

Figure 1. *Left:* the localized surface plasmon resonances of gold nanostructures (blue spectrum) are strongly coupled to excitons in light-harvesting complex II from green plants, LHCII, yielding new peaks above and below the energy of the uncoupled plasmon resonance (red spectrum). The absorption spectrum of LHCII is shown in green. *Right:* the coupling results from a linear combination of the plasmon and exciton states, which can be modelled as coupled harmonic oscillators to yield the splitting energy.



Figure 2. Arrays of nanostructures formed by interferometric lithography (a) are functionalised with monolayers of initiator-terminated adsorbates (b), enabling growth of poly(cysteine methacrylate) brushes via atom-transfer radical polymerisation (c). Attachment of N-hydroxysuccinimidyl ester derivatives of chlorophyll a (d) yields plexcitonic antenna complexes. The properties of the plexcitons are programmed by controlling the polymerisation time (molecular weight) grafting density and derivatisation of grafted polymers.



Figure 3. (a) In the strong coupling regime, macroscopically extended excited states are formed and the coupling energy depends on the concentration of excitons (chlorophylls) in the plasmon mode volume. By controlling the architectures of plexcitonic complexes, we can programme the coupling energy. Collapsed, reduced-density scaffolds (right-hand side) yield higher chlorophyll concentrations and hence a more pronounced avoided crossing than fully dense scaffolds, that yield lower exciton concentraions (left). (b) In the strong coupling regime, the scaled coupling energy (approximately equal to the Rabi energy) varies as the square root of the exciton T concentration, proportional to the fraction of repeat units in the polymer scaffold derivatised with chlorophyll.



Figure 4. Poly(a^{0.2}/_a¹⁰/_i) o acid methacrylate scaffolds display stimuluseres ponsive behavious enabling active control of plasmon-exciton coupling. By varying the temperature we can regulate the polymer swelling state 100 from swollen (23°C) to collapsed (34°C), switching the exciton density between high and low values and moving the system in and out of the strong coupling regime.

