12 Superconductivity and Magnetism

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We report on research projects in the field of high-temperature superconductors (HTS) and materials with novel electronic properties. Our studies involve various complementary techniques, such as x-ray diffraction (XRD), Resonant Inelastic x-ray Scattering (RIXS), and angle resolved photoemission spectroscopy (ARPES). Here we present some results from our recent investigations on cuprate high-temperature superconductors and ruthenate Mott insulators.

12.1 High-energy spin excitations in overdoped La_2CuO_4

Conventional superconductivity emerges as a result of electron-phonon interaction. Information about the phonon excitation spectrum (dispersions and lifetime effects) are therefore of great importance. Similarly, for magnetic superconductors [1], there is a strong interest in understanding and experimentally revealing the spin excitation spectrum. Mapping out the detailed evolution of the spin excitation spectrum across the high-temperature superconducting cuprate phase diagram, from the Mott insulator to the Fermi-liquid ground state, is hence important. Spin excitations have traditionally been studied by inelastic neutron scattering (INS). Studies of high-energy spin excitations have, however, been challenged by weak neutron cross sections. Over the last decade, resonant inelastic x-ray scattering (RIXS) has developed rapidly [2] and energy resolution now allows studies of spin excitations. RIXS is therefore an attractive complementary technique to neutron scattering. This has, in particular, lead to progress in understanding correlated low-dimensional $3d$ and $5d$ electron systems [3]. The spin excitation spectra of insulating one- and two-dimensional cuprates have, for example, been studied by soft x-ray RIXS using the copper L_3-edge. In recent years, spin excitations of doped cuprate and pnictide superconductors have also been investigated. These studies suggest that the high energy ($\omega > 100$ meV) spin excitation dispersion undergoes little change with doping [3]. This is in strong contrast to the low-energy part of the spectrum (studied by INS), that has a strong dependence on impurities, magnetic field and doping.

We have published [4] a systematic RIXS study of the spin and charge excitations found in overdoped $La_{2-x}Sr_xCuO_4$ (LSCO) $x = 0.23$. Examples of measured spectra are shown in Fig. 12.1. The line shape of the excitations in the spectra is analyzed using the response function of a damped harmonic oscillator. In this fashion, their dispersion and momentum dependence of the damping, γ, are extracted. Compared to previous RIXS reports [3], we obtain new information about these excitations along different high symmetry directions. We find that the spectral weight and damping γ are anisotropic in momentum space. The line shape is sharpest around the zone center. As reported for Bi-based cuprates [5], we also find a strong nodal / antinodal anisotropy of spectral weight. These observations are captured by susceptibility calculations based on the electronic band structure. The model calculation furthermore predicts a low-energy spin excitation branch, along the (π,π)-direction, which turns out to be particularly pronounced and dispersive in LSCO with $x = 0.23$ in comparison to other doped cuprates [5]. Future RIXS experiments with improved resolution should test this prediction.

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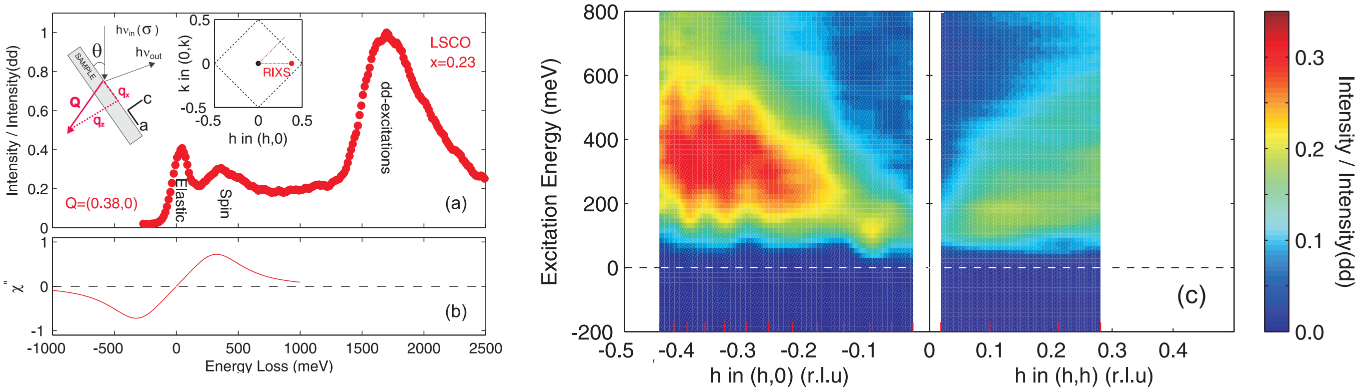

Fig. 12.1 – (a) RIXS spectrum, recorded on overdoped LSCO $x = 0.23$ using $\sigma-$polarized light, displaying elastic scattering, a low-energy excitation and a dd-excitation. The inset shows the scattering geometry and reciprocal space $(h,k,0)$ schematically. (b) Overdamped response function showing how $\chi'' \to 0$ for $\omega \to 0$. (c) Interpolated RIXS intensity, with elastic scattering subtracted, versus momentum $q = (h,0)$, (h,h) and photon energy loss ω. Red ticks indicate the grid of spectra used for the interpolation.

12.2 Magnetic field controlled charge density wave coupling in underdoped $YBa_2Cu_3O_{6+x}$

Charge density wave (CDW) correlations, that is, periodic modulations of the electronic charge density accompanied by a periodic distortion of the atomic lattice, have long been known to exist in underdoped La-based cuprate high-temperature superconductors [6]. More recently, it has been found that charge order is a universal property of underdoped high-temperature cuprate superconductors [7,8]. CDW correlations appear typically at temperatures well above the supraconducting transition temperature T_c. Cooling through T_c suppresses the CDW and leads to a state in which the superconducting and CDW order parameters are intertwined and competing [11].

The application of magnetic fields suppresses superconductivity. In the case of underdoped YBCO, a number of changes in electronic properties have been reported in the field range $B \approx 10 - 20$ T. For example, new splittings occur in nuclear magnetic resonance (NMR) spectra [9], ultrasound shows anomalies in the elastic constants [10] and the thermal Hall effect suggests that there is an electronic reconstruction. At larger fields, $B \gtrsim 25$ T a normal state with quantum oscillations (QO) [12] and coherent transport along the c-axis is observed. The existence of QO, combined with a high-field negative Hall and Seebeck effect, is most easily understood in terms of electron pockets.

Magnetic fields with $B \approx 10 - 20$ T also cause changes in the CDW order which can be seen by x-ray measurements. Initial experiments [14] showed that a magnetic field causes an enhancement of the diffuse CDW scattering [13, 14]. A recent x-ray free-electron-laser experiment [15] has shown that a magnetic field of $B \gtrsim 15$ T induces a new CDW Bragg peak, with a propagation vector along the b-axis, corresponding to an extended range of ordering along the c-axis and an in-phase correlation of the CDW modulation between neighbouring bilayers.

It is important to determine the nature of the CDW correlations induced by the magnetic field in YBCO and their relationship to the electronic properties. Of particular interest are the high-field CDW phase diagram and whether a field also induces a new CDW order propagating along the a-axis.

We have therefore used hard x-ray scattering measurements to determine the the evolution of the CDW correlations with magnetic fields up to 16.9 T for several doping levels [16]. We investigated the CDW for propagation vectors along the crystallographic a- and b-directions, allowing us to extend the pulsed-field measurements [15] and identify new field-induced anisotropies in the CDW. By measuring the profile of the diffuse CDW scattering as a function of field we show that the CDW inter-bilayer coupling along the c-axis is strongly field dependent. We also show that field-induced changes in the CDW can be associated with many of the anomalies observed in electronic properties. In particular, the $B - T$ phase diagram has two boundary lines associated with the formation of high-field CDW order (see Fig. 12.2). Our data also provides insight into the likely high-field structure of the CDW (in the normal state) which is relevant to describe the Fermi surface reconstruction leading to QO.

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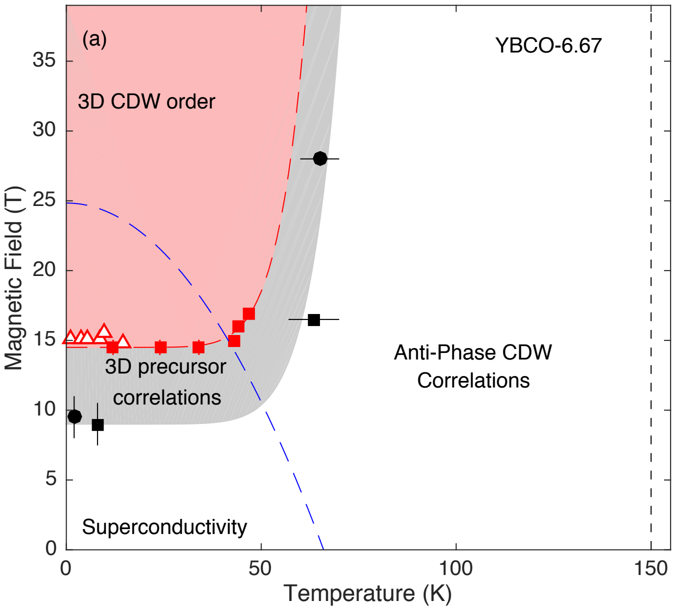

Fig. 12.2 – Temperature-magnetic field phase diagram. The pink shaded areas represent the regions where short range 3D CDW order exists. Grey bands indicate the regions where growing 3D CDW precursor correlations are observed. Solid red square points indicate the onset of a 3D CDW order with $\mathbf{q}_\mathrm{b}$ =(0,δ_b,0) determined from the variation of the $\xi_{\mathrm{c},\ell=1}$ correlation length and the intensity of the 3D peak.

12.3 Electron scattering, charge order, and pseudogap physics in $\mathrm{La}_{1.6-x}\mathrm{Nd}_{0.4}\mathrm{Sr}_x\mathrm{CuO}_4$

For the past 30 years, high-temperature superconductivity (SC) presented itself as one of the most important problems for physicists in the field of condensed matter physics. Layered copper-oxide compounds, which still hold the ambient pressure record of the maximum achievable T_c, exhibit a rich phase diagram including Mott and pseudogap physics along with CDW and spin-density wave (SDW) orders. An outstanding question is to understand how these phenomena are related. There exists compelling evidence for SC and charge-wave-order co-existing through an intertwined competing relation [17–19]. How this composite order (SC + CDW) relates to the pseudogap phase is, however, much less clear. For this reason, we have investigated [20] the charge stripe ordered system $\mathrm{La}_{1.6-x}\mathrm{Nd}_{0.4}\mathrm{Sr}_x\mathrm{CuO}_4$ (Nd-LSCO) by angle-resolved photoemission spectroscopy – one of the best probes for pseudogap physics.

In the system $\mathrm{La}_{1.6-x}\mathrm{Nd}_{0.4}\mathrm{Sr}_x\mathrm{CuO}_4$ (Nd-LSCO), CDW order around the special 1/8 doping suppresses strongly T_c. This allows a low-temperature spectroscopy study of the relation between pseudogap and CDW order [21]. These two effects are difficult to disentangle, as they both manifest themselves by a spectral gap [22]. By varying doping concentration $p = x$, photoemission-spectra were recorded in the overdoped metallic phase ($p > 0.25$), just inside the pseudogap phase ($p = 0.15, 0.2$) and at the so-called $p = 1/8$ doping where stripe order is the strongest. In the metallic phase, gap-less excitations have been observed all around the Fermi surface manifesting themselves as a single peak in the symmetrized energy distribution curve (EDC). By slightly reducing the doping just into the pseudogap phase a partial gap opens in the so-called anti-nodal region around the zone boundary. In this process spectral weight is conserved but shifted by the presence of the pseudogap. At $p = 1/8$ doping, a particularly strong enhancement of non-conservative, anti-nodal spectral-weight suppression is found inside the CDW (stripe ordered) phase. The suppression of spectral weight also extends up to much larger energies ($\approx$ 100 meV). It is thus a possibility that CDW order manifests itself in the antinodal spectra at the 1/8 doping. If so, the implication is that in Nd-LSCO, charge-stripe order and the pseudogap phase contributes differently to the suppression of anti-nodal spectral-weight. These results suggest that CDW order, which recently has been identified as a universal property of copper oxide compounds, is not directly linked to the pseudogap phase.

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12.4 Spin-Orbit-Induced Orbital Excitations in Sr_2RuO_4 and Ca_2RuO_4

The relativistic coupling between electronic spin and orbital momentum was long thought to have marginal influence on electrons in solids. Following the prediction and observation of topological surface states on Bi-based compounds [23], this paradigm has changed. Discovery of novel quantum phases realized through strong spin-orbit interaction is now a vivid field of research [24]. The demonstration of spin-orbit coupling driving a new type of Mott insulating state in layered iridates [25] is a good example of this. It has been proposed that doping of this effective $J_{1/2}$-Mott insulating state could lead to an exotic type of superconductivity [26], where Cooper pairs are composed of strongly spin-orbit coupled electrons.

In this context, it is interesting to study other systems that display Mott physics and superconductivity in conjunction with strong spin-orbit interaction. The $4d$-transition metal oxide system $Ca_{2-x}Sr_xRuO_4$ represents such a case. For $x = 0$, the system is a Mott insulator, whose exact nature is not clarified. At the opposite stoichiometric end ($x = 2$), the system has a superconducting ground state ($T_c = 1.5$ K) originating from a correlated Fermi liquid [27]. Although triplet p-wave superconductivity was proposed early on, the mechanism and symmetry class of the superconducting order parameter is still debated.

A fundamental question is how strongly spin-orbit interaction influences the electrons in these materials and whether it has an impact on the Mott insulating and superconducting ground states. Current experimental evidence for a strong spin-orbit interaction stems from absorption spectroscopy, that has revealed a considerable admixture of the Ruthenium t_{2g} orbitals. More recently, spin-resolved photoemission spectroscopy has reported spin-polarized bands in Sr_2RuO_4. However, the most direct consequence of strong spin-orbit interaction – the splitting of t_{2g} states – has not yet been probed directly by experiments. Orbital excitations transferred across this splitting are in fact not accessible to optical spectroscopies. Furthermore, the Ru L-edge (≈ 3 keV) is currently inaccessible to high-resolution RIXS instrumentation (as it lies right between soft and hard x-ray optics).

To overcome these experimental challenges, we accessed the Ru $4d$-orbital excitations through their hybridization with oxygen $p-$orbitals [28]. Exploiting a combination of x-ray absorption (XAS) and oxygen K-edge resonant inelastic x-ray spectroscopy (RIXS), we provide direct evidence for a splitting of the ruthenium t_{2g} states. This evidence is directly visible in the RIXS spectra shown in Fig. 12.3 where the 300 meV excitation stems from intra t_{2g} transitions. Our RIXS study of Ca_2RuO_4 and Sr_2RuO_4 reveals excitations that allow an estimation of the spin-orbit coupling, in the same fashion as for the iridates. These results suggest a spin-orbit coupling λ_{so} of ≈ 200 meV – only about two times weaker than in the iridates. The presence of strong spin-orbit interaction implications for the symmetry class of the p-wave superconducting ground state.

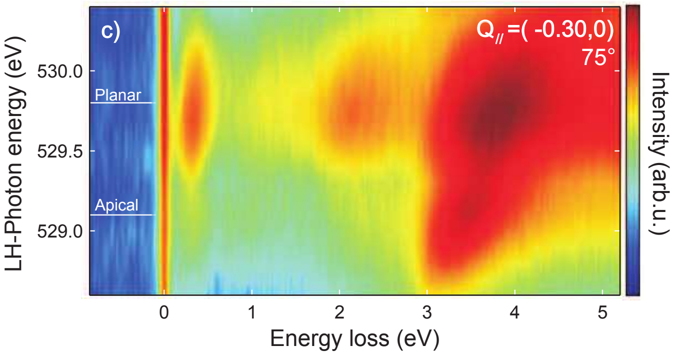

FIG. 12.3 – Resonant inelastic x-ray spectra collected with momentum transfer and polarization as indicated and displayed using a logarithmic color scale as a function of incident photon energy.

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