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Atomic-Scale Vibrational and Electronic Response of Interfaces in Heterostructures for Spintronics Applications

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Engineering the structural or chemical architecture of functional materials at the nano or even atomic level enables emergent properties that rely on the interplay between fundamental properties of matter such as charge, spin and local atomic-scale chemistry. A striking illustration of the relevance of this strategy is provided by placing Bi_2Se_3, a topological insulator (TI) with topologically-protected helical two-dimensional surface states and one-dimensional bulk states associated with crystal defects, in close proximity with graphene. The strong spin-orbit interaction and proximity effects result in subtle and controllable electronic band structure changes at and near the interface, with exciting potential for spintronic applications [1].

A particularly powerful means of characterization of these physico-chemical effects lies within a combination of high-resolution scanning transmission electron microscopy and energy-loss spectroscopy (STEM-EELS). Recent instrumentation advances have pushed the achievable energy resolution below 10meV while maintaining atomic-sized probes [2]. This further expands the imaging and spectroscopic capabilities of STEM-EELS. In addition to the obvious benefits of improved energy resolution for local electronic structure determination through core- or valence-loss spectroscopy, these new instruments now make it possible to probe phonon modes in materials at atomic resolution, including at single atom defects [3,4], provided the scattering geometry is chosen to maximize signal and interpretability [5].

Here we probe at high energy resolution the interfaces in a system consisting of Bi_2Se_3 films grown by chemical vapor deposition on epitaxial graphene/SiC(0001), where the number of carbon layers can be carefully controlled to tune possible proximity effects between the film and the substrate. All experiments were carried out on a monochromated Nion UltraSTEM100 MC operated at 60kV, with a probe convergence semi-angle of 31mrad and a beam current of approximately 4pA (after monochromation to ~12meV resolution). Chemical mapping confirms the atomic-level chemistry of the layers, while the analysis of the carbon *K* edge fine structure provides direct insights into the nature of bonding at the SiC/graphene and graphene/Bi_2Se_3 interfaces. Strikingly, the use of a dark-field EELS geometry reveals the emergence of locally resolved fine structure in the ultra-low loss region of the spectrum: Figure 1. In addition to a further direct interrogation of the chemical bonds between the layers, and a demonstration of chemically and atomically sensitive phonon mapping at interfaces in a bulk sample, it is thought these observations could also be linked to the interplay between the various phonon modes and the Dirac plasmons in the TI layers [6].

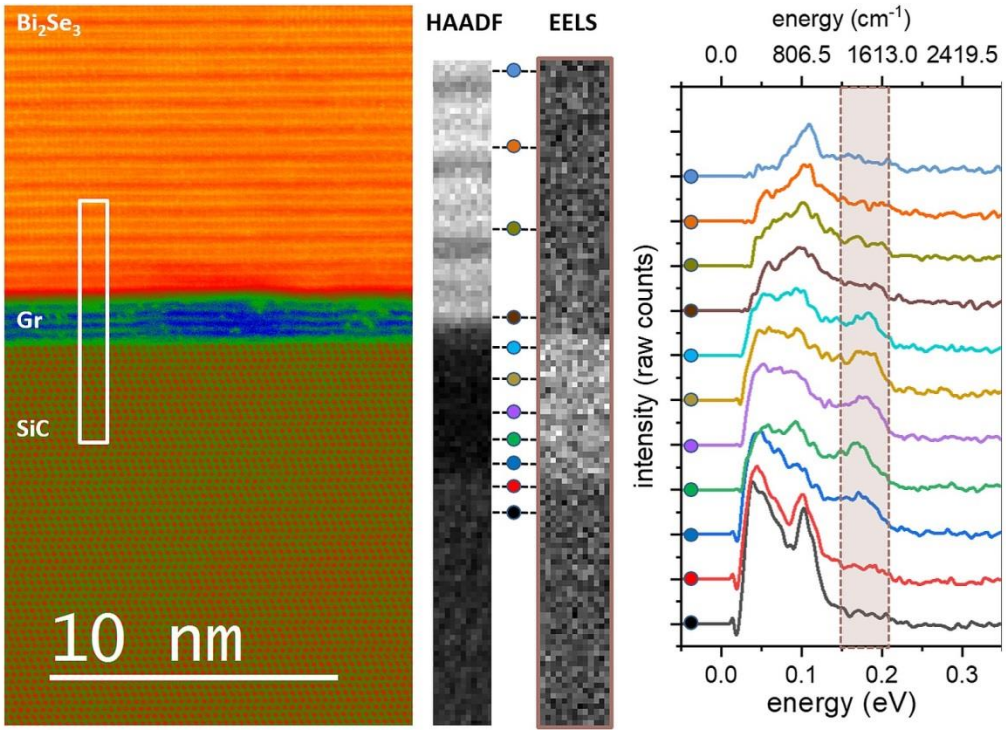

Figure 1. Left: High-angle annular dark field STEM image (false colored) of the SiC (in [11-20] zone axis)/Graphene (5 layers)/Bi2Se3 (in [1-100] zone axis) heterostructure. A white rectangle indicates the region across the interfaces over which an EELS spectrum image was recorded. Centre: simultaneous ADF and integrated EELS signal (over the energy window indicated in the extracted spectra, right), showing atomically-resolved phonon signal over the 5 graphene layers. The EELS was recorded in dark-field geometry, with a 44mrad collection aperture displaced 69mrad away from the optic axis. Right: selected background-subtracted spectra (using a standard power law model), corresponding to the positions indicated by colored dots.

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