Power-Law Conductivity inside the Mott Gap: Application to κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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The charge dynamics of spin-liquid states described by $U(1)$ gauge theory coupling to fermionic spinons is discussed in this paper. We find that the gapless spinons give rise to a power-law optical conductivity inside the charge gap. The theory is applied to explain the unusual optical conductivity observed recently in the organic compound κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. We also propose an optical experiment to search for the in-gap excitations in the kagome spin-liquid insulator.

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Recent work has shown that the organic compound κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [1–3] and the spin-1/2 kagome system ZnCu$_3$(OH)$_6$Cl$_2$ [4–6] hold great promise as the first two examples of spin-liquid states realized in dimensions greater than one [7–9]. Spin-liquid states are Mott insulators with an odd number of $S = \frac{1}{2}$ spin per unit cell, which shows no long-range magnetic order. They are proposed to exist in systems either in the vicinity of the Mott transition [7] or with frustrated lattice structures. In both cases the system can be modeled by an appropriate Hubbard model with on-site repulsion U and hopping integral t at half filling. For large enough U compared with t, charge excitations are gapped and the system is a spin liquid if long-range order is absent in the spin sector.

The two recently discovered systems are believed to be two-dimensional spin liquids. In the case of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ the system is described by a Hubbard model on a triangular lattice. Since the system can be driven metallic (indeed superconducting) under pressure, it is believed that U is not very large compared with t and the insulator is near the Mott transition [7,8]. In this case charge excitations acquire a gap and it is proposed that spin-charge separation occurs and spin $\frac{1}{2}$ excitations (spinons) form a Fermi surface [7,8]. In the case of ZnCu$_3$(OH)$_6$Cl$_2$, it is believed that $U \gg t$ and the spin dynamics are described by the antiferromagnetic Heisenberg model. The frustrated kagome lattice gives rise to a spin-liquid state with Dirac fermions excitation spectrum [9]. A common feature of these spin-liquid states is that the spin excitations are always coupled to internal $U(1)$ gauge fields representing spin-chirality fluctuations [7–9] in the spin systems.

It is often thought the Mott insulators are fully gapped in their optical (charge) responses. Furthermore, the spinons are neutral and do not absorb electromagnetic radiation. Here we point out that due to coupling with the internal gauge field, the spinons do contribute to optical conductivity, yielding a power-law absorption at low frequencies. This may explain some puzzling experimental observations recently reported in the organics [10].

The dynamics of the spin-liquid states can be studied in a slave-rotor representation of Hubbard models [7] with appropriate lattice structures. In this representation the electron operator is represented as $c(c^+)_{i\sigma} = f(f^+)_{i\sigma}e^{-(+)i\theta}$, where $f(f^+)_{i\sigma}$ is the spin annihilation (creation) operator and $e^{-(+)i\theta}$ lowers (raises) charge by one. After making a mean-field approximation, the low energy effective action of the system can be written in terms of θ and $f(f^+)$ fields separately, $L^{1(2)} = L_c + L_s^{1(2)}$, where L_c represents the charge dynamics and L_s represents the spin dynamics of the system. L_c is described by the strong coupling phase of a quantum $x - y$ model [11],

$$L_c \sim \sum_i \frac{1}{U}|[\partial_t - i(a_0 + A_0)]\theta_i|^2 - t_{\text{eff}}\sum_{\langle i,j\rangle}\cos[\theta_i - \theta_j - (\vec{a}_{ij} + A_{ij})], \tag{1a}$$

coupling to internal gauge fields $(a_0, \vec{a})$, where $t_{\text{eff}} \sim \alpha t$ with $\alpha < 1$ being a numerical factor determined self-consistently from the mean-field equation, $(A_0, \vec{A})$ represents the real electromagnetic field coupling to the system, and

$$L_s^{(1)} = \sum_\sigma \left(f_\sigma^+(\partial_t - ia_0 - \mu_f)f_\sigma - \frac{1}{2m_s}f_\sigma^+(-i\nabla - \vec{a})^2 f_\sigma \right) \tag{1b}$$

in the case of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, which is believed to possess a spinon Fermi surface. μ_f is the chemical potential; m_s^{-1} is expected to be of order of the exchange $J \sim t^2/U$. In the case of ZnCu$_3$(OH)$_6$Cl$_2$ where spinons have a Dirac fermion spectrum,

$$L_s^{(2)} = \sum_{\mu\sigma} \bar{\psi}_{+\sigma}[\partial_\mu - i(a_\mu + A_\mu)]\tau_\mu \psi_{+\sigma} + \bar{\psi}_{-\sigma}[\partial_\mu - (a_\mu + A_\mu)]\tau_\mu \psi_{-\sigma}, \tag{1c}$$

where $\mu = 0, 1, 2$, and τ_μ are Pauli matrices. The two-component Dirac spinor fields $\psi_{\pm\sigma}$ describe two inequivalent Dirac nodes in the spinon spectrum [9].

Effects of disorder and phonons can also be included in the actions. Their contributions can be included by adding a term

$$L' = \sum_{\vec{p},\vec{q}} (V(q) c^{+}_{\vec{p}+\vec{q}\sigma} c_{\vec{p}\sigma} + M(q) c^{+}_{\vec{p}+\vec{q}\sigma} c_{\vec{p}\sigma} (b_{\vec{q}} + b^{+}_{-\vec{q}}) + b^{+}_{\vec{q}} (\partial_0 - \omega_{\vec{q}}) b_{\vec{q}})$$

to $L_s^{(1)}$, where $V(q)$ is a disordered potential and $b(b^+)_{\vec{q}}$ are phonon annihilation (creation) operators with momentum $\vec{q}$. $M(q)$ is the electron-phonon coupling and $\omega_{\vec{q}}$ is the phonon dispersion. A corresponding term can also be added to $L_s^{(2)}$ for Dirac fermions.

The thermodynamic and magnetic properties of the above systems have been studied in several previous papers [7–9,12]. We shall concentrate on the charge dynamics of these spin liquids here. We assume a Mott insulator state with no broken symmetry and with isotropy in space. The current response function is given by the conductivity, which can be decomposed into longitudinal and transverse parts $\sigma_{\parallel}$ and $\sigma_{\perp}$. For a $U(1)$ spin liquid, the Ioffe-Larkin composition rule [13] relates the physical σ to the response function of the spin and charge components:

$$\sigma_{\perp}(q, \omega) = [\sigma^{-1}_{s\perp}(q, \omega) + \sigma^{-1}_{c\perp}(q, \omega)]^{-1} \tag{2}$$

and similarly for $\sigma_{\parallel}$. Here σ_s and σ_c are the proper response functions of the spin and charge (θ) fields appearing in the action L_s and L_c, respectively. The proper response functions represent sum of all diagrams which cannot be separated into two parts by cutting one interaction line associated with either the real or internal gauge field and represent the current response of the charges and spinons to the potential $\vec{a} + \vec{A}$ and $\vec{a}$, [13,14] respectively. Notice that both the phonon and impurity contributions can be included in the definition of the proper response functions. The origin of the Ioffe-Larkin rule is that an external $\vec{A}$ field induces a nonzero $\vec{a}$ field, which is needed to enforce the constraint $j_{c\mu} + j_{s\mu} = 0$ [14]. Thus even though the $\vec{A}$ field couples only to the θ field, the induced $\vec{a}$ field indirectly couples the gauge field to the gapless spinons.

We parametrize the longitudinal response of the charge field by a dielectric constant ε_c and ignore the analytic correction in q^2, ω^2 for small q and ω. Then

$$\varepsilon_c = 1 + \frac{4\pi i \sigma_{c\parallel}}{\omega}. \tag{3}$$

We expect $\varepsilon_c - 1$ to decrease with increasing charge gap. Furthermore, for small q there is no distinction between longitudinal and transverse response in an insulator. Using (3) for both, we find using Eq. (2)

$$\sigma_{\parallel(\perp)}(q, \omega) = \frac{\omega \sigma_{s,\parallel(\perp)}(q, \omega)}{\omega + i(\frac{4\pi}{\varepsilon_c - 1})\sigma_{s,\parallel(\perp)}(q, \omega)}. \tag{4}$$

We should point out that the replacement of the charge response by a dielectric constant is not as innocent as it appears. This step should be considered in the spirit of random phase approximation and justified using a $\frac{1}{N}$ expansion. The concern is the existence of Feynman diagrams involving multiple gauge field lines going across. In the language of proper response function, these become part of the charge vertex which couples to the external gauge field. Since the gauge field is gapless, the approximation of this vertex by a dielectric constant is not strictly correct except as leading order in $\frac{1}{N}$ [15].

Now we consider the optical conductivity given by $\sigma_{\perp}(q = 0, \omega)$. In this limit there is no distinction between longitudinal and transverse, and we can drop the $\perp$ subscript. The spinon conductivity is expected to be metallic-like. We can safely assume $\mathrm{Re}[\sigma_s(0, \omega)] \gg \omega$ and $\mathrm{Im}[\sigma_s] \ll \mathrm{Re}[\sigma_s]$ for small ω and obtain from Eq. (4)

$$\mathrm{Re}[\sigma(\omega)] = \omega^2 \left(\frac{\varepsilon_c - 1}{4\pi}\right)^2 \frac{1}{\mathrm{Re}[\sigma_s(\omega)]}. \tag{5}$$

Note that $\mathrm{Re}\sigma(\omega) = 0$ for $\omega = 0$ as expected for an insulator, but we find contribution inside the gap for small ω. First we consider the case when disorder scattering of the spin is weak. Then $\sigma_s(\omega) = ne^2\tau(\omega, T)/m_s$. The dominant contribution to τ^{-1} is inelastic scattering due to the gauge field [14], which is given by $\frac{1}{\tau} \sim [\max(\hbar\omega, k_B T)]^{4/3}$. The exponent $4/3$ comes from the scattering of the fermions from gauge fluctuations, whose frequency scale as $q^{1/3}$. These soft fluctuations give rise to a non-Fermi-liquid self-energy which scales as $\omega^{2/3}$. However, for transport scattering rate, two extra powers of q are needed to account for momentum relaxation, leading to the $4/3$ power law. For $\hbar\omega > \hbar/\tau_0$, $k_B T$, where τ_0 is the elastic scattering time, we find

$$\mathrm{Re}[\sigma(\omega)] = \omega^{3.33} \left(\frac{\varepsilon_c - 1}{4\pi}\right)^2 \frac{m_s}{n}, \tag{6}$$

in qualitative agreement with what is observed experimentally in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [10]. Our theory also predicts that $\mathrm{Re}[\sigma(\omega)]$ cross over to $\sim\omega^2$ for $\hbar\omega < k_B T$. The above results are strongly modified if localization effect is important and $\sigma(\omega)$ vanishes at $\omega \to 0$ faster than ω. In this case

$$\sigma(\omega) \sim \sigma_s(\omega)$$

will show similar behavior as observed in usual strongly disordered metals.

The above analysis can be generalized straightforwardly to the kagome system $ZnCu_3(OH)_6Cl_2$ with the "proper" response functions replaced by the corresponding functions for Dirac fermions. In this case $\sigma_s(q, \omega)$ has the universal form

$$\sigma_s(q, \omega) \sim \frac{e^2}{8} \frac{(\bar{c}^2 q^2 - \omega^2)^{(1+\beta)/2}}{i\omega}, \tag{7}$$

where $\beta = 0$ for noninteracting Dirac fermions and is nonzero in the presence of gauge field interaction. The value of β can be estimated perturbatively in $1/N$ expansion [15], but its exact value is unknown. Putting $\sigma_s(0, \omega)$ into Eq. (4), we predict $\mathrm{Re}[\sigma(\omega)] \propto \omega^{2-\beta}$ for $\beta < 1$ and $\mathrm{Re}[\sigma(\omega)] \propto \omega^{\beta}$ for $\beta > 1$, and the optical conductivity probes directly the unknown exponent β. Since the kagome system is deep in the Mott insulator regime, the observation of power-law conductivity inside the Mott gap stronger than ω^4 [see Eq. (14c) below] will be strong evidence for the existence of gapless spinons and the importance of gauge fields.

We shall now study in more detail the general dielectric response $\varepsilon(q, \omega)$ of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, which is believed to possess a spinon Fermi surface. We shall assume that the residual interactions are weak enough so that the spinons are in a Fermi liquid state.

The dielectric function of the spin liquid is given by

$$\varepsilon(q, \omega) = 1 - v_e(q)\chi_d(q, \omega),$$

where $\chi_d(q, \omega)$ is the proper density-density response function of the system that represents the sum of all polarization diagrams, which cannot be separated into two parts by cutting one Coulomb interaction line associated with the real electromagnetic field [13,15]. $v_e(q) = 4\pi e^2/q^2$ is the real Coulomb interaction. We assume here that the (3D) system is a sum of layers of spin liquid.

Charge conservation gives $\chi_d = (q^2/\omega^2)\chi_\parallel$, where $\chi_\parallel$ is the longitudinal current-current response function that is in turn given by $\sigma_\parallel = e^2\chi_\parallel/i\omega$. Combining these relations we obtain the usual formula

$$\varepsilon(q, \omega) = 1 + 4\pi i\sigma_\parallel(q, \omega)/\omega, \tag{8}$$

where $\sigma_\parallel$ is related to $\sigma_{s\parallel}$ by Eq. (4). In the absence of scattering, we expect the spinon density-density response function to be

$$\chi_{\mathrm{ds}} = \frac{dn}{d\mu} + \frac{i\gamma\omega}{v_F q}, \tag{9}$$

where γ is the quasiparticle density of states at the Fermi level and v_F is the Fermi velocity. Equation (9) is valid in Fermi liquid theory and has been shown to remain applicable for small q, ω when gauge fluctuations are treated to two loop order [15]. Particle conservation again allows us to write $\sigma_{s\parallel} = i\omega\chi_{\mathrm{ds}}/q^2$. Combining these results, we find

$$\begin{aligned}\sigma_\parallel(q, \omega) &= \left(\frac{\varepsilon_c - 1}{4\pi}\right)\frac{\omega}{i}\left[1 - \frac{i\omega}{\sigma_{s\parallel}}\left(\frac{\varepsilon_c - 1}{4\pi}\right)\right]^{-1} \\ &= \left(\frac{\varepsilon_c - 1}{4\pi}\right)\frac{\omega}{i}\left[1 - \frac{q^2}{\chi_{\mathrm{ds}}}\left(\frac{\varepsilon_c - 1}{4\pi}\right)\right]^{-1}.\end{aligned} \tag{10}$$

Using Eq. (8), we obtain at small q

$$\varepsilon(q, \omega) = \varepsilon_c + \frac{[(\varepsilon_c - 1)^2/4\pi]q^2}{\frac{dn}{d\mu} + \frac{i\gamma\omega}{v_F q}}. \tag{11}$$

The static dielectric constant is given by the charge part ε_c, and the full dielectric function is in principle measurable by electron diffraction.

Phonons have small effects on the above results. They only modify the interaction parameter γ and renormalize the compressibility $\partial n/\partial\mu$. The effect of disorder can be included by modifying $\chi_{\mathrm{ds}}(q, \omega)$ into a diffusive form $\frac{dn}{d\mu}\frac{Dq^2}{Dq^2 + i\omega}$ if localization effect is not important [16]. In this case, we obtain

$$\varepsilon(q, \omega) = \varepsilon_c + \frac{(\varepsilon_c - 1)^2(Dq^2 + i\omega)}{4\pi\sigma_{s,\parallel}}, \tag{12}$$

where D is the spinon diffusion constant and $\sigma_{s,\parallel} = e^2\frac{dn}{d\mu}D$. For $q = 0$, Eq. (12) is consistent with the ac conductivity given by Eq. (5) as expected.

It should be emphasized that the coupling of density and current responses to spin excitations exists rather generally in insulators and does not rely on existence of a spin-liquid state. Assuming that the electronic properties of the insulator are described by a Lagrangian with a one-particle term and an effective electron-spin coupling of form $L' = \vec{S}\cdot(\psi^+\vec{\tau}\psi)$, where $\psi = (c_\uparrow, c_\downarrow)$ is a two-component spinor, $c_{\sigma s}$ are electron operators, and $\vec{S}$ is an effective spin operator, the leading order coupling terms between spins and density/current fluctuations can be derived and are represented in the Feynman diagram shown in Fig. 1(a), where the solid lines are electron propagators. In real space-time, the diagrams are represented by expressions of form

$$\begin{aligned}\Gamma_\mu(x, x'_-, x'_+; \vec{S}) &= \sum_{\sigma\sigma'\nu\nu'} G_\sigma(x - x'_-)\hat{j}_\mu(x)G_\sigma(x'_+ - x)S^\nu(x'_-)\tau^\nu_{\sigma\sigma'}G_{\sigma'}(\delta x)S^{\nu'}(x'_+)\tau^{\nu'}_{\sigma'\sigma} \\ &= \sum_\sigma G_\sigma(x - x'_-)\hat{j}_\mu(x)G_\sigma(x'_+ - x)\{G_{-\sigma}(\delta x)[S^x(x'_-)S^x(x'_+) + S^y(x'_-)S^y(x'_+)] \\ &\quad + G_\sigma(\delta x)S^z(x'_-)S^z(x'_+) - i(\sigma)G_{-\sigma}(\delta x)[S^x(x'_-)S^y(x'_+) - S^y(x'_-)S^x(x'_+)]\},\end{aligned} \tag{13}$$

where $\hat{j}_\mu(\mu = 0, 1, 2)$ is the electron current operator and $x = (\vec{x}, t)$, $x'_{-(+)} = x' - (+)\delta x/2$. Assuming that the electrons have a gapped spectrum (insulator), the corresponding Green's function $G_\sigma(x)$ is short-ranged and the contributions mainly come from small δx region. Therefore we can expand $G_\sigma(x' \pm \delta x/2 - x) \sim G_\sigma(x' - x) \pm (\delta x/2)\partial_x G_\sigma(x' - x) + \ldots$, $S^\nu(x' \pm \delta x/2) \sim S^\nu(x') \pm (\delta x/2).\partial_{x'}S^\nu + \ldots$, etc., in Eq. (13) to derive the leading order spin-density (current) coupling terms in the insulating state in the continuum limit. A corresponding expansion for metallic ferromagnetic states has been reported previously [17]. By keeping two sites per unit cell, this procedure can be extended to derive the correction to

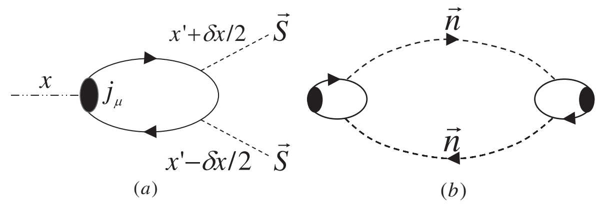

FIG. 1. (a) Leading order Feynman diagram representing coupling between spin and density or current fluctuations. Solid lines represent electron Green's functions. There is another diagram where electron lines reverse in direction. (b) Corresponding Feynman diagram representing correction to proper density-density response function.

optical conductivity in the antiferromagnetically ordered state in the Hubbard model, which is a competing state to the spin-liquid state observed in the organic compound κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [10]. In this case, $G(x) \rightarrow G^{ab}(x)$ and $\vec{S}(x) \rightarrow \vec{S}^a(x) = \vec{m}(\vec{x}) + (-1)^a \vec{n}(\vec{x})$, where a, $b = A$, and B are sublattice indices. $\vec{m}$ and $\vec{n}$ represent magnetization and staggered magnetization fluctuations, respectively. The low energy contribution to optical conductivity is dominated by coupling of density fluctuations to two spin wave processes represented by coupling to $\vec{n}$ fields. After some algebra, we obtain in the small wavevector limit

$$\Gamma_0(q, \omega; q', \Omega; q - q', \omega - \Omega; \vec{S}) \sim \omega(\vec{q}.\vec{q}')\vec{n}(\vec{q}', \Omega)\vec{n}(\vec{q} - \vec{q}', \omega - \Omega). \quad (14a)$$

We have assumed that the antiferromagnetic state is described by usual mean-field theory with staggered magnetization $\langle n \rangle \neq 0$. The corresponding correction to proper density-density response function [Fig. 1(b)] is

$$\delta\chi_d(0, \omega) \sim \frac{1}{V\beta}\sum_{q'\Omega}\frac{|\Gamma_0(0, \omega; q', \Omega; -q', \omega - \Omega; \vec{S})|^2}{(\Omega^2 - c_m^2 q'^2)[(\omega - \Omega)^2 - c_m^2 q']^2}, \quad (14b)$$

where $c_m \sim U\langle n \rangle$ is the spin wave velocity derived from the mean-field theory. Evaluating the integral, we find that the correction to optical conductivity is

$$\delta\sigma(\omega) \sim e^2\left(\frac{\omega}{c_m}\right)^{d+2}, \quad (14c)$$

for $\omega \ll U\langle n \rangle$, where d is the dimension. We note that the coupling vertex in Fig. 1(a) involves 3 fermion lines. The virtual excitation of these electrons implies a factor of $(t/U)^3$ if we are deep in the insulator where $U \gg t$. Thus the contribution to $\sigma(\omega)$ has a small factor of $(t/U)^6$. This is probably why the contributions from spin waves are usually ignored in the discussions of optical conductivity of Mott insulators. We note that this suppression factor does not appear for the spin liquid. Here we consider systems in proximity to the Mott transition, and the $(t/U)^6$ factor does not appear in Eq. (14c). Even so, we note that the optical conductivity is enhanced in the spin liquid compared with the antiferromagnetically ordered state, in agreement with what is observed experimentally [10].

In conclusion, we have shown that gapless spinons in a spin-liquid state give rise to a power-law optical absorption inside the Mott gap that is larger than that expected for two spin wave absorption in a Neel ordered insulator. Recent experiment has reported the surprising finding that the low temperature optical absorption in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is larger than another compound, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, which exhibits Neel ordering but is "closer" to the Mott transition in that it has a smaller Mott gap [10]. Our result gives a natural explanation of this puzzle. We believe that power-law absorption, especially if it can be observed in a large gap insulator such as the kagome system, is strong evidence for the existence of gapless spinons and gauge fields.

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