Fingerprints of Universal Spin-Stiffness Jump in Two-Dimensional Ferromagnets

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Motivated by recent progress on synthesizing two-dimensional magnetic van der Waals systems, we propose a setup for detecting the topological Berezinskii-Kosterlitz-Thouless phase transition in spin-transport experiments on such structures. We demonstrate that the spatial correlations of injected spin currents into a pair of metallic leads can be used to measure the predicted universal jump of $2/\pi$ in the ferromagnet spin stiffness as well as its predicted universal square root dependence on temperature as the transition is approached from below. Our setup provides a simple route to measuring this topological phase transition in two-dimensional magnetic systems, something which up to now has proven elusive. It is hoped that this will encourage experimental efforts to investigate critical phenomena beyond the standard Ginzburg-Landau paradigm in low-dimensional magnetic systems with no local order parameter.

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Introduction.-Phase transitions of matter are ubiquitous and fascinating phenomena in nature. In particular, transitions occurring in two dimensions (2D) have long attracted great interest because of their intriguing physics. Quantum or thermal fluctuations play a fundamental role in the stability of phases. The special features of low dimensional systems with continuous symmetries, are rooted in the celebrated Hohenberg-Mermin-Wagner theorem [1-3] stating that there can be no spontaneous breaking of a continuous symmetry at any finite temperature in dimensions $d \leq 2$. This prevents the existence of long-range order at any temperature T > 0, resulting in the absence of a local order parameter. Nonetheless, a lowtemperature phase free of topological defects can exist, featuring quasi-long-range order characterized by algebraically decaying correlations.

Phase transitions may quite generally be thought of as mediated by the proliferation of topological defects with a concomitant loss of some generalized stiffness [4]. Unfortunately, in most cases it is almost impossible to make these notions precise and quantitative, at least analytically. However, the precise mechanism by which this happens in low-dimensional magnets, superfluids, and crystals has been elucidated in a series of seminal works of Berezinskii, Kosterlitz, and Thouless (BKT) [5–8], describing a topological phase transition at the critical temperature T_c from tightly bound pairs of vortices and antivortices to an unpaired disordered phase.

A remarkable and unique prediction of this theory is a specific feature characterizing the vanishing of topological order, or stiffness, as the system approaches the critical temperature from below [9]. This is expressed by the long-wavelength relation

$$\frac{1}{\mathcal{K}_R(T)} = \frac{1}{\mathcal{K}} + 4\pi^2 \lim_{q \to 0} \frac{\langle n_q n_{-q} \rangle}{q^2}, \qquad (1)$$

where \mathcal{K} is the stiffness of system, e.g., superfluid density or spin stiffness in easy-plane ferromagnets, \mathcal{K}_R is the vortex-renormalized stiffness, and n_q is the vorticity in momentum space. When the system reaches the transition temperature, the stiffness \mathcal{K}_R jumps discontinuously to zero. The key feature of the transition is that this jump is *universal*, $\mathcal{K}_R(T_c)/k_BT_c = 2/\pi$. This was first verified experimentally in thin films of ⁴He [10] and later in other systems such as superconductors [11–15], colloidal crystals [16–18], Josephson-junction arrays [19,20], and ultracold atomic Bose gases [21].

Despite its great interest and recent efforts [22], the observation of BKT transitions has proven elusive in spin systems, mainly due to the difficulty of manufacturing twodimensional magnets. The discovery of graphene in 2004 [23] was a turning point for significant experimental progress in fabricating atomically thin magnetic films, also known as 2D magnetic van der Waals (vdW) materials [6,24–31].

In this Letter, we propose an experimental setup for the observation of the BKT transition in 2D magnetic vdW materials using standard methods in the study of spin transport. It is based on electrical measurements of charge-current cross-correlations in metal-magnet hybrid structures [32–35]. In this approach, distant metallic leads detect spin currents flowing into and out of the magnetic material, as displayed in Fig. 1. At the center of this proposal is the phenomenon of spin and charge conversion, an inherent property of materials having strong spin-orbit coupling manifested in the spin-Hall effect [36]. The characteristic spin dynamics in the magnetic insulator pump spin currents

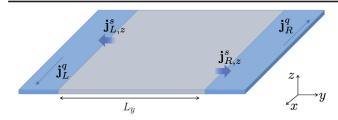


FIG. 1. Schematic plot for the direct observation of BKT transition. The setup consists of a 2D magnetic vdW material attached at its ends to a couple of identical nonmagnetic metallic leads. The detection is based on the measurement of charge-current cross-correlations between the left and right lead of the device geometry.

into the normal metal [37] resulting in a spin accumulation that, in turn, induces charge currents by the inverse spin-Hall effect. The electrical detection of spin-current signals, and their correlations, will provide direct access to thermally induced vortex proliferation and quasiordereddisordered phase transition. This work constitutes a first step to studying low-dimensional phenomena involving notions relevant to phase transitions, 2D magnetic material science, and spintronics.

Nonlocal spin-transport measurements in metal-magnetmetal heterostructures have proven useful for studying transport properties of pure spin currents [32], longdistance spin-transport [33,34,38], magnon-polaron transport [39], and viscosity in magnons systems [40]. Recently, Ref. [41] proposed a method of detecting the degree of coherence of magnon states by measuring spincurrent correlations. This detection scheme is particularly useful since the spin-current cross-correlation is related to the power spectral density. Although the ideas developed in Ref. [41] are general, the focus was on spatially homogeneous magnetization dynamics, e.g., ferromagnetic resonance and thermally induced magnons. In our approach, we tailor the detection scheme to a system that lacks long-range order. We show that spatially dependent charge-current cross-correlations allow characterizations of the BKT phase transition.

Our findings give direct access to track the evolution from algebraic- to exponential-decaying spin-spin correlations as the temperature increases. In particular, we provide an accessible route to measuring the universal jump of the spin stiffness in 2D magnets as the temperature approaches the critical temperature. Taking advantage of the spin to charge conversion, this enables a clear-cut experimental demonstration in a magnetic system of one of the remarkable predictions of the BKT theory.

Charge-current cross-correlation.—In the proposed setup, we consider a two-dimensional magnet, with dimensions L_x and L_y , coupled at its ends to a couple of identical nonmagnetic metallic leads, see Fig. 1. Pure spin currents into the left and right normal metals are related to electric signals via the inverse spin Hall effect

[36,42]. We introduce the charge-current cross-correlation, defined by $C^{(2)}(\tau) \equiv \langle I_L(\mathbf{r},t)I_R(\mathbf{r},t+\tau)\rangle$, where \langle,\rangle denotes a statistical average. Here, I_{ℓ} , with $\ell = L$, R, is the net charge current existing in the ℓ metallic lead. We are interested in the static correlation and thus in the evaluation of $C^{(2)} \equiv C^{(2)}(0)$. In the following, we relate $C^{(2)}$ to the cross-correlation of the injected spin currents using spin and charge conversion at the leads. This suffices to establish the connection between the characteristic spinspin correlations and the universal behavior predicted in the BKT theory.

Berezinskii-Kosterlitz-Thouless transition.—We consider a ferromagnetic insulator with axially symmetric exchange coupling around the \hat{z} direction. The nearestneighbor Hamiltonian is $H = \sum_{\langle ij \rangle} J_{\alpha\beta} S_{\alpha i} S_{\beta j}$, with *i*, *j* the position of spins and $\alpha, \beta = x, y, z$ labeling their components. The ferromagnetic exchange coupling is $J_{\alpha\beta} = -J_{\alpha}\delta_{\alpha\beta}$, with $J_x = J_y \equiv J$ and $J_z < J$, favoring spin ordering in the *xy* plane. Thus, we can express the normalized spin variable in terms of an angle θ_i on each lattice site *i*, as $S_i = (\cos \theta_i, \sin \theta_i, 0)^T$. The resulting Hamiltonian, known as the classical *XY* model [43], is given by

$$H = -J \sum_{\langle ij \rangle} \cos\left(\theta_i - \theta_j\right), \tag{2}$$

featuring a continuous SO(2) or U(1) symmetry. The XY model (2) is also a useful model for superfluid helium [10,44] and hexatic liquid crystals [45]. At low temperatures, we take into account only small spin fluctuations. Thus, in the continuum approximation, phase variations are described by the smooth Hamiltonian $H = J \int d\mathbf{r} (\nabla \theta)^2 / 2$. In two dimensions in the entire low-temperature *phase*, the spin-spin correlation function, defined by the statistical average $G(\mathbf{r} - \mathbf{r}') =$ $\langle S(\mathbf{r}) \cdot S(\mathbf{r}') \rangle$ decays algebraically, $1/|\mathbf{r} - \mathbf{r}'|^{\eta}$, with $\eta = k_B T / 2\pi J$, indicative of critical behavior. Here, the critical exponent η is the anomalous scaling dimension of the spin field. At long distances, the spin correlations vanish, $\lim_{|\mathbf{r}-\mathbf{r}'|\to\infty} G(\mathbf{r}-\mathbf{r}') = 0$, corresponding to the absence of long-range order in accordance with the Mermin-Wagner theorem [1]. Thus, spin waves suffice to destroy long-range order. The entire low-temperature phase is critical with an infinite correlation length and algebraic decay of spin-correlations.

In the high temperature phase, smooth spatial variations in $\theta(\mathbf{r})$ no longer suffice to accurately describe the fluctuations. Fluctuations beyond spin waves are included by separating the vector $\nabla \theta = \nabla \theta_L + \nabla \theta_T$ into a longitudinal and a transverse part, the spin waves and nonsmooth variations respectively, defined by $\nabla \cdot \nabla \theta_T = 0$ and $\nabla \times \nabla \theta_L = 0$. Thus, the model is generalized to $H = (J/2) \int d\mathbf{r} [(\nabla \theta_L)^2 + (\nabla \theta_T)^2]$, with the first term corresponding to the spin-wave part discussed above. The nonsmooth variations in θ describe vortices, topological defects associated with the first homotopy group [46] $\pi_1[U(1)] = \mathbb{Z}$ of the 2D XY model that have circulation $\oint dl \cdot \nabla \theta_T = 2\pi n$ with "topological charge" $n \in \mathbb{Z}$. At high temperature, the topological defects become important, and their statistical mechanics may be mapped to that of a 2D Coulomb gas with overall charge neutrality. The low-temperature phase where vortices and antivortices are tightly bound together is equivalent to an insulating dielectric state while the high temperature phase with dissociated pairs of vortex-antivortex pairs corresponds to a metallic phase. This transition is a topological phase transition. The temperature at which it takes place is found by noting that the energy of a vortexantivortex pair separated by a distance r, is $E = \pi J \ln (r/a)$ and the entropy $S = 2 \ln (r/a)$, where a is some shortdistance cutoff. A simple estimate for the Helmholtz free energy yield $F = (\pi J - 2T) \ln (r/a)$. It is clear that is energetically favorable to have free vortices when $T > T_c = \pi J/2k_B$. This estimate of T_c ignores screening of two test charges, and the description also ignores coupling between vortices and spin waves. Taking such effects onto account slightly reduces T_c without altering the universality class of the transition, which may be viewed as a special class of phase transition where the conformal invariance of standard critical points is lost, a "conformality lost" phase transition [47,48]. In modern terms, the precise mechanism for this loss of conformality is the annihilation of an ultraviolet and infrared fixed point as some marginal operator of the system is varied [47,48].

In general, a phase transition may be viewed as a proliferation of topological defects characterized by an appropriate homotopy group with a concomitant loss of an associated generalized stiffness. In the present context, thermally induced spin fluctuations (angle fluctuations) reduce and eventually destroy the existing topological order. The generalized stiffness for our system, the spin stiffness $\mathcal{K}_R(T)$, is a global order parameter for topological order.

We next provide the basics of how to measure $\mathcal{K}_R(T)$, and thereby detect the universal jump and the associated conformality lost transition in the recently discovered 2D vdW magnets, as the temperature is raised through $T > T_c$. We evaluate the spin current–current correlation function $C_{\mu\nu} = \langle j_{\mu}^s j_{\nu}^s \rangle / J^2$, where the spin current is $j_{\mu}^s(\mathbf{r}) = -J\partial_{\mu}\theta(\mathbf{r})$. The diagonal component, evaluated at the boundary of the magnet, correspond to the correlations between left and right spin currents (C_{yy}). In momentum space, the spin current is decomposed into two parts, one part originating with spin waves and another part with vortices, as follows:

$$C_{\mu\nu}(\boldsymbol{q}) = \frac{q_{\mu}q_{\nu}}{q^2} \frac{1}{\mathcal{K}} + \langle S_{q\mu}S_{-q\nu} \rangle_{\nu}.$$
 (3)

Here, $\mathcal{K} = J/k_B T$ and $\mathcal{S}_q = \mathcal{F}[(\nabla \theta)_v]$ is the Fourier transform of the vortex contribution. The second term in

the right-hand side in Eq. (3) may be expressed in terms of vortex correlators $\langle S_{q\mu}S_{-q\nu}\rangle_v = 4\pi^2 (\delta_{\mu\nu} - q_{\mu}q_{\nu})$ q^2 $\langle n_q n_{-q} \rangle / q^2$. Using this result in Eq. (3), we obtain $C_{\mu\nu}(q) = (q_{\mu}q_{\nu}/q^2)/\mathcal{K}_R$. The quantity \mathcal{K}_R represents the renormalized spin stiffness including the effects of thermally induced vortices. Thus, \mathcal{K}_R depends on temperature and obeys the relation given in Eq. (2). In the long-wavelength limit, $\mathcal{K}_R(T) =$ $\mathcal{K} - 4\pi^2 \mathcal{K}^2 \lim_{a \to 0} (\langle n_a n_{-a} \rangle / q^2)$, corresponding to the spin-wave and vortex part, respectively. Since, we are interested in the long-distance behavior of \mathcal{K}_R , we need to evaluate $\langle n_q n_{-q} \rangle$ when $q \to 0$. We first note that $\langle n_q n_{-q} \rangle = C_0 + C_2 q^2 + \cdots$, where C_0 vanishes by topological-charge neutrality. Thus, the only nonzero contribution in the long-wavelength limit is C_2 . The evaluation of this coefficient is standard and can be found in Ref. [43]. Below, we will relate the spin-current correlation $C_{\mu\nu}(q)$ with a measurable quantity, namely the charge-current cross-correlation.

Spin-charge conversion.—We assume that normal metals have a sufficiently strong spin-orbit coupling to support a considerable spin-Hall effect. The spin and charge transport in the bulk of metallic leads are captured by [49]

$$\mathbf{j}^{q} = \frac{\sigma}{e} \nabla \mu_{q} - \frac{\sigma'}{2e} \nabla \times \boldsymbol{\mu}_{s}, \qquad (4)$$

$$\frac{2e}{\hbar}\mathbf{j}_{n}^{s} = -\frac{\sigma}{2e}\nabla(\hat{\boldsymbol{n}}\cdot\boldsymbol{\mu}_{s}) - \frac{\sigma'}{e}(\hat{\boldsymbol{n}}\times\nabla)\boldsymbol{\mu}_{q}, \qquad (5)$$

where \mathbf{j}^{q} and \mathbf{j}_{n}^{s} are the charge and spin current (polarized in the \hat{n} direction), respectively. The electrical conductivity is σ , while σ' denotes the spin-Hall conductivity. The spin and charge accumulation, μ_s and μ_q , respectively, are described in the steady-state limit by the equations, $\nabla^2 \mu_q = 0$ and $\nabla^2 \mu_s = \mu_s / l_s^2$, where l_s is the spin-diffusion length in the normal metal. At the metal-magnet interface, the injected spin current (polarized along \hat{z}) is inhomogeneous since it originates from the spin's vortex at the magnet. As a result, a spin and charge accumulation, $\mu_s(x, y)$ and $\mu_a(x, y)$, are induced on the normal metal. The bulk equations, Eqs. (4) and (5), are complemented by the boundary conditions that enforce continuity for the spin current, $j_{z,y}^{s}(x,0) = j_{L,z}^{s}(x)$ and $j_{z,y}^{s}(x,-l) = 0$, and charge currents, $j_{y}^{q}(x,0) = j_{y}^{q}(x,-l) = 0$, at the left lead and where l is the width of the metal. Similar relations hold for the right lead. By simplicity we assume a spin-transparent interface, thus the injected spin current is $j_{L,z}^s(x) = -J\partial_y \theta(x, y)|_{y=0}$. The formal solution for the charge and spin accumulation are written as $\mu_{q,s}(x,y) = \int dx' K_{q,s}(x-x',y) j_{L,z}^s(x')$, with K_q and K_s the kernels, whose evaluation is detailed in the Supplemental Material [50].

We are interested on the induced charge current, averaged over the width of the metal, along the x direction at

each lead. We obtain $\bar{j}_L^q = 2el_s \vartheta \int_{-L_x/2}^{L_x/2} dx j_{L,z}^s(x)/l\hbar L_x$ in the limit $l \gg l_s$ and small spin Hall angle defined by $\vartheta = \tan^{-1} [\sigma'/\sigma]$. Note that the averaged charge current is independent on the position and proportional to the injected spin current averaged along the interface. Assuming that metals and magnet are in thermal equilibrium, the thermal average of charge current and spin current are zero. Their correlations, however, which are response functions of the system, will be nonzero and related by $\langle \bar{j}_L^q \bar{j}_R^q \rangle = (\xi/L_x) \int_{-L_x/2}^{L_x/2} dx C_{yy}(x, L_y)$, with $\xi = \pi G_0 l_s^2 \vartheta^2 J^2 \gamma(T)/l^2 \hbar$ and G_0 the quantum of conductance. The function $\gamma(T)$ is a numerical factor that varies in the range [4–3.5] when $0 < T < T_c$.

Measurement of universal jump in spin stiffness.—In the specific device geometry, the renormalized spin stiffness can be obtained from the measurement of the chargecurrent cross-correlation. Spin and charge conversion in the metallic leads allow us to relate the spin- and charge-current correlators, $C_{\mu\nu}$ and $C^{(2)}$, respectively. The detection of the spin stiffness can be realized by combined measurements of current correlations, since $Tr[\mathcal{C}_{\mu\nu}(\boldsymbol{q})] = 1/\mathcal{K}_R(T)$. The evaluation of Eq. (3) is carried out in the long-wavelength limit, thus we find that the temperature-dependent spin stiffness satisfies $1/\mathcal{K}_R(T) = 2 \int_{\mathcal{A}} d\mathbf{r} \mathcal{C}_{yy}(\mathbf{r})$. Here, we have considered axial symmetry and the integration is over the entire area, A, of the magnet. Although in actual measurements there is only access to the correlations at the boundary of the magnetic sample, this will not restrict the detection of $\mathcal{K}_R(T)$. In fact, its evaluation can be well approximated by repeating N measurements of $\langle \bar{j}_L^q \bar{j}_R^q \rangle$ for different lengths L_{y} . This series of measurements correspond to discretize the integration along the y direction. To make this approximation efficient, we employ the Gauss-Legendre quadrature method and find

$$\frac{\xi}{\mathcal{K}_R(T)\mathcal{A}} = \mathcal{C}^{(2)},\tag{6}$$

with $C^{(2)} = \sum_{i=1}^{N} c_i \langle \bar{j}_L^q \bar{j}_R^q \rangle_i$ the total charge-current correlator. The measurable correlations in a magnet with length L_{v}^{i} are denoted by $\langle \bar{j}_{L}^{q} \bar{j}_{R}^{q} \rangle_{i}$ and c_{i} are the weights of the approximation, see Supplemental Material [50] for details. It is expected that only a few measurements will be needed, due to the rapid convergence of this method. Note that to obtain Eq. (6) we have made two main assumptions. First, the flow of spin currents across the interface occurs with no resistance, i.e., a large value for the spin mixing conductance. Second, the charge current noise present in metallic leads is not considered. The observation of spin-current fluctuations requires a clear mapping to measurable charge currents, and hence the presence of current noise might result in additional complications. In practice, these side effects could lead to weaker signals, but we expect them to be less relevant for the cross-correlations between the left and right metal, and thus the sharp transition at the critical

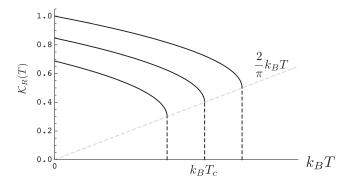


FIG. 2. Schematic plot of the (normalized) spin stiffness \mathcal{K}_R as a function of temperature. The discontinuity at the transition temperature T_c , represents the onset of a disordered phase of dissociated vortices and antivortices pairs. Various curves are plotted to make evident the universality in the jump of spin stiffness when normalized with the factor $k_B T_c$. As $T \to T_c^-$, $\mathcal{K}_R = (2/\pi)k_B T_c + c\sqrt{T_c - T}$, where *c* is a nonuniversal positive constant.

temperature should not be altered. Details of metal-magnet interface and a realistic treatment of noise are, however, open issues for the future.

The correlation (6), which constitutes the central result of this Letter, depends inversely on the renormalized spin stiffness. Thus, we expect that measurements of resistance fluctuations can display the temperature-dependent spin stiffness and the phase transition to a disordered phase of unbound vortices. In Fig. 2 we show the spin stiffness for various systems characterized by having different transition temperatures. In actual measurements, the universal behavior of the spin stiffness at transition temperature will be revealed as the jump occurs along a $2/\pi$ -slope straight line. For a specific setup, the jump in the spin stiffness is proportional to physical parameters related to the actual detector. It is therefore, convenient to normalize the correlation in order to isolate the intrinsic properties of the magnet.

Although the experimental realization of the device (Fig. 1) might be challenging, we consider it as a simple example to illustrate our proposal. A different option, beyond the scope of the present work, would be to consider the pair of metallic leads on top of the magnet. In this case, a spin accumulation is induced along, and polarized parallel to, the z direction. Accordingly, the observation of the spin stiffness would require the detection of spin-accumulation correlations, which could be done in a spin valve geometry. The latter requires a metallic ferromagnet on top of the leads for a voltage detection. The realizations of metal-magnet hybrids might be configured on state-of-the-art 2D ferromagnetic materials. The recent observation of spin waves [51] and spin valves [52] in CrI_3 -based heterostructures, the in-plane ferromagnetism in CrCl₃ [22], and the strong spin galvanic effect in graphene-transition metal dichalcogenide heterostructures [53,54], constitutes a real platform to guide future experiments to study the BTK transition.

Summary.—We have proposed a setup for measuring the BKT transition in a 2D magnetic-metal hydrid system. In a nonlocal geometry, measurements of voltage fluctuations can give access to the temperature dependence of spin stiffness. In particular, this approach provides direct evidence of the universal jump in the spin stiffness of the system, a global order parameter for topological order. We hope our proposal will encourage experimental efforts to detect this hallmark of the Berezinskii-Kosterlitz-Thouless topological phase transition in low-dimensional magnetic systems.

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