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Spinon Phonon Interaction and Ultrasonic Attenuation in Quantum Spin Liquids

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Several experimental candidates for quantum spin liquids have been discovered in the past few years which appear to support gapless fermionic excitations called spinons. The spinons may form a Fermi sea coupled to a U(1) gauge field, and may undergo a pairing instability. We show that despite being charge neutral, the spinons couple to phonons in exactly the same way that electrons do in the long wavelength limit. Therefore, we can use sound attenuation to measure the spinon mass and lifetime. Furthermore, transverse ultrasonic attenuation is a direct probe of the onset of pairing because the Meissner effect of the gauge field causes a “rapid fall” of the attenuation at \( T_c \), in addition to the reduction due to the opening of the energy gap. This phenomenon, well known in clean superconductors, may reveal the existence of the \( U(1) \) gauge field.

Quantum spin liquid in dimensions greater than one is a long sought state of matter which has eluded experimental investigation until recently [1]. We define the quantum spin liquid as an insulator with an odd number of electrons per unit cell which does not order magnetically down to zero temperature due to quantum fluctuations. The theory of quantum spin liquid is rather well developed, and it is expected that if such a state exists, it will have various exotic properties. For example, the low energy excitations may be objects which may carry spin \( \frac{1}{2} \) and no charge, called spinons. The spinons may be gapped or gapless and may obey either boson or fermion statistics.

Both materials are Mott insulators characterized by gapless excitations \([12,13]\). However, less detailed data are available and we shall focus our attention on the organics, even though the conceptual question we raise below will apply equally to these materials if fermionic spinons are found to be present. Spinons with Dirac spectra, however, may require a different treatment.

In this Letter we address two questions. First, how do the spinons couple to phonons, and, secondly, is there a way to unambiguously identify the pairing transition of spinons? As we shall see, the two questions are related because the attenuation of transverse sound turns out to be sensitive to the gauge magnetic field fluctuations and is a sensitive probe of the Meissner effect of gauge magnetic field at the onset of any pairing instability.

The coupling of electrons to phonons is often discussed in terms of the screened Coulomb coupling between

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Next we write a slight departure from Blount, we assume that the kinetic form to a frame moving with the lattice distortion. In a transformation and compute the first term in Eq. (3) to lowest order the sound velocity. We assume a slowly varying displacement following Blount’s discussion of the electron phonon coupling [14]. The present approach makes it clear that for a spinon Fermi surface the result does not rely on gauge fields in the long wavelength limit. We next consider the relaxation of ultrasound due to spinons in parallel with the standard treatment of electron relaxation, including the onset of superconductivity. Differences and similarities will be pointed out.

We begin with a derivation of the spinon-phonon coupling following Blount’s discussion of the electron phonon problem which was also used in Tsuneto’s theory of ultrasound attenuation [15,16]. Blount’s key insight is to transform to a frame moving with the lattice distortion. In a slight departure from Blount, we assume that the kinetic energy of the spinon is described by a mean field band $E_0(k)$. $E_0(k)$ can be a nearest neighbor tight binding band, for instance. The unperturbed Hamiltonian is

$$H_0 = E_0(p) + V_{\text{imp}}(r'),$$

where $p = -i\frac{\partial}{\partial x}$ is the momentum operator, $V_{\text{imp}}$ describes disorder scattering which relaxes the spinon distribution to the lattice, and $r'$ refers to the laboratory frame. The sound wave is described by $\delta R(r', t)$, which is a smoothly varying function of $r'$ such that it equals the displacement of the ions at the lattice points. The transformation to the moving frame $r = r' + \delta R(r', t)$ is accomplished by a canonical transformation $U = e^{iS}$, where $S = \frac{1}{2}(p \cdot \delta R + \delta R \cdot p)$. The transformed Hamiltonian is

$$H_0(r) + H_1(r) = U H_0(r') U^{-1} + i \frac{\partial U}{\partial t} U^{-1},$$

where $H_0(r)$ is the same as Eq. (1) with $r'$ replaced by $r$, and keeping only first order in $\delta R$,

$$H_1 = i[S, E_0(p)] + \frac{i}{2} \sum_{\alpha} \left\{ \nabla_{\alpha} \cdot \frac{\partial \delta R_{\alpha}}{\partial t} \right\}.$$

Next we write $\delta R \sim e^{i(q r - \omega t)}$, where $\omega = v_s q$ and $v_s$ is the sound velocity. We assume a slowly varying displacement and compute the first term in Eq. (3) to lowest order $\nabla \delta R$ and $\frac{\partial \delta R}{\partial t}$. We find

$$H_1 = \sum_{\alpha \beta} \frac{\partial \delta R_{\beta}}{\partial r_{\alpha}} p_{\beta} v_{\alpha} + i p \cdot \frac{\partial \delta R}{\partial t},$$

where $v_{\alpha}(p) = dE_0/dp_{\alpha}$ is the electron velocity. Equation (4) is derived by formally expanding $E_0(p)$ in power of $p$. The second term in Eq. (4) is of order $\omega k_F$, which is smaller than the first term by the ratio $\omega k_F/(q k_F v_F) = v_s/v_F$ and can be dropped.

Now we introduce the phonons

$$H_{\text{ph}} = \sum_{q,\lambda} \omega_{q\lambda} a_{q\lambda}^\dagger a_{q\lambda},$$

where $\lambda$ denotes the phonon branches with polarization $\epsilon_{q\lambda}$. Expanding $\delta R$ in terms of the phonon coordinate and substituting in (4) results in the spinon-phonon coupling term

$$H_{\text{s-phon}} = \sum_{k, q, \lambda, \sigma} M_{k\lambda}(q) f_{k+q\sigma}^\dagger f_{k\sigma}(a_{q\lambda} + a_{-q\lambda}^\dagger),$$

where $\rho_{\text{ion}}$ is the ion mass density. The spinons are coupled to a gauge field $\alpha$ which initially has no dynamics because it was introduced to enforce the constraint of no double occupation. We shall approximate $E_0(k) = k^2/2m$, and the spinon kinetic energy is

$$H_{0s} = \sum_{k, \sigma} \frac{1}{2m} (k + \alpha)^2 f_{k\sigma}^\dagger f_{k\sigma}.$$

In addition, we have the impurity scattering term $H_{\text{imp}} = \sum_{\alpha} v_{\text{imp}}(r) f_{\alpha}^\dagger(r) f_{\alpha}(r)$ and we consider non-spin-flip scattering only. We assume that impurity scattering gives an elastic scattering lifetime $\tau$ and mean free path $l = v_F \tau$ for the spinons.

From this point on we can discuss the sound attenuation in spin liquids in parallel with the theory for metals and superconductors. Historically, the first discussion was in the hydrodynamic regime valid for $q l \ll 1$ [17,18]. The fermions are treated as a viscous medium subject to strain fields set up by the sound waves. This picture is clearly independent of the charge of the fermions and can directly carry over to the spinon case. Starting from the linearized Navier-Stokes equation, the sound wave relaxation time $\tau = \frac{1}{\rho_{\text{ion}} v_s^2} \left( \frac{4}{3} \eta + \chi \right)$, $\eta = \frac{1}{\tau_s} N(0) m^2 v_s^3 \tau$, where $N(0)$ is the density of states at the Fermi level and we can take $\chi$ to be $\ll \eta$.

The hydrodynamic theory was extended by Pippard to all values of $q l$ using a Boltzmann equation approach [19]. Pippard pointed out that when $q l \gtrsim 1$, the electrons develop local charge and current fluctuations for the longitudinal and transverse phonons which contribute significantly to the sound attenuation. Here we rederive Pippard’s results using a diagrammatic method, because it can readily be extended to the pairing case. Our method is simpler than the work of
Tsuneto, who combined a diagrammatic and Boltzmann approach. Since the diagrammatic treatment is not readily available in the literature, we provide the details in the supplementary material [20].

Let us first redervie the results for metals. We compute the phonon self-energy $\Pi(q, \omega)$ due to the excitation of fermion particle-hole pairs. It is given by the diagrams shown in Fig. 1. The bold solid line is the fermion Green’s function with self-energy due to impurity scattering, $G_{\text{ret}}(k, \omega) = (\omega - \xi_k \mp i/2\tau)^{-1}$, where $\xi_k = k^2/2m - \mu$. The electromagnetic (EM) field propagation is screened by repeated bubbles representing density or current fluctuations for longitudinal and transverse sound, respectively. For longitudinal sound the Thomas-Fermi screening length $k^{-1}_T$ is much shorter than $q^{-1}$, and it can be shown (see supplementary material [20]) that the effect of screening is the same as calculating the unscreened bubble [Fig. 1(a)] with a coupling matrix $M_{kl}$ given by Eq. (7) with the trace subtracted, i.e., $M_{kl}(q) = \langle (k \cdot Caha)(k \cdot q) - \frac{1}{2} k^2 (q \cdot Caha) \rangle/m^2 \rho_m \omega \delta_{kq}$. This is the form suggested by Blount [15]. The standard results are reproduced, and the attenuation decreases in the superconducting state when the quasiparticles are gapped. In this rest of this letter we focus on the transverse sound. Here there is no vertex correction (impurity line across the bubbles) because the vertex given by Eq. (7) is odd in the $k$ component along $q$. $\Pi(0)$ describes the dissipation due to the creation of particle-hole excitation of fermions. In $\Pi(1)$ the particle-hole excitation creates current $j$ which couples to the electromagnetic gauge field via $j \cdot A$. The gauge field acquires a self-energy by coupling to current fluctuations, as shown in the third line in Fig. 1. The resulting retarded photon propagator (double wavy line) is given by

$$D_{\text{ret}} = \frac{1}{i\omega \sigma_\perp(q, \omega) + \omega^2 - c^2 q^2},$$

and $c$ is the speed of light. For ultrasound $\omega^2$ in Eq. (10) can safely be ignored. Following Pippard’s notation we write $\sigma_\perp(q, \omega) = g \sigma_0$, where $\sigma_0 = e^2 n \tau/m$ is the dc conductivity, and

$$\Pi^{(0)} = \frac{3}{2a} \text{Re} [s_2(\omega) - s_0(\omega)],$$

$$s_\mu(\omega) = \frac{1}{2i} \int_{-1}^{1} du \frac{u^n}{u + i/\omega},$$

where $a = q l/(1 + i\omega l)$. We can safely assume $\omega l \ll 1$ for the rest of the letter and set $a = q l$. Then $g = 1 - \frac{2}{\omega l}$ when $q l \ll l$ and $g = \frac{2\pi}{\omega l}$ when $q l \gg 1$. We see that in the clean limit ($q l \gg 1$) the first term in the denominator of Eq. (10) is nothing but Landau damping $N(0)/v_F q$, while in the opposite limit ($q l \ll 1$) it gives rise to the classical skin depth $k^{-1}_0$, where $k^{-1}_0 = \omega \sigma_0/c^2$.

The diagrams are evaluated to give (see supplementary material [20])

$$\text{Im} \Pi^{(0)} = \frac{\omega N(0) k_F^3}{4\rho_m v_F^2} \frac{s_1(ql) - s_3(ql)}{iq l}.$$  

(11)

On the other hand, $\Pi^{(1)}$ is proportional to the photon propagator, and depends on the relative size of the inverse skin depth $k^{-1}_0$, $q$, and $l^{-1}$. Let us consider the case when $c^2 q^2 \ll \omega \sigma_\perp(q, \omega)$, in which case $D_{\text{ret}} = \frac{i\omega \sigma_\perp(q, \omega)}{q l}$. This holds under the condition $q l \ll k^{-1}_0$ or $q l \ll k^{-1}_0$. We shall not discuss the extreme clean case, $q l \gg k^{-1}_0$ where $q l \ll k^{-1}_0$ [19], because it is never attained for the spinon case. We can show that $\Pi^{(1)}$ takes the remarkably simple form

$$\Pi^{(1)}_{\text{ret}} = \frac{1 - g}{1 - \text{Im} \Pi^{(0)}_{\text{ret}} + O(v_s/v_F)}.$$  

(12)

The ultrasound attenuation coefficient is given by

$$\alpha = -\frac{2}{v_s} \text{Im} \Pi^{(0)}_{\text{ret}} + \frac{nm}{\rho_m v_s \tau} \frac{1 - g}{g},$$

(13)

where the identity $s_1 - s_3 = -\frac{2}{\sqrt{3}} (1 - g) + O(v_s/v_F)$ has been used, and $n$ is the fermion density. Equation (13) agrees with Pippard’s result derived using Boltzmann’s equation. Using the limit $g = 1 - \frac{2}{\sqrt{3}} (ql)^2$ for $ql \ll 1$, we can verify that the hydrodynamic limit is reproduced.

Now consider the onset of superconductivity. $\Pi^{(0)}$ decreases below $T_c$, due to the opening of the energy gap (see supplementary material [20]), but $\Pi^{(1)}$ is affected much more dramatically. Physically the onset of Meissner effect suppresses the magnetic field fluctuations and $\Pi^{(1)}$ drops to zero rapidly below $T_c$. Mathematically this is because a constant term $e^2 n_s(T)/m$, where $n_s$ is the superfluid density, is added to the denominator of Eq. (10) and quickly dominates $i\omega \sigma_\perp(q, \omega)$. Since $\Pi^{(1)}$ is proportional to $D^{\text{EM}}_{\text{ret}}$, it drops rapidly to very small value. This is called the “rapid fall” and occurs over a millikelvin scale [21] in clean samples. We note from Eq. (12) that the fractional size of the drop is $(1 - g)$ which is very small ($-\frac{2}{\sqrt{3}} (ql)^2$) for $ql \ll 1$ but almost unity $(1 - \frac{3\pi}{4\sqrt{3}})$ in the clean limit of $ql \gg 1$.

Next we turn our attention to the attenuation of transverse sound by spinons. The main difference is that the spinons and gauge fields are treated in 2D. Furthermore, the
Maxwell term $\omega^2 - c^2 q^2$ is missing in the photon propagator. The dynamics of the gauge field is generated by spinon current fluctuation, and instead of Eq. (10) we have

$$D_T = \frac{1}{i o \sigma_\perp (q, \omega) - \chi q^2},$$

where $\sigma_\perp = \bar{g} \sigma_0$, $\sigma_0 = n \pi/m$, $n$ is the spinon density, and $\chi = 1/(24 \pi m)$ is the Landau diamagnetism [22]. Note that according to Eq. (8) the coupling constant to the gauge field has been set to unity instead of $e$. The factor $\bar{g}$ is calculated in 2D and is given by

$$\bar{g} = \frac{2}{a} \left[ t_2(a) - t_0(a) \right],$$

$$t_n(a) = \frac{1}{2i} \int_0^{2\pi} d \theta \frac{\cos^n \theta}{\cos \theta + i/a}.$$

Once again, $\bar{g}$ can be considered a function of $ql$, $\bar{g} = 1 - (ql)^2$ for $ql \ll 1$ and $\bar{g} = \frac{2}{ql}$ for $ql \gg 1$.

Just as in the EM case, we consider the case when $\chi q^2 \ll \omega \sigma_\perp$, i.e., $D_T = (i o \sigma_\perp)^{-1}$, and we conclude that $\alpha$ is given by Eq. (13) with $g$ replaced by $\bar{g}$. In the Fermi liquid state, we can use this formula to get information on the spinon mass and lifetime $\tau$ by studying the $q$ (i.e., $\omega$) dependence. At low temperature $\tau$ is a constant dominated by disorder [6] while at finite temperature $\tau$ may have interesting $T$ dependence due to scattering by other spinons or phonons. If the spinons are paired, the rapid fall also occurs just below $T_c$. Next we show that the condition $\chi q^2 \ll \omega \sigma_\perp$ mentioned above is the only relevant limit. For $ql \gtrsim 1$, $\omega \sigma_\perp$ is estimated to be $(v_s/v_F) k_F^2/m$ and will dominate $\chi q^2$ as long as $v_s/v_F \gg (q/k_F)^2$. Since $v_s/v_F = 10^{-3}$, this condition is satisfied for most accessible ultrasound frequencies. In the opposite case ($ql \ll 1$) the condition is $(v_s/v_F) q \gg (q/k_F)^2$ and is easier to violate. However, the rapid fall is very small in this case and difficult to detect and of little interest to us.

Finally we can estimate the temperature range $\Delta T = T_c - T$ of the rapid fall as sketched in Fig. 2. Since $i o \omega \sigma_\perp$ is replaced by $i o \omega \bar{g} - n_s(T)/m$ in Eq. (14) below $T_c$, we find that $\text{Im} \Pi^{(i)}_s = \text{Im} \Pi^{(i)}_N/[1 + (n_s(T)/m o \bar{g})^2]$, where $\Pi^{(i)}_s$ are the values in the superconducting and normal states. For $ql \gg 1$, attenuation is dominated by $\Pi^{(i)}_s$, and we estimate $\Delta T$ as the temperature when $\text{Im} \Pi^{(i)}_s$ has dropped half the normal state value. We assume the mean field (BCS) behavior $n_s(T) = 2n(\Delta T/T_c)$, and we find

$$\frac{\Delta T}{T_c} = \frac{v_s}{v_F}.$$