LARGE-SCALE INTEGRATION OF 2D MATERIAL HETEROSTRUCTURES BY ADHESIVE BONDING

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ABSTRACT

We report the integration of graphene/hexagonal boron nitride (hBN) heterostructure devices on large-areas by adhesive wafer bonding, a method suitable for industrial mass-production. In this new approach, we stack graphene and hBN by two consecutive bond transfers whereby the graphene and its interface to hBN is not in contact with potentially contaminating polymers or adhesives at any time. To show the feasibility of our approach for back end of the line (BEOL) integration of two-dimensional (2D) material heterostructures on standard silicon substrates, we fabricated graphene/hBN devices with electrical bottom contacts using only established semiconductor manufacturing tools, processes and materials.

KEYWORDS

2D Material Heterostructures, Integration, Transfer, Wafer Bonding, Graphene, Hexagonal Boron Nitride

INTRODUCTION

The exceptional properties of 2D material heterostructures, such as high charge carrier mobility, strong light absorption and synergetic effects between stacked layers, promise applications in high-performance sensors, photonics, and superconducting devices [1]–[3].

Integrating heterostructures into conventional semiconductor fabrication lines is crucial to exploit their outstanding electrical, mechanical and optical properties in future commercial NEMS devices while benefitting from established infrastructure for high-volume manufacturing on large substrates at low cost [4]. State-of-the-art integration approaches rely on stacking of 2D materials by consecutive transfers from dedicated growth substrates to the target substrate. However, available large-scale transfer methods degrade the material properties by causing defects, wrinkles and strain. Additionally, they contaminate the interface between the 2D materials by residuals from polymeric carrier layers and require manual handling of thin polymer sheets with the 2D materials. Consequently, none of the current integration approaches is compatible with existing infrastructure for large-scale semiconductor manufacturing. In earlier work, we presented a method for dry transfer of chemical vapor deposited (CVD) graphene by adhesive wafer bonding [5]. In contrast, here we use adhesive wafer bonding with conventional semiconductor tools to (a) transfer hBN, a 2D material that is electrically insulating, (b) stack graphene and hBN into van der Waals heterostructures, and (c) integrate electrical bottom contacts to the transferred 2D materials which demonstrates the compatibility with BEOL integration on preprocessed wafers.

Figure 1: Integration of two-dimensional material heterostructures by adhesive bonding. a) Schematics of the final device structure. b) Process flow for manufacturing the device in a): (1) Spin coating of BCB on target Si wafer. (2) Pre-cure of adhesive layer and deposition of Au electrodes. (3) 1st transfer of hexagonal boron nitride (hBN) from Cu foil to the target wafer by adhesive wafer bonding. (4) Patterning of the of hBN layer to open contact areas on top of the gold electrodes. (5) 2nd transfer of graphene from Cu foil. (6) Patterning of hBN/graphene heterostructures to final devices.

Figure 2: SEM images of fabricated graphene/hBN devices. a) After transfer of hBN (1st layer transfer) which covers the entire substrate. Femtosecond-laser ablation formed openings in the hBN layer which expose the underlying gold electrodes. b) After transfer of graphene (2nd transfer). The graphene forms a heterostructure with the underlying hBN and is contacted to the gold electrodes in rectangular openings in the hBN layer. c) Final graphene/hBN device. Femtosecond-laser ablation electrically isolated devices (dashed lines) and formed graphene patches connecting the outer electrodes. 2-terminal measurements of 7 devices yielded an average electrical device resistance of $1261 \pm 185 \Omega$.

DESCRIPTION OF INTEGRATION APPROACH

We report the integration of devices consisting of graphene/hexagonal boron nitride (hBN) heterostructures on large areas by adhesive wafer bonding (Figure 1a). First, we spin-coated a $2.5 \mu\text{m}$ thick layer of bisbenzocyclobutene (BCB) on a 100 mm diameter silicon wafer (Figure 1b-1). Baking at 190°C for 30 min on a hot plate partially cross-linked the thermosetting BCB, which increased its chemical stability and hence became insoluble in solvents and stable during the following process steps. Thermal evaporation of Ti/Au (20 nm / 100 nm, respectively) and a standard lift-off process structured bottom electrodes on top of the BCB (Figure 1b-2). To transfer the first 2D material to the target wafer, we bonded a $2.5 \times 2.5 \text{ cm}^2$ sheet of multilayer CVD hBN on copper foil, using a commercial wafer bonder (Suss-SB8, hBN facing the BCB, 190°C , 20 min, bond force: 250 N) (Figure 1b-3). Next, we etched the copper in FeCl_3 solution, which uncovered the hBN, lying on top of the BCB and the metal electrodes. Ablation by a femtosecond-laser patterned openings in the transferred hBN while the electrodes underneath remained intact (Figure 1b-4 and 2a). Our choice of bond temperature and time cross-linked the BCB only partly, which allowed a second transfer of another 2D material by reusing the existing adhesive below the first 2D material. To form a graphene/hBN heterostructure, we bonded a 4-inch sheet of monolayer CVD graphene on copper foil to the target wafer (Suss-SB8, graphene facing the target wafer, 190°C , 20 min, bond force: 3 kN) (Figure 1b-5) and removed the copper growth substrate by etching in FeCl_3 (Figure 2b). Finally, we patterned the graphene/hBN heterostructure to patches by ablation with a femtosecond-laser (Figure 1b-6). In the final device, the graphene was placed on top of the hBN and electrically connected the electrodes underneath (Figure 1a and 2c).

EXPERIMENTAL RESULTS

To characterize the electrical properties and to ensure uniform coverage of the transferred graphene on the target wafer, we performed terahertz (THz) spectroscopy. This non-contact inspection method achieves microscopic

resolution of the graphene sheet resistance (R_{sh}) on wafer-scale by using a THz near-field detector to scan over the wafer surface [6]. The spatially resolved map of R_{sh} on a test wafer with transferred graphene revealed a uniform coverage of graphene on the entire wafer (Figure 3). Note, for reference we removed the transferred graphene on the left side of the wafer. In the marked region, R_{sh} of 1681 measurements averaged to $967 \pm 30 \Omega/\text{sq.}$, which is a typical value for CVD graphene, transferred by manual transfer methods [7].

Raman spectroscopy gives information on the quality of graphene, by analyzing the position and width of the characteristic G and 2D peaks in the Raman spectrum. For pristine graphene, which is neither strained nor doped, these peaks are located at $\omega_{\text{G}} = 1582 \text{ cm}^{-1}$ and $\omega_{2\text{D}} = 2670 \text{ cm}^{-1}$ [8]. In Figure 4, we plot the correlation map of the G and 2D peak positions, and the full-width-at-half-maximum of the 2D peak ($I_{2\text{D}}$). The peak locations indicate minor compressive strain and strong hole doping. A low

Figure 3. Spatially resolved map of the graphene sheet resistance (R_{sh}) extracted from noncontact THz near-field spectroscopy. The right side of the wafer indicates a uniform coverage of graphene without major defects. The left side functioned as a reference and was not covered by graphene. The sheet resistance of the graphene heterostructure averages to $967 \pm 30 \Omega/\text{sq.}$ ($N = 1681$).

Figure 4: Correlation map of the Raman G and 2D peak positions (ω_G and ω_{2D} , respectively). The colormap represents the full-width-at-half-maximum of the 2D peak (Γ_{2D}). The peak locations indicate minor compressive strain and strong hole doping. The low values of Γ_{2D} hint at potentially high charge carrier mobility, hence, good quality of the graphene.

average of Γ_{2D} ($28.7 \pm 4.0 \text{ cm}^{-1}$) indicated little variations in strain, which hints at potentially high charge carrier mobility in the graphene – a crucial property for high-performance devices.

The SEM image in Figure 2c shows the final graphene/hBN device. The patterned graphene patch electrically connected the bottom electrodes, while the hBN was placed below the graphene. Hence, the graphene in the active device area was not in contact with the BCB. 2-terminal measurements of 7 devices yielded an average electrical device resistance of $1261 \pm 185 \Omega$, which is a typical value and spread for these type of devices and dimensions.

CONCLUSION

In this paper, we reported the successful integration of graphene/hBN heterostructure devices on large areas by adhesive wafer bonding. We tuned the degree of cross-linking of a thermosetting BCB adhesive layer to increase its chemical stability, which allowed (a) the formation of bottom electrodes, and (b) the stacking of 2D materials into heterostructures by repeated layer transfers, reusing the same adhesive layer. Layer and device characterizations indicated high 2D material quality after transfer. The compatibility of our approach with established semiconductor manufacturing processes makes it suitable for back end of the line (BEOL) integration of 2D material heterostructures on standard silicon substrates.

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