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Supporting Information

Passivation of Layered Gallium Telluride by Double Encapsulation with Graphene

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1. Images of GaTe flakes supported on different substrates

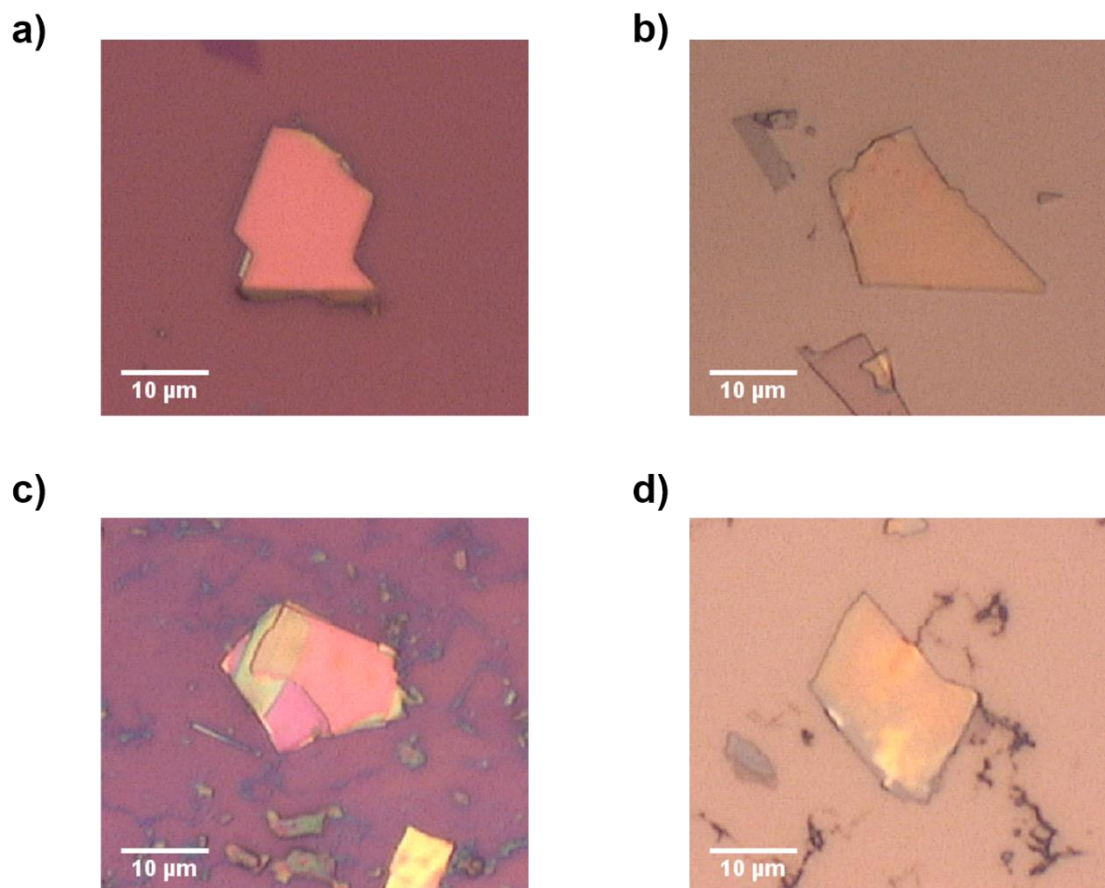


Figure S1. Optical images of GaTe flakes supported a) SiO_2/Si , b) $\text{Al}/\text{SiO}_2/\text{Si}$, c) graphene covered SiO_2/Si and d) graphene covered $\text{Al}/\text{SiO}_2/\text{Si}$ substrates.

2. SiO₂/Si and Al/SiO₂/Si substrate roughness

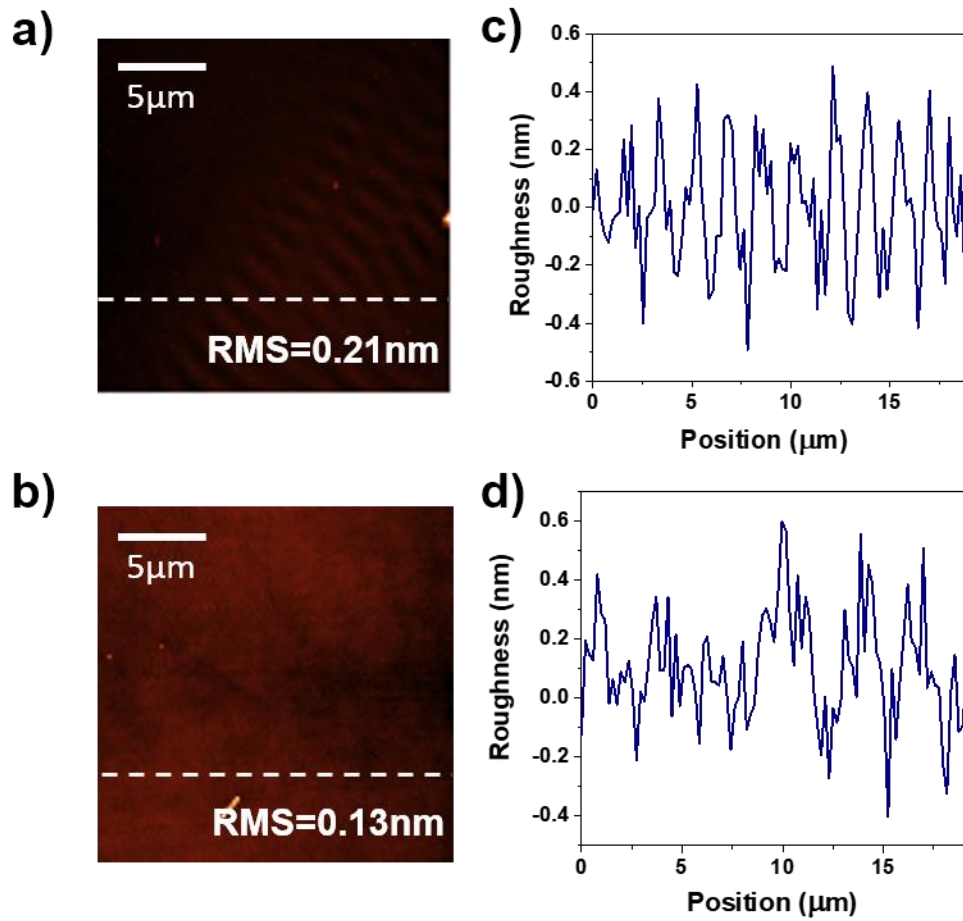


Figure S2. Roughness of SiO₂/Si and Al-coated SiO₂/Si substrates. AFM topography scans of a) SiO₂/Si and b) Al/SiO₂/Si. White dashed lines correspond to roughness profiles for SiO₂/Si and Al/SiO₂/Si shown in c) and d) respectively.

3. Comparison of GaTe degradation rate on CVD monolayer graphene substrates and mechanically exfoliated 7nm, 22nm graphene flakes

To confirm that the addition of a graphene buffer layer between GaTe/SiO₂ interfaces slowed degradation of GaTe we transferred 7nm and 22nm mechanically exfoliated graphene to SiO₂. GaTe flakes (~175nm thickness, 20μm by 20μm) were transferred on top. While the wet-transferred, CVD monolayer graphene used as a substrate in the main paper was convenient for generating uniform, large-area substrates, the use of mechanically exfoliated flakes allows replication of the bottom GaTe interface used for sandwiched samples (Figure 3-4). Two different graphene thicknesses were used to check for any potential thickness effects. It was not possible to reliably produce mechanically exfoliated graphene monolayers with a sufficiently large surface area to eliminate any contact between GaTe and SiO₂ for the purpose of comparison.

Figure S3a shows the evolution of the 115cm⁻¹ to 129cm⁻¹ peak intensity ratio over time for GaTe supported on wet-transferred monolayer graphene and 7nm, 22nm thick mechanically exfoliated graphene flakes. We observed no significant differences in the degradation rate for GaTe on 7nm versus 22nm graphene flakes, suggesting that the thickness of the graphene buffer layer is not important for this thickness range. However, GaTe degradation occurs more rapidly for flakes supported on wet-transferred graphene monolayers. There are several possible reasons for this. Despite the reported impermeability of pristine graphene layers^{1,2}, the introduction of structural defects allows permeation of gaseous species through the membranes. The presence of the defect-activated D peak (~1375cm⁻¹) in the wet-transferred monolayer Raman spectrum indicates the presence of structural disorder in the sample. The Raman spectra of the various graphene substrate layers studied here are shown together in figure S3b. There is no indication of defects in either of the mechanically exfoliated flakes.

This discrepancy between wet and dry-transferred samples could potentially lead to the increased degradation rate observed for GaTe supported on CVD monolayer graphene. Additionally, the presence of PMMA residues on the monolayer surface could potentially lead to further acceleration of the degradation process.

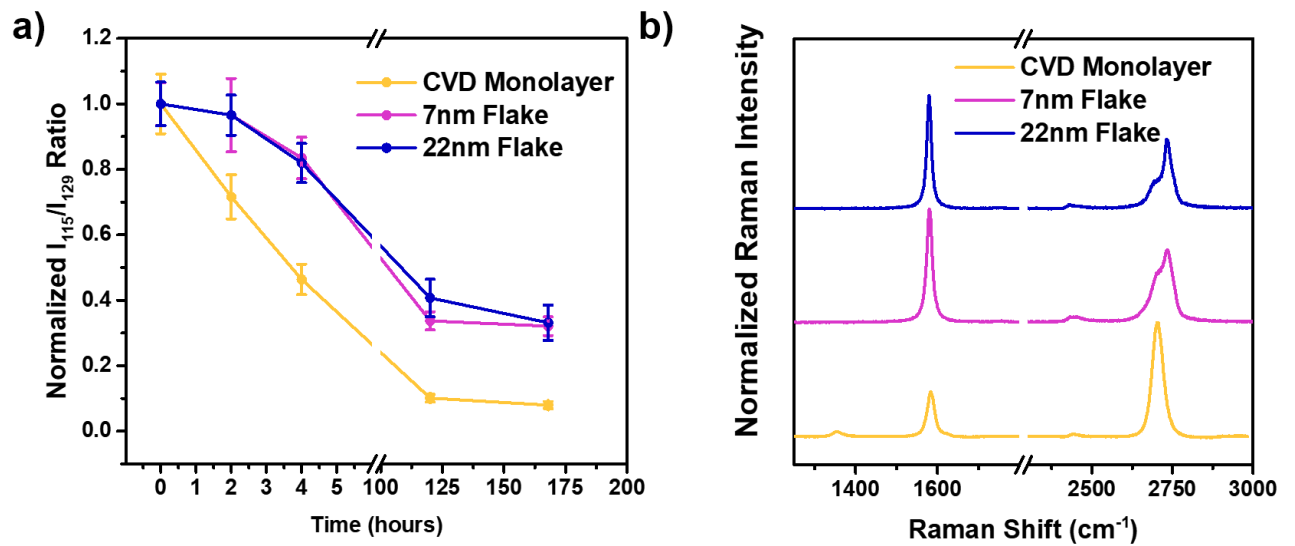


Figure S3. Comparison of degradation rate of GaTe on CVD wet-transferred graphene monolayers and 7nm, 22nm thick mechanically exfoliated graphene flakes. a) I_{115}/I_{129} Raman peak intensity ratio over time. b) Raman spectra of CVD monolayer graphene and mechanically exfoliated 7nm, 22nm graphene flakes.

4. Impact of PMMA residues on GaTe degradation rate

For CVD graphene transferred using the PMMA-mediated wet-transfer method it is very difficult to effectively remove PMMA residues from the surface.³⁻⁵ Therefore, we investigated whether the presence of residues on the bottom graphene surface could impact the degradation rate of GaTe flakes. Two CVD monolayer graphene samples were transferred to SiO₂/Si substrates as described in the Methods section of the manuscript. To remove the polymer support layer, one sample was left in acetone for 12 hours, the other for only 6 hours. This was intended to produce graphene substrates with low and high levels of PMMA residue respectively. Residue levels were confirmed using AFM (S4a, S4b). Bright spots show the locations of polymer residues. GaTe flakes measuring approximately 20 μ m by 20 μ m with a thickness of ~175nm were transferred on top of the graphene monolayers and the level of degradation in the samples monitored using Raman spectroscopy (S4c).

The sample with higher levels of PMMA surface residue showed an increased degradation rate compared with the cleaner graphene substrate and appeared fully degraded after 2 days. While PMMA residue on 2D material surfaces has previously been shown to significantly impact transport properties in devices,⁶⁻⁸ the impact of residues on the degradation rate of air unstable materials has not yet been studied. The origin of the effect observed here is therefore not known for certain. One possibility is that a combination of thermochemical and photochemical effects from laser exposure during measurements further breaks down the remaining residues to leave behind volatile, oxygen containing products that accelerate GaTe degradation.⁹ This effect has previously been successfully employed to clean graphene surfaces.¹⁰

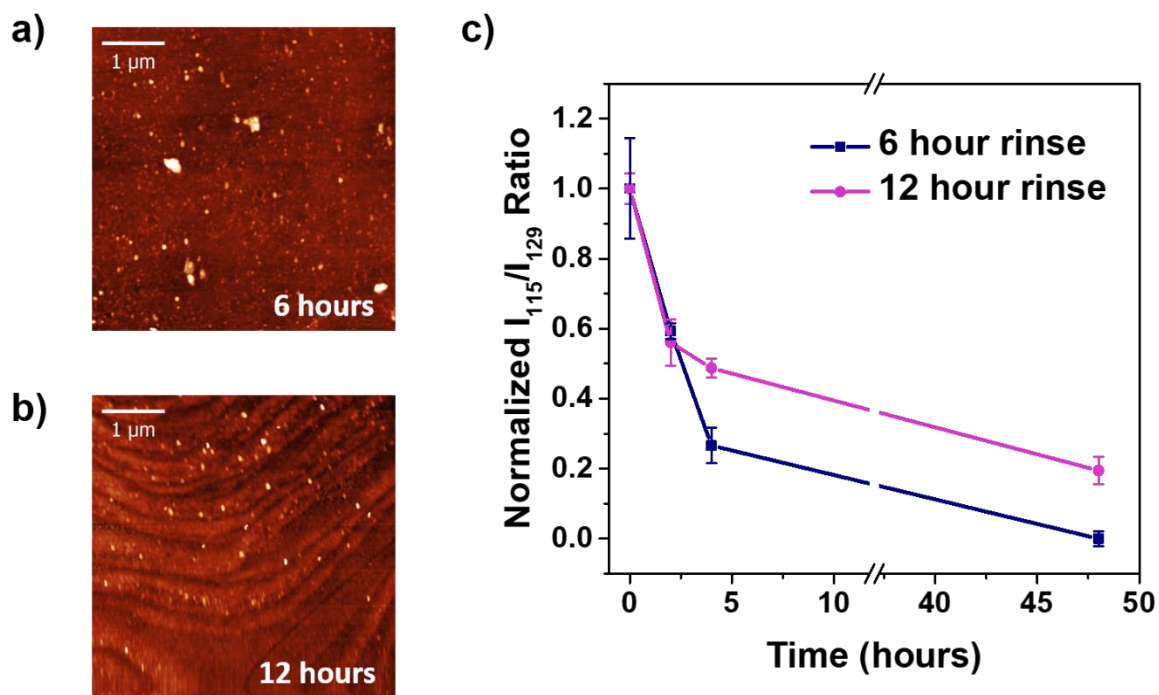


Figure S4. Influence of PMMA residue level on GaTe flake degradation. AFM topography scans of wet-transferred graphene monolayers rinsed for a) 6 hours and b) 12 hours in acetone. Scans show a high level and lower level of PMMA residue for a) and b) respectively. c) Evolution of Raman 115cm⁻¹ to 129cm⁻¹ peak ratios over time for GaTe supported on graphene with high (6 hours) and low (12 hours) levels of residue.

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