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Spiral Magnets as Gapless Mott Insulators.

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Abstract. – In the large-U limit, the ground state of the half-filled, nearest-neighbor Hubbard model on the triangular lattice is the three-sublattice antiferromagnet. In sharp contrast with the square-lattice case, where Goldstone modes never have a charge component, it is shown that beyond leading order in t/U the three Goldstone modes on the triangular lattice, at finite $\boldsymbol{q}$, are a linear combination of spin and charge. This leads to non-vanishing conductivity at any finite frequency, even though the magnet remains insulating at zero frequency. More generally, non-collinear spin order should lead to such gapless insulating behavior.

Non-bipartite lattices, such as the triangular or f.c.c. lattice, have a particular appeal for theoretical studies of correlated electrons because they lift many of the degeneracies present in bi-partite lattices such as simple cubic systems. On the triangular lattice, for example, the ground state of the Hubbard model at half-filling shows a large number of possible zero-temperature phases: paramagnetic, spiral magnetic order, and linear spin density waves in either metallic or insulating phases [1, 2]. This is to be contrasted with the same model on a square lattice which has an antiferromagnetically ordered ground state for all possible values of the interaction.

In this paper, we show that for lattices which do allow spiral (non-collinear) magnetic order in the ground state of the Hubbard Hamiltonian, a new phenomenon occurs. In the large-U limit, but for a finite value of the hopping integral, these is a coupling between charge and spin excitations in the non-collinear spin state. In other words, in such a magnetically ordered state the three Goldstone modes become a linear combination of spin and charge excitations and the poles appear in both spin *and* charge response functions. Since the Goldstone modes are gapless, this raises the issue of whether the system remains an insulator. In fact, if we define an insulator by the vanishing of the zero-temperature d.c. conductivity or equivalently of the Drude weight [3, 4], the Goldstone poles in the charge fluctuations do not change the fact that the system is insulating. Nevertheless, the dynamical conductivity is gapless and we conclude that the magnetically ordered state is an insulator but with no charged-particle–hole excitation gap. This illustrates very well the conclusion of Kohn [3] that the existence of an energy gap (in particle-hole excitations) is sufficient but not necessary to have an insulator. The non-collinearly ordered states studied in this letter are then unlike any other examples of Mott-Hubbard insulators where there is generally a gap in both single-particle and charged-particle–hole excitations. We first demonstrate that this mixed spin-charge character is due to the non-collinearity of the magnetic structure and is valid in an arbitrary number of dimensions (1). The case of the triangular lattice is then studied as a specific example.

To derive the spin-charge coupling, we start from the general one-band Hubbard model

$$H = -\sum_{\langle i,\,j\rangle,\,\sigma} t_{ij}(c^\dagger_{i\sigma}c_{j\sigma} + c^\dagger_{j\sigma}c_{i\sigma}) + U\sum_i n_{i\uparrow}\, n_{i\downarrow}\,, \tag{1}$$

where the first sum is over pairs of sites, t_{ij} is the hopping integral and U the on-site Coulomb interaction. We remark that since we are ultimately interested in the $q \to 0$ conductivity, it is crucial that we work with a conserving (gauge invariant) approximation. The simplest such approximation consists in describing the ordered state in the Hartree-Fock approximation and the fluctuations in the generalized random phase approximation (GRPA). The GRPA, in the ordered state, is an expansion in t/U and hence becomes more and more accurate as U becomes large, contrary to what happens in the paramagnetic state. Schrieffer *et al.* [6] have shown that this approach does indeed reproduce the spin wave results on the square lattice in the Heisenberg ($t \ll U$) limit. The main result of our paper, namely the mixed spin-charge character of the Goldstone modes, occurs in next-to-leading order in t/U when the ordered moments are *not* collinear and it remains valid in any dimension.

To proceed with the calculation, we first define $S^\mu(i) \equiv (1/2)\sum_{\alpha,\beta} c^\dagger_{i\alpha}\sigma^\mu_{\alpha\beta}c_{i\beta}$, a dimensionless four-component spin and charge operator. ($\mu = \rho, x, y, z$, and σ^ρ is the unit matrix.) We consider the general class of magnetically ordered states with planar spiral order

$$\langle S^z_i\rangle + i\langle S^x_i\rangle = S\exp[i\boldsymbol{Q}\cdot\boldsymbol{R}_i]\,, \tag{2}$$

where S is the order parameter representing the average moment on each site. Without loss of generality, we take the spiral in the (x, z)-plane. Examples of such states for two-dimensional lattices lying in the $(\hat{x}, \hat{y})$-plane include the collinear antiferromagnet on the square lattice, $\boldsymbol{Q} = \pi/a\,\hat{x} + \pi/a\,\hat{y}$, and the three-sublattice antiferromagnet on the triangular lattice, a 120° spiral with $\boldsymbol{Q} = 4\pi/3a\,\hat{x}$. As always when working with planar spiral order, the analysis is greatly simplified by using a rotating orthonormal basis in which the quantization axis at every site points in the same direction as the average spin density of eq. (2). In this rotating frame, the single-particle Green's function $G_{\alpha\beta}(i, j;\, \tau) \equiv -\langle Tc_{i\alpha}(\tau)c^\dagger_{j\beta}(0)\rangle$ becomes, in matrix notation, $\bar{G}(i, j;\, i\omega_n) = T^\dagger_i G(i, j;\, i\omega_n) T_j$, where the rotation matrix in spin space $T_i = \exp[-i(\boldsymbol{Q}\cdot\boldsymbol{R}_i)\sigma^y/2]$ depends on the site i. In this reference frame, the Green's function matrix $\bar{G}$ has the full underlying lattice periodicity, even for incommensurate spiral order. In the Hartree-Fock approximation, we find

$$\bar{G}(\boldsymbol{k}, i\omega_n) = \frac{A_+(\boldsymbol{k})/2}{i\omega_n + \mu - E_+(\boldsymbol{k})} + \frac{A_-(\boldsymbol{k})/2}{i\omega_n + \mu - E_-(\boldsymbol{k})}\,, \tag{3}$$

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(1) Coupling between the transverse spin excitations and charge excitations has been found in the *metallic* state of incommensurate spiral SDW states on the square lattice by John *et al.* [5].

where

$$A_{\pm}(\boldsymbol{k}) \equiv \begin{pmatrix} 1 \mp \dfrac{\Delta}{E(\boldsymbol{k})} & \pm i\,\dfrac{\eta(\boldsymbol{k})}{E(\boldsymbol{k})} \\ \mp i\,\dfrac{\eta(\boldsymbol{k})}{E(\boldsymbol{k})} & 1 \pm \dfrac{\Delta}{E(\boldsymbol{k})} \end{pmatrix},$$

with the definitions $\varepsilon_0(\boldsymbol{k}) \equiv -(1/N)\sum_{i,j} t_{ij} \cos \boldsymbol{k}\cdot(\boldsymbol{R}_i - \boldsymbol{R}_j)$ for the single-particle dispersion in the paramagnetic phase (N is the number of sites), and

$$\begin{cases} \varepsilon_{\pm} \equiv \varepsilon_0(\boldsymbol{k} \pm \boldsymbol{Q}/2)\,, & \eta(\boldsymbol{k}) \equiv \dfrac{\varepsilon_+(\boldsymbol{k}) - \varepsilon_-(\boldsymbol{k})}{2}\,, \\ \varepsilon(\boldsymbol{k}) \equiv \dfrac{\varepsilon_+(\boldsymbol{k}) + \varepsilon_-(\boldsymbol{k})}{2} + \dfrac{U}{2}\,, & \Delta \equiv US\,, \\ E(\boldsymbol{k}) \equiv \sqrt{\eta^2(\boldsymbol{k}) + \Delta^2}\,, & E_{\pm}(\boldsymbol{k}) \equiv \varepsilon(\boldsymbol{k}) \pm E(\boldsymbol{k})\,. \end{cases} \tag{4}$$

The gap equation is given by the self-consistency requirement

$$\frac{U}{2N}\sum_{\boldsymbol{k}} \frac{[f(E_-(\boldsymbol{k})) - f(E_+(\boldsymbol{k}))]}{E(\boldsymbol{k})} = 1\,, \tag{5}$$

where $f(x)$ is the Fermi function and the wave vector $\boldsymbol{k}$ spans the entire paramagnetic Brillouin zone of the crystal.

Proceeding to the collective excitations in the ordered state, we define the matrix response function

$$\chi^{\mu\nu}(i, j;\, \tau) \equiv -\langle TS^{\mu}(i;\,\tau)S^{\nu}(j;\,0)\rangle + \langle S^{\mu}(i)\rangle\langle S^{\nu}(j)\rangle\,.$$

χ is obtained in the GRPA by the usual summation of bubble and ladder diagrams. In the rotating frame where $\chi^{\mu\nu}(i, j;\, \tau) \to \tilde{\chi}^{\mu\nu}(i-j;\, \tau)$, the matrix GRPA equation takes the form ([2])

$$\tilde{\chi}(\boldsymbol{q}, i\Omega_n) = \tilde{\chi}^0(\boldsymbol{q}, i\Omega_n) + 2U\tilde{\chi}^0(\boldsymbol{q}, i\Omega_n)\Gamma\tilde{\chi}(\boldsymbol{q}, i\Omega_n)\,, \tag{6}$$

where Γ is a diagonal matrix with $\Gamma^{\rho\rho} \equiv 1$, $\Gamma^{xx} \equiv \Gamma^{yy} \equiv \Gamma^{zz} \equiv -1$. The density response in $\tilde{\chi}$ is related to the laboratory response χ by (see ([2])) $\chi^{\rho\rho}(\boldsymbol{q}, \boldsymbol{q}', \omega + i\delta) = \tilde{\chi}^{\rho\rho}(\boldsymbol{q}, \omega + i\delta)\,\delta_{\boldsymbol{q},\boldsymbol{q}'}$. The poles of $\tilde{\chi}$ coincide with those of χ, giving the position of the collective modes.

At $T = 0$ K, which we will consider from now on, the retarded zeroth-order matrix

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([2]) Details of the derivation will be given elsewhere.

susceptibility $\tilde{\chi}^0(\boldsymbol{q}, \omega + i\delta)$ is given by

$$\tilde{\chi}^0(\boldsymbol{q}, \omega + i\delta) = \frac{1}{8N} \cdot$$

$$\cdot \sum_{\boldsymbol{k}} \begin{bmatrix} \left(\frac{-\Delta^2 + EE' - \eta\eta'}{EE'}\right)\Lambda_- & i\Delta\left(\frac{\eta - \eta'}{EE'}\right)\Lambda_- & \left(\frac{-\eta}{E} + \frac{\eta'}{E'}\right)\Lambda_+ & \Delta\left(\frac{-1}{E} + \frac{1}{E'}\right)\Lambda_+ \\ -i\Delta\left(\frac{\eta - \eta'}{EE'}\right)\Lambda_- & \left(\frac{\Delta^2 + EE' - \eta\eta'}{EE'}\right)\Lambda_- & i\Delta\left(\frac{1}{E} + \frac{1}{E'}\right)\Lambda_+ & -i\left(\frac{\eta}{E} + \frac{\eta'}{E'}\right)\Lambda_+ \\ \left(\frac{-\eta}{E} + \frac{\eta'}{E'}\right)\Lambda_+ & -i\Delta\left(\frac{1}{E} + \frac{1}{E'}\right)\Lambda_+ & \left(\frac{\Delta^2 + EE' - \eta\eta'}{EE'}\right)\Lambda_- & -\Delta\left(\frac{\eta + \eta'}{EE'}\right)\Lambda_- \\ \Delta\left(\frac{-1}{E} + \frac{1}{E'}\right)\Lambda_+ & i\left(\frac{\eta}{E} + \frac{\eta'}{E'}\right)\Lambda_+ & -\Delta\left(\frac{\eta + \eta'}{EE'}\right)\Lambda_- & \left(\frac{-\Delta^2 + EE' + \eta\eta'}{EE'}\right)\Lambda_- \end{bmatrix}, \tag{7}$$

where unprimed functions are to be evaluated at $\boldsymbol{k} + \boldsymbol{q}/2$ and the primed functions at $\boldsymbol{k} - \boldsymbol{q}/2$, and where $\Lambda_\pm \equiv \gamma_- \pm \gamma_+$ with $\gamma_\pm = 1/(\omega + i\delta - (\varepsilon - \varepsilon') \pm (E + E'))$.

For hypercubic lattices in arbitrary dimension larger than one, symmetry arguments can be used to show that whatever the hopping matrix t_{ij} and the value of U, as long as there is long-range collinear antiferromagnetic order, the response matrix $\tilde{\chi}$ is block diagonal. There are then two Goldstone modes with purely transverse (x, y)-character. This is also the case for next-nearest-neighbor hopping on the square lattice where the resulting spin Hamiltonian is frustrated in the large-U limit.

From now on, we restrict ourselves to the nearest-neighbor triangular lattice at half-filling. At half-filling, as U is increased, the Hartree-Fock ground state evolves through a sequence of phase transitions to a SDW insulator with a three-sublattice 120° twist between spins on neighboring sites [2]. This spiral SDW state is the well-known ground state of the Heisenberg model with $J \sim 4t^2/U$ [7].

We go beyond previous studies by accounting for the collective excitations of the 120° spiral SDW phase. The electronic dispersion in the absence of interaction for the nearest-neighbor model on the triangular lattice is given by $\varepsilon_0(\boldsymbol{k}) = -2t(\cos(k_x a) + 2\cos(k_x a/2)\cos(\sqrt{3}k_y a/2))$. In the paramagnetic Brillouin zone, the single band $\varepsilon_0(\boldsymbol{k})$ is split into the two subbands $E_\pm(\boldsymbol{k})$ by the presence of spiral order, as can be seen from eqs. (3), (4). The direct single-particle energy gap is 2Δ but the indirect gap can be lower than this value. In the following, we restrict ourselves to the regime at half-filling where the indirect gap is positive and the 120° spiral SDW is the ground state. (Collective modes destabilize the spiral phase at $t/U \geqslant 0.146$ (see (2)).)

Expanding $\tilde{\chi}^0(\boldsymbol{q}, \omega)$ to second order in ω/U, t/U, and using the gap equation to eliminate S is favor of U, we find that the poles of $\tilde{\chi}(\boldsymbol{q}, \omega)$ correctly reproduce the spin wave result with $J = 4t^2/U$ [8] for the three Goldstone modes $\omega(\boldsymbol{q})$, $\omega(\boldsymbol{q} + \boldsymbol{Q})$, $\omega(\boldsymbol{q} - \boldsymbol{Q})$, of the triangular lattice. As the ratio t/U increases and the electrons become more itinerant, higher-order terms in t/U cannot be neglected and the coupling between charge and spin components becomes important. The matrix $\tilde{\chi}(\boldsymbol{q}, \omega)$ is no longer block diagonal and all response functions then share the same poles, although with different weights. As in the Heisenberg limit, there are still three Goldstone modes with a vanishing frequency at $\boldsymbol{q} = 0$ in the *magnetic* Brillouin zone. In fig. 1*a*) we plot the imaginary part of the response functions $\tilde{\chi}^{\rho\rho}$ and $\tilde{\chi}^{xx}$ for various values of $\boldsymbol{q}$. It is clear from the intensities in fig. 1*a*) that the coupling between spin and charge is very small. The intensity of this coupling increases with t/U.

Because the three Goldstone modes extend to *zero frequency* and now have charge as well

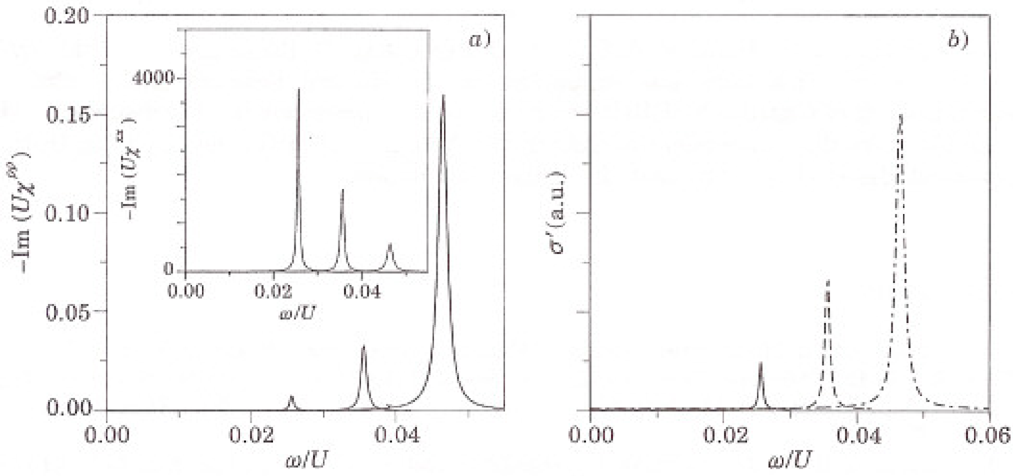

Fig. 1. – *a*) Imaginary part of the density and (inset) spin response functions at $t/U = 0.1$. From left to right the peaks correspond to $q_y = 0$, $q_x = 0.1$, 0.15, 0.25 (in units of $2\pi/a$). *b*) Real part of the conductivity at $t/U = 0.1$ for $q_y = 0$; $q_x = 0.1$, 0.15, 0.25 represented by the full, dashed and dot-dashed curves, respectively.

as spin character, it is natural to ask whether the system could become conducting despite the existence of a gap in the single-particle excitations. The appropriate definition of an insulator is that the coefficient (the Drude weight [3,4]) of the delta-function $\delta(\omega)$ in the zero-temperature d.c. conductivity $\sigma_{\text{d.c.}} = \lim_{\omega\to 0}\lim_{q\to 0}\sigma(\omega, \boldsymbol{q})$ vanishes. We obtained this quantity from the appropriate limit of

$$\sigma(\omega, \boldsymbol{q}) = ie^2\,\frac{(\omega + i\delta)}{q^2}\,\chi^{\rho\rho}(\omega + i\delta, \boldsymbol{q}, \boldsymbol{q})\,. \tag{8}$$

In this expression, one has to take the part of $\chi^{\rho\rho}(\omega + i\delta, \boldsymbol{q}, \boldsymbol{q}) = \widetilde{\chi}^{\rho\rho}(\omega + i\delta, \boldsymbol{q})$ which is irreducible with respect to the interaction. This accounts for the effect of screening. Expanding to second order in $\boldsymbol{q}$, ω, we found after lengthy algebra that the charge response function decreases faster than q^2 for $q \to 0$ so that the Goldstone-mode contribution to $\sigma(\omega, \boldsymbol{q})$ has effectively zero weight at $\boldsymbol{q} \to 0$. The same conclusion is reached by calculating the transverse current-current response function in the GRPA. It then follows that the system remains insulating even though there is no gap in the charge response function. This is not so surprising since at $\boldsymbol{q} = 0$ the Goldstone modes restore rotational invariance: hence they have the same parity as the ground state so that matrix elements of the current operator between the $\boldsymbol{q} = 0$ modes and the ground state vanish. At finite wave vector and frequency, however, the conductivity is finite and, as shown in fig. 1*b*), the absorption described by the real part is exactly at the Goldstone-mode position, as expected.

In conclusion, the triangular three-sublattice antiferromagnet described by the large-U half-filled one-hand Hubbard model provides an example of a gapless Mott insulator. In this system, the Goldstone modes acquire mixed spin and charge character at finite t/U, leading to the disappearance of the gap in the conductivity, despite a vanishing d.c. conductivity and the existence of a gap in the single-particle excitations. This phenomenon is generic for itinerant magnets with spiral order. An exception is when the ordering wave vector corresponds to collinear antiferromagnets.

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