Spectral Function in Mott Insulating Surfaces

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(Dated: February 7, 2020)

We show theoretically the fingerprints of short-range spiral magnetic correlations in the photoemission spectra of the Mott insulating ground states realized in the $\sqrt{3} \times \sqrt{3}$ triangular silicon surfaces K/Si(111)-B and SiC(0001). The calculated spectra present low energy features of magnetic origin with a reduced dispersion $\sim 10 - 40$ meV compared with the center-of-mass spectra bandwidth $\sim 0.2 - 0.3$ eV. Remarkably, we find that the quasiparticle signal survives only around the magnetic Goldstone modes. Our findings would position these silicon surfaces as new candidates to investigate non-conventional quasiparticle excitations.

It is well known that the strong correlation in electronic systems with an odd number of electrons per unit cell may give rise to Mott insulating (MI) ground states, in contradiction with band theory predictions. Generally, the presence of metal-transition ions with unpaired electrons are responsible for the MI properties of many compounds like the undoped high-Tc cuprates. However, it has also been shown that surfaces characterized by a $\sqrt{3} \times \sqrt{3}$ arrangement of silicon dangling bond surface orbitals can provide the ideal conditions to realize the MI phenomena without metal-transition ions. Although the on-site Coulomb potential $U$ is not so large, the surface state bandwidth is reduced due to reconstructions, becoming the correlation effect important. Recently, Weiting et al. used moment-resolved direct and inverse photoemission spectroscopy to demonstrate that the triangular interface K/Si(111)-(\sqrt{3} × \sqrt{3})-B (hereafter K/Si-B) has a MI ground state. Even though such an insulating state is not necessarily unstable toward antiferromagnetism due to the lack of perfect nesting, it has also been argued that K/Si-B is the first experimental realization of a triangular Heisenberg spin-1/2 model with a 120° Néel order. However, standard density-functional methods combined with exact diagonalization studies suggest a Mott insulator with a non-magnetic ground state. In addition, it was speculated that the reconstructed triangular surface SiC(0001)-(\sqrt{3} × \sqrt{3}) also presents a 120° Néel ordered ground state. Since direct measurements of magnetic order in such surfaces are difficult to implement, the magnetic properties of these MI ground states still remain unanswered.

On the other hand, nowadays it is possible to perform higher resolution photoemission experiments than the previous ones, which would allow a detailed analysis of the single-hole properties in these MI ground states. These issues motivated us to study theoretically the single-hole dynamics in a triangular antiferromagnet (AF) with two objectives: i) to investigate the effect of a frustrated magnetic order in the quasiparticle (QP) behavior, and ii) to obtain spectroscopic fingerprints of magnetic order through the photoemission spectra calculated for realistic parameters of the surfaces K/Si-B and SiC. A careful comparison with future higher resolution spectroscopy experiments could give some information about the underlying short-range magnetic structure.

As a consequence of the magnetic order, we find a strong k-dependence of the spectral function structures. In particular, we show the emergence of spectral weight of magnetic origin between the Fermi level and the measured surface state band, with a strongly reduced dispersion. For the surfaces K/Si-B and SiC, our results show a remarkable vanishing of the QP weight for a large region of the Brillouin zone (BZ) outside a neighborhood of the magnetic Goldstone modes.

To tackle this problem we assume the $t-J$ model, which is supposed to capture the low-energy physics involved in photoemission spectroscopy of antiferromagnetic Mott insulators. In order to solve the model, we use the self-consistent Born approximation (SCBA) complemented with exact diagonalization studies. For non-frustrated AF, it has already been established the success of the SCBA to reproduce exact diagonalization results on small clusters and angle-resolved photoemission spectroscopy (ARPES) experiments.

We assume a surface ground state with a magnetic wave vector $Q = (4\pi/3, 0)$ lying in the surface plane $x-z$, and spin waves as the low energy excitations. We perform a unitary transformation to local quantization axis so as to have a ferromagnetic ground state in the $z'$ direction. Then, we use the spinless fermion $c'_{\uparrow} = h_1^{\uparrow}$, $c'_{\downarrow} = h_2 S^z_1$, and the Holstein-Primakov $S^x_1 = \frac{1}{2}(a_1^+ + a_1)$, $S^y_1 = \frac{1}{2}(a_1^+ - a_1)$, $S^z_1 = \frac{1}{2} - a_1^+ a_1$ representations. These are replaced in the $t-J$ model keeping the relevant terms up to third order. After a lengthy but straightforward calculation, the Hamiltonian results

$$H = \sum_k \epsilon_k h_k^\dagger h_k + \sum_q \omega_q \alpha_q^\dagger \alpha_q$$
In the Hamiltonian (1), \( c_k = -t \gamma_k \) and \( \omega_q = \frac{4}{3} J \sqrt{(1-3 \gamma_q)(1+6 \gamma_q)} \) are the hole and magnon dispersions, respectively. \( \gamma_k = \sum_\delta \cos(k \delta) \) and \( \beta_k = \sum_\delta \sin(k \delta) \) are geometric factors, with \( \delta \) the positive vectors to nearest neighbors, and \( k \) varying in the first BZ of the \( \sqrt{3} \times \sqrt{3} \) surface. The bare hole-magnon vertex interaction is defined by \( M_{kq} = i \beta_k v_{-q} - \beta_k v_{-q} u_q \) with the Bogoliubov coefficients \( u_q = [(1+3 \gamma_q/2+\omega_q)/2\omega_q]^{1/2} \) and \( v_q = \text{sign}(\gamma_q) [(1+\gamma_q/2-3\omega_q)/2\omega_q]^{1/2} \). The free hopping hole term implies a finite probability of the hole to move without emission or absorption of magnons because of the underlying non-collinear magnetic structure. The hole-magnon vertex interaction adds another mechanism for the charge carrier motion which is magnon-assisted. The latter is the responsible for the spin-polaron formation when a hole is injected in a non-frustrated antiferromagnet \([7, 8]\). We will show the existence of a subtle interference between both processes that turns out to be dependent on the momenta. An important quantity that allows us to study the interplay between such processes is the retarded hole Green function that is defined as \( G^R_k(\omega) = \langle AF | h_k | \omega + \omega_q | AF \rangle \), where \( |AF\rangle \) is the undoped antiferromagnetic ground state with a 120° Néel order. In the SCBA the self energy at zero temperature results

\[
\Sigma_k(\omega) = \frac{3t^2}{N_s} \sum_q \frac{|M_{kq}|^2}{\omega - \omega_q - \varepsilon_{k-q} - \Sigma_{k-q}(\omega - \omega_q)}.
\]

We have solved numerically this self-consistent equation for \( \Sigma_k(\omega) \), and calculated the hole spectra function \( A_k(\omega) = -\frac{1}{\pi} \text{Im} G^R_k(\omega) \) and the quasiparticle (QP) weight \( z_k = \left( 1 - \frac{\partial \Sigma_k(\omega)}{\partial \omega} \right)^{-1} |E_k| \), where the QP energy is given by \( E_k = \Sigma_k(E_k) \). Before we discuss the results it is important to mention that, unlike previous works\([3]\), we will concentrate on the behavior of the photoemission spectra for realistic parameters of the surfaces K/Si-B and SiC, what implies a strong coupling regime and a careful extrapolation to the thermodynamic limit (we have studied cluster sizes up to 2700 sites).

For the K/Si-B (SiC) surface, photoemission spectra give a bandwidth \( W \sim 0.3 \text{ eV} \) (\( \sim 0.2 \text{ eV} \)) for the occupied surface electronic band\([1, 2]\). These experiments together with inverse photoemission indicate an effective on-site Coulomb repulsion \( U \sim 1-2 \text{ eV} \) (\( \sim 1.5-2 \text{ eV} \)). Electronic band calculations give a similar value for \( U \) and a wider surface bandwidth \( W \sim 0.61 \text{ eV} \) (\( \sim 0.35 \text{ eV} \)), due to the neglect of correlation effects, leading to a positive \( t \sim 0.07 \text{ eV} \) (\( \sim 0.04 \text{ eV} \))\([1, 4]\). The large \( U/W \) ratios indicate the presence of strong electronic correlations and a considerable suppression of charge fluctuations. In addition, scanning tunneling microscopy measurements on the SiC surface show a clear evidence of a Mott insulating state\([10]\). Theoretical works on the Hubbard model suggest that these surfaces would be located in the antiferromagnetic region of the phase diagram\([11]\). All these estimations point out to \( J/t \sim 0.1-0.4 \) for both surfaces. In that parameter range, we have observed negligible quantitative changes in our results, so we take \( J/t = 0.4 \) as a reference value.

\[
\sum_{k,q} \sum_{\omega} \left[ M_{kq} h_k h_{k-q} \alpha_q + h.c. \right].
\]

In order to test the validity of the SCBA we have compared its results for the single-hole dynamics with Lanczos exact diagonalization (ED) calculations on small clusters. In general, we have found a good agreement for all momenta of the BZ and for \( J/t \lesssim 1 \). In Fig. 1 we show the spectral functions for a cluster size of \( N = 21 \) and in the strong
coupling regime $J/t = 0.4$, for three momenta of the BZ. There is a very good agreement between ED and SCBA results in the whole range of energies. This concordance gives a strong support to our analytical approach and so, from now on, we will focus on the SCBA spectra in the thermodynamic limit.

In Fig. 2 we show the spectral function structures, which can be traced back to the distinct hole motion processes mentioned above. The spectra extend over a frequency range of $\sim 9t$, that is, the non-interacting electronic bandwidth. Similar values has been found in the experiments on the silicon surfaces [1, 2]. For momentum $\Gamma$ (upper panel) both processes contribute coherently to the quasiparticle excitation at the top of the spectra. There is also a small incoherent component centered around $6t$ below the QP peak. When moving away from $\Gamma$, there is a spectral weight transfer from the low energy coherent sector to higher energies. At $M'$ (middle panel) the coherence is completely lost, surviving only a magnetic tail at low energy and a structureless background. At $K$ (lower panel), corresponding to the magnetic vector $Q$, the coherence emerges once again, giving rise to the QP. In the background there is a broad resonance related to the free hopping hole motion with a finite lifetime of order $\sim 4J$.

In Fig. 3 we show the intensity of the QP peak along high symmetry axes and, in the inset, the region of the BZ where the quasiparticle weight is finite for $J/t = 0.4$. Besides the strong k-dependence, it can be observed that there is no QP for momenta outside the neighborhood of the magnetic Goldstone modes ($k = 0, Q$). The existence of the quasiparticle peak is also observed for an ample range of $J/t$ values. In particular, as $J/t$ increases the shaded areas in the inset of Fig. 3 where the quasiparticle weight is finite, increase and only for values $J/t > 2.5$ there is quasiparticle peak all over the Brillouin zone. The non existence of quasiparticle is a striking manifestation of the strong interference between the free and magnon-assisted hopping processes. It is expected that this feature could be observed in any system with short-range non-collinear correlations.

The available photoemission spectroscopy data for the silicon surfaces have a low energy resolution ($\sim 100 - 200$ meV) [1, 2] and it is impossible to discern the energy structure of the surface spectra. The surface bands obtained experimentally are correctly reproduced by the center-of-mass of our spectra. The latter coincides with the free hopping dispersion (see Fig. 4) as it is expected from the exact treatment of the first spectral moment of our theory. Using the transfer integral $t$ obtained from first-principles local-density-approximation calculations, our bandwidths compare very well with the experimental ones, reflecting the narrowing induced by the electronic correlation. For the K/Si-B (SiC) surface we have obtained $W \sim 0.3$ eV ($\sim 0.18$ eV). In Fig. 4 we show the center-of-mass and the photothreshold dispersions for the SiC surface. When the quasiparticle exists, the photothreshold corresponds to the QP energy excitation. As it can be observed, the underlying magnetic structure changes the center-of-mass minimum at the $K$ point to a QP energy local maximum, nearly degenerate with the hole ground state momentum $\Gamma$. There is also an appreciable reduced bandwidth $\sim 10$ meV of the photothreshold dispersion in comparison with the measured surface state bandwidth $\sim 200$ meV. Whereas for the K/Si-B surface the values are $\sim 40$ meV and $\sim 300$ meV, respectively.
In conclusion, we have studied theoretically the hole spectral function in the triangular $t-J$ model for realistic parameters relevant for the silicon surfaces SiC(0001)-($\sqrt{3} \times \sqrt{3}$) and K/Si(111)-$\sqrt{3} \times \sqrt{3}$. Assuming the presence of a long-range magnetic Néel order we have observed the emergence of low energy features of magnetic origin with a reduced dispersion band. As the photoemission spectrum is not sensitive to the asymptotic low energy magnetic properties of the system, we speculate however that it could give important information about the presence of short-range magnetic order. We have also obtained an unexpected vanishing of the QP weight for a large region of the Brillouin zone for these Mott insulating surfaces. Our theoretical predictions could provide useful ground to the analysis of future improved photoemission experiments. Using a simple and reliable analytical method (SCBA), we have found clear signatures of interesting physics caused by strong electronic correlation on simple silicon surfaces.

This work was done under PICT Grant No. N03-03833 (ANPCyT) and was partially supported by Fundación Antorchas.

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