



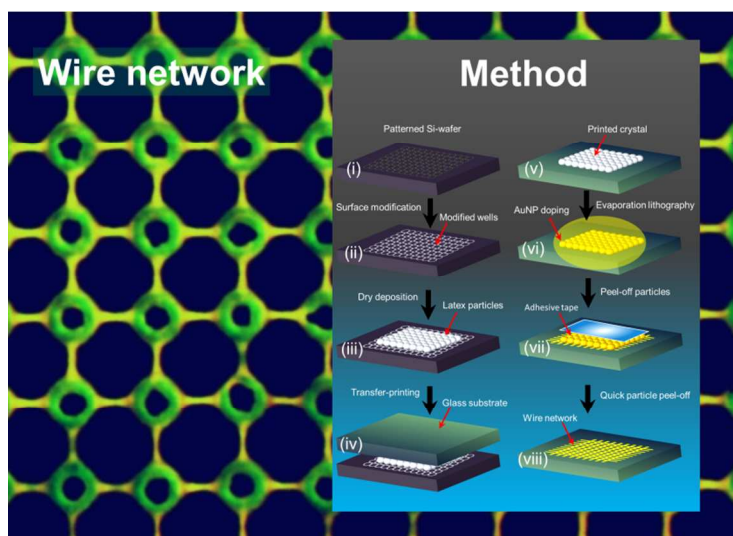
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Particle array template-based evaporation lithography is becoming increasingly important in micropatterning technologies.¹⁻⁴ The technique is based on using controlled solvent evaporation which leads to self-assembly of nanoparticles (dissolved in a suspension) on flat surfaces, driven by the reduction in the interfacial energy.⁵⁻⁸ The technique is considered as a viable alternative to overcome the challenges associated with transparent conductive films.⁹⁻¹³ In principle, the particle array template-based evaporation lithography is simple, cost effective and energy efficient. Improvements have been made in performance, by varying template design and investigating the evaporation mechanism.^{7,14,15} Nonetheless, the fabrication of square wire-networks via particle array-based evaporation lithography is quite challenging when the particle size approaches 1 μm . In earlier attempts, latex particle ($\geq 50 \mu\text{m}$) based evaporation lithography was highlighted, but the technique has not been ideally suitable for generating non-hexagonal metallic networks on an industrial scale.^{3,14} This was focused by fabricating defect-free photoresist templates over wafer-scale areas.⁷ Although this method generates metal wire-networks, it is still a challenge to reuse photoresist template to save large capital expenses involved in photolithography tools. Therefore, reusing the silicon templates repeatedly and to downscale the particle size in crystal preparations for the fabrication of square patterned wire-networks is highly desirable.

In this communication, we developed a hybrid methodology for fabricating gold wire networks by the combination of three major processes: 1) dry deposition of 2D monolayer of latex particles on a patterned silicon wafer in a square array arrangement, 2) lift-up of 2D monolayer template onto a flat glass or

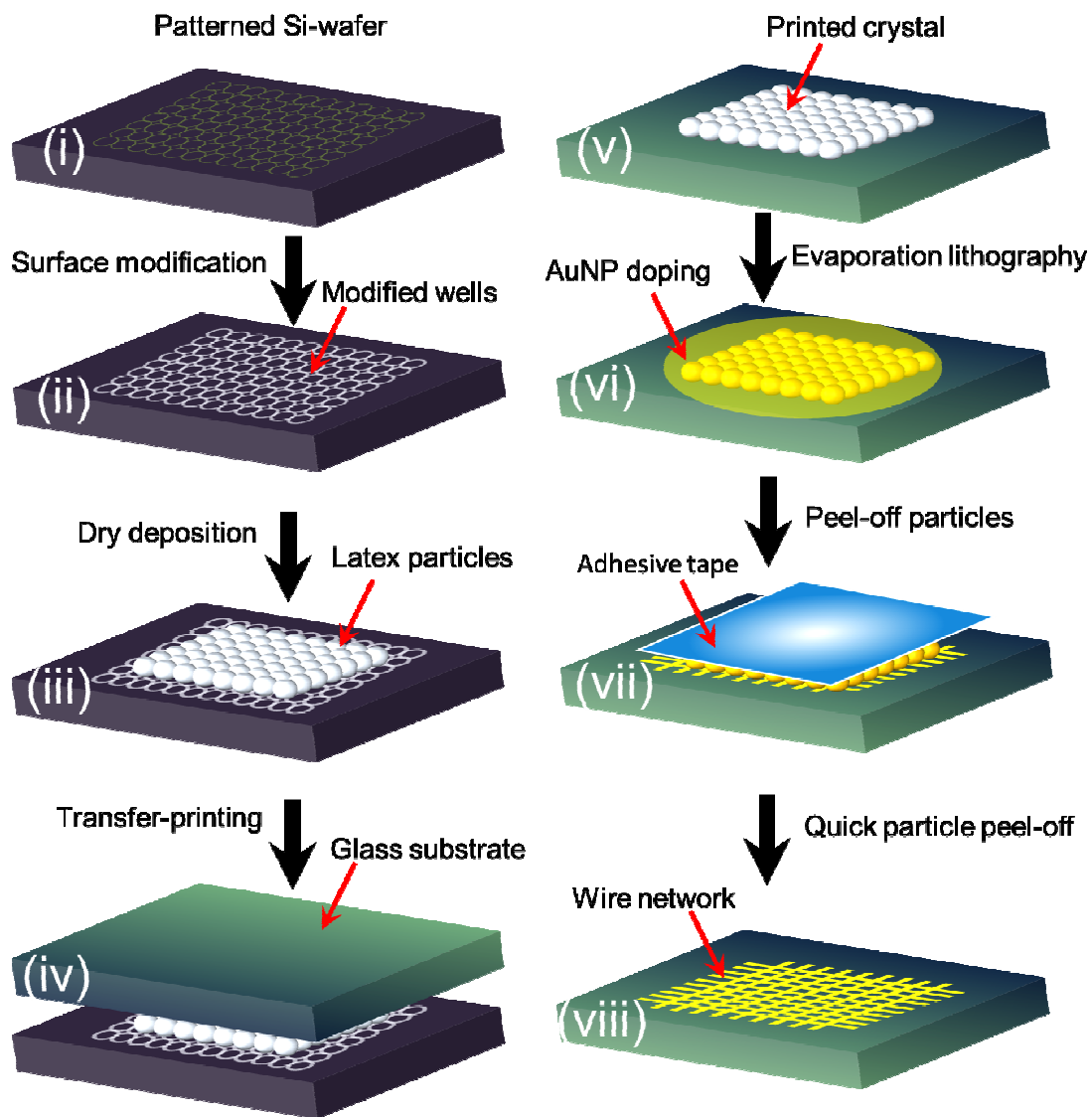


Fig. 1 Schematic diagram of the procedure employed in the fabrication of gold micro- and nanowire networks.

flexible substrate, and 3) evaporation lithography to undergo self-assembly of gold nanoparticles between the vertices of the 2D particle monolayer. One of the prime motivations behind this study is to down-scale the existing particle array template-based evaporation lithography process to fabricate connected gold wire networks at both micro- and submicron scale. Secondly, the idea of combining the patterned silicon wafer with lifted latex particle template creates an opportunity to clean and re-use the patterned wafer more often and thereby, saving fabrication time and resources. Finally, we illustrated the validity of this approach by creating an easy and high-speed approach to develop gold wire networks on a flexible substrate with a thin deposited adhesive. These advances will not only serve as a platform to scale up the production, but also demonstrated that the fabrication method can produce metallic wire networks of different scale and onto a variety of substrates.

The general experimental procedure adopted for the fabrication of gold wire networks is schematically presented in Fig. 1. The process begins with the manual dry deposition of spherical latex particles on a patterned silicon wafer with different pitch sizes (see Supporting Information, S1) to create 2D monolayer templates in a square symmetric arrangement. In this study, we used two different sizes of latex particles (5 and 1 μm in diameter) to prepare 2D monolayer templates. Instead of the natural hexagonal packing, the square symmetric arrangement of latex particles is chosen because it ensures larger interstitial spaces between the particles which promote the formation of stable network of liquid bridges as the de-wetting occurs in a square arrangement between every four particles. The liquid bridges are the precursors to the formation of large connected and regular networks of microwires. The size of wire-networks is tuned by the size of inter-particle spaces which solely depends on the size of latex particles used in the corresponding deposition process. Deposition of 5 μm latex particle-based template is prepared to fabricate microwire network, whereas, 1 μm latex particles produce nanowire network. We followed the dry deposition approach for latex particle ordering introduced by Khanh and Yoon.¹⁶ This approach of latex particle assembly into 1D and 2D arrays is highly advantageous over the conventional wet self-assembly techniques,¹⁷⁻²¹ since the approach does not depend on self-assembly in solution. The dry

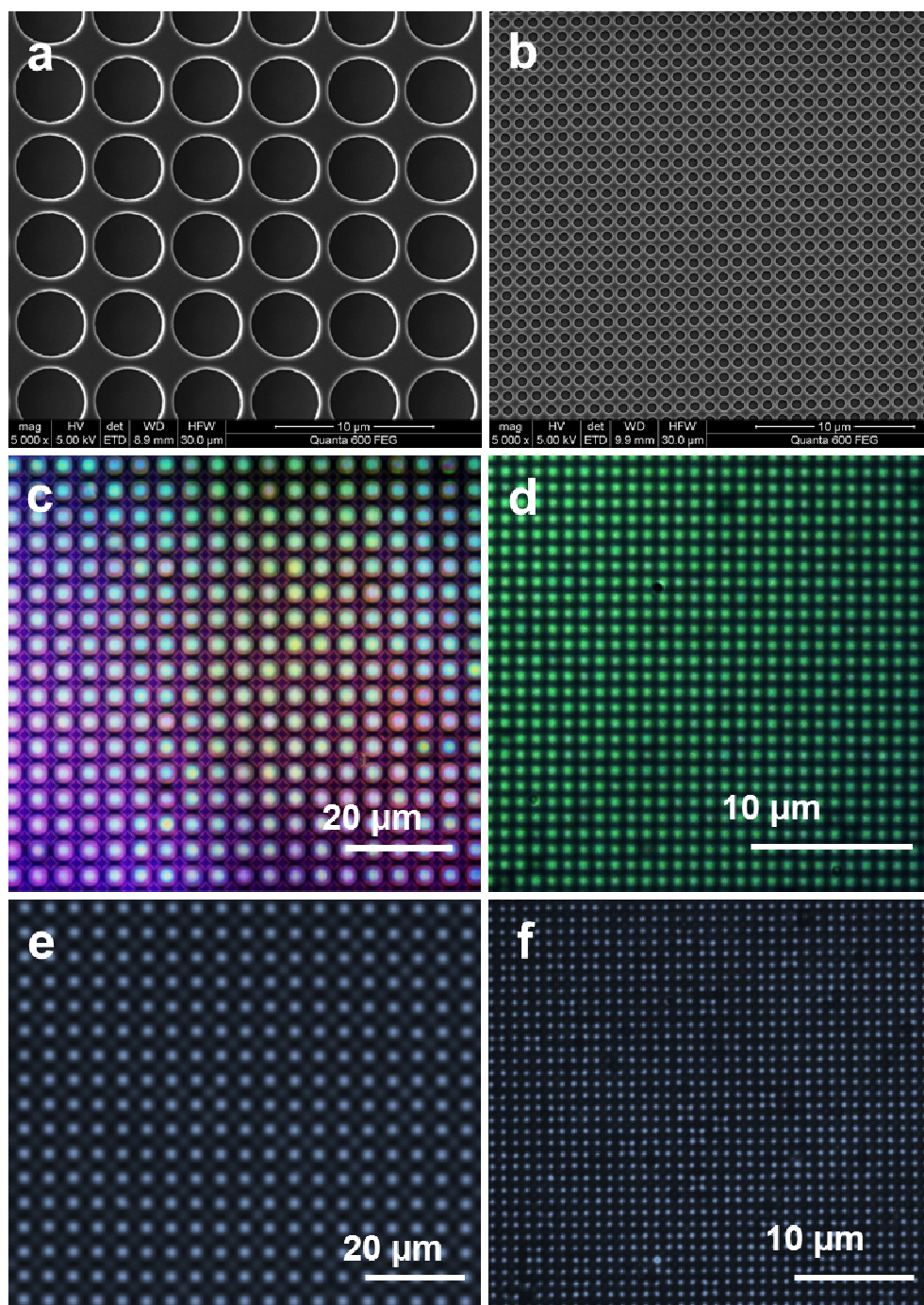


Fig. 2 Dry deposition and lift-up process. (a, c and e) represents the SEM image of silicon wafer used for the deposition of 5 μm latex particles, optical micrograph of 5 μm sized 2D monolayer latex particles deposited on silicon wafer and optical micrograph of lift-up of 5 μm size 2D monolayer on glass substrate respectively. Similarly, (b,d and f) shows the SEM image of silicon wafer used for the deposition of 1 μm latex particles, optical micrograph of dry of 1 μm sized 2D monolayer latex particles deposited on silicon wafer and optical micrograph of lift-up of 1 μm sized 2D monolayer on glass substrate respectively.

deposition technique involves repeated (forward and backward direction) rubbing of dry spherical colloidal particles into the patterned micro- and nanowell arrays and therefore, creating a very fast organization of the spherical colloidal particles into large and perfect 1D and 2D monolayer arrays. The freshly prepared elastomeric polydimethylsiloxane (PDMS) stamps are utilized to remove the randomly aggregated upper latex particles away from the main pattern. Fig. 2ab shows SEM images of patterned silicon wafers with two different pitch sizes on which we deposit 5 μm and 1 μm sizes of latex particles to create 2D monolayer latex templates for the preparation of micro- and nano wire networks respectively (Fig. 2cd and Supporting Information, S2). The latex particle suspensions used in this study were washed (2-3 times) by deionized water (D.I.), sonicated for 30 minutes in D. I. water to disperse the particles, dried under vacuum (overnight at room temperature) to remove the excess amount of surfactant that may cause particle aggregation during the deposition process. Prior to latex particle deposition, the silicon wafer was surface modified by trichloro(1*H*,1*H*,2*H*,2*H*-perfluorooctyl)silane (purchased from Sigma Aldrich) in a vacuum desiccator for 3 hours at room temperature, to facilitate an easy lift-up of latex particle template.

After the preparation of square symmetric 2D monolayer of latex particles, the process is followed by lift-up of latex particle template. The approach involves direct transfer of 2D monolayer of latex particles from patterned silicon wafer onto a rigid or flexible substrate without intermediate steps. Before lifting the particle template onto the glass substrate, the glass is plasma treated for 30 minutes for better lifting results. It is observed that the glass substrate which is not treated by plasma does not show any lifting capacity. After plasma exposure, the glass substrate is immediately pressed against the 2D monolayer on the silicon wafer on a flat surface. An appropriate pressure is evenly applied on the top to lift monolayer onto the glass substrate. This method successfully lifts 50-60 % of the overall 2D monolayer from silicon wafer onto the glass. Without using any glue or polymer/material deposition, the lift-up of latex particle crystal onto bare glass substrate by plasma treatment is a significant advancement. The undesired latex particles at the edges of the lifted crystal are removed by freshly prepared PDMS stamps by gently

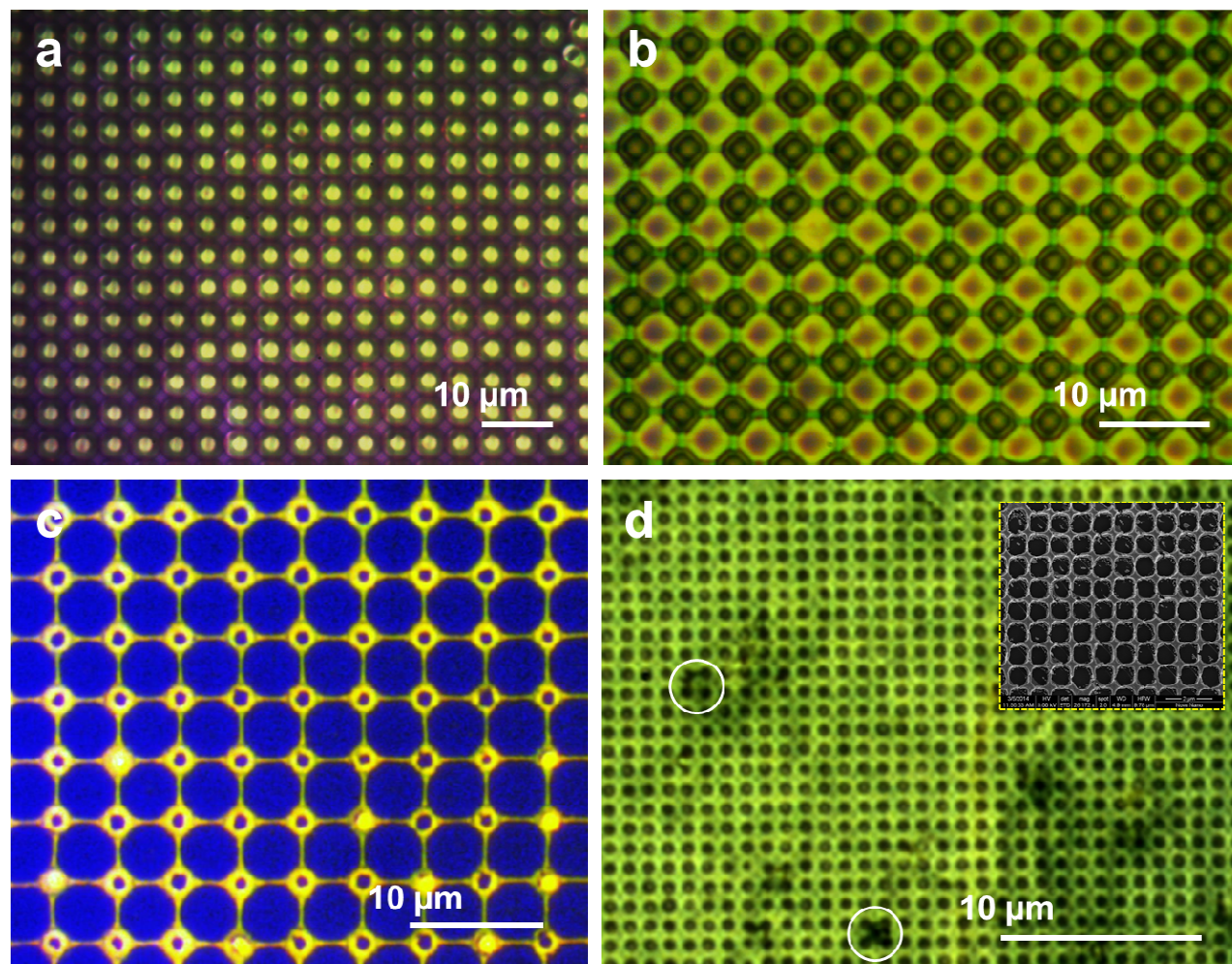


Fig. 3 Stepwise fabrication of gold micro- and nano- wire networks on flat glass substrate. Optical microscopic images of (a) 2D monolayer of latex particles (5 μm), submerged in gold nanoparticle suspension, (b) formation of liquid bridges on the substrate, (c) gold microwire network after the removal of 5 μm size latex particles, (d) gold sub-micron wire-pattern after the removal of 1 μm size latex particles, The inset in panel (d) shows the SEM image of respective sub-micron wire-pattern and the white circular rings shows the debris of lifted latex particles after oxygen plasma

placing them on the crystal. The cleaning is repeated as long as a perfect defect-free crystal template is obtained. This is to ensure that the liquid flows equally during the gold suspension doping and also evaporates smoothly to generate uniform wire network. The optical micrographs of lifted 2D monolayers of 5 μm and 1 μm sized latex particles are shown in Fig. 2e and Fig. 2f, respectively (also refer to Supporting Information, S3). After lifting the particle template, the silicon wafer is cleaned and prepared for another deposition.

Finally, for the fabrication of wire networks on glass substrate, gold nanoparticle (10 to 20 nm in diameter) suspension (2 wt.%) is doped on top of the lifted crystal on the glass substrate. The gold nanoparticle suspension is synthesized by the protocol reported previously.^[7] The suspension is stabilized by poly (vinyl pyrrolidone) (PVP) polymer and a small amount of 0.2 mM sodium dodecyl sulfate (SDS) surfactant to ensure stable liquid bridge formation. For rapid and uniform spreading of the aqueous gold suspension over glass substrate, the lifted latex particle crystal is plasma cleaned for 30 minutes and therefore, rendering crystal temporarily hydrophilic. The plasma treated crystal is then placed on a flat glass substrate before the nanoparticle suspension is doped on top of it. Both plasma cleaning and flat surface proves to have an important impact on the successful formation of regular wire networks. In addition, the excess amount of suspension is removed gently and the specimen is allowed to undergo delayed evaporation at low temperature (4-5 °C) to develop uniform wire networks. The formation of the wire network during the course of evaporation takes place in a series of stepwise processes. Firstly, metallic suspension spreads evenly over and within the latex crystal (Fig. 3a). Secondly, the evaporative-driven motion of the gold suspension between the particles forms a stable and regular liquid bridge networks around the base of the 2D monolayer of particles (Fig. 3b). Finally, on slow evaporation the liquid bridges transform into wire network (Fig. 3cd). The excess amount of nanoparticles in the suspension or fast evaporation gives slightly thicker wire networks (refer to Supporting Information, S4). Eventually, to yield wire networks, the latex particles are removed from glass substrate by one of the two

methods, depending upon the size of the particles used in the deposition process. To obtain microwire network, we used an adhesive tape to peel off the 5 μm particles and then followed by heating the specimen at 450 $^{\circ}\text{C}$ for 20 minutes. However, in case of nanowire network, the latex particles with a diameter of 1 μm were first etched by oxygen plasma for 30 minutes and then heated at 450 $^{\circ}\text{C}$ for 20 minutes to remove the latex particles. The specimen is cleaned by this procedure, however, some latex particle debris remains left behind even after heat and oxygen plasma treatment (refer to Fig. 3d). This factor has overall decreased the quality of large scale production of clean metallic patterns (see the Supporting Information, S5). The adhesive tape could not be used for peeling off the 1 μm size latex particles, because it not only removes the particle monolayer, but can also damages the nanowire network.

Replacing glass by a flexible substrate (Samsung screen protectors made from urethane with extremely thin one-sided adhesive surface) enhances the scale of lift-up and we can successfully lift up to 60-80% of the deposited 2D latex monolayers from the patterned silicon wafer. The lift-up of latex particle template on flexible substrate is rapid and result oriented. It consists of only two steps; 1) dry deposition of 2D latex monolayer on the patterned silicon wafer (the process is same as described previously, Fig. 2cd), and 2) lift-up of 2D monolayer crystal onto flat flexible substrate with extremely thin deposited adhesive (Fig. 4a). The presence of this thin layer of adhesive on the flexible film is critical to improve the lift-up percentage. For the removal of this adhesive, we etched the specimen by oxygen plasma before the gold suspension is doped onto the lifted crystal. Otherwise, the adhesive will prevent wetting and the formation of wire network at the base of latex particles. It is due to the presence of this adhesive surface, the wire structure is slightly irregular in case of flexible substrate (Fig. 4c). Until now, our approach of lift-up of latex template on flexible substrate is significantly easier and faster than earlier methods.^{22,23} The earlier methods involve multiple steps, making them laborious, time consuming, inefficient, and barely reproducible. Therefore, the advantages with single-step lift-up of 2D latex monolayer template reported in this paper may find applications in both established and new smart display gadgets.

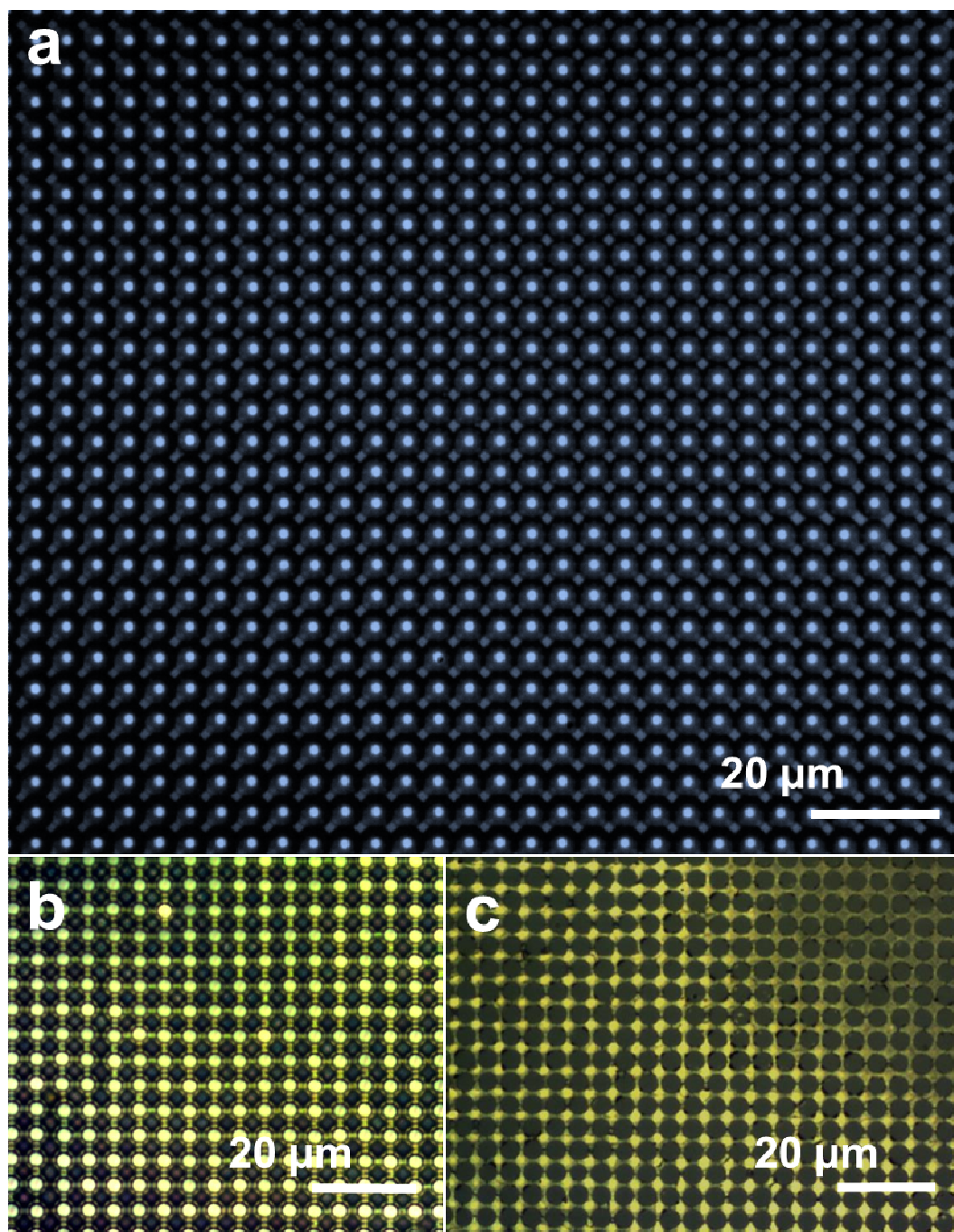


Fig. 4 Optical micrographs showing the latex particle template lift-up and fabrication of gold wire network on a flexible substrate by using 5 μm size latex particles, (a) latex particle lift-up of 2D monolayer on flexible substrate, (b) formation of liquid bridges and (c) the resulting microwire network imaged after the removal of latex particles.

Supporting Information

S1. Schematic illustration (top and side views) of the dimensions of micro- (ab) and nanowells (cd) on patterned silicon wafer used for the dry deposition of 5 μm and 1 μm sized latex particles. S2. Optical images of large area dry deposition of 2D monolayer latex particles onto patterned silicon wafer in a square array arrangement. S3. Optical images of large area lift-up of 2D monolayer latex particles on glass substrate. S4. Optical micrograph of gold microwire network fabricated at room temperature (25 °C). S5. Optical image of gold nano-pattern fabricated from evaporation lithography by exploiting 1 μm size latex particles.

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Notes and References

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