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Heterobilayer moiré magnets: moiré skyrmions and the commensurate-incommensurate transition

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We study untwisted heterobilayers of ferromagnetic and antiferromagnetic van der Waals materials, with in particular a Dzyaloshinskii-Moriya interaction in the ferromagnetic layer. A continuum low energy field theory is utilized to study such systems. We develop a phase diagram as a function of the strength of inter-layer exchange and Dzyaloshinskii-Moriya interactions, combining perturbative and strong coupling analyses with numerical simulations using Landau-Lifshitz-Gilbert equations. Various moiré-periodic commensurate phases are found, and the commensurate-incommensurate transition is discussed. Among the commensurate phases, we observe an interesting skyrmion lattice phase wherein each moiré unit cell hosts one skyrmion.

Magnetic skyrmions are long-lived, topologically protected spin-textures that were predicted to exist in chiral magnets [1–3], and experimentally observed in cubic, non-centrosymmetric materials such as MnSi [4], Fe_{1-x}Co_xSi [5, 6] and FeGe [7]. Non-collinear spin configurations occur in these materials due to the antisymmetric Dzyaloshinskii-Moriya (DM) interactions [8, 9]. In three-dimensional bulk magnets, skyrmion lattices are stabilized by thermal fluctuations above the helical state [4]. Interestingly, in the thin-film, two-dimensional limit, the skyrmion lattice is stable over a wide range of the phase diagram [10–12].

A new class of two-dimensional crystals, magnetic van der Waals (vdW) materials, has opened up numerous possibilities for both theoretical and experimental physics [13, 14]. All three fundamental spin Hamiltonians have been reported in these materials: the two-dimensional Heisenberg, Ising, and XY models [15, 16]. Furthermore, novel quantum phases are expected to appear in the heterostructures of these materials [17]. Recently, skyrmion crystals have been observed in the single-layered ferromagnetic vdW material Fe₃GeTe₂ [18–20], predicted to exist in CrI₃ [21] and Janus magnets [22], all attributed to the DM interaction, motivating us to study the moiré physics of such vdW systems.

We will focus on a nontwisted heterobilayer of ferromagnetic (harboring a DM interaction) and antiferromagnetic vdW materials. A similar construction was explored in [23], where dipolar interactions along with an external field were added to the moiré structure in contrast to the DM interaction of the present work. We show that a wealth of different phases featuring magnetic textures on the moiré scale and in particular a skyrmion lattice phase could form.

We will use the continuum formalism introduced in [17], which obviates the need to consider numerous lattice sites with complicated local environments. First, we review this continuum formalism and derive the Hamiltonian that we will focus on, followed by a perturba-

tive analysis. The competition between the DM interaction and the moiré potential induces a commensurate-incommensurate transition that will further be analyzed in the weak-coupling regime. Then different types of commensurate phases are introduced, supplemented furthermore by a numerical ground-state phase diagram, obtained from a Landau-Lifshitz-Gilbert analysis. Finally, we discuss the possible extensions and experimental relevance of the work. Details of the weak-coupling analysis will be presented in [24].

Setup of the problem—We study a heterobilayer system of honeycomb ferromagnetic and honeycomb antiferromagnetic vdW materials. This setting is similar to that in [23]. Both layers are assumed to exhibit long-range order, so a local description in terms of the order parameters $\mathbf{M}$, $\mathbf{N}$ for the ferromagnetic and the Néel antiferromagnetic layers will be employed, where $|\mathbf{M}| = |\mathbf{N}| = 1$. After [17], we will develop a continuum model to provide a low energy description of the system. When the interlayer coupling and the displacement gradients act as small perturbations on the intrinsic magnetism of the two layers, a continuum treatment is justified. Since we will only be concerned with ground state configurations (and possible other nearby states) in this work, it suffices to analyze the classical Hamiltonians only. This consists of intralayer and interlayer terms; we take the former as

$$\mathcal{H}_{\text{intra}} = \frac{\rho_1}{2}(\nabla \mathbf{M})^2 + \frac{\rho_2}{2}(\nabla \mathbf{N})^2 + D \mathbf{M} \cdot (\nabla \times \mathbf{M}) - C (N_z)^2, \quad (1)$$

where a single-ion anisotropy for the antiferromagnetic layer and a DM interaction for the ferromagnetic layer are assumed; the latter is predicted to be present in families of Janus magnets in particular [22]. We have utilized a Bloch-type DM interaction in this work, however the same results hold for a Néel-type DM interaction as well [24]. The anisotropy term is neglected in the ferromagnetic layer.

We now turn to the interlayer coupling: the spin

densities of the two layers are parametrized as: $\mathcal{S}_1 = m_0 \mathbf{M} [f_0 - \sum_a \cos(\mathbf{b}_a \cdot \mathbf{r})]$ and $\mathcal{S}_2 = n_0 \mathbf{N} \sum_a \sin(\mathbf{c}_a \cdot \mathbf{r})$, where $\mathbf{b}_a, \mathbf{c}_a$ denote the reciprocal lattice vectors of the microscopic lattices in the two layers. Furthermore, m_0 and n_0 are proportional to the ordered moments in the two layers, and the zeroth Fourier component f_0 is undetermined; its value will not matter for the low-energy physics. Taking a minimal Fourier expansion, we limit the summation over $\mathbf{c}_a$ and $\mathbf{b}_a$ to the three smallest reciprocal lattice vectors. Assuming a local inter-layer exchange of the form $H' = -J' \mathcal{S}_1 \cdot \mathcal{S}_2$ and that $|\mathbf{b}_a| - |\mathbf{c}_b| \ll |\mathbf{b}_a|, |\mathbf{c}_b|$, one arrives at the following inter-layer interaction: $H' = J' \mathbf{M} \cdot \mathbf{N} \sum_a \sin(\mathbf{d}_a \cdot \mathbf{r})$, where the three vectors $\mathbf{d}_a = \mathbf{b}_a - \mathbf{c}_a$ represent the reciprocal vectors of an emergent moiré lattice, and the fast-oscillating terms have been omitted. We would also like to mention in passing that considering a triangular lattice ferromagnet also leads to the same interlayer coupling.

$\mathcal{H} = \mathcal{H}_{\text{intra}} + \mathcal{H}'$ describes the system in the general setting described above. However, for simplicity, in the remainder of this work we make the assumption that spin stiffness and anisotropy parameters of the antiferromagnetic layer are large enough to render it essentially non-dynamical; this results in a uniform Néel vector in the $\pm \hat{z}$ directions; we choose $\mathbf{N} = +\hat{z}$. With these assumptions, the Hamiltonian reduces to an effective single-layer one, which, up to a constant rescaling, has the form:

$$\mathcal{H} = \frac{1}{2}(\nabla \mathbf{M})^2 + \beta \mathbf{M} \cdot (\nabla \times \mathbf{M}) + \alpha M_z \Phi(\mathbf{x}). \quad (2)$$

We have introduced the dimensionless parameters $\alpha = \frac{J'}{\rho_1 d^2}$, $\beta = \frac{D}{\rho_1 d}$ with $d = |\mathbf{d}_a|$, and adopted the dimensionless coordinates $\mathbf{x} = d \mathbf{r}$ and defined $\Phi(\mathbf{x}) = \sum_a \sin(\hat{\mathbf{d}}_a \cdot \mathbf{x})$.

When $\alpha = 0$, ground states of the system consist of spirals with wavenumber β (period $2\pi/\beta$). Generically, $\mathbf{M} = (\hat{\mathbf{q}} \times \hat{z} \sin[\mathbf{q} \cdot \mathbf{x}], \cos[\mathbf{q} \cdot \mathbf{x}])$ describes a spiral propagating in the $\hat{\mathbf{q}}$ -direction for $\beta > 0$ (and $|\mathbf{q}| = \beta$). In the strong-coupling limit when α is large and β is zero, the energy is minimized by a coplanar solution, where domains with $\mathbf{M}$ close to $(0, 0, \pm 1)$ form, separated by narrow domain walls. Each domain is identified as a region in which $\Phi(\mathbf{x})$ has a definite sign, resulting in the opposite sign for M_z . In Fig. 1a, we plot the structure of $\Phi(\mathbf{x})$, which forms a triangular lattice; $\Phi(\mathbf{x})$ has a positive or negative sign in each of the faces of the triangles.

Weak-coupling Analysis— In the small α and β regime, two possibilities can arise: when the α term is dominant over β , a *twisted* solution forms, where in each face of the triangular lattice the magnetization tends to bend towards $-\text{sign}[\Phi(\mathbf{x})]\hat{z}$; nonzero β causes distortions in the $\beta = 0$ coplanar solution. We call the phase containing these solutions as twisted-0 for a reason that will be explained later. Alternatively when the β term dominates, one expects a spiral configuration perturbed by the moiré potential. Since the spiral can have a period that is dif-

ferent from that of the moiré lattice, we call this the incommensurate phase. A commensurate-incommensurate (CI) transition can occur between these two phases. Below we perform a perturbative solution in the commensurate case, and then extend the analysis to the incommensurate phase and also the transition between the two phases.

We will use a polar coordinates parametrization $\mathbf{M} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ for the minimization of the classical Hamiltonian (2). To zeroth order in β , the solution to the Euler-Lagrange equation (see [24] for a complete discussion) yields a constant value for ϕ : $\phi(\mathbf{x}) = \phi^{(0)} + O(\beta\alpha)$. The simplified Euler-Lagrange equation for θ then reads $\nabla^2 \theta + \alpha \sin \theta \Phi(\mathbf{x}) = 0$, with the solution to zeroth order in β taking the form $\theta(\mathbf{x}) = \frac{\pi}{2} + \alpha \Phi(\mathbf{x}) + O(\alpha^3, \beta^2\alpha)$; this solution requires a definite mean value for θ , i.e. $\bar{\theta} = \pi/2$. It furthermore predicts that in the twisted-0 phase, fluctuations of θ from $\pi/2$ are captured by $\Phi(\mathbf{x})$ and hence triangular domains form in which spins prefer either up or down directions. We have checked numerically that the solution even with infinitesimal β , prefers only three values for $\phi^{(0)}$, i.e. $e^{i\phi^{(0)}} = [\hat{d}_y - i\hat{d}_x]^n$, with $n = 1, 2, 3$.

We now turn to a perturbative study of the incommensurate phase and the CI transition; we will focus on the incommensurate solutions close to the CI transition line. If α plays a subdominant role, the solution resembles a spiral with a period close to $\frac{2\pi}{\beta}$; this period increases as β is lowered towards the CI transition. Close to the transition point, locally the configuration looks similar to a commensurate one but small-width discommensurations (or solitons) can form with large separations. It is this separation distance that diverges at the CI transition [25]. We exploit this key information to find incommensurate solutions close to the transition by considering configurations that are locally very close to being commensurate but their local properties could change appreciably if a distance close to the long incommensurate periodicity is taken.

To this end, we make use of the commensurate solution found above, in which θ has the mean value $\bar{\theta} = \pi/2$; we now allow this mean value to fluctuate slowly as the position is varied. As a result, we work with a modified Euler-Lagrange equation $\nabla^2 \theta + \alpha \sin \theta \Phi(\mathbf{x}) = \lambda$, where λ is a Lagrange multiplier. This equation can be solved, and one finds the energy density per unit cell of such a periodic solution to have the form $-\frac{3}{4}\alpha^2 \sin^2 \bar{\theta}$; note that this is minimized for $\bar{\theta} = \pi/2$. Letting $\bar{\theta}$ fluctuate over long distances will result in an energy penalty; we require this energy penalty to be compensated by the DM interaction energy gain in the incommensurate solutions. In fact an effective one-dimensional Hamiltonian of the form $\mathcal{H}^{\text{eff}} = \frac{1}{2}(\partial_y \bar{\theta})^2 - \frac{3}{4}\alpha^2 \sin^2 \bar{\theta} - \beta \partial_y \bar{\theta}$, can be exploited to study this competition. This sine-Gordon Hamiltonian has been extensively studied, see e.g. [25], and is

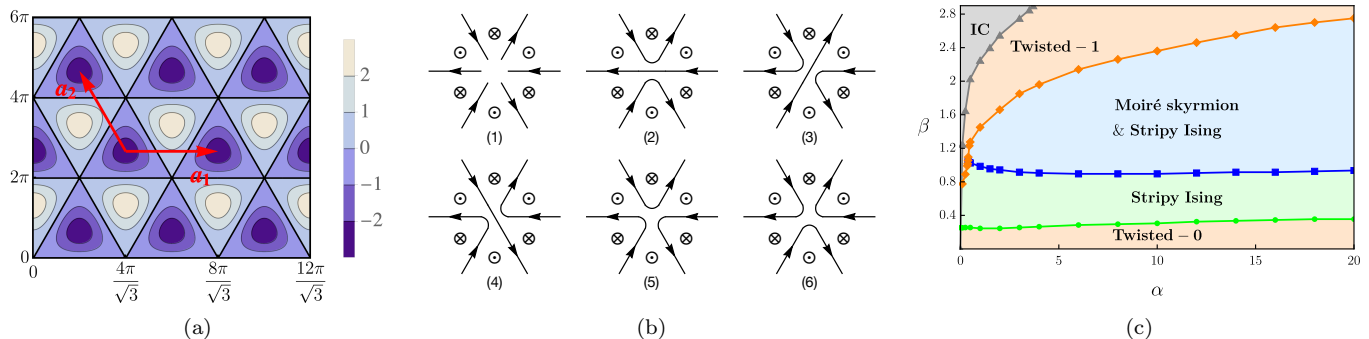


FIG. 1. (a) The moiré potential $\Phi(\mathbf{x})$. $\mathbf{a}_1$ and $\mathbf{a}_2$ form a basis for the emergent moiré lattice. (b) (1) An avoided vertex at large α and β . The magnetization vectors on the domain walls point along them. (2-4) The 3-state clock vertex. (5-6) The Ising vertex. (c) The ground-state phase diagram obtained from LLG equations. IC stands for incommensurate phases which can include states of various periodicities. In the blue region of intermediate β , the moiré skyrmion and the stripy Ising states are degenerate in energy. Different commensurate phases are separated by first-order transitions.

known to have a continuous CI transition at $\beta_c = \frac{\sqrt{6}}{\pi}\alpha$. This means that the phase separation line between the commensurate and the incommensurate phases has a linear form for small α and β . For $\beta \gtrsim \beta_c$, the ordering wavevectors deviate from the moiré ones (including the vanishing wavevector) with the asymptotic behavior $\delta k \sim \frac{\sqrt{3}\alpha}{2} \frac{1}{\log\left(\alpha^2 \frac{1}{\beta - \beta_c}\right)}$; the direction of this incommensurate wavevector correction is not fixed at this order of perturbation theory.

Possible Commensurate Phases— We now turn to the large- α regime where the properties of different commensurate phases are in most contrast. In this regime, triangular domains of spins mostly pointing in the $\pm\hat{z}$ directions form. These domains correspond to those in Fig. 1(a) with $\text{sign}[\Phi(x)] = \mp 1$, respectively. The widths of the domain walls (in dimensionless units) decrease as α increases, resulting from the competition of the kinetic and moiré potential terms. On the domain walls $\Phi(x) \approx 0$, thus the magnetization is not constrained by the α term. It is actually the different configurations that the magnetization could take on these domain walls that distinguish the different commensurate phases. Starting at $\beta = 0$, the energy is minimized by a coplanar configuration wherein for all points, the in-plane magnetization can take any direction, i.e. there is an $SO(2)$ rotational symmetry.

Next we consider the effect of nonzero β . The DM term, up to integration by parts, can be written as $-2\beta \mathbf{M} \cdot (\hat{z} \times \nabla M_z)$. Namely, $\mathbf{M}$ prefers to be perpendicular to the gradient of M_z , which lies normal to the domain walls. When β is not large, it does not affect configurations deep inside the triangular domains, but drives the magnetization vectors on the triangles' edges to point along them in the direction preferred by the DM interaction, as shown in Fig. 1b(1).

However, the configurations close to the vertices are frustrated, i.e., the three directions of magnetization vec-

tors along the edges of the triangles cannot be satisfied at the same time, and hence different vertex configurations could arise. One possibility is that the vertex magnetization follows one of the three directions on the edges, breaking the $SO(2)$ symmetry down to C_3 , as shown in Figs. 1b(2-4); we name this a 3-state clock vertex. Interestingly, a solution consisting of the same vertex configuration everywhere has the same symmetries as those of the twisted-0 solution discussed above. We call the phase containing this solution twisted-1 for reasons explained below.

Another possibility of the vertex configuration is Ising-like, i.e. spins pointing along $+\hat{z}$ or $-\hat{z}$ as in Figs. 1b(5) and 1b(6), enabling the possibility of skyrmions: if all the vertices choose the same configuration, say 1b(6), a skyrmion lattice of moiré scale is formed: triangular domains with up spin constitute a lattice of skyrmions in a down-spin sea (see also Fig. 2).

The skyrmion lattice phase can be viewed as stabilized due to an effective ferromagnetic interaction between neighboring Ising-type vertices, however it could happen that this interaction becomes antiferromagnetic, making a translational-symmetry-broken phase possible: Our numerical results (see below) indicate that one of such phases is possible, where the Ising vertices all point in the same direction along the direction specified by one of the moiré lattice unit vectors, while along the directions of other independent unit vectors, the Ising vertices alternatively point up and down (see Fig. 2). We name it as the Ising stripy phase.

Symmetries of these solutions are discussed in [24]. We go through the full Hamiltonian minimization next and determine the energetically favored solution for different parameter choices.

Numerical minimization—To minimize the energy functional in (2), we work with the Landau-Lifshitz-Gilbert (LLG) equations [26, 27]. The variation of the Hamilto-

nian with respect to the magnetization vector is treated as an effective external field, $\mathbf{B}^{\text{eff}} = -\delta H/\delta \mathbf{M}$, which results in the LLG equations: $\frac{d\mathbf{M}}{dt} = -g \mathbf{M} \times \mathbf{B}^{\text{eff}} + \eta \mathbf{M} \times \frac{d\mathbf{M}}{dt}$. Here g is the gyromagnetic ratio, which is not important since we are only interested in the late-time static configurations, and η is the Gilbert damping coefficient. Releasing from various trial configurations and comparing the energies of the final configurations, one can find the ground state with of the Hamiltonian. Fig. 1c shows the numerical phase diagram obtained from LLG equations using the *ubermag* and *OOMMF* packages [28–30]. Different periodic boundary conditions compatible with moiré lattice periodicities are imposed. More details on the numerics are presented in [24].

We find that apart from the incommensurate phases, the twisted-0, the twisted-1, the moiré skyrmion and the Ising stripy phases introduced above constitute the main body of the phase diagram in the strong coupling regime. In Fig. 2, we present examples of steady-state configurations for these three phases (note that the shown configurations are not necessarily ground states for the chosen parameters). In the moiré skyrmion phase, interestingly, there is one skyrmion per moiré unit cell with skyrmion number ± 1 and helicity $\pi/2$, i.e. $N_{sk} = \frac{1}{4\pi} \iint_{u.c.} d^2\mathbf{x} \mathbf{M} \cdot \left(\frac{\partial \mathbf{M}}{\partial x} \times \frac{\partial \mathbf{M}}{\partial y} \right) = \pm 1$; the sign is different for the two types of skyrmion lattices, i.e. it depends on the direction of magnetization ($+\hat{z}$ or $-\hat{z}$) at the centers of the skyrmions in each solution.

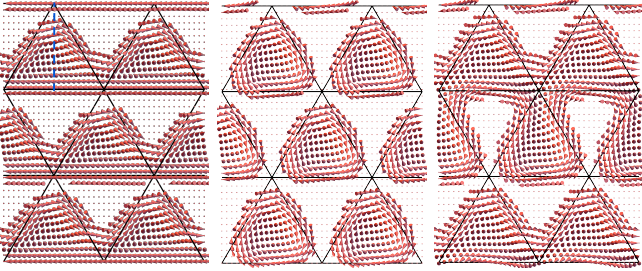


FIG. 2. Top view of the twisted-1 (left), skyrmion lattice (middle) and the stripy Ising (right) configurations generated at $(\alpha, \beta) = (1.25, 1)$, $(1.25, 1)$ and $(2.5, 0.5)$, respectively. The background $-\hat{z}$ pointing magnetization vectors are depicted as points. The vertices are of the type shown in Fig. 1b(2) for the twisted-1, of the Fig. 1b(6) type for the skyrmion lattice and of both 1b(5) and 1b(6) types for the Ising stripy. The solution on the left panel shows a nonzero winding in its magnetization as the dashed line is traversed (see the main text).

We see that the twisted-0 phase has the lowest energy for small β as expected. Upon increasing β , it is then replaced by the stripy Ising phase. Interestingly, at larger β , the stripy Ising and the moiré skyrmion states, despite having different symmetries, become degenerate in energy. Note that these two states both have Ising-type vertices that are only spatially arranged differently.

Then at higher values of β , the twisted-1 phase, that has vertex configuration 1b(2-4) everywhere, minimizes the energy. This phase has the same symmetries as the twisted-0 phase, but is different in the winding numbers along high symmetry lines: The magnetization vector rotates exactly by 2π in the $x - z$ plane along the dashed blue line segment specified in the left panel of Fig. 2. On the other hand, the net winding is zero in the twisted-0 phase (see [24] for a strong coupling figure of the twisted-0 phase), hence the names twisted-0 and twisted-1. In principle, other winding numbers can also be possible (similarly all other solutions can also have higher winding numbers as well). All transitions between these commensurate phases are first order, which can be observed by comparing the energies. More details can be found in [24].

In the upper left corner of the phase diagram, we have found different incommensurate phases, i.e., states incommensurate with the moiré potential. For example when $\alpha = 0$, the periodicity of the spirals is given by $2\pi/\beta$. In general, the competition between α and β can lead to configurations with various periods and symmetries, and even chaotic structures [31]. Since, in the LLG equations, we cannot exhaust all possible trial configurations and unit cells enlargements, and thus the extent of the incommensurate phases is underestimated; as a result, we take a cautious view and do not make definitive conclusions regarding the nature of this region or its phase boundary, except what can be extracted from the weak-coupling analysis presented above. In this weak-coupling regime, the prediction for the CI transition line is supported using numerics with large sample sizes, see the supplemental material [24].

Discussion—A considerable portion of the phase diagram Fig. 1c for $\alpha > 3$ is occupied by phases with the Ising type vertices. In this paper, we have ignored the single-ion anisotropy for the ferromagnetic layer; such anisotropy, if Ising-like, further stabilizes Ising-type vertices. On the other hand, a perpendicular magnetic field will similarly stabilize the Ising vertices, and in particular favor the skyrmion lattice phase over the stripy Ising phase in the blue region of the phase diagram in Fig. 1c; this is consistent with the well-known fact that external fields can stabilize skyrmion lattice phases [1, 2, 10–12].

We have made the assumption that the spin-stiffness and anisotropy parameters of the antiferromagnetic layer are large, such that the antiferromagnetic layer is in the collinear phase. We expect this assumption to be valid in the regime where $C/(\rho_2 d^2) \gtrsim \alpha^2$ and $\rho_2/\rho_1 \gtrsim 1$, as analyzed in our previous work [17]. Relaxing this assumption could lead to ground state solutions requiring nontrivial textures in both layers resembling those in [17]. Furthermore, let us mention that a small twisting between the two layers effectively increases the magnitude of the wavevector d introduced above. In the dimensionless parametrization, this corresponds to a modification

of the spatial structure of $\hat{\Phi}(\mathbf{x})$, and a decrease in α and β .

The material MnPS_3 , having an antiferromagnetic Heisenberg Hamiltonian and an Ising-like anisotropy [15, 32–34], may be a good candidate for the antiferromagnetic layer. The family of Janus transition metal dichalcogenides [35] could be a promising candidate for the FM layer; these materials have small lattice mismatches compared with MnPS_3 and also exhibit the DM interaction [22].

Interesting phenomena such as the topological Hall effect could arise due to the effect of moiré skyrmions on the conduction electrons through the emergent electromagnetic field (see for example [36, 37]). We leave the full description of the related physics to future work.

Note added: During the preparation of this draft, we noticed [38] which has some overlap with our work. Their focus is mainly on the LLG simulation of commensurate phases, however, other strong-coupling commensurate phases, incommensurate phases and in particular the commensurate-incommensurate transition studied here are not captured. In particular, the Ising stripy phase which we found to be degenerate with the skyrmion phase is absent their treatment.

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