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Current Perspectives

Superconductivity, antiferromagnetism, and neutron scattering

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ABSTRACT

High-temperature superconductivity in both the copper-oxide and the iron–pnictide/chalcogenide systems occurs in close proximity to antiferromagnetically ordered states. Neutron scattering has been an essential technique for characterizing the spin correlations in the antiferromagnetic phases and for demonstrating how the spin fluctuations persist in the superconductors. While the nature of the spin correlations in the superconductors remains controversial, the neutron scattering measurements of magnetic excitations over broad ranges of energy and momentum transfers provide important constraints on the theoretical options. We present an overview of the neutron scattering work on high-temperature superconductors and discuss some of the outstanding issues.

1. Introduction

Looking at temperature vs. composition phase diagrams of high-temperature superconductors, including both cuprate and iron–pnictide/chalcogenide systems, one finds antiferromagnetic (AF) order in close proximity to superconductivity [1–3]. This close association, together with experimental evidence for AF correlations in superconducting samples, has led many theorists to a common belief that AF fluctuations play an important role in the electron-pairing mechanism that underlies superconductivity [3–5]. The differences among theoretical perspectives only begin to appear when one considers the specifics of how the AF correlations impact pairing. To understand the source of these disagreements, one must step back and recognize that there is no consensus on how to describe the interaction between charge carriers and spin excitations in a metallic conducting system with strong AF correlations, especially when the system is close to a transition to a correlated (Mott) insulator state. In the absence of a common view on how to frame the problem, it should not be surprising that there is a lack of consensus on the approach to a solution.

Neutron scattering is the preeminent technique for studying AF spin fluctuations in solids. One can, of course, also obtain important information on local hyperfine fields, susceptibilities, and slow fluctuations from techniques such as nuclear magnetic resonance (NMR) and muon spin rotation, and magnetic order can be probed with resonant X-ray scattering. The defining feature of AF fluctuations in high-temperature superconductors, however, is their remarkably high energy scale. Magnetic excitations in these systems extend up to energies of several hundreds of meV, which easily exceeds the maximum energy of phonon excitations involved in the traditional mechanisms of superconductivity. This observation makes highly energetic AF fluctuations a primary suspect for mediating the unconventional high-temperature superconductivity.

To characterize the AF correlations in these and other strongly correlated metallic systems, it is therefore crucial to cover the energy range from fractions to hundreds of meV and to probe all of reciprocal space. Developments in spectrometers and sources over the last two decades have greatly improved the efficiency of such measurements. The throughput of triple-axis spectrometers has been enhanced through the use of multiple analyzers and detectors, beginning with developments such as RITA [6] (first at Risø and continuing at SINQ) and SPINS [7] and progressing to MACS [8] and BT7 [9] at the NIST Center for Neutron Research; of course, enhanced instruments are available at many other facilities, including the Institut Laue Langevin, FRM-II, and the Laboratoire Léon Brillouin. While most triple-axis instruments are optimized for excitations in the cold to thermal neutron range ($E \lesssim 30$ meV) time-of-flight spectrometers at spallation sources have enabled measurements with epithermal neutrons probing excitations up to ~ 1 eV, and covering a very large phase space. This began with the seminal MAPS spectrometer [10] (and later MERLIN [11]) at ISIS, and has become common place with instruments such as ARCS [12] and SEQUOIA [13] at the Spallation Neutron Source (SNS) and 4SEASONS [14] at the Japanese Proton Accelerator Research Complex. Very soon, the SNS spectrometer HYSPEC will have the capability of distinguishing magnetic from nuclear scattering through neutron-spin polarization analysis [15], a technique that has long been used to advantage on triple-axis instruments [16].

Ideally, experimentalists should be able to interpret their results by comparing with existing theoretical expressions or to simply measure the magnetic response in a superconductor family

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as a function of doping and temperature, and then let the theorists interpret the results. In practice, however, things are not so simple. The neutron scattering cross section is relatively weak, so that large crystals ($\gtrsim 1\ \mathrm{cm}^3$) or assemblies of crystals are typically required for each composition to be studied. In some cases, it has taken decades for crystal growth technology and know-how to evolve to the point that suitable crystals are available. Another aspect involves interpretation of the measurements. Frequently, there is a disconnect between theoretical interpretations and experimental results arising from various assumptions and idealizations adopted both in analyzing the data and in applying the theoretical model. To present results in a useful fashion, it is generally necessary to parameterize the data by fitting with a model. The models used, or the words used to describe them, are often based on specific theoretical perspectives. Without a general and accepted theoretical description of itinerant antiferromagnetism, there is no universally accepted and unbiased language for describing measurements of spin fluctuations in high-temperature superconductors.

In the following sections, we will briefly summarize what we consider to be the important results from neutron scattering studies on antiferromagnetism in hole-doped cuprates and in iron-based superconductors. We will also point to some of the open questions. Obviously, our choices and presentation reflect our own biases. The coverage of experiments and theory is necessarily incomplete, and we refer the interested reader to more extensive review articles on cuprates [4,17–27] and iron-based systems [2,28–32].

2. Notes on neutron scattering

Neutron scattering measures the product of the dynamical spin structure factor $\mathcal{S}(\mathbf{Q},\omega)$, which is the Fourier transform of the spin–spin correlation function, and the square of the magnetic form factor $F(\mathbf{Q})$, which is the Fourier transform of the density of the electronic magnetization cloud associated with each spin, normalized to one at $\mathbf{Q}=0$. Here $\hbar\mathbf{Q}$ is the neutron momentum transfer and $-\hbar\omega$ is the energy transfer. The dynamical structure factor describes the cooperative behavior of electronic spin variables, including spin order and excitations, whereas the form factor relates it to the behavior of the magnetization density in the crystal. This latter is the quantity which interacts with neutron's magnetic moment and is probed in experiment. The magnetic form factor is determined by the Wannier functions of magnetic electrons, which could be obtained from first-principles calculations. A prediction for the dynamical structure factor, $\mathcal{S}(\mathbf{Q},\omega)$, can in principle be obtained from the theoretical analysis of the model spin Hamiltonian that describes the system. For an ordered antiferromagnet, the excitations are spin waves that can be calculated in perturbation theory, where one assumes that the fluctuations are small compared to the ordered moment. As we will discuss, the fluctuations become more important when one reduces the size of the spin and the dimensionality. In the case of the weakly coupled planes with spin $S=1/2$ that occurs in cuprates, spin-wave theory is not a well-controlled approximation, though it remains a useful description.

$\mathcal{S}(\mathbf{Q},\omega)$ obeys a number of useful sum rules, which are helpful in analyzing the experimental neutron scattering data [7]. In particular, for a system of identical spins S, one has

$$C\int_{-\infty}^{\infty} d\omega \int_{\mathrm{BZ}} d\mathbf{Q}\ \mathcal{S}(\mathbf{Q},\omega) = S(S+1), \tag{1}$$

where $C=v_0/(2\pi)^3$ and v_0 is the unit cell volume. It is possible to convert $\mathcal{S}(\mathbf{Q},\omega)$ into the imaginary part of a generalized spin susceptibility with the formula

$$\chi''(\mathbf{Q},\omega)=\pi(1-e^{-\hbar\omega/kT})\mathcal{S}(\mathbf{Q},\omega). \tag{2}$$

By integrating $\chi''(\mathbf{Q},\omega)$ over $\mathbf{Q}$, one obtains the local susceptibility $\chi''(\omega)$.

3. Cuprates

3.1. Parent insulators

The parent materials of the cuprate superconductors are correlated insulators. Taking La_2CuO_4 as an example, it has a charge excitation gap of ~ 2 eV [18], so that the only low energy excitations involve spin fluctuations. These can be described by an effective spin Hamiltonian, where the nearest-neighbor Cu spins within the planes are coupled by a superexchange energy J that is quite large, while the effective coupling between planes is extremely weak, so that there are strong two-dimensional (2D) AF spin correlations at temperatures far above the magnetic ordering temperature T_N [33].

The Cu moments within the CuO_2 planes order antiferromagnetically below a Néel temperature $T_N=325$ K in stoichiometric La_2CuO_4 [34]. Neutron diffraction studies have shown that the ordered moment is quite small, $\approx 0.6\mu_B$ [35]. This corresponds to an ordered spin value of $\langle S\rangle \approx 0.27$ (assuming $g\approx 2.2$ for the spectroscopic Lande g-factor of Cu [36]), which is even smaller than the value of 0.30 predicted by spin-wave theory upon accounting for zero-point motion [37–39]. Similar results have been obtained for other cuprate antiferromagnets [22]. Consequently, the contribution of the static spin order, which is measured by elastic Bragg peaks (delta-functions in energy), $\langle S\rangle^2$, accounts for only $\sim 10\%$ of the total spin spectral weight, $S(S+1)=3/4$, in the sum rule for $S=1/2$. Hence, more than 90% of the spectrum is inelastic.

Fig. 1 shows a recent measurement of the spin excitations in antiferromagnetic La_2CuO_4 by Headings et al. [40]. The authors find that both the dispersion and the wave-vector dependence of the intensity are described rather well by spin-wave theory expressions. The absolute intensity, however, appears much lower than predicted by the linear spin-wave theory, requiring a downward renormalization by a factor of $Z_d=0.4\pm 0.04$. This is somewhat smaller than the value $Z_d\approx 0.6$ that is predicted from quantum corrections to linear spin waves [41].

A fit to the dispersion in La_2CuO_4 yields $J=143\pm 2$ meV, but also requires longer-range exchange couplings, second and third neighbor couplings J' and J'', which are relatively weak, and a significant 4-spin cyclic exchange term $J_c\approx 0.4J$. The overall bandwidth of the magnetic spectrum is $\sim 2J$. At the highest energies, there are some modest deviations in the spectral shape relative to the single-mode spectrum predicted by spin-wave theory. These seem to be consistent with corrections obtained in quantum Monte Carlo calculations [42]. Similar findings were recently reported for several other cuprates by Dalla Piazza et al. [43], where the spin excitation spectra can be well described by a perturbative (up to a second order) treatment of an effective 1-band Hubbard model with nearest- and next-nearest neighbor hopping, or by a spin-wave treatment of the local spin Heisenberg Hamiltonian with extended-range interactions J, J', J'' and J_c. Quantitative estimates for J require that one takes account of the bridging O atoms when evaluating the Cu to Cu hopping [44]. The 4-spin cyclic exchange can be obtained at the same order of approximation in such a multi-orbital model [45].

Evaluating the sum rule for La_2CuO_4 without any model assumptions for $\mathcal{S}(\mathbf{Q},\omega)$, the result is $\sim 60\%$ of the expected result