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Resonant inelastic light scattering micro-spectroscopy to probe inter-moiré miniband excitations in twisted 2D semiconductors

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Background incl. aims:
Resonant inelastic light scattering (RILS) is a powerful technique to study collective charge excitations in semiconductor quantum nanostructures including quantum wells and nanowires [1]. The application of this technique to probe collective behavior of electrons and holes in novel two-dimensional (2D) materials has not been explored yet. The moiré superlattice potential in twisted 2D semiconductors, e.g. WSe_2, leads to carrier localization and emergence of (flat) moiré bands in the electronic band structure, which host interesting correlation physics [2]. Although the signatures of such flat bands have been shown in recent studies including scanning tunneling spectroscopy (STS) [3] and angle resolved photoemission spectroscopy (ARPES) [4] , the observation of electronic transitions between them proving the emergence of these bands, especially near the K-point of the electronic structure, have remained elusive. In this work, we used RILS micro-spectroscopy to probe the inter-moiré band excitations (IMBE) in twisted bilayers of WSe_2 with a twist angle of ~3° and ~8°.

Methods:
The twisted WSe_2 bilayers were mechanically exfoliated from bulk crystals of WSe_2 using a dry deterministic transfer technique based on viscoelastic stamps. The bilayers were also encapsulated in multilayers of hBN to protect them from environment and to provide atomically flat interfaces. The multilayer stack was built upon a ~300nm silicon dioxide layer thermally grown on p-doped silicon substrate. After preparation, the samples were annealed in vacuum and then mounted inside a refrigerator and cooled down to 4K using liquid Helium. Different regions of the sample were identified using a white LED source and Köhler illumination in combination with piezoelectric x-y-z stages and imaged using a CMOS camera. The twisted bilayer regions of the sample were optically excited using a laser beam from a frequency tunable Ti : Sa laser (linewidth = 50 KHz), cleaned up using a monochromator (1nm bandpass) and coupled to the sample through a large numerical aperture (NA = 0.82) objective lens which is compatible with low temperatures. The excitation energies were chosen near the direct optical bandgap at the K-point pre-determined using photoluminescence spectroscopy. The laser spot size on the sample was ~2µm. The emitted and scattered light from the sample was collected using the same objective and guided to a triple-spectrometer operated in a subtractive mode to reject any stray light. The RILS spectra were recorded using a liquid nitrogen cooled CCD camera.

Results:
Our results show the signatures of transitions between the 1st and 2nd moiré bands for the twist angle of ~8° and between the 1st moiré band and 2nd, 3rd and higher moiré bands for a

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twist of ~3° [5] We further compare our results with the RILS spectra of natural bilayers of WSe_2 where the absence of IMBE as a consequence of absence of a moiré superlattice further consolidates our results. The IMBE transition energies are in excellent agreement with those expected from theoretical ab-initio calculations in spite of broadening resulting from twist angle disorder and non-vertical transitions [5].

Conclusions:
Our results quantify the IMBE energies at the K-point of the electronic structure, where the states relevant for correlation physics are hosted. They further establish low temperature RILS as a powerful non-invasive technique for probing collective charge excitations in twisted 2D semiconductors [5].

Keywords:

2D materials, RILS, moiré minibands

Reference:

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