Engineering 2D Diamond/Cu(111) Interfaces via Controlled Graphene Hydrogenation

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Copper/diamond composites are attractive for thermal management in high-power electronics due to diamond's exceptional thermal conductivity, yet their implementation is limited by poor Cu–diamond adhesion, high interfacial thermal resistance, and the lack of scalable near-net-shape processing. We demonstrate a chemically bonded Cu–diamond interface via controlled hydrogenation of graphene on Cu(111). Single- and bilayer graphene were grown by CVD on Cu(111) and rapidly cooled to reduce wrinkling. Hydrogenation (5–210 min) was carried out using a hot-filament-assisted process [1,2]. Multi-wavelength Raman spectroscopy combined with a quantitative sp^3/sp^2 model [3] shows that hydrogen uptake depends strongly on Cu surface state: oxidized Cu exhibits negligible functionalization, whereas clean Cu reaches $I_D/I_G \approx 2.6$ (λ = 488 nm) after 60 min. The evolution of I_D/I_G and D-band broadening indicates progressive defect generation, shrinking sp^2 domains, and saturation. Post-annealing (400 °C, 10 min, Ar) partially restores sp^2 character ($I_D/I_G < 1$) for samples with initial $I_D/I_G < 2.6$, while prolonged hydrogenation (210 min) lowers Raman intensity and yields broad features after annealing, consistent with extensive sp^3 bonding. UV Raman reveals a sharp diamond peak, directly evidencing 2D diamond stable up to at least 400 °C (Fig. 1a). Despite some sample inhomogeneity, the observed +33 cm^{-1} blue shift, opposite to the ≈ -30 cm^{-1} tensile softening expected for 2.35% biaxial strain ($\gamma \approx 1$), demonstrates that interfacial interactions dominate the phonon response. DFT predicts a thermodynamically stable chemisorbed interface ($\Delta E = -2.6$ eV per cell) (Fig. 1b). The 2D diamond layer retains a semiconducting band gap ($\approx$ 2.9 eV), while hybridization with Cu induces overall metallic behaviour at the interface. Projected DOS and differential charge density analyses confirm interfacial charge transfer and C–Cu bond formation (Fig. 1c). This hydrogenation-driven phase transformation offers a scalable route to chemically bridge Cu and diamond, expected to reduce thermal boundary resistance.

References

[1] F. Piazza et al., Carbon, 145 (2019) 10; Carbon 169 (2020) 129.
[2] T. Fournier et al., Carbon Trends 20 (2025) 100553.
[3] T. Fournier et al., Carbon 244 (2025) 120657.

Figures

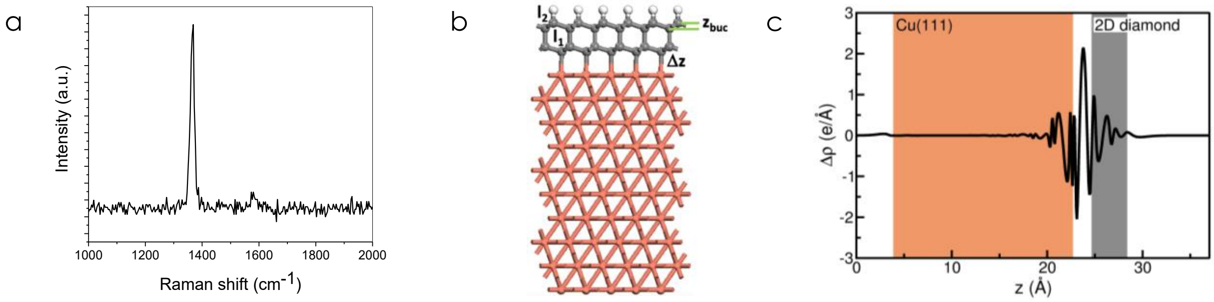

Figure 1: (a) 266 nm Raman spectrum confirming 2D diamond on Cu(111). (b) DFT-optimized 2D diamond/Cu(111) interface (C: grey; Cu: orange; H: white). (c) Differential charge density along the interface normal.