Layered 2D materials serve as a new class of substrates for templated synthesis of various nanomaterials even with highly dissimilar crystal structures; thus overcoming the lattice constraints of conventional epitaxial processes. Here, molybdenum disulfide (MoS₂) is used as a prototypical model substrate for oriented growth of in-plane Au nanowires (NWs) despite the nearly 8% lattice mismatch between MoS₂ and Au. Au NWs on the MoS₂ surface are oriented along three symmetrically equivalent directions within the substrate arising from the strong Au–S binding that templates the oriented growth. The kinetics of the growth process are explored through experiments and modeling. Strong charge transfer is observed between Au NWs and MoS₂, resulting in degenerate p-doping of MoS₂.

1. Introduction

The ability of one material to influence the crystallography of another at an interface is the foundation for constructing functional heterojunctions for a myriad of electronic, energy, and photonic applications. The ultimate limit of this process is epitaxy, whereby the second material, usually a thin layer, fully adopts the planar interface symmetry of the host substrate. Metal nanostructures with an epitaxial growth mode on semiconducting compounds [1] and metal oxides [2] have been widely studied for use in catalytic [2a,b] plasmonic [1a,2d] and nanoelectronic [1d,3] devices. Morphologies of the metal nanostructures should be taken into consideration to maximize the properties of the metal surface. In general, morphologies such as spheres (particles) [1d,4] platelets (squares, triangles etc) [1c,e] and vertically aligned pillars [5] have been systematically generated. Since the interface of the junction is often a 2D crystalline plane with high symmetry, growth of anisotropic shapes, such as horizontal nanowires (NWs), on the plane can be difficult. Previously, in-plane oriented epitaxial growth of Si [6], metal oxide [7], and III–V NWs [8] have been demonstrated via the vapor–liquid–solid (VLS) process with metal nanoparticles as catalysts. In the conventional epitaxial growth process, the crystal structure and lattice constants of the substrates and grown material need to be closely matched. Here, we explore the use of layered materials as template substrates for oriented growth of in-plane metal NWs. As a proof of concept, we demonstrate growth of in-plane oriented Au NWs with C3 symmetry on molybdenum disulfide (MoS₂) surfaces. The rich surface chemistry of MoS₂ with weakly bonded layered structure facilitates the oriented growth of Au NWs despite the highly mismatched crystal structures of Au and MoS₂ with a large lattice mismatch of 8%. This work presents a unique platform for growth of highly dissimilar heterostructures using layered substrates; overcoming the lattice constraints of conventional epitaxial processes. Furthermore, we demonstrate degenerate p-doping of MoS₂ as a result of charge transfer with the Au NWs.

Transition metal dichalcogenides (TMDCs) are promising materials for the next generation electro-optical devices due to their atomically thin layered nature [9] variety of band gaps ranging from metallic to insulating depending on the combination of metals and chalcogenide atoms [10] relatively high carrier mobilities (∼200 cm² V⁻¹ s⁻¹ for WSe₂) [11] and in some cases, high thermal and chemical stabilities [10,12] for device fabrication processing. The bonding within each layer of TMDCs is ionic/covalent while the adjacent layers are coupled via van der Waals (vdW) interactions. Due to this weak interlayer binding and very smooth surface, vdW epitaxy has been proposed to form functional crystals on the surface of layered materials [13]. Previously, vdW epitaxy of various 2D layered materials based on a combination of TMDCs, h-BN, and graphene has been reported [13a,14,15]. Since the interaction with the substrate is not strong, the vdW epitaxy has mainly generated 2D symmetric crystals due to the laterally highly-symmetrical plane at the
By casting AuCl₃ solution, Au atoms are supplied on the surface of MoS₂ (potential of Au ≈ 5.5 eV vs vacuum level).[17] With the valence band edge of MoS₂ lower than ≈ 5.5 eV, a redox reaction between AuCl₃ and MoS₂ would occur spontaneously on the surface (Figure 1b,c) to supply Au atoms effectively.[16,17b] Previously, formation of Au nanoparticles on MoS₂ has been reported using a similar AuCl₃ treatment.[1c,d,4,17a,18] Stimulated from the research, in-plane oriented Au NW formation was targeted in this research.

Figure 2a,b shows atomic force microscopy (AFM) images of a 6.4 nm thick MoS₂ flake before and after the AuCl₃ treatment, respectively. NWs are clearly observed on the surface after AuCl₃ treatment and thermal annealing. The length of the NWs is ≈ 500 nm with a diameter of ≈ 10 nm from the AFM observations. Since no NWs were observed on the surface of SiO₂ (Figure 2b), the growth of the NWs is expected to be controlled by the interaction between Au³⁺ and MoS₂. The NW formation is independent of MoS₂ flake thickness: NWs can also be formed on bilayers (1.5 nm thick, Figure 2c) and monolayers (0.7 nm thick, Figure 2d) of MoS₂. We analyzed the statistical distribution of the angles of the NWs shown in Figure 2d. We found that NWs were well oriented with peak distributions at approximately every 60°, along the +40°, 95°, and 154° directions, indicating the C₃ symmetry orientation (Figure 2e). Given the consideration of the C₃ symmetry of the ab plane of MoS₂ (Figure 1a) and no NW formation on the SiO₂ surface, the NW growth is strongly influenced from the MoS₂ substrate which is also suggested from the nuclei preferentially oriented along a lattice of MoS₂ (vide infra).

We further analyzed the effect of the AuCl₃ concentration and deposition time on NW morphology. Decreasing the concentration of AuCl₃ from 30 to 1 wt% by dilution with Milli-Q water decreases the diameter of NWs (d) from d = 17.5 nm (30 wt%) to d = 6.5 nm (1 wt%) (Figure 3a–d and Figure S1, Supporting Information) using the same deposition procedures as previously described (1 min deposition followed by 150 °C annealing for 2 min). Higher concentrations facilitate more Au deposition on the surface, leading to the generation of thicker Au NWs. We also found that there is a critical concentration for the generation of NWs based on the 1 min deposition procedure. Below 0.5 wt%, nanoparticles are formed instead (Figure 3e). However, by using a longer deposition time, even the 0.5 wt% AuCl₃ can be used to generate NWs (Figure 3f).

Figure 1. a) Schematic illustration of the growth of the in-plane oriented Au NWs on the MoS₂ surface. By casting AuCl₃ solution, Au atoms are supplied on the surface of MoS₂ (ab plane). Au atoms diffuse to form nuclei on the surface and eventually generate NWs by application of thermal energy. The NWs are in-plane oriented, reflecting the hexagonal lattice of the ab plane MoS₂. b) Schematic illustration of the redox reaction between Au³⁺ and MoS₂ to form an Au²⁻–MoS₂ complex. c) Qualitative energy diagram between MoS₂ and AuCl₃. The reduction potential of Au³⁺/Au²⁻ is ≈ 5.5 eV vs vacuum level. Since the valence band edge of MoS₂ is lower than ≈ 5.5 eV, electron transfer from MoS₂ to Au³⁺ occurs.

2. Results and Discussion

Figure 1a shows an illustrative image of the growth of Au NWs from nucleation to growth on the surface of MoS₂. Casting AuCl₃ solution onto MoS₂ supplies Au atoms on the surface. To form Au NWs on MoS₂, we cast a 30 wt% AuCl₃ solution in diluted HCl (Sigma-Aldrich) onto a mechanically exfoliated MoS₂ flake for 1 min and then blow off the extra solution with N₂ gas followed by annealing at 150 °C for 2 min on a hot plate in an ambient environment. During the thermal annealing process, Au atoms on the surface aggregate to form nuclei and further assemble to form NWs. Uniquely, the NWs are oriented with C₃ symmetry, suggesting coherence with the threefold symmetry[10] of the surface of MoS₂. Previous works have demonstrated self-assembled monolayers (SAM) of alkyl thiol molecules (R-SH) on Au surfaces.[16] In the SAM case, a strong interaction between S and Au facilitates the formation of the monolayer of thiolate molecules on top of Au.[16] The structure shown in Figure 1a is similar but opposite from the usual SAMs; Au atoms are instead assembled onto the S lattice of MoS₂, AuCl₃ was chosen as an Au source because it has a high reduction potential (≈ 5.5 eV vs vacuum level).[17]
This result also follows from the above discussion, indicating a critical amount of Au on MoS$_2$ needed to generate NWs. A longer deposition time of 60 min shows a similar distribution of Au NWs (≈10 nm thickness) as in the 10 min case (Figure 3g), indicating saturation of Au coverage on the MoS$_2$ surface by AuCl$_3$ treatment. Upon heating, these Au atoms diffuse and nucleate clusters. Higher concentrations lead to a higher nucleation rate for Au clusters. Faster growth rates are also expected for high concentration solutions.

Assembly of Au atoms on MoS$_2$ is found to show various morphologies from nanoparticles to NWs depending on the annealing conditions. Importantly, the NWs can be formed only in a limited annealing temperature condition. This is also observed under AFM before/after annealing of a MoS$_2$ flake treated with AuCl$_3$. Features were not observed before thermal annealing while on the other hand, NWs were clearly generated after annealing at 150 °C for 5 min (Figure S2, Supporting Information). Figure 4a shows a growth morphology diagram of Au on MoS$_2$ with various annealing times/temperatures. At the lower temperature region (below 100 °C), Au shows up only as small nanoparticles on MoS$_2$ (≈4 nm, Figure S3, Supporting Information). Increasing the temperature above 100 °C results in NW formation, suggesting that surface diffusion of Au is important to generate NWs (Figures S4 and S5, Supporting Information). With further annealing (either higher temperature or longer annealing time) the NWs display a pearling instability and form Au clusters (as noted in Figure 4). The observation of NW growth suggests an anisotropic growth rate along different Au crystalline facets. While the exact mechanism is intriguing and not well established, the presence of ions would play a critical role in the NW growth, similar to previously reported solution phase synthesis of NW growth. The importance of AuCl$_3$ existence on the MoS$_2$ surface is confirmed from a control experiment with evaporated Au (0 valence gold) that resulted in platelet structures instead of NWs after thermal annealing (Figure S7, Supporting Information). The C$_3$ symmetry of the NW growth orientation suggests strong binding interaction between Au and S atoms on the surface of MoS$_2$. Thus, despite the different crystal structure and large lattice mismatch (≈8%) of the two materials, the MoS$_2$ substrate acts as an effective template in guiding the orientation of the grown NWs on the surface.

The mechanism by which the MoS$_2$ substrate templates the growth process is not fully understood. Efforts to observe directly the relevant interface have been thwarted by the instability of the wires in the presence of an electron beam and/or light. When exposed to these forms of radiation, the NWs display a pearling instability and form Au clusters (as noted in Figure 4). One possible origin of the templating is that the wires are growing via vdW epitaxy. In this case, the growing wires would continue to feel the atomic scale corrugation of the substrate through the weak vdW bonding between the substrate and the wires, yielding a preferred orientation. Alternatively, more traditional epitaxy is also a possibility. As mentioned above, the lattice mismatch for {111} oriented growth is approximately 8% biaxially. (The lattice mismatch for {001} oriented growth would introduce lower, but still significant strain in the growing film.) Nevertheless, transmission electron microscopy (TEM) studies of the growth of Au on MoS$_2$, performed in situ, resulted in Au platelets that grew epitaxially on the substrate.
A recent theoretical analysis suggests that \{111\} oriented epitaxy is favored by the strength of the Au–S bond at the interface, and the elastic compliance of the MoS$_2$ substrate. Similar factors could be influencing the growth of the NWs, and an epitaxial relationship would help to explain the preferred growth direction.

To characterize the state of the Au clusters on MoS$_2$, we carried out X-ray photoelectron spectroscopy (XPS) analysis (Figure 5a,b). Figure 5a presents Au core levels (4f) from the treatment with 30 wt% AuCl$_3$ solution followed by thermal annealing as shown in Figure 2. The four peaks observed correspond to Au$^{3+}$ and Au$^{0}$ doublets, with both 4f$_{5/2}$ and the indicated 4f$_{7/2}$ components fit as Voight lineshapes. The peak area ratio corresponding to 4f$_{5/2}$ of Au$^{3+}$ to Au$^{0}$ is \approx 23\%. This suggests that close to 80% of the Au$^{3+}$ is converted into Au$^{0}$ on the MoS$_2$ surface, indicating strong electron transfer between MoS$_2$ and Au$^{3+}$. Figure 5b indicates the core levels of Cl (2p) on the same sample. Two types of Cl species are observed, corresponding to Cl–Au species originating in [AuCl$_4$]$^{-}$ and Cl atoms on MoS$_2$ which would be in the form of Cl$^{-}$ ions. From the relative area of each Cl core level peak, we estimate \approx 30\% of the total Cl atoms are Cl$^{-}$ ions on MoS$_2$. According to these results, the surface is mostly coated with elemental Au clusters with measurable [AuCl$_4$]$^{-}$ content. The X-ray diffraction (XRD) analysis in Figure S8 (Supporting Information) further identifies the assembly of crystalline Au on MoS$_2$ after AuCl$_3$ treatment and annealing.

The preferential crystallographic alignment of some of the Au nanoparticle nuclei prior to Au NW growth was observed by selected area electron diffraction (SAED) using TEM (Figure S9, Supporting Information). SAED diffraction patterns were taken from multiple Au nanoparticles on a free-standing MoS$_2$ flake and exhibited diffraction peaks consistent with \textit{d}-spacings of the bulk Au fcc phase. A consistent preferred orientation was not apparent for this sample size (\approx 20 particles), but aligned diffraction peaks were observed for Au(220)//MoS$_2$(\text{00\overline{1}}), Au(311)//MoS$_2$(\text{00\overline{1}}), Au(111)//MoS$_2$(100), and Au(200)//MoS$_2$(310). Given the variety of observed orientations and noting that not all particles in the set had visible diffraction peaks nor did any appear to share a principle zone axis with MoS$_2$, the MoS$_2$ surface in the TEM study did not

Figure 3. Concentration dependency of AuCl$_3$ on NW formation. a) AFM images of Au NWs/nanoparticles on MoS$_2$ obtained from treatment with various AuCl$_3$ concentrations followed by thermal annealing at 150 °C for 2 min. Concentrations and deposition times are labeled above each image. b–g) Histograms of the diameter of Au nanostructures obtained from AuCl$_3$ depositions with b) 30 wt% for 1 min, c) 3 wt% for 1 min, d) 1 wt% for 1 min, e) 0.5 wt% for 1 min, f) 0.5 wt% for 10 min, and g) 0.5 wt% for 60 min. Except for the case of 0.5 wt% for 1 min, all other conditions result in the formation of NWs. Red solid lines in the histograms are Gaussian fitting lines. In the case of the 30 wt% for 1 min case, the fitting failed due to the high dispersity.
appear to strongly enforce a crystallographic orientation of the Au nuclei. However, wherever the gold planes were visible by SAED they exhibited preferential orientation to a nearby MoS₂ plane. Unfortunately, a similar TEM study of NWs could not be performed due to pearling instability of the wires under the electron beam converting the NWs into nanoparticles.

Theoretical simulation using density functional theory (DFT) was also applied to better understand the nucleation from the interaction between Au atom and MoS₂ on the surface (Figure 5c and Supporting information). Compared to Au atom on the S atom of MoS₂, Au atom on the Mo atom of MoS₂ shows a higher energy (0.12 eV). The result is as expected; Au and S atoms have higher affinity. Another important point is that the binding energy of Au atom on S vacancies was found to be $-2.6$ eV. This interaction is found to be thermodynamically very strong, which would be the main nucleation site for Au clusters.

To analyze the electrical properties of MoS₂ after the generation of Au NWs, we examined the transfer characteristics of back gated MoS₂ field-effect transistors (FETs). FETs were fabricated by mechanically exfoliating $≈100$ nm thick MoS₂ flakes on $260$ nm SiO₂ on a heavily doped p⁺-Si wafer as a universal back gate (Figure 6a,b). Photolithography is used to define source and drain electrodes followed by evaporation of Au/Cr (40/0.5 nm). The channel length of the devices is 10 µm. Figure 6a and Figure S10 (Supporting Information) show transfer characteristic curves (drain voltage ($V_{DS}$) = +1 or $-1$ V for n and p type characteristics, respectively) before/after treatment of 1 wt% AuCl₃ on the device. The pristine device shows n-type behavior as similar to previous reports. After the treatment of the AuCl₃, the polarity of the device changes from n-type to p-type and the ON current level monotonically increases several orders of magnitude ($≈10^3$) with increasing deposition time of the AuCl₃. Previously, a report demonstrated...
AuCl₃ can act as an acceptor; the polarity transformation in our case is comparable. As discussed in Figure 3e,f, since a longer deposition time increases the surface coverage of Au on MoS₂, the hole concentration of MoS₂ is also increased as the deposition time increases. Interestingly, in the case of using 30 wt% AuCl₃ solution (Figure 6b), just a 30 s treatment shows a drastic change in the electrical properties, resulting in small VGS dependency, corresponding to a degenerately doped p⁺situation. Treatments longer than 30 s increase IDS monotonically. From these characteristic curves, we extracted the sheet charge density (nᵢD) using the Equation (1).²⁶

\[ nᵢD = \left( \frac{I_{DS}L}{qWV_{DS}μ} \right) \] (1)

where q is the electron charge, W and L represent width and length of the channel, respectively, I_DS is the drain current at VGS = 0 V, V_DS is the source to drain bias, and μ is the field-effect hole mobility. We used W = 45 μm by averaging the width of the two source and drain metal contacts. As shown in Figure 6c, the carrier density increases with increasing doping time to 2.5 × 10¹⁴ cm⁻² after 2 min. previously, NO₂ was used as a p-type degenerate dopant for WSe₂, with a doping concentration ceiling of ≈10¹³ cm⁻². The growth of Au on the surface of MoS₂ also induces the high carrier concentration due to its effective surface charge transfer interaction.

When measured under vacuum (≈10⁻⁵ Torr), the degenerate p-doped state of Au doped MoS₂ converts into a nondegenerate state with an ON/OFF ratio over 10² (Figure 6d). A reexposure of the device back to ambient environment immediately reverses the transfer characteristic back to a representative p⁺-type degenerate state without gate dependency. From these results, we attribute work function modulation of the Au due to air adsorbates as responsible for the demonstrated reversible transfer characteristic between the degenerate state and
nondegenerate states. Previously, devices with carbon nanotubes (CNTs) as a channel material showed similar reversible transfer characteristics.\(^{[27]}\) The work function of Au is easily affected by adsorbates such as water and oxygen, and in our case, these adsorbates would change the work function of the Au NWs (and nanoparticles) on MoS\(_2\).\(^{[28]}\) As a control experiment, these adsorbates would change the work function of the MoS\(_2\) by the STARnet phase of the Focus Center Research Program (FCRP), by the Joint Center for Artificial Photosynthesis, supported through the Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. XPS characterization was performed at the National Institute for Materials Science, supported through the Office of Science of the U.S. Department of Energy under Award Number DE-SC0004993.

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4. Experimental Section

Fabrication of the Devices: The devices shown in Figure 6 were fabricated via standard photolithographic techniques. Source and drain metal pads (Cr/Au = 0.5/40 nm) were made via electron beam evaporation followed by lift-off.

Treatment of AuCl\(_4^-\) on MoS\(_2\): The preparation of Au NWs was done with the following procedures. First, MoS\(_2\) flakes were exfoliated via mechanical exfoliation onto an Si wafer with 260 nm of thermal oxide. AuCl\(_4^-\) solution (AuCl\(_4^-\) in HCl, Sigma Aldrich) was then dropped onto the MoS\(_2\) flake and subsequently blown off with dry N\(_2\) gas. Finally, the sample was annealed at 150 °C for several minutes (usually 2 min). The test of AuCl\(_4^-\) concentrations ranged from 0.5 to 30 w%, diluted with Milli-Q water from an original concentration of 30 wt%. For electrical measurements (Figure 6), the AuCl\(_4^-\) treatment was done after fabrication of the back gated devices.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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Supporting Information


Oriented Growth of Gold Nanowires on MoS$_2$

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Supporting Information

Oriented Growth of Gold Nanowires on MoS₂

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Sample preparation for TEM observation

A MoS₂ flake was transferred onto a Si plate (a thin Si plate (~5 μm thickness) with 2 μm diameter holes) by a dry transfer technique (Ref. S1). The transferred flake was suspended on the hole in the plate. The flake was treated with a diluted AuCl₃ solution (0.5 wt%) for just several seconds (ca. 2 sec) and the extra liquid was sucked away with a paper cloth. Since the flake was in a suspended situation, dry N₂ gas blow was not used to remove the extra liquid to avoid any mechanical stresses that might damage the flake. The short treatment time was applied to reduce the probability of forming too many or stacked nuclei on the MoS₂. To form the nucleus, the treated sample was kept in a dry N₂ environment at r.t. for 1 month to grow it gradually in order to avoid annealing at a high temperature which may induce mechanical stress to the MoS₂ flake which can dissociate the flake on the Si plate.

Characterization

Optical microscope images are taken using an Olympus BX51 equipped with a digital camera (Olympus, QCOLOR3). Electrical measurements are done with an HP 4155C analyzer. Electrical measurements in vacuum (Figure 6d) are done with a vacuum probe station (Lake Shore Cryotronics Inc.). AFM observations are done with a Nanoscope Dimension 3100 atomic force microscope (Digital Instruments Inc.). XPS measurements are performed with a Kratos AXIS Ultra DLD system with a hemispherical analyzer and monochromatic Al Kα source, and fits were performed using typical Voigt lineshapes, a Shirley background, and
spin-orbit splitting values and ratios typical of the respective 4f and 2p orbitals. XRD patterns were taken on a Bruker AXS D8 Discover GADDS XRD Diffractometer system.

Transmission electron microscopy in Figure S9 was obtained by an FEI Titan microscope operating at 80 kV. The nanoparticles were confirmed to be chemically Au using energy dispersive x-ray spectroscopy. Selected Area Electron Diffraction (SAED) patterns were captured for regions with Au nanoparticles, and a reference MoS$_2$ SAED pattern was captured from an immediately adjacent region. Difference images were created by subtraction of raw SAED patterns taken with identical exposures and the result was normalized symmetrically about zero to a range of -1 to 1 using identical scaling parameters for all images so they remain directly comparable. Au and MoS$_2$ peak d-spacings were measured by Gaussian fit and d-spacings where by calibrated to the MoS$_2$ peaks using a value of $d = 2.73$ Å for (100) peaks. The Au peaks in Figure S9 had measured d-spacings of 2.36 Å, 1.98 Å, 1.40 Å, and 1.30 Å, which are close fits to nominal Au d-spacings for {111} ($d = 2.35$ Å), {200} ($d = 2.03$ Å), {220} ($d = 1.44$ Å), and {311} ($d = 1.23$ Å) planes.

**DFT simulations**

The DFT calculations are performed using the plane-wave code VASP [Ref.S2]. The exchange and correlation energy is described by the generalized gradient approximation (GGA) proposed by Perdew, Burke, and Ernzerhof (PBE) [Ref.S3]. Electron-ion interactions are treated with projector augmented wave (PAW) potentials [Ref.S4]. All calculations are performed using a plane-wave basis with a 280 eV energy cutoff. The van der Waals (vdW) interaction is also included following Grimme’s D2 method [Ref.S5]. The convergence criterion for self-consistent field (SCF) loop is $1 \times 10^{-6}$ eV. The building block to construct the supercell is the rectangular unit cell of MoS$_2$ which contains two primitive unit cells. The monolayer supercell contains 72 unit cells with a total of 216 atoms. The Au adatom (or molecule) is then placed onto different positions of the monolayer. A 50 Å vacuum slab is
added along the direction normal to the monolayer to ensure the separation of the system with its periodic boundary image and also the accuracy of vdW corrections. The Brillouin zone is sampled at the Γ point. Atomic positions are relaxed until the maximum Hellmann-Feynman force on any atom was below 0.01 eV/Å.

After relaxation, the Au adatoms in different positions stay close to the MoS₂ monolayer (all about 2 Å away from the monolayer). Their relative stability is further summarized in Figure 5c. For Au adatoms on perfect monolayer MoS₂, their stability can be directly compared from their total energies. The S top position (indicated by “3” in Figure 5c) is the most energetically favorable site of perfect monolayer MoS₂. For Au adatom in S vacancy, we further calculate the total energy of the system where the Au atom is displaced 20 Å away from the monolayer (still with a S vacancy). The energy difference between that of the original system (Au in S vacancy) quantitatively reflects the stability of Au in S vacancy. Similar calculations are done for Au on a perfect MoS₂ monolayer. In this way, we can compare Au on perfect MoS₂ and Au in S vacancy even though the numbers of S atoms are different.

References

Figure S1. AFM images (top) with corresponding reference images indicating the positions at which line scans were taken to determine NW diameters for the histograms shown in Figure 3. The samples are treated with (a) 1 min deposition of 30 wt% AuCl₃ solution, (b) 1 min deposition of 3 wt% AuCl₃ solution, (c) 1 min deposition of 1 wt% AuCl₃ solution, (d) 1 min deposition of 0.5 wt% AuCl₃ solution, (e) 10 min deposition of 0.5 wt% AuCl₃ solution, and (f) 60 min deposition of 0.5 wt% AuCl₃ solution. After the deposition of AuCl₃ solution, all samples are blown with dry N₂ gas to remove extra solution followed by annealing at 150 °C for 2 min.
Figure S2. AFM images before/after annealing the AuCl$_3$ treated MoS$_2$. In the top image no texture was observed on the MoS$_2$ flake (9.7 nm thickness) after 3 wt% AuCl$_3$ deposition for 1 min followed by N$_2$ blow dry of the solution. As shown in the bottom image however, after annealing the sample at 150 °C for 5 min, Au nanowires formed on the surface.

Figure S3. AFM images for MoS$_2$ flakes after treatments of 3 wt% AuCl$_3$ for 1 min followed by N$_2$ blow dry and annealing at (a) 70 °C for 2 min and (b) 100 °C for 2 min. No nanowires were generated and tiny Au dots are observed on the MoS$_2$ in both cases. The flake thicknesses are shown in parentheses above each image.
Figure S4. AFM images for MoS$_2$ flakes after treatments of 3 wt% AuCl$_3$ for 1 min followed by N$_2$ blow dry and annealing at (a) 130 °C for 10 sec, (b) 130 °C for 30 sec, (c) 130 °C for 2 min, (d) 130 °C for 5 min, (e) 130 °C for 20 min, and (f) 130 °C for 60 min. The flake thicknesses are shown in parentheses above each image. Transformation of nanowires into nanoparticles is observed in Figure S4f.
Figure S5. AFM images for MoS\(_2\) flakes after treatments of 3 wt% AuCl\(_3\) for 1 min followed by N\(_2\) blow dry and annealing at (a) 150 °C for 10 sec, (b) 150 °C for 2 min, (c) 150 °C for 10 min, (d) 150 °C for 20 min, (e) 150 °C for 60 min, (f) 175 °C for 2 min, (g) 175 °C for 5 min, and (h) 175 °C for 10 min. The flake thicknesses are shown in parentheses above each image. Transformation of nanowires into nanoparticles starts after 5 min annealing at 175 °C as shown in Figure S5g and completely changes after 10 min as shown in Figure S5h.
Figure S6. AFM images for MoS\textsubscript{2} flakes after treatments of 3 wt\% AuCl\textsubscript{3} for 1 min followed by N\textsubscript{2} blow dry and annealing at (a) 200 °C for 10 sec, (b) 200 °C for 30 sec, (c) 200 °C for 2 min, and (d) 250 °C for 2 min. The flake thicknesses are shown in parentheses above each image. Transformation of nanowires into nanoparticles starts after 30 sec annealing at 200 °C as shown in Figure S6b.
Figure S7. Au structure from e-beam evaporation of Au (2 nm) on MoS₂. The structure was obtained after annealing in forming gas (5% H₂/95% N₂) at 400°C for 2 min. (a) The AFM image of the evaporated Au on MoS₂ flakes after the annealing. Hexagonal platelet structures were observed. (b) A line profile on the dotted line in Figure S7a. A flat surface was confirmed. (c) XRD pattern of the evaporated Au structure on the MoS₂.
Figure S8. XRD pattern for a AuCl$_3$ treated MoS$_2$ sample (blue curve). Simulated patterns for MoS$_2$ (green curve) and Au (orange) are also shown. A broad peak around 20 degrees shown in the treated MoS$_2$ is from carbon tape underneath. Diffraction patterns are from Cif file of MoS$_2$ (9007660) is from Crystallography Open Database and Au (9008463) is from Crystallography Open Database.
Figure S9. Preferential alignments of Au nanoparticles on a MoS$_2$ surface is shown by TEM selected area electron diffraction (SAED) patterns. TEM images and diffraction patterns are taken from a free-standing MoS$_2$ flake after treatment with 0.5 wt% AuCl$_3$ solution. (a) The SAED pattern from a reference region of bare MoS$_2$ is shown with several of the principle planes labeled, taken from the circular region shown in the TEM image at the top right. The corresponding MoS$_2$ crystal structure for a single layer of the hexagonal P 63/mmc 2H polytype is depicted top left with the same three planes highlighted. (b) The SAED pattern for a region containing a single Au nanoparticle and the corresponding TEM image of this region are shown at top left and right respectively. The Au diffraction peaks are highlighted by subtracting the SAED pattern of bare MoS$_2$. In this case an Au peak with a d-spacing of 1.4 Å consistent with Au\{220\} planes ($d = 1.44$ Å) is shown aligned to the MoS$_2$ (\overline{1}10) peak. These two distinct peaks appearing in-line indicates that they are coplanar but with separate d-spacings consistent with a preferential alignment between these planes. (c) An SAED pattern (top left) taken from a region with multiple Au particles (top right) is shown with the Au peaks highlighted by again subtracting the SAED pattern from bare MoS$_2$. Multiple aligned Au and MoS$_2$ planes are present and labeled: Au\{311\}|\parallel MoS$_2$(\overline{2}10), Au\{111\}|\parallel MoS$_2$(100), and Au\{200\}|\parallel MoS$_2$(310).
Figure S10. Electronic property of the MoS$_2$ flake after 1 wt% AuCl$_3$ deposition for 5 min followed by N$_2$ blowdry and 150 °C annealing for 2 min. (a) An optical microscope image of the device. The thickness of the flake is 90 nm. (b) Transfer characteristics of the pristine device (before treatment) and after treatment of the AuCl$_3$ solution at drain voltages of 0.05 V (dark green) and 1 V (light green) for the pristine and -0.05 V (orange) and -1 V (red) for the AuCl$_3$ treated. (c) Output characteristics for the AuCl$_3$ treated sample. The gate voltage ($V_{GS}$) is varied from -80 V to 80 V with 10 V increments.
Figure S11. Control experiment to confirm HCl effect for transfer characteristics of MoS$_2$.

The device is similar to the one shown in Figure S10. Transfer characteristics of the pristine MoS$_2$ device (green), after deposition of 1.2 M HCl for 1 min (blue), and after annealing the HCl treated device at 150 °C for 2 min (red). Drain voltages are 1 V in all cases. The current level is increased after annealing at 150 °C due to improvements in the metal contacts. All of them show $n$-type behavior.