Reliable Exfoliation of Large-Area High-Quality Flakes of Graphene and Other Two-Dimensional Materials

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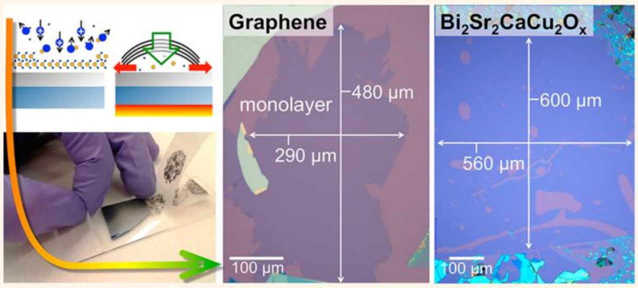
ABSTRACT Mechanical exfoliation has been a key enabler of the exploration of the properties of two-dimensional materials, such as graphene, by providing routine access to high-quality material. The original exfoliation method, which remained largely unchanged during the past decade, provides relatively small flakes with moderate yield. Here, we report a modified approach for exfoliating thin monolayer and few-layer flakes from layered crystals. Our method introduces two process steps that enhance and homogenize the adhesion force between the outermost sheet in contact with a substrate: Prior to exfoliation, ambient adsorbates are effectively removed from the substrate by oxygen plasma cleaning, and an additional heat treatment maximizes the uniform contact area at the interface between the source crystal and the substrate. For graphene exfoliation, these simple process steps increased the yield and the area of the transferred flakes by more than 50 times compared to the established exfoliation methods. Raman and AFM characterization shows that the graphene flakes are of similar high quality as those obtained in previous reports. Graphene field-effect devices were fabricated and measured with back-gating and solution top-gating, yielding mobilities of ∼4000 and 12 000 $cm^2/(V\ s)$, respectively, and thus demonstrating excellent electrical properties. Experiments with other layered crystals, *e.g.*, a bismuth strontium calcium copper oxide (BSCCO) superconductor, show enhancements in exfoliation yield and flake area similar to those for graphene, suggesting that our modified exfoliation method provides an effective way for producing large area, high-quality flakes of a wide range of 2D materials.



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Since the first successful exfoliation of monolayer graphene in 2004,[1] graphene and other 2D materials, such as h-BN, MoS_2, SnS_2, and others,[2−4] have gained large attention due to their distinctive properties. Graphene as the first discovered 2D material has provided access to new physics at single atomic thickness, such as unique electrical,[5,6] mechanical,[7] optical,[8] and sensing properties.[9,10] BN is widely used as an ideal electrically insulating substrate for vertical heterostructures with other 2D materials.[11−13] Layered metal dichalcogenides (MX_2) exhibit interesting properties when their thickness is reduced to single- or few layers,[3,14] and MX_2 field-effect transistors (FETs) demonstrated device properties such as current on/off ratio up to 10^8 (ref 3) and high charge carrier mobility.[15,16]

Exfoliation of monolayer or few-layer flakes from layered bulk crystals has played a central role in the development of 2D materials,[17] and it continues to be the primary method to rapidly gain access to high-quality flakes for exploring novel materials systems. For example, nearly all of the novel properties of graphene were first discovered and measured on exfoliated flakes: the unusual quantum Hall effects in monolayer and bilayer graphene,[6,18] Berry's phase,[6] the thickness-dependent Raman spectrum,[19] *etc.* Scalable growth techniques such as

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