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Breaking through the Mermin-Wagner limit in 2D van der Waals magnets

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¹⁸ The Mermin-Wagner theorem ¹ states that long-range magnetic order does not exist in one-¹⁹ or two-dimensional (2D) isotropic magnets with short-ranged interactions. The theorem has ²⁰ been a milestone on magnetism and has been driving the research of recently discovered 2D

van der Waals (vdW) magnetic materials^{2,3} from fundamentals up to potential applications⁴. 21 In such systems, the existence of magnetic ordering is typically attributed to the presence of a 22 significant magnetic anisotropy, which is known to introduce a spin-wave gap and circumvent 23 the core assumption of the theorem^{1,5,6}. Here we show that in finite-size 2D vdW magnets 24 typically found in lab setups (e.g., within millimetres), short-range interactions can be large 25 enough to allow the stabilisation of magnetic order at finite temperatures without any mag-26 netic anisotropy for practical implementations. We demonstrate that magnetic ordering can 27 be created in flakes of 2D materials independent of the lattice symmetry due to the intrinsic 28 nature of the spin exchange interactions and finite-size effects in two-dimensions. Surpris-29 ingly we find that the crossover temperature, where the intrinsic magnetisation changes from 30 superparamagnetic to a completely disordered paramagnetic regime, is weakly dependent on 31 the system length, requiring giant sizes (e.g., of the order of the observable universe $\sim 10^{26}$ 32 m) in order to observe the vanishing of the magnetic order at cryogenic temperatures as ex-33 pected from the Mermin-Wagner theorem. Our findings indicate exchange interactions as 34 the main driving force behind the stabilisation of short-range order in 2D magnetism and 35 broaden the horizons of possibilities for exploration of compounds with low anisotropy at an 36 atomically thin level. 37

38 Introduction

The demand for computational power is increasing exponentially, following the amount of data generated across different devices, applications and cloud platforms^{7,8}. To keep up with this trend,

smaller and increasingly energy-efficient devices must be developed which require the study of 41 compounds not yet explored in data-storage technologies. The discovery of magnetically stable 2D 42 vdW materials could allow for the development of spintronic devices with unprecedented power 43 efficiency and computing capabilities that would in principle address some of these challenges⁴. 44 Indeed, the magnetic stability of vdW layers has been one of the central limitations for finding 45 suitable candidates, given that strong thermal fluctuations are able to rule out any magnetism. 46 As it was initially pointed out by Hohenberg⁶ for a superfluid or a superconductor, and extended 47 by Mermin and Wagner¹ for spins on a lattice, long-range order should be suppressed at finite 48 temperatures in the 2D regime, when only short-range isotropic interactions exist. Importantly, 49 the theorem only excludes long-range magnetic order at finite temperature in the thermodynamic 50 limit¹, *i.e.*, for infinite system sizes. However, the common understanding is that the theorem also 51 excludes the alignment of spins in samples studied experimentally which are a few micrometers in 52 size^{9,10}, suggesting that such systems are indistinguishable from infinite. 53

The long-range order characterising infinite systems only becomes distinguishable from 54 short-range order describing the local alignment of the spins if the system size exceeds the cor-55 relation length at a given temperature¹¹. Previous numerical studies and the scaling analysis of 56 2D Heisenberg magnets¹²⁻¹⁵ have established that although only short-range order is observable at 57 finite temperature, the spin correlation length can be larger than the system size below some finite 58 crossover temperatures. An intriguing question on this long-range limit is how can we understand 59 real-life materials, which routinely have a finite size L (Fig. 1a), in light of the Mermin-Wagner 60 theorem. It is known that thermal fluctuations will affect the emergence of a spontaneous mag-61

netisation at low dimensionality. Nevertheless, it is unclear which kind of spin ordering can be foreseen in thin vdW layered compounds when finite-size effects and exchange interactions play together. With recent advances in computational power and parallelisation scalability, it is possible to directly model magnetic ordering processes and dynamics of 2D materials on the micrometre length-scale accessible experimentally. Here, we demonstrate that short-range order can exist in systems with no anisotropy even down to the 2D limit using computer-intensive atomistic simulations and analytical models.

69 **Results**

We start by defining the magnetization in our systems as:

$$\mathbf{m} = \frac{1}{N} \sum_{i} \mathbf{S}_{i},\tag{1}$$

where S_i denotes the classical spin unit vector at lattice site *i* and *N* is the number of sites. In the 70 absence of external magnetic fields, the expectation value of the magnetization $\langle \mathbf{m} \rangle$ vanishes in 71 any finite-size system due to time-reversal invariance. Yet, 3D systems of only a few nanometres 72 in size that are far from infinite have been studied for decades and exhibit a clear crossover from a 73 magnetically ordered to a paramagnetic phase ^{16,17}. The Mermin-Wagner theorem establishes that 74 $\langle \mathbf{m} \rangle$ must also be zero in infinite 2D systems with short-ranged isotropic interactions. However, 75 for practical implementations it is relevant to unveil whether the average magnetisation vanishes 76 because the spins are completely disordered at any point in time, or if they are still aligned on 77 short distances but the overall direction of the magnetisation m strongly suffers time-dependent 78

⁷⁹ variation. Short-range order may be characterised by the intrinsic magnetisation¹⁸:

$$\langle |\mathbf{m}| \rangle = \left\langle \sqrt{\left(\frac{1}{N}\sum_{i}\mathbf{S}_{i}\right)^{2}} \right\rangle,$$
(2)

which is always positive by definition. The intrinsic magnetisation is close to 1 ($\langle |\mathbf{m}| \rangle = 1$) in the short-range-ordered regime and converges to zero when the spins become completely disordered^{9,19,20}.

For simplicity we first consider a 2D honeycomb lattice (Fig. 1a) to model the magnetic ordering process for a large flake of $1000 \times 1000 \text{ nm}^2$. Such a symmetry is very common in several vdW materials holding magnetic properties and interfaces⁴, such as Cr₂Ge₂Te₆ (CGT) or CrI₃ in which 2D magnetic ordering was first discovered^{2,3}. The system consists of 8 million atoms with nearest-neighbor Heisenberg exchange interactions $(J_{ij}/k_{\rm B} = 70.8 \text{ K})$ and no magnetic anisotropy (K) using highly accurate Monte Carlo simulations (see Supplementary Sections 1-2 for details). The magnitude of $J_{ij}/k_{\rm B}$ is within the same range as those observed for CGT (with a critical temperature of 66 K)² where a negligible magnetic anisotropy (< 1 μ eV) was observed for thin layers but a stable magnetic signal was still measured at finite temperatures (~ 4.7 $(K)^2$. Indeed, as we show below, thermal fluctuations do not destroy the short-range magnetic order but rather induce the formation of strongly correlated spins at large spatial scale. We use an isotropic Heisenberg spin Hamiltonian $\mathscr{H} = -\sum_{i < j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$ as stated in the Mermin-Wagner theorem¹. We begin by assessing the existence of any magnetic order at non-zero temperatures by equilibrating the system for 39×10^6 Monte Carlo steps using a uniform sampling²¹ to avoid any potential bias before a final averaging at thermal equilibrium for a further 10⁶ Monte Carlo steps. Strikingly, a crossover between the low-temperature short-range-ordered regime and the completely disordered state ($\langle |\mathbf{m}| \rangle \approx 0$) is observed at nonzero temperatures (Fig. 1b) and zero magnetic anisotropy (K = 0). To estimate the crossover temperature (T_x), the simulation data was fitted by the Curie-Bloch equation in the classical limit⁹:

$$\langle |\mathbf{m}| \rangle(T) = \left(1 - \frac{T}{T_x}\right)^{\beta},$$
(3)

where T is the temperature and β is the crossover magnetization exponent. From the fitting one 82 obtains $T_x = 23.342 \pm 0.237$ K ($\beta = 0.54 \pm 0.020$), which is about one third of the mean-field (MF) 83 critical temperature $T_c^{\text{MF}} = zJ_{ij}/(3k_B) = 70.8 \text{ K}$ (where z = 3 is the number of nearest neighbours) 84 even for this considerable system size. The simulations were then repeated including magnetic 85 anisotropy ($K = 1 \times 10^{-24}$ J/atom) which resulted in a slight increase in the crossover temperature 86 $(T_x = 26.543 \pm 0.320 \text{ K}, \beta = 0.427 \pm 0.021)$ (Fig. 1b). We observed that this difference in T_x be-87 tween isotropic and anisotropic cases becomes negligible as the flake size is reduced (100×100 88 nm²) with minor variations of the curvature of the magnetisation versus temperature (Supplemen-89 tary Section 3 and Supplementary Figure 1). We also checked that different Monte Carlo sampling 90 algorithms (i.e., adaptive) and starting spin configurations (i.e., ordered, disordered) do not modify 91 the overall conclusions (Supplementary Section 4 and Supplementary Figure 2). Taking dipolar in-92 teractions into account only has a minor effect on the intrinsic magnetization curve (Supplementary 93 Figure 3). Although the magnetocrystalline anisotropy K or the dipolar interactions circumvent the 94 Mermin-Wagner theorem and lead to a finite critical temperature, this indicates that systems up to 95 lateral sizes of 1 μm are not suitable for observing the critical behaviour. Instead the crossover in 96 the short-range order defined by the isotropic interactions dominates in this regime, regardless of 97

⁹⁸ whether the anisotropy is present or absent. Previous studies on finite magnetic clusters on metallic ⁹⁹ surfaces^{22,23} suggested that anisotropy is not the key factor in the stabilisation of magnetic prop-¹⁰⁰ erties at low dimensionality and finite temperatures, but rather it determines the orientation of the ¹⁰¹ magnetisation.

Even though short-range interactions can stabilise short-range magnetic order in 2D vdW 102 magnetic materials, this does not necessarily imply that the direction or the magnitude of the 103 magnetisation is stable over time. As thermally activated magnetisation dynamics may poten-104 tially change spin directions²⁴, it is important to clarify whether angular variations of the spins 105 are present. Hence we compute the time evolution of the magnetisation along different directions 106 (x, y, z) and its angular dependence (Fig. 1c,d) through the numerical solution of the Landau-107 Lifshitz-Gilbert equation (see Methods for details). Over the whole simulation (40 ns), all compo-108 nents of the magnetisation assume approximately constant values which deviates by $\pm 5^{\circ}$ from the 109 mean direction θ_{av} . Similar analyses undertaken for different flake sizes ($L \times L$, L = 50, 100, 500 110 nm) show that the spin direction is very stable at each temperature considered (2.5 K, 10 K, 20 K, 111 30 K, 40 K) and follows a Boltzmann distribution (Supplementary Section 5 and Supplementary 112 Figure 4). These results show that the magnetisation in a 2D isotropic magnet is not only stable in 113 magnitude but its direction only negligibly varies over time. 114

An outstanding question raised by the modelling of the 2D finite flakes is whether other kind of common lattice symmetries (*i.e.*, hexagonal, square), lower dimensions (*i.e.*, 1D) and different sizes may follow similar behaviour to that found in the honeycomb lattice. Figure 2 shows that the effect is universal regardless of the details of the lattice or the dimension considered. We find persistent magnetic order for T > 0 K at zero magnetic anisotropy for the cases considered. There is a consistent reduction in the crossover temperature as a function of the system size $L \rightarrow \infty$ in agreement with the general trend of the temperature dependence of the correlation length discussed above (Fig. 2**a-c**). The 1D model (atomic chain) displays a similar trend (Fig. 2**d**) although the variation of $\langle |\mathbf{m}| \rangle$ with *T* is different due to the lower dimensionality.

To give an analytical description of these effects, we use the anisotropic spherical model 124 (ASM) for the calculation of the finite-size effects on the intrinsic magnetization^{18,25,26} (see Sup-125 plementary Section 7 for details). The ASM takes into account Goldstone modes in the system and 126 self-consistently generates a gap in the correlation functions which avoids infrared divergences re-127 sponsible for the absence of long-range order for isotropic systems in dimensions $d \le 2$ as $L \to \infty$ 128 as per the Mermin-Wagner theorem. We applied the formalism to 1D and 2D systems for the 129 isotropic Heisenberg Hamiltonian in the absence of an external magnetic field¹⁸. The results of our 130 analytical calculations are shown as shaded regions in Fig. 2 (see Supplementary Section 7 for the 131 definition of the regions). At low temperatures both limits agree well with our Monte Carlo calcu-132 lations within the statistical noise and clearly show the existence of a finite intrinsic magnetisation 133 at non-zero temperature for finite size. At higher temperatures there is a systematic difference be-134 tween the degree of magnetic ordering between the simulations and the analytical calculations due 135 to the ASM only becoming exact in the limit of infinitely many spin components. The large num-136 ber of Monte Carlo steps and strict convergence criteria to the same thermodynamic equilibrium 137 for ordered and disordered starting states (see Supplementary Section 4) rule out critical slowing 138

¹³⁹ down²⁷ as a source of difference between the analytical calculations and the simulations.

One may also argue in terms of the correlation length ξ which is comparable to the sys-140 tem size at the crossover temperature. It has been demonstrated¹³ that $\xi \propto \exp(cJ/T)$, where c 141 is a constant, meaning that the inverse crossover temperature T_x^{-1} only logarithmically increases 142 with the system size. Although our simulations are at the limit of the capabilities of current su-143 percomputer features, this effect is expected to persist for larger sizes of $2-10 \ \mu m$. These values 144 represent typical sizes of continuous 2D microflakes in experiments, and much larger than the ideal 145 nanoscale devices likely to be used in future 2D spintronic applications. Fitting a scaling function 146 to the crossover temperatures for different lattice symmetries (Fig. 2), we can plot the scaling of 147 the crossover temperature with size (Fig. 3a) which can then be extrapolated to larger scales. 148 The crossover temperature is still approximately 30 K for $2 - 10 \mu m$ flakes (Fig.3b). The graph 149 can be extrapolated to show that only at the $10^{15} - 10^{25}$ m range does the crossover temperature 150 become lower than ~ 1 K. To put these numbers in perspective to physical systems, these length 151 scales lie between the distance of the Earth to the Sun and the diameter of the observable universe. 152 Therefore, the often asserted notion⁴ that experimental 2D magnetic samples can be classified as 153 infinite and therefore display no net magnetic order at nonzero temperatures, as expected from the 154 Mermin-Wagner theorem, is not applicable. 155

The significance of the crossover temperature T_x in relation to the Curie temperature T_C is particularly important when discussing the nature of the magnetic ordering in 2D magnets at zero anisotropy for T > 0 K. We investigate this behaviour through colour maps of the spin ordering

after 40 million Monte Carlo steps comparing different system sizes and temperatures (Fig. 4). 159 At very low temperatures T = 2.5 K, where there is a high degree of order, the spin directions 160 are highly correlated, as indicated by a mostly uniform colouring. Although the temperatures are 161 near zero, the system is superparamagnetic indicating that over time the magnetization direction 162 fluctuates, and the effect is most apparent for the smallest sizes where the average direction has 163 moved significantly from the initial direction $S||_z$. At higher temperatures the deviation of the spin 164 directions within the sample increases as indicated by the more varied colouring. To quantitatively 165 assess the spin deviations we plot the statistical distribution of angle between the spin direction 166 and the mean direction for different temperatures for each size (Supplementary Figure 4). For an 167 isotropic distribution on the unit sphere there is a $\sin(\theta)$ weighting which is seen at the highest 168 temperature for all system sizes. For lower temperatures where the spin directions are more cor-169 related, the distribution is biased towards lower angles. Qualitatively there is little difference in 170 the spin distributions for the different samples. At T = 20 K there is however a systematic trend 171 in the peak angle increasing from $\theta = 40^{\circ}$ for the 50 × 50 nm² flake (Supplementary Figure 4a) 172 to around $\theta = 60^{\circ}$ at 1000×1000 nm² (Supplementary Figure 4d) indicating an increased level 173 of disorder averaged over the whole sample. This effect is straightforwardly explained by the size 174 dependence of spin-spin correlations. At small sizes the spins are strongly exchange coupled, pre-175 venting large local deviations of the spin directions. At longer length scales available for the larger 176 systems, the variations in the magnetisation direction are also larger. Surprisingly, our calculations 177 reveal that this effect is weak: even for very large flakes of a micrometre in size, only a small 178 increase can be observed in the position of the peak in the angle distribution at a fixed temperature. 179

Above the crossover temperature the spin-spin correlation length becomes very small compared to the system size with rapid local changes in the magnetisation direction, indicative of a completely disordered paramagnetic state. Our analysis reveals that the spins in finite-sized 2D isotropic magnets are strongly aligned due to short-range order at non-zero temperatures and up to the crossover temperature.

185 Discussion

Mathematically a phase transition is defined as a non-analytic change in the state variable for the 186 system, such as the particle density or the magnetization in the case of spin systems. For any finite 187 system the state variable is continuous by definition due to a finite number of particles, forming 188 a continuous path of intermediate states between two distinct physical phases²⁸. The same is true 189 for a magnetic system, forming a continuous path between an ordered and a paramagnetic state. A 190 priori then, it is impossible to have a true phase transition for any finite magnetic samples which are 191 routinely implemented in device platforms. Yet, nanoscale magnets that are far from infinite have 192 been studied for decades and exhibit a clear crossover from magnetically ordered to paramagnetic 193 phases, occurring for systems only a few nanometres in size^{16,17}. The crossover temperature in a 194 finite-size system hence can be described as an inflection point in M(T). The precise definition of 195 a phase transition is significant when considering the main conclusions of Mermin and Wagner¹, 196 which explicitly only apply in the case of an infinite system. As our results clearly show, sam-197 ple sizes measured experimentally are not classifiable as infinite and therefore not subject to the 198 Mermin-Wagner theorem. It is noteworthy that 3D compounds have weak dependence of their 199

critical temperature on the magnetic anisotropy²⁹. Similar analysis performed for a finite 3D honeycomb bulk system (Supplementary Figure 6a-b) show that the inclusion of anisotropy barely
change the results for Tc. This suggests that magnetism is an exchange-driven effect in both two
and three dimensions.

On the practical side, heterostructures with conventional metallic magnetic materials could 204 establish preferential directions of the magnetization through anisotropic exchange and dipolar 205 couplings. However, it is important to point out that the short-range order is enforced by the 206 isotropic exchange couplings and even a low anisotropy may suffice for stabilizing the direction of 207 the magnetization at the vdW layers, *i.e.*, from underlying magnetic substrates. We can imagine 208 micrometre-sized samples where all spins are still correlated at finite temperatures so it could 209 represent a single bit. However, for miniaturization purposes multiple nanometre-sized bits are 210 required on the same sample in order to be implemented in recording media. This is typically 211 achieved by magnetic domains, but there are no domains in an isotropic model since the domain 212 wall width is infinite. However, if vdW layers can be grown with grain boundaries, like in 2D 213 mosaics³⁰, which are large enough that each grain area would have an uniform magnetisation, 214 then a magnetic monolayer would have as many bits as available on the material surface. The 215 underlying substrate hence would set the magnetisation direction for further implementations. This 216 spin-interface engineering would be a considerable step towards on-demand magnetic properties 217 at the atomic level given the flexibility on the orientation of the magnetic moments without a 218 predefined direction at the layer. While the anisotropy circumvents the Mermin-Wagner theorem 219 and causes the critical temperature T_c to be nonzero in infinitely large systems, in finite samples 220

the short-range order persists up to much higher temperatures ($T_x > T_c$) since T_x is proportional to the isotropic exchange rather than the anisotropy^{31,32}.

In conclusion, we presented large-scale spin dynamic simulations and analytical calculations 223 of the temperature dependence of the intrinsic magnetization in 2D magnetic materials described 224 by an isotropic Heisenberg model. We found that short-range magnetic order at non-zero temper-225 ature is a robust feature of isotropic 2D magnets even at experimentally accessible length and time 226 scales. Our data show that the often asserted Mermin-Wagner limit¹ does not apply to 2D materi-227 als on real laboratory sample sizes. Since the spins are aligned due to the exchange interactions 228 already in the isotropic model, the direction of the magnetization may be stabilized by geometrical 229 factors or finite-size effects. These findings open up possibilities for a wider range of 2D magnetic 230 materials in device applications than previously envisioned. Furthermore, the limited applicabil-231 ity of the analytical Mermin-Wagner theorem opens similar possibilities in other fields such as 232 superconductivity and liquid crystal systems³³, where the relevant length scale of correlations is 233 known to be much greater than that required for experimental measurements and applications. Our 234 results suggest that if the magnetic anisotropy can be controlled to a certain degree³⁴ until it com-235 pletely vanishes, new effects of strongly correlated spins or more unusual disordered states may be 236 observed. 237

238 Methods

We used atomistic simulations methods^{9,35} to compute the magnetic properties of 2D magnetic materials. The energy of our system is calculated using the spin Hamiltonian:

$$\mathscr{H} = -\sum_{i < j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - K \sum_i (S_i^z)^2, \tag{4}$$

where $S_{i,j}$ are unit vectors describing the local spin directions on magnetic sites *i*, *j*, and J_{ij} is the 239 exchange constant between spins. An easy-axis magnetocrystalline anisotropy constant K can be 240 included as well, with negligible modifications of the results as described in the text. Simulations 241 were run for system sizes of 50 nm, 100 nm, 500 nm and 1000 nm laterally along the x and y 242 directions and 1 atomic layer thick along the z direction. For the honeycomb lattice, the simula-243 tions were initialized in either a perfectly ordered state aligned along the z direction or a random 244 state corresponding to infinite temperature. For these simulations the final $\langle |\mathbf{m}| \rangle(T)$ curves were 245 identical to each other. However, at low temperatures it took ten times as many steps to reach 246 the final equilibrium state from the random state, so for the remaining structures only simulations 247 starting from the ordered states were run. The systems were integrated using a Monte Carlo inte-248 grator using a uniform sampling algorithm³⁶ to remove any bias introduced from more advanced 240 algorithms²¹. To investigate the temperature dependence, the simulation temperature was varied 250 from 0 to 90 K in 2.5 K steps. 40×10^6 Monte Carlo steps were run for each temperature step. 251 This was split into 39×10^6 equilibration steps and then 10^6 time steps from which the statistics 252 were calculated. The Monte Carlo simulations use a pseudo-random number sequence generated 253 by the Mersenne Twister algorithm³⁷ due to its high quality, avoiding correlations in the generated 254

random numbers and with an exceptionally long period of $2^{19937} - 1 \sim 10^{6000}$. The parallel implementation generates different random seeds on each processor to ensure no correlation between the generated random numbers.

The time-dependent simulations in Fig. 1**c,d** were performed by solving the stochastic Landau-Lifshitz-Gilbert equation:

$$\frac{\partial \mathbf{S}_{i}}{\partial t} = -\frac{\gamma_{e}}{1+\lambda^{2}} \left[\mathbf{S}_{i} \times \mathbf{B}_{\text{eff}} + \lambda \mathbf{S}_{i} \left(\mathbf{S}_{i} \times \mathbf{B}_{\text{eff}} \right) \right], \tag{5}$$

which models the interaction of an atomic spin moment \mathbf{S}_i with an effective magnetic field $\mathbf{B}_{eff} = -\partial \mathcal{H}/\partial \mathbf{S}_i$. The effective field causes the atomic moments to precess around the field, where the frequency of precession is determined by the gyromagnetic ratio of an electron ($\gamma_e = 1.76 \times 10^{11}$ rad s⁻¹T⁻¹) and $\lambda = 1$ is the damping constant. The effect of temperature is taken into account using Langevin dynamics³⁸, where the thermal fluctuations are represented by a Gaussian white noise term. At each time step the instantaneous thermal field acting on each spin is given by

$$\mathbf{B}_{\rm th}^i = \sqrt{\frac{2\lambda k_B T}{\gamma \mu_s \Delta t}} \mathbf{\Gamma}(t) \tag{6}$$

where $k_{\rm B}$ is the Boltzmann constant, *T* is the system temperature and $\Gamma(t)$ is a vector of standard (mean 0, variance 1) normal variables which are independent in components and in time. The thermal field is added to the effective field in order to simulate a heat bath. The system was integrated using a Heun numerical scheme³⁶.

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270 Supplementary Materials

²⁷¹ Supplementary Sections 1-7, Supplementary Figures 1-5, and Supplementary References.

272 Data Availability

The data that support the findings of this study are available within the paper and its Supplementary
Information.

275 **Competing interests**

²⁷⁶ The Authors declare no conflict of interests.

277 Author Contributions

EJGS conceived the idea and supervised the project. SJ performed the atomistic simulations with inputs from EJGS and RFLE. LR and UA developed the semi-analytical model and undertook the numerical simulations. EJGS wrote the paper with a draft initially prepared by SJ and RFLE and also with inputs from KSN, UA and LR. All authors contributed to this work, read the manuscript, discussed the results, and agreed on the included contents.

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Figure 1: Short-range magnetic ordering at finite temperatures in a 2D isotropic magnet. a, Local view of the spin directions extracted from the atomistic simulations on a 2D honeycomb lattice. *a* is the atomic spacing (a = 0.4 nm), L is the length considered in the computations, and \mathbf{M}_{av} is the averaged magnetisation vector. θ corresponds to the angle between \mathbf{M}_{av} and the *z*-axis. $\theta_0 = 0$ denotes the initial configuration aligned with the *z*-axis. **b**, Temperature-dependent intrinsic magnetisation ($\langle |\mathbf{m}| \rangle$) with ($K = 1 \times 10^{-24}$ J/atom) and without (K = 0) anisotropy in a 1000 × 1000 nm² flake. Solid lines are the fit to Eq. (3). For K = 0, the fitting parameters are $\beta = 0.54 \pm 0.020$ and $T_x = 23.342 \pm 0.237$ K. For K > 0, $\beta = 0.427 \pm 0.021$ and $T_x = 26.543 \pm 0.320$ K. **c-d**, Temporal variation of the magnetisation (m/m_s) and angle $\theta - \theta_0$, respectively, at T = 10 K. All three spatial components (x, y, z) are considered in **c**. The dashed line in **d** shows the initial state in the simulations.



Figure 2: Temperature- and size-dependent properties of isotropic 1D and 2D materials with different crystal structures. a-d Comparative simulations of the temperature-dependent magnetisation for honeycomb, hexagonal, square lattices and an atomic chain (1D), respectively, for different system sizes. Points indicate the results of Monte Carlo simulations, the lines show fits to the Curie-Bloch Eq. (3) in the classical limit, and the shaded regions indicate the anisotropic spherical model calculations for different assumptions of the renormalisation factor for the Curie temperature arising from the mean-field approximation. See Supplementary Section 7 for details. The dashed and solid lines in **d** indicate the anisotropic spherical model calculations, and the exact solution, respectively. Both show a sound agreement with the atomistic simulations. The datasets in **a-c** clearly show the existence of short-range collinear magnetic order for all 2D lattices at the simulated sizes considered with nonzero crossover temperature. Zero magnetic anisotropy is included in all calculations.





Figure 4: **Temperature-dependent magnetic order.** Visualisations of the magnetic spin configurations for the honeycomb lattice starting from an ordered state as a function of system size (vertical row) and temperature (horizontal row). The spins are projected following the color scale shown in the sphere on the left. The bottom row shows a local view of the spins inside a 5 nm \times 5 nm area at the location outlined by the small boxes in the 1000 \times 1000 nm² snapshots.

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