Detection of XY Behavior in Weakly Anisotropic Quantum Antiferromagnets on the Square Lattice

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We consider the Heisenberg antiferromagnet on the square lattice with $S = 1/2$ and very weak easy-plane exchange anisotropy; by means of the quantum Monte Carlo method, based on the continuous-time loop algorithm, we find that the thermodynamics of the model is highly sensitive to the presence of tiny anisotropies and is characterized by a crossover between isotropic and planar behavior. We discuss the mechanism underlying the crossover phenomenon and show that it occurs at a temperature which is characteristic of the model. The expected Berezinskii-Kosterlitz-Thouless transition is observed below the crossover: a finite range of temperatures consequently opens for experimental detection of non-critical 2D XY behavior. Direct comparison is made with uniform susceptibility data relative to the $S = 1/2$ layered antiferromagnet $Sr_2CuO_2Cl_2$.

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Predictions of the Berezinskii-Kosterlitz-Thouless (BKT) theory [1] for topological ordering with zero order parameter have been verified in many real systems, such as superfluid or superconducting films [2] and Josephson junction arrays [3]. However, despite the BKT theory being originally formulated as referred to 2D planar magnets, evidence of XY behavior in real magnets is weak and limited to very peculiar cases [4]. On the other hand, for $S = 1/2$ there exist several cuprous oxides that, besides being excellent realizations of 2D antiferromagnets, are known [5] to display a weak exchange easy-plane (EP) anisotropy: unambiguous observations of XY critical behavior in such systems are not easy to achieve, due to both the weakness of the anisotropy and the existence of finite interlayer coupling. As for the former point, the EP anisotropy observed in real magnets is usually 10^{-4}–10^{-3} times the isotropic exchange coupling, and the signatures of BKT critical behavior are often either too weak to be extracted from the isotropic thermodynamics or too close to the critical temperature to be experimentally accessible. The residual interlayer coupling, even if orders of magnitudes smaller than the intralayer one, drives the system towards a 3D transition, which is actually triggered by the divergence of 2D ntralayer spin correlations. Therefore purely 2D critical behavior of diverging quantities is most often masked by the onset of 3D long-range order.

In this work we show that several nondiverging quantities are sensitive to the presence of EP anisotropy and display an evident and detectable crossover between isotropic and XY behavior above the expected BKT transition. Such crossover occurs at a temperature which is characteristic of the model and is marked by peculiar features in the temperature dependence of noncritical observables. In particular, we present quantum Monte Carlo (QMC) data for the uniform susceptibility, the finite-size staggered out-of-plane magnetization, the specific heat, and the density of in-plane vortices.

We consider the $S = 1/2$ easy-plane antiferromagnet on the square lattice described by the Hamiltonian

$$\hat{\mathcal{H}} = \frac{J}{2}\sum_{i,d}(\hat{S}_i^x\hat{S}_{i+d}^x + \hat{S}_i^y\hat{S}_{i+d}^y + (1-\Delta)\hat{S}_i^z\hat{S}_{i+d}^z), \qquad (1)$$

where $\boldsymbol{i} = (i_1, i_2)$ runs over the sites of a square lattice, $\boldsymbol{d}$ connects each site to its four nearest neighbors, $J > 0$ is the antiferromagnetic exchange coupling, and $\Delta \in (0, 1]$ is the EP anisotropy parameter. We use the reduced temperature $t = T/J$.

The above model is studied by means of QMC simulations based on the continuous-time loop algorithm [6–8] for $\Delta = 0.001$, 0.02, and 1, and on lattice sizes from $L = 64$ to $L = 200$. Each MC run consists of 10^4 thermalization steps, followed by (1–1.5) $\times 10^5$ MC steps for measurements. Whenever possible, we implement improved estimators [7] for the quantities of interest.

The class of EP antiferromagnets described by Eq. (1) is characterized by the possibility for the model to support nonlinear topological excitations in the form of in-plane vortices (V) and antivortices (AV). Such excitations play a fundamental role in determining the thermodynamics and critical behavior of the system, as described by the BKT theory relative to the classical planar rotator model. When both quantum and out-of-plane fluctuations are present, the BKT behavior persists even in strongly quantum ($S = 1/2$) nearly isotropic ($\Delta \simeq 10^{-3}$) models [8].

Nevertheless, there exist significant differences between the standard BKT phenomenology and the XY behavior actually observed in quantum nearly isotropic antiferromagnets, where the out-of-plane spin component plays an essential role above the transition.

Let us consider a strongly anisotropic EP model ($\Delta \lesssim 1$): At high temperatures the system, despite being disordered, already supports topological excitations in the form of well separated V and AV in the easy plane. As t decreases V and AV attract each other, till V-AV pairs begin to form and a maximum in the specific heat is correspondingly observed. At the critical temperature $t_{\rm BKT}$, V and AV are all paired and the topological transition occurs.

In the nearly isotropic case ($\Delta \ll 1$), the picture is modified as follows. At high temperature the system is isotropically disordered. When t decreases, the EP anisotropy becomes effective enough to stabilize planar configurations and allow for V and AV to appear in the easy plane. This phenomenon, occurring at a temperature hereafter indicated as $t_{\rm co}$, may be thought of as a crossover between the isotropic and a more genuine XY behavior, and suggests the temperature range $t_{\rm BKT} < t < t_{\rm co}$ as the most appropriate for experimental observations.

We consider the dimensionless uniform susceptibility $\chi_{\rm u}^{\alpha\alpha}$, defined as

$$\chi_{\rm u}^{\alpha\alpha} = \frac{J}{L^2}\sum_{ij}\int_0^\beta d\tau\langle\hat{S}_i^\alpha(0)\hat{S}_j^\alpha(\tau)\rangle. \tag{2}$$

Figure 1 shows its temperature dependence, which we find

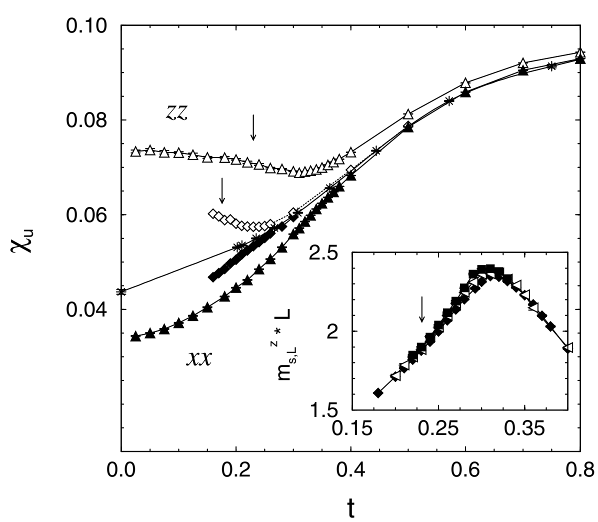

FIG. 1. Uniform susceptibility of the EP model for $\Delta = 0.02$ (triangles) and $\Delta = 0.001$ (diamonds). Open (full) symbols represent the out-of-plane (in-plane) susceptibility. The stars represent the uniform susceptibility in the isotropic case [6,9,10]. Inset: finite-size magnetization of the EP model for $\Delta = 0.02$ and $L = 64$ (diamonds), 96 (left triangles), and 128 (squares). Error bars are smaller than symbol sizes; arrows are the critical temperatures as estimated via finite-size scaling [8].

strongly characterized by the appearance of a minimum in the zz component. A similar minimum has indeed been experimentally observed in $Sr_2CuO_2Cl_2$ and suggested to be related to the onset of 2D XY behavior [11].

A sound argument in favor of this interpretation is the following. An infinitesimal uniform magnetic field causes a finite response in an antiferromagnet via the canting mechanism: adjacent spins antialigned in the plane perpendicular to the field cant out of such plane and give rise to a net magnetization, while a negligible response comes from spins antialigned along the field direction. On this basis, we interpret Fig. 1. At high temperature, $\chi_{\rm u}^{zz}$ and $\chi_{\rm u}^{xx}$ behave in the same way and decrease upon decreasing t as antiferromagnetic coupling gets more effective and canting consequently harder. As t is further lowered, the anisotropy starts to play a role, and most of the spins antialign in the easy-plane: therefore, compared to the isotropic case, the number of spins responding to a field applied in the plane (along z) decreases (increases). As a consequence, $\chi_{\rm u}^{xx}$ decreases faster, while $\chi_{\rm u}^{zz}$ slows down its decrease; the further reduction of out-of-plane fluctuations is eventually responsible for the low-temperature increase of $\chi_{\rm u}^{zz}$. A clear minimum consequently appears and marks the crossover to XY behavior at the temperature $t_{\rm co}$, where the out-of-plane component of the antiferromagnetic coupling becomes irrelevant. No particular feature is instead seen at the transition.

In order to check the direct relation between the crossover phenomenon and the out-of-plane fluctuations, we consider our data relative to finite-size out-of-plane staggered magnetization, $m_{\rm s,L}^z$; such quantity, which obviously vanishes at all temperatures in the thermodynamic limit, is quite useful when local spin configurations are under analysis, as in our case. In the inset of Fig. 1 we show data for different lattice sizes and $\Delta = 0.02$. By lowering t, $m_{\rm s,L}^z$ increases (as expected in the isotropic behavior) above $t_{\rm co}$, and decreases (as expected in XY behavior) below $t_{\rm co}$: a stable maximum is seen at $t_{\rm co}$. From the above evidence we obtain the estimates $t_{\rm co}(\Delta = 0.02) = 0.30(1)$ and $t_{\rm co}(\Delta = 0.001) = 0.225(10)$. Signatures of the crossover are also present in the staggered out-of-plane susceptibility and out-of-plane correlation length (shown in Ref. [8]), displaying a maximum at a temperature quite close to $t_{\rm co}$.

The above results suggest that the crossover phenomenon is peculiar to the system and is due to the suppression of out-of-plane fluctuations. In particular, $t_{\rm co}$ is the temperature below which the weakly anisotropic system behaves like a planar rotator model with a spin length effectively reduced by out-of-plane fluctuations. Other authors have referred to such crossover as due to a "spin-dimensionality reduction," meaning the loss of one spin component [11].

By the semiclassical reasoning in Appendix B of Ref. [8] we can predict $t_{\rm co}$ to depend on the anisotropy parameter Δ according to

$$t_{\rm co} \simeq \frac{4\pi\rho_{\rm s}/J}{\ln(C/\Delta)}, \tag{3}$$

where $\rho_{\rm s}$ is the renormalized spin stiffness of the quantum isotropic model, and C is a constant. A logarithmic fit to our data, shown in Fig. 2, gives $C = 160$ and $\rho_{\rm s} = 0.214$ J, which compares well with the known value [6] $\rho_{\rm s} = 0.180$ J.

The onset of XY behavior below $t_{\rm co}$ is further supported by the temperature dependence of the specific heat c. In the planar rotator model such quantity displays a maximum well above the BKT transition. Figure 3 shows our data for the weakly anisotropic model with $\Delta = 0.02$: an embryonic peak, out of the isotropic component, is seen at $t = 0.29(1)$, slightly below $t_{\rm co}$. To clarify the actual meaning of such a peak, we have also considered the density of in-plane vortices with unitary vorticity, defined as [13]

$$\rho_{\rm v} = \tfrac{1}{8}\langle 1 - 8\hat{S}_{\boldsymbol{i}}^x\hat{S}_{\boldsymbol{l}}^x + 16\hat{S}_{\boldsymbol{i}}^x\hat{S}_{\boldsymbol{j}}^y\hat{S}_{\boldsymbol{l}}^x\hat{S}_{\boldsymbol{m}}^y\rangle, \tag{4}$$

where $(\boldsymbol{i}, \boldsymbol{j}, \boldsymbol{l}, \boldsymbol{m})$ defines a plaquette of the square lattice (indices are ordered counterclockwise). Choosing z as the quantization axis, the above quantity is off-diagonal. The estimator of the bilinear term in Eq. (4) can be found in Ref. [7]. In the same spirit we introduce the estimator for the quartic term in the context of the loop algorithm [14].

By considering QMC data for $\Delta = 1$, shown in the inset of Fig. 3, we observe that the temperature derivative of $\rho_{\rm V}$ displays a maximum where the specific heat exhibits its peak. Both features mark therefore the onset of the formation of V-AV pairs. In Fig. 3 we show that this picture is fully reproduced by our data for $\Delta = 0.02$, thus confirming the XY character of our weakly anisotropic system.

From the experimental point of view, the above findings may first be used to easily characterize the anisotropy of the layered antiferromagnets: if a minimum in the out-of-plane component of the uniform susceptibility is observed above the transition, this is a signature of EP anisotropy (while a minimum *at* the transition suggests an easy-axis anisotropy, as shown in Ref. [8]). Once $t_{\rm co}$ has been experimentally determined, Eq. (3) allows one to get an independent estimate of the bare anisotropy parameter Δ of Eq. (1). As shown below, this is quite an essential point for interpreting the experimental data.

We now consider the layered cuprate $Sr_2CuO_2Cl_2$, whose intralayer spin-spin coupling of the Cu^{2+} ions has been proposed [15] to be governed by the Hamiltonian (1) with $J = 1450$ K. As for the anisotropy parameter Δ, the experimental analysis [15] hands us the renormalized value $\Delta^{\rm exp} = 0.00014$, extracted from low-temperature measurements of the spin gap in the out-of-plane branch of the 2D spin waves propagating in the Cu layers, by means of the approximated expression $G = 4JSZ_c\sqrt{2\Delta^{\rm exp}}$, where Z_c is the zero-temperature value of the spin-wave velocity renormalization coefficient of the isotropic Heisenberg system [16]. The above $\Delta^{\rm exp}$ is hence not the bare value appearing in Eq. (1), as it already contains quantum renormalizations. In fact, a more refined self-consistent spin-wave-theory calculation [14] leads to the expression $G = 4JSZ_c\sqrt{2Z_\Delta\Delta}$ with $Z_\Delta = 0.099$, and hence $\Delta = \Delta^{\rm exp}/Z_\Delta \simeq 0.0014$.

Experimental data relative to the uniform susceptibility of $Sr_2CuO_2Cl_2$ (from Refs. [11,17]) are shown in Fig. 4 together with our results for $\Delta = 0.001$; a constant offset for the experimental data has been introduced in order to take into account spurious temperature-independent contributions [18] to the measured values of $\chi_{\rm u}^{\alpha\alpha}$. The agreement is excellent for both the in-plane and the out-of-plane component; considering that no adjustment has

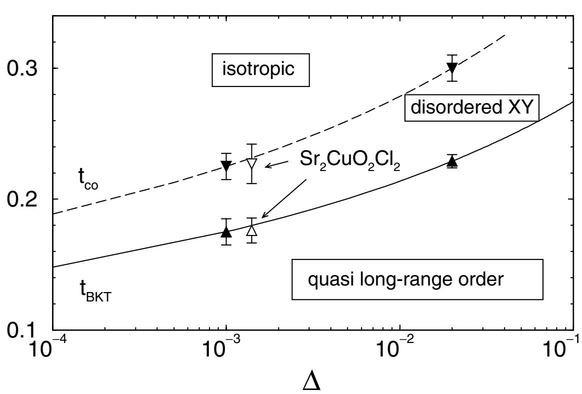

FIG. 2. Phase diagram of the weakly EP model on the square lattice. Down triangles: $t_{\rm co}$; up triangles: $t_{\rm BKT}$. Solid and dashed lines are logarithmic fits. Also reported are the transition and crossover temperatures of $Sr_2CuO_2Cl_2$ (see text).

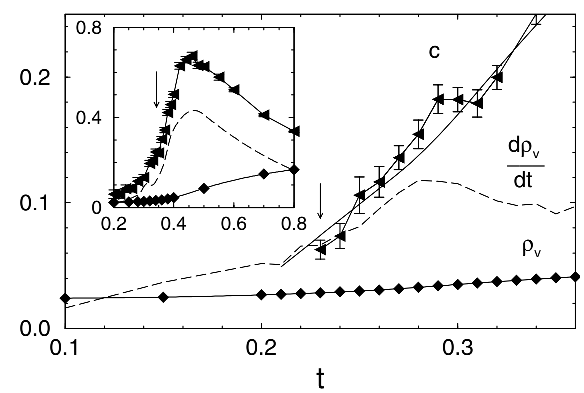

FIG. 3. Specific heat, vortex density, and its temperature derivative for the EP model with $\Delta = 0.02$. Dashed line: numerical derivative of splines for the vortex density data; solid line: specific heat of the isotropic model (from a numerical interpolation to the data of Ref. [9]). Inset: same picture for $\Delta = 1$ (vortex density data from Ref. [12]). When not visible, the error bars are smaller than the symbol size; arrows are the BKT critical temperatures.

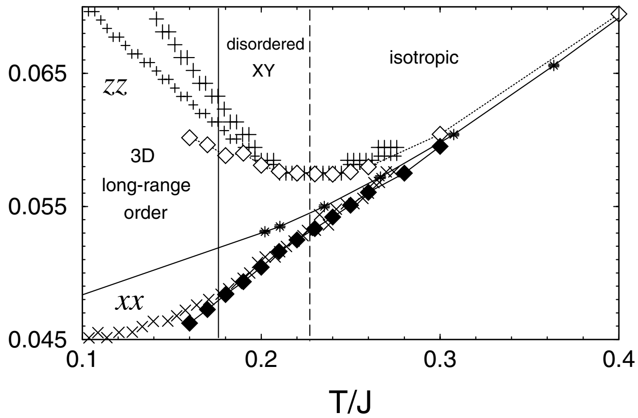

FIG. 4. Uniform susceptibility of $Sr_2CuO_2Cl_2$ compared to the theoretical predictions for the EP model with $\Delta = 0.001$. $\times$'s: χ_u^{xx} of $Sr_2CuO_2Cl_2$ (from Ref. [17]) (with gyromagnetic factor $g = 2$); thin and thick +'s: χ_u^{zz} of $Sr_2CuO_2Cl_2$ (from Ref. [11]) with $g = 2$ and $g = 2.46$, respectively. Other symbols and error bars as in Fig. 1.

been introduced for the temperature axis, the position of the minimum of χ_u^{zz} is nicely reproduced, and a crossover temperature $t_{co} \simeq 0.227$ for the real compound is determined. When the critical region is approached, experimental data deviate from the theoretical predictions due to the fact that a purely 2D model is no longer sufficient to capture the thermodynamic behavior of the real magnet, which is, in fact, characterized by an experimentally observed Néel transition at $t_N = 0.176$. Such transition temperature compares remarkably with the BKT transition temperature of the purely 2D model, $t_{BKT} = 0.175(10)$: this confirms that the 3D ordering is induced by the incipient intralayer BKT transition.

From our analysis we conclude that the experimental observation of a minimum in the transverse uniform susceptibility at a temperature which is well above the critical region, is a signature of EP character neatly detectable in real compounds even with very weak anisotropy. To this respect, we notice that the bare anisotropy value, needed for a quantitative comparison between experimental and theoretical results, significantly differs from the (measured) renormalized value, due to the relevance of quantum effects. Finally, two distinct regimes, separated by the crossover, can be identified in the disordered phase: an *isotropic* regime for $t > t_{co}$ and a *disordered XY* regime for $t_N < t < t_{co}$. It is relevant that the crossover temperature can be obtained by measuring nondiverging quantities above the critical region, which is an experimentally feasible task.

In summary, by means of quantum Monte Carlo simulations we have shown that two-dimensional $S = 1/2$ square-lattice antiferromagnets with a weak easy-plane anisotropy display a crossover from a high-temperature isotropic behavior to a genuinely 2D *XY* behavior at a temperature t_{co} which stays around 30% above the critical temperature t_{BKT}. We have, moreover, shown that $Sr_2CuO_2Cl_2$ realizes these predictions, and therefore stands as a very clean example of weakly planar quasi-2D antiferromagnet. This represents clear evidence of noncritical *XY* behavior in a real layered antiferromagnet. Other evidence for an isotropic-to-*XY* crossover in $Sr_2CuO_2Cl_2$ comes from neutron scattering [11] and NMR [19] experiments, and enforces our conclusions. Further work on such crossover effect in the above compound, as well as in other layered antiferromagnets with easy-plane anisotropy (as, e.g., Pr_2CuO_4), opens the perspective for a systematic investigation of the vortex phase in purely magnetic systems.

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