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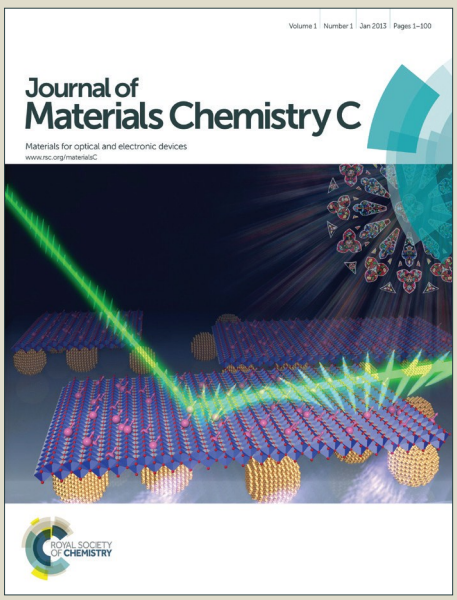





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# Enhanced Field Emission of Au Nanoparticle Decorated SiC Nanowires

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## ABSTRACT

Silicon carbide (SiC) nanostructures are considered as one of the excellent candidates for field emitters, owning to their versatile superior properties. The field emission with a low turn-on field ($E_{to}$) is crucial and highly desired for their practical application. In the present study, SiC nanowires (SiCNWs) were grown on the carbon fabrics *via* the pyrolysis of polymeric precursor, followed by surface decoration of Au nanoparticles by a sputtering process. The characterizations of their field emission (FE) properties revealed that the Au nanoparticles decorated SiC nanowires exhibit remarkably enhanced FE performances. Compared to those of the bare counterparts (*i.e.,* without the Au nanoparticle decoration), the $E_{to}$ of Au decorated SiCNWs was decreased drastically from 2.10 to 1.14 V/$\mu$m. The field enhancement factor ($\beta$) of the Au decorated SiCNWs are *ca.* 6244±50, which is nearly 6 times to that of the bare counterparts. The enhanced FE behaviors were mainly attributed to the synergistically increased $\beta$ and decreased $\Phi$ of the SiCNWs induced by Au decoration.

## 1. Introduction

Field emission (FE) has attracted many attentions over past two decades, due to their very promising prospect to be applied in displays and other electronic devices.[1,2] Among the family of the field emitters, SiC is regarded as one of the most important candidates for constructing advanced FE devices, owing to its excellent properties, such as superior mechanical properties, high electron mobility, low work function, high strength and stiffness, high-temperature stability,[3-10] as well as high corrosion resistance and thermal conductivity,[11] which enable them with the unique advantage to be serviced under high-temperature/high-voltage/high-power harsh environments. Up to date, many works have reported on the FE properties of SiC low-dimensional nanostrucutres in various morphologies with the typical turn-on fields ($E_{to}$, defined to the electric field required to produce a current density of 10 $\mu$A/cm$^2$) often falling in just several V/$\mu$m with the work stability up to 500 °C, suggesting their totally excellent FE performances.

However, obtaining a low $E_{to}$ is till one of the ground challenges to push forward the practical applications of the SiC field emitters. Based on the Fowler-Nordheim (F-N) theory,[12] there are mainly two effective strategies for enhancing the FE performance: one is to increase the field enhancement factor ($\beta$), and the other is to decrease the work function ($\Phi$). For examples, it was reported that the FE performance of SiC emitters was enhanced by decreasing the radii of curvature of the nanostructures with sharp tips to enhance the $\beta$ values based on the local field enhancement effect,[8-10,13,14]; The FE behaviors of the SiC nanowires was improved by tailoring the band gap of established nanomaterials *via* doping strategy (*e.g.,* incorporating Al,[8,14] N,[6,7,10,15,16] B[9,17] and P[18]), which could favor a more localized state near the Fermi energy level, thus leading to the reduce of work function. Recently, it was reported that the surface decoration of the emitters with metal nanoparticles (*e.g.,* Al, Cu, Mn, Ru, Cs，Ag and Pt

nanoparticles) could be effective to greatly increase the density of the emission sites and reduce the $\beta$ of the emitters, thus correspondingly enhance the FE properties.[1,19-23] For instance, Sridhar *et al.* reported that the Al nanoparticles decorated carbon nanotube arrays exhibit exciting excellent FE properties with ultralow $E_{to}$ and $E_{thr}$ (defined to the electric field required to produce a current density of 1 $m$A/cm$^2$) of 0.13 and 0.14 V/$\mu$m, respectively;[24] Gautier *et al.* proposed that the electric field of the carbon nanotubes for producing a current density of 0.1 $\mu$A/cm$^2$ was decreased from 2.68 to 0.96 V/$\mu$m once decorated by Au nanoparticles.[25] However, to the best of our knowledge, there are still no works shed light on decorating SiC nanostructures with metal nanoparticles for obtaining the enhanced FE performances.

In this work, we report the exploration of Au nanoparticles decorated SiCNWs field emitters for the enhanced FE behaviors. It is found that, as compared to those of the bare SiCNWs, the $E_{to}$ and $E_{thr}$ of the as-synthesized Au-SiCNWs have been decreased from 2.10 and 2.75 to 1.14 and 1.75 V/$\mu$m, respectively, with the $\beta$ improved for near 6 times up to 6244±50, suggesting that the metal nanoparticles decoration could be an effective route to significantly enhance the FE performances of the SiC field emitters.

## 2. Experimental Procedure

The SiC nanostructures were synthesized by catalyst-assisted pyrolysis of polysilazane (PSN, Institute of Chemistry, Chinese Academic of Science, China) precursor in a graphite-heater furnace. The PSN was commercially available and directly used without further purification. The PSN was first solidified by heat-treatment at 260 °C for 30 min under Ar atmosphere, and then subjected to be ball-milled into fine powders. The carbon fiber fabric substrate was immersed into the ethanol solution

of $Co(NO_3)_2$ with a concentration of 0.05 mol/L for 2 min. After being dried in the air at room temperature (RT), the substrate was placed on the top of a high-purity graphic crucible (purity: 99%) containing precursor powders (0.3 g), and then the crucible with the substrate was placed into the graphite-heater furnace. Subsequently, an atmosphere mixture of 5 vol% nitrogen and 95% vol% argon (both are 99.99% purity, 0.1 Mpa) was introduced into the chamber. The system was evacuated to $10^{-3}$ Pa and then purged for three times to reduce the $O_2$ to a negligible level. Finally, the furnace was heated from room temperature (RT) to 1450 °C within 48 min, and then cooled down to 1100 °C within 25 minutes followed by furnace cool to RT. The whole pyrolysis process was performed under the atmosphere mixture of 5% nitrogen and 95% argon with a flowing rate of 200 sccm. The resultant SiCNWs was then decorated with Au nanoparticles by a sputter coater for 15 s under RT.

The obtained product were characterized using field emission scanning electron microscopy (FESEM, S-4800, Hitachi, Japan), X-ray diffraction (XRD, D8 Advance, Bruker, Germany) with a Cu *Ka* radiation ($\lambda$=1.5406) and high-resolution transmission electron microscopy (HRTEM, JEM-2100F, JEOL, Japan). The FE properties of bare SiC and Au nanoparticles-decorated SiC emitters were performed on a home-built high vacuum FE setup with a base pressure of $1.5\times10^{-7}$ Pa at RT. The current–voltage (*I–V)* curves were recorded on a Keithley 248 unit with a detection resolution of 0.1 *f*A. The distance between the surface of SiC nanostructures and the anode of vacuum chamber was fixed at 700 $\mu$m

## 3. Results and Discussion

Fig. 1(a-b) show the representative SEM images of Au decorated SiCNWs grown on the carbon fabric substrate under different magnifications. Fig. 1(a) reveals the uniformly growth of SiC nanowires

on the substrate with the typical lengths of dozens of micrometers. The catalyst particles observed on the top of the nanowires (Fig. 1 (b)) suggest that the nanowires follow the typical vapor–liquid–solid (VLS) growth mechanism. Fig. 1(c) represents the enlarged surface of a single SiC nanowire, indicating that a large number of isolated nanoparticles, rather than the continuous film, are uniformly decorated around the surface of the SiC nanowire. Fig. 1(d) shows the typical XRD patterns of the as-synthesized SiCNWs with and without the decoration of the Au nanoparticles. It discloses that, besides the diffraction peaks of carbon fiber substrate, the other characteristic peaks are all assigned to the 3*C*-SiC (JCPDS Card No. 29-1129), suggesting that the grown SiCNWs with and without Au nanoparticle decoration are both of pure 3*C*-SiC phase. The strong and sharp diffraction peaks imply that the as-grown SiCNWs possess a good crystallinity. The absence of Au signals in the XRD pattern of Au-decorated nanowires may be attributed to the low Au content. Fig. 1(e-f) are the typical SEM images of the as-synthesized SiCNWs without the Au nanoparticle decoration. The similar morphologies of the obtained SiCNWs with and without the decoration of Au nanoparticles under this given magnification suggest that, the surface decoration of Au nanoparticles via the sputter coater has not destroyed the morphology of the SiCNWs.

Further characterization of the grown Au nanoparticle decorated SiCNWs is carried out under TEM. Fig. 2(a-b) shows the typical TEM images of a single SiC nanowire under different magnifications, suggesting the surface of nanowire was covered with numerous Au nanoparticles, which is consistent with the SEM characterizations. The dark islands observed around the nanowires surface further confirm that the Au nanoparticles are isolated ones, rather than continuous film. It discloses that the typical diameter of the SiCNWs is 100±10 nm with uniform decoration of nanoparticles around the surface. The density of Au nanoparticles is *ca.* $\sim 1\times10^{12}/cm^2$ and the nanoparticles average size is

estimated to 3.45 nm with a corresponding error of 0.8 nm, shown as the inset in Fig 2(b). For comparison, Fig. 2(c) presents the SiCNWs without the decoration of Au nanoparticles, showing their clear and smooth surface. It indicates that, except the rough surface, there are no obvious changes in the SiC nanowires caused by the deposited Au nanoparticles. Fig. 2(d) shows the corresponding HRTEM images under high magnifications, which suggest that the average diameter of the decorated Au nanoparticles is ~3.45 nm. The Au nanoparticles configuration is just after the sputter process, which could be mainly attributed to the used short time for the sputtering process (15 s). The amount of the deposited Au nanoparticles could be regulated by adjusting the deposited times. The growth of the Au nanoparticles might be dominated by a partial coalescence process,[26] which makes the sputtered metals tend to form three-dimensional islands once the deposited time is fixed at a short time. The inset in Fig. 2(d) provides the corresponding SAED pattern of the SiC nanowire, which is identical over the entire wire and indexed to be of 3*C*-SiC (JCPDS Card No. 29-1129). Fig. 2(e) is the enlarged HRTEM image of the Au nanoparticle decorated SiCNWs, which is recorded from the marked area of A in Fig. 2(d). The measured distances between two neighbored planes are of 0.24 and 0.25 nm, responding to the spacing of {111} planes of cubic Au and SiC, respectively, verifying that the growth of SiCNWs with the decoration of Au nanoparticles. Both the SAED and HRTEM characterizations imply that the as-synthesized SiC nanowire is single crystalline with the growth direction of [111]. The EDX spectrum (the experimental setup not shown here) recorded from the nanowire body for more than 10 wires reveal that it mainly consists of Si, C, Au, O and Cu. The detected Au signals further demonstrate the existence of Au nanoparticles decorated around the SiC nanowire surface. The O is from the amorphous $SiO_x$ outlayer around the SiC wires,[14] and the Cu comes from the copper grid used to support the TEM sample. The atomic ratio of Si to C, within the experimental limit, is close to 1:1, suggesting the

as-synthesized nanowire is SiC. The average N doping level of ~2.43 *at.*% of the SiC nanowire indicates that the resultant product is *n*-type SiCNWs.[6,15,16]

The FE characteristics of the bare and Au nanoparticles-decorated SiCNWs are revealed by emission current density (*J*) *versus* the applied electric field (*E*) plots, as shown in Fig. 3(a). The consistent and relatively smooth curves imply their stable electron emission. To understand the FE behavior, the *J*-*E* data are analyzed by the *F*-*N* equation:[12]

$$J = (A\beta^2 E^2 / \Phi) \cdot \exp(-B\Phi^{3/2} / \beta E)$$

Where *J* is the emission current density, *E* is the applied field, $\Phi$ is the emitter material work function, *A* and *B* are constants, and $\beta$ is the field enhancement factor. The corresponding *F*-*N* plot obtained by plotting ln ($J/E^2$) *versus* 1/*E* is displayed in Fig. 3(b). The measurements of the FE performance in our case are performed under RT, suggesting that the electron emission should be mainly caused by the applied voltages, and the thermal-induced electron emission could be excluded. The approximately linearity behaviors indicate that the electron emission from these two samples both follow the traditional *F*-*N* rule. By taking the work function of 4.0 eV for SiC at RT,[3] the field enhancement ($\beta$) are *ca.* 1150±50 and 6244±50 for the bare and Au nanoparticle decorated SiCNWs, respectively. It seems that the $\beta$ of Au-decorated SiCNWs are nearly 6 times to that of the bare counterparts, which could facilitate the electron emission from the SiCNWs.[27] Fig. 3(c-d) show the variation of the $E_{to}$ and $E_{thr}$ of the bare and Au-decorated SiCNWs, which are 2.1, 1.14 V/$\mu$m and 2.75, 1.75 V/$\mu$m, respectively. It implies that, as compared to that of bare SiCNWs, the $E_{to}$ of the Au-decorated SiCNWs is remarkably reduced for nearly one times (*i.e.,* 2.1 *vs* 1.14 V/$\mu$m), suggesting the significantly enhanced field emission caused by the decoration of the Au nanoparticles. It is also promising that the FE performance of the SiC

nanowires could be further enhanced once the decoration of the Au nanoparticles has been optimized in the size, density and spacing by tailoring the sputtering process.[26,28-30]

The enhanced FE properties of the Au nanoparticle decorated SiCNWs could be attributed to the following issues. i) The significantly increased emission sites. As schematically shown in Fig. 4(a), numerous Au nanoparticles have been fixed around the surface of the nanowires after the decoration treatment. Owing to their geometries of small curvature radius with the average diameter sized in just ~3.5 nm, these nanoparticles could be acted as the effective electron emission sites, which could bring a significantly increased density of the electron emission sites of the SiC nanowires.[17,23,31] Accordingly, the ability of the electron emission from one SiC nanowire with the Au nanoparticle decoration can be comparable to those of numerous bare SiC nanowires, leading to the greatly enhanced field emission. ii) The local field enhancement effect. As seen from Fig. 1(f) and Fig. 2(d-e), the diameter sizes of the bare SiC nanowire body and Au nanoparticles are 100±10 and 3.45±0.8 nm, suggesting that the curvature radius of the nanoparticles are much smaller than those of the bare SiC nanowires. The emission site with a smaller curvature radius could benefit to enhancing the electron emission due to the strong local electric field at the tips and the unique direction of electron emission.[32] That is to say, the decorated Au nanoparticles favor the electron emission from the SiC nanowires with a significantly enhanced local field enhancement effect, thus leading to the enhanced field emission. The stronger local field enhancement effect, together with the remarkably increased electron emission sites as mentioned above, makes the Au nanoparticle decorated SiC nanowires with profoundly improved field enhancement factor ($\beta$) (*i.e.,* increased from 1150±50 to 6244±50 for nearly 6 times after the Au decoration). iii) The reduced work function ($\Phi$). The reported works proposed that the electrical property of Au nanoparticles/SiC interface, especially the existed Schottky barrier height (SBH), is critically important

for the exploration of their potential devices applications.[33,34] It is known that the field emission is mainly due to the tunneling of electrons through the surface barrier (*i.e.,* from the Fermi level ($E_f$) to the vacuum level ($E_0$)), which can be attributed to the quantum mechanical tunneling mechanism, namely, the Fermi energy determines the field emission current. The $\Phi$ is defined to the energy barrier between the $E_f$ and $E_0$ (as schematically shown in Fig. 4(b), *i.e.,* $\Phi = E_0 - E_f$), implying that a lower $E_f$ would result in a larger $\Phi$ with a weaker FE ability. It was reported that the $\Phi$ of Au nanoparticles would be decreased from 5.1 to 3.6 eV once the size of Au nanoparticles is scaled down to 10 nm.[33,35] In the present work, the average size of decorated Au nanoparticles is 3.45±0.8 nm, implying that the $\Phi$ of the Au nanoparticles should be lower than 3.6 eV. It is smaller than that of SiC (4.0 eV),[3] implying the $E_f$ of the Au nanoparticles (referred to $E_{fa}$ in Fig.4(b)) should be higher than that of the SiC (referred to $E_{fs}$ in Fig.4(b)). Once the Au nanoparticles has been decorated around the surface of the SiC nanowires, the SiC-Au heterojunction could be formed, which leads to the formation of the forward-biased built-in electric field.[36] Accordingly, the electrons would migrate from Au nanoparticles to SiC driven by the formed built-in electric field, leading to the formation of a balanced Fermi level (referred to $E_{fb}$ in Fig.4(b)),[25] which should be located between the $E_{fs}$ and $E_{fa}$, and thereby make the reduction of the $\Phi$, consequently leading to the enhanced field emission of SiC nanowires.[6,25,35] In a brief word, the decorated Au nanoparticles around the surface of the SiC nanowires could favor them simultaneously with increased $\beta$ and reduced $\Phi$, which result in the significantly enhanced field emission from the SiC nanowires.

## 4. Conclusions

In summary, we have demonstrated the exploration of SiC nanowire field emitters *via* the pyrolysis

of polymeric precursor followed by the decoration of Au nanoparticles by sputtering. The diameters of the grown SiCNWs and decorated Au nanoparticles are 100±10 and 3.45±0.8 nm, respectively. The FE property measurements suggest that their electron emission could be profoundly enhanced by the decorated Au nanoparticles. The Au decorated SiCNWs exhibit a low $E_{to}$ of 1.14 V/$\mu$m, which has been reduced for ~1 times as compared to that of the bare SiCNWs. The $\beta$ of the Au decorated SiCNWs is *ca.* 6244±50, which is almost 6 times to that of the bare counterparts. The enhanced FE behaviors could be mainly attributed to the significantly increased emission site density, stronger local field enhancement effect, and decreased work function ($\Phi$), owing to the decorated Au nanoparticles with small curvature radius. It is promising that current work could be an efficient strategy for profoundly enhancing the FE performances of the SiC nanostructures with a lower $E_{to}$, which could push forward them to be potentially applied in electron emission guns, flexible field emission displays and e-papers.

## *Acknowledgements*

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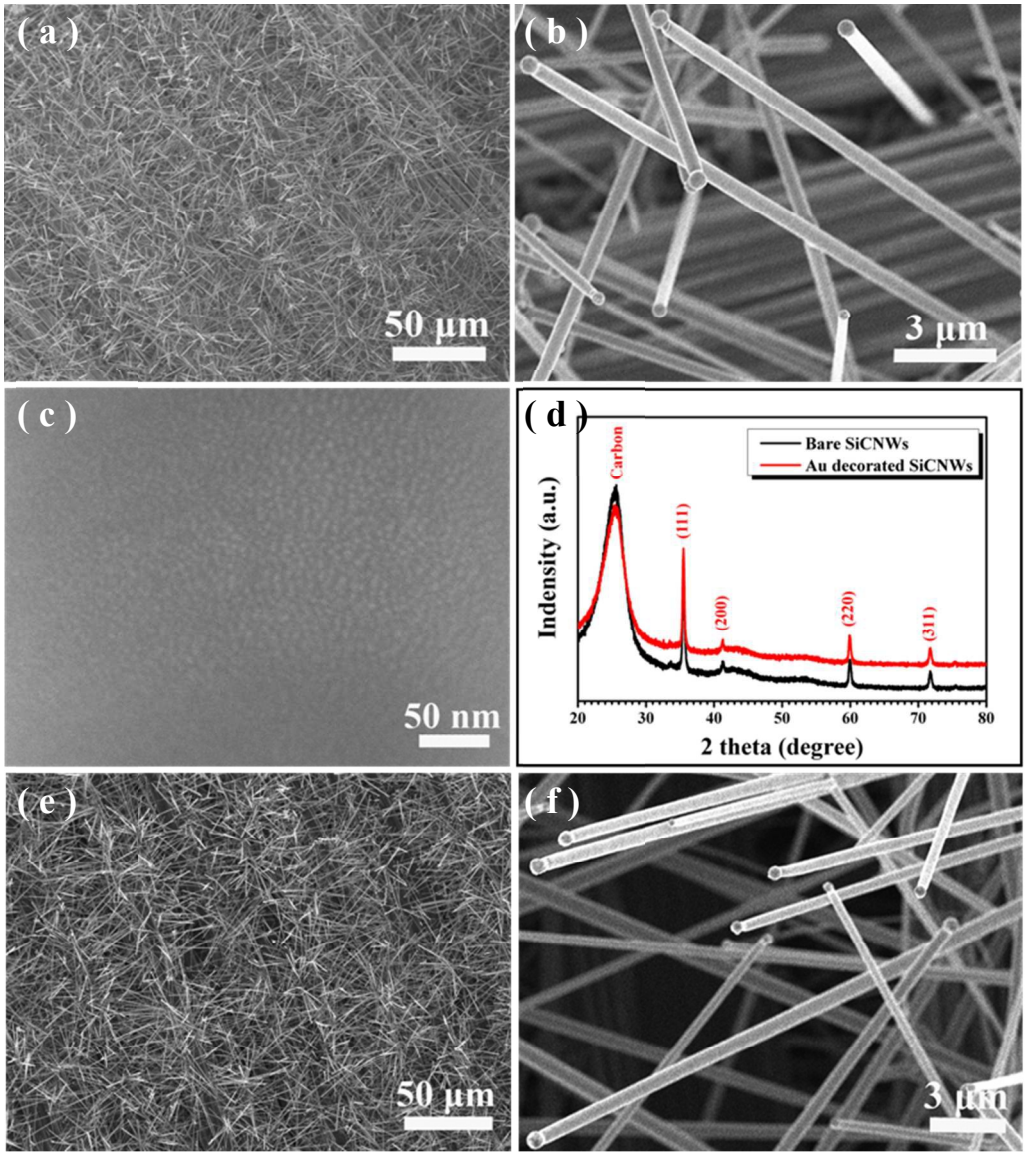



**Fig. 1** (a-c) Typical SEM images of Au nanoparticles decorated SiCNWs under different magnifications. (d) Typical XRD patterns of the grown SiCNWs with and without Au decoration. (e-f) Typical SEM images of the SiCNWs without the decoration under different magnifications.

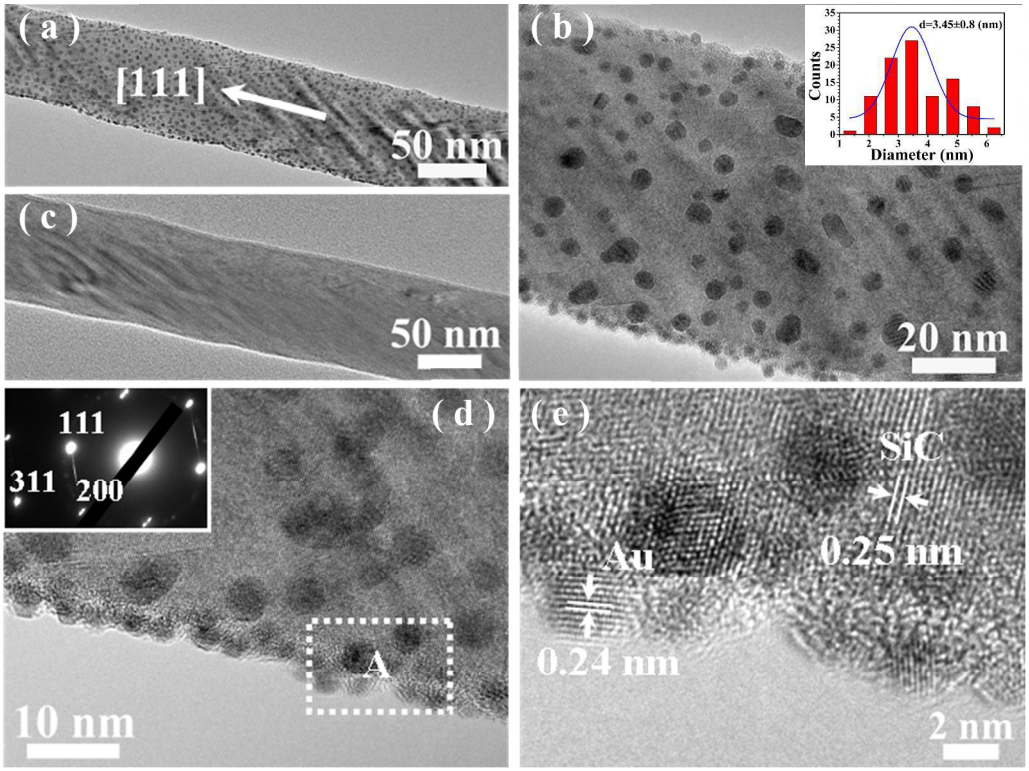



**Fig. 2** (a) Representative TEM images of a single Au nanoparticle decorated SiCNW under low magnification. (b) TEM images of a single Au nanoparticle decorated SiCNW under high magnification. The inset is the size distribution of Au nanoparticles on the surface of SiCNW. (c) Reprentative TEM image of a single SiCNW without the decoration. (d-e) Typical HRTEM image of Au nanoparticle decorated SiCNWs under different magnifications. The inset in (e) is the corresponding SAED pattern.

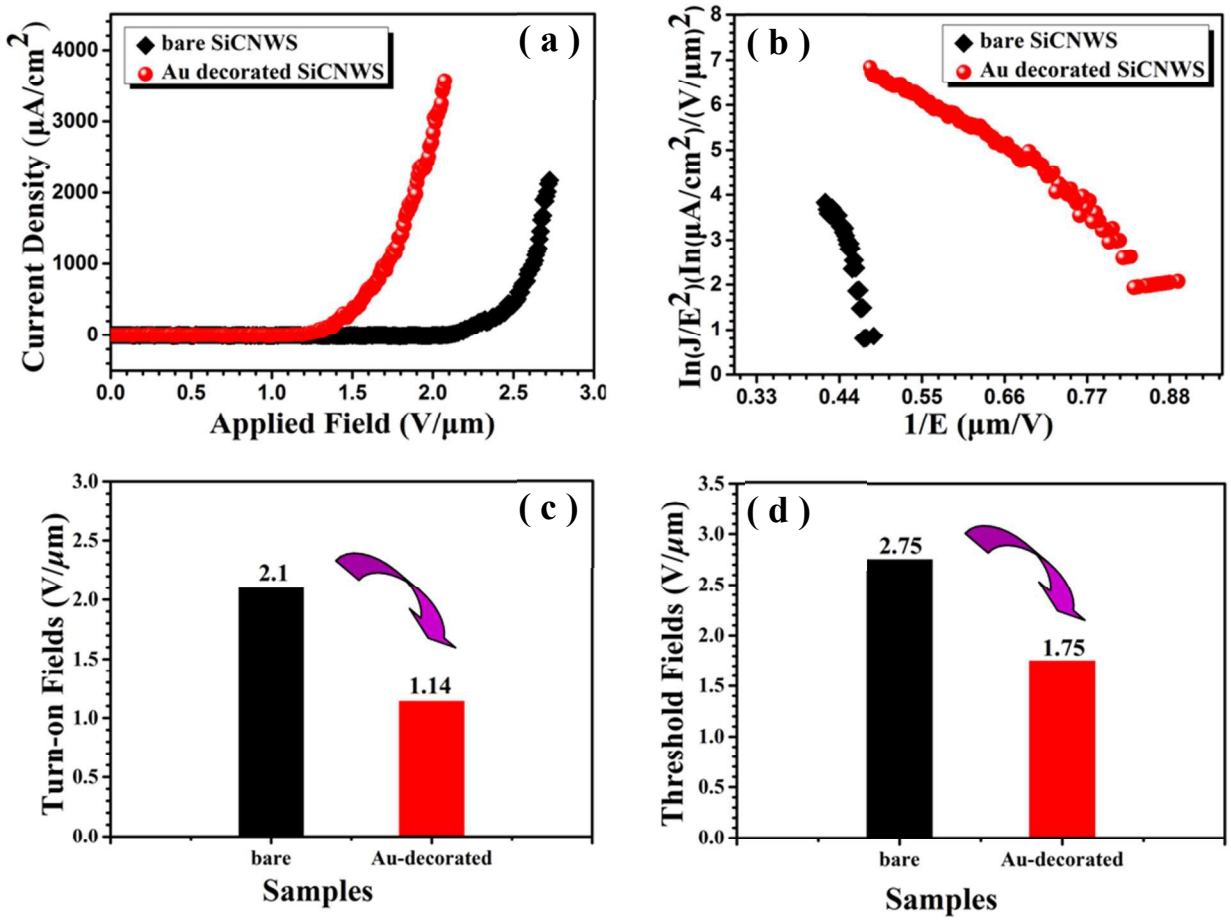



**Fig. 3** (a) *J-E* plots of the SiCNWs with and without Au nanoparticles decorated. (b) The corresponding *F-N* plots. (c-d) The variations of $E_{to}$ and $E_{thr}$ between the SiCNWs with and without the Au decoration, respectively.

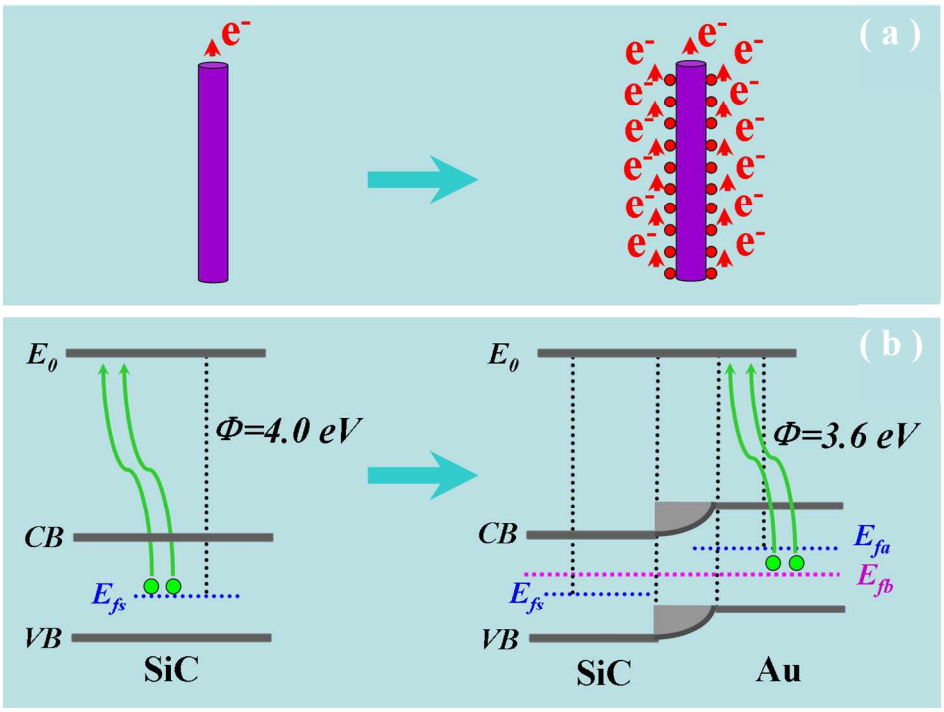
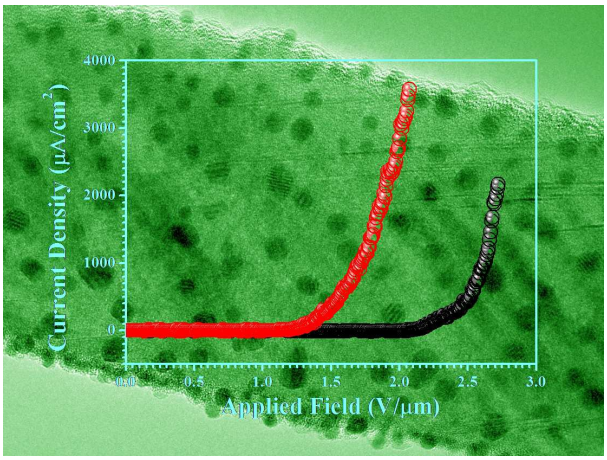



**Fig. 4** (a) Schematic illustration of the electron emissions of the SiCNWs after the Au decoration. (b) Schematic illustration of the change of the $E_f$ and electrons transfer induced by the Au decoration.

## Graphical Table of Contents




We reported Au nanoparticle decorated SiC nanowire field emitters with enhanced $E_{to}$ and $\beta$ of 1.14 V/$\mu$m and 6244±50, respectively.