9 Recent Advances in Experimental Research on High-Temperature Superconductivity

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1 Introduction

The theoretical description of electronic materials in the crossover regime between fully localized and fully itinerant electrons continues to be one of the greatest challenges in theoretical physics. The most profound problem in this field – the origin of high-temperature superconductivity – remains unsolved despite more than a quarter century of research. Recently, however, the experimental investigation of correlated-electron materials has made astounding progress, based on advances in materials synthesis and experimental methodology. As a result, the overall situation in this research field now looks very different than it did just ten years ago. Whereas some theoretical challenges appear less formidable than they did at that time, unforeseen new issues have been raised by the latest experiments. In this chapter, we will briefly summarize some of these developments, and then discuss some concrete challenges for theoretical research on cuprate superconductors in more detail. The emphasis will be on results obtained by spectroscopic methods.

A particularly influential development on the materials front has been the discovery and subsequent exploration of high-temperature superconductivity in iron pnictides and chalcogenides. Although these compounds exhibit a completely different chemical composition and lattice structure from the copper oxides, the phase diagrams of both classes of materials are closely related [1]. In particular, antiferromagnetically ordered phases at commensurate valence electron configuration are surrounded by superconducting phases at both lower and higher electron density. The observation of closely analogous low-energy spin fluctuations (including the so-called "resonant mode") in the superconducting regimes of the phase diagram [2] has further highlighted the analogy to the cuprates and the case for magnetic mechanisms of Cooper pairing. At the same time, the antiferromagnetic state in the iron-based materials is a metallic spin density wave, rather than a Mott insulator with fully localized electrons. This implies that high-temperature superconductivity is not confined to "doped Mott insulators" – a class of systems in which the combination of strong correlations and disorder poses particularly profound theoretical problems. A theoretical approach to the high-T_c problem from the metallic limit with more effectively screened Coulomb interactions and a well-defined Fermi surface now appears much more promising than it did before the discovery of the iron-based superconductors.

A related breakthrough on the experimental front was made possible by the recently developed capability of carrying out transport and thermodynamic measurements in magnetic fields up to 100 T. Combined with the availability of single-crystal samples with very long transport mean free paths (including especially the stoichiometric underdoped compounds $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_4O_8$), such experiments have led to the discovery of quantum oscillations indicative of Landau quasiparticles in underdoped cuprates [3–8], adding to earlier data on overdoped compounds [9, 10] above their respective critical fields. Quantum oscillations have also been observed in several iron pnictide superconductors, including the stoichiometric compounds LiFeAs and LiFeP [11]. Fermionic quasiparticles are thus a generic feature of high-temperature superconductors over a wide range of doping levels. This speaks in favor of theories based on Cooper pairing of conventional quasiparticles, and against various more exotic models of high-T_c superconductivity.

The last decade also saw increasingly insightful experiments with scanning tunneling spectroscopy on correlated-electron materials, including the copper-based, iron-based, and heavy-fermion superconductors. These experiments opened our eyes to nanoscale inhomogeneities of the electron density induced by randomly placed dopant atoms in some of the most extensively investigated materials including superconducting $Bi_2Sr_2CuO_{6+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ [12,13]. Because of the low carrier density, the Coulomb potential of the ionized donor and acceptor atoms is much more poorly screened than in conventional metals. This can lead to a pronounced inhomogeneous broadening of spectroscopic features in volume-averaging experiments (including especially photoemission spectroscopy) on non-stoichiometric materials. These extrinsic effects must be considered before interpreting broad spectroscopic features as evidence of non-Fermi-liquid behavior. Disorder and inhomogeneity have also led to the development of a new technique, quasiparticle interference (QPI) spectroscopy [14], as a powerful phase-sensitive probe of the superconducting order parameter in both copper- [15] and iron-based [16] superconductors.

Further, the research field has benefitted greatly from the increase in energy resolution of spectroscopic probes such as inelastic neutron scattering (INS), angle-resolved photoemission spectroscopy (ARPES), and (non-resonant) inelastic x-ray scattering (IXS), which can now be performed with resolutions of 1 μeV in the former and 1 meV in the latter two. While high-resolution INS is yielding new insights into "spin freezing" phenomena in both copper oxides [17] and iron arsenides [18], high-resolution ARPES has not only led to a detailed description of the energy and momentum dependence of the superconducting gap function in high-temperature superconductors [19], but has recently also provided equivalent data sets for $NbSe_2$ and other classical charge density wave materials with smaller gaps [20, 21]. Notably, these experiments have led to the discovery of a "pseudogap" and "Fermi arcs" above the CDW transition temperature in $TaSe_2$ and $NbSe_2$ [20, 21]. High-resolution IXS experiments provide detailed insights into the role of the electron-phonon interaction in driving charge-density-wave formation [22–24]. Following these developments, spectroscopic data on cuprates [22] can now be calibrated against the behavior of their more conventional cousins [23, 24].

Over the past five years, resonant elastic (REXS) and inelastic (RIXS) x-ray scattering have had a tremendous impact in research on correlated-electron systems. REXS allows the determination of spin, charge, and orbital order of the valence electron system with very high sensitivity [25], and has been instrumental for the recent discovery of charge density waves in bulk copper-oxide superconductors [26–30]. RIXS, on the other hand, was known ten years ago mainly as a momentum-resolved probe of interband transitions in the 1-5 eV range, quite similar to electron energy loss spectroscopy. Following a phenomenal improvement of the energy resolution by about an order of magnitude [31], RIXS experiments have resolved orbital and spin excitations in a variety of metal oxides, including recently the copper-oxide [32–35] and iron-pnictide [36] superconductors. With its high sensitivity to high-energy excitations, which is complementary to INS, RIXS is becoming an increasingly powerful spectroscopic probe of correlated-electron materials.

These developments on the experimental front have opened up various new theoretical challenges that could hardly be foreseen a decade ago. First, the theoretical foundation for some of the new techniques is far from complete, and it will sometimes go hand-in-hand with the understanding of the systems to be investigated. Whereas the expression of the non-resonant x-ray and neutron scattering cross sections in terms of density-density and spin-spin correlations is generally understood and accepted, an analogous formalism for REXS and RIXS is still under development. It is already clear that a comprehensive description of the photon energy dependence of the REXS and RIXS cross sections will have to take Coulomb interactions into account – the same interactions whose low-energy manifestation are the subject of investigation. Similarly, a comprehensive understanding of QPI in tunneling spectroscopy will require detailed information about the defects that scatter the quasiparticles. Even more fundamental challenges are raised by the theoretical description of pump-probe techniques that take the correlated-electron system far out of thermal equilibrium.
Here we will highlight some theoretical challenges from recent experiments that can be spelled out independent of technical details of the experimental probes.

2 Magnetic order

The generic phase diagram of the copper oxides (Fig. 1) includes a Mott-insulating phase centered around the doping level $p = 0$ corresponding to a single hole per Cu site in the CuO_2 planes, and a d-wave superconducting phase extending from $p \sim 0.05$ to ~ 0.25. In the Mott-insulating phase, commensurate, collinear antiferromagnetic order is observed with ordering wave vector $\mathbf{q} = (\pi, \pi)$ (in a notation in which the nearest-neighbor lattice parameter $a \sim 3.8$ Å is set to unity). Static magnetic order persists at low temperatures over some range of p, but the ordering wave vector is shifted away from (π, π). In both $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$, the two systems where the doping-induced commensurate-incommensurate transition has been studied in detail, the amplitude of the magnetization in this state remains a substantial fraction of the antiferromagnetic sublattice magnetization at $p = 0$, and the incommensurability δ increases monotonically with p [17, 39, 40]. The direction of the propagation vector in both compounds is different (along the Cu-O bond in $YBa_2Cu_3O_{6+x}$, and 45° away from it in $La_{2-x}Sr_xCuO_4$ for $0.02 \leq p \leq 0.05$).
High-resolution neutron scattering and muon spin rotation (μSR) studies of $YBa_2Cu_3O_{6.35}$ with $p \sim 0.06$ have demonstrated that the incommensurate magnetic order at wave vector $(\pi \pm \delta, \pi)$ is highly unstable to thermal fluctuations [17]. Even at temperatures of a few Kelvin, the signatures of static magnetic order in both sets of experiments are replaced by those of a slowly relaxing local magnetization. The "wipeout" of the nuclear magnetic resonance signals in other underdoped cuprates at low temperatures has also been attributed to slow spin fluctuations of this kind [41, 42]. This behavior is consistent with the "critical slowing down" expected in proximity to a zero-temperature phase transition in two-dimensional Heisenberg systems [43] in combination with disorder due to dopant atoms that limit the exponential divergence of the spin-spin correlation length at low temperatures [44].

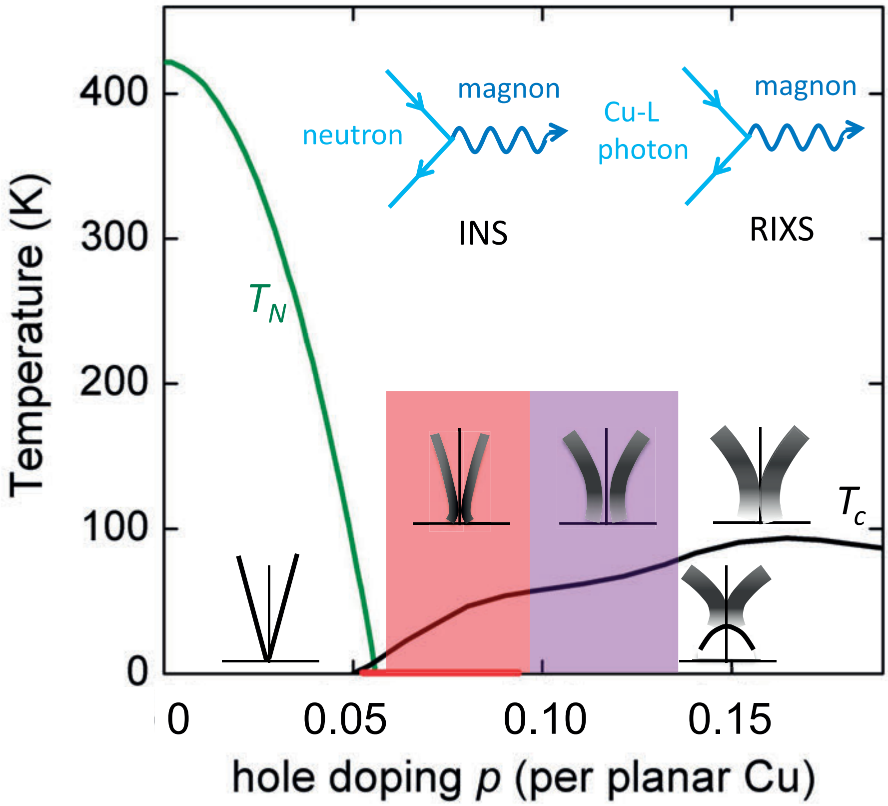

Fig. 1: *Phase diagram of $YBa_2Cu_3O_{6+x}$. The Néel temperature, T_N, and superconducting transition temperature, T_c, were taken from Refs. [37] and [38], respectively. The red line indicates the stability range of static incommensurate magnetic order. Pink and purple shaded regions indicate temperature and doping regimes with low-energy incommensurate spin and charge correlations, respectively. The insets show diagrams illustrating INS and RIXS from spin excitations, as well as sketches of the dispersion and spectral weight distribution of the spin excitations around the antiferromagnetic ordering wavevector $\mathbf{q} = (\pi, \pi)$ in the different regimes of the phase diagram.*

Upon further heating, low-energy spin fluctuations persist, but the incommensurability δ decreases continuously with increasing temperature. At temperatures exceeding $T \sim 150$ K, the signature of the uniaxial magnetic modulation is no longer visible in the neutron scattering data, and the magnetic response is centered at $\mathbf{q} = (\pi, \pi)$. The order-parameter-like temperature dependence of δ is consistent with a proximate "nematic" phase transition where the fourfold rotational symmetry of the CuO_2 layers is spontaneously broken [45]. In the orthorhombic crystal structure generated by the oxygen dopant atoms in $YBa_2Cu_3O_{6+x}$ with $x \geq 0.2$, this transition is expected to be broadened into a crossover. Thermodynamic singularities akin to those associated with a nematic transition in tetragonal $Sr_3Ru_2O_7$ [46] are indeed not observed in the cuprates. However, the observation of a similar temperature-driven incommensurate-commensurate transition in lightly doped $La_{2-x}Ba_xCuO_4$ [47] shows that it reflects an intrinsic trend of the correlated electrons in the CuO_2 planes, rather than subtleties of the crystal structure of specific compounds such as the chains of oxygen dopant atoms in $YBa_2Cu_3O_{6+x}$. Uniaxially modulated spin structures with unusual thermal properties thus appear to be a generic feature of all cuprates at doping levels close to the Mott-insulating phase.

In some compounds with intrinsically low superconducting T_c, including especially materials with composition $La_{2-x}Ba_xCuO_4$ and $La_{2-x-y}Sr_x(Nd,Eu)_yCuO_4$ ("214 compounds") that exhibit the "low-temperature tetragonal" (LTT) crystal structure, uniaxial incommensurate magnetic order with wave vectors $\mathbf{q} = (\pi \pm \delta, \pi)$ and $(\pi, \pi \pm \delta)$ has also been observed at higher doping levels, in some cases up to $p \sim 0.15$ [48–55]. Corresponding charge-modulation peaks at $\mathbf{q} = (2\delta, 0)$ and $\mathbf{q} = (0, 2\delta)$ indicate that these magnetic peaks arise from a uniaxial ("stripe") modulation of the spin amplitude. Static stripe order is only observed in compounds with the LTT structure whose primitive vectors are parallel to the stripe propagation vector – a situation that favors pinning to the lattice. Orthorhombic $La_{2-x}Sr_xCuO_4$ does not exhibit stripe order, but low-energy spin fluctuations with the same momentum-space signature have been interpreted as evidence of fluctuating stripes [49, 51].

In all other cuprates investigated so far, static magnetic order disappears for $p \gtrsim 0.07$, and the magnetic excitation spectrum determined by neutron scattering develops a sizable spin gap [56], as expected for a magnetic quantum critical point. However, spinless Zn impurities in the CuO_2 planes nucleate static incommensurate magnetic order with a correlation length of a few unit cells in the spin-gap regime [57, 58]. Even at an impurity concentration of less than 1%, this leads to inhomogeneous coexistence between different electronic phases. Dopant-induced disorder, which is particularly pronounced in the 214 compounds [59], may contribute to the stability of the "stripe" phase in nominally pristine members of this family.

3 Charge order

Resonant [26, 28, 30] and nonresonant [27, 29] x-ray diffraction experiments on $YBa_2Cu_3O_{6+x}$ samples in the doping regime $0.07 \leq p \leq 0.13$ have revealed biaxial, incommensurate charge density wave (CDW) correlations. Similar CDW correlations were demonstrated very recently in REXS experiments [60] on $Bi_2Sr_2CuO_{6+\delta}$, $Bi_2Sr_2CaCu_2O_{8+\delta}$, and $HgBa_2CuO_{4+\delta}$, indicating that incommensurate biaxial CDW correlations are a universal feature of the underdoped cuprates. The recent REXS experiments are qualitatively consistent with prior STS measurements that have revealed charge modulations in several cuprate families [61–63]. However, since the CDW features are superposed by electronic reconstructions induced by incommensurate lattice modulations in the former two compounds (some of which have turned out to be surface sensitive [64]), they are more difficult to interpret than those in $YBa_2Cu_3O_{6+x}$.

The compilation of REXS data on $YBa_2Cu_3O_{6+x}$ in Fig. 2 shows a continuous increase of the amplitude and a reduction of the wave vector of these correlations with increasing p. On a qualitative level, the doping dependence of the CDW wave vector (Fig. 3) tracks the distance between the antinodal regions of the Fermi surface (Fig. 4), which shrinks as the Fermi surface expands with increasing doping level. A quantitative comparison with ARPES data, however, reveals a better agreement with the distance between the tips of the "Fermi arcs" where the density of states is enhanced due to the opening of the pseudogap [60]. This suggests an intimate rela-

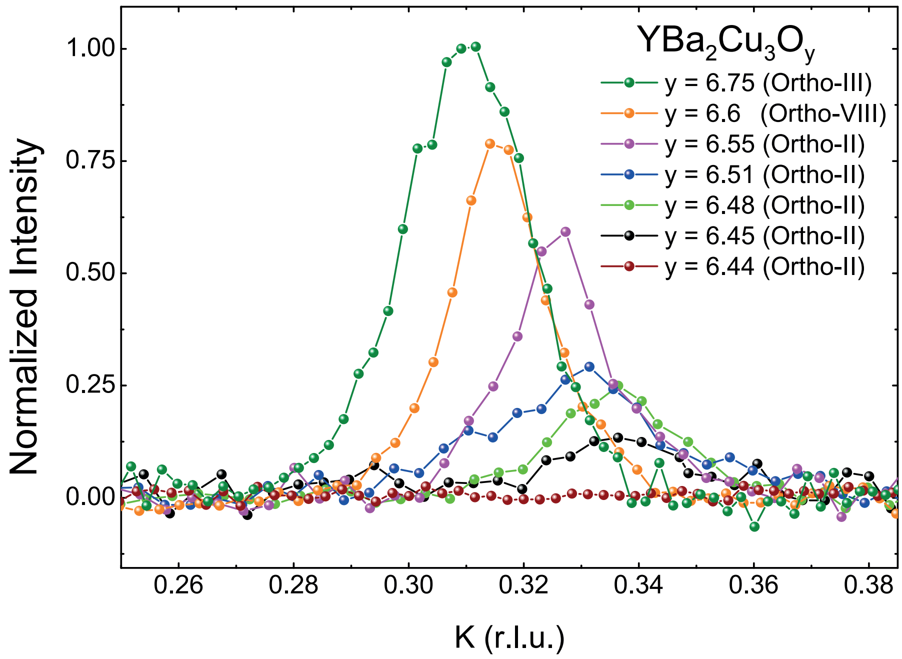

Fig. 2: *REXS scans with photon energy tuned to the L-absorption edges of planar copper atoms in $YBa_2Cu_3O_{6+x}$. The scans are along $\mathbf{q} = (0, K)$ in the CuO_2 layers, where K is measured in reciprocal lattice units (r.l.u.). The labels Ortho-II, III, and VIII refer to the arrangement of oxygen dopant atoms [26, 28, 30].*

tionship between the "Fermi arc" phenomenon and the CDW that should be further explored. Both the intensity and the correlation length of the CDW peaks grow upon cooling and exhibit pronounced maxima at the superconducting T_c [26–30]. This directly demonstrates competition between superconductivity and CDW formation, which predominantly affects electronic states near the antinodal regions of the Fermi surface. The strong competition between both types of order implied by these findings also explains the well-known plateau in the T_c-versus-p relation.

Very recent high-resolution IXS data [22] have shown large anomalies of acoustic phonons associated with CDW formation, as well as a "central peak" indicative of static CDW regions nucleated by lattice defects. Pinning of soft phonons associated with structural phase transitions has also been observed in classical materials such as $SrTiO_3$ and Nb_3Sn, albeit over a much narrower temperature range. The persistence of this domain state over a much wider temperature range than corresponding phenomena in classical materials [65, 66] probably reflects the strong competition between CDW correlations and superconductivity. The nanoscale CDW domains revealed by these experiments will certainly contribute to the anomalous transport and thermodynamic properties of the underdoped cuprates. Both the central peak and the acoustic-phonon anomalies abruptly disappear in optimally doped $YBa_2Cu_3O_7$ [22], indicating that the nanoscale inhomogeneity is detrimental to high-temperature superconductivity. We note, however, that superconductivity-induced anomalies of the Cu-O bond-bending phonon around the CDW wave vector have also been observed in $YBa_2Cu_3O_7$ [67]. These findings indicate an underlying zero-temperature CDW critical point [68], which deserves further experimental and theoretical investigation.

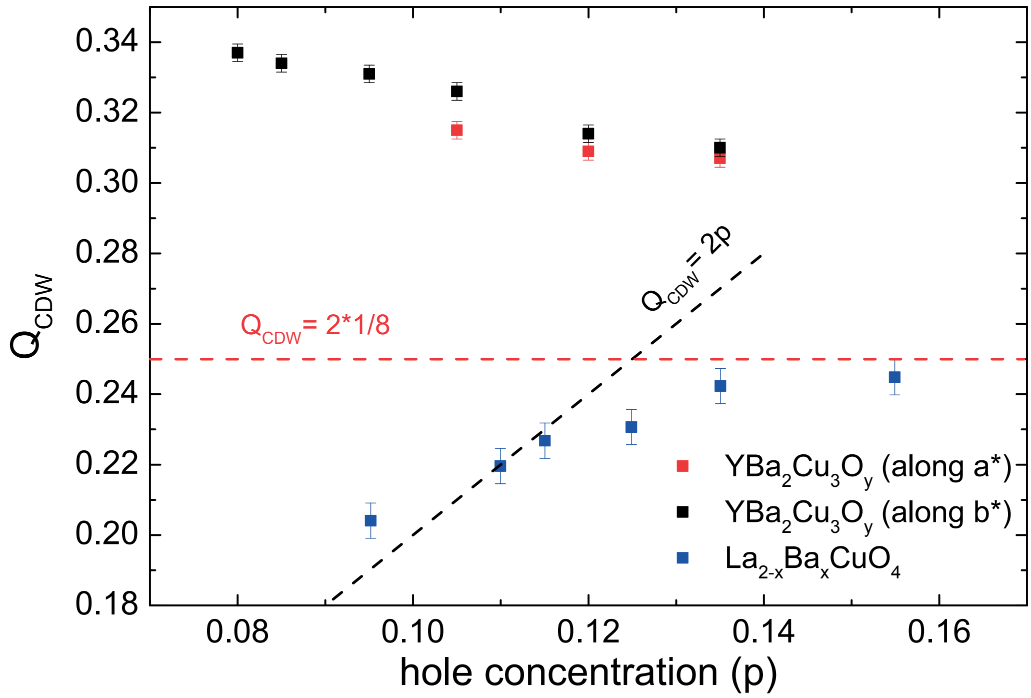

Fig. 3: *Doping dependence of the CDW wavevector in $YBa_2Cu_3O_{6+x}$ [26, 28, 30], compared to the wave vector characterizing charge order in the "striped" state of $La_{2-x}Ba_xCuO_4$ [55].*

Whereas in zero magnetic field the competition between superconductivity and CDW order appears to preclude true CDW long-range order, recent NMR [69] and ultrasound [70] experiments have provided evidence of static CDW order and a thermodynamic CDW phase transition in high magnetic fields, where superconductivity is either greatly weakened or entirely obliterated. This is consistent with the reduction of the CDW peak intensity observed in REXS experiments in moderate magnetic fields [27, 29, 30]. In the presence of CDW long-range order, a Fermi surface reconstruction leading to the formation of small pockets is expected. Although this is an appealing explanation of the recent quantum oscillation data on stoichiometric $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_4O_8$ [3, 7], a comprehensive, quantitatively consistent explanation of REXS and quantum oscillation data has not yet been reported.

4 Spin fluctuations

Since the discovery of the d-wave symmetry of the superconducting gap function, spin-fluctuation-mediated Cooper pairing has been one of the leading contenders in the quest for the mechanism of high-temperature superconductivity [1]. By combining INS and RIXS data, we now have detailed and comprehensive information about the spin fluctuation spectrum, which can be used as a basis for stringent tests of these models. We provide a brief survey of recent results on the doping evolution of the spin excitations, before discussing their implications for spin-fluctuation-mediated pairing theories.

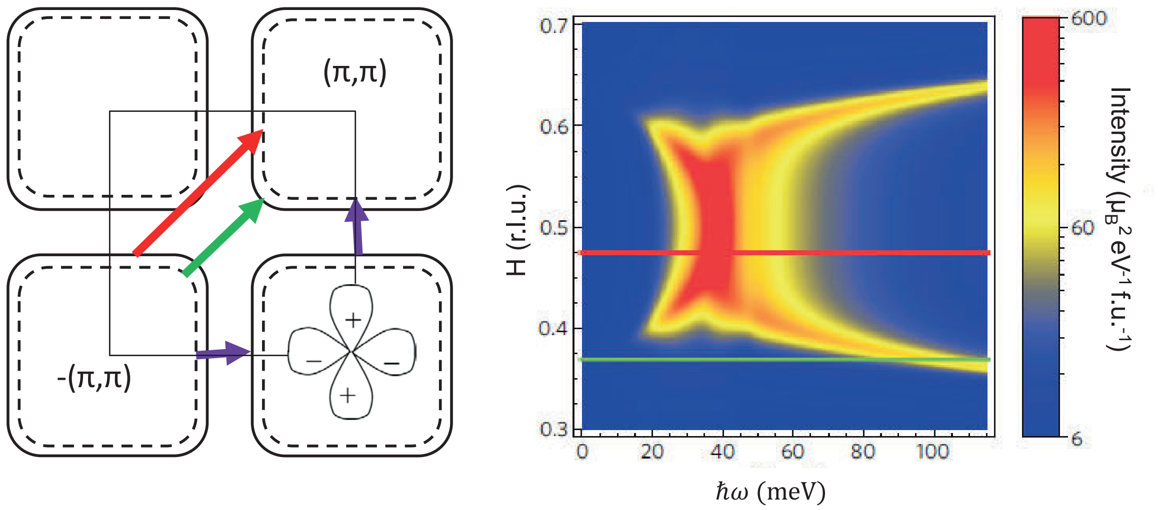

Fig. 4: *Left panel: Kinematics of spin (red and green arrows) and charge (purple arrows) fluctuation scattering of quasiparticles on the Fermi surface of a bilayer compound such as* $YBa_2Cu_3O_{6+x}$*. Black solid and dashed lines correspond to Fermi surfaces in antibonding and bonding bands, respectively. The inset shows a sketch of the* d*-wave superconducting gap function. Right panel: Spin fluctuation intensity in* $YBa_2Cu_3O_{6.6}$ *along the* (H, H) *direction in the* CuO_2 *planes.* H *is measured in reciprocal lattice units, so that* $H = 0.5$ *corresponds to* $\mathbf{q} = (\pi, \pi)$ *[121].*

In the Mott insulating state, the spin excitations determined by INS are well described as magnon modes of the 2D Heisenberg model with nearest-neighbor superexchange interaction $J \sim 130 - 140$ meV. The magnons emerge from the antiferromagnetic ordering wave vector $\mathbf{q} = (\pi, \pi)$ and are nearly gapless due to the weak magneto-crystalline anisotropy of the Cu ions (Fig. 1) [39,40]. In bilayer compounds such as $YBa_2Cu_3O_6$, an additional superexchange interaction between spins in directly adjacent CuO_2 layers, $J_\perp \sim 0.1J$, has to be taken into account. This leads to a non-generic optical magnon branch with a gap of 70 meV at $\mathbf{q} = (\pi, \pi)$ [71,72]. All other exchange interactions are significantly weaker.

In compounds with incommensurate magnetic order, including lightly doped $YBa_2Cu_3O_{6+x}$ and $La_{2-x}Sr_xCuO_4$, as well as moderately doped, stripe-ordered $La_{2-x}Ba_xCuO_4$ with $x = 1/8$, gapless spin excitations emerge from the magnetic Bragg reflections (Fig. 1). Contrary to the magnon excitations in the commensurate antiferromagnet, however, these excitations do not follow the predictions of the linear spin wave theory, which for incommensurate magnets predicts spin wave "cones" with approximately uniform spectral weights along their rims, at least at low energies [76]. Instead, intense low-energy excitations emanating from the incommensurate magnetic reflections first disperse towards $\mathbf{q} = (\pi, \pi)$, then bend over and approach the magnon branch of the insulating cuprates. With increasing excitation energy, cuts of the magnetic dispersion surface along the ordering wavevector thus resemble an "hourglass" with an open neck around $\mathbf{q} = (\pi, \pi)$ at $\hbar\omega \sim 30 - 50$ meV [73, 75]. Similar hourglass dispersions have recently also been observed in insulating manganates [83] and cobaltates [84] with incommensurate magnetic order.

Whereas in the cuprates with 214 structure the magnetic excitations in the normal state remain nearly gapless even when static magnetic order is not present, the spectral weight of low-energy spin excitations in $YBa_2Cu_3O_{6+x}$ and other compounds with higher maximal T_c is strongly reduced, so that the excitation spectrum can be approximated as a gapped version of the "open hourglass" spectrum in the state with incommensurate magnetic order (Fig. 1) [56, 74]. This is consistent with a quantum phase transition between magnetically ordered and quantum-disordered phases.

The momentum distribution of the spin excitation intensity is similar in underdoped cuprates with and without magnetic order. Below the neck of the hourglass, it exhibits a uniaxial anisotropy, reflecting the uniaxial nature of the (real or proximate) ground state [74]. At excitation energies above the neck, it displays fourfold symmetry. This aspect cannot be reproduced by calculations attributing the spin modulation to static "stripes" [77, 78], but it is correctly captured by models incorporating strong charge fluctuations on top of the "striped" background [79, 80], and by models of spin fluctuations in metals near a nematic instability [81]. Models based on a spiral ground state also reproduce many of the salient features of the magnetic excitations [82].

In the superconducting state of $YBa_2Cu_3O_{6+x}$, the neck of the hourglass closes, and a sharp "resonant" mode with a downward dispersion is formed below the superconducting energy gap (Fig. 1). As a prominent signature of magnetically mediated Cooper pairing, this mode has been studied very extensively [2,85–97]. Its energy increases with increasing p up to optimal doping, and decreases in the overdoped state, qualitatively following the "dome" in the T_c-versus-p relation. Similar observations have been made in other compounds with optimal T_c around 90 K, including $Bi_2Sr_2CaCu_2O_{8+\delta}$ [98–100], $Tl_2Ba_2CuO_{6+\delta}$ [101], and $HgBa_2CuO_{4+\delta}$ [102]. Magnetic resonant modes have also been observed in iron pnictide [106–108] and heavy-fermion superconductors [109–111], and thus appear generic to superconductors near an antiferromagnetic instability. In overdoped cuprates, the spectral weight of the low-energy spin excitations around $\mathbf{q} = (\pi, \pi)$, including the one of the resonant modes below T_c, is gradually reduced [103–105], and disappears entirely (to within the sensitivity of INS) in strongly overdoped $La_{1.78}Sr_{0.22}CuO_4$ ($p = 0.22$) [116, 117].

Based on these observations, and on related anomalies in fermionic spectral functions, the resonant mode has been attributed to a feedback effect of the Cooper pairing interaction on low-energy spin fluctuations that mediate the pairing interactions [1,112,121]. Note that the BCS coherence factors in the dynamical spin susceptibility extinguish the spectral weight of the mode, unless the sign of the superconducting gap function changes sign at the Fermi surface. The observation of the resonant mode has therefore been taken as evidence for d-wave pairing symmetry in the cuprates [1,87,113], and of $s_\pm$ symmetry in the iron pnictides [106–108,114,115].

The spin dynamics for $\hbar\omega \lesssim 100$ meV thus reflects the ground state of the spin system, which strongly evolves with temperature and doping and is influenced by details of the crystal and electronic structure of different cuprate families. On the other hand, recent RIXS measurements on $YBa_2Cu_3O_{6+x}$ over a wide range of doping levels have shown that spin fluctuations with excitation energies $\hbar\omega \gtrsim 100$ meV are weakly doping and temperature dependent, and that their dispersion relations and energy-integrated spectral weights remain closely similar to antiferromagnetic magnons in Mott-insulating $YBa_2Cu_3O_{6+x}$ [33]. Very recently, we have extended these experiments to highly overdoped $Tl_2Ba_2CuO_{6+\delta}$ ($p = 0.27$), a compound that features a single, isolated CuO_2 plane per formula unit and very low intrinsic disorder [34]. In analogy to the spin excitations above the ordering temperature of insulating magnets, the "paramagnon" excitations in the cuprates are indicative of short-range correlations between localized spins. Since ARPES [118], angle-dependent magnetoresistance [9], and quantum oscillation [10] experiments on $Tl_2Ba_2CuO_{6+\delta}$ have demonstrated canonical Fermi-liquid behavior with a Fermi surface that agrees quantitatively with the predictions of density functional theory, the persistence of magnon-like excitations well into the Fermi-liquid regime of the cuprates is highly surprising. Related results have been reported for highly overdoped $La_{2-x}Ca_xCuO_4$ [35].

5 Spin-fluctuation-mediated superconductivity

Are these spin excitations mediators of the Cooper pairing interaction, or are they simply bystanders? The appearance of the magnetic resonant mode in the superconducting state indicates that the low-energy excitations near $\mathbf{q} = (\pi, \pi)$ are intimately involved in the formation of the superconducting state. The absolute spectral weight of the resonant mode translates into an exchange energy that is comparable to the superconducting condensation energy [88,97,119]. The recently detected superconductivity-induced enhancement of the two-magnon Raman scattering cross section (which is dominated by high-energy spin excitations near the antiferromagnetic zone boundary) in $HgBa_2CuO_{4+\delta}$ indicates that higher-energy excitations also experience significant feedback effects [120]. Feedback effects over the entire paramagnon spectrum are not unexpected, because (unlike phonons in conventional superconductors) the spin excitations are generated by the same electrons that form the Cooper pairs. Converse evidence gleaned from photoemission [123] and optical [124–126] spectroscopies also indicate substantial coupling between conduction electrons and bosonic excitations with a bandwidth of ~ 300 meV (comparable to that of the paramagnon spectrum), although based on these data alone it is hard to determine whether or not this coupling contributes positively to the d-wave Cooper pairing interaction.

These results have encouraged us and our collaborators to go a step further towards a quantitative description of spin-fluctuation mediated Cooper pairing. Specifically, we have taken the experimentally measured spin fluctuation spectra of $YBa_2Cu_3O_{6.6}$ [121] and $YBa_2Cu_3O_7$ [33] as input for Eliashberg calculations of the superconducting gap, Δ, and T_c. These calculations are carried out in the framework of the "spin-fermion" model, which treats the spin excitations in a manner analogous to phonons in conventional superconductors. Given the failure of similar calculations to quantitatively describe the microscopic parameters of conventional superconductors, it is not unreasonable to be skeptical about their prospects for these highly correlated materials. One has to keep in mind, however, that most conventional superconductors have complex Fermi surfaces and many phonon branches, and that the momentum-dependent electron-phonon interaction is difficult to measure or compute accurately. Uncertainties in either of these quantities translate into large errors in Δ and T_c. In the cuprates, only a single electronic band and a single excitation branch is relevant near the Fermi surface (apart from minor complications in materials with multilayer structures), and information about the spin-fermion coupling can be extracted from spectroscopic data, greatly reducing the associated uncertainties.

The spin-fermion model ultimately needs to be rigorously justified. On a qualitative level, however, it appears promising at least for optimally doped and overdoped materials, whose spin excitations are gapped and do not exhibit quantum-critical behavior, and whose Fermi surfaces are not strongly affected by the "pseudogap". Herein lies the importance of the recent RIXS experiments, which have supplied detailed information about the spin excitations in the optimally and overdoped regimes of materials with intrinsically high T_c [33, 34]. Lightly doped materials with incommensurate magnetic order and 214 materials close to a "stripe" instability, which have been extensively characterized by INS [39], clearly require a different theoretical treatment.

In the Eliashberg calculations for $YBa_2Cu_3O_{6+x}$, spin excitations extending from the spin gap at $\mathbf{q} = (\pi, \pi)$ up to $\hbar\omega \sim 200$ meV, which scatter electrons between states in lobes of the d-wave superconducting gap function with opposite sign, are pair forming [33, 34]. The highest-energy excitations near the antiferromagnetic zone boundary are indifferent to d-wave pairing, while those near $\mathbf{q} = 0$, which scatter electrons within the same lobe of the gap function, are pair breaking. Since the latter excitations have much lower spectral weight than those near $\mathbf{q} = (\pi, \pi)$, this pair-breaking effect only leads to a minor reduction of T_c. Because of kinematical constraints, RIXS detects predominantly low-$\mathbf{q}$ spin excitations that are only weakly involved in Cooper pairing. The recently reported weak doping-dependence of the RIXS cross section in the overdoped regime [34, 35], where T_c depends strongly on p, therefore does not contradict spin fluctuation mediated Cooper pairing models. We note that INS data do in fact show a strong decrease of the spectral weight of spin fluctuations near $\mathbf{q} = (\pi, \pi)$ at high p, consistent with a reduction of the Cooper pairing strength [116, 117].

The Eliashberg calculations predict a superconducting transition temperature $T_c \sim 170$ K and gap $\Delta \sim 60$ meV, about a factor of two larger than the experimental observations for $YBa_2Cu_3O_7$ [33, 121]. Interestingly, infrared conductivity experiments have provided evidence of superconducting fluctuations in underdoped $YBa_2Cu_3O_{6+x}$ up to temperatures of order 180 K [127], suggesting that the outcome of the mean-field Eliashberg calculations is physically meaningful. In view of the experimental data presented in the previous section, competition from CDW order is likely to be a major factor in the suppression of T_c from its mean-field value. The calculations also account for the "kinks" in the electronic band dispersions in a quantitatively consistent manner [121, 122]. These results, along with other efforts along the same lines [128, 129] give rise to the hope that a systematic improvement of these calculations – combined with more complete and accurate experimental data – will finally lead to a quantitative understanding of high-T_c superconductivity.

6 Challenges for theory

In summary, advances in experimental research have yielded a detailed, consistent picture of the phase behavior and spin dynamics of the cuprates over a wide range of doping levels. Although our knowledge is still far from complete – further experimental work is required especially in the overdoped regime – the data now at hand are already an excellent basis for the assessment of spin-fluctuation-mediated pairing models. The recent experimental results present the following specific challenges for theoretical research.

First, based on the most recent experimental data, the interplay between spin and charge excitations, as well as the origin of the "pseudogap" and "Fermi arc" phenomena and their relationship to the phase behavior of underdoped cuprates can now be addressed in a well defined and quantitative manner. Recent analytical calculations have already yielded interesting insights in this regard [128, 130]. Numerical work on strong-correlation models, which has shown that a momentum dependent pseudogap can arise as a consequence of on-site Coulomb interactions, should now be able to compute the momentum dependent charge susceptibility, and compare the results with experimental data. Corresponding data on more weakly correlated metals such as $NbSe_2$ are available for comparison [20, 21].

Consideration of the competition between CDW order and superconductivity should lead to a systematic improvement of the Eliashberg calculations for spin-fluctuation-mediated superconductivity [33, 121]. Along the same lines, theoretical work is required to explore the suitability and limitations of the spin-fermion model underlying these calculations from microscopic Hamiltonians such as the Hubbard model [1]. Analytical or numerical calculations of the Hubbard model should explore whether such models can explain the experimentally observed persistence of magnon-like excitations up to at least $p \sim 0.3$ [34].

The latest set of IXS measurements on acoustic-phonon energies and linewidths [22] show that a complete understanding of the underdoped cuprates will require detailed knowledge of the electron-phonon interaction. Combined with ARPES data on low-energy anomalies in the electronic band dispersions, the IXS data will enable a new approach to the evaluation of the electron-phonon coupling strength. Although earlier INS measurements on Cu-O bond-bending and -stretching vibrations at energies 40-80 meV had yielded evidence of substantial electron-phonon interactions [131–135], it has been difficult to correlate these data quantitatively with ARPES, because multiple closely spaced phonon modes and spin fluctuations are present in this energy range. The data on acoustic phonons now allow a detailed evaluation of the coupling constants in the different channels (s- and d-wave Cooper pairing and CDW formation).

Collective excitations (such as phasons and amplitudons) associated with charge density wave order, their hybridization with phonons, and their influence on thermodynamic and transport properties should be explored theoretically. Specific predictions for the RIXS cross section of such excitations will also be useful. The influence of disorder on incommensurate CDW correlations and on the thermodynamic and transport properties should be addressed, in a manner analogous to recent work on disordered incommensurate magnets [44, 57]. Specifically, it should be interesting to explore how disorder influences the nature of the thermodynamic

singularities associated with CDW formation.
What is the origin of the differences in spin- and charge-ordering patterns in the different cuprate families? The "striped" phase with combined uniaxial spin and charge order in the 214 materials, which has drawn an enormous amount of attention over the past two decades, now appears to be atypical for the cuprates, whereas a spin-gapped state with biaxial incommensurate charge order appears to be generic. Note, however, that the total amplitude of the charge modulation in the striped and CDW states is closely similar [137]. In addition to differences in the Fermi surface geometry (especially the nesting conditions) in different compounds, theoretical work should address the role of soft phonons associated with low-temperature structural phase transitions, which are unique to the 214 family [136].
Finally, both experimental and theoretical research is required to finally settle the issue of the theoretically predicted loop-current order with $\mathbf{q} = 0$ [138]. Progress in experimental research is required to resolve the apparent contradiction between elastic neutron scattering experiments (based on which discovery claims have been made), and NMR and μSR experiments (which have led to null results). Likewise, theoretical work has led to divergent claims about the stability of loop-current order, which need to be conclusively resolved.

7 Outlook

This brief review shows that we have come a long way in our understanding of the electronic properties of the cuprates, and of the mechanism of high-T_c superconductivity. Of course, bona-fide predictions will be the ultimate test of any theoretical understanding we believe to have gained. The recently acquired capability to synthesize transition-metal oxide heterostructures and superlattices with atomic-scale precision [139, 140] now offers many new perspectives in this regard. Promising proposals include the manipulation of charge transfer across metal-oxide interfaces [141, 142], and "re-engineering" of the cuprate Hamiltonian in nickelate perovskites by strain and spatial confinement [143]. The latter proposal has stimulated efforts to control the effective dimensionality [144], orbital degeneracy [145], and phase behavior [146] in nickelates. Superconductivity has not yet been observed, but it is still early days.
Dynamical control of the cuprates by light stimulation is another field that is just beginning to be explored. Among the early achievements in this newly emerging field are photoinduced superconductivity in a stripe-ordered 214 compound [147] and the enhancement of T_c in underdoped $YBa_2Cu_3O_{6+x}$ [148]. With further development, controlled pumping of single excitations may ultimately develop into a powerful new way of testing theories of correlated-electron materials.

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