Nanometer-thick copper films with low resistivity grown on 2D material surfaces

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Thin Copper (Cu) films (15 nm) are deposited on different 2D material surfaces through e-beam deposition. With the assist of van der Waals epitaxy growth mode on 2D material surfaces, preferential planar growth is observed for Cu films on both MoS2 and WSe2 surfaces at room temperature, which will induce a polycrystalline and continuous Cu film formation. Relative low resistivity values 6.07 (MoS2) and 6.66 (WSe2) μΩ·cm are observed for the thin Cu films. At higher growth temperature 200 °C, Cu diffusion into the MoS2 layers is observed while the non-sulfur 2D material WSe2 can prevent Cu diffusion at the same growth temperature. By further increasing the deposition rates, a record-low resistivity value 4.62 μΩ·cm for thin Cu films is observed for the sample grown on the WSe2 surface. The low resistivity values and the continuous Cu films suggest a good wettability of Cu films on 2D material surfaces. The thin body nature, the capability to prevent Cu diffusion and the unique van der Waals epitaxy growth mode of 2D materials will make non-sulfur 2D materials such as WSe2 a promising candidate to replace the liner/barrier stack in interconnects with reducing linewidths.

In silicon (Si) industry, copper (Cu) has been widely used for the application of interconnects due to its low resistivity1–3. On the other hand, the high diffusivity of Cu will result in Cu atom diffusion into the dielectric layers such that short circuit and therefore, system failure will be observed for Si chips4–7. Therefore, to avoid the problem of Cu diffusion, diffusion barrier layers between Cu wires and dielectrics are required for Cu interconnects. In nowadays, thin TaN layers (~ 3 nm) are commonly adopted as the diffusion barrier layer to isolate the Cu wires (~ 20 nm in widths/heights) from the surrounding dielectrics7,8. For a better adhesion between Cu and the TaN diffusion barrier layer, an additional liner layer Ta (~ 1 nm) is also required9. In this case, the Cu/Ta/TaN stack has become the standard interconnect structure in modern semiconductor industries. However, with the further shrinkage in the device line widths below 3 nm, interconnect scaling down to sub-20 nm will be required in the future. With the reduction of Cu wire thicknesses, its resistivity will increase remarkably due to the electron scattering by phonons, point defects, impurities and grain boundaries8–14. To avoid significant resistivity increasing with reduced Cu wire thickness, an alternate choice would be the thinner barrier/liner layers. However, when the TaN film thickness is reduced to below 3 nm, the barrier layer is no longer able to prevent the Cu diffusion to the surrounding dielectrics14. In this case, a new material with the capability of blocking Cu diffusion in a few nanometer thicknesses will become an urgent need for further interconnect scaling. Under this scenario, since mono-layer 2D materials are of few angstroms in thicknesses, they will become a promising candidate for this application. Besides the thin body nature of 2D materials, in previous publications, we have demonstrated vertical 2D material hetero-structures with epitaxially grown elemental 2D materials such as antimony (Sb), germanium (Ge) and tin (Sn) grown on molybdenum disulfide (MoS2) surfaces15,16. The results have revealed that the epi-layer formation through the van der Waals epitaxy growth mode on 2D material surfaces has less dependent to the substrate lattice structures. By further expanding this concept to metal crystals, van der Waals epitaxy on 2D material surfaces will help to improve the crystallinity of thick gold (Au) films grown on MoS2 surfaces17. The same mechanism will also help to improve the continuity of the nanometer-thick Au film such that close-to-theory sheet resistance will be obtained for 10 nm Au film grown on MoS2 surfaces18. The continuous thin Au films grown on MoS2 surfaces have also demonstrated good adhesion and wettability at the metal/2D material interfaces. Therefore, by using the thin 2D materials to replace the Ta/TaN stack, further interconnect scaling may be achieved. The unique van der Waals epitaxy growth mode of 2D material surfaces

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In this paper, we have deposited 15 nm Cu films on MoS$_2$ and WSe$_2$ surfaces by using the e-beam deposition system. With the assist of van der Waals epitaxy growth mode, polycrystalline Cu films are obtained on 2D material surfaces at room temperature (RT). By using the non-sulfur 2D material such as WSe$_2$, Cu diffusion can be avoided at higher growth temperatures. By further increasing the deposition rates, a record-low resistivity value 4.62 $\mu$Ω-cm for thin Cu films is observed for the sample grown on the WSe$_2$ surface. The thin thicknesses down to few atomic layers and good wettability of WSe$_2$ to Cu have made it a promising candidate to replace the liner/barrier stack in interconnects with reducing linewidths.

**Results and discussions**

**Thin copper films grown on different substrates.** To investigate the influence of van der Waals epitaxy to the thin Cu films, 15 nm Cu were deposited on 300 nm SiO$_2$/Si and tri-layer MoS$_2$/c-plane sapphire substrates at RT by using an e-beam deposition system with 0.1 Å/s deposition rate. The 500 × 500 nm$^2$ atomic force microscopy (AFM) images of the two samples are shown in Fig. 1a. As shown in the figure, Cu clusters with deep trenches are observed for the sample grown on the SiO$_2$/Si substrate, which indicates that the dangling bonds on SiO$_2$ surfaces may hinder the Cu atom migration and therefore, island formation instead of planar film growth will be observed on SiO$_2$ surfaces. Different from the Cu film grown on SiO$_2$ surfaces, a more continuous Cu film is observed on the MoS$_2$ surface. Although Cu clusters are still observed for the sample, the shallower trenches between the clusters suggest that the van der Waals epitaxy growth mode on 2D material surfaces at room temperature (RT) will help the planar growth of thin metal films. The root-mean-square (RMS) roughness values of the Cu films deposited on SiO$_2$ and MoS$_2$ surfaces are 1.67 and 1.04 nm, respectively, which is consistent with the observation that deeper trenches are observed for the Cu film deposited on the SiO$_2$ surface. As a result, compared with the resistivity value 19.71 $\mu$Ω-cm for the sample grown on the SiO$_2$/Si substrate, a much lower resistivity 6.07 $\mu$Ω-cm will be observed for the sample grown on the MoS$_2$ surface, which is close to ~ 5 $\mu$Ω-cm resistivity obtained from 15 nm Cu film grown on mechanically exfoliated MoS$_2$ surfaces$^{19}$. The 2θ − θ curves of the two samples measured by the X-ray diffraction system (XRD) are shown in Fig. 1b. Except for the substrate peaks, an additional Cu (111) peak is observed only for the Cu film grown on the MoS$_2$ surface, which suggests that besides planar film growth, a better crystallinity is also obtained for the Cu film grown on MoS$_2$ surfaces. The observation of Cu (111) peak alone may suggest that most of the Cu domains grown on MoS$_2$ surfaces are well-aligned with (111) direction. Further investigation is required in the future to verify this point. On the other hand, it is expected that different orientations of the Cu domains are obtained on the SiO$_2$ surface. Therefore, compared with the well-aligned (111) direction for the Cu domains on the MoS$_2$ surface, the weakened XRD peaks of the Cu domains are not observed for the Cu film deposited on the SiO$_2$ surface. The improved crystallinity of the Cu films grown on 2D material surfaces may help to reduce the inelastic electron scatterings on the Cu grain boundaries.

**Copper films grown on MoS$_2$ surfaces at different temperatures.** To further investigate the influence of growth temperatures to the morphologies and crystallinity of the thin Cu films, two more samples with 15 nm Cu deposited on MoS$_2$ surfaces at 100 and 200 °C are prepared. The 1 × 1 µm$^2$ AFM images of the sample prepared at RT is also shown in the figure. With increasing growth temperatures, significant Cu grain coalescence is observed. Flattened plateau with deeper trenches in between is also observed at higher growth temperatures. The phenomenon of Cu grain coalescence will also result in an increasing RMS roughness values from 1.04, 1.44 to 2.23 nm for the Cu films grown at RT, 100 and 200 °C, respectively. The results may indicate improved crystallinity of the thin Cu films with increasing growth temperatures. The normalized Cu (111) XRD peaks of the three samples are shown in Fig. 2b. As shown in the figure, the full widths at the half maximum (FWHM) of the Cu (111) XRD peak decreases from 0.69, 0.64 to 0.61 degree for the three samples prepared at RT, 100 °C and 200 °C, respectively, which suggests that improved crystallinity/larger Cu grains are obtained with increasing growth temperatures. The results are consistent with the observation of enlarged Cu grains with increasing growth temperatures from the AFM images. However, although improved crystallinity is obtained at higher growth temperatures, the resistivity values of the thin Cu films will increase from 6.07 (RT), 8.16 (100 °C) to 40.2 (200 °C) $\mu$Ω-cm with increas-

![Figure 1](https://www.nature.com/scientificreports/)
ing growth temperatures. The less continuous Cu films resulted from the Cu grain coalescence with increasing growth temperatures may be the main mechanism responsible for this phenomenon. Since the role of MoS₂ is to replace the Ta/TaN stack in interconnects, its effect to block the Cu diffusion is a critical issue for this purpose. The cross-sectional high-resolution transmission electron microscope (HRTEM) images of the three samples are shown in Fig. 2c. As shown in the figure, clear tri-layer MoS₂ with polycrystalline Cu films are observed for the two samples prepared at RT and 100 °C, respectively. However, for the sample prepared at 200 °C, the Cu/MoS₂ interfaces are less clear. Some MoS₂ layers seem to be lifted away from the sapphire substrates, which may indicate Cu diffusion into the MoS₂ films at 200 °C. To investigate this phenomenon, the high-angle annular dark field (HAADF) mappings of Cu, S and Mo elements for the sample grown at 200 °C are shown in Fig. 2d. The observation of Cu signals in the MoS₂ layer suggests that Cu atoms will diffuse into MoS₂ at 200 °C, which is also observed for the sample grown at 100 °C. The HAADF mappings of Cu, S and Mo elements for the sample grown at 100 °C are shown in the supplementary information Fig. S1. As shown in Fig. 2c, the diffused Cu atoms may lift some MoS₂ layers away from the sapphire substrates. In this case, both Mo and S signals will also be observed in the Cu layer near the Cu/MoS₂ interface.

Copper films grown on WSe₂ surfaces. Since Cu atoms will easily react with sulfur, it is possible that the defects with Mo vacancies will induce Cu diffusion into the MoS₂ layers. The element sulfur is also a potential contaminant for semiconductor fabrication lines. In this case, a non-sulfur 2D material should be adopted for the application of liner/barrier layer in interconnects. To demonstrate this point, wafer-scale tri-layer tungsten diselenide (WSe₂) films are grown on sapphire substrates by selenizing pre-deposited tungsten films at 950 °C. The picture and the cross-sectional HRTEM image of the standalone WSe₂ sample showing its uniformity are shown in the supporting information (Fig. S2). By using the WSe₂/sapphire samples as the new substrates, a 15 nm Cu film is deposited on the WSe₂ surface at RT. The 2θ–θ curve of the sample measured by XRD is shown in Fig. 3a. The X-ray curve of the Cu film grown on the MoS₂ films at RT is also shown in the figure for comparison. Besides the substrate peak, the Cu (111) peak is observed on both MoS₂ and WSe₂ samples. The results indicate that compared with the Cu film grown on the SiO₂ surface (Fig. 1), better crystallinity is obtained for the thin Cu films grown on both MoS₂ and WSe₂ surfaces. The same van der Waals epitaxy of the thin Cu film grown on the MoS₂ surface may also take place on the WSe₂ surface. The 1 × 1 μm² AFM image of the sample grown on the WSe₂ surface is shown in Fig. 3b. Compared with the sample grown on the MoS₂ surface (Fig. 2a), a similar morphology is observed for the sample grown on the WSe₂ surface, which indicates that the unique van der Waals epitaxy growth mode will take place on different 2D material surfaces. The preferential planar film growth instead of island formation will improve the continuity of the thin Cu films deposited on both MoS₂ and
WSe₂ surfaces. In this case, a similar resistivity value 6.66 μΩ-cm with the sample grown the MoS₂ surface (6.07 μΩ-cm) will be observed for the 15 nm Cu film grown on the WSe₂ surface.

To investigate the influence of growth temperatures to the Cu diffusion in the WSe₂ samples, two more samples with 15 nm Cu deposited on WSe₂ surfaces at 100 and 200 °C are prepared by using the e-beam deposition system with 0.1 Å/s deposition rate. Similar with the samples grown on MoS₂ surfaces, Cu grain coalescence and decreasing FWHMs of the Cu (111) XRD peak are observed for the WSe₂ samples with increasing growth temperatures (Fig. S3 in the supporting information). The results confirm again that the same van der Waals epitaxy occurs on both MoS₂ and WSe₂ surfaces. The cross-sectional HRTEM images of 15 nm Cu films grown on WSe₂ surfaces at RT, 100 and 200 °C are shown in Fig. 4a. Unlike partial MoS₂ liftoff from the sapphire substrates for the sample grown on the MoS₂ surface at 200 °C, polycrystalline Cu films with clear tri-layer WSe₂ are observed for all the three samples. The results suggest that WSe₂ layers may effectively prevent Cu diffusion. The HAADF mappings of Cu and W elements for the sample grown at 200 °C are shown in Fig. 4b. A clear separation of Cu and Se signals are observed in the figure. The results indicate that by using non-sulfur 2D materials as the barrier layer, Cu diffusion can be effectively avoided. Compared with the ~ 4 nm thick Ta/TaN stacks, the thickness of tri-layer WSe₂ is around 2.0 nm. Further thickness reduction is possible by reducing the layer numbers of the WSe₂ films. The thinner line/barrier stacks of interconnects will leave a large volume for Cu films such that

Figure 3. (a) The 2θ–θ curve measured by XRD and (b) the 1 × 1 μm² AFM image of the 15 nm Cu grown on the WSe₂ surface. The X-ray curve of the Cu film grown on the MoS₂ surface at RT is also shown in (a) for comparison.

Figure 4. (a) The cross-sectional HRTEM images of 15 nm Cu deposited on tri-layer WSe₂/sapphire substrates at RT, 100 and 200 °C. (b) The HAADF mappings of Cu and Se elements for the sample grown at 200 °C. The white lines on the figure depict the actual WSe₂/sapphire and Cu/WSe₂ interfaces.
further size reduction is possible for the interconnects. The relatively low resistivity values 6.07 and 6.66 μΩ-cm and the continuous Cu films observed on MoS₂ and WSe₂ surfaces at RT suggest a good wettability of Cu films on 2D material surfaces. The thin body nature, the capability to prevent Cu diffusion and the unique van der Waals epitaxy growth mode of 2D materials will make WSe₂ a promising candidate to replace the Ta/TaN stack in interconnects with reducing linewidths.

Although the van der Waals epitaxy growth mode on 2D material surfaces is advantageous for planar growth of metal films. The polycrystalline nature of grown 2D materials with wafer scale may hinder the adatom migration and induce local island formation on the 2D material surfaces. In this case, compared with exfoliated 2D material flakes, higher resistivity values will be observed on grown 2D material surfaces with the same metal film thicknesses. As discussed in the previous section, although a resistivity value 6.07 μΩ-cm can be achieved for the 15 nm Cu film grown on the MoS₂ surface at RT, a lower resistivity value ~ 5 μΩ-cm is still observed on exfoliated MoS₂ flakes with the same Cu thickness. As we have observed for the samples with different growth temperatures, the Cu grain coalescence will improve the crystallinity of the film. The same mechanism will also induce a less continuous film such that higher resistivity values will be observed. Therefore, one possible approach to solve this problem is to increase the adatom density on the substrate surfaces such that a more continuous film can be obtained. 15 nm Cu films with deposition rates 0.5 and 1.0 Å/s are deposited on WSe₂ surfaces by using the e-beam deposition system. The resistivity values obtained for the two samples are 5.22 and 4.62 μΩ-cm, respectively. The resistivity values of Cu films reported in previous publications are shown in Fig. 5. The resistivity value 1.68 μΩ-cm of Cu in nature is also shown in the figure. As shown in the figure, the resistivity value 4.62 μΩ-cm is the lowest value reported in literature. Given the fact that polycrystalline instead of single-crystal films are obtained for wafer-scale 2D materials at this stage, it is possible to achieve further reduction in the resistivity value for the thin Cu films if improved crystallinity can be achieved for grown 2D materials in the future. Nevertheless, the recorded low resistivity value of the thin Cu film grown on the WSe₂ surface has already demonstrated its potential to replace the Ta/TaN stack in interconnects. Besides the direct growth of 2D materials on different substrates, the film transferring can be an easy approach to facilitate the integration of 2D materials with other substrates. After transferring the tri-layer WSe₂ to a 300 nm SiO₂/Si substrate, 15 nm Cu is deposited on the WSe₂ surface by using the e-beam deposition system with the deposition rate 1.0 Å/s at RT. Compared with the value 4.62 μΩ-cm obtained for the sample grown on the un-transferred WSe₂ surface, a similar resistivity value 5.43 μΩ-cm is observed for this sample. Despite the contamination and damages introduced during the transferring procedure, the results have demonstrated that van der Waals epitaxy is still the main mechanism of epi-layer growth on 2D material surfaces.

**Conclusion**

We have demonstrated 15 nm Cu film deposited on different 2D material surfaces. With the assist of van der Waals epitaxy growth mode on 2D material surfaces, preferential planar growth is observed for Cu films on both MoS₂ and WSe₂ surfaces at room temperature, which will induce a polycrystalline and continuous Cu film formation. At higher growth temperature 200 °C, Cu diffusion into the MoS₂ layers is observed while the non-sulfur 2D material WSe₂ can prevent Cu diffusion at the same growth temperature. By further increasing the deposition rates, a record-low resistivity value 4.62 μΩ-cm for thin Cu films is observed for the sample grown on the WSe₂ surface. After transferring the tri-layer WSe₂ to a 300 nm SiO₂/Si substrate, a similar resistivity value 5.43 μΩ-cm is obtained with 15 nm Cu deposited on the WSe₂ surface. The low resistivity values of thin Cu films grown on transferred or un-transferred WSe₂ surfaces have provided varieties for the 2D material integration with other materials in practical applications. These results have demonstrated that non-sulfur 2D materials such as WSe₂ can be a promising candidate to replace the liner/barrier stack in interconnects with reducing linewidths.
Methods
For the growth of tri-layer MoS$_2$, the Mo metal was deposited on the sapphire substrates by using a radiofrequency (RF) sputtering system. We used 30 W for constant sputtering power and 5 × 10$^{-3}$ Torr with a 30 sccm gas flow of argon (Ar) for background pressure. The deposition time is 37 s for the preparations of MoS$_2$ layer. After the metal deposition, the sample was placed at the center of the hot furnace for sulfurization. 200 sccm Ar gas was used as carrier gas while the pressure was kept at 50 Torr. The sulfurization temperature was kept at 850 °C with 0.25 g of sulfur (S) powder for 20 min. For the growth of tri-layer WSe$_2$, a similar transition metal deposition and selenization procedure is adopted. The W metal film was deposited on the sapphire substrate by using the same RF sputtering system. Similar deposition conditions as the Mo deposition are adopted for the W deposition except for the higher sputtering power 40 W. The deposition time is 25 s for the W deposition. After the metal deposition, the sample was placed at the center of the hot furnace for selenization. 85 sccm Ar gas was used as carrier gas while the pressure was kept at 100 Torr. The selenization temperature was kept at 950 °C with 0.2 g of Se powder for 30 min. The deposition rate and the thickness of the Cu films are measured by the quartz crystal microbalance (QCM) which is equipped with the e-beam system. The resistivity values were obtained through the equation $\rho = R_s \times$ (film thickness), where $R_s$ is the sheet resistance obtained through four-point measurements. For the four-point measurements, a four-pin probe station KSR-4 with BeCu probe tip and tip pitch 1 mm is adopted. Keithley 2400 SourceMeter is used to measure the sheet resistance of the samples. The cross-sectional HRTEM images measurement are obtained by using a JEOL JEM-2800F transmission electron microscope.

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Author contributions
Y.-W.L., D.-J.Z., P.-C.T. and C.-T.C. did the material growth/characterizations and electrical measurements of the samples. Y.-W.L., W.-C.T., and S.-Y.L. clarified the experimental data and came out with the manuscript. S.-Y.L. is the group leader and conducted the investigation of this work.

Competing interests
The authors declare no competing interests.

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