Itineracy effects on spin correlations in 1D Mott insulators

M. J. Bhaseen,1 F. H. L. Essler,1 and A. Grage2
1Department of Physics, Brookhaven National Laboratory, Upton, NY 11973-5000, USA
2Fachbereich Physik, Philipps-Universit"at Marburg, D-35032 Marburg, Germany
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We consider spin correlations in the one dimensional half filled repulsive Hubbard model. For very large values of the on-site repulsion \( U \) the spin correlations are dominated by virtual hopping processes of electrons and are described in terms of a spin-1/2 Heisenberg chain. As \( U \) is decreased real hopping processes of electrons become important and eventually dominate the spin response. We discuss the evolution of the dynamical structure factor as a function of \( U \). We comment on the relevance of our results for inelastic neutron scattering experiments on quasi-1D materials.

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Quasi-one-dimensional (Q1D) Mott insulators are a paradigm for the importance of strong correlations and are known to exhibit a wide variety of unusual physical phenomena such as spin-charge separation and quantum number fractionalization. Experimental realizations include the chain cuprates SrCuO\(_2\) and Sr\(_2\)CuO\(_3\) and a number of organic compounds \[1, 2\]. It has been known for a long time that the magnetic properties of Q1D Mott insulators are rather unusual. The dynamical structure factor is entirely incoherent and reflects the fact that the elementary spin excitations carry spin 1/2. They are very different in nature from antiferromagnetic spin waves. In the limit of very strong Coulomb repulsion the spin degrees of freedom are commonly modelled by the spin-1/2 Heisenberg chain. This has proved to provide an adequate description of recent inelastic neutron scattering experiments on SrCuO\(_2\) \[3, 4\]. In this limit the low energy dynamics is dominated by spin flips generated through virtual hopping processes of electrons. However, for general Mott insulating materials there is no reason for the Coulomb repulsion to be much stronger than the electron hopping amplitude. Indeed, this situation is realized in the Bechgaard salts. A natural question to ask then is how electron itineracy affects the spin dynamics. This issue has recently attracted much attention in the context of the two-dimensional Mott insulating cuprate La\(_2\)CuO\(_4\) \[5\]. The experimentally observed spin wave dispersion was found to depart significantly from the Heisenberg form, revealing the presence of ring-exchange interactions. This is a direct manifestation of electron itineracy in the sense that ring-exchange interactions are generated by higher order virtual electron hopping processes.

Motivated by these findings we investigate the effects of electron itineracy on the unusual spin correlations in Q1D Mott insulators. We do so in the simplest model that incorporates the relevant physics, the one dimensional half filled Hubbard model. The Hamiltonian is given by

\[
H = -t \sum_{l, \sigma} (c_{l, \sigma}^+ c_{l+1, \sigma} + c_{l+1, \sigma}^+ c_{l, \sigma}) + U \sum_l (n_{l, \uparrow} - \frac{1}{2}) (n_{l, \downarrow} - \frac{1}{2}),
\]

where \( c_{l, \sigma}^+ \) creates an electron with spin \( \sigma = \uparrow, \downarrow \) at site \( l \), \( n_{l, \sigma} \equiv c_{l, \sigma}^+ c_{l, \sigma} \), and \( U \) is the on-site Coulomb repulsion. In momentum space, the kinetic term yields a cosine band,

\[
E_k = 2t \sum_{\sigma} \left| J_1(x) \cos(xU/4t) \right|,
\]

where \( J_1(x) \) and \( J_3(x) \) are Bessel functions, and \( E_k(0) \) plays the role of a “spinon bandwidth”. The holon and antiholon, are gapped, spinless, and carry charge \( |e| \) and \(-|e|\) respectively. The “charge gap” \( \Delta \), defined as the minimum of the holon energy, increases with \( U \) and is given by:

\[
\Delta = -2t + \frac{U}{2} + 2t \int_0^\infty dx \frac{J_1(x) e^{-xU/4t}}{x \cosh(xU/4t)}.
\]

These excitations form a basis of scattering states \[7\] in which one may explore the dynamical response of 1D Mott insulators.

As usual, the dynamical structure factor is obtained from the imaginary part of the Fourier transform of the spin-spin correlation function. In view of the SU(2) spin symmetry of the Hamiltonian we consider:

\[
S_{\uparrow \uparrow}(\omega, q) = -\Im \langle \chi^\uparrow_\uparrow(\omega, q) | \omega \rightarrow \eta - i \omega \rangle,
\]
where \( \eta \to 0^+ \) and the susceptibility (in Euclidean time) is given by

\[
\chi_{EE}^{zz}(\tilde{\omega}, q) = -\int_{-\infty}^{\infty} dx \, dr \, e^{i\tilde{\omega}r - iqx} \langle S^z(\tau, x)S^z(0, 0) \rangle. \tag{6}
\]

The spin operator is given by \( S^z \equiv (n_+ - n_-)/2 \). In the first instance, it is instructive to study the known limits of large and small \( U/t \). In the limit \( U \gg t \), the half filled Hubbard model reduces to an isotropic Heisenberg antiferromagnet with exchange \( J \approx 4t^2/U \) \([8]\). In this case, the structure factor is well approximated by the so-called Müller ansatz \([3]\):

\[
S^{zz}(\omega, q) \propto \frac{\Theta(\omega - \omega_L)\Theta(\omega_L - \omega)}{\sqrt{\omega^2 - \omega_L^2}}; \quad U \gg t. \tag{7}
\]

Whilst more detailed results can be obtained using integrability \([10]\), this approximation is sufficient for our current discussion. The lower and upper spinon boundaries, \( \omega_L = (\pi/2)J/\sin q_0 \) and \( \omega_U = \pi J/\sin (q_0/2) \), follow from the spinon dispersion relations \([2]\) and \([3]\). These boundaries delimit a continuum of two spinon excitations, and we plot this in Fig. 1(a). It is readily seen that the spectral weight (plotted darker) is concentrated on the lower spinon boundary. The power law singularity extends down to \( \omega = 0 \) in the vicinity of \( q = \pi \), and reflects the gapless nature of spinons.

In the limit of large external frequencies and \( U \ll t \), one may treat the Hubbard interaction perturbatively. Using the tightbinding propagator \( G(\tilde{\omega}, q) = a_0[\tilde{\omega} + 2t \cos q]^{-1} \), a summation over “bubble” diagrams yields:

\[
\chi_{EE}^{zz}(\tilde{\omega}, q) = \frac{\chi_{0E}^{zz}(\tilde{\omega}, q)}{1 + 2(U/a_0)\chi_{0E}^{zz}(\tilde{\omega}, q)}; \quad U \ll t, \tag{8}
\]

where the \( U = 0 \) single bubble is given by

\[
\chi_{0E}^{zz}(\tilde{\omega}, q) = -\frac{a_0}{\pi} \arctan \left[ \frac{4t(\sin q_0/2)^2}{\sqrt{\omega^2 + (4t \sin q_0/2)^2}} \right]. \tag{9}
\]

We plot the corresponding structure factor in Fig. 1(b). In this limit, significant spectral weight is concentrated on the upper spinon boundary. (It is evident that this large \( \omega \) approximation fails to capture the low frequency spinon divergence; see discussion below.) Thus apart from a rescaling of the spinon bandwidth, to which we shall return below and in Fig. 3 there is a manifest redistribution of spectral weight as one changes the ratio \( U/t \). It is therefore highly desirable to build up a better picture for what happens in the intermediate regime \( U \approx t \). We address this pertinent issue using the techniques of integrable field theory.

At low energies, we may linearize the non-interacting fermionic spectrum around \( \pm |k_F| \). The resulting model may be bosonized \([11]\) in terms of two Bose fields, \( \Phi_s \) and \( \Phi_c \). These correspond to the collective spin and charge degrees of freedom respectively. The continuum Hamiltonian density separates in two commuting pieces:

\[
\mathcal{H} = \mathcal{H}_s + \mathcal{H}_c,
\]

where

\[
\mathcal{H}_s = \frac{v_s}{16\pi} \left[ (\partial_x \Phi_s)^2 - v_s^{-2}(\partial_t \Phi_s)^2 \right] - g : \mathbf{J} \cdot \mathbf{J} :, \quad \mathcal{H}_c = \frac{v_c}{16\pi} \left[ (\partial_x \Phi_c)^2 - v_c^{-2}(\partial_t \Phi_c)^2 \right] + g : \mathbf{I} \cdot \mathbf{I} :.
\]

Here \( \mathbf{J} \) and \( \mathbf{I} \) are the SU(2) spin and isospin currents, \( v_s = v_F - Ua_0/2\pi \), \( v_c = v_F + Ua_0/2\pi \) and \( g = 2Ua_0 \). The interaction in the charge sector is marginally relevant and leads to the formation of the charge gap. The interaction in the spin sector is marginally irrelevant and will be neglected. This description is valid for \( \Delta \ll 4t \) and for \( U \lesssim 3t \). In order to enhance the clarity of presentation, we will mainly work in the so-called scaling limit:

\[
t \to \infty, \quad U/t \to 0, \quad \Delta \to m \equiv \frac{4}{\pi} \sqrt{U/t} e^{-2\pi t/U} \text{ fixed.}
\]

In this limit \( v_s \) coincides with \( v_c \) and Lorentz invariance emerges. The charge sector is governed by the SU(2) invariant sine Gordon model, and the spin sector by a massless Gaussian model. This simplifies the ensuing formulæ, whilst capturing the relevant physics. Where appropriate, we give the results for \( v_s \neq v_c \).

In this low energy description, the \( z \) component of the spin operator takes the form:

\[
S^z = -\frac{a_0}{4\pi} \partial_x \Phi_s + (-1)^n \frac{a_0}{\pi} \cos \Phi_s \sin \frac{\Phi_s}{2}. \tag{10}
\]

We draw attention to the fact that both the “spin” and the “charge” fields enter the staggered component of the spin operator. Excitations of the charge field, and thus electron itineracy, may therefore influence magnetic correlations in the vicinity of \( q \sim \pi \). In the first, crude
approximation, the operator $\cos \frac{\Phi}{2}$ is replaced by its constant vacuum expectation value $\langle 0 | \cos \frac{\Phi}{2} | 0 \rangle$, and correlators are given by the critical spin sector alone. However, this is merely the leading term in a systematic expansion of the charge sector. In contrast to naive expectations, these terms are shown to be significant. In the subsequent analysis, we shall calculate the “itinerancy corrections” to the magnetic structure factor arising from these charge excitations. We employ a number of results developed in the study of density-density correlations [12]. As follows from equation [16] the staggered part of the spin-spin correlation function is given by

$$\langle S^z(\tau, x)S^z(0, 0) \rangle_{\text{stagg}, \epsilon} = (-1)^{\epsilon_0} \frac{1}{2\pi^2 v_F} \frac{a_0^2 \mathcal{F}(r)}{r},$$  

where $r = \sqrt{x^2 + \tau^2}/v_F$ and we have defined the contribution from the vacuum expectation sector

$$\mathcal{F}(r) \equiv \langle 0 | \cos \frac{\Phi_c}{2} | \tau, x \rangle \cos \frac{\Phi_c}{2}(0, 0) | 0 \rangle.$$  

We introduce a basis of states $| \theta_n \rangle = | \epsilon_1 \rangle$, where the index $\epsilon = \pm$ denotes holon and antiholon, and $\theta$ are their rapidities. Inserting the resolution of the identity

$$1 = | 0 \rangle \langle 0 | + \sum_{n=1}^{\infty} \sum_{\epsilon_1} \int \frac{d\theta_1 \cdots d\theta_n}{(2\pi)^n n!} | \theta_1 \cdots \theta_n \rangle_{\epsilon_1 \cdots \epsilon_1} \langle \theta_1 \cdots \theta_n |,$$

one may develop [12] in a systematic expansion over multi-particle intermediate states:

$$\mathcal{F}(r) = \mathcal{F}_0(r) + \mathcal{F}_2(r) + \ldots$$  

The leading term is obtained from the vacuum expectation value of the operator

$$\mathcal{F}_0(r) = \langle 0 | \cos \frac{\Phi_c}{2} | 0 \rangle^2 \frac{m}{v_F} | C |^2$$  

and is a constant. The dimension of $\langle 0 | \cos \frac{\Phi_c}{2} | 0 \rangle$ is $[L]^{-1/2}$. The dimensionless constant $C$ is pinned by the short distance properties of the charge sector [13] and fixes the normalization of the structure factor. That is to say, to leading order, the staggered spin-spin correlation function is approximated by the conformal $1/r$ decay. Since the operator $\cos \Phi_c/2$ does not couple to single holon states the next contribution may be written:

$$\mathcal{F}_2(r) = \sum_{\epsilon_1 \epsilon_2} \int \frac{d\theta_1 d\theta_2}{(2\pi)^2} \left[ \langle 0 | \cos \frac{\Phi_c}{2} | \theta_1 \theta_2 \rangle_{\epsilon_1 \epsilon_2} \right]^2 \times \exp \left[ -m \left( \text{ch} \theta_1 + \text{ch} \theta_2 \right) \right] - im \left( \text{sh} \theta_1 + \text{sh} \theta_2 \right) x/v_F].$$

Defining $\theta = \theta_1 - \theta_2$, the form factor is given by [14, 15]

$$f \cos \frac{\Phi_c}{2}(\theta)_{+} \equiv \langle 0 | \cos \frac{\Phi_c}{2} | \theta_1 \theta_2 \rangle_{+} = \sqrt{\mathcal{F}_0} \frac{2m}{\gamma} \frac{\text{sh} \theta}{\theta + i\pi} E(\theta),$$  

where $E(\theta) = \exp \left( -\int_0^\infty \frac{dx}{x} \frac{\sin^2(\theta + i\pi \frac{x}{\pi}) e^{-x}}{\text{sh}(2x) \text{ch}(x)} \right).$  

In view of [13] we may expand the structure factor

$$S^{zz}(\omega, q) = \sum_{n=0}^\infty S^{zz}_{2n}(\omega, q),$$  

where $S^{zz}_{2n}(\omega, q)$ denotes the contribution due to intermediate states with $2n$ (anti)holons. The “conformal” contribution is easily evaluated:

$$S^{zz}_0(\omega, \pi \frac{A}{a_0} + q) = \frac{a_0^2}{v_F} \frac{| C |^2}{| \pi \frac{A}{a_0} + q |} \times \int_0^{\theta'} \frac{d\theta}{\gamma} \frac{8 \text{sh}^2 \theta}{\gamma^2} (\text{E}(\theta))^2 F \left[ \frac{1}{2}, \frac{1}{2}, 1; 1 - \frac{\gamma^2}{2} \right],$$

where $\Theta(y) = 1$ for $y \geq 0$ is the Heaviside step function, and $s \equiv \sqrt{\omega^2 - v_F^2 q^2}$. Performing the sum over isotopic indices and the necessary integrals [12], the “itinerancy correction” is found to be

$$S^{zz}_2(\omega, \pi \frac{A}{a_0} + q) = \frac{a_0^2}{v_F} \frac{| C |^2}{(2\pi)^2} \frac{8 \text{sh}^2 \theta}{\gamma^2} \frac{\text{E}(\theta)^2 F \left[ \frac{1}{2}, \frac{1}{2}, 1 - \frac{\gamma^2}{2} \right]}{\sqrt{\Omega_+ \Omega_-}} ,$$

where $\Omega_\pm = \omega \pm v_s q + 2\Delta \text{ch} \theta \left( \text{ch} \theta \pm \left( v_s/v_c \right) \text{sh} \theta \right)$. In the vicinity of the threshold ($s = 2m$), the integral in (19) may be obtained by Taylor expanding the integrand. The leading threshold behavior as $s \to 2m^+$ is

$$S^{zz}_2(\omega, \pi \frac{A}{a_0} + q) \to \frac{a_0^2}{v_F} \frac{| C |^2}{(2\pi)^2} \frac{8 \text{sh}^2 \theta}{\gamma^2} \frac{(\text{E}(0))^2 F \left[ \frac{1}{2}, \frac{1}{2}, 1 - \frac{\gamma^2}{2} \right]}{\sqrt{\Omega_+ \Omega_-}} \frac{s - 2m}{m}^{3/2}.$$

In contrast to the conformal correction, this term goes to zero at its threshold and is free of singularities. At high energies $s \gg 2m$, $S^{zz}_2(\omega, \pi \frac{A}{a_0} + q) \to$ constant. In real space, the asymptotics of this correction (evaluated in the saddle point approximation) yield:

$$\langle S^z(\tau, x)S^z(0, 0) \rangle_{\text{stagg}, \epsilon} = (-1)^{\epsilon_0} \frac{1}{2\pi^2 v_F} \frac{a_0^2 m}{v_F^2} \text{E}(0) + A e^{-2mr} + \ldots,$$

for $mr \gg 1$, where

$$A = \frac{2\sqrt{2}}{\pi} \text{E}(0) \exp \left( -1 + \int_0^\infty \frac{dx}{x} \frac{e^{-x}}{\text{ch}^2 x} \right).$$

It is apparent from Fig. 2 that the itinerancy correction becomes significant for $s \sim 10m$; for $v_s \neq v_c$,
s ∼ 10Δ. In order to observe this feature within the two spinon continuum, we seek to maximize ∆ subject to 10Δ ≲ 2Es(0) ≪ 40t. As may be seen from the inset of Fig. 2 this condition is satisfied for 2 ≲ U/t ≲ 3. In particular, the spinon bandwidth Eσ(0) decreases with increasing U/t. In view of this we rescale the spinon energies by this factor. As shown in Fig. 3 the rescaled dispersion relations collapse (approximately, but remarkably well) onto a single curve. We may therefore compare systems with different values of U/t on a single plot. In conjunction with the limiting cases, U ≪ t and U ≫ t, our detailed analytic results support the simple yet compelling view depicted in Fig. 3. As U/t increases spectral weight is transferred from the upper rescaled boundary, and builds up in both the conformal divergence and the itineracy correction. The downward shift at high frequencies is evident from the bubble summation and the build up at low frequencies originates in the increase of the charge gap.

In this letter we have determined dynamical spin correlations in 1D Mott insulators. Away from the well-understood “Heisenberg limit” of very large Mott gaps, electron itineracy effects are shown to be quite significant. On the basis of our calculations we have put forward a simple picture describing the spectral weight transfer in the dynamical structure factor as a function of the strength of the Coulomb repulsion. We expect our findings to be of relevance for inelastic neutron scattering experiments on quasi-1D small-gap Mott insulators.

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