Magnetic resonant excitations in High-$T_c$ superconductors

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The observation of an unusual spin resonant excitation in the superconducting state of various High-$T_c$ copper oxides by inelastic neutron scattering measurements is reviewed. This magnetic mode is discussed in light of a few theoretical models and likely corresponds to a spin-1 collective mode.

More than fifteen years after the high temperature superconductivity discovery, antiferromagnetic (AF) fluctuations pairing mechanism$^1$ is still highly controversial. However, inelastic neutron scattering (INS) measurements have successfully brought to light the existence of unusual AF excitations that develop below $T_c$ and could be the hallmark of an unexpected spin-1 collective mode, tightly bound to the superconducting (SC) state. Whatever the role of that mode for superconductivity, it has to be derived from the same microscopic model used to discuss superconductivity. We here review its characteristic features in a few cuprates and discuss its possible origin in light of different theoretical models.

In optimally doped YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) ($T_c$=93 K) where it has been discovered$^2$ (Fig. 1a), the spin excitation spectrum is dominated in the SC state by a sharp magnetic excitation at an energy of $\sim$40 meV and at the planar antiferromagnetic wave vector $\mathbf{q}_{AF} = (\pi/a, \pi/a)$, the so-called magnetic resonance peak$^2$ $^3$ $^4$ $^5$. Its intensity decreases with increasing temperature and vanishes steeply at $T_c$, without any significant shift of its characteristic energy $E_r$. In the underdoped regime, $E_r$ monotonically decreases with decreasing hole concentration$^6$ $^7$ so that $E_r \simeq 5 k_B T_c$ (Fig. 2). Besides, it is possible to vary $T_c$ without changing the carrier concentration through impurity substitutions of Cu in the CuO$_2$ planes. This is the case in YBa$_2$(Cu$_{1−y}$ Ni$_y$)$_3$O$_7$ (y=1%, $T_c$=80 K), where the magnetic resonance peak shifts to lower energy with a preserved $E_r/k_B T_c$ ratio (Fig. 1b) $^8$.

In optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCO) ($T_c$=91 K), a similar magnetic resonance peak has been observed at 43 meV (Fig. 1d)$^9$). Furthermore, $E_r$ shifts down to 38 meV in the overdoped regime ($T_c$=80 K) $^{10}$, preserving a constant ratio with $T_c$: $E_r \simeq 5.4 k_B T_c$ (Fig. 2). Thus, whatever the hole doping, the energy position of the magnetic resonance peak always scales with $T_c$. In contrast to YBCO, where the resonance peak is resolution limited in energy, the resonance peak in BSCO exhibits an energy width of $\sim 13$ meV. In addition, the momentum width of the excitation is twice broader. A similar energy and momentum broadening has been also reported in YBa$_2$(Cu$_{1−y}$ Ni$_y$)$_3$O$_7$ $^8$ (Fig. 1b) and can therefore be ascribed to disorder, such as impurities or inhomogeneities. Furthermore, the observation of a spatial distribution of the SC gap in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ by Scanning Tunneling Microscopy measurements$^{11}$ provides evidence in favor of an intrinsic disorder in this system.

Further, the magnetic resonance peak has been observed in optimally doped Tl$_2$Ba$_2$CuO$_{6+\delta}$($T_c$=90 K) at $E_r$=47 meV (Fig. 1c) $^{12}$. This yields a ratio $E_r/k_B T_c \simeq 6$ slightly larger than YBa$_2$Cu$_3$O$_{6+x}$. Nevertheless, as in YBa$_2$Cu$_3$O$_7$, the excitation is limited by the resolution in energy and displays a momentum width of 0.25 Å$^{-1}$ (half width at half maximum). Meanwhile, the energy integrated intensity of
the magnetic resonance peak is almost the same in both systems: 0.7-0.8 \mu_0^2 eV^{-1}/CuO_2 plane. Thus, the magnetic resonance peak appears as a common excitation to the SC state of all High-\(T_c\) superconductors, investigated so far by INS measurements, whose maximum \(T_c\) can be as high as \(\sim 90\) K. Furthermore, the existence of this excitation does not depend on the number of CuO_2 planes per unit cell: one for Tl_2Ba_2CuO_B and two for YBa_2Cu_3O_{6+x} and Bi_2Sr_2CaCu_2O_{8+\delta}.

While the magnetic resonance peak exists in cuprates whose maximum \(T_c\) is about 90 K, it has never been observed in the mono-layer system La_{2-x}Sr_xCuO_4 with a maximum of \(T_c\) of \(\sim 40\) K. Furthermore, the magnetic excitations in that compound are rather strong even in the normal state and located at incommensurate planar wave vector \(Q_{mag} = (\pi/a(1 \pm \delta_{inc}), \pi/a)\) and \(\pi/a, \pi/a(1 \pm \delta_{inc})\) [16]. This is in a marked contrast with the systems mentioned above, for which the normal state magnetic fluctuations (if observable) remain centered around \(\pi/a, \pi/a\). However, passing through \(T_c\), the incommensurate spin fluctuations of La_{2-x}Sr_xCuO_4 are enhanced and become narrower in momentum space, in an energy range which is about 5\(k_B T_c\) [17]. This phenomenon, usually referred to as a "coherence effect" and the resonance peak could eventually share a common origin.

In underdoped YBa_2Cu_{3+x}O_{6+x} (\(x=0.6, T_c=63\) K, \(E_r=34\) meV), INS measurements provide evidence for incommensurate-like spin fluctuations at 24 meV and low temperature (seemingly similar to those observed La_{2-x}Sr_xCuO_4) [18]. These incommensurate-like spin fluctuations are also observed at higher oxygen concentrations: \(x=0.7\) [19], \(x=0.85\) [20]. As a function of temperature and energy [20], the incommensurability \((\delta_{inc})\) increases below \(T_c\) with decreasing temperature and decreases upon approaching \(E_r\) in the...
SC state (Fig. 3a). The simultaneous disappearance of $\delta_{\text{inc}}$ at $E_c$ and $T_c$ indicates that the resonance peak and the incommensurate-like spin fluctuations are intrinsic features of the SC state and that they can be viewed as continuously connected (Fig 3a). In other words, these results lead to an unified description of both the incommensurate spin excitations and the magnetic resonance peak in terms of a unique collective spin excitation mode with a downward dispersion [20]. Recently, the actual symmetry of this dispersion for an optimally doped YBCO sample has been looked at carefully [21] and was found basically circular within the 2D copper-oxygen plane. In addition, a second magnetic mode with much weaker intensity is reported dispersing upward above the $(\pi/a, \pi/a)$ peak [21]. The deep underdoped state YBCO$_{6.5}$ has been also recently re-investigated in partly detwinned sample with an ortho-II structure [22]. In contrast to the dispersive mode picture, it is claimed [22] that the low energy magnetic excitations are essentially one-dimensional as expected for hydrodynamic stripes. Therefore, the detailed doping dependence of the spin fluctuations needs to be clarified to reconcile these conclusions by studying fully detwinned samples. Indeed, one needs to determine the specific role of the Cu-O chains in YBCO for the magnetic anisotropy.

The main difference between mono-layer and bilayer systems shows up in the momentum dependence of the magnetic resonance peak along the (0 0 1) direction. In Tl$_2$Ba$_2$CuO$_{6+\delta}$, the excitation remains purely bidimensional. In contrast in bilayer systems, the two CuO$_2$ planes correlate antiferromagnetically within the bilayer. That interlayer AF coupling is responsible in insulating parent compounds for both acoustic and optic magnons, whose counterpart in the metallic state are the odd (o) and even (e) excitations. The neutron scattering cross section then reads [6]:

$$\frac{d^2\sigma(Q,\omega)}{d\Omega d\omega} \propto \sin^2(Qz d/2) Im[\chi_o(Q,\omega)] + \cos^2(Qz d/2) Im[\chi_e(Q,\omega)]$$

where $\text{Im}[\chi_{o,e}(Q,\omega)]$ corresponds to the imaginary part of the dynamical magnetic susceptibility in each channel and $d$ ($=3.3$ Å) stands for the distance between CuO$_2$ planes within the bilayer. Intuitively, one could expect a splitting of the magnetic resonance peak under the interlayer AF coupling, leading to a magnetic resonance in each channel. For a long time, the magnetic resonance peak was observed only in the odd channel. However, we could recently observe a resonant mode in each channel in slightly overdoped YBCO through 10 % substitution of Y by Ca [23]. They occur at two different energies $E_c^o = 36$ meV and $E_c^e = 43$ meV and the even mode exhibits an intensity one third times less than the odd one. The question why the even mode is now sizeable in this overdoped regime and not in previous studies remains open as it could be simply due to improvement of neutron instruments. However, it might also be related to the electronic transport between closely spaced CuO$_2$ layers which becomes coherent in the overdoped regime, as demonstrated by recent experiments showing well-defined bonding and antibonding bands.

Considering the different models for the magnetic resonance peak, we focus on models where electron-electron interactions play the central role, despite the still possible existence of electron-phonon couplings in cuprates. Essentially, there is no indication of an effect of the lattice on the magnetic resonance peak. Secondly, we consider here models where the downward dispersion of the resonant mode would naturally emerge. For instance, approaches [24] [25] which associate the resonance peak to a pre-existing soft mode reminiscent of nearby (commensurate or incommensurate) AF phase would yield a collective mode dispersing predominantly upward.

The existence of a spin 1-collective in d-wave superconductors such as High-T$_c$ cuprates, is derived from an itinerant description of the magnetic properties of the system and of strong correlation effects [26] [27] [28] [29] [30] [31] and references therein). This leads to a particle-hole ($p-h$) bound state, usually referred as a spin exciton. In these strong coupling models (see also Refs. [32] [33] in the framework of the $t-J$ model), the generalized spin susceptibility $\chi(q,\omega)$ is expressed as a function of the non interaction spin susceptibility $\chi_0(q,\omega)$ and the magnetic interaction. $\chi(q,\omega)$ has an RPA-like form:

$$\chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 + J(q)\chi_0(q,\omega)}$$
magnetic interaction (Fig 3.b). The mode vanishes when approaching the continuum by changing wave vector from \( \mathbf{q} \) by a downward dispersion controlled by the momentum dependences of the continuum threshold and the smaller that the threshold of the continuum at wave vector \( \mathbf{q} \).

So far, one can deduce the continuum threshold, \([29, 31]\) which might correspond to the recently observed high energy mode \([21]\).

In Eq. 2, the dynamical Stoner criterion, i.e. the continuum, a spin triplet \( 1 \)-collective mode is characterized in an itinerant system the continuum of spin flip particle-hole \( (p-h) \) excitations, given by the Lindhard function in the normal state or the BCS function in the SC state. In the SC state, due to the opening of the SC gap, the continuum becomes gaped (Fig. 3b) below a threshold energy at \( \omega_c = 2\Delta_m \), where \( \Delta_k \) is the momentum dependent superconducting \( d \)-wave energy gap and \( k_s \) is the so-called hot-spot wave vector defined as both \( k_s \) and \( k_s + Q_{AF} \) are lying on the Fermi surface. In addition to the \( p-h \) excitations within the continuum, a spin triplet \( p-h \) bound state can form below the continuum thank to the AF interaction.

In Eq. 2 the dynamical Stoner criterion, i.e. \( 1 + J(q)Re[\chi_0(q, \omega)] = 0 \), is then fulfilled for an energy smaller that the threshold of the continuum at wave vector \( q \). This spin \( 1 \)-collective mode is characterized by a downward dispersion controlled by the momentum dependences of the continuum threshold and the magnetic interaction (Fig 3b).

The mode vanishes when approaching the continuum by changing wave vector from \( (\pi/a, \pi/a) \). The model also predicted an excitation dispersing upward within the continuum \([29, 31]\) which might correspond to the recently observed high energy mode \([21]\).

From experimental point of view, a major challenge for future INS experiments would be to observe the magnetic continuum. So far, one can deduce the continuum threshold, \( \omega_c \), from i) the measurement of the maximum of the SC gap, \( \Delta_m \) (determined by angle resolved photo-emission spectroscopy \([13]\), by the measurement of the \( B_{1g} \) mode in Raman scattering \([34]\) or by tunneling data \([14]\), see Fig. 3) and ii) from the Fermi surface topology \([35]\). At optimal doping in BSCO, one can show that the magnetic resonance peak lies well below the continuum: \( \Delta_m \approx 35 \text{ meV}, \omega_c \approx 1.8\Delta_m \) and \( E_T \approx 1.2\Delta_m \). From optimal doping to the overdoped regime, one may expect \( \omega_c \) to reach the limit \( \sim 2\Delta_m \). Simultaneously, the ratio \( 2\Delta_m/k_B T_c \) evolves from 7-8 to 5-6, while according to INS data, the ratio \( E_T/k_B T_c \) seems to be preserved (Fig. 2). These evolutions suggest that in the overdoped regime the binding energy of the spin exciton, \( \omega_c - E_T \), weakens, leading to the possible disappearance of the spin \( 1 \)-collective mode. Further INS experiments, in the deeply overdoped regime are required to test such a possibility.

For a bilayer system \([26, 27, 36]\), the interlayer AF coupling, \( J_\perp \), is treated in perturbation, such that the odd \( (o) \) and even \( (e) \) spin susceptibilities are given by:

\[
\chi_o(q, \omega) = \frac{\chi(q, \omega)}{1 - J_\perp \chi(q, \omega)} \quad \text{and} \quad \chi_e(q, \omega) = \frac{\chi(q, \omega)}{1 + J_\perp \chi(q, \omega)}.
\]
In Fig. 4, we calculated these susceptibilities with a realistic value of $J_{\perp}$ ($\sim 0.1$ J) \cite{27} and with other parameters identical to those of ref. \cite{31}. Because of $J_{\perp}$, the odd spin exciton is likely pushed to lower energy, while the even one merges into the continuum. Actually, this explains why the odd channel mode is mainly observed as well as why the resonant mode appears to shift to lower energy in a bilayer system in regard with a mono-layer system. Going further, their respective spectral weights $W_{o,e}$ are predicted to be approximately proportional to their binding energies $W_{o,e} \sim \omega_r - E_{r,e}^0$. This is found in the calculated susceptibilities in Fig. 4 and sketched in the figure inset. Using this property in overdoped YBCO-Ca\cite{23}, one could directly estimate of continuum threshold at $\omega_r(Q_{AF}) \approx 49$ meV.

If the spin exciton scenario provides a plausible explanation for the resonance peak around the optimal doping, more theoretical work is needed to fully account for its evolution as a function of hole doping. In particular, there is no explicit relationship between the energy of the collective mode at $(\pi/a, \pi/a)$ and the value of $T_c$. The phenomenological relationship $E_r/k_B T_c \approx 5-6$ remains to be explained. Furthermore, the same model must simultaneously describe the unusual features observed below $T_c$ as well as the spin excitation spectrum of the normal state. In the underdoped regime, the magnitude and the energy dependence of spin fluctuations observed by INS are still difficult to reproduce quantitatively. Thus, most likely, the model needs to go beyond a purely itinerant picture in order to capture the deep underdoped state.

However, the spin-exciton interpretation for the magnetic neutron resonance and its downward dispersion is not unique. Indeed, besides such an approach the spin 1-collective mode corresponds to a $\pi$-excitation\cite{42} as the SO(5) model (spin itinerant picture) or a magnon-like excitation (spin localized picture) of a disordered stripe phase.

The SO(5) model\cite{38} considers as a starting point that, in the AF insulating state of cuprates, there exists a super-symmetry that allows the system to switch from the AF state to the d-wave SC state. This symmetry involves the existence of a Goldstone mode, the $\pi$-excitation\cite{39}. This excitation can be depicted as an excitation that creates a $p-p$ pair carrying a charge $2e$, a spin $S=1$ and with total momentum $(\pi/a, \pi/a)$. Upon doping the symmetry is broken, the $\pi$-excitation survives, but becomes massive. While it exists already in the normal state, it can only be observed by INS in the SC state due to the $p-h$ mixing. Its characteristic energy is roughly linear with hole doping and its intensity in the SC state scales with $|\Delta_m|^2$. Moreover, a downward dispersion is obtained due to by the phase slips of the SC order parameter induced by the propagation of the $\pi$-excitation\cite{40}. Such an approach accounts for several features observed by INS, especially in the underdoped regime, where $T_c$ and $E_r$ increase with hole doping. The $\pi$-excitation should decrease in magnitude, but remains almost at the same energy, as observed experimentally. However this scenario cannot explain the decrease of $E_r$ in the overdoped regime. Furthermore, recent calculations have shown that, if the $\pi$-excitation existed, it should be observed at high energy, above $2\Delta_m$\cite{41}: this casts some doubt about the interpretation of the resonance as a $\pi$-triplet excitation.

Alternatively, the stripe model considers that in a S=1/2 AF Heisenberg system, doped holes segregates to form lines of charges, separating AF domains in anti-phase. The metallic state is viewed as a disordered stripe phase, where charged lines can fluctuate. While there is not a general interpretation of the magnetic resonance peak in stripe models, it has been for instance proposed that the resonance peak and the incommensurate spin fluctuations observed by INS in the SC state could be viewed as magnon-like excitations reminiscent of the ordered stripe phase\cite{42} or could correspond to the eigen magnetic modes of the liquid stripe phase\cite{43}. Magnons, developing symmetrically around the magnetic incommensurate wave-vector, $Q_{mag}$, of the stripe ordered phase and merging at $(\pi/a, \pi/a)$, actually describe correctly the spin dynamics observed in stripe-ordered nickelates\cite{45} as predicted in the spin-only model\cite{42} \cite{43}. In cuprates, the lack of symmetric peaks around the incommensurate wave-vector $Q_{mag}$ (see Fig. 5) does not seem to validate these approaches. In addition, this model, if interesting, fails to explain why the resonance peak and the incommensurate spin fluctuations exist basically only in the SC state. Independently, it could be also interesting to understand how, in bilayer compounds, the adjacent CuO$_2$ planes succeed in accommodating the Coulomb repulsion between charged lines and the AF interlayer coupling. This is a central issue to account for as the magnetic resonance peak exists mostly in the odd channel.
Finally, INS experiments have shown the existence of an unusual enhancement of spin fluctuations in the SC state around the vector $(\pi/a, \pi/a)$ and at an energy $E_c$ which is found experimentally to scale with $T_c$. Combined with the observation at lower energy of incommensurate spin fluctuations, that develop also below $T_c$, INS data point toward the existence of a dispersive spin 1-collective mode deep inside the SC state. The observation of that mode, first discovered in YBa$_2$Cu$_3$O$_7$, has been then extended to other systems with one or two CuO$_2$ planes per unit cell, such as Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and Tl$_2$Ba$_2$CuO$_{6+\delta}$. This establishes the magnetic resonance peak as a generic excitation of the SC state of cuprates whose maximum $T_c$ can be as high as 90 K. In the strongly under- and overdoped regimes ($T_c \leq 50$ K) or in other cuprate families with lower maximum $T_c$ (such as La$_{2-x}$Sr$_x$CuO$_4$), the observation of the the spin 1-collective mode (if any) still requires more experimental work. In any case, the observation of such an excitation, thank to inelastic neutron scattering, is one of the most persuasive experimental indication of the crucial role of magnetic interactions in for the physics of High-$T_c$ copper oxides.

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