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Enhanced charge stripe order of superconducting $La_{2-x}Ba_xCuO_4$ in a magnetic field

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The effect of a magnetic field on the charge stripe order in $La_{2-x}Ba_xCuO_4$ has been studied by means of high-energy (100 keV) x-ray diffraction for charge carrier concentrations ranging from strongly underdoped to optimally doped. We find that charge stripe order can be significantly enhanced by a magnetic field applied along the c axis, but only at temperatures and dopings where it coexists with bulk superconductivity at zero field. The field also increases stripe correlations between the planes, which can result in an enhanced frustration of the interlayer Josephson coupling. Close to the famous $x = \frac{1}{8}$ compound, where zero field stripe order is pronounced and bulk superconductivity is suppressed, charge stripe order is independent of a magnetic field. The results for $La_{2-x}Ba_xCuO_4$ resemble recent observations in $YBa_2Cu_3O_{6+\delta}$ and, independent of potential differences in the microscopic origin of charge order in these two compounds, imply a very similar competition with three-dimensionally coherent superconductivity.

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There is mounting evidence for proximity of the superconducting (SC) ground state in the cuprates to competing states with static spin and/or charge density modulations.[1–6] A very interesting example was recently observed with soft and hard x-ray diffraction in $YBa_2Cu_3O_{6+\delta}$-based cuprates.[7–10] Around a hole concentration in the CuO_2 planes of $p = \frac{1}{8}$, both techniques detect the onset of an incommensurate charge density modulation at ∼140 K that decreases below the SC transition at $T_c \sim 65$ K, but can be enhanced if SC is weakened by a magnetic field applied perpendicular to the CuO_2 planes ($H \parallel c$). One much-discussed possibility is that the order is caused by a nesting instability associated with a reconstruction of the Fermi surface, for which there is evidence from quantum oscillation measurements.[11–13] $YBa_2Cu_3O_{6+\delta}$ also exhibits incommensurate spin correlations;[14–16] however, the magnetic wave vectors seem to be unrelated to those of the charge modulations. This conclusion is corroborated by the fact that spin excitations are gapped for $p \gtrsim 0.08$, which includes the region showing the charge modulations and quantum oscillations.[17,18]

Another competing state is the stripe phase in the La-based cuprates, which also is most stable at a hole content of $p \sim \frac{1}{8}$, where $p = x$.[19] Famous examples are $La_{1.875}Ba_{0.125}CuO_4$ and $La_{1.88-y}(Nd,Eu)_ySr_{0.12}CuO_4$ where bulk SC is strongly suppressed and replaced by an incommensurate order that has been described as an arrangement of charge stripes (or charge order, CO) separating antiferromagnetic spin stripes (spin order, SO).[4,19–21] The spin correlations resemble those in $YBa_2Cu_3O_{6+\delta}$ at lower doping;[16] however, the CO wave vector is uniquely related to the SO wave vector.[22–24] Does this mean that the charge modulations in the Y- and La-based cuprates have different origins? Understanding their physics seems crucial and may provide important clues about the SC itself.

To make progress on the stripes frontier, recent studies have focused on $La_{2-x}Ba_xCuO_4$ in high magnetic fields.[25–28] If SO and CO are indeed coupled and compete with SC, both stripe orders should increase by similar amounts in a magnetic field $H \parallel c$. The first clear evidence that this is indeed the case was obtained in strongly underdoped $La_{1.905}Ba_{0.095}CuO_4$, which is a bulk SC with weak zero-field stripe order.[26,27] This observation makes $La_{2-x}Ba_xCuO_4$ an excellent system in which to study the field effect on the CO as a function of doping.

Here, we report x-ray diffraction experiments on $La_{2-x}Ba_xCuO_4$ for $0.095 \leqslant x \leqslant 0.155$ and fields up to 10 T. We show that CO can be enhanced in a broad range of doping. The effect is particularly large in samples far away from $x = \frac{1}{8}$ where CO is weak and bulk SC strong, and is absent close to $x = \frac{1}{8}$ where CO is strong and bulk SC suppressed. It is observed only below T_c and for $H \parallel c$, which implies that stripe order emerges as the new ground state when bulk SC is suppressed. For the compositions showing the strongest effect, $x = 0.095$ and 0.155, even at $H = 10$ T the CO order parameter remains much below that at $x = \frac{1}{8}$. We have also analyzed the CO correlations between the planes, and for $x = 0.095$ we find a clear enhancement due to the field.

The $La_{2-x}Ba_xCuO_4$ single crystals with nominal Ba contents $x = 0.095$, 0.11, 0.115, 0.125, 0.135, and 0.155 are the same as in our zero-field study;[24] some of these compositions have been the subject of further characterizations.[27,28,30–35] Figure 1(f) shows the crystal structure of $La_{2-x}Ba_xCuO_4$, which differs from that of $La_{2-x}Sr_xCuO_4$ in a subtle fashion that explains their distinct behaviors.[36–38] At low temperature, $La_{2-x}Sr_xCuO_4$ assumes orthorhombic (LTO) symmetry ($Bmab$), whereas $La_{2-x}Ba_xCuO_4$ transforms from LTO to tetragonal (LTT) symmetry ($P4_2/ncm$), or a less orthorhombic (LTLO) symmetry ($Pccn$), which is a structure between LTO and LTT.[24,27] In the LTO phase, the CuO_6 octahedra tilt about [110], causing all in-plane Cu-O-Cu bonds to bend, whereas in the LTT phase they alternately tilt about [100] and [010] in adjacent planes, causing only half of all bonds to bend. This locally broken rotational symmetry of the CuO_2 planes in the LTT phase is believed to pin stripes more strongly.[19]

The x-ray diffraction experiments were performed with the triple-axis diffractometer at beamline BW5 at DESY at a photon energy of 100 keV.[39] The crystals were mounted with the $(h,0,\ell)$ zone in the scattering plane, and the magnetic field was

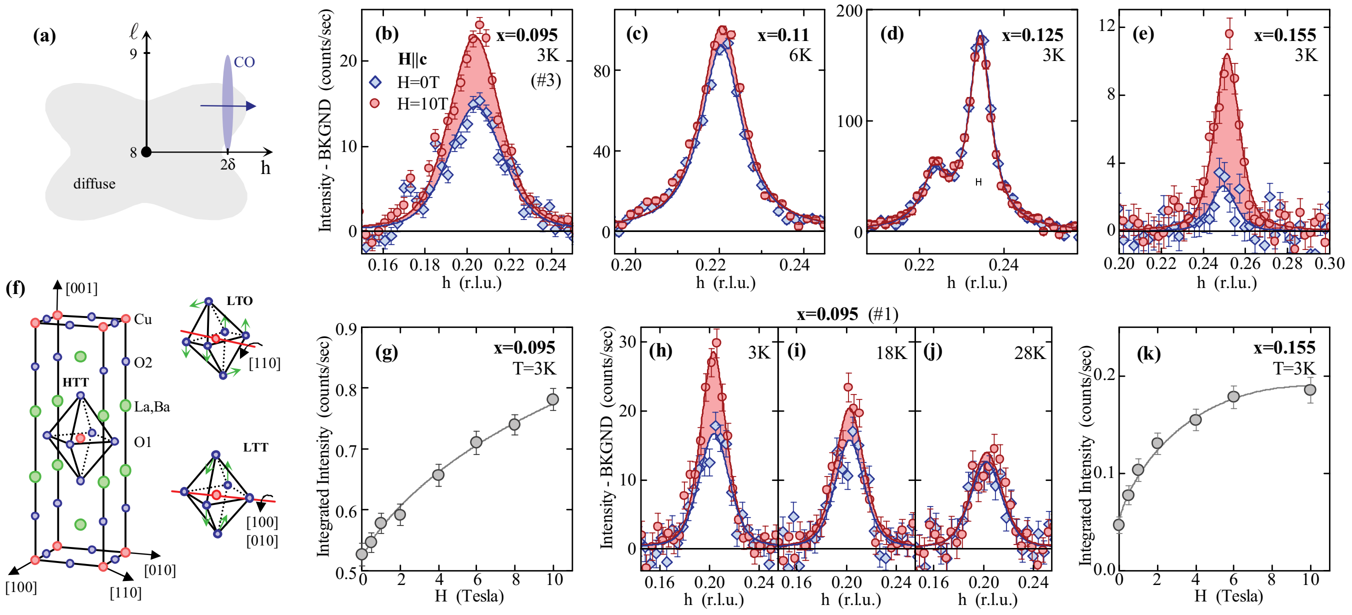

FIG. 1. (Color online) In-plane CO correlations at $T \sim 3$ K for $0.095 \leqslant x \leqslant 0.155$ and $H \parallel c$. (a) $(h,0,\ell)$ zone with CO peak at (2δ, 0, 8.5), diffuse intensity around (0, 0, 8), and typical h scan. (f) Unit cell in the HTT phase. Tilt directions of the CuO_6 octahedra in the LTO and LTT phases. (b)–(e) h scans at $H = 0$ and 10 T for different x. (h)–(j) h scans for $x = 0.095$ at different temperatures. (g), (k) Integrated intensity for $x = 0.095$ and 0.155. The solid lines are least-squares fits using $I(H) = I_0 + I_1(H/H_{c2})\ln(H_{c2}/H)$ of Ref. 29, where the upper critical field of the SC state H_{c2}, and I_0 and I_1 are parameters. In agreement with expectations, we find that H_{c2} is larger for $x = 0.095$ than for $x = 0.155$. (d) The split of the CO peak for $x = 0.125$ is caused by the crystal's mosaic (Ref. 24). The horizontal bar indicates the instrumental resolution full width at half maximum. Solid lines through the h scans are least-squares fits using a pseudo-Voigt line shape.

applied parallel to the c axes. Counting rates are normalized to a storage ring current of 100 mA. Further experimental details have been described in Ref. 24. Scattering vectors $\mathbf{Q} = (h,k,\ell)$ are specified in units of $(2\pi/a,2\pi/a,2\pi/c)$, where $a = 3.78$ Å and $c = 13.2$ Å are the lattice parameters of the high-temperature tetragonal (HTT) phase ($I4/mmm$) in Fig. 1(f). The data for $x = 0.095$ were obtained in three experiments, which we indicate by numbers (#) in the figures.

The CO leads to weak satellites about the fundamental reflections with ordering wave vectors $\mathbf{Q}_{\mathrm{CO}} = (2\delta,0,0.5)$ and $(0,2\delta,0.5)$. To study the CO within the CuO_2 planes, we have performed h scans through the satellite at $(2\delta,0,8.5)$, indicated in Fig. 1(a). Figures 1(b)–1(e) display data at $H = 0$ and 10 T applied $\parallel c$ for four dopings at base temperature. Obviously, 10 T results in large intensity gains for $x < \frac{1}{8}$ and $x > \frac{1}{8}$, but does not affect peak positions. In particular, for $x = 0.095$ the CO peak increases by ~50%, and for $x = 0.155$ by ~200%. The field effect decreases toward zero as $x \to \frac{1}{8}$, as is shown in Figs. 1(c) and 1(d). Already at $x = 0.11$ the enhancement is very small (< 10%). But, it was confirmed to be finite in a second experiment. Additional data (not shown) for $x = 0.115$ and 0.135 show no effect.

The detailed field dependence of the integrated intensity for $x = 0.095$ and 0.155 in Figs. 1(g) and 1(k) shows a strong initial increase followed by a tendency to saturate, which is similar to the SO in $La_{2-x}Sr_xCuO_4$.[1] In Ref. 29, it was predicted for a state of coexisting spin density wave and superconducting order that the SO intensity should grow as $(H/H_{c2})\ln(H_{c2}/H)$, a form that is consistent with experimental studies.[1,40,41] We find that the same functional form describes the CO data; we emphasize, however, that the model in Ref. 29 did not explicitly include any charge stripe order.

To determine whether the field affects the stripe stacking order along the c axis, we performed ℓ scans through the CO peaks at $(2\delta,0,8.5)$ and $(2\delta,0,9.5)$, as is indicated in Fig. 2(a). Again, $x = 0.095$, in Fig. 2(b), shows a strong field effect while $x = 0.125$, in Fig. 2(f), is constant. Additional ℓ scans at $h = 2\delta \pm 0.03$ were performed to estimate the background signal at $h = 2\delta$. This is particularly important for $x = 0.095$ where the CO peak is small and the background has a similar ℓ dependence due to a contribution from diffuse scattering around the Bragg peaks. The corrected data in Figs. 2(c) and 2(g) were fit to extract the peak widths, and from that the c-axis correlation lengths ξ_c in Fig. 2(e). While $\xi_c \sim 10$ Å for $x = 0.125$, which is slightly below one lattice constant, it is only half of that for $x = 0.095$ at zero field, but here can be enhanced by 50% at 10 T.

To confirm this result for $x = 0.095$, h scans at different ℓ were performed [see Figs. 2(d) and 2(h)]. If the field were to increase only the peak intensity, percentage wise it should be the same at any ℓ. This is clearly not the case. Intensity increases at the peak positions $\ell = 8.5$ and 9.5, but not at $\ell = 9$ where the tails of the peaks overlap, which implies that the peaks indeed narrow in ℓ and that ξ_c grows.

Next, we look at the T dependence. Representative h scans for $x = 0.095$ in Figs. 1(h)–1(j) indicate that the field effect is maximum at low T and disappears upon warming. Figure 3 presents more detailed data for the three dopings that show a field effect. The top panels display peak intensities at 0 (1 T for $x = 0.155$) and 10 T, the lower panels the

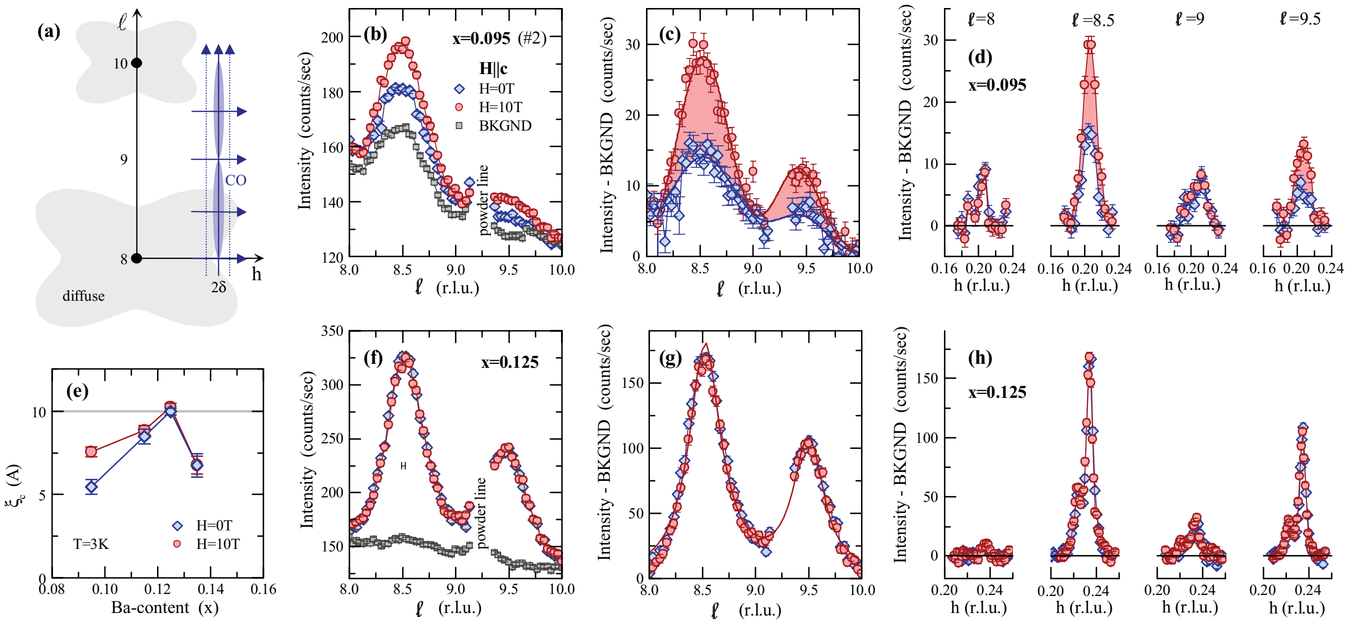

FIG. 2. (Color online) Out-of-plane CO correlations at $T = 3$ K for $x = 0.095$ and 0.155 vs field $H \parallel c$. (a) $(h,0,\ell)$ zone with CO peaks at $\ell = 8.5$ and 9.5, diffuse intensity around fundamental peaks, and typical ℓ and h scans. (b), (f) ℓ scans for $h = 2\delta$ at $H = 0$ and 10 T, including background from average of ℓ scans at $h = 2\delta \pm 0.03$. The horizontal bar in (f) indicates the instrumental resolution full width at half maximum. (c), (g) Same data after background subtraction, and with least-squares fits using a pseudo-Voigt line shape. (d), (h) h scans at various ℓ values. (e) Correlation length ξ_c vs x.

Meissner effect at 0.01 T (and 1 T for $x = 0.155$). For all three dopings, the CO depends on the field only below T_c. The $x = 0.095$ crystal displays a particularly interesting SC transition that is interrupted by the CO transition.[24,26,27,33] SC first appears at $T_{c1} = 32$ K in the non-stripe-ordered LTO phase. This SC state weakens when CO sets in at the LTO → LTLO transition at 30 K, with a corresponding reduction of the interlayer Josephson coupling.[32] (Note that $x = 0.095$ is nearly LTT.[24,27]) Then, at $T_{c2} = 27$ K, SC reappears, at which point the zero field CO saturates. Only when suppressing the SC state with 10 T, the CO peak continues to increase below T_{c2}. Note that the T dependence in Fig. 3(a) was measured with higher precision than in Ref. 27, and now reveals the impact of SC on the CO below T_{c2}. Also in the case of $x = 0.155$, where the CO was measured with a minimum field of 1 T, the onset of the field effect coincides with the SC transition measured in the same field.

Could all these effects be the result of a magneto-elastic mechanism that enhances stripe pinning? The most relevant pinning parameter of the LTT and LTLO phases is the CuO$_6$ tilt angle Φ.[37] If Φ were to increase with field, certain superstructure reflections, such as (1, 0, 0), would become stronger. We find these peaks to be independent of H, which leads us to conclude that the CO enhancement is a purely electronic effect.

In Fig. 4(a), we plot the doping and field dependence of the CO order parameter, normalized to $x = \frac{1}{8}$ in zero field. The data represent the square root of the integrated intensity of the h scans. Strongly underdoped $x = 0.095$ and optimally doped $x = 0.155$ display a strong initial increase, but tend to saturate at high fields at ∼65% and ∼30% of the full order at $x = \frac{1}{8}$, respectively. The crystals closer to $x = \frac{1}{8}$ show order parameters larger than ∼90% and either no or just a weak increase with field. (The $x = 0.115$ sample also shows no field effect, but that crystal was measured under different conditions which impedes a direct comparison.) In Figs. 4(b) and 4(c) we compare the doping dependence of the CO order parameter at $H = 0$ and 10 T with that of T_c, T_{CO}, and T_{SO} in zero field.[24] Clearly, the CO enhancement is maximum where bulk SC is strong and CO is weak. This corresponds well with the neutron diffraction data for $x = 0.095$ and 0.125 which show a similar H dependence of the SO.[33,42] The weak enhancement of the SO close to T_{SO} reported in Ref. 42 for $x = 0.125$ in high fields is not observed for the CO close to either T_{SO} or T_{CO}. We assume that the SO is stabilized not only through the suppression of SC, but also through the gain of Zeeman energy.

We note that the observed field effect could also represent a change of the stripe ordered volume fraction proportional to that of the integrated intensity. For example, if the stripe order is induced in the vicinity of magnetic vortices,[43] then the intensity might grow with the vortex density (proportional to H) until the CO correlation length[24] becomes comparable to half of the vortex spacing, which occurs near 10 T for $x = 0.095$ and 0.155. Of course, any evaluation of volume fractions would depend on the local maximum order parameter, which we do not have independent knowledge of.

The enhancement of CO at high magnetic fields in La$_{2-x}$Ba$_x$CuO$_4$ over a broad range of x is a long-sought-for confirmation of the strong coupling between stripe type charge and spin orders in La-based cuprates. In the case of La$_{2-x}$Sr$_x$CuO$_4$ several neutron scattering experiments of the past decade have shown that SO can be enhanced by a field $H \parallel c$.[1,40,44] However, there had been no evidence of CO

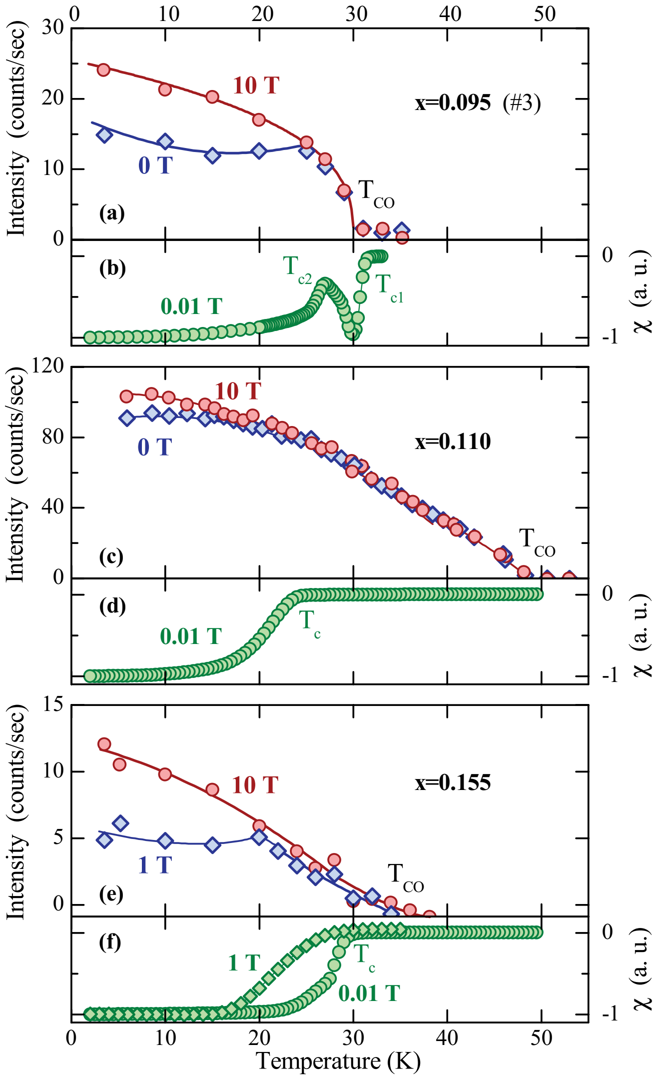

FIG. 3. (Color online) T dependence of CO and SC for $x = 0.095$, 0.11, and 0.155 and $H \parallel c$. (a), (c), (e) Peak intensity of (2δ, 0, 8.5) CO peak at $H = 0$ (1 T for $x = 0.155$) and 10 T. (b), (d), (f) Normalized Meissner effect at $H = 0.01$ T (and 1 T for $x = 0.155$).

until very recent resonant soft x-ray scattering experiments revealed CO near the sample surface, but not in the bulk.[45] Our high-energy x-ray data prove that in $La_{2-x}Ba_xCuO_4$ the zero field CO and its enhancement in the field are bulk properties. The exclusive occurrence of the field effect in bulk SC below T_c, as well as on both sides of $x = \frac{1}{8}$ doping, clearly implies a competition between stripe order and SC.

Recently, it was proposed that stripe order does not suppress SC pairing correlations in the planes, but prevents three-dimensional phase coherence by frustrating the interlayer Josephson coupling.[27,30,46–48] Thus, it is possible that the field not only suppresses SC, but also enhances the interlayer CO correlations. The field-driven increase of the CO correlation length ξ_c for $x = 0.095$ is clear evidence of such an effect.

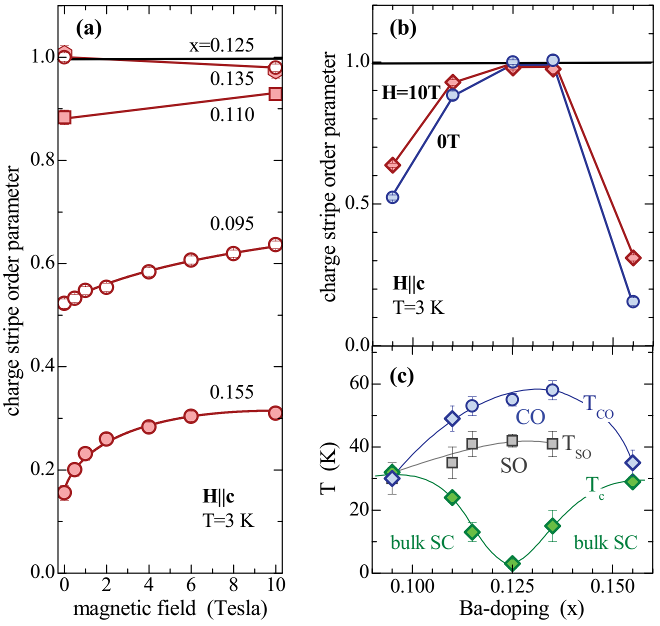

FIG. 4. (Color online) CO order parameter for $H \parallel c$ and different x at $T = 3$ K. (a) As a function of H. (b) As a function of doping at $H = 0$ and 10 T. (c) Zero-field phase diagram with critical temperatures T_c, T_{CO}, and T_{SO} from Ref. 24, except for three new T_{CO} values (diamonds) from this study. The solid lines for $x = 0.095$ and 0.155 in (a) are fits using the square root of the expression in Fig. 1. All other solid lines are guides to the eye.

In $La_{2-x}Ba_xCuO_4$, the zero-field CO wave vector is tightly linked to that of the SO, and increases with x, in agreement with the trend predicted by the stripe model.[19,24,49] Our study shows that this trend is independent of the magnetic field. Furthermore, the increase of the CO wave vector is incompatible with the decrease of the antinodal nesting vector, as measured with angle-resolved photoemission spectroscopy.[50] This is different for the checkerboard-type charge modulation in the Bi-based cuprates, and the recently discovered modulations in Y-based compounds.[2,8,51,52] There, the charge modulation wave vectors decrease with doping, and tend to agree with a Fermi-surface nesting scenario.[7–9,51,52] Thus, the sum of experiments seems to indicate a distinct nature for the stripe order in La-based cuprates, and the nesting-related charge modulations in Bi- and Y-based cuprates. However, the qualitatively same field dependence of these two states in the normal state as well as below T_c, as observed here and in Ref. 8, suggests that they depend in a similar way on the suppression of the competing bulk SC state. This makes one wonder if and how these charge modulated states are connected, which is the next piece of the cuprate puzzle to understand.

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