



Nano-assembly and welding of gold nanorods based on DNA origami and plasmon-induced laser irradiation

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Abstract

The bottom-up organization of noble metal nanostructures with nanometer-scale precision is an important goal in nanotechnology. Owing to their unique localized surface plasmon resonance, well-defined metal nanostructures arrays could be used to develop applications in nano-photonics, nano-plasmonics, and nano-electronics. This article proposes an alternative pathway of a controllable approach to assemble and weld together the gold nanostructures. As a typical plasmonic nanostructure, the gold nanorods (Au NRs) was synthesized by the classical seed-mediated growth method. Based on the recognition of biomolecules through complementary DNA hybridization, we used DNA origami strategy for controllable assembly of Au NRs. Rectangular DNA origami as a template can induce the geometrically assembled of Au NRs. We designed and fabricated tip-to-tip Au NRs dimers on the DNA templates. Then, the follow-up formation of nanojunctions between assembled tip-to-tip Au NRs dimers Au NRs was conducted by irradiating infrared femtosecond pulses laser. The ability to coupling plasmonic nanostructures by assembly and nano-welding could be fundamental to developing novel optical properties and ensuring materials.

Keywords Gold nanorods · DNA origami · Assembly · Nano-welding

1 Introduction

Currently, there are mainly two ways to assemble nanomaterials: top-down method and bottom-up method (Ozin et al. 2009; Yu et al. 2013). The top-down method at the nanoscale encountered great obstacles and challenges due to limitations of the fabrication size. The strategies based on the bottom-up method are more accessible to prepare complex and exquisite nanostructures. The precise assembly of bottom-up technology has advantages in the spatial assembly resolution as low as 10 nm (Biswas et al. 2012). Especially, the self-assembly by DNA nanotechnology which is one of the hot issues in the field of the bottom-up molecular self-assembly technology (Andersen et al. 2009; Liu and Liedl 2018). Over the past decades, rapid progress has been made in the

structure of DNA nanotechnology. A large variety of DNA nanostructures with different geometric shapes and topological features have been constructed in high yield. DNA nanotechnology, particularly, the DNA origami method, offers a robust method for nanoscale configuration (Steinhauer et al. 2009; Tan et al. 2011; Xu et al. 2017). Enormous progress has been made in the DNA guided organization of nanomaterials into discrete, one dimensional, two-dimensional and three-dimensional architectures (Lan et al. 2013; Schreiber 2014). DNA nanotechnology is a vehicle for the controllable assembly of nanostructures because it enables the positioning of nanoparticles with nanoscale precision and the tailoring of their binding interactions. DNA origami, which is based on the folding a long single-stranded DNA scaffold with the assist of hundreds of short complementary staple strands, can create almost any arbitrary two-dimensional even three-dimensional shapes nanostructures. Every stable strand is unique on the DNA origami, which makes it nano-addressable and a perfect template for nanoparticles self-assembly (Rothenmund 2006). Great efforts have been made to use self-assembled DNA nanostructures as scaffolds to constructing advanced and unique plasmonic architectures, such as the ability to self-assembly of noble metal

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nanoparticles into complex and discrete nanostructures at specific locations (Pal et al. 2011).

Using DNA origami templates as nanorobots to assemble nanostructures and thus fabricate nanoscale devices, because it is capable of some robotic tasks like the connection at the nanoscale (Hung et al. 2010; Kershner et al. 2009). For example, Harb et al. reported that T shape DNA origami could act as a template to assemble small gold nanospheres with 4.1 nm gap. And then the gold nanospheres were grown into larger size by electrodeposition, so the gold nanospheres could connect with each other and form the T shape conductive nanowires (Klein et al. 2013). In addition, Kuang et al. have successfully assembled a gold linear waveguide nanostructure with a diameter of 10 nm using multi-scaffold DNA origami (Pearson et al. 2012). These methods provide another way for the fabrication of various kinds of collection configuration conductive devices at the nanoscale, which is expected to become a new path to design large-scale electronic circuit nanodevices (Liu et al. 2011). This technique holds great promise for exploiting applications in the field of photonics, electronics, and biosensing and biomedicine (Li et al. 2014; Thacker et al. 2014).

Gold nanomaterials have drawn much attention due to their unique optical properties which called localized surface plasmon resonance (LSPR) when the free conduction electrons interact with incident light (Hu et al. 2006; Wang et al. 2013). Due to their excellent plasmonic properties, the gold nanomaterials exhibit great potential applications in the fields of optoelectronics, sensing, and catalysis (Tran and Nguyen 2011; Wang and Shen 2006). Among the various shape of gold nanocrystals, gold nanorods (Au NRs) have anisotropy geometrical structure. Au NRs possess two LSPR band: the transverse resonance absorption peak (TPR) and the longitudinal resonance absorption peak (LPR) corresponds to its longitudinal and transverse axes, respectively (Chen et al. 2013; Yang et al. 2015). Furthermore, the LPR band can be tunable depending on its aspect ratio (AR) (Pérez-Juste et al. 2005). When the interparticle distance between two Au NRs are sufficiently close to each other, Au NRs will exhibit strong plasmonic coupling interaction and generate a collective effect such as hotspots, which is benefits for their application (Vigderman et al. 2012; Xu et al. 2011; Zhang et al. 2014).

Using DNA origami as scaffold templates to guide different sizes and shapes plasmonic nanoparticles in an orderly arrangement at the nanoscale have great significance in the research of the interaction between nanoparticles such as Au NRs (Lan et al. 2013) and their application in novel nanodevices, biosensors and drug delivery (Chen et al. 2015; Song et al. 2017). Here, this paper provides a controllable approach to assemble and weld together the gold nanorods (Au NRs). First, the Au NRs were fabricated by the seed-mediated growth method. Then, by using rectangular DNA

origami as a template to arrange the anisotropic nanorods, Au NRs were designed tip-to-tip assembling into dimer structures. Finally, irradiation of these dimers with femto-second radiation forms a nanojunction between them and welds the dimers into fused dimers.

2 Experimental

2.1 Materials

Gold(III) chloride trihydrate ($\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$), silver nitrate (AgNO_3) and cetyltrimethylammonium bromide (CTAB) were acquired from Sigma Aldrich. And sodium dodecyl sulfate (SDS) were purchased from Aladdin. Bis (psulfanato-phenyl) phenyl-phosphine (BSPP) was bought from Strem Chemicals. L-ascorbic acid (AA) and sodium borohydride (NaBH_4) was obtained from Alfa Aesar. Non-thiolated DNA sequences were bought from Bioneer. Thiolated DNA sequences of HPLC grade were bought from Invitrogen. All the chemicals were commercially obtained and used without further purification.

2.2 Fabrication

Au NRs were fabricated according to the classical seed-mediated growth method in aqueous solutions (Pérez-Juste et al. 2005). The seed solution was generated by adding 600 μL aliquot of the ice-cold 10 mM NaBH_4 solution to the 5 mL mixture of 10 mM HAuCl_4 and 100 mM CTAB at 28 °C, and then mixing and stirring the reaction solution for 2 min intensely. Next, the growth solution was prepared by adding 32 μL aliquot of 100 mM AA into containing 10 mM HAuCl_4 , 10 mM AgNO_3 , and 100 mM CTAB. After AA adding, the growth solution changed to colorless immediately. Finally, 10 μL aliquot of seed solution was added to the growth solution, and the reaction mixture was gently agitated for 20 s and left undisturbed and aged for 12 h. Afterwards, the Au NRs was successfully synthesized, and the colloidal solution was centrifugated at 11,000 rpm for 30 min. And finally, the as-synthesized Au NRs were re-dispersed in deionized water for further use.

Following, the Au NRs were functionalized with thiolated DNA. First, Au NRs were stabilized and capped with BSPP (2.5 mM) through replacement by CTAB molecules. Then, the BSPP-capped Au NRs was mixed and incubated with thiolated DNA solution (500 μM) with the molar ratio over 500 in $1 \times \text{TBE}$ buffer containing 0.01% SDS and incubated at room temperature for several hours. Next, 10 μL aliquot of 5 M NaCl was intervals added to the reaction mixture solution until the concentration of NaCl became 500 mM over 20 h. Finally, the DNA functionalized Au NRs were purified to remove excess free thiolated DNA strands through

2.5% agarose gel electrophoresis in $0.5\times$ TBE running buffer which contained Acetic acid, 10 mM; EDTA, 1 mM; Tris, 20 mM; and Magnesium acetate, 6.25 mM with pH 8.0. The rectangular DNA origami template with dimensions $90\text{ nm}\times 60\text{ nm}\times 2\text{ nm}$ is self-assembled from a long single-stranded M13 viral genomic DNA (M13mp18) which folded by a set of ~ 200 short staple strands through sequence-specific hybridization and formation of multiples of DNA crossover according to the protocol (Rothemund 2006).

The rectangular DNA origami was obtained by annealing the mixtures of single-stranded M13mp18 DNA with staple strands and capturing strands with a ratio of 1:10:10 by PCR from 94 to 25 °C over 12 h. For the purpose of removing excess staple strands and capturing strands, the synthesized DNA origami products were stained using SYBR-Green and then purified through 1% agarose gel electrophoresis using $0.5\times$ TAE- Mg^{2+} as running buffer. After that, the obtained agarose gel band of the DNA origami structure was cut out under UV light and recovered by electroelution with a dialysis membrane (8000–14,000 MWCO). The purified DNA origami templates were mixed with DNA-functionalized Au NRs at the molar ratio of DNA origami: AuNR₁: AuNR₂ of 1:5:5. Finally, the Au NR dimers were assembled by annealing DNA-functionalized Au NRs with capturing strands from 45 to 25 °C with 12 h.

2.3 Characterization

The optical absorption spectra were collected by using a Perkin–Elmer Lambda 35 UV–vis–NIR spectrometer. The scanning electron microscopy (SEM) images of the samples were recorded with by FEI Quanta 450 FESEM instrument at an accelerating voltage of 10 kV. Transmission electron

microscope (TEM) images were obtained with JEOL JEM-2011F TEM instrument and operated at an accelerating voltage of 200 kV. The 100 fs laser pulses were generated by the laser system (Spectra-Physics) and operated at a repetition rate of 50 kHz which were amplified by Ti–Sapphire and centered at 800 nm.

3 Results and discussions

The Au NRs were synthesized by a typical seed-mediated growth method with the help of surfactant-directed shape as shown in Fig. 1a. It is well known that the CTAB encapsulated on Au NRs can act as shape-direct agent and stabilize the nanocrystal. The CTAB molecules adsorbed on Au NR and formed bilayers steric interaction (Hung et al. 2010). And the SEM and TEM images of Au NR crystal structure are shown in Fig. 1b, c, respectively. The as-synthesized Au NRs possess an average length of $41\text{ nm}\pm 0.3$, and the average aspect ratio is about 3. The optical absorption spectrum of Au NRs colloidal solution is shown in Fig. 1d, as expected, the Au NRs colloid display two characteristic plasmon absorption bands, a strong longitudinal plasmon resonance (LPR) band at 738 nm and a weak transverse plasmon resonance band at 521 nm. In general, during the preparation of Au NRs, the CTAB as a surfactant is coated on the surface of the nanorods. However, the CTAB usually blocks the nanorods to bind biological molecules due to the high concentration of CTAB the stable double layer of CTAB capped on the surface of Au NRs (Pérez-Juste et al. 2005). Therefore, the functionalized modification of the Au NRs is necessary for their next step applications. The thiolated DNA and Au NR cannot be easy to form Au–S bond

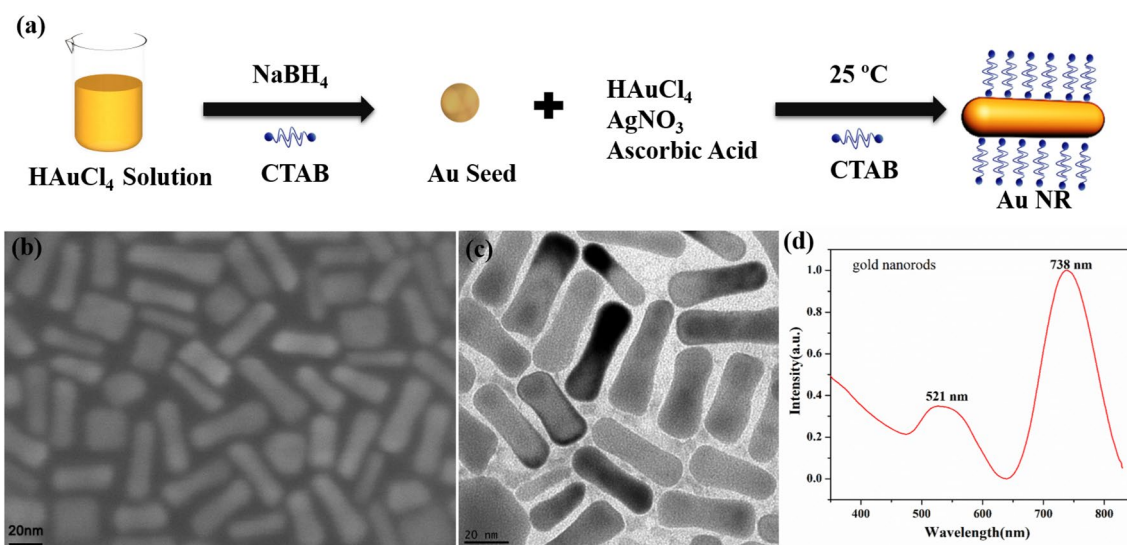


Fig. 1 a The growth process, b SEM image, c TEM image, and d UV–vis spectrum of the Au NRs

because of CTAB, so BPSS is used to replace the surfactant CTAB on the surface of Au NRs to assist the modification of DNA molecules.

In our research, two different sequence thiolated DNA molecules are chosen as the modifier to functionalize the Au NRs, respectively. Au NRs were functionalized with two different sets of thiolated DNA, respectively, as illustrated in Fig. 2. Then the thiolated DNA modified Au NRs are purified through 2.5% agarose gel electrophoresis to remove the excess unbound thiolated DNA. Next, the DNA origami templates (90 nm × 60 nm × 2 nm) were obtained by annealing a single-stranded M13mpl8 DNA with a set of ~200 short staple strands (Rothemund 2006). The rectangular DNA origami template, which has a relatively large utilization area, is conducive to assemble various nanoscale building blocks. The rectangular DNA origami structure schemes in Fig. 3a and the green lines are short staple strands; the red line is single-stranded M13mpl8 DNA. By fixing 226 different short staple strands at different locations in a long single strand M13mpl8 scaffold based on the DNA base pairs complementary principle to fold it into the rectangular structure. In rectangular DNA origami structure, each of the staple strands has a unique position which means that the DNA origami nanostructures are addressable in nanoscale. This property makes the DNA origami as a good template for positioning and assembling of the Au NRs. Then, the staple of some location strands ends extend 15 bases, which can be used to DNA complementary pairing with Au NRs surface. These extend strands called capture strand, one set of seven capture strands on the templates are used to fix Au NR₁, and another set of seven capture strands are used to fix

the Au NR₂. Through the design to the position of capture strands on the DNA origami, the relative position between the two Au NRs can be fixed as shown in Fig. 3b. Then, following by extending anchoring two different sets of capturing strands on one side of the rectangular DNA origami to anchor functionalized with Au NR₁ and Au NR₂ which are complementary to the thiolated DNA functionalized on Au NRs surface. The thermal annealing procedure was carried out by PCR protocol to ensure a high positioning accuracy of the Au NRs dimers on DNA origami templates. At last, the pre-designed tip-to-tip Au NRs dimer nanostructures were fabricated by hybridizing the DNA origami and Au NRs. The above steps were used to guide the assembly of Au NRs on the templates as schemed in Fig. 2.

The assembled tip-to-tip Au NRs dimer was collected by gel electrophoresis. And then 20 μL of the collected Au NRs dimers were dropped on the 1 × 1 cm silicon (Si) wafer substrate, as displayed in Fig. 4. Based on the different magnification of the SEM images, the assembled productivity of tip-to-tip Au NRs can be estimated to be about 30% percent in the products. In Fig. 4b, the tip-to-tip assembled Au NRs were marked with the red dotted line to show the assembled Au NRs dimers clearly and intuitively. In addition, to accurately assess the tip-to-tip assembled Au NRs nanostructures, 5 μL of the collected Au NRs dimers drop on the copper grid to clearly see the formation of Au NR dimers which was manifested by TEM, and the images are shown in Fig. 5. Because the thickness of a single rectangular DNA origami is only 2 nm, this ultra-thin two-dimensional structure of DNA scaffold is difficult to observe under TEM even after negative staining. When it is linked with Au

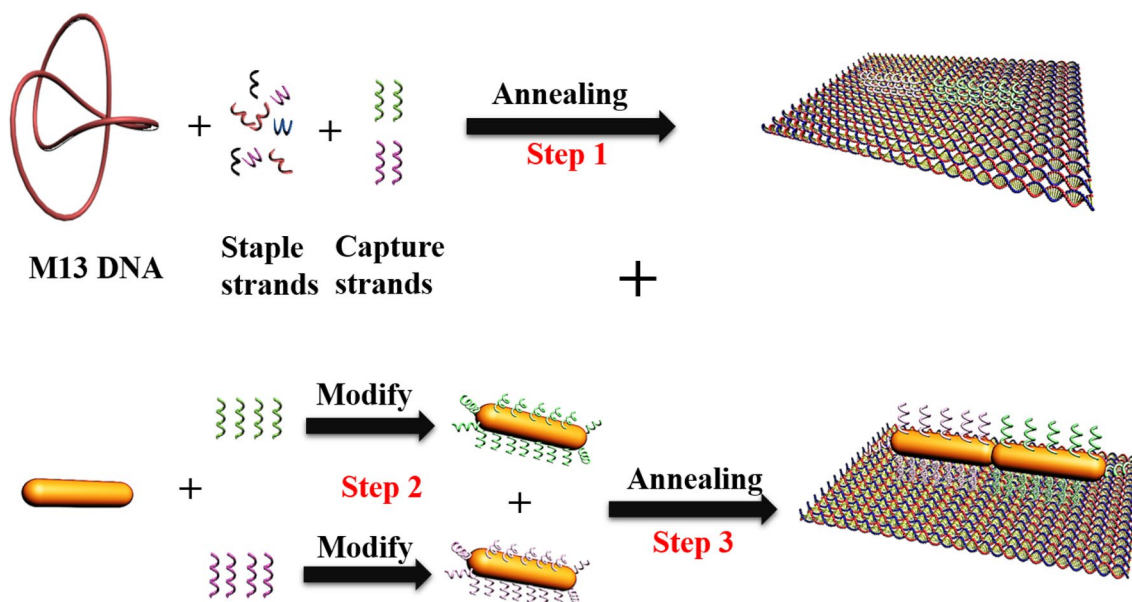


Fig. 2 Schematic illustration of the assembly of dimer Au NRs on rectangle DNA origami template

Fig. 3 Schematic illustration of **a** each sequence of the rectangular DNA origami template and **b** the location of the tip-to-tip Au NRs on the DNA origami template

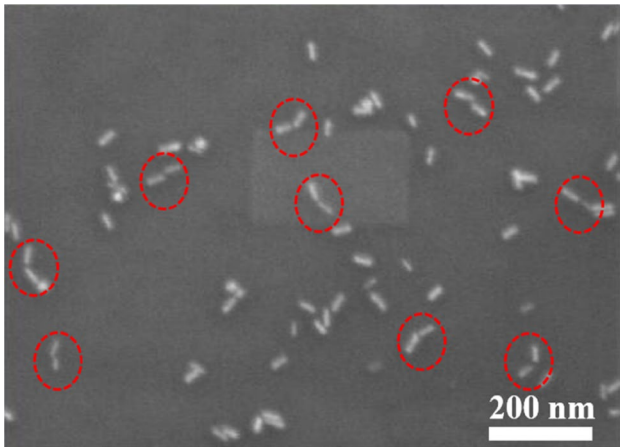
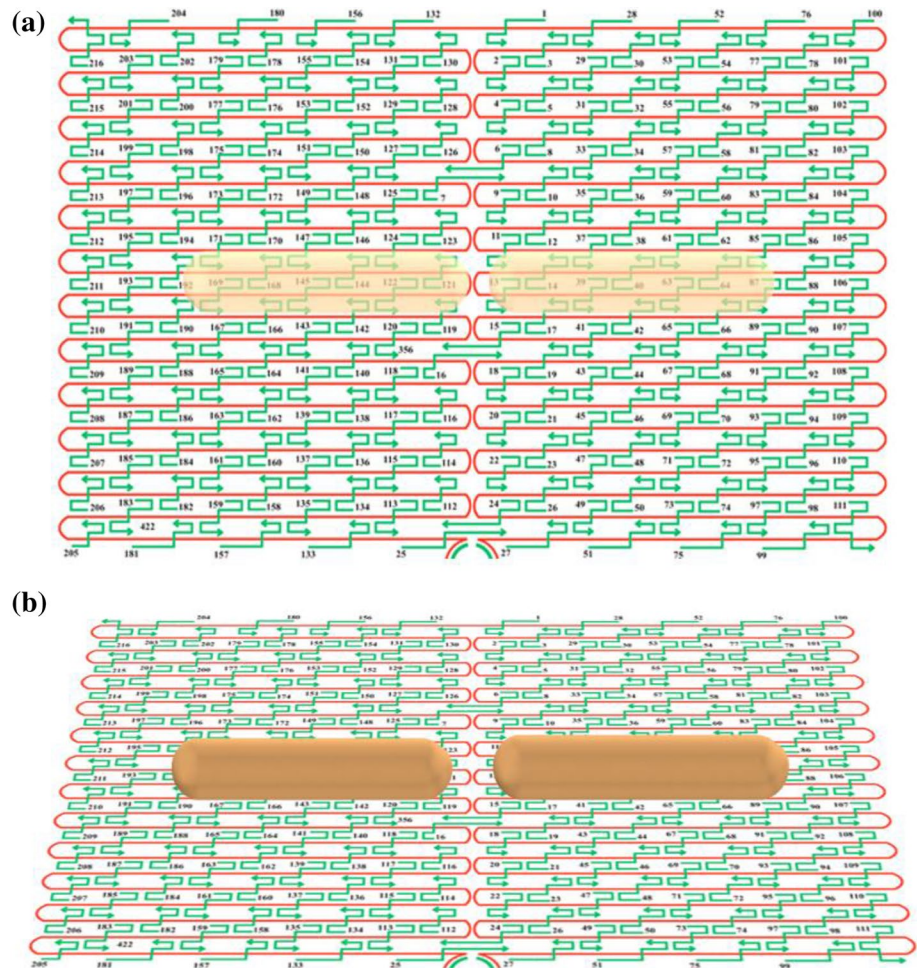


Fig. 4 SEM image of assembled tip-to-tip Au NRs by DNA origami on Si substrate

NRs, the contrast between the DNA template and Au NRs is very different. As a result, it completely disappeared under TEM, and only the structure of the Au NRs can be observed under TEM. And the enlarged image of a pair of tip-to-tip

Au dimer which was displayed by TEM in Fig. 5b, the tip-to-tip distances are about 4.5 nm which is very close to each other due to the DNA origami assembling technique. Ideally, if Au NRs are perfectly assembled on the rectangular DNA origami template to form the tip-to-tip structure according to the experimental design, then the angle between the Au NRs should be theoretically 180° . But according to the observation of SEM images on a large number of assembled Au NRs, the orientation arrangement of the Au NRs dimers displays a certain deviation from the original design, which the Au NRs are tip-to-tip parallelled with each other. Some degree of angle between the two Au NRs in the dimer can be observed from SEM and TEM images in Figs. 4 and 5a. The angle between the two Au NRs is not always 180° , and the angles between Au NRs dimers can range from 80° to 180° . We have analyzed the following reasons for the angle deviation: First, the rectangular DNA origami whose height is 2 nm is not a rigid structure and could occur curling (Lan et al. 2013). The capture strands extending on the DNA origami template are assembled with the thiolated DNA on the Au NRs. Theoretically, the origami template and the assembled Au NRs could exhibit a few nanometers distance

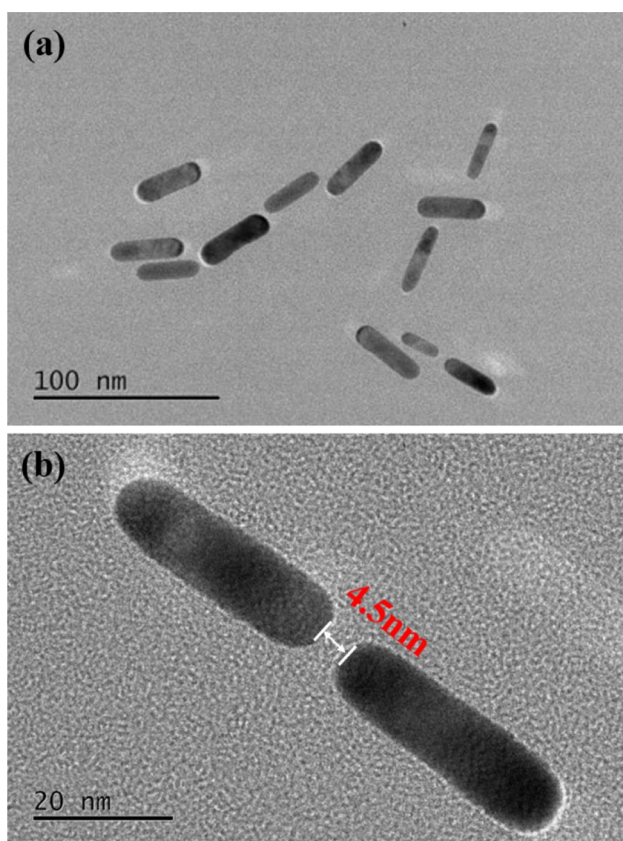


Fig. 5 TEM images of **a** tip-to-tip Au NRs after assembling on the DNA origami template and **b** a pair of assembled Au NRs dimer

between them. Therefore, it is very easy to produce a deviation when assembling (Chen et al. 2015). Secondly, the size of Au NRs used in the experiment is relatively large. Even if there are seven capture strands to link the Au NRs, it cannot guarantee the effective assembly of Au NRs with DNA origami. Besides, the thiols group of the thiolated DNA molecules also possibly allow a certain degree of flexibility at the connecting point. Finally, the SEM and TEM images were taken after the sample was dried in the air. Due to the shrinking effect, the angle between the Au NRs may also change. Although productivity of the Au NRs dimers on the DNA origami template was not high, the assembly reaction still successful to some extent. That is, the capture strands extending from the rectangular origami were successfully linked with the Au NRs by DNA hybridization. Still, these experiments data depict that the assembled configurations roughly agree with our original design. And the assembly productivity may need further adjust the parameter during the synthesis process in detail.

Afterward, the assembled Au NRs dimers by DNA origami templates on the Si substrate are then gone through irradiation under the femtosecond laser for 3 min. The parameters of the femtosecond laser system are wavelength = 800 nm, repetition rate = 50 kHz, pulse energy = 500 $\mu\text{J}/\text{cm}^2$, pulse width = 100 fs, and the spot diameter of irradiated beam area ≈ 5 mm. Figure 6a displays the formation process of the welded Au NRs dimers by femtosecond laser pulses exposure. And the nanojunction connecting the nanorods can be observed by the magnified TEM images and the welded tip-to-tip Au NRs dimers are shown

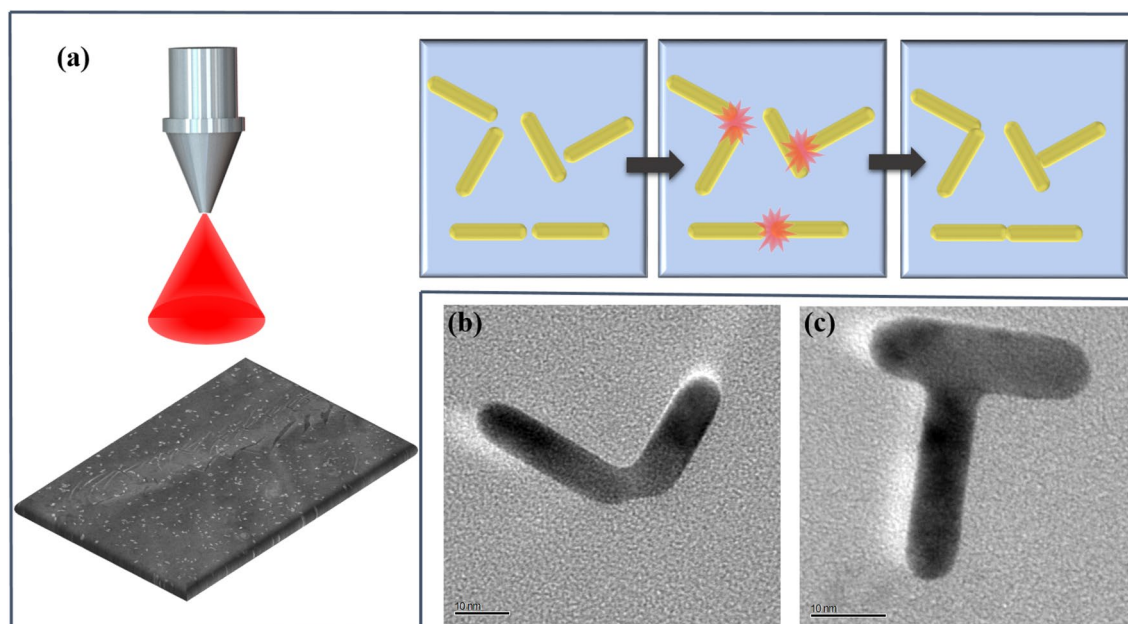


Fig. 6 **a** Schematic of the laser-induced nano-welding process and TEM images of the welding Au NRs dimers with different angle **b** 60° and **c** 90° by fs laser

in Fig. 6b, c. The welded nanojunction was shown with the 60° and 90° angle Au NRs dimer as examples which have about -2 to -5 nm gap over-lap with Au NRs dimers. When the incident laser irradiation on the assembled nanostructures, the excitation of LSPR oscillations of Au NRs with a femtosecond pulse laser which is approached to the LPR (738 nm) resonance wavelength of Au NRs can coupling with the incident laser. The ignited electrons of Au NRs were resonance coupling with the laser wavelength and form relaxation dynamic system (González-Rubio et al. 2015). And this dynamic relaxation system will thermalize the electrons on the surface of Au NRs. Therefore, the hotspot will generate in the gap between the Au NRs dimers, which will result in the local electric field enhancements. Thus, the assembled tip-to-tip Au NRs dimers have been welded together by femtosecond laser pulses which have nanocontact (Fontana et al. 2017). To illustrate the strong electric enhancement properties of Au NRs in the hotspot region. The local electric field intensity distributions around the Au NRs dimer are simulated by the finite element method (COMSOL Multiphysics 4.3 software) with an incident laser at 800 nm-wavelength. In our calculations, the dimensions of the Au NRs were extracted from the TEM images. Three types of Au NRs dimers with different angle 180°, 90°, and 60° with 5 nm gap were constructed, and the incident 800 nm wavelength laser was propagated along the z -direction and polarized along the y -direction. The distribution of enhanced electric field around Au NRs dimer as shown in Fig. 7 are not homogeneous around the whole surface of the structures, the hot spot between the gap has greater enhancement than another area of Au NR (Lin et al. 2016).

Furthermore, the 100 fs laser irradiation which has extremely short timescale and very low energy pulse is a non-thermal welding process. As a result, the excited electrons could only instantaneously weaken the lattice rather than transfer the absorbed energy to the lattice (Herrmann et al. 2014). Owing to the increase of surface energy at the tips of Au NRs by incident femtosecond pulse laser, the gold atoms are locally weakened could be more mobility near the hotspot region, and then flow the gap and form the nanojunction (Ekici et al. 2008; Petrova et al. 2006). Therefore, the Au NRs dimers can be welded and formed coalescence nanostructure (González-Rubio et al. 2016;

Son et al. 2014). Besides, when increasing pulse energy of fs laser up to 1 mJ/cm^2 and 1 mJ/cm^2 , the shape of Au dimers can evolve from locally tip welds to melted into nanosphere, respectively, and finally to melted to large a nanosphere as shown in the Fig. 8. Because with increase the energy of irradiation energy, it will increase the mobility of the surface atoms and facilitated the reshaping to nanosphere, which is driven by the thermodynamically due to the minimum surface energy (Link et al. 1999a, b). The laser radiation provides an alternative way to weld the tip-to-tip Au NRs dimers. Even though the tip-to-tip Au NRs dimers did not have an ohmic contact with each other when assembled by DNA origami, femtosecond laser irradiation is capable of welding the dimers with the nano-joined together.

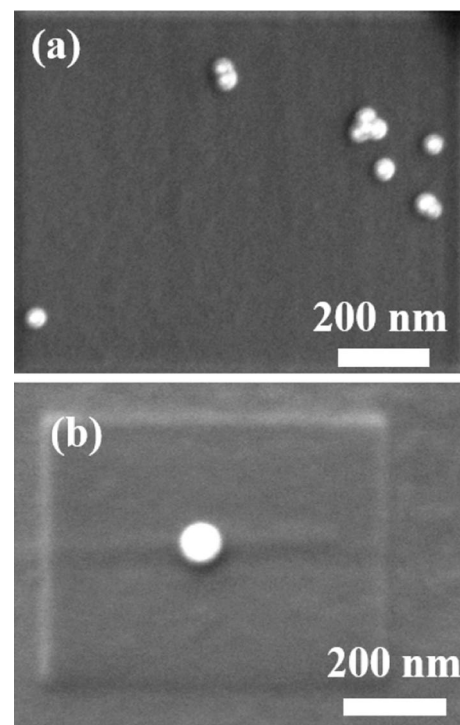
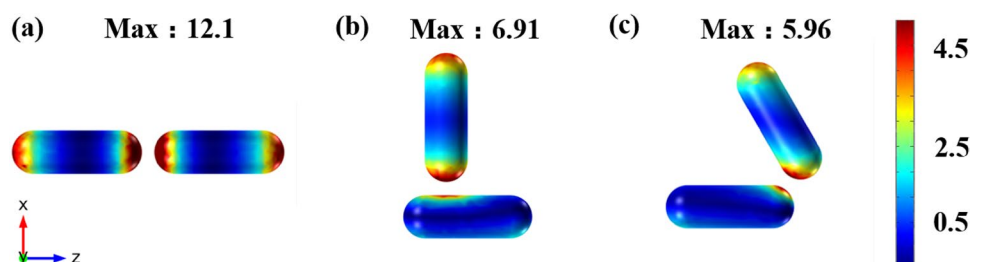


Fig. 8 SEM images of the welding Au NRs dimers with different pulse energy **a** 1 mJ/cm^2 and **b** 1.5 mJ/cm^2 by fs laser

Fig. 7 FEM simulation of the electric field enhancement of Au NRs dimers ($40 \times 13 \text{ nm}$) with different angel **a** 180°, **b** 90°, and **c** 60°, the gap distance between two Au NRs $d=5 \text{ nm}$, the incident laser was propagated along the z direction and polarized along the y direction



4 Conclusion

In this paper, Au NRs have been fabricated at room temperature simply by the seed-mediated growth method. Then Au NRs have been self-assembled in the manner of tip-to-tip dimers by DNA origami technique. Finally, the formed tip-to-tip Au NRs dimers on the substrate are under irradiating 800 nm femtosecond pulses with pulse energy of 500 $\mu\text{J}/\text{cm}^2$ just for 3 min which produced nano-weld tip-to-tip Au NRs dimers. Based on the rapid development of chemical synthesis, the fabrication of arbitrary shape and size of the nanoparticles can be achieved. And with the structural diversity and nanoscale addressability of DNA origami technique, various nanoscale plasmonic nanostructures can theoretically be constructed. Furthermore, building plasmonic nanostructures which can interact with light to produce various effects such as photothermal effects, photoelectric effects. And combing the DNA origami technique with welding the nanostructures through femtosecond laser which can manufacture nanodevices with unique plasmonic effects. For example, the control over assembling and welding of nanostructures could potentially be used for applications in the related field such as the manufacture of electronic and optoelectronic devices in nanoscale. This technical route offers a potential expectation for programmable large-scale nanocircuits generation which may able to be incorporated with the electrical and optical and biological or chemical components to applied in the related field such as sensing, drug delivery, cancer treatment, and information processing.

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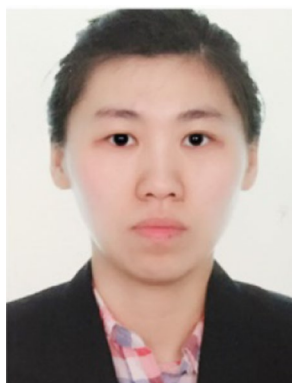
Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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