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Resonant spin excitations in $YBa_2Cu_3O_{6+x}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$

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Abstract

A summary of some recent results of neutron scattering studies of high temperature superconductors is given, with a focus on resonant spin excitations in the superconducting states of $YBa_2Cu_3O_{6+x}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$. The opportunities offered by advances in neutron scattering instrumentation, such as focusing and polarization techniques, for these experiments are discussed.

Keywords: A. Superconductors; C. Neutron scattering; D. Magnetic properties; D. Electronic structure

1. Introduction

Inelastic neutron scattering provides incisive information about collective magnetic excitations that is indispensable for the development of theories of high temperature superconductivity in the cuprates. The first inelastic neutron scattering measurements on the layered cuprates were reported in 1987 for $La_{2-x}Sr_xCuO_4$ [1] and in 1988 for $YBa_2Cu_3O_{6+x}$ [2]. These measurements have been greatly refined and extended over the past decade, but no inelastic magnetic neutron scattering experiments on the many other families of high temperature superconductors have been reported since then. As the data on $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ differ in many respects, this situation is unsatisfactory. In particular, the magnetic resonance peak, one of the most striking phenomena discovered by neutron scattering in these materials [3–10] and the subject of numerous recent theoretical studies [11–22], has only been observed in one of these two materials ($YBa_2Cu_3O_{6+x}$).

Several circumstances have conspired to impede progress on this front. First, because of the chemical complexity of these materials, it is difficult to grow large single crystals that are necessary for the neutron experiments. These difficulties are exacerbated by the weakness of the magnetic cross section, for even in the antiferromagnetically long range ordered precursor compounds the magnetic copper atoms only carry a spin-$\frac{1}{2}$. In the metallic regime, the spin excitations are broadened and the amplitude of the scattering cross section is further reduced. Finally, the large chemical unit cells of these materials give rise to complicated phonon spectra. Even a relatively simple cuprate such as $YBa_2Cu_3O_7$ contains 13 atoms per unit cell, and the separation of the magnetic excitations from the 39 phonon branches is challenging. Here we summarize some of the results of neutron scattering work on $YBa_2Cu_3O_{6+x}$ over the past few years, with a focus on resonant spin excitations observed in the superconducting state of this material. Opportunities provided in the coming years by new instrumentation, in particular by advances in focusing techniques and polarization analysis, will also be outlined. Focusing techniques have recently allowed our group to detect magnetic excitations in a single crystal of optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ ($T_c = 91$ K).

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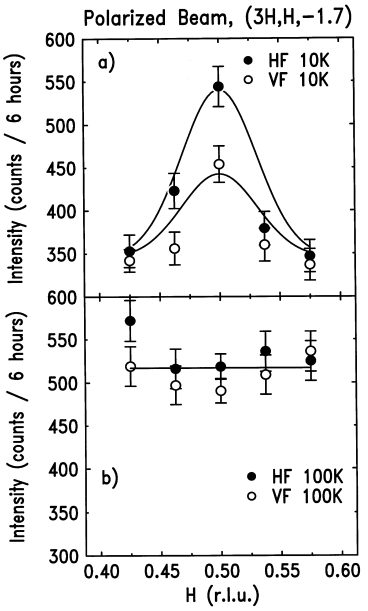

Fig. 1. Constant-energy scans at excitation energy 41 meV, of the magnetic (spin-flip) neutron scattering intensity as a function of the wave vector parallel to the CuO_2 planes, at temperatures (a) 10 K ($<T_c$) and (b) 100 K ($>T_c$) for $YBa_2Cu_3O_7$ [7]. The data were taken in the direction $\mathbf{Q} = (3H, H, -1.7)$, with the wave vector $\mathbf{Q} = (H,K,L)$ given in reciprocal lattice units (r.l.u.), that is, in units of the reciprocal lattice vectors $a^* \sim b^* = 1.62\ \text{Å}^{-1}$ and $c^* = 0.57\ \text{Å}^{-1}$. In these units, $Q_\parallel = (1.5, 0.5)$ is equivalent to the antiferromagnetic ordering wave vector $(\pi/a,\pi/a)$. The wave vector component perpendicular to the CuO_2 planes is chosen as described in Ref. [7]. The magnetic cross section is proportional to the difference between the intensities measured with horizontal (HF) and vertical (VF) guide fields at the sample. No magnetic intensity is detected above T_c to within the experimental error.

2. Experimental results

Arguably the best method of separating lattice vibrational and magnetic excitations is polarization analysis. The time-honored technique involves polarizing (typically Heusler alloy) crystals as both monochromator and analyser. The magnetic cross section is determined by subtracting the neutron intensity measured with a horizontal guide field at the sample position (parallel to the momentum transfer Q) from that measured with a vertical guide field (perpendicular to Q). Fig. 1 shows polarized beam data [7] taken in this way on a $\sim$10 cm^3 single crystal of $YBa_2Cu_3O_7$. Clearly, a magnetic excitation is present at an energy of 40 meV and a reduced wave vector $\mathbf{q} = (\pi/a, \pi/a)$ (where a is the nearest-neighbor Cu–Cu distance in the CuO_2 planes) in the superconducting state, and it disappears in the normal state. Analogous data [7] show that the excitation is sharply peaked also as a function of energy. It has hence been termed "magnetic resonance peak". The data of Fig. 1 also show that it is impractical at this time to use polarized-beam

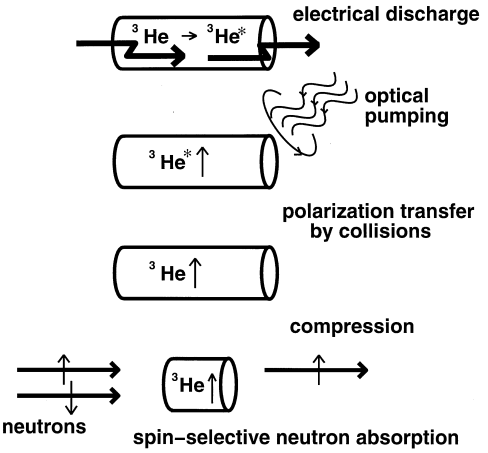

Fig. 2. Sketch of the ^{3}He polarizer, following Ref. [23]. Some of the ^{3}He atoms are promoted into the excited state ^{3}He* by electrical discharge. Atoms in the excited state are optically pumped by circularly polarized light into a state with a net magnetization. The magnetized ^{3}He* atoms then transfer their polarization to the remaining ^{3}He atoms in collisions. The gas is subsequently compressed, and the neutron beam is polarized in transmission by spin selective absorption, according to the reaction $^3\text{He} + \text{n} \rightarrow {}^4\text{He}^* \rightarrow {}^3\text{H} + \text{p}$.

measurements as an exploratory tool to determine the magnetic excitation spectra of new cuprate systems, as the counting time required to obtain adequate statistics was six hours per point even for a very large single crystal. Such crystals are not available for most cuprates. Fig. 2 shows a sketch of a new polarization technique currently under development at the Institut Laue Langevin in Grenoble, France [23]. Its operating principle is the spin-selective neutron absorption by a polarized gas of ^{3}He via the nuclear reaction $^3\text{He} + \text{n} \rightarrow {}^4\text{He}^* \rightarrow {}^3\text{H} + \text{p}$, which takes place only when the ^{3}He and neutron spins are antiparallel. This

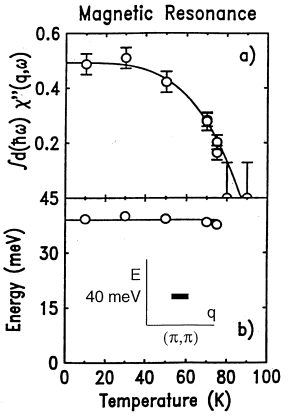

Fig. 3. (a) Absolute spectral weight and (b) energy of the resonance peak as a function of temperature. The inset is a sketch of the resonance peak intensity in an energy vs. momentum diagram.

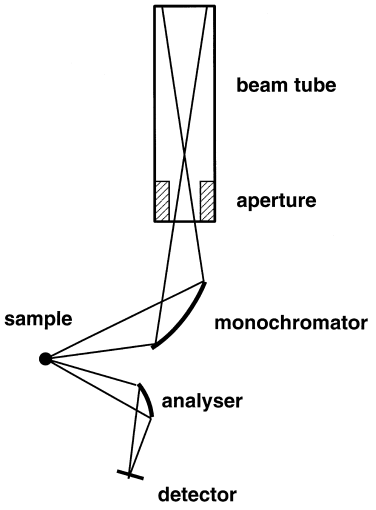

Fig. 4. Sketch of a triple axis spectrometer that focuses the neutron beam horizontally (in the scattering plane), in addition to the conventional vertical focusing, following Ref. [27].

method avoids the losses due the low reflectivity of the Heusler crystals as well as geometrical constraints associated with the need to magnetize these crystals. When it is fully implemented, it will offer new opportunities to use polarization analysis as a routine tool to investigate new and increasingly complex materials.

Since conventional polarization analysis is extremely time consuming, and large $YBa_2Cu_3O_7$ crystals were initially not available, a variety of approximate methods were used over the years to extract the magnetic cross section from unpolarized-beam data. The resonance peak was therefore first discovered in unpolarized-beam experiments [3]. (Simplified polarized-beam techniques were also used [4], but have their own pitfalls [7].) Fig. 3 gives a summary of the behavior of the magnetic resonance peak in optimally doped $YBa_2Cu_3O_7$, obtained from unpolarized-beam data [7]. Its intensity is sharply concentrated around a single point in an energy-momentum diagram. This in itself is highly unusual, because magnetic excitations in conventional metals typically form broad continua with amplitudes much too weak to be observable by inelastic neutron scattering. Further, the intensity decreases continuously with increasing temperature and vanishes above T_c. The mode has also been observed in underdoped $YBa_2Cu_3O_{6+x}$, and it was found that its energy decreases monotonically with decreasing hole concentration [8–10].

The fact that such a collective mode has not been observed in conventional superconductors is partly explained by the so-called coherence factor which multiplies its cross section in the superconducting state. It can be shown that the coherence factor is nonzero only in superconductors with a sign change of the gap function on the Fermi surface [5]. Neutron spectroscopy is thus a *phase-sensitive* probe of superconductivity. However, even with the *d*-wave coherence factor, a simple band model with noninteracting electrons can neither account for the sharpness of resonance peak in energy and momentum [11] nor for its spectral weight which has now been experimentally determined in abolute units [7]. Although band structure effects may play some role [12–14], it has become apparent that these features can only be described by models that involve electron–electron interactions at an essential level. There are various different models that attempt to accomplish this. Many of these begin with the band susceptibility appropriate for $YBa_2Cu_3O_{6+x}$ and incorporate electron correlations at the mean field level (akin to the Stoner model of ferromagnetism). Below the superconducting transition temperature, the renormalized susceptibility can develop a bound state that corresponds to the resonance peak [11–18]. Others begin by the spin wave excitations of an antiferromagnetic insulator and hypothesize that these are overdamped in the normal state through interaction with fermionic quasiparticles. In such models, the resonance peak corresponds to an undamped spin wave that becomes visible as decay channels are removed below the superconducting energy gap [19]. There are also models that invoke radically new physics such as interlayer pair tunneling [20] and a new SO(5) symmetry group linking antiferromagnetism and superconductivity [21,22].

In the light of its theoretical importance, it is imperative to establish experimentally that the resonance peak is a generic feature of the high-T_c superconductors by demonstrating its existence in other cuprates systems. Among the many unexplored cuprate families, $Bi_2Sr_2CaCu_2O_{8+\delta}$ is particularly interesting, because the excellent surface quality of single crystals of this material has allowed experimentalists using surface sensitive techniques such as angle resolved photoemission spectroscopy (ARPES) to collect a wealth of detailed, high quality data on the Fermi surface and band dispersions (see, e.g. Refs. [24,25]). In the materials that have been investigated by inelastic neutron scattering, ARPES data have been much more controversial. It appears that this situation is a consequence of the basic lattice structure of the cuprates: The most robust structural unit, common to all high temperature superconductors, is the CuO_2 layer. Materials in which the effective bonding between successive layers is weak can be cleaved easily and give excellent surfaces; this same feature, however, results in a platelike crystal growth habit and has until now prevented the formation of large, bulk single crystals required for inelastic neutron scattering.

Very recently, a $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal of volume 0.08 cm^3 was successfully synthesized [26]. Given the weak magnetic cross section, this crystal is still too small to perform inelastic magnetic neutron scattering at a conventional triple axis spectrometer within a reasonable time period. The experiment proved feasible only by using horizontally as well as vertically focusing neutron optics recently developed at the Laboratoire Léon Brillouin [27], and implemented both on the 2T spectrometer there and on

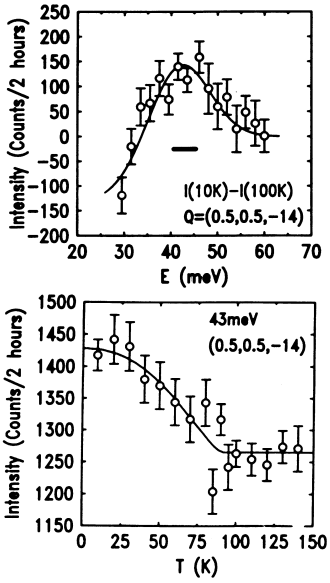

Fig. 5. (Upper panel) difference spectrum of the neutron intensities at $T = 10$ K ($<T_c$) and $T = 100$ K ($>T_c$) in $Bi_2Sr_2CaCu_2O_{8+\delta}$, at wave vector $\mathbf{Q} = (0.5, 0.5, -14)$. The bar respresents the instrumental energy resolution, the line is a guide-to-the-eye. (Lower panel) temperature dependence of the neutron intensity at energy 43 meV and wave vector $\mathbf{Q} = (0.5, 0.5, -14)$. The intensity falls to background level above $T_c = 91$ K, as revealed by **Q**-scans at this temperature. The line is a guide-to-the-eye. From Ref. [28].

the IN8 spectrometer at the Institut Laue–Langevin. A sketch of these spectrometers is given in Fig. 4, and Fig. 5 shows representative data collected on $Bi_2Sr_2CaCu_2O_{8+\delta}$ [28]. Although somewhat broader than in $YBa_2Cu_3O_7$ (where the response is resolution limited in energy), the data clearly demonstrate the existence of a resonance peak in the superconducting state of this material as well. These observations will, for the first time, allow a simultaneous description of the single-particle spectral function as measured by ARPES and the collective spin dynamics as measured by neutron scattering in the *same* material. There are in fact theoretical models according to which the resonance peak is responsible for some of the unusual features of the ARPES spectra of $Bi_2Sr_2CaCu_2O_{8+\delta}$ [29,30]. Given the very different crystal structures of $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$, this observation also rules out scenarios in which the resonance peak is brought about by a delicate combination of microscopic parameters that is only found in one specific material. Rather, it is a robust feature of the cuprates, and its explanation must be an integral part of any theory of high temperature superconductivity.

There are two other possible reasons for the apparent *absence* of the resonance peak in the $La_{2-x}Sr_xCuO_4$ family. First, in both the $YBa_2Cu_3O_7$ and the $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystal structures, two CuO_2 layers are located in close proximity, and the neutron experiments show clear evidence of strong magnetic interactions between directly adjacent

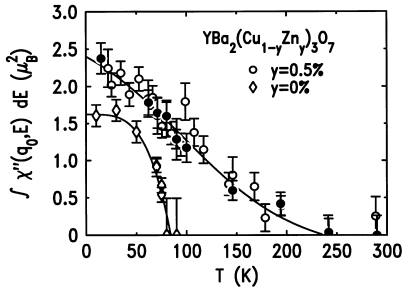

Fig. 6. Absolute spectral weight of the magnetic resonance peak in pure $YBa_2Cu_3O_7$ (Fig. 3), and of the broadened spectral distribution in $YBa_2(Cu_{0.995}Zn_{0.005})_3O_7$, following Ref. [31].

layers. If strong interlayer interactions are essential for the resonance peak [18], its absence in $La_{2-x}Sr_xCuO_4$ may be a consequence of the much larger layer spacing in this material. Alternatively, the resonance peak may be suppressed by the random potential of the Sr acceptor ions that is known to significantly affect other physical properties of $La_{2-x}Sr_xCuO_4$.

We have recently investigated the effect of a dilute concentration of nonmagnetic impurities on the magnetic resonance peak in a controlled fashion by substituting 0.5% of the magnetic copper ions in $YBa_2Cu_3O_7$ by Zn [31]. The Zn^{2+} ion has a full *d*-shell and therefore does not carry a magnetic moment. In ordinary superconductors, nonmagnetic impurities have only weak effects because the electrons within a Cooper pair have opposite spins; pair breaking therefore requires magnetic impurities. Surprisingly, the effects of Zn substitution on the superconducting transition temperature of the cuprates is comparable or larger than that of many magnetic impurities. NMR and magnetization experiments have revealed a local staggered magnetization induced on copper ions that surround the Zn impurity (see e.g. Ref. [32]), so that Zn cannot be regarded as purely nonmagnetic in the materials. Previous neutron scattering experiments for Zn concentrations of 2% and larger [33–35] had revealed low energy magnetic fluctuations associated with the impurities. However, a very dilute concentration of 0.5% has only a small effect on the transport properties. For instance, the superconducting transition temperature T_c is only lowered by a few degrees ($T_c = 87$ K for $YBa_2(Cu_{0.995}Zn_{0.005})_3O_7$, compared to $T_c = 93$ K for pure $YBa_2Cu_3O_7$). By contrast, the effect on the magnetic excitations is dramatic [31]. The magnetic response, which is resolution-limited in the pure system, is broadened to about 8 meV in the Zn-substituted system, though it remains centered around 40 meV. (At this level of impurity concentration, no low energy fluctuations could be detected.) Even more surprisingly, a large fraction (about 50%) of the magnetic intensity in $YBa_2(Cu_{0.995}Zn_{0.005})_3O_7$ persists above T_c, while in the pure system no magnetic intensity is detectable in the normal state (Fig. 6). The extreme sensitivity of the resonance peak to Zn substitution hints at a

hitherto unappreciated delicate coherence that has not been anticipated by any of the models of the resonance peak [11–22]. It is worth mentioning, however, that such dramatic effects of nominally nonmagnetic impurities are well known in collective singlet states of *one-dimensional* quantum magnets such as $CuGeO_3$ (for a review see Ref. [36]). It may be possible to find common theoretical ground between these one-dimensional systems and the two-dimensional cuprates [37].

3. Conclusion

The strikingly simple phenomenology of the magnetic resonance peak in this seemingly very complex system leaves much room for further theoretical and experimental work. From a theoretical perspective, the neutron data, especially when combined with photoemission data on the same material, now present a remarkably detailed, microscopic picture of these materials that poses a fundamental challenge to our theoretical understanding of metals. On the experimental side, it is likely that new experimental techniques such as double-focusing spectrometers and ^{3}He polarizers will make a variety of other cuprate systems accessible to neutron spectroscopy in the coming years.

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