Detection of XY behaviour 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 behaviour. 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 behaviour. Direct comparison is made with uniform susceptibility data relative to the $S = 1/2$ layered antiferromagnet Sr$_2$CuO$_2$Cl$_2$.

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 [1] for topological ordering with zero order parameter has been verified in many real systems, such as superfluid or superconducting films [2] and Josephson junction arrays [3]. Nevertheless, there exist significant differences between the standard BKT phenomenology and the XY behaviour actually observed in quantum nearly isotropic antiferromagnets, where the out-of-plane spin component plays a fundamental role in determining the thermodynamics and critical behaviour 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 behaviour persists even in strongly quantum ($S = 1/2$) nearly isotropic ($\Delta \approx 10^{-3}$) models [5]. Nevertheless, there exist significant differences between the standard BKT phenomenology and the XY behaviour actually observed in quantum nearly isotropic antiferromagnets, where the out-of-plane spin component plays a fundamental role in determining the thermodynamics and critical behaviour 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 behaviour persists even in strongly quantum ($S = 1/2$) nearly isotropic ($\Delta \approx 10^{-3}$) models [5].
an essential role above the transition.

Let us consider a strongly anisotropic EP model ($\Delta \leq 1$): 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 $t_{\text{BKT}}$ $V$ 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 $t_{\text{co}}$, may be thought of as a crossover between the isotropic and a more genuine XY behaviour, and suggests the temperature range $t_{\text{BKT}} < t < t_{\text{co}}$ as the most appropriate for experimental observations.

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

$$\chi_{u}^{\alpha\alpha} = \frac{J}{L^2} \sum_{ij} \int_{0}^{\beta} d\tau \left\langle \hat{S}_{i}^{\alpha}(0)\hat{S}_{j}^{\alpha}(\tau) \right\rangle .$$

Fig. 1 shows its temperature dependence, which we find strongly characterized by the appearance of a minimum in the $zz$ component. A similar minimum has been indeed experimentally observed in Sr$_2$CuO$_2$Cl$_2$ and suggested to be related to the onset of 2D XY behaviour [11].

A sound argument in favour 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 anti-aligned along the field direction. On this basis, we interpret Fig. 1. At high temperature, $\chi_{u}^{zz}$ and $\chi_{u}^{zx}$ 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 antialias 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, $\chi_{u}^{zz}$ decreases faster, while $\chi_{u}^{zx}$ slows down its decrease; the further reduction of out-of-plane fluctuations is eventually responsible for the low-temperature increase of $\chi_{u}^{zz}$. A clear minimum consequently appears and marks the crossover to XY behaviour at the temperature $t_{\text{co}}$, 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, $m_{s_L}^{z}$: 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 $t$, $m_{s_L}^{z}$ increases (as expected in the isotropic behaviour) above $t_{\text{co}}$, and decreases (as expected in XY behaviour) below $t_{\text{co}}$: a stable maximum is seen at $t_{\text{co}}$. From the above evidences we obtain the estimates $t_{\text{co}}(\Delta = 0.02) = 0.30(1)$ and $t_{\text{co}}(\Delta = 0.001) = 0.225(10)$. Signatures of the crossover are also present in the staggered out-of-plane susceptibility and out-of-plane correlation length (shown in Ref. [3]), displaying a maximum at a temperature quite close to $t_{\text{co}}$.

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, $t_{\text{co}}$ 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 $t_{\text{co}}$ to depend on the anisotropy parameter

\[ t_{\text{co}} = \frac{\Delta}{2} \frac{L^2}{N^2} \]
the formation of V-AV pairs. In Fig. 3 we show that this exhibits its peak. Both features mark therefore the onset of the quartic term in the context of the loop algorithm [14].

Ref. 7. In the same spirit we introduce the estimator for the estimator of the bilinear term in Eq. (4) can be found in [13].

where $\rho_\alpha$ 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 $\rho_\alpha = 0.214 J$, which compares well with the known value $\rho_\alpha = 0.180 J$.

The onset of XY behaviour below $t_{co}$ 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. Fig. 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 $t_{co}$. To clarify the actual meaning of such peak, we have also considered the density of in-plane vortices with unitary vorticity, defined as

$$\rho_v = \frac{1}{8} \left\langle 1 - 8 \hat{S}_i^x \hat{S}_j^x + 16 \hat{S}_i^y \hat{S}_j^y \hat{S}_k^z \hat{S}_m^z \right\rangle,$$

where $(i, j, l, m)$ defines a plaquette of the square lattice (indices are ordered counterclockwise). Choosing $z$ as 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 $\rho_v$ 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 be firstly 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. [5]). Once $t_{co}$ has been experimentally determined, Eq. (3) allows one to get an independent estimate of the bare anisotropy parameter $\Delta$ of Eq. (1). As shown below, this is quite an essential point for interpreting the experimental data.

We now consider the layered cuprate Sr$_2$CuO$_2$Cl$_2$, whose intra-layer spin-spin coupling of the Cu$^{2+}$ ions has been proposed [18] to be governed by the Hamiltonian [19] with $J = 1450$ K. As for the anisotropy parameter $\Delta$, the experimental analysis [15] hands us the renormalized value $\Delta^{\exp} = 0.00014$, 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 $G = 4JSZ_c\sqrt{2\Delta^{\exp}}$, where $Z_c$ is the zero-temperature value of the spin-wave velocity renormalization coefficient of the isotropic Heisenberg system [16]. The above $\Delta^{\exp}$ is not hence the bare value appearing in Eq. (4), as it already contains quantum renormalizations. In fact, a more refined self-consistent spin-wave-theory calculation [14] leads to the expression $G = 4JSZ_c\sqrt{2\Delta}$ with $Z_\Delta = 0.099$, and hence $\Delta = \Delta^{\exp}/Z_\Delta \simeq 0.0014$.

Experimental data relative to the uniform susceptibility of Sr$_2$CuO$_2$Cl$_2$ (from Refs. [11] and [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 $\chi_{uu}^{\alpha \alpha}$. The agreement is excellent for both the in-plane and the out-of-plane component; considering that no ad-
justment has been introduced for the temperature axis, the position of the minimum of $\chi^{zz}_u$ is nicely reproduced, and a crossover temperature $t_{co} \approx 0.227$ 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 behaviour 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 intra-layer 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 identifiable in the disordered phase: an $\chi^{zz}_u$ of Sr$_2$CuO$_2$Cl$_2$ (from Ref. 11) with $g=2$; thin and thick $+\cdot$ of Sr$_2$CuO$_2$Cl$_2$ (from Ref. 11) with $g=2$ and $g=2.46$, respectively. Other symbols and error bars as in Fig. 1.

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