Misorientation-angle-dependent electrical transport across molybdenum disulfide grain boundaries

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Grain boundaries in monolayer transition metal dichalcogenides have unique atomic defect structures and band dispersion relations that depend on the inter-domain misorientation angle. Here, we explore misorientation angle-dependent electrical transport at grain boundaries in monolayer MoS2 by correlating the atomic defect structures of measured devices analysed with transmission electron microscopy and first-principles calculations. Transmission electron microscopy indicates that grain boundaries are primarily composed of 5–7 dislocation cores with periodicity and additional complex defects formed at high angles, obeying the classical low-angle theory for angles <22°. The inter-domain mobility is minimized for angles <9° and increases nonlinearly by two orders of magnitude before saturating at $\sim 16 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ around misorientation angle $\approx 20°$. This trend is explained via grain-boundary electrostatic barriers estimated from density functional calculations and experimental tunnelling barrier heights, which are $\approx 0.5 \text{ eV}$ at low angles and $\approx 0.15 \text{ eV}$ at high angles ($\geq 20°$).
Understanding the atomic structures of grain boundaries (GBs) is essential to controlling and improving electrical transport properties in both bulk and low-dimensional materials. In traditional mesoscopic semiconductors, GBs are scattering sites for majority carriers and degrade transport via the formation of electrostatic barriers 1,2. Such degradation is amplified in low-dimensional materials due to the limited phase space and reduction in Coulomb screening 3. Two-dimensional (2D) transition metal dichalcogenides are one class of low-dimensional materials with unique electrical and electro-optical properties showing promise for emerging valleytronics 4 and soft electronics applications 5. However, large-area monolayer transition metal dichalcogenides synthesized via chemical vapour deposition (CVD) inevitably possess GBs between randomly oriented crystal domains. Early theoretical research involving density functional theory (DFT) calculations for MoS2 predicted the possibility of both metal and semiconductor GB characteristics, depending on the local atomic structures 6-8. However, experimental assessment of MoS2 GB transport is still under debate in the literature 6-11 due to a large device-to-device variation, poor single-domain carrier mobility, and, most importantly, lack of correlation between transport properties and GB atomic structures.

Here, we overcome these difficulties by directly correlating four-probe transport measurements across single GBs with both high-resolution transmission electron microscopy (TEM) imaging of the measured devices and first-principles calculations. Using an inert-environment fabrication procedure, we observe a positive and nonlinear correlation between field effect mobility ($\mu_{FE}$) and misorientation angle (z). The precise boundary location, orientation and atomic structure of transistor devices were also examined by TEM, results of which were cross-referenced with further characterization and imaging of pristine MoS2 GBs. On using DFT to calculate the z-dependent local GB band structures, equilibrium electrostatic GB barriers were found to be correlated with experimentally determined barriers, which gives a clear understanding of the underlying mobility dependences.

**Results**

**Electrical characterization.** Monolayer MoS2 flakes (typical sizes of 20–40 $\mu$m) were grown by CVD and subsequently transferred onto SiO2 (100 nm)/Si substrates (see Methods). GB positions were then initially determined by dark-field optical microscopy images while considering the growth kinetics (Supplementary Fig. 1). Regions with no sign of wrinkling or multilayers were then selected to prevent misinterpretation of the transport mechanism. To suppress interaction with oxygen-related functional groups, further device fabrication was performed in a glove box ($H_2O$, $O_2$< 1 p.p.m.) and only acetone solvent was used during the electron-beam lithography procedures to minimize additional GB functionalization.

$V_{gs}$-dependent transport measurements were performed on an elongated Hall-bar geometry to allow single-crystal and GB four-probe sheet resistance ($R_s$) and $\mu_{FE}$ comparison. Figure 1a shows the schematic of a typical carrier transport across a GB, while a representative multi-terminal device tested in this work is displayed in Fig. 1b. A GB with an optical microscopy-determined misorientation angle of 19° is located near the centre of the monolayer MoS2 film (red-dashed line), where the white dashed lines (inset) denote the pre-etch shape of MoS2 flakes (Supplementary Fig. 2). The $R_s$ and $\mu_{FE}$ associated with intra-domain and inter-domain transport were measured as a function of $V_{gs}$ by choosing appropriate electrodes either within the domain or across the GB line, respectively (Fig. 1c and Supplementary Fig. 7a). The selected-area electron-diffraction (SAED) pattern (right inset of Supplementary Fig. 3b) and scanning (STEM) findings of the exact device in Fig. 1b both revealed that z = 19°, which further agrees with the optical estimates within a statistical error of ±2° (Supplementary Fig. 3).

In this 19° sample, the intra-domain (inter-domain) $\mu_{FE}$ = 24–60 (16) cm2 V−1 s−1 and $R_s$ = 30–85 (95) kΩ cm−2. After completing the electrical measurements, the devices were expeditiously transferred to an Au TEM grid (an Au grid was used to suppress additional oxidation of the sample as the moisture level should be minimized to prevent sample damage). STEM analysis of the boundary revealed that the GB line consists of a series of 5–7 membered ring cores, as indicated by the orange-dashed line in the inset of Fig. 1c. (Mo atoms are denoted in blue, while S atoms are displayed in yellow.)

For further angle-dependent transport analysis, a statistical distribution of the mobility was obtained from 43 devices (29 intra-domain and 14 inter-domain, refer to Supplementary Fig. 3 for raw mobility data), as shown in Fig. 1d. A decisive cutoff separates intra-domain and inter-domain data at $\mu_{FE}$ = 16 cm2 V−1 s−1 . The intra-domain $\mu_{FE}$ dataset forms an ordered but highly dispersive statistical distribution with a maximum (minimum) of $\mu_{FE}$ = 17 (115) cm2 V−1 s−1 , which results from conduction band-tail states localized near the conduction band edge 12 (Supplementary Fig. 5). Figure 1d shows the raw data and natural-log of $\mu_{FE}$ histograms (inset); excellent fitting to a log-normal distribution was observed, with an expected $\mu_{FE}$ = 44 cm2 V−1 s−1 . Notably, seven devices had a $\mu_{FE}$ > 75 cm2 V−1 s−1 and on-state $R_s$ was consistent with the best-reported exfoliated results 13 (also see Supplementary Fig. 6).

With statistically reliable intra-domain mobility data to contrast, the inter-domain misorientation angle-dependent mobility was measured. Results are shown in Fig. 1e ($R_s$ data are presented in Supplementary Fig. 7). Interestingly, the inter-domain mobility increases for small angles (8–20°), although some variability exists in the low-angle data. Above 20°, $\mu_{FE}$ saturates below the 16 cm2 V−1 s−1 intra-domain cutoff. Overall, the inter-domain mobilities vary by 2 orders of magnitude across the tested range of z. It was further observed that although devices with 8–20° inter-domain misorientation display poor transport, these samples consistently conduct, as demonstrated by the large number of inter-domain data points within this range. Conversely, samples within the 20–60° range show better transport properties; many GBs were insulating (broken) due to a torn/poor interface. One example is the 34° sample (Fig. 1e), which was measurable only after annealing at 420 K, and then quickly became insulating and unmeasurable at 300 K. Other missing inter-domain data of Fig. 1e resulted from similar insulating behavior, likely due to tearing during substrate transfer (Supplementary Fig. 8). This observation agrees with previous experiments on 3D crystals, where high-angle samples frequently shattered after growth 14. The peculiarly high mobility of the inter-domain 19° device results from the boundary crossing the corner of the contact (Fig. 1b,c), which was discovered during post-transfer scanning electron microscopy or STEM analysis.

**Structural characterization.** To directly correlate the transport results to the atomic defect structures, we performed extensive TEM/STEM analysis of the aforementioned devices and many other representative samples; the results are summarized in Fig. 2. The diffraction contrast in the dark field (DF)-TEM images (Figs 2a–d) clearly reveals the morphology and location of the GBs. Low angle (LA)-GBs (0–13°) are rather straight, and formed by aligned dislocation cores, visible as dotted lines in the DF images (Fig. 2a). The SAED is presented in the inset of Fig. 2a,
while the high-resolution STEM annular dark field (ADF) image is shown in Fig. 2c.

In bulk specimens, LA-GBs are composed of periodically piled edge dislocations. The distance between the edge dislocation cores can be estimated by \( d = \frac{b}{\sin(z)} \), where \( b \) is the Burgers vector. The dislocations are composed of ‘Up’ or ‘Down’ types (Figs 2j–m) that correspond to S–S bonding or Mo–Mo bonding at the core, respectively. Due to the three-fold (not six-fold) symmetry of the MoS\(_2\) monolayer, a lack of inversion symmetry exists. Such a scenario induces the formation of two types of LA-GBs with the aforementioned ‘Up’ or ‘Down’ dislocations along the armchair directions, as highlighted in Fig. 2n. All devices investigated for electrical transport were of the ‘Up’-type boundary variety (except for \( 19^\circ \), which is not utilized in the comparison) and thus comparison can be made. Furthermore, the energies associated with ‘Up/Down’ dislocations are similar, and both are energetically favourable since they both have the smallest Burgers vector [11–20]. Due to kinetic growth limitations, GBs with overall relative orientation between the armchair directions of MoS\(_2\) are also composed of [11–20] dislocations (‘Up/Down’). The boundary is decomposed into short armchair-oriented sections and is slightly kinked on the mesoscopic scale (Fig. 2a). For bulk materials, LA-GBs usually imply \( \alpha < 6^\circ \) due to strong interactions between the strain fields generated by adjacent dislocation cores at higher angles [15]. However, basal plane strain relaxation can occur in 2D materials via local rippling or buckling [16], thereby allowing for an extension of LA-GB theory to larger angles. The periodicity of the dislocation core distance is inversely proportional to \( \alpha \), in accordance with the classical LA-GB theory (Fig. 2i and Supplementary Fig. 9), but the range greatly exceeds the classical limit and is extended up to \( 13^\circ \), at which point the GB lines are bent by intense strain fields. Thermodynamically, these LA-GBs should follow a straight line in a direction perpendicular to the Burgers vector (armchair directions in MoS\(_2\)), which is consistent with our observations (Supplementary Fig. 10).

High-angle (HA)-GBs between two domains can be classified as either coherent or incoherent, the former of which refers to GBs that possess a periodic lattice. Using notation from coincident site lattice theory [17], the most favourable coherent boundaries for monolayer MoS\(_2\) are \( \Sigma = 22^\circ \), \( \Sigma = 28^\circ \), \( \Sigma = 19^\circ \), and so on (Supplementary Fig. 11). From the results of TEM/STEM characterizations and DFT calculations (Supplementary Fig. 12), we observed that most HA-coherent GBs also follow straight lines composed of continuous 5–7 ring cores, in a manner similar to LA-GBs (Figs 2c,g). Any GB with an \( \alpha \) that deviates from coherent angles will be an incoherent GB (Fig. 2b). A curved (non-straight) GB can be identified in the atomic scale images (Fig. 2f). When compared to coherent GBs, the incoherent GBs have a larger aperiodic width of defects, whereas the mirror GBs (60° GB) are mostly straight along the zigzag direction and composed of a series of 4–4 cores with few kinks (Figs 2d,h). However, when the mirror GB follows the armchair direction (due to growth kinetics), it is decomposed into a series of 4–8 cores (Supplementary Fig. 13).

In terms of mechanical stability, LA-GBs are mostly stable, regardless of the GB type. This explains how we were able to accumulate a large set of data points, as shown in Fig. 1e. However, in the HA region, only coherent GBs with \( \alpha = 22^\circ \), \( 38^\circ \) and 60° were stable. While many samples contained boundaries with intermediate angles, transport data were not collected for these specimens because GBs were easily fragmented during the transfer process. Since incoherent GBs are generally wider with a complex array of defect structures (see Fig. 2f), they are
energetically less stable when compared to coherent GBs. This agrees well with the theoretical prediction that the total energy usually increases in response to the misorientation angle\textsuperscript{18}.

**Discussion**

Dislocations and GBs in III–V systems (e.g., GaN) have previously been modeled by a generalized line charge with density \( \rho_L \propto \text{defect density} \). Increasing \( \rho_L \) induces a larger electrostatic potential and results in a lower effective mobility\textsuperscript{19,20}. In comparison, here we observe a positive and nonlinear correlation between \( \mu_{FE} \), the dislocation core density and \( \alpha \). To explain why greater dislocation core density permits greater inter-domain mobility, and if results are consistent with the aforementioned theoretical models, we first consider whether the boundary is charged. \( I_{ds} \)–\( V_{gs} \) curves from 26 two-terminal FETs (Fig. 3a) reveal that \( I_{ds} \) is rather invariant and limited by leakage current, while hysteresis is trivial and a uniform 0.25 V (300 K), equal to the \( V_{gs} \) point spacing. Boundary charge must therefore be fixed, which is further examined by comparing respective \( V_{th} \) values. As transport is band-tail limited and a well-defined saturation regime is absent in the \( I_{ds} \)–\( V_{gs} \) curves (Fig. 3a), \( V_{th} \) is defined by the \( V_{gs} \) logarithmic turn-on point corresponding to the peak subthreshold swing (SS). This point is denoted at \( V_{gs} - V_{th} = 0 \) by a dotted line in Fig. 3a. We found \( -11 \text{ V} < V_{th} < -5 \text{ V} \) for all samples (Fig. 3b), which is comparable to that obtained from STM studies of atomically clean exfoliated samples\textsuperscript{21}. A positive 1.6-V shift (in median value) is observed in the inter-domain samples compared with intra-domain, which is clear evidence for the existence of an electrostatic potential barrier. This potential barrier can be estimated by noting that, at \( V_{th} \), the electrostatic potential \( \Delta V_{th} \) is measured at the Fermi level (\( E_F \)) within the bandgap, \( C_{SiO_2} \approx 0 \). Substituting \( C_{SiO_2} \), and SS above, we observe that \( C_{SiO_2} = 76 \text{ nF cm}^{-2} \). An order-of-magnitude estimate for the potential barrier height \( \Phi_0 \) is obtained from an equivalent circuit model with SiO\textsubscript{2} capacitance in series with the MoS\textsubscript{2} self-capacitance, \( \Delta E_F = \Delta V_{th} (C_{SiO_2}/C_{loc}) = 190 \text{ meV} \) (see Methods).

However, in accordance with \( \mu_{FE} \), inter-domain \( I_{ds}/I_{off} \), and \( R_s \) results (Figs 3a and 1), \( \Phi_0 \) is \( \alpha \)-dependent. For \( 9^\circ \leq \alpha \leq 60^\circ \) the core-density periodicity is less than 2 nm, and thus electrostatic
Intra-domain devices have an average tunneling barrier height in the vicinity of dislocation cores for smaller crosses the completely delocalized state in the middle of (supercell). In contrast, at the mirror boundary, the Fermi level is surmised from the strongly localized charges at the GBs (Fig. 4c, resonant charge-scattering centres, a notion that can be similarly come mostly from the Mo 4d orbital at the 5–7 dislocation cores pinning. The first two bands above the Fermi level (red line) negligible dispersion for all three angles, indicating strong Fermi level pinning. The second two bands above the Fermi level (red line) come mostly from the Mo 4d orbital at the 5–7 dislocation cores (‘Down’ core for the first band and ‘Up’ core for the second band). These energy levels near the Fermi level may serve as resonant charge-scattering centres, a notion that can be similarly surmised from the strongly localized charges at the GBs (Fig. 4c, ‘Down’ core at the centre and ‘Up’ core at the right edge of the supercell). In contrast, at the mirror boundary, the Fermi level crosses the completely delocalized state in the middle of (Γ–Y), revealing metallic character due to the 4–4 ring dislocation cores (Supplementary Fig. 16). The delocalized metallic feature with weak pinning provides a likely explanation for the high mobility of the mirror GBs.

Angle-dependent transport was then modelled by considering local band bending at the boundary induced during charge equilibration, as shown schematically in Fig. 4d. At equilibrium, the interfacial $\Phi_b$ is approximated by $\Phi_b = W_{GB} - W_{MoS_2} - E_d$, where $W_{GB}$ is the GB work function, $W_{MoS_2}$ is the MoS$_2$ work function and $E_d$ is the trap energy with respect to the GB work function. $E_d$ is fixed due to Fermi level pinning, resulting in the equilibrium band diagram displayed at the bottom of Fig. 4d. The work functions for the three GB structures obtained by DFT are 5.67 eV (7°), 5.71 eV (13°) and 5.50 eV (22°). Due to the presence of band-tail dispersion in the pristine MoS$_2$, $W_{MoS_2}$ is unknown and $V_F$-dependent, and an exact $\Phi_b$ is undefined. However, relative values of $\Phi_b$ are compared with the JWKB experimental results in Fig. 3c; an offset of 150 meV was added to the calculated values. Both the ‘Up’ and ‘Down’ dislocation types show nearly identical trend, with $\Phi_b$ decreasing at higher $\phi$. The localization of charges at the boundary and subsequent Fermi level pinning determine the effective GB potential barrier height. GBs with defect states that are energetically farther from the conduction band (smaller $E_d$) will have greater $\Phi_b$ values. Consequently, in MoS$_2$ the exact GB atomic structure determines the barrier, and, although the density of dislocation cores increases with $\phi$, the charge density per unit length actually decreases as dislocation density increases. These results indicate that boundary transport is consistent with the previous theoretical treatment of 2D systems, although no exploration of the exact scattering mechanism at the GB is possible here.

For incoherent HA-GBs, more complicated defects than 5–7 cores exist, along with a greater number of dangling bonds in a zigzag shape or containing holes (Fig. 2), which cannot be explained by our JWKB tunnelling barrier treatment. As opposed to a straight potential barrier found in coherent GBs, a non-periodic kinked boundary likely forms a wider depletion region with regions of greater localized charge density. Furthermore, due to growth limitations for LA-GBs (<7°), we were unable to characterize the transport properties for very small $\phi$. In this regime, the induced potential perturbations from individual dislocation cores will become more isolated as $\phi$ decreases. We postulate that carriers will simply pass within the sawtooth-like GB potential, leading to improved mobility with reduced $\phi$.

In conclusion, we have provided a more unified picture of the relationship between mobility, merging angle and atomistic structures of the GBs of monolayer MoS$_2$. The results provide practical expectations regarding transport properties in large-area films, which will be restricted largely by the poor mobility across LA GBs. The results obtained in this work are applicable to other similar 2D systems, and contribute to the fundamental understanding of transport in semiconductors.

Methods

MoS$_2$ grown on SiO$_2$/Si wafer by CVD. OptiPrep density gradient medium (Sigma-Aldrich, D1556, 60% (w/v) solution of iodixanol in water) (defined as solution A) was purchased and used as a medium to dissolve promoter and precursor. It has no influence on growth. The promoter was prepared by dissolving sodium cholate (SC) hydrate (Sigma-Aldrich, C6445) in deionized (DI) water (0.6 g of SC in 30 ml of DI water), defined as solution B. A solution-phase ammonium heptamolybdate (AHM) precursor was also prepared by dissolving ammonium molybdate tetrahydrate (Sigma-Aldrich, 431346) into DI water (defined as solution.
device fabrication. The synthesized MoS₂ on an SiO₂/Si substrate was transferred onto a highly doped (10¹⁸ cm⁻³) silicon substrate with a 100-nm-thick thermal oxide layer by a conventional transfer method mentioned in the previous TEM sample preparation. Glovebox preparation was employed for baking samples at 120°C for SiO₂ and MoS₂ surface trap passivation for 2 h before beginning fabrication. All further baking steps during fabrication were also performed in the glovebox. Electron beam lithography and evaporation (Ti/Au 7/35 nm) were used to define the MoS₂ electrode contacts for each sample. MoS₂ was patterned into an elongated Hall bar structure via a final electron beam lithography step, followed by a weak 10-W plasma etch for 10 s under a 30-sccm flow of SF₆.

TEM and DF-TEM. TEM imaging was conducted using a JEM ARM 200F machine operated at 80 kV; no damage was apparent in the samples at this accelerating voltage. The acquisition time for DF imaging was 1 s using the smallest objective lens aperture. The HR-TEM imaging acquisition time was also 1 s.

Figure 4 | DFT calculations of the band structure for GBs with various misorientation angles. Band structure near the Fermi level (red dash line) of (a) tilt and (b) mirror GBs. Inset shows the first Brillouin zone. The first band above the Fermi level is highlighted by a green-dashed line. (c) The charge densities for the first band above the Fermi level, as indicated by the ‘Down’ type of 5–7 dislocation cores for 7°, 13° and 22°. (d) The expected flat band alignment near the GB with defect states extracted from the band structures and the corresponding band bending after Fermi level equilibrium. The Schottky barrier height decreases in response to α.
ADF-STEM. ADF-STEM imaging was conducted using a probe aberration-corrected JEM ARM200F operated at 80 kV. High-angle ADF images were acquired at 20 mrad convergence angle. Because the energy of the low-voltage electron beam was below the damage threshold energy, the pristine MoS2 lattice remains stable and defect-free with regular scanning. Images presented in the main text were acquired with a medium-angle ADF detector with a collection angle ranging from 50 to 180 mrad and an acquisition time of 40 µs per pixel. We have used controlled imaging time (within 1 min) under high-magnification STEM node to avoid beam damage on the MoS2 sample.

Density functional calculations. DFT calculations performed with DMOL3 code were used to study the geometric structures and electronic states of different MoS2 monolayers. Geometries were fully optimized within a local spin density approximation with the Perdew–Wang correlational (LDA/PWC) until the convergence criteria for energy, force and displacement were less than 10⁻³ Hartree, 4 x 10⁻⁴ Hartree Å⁻¹ and 5 x 10⁻⁴ Å⁻¹, respectively. All-electron basis sets were utilized for all elements. A Monkhorst–Pack sampling scheme with a 2 x 2 x 1 k-point mesh was used for geometrical optimizations, and a 4 x 8 x 2 k-point mesh was employed for electronic structure calculations. All models for structure optimization were supercells with a sufficiently large size and minimal defect interactions. The supercell sizes of all GB types were the same, as shown in Fig. 4c. For example, the supercell size is 43 x 18 x 20 Å for the 7th GBs.

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Author contributions

T.H.L., D.J.P. and J.Z. contributed to this work in the experimental planning, experimental measurements, data analysis and manuscript preparation. Q.D. contributed to the theoretical calculations. H.K. and G.H.H. synthesized MoS2 on SiO2/Si wafer. S.H.C. and H.Y.J. aided with device fabrication. Y.H.L. contributed to the experimental planning, data analysis and manuscript preparation.

Additional information

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