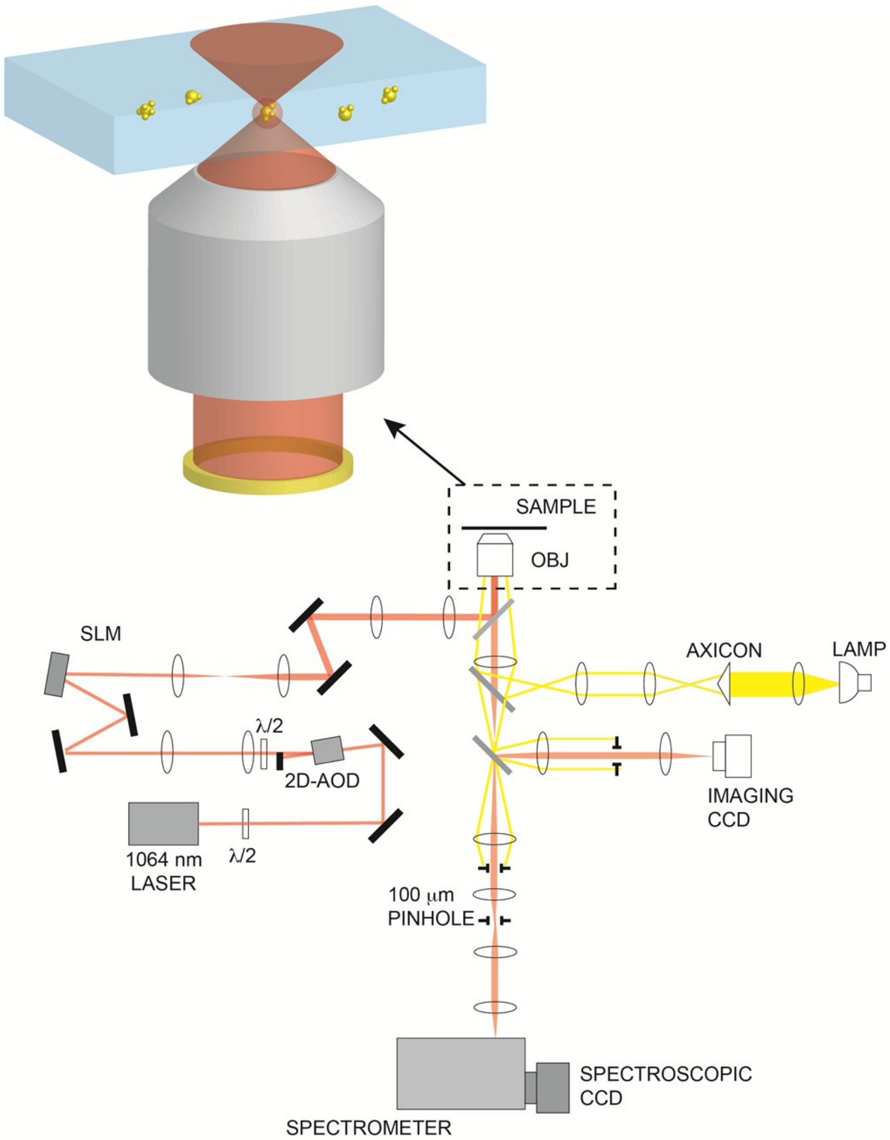

Figure S1. Schematic diagram of the optical tweezers combined with dark-field imaging and spectroscopy. Trapping is achieved using an infrared emitting solid state laser that is focused through a high-numerical aperture objects into a fluid filled chamber containing the nanoparticles. The heating in the trap is controlled by the strength of the trapping laser. Dark field imaging is achieved by illuminating back aperture of the objective with a ring illumination and then collecting only the scattered light through a confocal aperture; the scattered light is passed to a spectrometer to record the spectrum of the trapped object.

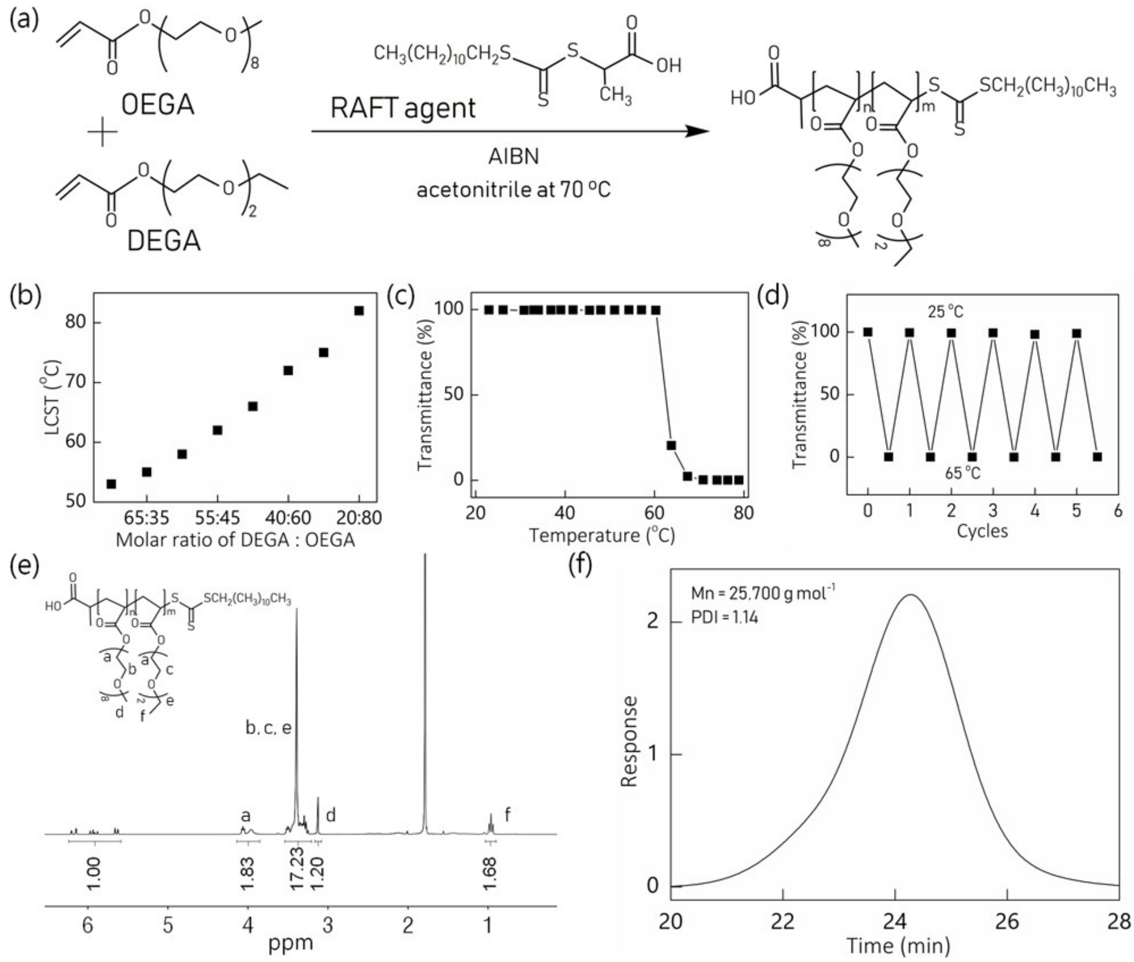

Figure S2. Thermoresponsive copolymer synthesis and characterization. (a) Schematic presentation of poly(DEGA-*co*-OEGA) synthesis *via* RAFT polymerization. (b) The LCST measurement of poly(DEGA-*co*-OEGA) at a different molar ratio of DEGA:OEGA (The data was shown in Table S1). (c) Thermal Profile (2 °C/min) of poly(DEGA-*co*-OEGA) (entry 9, M_w = 25,700 g/mol, [DEGA]:[OEGA] = 60:40) in Milli-Q water as measured by UV-Vis measurement (λ = 500 nm, [copolymer] = 5.0 mg/mL). The LCST of copolymer is around 60 °C. (d) Thermal cycling of poly(DEGA-*co*-OEGA) (entry 9) after 6 cycles of heating and cooling. (e) NMR data of poly(DEGA-*co*-OEGA) (entry 9). The ^{1}HNMR spectra and peak labels for (top panel) before purification (f) GPC data of poly(DEGA-*co*-OEGA) (entry 9).

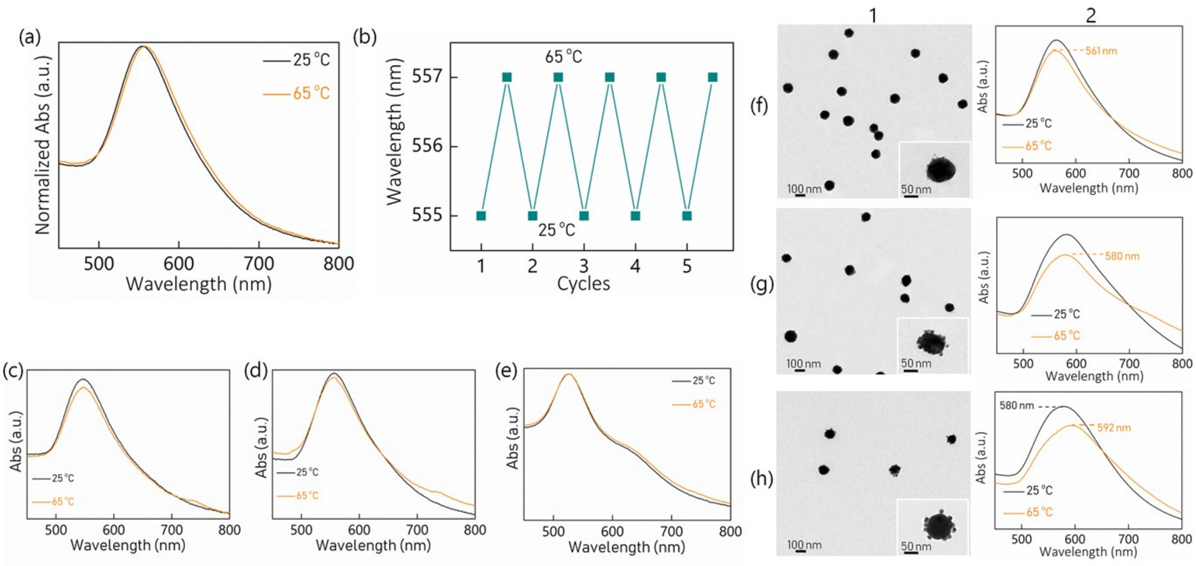

Figure S3. (a)-(b) Representative UV-Vis characterizing the thermal behaviour of the AuNP$_{80\ nm}$@poly(DEGA-*co*-OEGA) and thermal cycling of the AuNP$_{80\ nm}$@poly(DEGA-*co*-OEGA). (c)-(e) The effect of different amount of MPTMS: (c) 0.5 µL, 1% MPTMS, (d) 1 µL, 1% MPTMS and (e) 3 µL, 1% MPTMS ethanol solution on the responsive behaviour of the core$_{80\ nm}$-satellite$_{40\ nm.}$ (e)-(h) TEM images (Panel 1) and UV-Vis spectra (Panel 2) (e) for core$_{80\ nm}$-satellite$_{10\ nm}$; (f) core$_{80\ nm}$-satellite$_{15\ nm}$; (h) core$_{80\ nm}$-satellite$_{20\ nm}$.

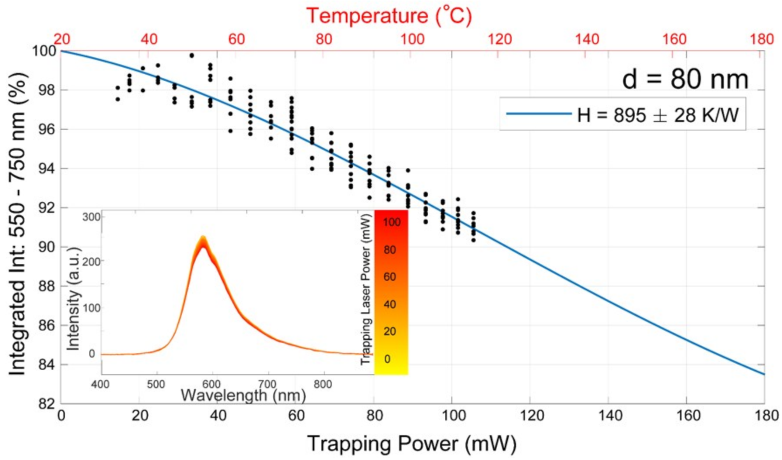

Figure S4. Backscattering spectra of 80 nm gold nanoparticles in the optical tweezers as the trapping power is cycled, 550 nm peak decreases in integrated intensity as the power is increased due to change in refractive index of surrounding water as it is heated. Change in integrated intensity vs trapping laser power is used to determine the temperature of the nanoparticles based on the temperature of the surrounding water. The measurements are used to give an indicative measure of the expected heating in the trap for the core-satellite nano-assemblies.

Table S1 Summary of copolymers used in this paper (Entry 9 was chosen as the copolymer to react with the AuNP).

Entry	Feed Ratio (mol %)		Reaction hours (h)	Reaction temperature (°C)	M_w (g/mol)	Temperature (LCST) (°C)
	DEGA	OEGA				
1	70	30	2	70	20,000	53
2	70	30	3	70	25,000	54
3	70	30	4	70	24,500	54
4	65	35	2	70	10,000	55
5	65	35	3	70	22,000	56
6	65	35	4	70	20,500	56
7	60	40	2	70	10,000	58
8	60	40	3	70	22,400	58
9	60	40	4	70	25,700	58
10	55	45	2	70	10,000	62
11	55	45	3	70	17,100	62
12	55	45	4	70	28,180	62
13	50	50	2	70	12,100	66
14	50	50	3	70	15,218	65
15	50	50	4	70	24,857	66
16	40	60	2	70	15,121	72
17	40	60	3	70	24,578	72
18	40	60	4	70	26,173	73
19	35	65	2	70	25,821	75
20	35	65	3	70	20,125	75
21	35	65	4	70	29,466	75
22	20	80	2	70	18,080	82
23	20	80	3	70	24,198	83
24	20	80	4	70	26,917	82