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Toward ultraclean 2D heterostructure interfaces: h-BN as a novel tool to visualize organic residues

Mechanical exfoliation of layered bulk materials, as well as 2D growth techniques provide a large number of 2D Van der Waals materials that can be prepared in monolayer and few-layer form. Restacking these materials by a variety of transfer methods results in 2D heterostructures, which typically feature a combination of graphene with semiconductors such as MoS_2 or black phosphorus, as well as insulators such as hexagonal boron nitride (h-BN).

Previously, 2D sample quality was often limited by the SiO_2 substrate, which is rough and contains a high density of charged impurities. This limitation can be overcome by using h-BN as a substrate: h-BN is ultraflat and free from charged impurities, typically resulting in dramatically enhanced sample quality, and this approach has enjoyed enormous popularity in recent years. Nonetheless, frequently extrinsic factors continue to limit the quality of 2D heterostructures, and one of these factors is the presence of organic processing residues, such as adhesives from mechanical exfoliation or residues from transfer and lithography steps, which can be left on sample surfaces and trapped at interfaces.

A number of approaches have been developed to reduce and remove such organic polymer residues, but many techniques suffer from drawbacks, and their applicability tends to be highly specific to the sample material. For example, acetone cleaning approaches most often lead to incomplete residue removal only; recipes involving heating the sample in vacuum or inert gas, instead of evaporating organic residues often spread the residues across the sample; and oxidative heating methods are incompatible with reactive 2D materials such as black phosphorus. Thus, development of new 2D material cleaning recipes that address these shortcomings is highly desirable.

A key obstacle in the development of new cleaning recipes is the difficulty in detecting organic residues, which complicates the evaluation of cleaning efficacy; organic polymer films of nanometer thickness are typically transparent and liquid-like, and common sample characterization methods such as optical microscopy, atomic force microscopy, or Raman spectroscopy are not suitable to detect their presence. To overcome this obstacle, we exploit the observation that thin organic polymer films deposited on h-BN lead to very bright photoluminescence [1], enabling us to visualize even minute amounts of organic residue. Our approach (schematically shown in Fig. 1) involves the preparation of exfoliated h-BN samples, which we deliberately contaminate with the target organic polymer of interest, such as adhesive residue. For a given candidate cleaning method, we then aquire photoluminescence maps before and after the cleaning step, enabling a clear assessment of the recipe's effectiveness.

Initial results indicate that the adhesive contained in a dicing tape widely used for mechanical exfoliation (Ultron Systems 1007R) is very difficult to remove fully. Figure 2 illustrates the photoluminescence characterization of a cleaning recipe involving heat treatment at 500 °C in partial oxygen atmosphere. The efficacy of cleaning treatments tested for this case increases as follows: acetone < hexane ≈ trichloroethylene < heating in partial oxygen atmosphere at 500 °C. Heating in partial hydrogen atmosphere at 300 °C results in spreading, not removal of this residue on h-BN samples; immersion in anisole does not remove the residue either, and instead leads to the deposition of a chemically altered polymer. We discuss the implications of our findings.

Separately, h-BN has recently been reported as a host to room temperature quantum emitters a phenomenon previously observed in wide-bandgap 3D semiconductors such as diamond and nanocrystal quantum dots. In this work, I study the room temperature emission properties of h-BN flakes under green laser excitation (532 nm). Using photoluminescence spectroscopy, I observe emission from isolated defect sites and characterize their spatial distribution and temporal evolution in detail, with the goal to understand the physical mechanism of this phenomenon.

Reference

[1] Garcia *et al.* *Nano Lett.* **12**, (2012) 4449

Figures

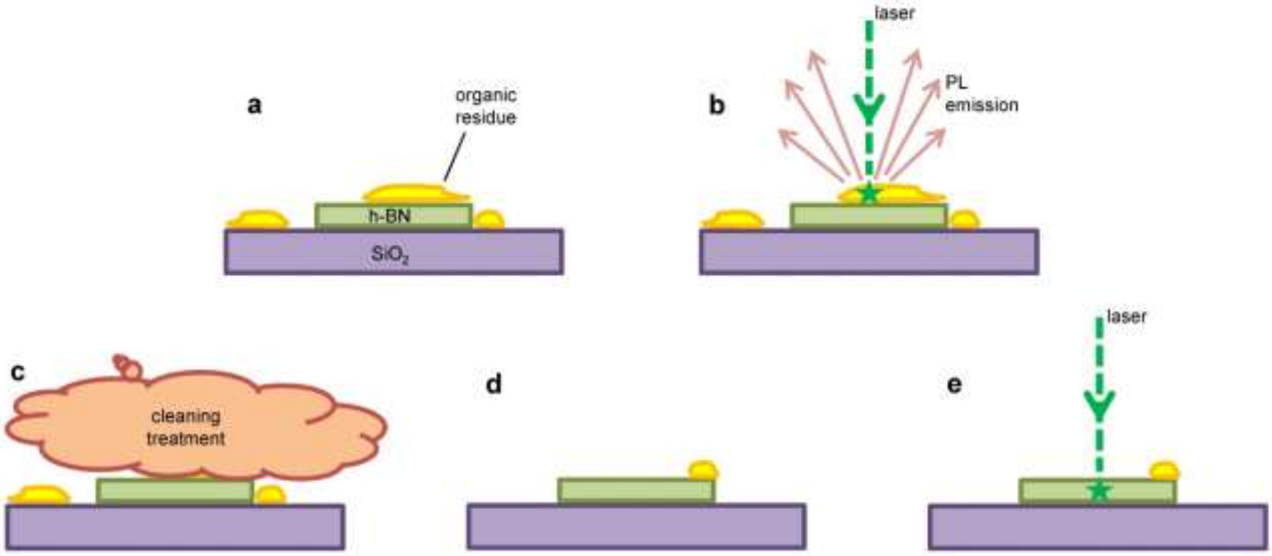

Figure 1: Schematic process flow for cleaning method efficacy assessment. **a.** Exfoliated h-BN sample, deliberately contaminated with target polymer of interest. **b.** Visualization of organic residue via photoluminescence (PL) mapping. **c.** Application of candidate cleaning method. **d.** h-BN sample after cleaning, typically with reduced organic contamination. **e.** Visualization of organic residue post-cleaning via PL mapping.