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Charge Friedel oscillations in a Mott insulator

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When a metal undergoes a transition to an insulator it will lose its electronic Fermi surface. Interestingly, in some situations a “ghost” Fermi surface of electrically neutral spin carrying fermions may survive into the insulator. Such a novel ghost Fermi surface has been proposed to underlie the properties of a few different materials but its direct detection has proven elusive. In this paper, we show that the ghost Fermi surface leads to slowly decaying spatial oscillations of the electron density near impurities or other defects. These and related oscillations stem from the sharpness of the ghost Fermi surface and are direct analogs of the familiar Friedel oscillations in metals. The oscillation period contains geometric information about the shape of the ghost Fermi surface, which can be potentially exploited to detect its existence.

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Introduction. In recent years it has become clear that the insulating side in the vicinity of the Mott metal-insulator transition may, in some frustrated lattices, provide a realization of the long sought quantum spin-liquid (SL) state. These are insulators with an odd number of electrons per unit cell that do not order magnetically or break lattice symmetries. Candidate materials for SL phases of electronic Mott insulators are the layered quasi-two dimensional organic materials κ -(ET)₂Cu₂(CN)₃ and EtMe₃Sb[Pd(dmit)₂]₂,^{1,2} and the three-dimensional “hyperkagome” material Na₄Ir₃O₈.³ These materials all become metallic under moderate pressure^{4,5} and hence are appropriately thought of as weak Mott insulators. While SL behavior may well be a common feature of weak Mott insulators, it remains much more elusive in strong Mott insulators. Notable exceptions are the spin-1/2 Kagome magnets⁶ ZnCu₃(OH)₆Cl₂ and Cu₃V₂O₇(OH)₂ · 2H₂O.⁷

A fundamental question about the Mott transition is the fate of the metallic Fermi surface (FS) as it transitions into the Mott insulator. Theoretically the natural SL state in the vicinity of the Mott transition retains a “ghost” Fermi surface (FS) of mobile neutral spin carrying fermionic excitations (dubbed spinons) even as the charge of the electron gets pinned to the lattice to form an insulator. Such a state has been proposed to describe the intermediate temperature physics of many of the candidate SL materials.^{8–10} Experimentally, both organic salts and Na₄Ir₃O₈ have gapless spin excitations with low-temperature specific heat¹¹ and spin susceptibility^{1,2} resembling those of a metal even though they are electric insulators. Further recent experiments¹² in the insulator EtMe₃Sb[Pd(dmit)₂]₂ show metallic thermal transport, suggesting that the gapless excitations are mobile.

What are experimental signatures of such a ghost FS? The possibility of unusual quantum oscillations at low-*T* has been discussed,¹³ but it is likely that the spinon FS undergoes an instability at low-*T*, possibly related to spinon pairing.^{14–16} Another idea is to look for an oscillatory spinon mediated Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling between two ferromagnets separated by a spin liquid material.¹⁷ However, unlike the standard giant magnetoresistance effect, the coupling cannot be measured via the electrical resistance. This proposal also requires controlling the thickness of the SL layer to atomic precision.

In this paper, we identify surprising aspects of such a SL Mott insulator that may offer a simpler route to detecting the spinon FS. First, we show that the electrical charge density induced by an external potential has Friedel oscillations at the “ $2\mathbf{K}_F$ ” wave vectors that connect tangential antipodal portions of the spinon FS. Friedel oscillations in the charge density are well known properties of metals with a sharp electron Fermi surface. Remarkably these oscillations survive the transition into the SL Mott insulating state. The spinons that form the FS in this SL carry spin, and hence there are $2\mathbf{K}_F$ singularities in the spin response. Although spinons are electrically neutral, the $2\mathbf{K}_F$ singularities also show up in the charge response.

A closely related effect is the existence of a Kohn anomaly in the phonon spectrum arising from the coupling of the phonons to the spinon density. A similar signature will appear in scanning-tunneling microscopy (STM) spectra above the threshold bias for tunneling into the Mott insulator, which will show spatial modulation near impurities with wavelength set by the $2\mathbf{K}_F$ wave vectors of the spinon Fermi surface. These related effects are more directly measurable in experiments and can serve to detect the spinon FS.

We provide an estimate of the oscillation amplitude of the charge density and show that it is essentially unchanged as the material undergoes the metal-insulator transition. However, its magnitude is small compared to the free Fermi gas so that its direct measurement may be even more challenging than in a metal. In contrast, the Kohn anomaly is not weakened and survives even in the strong Mott insulator limit. Just as in a normal metal, it is thus easier to detect the Friedel oscillations through their effects on the phonon and STM spectra.

Spinon Fermi surfaces and $2\mathbf{K}_F$ singularities in a weak Mott insulator. A precise characterization of the ghost Fermi surface of spinons in an insulator is the presence of sharp singularities in the spin correlations at the $2\mathbf{K}_F$ wave vectors. In this paper, we explore the consequences of similar sharp singularities in the spinon density response. Specifically in the presence of a FS the spinon density response χ_f will have a singularity of the form

$$\chi_{f,\mathbf{Q}}^s(\mathbf{q}) \equiv \chi_f(\mathbf{Q} + \mathbf{q}) - \chi_f(\mathbf{Q}) = |q_{\parallel}|^{\phi} F\left(\frac{q_{\perp}^2}{|q_{\parallel}|}\right), \quad (1)$$

where \mathbf{Q} is any $2\mathbf{K}_F$ vector, $q_{\parallel} = \mathbf{q} \cdot \hat{\mathbf{Q}}$, $q_{\perp} = \mathbf{q} \times \hat{\mathbf{Q}}$, $F(x)$ is a universal nonsingular function, and the exponent ϕ is 1/2 for a Fermi liquid (FL) in two spatial dimensions but will be modified in a non-Fermi liquid as we briefly discuss below.

Consider a one band Hubbard model at half-filling on a nonbipartite lattice such as the triangular lattice:

$$H = -t \sum_{\langle rr' \rangle \alpha} (c_{r\alpha}^{\dagger} c_{r'\alpha} + \text{h.c.}) + U \sum_r (n_r - 1)^2, \quad (2)$$

where $c_{r\alpha}$ destroys an electron with spin $\alpha = \uparrow, \downarrow$ at site r , $n_r = c_r^{\dagger} c_r$ is the electron number at site r , and $U > 0$ is an on-site repulsion. As the ratio t/U decreases the system undergoes a Mott transition from a FL metal to a Mott insulator. We are interested in situations in which this Mott insulator is a nonmagnetic SL with gapless excitations. The Mott transition and the SL phase are conveniently discussed using the slave rotor representation of Ref. 18. We write $c_{r\alpha} = b_r f_{r\alpha}$ with $b_r \equiv e^{i\phi_r}$ a spin-0 charge- e boson, and $f_{r\alpha}$ a spin-1/2 charge-0 fermionic spinon. The electrical charge at a site n_r is identified with the boson number and is conjugate to the boson phase ϕ_r . The physical electron operator is manifestly invariant under local opposite phase rotations of b_r and $f_{r\alpha}$. Thus a proper reformulation of the Hubbard model will include an emergent $U(1)$ gauge field.

An effective theory⁹ for the phases of the Hubbard model is given by an imaginary time path integral with action:

$$S = \int d\tau \mathcal{L}_b(b, a_{\mu}) + \mathcal{L}_f(f, a_{\mu}), \quad (3)$$

$$\begin{aligned} \mathcal{L}_b = & \sum_r \frac{1}{2U} |(\partial_{\tau} + ia_o + \lambda)b_r|^2 \\ & - \sum_{\langle rr' \rangle} t_b (b_r^* b_{r'} e^{ia_{rr'}} + \text{c.c.}), \end{aligned} \quad (4)$$

$$\begin{aligned} \mathcal{L}_f = & \sum_r \bar{f}_r (\partial_{\tau} - (ia_o + \lambda)) f_r \\ & - \sum_{\langle rr' \rangle} t_f (\bar{f}_r f_{r'} e^{-ia_{rr'}} + \text{c.c.}). \end{aligned} \quad (5)$$

Here, a_0 and $a_{rr'}$ are the temporal and spatial components of the emergent $U(1)$ gauge field, $t_{b,f}$ are boson and spinon hopping amplitudes, and λ is adjusted to ensure that $\langle n_b - n_f \rangle = 0$.

In a mean field approximation, which ignores the gauge but not other interactions, the charge sector is mapped to a boson Hubbard model with hopping t_b and repulsion U while the spin sector is described by free fermionic spinons that form a FS. When t/U is large, the boson hopping t_b dominates, leading to a state with $\langle b_r \rangle \neq 0$. In terms of the original electrons, this is a Fermi liquid phase. As t/U is decreased, the bosons form a Mott insulating state with a charge gap. This corresponds to a SL electronic Mott insulating state, which retains a FS of gapless neutral spin-1/2 spinons. To go beyond mean field, in the Mott insulating phase the bosons may be integrated out, resulting in an effective low-energy theory of a spinon FS coupled to a $U(1)$ gauge field. Based on this theory, an estimate for the $2\mathbf{K}_F$ exponent ϕ was given in an early work.¹⁹ Controlled calculations²⁰ of $2\mathbf{K}_F$ singularities in the spinon number $n_{f,r} = f_r^{\dagger} f_r$ have been performed for

the two-dimensional system and found $\phi \approx 0.4$. Exactly the same exponent ϕ and scaling function F also describe the $2\mathbf{K}_F$ singularities in the spin density $\vec{S}_r = \frac{f_r^{\dagger} \vec{\sigma} f_r}{2}$ correlations due to an emergent symmetry of the spinon FS state.²⁰ In three dimensions, the gauge field plays a more innocuous role and the exponent is the same as in the three dimensional Fermi liquid.

Charge density correlations. The spinon number $n_{f,r}$ is a spin-singlet operator that transforms under lattice and time reversal symmetries in the same way as the electrical charge $n_r = c_r^{\dagger} c_r$, thus we might expect a linear coupling between $n_{f,r}$ and n_r . For long wavelength, external perturbations that couple to the charge density, the boson system is gapped and the response is that of an incompressible insulator. However, for short wavelength perturbations, the boson density has a nonzero response, which, in turn, induces a response of the spinon density. At a $2\mathbf{K}_F$ wave vector of the spinon FS, there are long range oscillations of the fermion density that couple back to the physical charge density, which will therefore show Friedel oscillations.

An approximate self-consistent calculation of the magnitude of this effect may be done within the slave-particle mean-field theory²³. An external potential with Fourier components $V(\mathbf{p})$ induces some change $\delta\lambda$ in the mean-field parameter λ . In general, the hopping parameters t_b and t_f will also change, for a rough estimate we ignore these below. The resulting change in the boson and fermion densities in linear response theory is

$$\delta\langle n \rangle(\mathbf{p}) = \chi_b(\mathbf{p})[V(\mathbf{p}) + \delta\lambda(\mathbf{p})], \quad (6)$$

$$\delta\langle n_f \rangle(\mathbf{p}) = -\chi_f(\mathbf{p})\delta\lambda(\mathbf{p}). \quad (7)$$

Here, $\chi_{b,f}$ are the density response functions of the boson and fermion, respectively. The condition $\delta\langle n \rangle(\mathbf{p}) = \delta\langle n_f \rangle(\mathbf{p})$ determines $\delta\lambda$. The charge density response function defined through $\delta\langle n \rangle(\mathbf{p}) = \chi(\mathbf{p})V(\mathbf{p})$ is then

$$\chi = \frac{\chi_b \chi_f}{\chi_b + \chi_f}. \quad (8)$$

This is the well-known Ioffe-Larkin composition rule, which we now use to evaluate the $2\mathbf{K}_F$ response.

Near a $2\mathbf{K}_F$ wave vector we write the fermion response $\chi_f(2\mathbf{K}_F + \mathbf{q}) = \chi_{f,2\mathbf{K}_F}^0 + \chi_{f,2\mathbf{K}_F}^s(\mathbf{q})$, where $\chi_{f,2\mathbf{K}_F}^0$ is the smooth background response at $2\mathbf{K}_F$ and $\chi_{f,2\mathbf{K}_F}^s(\mathbf{q})$ is the singular part. Denoting the (nonsingular) boson response $\chi_b(2\mathbf{K}_F + \mathbf{q}) = \chi_{b,2\mathbf{K}_F}^0$, we see that the singular part $\chi_{f,2\mathbf{K}_F}^s$ of the $2\mathbf{K}_F$ electrical density response is reduced from that of the spinon density $\chi_{f,2\mathbf{K}_F}^s$ by a factor

$$R = \left(1 + \frac{\chi_{f,2\mathbf{K}_F}^0}{\chi_{b,2\mathbf{K}_F}^0} \right)^{-2}. \quad (9)$$

As t/U is varied across the Mott transition, both $\chi_{b,2\mathbf{K}_F}^0$ and $\chi_{f,2\mathbf{K}_F}^0$ will be essentially unchanged so that the factor R , and hence the magnitude of the charge Friedel oscillations is more or less the same on either side of the transition. The factor R may be estimated within the slave-rotor mean-field theory combined with a large- N approximation to the quantum rotor model that describes the bosons.²³ Near the Mott transition, we find $R = 0.0024$. Though the magnitude of the charge Friedel oscillations is very small it is interesting conceptually that it is

nonzero. Deep in the Mott insulator, R decreases further and vanishes in the extreme limit where $\chi_b(q) = 0$ for all q .

Actually, as discussed above, the detailed nature of the $2\mathbf{K}_F$ singularity for the spinons is modified and even potentially enhanced²⁰ in the Mott insulator as compared to the Fermi liquid due to the gapless $U(1)$ gauge field. Thus we reach the remarkable conclusion that there are charge Friedel oscillations in the insulator. In fact, they may even be more slowly decaying than in the metal.

Detection of spinon density Friedel oscillations. The presence of the $2\mathbf{K}_F$ singularities in the charge density response relies on the $2\mathbf{K}_F$ singularities in the spinon density response. We now argue that this latter singularity directly affects the phonon and STM spectra which could be more useful to detect the spinon FS.

In the SL, the charge of the electron gets pinned to the ions that make up the underlying lattice, while the spin continues to be mobile and forms a FS. Thus the oscillating entity in a phonon mode is the electrically neutral ion-boson composite (see Figs. 1 and 2). While electrically neutral, this object carries gauge charge of the boson b_r , and hence there will be long range interactions between different ion-boson composites. To properly describe the phonon spectrum, we must take into account the screening of the gauge interaction by the spinon fluid. Specifically, there will be an emergent ‘‘Coulomb’’ interaction between the ion-boson composites that will be screened by the mobile spinons. As in the usual discussions of the Kohn anomaly the ability of the spinon fluid to screen the gauge interaction is sharply changed as the phonon wave vector passes through any $2\mathbf{K}_F$ wave vector. Consequently, the Kohn anomaly survives deep in the Mott insulator so long as the system retains a spinon FS. It is not weakened by the suppression of electric charge fluctuations, and remains similar to a weakly interacting metal.

Alternatively, strong Mott insulators are well described by simple spin-only models with dominant nearest neighbor exchange. Here, the $2\mathbf{K}_F$ singularities in the spinon density should be interpreted as slow power correlations of incommensurate valence-bond solid order. Such correlations have

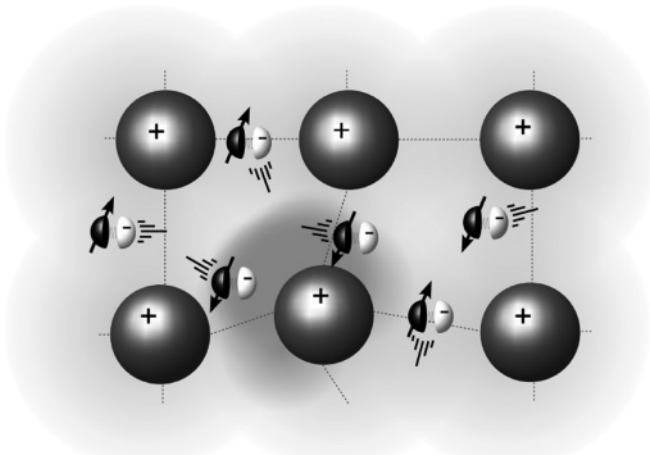


FIG. 1. Screening in a Fermi liquid. In a regular metal, charge (represented by white half-spheres) and spin (black half-spheres with arrows) of the electrons are confined, and the electrons move to screen any displaced positively charge ions (big spheres).

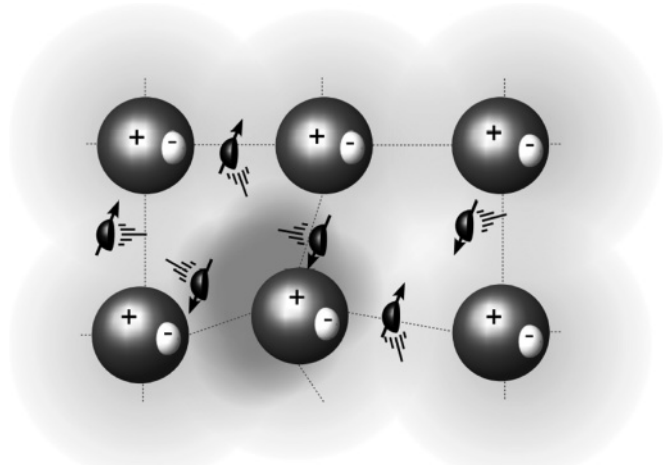


FIG. 2. Screening in a spin liquid. In the spin liquid Mott insulator, the charge of the electron is bound to the ions while the spins stay mobile. The electrically neutral ion-chargeon composite carry internal gauge charge and their displacement is screened by the (oppositely) gauge-charged spinon fluid.

been found in a spin-1/2 system on triangular two-leg²¹ and four-leg²² ladders that realize quasi-1-dimensional versions of the spinon FS state. The above argument shows that phonons couple to these incommensurate valence-bond fluctuations, and consequently, there is a Kohn anomaly in the phonon spectrum.

In metals, Friedel-like oscillations have been imaged through STM. The most common strategy is to measure the spatial modulation of the tunneling spectrum at fixed bias. In the Mott insulator, we are faced with the immediate problem that (at $T = 0$) there is a gap in the single electron spectrum. However, if the bias exceeds the gap it is possible to tunnel into the sample. The tunneling spectrum near the threshold is readily calculated within the slave-rotor mean-field theory. We find $N(\omega) = 0$ for $\omega < \Delta$ ²³ and

$$N(\omega > \Delta) \sim (\omega - \Delta)N_f(0). \quad (10)$$

In the presence of a defect such as a step edge or an impurity, the local spinon density of states $N_f(\omega \rightarrow 0; \mathbf{x})$ oscillates as a function of spatial location \mathbf{x} at the $2\mathbf{K}_F$ wave vectors of the spinon FS. Thus the electron tunneling spectrum at a bias voltage close to the gap acquires spatial modulations at the $2\mathbf{K}_F$ wave vectors of the spinon Fermi surface.

We emphasize that the suppression factor (9) for the oscillations in the charge density response plays no role in the Kohn anomaly or the STM oscillations. For the Kohn anomaly, our discussion of the strong Mott insulator, where charge oscillations are completely absent, shows that it is not

TABLE I. Smallest wave vectors connecting antipodal tangential portions of the Fermi surface in the spin-liquid candidates.

$\kappa - (\text{ET})_2\text{Cu}_2(\text{CN})_3$	$\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$	$\text{Na}_4\text{Ir}_3\text{O}_8$
Triangular	Kagome	Hyperkagome
$\frac{0.58\pi}{a} \approx 0.22 \text{ \AA}^{-1}$	$\frac{0.58\pi}{a} \approx 0.265 \text{ \AA}^{-1}$	hole: $\frac{0.52\pi}{a} \approx 0.18 \text{ \AA}^{-1}$ el.: $\frac{0.62\pi}{a} \approx 0.21 \text{ \AA}^{-1}$

weakened. For the STM spectrum, we note that the ratio of the oscillatory signal, N_{osc} , to the background, N_{bg} , is given by,

$$\frac{N_{\text{osc}}(\omega \gtrsim \Delta)}{N_{\text{bg}}(\omega \gtrsim \Delta)} = \frac{N_{\text{osc}}}{N_{\text{bg}}|_{\text{free fermions}}} \quad (11)$$

as is clear from Eq. (10) (see also Ref. 23).

The effects we describe will only be visible at temperature much lower than the Fermi temperature (estimated to be ≈ 300 K for the organic spin liquids). The spinon Fermi surface may be unstable at very low- T (≈ 5 K in organics) so that experiments are best performed at low- T just above this instability. In both the organic spin liquids [κ -(ET)₂Cu₂(CN)₃ and EtMe₃Sb[Pd(dmit)₂]₂] and in Na₄Ir₃O₈, determination of the phonon spectrum through neutron scattering is challenging but may possibly be done through inelastic x-ray scattering techniques. The smallest $2\mathbf{K}_F$ wave vectors for the Kohn anomaly are given in Table I. The hyper-kagome material Na₄Ir₃O₈ has a rather complicated band structure in its metallic state, which will be inherited by the spinons in the proposed SL Mott insulator. Some representative $2\mathbf{K}_F$ wave vectors for this material¹⁷ are listed for completeness in Table I. Previous

STM studies²⁴ of the κ -ET organics have focused on a superconducting material. STM studies of the insulating SL will be useful in both determining the single particle gap and detecting the possible spinon FS. In this context, we note that, in κ -(ET)₂Cu₂(CN)₃, the charge gap as measured by optical conductivity and dc transport is small (in the 15–100 meV range) though it is hard to extract a precise number. This range should be readily accessible in STM experiments.

Finally we briefly turn to strong Mott insulators with negligible virtual charge fluctuations but which nevertheless are in SL phases. This is the situation, for instance, in the Kagome materials ZnCu₃(OH)₆Cl₂ and Cu₃V₂O₇(OH)₂ · 2H₂O. Indeed, a SL with a spinon FS has been proposed for ZnCu₃(OH)₆Cl₂ in Ref. 25. From the arguments presented above, it follows that there will again be a Kohn anomaly at the $2\mathbf{K}_F$ wave vectors. Detecting such an anomaly will then be a nontrivial proof of the proposal that a spinon FS exists in this material.

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