

**YY6.04/W6.04**

Revealing Angular Dependence on the Optical Response of Bilayer Graphene by Electron Energy-loss Spectroscopy

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We present a systematic study of the optical response of bilayer graphene as a function of misorientation angle using a combination of electron energy-loss spectroscopy in an aberration-corrected scanning transmission electron microscope. We find that an additional absorption peak ( $\sim 4.40$  eV), which has not been reported before, emerges in the ultraviolet region of the energy loss function of bilayer graphene for large misorientation angles ( $\sim 30$  degrees). The additional absorption peak is below the nominal  $\pi$  peak of bilayer graphene ( $\sim 5.10$  eV) and also at a different energy position of the  $\pi$  peak for monolayer graphene (4.95 eV). The analysis of the data also reveals that  $\pi$  peak as well as the  $\pi+\sigma$  peak ( $\sim 15$  eV) do not shift in energy as function of misorientation angle. The observations will be explained using total-energy first-principles calculations based on density functional theory within the random phase approximation. The results obtained in this study indicate that the misorientation angle between graphene layers can affect the optical properties, therefore suggesting that the misorientation angle between layers could also be used as a variable in the designing of novel optoelectronic devices based on hybrid two-dimensional materials. This research was supported by Oak Ridge National Laboratory's Shared Research Equipment (ShaRE) User Facility (JCI), which is sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy, and by the National Science Foundation grant No. DMR-0938330 (WZ).