Spectroscopic fingerprints of the frustrated magnetic order in $\text{Li}_2\text{VOSiO}_4$: a $t-J$ model study

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Abstract

We have analyzed theoretically the photoemission spectra of the insulating compound $\text{Li}_2\text{VOSiO}_4$. Recently, this compound has been proposed as the first experimental realization of the frustrated $J_1-J_2$ Heisenberg model. Although it is well known that $\text{Li}_2\text{VOSiO}_4$ is magnetically ordered in a collinear arrangement below $T_N = 2.8K$, there is some controversy about the coexistence of two collinear phases above $T_N$. Using a generalized $t-J$ model we have obtained a complex spectral structure that can be traced back to the underlying collinear magnetic structures. We discuss the possibility to use ARPES experiments as a way to discern among the different scenarios proposed in the literature.

Key words: quantum magnetism, frustration, ARPES
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The Heisenberg model on a square lattice with antiferromagnetic (AF) exchange interactions to first ($J_1$) and second nearest neighbors ($J_2$) is the so-called $J_1-J_2$ model. It has all the necessary ingredients to show a rich $T = 0$ phase diagram: frustrating exchange interactions, low spin $S=1/2$, and low dimensionality. For $J_2/J_1 \geq 0.55$ the ground state is a twofold degenerate collinear phase (see Fig. 1), with nearest neighboring spins aligned ferromagnetically along the $x$ direction and antiferromagnetically along the $y$ direction, or vice versa, characterized by the magnetic wave vector $\mathbf{Q} = (0, \pi)$ or $(\pi, 0)$. So, the $90^\circ$ rotational symmetry of the square lattice is broken. Even if in 2D models with continuous symmetry there cannot be a phase transition at finite temperature, Chandra, Coleman and Larkin (CCL)[2] argued that the twofold degeneracy of the collinear ground state could generate a finite temperature

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Ising-like phase transition, since now there is a discrete symmetry that can be broken, although preserving the spin rotational symmetry. Recent series expansion calculations[3], however, do not find evidence for such a finite $T$ transition. On the other hand, it has been proposed[4] another mechanism in which the coupling of spins with the lattice is taken into account and a finite temperature Peierls-like transition may remove the twofold magnetic degeneracy.

Fig. 1. Collinear ordered ground states of the $J_1 - J_2$ model for $J_2/J_1 \geq 0.55$.

These theoretical issues can be explored in the quasi-2D vanadium oxide $Li_2VOSiO_4$, which has been found to be good experimental realizations of the $J_1 - J_2$ model[5]. In $Li_2VOSiO_4$, the $S = 1/2$ can be assumed localized at $V^{4+}(3d^1)$ ions with competing superexchange interactions to first and second neighbors on a square lattice. NMR[5], neutron scattering[6], resonant x-ray scattering[6], and magnetization measurements[5] indicate that a collinear structure is established below $T_N = 2.8K$. It should be noticed that the collinear transition at $T_N$ is triggered by the diverging 2D spin correlations present in the vanadium layers. Consequently, the $J_1 - J_2$ model is the appropriate one for the analysis of the magnetic properties of $Li_2VOSiO_4[5]$, though the finite Néel temperature is due to 3D residual interactions. Above $T_N$ the situation is less clear. It has been suggested by NMR experiments[5] that just above the Néel temperature the degeneracy between the two possible collinear phases is relieved by a structural distortion. Furthermore, it has been pointed out[7] that magnetic domains of both kind of collinear phases coexist for $T_N < T < J_1 + J_2$. This coexistence has been deduced quite indirectly from the very low spin dynamics observed in NMR measurements. However, neutron scattering diffraction experiments[6] do not reveal any structural phase transition. So, one question that remains to be answered is whether the rupture of the discrete degeneracy between the two collinear phases occurs at $T_N$ or at a higher temperature $T \sim J_1 + J_2$, driven by a purely magnetic mechanism[2].

Consequently, it is worth to look for an alternative experiment able to detect different magnetic structures. For this reason, we propose that angle-resolved
photoemission experiments (ARPES), in which the induced photohole probes the underlying magnetic structure, can give relevant information that can be contrasted with the already existing experimental data. In general, photoemission experiments are not used to extract magnetic information of a compound since the spectrum is not able to distinguish between short and true long range order. However, it has been recently observed\cite{9} that the photohole propagates at different energy scales that are closely related to the underlying magnetic correlations, so that ARPES experiments could be used to discern among different magnetic structures, even short ranged ones. In particular, for magnetic structures where ferro and AF correlations coexist, we have found\cite{9} a many-body quasiparticle excitation that results from the coherent coupling of the hole with the AF magnon excitations, while the ferromagnetic component gives rise to a free-like hole motion at higher energy.

In the present article, we compute hole spectral functions using a generalized $t-J$ model appropriate for $Li_2VOSiO_4$. Using realistic parameters\cite{8} we have found that the photoemission spectra clearly show different features when both collinear phases coexist, or not. This result could be helpful in elucidating the controversy about the magnetic properties of $Li_2VOSiO_4$ in the regime $T_N < T < J_1 + J_2$.

To compute the spectral function corresponding to a photohole induced in a photoemission experiment for $Li_2VOSiO_4$, we use a generalized $t-J$ model that naturally takes into account the dynamics of a hole injected in the $J_1-J_2$ model. This generalized $t-J$ model can be written as

$$
H = -t_1 \sum_{\langle i,j \rangle} (\hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} + h.c.) - t_2 \sum_{\langle i,k \rangle} (\hat{c}_{i,\sigma}^\dagger \hat{c}_{k,\sigma} + h.c.) +
+ J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + + J_2 \sum_{\langle i,k \rangle} \mathbf{S}_i \cdot \mathbf{S}_k,
$$

where $t_1$ ($t_2$) and $J_1$ ($J_2$) correspond to the hopping and the exchange integrals, respectively, to first (second) neighbors. The importance of hopping and exchange interactions to second neighbors is evidenced by the existence of a collinear magnetic order below $T_N$\cite{5} and it is theoretically supported by LDA calculations\cite{8}. In order to calculate the spectral function we use the spinless fermion representation\cite{10}. This is a standard procedure that leads to an effective Hamiltonian that explicitly includes the coupling between the hole and the magnon excitations of the frustrated magnetic order. The effective Hamiltonian can be written as:

$$H =$$

$$+ J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + + J_2 \sum_{\langle i,k \rangle} \mathbf{S}_i \cdot \mathbf{S}_k,$$

$$\text{(1)}$$

$$\text{where } t_1 (t_2) \text{ and } J_1 (J_2) \text{ correspond to the hopping and the exchange integrals, respectively, to first (second) neighbors. The importance of hopping and exchange interactions to second neighbors is evidenced by the existence of a collinear magnetic order below } T_N \text{[5] and it is theoretically supported by LDA calculations[8]. In order to calculate the spectral function we use the spinless fermion representation[10]. This is a standard procedure that leads to an effective Hamiltonian that explicitly includes the coupling between the hole and the magnon excitations of the frustrated magnetic order. The effective Hamiltonian can be written as:}$$
\[ H_{ef} = \frac{1}{N} \sum_k \epsilon_k h_k^\dagger h_k + \frac{1}{\sqrt{N}} \sum_{kk'} M_{kk'} h_k^\dagger h_{k'} \alpha_{k-k'} + \sum_k \omega_k \alpha_k^\dagger \alpha_k \]  
\[ (2) \]

where the operator \( h_k^\dagger (\alpha_k^\dagger) \) creates a hole (magnon) with momentum \( k \). 
\( \epsilon_k = 2t_1 \cos k_y \) is the bare hole energy, 
\( M_{kk'} = \beta_{kk'} v_{kk'} - \beta_{kk'} u_{kk'} \) is the hole-magnon vertex interaction, where 
\( \beta_k = 2t_1 \sin k_x + 4t_2 \sin k_x \cos k_y \), and 
\( u_k, v_k \) are the usual Bogoliubov coefficients. 
\( \omega_k \) is the spin wave dispersion in a collinear order[2]. The expressions for \( \epsilon_k \) and \( \beta_k \) have been given for the collinear order \((\pi,0)\), similar expressions hold for \((0,\pi)\). Treating \( H_{ef} \) within the self-consistent Born approximation (SCBA)[10], a self-consistent equation for the self energy \( \Sigma_k(\omega) \), at \( T = 0 \), can be derived

\[ \Sigma_k(\omega) = \frac{1}{N} \sum_q |M_{kq}|^2 G_{k-q}(\omega - \omega_q), \]

where the Green function is defined as \( G_k(\omega) = (\omega - \epsilon_k - \Sigma_k(\omega))^{-1} \). Once the self energy is calculated, the spectral function can be computed as \( A_k(\omega) = -\frac{1}{\pi} \text{Im} G_k(\omega) \).

Theoretically, at finite low temperatures, \( T < J_1 + J_2 \), it is expected a considerable 2D magnetic correlation length \( \xi \sim \exp(2\pi \rho_s/T) \), where \( \rho_s \) is the spin stiffness. Therefore, the low energy excitations at finite temperatures still resemble the magnon excitations of the ordered ground state. As the spectral function is insensitive to the long range order, a \( T = 0 \) calculation that considers the coupling between the photohole and magnons will be valid for \( T_N < T < J_1 + J_2 \).

Regarding the coexistence of both collinear phases in the range \( T_N < T < J_1 + J_2 \), it has been suggested[7] that the size of the magnetic domains is proportional to the correlation length \( \xi \). If we assume that the domain sizes are sufficiently large, it is a reasonable approximation to consider that the photohole dynamics is only affected by the magnetic domain where it was induced. As a consequence, the expected spectral function in the case of coexistence of both orders will be the incoherent sum of the contribution of each order, that is, 
\( A_k(\omega) = \frac{1}{2}[A_{k}^{(\pi,0)}(\omega) + A_{k}^{(0,\pi)}(\omega)] \), as we assume that the photoelectron is extracted with equal probability from both kind of magnetic domains.

To obtain the spectroscopic fingerprints corresponding to the coexistence, or not, of different magnetic domains for \( T_N < T < J_1 + J_2 \), we have evaluated the spectral functions for each case using the LDA estimated microscopic parameters for \( \text{Li}_2\text{VOSiO}_4 \): \( t_1 = -8.5meV, \ t_2 = -29.1meV, \ J_1 = 0.83K \) and \( J_2 = 9.81K \)[8]. In the upper panel of Figure 2 it is shown the spectral function for a hole with momentum \( k = (\pi,0) \) propagating in the collinear order characterized by the wave vector \( Q = (\pi,0) \). This spectral function
has a finite lifetime resonance located near $\omega = 2t_1$, that can be associated with the hole propagation along the ferromagnetic chains in the $y$ direction. At the bottom of the spectral function there is a weak quasiparticle peak, whose origin can be traced back to the coherent coupling of the hole with the magnons\cite{10,9}. We have observed that the broad resonance disperses along the $k_y$ direction like $\epsilon_k$, with a bandwidth $\sim 4t_1$, while the quasiparticle peak exhibits a much smaller dispersion of order $2J_2$.

As we mentioned above, for the case in which $(\pi,0)$ and $(0,\pi)$ orders coexist (bottom panel of Fig. 2) we have computed the spectral function as the incoherent sum of the contribution of each order. Due to the presence of the two kinds of collinear arrangements, the selected photohole with momentum $k = (\pi,0)$, propagates preferently at two different energy scales, separated by an energy $\sim 6t_1$, and which are related to the propagation along the ferromagnetic chains of each magnetic background. The differences between the spectral function for $(\pi,0)$, $A(k,\omega)_{(\pi,0)}$, and the incoherent sum are most clearly observable for momenta close to $(\pi,0)$ or $(0,\pi)$, while along the Brillouin zone diagonal, $\Gamma \rightarrow (\pi,\pi)$, the spectral functions are equal for both orders. It should be noticed that, in the case of coexistence of phases, the discrete broken lattice symmetry is restored and, consequently, the spectral functions along the high-symmetry directions $\Gamma \rightarrow (\pi,0)$ and $\Gamma \rightarrow (0,\pi)$ coincide.

Another problem that still remains unsolved in $Li_2VOSiO_4$ is the lack of an accurate estimation of the degree of frustration $J_2/J_1$. While there is a general consensus that $J_1 + J_2 \sim 8.2K$, there is no agreement about the ratio $J_2/J_1$, with proposed values ranging from $1.1$\cite{5} to $12$ \cite{8}. Contrary to what happens with the magnetic susceptibility and the specific heat\cite{8}, we have observed that several observable features of the spectra are sensitive to the Hamiltonian pa-
rameters. For instance, within our scheme, the first moment of the spectrum for momentum $k$, that is, its center of gravity $<\epsilon>_k=\int \omega A(k,\omega) d\omega$, is equal to the hole free hopping energy $\epsilon_k$, with a bandwidth $W=4t_1$. The second moment of the spectrum $<\Delta\epsilon>_k=\sqrt{\int (\omega-<\epsilon>_k)^2 A(k,\omega) d\omega}$ characterizes its energy extension and it is equal to the average hole-magnon vertex interaction $\frac{1}{N} \sum_{q} |M_{k,q}|^2 \sim 3t_1 + 5t_2$. Finally, we have found that the weak quasiparticle signal at the threshold of the spectra disperses with a bandwidth $\sim 2J_2$, because of the predominance of AF correlations between second nearest neighbors. As we can see, all the above spectral features are intimately related to the presence of collinear magnetic order, and, in principle, they can be read off directly from ARPES experiments, allowing an experimental estimation of the microscopic parameters.

In conclusion, we have presented a theoretical calculation of the photoemission spectra for $Li_2VOSiO_4$, the first experimental realization of the $J_1-J_2$ model. We have used a generalized $t-J$ model that takes proper account of the photohole dynamics in a collinear magnetic background. We have shown that it is possible to extract spectral fingerprints of the underlying magnetic structures, directly associated with the ferro and AF components of the magnetic order. Therefore, we suggest that ARPES experiments would give further insight into open questions about this compound, complementing already available data. Of particular theoretical interest is the coexistence or not of the two collinear orders, $(\pi,0)$ and $(0,\pi)$, in the temperature range $T_N<T<J_1+J_2$. Our results point out that ARPES spectra can clearly distinguish between both situations, because the presence and dispersion of broad resonances in the spectral functions depend crucially on the underlying magnetic structures. We have shown that these broad resonances correspond to the photohole propagation along the ferromagnetic chains of the collinear domains. Moreover, we have found that several spectral features, like the quasiparticle and the center of gravity dispersions, would allow an experimental determination of the hopping and exchange energies.

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