Defect-Enabled Phase Programming of Transition Metal Dichalcogenide Monolayers

Yang Xia, Joel Berry, and Mikko P. Haataja*

ABSTRACT: The ability to tune the local electronic transport properties of group VI transition metal dichalcogenide (TMD) monolayers by strain-induced structural phase transformations (“phase programming”) has stimulated much interest in the potential applications of such layers as ultrathin programmable and dynamically switchable nanoelectronics components. In this manuscript, we propose a new approach toward controlling TMD monolayer phases by employing macroscopic in-plane strains to amplify heterogeneous strains arising from tailored, spatially extended defects within the monolayer. The efficacy of our proposed approach is demonstrated via numerical simulations of emerging domains localized around arrays of holes, grain boundaries, and compositional heterointerfaces. Quantitative relations between the macroscopic strains required, spatial resolution of domain patterns, and defect configurations are developed. In particular, the introduction of arrays of holes is identified as the most feasible phase programming route.

KEYWORDS: 2D materials, defects, transition metal dichalcogenides, phase engineering, strain engineering

Transition metal dichalcogenide (TMD) monolayers, a class of two-dimensional (2D) materials, have received much attention over the past decade in the pursuit of new technologies including ultrathin, flexible electronic and optoelectronic devices due to their tunable mechanical and electronic properties. Group VI TMDs, the materials of interest in the present work, are comprised of chemical compounds with the formula MX₂, where M denotes a transition metal atom (Mo or W), while X denotes a chalcogen atom (S, Se or Te). In their monolayer form, these materials in fact consist of three covalently bonded atomic layers with two chalcogen atom layers separated by a layer of transition metal atoms. According to first-principles calculations, at low temperatures and under strain-free conditions, the ground state crystal structure of these TMD monolayers (with the exception of WTe₂) is the semiconducting 2H structure. Intriguingly, both experiments and simulations have demonstrated that a phase transformation from the semiconducting 2H to the (semi)metallic 1T structure can be induced upon subjecting the TMD monolayers to in-plane elastic strains. The existence of such a semiconducting-to-metallic transformation is central to the quest to develop Dynamically Programmable Electromechanical 2D Materials (DPE2DMs), in which the local phases—and thus properties—can be manipulated by mechanical deformations.

A challenge in the quest for DPE2DMs, however, resides in how the local elastic strains are introduced. Externally applied macroscopic homogeneous strains alone would lead to the phase transition occurring across the sheet with limited spatial controllability. Thus, to systematically engineer the microstructure of the monolayer, designable heterogeneous local strain fields are required. In our earlier work, such heterogeneous in-plane strains were generated by applying out-of-plane deformations via actuation of multiple indentor tips. In the present work, we take advantage of the fact that tailored, spatially extended defects within the TMD monolayer induce local deformations, and homogeneous macroscopic deformations can be employed to induce phase transitions in the vicinity of such defects by amplifying local strains.

Indeed, it is widely recognized that 2D materials are not structurally perfect, and hence defect engineering has become accepted as a potentially fruitful approach to modulate the properties of these materials. Among all possible defects, holes or notches, which can readily be patterned in MoS₂ monolayers and other 2D materials via electron-beam irradiation or block copolymer lithography, are perhaps the most readily realizable and controllable kinds of defects that can be employed as sources of heterogeneous local strain fields. In addition, recently reported substrate topography-induced grain boundaries in graphene as well as misfit dislocations along compositional (hetero)interfaces within chemically heterogeneous monolayer TMD superlattices,
comprise other classes of spatially extended defects with associated local strains. As we demonstrate below, all of these defects, when subjected to macroscopic homogeneous strain fields, locally nucleate and spatially confine the conducting 1T' domains in a predictable and controllable manner.

Our main findings can be summarized as follows. Numerical simulations and analytical arguments show that arrays of holes, grain boundaries, and heterointerfaces, combined with macroscopic strains, can be used as inputs to generate designable, connected 1T' phase patterns (“channels”) in atomically thin group VI TMD monolayers. The mapping between the input defect configurations and the output phase patterning is quantitatively investigated and characterized. In particular, the creation of arrays of holes provides the most flexibility toward phase programming, while grain boundaries and heterointerfaces can better maintain the electrical and mechanical performance of the monolayer.

THEORETICAL APPROACH

We employ a continuum model based on the phase-field microelasticity (PFM) approach, which builds on and extends previously introduced PFM models for TMD monolayers. Our atomistically-informed PFM model (details of which can be found in the SI) seamlessly incorporates defects and their strain fields, external strains, transformation strains between the 2H and 1T' domains, and spatially varying elastic properties associated with chemically heterogeneous TMD monolayers (implemented via the so-called eigenstrain method), as shown schematically in Figure 1. While the elastic properties of the 1T' domains are known to be anisotropic, in the present work they are treated as isotropic to facilitate large-scale computations; the validity of this assumption will be discussed at the end of the paper. The total (free) energy of the system is expressed as $E_{\text{total}} = E_{\text{el}} + E_{\text{chem}} + E_{\text{sub}}$, where $E_{\text{el}}$, $E_{\text{chem}}$, and $E_{\text{sub}}$ denote elastic, chemical, and substrate interaction energies, respectively. For a given applied strain and defect configuration, $E_{\text{total}}$ is numerically minimized with respect to elastic deformations and 1T' phase order parameters.

PROGRAMMING VIA HOLES

According to the theory of elasticity, regions close to holes or inclusions may be expected to act as preferred nucleation sites of 1T' domains due to strain amplification. As predicted by the
classic Eshelby solution and verified by molecular dynamics simulations of graphene strains reach their maxima along the edges of the inclusions. Inspired by these observations, we consider arrays of circular holes of radii \( R_h \) and separations \( D_h \), respectively, under biaxial macroscopic tensile strain (i.e., \( \varepsilon_{xx} = \varepsilon_{yy} \) and \( \varepsilon_{xy} = 0 \)), to test the feasibility of this phase programming strategy (cf. Figure 1d). Numerical simulations were carried out for a perfectly planar MoTe\(_2\) monolayer. As shown in Figure 2a,b, the 1T’ domains nucleate and grow in the vicinity of the holes and eventually form quasi-1D channels along the arrays of holes once \( \varepsilon_{ij} \) reaches a threshold value \( \varepsilon_{ij}^{cr} \).

Interestingly, when the holes are far apart (\( D_h/R_h \geq 12 \)), 1T’ domains first decorate each hole before connecting across the holes as external strain is increased, whereas when the holes are much closer (\( D_h/R_h \sim 4\text{--}10 \)), connections across the holes form prior to the decoration of the hole edges, thus completing the formation of the channel.

More quantitatively, the dependencies of output resolution (i.e., the minimum channel width \( W_{min} \)) and \( \varepsilon_{ij}^{cr} \) on \( D_h/R_h \) are shown in Figure 2c,d, respectively. It can be seen that there are two regimes for both \( \varepsilon_{ij}^{cr} \) and \( W_{min} \), with \( \varepsilon_{ij}^{cr} \) denoting the strain required for the phase transition in a defect-free system, as indicated by the phase diagram in Figure S1b. For large \( D_h/R_h \), \( \varepsilon_{ij}^{cr} \) and \( W_{min} \) vary linearly with \( D_h/R_h \), respectively, while for small \( D_h/R_h \), both approach constants. As shown in the SI, these asymptotic behaviors can be quantitatively rationalized by analyzing the spatially varying local strains in the vicinity of the holes. We also find that the dependence of \( \varepsilon_{ij}^{cr} \) and \( W_{min} \) on \( D_h/R_h \) is the same for other defect shapes, such as squares (see SI).

Based on the above scaling arguments, we conclude that the spatial resolution \( W_{min} \) and \( \varepsilon_{ij}^{cr} \) are controlled by the ratio \( D_h/R_h \) for...
circular holes. On the one hand, in order to generate high resolution “output” (i.e., small $W_{\text{min}}$) with gentle applied “input” strains, the limit $D/R_h \rightarrow 2$ is required. On the other hand, such closely packed holes may amplify the strains beyond the fracture limit of the monolayer. Therefore, the optimal $D/R_h$ should lie around the transition region $D/R_h \approx 10$ where $W_{\text{min}}$ begins to saturate. To illustrate the efficacy of our proposed approach, we programmed a set of more complex $1T'$ channels by way of arrays of circular holes as depicted in Figure 2e–g.

### PROGRAMMING VIA GRAIN BOUNDARIES

It is now possible to precisely control the location, length, misorientation angle, and orientation of grain boundaries (GBs) in 2D materials via “topological engineering” by growing the monolayer on substrates with nonzero Gaussian curvature. Low-angle GBs (LAGBs), comprised of arrays of edge dislocations, are particularly promising candidates for inducing local phase transitions in light of the heterogeneous strain fields associated with the dislocations. To study the efficacy of LAGBs as a phase programming strategy, we have constructed perfectly planar polycrystalline MoTe$_2$ monolayers with periodic, infinitely long GBs represented by arrays of discrete dislocations as depicted in Figure 1e and further discussed in the SI. Since the Burgers vectors $b$ of possible stable types of dislocations (5/7, 4/6, and 6/8$^{25}$) are all in the zigzag direction(s), the orientation of a GB should always align with one of the armchair ones. Therefore, we will only focus on GBs oriented along one of these armchair directions ($y$-axis). Furthermore, this system can be simplified to a single GB with periodic strain-controlled boundaries in both directions without loss of generality (cf. Figure 1e). Finally, the GB misorientation angle $\theta$ is related to $b$ and dislocation spacing $d$ via $\theta = |b|/d$.

Now, upon applying a tensile strain transverse to the GB ($\epsilon_{xx}$), small $1T'$ domains first form near the dislocation cores, and increasing the strain induces further growth of the domains. According to the MoTe$_2$ phase diagram in the SI, $1T'$ domain variants $p=2$ and 3 are equally favored under such strain. These domains were shown$^7$ to possess habit inclinations parallel to the $y$-axis, which facilitates the formation of straight channels of width $W_{\text{min}}$ centered at the GB beyond a threshold strain $\epsilon_{xx}^{\text{cri}}$. Sequences of such events are depicted in Figure 3a,b.

In more quantitative terms, the dependencies of $\epsilon_{xx}^{\text{cri}}$ and $W_{\text{min}}$ on the misorientation angle $\theta$ are shown in Figure 3c,d, respectively. According to these data, $\epsilon_{xx}^{\text{cri}} \sim \epsilon_{xx}^\theta - \theta$ while $W_{\text{min}}$...
Taken together, the numerical data and corresponding analytical arguments in the SI thus suggest that $W_{\text{min}}$ can in principle be tuned over a relatively wide range, but unlike in the case of arrays of holes, there is only one degree of freedom, $\theta$. Burgers vector $b$ is a fixed intrinsic parameter that cannot be externally controlled, so large $W_{\text{min}}$ requires proportionally small $\theta$ (e.g., $W_{\text{min}} \gtrsim 100$ nm requires $\theta \lesssim 0.1^\circ$). Practical limitations on material control and quality will therefore likely impose $W_{\text{min}} < 100$ nm for GBs.

**PROGRAMMING VIA MISFIT DISLOCATIONS ALONG HETEROINTERFACES**

Finally, in addition to the above-mentioned defects in chemically homogeneous TMD monolayers, chemical heterogeneity by way of distinct chalcogen or transition metal species can be imprinted within 2D heterostructures during monolayer synthesis via precursors, or substrate control. In experiments, both chemically diffuse and sharp heterointerfaces have been observed. In the case of chemically sharp heterointerfaces, the composition-dependent misfit strains can be partially relaxed by a combination of wrinkling and misfit dislocation formation. In particular, we have previously shown that the equilibrium misfit dislocation density $\rho$ is controlled by the chemical species, substrate interaction, and periodicity of the TMD superlattices.

To test the efficacy of such heterointerfaces as a phase programming strategy, simulations were carried out for MoTe$_2$/MoS$_2$ systems with prefixed chemical heterogeneity and misfit dislocations, as illustrated schematically in Figure 1f. Similar to dislocations comprising LAGBs, the Burgers vectors of the misfit dislocations should align with the zigzag directions, and hence the heterointerfaces will be confined to one of these directions ($x$-axis in this case). Now, as can be seen in Figure 4a–c, the 1T' domains first form near the cores of the misfit dislocations in MoTe$_2$ under small applied strain $\varepsilon_{yy}$ in perfectly planar, free-standing, and substrate-supported MoTe$_2$/MoS$_2$ superlattices. The domains subsequently grow and coalesce to form channels along the $x$-direction upon applying a sufficiently large strain. Due to the angle between the habit inclinations and the heterointerfaces, the 1T' channels do not always form straight but “zigzag” instead. Thus, we have not attempted to quantify a minimum channel width for heterointerfaces.

More quantitatively, the dependence of the threshold strain $\varepsilon_{yy}^{\text{th}}$ on misfit dislocation density $\rho$ is shown in Figure 4d for
free-standing, substrate-supported, and perfectly planar superlattices. At large $\rho$, $\epsilon_{yy}^{\text{ref}} \approx 0.6|b| + \text{const.}$ in all such superlattices. As shown in the SI, this is due to the fact that, in this limit, the superlattice remains planar. Upon accounting for the misfit strain relaxed by the dislocations, analysis of the phase diagram for MoTe$_2$ [cf. Figure S1b] immediately leads to the observed linear dependence. On the other hand, for sufficiently wide free-standing or substrate-supported superlattices with large misfit strains and small $\rho$, wrinkling provides an effective means to relax strains within the compressive regions of the monolayer. Thus, channel completion behavior in this limit should be similar to the perfectly planar MoTe$_2$/MoS$_2$ superlattices with $\rho \approx \rho_{\text{max}}$, implying that $\lim_{\rho \to 0} \epsilon_{yy}^{\text{ref}} \approx \epsilon_{yy}^{\text{ref}}(\rho_{\text{max}})$ in agreement with our numerical results.

**CONCLUSION**

In this work, we have proposed a new approach toward reversibly controlling the formation of semimetallic 1T’ channels within atomically thin group VI TMD monolayers via a combination of strain-inducing defects and applied in-plane strains. Both geometric (arrays of holes) and material defects (low angle grain boundaries and misfitting heterointerfaces) have been considered and investigated via a combination of numerical simulations and analytical arguments. Each phase programming strategy has its benefits and drawbacks, as will be discussed next.

Arrays of holes were found to be the most promising phase programming strategy. In particular, the isotropic amplification of the applied strains provides flexibility as far as the orientations and feature sizes of the target output patterns are concerned. The spatial resolution of the phase domains and critical strains were found to only depend on the relative distance between the holes, and the optimal density of circular holes was estimated for practical applications. It was also demonstrated that rather complex phase patterns can be programmed with this approach. On the other hand, perforation of the monolayer with holes will negatively affect the overall mechanical properties of the monolayer and may even compromise its mechanical integrity. In addition, since all 1T’ variants are equivalently favored under isotropic strains, variant interfaces will be always present. Such interfaces will inevitably have some influence on the charge transport properties of the monolayer.

In the case of low angle grain boundaries (LAGBs), upon applying strains normal to their orientation in polycrystalline TMDs, it was found that formation of 1T’ phase channels could be induced along the GBs, with critical strains and channel widths depending only on the GB misorientation angle. On the plus side, a polycrystalline TMD monolayer will be able to retain its mechanical integrity much better than a perforated one. On the negative side, the restriction of the channels running along the LAGBs will limit this phase programming strategy to relatively simple patterns.

Finally, in the case of heterointerfaces, when uniaxial strains were applied parallel to the heterointerface, zigzag channels formed beyond a threshold strain whose magnitude depended on both the misfit dislocation density and substrate interaction. In comparison with arrays of holes and LAGBs, formation of a single 1T’ variant is expected, thus eliminating any variant interfaces which may negatively affect electronic transport properties. On the other hand, the channels will always align with the heterointerfaces, which restricts the range of possible phase patterns achievable with this programming strategy.

It should be noted that all of the quantitative analysis and simulations reported in this work focus on the formation of 1T’ channels in MoTe$_2$, in light of the fact that it possesses the smallest bulk transformation—and hence threshold—strain $\epsilon^* \sim 0.05$ among all group VI TMDs at $T = 0$ K. For other group VI TMDs, transformation strains are higher with $\epsilon^* \sim 0.2$ (with the exception of WTe$_2$, which has a stable 1T’ structure in the absence of external strain). However, $\epsilon^*$ can be tuned by alloying the transition metal or chalcogen elements to facilitate the proposed phase programming protocols under even smaller applied strains. In fact, we expect the threshold strains required for all of the aforementioned phase programming strategies to be significantly smaller at finite temperatures. Indeed, it has been reported experimentally that, at room temperature, $\epsilon^*$ for MoTe$_2$ thin films can be as small as 0.002. With such small threshold strains, maximum strains in the percolated thin film will be approximately 0.005 (cf. Figure 2a,b). Therefore, phase programming via arrays of holes should be possible without any compromises in the mechanical integrity of the monolayer, given the fracture strain of 1T’-MoTe$_2$ of $\sim 0.03$ at 250 K. We also note that homogeneous macroscopic tensile strains as large as $\sim 0.01$ can be readily applied to TMD mono- and multilayers by depositing them on polymeric substrates and thus subjecting them to a temperature change or employing a two-point bending apparatus.

Our numerical studies were conducted under the assumption of identical stiffness tensors for the 2H and 1T’ phases to facilitate large-scale computations. To test the validity of this assumption, we implemented the anisotropic 1T’ stiffness tensor in the case of perfectly planar MoTe$_2$/MoS$_2$ superlattices, for which a single 1T’ variant emerges via the phase programming protocol. Our numerical results (cf. Figure S4 in the SI) demonstrate that the threshold applied strain $\epsilon_{xx}^*$ does not differ significantly from the homogeneous and isotropic case. This is reasonable as threshold strains are mainly determined by the 2H-1T’ phase diagram in strain space, which is not significantly affected by elastic anisotropy of the 1T’ domains (cf. Figure S1 in the SI).

Finally, regarding the reversibility of the strain-induced transformations at the heart of our proposed phase programming protocols, we note that some hysteresis was observed in the simulations where the applied strain was ramped up past the threshold strain and then ramped back down. The magnitude of the hysteresis, however, was comparable to the estimated error bars, suggesting that the protocols can be applied repeatedly and reversibly. It is possible that, during cyclic transformations between the 2H and 1T’ structures, defects not accounted for in our phase-field modeling framework (e.g., 1D twin boundaries) may accumulate. Careful experiments and/or more microscopic modeling approaches are required to conclusively address this important question.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.1c00742.

Details of the phase-field model, analysis, and numerical calculations reported in the main text (PDF).
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Supplementary Information for “Defect-Enabled Phase Programming of Transition Metal Dichalcogenide Monolayers”

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1 Governing equations for chemically heterogeneous TMD monolayers

The total (free) energy of the system $E_{total}$ in our phase field microelasticity (PFM) model is written

$$E_{total} = E_{el} + E_{chem} + E_{sub},$$  \hspace{1cm} \text{[1]}

where $E_{el}$, $E_{chem}$ and $E_{sub}$ denote elastic, chemical and substrate interaction energies, respectively. The elastic energy $E_{el}$ is written $E_{el} = E_{strain} + E_{bend}$, where $E_{strain}$ and $E_{bend}$ denote in-plane deformation and out-of-plane bending deformation energies, respectively. Below, we will discuss each contribution to $E_{total}$ separately.

1.1 In-plane elastic energy

To describe chemical and elastic heterogeneities, two order parameters $\phi(r)$ and $\psi(r)$ are used to represent different chemical components [$\phi(r) = 0$, say, for MoS$_2$ and 1 for MoTe$_2$] and free
boundaries \[\psi(r) = 0(1)\] in regions outside (inside) the monolayer, respectively, such that different bulk energies, lattice constants, and elastic constants can be assigned to chemically or structurally distinct TMDs and perforations.

As stated in the main text, the in-plane strain energy in our phase field model (PFM) is expressed as

\[
E_{\text{strain}} = \frac{1}{2} \int_A C_{ijkl}(r)[(\epsilon_{ij}(r) - \epsilon^{\text{mis}}_{ij}(r)) \times (\epsilon_{kl}(r) - \epsilon^{\text{mis}}_{kl}(r))]d^2r, \tag{2}
\]

where \(r = [x, y]^T\), \(C_{ijkl}(r) = \{[1-\phi(r)][(1-\sum_p \eta^2_p(r))C_{ijkl}^{(1)} + \sum_p \eta^2_p(r)C_{ijkl}^{(1,p)} + \phi(r)[(1-\sum_p \eta^2_p(r))C_{ijkl}^{(2)} + \sum_p \eta^2_p(r)C_{ijkl}^{(2,p)}]\}\psi(r)\) denotes the local stiffness tensor, \(\epsilon_{ij}(r) = \bar{\epsilon}_{ij} + \frac{1}{2}(\frac{\partial u_i(r)}{\partial r_j} + \frac{\partial u_j(r)}{\partial r_i} + \frac{\partial \omega(r)}{\partial r_i} - \frac{\partial \omega(r)}{\partial r_j})\) is the strain tensor with \(\bar{\epsilon}_{ij}\) denoting the applied strain, and \(\epsilon^{\text{mis}}_{ij}(r) = \epsilon^{H}_{ij}(r) + \sum_{p=1}^v \eta^2_p(r)\epsilon^{T}_p(p, r) + \epsilon^{\text{dis}}_{ij}(r))\) is a generalized misfit strain tensor arising from lattice mismatch, phase transformation and presence of dislocations. Furthermore, \(\epsilon^{H}_{ij}(r)\) denotes the misfit strain tensor arising from lattice mismatch between distinct group VI TMDs in the 2H phase such that \(\epsilon^{H}_{xx}(r) = \phi(r)\frac{a_1^H-a_1^H}{a_1^H}\), \(\epsilon^{H}_{yy}(r) = \phi(r)\frac{b_2^H-b_2^H}{b_2^H}\), and \(\epsilon^{H}_{xy}(r) = 0\). In addition, \(\epsilon^{T}_p(p, r) = (1-\phi(r))\epsilon^{T(1)}_p(p) + \phi(r)\epsilon^{T(2)}_p(p)\) denotes the transformation strain tensor for 1T variant \(p\), which can be written as \(\epsilon^{T(k)}_p(p) = R[\theta_p]^{(k)}\epsilon^{(k)}_p R^T[\theta_p]\), with \(R[\theta_p]\) denoting the 2D rotation matrix and \(\epsilon^{T(k)}_{xx} = \frac{a_1^H-a_1^H}{a_1^H}, \epsilon^{T(k)}_{yy} = \frac{b_2^H-b_2^H}{b_2^H}, \epsilon^{T(k)}_{xy} = 0\). Here \(\theta_1 = 0, \theta_2 = \frac{2\pi}{3}\), and \(\theta_3 = \frac{-2\pi}{3}\) are the rotation angles corresponding to each 1T variant orientation. \(\epsilon^{\text{dis}}_{ij}(r)\) in turn denotes the strain due to dislocations, which is defined in Eq. [4] below. \(C_{ijkl}^{(1)}\) and \(C_{ijkl}^{(2)}\) denote the stiffness tensors of TMDs 1 and 2 in the 2H crystal structure, while \(C_{ijkl}^{(1,p)}\) and \(C_{ijkl}^{(2,p)}\) denote the stiffness tensors of the \(p\)th 1T variant. \(a_1^H, a_2^H, b_1^H, b_2^H, a_1^{T'}, a_2^{T'}, b_1^{T'}, \) and \(b_2^{T'}\) are the lattice constants of TMDs 1 and 2 in 2H and 1T crystal structures, respectively. \(\bar{\epsilon}_{ij}\) denotes the applied macroscopic strain, \(u_i(r)\) is the in-plane displacement, and \(\omega(r)\) represents the out-of-plane displacement.

### 1.2 Dislocations in PFM model

Dislocation loops in 3D phase field models can be considered as misfitting platelets associated with the slipped regions in the crystal.\[11\] Upon introducing order parameters \(\zeta_q(r)\), where the index \(q\) refers to the type of dislocation (e.g., its Burgers vector and slip plane normal), the effective misfit strain carried by dislocations can be expressed as

\[
\epsilon^{\text{dis}}_{ij}(r) = \sum_q \epsilon^{\text{dis}}_{ij}(q)\zeta_q(r); \tag{3}
\]

\[
\epsilon^{\text{dis}}_{ij}(q) = \frac{b_i(q)n_j(q) + b_j(q)n_i(q)}{2d(q)}, \tag{4}
\]
where $\mathbf{b}(q)$, $\mathbf{n}(q)$, and $d(q)$ denote the Burgers vector, slip plane normal, and interplanar distance of $q$-type dislocations, respectively. In our quasi-2D model, the dislocation loops are transformed into dislocation segments, such that an edge dislocation dipole can be described by a line segment of non-zero $\zeta_q(r)$ with the two dislocations located at the ends of the segment. We will employ this formulation to incorporate misfit dislocations along heterointerfaces in binary TMD superlattices.

To create infinitely long 1D arrays of dislocations (corresponding to low angle grain boundaries), on the other hand, the above method requires overlapping dislocation loops (lines) to represent the displacements induced by the dislocations. Thus, it becomes impossible to make the system periodic as the displacement is not periodic along the GB. As a convenient alternative, the analytical solution for the strain field induced by a periodic 1D dislocation array\cite{2} is employed to determine $\epsilon^{\text{dis}}_{ij}(r)$. That is, the stress field induced by a line of periodic edge dislocations along $y$ is given by

$$
\begin{align*}
\sigma^{\text{dis}}_{xx}(r) &= -\frac{\mu |\mathbf{b}|}{2d(1-\nu)} \sin \frac{2\pi y}{d} \left(\cosh \frac{2\pi x}{d} - \cos \frac{2\pi y}{d} + \frac{2\pi x}{d} \sinh \frac{2\pi x}{d}\right); \\
\sigma^{\text{dis}}_{yy}(r) &= -\frac{\mu |\mathbf{b}|}{2d(1-\nu)} \sin \frac{2\pi y}{d} \left(\cosh \frac{2\pi x}{d} - \cos \frac{2\pi y}{d} - \frac{2\pi x}{d} \sinh \frac{2\pi x}{d}\right); \\
\sigma^{\text{dis}}_{xy}(r) &= \frac{2\pi \mu |\mathbf{b}| x}{d(1-\nu)} \exp \left(-\frac{2\pi x}{d} \cos \frac{2\pi y}{d}\right),
\end{align*}
$$

[5]

where $d$ denotes the distance between the dislocations, $\mathbf{b}$ is the Burgers vector, and $\mu$ is the shear modulus. The misfit strain tensor due to dislocations $\epsilon^{\text{dis}}_{ij}(r)$ can then be evaluated from $\epsilon^{\text{dis}}_{ij}(r) = S_{ijkl}(r)\sigma^{\text{dis}}_{kl}(r)$, where $S_{ijkl}(r)$ denotes the matrix inverse of $C_{ijkl}(r)$.

1.3 Bending energy

The bending energy term $E_{\text{bend}}$ is expressed as

$$
E_{\text{bend}} = \int_A \kappa(r) \left[ \left( \frac{\partial^2 \omega(r)}{\partial x^2} + \frac{\partial^2 \omega(r)}{\partial y^2} \right)^2 + 2(1-\nu(r)) \times \left\{ \left( \frac{\partial^2 \omega(r)}{\partial x \partial y} \right)^2 - \frac{\partial^2 \omega(r)}{\partial x^2} \frac{\partial^2 \omega(r)}{\partial y^2} \right\} \right] d^2r, \quad [6]
$$

where $\kappa(r) = \frac{Y(r)h^3}{24(1-\nu(r))^2}$ denotes the local bending rigidity. Furthermore, $\nu(r) = \{1 - \phi(r)\nu_1 + \phi(r)\nu_2\} \psi(r)$ is the local Poisson’s ratio, $Y(r) = \{1 - \phi(r)\} Y_1 + \phi(r) Y_2 \psi(r)$ is the local Young’s modulus, and $h$ is the thickness of the sheet. Finally, $\nu_1, \nu_2, Y_1$ and $Y_2$ denote the Poisson’s ratios and Young’s moduli of TMDs 1 and 2, respectively.
1.4 Chemical energy

The chemical energy term $E_{\text{chem}}$ accounts for the relative stability of the 2H and 1T' phases under strain-free conditions, and consists of interfacial ($E_{\text{inter}}$) and bulk free energy ($E_{\text{bulk}}$) components, which we will discuss next.

1.4.1 Interfacial energy

For simplicity, we first assume the length scale of our simulation system is sufficiently large such that the anisotropy of the interfacial energy can be ignored. Under this assumption, $E_{\text{inter}}$ can be expressed as

$$E_{\text{inter}} = \int_A \frac{1}{2} \sum_p |\beta \nabla \eta_p(r)|^2 d^2 r. \quad [7]$$

In cases where an anisotropy of interfacial energy needs to be included, it can be incorporated by introducing a dependence on the interface orientations to the coefficient $\beta(r)$ as in Ref.\textsuperscript{3}

1.4.2 Bulk free energy

In contrast to existing 2D PFM models for hexagonal to orthorhombic transformations\textsuperscript{3,4} an eighth order polynomial is employed in the present work to construct the bulk free energy. This facilitates the adjustment of energy differences and barriers for the structural transformations of different TMDs. More specifically,

$$E_{\text{bulk}} = \int_A \left[ \frac{a(r)}{2} \sum_p \eta_p^2(r) - \frac{b(r)}{4} \sum_p \eta_p^4(r) + \frac{c(r)}{6} \sum_p \eta_p^6(r) + \frac{d(r)}{8} \left( \sum_p \eta_p^2 \right)^4 \right] d^2 r, \quad [8]$$

where $a(r) = \{[1-\phi(r)]a_1+\phi(r)a_2\} \psi(r)$, $b(r) = \{[1-\phi(r)]b_1+\phi(r)b_2\} \psi(r)$, $c(r) = \{[1-\phi(r)]c_1+\phi(r)c_2\} \psi(r)$, and $d(r) = \{[1-\phi(r)]d_1+\phi(r)d_2\} \psi(r)$. Here, $a_1, a_2, b_1, b_2, c_1, c_2, d_1$ and $d_2$ denote adjustable parameters that set the chemical driving force for the structural transformations of TMDs 1 and 2, respectively.

It should be noted that three order parameters $\eta_p(r)$ ($p = 1, 2$ or 3) are employed to distinguish six variants of the 1T' structure, with $\eta_p(r) = +1(-1)$ denoting the $p^+(p^-)$ variant. Following our earlier work\textsuperscript{3,4} we do not distinguish between the $p_A$ and $p_B$ variants, and hence the 12 possible 1T' variants are captured by the three order parameters [see Figs. 1 a and b in the main text]. Results from existing first principles calculations are employed to fit and constrain the model parameters, as will be discussed below.
Following the strategy of Berry et al.\textsuperscript{3,4} the transformation strains $\bar{\epsilon}_{ij}^{(k)}(p)$ for the phase transformations obtained from first principles calculations\textsuperscript{3} are incorporated in $E_{\text{chem}}$ as follows. Values of $a_k$, $b_k$, $c_k$ and $d_k$ are chosen such that $E_{\text{total}}$ has identical local minima at $\eta_0(r) = 0$ and $\pm 1$ under $\bar{\epsilon}_{ij} = \bar{\epsilon}_{ij}^{(k)}(p)$, with $\bar{\epsilon}_{ij}^{(k)}(p)$ denoting the strains required for the 2H $\rightarrow$ 1T\textsuperscript{'} transformation of a TMD indexed by $k$. In practice, we only equate $E_{\text{total}}(\eta_1 = 0)$ and $E_{\text{total}}(\eta_1 = \pm 1)$ at $\epsilon_{yy} = \epsilon_{yy}^{(k)}$ [cf. Fig. S1 (a)]. Once the parameters $a_k$, $b_k$, $c_k$ and $d_k$ have been fixed, a phase diagram for the $k^{th}$ TMD can be readily constructed, as shown in Figs. S1 (b)-(d). In Fig. S1 (b), the elastic stiffness tensors are taken to be homogeneous and isotropic for both 2H and 1T\textsuperscript{'} structures. In this case, the phase boundaries between the 2H phase and all three 1T\textsuperscript{'} variants can be readily determined by computing and comparing the bulk free energies of all structures as functions of the applied strain. However, in order to better reproduce the phase diagram constructed from first principles calculations,\textsuperscript{5} heterogeneous and anisotropic stiffness tensors for 1T\textsuperscript{'} structures\textsuperscript{6,7} should be considered. When such a stiffness tensor is employed, the phase diagram for a single variant in Fig. S1 (c) emerges. [If an isotropic and homogeneous stiffness tensor for the 1T\textsuperscript{'} phase is employed instead, the phase boundary is given by a straight line as shown in the figure.] To account for the other two 1T\textsuperscript{'} variants, anisotropic stiffness tensors and transformation strain tensors are rotated, and the bulk free energies are again evaluated as functions of strain. The results of such computations are recapitulated in the phase diagram in Fig. S1 (d). Importantly, when all three 1T\textsuperscript{'} variants are considered, assuming elastic isotropy for both phases has very little quantitative effect on the 2H/1T\textsuperscript{'} phase boundary for MoTe\textsubscript{2}. Finally, it is interesting to note that there is a crossing of phase boundaries for different 1T\textsuperscript{'} phases under large applied strains in Fig. S1 (d). Intuitively, this can be explained by the anisotropy of the stiffness tensor, which selects certain variants under large, effectively uniaxial strains based on the “softer” elastic modulus along that direction.

1.5 Substrate interaction

A simple phenomenological form for the substrate interaction energy $E_{\text{sub}}$ is employed in the present work. A harmonic form is assumed, such that

$$E_{\text{sub}} = k_{\text{eff}} \int_A \left[ \omega(r) - \omega_{eq}(r) \right]^2 d^2r. \quad [9]$$

Here, $k_{\text{eff}}$ denotes an effective substrate interaction coefficient, while $\omega_{eq}(r)$ denotes the equilibrium distance between the monolayer and the planar substrate. Without loss of generality, we set $\omega_{eq}(r) = 0$, such that $E_{\text{sub}} = k_{\text{eff}} \int_A \omega^2(r) d^2r$. When $k_{\text{eff}} = 0$, the TMD monolayer is free-standing, while in the limit $k_{\text{eff}} \rightarrow \infty$, all nonzero out-of-plane deflections are eliminated, resulting in a perfectly planar monolayer. Finally, for substrate supported monolayers, a finite $k_{\text{eff}} > 0$ is
Figure S 1: (a): $E_{total}$ versus $\eta_p$ under different $\tau_{yy}$ for MoTe$_2$ with $\tau_{yy}^* = 0.05$. (b): Phase diagram for MoTe$_2$ in ($\tau_{xx}$, $\tau_{yy}$) space, with the stable phases indicated by color. Specifically, the gray region refers to 2H, blue region refers to one 1T' variant (0°), and orange region refers to coexistence of two 1T' variants (120° and 240°). Points along the 2H/1T' phase boundaries correspond to homogeneous applied strains ($\epsilon_{xx}^*$, $\epsilon_{yy}^*$) at which the phase transformation is triggered in a macroscopic, homogeneous system. (c) and (d): Phase diagram for MoTe$_2$ in ($\tau_{xx}$, $\tau_{yy}$) space with heterogeneous and anisotropic stiffness tensors. Only one 1T' variant (0°) is shown in (c) while all three variants are taken into account in (d). The red dashed line in (c) indicates the phase boundary obtained with a homogeneous and isotropic 1T' stiffness tensor.
employed. We expect that the above choice for $E_{\text{sub}}$ quantitatively captures the leading order behavior of substrate-supported TMDs with finite out-of-plane deflections.

1.6 Total energy in dimensionless form

To facilitate the numerical simulations, a dimensionless form of the total energy of the system is employed by introducing the scaled quantities $\tilde{E}_{\text{total}} = E_{\text{total}} Y_1^{-1}|b|^3$, $\tilde{E}_{\text{el}} = E_{\text{el}} Y_1^{-1}|b|^3$, $\tilde{E}_{\text{chem}} = E_{\text{chem}} Y_1^{-1}|b|^3$, $\tilde{E}_{\text{sub}} = E_{\text{sub}} Y_1^{-1}|b|^3$, $\tilde{E}_{\text{strain}} = E_{\text{strain}} Y_1^{-1}|b|^3$, $\tilde{E}_{\text{bend}} = E_{\text{bend}} Y_1^{-1}|b|^3$, $\bar{r} = r|b|$, $\bar{\kappa} = \kappa Y_1^{-1}|b|^3$, $\bar{\omega} = \omega|b|$, $\bar{\beta} = \beta Y_1^{-1}|b|^3$, $\tilde{\eta} = a Y_1^{-1}|b|$, $\tilde{\beta} = b Y_1^{-1}|b|$, $\tilde{\upsilon} = c Y_1^{-1}|b|$, $\tilde{d} = d Y_1^{-1}|b|$, $\tilde{k}_{\text{eff}} = k_{\text{eff}}|b|$, and $\tilde{\omega}_{\text{eq}} = \omega_{\text{eq}}|b|$. Now, the dimensionless total energy can be written as

$$\tilde{E}_{\text{total}} = \tilde{E}_{\text{el}} + \tilde{E}_{\text{chem}} + \tilde{E}_{\text{sub}}, \quad [10]$$

where

$$\begin{align*}
\tilde{E}_{\text{el}} &= \tilde{E}_{\text{strain}} + \tilde{E}_{\text{bend}}; \\
\tilde{E}_{\text{strain}} &= \int_A \frac{1}{2} \tilde{C}_{ijkl}(\bar{r})[\epsilon_{ij}(\bar{r}) - \epsilon_{ij}^{\text{mis}}(\bar{r})][\epsilon_{kl}(\bar{r}) - \epsilon_{kl}^{\text{mis}}(\bar{r})] d^2\bar{r}; \\
\tilde{E}_{\text{bend}} &= \int_A \tilde{\kappa}(\bar{r}) \left\{ \left( \tilde{\nabla}^2 \tilde{\omega}(\bar{r}) \right)^2 + [1 - \nu(\bar{r})] \left[ \left( \frac{\partial^2 \tilde{\omega}(\bar{r})}{\partial x^2} - \frac{\partial^2 \tilde{\omega}(\bar{r})}{\partial y^2} \right) \right] \right\} d^2\bar{r}; \\
\tilde{E}_{\text{chem}} &= \int_A \left[ \frac{1}{2} \sum_p \tilde{\beta}(\bar{r}) \nabla \eta_p(\bar{r}) \right]^2 \\
&+ \frac{\tilde{a}(\bar{r})}{2} \sum_p \eta_p^2(\bar{r}) - \frac{\tilde{b}(\bar{r})}{4} \sum_p \eta_p^4(\bar{r}) + \frac{\tilde{c}(\bar{r})}{6} \sum_p \eta_p^6(\bar{r}) + \frac{\tilde{d}(\bar{r})}{8} \left( \sum_p \eta_p^4 \right)^2 \right] d^2\bar{r}; \\
\tilde{E}_{\text{sub}} &= \tilde{k}_{\text{eff}} \int_A \left[ \tilde{\omega}(\bar{r}) - \tilde{\omega}_{\text{sub}} \right]^2 d^2\bar{r}.
\end{align*}$$

Assuming a fixed spatial distribution of chemical components, the mechanical and structural equilibrium state of the TMD monolayer can be computed by minimizing Eq. [10] with respect to $u_i$, $\eta_p$, and $\omega$, subject to certain boundary conditions (e.g., strain-controlled or stress-free).

1.7 Modified eigenstrain method for solving mechanical equilibrium equations

An eigenstrain method developed by Wang et al.\(^8\) has been adapted to relax the in-plane displacements efficiently in systems with free boundaries (corresponding to stress-free boundary conditions). A few modifications to the original method have been made due to the presence of out-of-plane displacements in the present system. First, the inhomogeneous part of stiffness tensor
is defined as \( \Delta \tilde{C}_{ijkl}(\tilde{r}) = \tilde{C}^{(1)}_{ijkl} - \tilde{C}_{ijkl}(\tilde{r}) \) with \( \tilde{C}^{(1)}_{ijkl} = \frac{C^{(1)}_{ijkl}}{V_{ijkl}} \). A new effective eigenstrain tensor \( \epsilon_{ij}^{0}(\tilde{r}) \) is then introduced such that

\[
\tilde{C}_{ijkl}\epsilon_{kl}(\tilde{r}) = \tilde{C}^{(1)}_{ijkl}\epsilon_{kl}^{0}(\tilde{r}) + \Delta \tilde{C}_{ijkl}(\tilde{r})[\epsilon_{kl}(\tilde{r}) - \epsilon_{kl}^{mis}(\tilde{r})],
\]

where \( \tilde{C}^{(1)}_{ijkl} = \frac{C^{(1)}_{ijkl}}{C^{(0)}_{ijkl}} \), \( \Delta \tilde{C}_{ijkl}(\tilde{r}) = \tilde{C}^{(1)}_{ijkl} - \tilde{C}_{ijkl}(\tilde{r}) \) is the difference between the homogeneous and heterogeneous stiffness tensors. With the above definition of \( \epsilon_{ij}^{0}(\tilde{r}) \), the strain energy of an inhomogeneous system can be related to a homogeneous one:

\[
\tilde{E}_{strain} = \frac{1}{2} \int_{A} \tilde{C}^{(1)}_{ijkl}[\epsilon_{ij}(\tilde{r}) - \epsilon_{ij}^{0}(\tilde{r})][\epsilon_{kl}(\tilde{r}) - \epsilon_{kl}^{0}(\tilde{r})]d^{2}\tilde{r} + \frac{1}{2} \int_{A} [\tilde{C}^{(1)}_{ijmn}\Delta \tilde{S}_{mnpq}(\tilde{r})\tilde{C}^{(1)}_{pqkl}][\epsilon_{ij}^{0}(\tilde{r}) - \epsilon_{ij}^{mis}(\tilde{r})][\epsilon_{kl}^{0}(\tilde{r}) - \epsilon_{kl}^{mis}(\tilde{r})]d^{2}\tilde{r},
\]

where \( \Delta S_{ijkl}(\tilde{r}) \) is the inverse tensor of \( \Delta C_{ijkl}(\tilde{r}) \).

The first integral in Eq. [12] has the form of a strain energy in an elastically homogeneous system with an a priori unknown effective misfit strain \( \epsilon_{ij}^{0}(\tilde{r}) \); this strain is obtained numerically via another relaxation scheme as will be discussed shortly. Note that the second integral in Eq. [12] is independent of the in-plane strain tensor \( \epsilon_{ij}(\tilde{r}) \), and thus it can be evaluated as in an elastically homogeneous system following the approach of Khachaturyan once \( \epsilon_{ij}^{0}(\tilde{r}) \) has been determined.

### 1.8 Mechanical and microstructural equilibrium

Depending on the type of boundary conditions applied in the simulations, the macroscopic strain \( \epsilon_{ij} \) can be either prefixed (for strain controlled boundary conditions), or computed at mechanical equilibrium (for stress-free scenarios). The equations of mechanical equilibrium for the macroscopic strain \( \epsilon_{ij} \) (stress controlled), eigenstrain \( \epsilon_{ij}^{0}(\tilde{r}) \), in-plane displacements \( \tilde{u}_{i}(\tilde{r}) \), out-of-plane displacements \( \tilde{\omega}(\tilde{r}) \), and structural equilibrium for order parameters \( \eta_{p}(\tilde{r}) \) are given by

\[
\tilde{\epsilon}_{ij} = \frac{1}{A} \int_{A} \left[ \epsilon_{ij}^{0}(\tilde{r}) - \frac{1}{2} \frac{\partial \tilde{\omega}}{\partial \tilde{r}_{i}} \frac{\partial \tilde{\omega}}{\partial \tilde{r}_{j}} \right] d^{2}\tilde{r};
\]

\[
\frac{\delta \tilde{E}_{total}}{\delta \epsilon_{ij}^{0}(\tilde{r})} = \frac{\delta \tilde{E}_{strain}}{\delta \epsilon_{ij}^{0}(\tilde{r})} = \tilde{C}^{(1)}_{ijkl}\Delta \tilde{S}_{klmn}(\tilde{r})\tilde{C}^{(1)}_{mnst}[\epsilon_{st}^{0}(\tilde{r}) - \epsilon_{st}^{mis}(\tilde{r})] - \tilde{C}^{(1)}_{ijkl}[\epsilon_{kl}(\tilde{r}) - \epsilon_{kl}^{mis}(\tilde{r})] = 0;
\]

\[
\tilde{u}_{i}(\tilde{k}) = -iG_{ij}(\tilde{k})\tilde{C}^{(1)}_{ijkl}\epsilon_{lm}^{0}(\tilde{k})k_{k} + G_{ij}(\tilde{k})\tilde{C}^{(1)}_{ijkl}\tilde{\eta}_{klm}(\tilde{k});
\]
In the system are taken to be stationary, implying that the order parameters are prefixed and do not evolve during the numerical relaxation process. Similarly, all dislocations \( \tilde{\epsilon}_{ij}(\vec{r}) \) with \( 
abla^{2} \tilde{\epsilon}_{ij}(\vec{r}) = 0 \), and 

\[
\delta \tilde{E}_{\text{total}}(\omega(\vec{r})) = -2 \frac{\partial}{\partial \tilde{r}_{i}} \left\{ C_{ijkl}(\tilde{\epsilon}) \frac{\partial \omega}{\partial \tilde{r}_{j}} [\epsilon_{kl}(\tilde{\epsilon}) - \epsilon_{kl}^{\text{mis}}(\tilde{\epsilon})] \right\}
\]

\[
+ \frac{\partial^{2}}{\partial \tilde{x}^{2}}[\kappa(\tilde{\epsilon}) \frac{\partial^{2} \omega}{\partial \tilde{x}^{2}} + \kappa(\tilde{\epsilon}) \frac{\partial^{2} \omega}{\partial \tilde{y}^{2}}] + \frac{\partial^{2}}{\partial \tilde{y}^{2}}[\kappa(\tilde{\epsilon}) \frac{\partial^{2} \omega}{\partial \tilde{y}^{2}}] + \tilde{\kappa}(\tilde{\epsilon}) \frac{\partial^{2} \omega}{\partial \tilde{y}^{2}}]
\]

\[
+ 2 \frac{\partial^{2}}{\partial \tilde{x} \partial \tilde{y}}[\kappa(\tilde{\epsilon})(1 - \nu(\tilde{\epsilon})) \frac{\partial^{2} \omega}{\partial \tilde{x} \partial \tilde{y}}] = 0;
\]  

\[\tag{16}\]

\[\delta \tilde{E}_{\text{total}}(\eta(\vec{r})) = 2 \eta_{q}(\tilde{\epsilon}) \left\{ [1 - \phi(\tilde{\epsilon})] \tilde{C}_{ijkl}^{(1)} - \tilde{C}_{ijkl}^{(2)} \right\} + \phi(\tilde{\epsilon}) \left\{ \tilde{C}_{ijkl}^{(1)} - \tilde{C}_{ijkl}^{(2)} \right\} \psi(\tilde{\epsilon}) [\epsilon_{ij}(\tilde{\epsilon}) - \epsilon_{ij}^{\text{mis}}(\tilde{\epsilon})]
\]

\[\times \left\{ [\epsilon_{kl}(\tilde{\epsilon}) - \epsilon_{kl}^{\text{mis}}(\tilde{\epsilon})] - 2 \eta_{q}(\tilde{\epsilon}) \tilde{C}_{ijkl}(\tilde{\epsilon}) \epsilon_{ij}^{T}(q, \tilde{\epsilon}) \left[ \epsilon_{kl}(\tilde{\epsilon}) - \epsilon_{kl}^{\text{mis}}(\tilde{\epsilon}) \right] \right\}
\]

\[\times \beta(\tilde{\epsilon}) \nabla^{2} \eta_{q}(\tilde{\epsilon}) + \tilde{a}(\tilde{\epsilon}) \eta_{q}(\tilde{\epsilon}) - \tilde{b}(\tilde{\epsilon}) \eta_{q}^{3}(\tilde{\epsilon}) + \tilde{c}(\tilde{\epsilon}) \eta_{q}^{5}(\tilde{\epsilon})
\]

\[+ \tilde{d}(\tilde{\epsilon}) \eta_{q}(\tilde{\epsilon}) \left[ \sum_{p} n_{p}^{2}(\tilde{\epsilon}) \right]^{3} = 0;
\]  

\[\tag{17}\]

\[\tag{18}\]

with \( \tilde{\epsilon}_{ij}(\vec{k}) \) the Fourier transform of \( \tilde{\epsilon}_{ij}(\vec{r}) \), \( \Omega_{ij}(\vec{n})^{-1} = \tilde{C}_{ijkl}^{T} n_{k} n_{l} \), \( \vec{n} = \vec{k}/|\vec{k}| \), \( \tilde{\eta}(\vec{k}) \) the Fourier transform of \( \tilde{\eta}(\vec{r}) \), \( G_{ij}(\vec{k}) = \Omega_{ij}(\vec{n})/k^{2} \), and

\[
\tilde{N}_{kln}(\vec{k}) = \left\{ \frac{\partial \omega}{\partial \tilde{r}_{k}} \frac{\partial^{2} \omega}{\partial \tilde{r}_{l} \partial \tilde{r}_{m}} \right\}_{\vec{k}}
\]

\[\tag{19}\]

where \( \{ \}_{\vec{k}} \) denotes the Fourier transform.

\section{Numerical relaxation method}

In order to solve the above equations for mechanical and structural equilibrium, we note that the analytical solution of \( \tilde{u}_{i}(\vec{r}) \) is explicitly given in Eq. \( \ref{15} \). However, Eqs. \( \ref{14}, \ref{16} \) and \( \ref{18} \) are either non-linear partial differential equations (PDEs) or linear ones with non-constant coefficients. To solve them expeditiously, the following iterative numerical scheme was employed:

1. Assuming that the diffusion of chemical species is negligible, the order parameters \( \phi(\tilde{\epsilon}) \) and \( \psi(\tilde{\epsilon}) \) are prefixed and do not evolve during the numerical relaxation process. Similarly, all dislocations in the system are taken to be stationary, implying that the order parameters \( \zeta_{q}(\tilde{\epsilon}) \) are also non-evolving.

2. The macroscopic strain \( \tilde{\epsilon}_{ij} \) can either be prefixed (strain controlled boundary condition) or computed from Eq. \( \ref{13} \) for stress-free boundary conditions.
3. Iterate \( \epsilon_{ij}^0(\vec{r}) \) toward the steady-state given by Eq. \([21]\) via
\[
\frac{\partial^2 \epsilon_{ij}^0(\vec{r}, \tilde{t})}{\partial \tilde{t}^2} + (\gamma_0, \varepsilon^0 - \gamma_{1, \varepsilon^0} \nabla^2) \frac{\partial \epsilon_{ij}^0(\vec{r}, \tilde{t})}{\partial \tilde{t}} = -\alpha_{\varepsilon^0} \frac{\delta E_{\text{total}}}{\delta \epsilon_{ij}^0(\vec{r}, \tilde{t})},
\]  
where \( \tilde{t} \) denotes a fictitious dimensionless “time” used in the numerical relaxation process, \( \gamma_{0, \varepsilon^0} \) is a uniform damping parameter, \( \gamma_{1, \varepsilon^0} \) is a damping parameter that preferentially suppresses large wavenumber oscillations, and \( \alpha_{\varepsilon^0} \) is the wave speed.

4. Compute \( \tilde{u}_i(\vec{r}) \) by way of Eq. \([15]\).

5. Evaluate the in-plane strain from \( \epsilon_{ij}(\vec{r}) = \varepsilon_{ij} + \frac{1}{2} \left[ \frac{\partial \tilde{u}_i(\vec{r})}{\partial r_j} + \frac{\partial \tilde{u}_j(\vec{r})}{\partial r_i} - \frac{\partial \tilde{u}_k(\vec{r})}{\partial r_i} \frac{\partial \tilde{u}_l(\vec{r})}{\partial r_j} \right] \).

6. Iterate \( \eta_p(\vec{r}) \) to equilibrium using
\[
\frac{\partial^2 \eta_p(\vec{r}, \tilde{t})}{\partial \tilde{t}^2} + (\gamma_0, \eta_p - \gamma_{1, \eta_p} \nabla^2) \frac{\partial \eta_p(\vec{r}, \tilde{t})}{\partial \tilde{t}} = -\alpha_{\eta_p} \frac{\delta E_{\text{total}}}{\delta \eta_p(\vec{r}, \tilde{t})},
\]  
where \( \gamma_0, \eta_p, \gamma_{1, \eta_p} \) and \( \alpha_{\eta_p} \) denote effective damping parameters and wave speed, akin to step #3 above.

7. Iterate \( \tilde{\omega}(\vec{r}) \) to equilibrium using
\[
\frac{\partial^2 \tilde{\omega}(\vec{r}, \tilde{t})}{\partial \tilde{t}^2} + (\gamma_0, \omega - \gamma_{1, \omega} \nabla^2) \frac{\partial \tilde{\omega}(\vec{r}, \tilde{t})}{\partial \tilde{t}} = -\alpha_{\omega} \frac{\delta E_{\text{total}}}{\delta \tilde{\omega}(\vec{r}, \tilde{t})},
\]  
Again, \( \gamma_0, \omega, \gamma_{1, \omega} \) and \( \alpha_{\omega} \) denote effective damping parameters and wave speed. During the simulations, Eq. \([22]\) is first iterated until \( \tilde{\omega}(\vec{r}) \) converges, after which we return to step #6 and repeat. Once both \( \tilde{\omega}(\vec{r}) \) and \( \eta_p(\vec{r}) \) have converged, we return to step #3 and repeat the above steps until the variables \( \tilde{\omega}(\vec{r}), \eta_p(\vec{r}), \) and \( \epsilon_{ij}^0(\vec{r}) \) have converged.

### 3 Parameter values employed in simulations

The elastic and lattice constants for TMDs are listed in Tables S1 and S2 as well as plotted in Fig. S2 for convenience. The parameters \( a, b, c \) and \( d \) for the bulk energy terms are tabulated in Table S3. The interfacial energy parameter \( \beta \) is set at 0.003 for all TMDs. The dynamic relaxation parameters used in the simulations are \( \alpha_{\varepsilon^0} = 0.5, \gamma_{0, \varepsilon^0} = 1, \gamma_{1, \varepsilon^0} = 1, \alpha_{\eta_p} = 10, \gamma_{0, \eta_p} = 1, \gamma_{1, \eta_p} = 1, \alpha_{\omega} = 5, \gamma_{0, \omega} = 1, \) and \( \gamma_{1, \omega} = 1 \) with a discrete time step \( \Delta \tilde{t} = 0.075 \) and grid spacing \( \Delta \tilde{x} = 1 \). The order parameter \( \phi(\vec{r}) \) is set to 0.05 for TMD #1 and 1 for TMD #2 with a diffuse interface described by a hyperbolic tangent function. [We note that \( \phi(\vec{r}) \) should be non-zero, otherwise \( \tilde{C}_{ijkl} = 0 \) and \( \tilde{S}_{ijkl} \) becomes singular. Thus, we set the minimum value of \( \phi(\vec{r}) \) to be 0.05 in all...
Figure S2: Structural and elastic heterogeneity of group VI TMDs. (a) Lattice constants $a$ and $b$. (b) Young’s moduli $Y$ and Poisson’s ratios $\nu$.

Table S1: Young’s moduli $Y$ and Poisson’s ratios $\nu$ of group VI TMDs for the 2H phase.

<table>
<thead>
<tr>
<th>TMDs</th>
<th>$Y$ (N/m)</th>
<th>$\nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MoS$_2$</td>
<td>124.5</td>
<td>0.25</td>
</tr>
<tr>
<td>MoSe$_2$</td>
<td>103.9</td>
<td>0.23</td>
</tr>
<tr>
<td>MoTe$_2$</td>
<td>79.4</td>
<td>0.24</td>
</tr>
<tr>
<td>WS$_2$</td>
<td>139.6</td>
<td>0.22</td>
</tr>
<tr>
<td>WSe$_2$</td>
<td>116.0</td>
<td>0.19</td>
</tr>
<tr>
<td>WTe$_2$</td>
<td>86.4</td>
<td>0.18</td>
</tr>
</tbody>
</table>

our simulations.]

4 Analysis of threshold strains and channel widths for phase patterning protocols

4.1 Linear arrays of holes

To rationalize the behaviors of $\epsilon_{ij}^{cri}$ and $W_{min}$ in the case of linear arrays of circular holes, we will consider two limits, namely $D_h/R_h \gg 1$ and $D_h/R_h \sim 2$. In the former case, we will focus on an isolated circular hole under biaxial strain $\epsilon_{xx} = \epsilon_{yy} = \epsilon$ embedded in an infinitely large, elastically homogeneous sheet, and ignore the effects of all other holes. The radial and hoop strains in this case are given by $\epsilon_{rr} = \epsilon \left[1 - (1+\nu)/(1-\nu) \frac{R^2}{R^2} \right]$ and $\epsilon_{\theta\theta} = \epsilon \left[1 + (1+\nu)/(1-\nu) \frac{R^2}{r^2} \right]$, where $r$ denotes the distance
Table S2: Lattice constants of group VI TMDs for both 2H and 1T′ phases.

<table>
<thead>
<tr>
<th>TMDs</th>
<th>a,H(Å)</th>
<th>b,H(Å)</th>
<th>a,T′(Å)</th>
<th>b,T′(Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MoS$_2$</td>
<td>3.183</td>
<td>5.513</td>
<td>3.179</td>
<td>5.717</td>
</tr>
<tr>
<td>MoSe$_2$</td>
<td>3.318</td>
<td>5.747</td>
<td>3.280</td>
<td>5.971</td>
</tr>
<tr>
<td>MoTe$_2$</td>
<td>3.550</td>
<td>6.149</td>
<td>3.455</td>
<td>6.380</td>
</tr>
<tr>
<td>WS$_2$</td>
<td>3.182</td>
<td>5.511</td>
<td>3.197</td>
<td>5.707</td>
</tr>
<tr>
<td>WSe$_2$</td>
<td>3.315</td>
<td>5.744</td>
<td>3.300</td>
<td>5.944</td>
</tr>
<tr>
<td>WTe$_2$</td>
<td>3.552</td>
<td>6.154</td>
<td>3.491</td>
<td>6.320</td>
</tr>
</tbody>
</table>

to the center of the hole. A conductive channel should form when $\epsilon_{rr} \gtrsim \epsilon_{ij}^*$ and $\epsilon_{\theta\theta} \gtrsim \epsilon_{ij}^*$ everywhere between the holes. Since the smallest strains occur between the holes at midpoint $r = \frac{D_h}{2}$, we immediately obtain the numerically observed scaling relation $\epsilon_{ij}^{cri}/\epsilon_{ij}^* = 1 - const \times (D_h^2/R_h^2)^{-1}$. Furthermore, given that the strains exceed $\epsilon_{ij}^*$ within an annulus of outer radius $\frac{D_h}{2}$, the channel width scaling is given by $W_{min} \sim D_h$, consistent with the data in Fig. 2d in the main text.

On the other hand, in the limit $D_h/R_h \sim 2 - 10$, elastic interactions between holes can no longer be neglected. In this limit, amplification of the transverse strain $\epsilon_{yy}$ readily leads to the formation of 1T′ domains which span two consecutive holes at moderate biaxial strains. In order to complete the channels, however, the edges of each hole must be decorated by the 1T′ domains. This is achieved when $\epsilon_{xx} \gtrsim \epsilon_{xx}^*$, with $\epsilon_{xx}$ denoting the longitudinal strain. As can be seen in the inset of Fig. 2c in the main text, the amplification of $\epsilon_{xx}$ is only weakly dependent on $D_h$, thus leading to the saturation of $\epsilon_{xx}^{cri}$. Furthermore, $W_{min}$ should be proportional to $R_h$ and independent of $D_h$, in agreement with our data in Fig. 2d in the main text.

### 4.2 Grain boundaries

For the case of phase patterning facilitated by grain boundaries, we will employ similar arguments as in the case of linear arrays of holes. Specifically, channel formation should be completed when

Table S3: Parameters $a$, $b$, $c$ and $d$ appearing in bulk energy term for group VI TMDs.

<table>
<thead>
<tr>
<th>TMDs</th>
<th>$a$</th>
<th>$b$</th>
<th>$c$</th>
<th>$d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MoS$_2$</td>
<td>0.125</td>
<td>0.04</td>
<td>-0.339</td>
<td>0.32</td>
</tr>
<tr>
<td>MoSe$_2$</td>
<td>0.097</td>
<td>0.04</td>
<td>-0.337</td>
<td>0.32</td>
</tr>
<tr>
<td>MoTe$_2$</td>
<td>0.015</td>
<td>0.02</td>
<td>-0.0255</td>
<td>0.03</td>
</tr>
<tr>
<td>WS$_2$</td>
<td>0.115</td>
<td>0.04</td>
<td>-0.341</td>
<td>0.32</td>
</tr>
<tr>
<td>WSe$_2$</td>
<td>0.09</td>
<td>0.04</td>
<td>-0.335</td>
<td>0.32</td>
</tr>
<tr>
<td>WTe$_2$</td>
<td>0</td>
<td>0.016</td>
<td>-0.025</td>
<td>0.03</td>
</tr>
</tbody>
</table>
the strain at midpoint between dislocations, \( \epsilon_{xx}(0, d/2) \), reaches the defect-free transformation strain \( \epsilon^*_{xx} \). Now, \( \epsilon_{xx} = \tau_{xx} + \epsilon_{xx}^{\text{disloc}} \), where \( \epsilon_{xx}^{\text{disloc}} \) denotes the additional strain due to dislocations. Given that \( \epsilon_{xx}^{\text{disloc}}(0, y) \sim b/y \) for LAGBs\(^2\) we immediately obtain \( \epsilon^{\text{cri}}_{yy} \sim \epsilon^*_{xx} - b/d \sim \epsilon^*_{xx} - \theta \), in agreement with our numerical data. Furthermore, since the strains emanating from the dislocations become screened at distances \( \gtrsim d \) from the GB, the minimum channel width \( W_{\text{min}} \sim d \sim 1/\theta \), again consistent with our data.

4.3 Heterointerfaces

Let us first consider perfectly planar MoTe\(_2\)/MoS\(_2\) superlattices. First, as far as the effects of misfit dislocations on the threshold strains in perfectly planar monolayers are concerned, on the one hand, a larger \( \rho \) translates to higher density of nucleation sites for the 1T\('\) domains. On the other hand, by relaxing misfit strains, dislocations reduce the driving force for the subsequent growth and coalescence of the 1T\('\) domains. Now, we note that, as per the phase diagram for the 2H/1T\('\) transformation for MoTe\(_2\) in Fig. S1 (b), the predicted bulk transformation strain \( \epsilon^*_y \approx 0.6\epsilon_{xx} + 0.06 \) with tensile (compressive) strain in the \( y \)-direction (\( x \)-direction) and \(-0.1 \leq \epsilon_{xx} \leq 0.1 \). The compressive strain \( \epsilon_{xx} \) arises from the misfit between MoS\(_2\) and MoTe\(_2\), and is relaxed – either partially or fully – by misfit dislocations. In particular, the net compressive strain can be approximated as \( \epsilon_{xx} \approx \epsilon^{\text{mis}} + \rho |b| \). Therefore, it follows that

\[
\epsilon^{\text{cri}}_{yy} \approx 0.6\rho |b| + 0.06 + 0.6\epsilon^{\text{mis}} = 0.6\rho |b| + \text{const},
\]

in very good quantitative agreement with the numerical data in Fig. 4 d in the main text.

Next, in the case of free-standing monolayers (cf. Fig. 4 b in the main text), for sufficiently wide superlattices with large misfit strains \( \epsilon_{ij}^H \) and small \( \rho \), wrinkling provides an effective means to relax strains within the compressive regions of the monolayer\(^{12}\) Thus, channel completion behavior in this limit should be similar to the perfectly planar MoTe\(_2\)/MoS\(_2\) superlattices with \( \rho \approx \rho_{\text{max}} \). However, for large \( \rho \), the superlattice will remain planar as wrinkling becomes energetically too costly\(^{12}\) Therefore, \( \epsilon^{\text{cri}}_{yy} \) can be estimated in these two limits via

\[
\epsilon_{yy}^{\text{cri}} = \begin{cases} 
0.6\rho_{\text{max}} |b| + \text{const}, & \rho \to 0 \\
0.6|b| + \text{const}, & \rho \to \rho_{\text{max}}
\end{cases}
\]

while for intermediate values of \( \rho \), smooth interpolation behavior is expected. The numerical data in Fig. 4 d in the main text for \( \epsilon_{yy}^{\text{cri}} \) are in very good agreement with the above arguments.

Finally, for superlattices with finite substrate interaction (see Fig. 4 c in the main text), it is reasonable to expect that the dependence of \( \epsilon_{yy}^{\text{cri}} \) on \( \rho \) should interpolate between the two extreme cases discussed above. Specifically, for \( \rho \to \rho_{\text{max}} \), \( \epsilon_{yy}^{\text{cri}} \approx 0.6\rho |b| + \text{const} \), while for \( \rho \to 0 \), a non-zero
substrate interaction asymptotically yields $\epsilon_{yy}^{cri} \sim 0.6\rho_{max}|b| + \text{const}$. Again, our numerical data in Fig. 4 d in the main text for $\epsilon_{yy}^{cri}$ are in good agreement with the above considerations.

5 Phase programming via arrays of square holes

As discussed in the main text, the critical strain $\epsilon_{ij}^{cri}$ and the minimum width $W_{min}$ for the formation of connected 1T’ channels are controlled by the ratio $D_h/R_h$ for circular holes. Since the magnitude of strain amplification of holes is directly related to their geometry, we have also investigated MoTe$_2$ monolayers with arrays of square holes of edge length $a_h$ and separation $D_h$ to verify the generality of our main conclusions.

As per the simulation results in Fig. S3, the formation and growth of the 1T’ domains as the external strain is increased proceeds in a manner analogous to the circular hole case discussed in the main text. More quantitatively, $\epsilon_{xx}^{cri}$ and $W_{min}$ are controlled by $D_h/a_h$, such that $\epsilon_{xx}^{cri}$ and $W_{min}/a_h$ essentially saturate for $D_h/a_h \sim 2 - 4$, while $\epsilon_{xx}^{cri}/\epsilon_{xx}^{*} \sim 1 - \text{const.} \times \frac{1}{D_h/a_h^2}$ and $\frac{W_{min}}{a_h} \sim \frac{D_h}{a_h}$ for $D_h/a_h \gtrsim 6$.

6 Effects of anisotropic 1T’ phase stiffness tensor and elastic heterogeneity on phase patterning via misfit dislocations

Finally, in order to assess the effects of the isotropic and homogeneous elastic stiffness tensor adopted for the 1T’ variants on phase patterning, we have implemented the fully anisotropic and heterogeneous stiffness tensor for a specific phase patterning protocol, namely that with misfit dislocations. More specifically, we considered the case of perfectly planar monolayers, and evaluated the threshold strain $\epsilon_{yy}^{cri}$ at varying misfit dislocation densities $\rho$. The data for both the isotropic and anisotropic systems are shown in Fig. S4 (a). It can be seen that quantitative differences between the isotropic and fully anisotropic systems are very small. Not unexpectedly, the emerging 1T’ domain morphologies do show some differences, as shown in Figs. S4 (b) and (c), but they appear to have a rather small effect on the threshold strain at which connected paths form.

References


Figure S3: (a), (b): Formation of connected 1T’ phase channels in a MoTe$_2$ monolayer under externally applied biaxial tensile strain in the presence of a linear array of square holes. $D_h/a_h = 2$ and 7.5 for (a) and (b), respectively. Applied strain ($\epsilon_{xx}, \epsilon_{yy}$) is indicated underneath each panel. First and second columns represent the spatial distribution of the 1T’-MoTe$_2$ variants, while the last columns show the scaled maximum principal strains $\epsilon^{\text{max}}/\epsilon^{*}_{xx}$, with $\epsilon^{*}_{xx} \approx 0.11$ as per the phase diagram in Fig. S1 (b). (c): Threshold strain $\epsilon^{\text{cri}}_{xx}/\epsilon^{*}_{xx}$ versus $1/(D_h/a_h)^2$. An error bar of 0.03 is included to represent a strain range for the connection to complete. The dashed red line is a guide to the eye. (d): Minimum width $W_{\text{min}}/a_h$ vs. $D_h/a_h$. Error bars represent 10% confidence intervals.
Figure S4: (a): Quantitative comparison of the threshold strains $\varepsilon_{yy}^{\text{cri}}$ vs. misfit dislocation density $\rho$ for a perfectly planar MoS$_2$/MoTe$_2$ heterostructure under applied uniaxial tensile strain, computed for both elastically isotropic and anisotropic systems. Symbol sizes represent confidence intervals. The red dashed line is a guide to the eye. (b) and (c): 1T$'$ domain morphologies before (b) and after (c) the formation of a 1T$'$ channel in both elastically isotropic and anisotropic systems at $\rho = 0.19 \text{nm}^{-1}$ [vertical dashed line in (a)]. Applied strain $(\varepsilon_{xx}, \varepsilon_{yy})$ is indicated underneath each panel.