

Optically probing the asymmetric interlayer coupling in rhombohedral-stacked MoS₂ bilayer

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Abstract

The interlayer coupling is emerging as a new parameter for tuning the physical properties of two-dimensional (2D) van der Waals materials. When two identical semiconductor monolayers are stacked with a twist angle, the periodic interlayer coupling modulation due to the moiré superlattice may endow exotic physical phenomena, such as moiré excitons and correlated electronic phases. To gain insight into these new phenomena, it is crucial to unveil the underlying coupling between atomic layers. Recently, the rhombohedral-stacked transition metal dichalcogenide (TMD) bilayer has attracted significant interest because of the emergence of an out-of-plane polarization from non-ferroelectric monolayer constituents. However, as a key parameter responsible for the physical properties, the interlayer coupling and its relationship with ferroelectricity in them remain elusive. Here we probe the asymmetric interlayer coupling between the conduction band of one layer and the valence band from the other layer in a 3R-MoS₂ bilayer, which can be understood as a result of a layer-dependent Berry phase winding. By performing optical spectroscopy in a dual-gated device, we show a type-II band alignment exists at K points in the 3R-MoS₂ bilayer. Furthermore, by unraveling various contributions to the band offset, we quantitatively determine the asymmetric interlayer coupling and spontaneous polarization in 3R-MoS₂. Our results unveil the physical nature of stacking-induced ferroelectricity in TMD homostructures and have important implications for many-body states in the moiré superlattice of 2D semiconductors.