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Energy Resolved Photoconductivity Mapping in a Monolayer Bilayer WSe_2 Lateral Heterostructure

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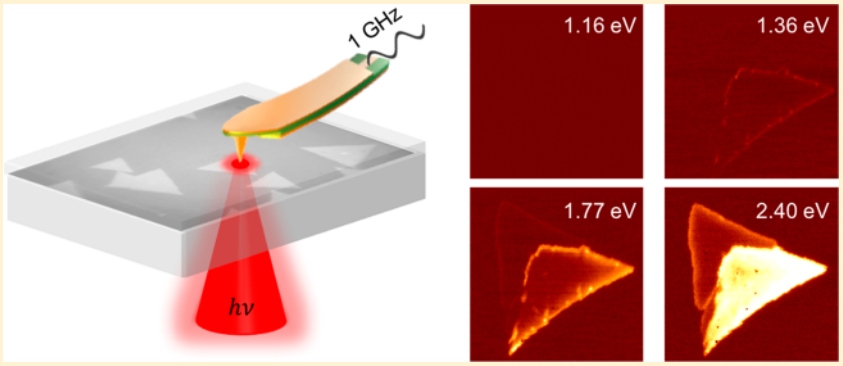
Supporting nformation



ABSTRACT: Vertical and lateral heterostructures of van der Waals materials provide tremendous exibility for band-structure engineering. Because electronic bands are sensitively affected by defects, strain, and interlayer coupling, the edge and heterojunction of these two-dimensional (2D) systems may exhibit novel physical properties, which can be fully revealed only by spatially resolved probes. Here, we report the spatial mapping of photoconductivity in a monolayer bilayer WSe_2 lateral heterostructure under multiple excitation lasers. As the photon energy increases, the light-induced conductivity detected by microwave impedance microscopy first appears along the heterointerface and bilayer edge, then along the monolayer edge, inside the bilayer area, and finally in the interior of the monolayer region. The sequential emergence of mobile carriers in different sections of the sample is consistent with the theoretical calculation of local energy gaps. Quantitative analysis of the microscopy and transport data also reveals the linear dependence of photoconductivity on the laser intensity and the in uence of interlayer coupling on carrier recombination. Combining theoretical modeling, atomic-scale imaging, mesoscale impedance microscopy, and device-level characterization, our work suggests an exciting perspective for controlling the intrinsic band gap variation in 2D heterostructures down to a regime of a few nanometers.

KEYWORDS: *van der Waals materials monolayer bilayer interface edge states microwave impedance microscopy photoconductivity imaging*

The electronic properties of two-dimensional (2D) van der Waals (vdW) materials are highly sensitive to the local atomic arrangement,[1,2] which offers tremendous opportunities for band structure engineering. In transition metal dichalcogenides (TMDs), for instance, the band gap can be tuned by choosing different MX_2 (M = Mo, W···; X = S, Se, or Te) compounds and alloys,[3,4] changing the number of layers,[5,6] vertical stacking, or lateral stitching of different materials[1,2,7] and, in the case of vertical heterostructures, varying the twist angle between the atomic layers.[8,9] Moreover, broken crystalline symmetry at the edges and heterointerfaces of vdW materials may lead to the emergence of new electronic states in the band structure,[10,11] which could even become topologically nontrivial in certain cases.[12 14] When these possibilities are taken into account, the local band structure in 2D materials and heterostructures can be readily explored for electronic applications, provided that such spatial variations are well-understood and controlled.[15 17]

Many optoelectronic applications such as photodetectors and photovoltaic devices require the photoexcitation of mobile carriers by above-gap illumination and the subsequent charge transport.[18] In conventional photocurrent spectroscopy (PCS) experiments,[19] the sample is illuminated by a wavelength-tunable

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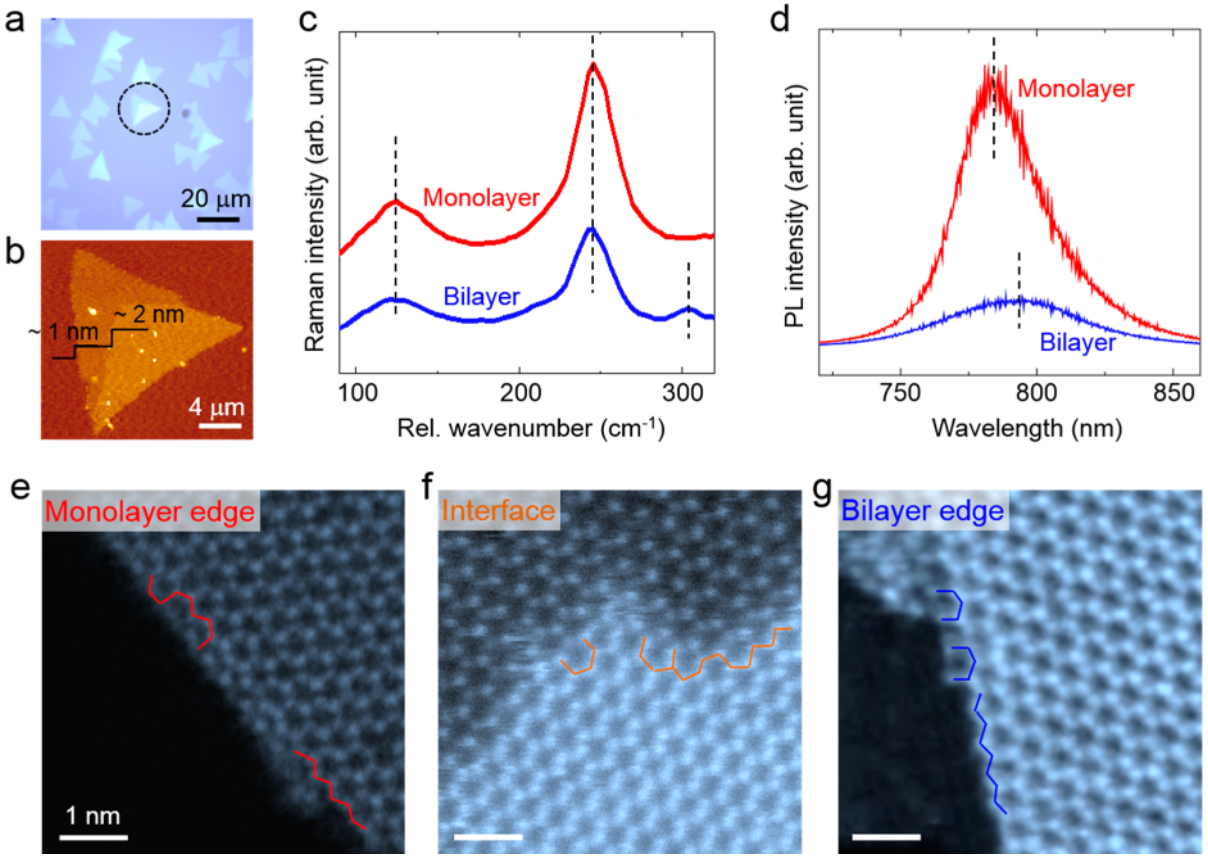

Figure 1 Characterization of the WSe_2 sample. (a) Optical image of the CVD-grown WSe_2 sample, showing several akes with the desired monolayer bilayer heterostructure. (b) AFM image of the ake inside the dashed circle in panel a. (c) Raman spectra in the monolayer and bilayer regions. (d) Photoluminescence spectra in the monolayer and bilayer regions. The dashed lines in panels c and d indicate the Raman and PL peaks. (e g) HAADF-STEM images near the monolayer edge, monolayer bilayer interface, and bilayer edge, respectively. Some zigzag and armchair sections are highlighted at the disordered edge and heterointerface. The scale bars in panels e g are 1 nm.

light source, and the induced photocurrent is measured as a function of the photon energy. For spatially resolved studies, scanning photocurrent microscopy (SPCM) with a diffraction-limited resolution of 0.5 1 m can map out the photo-generated current across the source and drain electrodes when a focused laser beam scans over the device.[20] The SPCM signal is usually strongest near the electrodes because the current is most effectively generated in regions with a large electric field. Its interpretation can be further complicated by carrier diffusion, Schottky contact, thermoelectric, and bolometric effects,[20 23] making it unsuitable to investigate spatial variations of intrinsic material properties. For thin films deposited on metallic substrates, the light-induced conductance can also be detected by conductive atomic-force microscopy (C-AFM),[24] which again suffers from the extrinsic Schottky effect. Here, we report the imaging of intrinsic photoconductivity in a monolayer bilayer (ML-BL) WSe_2 lateral heterostructure by microwave impedance microscopy (MIM)[25] with light stimulation at multiple wavelengths. Taking advantage of the capacitive coupling, the MIM can perform non-invasive nanoscale photoconductivity imaging without the need for electrical contacts.[26,27] As the photon energy of excitation lasers increases from 1.16 to 2.78 eV, the photo response first appears along the ML BL interface and the BL edge, then along the ML edge and inside the BL bulk, and finally in the interior of the ML. The spatial evolution of photo response is consistent with the variation of local band gaps calculated by the first-principles method. The mesoscale photoconductivity mapping fills an important void between atomic scale characterization and device-level transport measurements. Our results suggest the exciting possibility of controlled in-plane band gap engineering on the length scale of nanometer, which is uniquely enabled by atomically thin lateral heterostructures and may lead to completely new integrated devices.

The WSe_2 nano akes were grown on double-side polished sapphire substrates by chemical-vapor deposition (CVD). Figure 1a shows an optical microscopy image of the as-grown akes, where the monolayer and bilayer regions display different optical contrast. In this work, we focus on akes with both regions, i.e., on a ML-BL lateral heterostructure whose layer thickness was confirmed by the AFM image in Figure 1b. The Raman and photoluminescence (PL) spectra of the ML and BL regions (Figure 1c,d) were measured by a confocal Raman microscope. The Raman intensities of both the in-plane E^1_{2g} (~128 cm ^{1}) and out-of-plane A_{1g} (~246 cm ^{1}) phonon modes in bilayer WSe_2 are weaker than that in the monolayer, whereas the out-of-plane B^1_{2g} (~304 cm ^{1}) mode only appears in the bilayer region, consistent with previous investigations.[6,28] The Raman maps by integrating these three modes are shown in Figure S1. The PL signal of monolayer WSe_2 at ~780 nm is much stronger than that of the bilayer area at ~794 nm due to the direct-to-indirect-gap transition.[5,6,29,30] To determine the atomic arrangement of the sample, we carried out high-resolution imaging by transmission electron

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