance was recorded at 465 nm.

Table S1 SPR peak positions and corresponding alloy composition of the Au, Ag and Au/Ag embedded SiO₂ films.

Film	SPR (nm)	Alloy composition (calculated based on SPR position) ²⁴
Au (500 °C; 10% H ₂ –90% Ar)	524	Au
Au–Ag (550 °C; air)	496	Au ₇₄ Ag ₂₆
Au–Ag (500 °C; 10% H ₂ –90% Ar)	466	Au ₄₈ Ag ₅₂
Ag (500 °C; 10% H ₂ –90% Ar)	415	Ag

Table S2 Refractive index (RI) values of the final heat-treated films deposited on one side polished Si wafer measured at room temperature and 400 °C in air and reducing (10% H₂–90% Ar) atmospheres.

Films	Refractive index at $\lambda = 800$ nm		
	Room temperature	400 °C (air)	400 °C (10% H ₂ –90% Ar)
Pure SiO ₂	1.417±0.003 ^a	1.324±0.002	1.328±0.002
Au doped SiO ₂	1.431±0.003	1.332±0.002	1.330±0.002
Ag doped SiO ₂	1.438±0.003	1.343±0.002	1.340±0.002
Au/Ag doped SiO ₂	1.449±0.003	1.380±0.002	1.378±0.002

^aRI value at 633 nm (λ): 1.420±0.003.

Table S3 Binding energy (Ag 3d_{5/2}) values of pure Ag and Au–Ag incorporated films heat-treated under different conditions.

Film	SPR (nm)	Composition	Ag 3d _{5/2} (eV)
Ag(0) (500 °C; 10% H ₂ –90% Ar)	415	Ag	368.1
Au–Ag (500 °C; 10% H ₂ –90% Ar)	466	Au ⁰ ₄₈ Ag ⁰ ₅₂	367.2
Au–Ag (300 °C; air)	490	Au ⁰ ₄₈ Ag ⁰ ₂₂ Ag ⁺ ₃₀	367.7