Chiral Atomically Thin Films

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Chiral materials possess left- and right-handed counterparts linked by mirror symmetry. These materials are useful for advanced applications in polarization optics[1,2], stereochemistry[3,4] and spintronics[5]. In particular, the realization of spatially uniform chiral films with atomic-scale control of their handedness could provide a powerful means for developing nanodevices with novel chiral properties. However, previous approaches based on natural or grown films[1,2], or arrays of fabricated building blocks[6–8], could not offer a direct means to program intrinsic chiral properties of the film on the atomic scale. Here, we report a chiral stacking approach, where two-dimensional materials are positioned layer-by-layer with precise control of the interlayer rotation (θ) and polarity, resulting in tunable chiral properties of the final stack.[9] Using this method, we produce left- and right-handed bilayer graphene, that is, a two-atom-thick chiral film. The film displays one of the highest intrinsic ellipticity values (6.5 deg μm^{-1}) ever reported, and a remarkably strong circular dichroism (CD) with the peak energy and sign tuned by θ and polarity. We show that these chiral properties originate from the large in-plane magnetic moment associated with the interlayer optical transition.[10] Furthermore, we show that we can program the chiral properties of atomically thin films layer-by-layer by producing three-layer graphene films with structurally controlled CD spectra. Our approach, which is generally applicable to other layered materials, can provide a powerful platform for generating and integrating ultrathin devices based on chiral metamaterials with programmed interactions with other chiral objects, including photons, molecules, and spin polarized electrons.

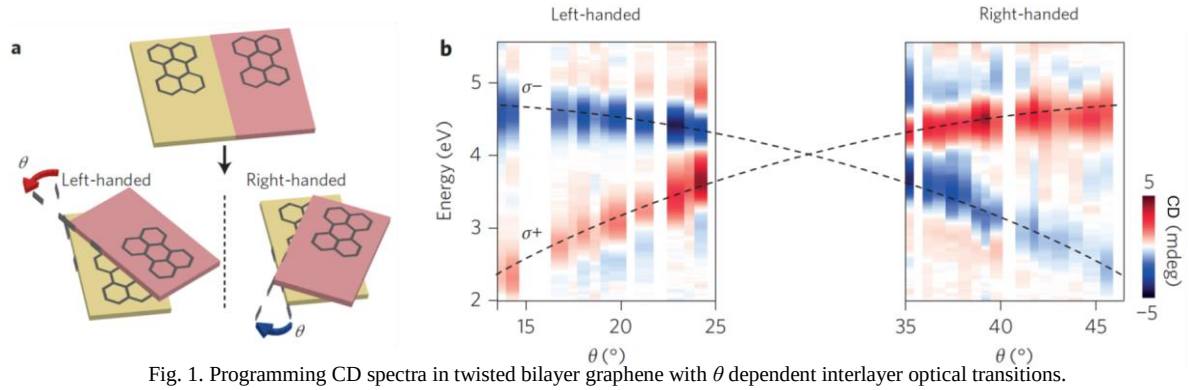

Fig. 1. Programming CD spectra in twisted bilayer graphene with θ dependent interlayer optical transitions.

Acknowledgement

This work was supported by the National Science Foundation (NSF) through the Cornell Center for Materials Research (NSF DMR-1120296), the AFOSR (FA2386-13-1-4118), and the Nano Material Technology Development Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT, and Future Planning (2012M3A7B4049887). Y. Ogawa was partially supported by Grant-in-Aid for JAPS Fellows. A. Sánchez-Castillo and C. Noguez were supported by DGAPA-UNAM(PAPIIT IN107615) and CONACyT (179454). Sample fabrication was performed at the Cornell Nanoscale Science & Technology Facility, a member of the National Nanotechnology Infrastructure Network, which is supported by the National Science Foundation (ECS-0335765).

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