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Christensen, Niels Bech; Chang, J.; Razzoli, E.; Bator, M.; Niedermayer, C.; Lefmann, K.; Ronnow, H.M.; McMorrow, D.F.; Schneidewind, A.; Link, P.; Hiess, A.; Boehm, M.; Mottl, R.; Pailhes, S.; Oda, M.; Ido, M.; Momono, N.; Mesot, J.

Published in:
Physical Society of Japan. Journal. Supplement

Publication date:
2011

Document Version
Publisher's PDF, also known as Version of record

Proc. Int. Workshop NASCES11
J. Phys. Soc. Jpn. **80** (2011) SB030

Magnetic Field-Induced Closure of the Spin Excitation Gap near Optimal Doping in $La_{2-x}Sr_xCuO_4$

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(Received April 30, 2011)

We have investigated how a magnetic field applied perpendicular to the CuO_2 planes of the near-optimally hole-doped high-temperature superconductor $La_{1.855}Sr_{0.145}CuO_4$ ($T_c \approx 36$ K) influences the low-energy magnetic excitation spectrum. Our detailed single-crystal neutron scattering experiments reveal that the gap to magnetic excitations falls off linearly with increasing field and reaches zero at the magnetic field $\mu_0 H_s$=7±1 T required to induce long-range incommensurate magnetic order. A comparison with the electron-doped cuprate $Nd_{1.85}Ce_{0.15}CuO_4$ is made and the possible link between field-induced magnetic order and Fermi surface reconstruction in cuprates is discussed.

KEYWORDS: neutron scattering, high-temperature superconductors, magnetic excitations, quantum oscillations, quantum phase transitions

1. Introduction

In the past decade, many neutron scattering studies focused on magnetic field effects on magnetic order and low-energy spin fluctuations in superconducting (SC) $La_{2-x}Sr_xCuO_4$ (LSCO).[1–7] These experiments probed the field-dependence of the intensities $S(Q, \omega)$ of a quartet of incommensurate (IC) peaks at wavevectors Q_{IC}, which in square lattice notation are $\pi(1\pm\delta, 1)$ and $\pi(1, 1\pm\delta)$. In the underdoped regime of LSCO, the incommensurability $\delta \approx x$.[8] For LSCO, x equals the concentration p of holes per copper, i.e. $\delta \approx p$. The magnetic field studies have promoted an improved understanding of the role of competing order parameters in cuprate high-temperature superconductors and may also provide a crucial link in attempts to understand emerging evidence[9–16] for Fermi surface reconstruction.

For hole-concentrations $p = x > x_s = 0.13$, the zero-field ground state of LSCO is superconducting and non-magnetic with a gap Δ_{SG} to the lowest-energy IC spin fluctuations: For $T << T_c$ no intensity is observed for neutron energy transfers $\hbar\omega < \Delta_{SG}$. This so-called *spin gap* is commonly associated with superconductivity since (i) signatures of the opening of a gap are observed below T_c[17,18] and since (ii) the spin-gap energy Δ_{SG} scales with T_c for optimally doped and overdoped samples (See e.g. ref. 19). For $x < x_s$, on the other hand, superconductivity coexists with IC magnetic order in the zero-field ground state. The magnetic order is similar to that which was originally observed in the *stripe* state of $La_{1.48}Nd_{0.40}Sr_{0.12}CuO_4$[20] and subsequently in $La_{1.875}Ba_{0.125}CuO_4$,[21] but is characterized by a smaller value of the ordered magnetic moment.[6] In this underdoped regime, any gap to IC magnetic excitations is, at the very least, strongly suppressed towards low energies.[19,22,23)]

Application of a magnetic field perpendicular to the CuO_2 planes of LSCO enhances magnetic Bragg peak intensities for $x < x_s$[4,6)] while samples in the range between x_s and optimal doping $x_{opt} \approx 0.16$ have been shown to exhibit quantum phase transitions (QPT's) at finite transition fields H_s to the state with coexisting superconductivity and IC magnetic order.[5,6)]

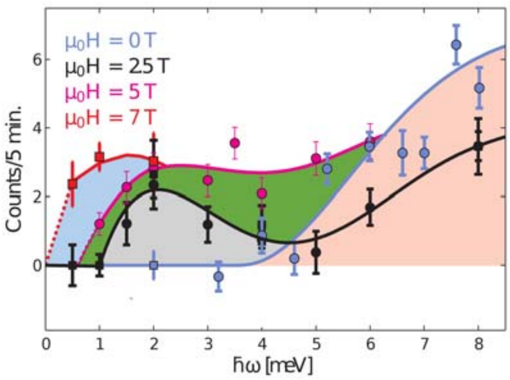

Fig. 1. Inelastic neutron scattering response at Q_{IC} as a function of energy transfer $\hbar\omega$ for $\mu_0 H$ = 0 T (blue), 2.5 T (black), 5 T (pink) and 7 T (red). All data are taken at base temperature, T = 1.5 K. Square points are from Q-scans and circular points are from 3-point scans. All lines are guides to the eye. We define the gap as the energy scale below which no intensity can be observed. The $\mu_0 H$ = 0, 2.5 and 7 T data are reproduced from ref. 7.

Using triple-axis neutron scattering, we have previously[7)] studied the relation between the regimes above and below x_s by probing the low-energy IC magnetic fluctuations of $La_{1.855}Sr_{0.145}CuO_4$ [$T_c(\mu_0 H\text{=0T}) \approx 36$ K] as it approaches its QPT at $\mu_0 H_s = 7\pm1$ T[6)] to the coexistence state. Starting from a fully gapped, magnetically disordered state at 0T, our data showed that the gap $\Delta(H)$ to excitations drops from $\Delta_{SG} = \Delta(H\text{=0T}) = 4\pm0.5$ meV to zero at a field which within the sensitivity of the experiment equals H_s.[7)] Given that H_{c2} can exceed 50 T for LSCO near optimal doping,[24)] the vanishing of $\Delta(H)$ at $H_s \ll H_{c2}$ proves that the gap is very strongly affected by proximity—as a function of carrier doping and magnetic field—to a phase with magnetic order.

We interpreted[7)] the closure of the spin excitation gap at H_s as a consequence of Goldstone's theorem which requires gapless excitations when continuous spin rotation symmetry is broken by the onset of magnetic order. For a second order phase transition, the gap $\Delta(H)$ is expected to close gradually as a function of field in such a scenario. An alternative view was suggested by recent QMC calculations in a spin-only model.[25)] Inspired by insights from a Ginzburg-Landau theory for coupled SC and spin density wave (SDW) orders[26)], the QMC work showed how vortices can nucleate slow magnetic fluctuations that are reflected in the emergence and growth of a spectral peak at energies below the zero-field gap Δ_{SG}. On the experimental side, a careful study of the evolution of the low-energy spin excitation spectrum across x_s in zero magnetic field also lead to a view of the spin excitation spectrum as consisting of two components—one spin-gapped and the other (observable below x_s) gapless.[22)] The spectrum $S(Q_{IC}, \hbar\omega)$ at 2.5 T obtained in ref. 7 and reproduced in Fig. 1, indeed displays an apparent local maximum near 2 meV and a local minimum near 4 meV, possibly reflecting the presence of two spectral components with different H-dependences. It is however also possible that the detailed form of the spectrum at 2.5 T is affected by limited counting statistics in ref. 7. To further complicate matters, similar in-gap states were previously reported for x = 0.163 (ref. 1) but were not observed at higher doping levels.[2,3)]

2. Data and Discussion

To test whether one or two components are present in the spin excitation spectrum of $La_{1.855}Sr_{0.145}CuO_4$, we performed further neutron scattering experiments using the same crystals and experimental setup of the IN14 triple-axis spectrometer at ILL, Grenoble, as in our previously published work.[7)] Our new data with improved statistical quality for the spectrum $S(Q_{IC}, \hbar\omega)$ at 5 T and for the H-dependent intensity $S(Q_{IC}$, 4 meV) are presented in Figs. 1 and 2, respectively. The smooth H-dependence at 4 meV and the absence of a low-energy peak in the 5 T spectrum both point towards a gradual closure of the gap without a low-energy spectral feature—at least for fields close to H_s. From the 5 T data in Fig. 1, we deduce a small but finite gap $\Delta(H\text{=5T}) = 0.5\pm0.5$ meV, while Fig. 2 shows that the spin gap $\Delta_{SG} = \Delta(H\text{=0T})$ must be lower than given in ref. 7. Combining the data in Figs. 1 and 2, our best estimate for the spin gap is a slightly lower value, $\Delta_{SG} = 3.25\pm0.75$ meV. Considering the available data from this experiment and ref. 7 in their totality, we plot the H-dependence of the excitation gap in Fig. 3. The gap falls off linearly with slope $d\Delta/d(\mu_0 H) = -0.35(7)$ meV/T. It is natural to compare this observation with the case of optimally electron-doped $Nd_{1.85}Ce_{0.15}CuO_4$ (NCCO) where the excitation gap at $Q=(\pi,\pi)$ extrapolates to zero near the mean field upper critical field H_{c2},[27)] leading to a more gradual drop of the excitation gap $d\Delta/d(\mu_0 H) \approx -0.2$ meV/T (See Fig. 3).

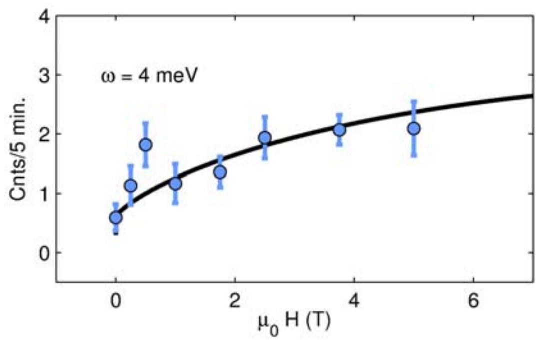

Fig. 2. Magnetic field dependence of the inelastic neutron response at Q_{IC} with $\hbar\omega$ = 4 meV. The symbols have the same significance as in Fig. 1 and data are taken at T = 1.5 K.

The Ginzburg-Landau framework relates the appearance of SDW order to changes in Fermi surface topology.[28]. We conclude by discussing how our observations of field-induced spin-density-wave order and excitation gap closure may also be relevant for the understanding of transport properties in hole-doped cuprates with $p \approx 1/8$. The combination of high-field quantum oscillations with negative Hall and thermopower coefficients has been interpreted in favor of small electron pockets in $YBa_2Cu_4O_8$ ($p = 0.14$).[15] and in $YBa_2Cu_3O_{7-y}$ (YBCO), with the oxygen content tuned such that the hole-doping level $p = 0.10$–0.12.[9–14] At $p \approx 1/8$, there is a striking similarity of the thermopowers S observed in YBCO and the stripe-ordered compounds $La_{2-x}Ba_xCuO_4$, $La_{2-x-y}Nd_ySr_xCuO_4$ and $La_{2-x-y}Eu_ySr_xCuO_4$. In all these compounds the thermopower undergoes a sign change around T=50 K and $S/T \rightarrow -0.5\pm0.2$ μV/K for $T \rightarrow 0$.[11] Notice also that the doping dependence of the thermopower is similar in YBCO and $La_{2-x-y}Eu_ySr_xCuO_4$.[12] It was therefore suggested that the mechanism responsible for the formation of electron pockets is similar in Yttrium and Lanthanum-based cuprates.[11,12]

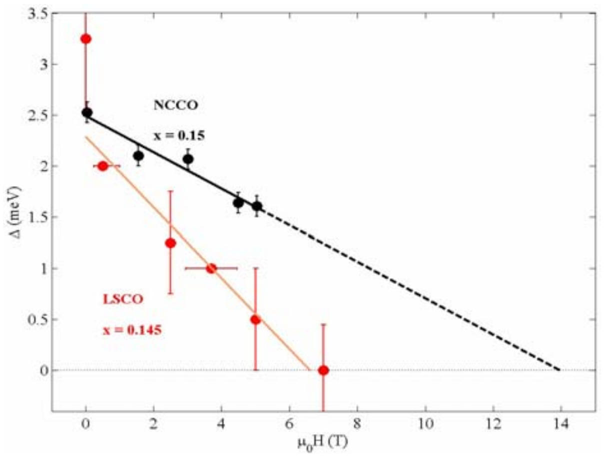

Fig. 3. Field-dependence $\Delta(H)$ of the excitation gap of LSCO (this work) and NCCO (ref. 27). The cutoff of intensity at $\hbar\omega = \Delta(H)$ is much cleaner in NCCO, as reflected in error bars which are smaller for the NCCO data despite the worse experimental energy resolution of that experiment.

Broken symmetries can introduce new periodicities that may in turn lead to Fermi surface reconstruction. In stripe-ordered compounds the IC spin and charge modulations[20,21,29] break the translational symmetry of the lattice and either can offer a plausible explanation for observations of electron pockets.[30] Indeed, recent NMR results on YBCO (p=0.108 and p=0.12) suggest that charge stripe order without static magnetic order is induced at fields matching those at which quantum oscillations are observed.[31] Two sets of neutron scattering experiments on LSCO samples with nominal hole-doping level close to 0.145[5,6–7] demonstrate that the application of a magnetic field can induce long-range spin-density-wave order in samples where no such order exists in zero field. Neutron scattering studies of YBCO reveal IC short-range order that can be enhanced by a magnetic field for $p \leq 0.082$,[32–34] but in the doping range relevant for understanding quantum oscillation experiments (p=0.10–0.14), spin modulations that are static on the time-scale probed by neutrons have yet to be uncovered. It is conceivable that field-induced spin stripe order exists for p near 1/8 in YBCO as well, but that the relevant critical field H_s is beyond reach of current magnet technology at neutron scattering facilities. Note, however, that our results on near-optimally doped LSCO show that a tendency towards SDW ordering is clearly visible already at fields below H_s, namely in the field-dependence of the excitation gap $\Delta(H)$. If related physics governs YBCO around $p = 1/8$, it is then plausible that signatures of gap closure in a field is already within experimental reach.

Turning to electron-doped cuprates, the field-induced softening of the excitation gap in NCCO with $p = 0.15$ (see Fig. 3) was interpreted[27] in terms of a weakening of the SC order parameter as the gap extrapolates to zero around H_{c2}, but with no role played by magnetic order. From the similarity with LSCO it is, however, also possible that the softening of the spin excitation gap is the consequence of approaching a magnetically ordered phase as the field is increased. This would be consistent with transport experiments[16] performed in the field-induced normal state of NCCO, which do in fact provide strong evidence for Fermi surface reconstruction consistent with the commensurate $Q = (\pi, \pi)$ order parameter characterizing the antiferromagnetic phase of NCCO with $p < 0.134$[35] as well as the low-energy magnetic excitations in superconducting NCCO.[27]

The possibility of tuning a homogeneous superconductor through a QPT to a state with coexisting SC and stripe/SDW order parameters was anticipated by Ginzburg-Landau theory.[26] On approaching the QPT the excitation gap was predicted to drop with a slope related to the strength of the repulsive coupling between the order parameters.[26] In turn, this suggests a stronger repulsion for nearly optimally doped LSCO than for optimally doped NCCO. On the other hand the available experimental evidence[16,27] suggests that for optimally doped NCCO, magnetic order sets in only at fields larger than H_{c2}, possibly with an intervening non-superconducting, paramagnetic phase. Naively, this experimental scenario indicates that the repulsion between SDW and SC order parameters is stronger —not weaker—in NCCO than in LSCO. To resolve this discrepancy and to arrive at a satisfying overall understanding of the relative importance of SC and SDW order parameters and the couplings between them in cuprates such as NCCO, YBCO and LSCO, an extension of the Ginzburg-Landau approach[26] to the high-field regime may be needed in addition to further experimental work on the doping and field-dependence of both magnetic order and spin excitations gaps.

3. Summary and Conclusions

In order to clarify whether the spin excitation spectrum of nearly optimally doped LSCO consists of one or two components, we revisited its dependence on a magnetic field applied perpendicular to the CuO_2 planes. Our results support a gradual closure of the excitation gap without the emergence of a second low-energy component. The excitation gap drops more rapidly in LSCO near optimal hole-doping than in NCCO at optimal electron-doping.

Taking Ginzburg-Landau theory as a point of departure, we discussed how this observation may be related to the relative strength of the coupling between magnetism and superconductivity in LSCO and NCCO. Finally, we argued that neutron scattering observations of field-induced magnetic order and excitation gap softening provide an important link in attempts to understand the evidence for Fermi surface reconstruction in the cuprates.

Acknowledgements

NBC was supported by the Danish Agency for Science, Technology and Innovation under DANSCATT. JC was supported by fellowships from the Swiss National Science Foundation and the Fonds Québécois de la Recherche sur la Nature et les Technologies (FQRNT). We thank Brian Møller Andersen for many fruitful discussions.

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