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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 $S = \frac{1}{2}$ 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.

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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. They will be accompanied by gauge fields, which may be of the $U(1)$ or Z_2 variety. In the past few years, several candidates for the quantum spin liquid have emerged. The best studied example is a family of organic compounds. The original κ-$(ET)_2Cu_2(CN)_3$ salt (abbreviated as ET) [2] has recently been joined by a second material [3], the $Pd(dmit)_2(EtMe_3Sb)$, which we shall refer to as dmit. Both materials are Mott insulators on an approximate triangular lattice with spin $\frac{1}{2}$ per unit cell, but are not far from the Mott transition because they become superconductor (ET) or metal (dmit) under modest pressure. There is no sign of magnetic ordering down to 30 mK despite an exchange interaction $J \approx 250$ K. Both materials show a linear T coefficient of the specific heat at low temperatures and a finite spin susceptibility [4]. The Wilson ratio is close to one, usually associated with metals. and is highly unusual for an insulator. The thermal conductivity κ is a good probe of these low lying excitations. Experiments on the ET salts indeed found a large contribution, but κ/T is reduced below 0.3 K [5]. On the other hand, recent experiments on dmit found that κ/T extrapolates to a constant down to the lowest temperature [6]. These data strongly support the picture that the low lying excitations are mobile fermionic particles, called spinons.

Initial theoretical work pointed to a state where spinons form a Fermi surface and are coupled to $U(1)$ gauge fields [7,8]. However, a peak in the specific heat around 6 K in ET and 4 K in dmit suggests a phase transition, which, in the case of ET, has been confirmed by thermal expansion measurements [9]. Furthermore, the nuclear spin relaxation rate $1/T_1T$ shows a power law decrease below 1 K [10]. These data led to the suggestion that the Fermi surface may be unstable to a pairing instability which nevertheless leaves a finite density of states intrinsically or due to impurities [11]. Thus the true ground state in the organic salts remains unknown at present.

Two other examples, the kagome compound $ZnCu_3(OH)_6Cl_2$ and the three-dimensional hyper-kagome $Na_4Ir_3O_8$ also satisfy the condition of being spin liquids in that they do not show magnetic order and both are 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

electrons and nuclei, and one may have the impression that the charge neutral spinon may couple differently. It turns out that in the long wavelength limit, the coupling matrix elements are exactly the same. This is because in this limit the coupling can be viewed as a distortion of the Fermi surface by the local stress of the unit cell and is the same whether the fermions are charged or not. Recently the spin phonon coupling was discussed in terms of interactions mediated by gauge fields with the conclusion that the coupling is comparable to 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 $E_0(\boldsymbol{k})$. $E_0(\boldsymbol{k})$ can be a nearest neighbor tight binding band, for instance. The unperturbed Hamiltonian is

$$H_0 = E_0(\boldsymbol{p}) + V_{\rm imp}(\boldsymbol{r}'), \tag{1}$$

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

$$H_0(\boldsymbol{r}) + H_1(\boldsymbol{r}) = UH_0(\boldsymbol{r}')U^{-1} + i\frac{\partial U}{\partial t}U^{-1}, \tag{2}$$

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

$$H_1 = i[S, E_0(\boldsymbol{p})] + \frac{i}{2}\sum_\alpha \left\{\nabla_\alpha, \frac{\partial \delta R_\alpha}{\partial t}\right\}. \tag{3}$$

Next we write $\delta\boldsymbol{R} \sim e^{i(\boldsymbol{q}\cdot\boldsymbol{r}-\omega t)}$, 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 $\boldsymbol{\nabla}\delta\boldsymbol{R}$ and $\frac{\partial\delta\boldsymbol{R}}{\partial t}$. We find

$$H_1 = \sum_{\alpha\beta}\frac{\partial \delta R_\beta}{\partial r_\alpha} p_\beta v_\alpha + i\boldsymbol{p}\cdot\frac{\partial\delta\boldsymbol{R}}{\partial t}, \tag{4}$$

where $v_\alpha(\boldsymbol{p}) = dE_0/d\boldsymbol{p}_\alpha$ is the electron velocity. Equation (4) is derived by formally expanding $E_0(\boldsymbol{p})$ in power of $\boldsymbol{p}$. The second term in Eq. (4) is of order ωk_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

$$H_{\rm ph} = \sum_{\boldsymbol{q},\lambda,\sigma}\omega_{\boldsymbol{q}\lambda}a^\dagger_{\boldsymbol{q}\lambda}a_{\boldsymbol{q}\lambda}, \tag{5}$$

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

$$H_{s\text{-ph}} = \sum_{\boldsymbol{k},\boldsymbol{q},\lambda,\sigma} M_{\boldsymbol{k}\lambda}(\boldsymbol{q}) f^\dagger_{\boldsymbol{k}+\boldsymbol{q}\sigma} f_{\boldsymbol{k}\sigma}(a_{\boldsymbol{q}\lambda} + a^\dagger_{-\boldsymbol{q}\lambda}), \tag{6}$$

$$M_{\boldsymbol{k}\lambda}(\boldsymbol{q}) = (\boldsymbol{k}\cdot\hat{\varepsilon}_{\boldsymbol{q}\lambda})[\boldsymbol{q}\cdot\boldsymbol{v}(\boldsymbol{k})](2\rho_{\rm ion}\omega_{\boldsymbol{q}\lambda})^{-1/2}, \tag{7}$$

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

$$H_{0s} = \sum_{\boldsymbol{k},\sigma}\frac{1}{2m}(\boldsymbol{k}+\boldsymbol{a})^2 f^\dagger_{\boldsymbol{k}\sigma}f_{\boldsymbol{k}\sigma}. \tag{8}$$

In addition, we have the impurity scattering term $H_{\rm imp} = \sum_{i\sigma} v_{\rm imp}(\boldsymbol{r}_i) f^\dagger_\sigma(\boldsymbol{r}_i) f_\sigma(\boldsymbol{r}_i)$ and we consider non-spin-flip scattering only. We assume that impurity scattering gives an elastic scattering lifetime τ 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 $ql \ll 1$ [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 τ_s is

$$\tau_s^L = \frac{1}{\rho_{\rm ion}v_s^2}\left(\frac{4}{3}\eta + \chi\right), \qquad \tau_s^T = \frac{1}{\rho_{\rm ion}v_s^2}\eta \tag{9}$$

for the longitudinal and transverse sound, respectively, where η, χ are the shear and compressional viscosities. The sound attenuation constant α defined as the inverse of the phonon mean free path $l_{\rm ph}$ is the imaginary part of $k = (\omega/v_s)(1 + i\omega\tau_s)^{-1/2}$ and given by $\alpha = \frac{\omega^2\tau_s}{2v_s}$ in the limit $\omega\tau_s \ll 1$. The fermion viscosity is given by $\eta = \frac{1}{15}N(0)m^2v_F^4\tau$, where $N(0)$ is the density of states at the Fermi level and we can take χ to be $\ll \eta$.

The hydrodynamic theory was extended by Pippard to all values of ql using a Boltzmann equation approach [19]. Pippard pointed out that when $ql \gtrsim 1$, 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 rederive 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, $G_{\mathrm{ret(adv)}}(k,\omega) = (\omega - \xi_k \pm i/2\tau)^{-1}$, 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_{TF}^{-1} 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 $\tilde{M}_{k\lambda}$ given by Eq. (7) with the trace subtracted, i.e., $\tilde{M}_{\boldsymbol{k}\lambda}(\boldsymbol{q}) = [(\boldsymbol{k}\cdot\hat{\varepsilon}_{\boldsymbol{q}\lambda})(\boldsymbol{k}\cdot\boldsymbol{q}) - \frac{1}{3}k^2(\boldsymbol{q}\cdot\hat{\varepsilon}_{\boldsymbol{q}\lambda})]/m\sqrt{2\rho_{\mathrm{ion}}\omega_{\boldsymbol{q}\lambda}}$. 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 the 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 $\boldsymbol{k}$ component along $\boldsymbol{q}$. $\Pi^{(0)}$ describes the dissipation due to the creation of particle-hole excitation of fermions. In $\Pi^{(1)}$ the particle-hole excitation creates current $\boldsymbol{j}$ which couples to the electromagnetic gauge field via $\boldsymbol{j}\cdot\boldsymbol{A}$. 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

$$D_{\mathrm{ret}}^{\mathrm{EM}} = \frac{1}{i\omega\sigma_\perp(q,\omega) + \omega^2 - c^2q^2}, \tag{10}$$

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

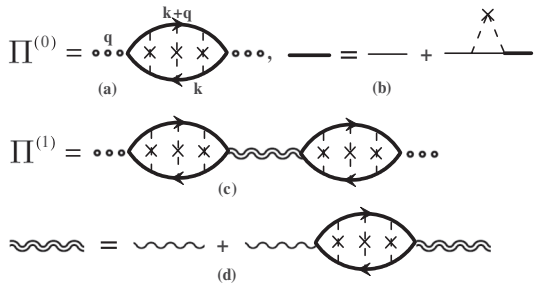

FIG. 1. Feynman diagrams for phonon self-energy. (a) Phonon self-energy due to fermion particle-hole excitations. (b) Fermion Green's function with self-energy due to impurity scattering. (c) Phonon self-energy due to current fluctuation. (d) The gauge field acquires a self-energy by coupling to current or charge fluctuations.

$$g = \frac{3}{2a}\,\mathrm{Re}[s_2(a) - s_0(a)],$$
$$s_n(a) = \frac{1}{2i}\int_{-1}^{1} du\,\frac{u^n}{u + i/a},$$

where $a = ql/(1 + i\omega\tau)$. We can safely assume $\omega\tau \ll 1$ for the rest of the Letter and set $a = ql$. Then $g \to 1 - \frac{2}{15}(ql)^2$ when $ql \ll 1$ and $g \to \frac{3\pi}{4}(ql)^{-1}$ when $ql \gg 1$. We see that in the clean limit ($ql \gg 1$) the first term in the denominator of Eq. (10) is nothing but Landau damping $N(0)\omega/v_Fq$, while in the opposite limit ($ql \ll 1$) it gives rise to the classical skin depth k_0^{-1} where $k_0^2 = \omega\sigma_0/c^2$.

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

$$\mathrm{Im}\,\Pi_{\mathrm{ret}}^{(0)} = \frac{\omega N(0)k_F^3}{4\rho_{\mathrm{ion}}mv_s^2}\,\frac{s_1(ql) - s_3(ql)}{iql}. \tag{11}$$

On the other hand, $\Pi_{\mathrm{ret}}^{(1)}$ is proportional to the photon propagator, and depends on the relative size of the inverse skin depth k_0, q, and l^{-1}. Let us consider the case when $c^2q^2 \ll \omega\sigma_\perp(q,\omega)$, in which case $D_{\mathrm{ret}}^{\mathrm{EM}} = (i\omega\sigma_\perp(q,\omega))^{-1}$. This holds under the condition $q \ll k_0$ if $ql \ll 1$ and $q^2 \ll k_0^2/(ql)$ if $ql \gg 1$. (We shall not discus the extreme clean case, $q^2 \gg k_0^2/ql$ while $q \ll k_0$ [19], because it is never attained for the spinon case.) We can show that $\Pi_{\mathrm{net}}^{(1)}$ takes the remarkably simple form

$$\Pi_{\mathrm{ret}}^{(1)} = \frac{1-g}{g}\,\mathrm{Im}\Pi_{\mathrm{ret}}^{(0)} + O(v_s/v_F). \tag{12}$$

The ultrasound attenuation coefficient is given by

$$\alpha = -\frac{2}{v_s}\,\mathrm{Im}(\Pi_{\mathrm{ret}}^{(0)} + \Pi_{\mathrm{ret}}^{(1)}) = \frac{nm}{\rho_{\mathrm{ion}}v_s\tau}\,\frac{1-g}{g}, \tag{13}$$

where the identity $s_1 - s_3 = -\frac{2i}{3}(1-g) + O(v_s/v_F)$ has been used, and n is the fermion density. Equation (13) agrees with Pippard's result derived using Boltzmann's equation. Using the limit $g = 1 - \frac{2}{15}(ql)^2$ 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 $e^2n_s(T)/m$, 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 $D_{\mathrm{ret}}^{\mathrm{EM}}$, 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 ($\sim\frac{2}{15}(ql)^2$) for $ql \ll 1$ but almost unity ($1 - \frac{3\pi}{4ql}$) 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

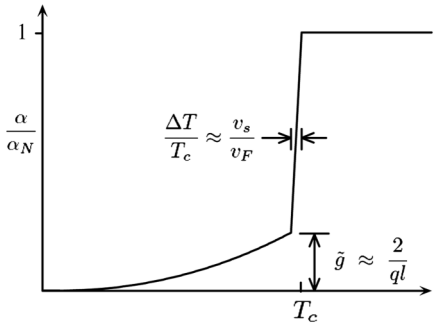

FIG. 2. Schematic plot of the attenuation of transverse sound (for $ql \gtrsim 1$) in a spin liquid normalized to α_N, the value in the Fermi liquid state. The "rapid fall" over a narrow temperature range ΔT at the transition to the spinon paired state is due to the Meissner effect of the gauge field.

Maxwell term $\omega^2 - c^2q^2$ 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

$$D^T_{\text{ret}} = \frac{1}{i\omega\tilde{\sigma}_\perp(q,\omega) - \chi q^2}, \tag{14}$$

where $\tilde{\sigma}_\perp = \tilde{g}\tilde{\sigma}_0$, $\tilde{\sigma}_0 = n\tau/m$, 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 $\tilde{g}$ is calculated in 2D and is given by

$$\tilde{g} = \frac{2}{a}[t_2(a) - t_0(a)],$$
$$t_n(a) = \frac{1}{2i}\int_0^{2\pi} d\theta \frac{\cos^n\theta}{\cos\theta + i/a}.$$

Once again, $\tilde{g}$ can be considered a function of ql, $\tilde{g} = 1 - \frac{(ql)^2}{4}$ for $ql \ll 1$ and $\tilde{g} = \frac{2}{ql}$ for $ql \gg 1$.

Just as in the EM case, we consider the case when $\chi q^2 \ll \omega\tilde{\sigma}_\perp$, i.e., $D^T_{\text{ret}} = (i\omega\tilde{\sigma}_\perp)^{-1}$, and we conclude that α is given by Eq. (13) with g replaced by $\tilde{g}$. In the Fermi liquid state, we can use this formula to get information on the spinon mass and lifetime τ by studying the q (i.e., ω) dependence. At low temperature τ is a constant dominated by disorder [6] while at finite temperature τ 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 $\chi q^2 \ll \omega\tilde{\sigma}_\perp$ mentioned above is the only relevant limit. For $ql \gtrsim 1$, $\omega\tilde{\sigma}_\perp$ is estimated to be $(v_s/v_F)k_F^2/m$ and will dominate χq^2 as long as $v_s/v_F \gg (q/k_F)^2$. Since $v_s/v_F \approx 10^{-3}$, this condition is satisfied for most accessible ultrasound frequencies. In the opposite case ($ql \ll 1$) the condition is $(v_s/v_F)ql \gg (q/k_F)^2$ 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 $i\omega\tilde{\sigma}_\perp$ is replaced by $i\omega\tilde{\sigma}_\perp - n_s(T)/m$ in Eq. (14) below T_c we find that $\text{Im}\Pi^{(1)}_s = \text{Im}\Pi^{(1)}_N/\{1 + [n_s(T)/m\omega\tilde{\sigma}_0\tilde{g}]^2\}$, where $\Pi^{(1)}_{s,N}$ are the values in the superconducting and normal states. For $ql \gg 1$, attenuation is dominated by $\Pi^{(1)}$, and we estimate ΔT as the temperature when $\text{Im}\Pi^{(1)}_s$ 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

$$\frac{\Delta T}{T_c} \approx \frac{v_s}{v_F}. \tag{15}$$

We conclude that fermionic spinons in a fermion couple to phonons in a way that is identical to electron phonon coupling in the long wavelength limit. For $ql \gg 1$ the attenuation of transverse ultrasound is dominated by a component which is due to the fluctuations of transverse gauge fields. At the pairing transition of the spinon, this component is suppressed by the onset of the Meissner effect for the gauge field, leading to a rapid drop in attenuation in a very narrow temperature range below T_c given by Eq. (15). After the rapid fall, the attenuation is reduced in the usual way by the gapping of the quasiparticles. We believe the phenomenon of the rapid fall in the attenuation of transverse ultrasound gives a clear signature of spinon pairing and the existence of $U(1)$ gauge fields.

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