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Current Perspectives

Insights into the high temperature superconducting cuprates from resonant inelastic X-ray scattering

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ABSTRACT

Recent improvements in instrumentation have established resonant inelastic X-ray scattering (RIXS) as a valuable new probe of the magnetic excitations in the cuprates. This paper introduces RIXS, focusing on the Cu L_3 resonance, and reviews recent experiments using this technique. These are discussed in light of other experimental probes such as inelastic neutron scattering and Raman scattering. The success of these studies has motivated the development of several new RIXS spectrometers at synchrotrons around the world that promise, among other improvements, 5–10 times better energy resolution. We finish by outlining several key areas which hold promise for further important discoveries in this emerging field.

1. Introduction

Understanding high temperature superconductivity (HTS) in the cuprates has been one of the defining problems of condensed matter physics for the last quarter of a century [1]. At the core of the problem is the quest to characterize the nature of the ground state and the low energy excitations that define the normal state from which HTS emerges. This has driven the development of several spectroscopic techniques including angle-resolved photoemission (ARPES) [2] and scanning tunneling spectroscopy (STS) [3] as probes of electronic structure, and inelastic neutron scattering as a probe of magnetism [4]. Indeed, these techniques have provided numerous important insights into the physics of the cuprates, including the emergence of Fermi arc features in the electronic spectral function [3] and the resonance phenomenon in which the dynamical magnetic susceptibility changes through the superconducting transition [4]. In recent years, instrumentation for resonant inelastic X-ray scattering (RIXS) techniques with both soft and hard X-rays has also improved dramatically [5], allowing this technique to directly measure magnetic excitations in several materials such as the cuprates [6–8], nickelates [9], pnictides [10] and iridates [11–13].

This Current Perspectives paper describes recent experimental progress in soft X-ray RIXS studies of magnetic excitations in the cuprates with a particular focus on the doping dependence of the magnetic excitation spectrum and how this relates to superconductivity [8,14–19]. We start by outlining the RIXS technique and the basics of the cuprate phase diagram. This paper then describes some of the insights gained into the cuprates; first in the underdoped and optimally doped cases, before moving onto the overdoped case, including both the measurements themselves and also their relationship with other experimental probes such as inelastic neutron scattering and Raman scattering. We end by discussing upcoming improvements in RIXS instrumentation and outline some opportunities for future experiments on stripe-ordered cuprates and heterostructures, as well as extending RIXS to probing phonons.

1.1. Resonant inelastic X-ray scattering

Resonant inelastic X-ray scattering, abbreviated RIXS, is an X-ray spectroscopic technique in which one measures the change in energy and momentum of X-rays that scatter from a material. This technique has great potential for probing the low energy excitations in correlated electron systems such as the cuprates, as it can be used to study charge, magnetic, lattice and orbital excitations. Furthermore, it is element and orbital resolved, bulk sensitive and compatible with small samples [5]. Fig. 1 illustrates the RIXS process for the case that will form the focus of this paper: the L_3 resonance in the Cu $3d^9$ ion present in the cuprates. In the initial state, $|i\rangle$, there is one hole in the Cu $3d$ valence band, which is filled by the incident X-ray exciting a $2p_{3/2}$ core electron to form a highly energetic intermediate state, $|n\rangle$. Due to the strong spin–orbit coupling of the core hole, the orbital angular momentum of the photon can be exchanged with the spin angular momentum of the valence hole in order to create a spin flip excitation while conserving total (spin + orbital) angular momentum [20].

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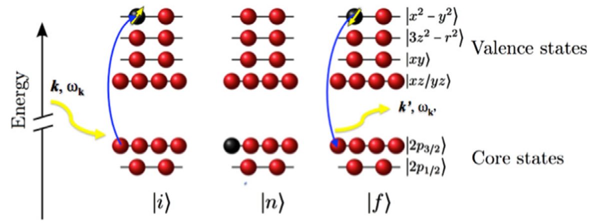

Fig. 1. A schematic of the direct RIXS process showing the case of the L_3-edge resonance for a Cu $3d^9$ ion at 931 eV. The initial $|i\rangle$, intermediate $|n\rangle$, and final $|f\rangle$, states are shown from left to right. Red spheres denote states filled by electrons and black spheres denote holes in up or down spin states. Incoming and outgoing photons are represented as wavy yellow lines and the blue arrows depict transitions. In this process spin excitations are created with energy, $\hbar\omega_{\mathbf{k}} - \hbar\omega_{\mathbf{k}'}$, and momentum, $\hbar\mathbf{q} = \hbar\mathbf{k} - \hbar\mathbf{k}'$.

The core hole is then filled to form the final state, $|f\rangle$, which contains a spin flip magnetic excitation distributed throughout the lattice. Such an experiment, in which the core electron is promoted into the valance band, and an electron from a *different* state fills the core hole, is called *direct* RIXS. This is distinct from indirect RIXS where an electron is excited into a high energy vacant state well above the chemical potential and an electron from the *same* state fills the core hole [21]. When direct RIXS is not forbidden it is the dominant process and it is this process that we focus on in this paper.[1] For many years, it was believed that single magnons could not be observed in L-edge RIXS studies of the cuprates without a simultaneous orbital excitation [23]. Only in 2009 it was demonstrated that single magnon excitations are only forbidden when the spin orientation is perpendicular to the CuO_2 planes [20]. Given that the spins in magnetically ordered cuprates lie parallel to the CuO_2 planes, this implied that given sufficient energy resolution and signal-to-noise ratio, these excitations should be experimentally accessible. The energy resolution for soft X-ray RIXS at 931 eV (the energy of the Cu L_3-edge resonance) has improved by over a factor of 10 from 1996 to 2008 to the current value of 130 meV at the Swiss Light Source [24,25]. This has made it possible to access spin flip excitations in the cuprates [8]. Fig. 2 plots RIXS measurements of the magnon dispersion of La_2CuO_4 taken from Ref. [8], which are in excellent agreement with a spin wave fit to inelastic neutron scattering (INS) measurements [26]. Shortly after the observation of magnons in La_2CuO_4, magnons were also observed in RIXS measurements of other square lattice antiferromagnetic insulating cuprates $Sr_2CuO_2Cl_2$ [27] and $Bi_2Sr_2YCu_2O_8$ [28]. Thus RIXS provides an alternative to INS for measuring the magnetic excitations in the cuprates.

INS is a very well established experimental technique and it can be performed with excellent energy resolution – well below 1 meV, although the best practical energy resolution is usually limited by count rate considerations [29]. Due to the weak interaction of the neutron with matter the INS cross section is relatively simple and well understood [29], which allows researchers to determine the magnetic dynamical structure factor, $S(\mathbf{Q}, \omega)$ in absolute units [30]. However, this weak interaction also means that neutrons travel several cm through matter before scattering and that large single crystals, several cm^3 in volume, are required for most INS experiments on cuprates.

In RIXS, on the other hand, the interactions are both stronger and more complicated and most theories for direct RIXS are based on operator treatments [23,31,21,20,32,33]. This strong interaction also means that the penetration depth of soft X-rays is of the order of 1000 Å – far shorter than that of thermal neutrons. This facilitates studies of small samples, indeed, heterostructures based on 1 unit cell thick cuprate layers can even be measured [15,34]. In point of fact, comparing the count rate, normalized to the probed volume, in state-of-the-art RIXS [15] and INS [26] one finds that RIXS is $\sim 10^{11}$ times more sensitive. This high sensitivity, even in small samples, is perhaps the key advantage of RIXS compared to INS, and it is this that has given Cu L_3-edge RIXS an important niche for measuring magnetic excitations, despite the relatively coarse energy resolution of the current RIXS spectrometers and the less well understood cross section.

In terms of measuring magnetic excitations it is important to note that Raman scattering can also be used to measure magnetic excitations – a field that is reviewed in, for example, Refs. [36,37]. In Raman scattering measurements on the cuprates, two magnon excitations appear most strongly (see for example Ref. [38]) through the spin exchange scattering mechanism [39]. Single magnon excitations are only very weakly allowed due to finite spin–orbit coupling in the valance band [39,40]. In Raman scattering, however, the incident light carries negligible momentum so it can only probe the Brillouin zone center. Cu L_3-edge RIXS has a less strict limitation: it can, in principle, probe magnetic excitations out to $(0.5, 0)$, though it cannot reach the antiferromagnetic ordering wavevector $(0.5, 0.5)$. Cu K-edge RIXS has also been used to measure multimagnon excitations with a total summed spin of zero [41,6,42], covering several Brillouin zones in reciprocal space. Cu M-edge [43,44] and O K-edge RIXS [45,46] can also detect multimagnon excitations although these have even more severe momentum restrictions than Cu L-edge RIXS.

1.2. Cuprates

The phase diagram of the cuprates is shown as a function of doping, x, in Fig. 3(a). Much of the physics of the cuprates, especially at low dopings, is dominated by a strong on-site Coulomb repulsion, U, between electrons, and many researchers believe that the three band Hubbard model, or simplified effective models based on this starting point, contains the physics required to describe the intrinsic properties of the normal state and the resulting HTS [47]. The parent compounds of the cuprates such as La_2CuO_4 have one hole per Cu site and the strong U leads to insulating behavior with a charge excitation gap of 2 eV [48]. These localized holes, of predominantly Cu character [49], order antiferromagnetically below 325 K. The resulting spin dynamics is well described in terms of spin wave, or magnon, excitations [35,26] within the 2 dimensional spin $\frac{1}{2}$ Heisenberg model on the square lattice. Indeed, the Heisenberg model can be derived from the Hubbard model at zero doping (also often referred to as half filling) [47]. At the other extreme, for $x \gtrsim 0.3$ the ground state and low energy electronic excitations have Landau Fermi liquid-like properties with resistivity that scales as T^2 and a fully connected Fermi surface [50]. The superconducting dome extends over $0.05 < x < 0.3$ where the superconducting state has a $d_{x^2-y^2}$ gap symmetry. Most of the complexity of the cuprates lies in the normal state from which HTS emerges.

At a doping of $x = 0.03$ antiferromagnetic order gives way to the pseudogap phase [51–54,2,1]. This phase is associated with a partial reduction of the electronic density of states as the sample is cooled from high temperature through the pseudogap temperature. The electronic spectral function in this state has been extensively measured by ARPES [2] which observes arcs of photoemission intensity, with strong spectral weight along the nodal $((0,0) \rightarrow (0.5, 0.5))$ direction and vanishing intensity along the antinodal $((0,0) \rightarrow (0.5, 0))$ direction. The origin of the pseudogap remains controversial, some popular interpretations are that it arises from preformed Cooper pairs that have not gained phase coherence [55], or that it comes from a competing ordering tendency such as charge/spin density waves [56,16] or loop

[1] The reader is referred to Refs. [22,5] for a discussion of indirect RIXS.