Vector chiral spin liquid phase in absence of geometrical frustration

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(Dated: June 28, 2010)

Making use of detailed classical Monte Carlo simulations, we study the critical properties of a two dimensional planar spin model on a square lattice composed by weakly interacting helimagnetic chains. We find a large temperature window where the vector chirality order parameter \( \langle \kappa_{jk} \rangle = \langle S_j \times S_k \rangle \), the key quantity in multiferroic systems, takes nonzero value in absence of long-range order or quasi-long-range order, so that, our model is the first example where, at finite temperatures, a vector chiral spin liquid phase in absence of geometrical frustration is explicitly reported. We also show that the strength of interchain interaction is fundamental in order to obtain the vector chiral spin liquid phase. The relevance of our results for three-dimensional models is also discussed.

PACS numbers: 75.30.Kz, 75.10.-b, 75.40.Mg, 77.80.-e

Geometrical frustration and/or competition between interactions can lead to exotic noncollinear magnetic thermodynamic phases, which can be characterized by unusual order parameters. Particularly relevant are two order parameters: scalar chirality \( \langle \chi_{jkl} \rangle = \langle S_j \times S_k \cdot S_l \rangle \) and vector chirality (or spin current) \( \langle \kappa_{jk} \rangle = \langle S_j \times S_k \rangle \). These two chiralities present different symmetries: nonzero value of \( \langle \chi_{jkl} \rangle \) implies that the time-reversal symmetry is broken, while parity symmetry breaking comes with \( \langle \kappa_{jk} \rangle \neq 0 \). Both of them are relevant in strongly correlated electron systems: a nonzero \( \langle \chi_{jkl} \rangle \) gives rise to large anomalous Hall effect \cite{1} and leads to orbital electric currents in frustrated geometries \cite{2}, while new electromagnetic phenomena emerge in Mott insulators as a consequence of induced \( \langle \chi_{jkl} \rangle \), generated by the coupling between the \( \langle \kappa_{jk} \rangle \) and an external homogeneous magnetic field \cite{3}. On the other hand, relativistic spin-orbit interaction leads to a coupling between the vector chirality and the electric polarization \cite{3,4} which play a fundamental role in magneto-electric properties. This coupling permits also to obtain experimental informations about the vector chirality (which is difficult to measure owing to the absence of external physical fields that couple directly to \( \kappa_{jk} \)): the chiral components in multiferroic MnWO\(_4\) have been detected by neutron diffraction using spherical polarization analysis as a function of temperature and of external electric field \cite{5}. The vector chirality, which is the argument of this letter, always accompanies helical magnetic order, and it can arise from spontaneous \( Z_2 \) symmetry breaking in systems with competitive exchange interactions \cite{6}, or it can be stabilized by the Dzyaloshinskii-Moriya antisymmetric exchange interaction in noncentrosymmetric compounds, \cite{6} \cite{10} \cite{11} \cite{12}. However, the vector chiral symmetry can be broken also in a magnetically disordered state. Such phase is named a vector chiral spin liquid phase and has been intensively studied in the last years. It has been predicted to occur in one-dimensional (1\( d \)) frustrated quantum magnetic systems \cite{12} \cite{14}. For higher dimension \( d \) it is crucial to understand if the vector chiral spin liquid phase is stable also in presence of thermal fluctuations \cite{14}. For \( d=2 \), this phase has been clearly obtained at finite temperature \( T \) by classical Monte Carlo (MC) simulation of a triangular lattice of spins with bilinear and biquadratic interactions \cite{15}. However, for models without geometrical frustration and \( d=2,3 \), a clear evidence of this exotic phase is yet lacking, even if Onoda and Nagaosa \cite{16} \cite{17}, investigating a Ginzburg-Landau Hamiltonian describing helical magnets, suggest that a vector chiral spin liquid phase can be stabilized even in \( d=3 \). This prediction was questioned by Okubo and Kawamura \cite{18}, because the results of their classical MC simulations do not show any evidence of such phase, but only a first order phase transition to a helimagnetic order. In this context, it is important to note that in the quasi-1\( d \) \( XY \) organic magnet Gd(hfac)\(_3\)NITEt \cite{19} (a compound with high value of spin, \( S=7/2 \)) a 3\( d \) vector chiral spin liquid phase has been experimentally observed. This result is due to the fact that \( d=1 \) is the lower critical dimension for an Ising order parameter, like \( \langle \kappa_{jk} \rangle \), so that the chiral correlation length, which diverges exponentially at low \( T \), is much larger than the spin correlation length, which diverges with a power law. Taking into account the interchain interaction within mean field approximation a 3\( d \) vector chiral spin-liquid phase results at intermediate \( T \) \cite{20}. However, theoretical results obtained considering also the interchain fluctuations are still lacking, and a direct numerical evidence of such a chiral phase in quasi-1\( d \) system will be relevant.

In this letter we present the results obtained by employing accurate classical MC simulation techniques to investigate a 2\( d \) spin system composed by weakly interacting helimagnetic chains. Despite the absence of geometrical frustration (the model being defined on a square lattice), a clear separation can be observed between the chiral phase transition temperature \( T_c \) and the Kosterlitz-Thouless (\( KT \)) one \( T_{KT} \) separating the quasi-long-range spin ordered phase from the disordered one.
We consider a simple square lattice on the \((x,y)\)-plane composed of \(N=L \times L\) planar spins \(\vec{S}_{i,j}\) (\(|\vec{S}|=1\)), whose interactions are described by the Hamiltonian:

\[
H = - \sum_{i=1}^{L} \sum_{j=1}^{L} \left\{ J_1 \vec{S}_{i,j} \cdot \vec{S}_{i,j+1} + J_2 \vec{S}_{i,j} \cdot \vec{S}_{i,j+2} + J' \vec{S}_{i,j} \cdot \vec{S}_{i+1,j} \right\} .
\]  

(1)

\(j\) labels spins along each chain, while \(i\) is the chain label. Intra-chain exchange interactions are ruled by a nearest neighbour (NN), ferromagnetic (FM) coupling constant \(J_1\) and a next-nearest neighbour (NNN), antiferromagnetic (AFM) coupling \(J_2\); interchain NN spin interactions are ruled by the FM coupling constant \(J'\). If the condition \(\delta=|J_2|/J_1 > 1/4\) is fulfilled, the ground state corresponds to a helical order along the chains, with a pitch vector \(q_0 = \pm \cos^{-1}(-1/\delta)\). In the following we take \(\delta=0.3\) (i.e. \(J_1=1\), and \(J_2=0.3\)), while \(T\) will be given in units \(J_1\). Periodic boundary conditions have been applied along the direction perpendicular to the chains while, due to the incommensurate helix modulation, free boundary conditions were taken along the chain direction. Configuration sampling has been carried on making use of the usual Metropolis technique, while correlations between sampled configurations were mitigated by microcanonical over-relaxed moves\[21\]. For each \(T\) at least three different runs have been performed, each run being composed of \(24 \times 10^6\) MC sweeps, with the first \(4 \times 10^6\) thermalization steps being discarded. Near the critical regions the multiple-histogram (MH) methods \[21\] were employed. Results for \(L=24-128\) are reported.

The weakly interacting chain system has been investigated by setting \(J'=0.1J\): In Fig. 1a the obtained results for the specific heat, \(c_v\), vs. \(T\) are reported. We observe a well defined narrow and sharp peak at \(T \approx 0.12\), which should be ascribed to the onset of a vector chiral spin liquid phase. In this \(T\) range, the scaling behavior of \(c_v\) for different \(L\) is reported in Fig. 1a: We immediately observe as increasing \(L\) the peak more and more acquires the typical features associated with a second order phase transition in the thermodynamic limit. Increasing \(T\), a second broad and size-independent peak is observed at \(T \approx 0.2\), which is consistent with a \(KT\) scenario. In order to estimate the critical temperatures, we employ the Binder’s fourth cumulant \(u_4=1−\langle O^4 \rangle/3\langle O^2 \rangle\)\[21\], where \(O\) will be the chirality, \(\kappa=K'\sum_{ij} \left[ \vec{S}_{ij} \times \vec{S}_{i,j+1} \right]^2\) (where \(K'=[L(L-1)\sin q_0]^{-1}\)), or the helical order parameter, \(m_{HM}\), defined as \(m_{HM}=K'' q q_0 S(\vec{q})\), where \(S(\vec{q})\) is the structure factor, with \(\vec{q}=(0,q_0)\), and the normalization factor \(K''\) is the reciprocal of the structure factor integral at \(T=0\)\[22\]. The Binder cumulant for different \(L\) is reported in Fig. 2a and b for the chirality and the helical order parameters, respectively. For the chirality we can evaluate \(T_\kappa=0.1176(6)\), while for the helical order parameter, we obtain \(T_{KT}=0.1095(5)\). The data in Fig. 2 allows us to assert that the two critical temperatures are well distinguishable with \((T_\kappa-T_{KT})/T_{KT} \approx 7.4\%\). The identification of the crossing temperature in Fig. 2 as the \(KT\) transition temperature can be further validated by the identification of the finite-size scaling (FSS) relation \(\chi_m(L) \propto L^{\nu/\nu'}\); a best fit procedure gives \(\gamma/\nu=1.77(3)\) (Fig. 3a), fully consistent with the \(KT\) behaviour of a 2d planar system. Another evidence of the presence of two distinct critical points comes from the scrutiny of
the vortex density, $\rho$ \cite{28}. In the dilute-gas approximation, we have $\rho \sim \exp(-2\mu/T)$, where $2\mu$ is the energy required to create a pair of vortices \cite{23}, and it can be obtained by linear fit of $-\ln \rho$ as a function of $T^{-1}$ (Fig. 3b). Three different regimes can be identified: low-$T$ ($T<T_{KT}$), intermediate-$T$ ($T_{KT}<T<T_{\kappa}$), and high-$T$ regime ($T>T_{\kappa}$). The linear fit in the range $T_{KT}<T<T_{\kappa}$ (solid line) shows an activation energy of dissociated vortex pairs greater than that obtained for lower temperatures $T<T_{KT}$, where all vortex pairs are bounded, with a clear slope change at $T\sim T_{KT}$. Finally, the creation of other dissociated vortex pairs appears again easier in the region $T>T_{\kappa}$, where $\mu$ strongly decreases signaling the onset of a complete disorder \cite{24}.

A proper characterization of the vector chiral spin liquid transition involves several aspects. A first issue, concerning the order of the transition, can be coped with by analyzing the equilibrium energy distribution at $T=T_{\kappa}$. Even at the largest simulated size of the lattice, $L=128$, no double-peaked structure was observed, so that we have no explicit indication in favor of classifying the chiral transition as a first-order one.

The universality class pertaining to the vector chiral spin liquid transition has been investigated by an accurate FSS analysis. In Fig. 4, the chiral susceptibility, $\chi_\kappa$, is displayed for different values of $L$. From the expected dependence of the peak position temperature on $L$, $T_c(L)=T_c + cL^{-1/\nu}$, we can estimate the critical exponent $\nu$, making use of the value of $T_c$ previously obtained from the Binder’s cumulant discussed above, getting $\nu=1.02(5)$ (Fig. 4a). Analyzing the peak values of $\chi_\kappa$ with the FSS relation $\chi_\kappa(L) \propto L^{\gamma/\nu}$, we obtain the ratio $\gamma/\nu = 1.66(7)$ (Fig. 4b), which implies $\gamma=1.70(8)$. These values of $\gamma$ and $\nu$ are in very fair agreement with $\gamma=7/4$ and $\nu=1$, i.e. the proper values of the Ising universality class in 2$d$. Concerning the critical exponent $\alpha$ for $c_v$, which for the Ising universality class in 2$d$ is $0$, the $c_v$-peak values, $c_v^{\text{max}}(L)$, vs. $L$ are very well fitted (see Fig. 1c) by the FSS relation proper of the 2$d$ Ising model $c_v^{\text{max}}(L)=A+B\ln(L)+CL^{-1}$ \cite{22}. This, allows us to conclude that $\alpha=0$, confirming the Ising character of the vector chiral spin liquid transition.

We point out that the quasi-1$d$ nature of the model is fundamental in order to obtain a vector chiral spin liquid phase in absence of (quasi-)long-range order. This has been explicitly checked by MC simulations we have performed assuming $J'=J_1$, a model investigated by Garel and Doniach \cite{26} many years ago. In Fig. 5 a summary of the obtained results is reported.

For the specific heat, $c_v$, reported vs. $T$, we observe a size-independent broad peak at $T\approx 0.75$, consistent with a $KT$ scenario, while a size-dependent, narrow peak at $T\approx 0.34$ is found. Using the helicity modulus $\Upsilon(T)$ \cite{27}, one is able to evaluate $T_{KT}$ taking advantage of the universal jump: $\Upsilon(T)/T \to 2/\pi$ for $T \to T_{KT}$. We estimate $T_{KT}\approx 0.45$. For the largest simulated size ($L=108$), $\chi_\kappa$ shows a narrow peak at the same temperature $T_{\kappa}$ of the narrow, size-dependent, $c_v$ peak and, above all, $T_{\kappa}$ is significantly lower than $T_{KT}$. These observations are corroborated by the data obtained for a new parameter defined as $M=\frac{1}{L} \sum_{i=1}^{L} x_i$ where $m_i=\sqrt{\left(\frac{1}{L} \sum_{j=1}^{L} S_{i,j}x\right)^2 + \left(\frac{1}{L} \sum_{j=1}^{L} S_{i,j}y\right)^2}$ is the columnar magnetization perpendicular to the helical displacement. This observable turns out to be relevant both for the
chiral phase and for the establishment of the $KT$ phase: Indeed, both two- and four-points correlations contribute to its susceptibility $\chi_M$ (Fig. 3), which displays a first anomaly, which progressively stabilizes at $T_{KT}$ when $L$ increases, signaling the onset of a quasi-order; subsequently, at lower-$T$, $\chi_M$ has a second anomaly consistent with those already displayed by $c_v$ and $\chi_c$. So, we can definitely estimate $T_c\simeq0.34$. We conclude that, for $J'=J_1$, we have a clear separation between the $KT$ behaviour and chiral setup, but, at variance with the quasi-1$d$ case, the onset of the chiral order is established at a temperature $T_c$ lower than $T_{KT}$.

In conclusion, we have presented the outcomes of intensive MC simulations for a 2$d$ $XY$ classical spin system, defined on a square lattice, composed by weakly interacting frustrated chains. We observe a clear separation between the vector chiral spin liquid phase and the quasi-long-range ordered phase, with $T_c<T_{KT}$. We have found that, in a system without geometrical frustration, the chirality displays a second order phase transition, consistent with the 2$d$ Ising universality class. This result confirms the intriguing possibility of an emergent finite-temperature phase showing chiral long range order in the absence of the helical one as investigated by many authors in the multiferroic context [14–17]. We found the quasi-1$d$ nature of the system being fundamental in order to observe such an exotic phase in absence of (quasi-)long-range order: indeed, assuming the same NN exchange constants in both directions ($J=J_1$) we find that the sequence of phase transitions can be reversed $T_c<T_{KT}$. The opposite sequence of the two phase transitions for the investigated 2$d$ models can also give indications for the behavior of their 3$d$ counterparts. As we move from $d=2$ to $d=3$, the $KT$ phase transition is replaced by a proper second order phase transition to a helimagnetic spin arrangement which implies an underlying chiral one, so that it is not surprising that in their MC simulation Okubo and Kawamura [18] do not observe a chiral phase but only the phase transition to the helical order, which also entails chiral order. On the contrary, for a 3$d$ collection of weakly interacting helimagnetic chains, it is reasonable to hypothesize that the phase transition to helimagnetic order occurs at a lower $T$ than the chiral one, and the vector chiral spin-liquid phase can manifest [20] according to the scenario recently emerged from experiments on the quasi-1$d$ organic high-spin magnet Gd(hfac)$_3$NITet [19].

Acknowledgments: F.C. thanks INSTM, the Natural Science and Engineering Research Council of Canada under Research Grant 121210893, and the Alberta Informatics Circle of Research Excellence (iCore); financial support was also given by the Italian Ministry of University within the 2008 PRIN program (contract N. 2008PARRTS_003).

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