Spiral order induced by distortion in a frustrated square-lattice antiferromagnet.

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In a strongly frustrated square-lattice antiferromagnet with diagonal coupling \( J' \), for \( \alpha = J/(2J') \lesssim 1 \), an incommensurate spiral state with propagation vector \( \mathbf{Q} = (\pi \pm \delta, \pi \pm \delta) \) near \((\pi, \pi)\) competes closely with the Néel collinear antiferromagnetic ground state. For classical Heisenberg spins the energy of the spiral state can be lowered as it adapts to a distortion of the crystal lattice. As a result, a weak superstructural modulation such as exists in doped cuprates might stabilize an incommensurate spiral phase for some range of the parameter \( \alpha \) close to 1.

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An interplay between small distortion of the crystal lattice and the magnetic properties of the material is currently a subject of intense research. One problem which supplies strong motivation for such studies is that of stripe order in the lightly doped high-\( T_c \) cuprates \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4+y \) (LSCO) and in related nickelates \([1,2]\). These phases are always associated with a weak superstructural distortion of the original “stacked square lattice” structure of the un-doped parent material. Incommensurate magnetism in these compounds is usually interpreted in terms of a segregation of the doped charges into lines which separate the antiferromagnetic domains (“stripes”) characteristic of the un-doped material. Although modulation of the crystal structure which is induced by charge-stripe segregation is often too small to be observed in experiment \([3]\), it is clear that essential result of the stripe order for the spin system of cuprates is a periodic modulation of the exchange coupling in the Heisenberg spin Hamiltonian which describes their magnetic properties \([3]\). So far, though, only the simplest “average” consequence of the stripe superstructure, in the form of the effective weakening of exchange coupling in the direction perpendicular to the stripes, has been considered \([3]\). A similar problem, of an interplay between the spin order and the cooperative Jahn-Teller distortion accompanying the charge order, arises in the context of the charge-ordered phases in doped manganites \([3]\).

Because the low-energy magnetic properties of layered LSCO cuprates are believed to be adequately described by the two-dimensional (2D) Heisenberg spin Hamiltonian, this model has recently become a focus of intense research. Special attention was devoted to the frustrated square lattice, where in addition to the nearest-neighbor exchange interaction, \( J > 0 \), there is a diagonal coupling, \( J' > 0 \), such that \( \alpha = J/(2J') \) is close to 1. It was originally motivated by the predictions that non-Néel resonating valence bond states \([4,5]\) and quantum-critical behavior \([5]\) associated with the \( T = 0 \) order-disorder phase transitions which may occur in this case might be important for the physics of the superconductivity in cuprates.

Despite RVB spin-liquid state and quantum criticality are strongly predicated upon the quantum nature of the spins (\( S=1/2 \) in cuprates), a semiclassical spin-wave theory appears to provide a surprisingly good guidance to the behavior of the frustrated square-lattice antiferromagnet (FSLA) \([6,7,10,11,12,13,14,15]\). Perhaps, this is because the phenomenon of frustration mainly rests on the ground state degeneracy which exists for classical, as well as for quantum spins. In fact, existence of the spin-liquid phase possibly related to the RVB state in the FSLA for the range of the parameter \( \alpha \) around \( \alpha = 1 \) was first conjectured in Ref. \([6]\) on the basis of the conventional spin-wave calculation to the order 1/\( S \). This suggestion was then supported by the field-theory methods \([16]\), numerical calculations \([11,13,12]\) and other studies \([12,14,15]\). It was established that a disordered phase, whose nature is still controversial, is realized for \( 0.8 \lesssim \alpha \lesssim 1.1 \). Although these studies were essentially aimed at understanding the physics of doped LSCO and related materials, the lattice modulation was generally ignored. One reason for this is that, traditionally, the lattice distortion in a spin system is treated by switching to a larger unit cell, with multiple different spin sites. This approach is not viable for the long-periodic superstructures, and is not possible for the charge-ordered states with incommensurate modulation.

In contrast with the previous studies, present paper addresses the consequences of the superstructural lattice distortion for the ground state of the 2D Heisenberg spin Hamiltonian with classical spins, ie essentially presents an “unrealistic” mean-field (MF) treatment of the realistic spin model. Although MF results are subject to significant quantum corrections, especially for small spins \( S \lesssim 1 \), they nevertheless provide useful guidance about the hierarchy of the competing ground states (GS) in the system. In fact, the MF ground state very often survives account for quantum and thermal fluctuations, as it does for the un-frustrated 2D antiferromagnet.

Main finding of this paper is that a weak superstructural modulation of the crystal lattice in the FSLA may stabilize an incommensurate spin-spiral ground state with the propagation vector \( \mathbf{Q} = (\pi \pm \delta, \pi \pm \delta) \) close to \((\pi, \pi)\) for \( \alpha \leq 1 \). Although in the absence of a structural modulation the energy of the spiral states is higher
than that of the collinear Néel states illustrated in Fig. 1(a) (except for $\alpha = 1$), they are in close competition for $\alpha$ near 1. While spin spiral is usually ignored in the analysis of the possible phases in FSLA, in presence of a superlattice modulation it might actually win the competition for some range of $\alpha \lesssim 1$. Here this is shown explicitly on the mean field level, by treating the effect of a small but otherwise quite arbitrary lattice distortion, as a perturbation in the microscopic classical-spin Heisenberg Hamiltonian.

Consider a system of $N$ equivalent spins on a square lattice, Fig. 1 (a), coupled by Heisenberg exchange interaction, $\mathcal{H} = \sum_{i,j} J_{ij} (S_i S_j)$. While only coupling between the nearest neighbors along the side ($J$) and along diagonal ($J'$) will be of interest in this paper, here $J_{ij} = J_{ji}$ parameterize a general exchange coupling between the spins at arbitrary lattice sites $i$ and $j$. In the absence of a distortion the MF classical ground state is a planar transverse spin spiral, $S_j = (S \cos(Q r_j), S \sin(Q r_j), 0)$, [17, 18]. The ordering wave vector $Q$ corresponds to the minimum of the lattice Fourier transform of the exchange interaction, $J_Q = \sum_{r_{ij}} J_{ij} \exp(-i q r_{ij}), \ r_{ij} = r_j - r_i$. This GS is obtained by finding the minimum-energy configuration for the Heisenberg Hamiltonian with classical spins under the constraint that $S_{ij}^2 = S^2$ for all sites $j$. In general case, spontaneous symmetry breaking is defined by the two mutually perpendicular spin vectors which determine the polarization of the spiral, i.e by the Fourier transform of the lattice spin distribution, $S_Q = S' + i S''$. For collinear situations, such as ferro- or antiferromagnet, corresponding to $Q = 0$ and, e.g. $Q = (\pi, \pi)$, respectively, only a single vector is needed for the order parameter.

A slight distortion of the crystal structure which is characterized by the appearance of the additional, weak superlattice Bragg reflections at wavevectors $\pm Q_c$, corresponds to a small harmonic modulation of the ionic positions, $(r_j)^{'} = r_j + \epsilon_1 \cos(Q_j r_j) + \epsilon_2 \sin(Q_j r_j)$. In most general case, this results in a harmonic modulation of the exchange coupling. It has either the same wavevector $Q_c$, if it appears as a first-order correction to $J_{ij}$ in small parameter $\epsilon \sim \frac{(Q_{\alpha})}{(Q_{\alpha})} \ll 1$, or the wavevector $2Q_c$, if it appears only in the second order, $\sim \epsilon^2$, [19]. There is also a second-order correction to the bond energy, $J_{ij} = J_{ij} + \delta J_{ij}$. The spin Hamiltonian becomes,

$$\mathcal{H} = \sum_{i,j} \left( J_{ij} + j_{ij} e^{iQ_c r_{ij}} + j_{ij}^* e^{-iQ_c r_{ij}} \right) S_i S_j . \tag{1}$$

Here the tildes were omitted, and the complex $j_{ij} = j_{ij}' + j_{ij}''$ was introduced. While without distortion $J_{ij}$ would satisfy all symmetries of the lattice, exchange constants in Eq. (1) possess only those symmetries of the un-distorted lattice which preserve $Q_c$ and the distortion polarizations $\epsilon_1, \epsilon_2$ (this includes all translations).

The modulated-exchange terms allow umklapp processes which couple $S_Q$ and $S_{Q \pm Q_c}$ in the spin Hamiltonian, and couple these Fourier-components in the equations expressing the conditional minimum of the classical exchange energy. As a result, additional Fourier harmonics, at wavevectors $Q + n Q_c, n = \pm 1, \pm 2, \ldots$, appear in the GS spin structure. It has the form of expansion,

$$S_Q = \sum_n S_{Q + n Q_c} \delta Q + n Q_c + S^{*}_{Q + n Q_c} \delta - Q + n Q_c ,$$

where $S_Q = O(\epsilon^{|n|})$. This corresponds to a bunched spiral [17, 18, 19], based on very general exchange symmetry arguments [18, 19]. Without additional symmetry breaking, the perturbing terms have to be proportional to the non-perturbed order parameter. As a result, the leading new Fourier-components, $S_{Q \pm Q_c}$, are,

$$S_{Q + Q_c} = \frac{\frac{J_Q Q_c + J_Q + Q_c}{\chi_{\perp}(Q_c) \omega_{Q_c}^2 S_{Q_c}^2}}{S_Q + O(\epsilon^3)} , \tag{2}$$

$$S_{Q - Q_c} = \frac{\frac{-J_Q Q_c - J_Q + Q_c}{\chi_{\perp}(Q_c) \omega_{Q_c}^2 S_{Q_c}^2}}{S_Q + O(\epsilon^3)} . \tag{3}$$

Here $\omega_q = \sqrt{2(J_{ij} Q + J_{ij} Q - 2J_{ij})}$ is the spin-wave spectrum in the initial, non-distorted,
single-\(Q\) exchange spiral, \(\chi_{\perp}(q) = \left[ 2(J_q - J_{Q}) \right]^{-1}\)
is its transverse (perpendicular to the spin plane), \(q\)-dependent staggered static spin susceptibility, and
\(J_q = \sum_{\alpha,\beta} J_{ij} \exp(-iqr_{ij}) = \vec{J} - \vec{q}\) is a lattice Fourier-transform of the modulated exchange term. Neglecting the \(O(\epsilon^4)\) terms, the corrected ground state energy is obtained in the second order of the perturbation theory,

\[
E_{GS} = \frac{J_Q S^2}{N} - \frac{\left| \vec{J}_{Q} - \vec{J}_{Q+c} \right|^2}{\chi_{\perp}(Q,c)} S^4.
\] (4)

and, correspondingly, is, in general, lowered by the exchange modulation. This occurs as a result of the appropriate adjustment (bunching) of the initial single-\(Q\) spiral spin structure, through appearance of the additional Fourier-harmonics, \(S_{Q+c,Q}\). In addition, the pitch of the primary spiral component, \(S_{Q}\), may also change, \(Q \to \tilde{Q}\), because the spiral propagation vector, \(\tilde{Q}\), is now defined by the minimum of the corrected energy, Eq. (4).

A singular situation occurs when the lattice modulation has the wavevector \(\tilde{Q}\), which is near the dispersion soft spot of the initial spiral, eg close to its Goldstone mode. In this case corrections (4) - (4) diverge, and the perturbation approach fails, highlighting the sensitivity of spin system to such modulations. In frustrated spin systems, entire soft regions, such as the lines of soft modes, often appear due to the accidental cancellation of the interactions. As a result, such systems must be extremely sensitive to structural distortions. On the other hand, in many important cases, such as a nearest-neighbor non-frustrated antiferromagnet, \(Q\) is a special symmetry point of \(\vec{J}_{Q}\) (\(\vec{J}_Q \sim \epsilon \vec{J}_{Q}\)), and the correction term vanishes. Therefore, simple structures, such as collinear antiferromagnets, are, in general, not sensitive to small lattice modulations. In what follows, the singular cases will be excluded from the consideration.

The results obtained above can now be applied to analyze the effect of lattice modulation in a square-lattice antiferromagnet, which may be of direct relevance for the charge-ordered phases in doped LSCO cuprates and related perovskites. For definitiveness, consider the case of \(n\)-periodic diagonal modulation with \(Q_n = (\frac{\pi}{n}, \frac{\pi}{n})\), where \(n = 2, 3, 4, \ldots\), illustrated in Fig. 1(b) (in LSCO the most stable superstructure occurs for \(n = 8\)).

Without frustration, \(J_q = \epsilon J_q\), and, upon switching to \(Q = q(\alpha + \alpha_2)\) and \(Q' = q(\alpha - \alpha_2)\), the problem is factorized and corrections are essentially the same as for 1D chain. There is no change of the global minimum of classical spin energy, so the nearest-neighbor antiferromagnetism is stable with respect to the bond modulation.

In the frustrated case, \(J_q = 4J \cos Q \cos Q' + 2J' \cos 2Q + \cos 2Q'\), and, if both side and diagonal bonds are modulated, \(J_q = 4J \cos Q \cos Q' + 2J' \cos 2Q + \cos 2Q'\). Upon account for distortion the GS energy is,

\[
E_{GS} = \frac{J_Q S^2}{N} - \frac{\sin^2 Q}{\frac{J}{2} \cos Q \cos Q' + 2J' \cos^2 \frac{\pi}{n} \cos 2Q}. \tag{5}
\]

In the absence of bond modulation, the ground state is determined by the hierarchy of the local minima of \(J_Q\), which only depends on \(\alpha = \frac{\pi}{J'}\). For weak frustration, \(\alpha > 1\), the global minimum is that with \(\sin Q = \sin Q' = 0\). It corresponds to the conventional, collinear Néel antiferromagnetic order with a single propagation vector \(Q = (\pi, \pi)\), and the ground state energy \(\frac{J}{2} E_{(\pi,\pi)} = -4JS^2(1 - \frac{1}{4\pi})\). Although there are four equivalent \(Q\)-points in the Brillouin zone (BZ), \((\pm \pi, \pm \pi), (\pm \pi, \pm \pi)\), which restore the lattice \(C_4\) rotational symmetry, they are related through addition of the appropriate reciprocal lattice vectors \(\tau\), so there is no true GS degeneracy in \(Q\)-space. The only degeneracy is the GS rotational symmetry in spin space, which is left of the \(O(3)\) symmetry of the Heisenberg spin Hamiltonian.

For strong frustration, \(\alpha < 1\), there are two non-equivalent lowest-energy minima of \(J_Q\), they satisfy \(\cos Q = \cos Q' = 0\) and have the GS energy \(\frac{J}{2} E_{(\pi,\pi)} = -4JS^2 \frac{1}{2\pi}\). They correspond to two pairs of equivalent \(Q\)-points in the BZ, \((\pm \pi, 0)\) and \((0, \pm \pi)\), which represent the antiferromagnetic order propagating along the \(X\) and \(Y\) axis, respectively. This double degeneracy in \(Q\)-space can be used to construct a continuum of states which are the linear combinations of the above two. This continuous GS degeneracy is usually described in terms of two decoupled antiferromagnetic sublattices based on the diagonals of the original square lattice, which is transparent for \(J' \gg J\). Each sublattice has an antiferromagnetic order, but there may be an arbitrary angle between the two, because the mean field from one sublattice cancels on the sites of the other, Fig. 1a. This continuous degeneracy is lifted by zero-point or thermal spin fluctuations which prefer collinear arrangements of the two sublattices in the GS. This phenomenon is known as “order from disorder” [21, 22].

Although it is not the focus of this paper, an interesting situation occurs for \(\alpha = 1\), when, on the MF level, there is also a continuous GS degeneracy in the \(Q\)-space. The minimum condition for \(J_Q\) becomes \(\cos Q = \cos Q'\), and is satisfied for any spiral with the propagation vector \(Q\) that belongs to the square with the vertices at \((\pm \pi, \pm \pi), (\mp \pi, \mp \pi)\). They all have the same energy, \(\frac{J}{2} E_{(\pi,\pi)} = -2JS^2 = -4J'S^2\). This continuous \(Q\)-space degeneracy is at the origin of the spin-liquid phase conjectured in FSLA for \(\alpha \leq 1\) [10, 11, 12, 13, 14, 15, 16].

What is important here, is that the spiral states with \(Q \approx (\pi, \pi)\) are in close competition with the collinear states for \(\alpha \leq 1\). In particular, the spiral with the propagation vector defined from \(Q' = 0\), \(\cos Q = -\frac{J}{2\pi}\), i.e. \(Q = (\cos^{-1}(\frac{J}{2\pi}), \cos^{-1}(\frac{J}{2\pi}))\), is a local minimum of
J_Q along the diagonal, \((q,q)\), direction, whose energy in the absence of modulation is \(\pm E_Q = -2\alpha JS^2\). Except for \(\alpha = 1\), though, the energy of this local extremum (and of all other spiral states) is higher than that for the decoupled antiferromagnetic sublattices, \(E_{(\pi,0)}\), and for this reason they are usually ignored. However, it is clear from the Eq. (3) that, while the energy of the antiferromagnetic states is insensitive to the bond modulation, the energy of the spiral state can be lowered as it adapts to the lattice distortion! Therefore, at least on the MF level, a spiral may become the lowest energy state (ie the ground state) for some range of the parameter \(\alpha\) in the vicinity of 1 (whose width is \(\sim O(\epsilon^2)\)). For a long-periodic modulation, \(Q_c \ll 1\), and for \(j' = 0\), it is easy to find that the spiral phase is stable for \(1 - |j/J|^2 < \alpha < 1\). The principal propagation vector of the spiral is obtained by minimizing Eq. (4).

While it would be interesting to study the modulated-exchange Hamiltonian \(\mathbf{1}\) for quantum spins and for the arbitrary values of \(|J_{ij}/J_{ij}|\), this is a formidable task which is beyond the scope of this paper. Here a perturbative scheme is used to find the mean field ground state. It is valid for classical spins, \(S \gg 1\), and for small exchange modulation, \(|J_{ij}/J_{ij}| \sim \epsilon \ll 1\). Nevertheless, it provides an important insight into behavior of the frustrated square-lattice antiferromagnet. A finding that (by selecting the spiral order) exchange modulation effectively destabilizes collinear Néel states preferred by the fluctuations clearly supports the instability of the frustrated square-lattice antiferromagnet with \(J/(2J')\) close to 1 with respect to the bond-modulated states. \(\mathbf{13, 14, 15}\).

The essential results of this paper are summarized by Eqs. (2) - (4). The main finding is that the energy of the equal-spin transverse spiral state can be lowered by the exchange modulation in the Heisenberg spin Hamiltonian. This happens as spiral adapts to the modulation through appearance of the additional Fourier-harmonics, \(S_{Q+nQ'}\), \(n = \pm 1, \pm 2, \ldots\) (bunching). As a result, in frustrated square-lattice antiferromagnet with diagonal coupling \(J'\), such, that \(\alpha = J/(2J')\) is close to 1, lattice modulation may open a region of stability of the incommensurate spiral phase. This “order by distortion” phenomenon competes with “order by disorder”, which prefers collinear arrangements of two antiferromagnetic sublattices. Incommensurate spiral phase with the propagation vector \(Q = (\pi \pm \delta, \pi \pm \delta)\) close to \((\pi, \pi)\) wins for the range \(O(\epsilon^2)\) of \(\delta\) around \(\alpha = 1\).

The arguments presented here provide plausible explanation for the incommensurate spin-ordered phases, which are among the most interesting and puzzling features observed in the doped perovskites, and may also be of direct relevance for the doped LSCO materials. For the Heisenberg spin Hamiltonian on square lattice in the absence of distortion, one needs at least a third-neighbor coupling in order to stabilize the MF spiral ground state.

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