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Stacking of Two-Dimensional Materials to Large-Area Heterostructures by Wafer Bonding

Arne Quellmalz^{1*}, Simon Sawallich^{2,3}, Maximilian Precht⁴, Oliver Hartwig⁴, Siwei Luo⁴, Stefan Wagner⁵, Georg S. Duesberg⁴, Max C. Lemme⁵, Frank Niklaus¹, and Kristinn B. Gylfason¹

¹Micro and Nanosystems, KTH Royal Institute of Technology, Malvinas väg 10, 10044 Stockholm, Sweden; ²Protemics GmbH, Otto-Blumenthal-Str. 25, 52074 Aachen, Germany; ³Chair of Electronic Devices, RWTH Aachen University, Otto-Blumenthal-Str. 2, 52074 Aachen, Germany; ⁴Institute of Physics, Universität der Bundeswehr München, Werner-Heisenberg-Weg 39, 85577 Neubiberg, Germany; ⁵AMO GmbH, Advanced Microelectronic Center Aachen (AMICA), Otto-Blumenthal-Str. 25, 52074 Aachen, Germany; *E-mail: arne.quellmalz@eecs.kth.se

Abstract: The integration of 2D materials for photonic applications is not compatible with high-volume manufacturing. We report a generic methodology that uses only readily available semiconductor equipment and experimentally demonstrate the stacking of graphene and molybdenum disulfide (MoS₂). © 2020 The Authors(s)

1. Introduction

The outstanding properties of two-dimensional (2D) materials such as graphene and transition metal dichalcogenides enabled the demonstration of high-performance devices for photonic and optoelectronic applications [1], [2]. Vertical stacking of 2D materials forms van der Waals heterostructures that show synergetic effects between the stacked layers [3], [4]. Integrating these materials into conventional semiconductor and photonic manufacturing lines is critically important to exploit their material properties in commercial devices while leveraging established infrastructures with low-cost and high-volume manufacturing on large substrates. However, current integration approaches are not compatible with industrial processing on wafer-level. Here, we present a generic methodology for the large-area transfer and stacking of 2D materials to heterostructures by adhesive wafer bonding [5], [6].

2. Description of Methodology

Figure 1: Schematic illustration of the methodology for large-area transfer of 2D materials (a) and formation of 2D material heterostructures (a and b).

Our approach for transferring 2D materials from their growth substrate to a target wafer comprises four consecutive steps (Figure 1a): First, the target wafer is spin-coated with an adhesive polymer layer of thermosetting bisbenzocyclobutene (BCB). Next, the growth substrate is placed on top of the target wafer such that the 2D material faces the adhesive layer. Inside a commercial wafer bonder, heating temporarily decreases the viscosity of the adhesive layer while the bond chucks apply a uniform force to the wafer stack. Thus, the adhesive layer molds against the 2D material and forms a stable bond between the 2D material and the target wafer. Finally, etching or delamination removes the growth substrate, which leaves the 2D material transferred on the target wafer. Partially cross-linked BCB allows repeated molding. Hence, repeating the steps of (2) material placement, (3) wafer bonding, and (4) growth substrate removal, transfers a second 2D material to the target wafer and forms van der Waals heterostructures (Figure 1b). In this process, the interface between the first and second 2D material is not exposed to any adhesive. Partially cross-linked BCB is chemically stable and sustains conventional wafer processing such as photolithography, material deposition, and etching. This feature allows structuring of the 2D material and integration of metal contacts.

3. Results

We formed molybdenum disulfide (MoS₂)/graphene heterostructures by stacking a layer of MoS₂ onto a sheet of graphene in two consecutive transfers. The map of the graphene sheet resistance (R_{sh}) from Terahertz (THz) near-field spectroscopy (Figure 2a) indicated the presence of graphene in both the heterostructure and its surroundings [7]. R_{sh} was significantly reduced in the MoS₂/graphene heterostructure. Since a sole MoS₂ layer was not directly detectable in this measurement, we speculate that interactions between the layers caused this modification. Raman

and photoluminescence (PL) spectroscopy of the MoS₂/graphene heterostructure confirmed the presence and integrity of both materials (Figure 2b and 2c). Two consecutive transfers of monolayer (1-layer) CVD graphene to a 100 mm diameter silicon wafer formed double-layer (2-layer) graphene on a target wafer. The map of R_{sh} (resolution: 500 μ m) revealed a uniform coverage of graphene on the target wafer without major defects (Figure 2d). Raman spectroscopy yielded the correlation map of the G and 2D peak positions (Figure 2e). The full width at half maximum of the 2D peak (Γ_{2D}) indicated minor strain variations in the layers, which suggested potentially high charge carrier mobility in the graphene.

Figure 2: a) Sheet resistance (R_{sh}) of graphene in the MoS₂/graphene heterostructures (center) and monolayer graphene (surrounding). b) Averaged Raman spectrum of MoS₂ and graphene (shaded area: standard deviation) acquired in a 1 x 1 mm² region (white rectangle in a). E'_{2g} = 385.16 $\pm$ 0.35 cm⁻¹, A_{1g} = 408.06 $\pm$ 0.28 cm⁻¹. Full width at half maximum of the 2D peak (Γ_{2D}). c) Averaged photoluminescence (PL) spectrum of MoS₂ (same acquisition site) with A and B excitonic transitions. d) R_{sh} of 1-layer and 2-layer graphene transferred to a 100 mm silicon wafer. e) Correlation map of the Raman G and 2D peak positions in 1-layer graphene (triangles) and 2-layer graphene (circles).

4. Conclusion

We introduced a methodology for large-area transfer and stacking of 2D materials to heterostructures by adhesive wafer bonding. Our approach avoids manual handling of the 2D material and exclusively uses processes and materials that are readily available in most large-scale semiconductor manufacturing lines. Experimentally, we demonstrated the stacking of two monolayers of graphene to 2-layer graphene and the formation of MoS₂/graphene heterostructures by two consecutive transfers. Hence, we expect that our methodology has the potential to accelerate advancements in fundamental 2D material science and a wide range of application areas in optics and photonics.

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6. References

- [1] M. Romagnoli *et al.*, "Graphene-based integrated photonics for next-generation datacom and telecom," *Nature Reviews Materials*, vol. 3, no. 10, p. 392, Oct. 2018, doi: 10.1038/s41578-018-0040-9.
- [2] K. F. Mak and J. Shan, "Photonics and optoelectronics of 2D semiconductor transition metal dichalcogenides," *Nature Photonics*, vol. 10, no. 4, pp. 216–226, Apr. 2016, doi: 10.1038/nphoton.2015.282.
- [3] Y. Cao *et al.*, "Unconventional superconductivity in magic-angle graphene superlattices," *Nature*, Mar. 2018, doi: 10.1038/nature26160.
- [4] A. L. Sharpe *et al.*, "Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene," *Science*, vol. 365, no. 6453, pp. 605–608, Aug. 2019, doi: 10.1126/science.aaw3780.
- [5] A. Quellmalz *et al.*, "Wafer-Scale Transfer of Graphene by Adhesive Wafer Bonding," in *2019 IEEE 32nd International Conference on Micro Electro Mechanical Systems (MEMS)*, Jan. 2019, pp. 257–259, doi: 10.1109/MEMSYS.2019.8870682.
- [6] A. Quellmalz *et al.*, "Large-Scale Integration of 2D Material Heterostructures by Adhesive Bonding," presented at the 2020 IEEE 33rd International Conference on Micro Electro Mechanical Systems (MEMS), Jan. 2020.
- [7] M. Wächter, M. Nagel, and H. Kurz, "Tapered photoconductive terahertz field probe tip with subwavelength spatial resolution," *Appl. Phys. Lett.*, vol. 95, no. 4, p. 041112, Jul. 2009, doi: 10.1063/1.3189702.