

<https://doi.org/10.1038/s41524-026-02346-w>

Predicting moiré physics from local proximity effects in van der Waals heterostructures

Lukas Cvitkovich , Klaus Zollner & Jaroslav Fabian

While proximity-induced magnetism in van der Waals (vdW) heterostructures is often treated as uniform or analytically predictable, we reveal significant spatial fluctuations that challenge conventional modeling. Through a case study of graphene on $\text{Cr}_2\text{Ge}_2\text{Te}_6$ (CGT), we reveal a profound decoupling between pseudospin-governed polarization and the highly localized nature of induced atomic moments. We resolve this dependency using a machine learning framework trained on density functional theory (DFT) data. By employing atomic environment descriptors, we demonstrate that proximity effects are dictated by the local stacking configuration within $\sim 2 \text{ nm}^2$, enabling the discovery of complex moiré patterns and dodecagonal states—features that remain computationally prohibitive for standard DFT. This study provides a scalable method for the precise engineering of proximity-driven phenomena in next-generation quantum materials.

Integration of twisted 2D materials into van der Waals (vdW) heterostructures allows tailoring of electronic, optical, and magnetic properties^{1–7} for ultrafast and low-power electronic and spintronic⁸ devices. One particularly promising strategy is to harness proximity effects, enabling superconductivity^{9,10} or strong spin-orbit coupling^{11–21} in materials lacking these features. Furthermore, when bringing non-magnetic 2D materials, such as graphene or transition metal dichalcogenides²², in contact with a magnetic material, a proximity-induced magnetic exchange interaction can arise^{21,23–31}. Once a non-magnetic channel acquires an exchange field via proximity, it can support spin precession^{32–34}, spin filtering^{23,35}, or valley-selective spin dynamics³⁶, enabling spintronic device platforms.

Among 2D materials for proximity-induced phenomena, graphene stands out for its exceptional spin transport^{32,37–39}. Magnetism in graphene can be induced via proximity exchange coupling to ferro- or antiferromagnets, ideally preserving its intrinsic transport by avoiding charge transfer and parasitic channels. Suitable magnetic materials include semiconductors or insulators such as $\text{Cr}_2\text{Ge}_2\text{Te}_6$ ^{16,40,41}, EuO ^{23,42–44}, or CrI_3 ^{36,44–49}, but also ferromagnetic metals such as Co or Ni, separated from graphene by hBN tunnel barriers^{50,51}.

Proximity exchange coupling² has been studied theoretically^{23,26,50,52–55} and experimentally^{55–57}, showing sensitivity to interlayer distance^{23,26} and tunability by twisting^{4,5,26,28} and gating^{50,52,56,57}. Complementary to scanning tunneling microscopy⁵⁸, density functional theory (DFT) currently provides the only reliable means to investigate proximity effects at the atomic scale^{23,26,50,52–54}, but its high computational cost limits studies to small supercells (a few hundred atoms), making predictions for arbitrary twist angles and large-scale systems infeasible. As a result, phenomena inherently

requiring large simulation cells, such as twist-dependent magnetic moiré patterns^{5,59}, as well as spatial variations from local structural inhomogeneities and the role of local stacking configurations ("registries") in proximity effects²⁶ remain largely unexplored.

In this work, we reveal pronounced atomic-scale fluctuations of proximity-induced magnetic moments in graphene stacked onto a monolayer of the ferromagnetic semiconductor $\text{Cr}_2\text{Ge}_2\text{Te}_6$ (CGT). This behavior contrasts sharply with the spin polarization of Dirac electrons, which is locked to the pseudospin, leading to the following classification of proximity effects in graphene: (i) *pseudospin-preserving* proximity, producing a largely uniform and predictable texture captured by a lattice Hamiltonian; and (ii) *pseudospin-breaking* proximity, which is inherently local and defies simple atomistic modeling. Spin polarization at the Fermi level falls into type (i), where the Dirac states are only slightly perturbed²⁶. But whenever the hybridization with the proximitizing layer is resonant—as in graphene/hBN/Co⁵¹ or, as in our case, where the deep p_z band states in graphene/CGT affect the carbon magnetic moments—the proximity effects are of type (ii).

How can we model non-perturbative, class (ii), proximity effects? Can we, as for class (i), go beyond DFT and develop a methodology to efficiently predict pseudospin-breaking quantities for arbitrary stackings, by mapping the local atomic-scale geometry to the induced response? As we demonstrate here, machine learning (ML) provides a compelling, computationally efficient alternative to expensive DFT simulations. Our premise is simple: assuming that the magnetic moment of each carbon atom is fully determined by its local atomic environment, we train an ML model using atomic geometries as input and DFT-calculated magnetic moments as targets, see Fig. 1. Remarkably, besides accurate predictions, this model also gives a

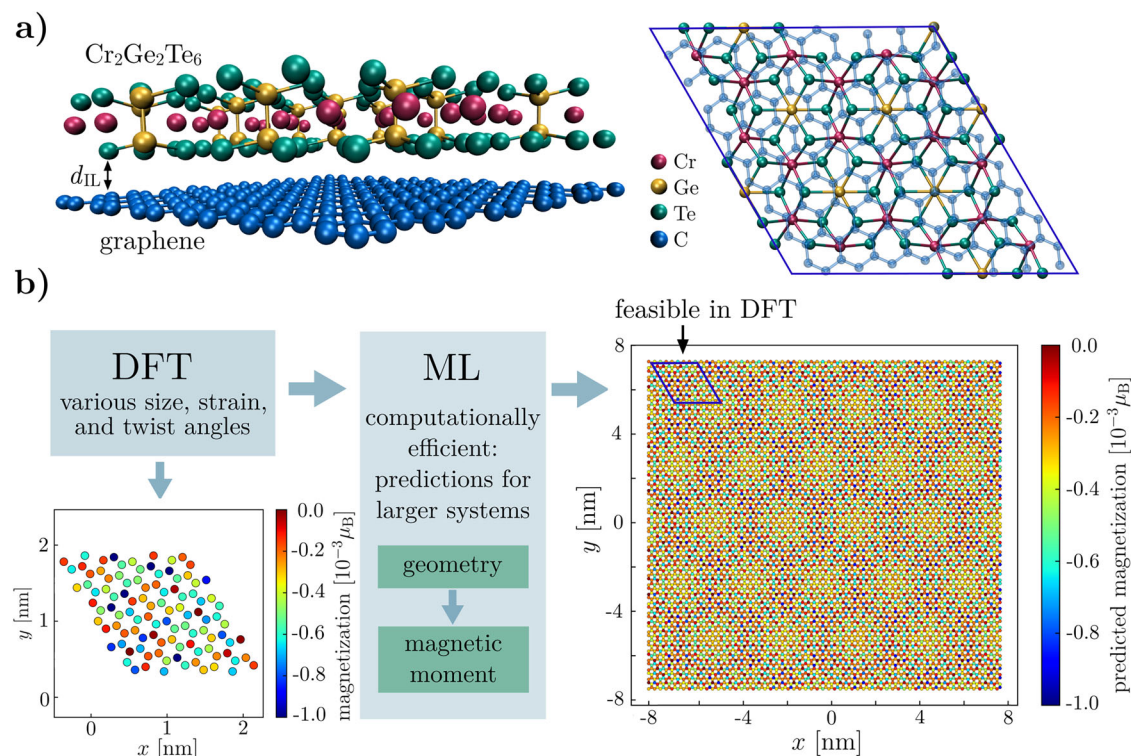


Fig. 1 | ML workflow for modeling magnetic proximity effects in graphene/Cr₂Ge₂Te₆. **a** 3D and top view of a representative DFT simulation cell. Proximity to the 2D Ising ferromagnet Cr₂Ge₂Te₆ induces a magnetic moment in the carbon atoms. Images created by VMD software⁷⁸. **b** We employ DFT to determine the proximitized magnetization of the graphene layer in a set of heterostructures with

varying sizes, twist angles θ , and interlayer distances d_{IL} . Together with the structural/geometrical information encoded by the SOAP descriptor, the data set is then used to train an ML regression model to predict the proximitized magnetization in much larger structures, as indicated.

measure of how local the proximity effect is (and in doing so, validating the underlying assumption of locality).

We apply the ML framework to twisted graphene/CGT, which is experimentally relevant for spin transport^{16,27,31}, to model the induced magnetic moments in graphene, a type (ii) quantity. We show that the magnetic moment of a carbon atom depends only on a small surrounding region of ~ 2 nm². The ML model predicts large-scale magnetic textures and generalizes to much larger systems, enabling predictions for structures with millions of atoms and arbitrary twist angles. Based on the ML predictions, we demonstrate that the proximity-induced magnetic landscape remains strictly locked to the local atomic registry even in the limit of maximum incommensurability. This is evidenced by the transition into a dodecagonal magnetic state at a twist angle of 30°, a regime where interlayer interactions are typically expected to self-average or vanish.

Results

Two types of proximity effects

The initial observation that motivates this study is strongly fluctuating proximity-induced magnetic moments in the graphene layer of commensurate graphene/CGT supercells, as shown in Fig. 1. The fairly wide distribution of induced magnetic moments reveals a previously unrecognized dichotomy: Proximitized quantities may or may not be sensitive to the local atomic environment, and in some systems, both types of proximity effects can occur at the same time. The heterobilayer CGT/graphene is a prime example of a system showing both types of proximitized quantities: While the spin polarization at the Fermi level is staggered, i.e., locked to the pseudospin, the proximity-induced magnetic moments vary across the graphene layer, as illustrated in panels a and b of Fig. 2.

In general, the two regimes can be mathematically described as follows. By setting up a basic perturbation theory, one can show that the effective

induced magnetic moment (or exchange field) on a carbon atom i is

$$\lambda_{\text{eff},i} \approx \sum_m \frac{|t_{i,m}|^2}{E - \epsilon_m} \Delta_m, \quad (1)$$

where $t_{i,m}$ is the position-dependent interlayer hopping between carbon atom i and another atom m in the neighboring layer, E is the energy of the graphene state, ϵ_m is the energy (band) of the neighboring layer (which may correspond to the Te states in CGT in the present work), and Δ_m is the intrinsic exchange splitting of the substrate.

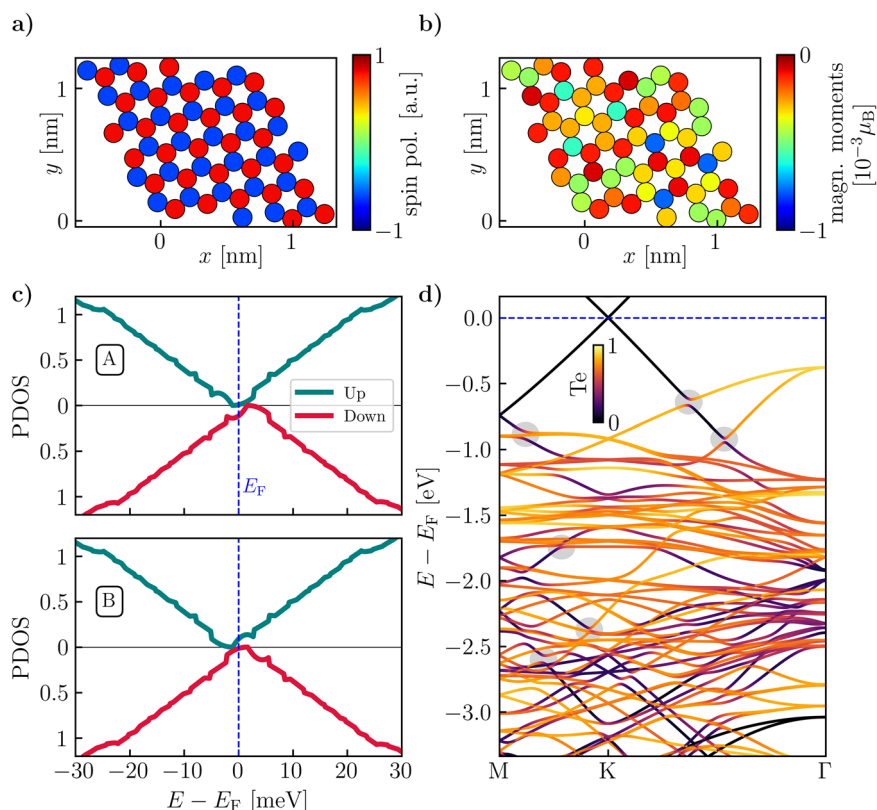
First, the pseudospin-conserving regime. At the Fermi level, the graphene states are energetically very far away from the Te bands. Mathematically, the denominator in Eq. (1) is large; therefore, the interaction becomes weak. One can then adopt a picture of a sublattice basis with an effective staggered potential coming from the different average energy (over the commensurate unit cell) of the orbitals on A and B sublattice. This is a complementary, real-space view of the perturbation theory in the momentum space^{60,61}.

The situation is fundamentally different in the pseudospin-breaking regime. At lower energies, in the case of graphene on CGT in the range from -3 to -0.5 eV, the interaction becomes resonant $E \approx \epsilon_m$, meaning that the perturbation blows up (the denominator $E - \epsilon_m \rightarrow 0$). The induced magnetization is no longer a weak background; it becomes highly sensitive to the exact, local environment and therefore also to the exact hopping amplitude $|t_{i,m}|^2$. In an incommensurate lattice, like graphene on CGT, $|t_{i,m}|^2$ varies strongly from one carbon atom to the next. This breaks the sublattice (pseudospin) symmetry locally.

DFT perspective and microscopic origins

In the following, we further explain this behavior by means of a deeper analysis of the DFT calculations. In order to resolve exchange splittings on

Fig. 2 | Two types of proximity effects. Presented are DFT simulations for graphene/CGT at twist angle $\theta = 8.948^\circ$. **a** Spin polarization $P(E_F)$ over the carbon atoms in the supercell. **c** The pseudospin character of $P(E_F)$ is reflected in the calculated projected density of states (PDOS). **b** Proximity-induced magnetic moment of C atoms, showing a wide distribution. **d** Band structure for spin-down bands with color-coded projections on Te states. Anti-crossings (gray circles) indicate resonant hybridization.



the order of a few meV, sampling of the Brillouin zone (BZ) around the K point requires a dense k -point mesh. We therefore implemented an adaptive k -point mesh (detailed in the “Methods” section). This approach allows for a trade-off between sufficient sampling around K, while also delivering accurate states at lower energy and across the full BZ. We applied this method to one exemplary structure (102 atoms), using 48×48 k -points across the BZ, while a 15 times denser mesh is used around K.

Our analysis begins with spin polarization at the Fermi energy, the regime that governs spin transport. After obtaining the electronic structure from DFT, we project onto a localized atomic basis and integrate across the BZ using a linear triangular method⁶² to obtain the projected density of states (PDOS) of each C atom. As shown in Fig. 2c, only two types of PDOS appear, which can readily be associated with the sublattice of the graphene layer. As noted before, this is a sharp contrast to the wide spread in magnetization among the atoms. Furthermore, while in this example the magnetic moments all align in the same direction, at the considered twist angle $\theta = 8.948^\circ$, the spin polarization is ferrimagnetic. Validating the simple Hamiltonian from refs. 26,50, which treats the sublattices of graphene separately, the PDOS is precisely mirrored about the Dirac point between sublattices A and B. The local atomic environment leaves the spin polarization at the Fermi level unchanged.

Next, we focus on the induced magnetic moments. An atom’s magnetic moment reflects the net spin polarization accumulated across all occupied electronic states up to the Fermi level. In order to clarify the origins of the distribution of magnetic moments, we consider the band structure in Fig. 2d. It shows clear anti-crossings (some marked by gray circles) between the graphene p_z bands and—as apparent upon projection on Te atoms—Te-dominated bands of CGT (the projections on other atom types are shown in Supplementary Note 1).

For an atom-resolved analysis, we calculate the atom-projected spin polarization $P(E) = \text{PDOS}_{\text{up}}(E) - \text{PDOS}_{\text{dn}}(E)$ along the full energy range which gives the total magnetization of each atom by $m(E) = \int_{-\infty}^E P(E') dE'$. As shown in Fig. 3, close to the Fermi energy, $m(E)$ is virtually the same for each C atom. The difference in magnetization between the C atoms is

mainly accumulated at the energy range from -3 to -0.5 eV, directly corresponding to the energy window where graphene bands hybridize with neighboring Te valence states.

Local description of proximity-induced magnetization

As shown above, the distribution of induced magnetic moments is mainly driven by hybridization with Te atoms. Given that even directly neighboring C atoms of a shared sublattice may have very different atomic environments—a consequence of the lattice mismatch—an intuitive conjecture is that these fluctuations are caused by spatial inhomogeneities. For further evidence, we have verified that the induced magnetic moments are highly sensitive to even small displacements of the graphene layer (see Supplementary Note 2). We therefore arrive at the central assumption of this work, namely that the magnetization of a C atom in the CGT/Gr heterostructure is governed solely by its local atomic environment. This assumption is implemented into our ML framework by the use of local atomic descriptors (as detailed in the “Methods” section). The results obtained by this ML model are discussed in the following.

We assess the accuracy of the ML model using testing data from our DFT dataset. In total, the dataset comprises 9498 samples, each consisting of a C atom, its local atomic environment, and the corresponding induced magnetic moment. 10% of this data has been isolated for the testing process and is used for predictions by the trained ML model. The model performance is quantified using cross-validation, yielding a mean squared error (MSE) of $2 \times 10^{-9} \mu_B^2$, corresponding to an average absolute prediction error of approximately $4.5 \times 10^{-5} \mu_B$. Given average magnetic moments of $0.4 \times 10^{-3} \mu_B$, the deviation between predicted and DFT-computed magnetic moments is around 10%. The correlation plot of this data is shown in Fig. 4.

In order to determine the extent of the region which ultimately determines the induced magnetic moment, we analyze the dependence of the prediction accuracy on the cut-off radius r_{cut} of the SOAP descriptor. Due to a polynomial weighting scheme, all atomic orbitals effectively vanish beyond r_{cut} meaning that overlap between two orbitals is only captured if the atoms are within a distance of $2r_{\text{cut}}$. As shown in Fig. 5, the MSE saturates

Fig. 3 | Magnetization $m(E)$ obtained from the DFT-derived spin polarization for some representative C atoms at $\theta = 8.948^\circ$. **a Differences are mostly picked up at energies between -3 and -0.5 eV, where Te hybridizes with the graphene p_z bands. **b** Magnetization across the graphene layer. The associated C atoms of **a** are labeled with letters A to D.**

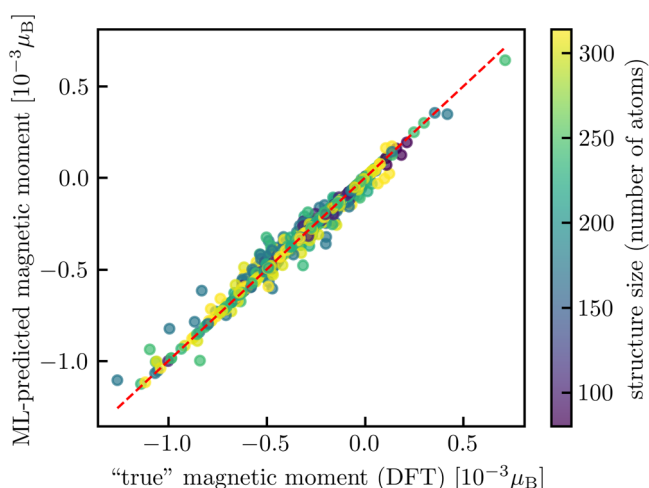
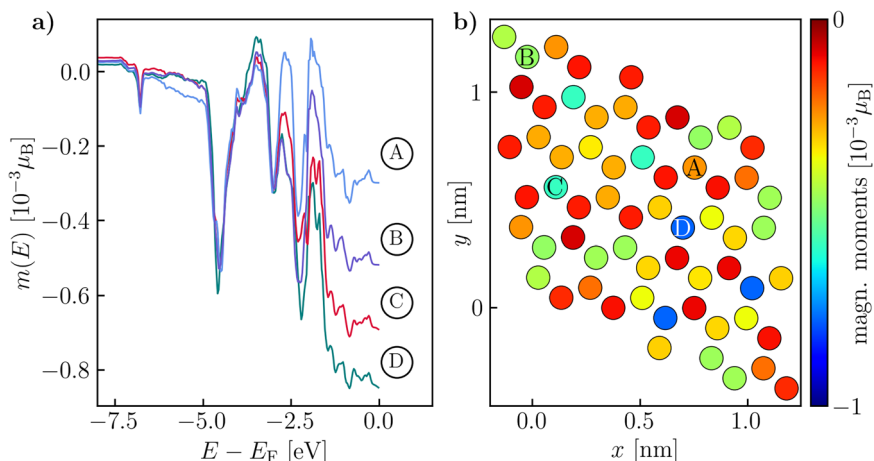


Fig. 4 | Verification of the ML model within the DFT dataset. 90% of the data has been used for training, and the remaining 10% serve as the testing data.

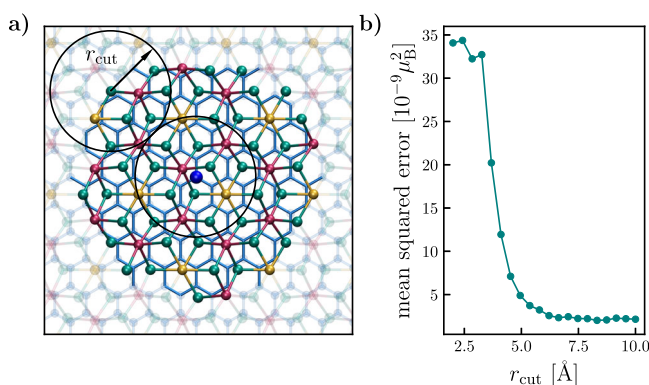


Fig. 5 | Saturation of ML prediction accuracy at increased cut-off radius. **a** The local environment of the central C atom is defined by r_{cut} . **b** The prediction error saturates at $r_{\text{cut}} = 6$ Å.

for $r_{\text{cut}} > 6$ Å, indicating that increasing the descriptor range beyond this point provides no additional information. We therefore conclude that the proximitized magnetic moment of each C atom is determined by its local atomic environment within 12 Å, a region that contains about 100 CGT atoms.

Large-scale predictions

Finally, we apply the ML model to predict magnetic textures of practically relevant system sizes. For this purpose, we combine completely unstrained layers of CGT and graphene and overlay them with varying twist angles, considering the equilibrium d_{IL} of 3.55 Å. Most importantly, we find magnetic moiré patterns arising as a result of periodically recurring local environments of the heterostructures, see Fig. 6. The extent as well as the quantitative fluctuations of these patterns are highly dependent on the twist angle θ . The moiré patterns appear on scales that are far from feasible DFT calculations.

We further analyze the spatial modulation of the proximity-induced magnetic moments by performing a Fourier transform into reciprocal space. This procedure allows for the precise extraction of the Moiré wave-vector and its associated periodicity. In the commensurate and near-commensurate regimes, the hexagonal symmetry of the constituent layers dictates a hexagonal Moiré pattern. In reciprocal space, this is manifested as six primary peaks arranged with C_6 symmetry, see Fig. 7a. Note that in this representation, the expected circular arrangement of these peaks appears elongated, which is a projection artifact arising from representing the data in the fractional basis of the graphene reciprocal lattice vectors (k_n, k_m) rather than in Cartesian reciprocal coordinates.

Each peak corresponds to a Moiré modulation defined by the difference vector:

$$\vec{q}(\theta) = \vec{G}_1(h, k, \theta) - \vec{G}_2(l, m) = k_n \vec{b}_1(\theta) + k_m \vec{b}_2(\theta) \quad (2)$$

where the reciprocal lattice vectors of each layer are given by linear combinations of their primitive basis vectors, $\vec{G}_1(h, k, \theta) = h\vec{b}_1(\theta) + k\vec{b}_2(\theta)$ for graphene and $\vec{G}_2(l, m) = l\vec{g}_1 + m\vec{g}_2$ for CGT, with $(h, k, l, m) \in \mathbb{Z}$. The reciprocal lattice vectors are defined as $\vec{b}_1 = 2\pi[a_{\text{Gr}}^{-1}, (-\sqrt{3}a_{\text{Gr}})^{-1}]$, $\vec{b}_2 = 2\pi[0, 2(\sqrt{3}a_{\text{Gr}})^{-1}]$ and $\vec{g}_1 = 2\pi[a_{\text{CGT}}^{-1}, (-\sqrt{3}a_{\text{CGT}})^{-1}]$, $\vec{g}_2 = 2\pi[0, 2(\sqrt{3}a_{\text{CGT}})^{-1}]$, where $a_{\text{Gr}} = 2.46$ Å and $a_{\text{CGT}} = 6.8275$ Å. The twist angle dependency enters through the rotation of the graphene lattice relative to the stationary substrate, such that $\vec{b}_i(\theta) = \mathbf{R}(\theta)\vec{b}_i$, where $\mathbf{R}(\theta)$ is the 2D rotation matrix. The fractional coordinates k_n and k_m of the moiré wave-vector describe the Moiré modulations as a linear combination of reciprocal lattice vectors of the twisted graphene layer.

The magnitude $|\vec{q}(\theta)|$ determines the real-space Moiré periodicity, $L_M = 2\pi/|\vec{q}|$. The evolution of L_M as a function of the twist angle θ is plotted for the fundamental interval $[0^\circ, 30^\circ]$ in Fig. 7b. While an infinite set of difference vectors is mathematically possible, the physically observable Moiré pattern is governed by a selection rule: the dominant peaks must involve low-index reciprocal lattice vectors. High-order Fourier components of the atomic potential decay rapidly due to the finite width of the

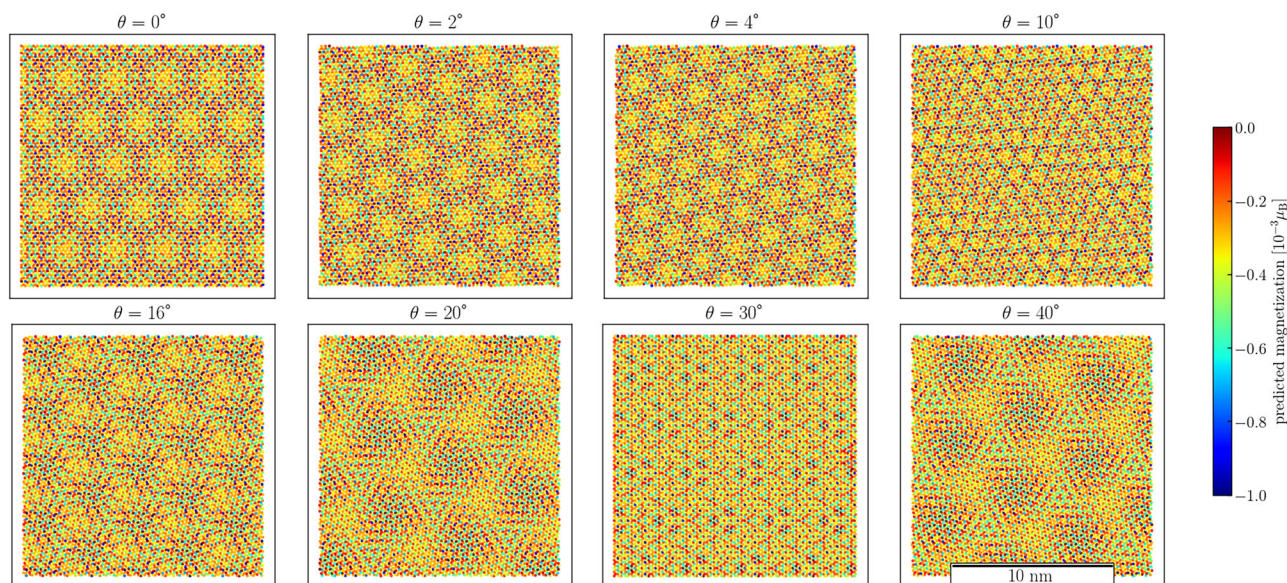


Fig. 6 | Magnetic moiré patterns emerging in larger simulation cells for various twist angles θ as predicted by the ML model. The scale bar in the last panel holds for all panels. Due to the hexagonal shape of both material layers, the pattern repeats with a 60° periodicity and is symmetric about $\theta = 30^\circ$. Thus, $\theta = 20^\circ$ corresponds to $\theta = 40^\circ$.

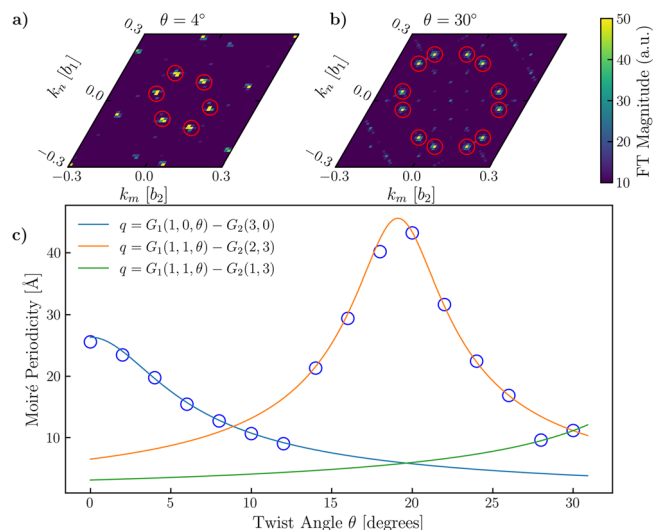


Fig. 7 | Fourier analysis of magnetic moiré patterns. **a** At $\theta = 4^\circ$, six peaks (red circles) indicate a hexagonal moiré lattice. **b** At $\theta = 30^\circ$, 12 peaks of similar intensity signify a dodecagonal state. **c** Moiré periodicity L_M (blue circles) compared to theoretical predictions $q = |\vec{G}_1(h, k, \theta) - \vec{G}_2(l, m)|$. Crossing points at $\approx 9^\circ$ and 30° mark transitions between dominant moiré harmonics.

atomic orbitals and the smoothness of the interlayer potential. Quantitatively, the spectral intensity of second harmonics (at $2\vec{q}$) is one to two orders of magnitude weaker than the primary Moiré peaks, and therefore hardly visible in Fig. 7.

Consequently, only a few combinations of (h, k, l, m) indices possess sufficient Fourier amplitude to be detected. We identify three specific cases resulting from the index sets $(1, 0, 3, 0)$, $(1, 1, 2, 3)$, and $(1, 1, 1, 3)$. As shown in Fig. 7c, these three combinations provide a comprehensive explanation for the ML-predicted data across the entire investigated angular range. The dominance of a specific combination depends on the twist angle, with crossing points (here at $\theta \approx 9^\circ$ and 30°) marking transitions where the system's characteristic spatial frequency shifts. At these angles, multiple Moiré harmonics coexist, leading to complex interference or “super-Moiré” features, cf. Fig. 6. In the vicinity of these crossing points, the numerically extracted L_M may deviate from the theoretical maximum periodicity; this

occurs because our automated extraction identifies the Fourier component with the highest spectral intensity, which may belong to a competing track of higher amplitude but shorter wavelength.

The formation of the magnetic dodecagonal state is specifically intriguing because the underlying structure, the hetero-bilayer at $\theta = 30^\circ$, is not dodecagonal. Why do we still observe a magnetic dodecagonal pattern? The answer is that the dodecagonal state is an outcome of the intersection of two Moiré wavevector trajectories at this specific twist angle (see also Fig. 7c). This intersection (and therefore the formation of a dodecagonal state) is enforced by symmetry, which can be shown analytically. Starting from Eq. (2) together with the definitions in the paragraph below, one can show that two trajectories corresponding to index sets (h_1, k_1, l_1, m_1) and (h_2, k_2, l_2, m_2) where $h_1 = h_2 = k_1 = k_2$ intersect at 30° if and only if $m_1 = m_2 = m$ and $l_1 + l_2 = m$. In other words, the moiré periodicity at $\theta = 30^\circ$ exhibits doubling for some index configurations:

$$L_M^{30^\circ}(h, h, l, m) = L_M^{30^\circ}(h, h, m - l, m). \quad (3)$$

As shown in Fig. 7, we do find an intersection for the index sets $(1, 1, 2, 3)$ and $(1, 1, 1, 3)$ which fulfill these criteria. Crucially, this intersection is enforced by the symmetry of the hexagonal lattices at the specific twist angle of 30° and is independent of the lattice vectors of the two layers. We stress that this argument is fully agnostic to both the ML and DFT data, yet this result is contained in the ML predictions (Fig. 7b).

Next, we discuss the amplitude of the induced magnetic Moiré pattern. While the Moiré periodicity is simply a result of the underlying atom structure, which meets our expectations (see Fig. 7), its amplitude is predicted by the ML model specifically for the magnetic proximity data it is trained on. We present the dependency of the Moiré peak intensity on the twist angle in Fig. 8. We focus on the innermost circle of Moiré peaks as marked in Fig. 7. The data reveal a pronounced maximum at 14° , indicating that the amplitude of the magnetic moiré modulation is strongest at this specific structural alignment. Furthermore, the dodecagonal peaks at 30° exhibit an intensity comparable to those at other twist angles. This demonstrates that the induced magnetic order does not average out, remaining robust even at maximal incommensurability. Finally, we note that these results are independent of the initial stacking registry; translating the graphene layer by an in-plane vector—such as $\vec{T} = 0.5\vec{b}_1 + 0.5\vec{b}_2$ —merely shifts the moiré pattern in real space without altering the observed intensities significantly.

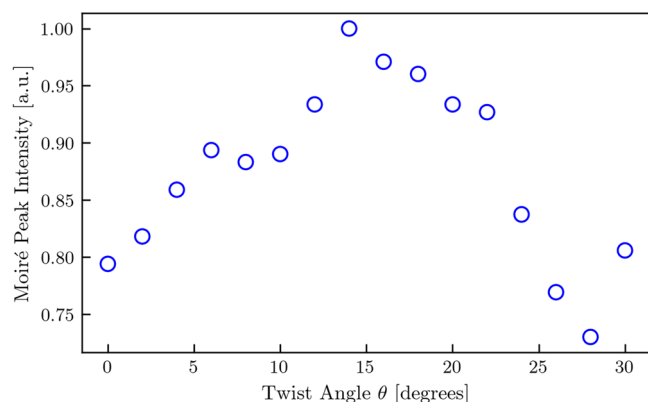


Fig. 8 | Dependence of moiré superlattice peak intensity on interlayer twist angle. The shown variation in peak intensity illustrates the angle-dependent amplitude of the proximitized magnetic moiré potential as predicted by our ML model.

The crossing point at 30° represents a unique high-symmetry state. At this angle, the heterostructure achieves maximal incommensurability, and the translational symmetry of a periodic Moiré crystal is replaced by a dodecagonal pattern. In this regime, 12 peaks of equivalent Fourier amplitude emerge on a single ring in reciprocal space, see Fig. 7b. The extracted periodicity of 11.16 Å represents the characteristic length scale of the quasi-periodic interference. In real space, this corresponds to the distance between local high-symmetry motifs that, while ordered, never repeat on a periodic grid.

Crucially, our results demonstrate that the magnetic proximity effect remains robust even in the absence of translational symmetry. We have also observed that the intensity of the Moiré peaks at 30° remains comparable to those in the periodic regimes. This further indicates that the induction of magnetic moments is driven by local atomic registry rather than global commensurability, opening a pathway to exploring exotic spin transport and frustrated magnetism in quasi-periodic landscapes.

Discussion

Proximity-induced magnetism in twisted graphene/CGT heterostructures reveals a complex multi-scale landscape that defies conventional coarse-grained modeling. Our findings demonstrate that these effects are partitioned into two distinct regimes: a pseudospin-governed polarization at the Fermi level and environment-sensitive magnetic moments arising from deep-lying orbital hybridization.

The fluctuations observed at the atomic scale (Fig. 1) indicate that the induced magnetic moment is not a smooth function of the Moiré coordinate, but rather a discrete response to the local chemical environment. Unlike the exchange splitting at the Dirac point, which is largely protected by the sublattice symmetry, the total atomic moment is a quantity determined by the local stacking. It is primarily accumulated through resonant hybridization between the graphene p_z orbitals and the Te bands in the -3 to -0.5 eV range (Fig. 3). This energy-dependent coupling is hypersensitive to the orbital overlap, which varies significantly even between nearest-neighbor carbon atoms in an incommensurate stacking.

While long-range Moiré potentials are used to describe the periodic recurrence of these specific local atomic registries on a global scale, the actual magnetic moment induced at any specific carbon atom is entirely determined by its immediate atomic neighborhood. The periodic magnetic moiré pattern arises because these local registries repeat in the large moiré structure as well.

In order to model proximity-induced magnetic moments on an experimentally relevant scale, we employ Machine Learning. Using a SOAP-based ML model (see Supplementary Note 3 for a quantitative comparison of several ML algorithms) trained on an extensive DFT data set, we achieve accurate predictions of magnetic moments across a diverse set of structures.

The model generalizes to large-scale systems and enables rapid exploration of the local magnetic landscape. While simple analytic models based on interatomic distances fail to capture the underlying physics (see Supplementary Note 4) due to highly complex hybridization (see Supplementary Note 5), the ML approach successfully encodes complex geometric dependencies.

This locality is quantitatively bounded by our ML analysis. The saturation of the prediction error at a cut-off radius of $r_{\text{cut}} \approx 6$ Å (Fig. 5) provides a physical length scale for magnetic proximity: a carbon atom “feels” the magnetic substrate only within this radius. That finding has two consequences. First, due to the geometric periodicity within the hetero-bilayer on a large scale, magnetic Moiré patterns appear on top of the strong fluctuations on the atomic scale. Second, it suggests that while Moiré physics is often discussed in terms of long-range potentials, the underlying magnetic induction is an inherently short-ranged, quasi-chemical process.

The emergence of a dodecagonal magnetic state at 30° is a striking consequence of this locality. In traditional Moiré theory, the “self-averaging” limit at large twist angles is expected to wash out interlayer effects⁶³. However, the emergence of quasicrystalline states with dodecagonal symmetry has been reported for twisted bilayer graphene at 30°⁶⁴, indicating that this averaging effect is not always dominant. Our model shows that because the induction is locked to the local registry, the graphene layer remains magnetically “templated” even when global translational periodicity is lost. The spectral weight of the magnetic Fourier components at 30° remains robust, overcoming the self-averaging limit.

Finally, the scalability of our ML framework offers a paradigm shift for high-throughput materials design. Because the atomic environment descriptor relies solely on nuclear positions, the framework is intrinsically agnostic to the underlying magnetic order. This allows the model to be generalized to complex magnetic textures beyond the Ising ferromagnetism considered here, including antiferromagnetic or non-collinear substrates or to fundamentally different quantities like the induced charge (see Supplementary Note 6). While DFT is restricted to small, commensurate “approximants”, our approach allows for the simulation of experimental-scale flakes (millions of atoms). This toolkit provides a high-fidelity bridge between ab initio theory and macroscopic device physics in next-generation spintronics.

Methods

Crystal structures and DFT setup

The DFT-generated dataset contains van der Waals heterostructures composed of graphene and CGT with twist angles from 0° to 30°, sampled in steps of approximately 3°, following ref. 26. To construct commensurate supercells, the monolayers are strained by up to 3%. Initially, we fully relaxed the 9 structures at these various twist angles to consider correct interlayer distances d_{IL} and rippling of the graphene layer (<1 pm)²⁶.

The dataset was subsequently extended by translation of the graphene layer (retaining three-fold rotation symmetry), rigid randomized lateral shifts (symmetry-breaking), and vertical interlayer displacements of ± 0.1 and ± 0.2 Å. However, for these structures with rigid shifts (vertical and lateral), we have not relaxed the structure subsequently to stay within reasonable computational limits. Thus, it is fair to say that relaxation is indirectly included in our diverse training set.

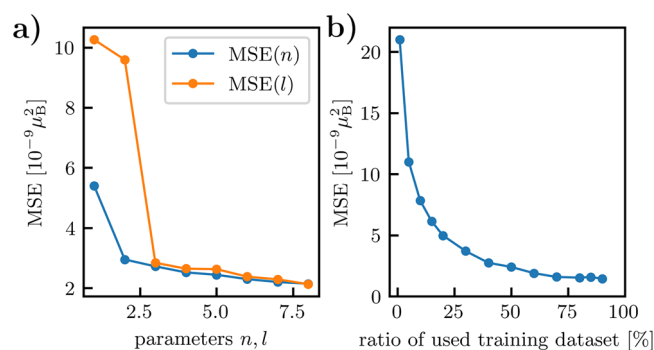
In total, 75 different structures with 9498 C atoms are contained in the data set; however, in structures with a remaining three-fold rotational symmetry, the number of distinct environments is reduced by a factor of three. An overview of the complete dataset used for training of the ML model is given in Table 1. Since we find that the prediction of magnetic moments can be based solely on descriptions of the local atomic environment, larger supercells do not need to be included in the training dataset. From the perspective of the ML model, larger supercells are only a larger collection of local environments. However, such calculations are currently computationally out of reach, initially motivating the use of ML.

The calculations were carried out using plane-wave DFT as implemented in the code *Quantum ESPRESSO*⁶⁵. According to the system size, the

Table 1 | CGT/Gr structures in the training dataset of the ML model

# atoms	# C atoms	# Structures	θ [°]	sym. shift	sym. broken	δ_{IL} [Å]	n_k
80	50	4	30.00	1	0	± 0.1	42
102	62	14	8.94	2	5	$\pm 0.1, \pm 0.2$	42
174	104	14	26.99	2	5	$\pm 0.1, \pm 0.2$	18
218	128	1	0.00	0	0		18
224	134	9	12.21	1	1	$\pm 0.1, \pm 0.2$	12
236	146	9	5.81	1	1	$\pm 0.1, \pm 0.2$	12
242	152	5	23.41	1	1	$\pm 0.1, \pm 0.2$	12
302	182	10	3.00	2	2	$\pm 0.1, \pm 0.2$	9
314	194	9	14.7	2	1	$\pm 0.1, \pm 0.2$	9

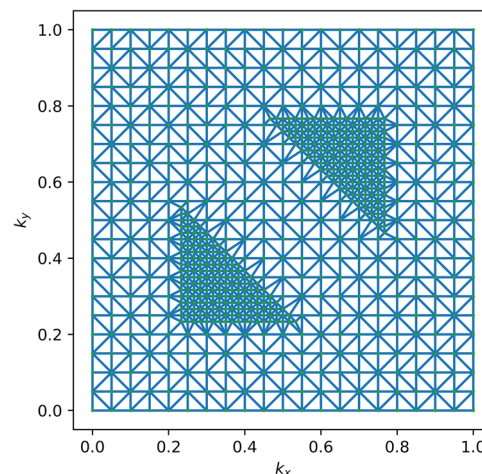
In the first two columns, the number of (carbon) atoms is given. The third column lists the number of considered structures. This number results from the changes (relaxation, lateral shifts, interlayer displacements) that were additionally applied to each structure and added to the dataset. The twist angle θ denotes the rotational angle between the two layers. Lateral shifts are applied in two ways, symmetry-retaining and symmetry-breaking shifts. The number of such operations applied to each structure is given in columns 5 and 6. Next, the considered vertical shifts δ_{IL} from the equilibrium $d_{\text{IL}} = 3.55$ Å, and finally, the number of used kpoints n_k are indicated.

**Fig. 9 | Model convergence and error scaling.** MSE as a function of **a** the SOAP basis set size and **b** the used training set size.

k -point mesh has been chosen as listed in Table 1. Data for the ML training set is calculated based on standard uniform meshes. We used PAW pseudopotentials with scalar relativistic non-linear core correction⁶⁶ in combination with the PBE functional⁶⁷. We utilized the DFT-D2⁶⁸ dispersion correction in all calculations. Correlation effects of Cr 3d electrons are considered by a Hubbard correction of $U = 1.0$ eV. In ref. 69, it has been shown that the system becomes in-plane anisotropic for $U < 0.2$ eV, and that the interlayer coupling becomes antiferromagnetic for $U > 1.7$ eV, contradicting experimental observations. Therefore, the U values within the range of 0.2–1.7 eV are argued to be a reasonable choice. Our Hubbard U is in the middle of this range. We have tested for the stability of our main claims with respect to the chosen Hubbard U and find that it rescales the magnitude of the magnetic moments but keeps the results and conclusions qualitatively unaffected, as shown in Supplementary Note 7.

Machine learning framework and descriptors

To ensure that the machine learning model captures only the intrinsic geometry-magnetization relationship (independent of arbitrary rotations or translations), we represent atomic environments using the Smooth Overlap of Atomic Positions (SOAP) descriptor⁷⁰ as implemented in the *DSDcribe* Python library^{71,72}. It maps an input atomic structure to a column vector,

**Fig. 10 | An exemplary adaptive k -mesh with 20×20 kpoints across the full BZ.** The region around the K point is replaced with a three times denser mesh.

determined by the overlap of hydrogen-like orbitals placed on each atom within a distance of $2r_{\text{cut}}$.

The descriptor uses a basis of radial functions and spherical harmonics, controlled by parameters n and l . As shown in Fig. 9, a minimum of $n = 2$ and $l = 3$ is necessary to qualitatively capture the induced magnetization. The lowest MSE is achieved with $n = l = 8$. We find increased accuracy when weighting the atomic density by a polynomial function $[1 + 2(r/r_{\text{cut}})^3 - 3(r/r_{\text{cut}})]^m$ as suggested in ref. 73. We chose $m = 1$, a cutoff radius of $r_{\text{cut}} = 8$ Å, and a Gaussian smearing width of 0.5 Å.

We note that the optimal orbital basis found above, characterized by $n = l = 8$, lacks explicit chemical meaning and primarily reflects the internal workings of SOAP, whose description is limited to 3-body interactions. Other descriptors may reach comparable accuracy using smaller orbital sets. The atomic cluster expansion (ACE) from ref. 74 builds a systematic expansion over multi-body terms. As a result, a smaller orbital basis can suffice to reach similar prediction accuracy. However, as SOAP provides satisfactory accuracy, we leave further optimization of the ML framework for future studies.

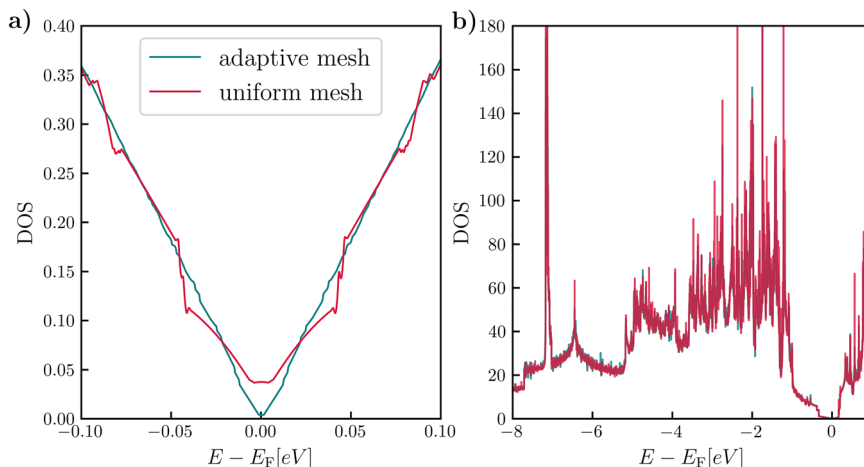
Each datapoint gets a SOAP vector assigned. The target is the magnetic moment from DFT. We train a Random Forest regression model⁷⁵ implemented via the *scikit-learn* Python library⁷⁶. The model was trained with 400 decision trees, using a square root strategy for feature selection at each split, a maximum tree depth of 30, and no bootstrapping. Alternative ML models may yield similar performance and may be used as well, see Supplementary Note 3.

The fully trained ML model is available in an open-source repository⁷⁷ as a *pickle* object. The full training dataset, including all atomic structures and corresponding DFT-computed magnetic moments, is also publicly accessible there.

Adaptive k -point mesh

Obtaining Dirac cones with exchange splittings on the order of a few meV is computationally challenging, given our large supercells, requiring dense k -space sampling around K. To circumvent full BZ sampling with a dense mesh, we implemented a Python code to construct an adaptive mesh (Fig. 10). On this adaptive mesh, we obtain DOS and PDOS by numerically integrating the band energies using a triangular method⁶². Our code generates irreducible meshes at two specified grid densities, replaces a predefined region around K of the coarse mesh with the dense one, and passes the resulting k -points to *Quantum Espresso*. As shown in Fig. 11, this successfully recovers the expected DOS of Dirac cones.

Fig. 11 | Comparison between uniform and adaptive k -grid. **a** The DOS around the Dirac point is captured with significantly enhanced accuracy. Here, the adaptive k -grid uses a 5-fold denser mesh around K . **b** Both methods yield the same results for a broader energy range around E_F .



Data availability

The fully trained ML model is available in an open-source repository (https://github.com/lukas-cvitkovich/CGT_Gr) as a *pickle* object. The full training dataset, including all atomic structures and corresponding DFT-computed magnetic moments, is also publicly accessible there.

Received: 23 April 2026; Accepted: 14 September 2026;

Published online: 29 September 2026

References

- Geim, A. K. & Grigorieva, I. V. Van der Waals heterostructures. *Nature* **499**, 419–425 <https://doi.org/10.1038/nature12385> (2013).
- Sierra, J. F., Fabian, J., Kawakami, R. K., Roche, S. & Valenzuela, S. O. Van der Waals heterostructures for spintronics and opto-spintronics. *Nat. Nanotechnol.* **16**, 856–868 <https://doi.org/10.1038/s41565-021-00936-x> (2021).
- Novoselov, K. S., Mishchenko, A., Carvalho, A. & Neto, A. H. C. 2d materials and van der Waals heterostructures. *Science* **353**, aac9439 <https://doi.org/10.1126/science.aac9439> (2016).
- Carr, S. et al. Twistronics: manipulating the electronic properties of two-dimensional layered structures through their twist angle. *Phys. Rev. B* **95**, 075420 <https://doi.org/10.1103/PhysRevB.95.075420> (2017).
- Carr, S., Fang, S. & Kaxiras, E. Electronic-structure methods for twisted moiré layers. *Nat. Rev. Mater.* **5**, 748–763 <https://doi.org/10.1038/s41578-020-0214-0> (2020).
- Hennighausen, Z. & Kar, S. Twistronics: a turning point in 2d quantum materials. *Electron. Struct.* **3**, 014004 <https://doi.org/10.1088/2516-1075/abd957> (2021).
- Ribeiro-Palau, R. et al. Twistable electronics with dynamically rotatable heterostructures. *Science* **361**, 690–693 <https://doi.org/10.1126/science.aat6981> (2018).
- Žutić, I., Fabian, J. & Das Sarma, S. Spintronics: fundamentals and applications. *Rev. Mod. Phys.* **76**, 323–410 <https://doi.org/10.1103/RevModPhys.76.323> (2004).
- Moriya, R., Yabuki, N. & Machida, T. Superconducting proximity effect in a NbSe₂/graphene van der Waals junction. *Phys. Rev. B* **101**, 054503 <https://doi.org/10.1103/PhysRevB.101.054503> (2020).
- Han, H. et al. Superconducting proximity effect in a van der Waals 2h-tas2/nbse2 heterostructure. *Appl. Phys. Lett.* **118**, 253101 <https://doi.org/10.1063/5.0051968> (2021).
- Gmitra, M. & Fabian, J. Graphene on transition-metal dichalcogenides: a platform for proximity spin-orbit physics and optospintronics. *Phys. Rev. B* **92**, 155403 <https://doi.org/10.1103/PhysRevB.92.155403> (2015).
- Gmitra, M., Kochan, D., Högl, P. & Fabian, J. Trivial and inverted Dirac bands and the emergence of quantum spin hall states in graphene on transition-metal dichalcogenides. *Phys. Rev. B* **93**, 155104 <https://doi.org/10.1103/PhysRevB.93.155104> (2016).
- Khokhriakov, D. et al. Tailoring emergent spin phenomena in Dirac material heterostructures. *Sci. Adv.* **4**, eaat9349 <https://doi.org/10.1126/sciadv.aat9349> (2018).
- Zollner, K. & Fabian, J. Single and bilayer graphene on the topological insulator Bi₂Se₃: electronic and spin-orbit properties from first principles. *Phys. Rev. B* **100**, 165141 <https://doi.org/10.1103/PhysRevB.100.165141> (2019).
- Khokhriakov, D., Hoque, A. M., Karpik, B. & Dash, S. P. Gate-tunable spin-galvanic effect in graphene-topological insulator van der Waals heterostructures at room temperature. *Nat. Commun.* **11**, 3657 <https://doi.org/10.1038/s41467-020-17481-1> (2020).
- Karpik, B. et al. Magnetic proximity in a van der Waals heterostructure of magnetic insulator and graphene. *2D Mater.* **7**, 015026 <https://doi.org/10.1088/2053-1583/ab5915> (2019).
- Zihlmann, S. et al. Large spin relaxation anisotropy and valley-zeeman spin-orbit coupling in wse₂/graphene/h-bn heterostructures. *Phys. Rev. B* **97**, 075434 <https://doi.org/10.1103/PhysRevB.97.075434> (2018).
- Song, K. et al. Spin proximity effects in graphene/topological insulator heterostructures. *Nano Lett.* **18**, 2033–2039 <https://doi.org/10.1021/acs.nanolett.7b05482> (2018).
- Safeer, C. K. et al. Room-temperature spin Hall effect in graphene/mos2 van der Waals heterostructures. *Nano Lett.* **19**, 1074–1082 <https://doi.org/10.1021/acs.nanolett.8b04368> (2019).
- Herling, F. et al. Gate tunability of highly efficient spin-to-charge conversion by spin Hall effect in graphene proximitized with WSe₂. *APL Mater.* **8**, 071103 <https://doi.org/10.1063/5.0006101> (2020).
- Pezo, A. et al. Manipulation of spin transport in graphene/transition metal dichalcogenide heterobilayers upon twisting. *2D Mater.* **9**, 015008 <https://doi.org/10.1088/2053-1583/ac3378> (2021).
- Manzeli, S., Ovchinnikov, D., Pasquier, D., Yazyev, O. V. & Kis, A. 2d transition metal dichalcogenides. *Nat. Rev. Mater.* **2**, 17033 <https://doi.org/10.1038/natrevmats.2017.33> (2017).
- Yang, H. X. et al. Proximity effects induced in graphene by magnetic insulators: first-principles calculations on spin filtering and exchange-splitting gaps. *Phys. Rev. Lett.* **110**, 046603 <https://doi.org/10.1103/PhysRevLett.110.046603> (2013).
- Hallal, A., Ibrahim, F., Yang, H., Roche, S. & Chshiev, M. Tailoring magnetic insulator proximity effects in graphene: first-principles calculations. *2D Mater.* **4**, 025074 <https://doi.org/10.1088/2053-1583/aa6663> (2017).
- Zollner, K., Faria Junior, P. E. & Fabian, J. Proximity exchange effects in mose₂ and wse₂ heterostructures with cr₂: twist angle, layer, and gate dependence. *Phys. Rev. B* **100**, 085128 <https://doi.org/10.1103/PhysRevB.100.085128> (2019).

26. Zollner, K. & Fabian, J. Engineering proximity exchange by twisting: reversal of ferromagnetic and emergence of antiferromagnetic Dirac bands in graphene/cr₂ge₂te₆. *Phys. Rev. Lett.* **128**, 106401 <https://doi.org/10.1103/PhysRevLett.128.106401> (2022).
27. Chau, T. K., Hong, S. J., Kang, H. & Suh, D. Two-dimensional ferromagnetism detected by proximity-coupled quantum Hall effect of graphene. *npj Quantum Mater.* **7**, 27 <https://doi.org/10.1038/s41535-022-00435-9> (2022).
28. Choi, E.-M., Sim, K. I., Burch, K. S. & Lee, Y. H. Emergent multifunctional magnetic proximity in van der Waals layered heterostructures. *Adv. Sci.* **9**, 2200186 <https://doi.org/10.1002/adv.202200186> (2022).
29. Ghiasi, T. S. et al. Quantum spin Hall effect in magnetic graphene. *Nat. Commun.* **16**, 5336 <https://doi.org/10.1038/s41467-025-60377-1> (2025).
30. Yang, H. et al. A seamless graphene spin valve based on proximity to van der Waals magnet Cr₂Ge₂Te₆. *Nat. Electron.* **8**, 15–23 <https://doi.org/10.1038/s41928-024-01267-0> (2025).
31. Sahani, D. et al. Giant gate-controlled odd-parity magnetoresistance in magnetized bilayer graphene at room temperature. *Phys. Rev. Lett.* **134**, 106301 <https://doi.org/10.1103/PhysRevLett.134.106301> (2025).
32. Tombros, N., Jozsa, C., Popinciuc, M., Jonkman, H. T. & van Wees, B. J. Electronic spin transport and spin precession in single graphene layers at room temperature. *Nature* **448**, 571–574 <https://doi.org/10.1038/nature06037> (2007).
33. Raes, B. et al. Determination of the spin-lifetime anisotropy in graphene using oblique spin precession. *Nat. Commun.* **7**, 11444 <https://doi.org/10.1038/ncomms11444> (2016).
34. Savero Torres, W. et al. Spin precession and spin Hall effect in monolayer graphene/Pt nanostructures. *2D Mater.* **4**, 041008 <https://doi.org/10.1088/2053-1583/aa8823> (2017).
35. Piquemal-Banci, M. et al. Spin filtering by proximity effects at hybridized interfaces in spin-valves with 2d graphene barriers. *Nat. Commun.* **11**, 5670 <https://doi.org/10.1038/s41467-020-19420-6> (2020).
36. Jeong, J. et al. Spin-selective memtransistors with magnetized graphene. *Adv. Mater.* **36**, 2310291 <https://doi.org/10.1002/adma.202310291> (2024).
37. Han, W. et al. Spin transport and relaxation in graphene. *J. Magn. Magn. Mater.* **324**, 369–381 (2012).
38. Zollner, K., Kurpas, M., Gmitra, M. & Fabian, J. First-principles determination of spin–orbit coupling parameters in two-dimensional materials. *Nat. Rev. Phys.* **7**, 255–269 <https://doi.org/10.1038/s42254-025-00818-4> (2025).
39. Han, W., Kawakami, R. K., Gmitra, M. & Fabian, J. Graphene spintronics. *Nat. Nanotechnol.* **9**, 794–807 <https://doi.org/10.1038/nnano.2014.214> (2014).
40. Zhang, J., Zhao, B., Yao, Y. & Yang, Z. Robust quantum anomalous Hall effect in graphene-based van der Waals heterostructures. *Phys. Rev. B* **92**, 165418 <https://doi.org/10.1103/PhysRevB.92.165418> (2015).
41. Zollner, K. et al. Scattering-induced and highly tunable by gate damping-like spin-orbit torque in graphene doubly proximitized by two-dimensional magnet cr₂ge₂te₆ and monolayer ws₂. *Phys. Rev. Res.* **2**, 043057 <https://doi.org/10.1103/PhysRevResearch.2.043057> (2020).
42. Averyanov, D. V. et al. High-temperature magnetism in graphene induced by proximity to EuO. *ACS Appl. Mater. Interfaces* **10**, 20767–20774 <https://doi.org/10.1021/acsami.8b04289> (2018).
43. Pandey, S. et al. Room-temperature anomalous Hall effect in graphene in interfacial magnetic proximity to EuO grown by topotactic reduction. *Phys. Rev. B* **108**, 144423 <https://doi.org/10.1103/PhysRevB.108.144423> (2023).
44. Pandey, S. et al. Proximity-mediated multi-ferroelectric coupling in highly strained EuO-graphene heterostructures. *Adv. Mater.* **37**, 2417669 <https://doi.org/10.1002/adma.202417669> (2025).
45. Zhang, J. et al. Strong magnetization and Chern insulators in compressed graphene/CrI₃ van der Waals heterostructures. *Phys. Rev. B* **97**, 085401 <https://doi.org/10.1103/PhysRevB.97.085401> (2018).
46. Cardoso, C., Soriano, D., García-Martínez, N. A. & Fernández-Rossier, J. Van der Waals spin valves. *Phys. Rev. Lett.* **121**, 067701 <https://doi.org/10.1103/PhysRevLett.121.067701> (2018).
47. Seyler, K. L. et al. Valley manipulation by optically tuning the magnetic proximity effect in WSe₂/CrI₃ heterostructures. *Nano Lett.* **18**, 3823–3828 <https://doi.org/10.1021/acs.nanolett.8b01105> (2018).
48. Farooq, M. U. & Hong, J. Switchable valley splitting by external electric field effect in graphene/crI₃ heterostructures. *npj 2D Mater. Appl.* **3**, 3 <https://doi.org/10.1038/s41699-019-0086-6> (2019).
49. Cardoso, C., Costa, A. T., MacDonald, A. H. & Fernández-Rossier, J. Strong magnetic proximity effect in van der Waals heterostructures driven by direct hybridization. *Phys. Rev. B* **108**, 184423 <https://doi.org/10.1103/PhysRevB.108.184423> (2023).
50. Zollner, K., Gmitra, M., Frank, T. & Fabian, J. Theory of proximity-induced exchange coupling in graphene on hbn/(co, ni). *Phys. Rev. B* **94**, 155441 <https://doi.org/10.1103/PhysRevB.94.155441> (2016).
51. Zollner, K., Cvitkovich, L., Silvili, R., Stier, A. V. & Fabian, J. Resonant magnetic proximity hot spots in co/hbn/graphene. *arXiv* <https://arxiv.org/abs/2601.07781> (2026).
52. Lazić, P., Belashchenko, K. D. & Žutić, I. Effective gating and tunable magnetic proximity effects in two-dimensional heterostructures. *Phys. Rev. B* **93**, 241401 <https://doi.org/10.1103/PhysRevB.93.241401> (2016).
53. Zanolli, Z. Graphene-multiferroic interfaces for spintronics applications. *Sci. Rep.* **6**, 31346 <https://doi.org/10.1038/srep31346> (2016).
54. Zollner, K. & Fabian, J. Bilayer graphene encapsulated within monolayers of ws₂ or cr₂ge₂te₆: tunable proximity spin-orbit or exchange coupling. *Phys. Rev. B* **104**, 075126 <https://doi.org/10.1103/PhysRevB.104.075126> (2021).
55. Beer, A. et al. Proximity-induced exchange interaction and prolonged valley lifetime in mose2/crsbr van-der-Waals heterostructure with orthogonal spin textures. *ACS Nano* **18**, 31044–31054 <https://doi.org/10.1021/acs.nano.4c07336> (2024).
56. Liu, T. J., Prestigiacomo, J. C. & Adams, P. W. Electrostatic tuning of the proximity-induced exchange field in EuS/Al bilayers. *Phys. Rev. Lett.* **111**, 027207 <https://doi.org/10.1103/PhysRevLett.111.027207> (2013).
57. Yang, B. et al. Electrostatically controlled spin polarization in graphene-crsbr magnetic proximity heterostructures. *Nat. Commun.* **15**, 4459 <https://doi.org/10.1038/s41467-024-48809-w> (2024).
58. Qiu, Z. et al. Visualizing atomic structure and magnetism of 2d magnetic insulators via tunneling through graphene. *Nat. Commun.* **12**, 70 <https://doi.org/10.1038/s41467-020-20376-w> (2021).
59. Tong, Q., Chen, M. & Yao, W. Magnetic proximity effect in a van der Waals moiré superlattice. *Phys. Rev. Appl.* **12**, 024031 <https://doi.org/10.1103/PhysRevApplied.12.024031> (2019).
60. Li, Y. & Koshino, M. Twist-angle dependence of the proximity spin-orbit coupling in graphene on transition-metal dichalcogenides. *Phys. Rev. B* **99**, 075438 <https://doi.org/10.1103/PhysRevB.99.075438> (2019).
61. David, A., Rakyta, P., Kormányos, A. & Burkard, G. Induced spin-orbit coupling in twisted graphene–transition metal dichalcogenide heterobilayers: twistrionics meets spintronics. *Phys. Rev. B* **100**, 085412 <https://doi.org/10.1103/PhysRevB.100.085412> (2019).
62. Kurganskii, S. I., Dubrovskii, O. I. & Domashevskaya, E. P. Integration over the two-dimensional Brillouin zone. *Phys. Status Solidi (b)* **129**, 293–299 <https://doi.org/10.1002/pssb.2221290129> (1985).

63. Bistritzer, R. & MacDonald, A. H. Moiré bands in twisted double-layer graphene. *Proc. Natl. Acad. Sci. USA* **108**, 12233–12237 <https://doi.org/10.1073/pnas.1108174108> (2011).
64. Pezzini, S. et al. Twisted bilayer graphene quasicrystals from chemical vapor deposition. *Nano Lett.* **20**, 3313–3319 <https://doi.org/10.1021/acs.nanolett.0c00172> (2020).
65. Giannozzi, P. et al. Quantum espresso: a modular and open-source software project for quantum simulations of materials. *J. Phys. Condens. Matter* **21**, 395502 <https://doi.org/10.1088/0953-8984/21/39/395502> (2009).
66. Dal Corso, A. Pseudopotentials periodic table: from h to pu. *Comput. Mater. Sci.* **95**, 337–350 (2014).
67. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **77**, 3865–3868 <https://doi.org/10.1103/PhysRevLett.77.3865> (1996).
68. Grimme, S., Antony, J., Ehrlich, S. & Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (dft-d) for the 94 elements h-pu. *J. Chem. Phys.* **132**, 154104 <https://doi.org/10.1063/1.3382344> (2010).
69. Gong, C. et al. Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals. *Nature* **546**, 265–269 <https://doi.org/10.1038/nature22060> (2017).
70. Bartók, A. P., Kondor, R. & Csányi, G. On representing chemical environments. *Phys. Rev. B* **87**, 184115 <https://doi.org/10.1103/PhysRevB.87.184115> (2013).
71. Himanen, L. et al. DScribe: library of descriptors for machine learning in materials science. *Comput. Phys. Commun.* **247**, 106949 <https://doi.org/10.1016/j.cpc.2019.106949> (2020).
72. Laakso, J. et al. Updates to the dscribe library: new descriptors and derivatives. *J. Chem. Phys.* **158**, 234802 (2023).
73. Caro, M. A. Optimizing many-body atomic descriptors for enhanced computational performance of machine learning based interatomic potentials. *Phys. Rev. B* **100**, 024112 <https://doi.org/10.1103/PhysRevB.100.024112> (2019).
74. Drautz, R. Atomic cluster expansion for accurate and transferable interatomic potentials. *Phys. Rev. B* **99**, 014104 <https://doi.org/10.1103/PhysRevB.99.014104> (2019).
75. Breiman, L. Random forests. *Mach. Learn.* **45**, 5–32 <https://doi.org/10.1023/A:1010933404324> (2001).
76. Pedregosa, F. et al. Scikit-learn: machine learning in Python. *J. Mach. Learn. Res.* **12**, 2825–2830 (2011).
77. Cvitkovich L. & Zollner K. CGT_Gr https://github.com/lukas-cvitkovich/CGT_Gr (Github 2026).
78. Humphrey, W., Dalke, A. & Schulten, K. VMD—visual molecular dynamics. *J. Mol. Graph.* **14**, 33–38 (1996).

Acknowledgements

This project has received funding from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) SPP 2244 (Project No. 443416183), SFB 1277 (Project No. 314695032), and the EU 2DSPIN-TECH Program (Graphene Flagship).

Author contributions

L.C. orchestrated improvements in the DFT training set, trained the ML model, analyzed the data, prepared all figures, and wrote the manuscript. K.C. generated the DFT training set and reviewed the ML predictions and the manuscript. J.F. supervised the project and reviewed the manuscript.

Funding

Open Access funding enabled and organized by Projekt DEAL.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41524-026-02346-w>.

Correspondence and requests for materials should be addressed to Lukas Cvitkovich.

Reprints and permissions information is available at <http://www.nature.com/reprints>

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

© The Author(s) 2026