Spin–1 Neutron Resonance Peak Cannot Account for Electronic Anomalies in the Cuprate Superconductors

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In certain cuprates, a spin-1 resonance mode is prominent in the magnetic structure measured by neutron scattering. It has been proposed that this mode is responsible for significant features seen in other spectroscopies, such as photoemission and optical absorption, which are sensitive to the charge dynamics, and even that this mode is the boson responsible for "mediating" the superconducting pairing. We show that its small (measured) intensity and weak coupling to electron-hole pairs (as deduced from the measured lifetime) disqualifies the resonant mode from either proposed role.

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Introduction.—One of the most striking features of the high temperature superconducting cuprates is the sharp resonance peak observed in inelastic neutron scattering measurements [1–6]. This phenomenon has a clear and intimate relation to the superconductivity that occurs in these materials—the resonance grows in intensity and narrows at temperatures less than the superconducting T_c, and its intensity is suppressed by perpendicular magnetic fields in a way that correlates directly with the suppression of superconductivity [7].

Since its discovery, there have been many interesting theoretical proposals concerning the origin and implications of the resonance. One class of proposals identifies the resonance peak as a signature of superconductivity, relating its intensity to the condensation energy [8] or condensate fraction [8,9] of the superconducting state. Other proposals focus on the effect that scattering of quasiparticles from the resonance has on various other experimentally accessible properties of the cuprates, especially those that show dramatic changes as the temperature is lowered from above to below T_c. For instance, this idea has been invoked to explain the "peak-dip-hump" structure [10,11] and the "kink" [12] in the quasiparticle dispersion measured by angle resolved photoemission spectroscopy (ARPES), and the pseudogap structure seen in optical conductivity [13]. Finally, there are proposals which consider the resonance mode to be the boson which "mediates" an effective attraction between electrons which is responsible for the high-temperature superconducting pairing [11,14,15].

In this Letter, we address what the resonance mode can and cannot do. In particular, we will show that the first set of ideas requires that the integrated intensity associated with the resonance, I_0, be small in units of the total integrated spin structure factor, while the latter require $I_0 \sim 1$. While apparently contradictory numbers exist in the experimental literature, a careful analysis [16–19] shows that values of I_0 in the few percent range can be deduced from all the absolute intensity measurements. (See Table I.) Thus, the resonance peak may be a unique probe of the superconducting condensate [9], and might account for the condensation energy [8,17], but it cannot cause any significant structure in ARPES and optical conductivity.

Possible relation to the condensation energy.—The concept of condensation energy is not well defined when fluctuation effects are important [20]. In the absence of better estimates, we adopt the mean-field expression for the condensation energy:

$$U = \tfrac{1}{2}\rho\Delta^2 \sim 2\rho T_c^2 . \quad (1)$$

If the density of states is proportional to $1/(2J)$ where J is the exchange interaction in the t-J model invoked in Ref. [8], the condensation energy can be expressed as

$$U \sim \frac{T_c^2}{J} = J\left(\frac{T_c}{J}\right)^2. \quad (2)$$

In this context, Scalapino and White [21] have pointed out that there is an exact relation between the nearest-neighbor exchange contribution to the internal energy and the magnetic structure factor $S(\vec{k},\omega)$ which in spatial dimension $d = 2$ is

$$U_{\mathrm{mag}} = J\int \frac{d\omega\, d\vec{k}}{(2\pi)^{d+1}}[2 - \cos(k_x) - \cos(k_y)]S(\vec{k},\omega). \quad (3)$$

TABLE I. Integrated spectral weight in the resonant peak well below T_c in units such that a spin 1/2 per planar Cu atom would have integrated weight equal to 1.

Material	T_c (in K)	I_0	Reference
$YBa_2Cu_3O_{6.5}$	52	0.017	[6]
$YBa_2Cu_3O_{6.6}$	62.7	0.01 ± 0.007	[16,17]
$YBa_2Cu_3O_{6.7}$	67	0.014	[6]
$YBa_2Cu_3O_{6.85}$	87	0.017	[6]
$YBa_2Cu_3O_{6.99}$	93	0.011	[6]
$Bi_2Sr_2CaCu_2O_{8+\delta}$	91	0.057	[5]

Therefore, it is clear that if the condensation energy comes principally from this term, and if it is due to the transfer of spectral weight from a broad (in $\vec{k}$) background into the resonant peak, then its intensity must be very small, $I_0 \sim (T_c/J)^2$. Also in accord with these ideas, Dai *et al.* [17] have shown that, in absolute units, the specific heat is roughly equal to the temperature derivative of the resonance intensity.

Scattering from a collective mode.—A collective mode with a strongly temperature-dependent intensity is unusual, so it is natural to attribute other strongly temperature-dependent spectral features to the coupling between quasiparticles and the collective mode. However, unless the mode has large weight, it is as if it were hardly there at all, however prominent it may appear in a scattering experiment. Consider, for example, the electron-phonon coupling in a weakly correlated metal with a very large number, N, of atoms per unit cell. The scattering of electrons from any one optical mode will not, generally, have a significant effect on the electron dynamics—its effects will be reduced by a factor of $1/N$.

Several prominent features of the ARPES spectra of the high-temperature superconductors have been attributed to scattering of electrons from the resonant mode, in particular the pronounced peak-dip-hump structure in the "antinodal region" of the Brillouin zone (near $\vec{k} = \langle \pi, 0 \rangle$) and the kink in the electron dispersion seen especially in the "nodal direction" ($\langle 0,0 \rangle$ to $\langle \pi, \pi \rangle$). These are order 1 effects, and so require a large intensity of the resonance peak unless the coupling to quasiparticles is extremely large [10]. On the same grounds, the pseudogap features in the optical conductivity require a large scattering from the resonant peak.

Therefore, it is clear that the resonance peak cannot be responsible for both the condensation energy *and* the pronounced structures in the scattering rates.

The spectral intensity of resonance peak is small.—The resonance peak is the most prominent feature seen in inelastic neutron scattering of the high-temperature superconductors, $YBa_2Cu_3O_{7-\delta}$ (YBCO) [1–4,17]. Even though it turns on at a temperature which increases with decreasing T_c and thus tracks the celebrated pseudogap phenomenon, its most rapid evolution in intensity, lifetime, and width in k space occurs near T_c. In addition, its frequency scales with T_c and its field dependence [7], both in magnitude and anisotropy, is linked to the upper critical field H_{c2}. These experiments establish a strong connection between the resonance and both the spin and orbital aspects of superconductivity. However, the resonance is so prominent only because its spectral weight is concentrated in a narrow range of frequency and $\vec{k}$.

Absolute intensity measurements reveal that the intensity, when properly integrated over frequency and the Brillouin zone, is always rather small [16]. (See Table I.) Simple considerations of the chemistry and physics of the copper-oxide planes leads to the conclusion that each planar copper is in a d^9 configuration with its orbital angular momentum quenched by the crystal field, leaving only a $S = 1/2$ degree of freedom. We therefore expect the total integrated spectral weight per planar copper to be $\hbar^2 S(S+1) = \hbar^2(3/4)$. In YBCO, the measured spectral weight in the resonance is [16] of the order of 1%–2% of this. In $Bi_2Sr_2CaCu_2O_{8+\delta}$, because the resonant peak appears broader [5] in $\vec{k}$, its integrated strength might be somewhat larger, but still at the 5% level. These ratios are not subject to significant uncertainties. For example, the same experimental methods show that in the undoped, antiferromagnetic "parent" materials, the measured spectral weights [22,23] are in quantitative agreement with the results of spin-wave theory for a $S = 1/2$ system. Furthermore, the relatively low doped hole densities, even at "optimal" doping, implies that the total magnetic spectral weight (below the charge transfer gap) cannot differ greatly from $S(S+1)$.

A little mathematics.—Let us now carry out the simplest calculation to illustrate our argument. The Hamiltonian which represents the coupling between the conduction electrons and the spin mode is as follows:

$$H = H_0 + g\mathbf{S} \cdot \psi^\dagger \vec{\sigma} \psi + H_s, \tag{4}$$

where H_0 and H_s are the bare Hamiltonians for the conduction electrons and spins, respectively.

We approximate the imaginary part of the zero temperature spin susceptibility, measured via neutrons, as

$$\begin{aligned} \mathrm{Im}\chi(\mathbf{q}, \omega) = (\pi/3) g_L^2 \mu_B^2 S(S+1) \\ \times \Big\{ I_0(2\pi)[\delta(\omega - \Omega) - \delta(\omega + \Omega)] f(\mathbf{q}) \\ + \frac{(1 - I_0)\mathrm{sgn}(\omega)}{\Lambda_\omega \Lambda_q^d} \Big\}, \end{aligned} \tag{5}$$

where Ω is the resonance frequency, g_L is the Lande g factor, and Λ_ω and Λ_q are, respectively, the frequency and momentum cutoffs. The structure factor of the resonant mode, $f(\mathbf{q})$, is known to be peaked at $\vec{q}$ in the neighborhood of $\mathbf{Q} = (\pi, \pi)$; for simplicity we will take $f(\mathbf{q}) = (2\pi)^d \delta(\mathbf{q} - \mathbf{Q})$, although the results are easily generalized to the case in which f is a Lorentzian or Gaussian. We will also take $d = 2$, although, of course, the real cuprate superconductors are anisotropic three-dimensional systems.

ARPES: The leading perturbative contribution to the self-energy from the resonance peak for $\omega > 0$ is written as

$$\begin{aligned} \Sigma(\mathbf{k}, \omega) = I_0 g^2 \int d^2q \left(\frac{1}{\omega - \Omega - \xi_{\mathbf{k}-\mathbf{q}} + i\eta} \right) \\ \times \delta(\mathbf{q} - \mathbf{Q}), \end{aligned} \tag{6}$$

where $\xi_{\mathbf{k}}$ is the quasiparticle dispersion. Therefore, the single particle spectrum has two poles located at

$$\begin{aligned} \omega_1 = \xi_{\mathbf{k}} - \frac{I_0 g^2}{\Omega} + \ldots, \\ \omega_2 = \Omega + \xi_{\mathbf{k}-\mathbf{Q}} + \frac{I_0 g^2}{\Omega} + \ldots, \end{aligned} \tag{7}$$

where ... refers to terms of order $\mathcal{O}(\frac{I_0^2 g^4}{\Omega^3})$. The weight of each pole is

$$Z_{\omega_1} = 1 - \frac{I_0 g^2}{\Omega^2} + \ldots, \qquad Z_{\omega_2} = \frac{I_0 g^2}{\Omega^2} + \ldots. \tag{8}$$

To the same order, the scattering from the remaining (nonresonant) spin fluctuations produces an additive contribution to Σ proportional to $g^2(1 - I_0)$ which is of the marginal Fermi liquid form, discussed elsewhere [24].

Optical conductivity: While a perturbative expression for the conductivity, $\sigma(\omega)$, itself is impossible, due to its singular behavior at small ω, it is straightforward to obtain a perturbative expression for the so-called frequency-dependent scattering rate [25], defined in terms of the real and imaginary parts, σ' and σ'', as

$$1/\tau^*(\omega) \equiv \omega\sigma'/\sigma'' = 1/\tau_0^*(\omega) + 1/\tau_1^*(\omega). \tag{9}$$

To lowest order, the contribution to the $T = 0$ scattering rate from the resonance mode is

$$1/\tau_0^*(\omega) = \frac{m\omega^2}{ne^2} g^2 I_0 F(\omega), \tag{10}$$

where $4\pi e^2 n/m$ is the Drude weight and

$$F(\omega) = \frac{\pi^2 e^2}{\omega^3 m^2 v_F v_\Delta}(\omega - \Omega)\theta(\omega - \Omega). \tag{11}$$

To obtain the explicit expression for $F(\omega)$, we have used the dispersion relation for the nodal quasiparticle with two different velocities, where v_F and v_Δ are, respectively, the velocities perpendicular and parallel to the Fermi surface. A different assumed dispersion relation would not change the overall conclusion of this paper, although it would change the detailed structure of $F(\omega)$. The contribution of the constant part of the spin susceptibility is, unsurprisingly, as in a marginal Fermi liquid, linear in the frequency

$$1/\tau_1^*(\omega) \propto g^2(1 - I_0)|\omega|. \tag{12}$$

Since Matthiessen's rule holds to this order, these scattering rates should simply be added (and so should the scattering due to any other process).

Our analytic results are consistent with the more complicated results obtained for more detailed and realistic models previously [10,13]. However, the present results highlight the fact that all effects of the resonance mode are proportional to I_0, and so are negligible if I_0 is small.

What about the coupling constant?—The effects of the resonant mode are not just proportional to I_0, but depend on the coupling strength g. Could we imagine obtaining a large effect with a small I_0 but a large g? Of course, a large g is incompatible with any sort of perturbative treatment, so such an approach probably does not make sense. However, it also turns out that one can obtain a reasonable estimate of the coupling constant from the experimentally measured frequency width (lifetime) of the resonance peak. The resonance mode in YBCO in the superconducting state is very sharp, with an intrinsic linewidth (Γ) of about 2 meV. In optimally doped material, the mode is unobservable above T_c, but in underdoped material it persists to higher temperatures. Here, the line broadens [17] so that $\Gamma \approx 10$ meV. Such a broadening is expected whenever the resonance mode can decay into electron-hole pairs. This decay channel is somewhat suppressed well below T_c due to the limited phase space available for such electron-hole pairs.

An analogous problem was solved long ago, where instead of the resonant mode, the crystal-field excitations in metallic rare-earth systems were investigated. Simply adopting the expressions obtained there for the damping of an electronic collective mode due to electron-hole excitations, one obtains [27,28]

$$\Gamma = 4\pi[gN(0)]^2\Omega, \tag{13}$$

where $N(0)$ is the density of states at the Fermi energy, or more precisely the density of particle-hole states with momentum $\vec{Q}$ and energy Ω. In order to invert this equation to obtain an estimate of g, we can use the measured values of Γ and Ω, but we need an estimate of $N(0)$. In the normal state, this can be done in several ways. First, on the basis of the theoretical expectation that the bandwidth of the electrons is renormalized down to something of order J, or for that matter from the measured ARPES spectra, it is possible to obtain a rough dimensional estimate of the normal state density of states, $N(0) = 1/W \sim 1/(100\text{ meV})$, to obtain an estimate of g:

$$g \sim 14\text{ meV}. \tag{14}$$

This is not a large coupling. The conventionally defined dimensionless coupling constant $\lambda = 2I_0 g^2 N(0)/\Omega$ is only $\lambda \sim I_0/10$. Needless to say, such a feeble (small I_0) boson coupled so weakly (small g) to electron-hole pairs cannot mediate a strong pairing interaction; searches for the mechanism of high T_c must begin elsewhere.

One might worry that our estimate of $N(0)$ is somewhat too large, as it does not take into account any suppression of the density of states due to the pseudogap observed above T_c in underdoped materials—a smaller assumed $N(0)$ would give rise to a larger estimate of g. However, a more direct estimate of the density of states can be obtained from the measured [29] specific heat in the normal state; for YBCO, $\gamma \equiv C_v/T$ approaches a normal state value of around 2 mJ/gm-at K^2, which corresponds to a density of states per copper of $N(0) = 11\text{ eV}^{-1}$, in good agreement with our dimensional estimate.

Finally, an independent estimate of the coupling constant can be obtained from the measured lifetime in the superconducting state. Here the particle-hole continuum is dominated by the nodal quasiparticles, whose dispersion is presumably known. It is straightforward to see that the appropriate density of states per spin with momentum $\vec{Q}$ and energy Ω computed within this model is $\pi\omega/(v_F v_\Delta k_n^2)$, where $\vec{k}_{\text{node}} = \langle k_n, k_n\rangle$ is the position of the nodal point measured from $(\pi/2, \pi/2)$. With this expression for the density of states, and taking the canonical values of $v_\Delta \sim 1.2 \times 10^6$ cm/s [30], $v_F \sim 1.7 \times 10^7$ cm/s, and $k_n \sim 1/8 \times 10^{-8}$ cm [31], we obtain an estimate $g \sim 2$ meV and $\lambda \sim 0.01 I_0$.

Although it takes us a bit into the realm of speculation, it is worth noting that the remarkably small value of the coupling to the resonant mode is not, altogether, unexpected. If we think of the resonant mode, in some loose sense, as a would-be antiferromagnetic magnon [11], then an argument due to Schrieffer [32] implies that it couples only through gradient couplings to particle-hole pairs. In particular, one might expect the average coupling to be roughly proportional to the reciprocal space width around $\vec{k} = \vec{Q}$ occupied by the resonant peak. Since this width is of order 20% of the width of the Brillouin zone, it is reasonable to expect the coupling to the antiferromagnetic resonance itself to be correspondingly reduced relative to an order 1 microscopic coupling between electrons and spins.

Conclusion.—The resonance mode is important because it is the most prominent feature of an especially simple correlation function. It is one of the salient features of high temperature superconductivity whose understanding will eventually result in significant insight into the mechanism of high-temperature superconductivity. However, to the best of our knowledge, its spectral weight is always small. Therefore, the existence and character of the resonance mode may well be a direct consequence of the high-temperature superconductivity in the cuprates but it cannot be the "glue" in any conventional pairing theory, nor can it account for anomalies in photoemission and optical absorption data. This conclusion is reinforced by the observation that many [33,34] of the putative signatures of scattering from the resonant peak are observable in $La_{2-x}Sr_xCuO_4$, where no resonant mode has been seen in neutron scattering.

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