Disentanglement of pseudogaps and charge stripe order in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$

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High-temperature superconductivity, pseudogap phenomenology, and charge ordering are universal features of underdoped cuprate materials. Whereas superconductivity and charge ordering display an intertwinned competing relation, the role of the pseudogap phase remains elusive. Here we show how two distinct spectroscopic energy scales, in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, can be associated respectively to charge stripe order and pseudogap physics. This disentanglement enables an investigation of the low temperature pseudogap phase without interference from charge order. Under these conditions, a positive correlation between electron scattering and the pseudogap is found. This link to single-particle scattering suggests that the pseudogap is connected to Mott physics rather than superconductivity or charge ordering.

Partial gapping of spectral weight is found in many correlated metals[1–3]. These so-called pseudogaps are often accompanied by instabilities towards superconductivity and/or spin/charge modulations. This is, for example, the case of $NbSe_2$[4] and hole-doped cuprates[5–8] that display pseudogap phenomenology, charge-density-wave ordering and superconductivity. A fundamental question is therefore whether pseudogaps are precursors or seeds to these metal instabilities? In the high-T_c cuprates there are ongoing efforts to understand the possible relation between the pseudogap phase and superconductivity[9]. Despite relentless efforts, no consensus has been reached[10–14].

Much less attention has been given to spectroscopic signatures of density-wave orders. Recent discoveries of charge-density-wave order in $YBa_2Cu_3O_{7-\delta}$ (YBCO)[5–8] and Bi-based cuprates[20,21] has, however, made clear that charge ordering is a universal property. Here, we report an angle-resolved photoemission spectroscopy (ARPES) study of the charge stripe ordered system $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ (Nd-LSCO)[16,22]. As shown in the phase diagram (Fig. 1), this material has a strongly suppressed superconducting transition temperature, which allows a low temperature study of the pseudogap phase without complications from superconductivity. In the normal state ($T \geq T_c$), we identify two distinct spectral gaps and associate them to stripe order and the pseudogap phase, respectively. The disentanglement of these two contributions enables us to show that the pseudogap Δ^* correlates with the electron scattering rate Γ.

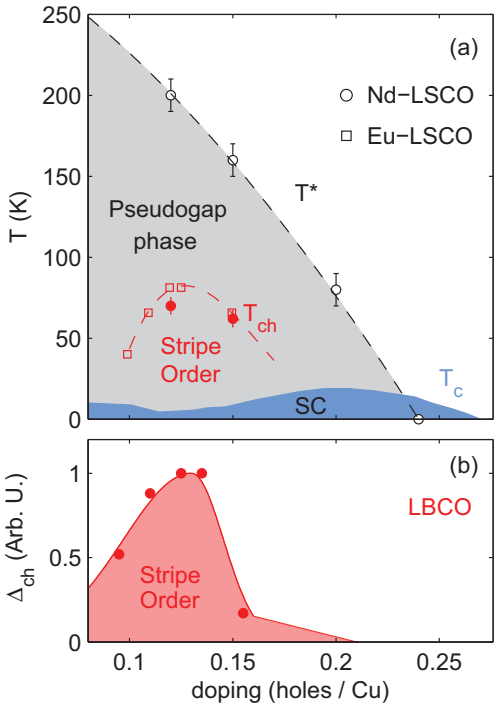

FIG. 1: (a) Temperature-doping phase diagram of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ (Nd-LSCO), established by diffraction and resistivity experiments.[15–17] The pseudogap temperature T^* is determined by the deviation from the high-temperature linear resistivity.[15] The charge ordering temperature (T_{ch}) is obtained from x-ray diffraction.[16–18] All lines are guides to the eye. (b) Charge stripe order parameter Δ_{ch} derived from hard x-ray diffraction experiments on LBCO.[19]

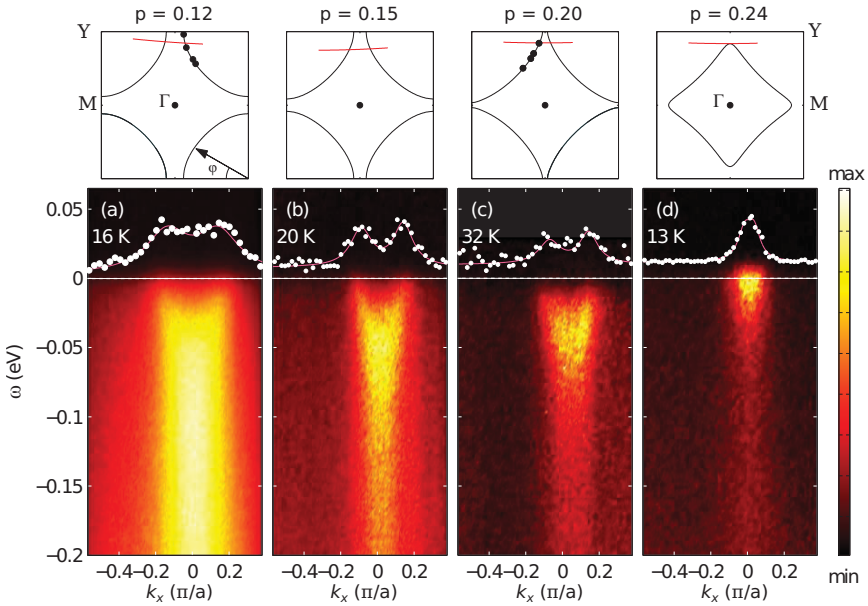

FIG. 2: (a)-(d) Anti-nodal angle-resolved photoemission spectra taken in the normal state of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ for different dopings $p = x$ as indicated. White points are momentum disctribution curves at the Fermi level indicated by horizontal dashed lines. Top panels schematically show the Fermi surface topology for each of the doping concentrations. The red lines indicate the trajectory along which the anti-nodal spectra were recorded. Solid black points indicate the underlying Fermi momenta at which symmetrized EDC's are shown in Fig. 3(b-c).

Results

Electronic structure: Normal state ($T \gtrsim T_c$) spectra taken in the anti-nodal (π,0)-region of Nd-LSCO $x = p = 0.12$, 0.15, 0.20, and 0.24 are shown in Fig. 2. As doping is reduced, spectral weight is gradually shifted to larger energies. Finite spectral weight is, however, found for all compositions at the Fermi level E_F ($\omega = 0$) even deep inside the pseudogap phase[23]. For particle-hole symmetric systems, it is possible to define the underlying Fermi momenta k_F from the maximum intensity of the momentum distribution curves (MDC) at $\omega = 0$. The Fermi surface topology of Nd-LSCO, shown schematically in Fig. 2, is similar to that of $La_{2-x}Sr_xCuO_4$ (LSCO)[24]. A van-Hove singularity crosses E_F at a doping concentration near $x = p = 0.20$, separating electron- from hole-like Fermi surfaces.

Pseudogap probed by photoemission: It is common practice to define single-particle gaps by the energy scale below which suppression of spectral weight takes place. Analysis of symmetrized energy distribution curves (EDCs) at $k = k_F$ is a standard method to demonstrate suppression of spectral weight near the Fermi level[25].

For overdoped Nd-LSCO $p = 0.24$, even the anti-nodal spectra have a Voigt-like profile (see top spectrum of Fig. 3a) at $T = 20$ K, suggesting resolution limited gapless excitations. Therefore, either the pseudogap energy scale Δ^* is much smaller than the applied resolution (15 meV - FWHM), or the pseudogap temperature T^* is lower than T_c. The latter would be consistent with resistivity measurements, that found $T^* < 1$ K for this compound[15].

At slightly lower doping ($p = 0.20$), clear suppression of spectral weight is found, for energies below $\Delta^* \sim 25-30$ meV, in the anti-nodal region for $T \sim T_c$ (Fig. 3c). Again, this is fully consistent with resistivity experiments, that established $T^* \approx 80$ K for this composition[15]. Similar ARPES spectra were obtained on Nd-LSCO with $p = 0.15$, see Fig. 3a.

Pseudogap and stripe charge order: A dramatic change of anti-nodal line shape appears when carrier concentration is tuned near to 1/8-doping. Suppression of spectral weight is greatly enhanced. As a result, the double peak structure – found for Nd-LSCO $p = 0.15$ and 0.20 – is depleted completely. This obviously complicates the interpretation of the spectra.

Another complication is that the detailed spectral line shape becomes more susceptible to the background. From one experiment to another, different signal-to-background ratios were found. As a consequence, slightly different anti-nodal line shapes were extracted, see Supplementary Information. In particular, the high-energy ($\omega >$100 meV) tails vary slightly across different experiments. Despite these variations, strong suppression of spectral weight is always observed for energies lower than $\Delta^* \sim$40 meV (indicated by black bars in Fig. 3a). However, spectral weight suppression is also present for higher energies. This additional suppression is revealed by comparison with ND-LSCO $p = 0.15$ and 0.20 (black curves

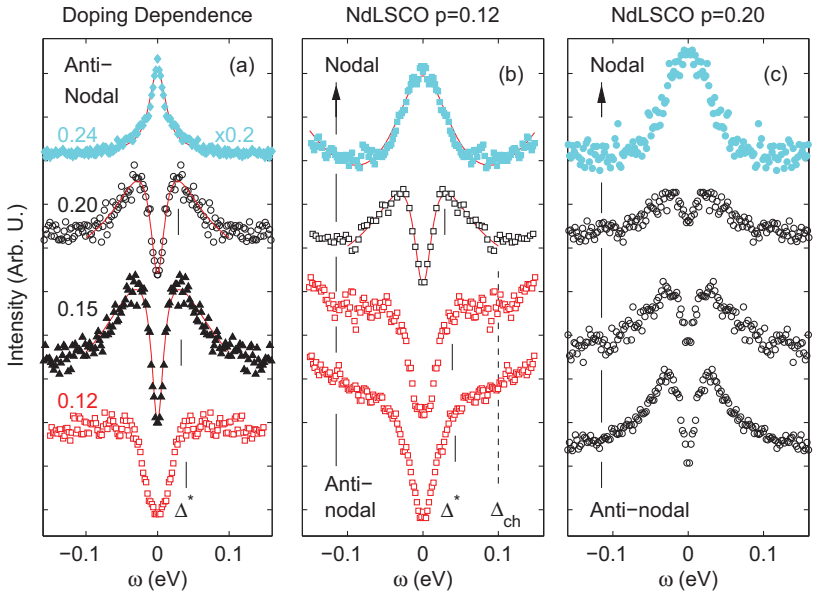

FIG. 3: Symmetrized normal state energy distribution curves (EDCs) recorded on $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. (a) Symmetrized EDC's taken in the anti-nodal region for doping concentrations as indicated. (b-c) Energy distribution curves (EDCs) symmetrized and displayed for different Fermi momenta k_F along the underlying Fermi surface of Nd-LSCO $x = p = 0.12$ and 0.20 – see top inset of Fig. 2. No background subtraction was performed. Differences between anti-nodal spectral of Nd-LSCO $x = p = 0.12$ (bottom of (a) and (b)) originate from slightly different background-to-signal ratios, see Supplementary Information. Solid red lines are guides to the eye. Blue points are used for spectra displaying gapless excitations, spectra where one gap energy-scale can be resolved are featured in black, while spectra interpreted in terms of two gaps are shown in red.

in Fig. 3a), where the spectral intensity decays with increasing $\omega > \Delta^*$. In contrast, near the 1/8-doping, the spectral intensity is either flat or displays a slight increase versus ω when $\omega > \Delta^*$. We interpret this as a consequence of spectral weight suppression and hence we conclude that two distinct gap energy scales are present at the 1/8 doping.

This is further supported by the momentum dependence shown in Fig. 3b. A systematic evolution of the line shape is found when moving from the anti-nodal to the nodal region. It resembles the doping dependence (Fig. 3a): first the double peak structure is recovered and second, upon entering the Fermi arc, gapless excitations are found[23]. For comparison, the momentum dependence of the EDC lineshapes in Nd-LSCO $p = 0.20$ is shown in Fig. 3c.

Discussion:

It is commonly assumed that the stripe ordered ground state found in Nd-LSCO is identical to that of $La_{2-x}Ba_xCuO_4$ (LBCO) and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ (Eu-LSCO) with $p = x \simeq 1/8$. All three systems have the same low-temperature tetragonal crystal structure, similar thermopower[27], and the same spin/charge stripe structure[28]. Previous ARPES studies of these stripe ordered systems[2,23,29,30] have, however, been interpreted differently[29,30]. One ARPES study on LBCO concluded, based on leading edge analysis, that the pseudogap has a maximum at the 1/8 doping[29]. This is not inconsistent with the present observation of a strongly enhanced suppression of spectral weight near 1/8-doping. However, hard x-ray diffraction studies[19] on LBCO – providing insight into the doping dependence of the stripe order parameter Δ_{ch} – lead us to re-consider the interpretation of this effect. In fact, for conventional CDW systems, the order parameter is identical to the single-particle gap[31], and Δ_{ch} scales with the lattice distortion u[31]. By measuring this distortion, it was found that Δ_{ch} has a strong doping dependence[19] (reproduced in Fig. 1b) – peaking sharply at the 1/8-doping. Just a slight increase of doping, to say $p = 0.15$, results in a single-particle gap Δ_{ch} dwarfed by a factor of five[19] (compared to 1/8-doping).

We therefore suggest that the greatly enhanced suppression of spectral weight near 1/8 doping stems from stripe order. The suppression of spectral weight, at this doping, seems to extend up to energies as large as ∼100 meV (Fig. 3b). If Δ_{ch} is five times smaller at $p = 0.15$, as suggested by x-ray diffraction[19], one would have $\Delta_{ch}(0.15) \approx 20$ meV. This energy scale is smaller but comparable to the pseudogap. For $p \geq 0.15$ it is hence difficult to extract Δ_{ch} from ARPES experiments, since this energy scale is inside the pseudogap.

Recent x-ray diffraction experiments have made clear that charge order is a common property of underdoped cuprate superconductors[5–8,16,20,21]. As discussed, charge order complicates the interpretation of the photoemis-

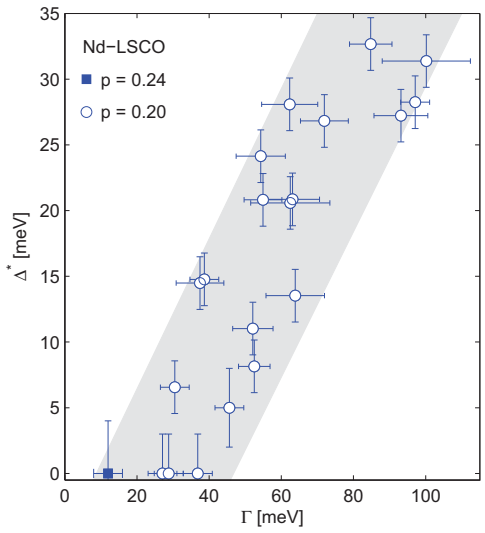

FIG. 4: Pseudogap Δ^* versus the scattering rate Γ. Both quantities were extracted by fitting symmetrized energy distribution curves along the underlying Fermi surface of Nd-LSCO $p = 0.20$ and 0.24. The fitting procedure is explained in the text. The grey shaded area indicates schematically the correlation between the pseudogap and the electron scattering.

sion spectra. Due to this fact, spectroscopic efforts to reveal the nature of the pseudogap are best carried out in regions of the phase diagram where charge / spin order is absent or negligibly weak. Nd-LSCO with $p = 0.20$ provides such a case. Although evidence for short range charge order has been observed from NQR experiments[32,33], the CDW order parameter is significantly weakened compared to 1/8 doping[19].

In absence of matrix element effects, the symmetrized intensity $I(k_F, \omega)$ is given by the spectral function $A(k_F, \omega) \sim -\mathrm{Im}\Sigma/[(\omega - \mathrm{Re}\Sigma)^2 + \mathrm{Im}\Sigma^2]$[25]. To model a spectral gap $\tilde{\Delta}$, $\mathrm{Re}\Sigma = \tilde{\Delta}^2/\omega$[35] is commonly used and $\mathrm{Im}\Sigma$ is approximated by a constant Γ[34–37]. The spectral function $A(x) \sim \gamma/((x - 1/x)^2 + \gamma^2)$ can hence be expressed by the two dimension-less quantities $x = \omega/\tilde{\Delta}$ and $\gamma = \Gamma/\tilde{\Delta}$. Our analysis is restricted to spectra for which background subtraction has little overall impact on the spectral line shape (See Supplementary Information). Furthermore, resolution effects are modelled by Gaussian convolution of the model function $A(k_F, \omega)$. In this fashion, Γ and $\tilde{\Delta}$ were extracted along the underlying Fermi surface in Nd-LSCO $p = 0.20$. The single-particle gap $\tilde{\Delta}$ is essentially identical to the energy scale Δ^* below which suppression of spectral weight is observed. To simplify the notation, we therefore use $\Delta^* = \tilde{\Delta}$. As shown in Fig. 4, a positive correlation between the pseudogap Δ^* and the scattering rate Γ is found.

We conclude by discussing possible scenarios for the pseudogap phase[9,38,39]. Experimental efforts have been made to explore possible connections between the pseudogap phase and a symmetry breaking phase transition. Evidence for broken time reversal[40,42], translational[13] and rotational symmetries[12] have previously been reported. However, to date experiments have not been able to associate a universal order parameter to the pseudogap phase. The $Q = 0$ magnetic order observed by neutron scattering[40] has, for example, not been confirmed by NMR experiments[41]. It has therefore been suggested that the neutron scattering experiments are detecting a fluctuating order[41]. In fact, pseudogaps may for two dimensional systems emerge due to strong fluctuations[9]. In this case, the pseudogap appears as a precursor to a "real" instability, at a lower temperature. Charge order and superconductivity are – as universal features of the phase diagram – candidates in this context. There exist experimental evidences suggestive of a smooth connection between superconducting gap Δ_{sc} and Δ^*[36,43]. The finite length of the Fermi arc at $T \sim T_c$ found in Nd-LSCO provides, however, no direct link between the d-wave superconducting order parameter and the pseudogap. A connection between the end-points of the Fermi-arcs and CDW scattering vectors found in Bi-based compounds[21] is also not confirmed in the Nd-LSCO system. In fact, here the energy scales associated with charge order and pseudogap physics have different doping dependence.

Finally, pseudogap phenomenology can also emerge as a result of Mott physics[47]. This scenario has been explored by different types of dynamical cluster approaches to solve the Hubbard model[45–47]. Except for charge ordering, these models capture the universal features of the phase diagram[45,47]. Furthermore, the experimental facts that the pseudogap appears in the antinodal region and decreases in strength with increased hole doping are reproduced by these models[45–47]. A connection between $\mathrm{Im}\Sigma$ and the pseudogap has also been predicted[47]. Our experimental observation of a correlation between the pseudogap and single-particle scattering is therefore consistent with the scenario where the pseudogap appears as a result of Mott physics.

Methods: Our ARPES experiments were carried out at the Swiss Light Source (SLS) on the Surface and Interface Spectroscopy (SIS) beam line[44] using 55 eV circular polarized photons. Samples of Nd-LSCO with $x = p = 0.12$, 0.15, 0.20 and 0.24 – grown by the traveling zone method – were cleaved *in-situ* under ultra-high vacuum (UHV) conditions ($\sim 0.5^{-10}$ mbar) using a top-post technique or a special designed cleaving tool[26]. Photo-emitted electrons were analyzed using a SCIENTA 2002 or a R4000 analyzer. A total energy resolution of $\sim$ 15 meV was achieved with this setup. Due to matrix element effects, all data were recorded in the second Brillouin zone but represented by the equivalent points in the first zone. The Fermi level was obtained by measuring poly-crystalline copper in thermal and electric contact with our sample. Copper spectra were also used to normalize detector efficiencies.

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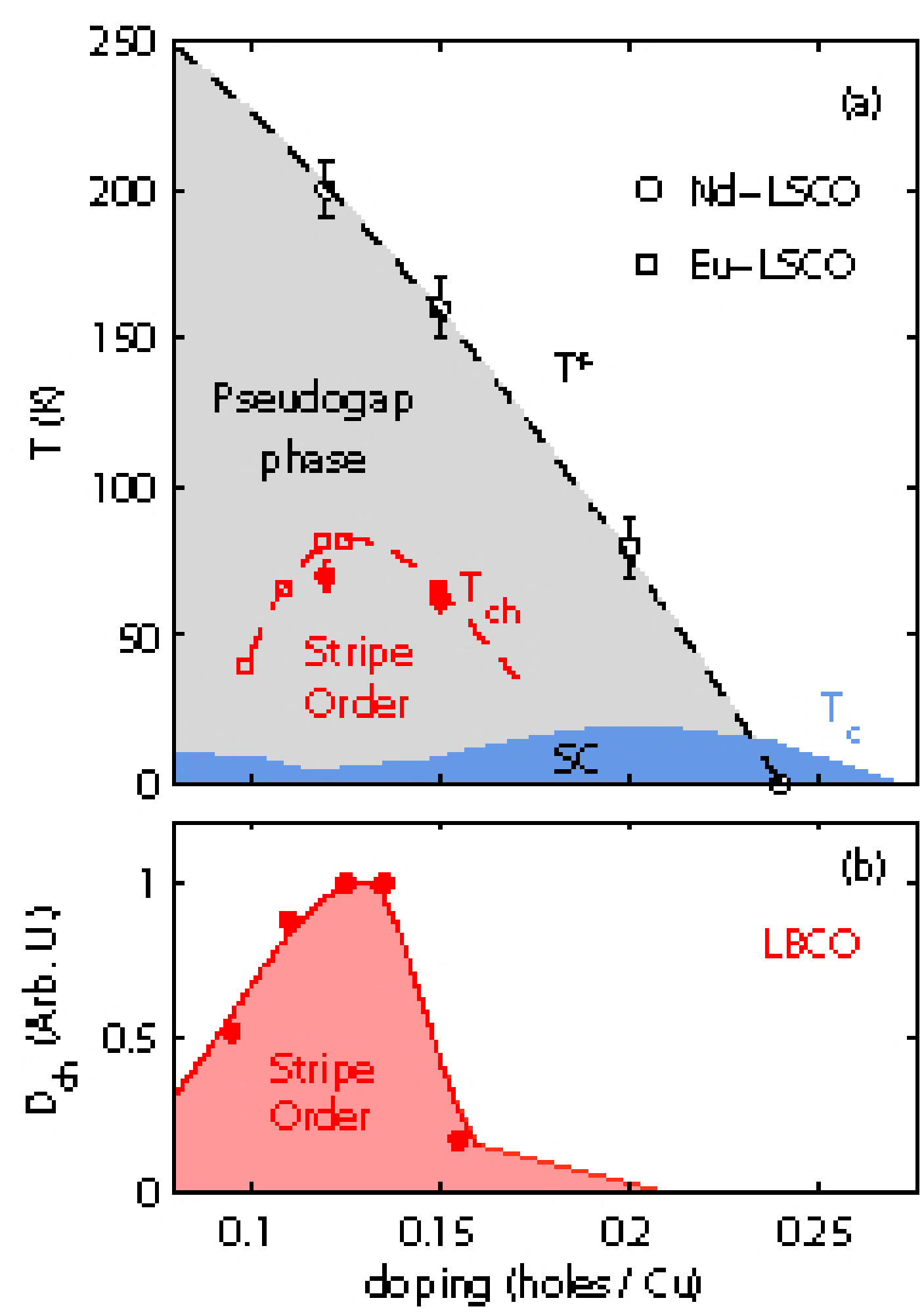
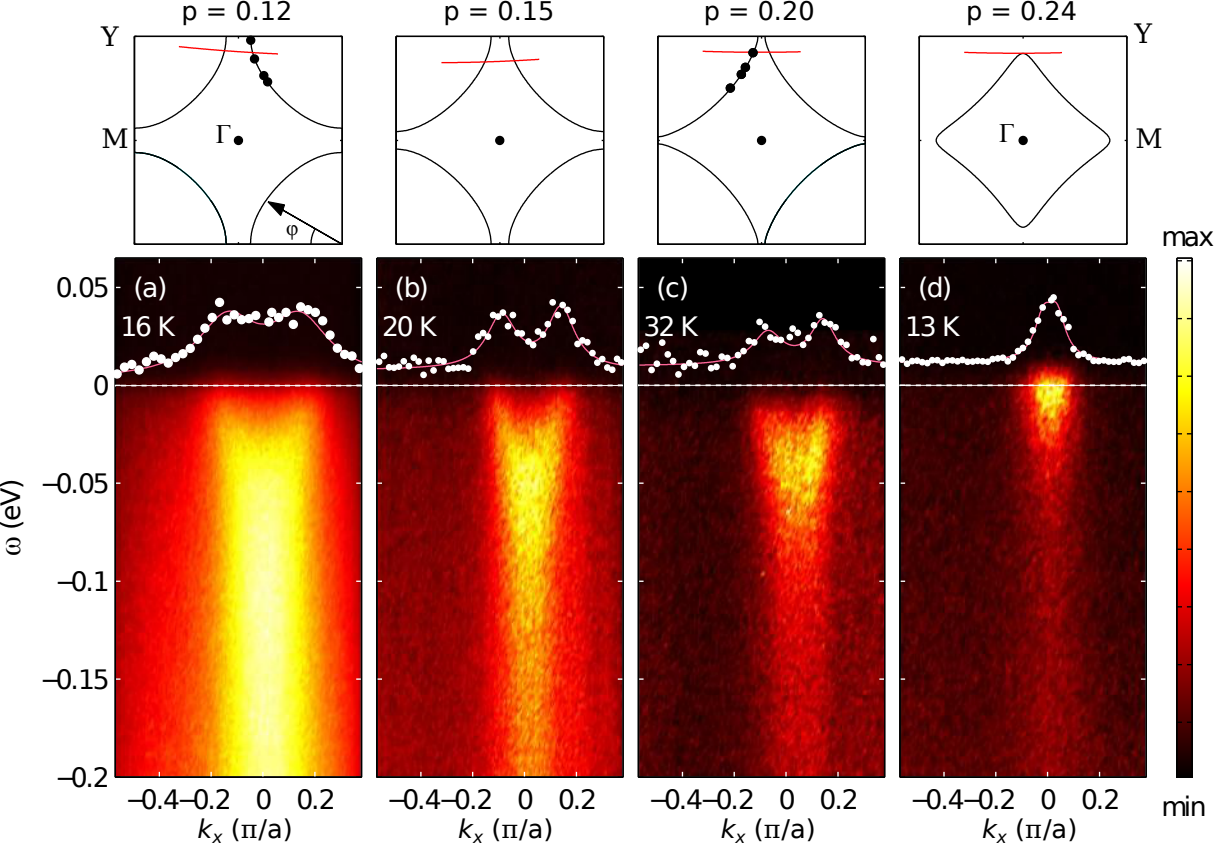
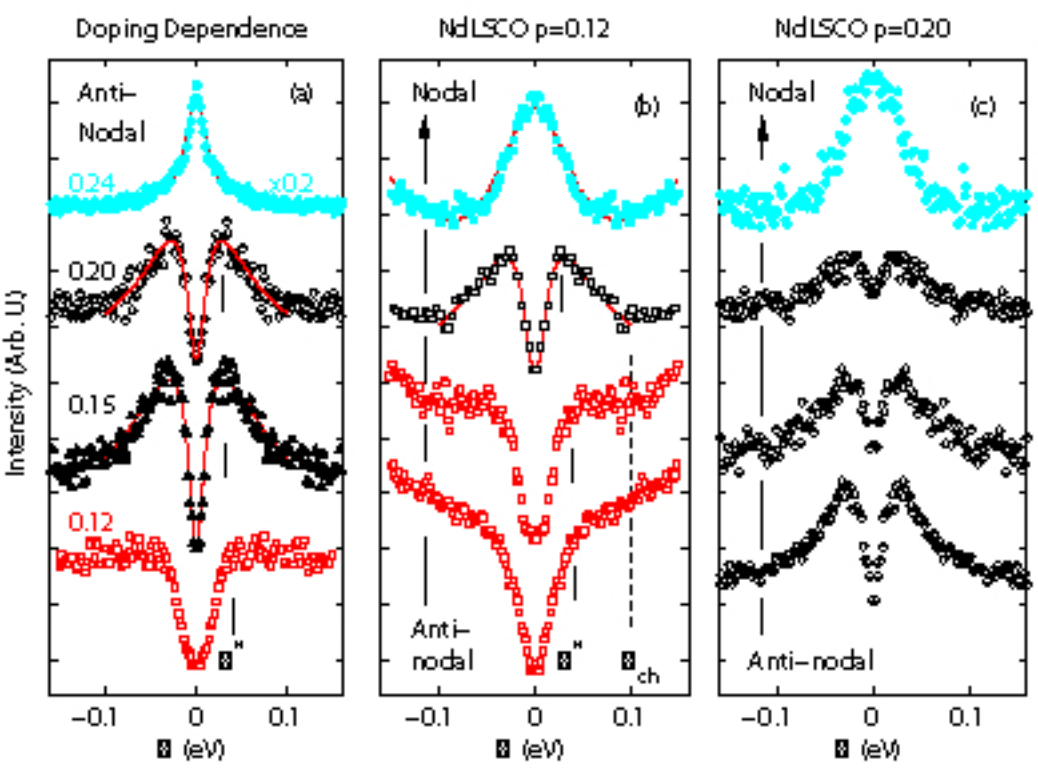
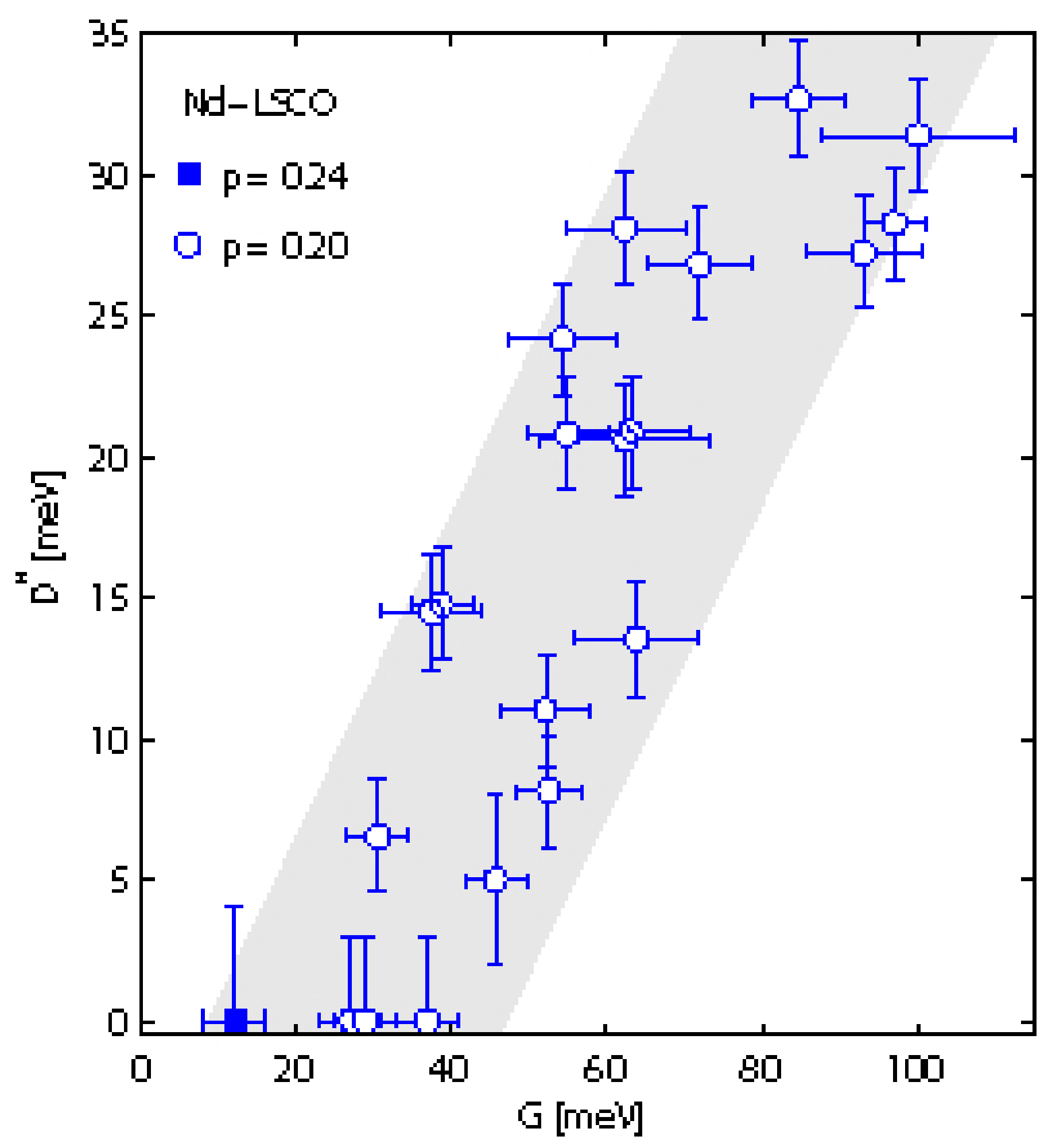

(a)
Nd–LSCO
Eu–LSCO
T*
Pseudogap
phase
T_ch
Stripe
Order
T_c
SC
T (K)
250
200
150
100
50
0
(b)
LBCO
Stripe
Order
1
0.5
0
0.1
0.15
0.2
0.25
doping (holes / Cu)


p = 0.12
p = 0.15
p = 0.20
p = 0.24
Y
M
Γ
φ
(a)
16 K
(b)
20 K
(c)
32 K
(d)
13 K
0.05
0
−0.05
−0.1
−0.15
−0.2
ω (eV)
−0.4 −0.2 0 0.2
k_x (π/a)
max
min


Doping Dependence
Nd LSCO p=0.12
Nd LSCO p=0.20
Anti-
Nodal
(a)
024
x02
0.20
0.15
0.12
Intensity (Arb. U)
Nodal
(b)
Anti-
nodal
Nodal
(c)
Anti-nodal
−0.1
0
0.1
(eV)


Nd-LSCO
p = 0.24
p = 0.20
Δ* [meV]
Γ [meV]
0
5
10
15
20
25
30
35
0
20
40
60
80
100