Assemblies and composites of gold nanostructures for functional devices

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Abstract
Gold has been one of the most vastly used noble metals due to its unique properties. In modern manufacturing, gold is extensively used in electronics industry as electrical connectors due to the high conductivity and corrosion resistance. With advancements in gold chemistry and nanofabrication technologies, gold materials can be tailored down to the dimension of nanoscale, which enables various novel properties. However, it is still a huge challenge to realize practical functional devices by rational utilizing these gold nanostructures-based materials. Herein, the recent developments in the design and fabrication of various functional devices based on assemblies and composites of gold nanostructures are summarized. Starting from the plasmonic effect, optical colorimetric sensors, optoelectronics and surface-enhanced Raman spectroscopy sensors are introduced. Followed by conductive devices with novel properties, flexible transparent conductors, stretchable electronics, wearable and implantable devices are discussed. Both bottom-up and top-down approaches to prepare assemblies and composites are covered. In addition, the challenges and future developments in the field are also addressed. It is believed that further developments in gold nanostructures-based materials will greatly contribute to the next-generation biosensors, optoelectronics, wearable and implantable electronics.

KEYWORDS
assemblies, composites, functional devices, gold materials, nanostructures

1 | INTRODUCTION

As the most widely used noble metals, gold has played an irreplaceable part throughout human civilisation history due to its unique physical and chemical properties.[1,2] The Lycurgus Cup exhibiting dichroic effect was made in the 4th century by blending gold nanoparticles with glass and other materials.[2] Gold is soft, malleable, ductile and resistant to corrosion, oxidation and many other chemical reactions.[2,3] In modern manufacturing, large amount of gold is consumed in electronic industry owing to its high conductivity.[4] With the advancements in chemistry and nanotechnology, materials can be precisely synthesized and controlled in the dimension of nanoscale.[5–7] As a result, lots of nanostructured gold materials have been developed, which offer new properties and application scopes for such an antique noble metal. Although other nanomaterials are also used for device fabrication, such as silver, copper nanomaterials, carbon nanotubes and graphene, gold nanomaterials possess some unique advantages for devices requiring high biocompatibility, low cytotoxicity, long-term durability and simple synthesis procedures.[1–3] Typical nanostructures from bottom-up fabrication approaches include various structures, such as nanoparticles, nanorods, nanoplates, nanowires, etc.[3,5,8–11] While nanostructures fabricated via top-down approaches, such as nanomeshes and nanobelts, are also included in the review to introduce the latest progress and advancements in flexible and stretchable devices.[4,12–15]. To construct practical functional devices, nanostructures are required to be assembled or incorporated with other materials to form assemblies or composites (Figure 1). Assemblies are obtained by self-assembly of bottom-up prepared nanostructures. And composites are acquired by the nanostructures embedded in or incorporated with polymers or elastomers, which is realized by top-down approaches. Based on these assemblies and composites of nanostructures, various devices...
FIGURE 1 Illustration of assemblies and composites of gold nanostructures and their applications in the fabrication of various functional devices. (A) Devices based on the plasmonic effect: colorimetric sensors, optoelectronic devices and surface-enhanced Raman spectroscopy (SERS) sensors. (B) Devices based on the conductivity: Stretchable electronics, transparent conductors and wearable devices.

are fabricated depending on the two major characteristics of gold materials: plasmonic effect and conductivity. The plasmonic effect of noble metal nanostructures comes from the localized surface plasmon resonance (LSPR), which leads to an intense absorption of light, as well as the enhancement of the local electromagnetic field (Figure 1A).[5,16,17] The LSPR property depends on a few factors, including geometry factors of the plasmonic particles, the dielectric properties of mediums and the interparticle coupling effect. As a result, by tuning the plasmonic effect, assemblies and composites of gold nanostructures have been utilized for the fabrication of colorimetric sensors, optoelectronics and surface-enhanced Raman spectroscopy (SERS) devices. As for conductive devices, gold has been extensively used in electronics industry as electrical connectors, such as printed circuit boards and gold bonding wires, owing to the excellent corrosion resistance and high conductivity. Recently, gold materials in micro/nanoscale have enabled conductors with novel properties, such as flexibility, stretchability and transparency.[18–25] To tailor gold materials, both bottom-up and top-down approaches have been used. Bottom-up methods obtain micro/nanostructures by wet chemistry reactions and the self-assembly of colloidal nanostructures.[126–28] While top-down methods including physical vapor deposition (PVD), photolithography and etching are used to acquire patterned surfaces and micro/nanostructures.[129–33] By using bottom-up, top-down or the combination of the two methods, a variety of state-of-the-art devices have been developed, which brought us to the next-generation electronics, such as stretchable conductors, transparent conductors and wearable and implantable devices (Figure 1B).[34–40]

Herein, we discussed the recent advances in the design and fabrication of functional devices based on assemblies and composites of gold nanostructures. Devices utilizing the plasmonic effect or conductivity of gold materials are introduced, respectively. The challenges and perspectives for the future device fabrication are identified and discussed. With the unique merits, nanostructures-based gold materials will bring new opportunities for this ancient noble metal in a myriad of applications in biosensors, optoelectronics, stretchable electronics and wearable and implantable devices.[41–47]

2 DEVICES BASED ON PLASMONIC EFFECT OF GOLD NANOSTRUCTURES-BASED MATERIALS

In this section, we mainly discuss the devices utilizing the plasmonic effect of gold nanoparticles (AuNPs) fabricated by bottom-up methods. We also introduced a SERS sensor based on nanostructures constructed by a top-down approach. As for bottom-up prepared AuNPs, it is a common approach to induce self-assembly of the nanostructures to realize the modulation of the LSPR due to the interparticle coupling effect.[48,49] Both ordered and disordered structures can be obtained by the self-assembly process.[50,51] Disordered assemblies are also known as aggregations, which can be triggered by various interactions such as electrostatic attractions, hydrogen bonding, coordination interactions and biological recognitions.[52,53] Such assemblies/aggregations are widely used for biosensing and point-of-care diagnosis. Ordered assemblies, such as one- or two-dimensional arrays,
lead to tunable and well-controlled physical properties, which are frequently applied for optoelectronics and plasmonic-enhanced devices.\[^{50,54–56}\] Although the driven force is similar, it requires well-designed templates or patterned structures to realize the assembly process.

### 2.1 Colorimetric sensors based on the assemblies/aggregations

The colloidal gold nanoparticles (AuNPs) exhibit distinguishable colour owing to the large molar extinction coefficients of nanoparticles in the visible region.\[^{16,57}\] Such optical property is tunable by the modulation of the LSPR. The most frequently used strategy is to modify the interparticle distance by inducing assembly or aggregation of the nanoparticles, resulting in a colorimetric change that is detectable by instruments or even observable by naked eyes.\[^{57–60}\] Such strategy is widely used for the fabrication of a variety of biomedical sensors.\[^{61,62}\] Figure 2A illustrates a colorimetric assay of utilizing the assembly of AuNPs to detect creatinine, which is an important clinical marker for chronic kidney disease.\[^{63}\] The citrate-stabilized colloidal AuNPs with a size of 13 nm exhibit a red colour, corresponding to the SPR absorption band at ∼520 nm. The pre-decorating of the particles with uric acid and Hg\(^{2+}\) ions resulted in little change to the colloid solution. Only the presence of creatinine significantly changed the colour of the colloids from red to blue. Correspondingly, the original SPR band at 520 nm underwent an intensity decreasing with a position red-shifting. In addition, a new absorption band emerged above 600 nm (Figure 2B). Such change is caused by the assembly/aggregation of the nanoparticles due to the synergistic coordinative binding of creatinine and uric acid towards Hg\(^{2+}\). The optical image of the colloid suspension and the transmission electron microscopy images before and after the adding of creatinine are shown in Figure 2C. To demonstrate the selectivity of this method, the system was challenged by a series of small molecules with similar chemical structures to creatinine (Figure 2D). No significant absorption change was found for all the other interferents, indicating a high selectivity of this method, the system was challenged by a series of small molecules with similar chemical structures to creatinine (Figure 2D). No significant absorption change was found for all the other interferents, indicating a high selectivity of this method. The citrate-stabilized colloidal AuNPs with a size of 13 nm exhibit a red colour, corresponding to the SPR absorption band at ∼520 nm. The pre-decorating of the particles with uric acid and Hg\(^{2+}\) ions resulted in little change to the colloid solution. Only the presence of creatinine significantly changed the colour of the colloids from red to blue.

### 2.2 Plasmonic-enhanced optoelectronics and SERS sensors based on the assemblies and the composites

For plasmonic-enhanced devices, Au nanostructures are integrated into optoelectronic devices as an additive to improve the photo-response performance.\[^{5,71}\] Significant improvement is shown in the visible range when plasmonic AuNPs are incorporated with optoelectronic materials due to the enhanced visible light absorption arose from LSPR excitation. For example, AuNPs were assembled orderly with conductive polymer polypyrrole (PPy) into one-dimensional (1D) trench structure.\[^{72}\] The fabrication process is illustrated in Figure 3A. The pair gold electrodes were fabricated using photolithography on silicon substrates with an interdistance of 2.5 µm (Figures 3B and 3C). Prior to the assembly of AuNPs, the substrate was first functionalized by aminopropyltriethoxysilane (APTES). The free amine groups from the APTES provided the positively charged surface for the binding with negatively charged AuNPs. The trench structure was constructed by e-beam lithography method using polymethylmethacrylate as the resist. To assemble the AuNPs, the substrate with 1D groove structure was immersed into colloidal AuNPs suspension (∼29.8 nm) and 1D chains of AuNPs were obtained (Figure 3D). PPy nanowires were further deposited in a lift-off process using a template-directed growth method (Figure 3E). Subsequently, the performance as a photodetector of the as-prepared device was tested (Figure 3F). The generation of photocurrent was observed for the devices with 1D assembled AuNPs. In addition, the photo-response exhibited a spectral selectivity towards an excitation wavelength of 530 nm, which is associated with the LSPR band of the nanoparticles. In contrast, the devices of PPy nanowires without incorporating AuNPs showed no significant photocurrent in the testing wavelengths. Gold nanostructures have been extensively used for the construction of surface-enhanced Raman spectroscopy (SERS) sensors owing to the plasmonic effects. It is known that anisotropic nanostructures exhibit better SERS performance, such as gold nanorod, nanoplates etc. and nanostructure assemblies with small gaps.\[^{73,74}\] Figure 3G illustrates a SERS sensor based on AuNPs assembled on porous block copolymer polystyrene-block-poly(2-vinyl pyridine) (PS-b-P2VP) nanorod arrays with an analyte preconcentrating function.\[^{75}\] The porous copolymer structures worked as a continuous nanopore sponge system, which can automatically absorb and bring the analyte liquids up to the upper surface of the SERS sensor due to the disturbance to the electrostatic double layers by the change of pH values, ionic strength and the presence of foreign molecules that neutralize or replace the citrate ligands. In the scenario of a competitive format assay, where reducing the concentration of bioprobes on the AuNPs is necessary to decrease the limit of detection, the aforementioned drawback significantly affects the labelling and leads to unclear and unstable results (Figures 2F and 2G). To solve this problem, AuNPs were prepared with thiolated PEG as the protective capping. The performance comparison of the PEG-coated (Group I) and regular citrate (Group II) AuNPs are shown in Figure 2H. The results from citrate AuNPs displayed strong reddish colour outside of the control and testing regions. In contrast, PEG-coated particles displayed no colour in the background areas. In addition, the testing areas of Strip 1, 2 and 3 in Group I were discernible. While, the testing areas of Strip 1, 2 and 3 in Group II are unable to be distinguished using naked eyes.
Colorimetric sensors fabricated based on the assembly/aggregation of gold nanoparticles (AuNPs). (A) Schematic illustration of colorimetric detection of creatinine. (B) Evolution of the absorption spectra of the AuNPs in the presence of different concentration of creatinine: 15–35 µM. (C) Optical and transmission electron microscopic (TEM) images of the AuNPs before and after assembly/aggregation. (D) Demonstration of the detection selectivity towards creatine over other molecules with similar structures. (A–D) Reproduced with permission.[63] Copyright 2015, Wiley-VCH. (E) Configuration of a lateral flow assay device to detect bisphenol A (BPA) using poly(ethylene glycol) (PEG)-covered AuNPs. (F and G) Schematic comparison of AuNPs with or without PEG modification in the lateral flow assays. The PEG-modified particles are able to locate the target and suppress the possibility of false negative/positive results due to anti-aggregation effect of PEG capping. While regular particles aggregate easily and increase the possibility of false results. (H) Comparison of the lateral flow assay performance of PEG-coated AuNPs (I) and AuNPs without coating (II). BPA concentrations: (1) 0 µg mL⁻¹, (2) 0.0008 µg mL⁻¹, (3) 0.004 µg mL⁻¹, (4) 0.01 µg mL⁻¹, (5) 0.03 µg mL⁻¹, and (6) 0.06 µg mL⁻¹. (E–H) Reproduced with permission.[70] Copyright 2017, Wiley-VCH.

to the capillary force combined with directional evaporation. The assembly and attachment of the AuNPs were performed by floating the nanorods in the suspension of citrate AuNPs (∼35.5 nm) with only the tips immersed into the suspension. Figure 3H shows the side-view and top-view SEM images of the nanoparticles assembled and attached onto the nanorod structures. The as-prepared preconcentrating SERS sensor can be used for the detection of analytes in form of liquid droplets, thin liquid films and even analytes contained in a piece of tissue paper. Next we also introduce a SERS sensor fabricated using top-down approach. Figure 3I illustrates the SERS sensor based on Au-capped polymeric nanocones inte-
FIGURE 3  Optoelectronic device and surface-enhanced Raman spectroscopy (SERS) sensors based on the guided assembly of gold nanostructures. (A) Fabrication process of individual 1-dimensional (1D) polypyrrole (PPy) nanowires embedded with AuNPs. (B and C) Microscopic images of the Au electrode pair. (D and E) Scanning electron microscope (SEM) images of the 1D assembled AuNPs chain in the 1D trench structure and PPy nanowires embedded with the 1D assembled AuNPs. (F) Comparison of photo-response in typical on-off I–t curves for PPy nanowires without (top) and with (bottom) the embedment of 1D assembled AuNPs at various wavelengths. (A–F) Reproduced with permission. Copyright 2016, Wiley-VCH. (G) Schematic illustration of a SERS sensor with AuNPs assembled on porous diblock copolymer polystyrene-block-poly(2-vinyl pyridine) (PS-b-P2VP) nanorods enabling capillarity-assisted preconcentration. (H) Side-view and top-view SEM images of the SERS sensor showing the PS-b-P2VP nanorods functionalized with assembled AuNPs. (G–H) Reproduced with permission. Copyright 2017, Wiley-VCH. (I) Illustration of a SERS sensor based on Au-capped polymer nanocones embedded in a microfluidic device. (J) Optical image of the microfluidic device showing the microfluidic lid and the embedded SERS sensor arrays. (K) SERS mapping recorded using the sensor array in a continuous flow measurement of 1 mM bis-(4-pyridyl)-ethylene (BPE) at a flow rate of 100 µL min⁻¹. The mapping refers to the 1198 cm⁻¹ peak. (L) Typical SERS data collected from the sensor array. (I–L) Reproduced with permission. Copyright 2018, American Chemical Society.
grated into a microfluidic device. The polymer nanocones were obtained by a replication method. The Au capping was subsequently conducted by e-beam evaporation, resulting in closely packed Au caps with an average diameter of ~116.6 nm and a height of ~225.5 nm. The SERS structures were further integrated with microfluidic lid consisting of a main channel and Y-shaped inlet and outlet for the sample liquid (Figure 3A). After integration with the microfluidic system, a dynamic measurement can be achieved. Figure 3K shows a SERS data mapping of a continuous flowing measurement of 1 mM trans-1,2-bis-(4-pyridyl)-ethylene (BPE) solution in the microchannel at a flow rate of 100 µL min⁻¹. The mapping refers to the intensity of the 1198 cm⁻¹ peak. The typical SERS spectra collected from the five sensors show the variation across the sensor array (Figure 3L).

3 | DEVICES BASED ON CONDUCTIVITY OF GOLD NANOSTRUCTURES-BASED MATERIALS

In this section, we focus on the novel properties and features for gold nanostructures-based materials as conductors, such as stretchability, conformability, transparency and biocompatibility. Different from the existing silicon-based rigid electronic devices, the next-generation transparent conductors or electrodes must have Young’s moduli matching human skin or tissues to mitigate discomfort and to minimize immune response. Both bottom-up and top-down approaches have successfully been applied to achieve gold nanostructures-based high performance conductors with fascinating mechanical, electrical and optical properties.

3.1 | Conductors with flexibility and transparency based on the assemblies

Although conventional gold conductors are rigid, they can become deformable if the bulk gold materials are thinned and tailored into nanostructures. Therefore, bottom-up prepared nanomaterials were assembled to fabricate conductors with mechanical deformability. For instance, AuNPs (~13 nm) were assembled into polyurethane films by using both a layer-by-layer (LBL) assembly and a vacuum-assisted floculation (VAF) approaches (Figures 4A and 4B). A free-standing composite films with a thickness of ~2.0 µm can be obtained after 500 LBL cycles. While a free-standing film prepared by VAF approach showed a typical thickness of ~30 µm. The LBL films exhibited well-dispersed AuNPs in the composites, leading to higher conductivity. In contrast, the VAF films showed smaller conductivity due to the presence of more aggregated nanoparticles (Figure 4C). However, the larger domains of polyurethane in the VAF films resulted in a much higher stretchability, which is ~480% of strain, compared to ~110% of strain for LBL films (Figure 4D). The high conductivity and stretchability achieved in this study despite the minimal aspect ratio of the spherical nanoparticles were attributed to the dynamic self-organization of the nanoparticles under tensile stress. Compared to spherical nanoparticles, metal nanowires usually show improved mechanical stability of the fabricated flexible conductors, which can withstand greater deformations while maintaining the electrical conductivity. Such an advantage is due to the high aspect ratio of the nanowires, which also enables the realization of flexible conductors with transparency. A transparent flexible conductor was fabricated by a solution-based self-assembly approach. By drop-casting aged gold nanowires (AuNWs) suspension onto an air-water interface, self-assembled mesh structures were obtained due to the partial removal of the oleylamine ligands (Figure 4E). The AuNWs bundles showed a typical thickness of 193.7 ± 67.6 nm determined by atomic force microscopy (Figure 4F). Such assembled AuNWs structures were used to fabricate patterned transparent electrodes on polyethylene terephthalate (PET) film using a simple mask coating method (Figure 4G). The AuNWs mesh structures showed excellent transparency (Figure 4H). The mesh film exhibited high transmittance (~92%) and conductivity (sheet resistance of ~130.1 Ω sq⁻¹) with an aging time of 12 h (one-layer). The prepared transparent conductor on PET exhibited a very low resistance variation even under bending radius of 2 mm, indicating a high flexibility (Figure 4I). As a proof-of-concept, a circuit on PET film was designed to power four light-emitting diodes (LEDs) in parallel. The lighting of the LEDs was not affected by either tensile or compressive stresses applied when the device was attached to a human wrist (Figure 4J).

3.2 | Stretchable electrodes for wearable and implantable devices based on the composites

Top-down fabrication approaches have the advantages of achieving complex structural and patterning designs with accessible process, as well as large-scale production capability. Therefore, gold nanostructures were embedded in or incorporated with polymers/elastomers using top-down approaches to enable highly stretchable conductors and devices. A stretchable on-skin device was fabricated based on polyanvinil alcohol (PVA)/Au composite nanomesh structures (Figure 5A). Firstly, PVA nanomesh sheets were prepared by using electrospininning method. Then the deposition of a layer of gold (70–100 nm) was performed by PVD method using a shadow mask for patterning. When the as-prepared nanomesh conductors were placed on the skin and wetted with water, the PVA nanofibres dissolved and left the nanomesh conductor attached to the skin. Figure 5B shows the conformable attaching of the nanomesh conductor on to a fingertip and the SEM image of the nanomesh conductor. For demonstration, an on-skin wireless sensor system with touch, temperature and pressure sensing functions was constructed using the nanomesh conductors (Figure 5C). The touch sensing was realized by simple nanomesh conductor arrays. Periodical resistance change of the touch sensor responding to the repeat contact of the finger with an object was observed, indicating a reliable touch sensing performance of the device (Figure 5D). As is known, wrinkled gold
Flexible and transparent conductors were fabricated based on assemblies of gold nanostructures. (A and B) SEM and optical images of conductive polyurethane/AuNPs films prepared by layer-by-layer assembly (left) or vacuum-assisted flocculation (VAF) (right). (C) Evolution of conductivity as a function of the number of films laminated. (D) Change of conductivity as a function of tensile strain of films prepared by LBL and VAF methods. (A-D) Reproduced with permission. Copyright 2013, Springer Nature. (E) Illustration of the fabrication process of the transparent nanomesh conductor via self-assembly of ultrathin gold nanowires (AuNWs) and the SEM image of the AuNWs meshes obtained. (F) Microscopic imaging of the AuNWs mesh film (scale bar: 100 µm) and atomic force microscopy (AFM) imaging of the film transferred to a mica substrate (scale bar: 10 µm). (G) Illustration of the patterning of the AuNWs mesh. (H) Transmittance and sheet resistance of AuNWs multilayer structures with different aging time. (I) Comparison of resistance change of an AuNWs mesh film (red star) and an ITO (blue triangle) on PET substrate at different bending radius. (J) Demonstration of using the AuNWs mesh films as connections to light up four LEDs under convex and concave bending states. (E-J) Reproduced with permission. Copyright 2016, Wiley-VCH

Films or belts have been incorporated with elastomers to realize stretchable conductors. A novel design of suspended gold nanobelts on polydimethylsiloxane (PDMS) tripod structures was reported (Figure 5E). Owing to tripod structures (bottom), the stress from physical contact of two materials with mismatching Young’s modulus can be significantly reduced comparing to a normal wrinkled structure (top). A highly stretchable conductor was fabricated, and the electrode could be stretched to 130% of strain without a significant decrease in conductivity (Figures 5F and 5G). Besides, the deposition of metal thin-films on elastomers is another frequently used approach to enable flexible/stretchable electronics. However, failures frequently happen due to the low adhesion between metal films and elastomers. Thus, the interfacial adhesion is critical for the stretchable electronics aiming for long-term use. To provide accessible method to achieve high interfacial adhesion and to enable large-scale fabrication possibility, a method of using thermal radiation assisted metal
FIGURE 5 Wearable and implantable electrodes fabricated based on gold nanostructures/polymers composites. (A) Schematic illustration of attaching the stretchable PVA/Au nanomesh on-skin conductor on to the skin by water wetting. (B) A photograph of the nanomesh conductor forming conformable attaching onto a fingertip (scale bar: 1 mm) and an SEM image of a nanomesh conductor on a silicone skin replica (scale bar: 5 µm). (C) Photographs of the on-skin wireless sensor system. (D) Resistance change of the touch sensor in response of the touching movements of the fingertip. (A–D) Reproduced with permission.[12] Copyright 2017, Springer Nature. (E) Schematic illustration of the Au nanobelt on a normal wrinkled structure (top) and on a tripod suspended structure (bottom) and the analysis of the strain distribution by finite element modelling (FEM) method. (F) A photograph of the prepared highly stretchable electrode based on Au nanobelts on tripod PDMS structures. (G) I–V curves of the stretchable electrode under different tensile strains. (E–G) Reproduced with permission.[4] Copyright 2015, Wiley-VCH. (H) Illustration of the thermal radiation-assisted metal encapsulation (TRAP) in partially cured PDMS. (I) Cross-section SEM imaging of the composite film with gold structures embedded in the PDMS substrate forming interlocking structure. (J) Comparison of resistance change as a function of the strain for TRAP Au film and a neat Au film without interlocking layer. (K and L) Photographs showing the long-term implantation testing in rats for intramuscular monitoring. (M) Intramuscular electromyography (EMG) signals recorded and the corresponding spectrum analysis. (H–M) Reproduced with permission.[102] Copyright 2019, Wiley-VCH

encapsulation was reported to fabricate highly stretchable gold electrodes.[102] For the fabrication process, a partially cured PDMS film prepared by precise controlling of the pre-curing time was mounted in the evaporation chamber to conduct the PVD process (Figure 5H). When gold atoms reached the soft partially polymerized PDMS, metal diffusion occurred due to fluidic physical property of the PDMS substrate. Subsequently, heat transfer from the source facilitates the fully curing of the PDMS film, resulting in a composite interlocking layer between the gold film and the PDMS, which greatly enhanced the mechanical property of the conductor with an interfacial adhesion of larger than 2 MPa (Figure 5I). The size of the gold nanoparticles is ~20 nm (grain size). And the PDMS layer has a typical size of ~500 µm. As a result, the films exhibited greatly enhanced stretchability compared to normal thin-film-based conductors. The
maximum tensile stretchability of the interlocking films was measured to be up to 130% of the strain (Figure 51), while a gold film with similar thickness (~300 nm) of Au deposited using conventional PVD method displayed a maximum stretchability of no more than 10% of strain. Using this approach, conductive gold films can be fabricated as large as of hundreds of square centimeters. Owing to the high stretchability and stability, the capability of the stretchable electrodes working as long-term implantable devices was tested in animals (Figures 5K and 5L). Electrodes were implanted in rats to collect the intramuscular electromyography signals. The signals collected in this way displayed low noise as revealed by the spectrum analysis (Figure 5M). Furthermore, the implantation and signal acquisition were successfully conducted for over 4 months, indicating a great potential of the electrodes for practical long-term use. In spite of its high cost, gold nanomaterials are still highly desirable for wearable healthcare electronics and implantable devices, where chemical stability and biocompatibility are greatly demanded.

4 PERSPECTIVE

Through both bottom-up and top-down approaches, assemblies and composites of gold nanostructures have been used to fabricate various functional devices, providing potential applications in biosensing, medical diagnosis, photocatalysis, stretchable electronics, wearable and implantable devices. However, there are still challenges and barriers to be overcome. From the fabrication aspect, large-scale production of nanostructured gold materials is a requisite for practical industrial applications. At the current stage, most bottom-up approaches for nanostructures synthesis require precise controlling of temperature, pressure, concentration, pH, inert atmosphere and so forth, which can hardly be conducted outside a laboratory. In addition, significant disparity between different batches is another big issue for nanomaterials synthesis. Top-down approaches could have an advantage in terms of production scaling-up and reproducibility, but there is still a lack of standard criteria for the fabrication protocols. And the cost is also an issue for top-down-based methods. Apart from fabrication, rational device integration to realize complex or multiple functions is necessary for the next-generation electronics and devices. Currently, most designs only focus on one particular function based on either plasmonic property or conductivity. The combination of the two properties will definitely enable novel devices, such as flexible photo-detectors, wearable colorimetric sensors for healthcare monitoring and chronic disease diagnosis. It is expected that gold nanostructures-based materials will make great contribution to the future electronics in E-skins, bio-inspired electronics and wearable healthcare devices.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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