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The spectral function for Mott insulating surfaces

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Abstract
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 ~10–40 meV compared with the centre-of-mass spectra bandwidth ~0.2–0.3 eV. Remarkably, we find that the quasiparticle (QP) signal survives only around the magnetic Goldstone modes. Our findings position these silicon surfaces as new candidates for investigation in the search for non-conventional QP excitations.

It is well known that the strong correlations 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 transition metal ions with unpaired electrons is responsible for the MI properties of many compounds such as the undoped high-T_c 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 for realizing MI phenomena without transition metal ions [1–4]. Although the on-site Coulomb potential U is not very large, the surface state bandwidth is reduced due to reconstructions, the correlation effect becoming important. Recently, Weitering *et al* [1] used moment-resolved direct and inverse photoemission spectroscopy to demonstrate that the triangular interface K/Si(111)-($\sqrt{3}\times\sqrt{3}$)–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 (ED) studies suggest a Mott insulator with a non-magnetic ground state [5]. In addition, it was speculated that the reconstructed triangular surface SiC(0001)-($\sqrt{3}\times\sqrt{3}$) [2] (hereafter SiC) also presents a 120° Néel ordered ground state [3]. Since direct measurements of magnetic order in such surfaces are difficult to implement, the magnetic properties of these MI ground states still remain to be established [3].

On the other hand, nowadays it is possible to perform photoemission experiments with higher resolution than the previous ones, which should 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 on the quasiparticle (QP) behaviour; 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 the neighbourhood 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 [6]. In order to solve the model, we use the self-consistent Born approximation (SCBA) complemented with ED studies. For non-frustrated AF, the success of the SCBA in reproducing ED results on small clusters and results from angle-resolved photoemission spectroscopy (ARPES) experiments [6] has already been established [7].

We assume a surface ground state with a magnetic wavevector $\boldsymbol{Q} = (4\pi/3, 0)$ lying in the surface plane x–z, and spin waves as the low-energy excitations. We perform a unitary transformation to the local quantization axis so as to have a ferromagnetic ground state in the z'-direction. Then, we use the spinless fermion ($\hat{c}'_{i\uparrow} = h_i^\dagger$, $\hat{c}^{\dagger\prime}_{i\downarrow} = h_i S_i^-$) and the Holstein–Primakov ($S_i^{x'} \sim \frac{1}{2}(a_i^\dagger + a_i)$, $S_i^{y'} \sim \frac{\mathrm{i}}{2}(a_i^\dagger - a_i)$, $S_i^{z'} = \frac{1}{2} - a_i^\dagger a_i$) representations. These are substituted in the t–J model, keeping the relevant terms up to third order. After a lengthy but straightforward calculation, the Hamiltonian results:

$$H = \sum_{\boldsymbol{k}} \epsilon_{\boldsymbol{k}} h_{\boldsymbol{k}}^\dagger h_{\boldsymbol{k}} + \sum_{\boldsymbol{q}} \omega_{\boldsymbol{q}} \alpha_{\boldsymbol{q}}^\dagger \alpha_{\boldsymbol{q}} - t\sqrt{\frac{3}{N_s}} \sum_{\boldsymbol{k},\boldsymbol{q}} [M_{\boldsymbol{kq}} h_{\boldsymbol{k}}^\dagger h_{\boldsymbol{k}-\boldsymbol{q}} \alpha_{\boldsymbol{q}} + \text{h.c.}]. \tag{1}$$

In Hamiltonian (1), $\epsilon_{\boldsymbol{k}} = -t\gamma_{\boldsymbol{k}}$ and $\omega_{\boldsymbol{q}} = \frac{3}{2}J\sqrt{(1-3\gamma_{\boldsymbol{q}})(1+6\gamma_{\boldsymbol{q}})}$ are the hole and magnon dispersions, respectively. $\gamma_{\boldsymbol{k}} = \sum_\delta \cos(\boldsymbol{k} \cdot \boldsymbol{\delta})$ and $\beta_{\boldsymbol{k}} = \sum_\delta \sin(\boldsymbol{k} \cdot \boldsymbol{\delta})$ are geometric factors, with the $\boldsymbol{\delta}$ the positive vectors to nearest neighbours, and the $\boldsymbol{k}$ varying in the first BZ of the $\sqrt{3} \times \sqrt{3}$ surface. The bare hole–magnon vertex interaction is defined by $M_{\boldsymbol{kq}} = \mathrm{i}(\beta_{\boldsymbol{k}} v_{-\boldsymbol{q}} - \beta_{\boldsymbol{k}-\boldsymbol{q}} u_{\boldsymbol{q}})$ with the Bogoliubov coefficients $u_{\boldsymbol{q}} = [(1+3\gamma_{\boldsymbol{q}}/2+\omega_{\boldsymbol{q}})/2\omega_{\boldsymbol{q}}]^{\frac{1}{2}}$ and $v_{\boldsymbol{q}} = \mathrm{sgn}(\gamma_{\boldsymbol{q}})[(1+\gamma_{\boldsymbol{q}}/2-3\omega_{\boldsymbol{q}})/2\omega_{\boldsymbol{q}}]^{\frac{1}{2}}$. The free hopping hole term implies a finite probability of the hole moving without emission or absorption of magnons because of the underlying *non-collinear* magnetic structure. The hole–magnon vertex interaction adds another mechanism for charge carrier motion which is magnon assisted. The latter is responsible for the spin-polaron formation when a hole is injected in a non-frustrated AF [7, 8]. We will show the existence of a subtle interference between the two 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 $G_{\boldsymbol{k}}^h(\omega) = \langle \mathrm{AF}| h_{\boldsymbol{k}} \frac{1}{(\omega + \mathrm{i}\eta^+ - H)} h_{\boldsymbol{k}}^\dagger |\mathrm{AF}\rangle$, where $|\mathrm{AF}\rangle$ is the undoped antiferromagnetic ground state with a 120° Néel order. In the SCBA the self-energy at zero temperature results as

$$\Sigma_{\boldsymbol{k}}(\omega) = \frac{3t^2}{N_s} \sum_{\boldsymbol{q}} \frac{|M_{\boldsymbol{kq}}|^2}{\omega - \omega_{\boldsymbol{q}} - \epsilon_{\boldsymbol{k}-\boldsymbol{q}} - \Sigma_{\boldsymbol{k}-\boldsymbol{q}}(\omega - \omega_{\boldsymbol{q}})}.$$

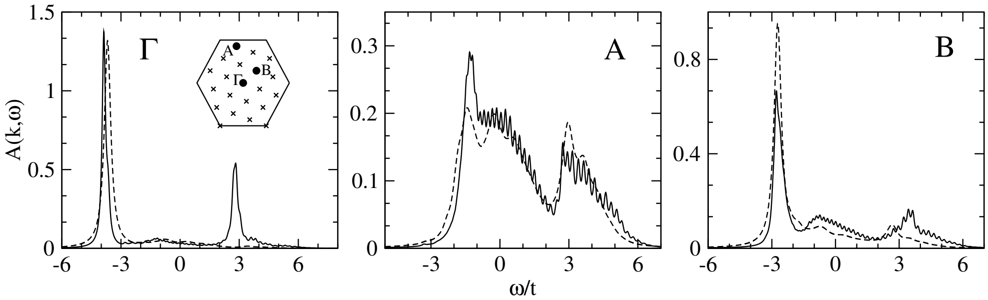

Figure 1. Spectral functions versus frequency for $J/t = 0.4$ and $N = 21$ corresponding to the momenta at BZ points $\Gamma = (0, 0)$, $\mathrm{A} = \frac{4\pi}{21}(-1, 3\sqrt{3})$, and $\mathrm{B} = \frac{4\pi}{21}(2, \sqrt{3})$, shown as filled circles in the inset of the left panel (the crosses represent the other momenta). The solid and dashed curves are the exact and SCBA results, respectively.

We have solved this self-consistent equation numerically for $\Sigma_{\boldsymbol{k}}(\omega)$, and calculated the hole spectra function $A_{\boldsymbol{k}}(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\,G^h_{\boldsymbol{k}}(\omega)$ and the QP weight $z_{\boldsymbol{k}} = \left(1 - \frac{\partial \Sigma_{\boldsymbol{k}}(\omega)}{\partial \omega}\right)^{-1}\Big|_{E_{\boldsymbol{k}}}$, where the QP energy is given by $E_{\boldsymbol{k}} = \Sigma_{\boldsymbol{k}}(E_{\boldsymbol{k}})$. Before we discuss the results it is important to mention that, unlike previous researchers [9], we will concentrate on the behaviour of the photoemission spectra for realistic parameters of the surfaces K/Si–B and SiC, which 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 $W \sim 0.3$ eV ($\sim$0.2 eV) for the occupied surface electronic band [1, 2]. These experiments together with inverse photoemission studies indicate an effective on-site Coulomb repulsion $U \sim$ 1–2 eV ($\sim$1.5–2 eV). Electronic band calculations give a similar value for U and a wider surface bandwidth $W \sim 0.61$ eV ($\sim$0.35 eV), due to the neglect of correlation effects, leading to a *positive* $t \sim 0.07$ eV ($\sim$0.04 eV) [4, 5]. The large U/W ratios indicate the presence of strong electronic correlations and a considerable suppression of charge fluctuations. In addition, scanning tunnelling microscopy measurements on the SiC surface show clear evidence of a MI 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 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.

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

In figure 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 the momentum at Γ (upper panel) the two processes contribute coherently to the QP excitation at the top of the spectra. There is also a small incoherent component centred around $6t$ below the QP peak. On moving away from Γ, there is a spectral weight transfer from the low-energy coherent sector to higher energies. At

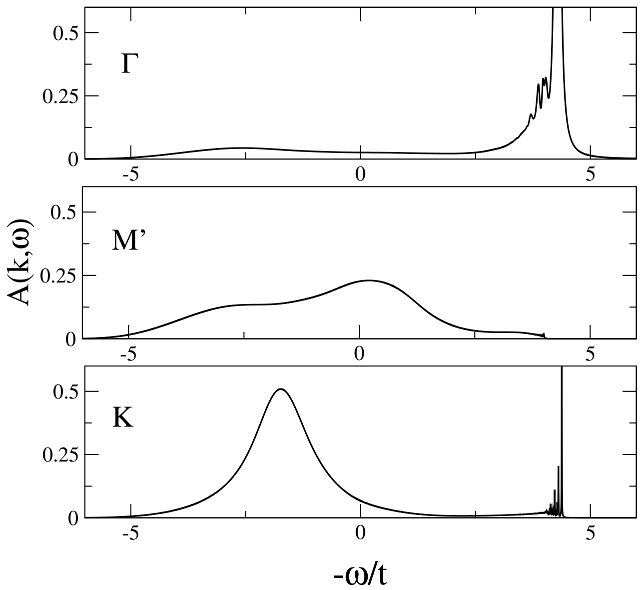

Figure 2. Spectral functions for realistic parameters of the silicon surfaces ($J/t = 0.4$) calculated at three different points of the BZ (see the inset of figure 3). Notice that the Fermi level has been placed on the right.

M′ (middle panel) the coherence is completely lost, only a magnetic tail at low energy and a structureless background surviving. At K (lower panel), corresponding to the magnetic vector $\boldsymbol{Q}$, 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 figure 3 we show the intensity of the QP peak along high-symmetry axes and, in the inset, the region of the BZ where the QP 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 neighbourhood of the magnetic Goldstone modes ($\boldsymbol{k} = \boldsymbol{0}, \boldsymbol{Q}$). The existence of the QP peak is also observed for a large range of J/t values. In particular, as J/t increases the shaded areas in the inset of figure 3, where the QP weight is finite, increase and only for values $J/t \gtrsim 2.5$ is there a QP peak anywhere in the BZ. The non-existence of QP is a striking manifestation of the strong interference between the free and magnon-assisted hopping processes. It is thought 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 (~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 centre of mass of our spectra. The latter coincides with the free hopping dispersion (see figure 4) as 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 [4], 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 (~ 0.18 eV). In figure 4 we show the centre-of-mass and photothreshold dispersions for the SiC surface. When the QP exists, the photothreshold corresponds to the QP energy excitation. As can be observed, the underlying magnetic structure changes the centre-of-mass minimum at the K point to a QP energy local maximum, nearly degenerate

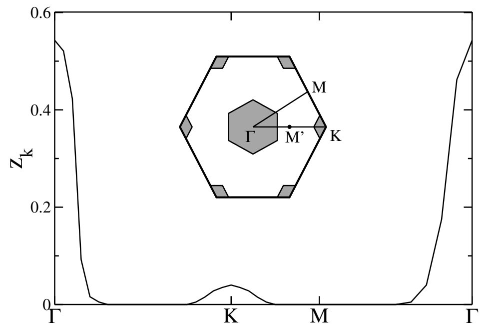

Figure 3. QP intensity along the Γ–K–M–Γ path for $J/t = 0.4$. Inset: the $\sqrt{3} \times \sqrt{3}$ BZ. In the shaded areas, the QP weight is finite.

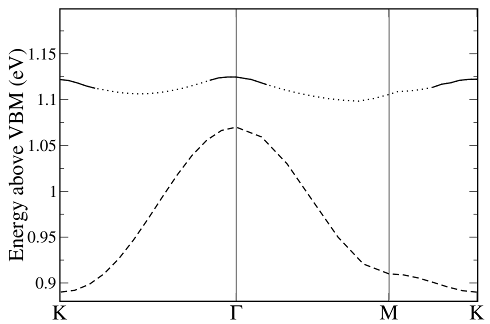

Figure 4. Surface band structure for realistic parameters ($J/t = 0.1$, $t = 0.04$ eV) of the SiC(0001)-($\sqrt{3} \times \sqrt{3}$) surface. The dotted line is the photothreshold energy, the solid one is the QP dispersion, and the dashed one is the centre-of-mass spectra band. Energies are given relative to the measured bulk valence band maximum (VBM). Experimentally the Fermi level is located at 2.3 eV above the VBM.

with the hole ground state momentum at Γ. There is also an appreciably reduced bandwidth, ∼10 meV, of the photothreshold dispersion in comparison with the measured surface state bandwidth ∼200 meV—whereas for the K/Si–B surface the values are ∼40 and ∼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 BZ for these MI surfaces. Our theoretical predictions could provide a useful basis for 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.

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