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Zn-Doping Dependence of Stripe Order in $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$

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Abstract The effect of Zn-doping on the stripe order in $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$ has been studied by means of x-ray and neutron diffraction as well as magnetization measurements. While 1% Zn leads to an increase of the spin stripe order, it unexpectedly causes a wipe out of the visibility of the charge stripe order. A magnetic field of 10 Tesla applied along the c-axis has no reversing effect on the charge order. We compare this observation with the Zn-doping dependence of the crystal structure, superconductivity, and normal state magnetism.

Keywords Stripes · Cuprates · Zn doping

1 Introduction

Numerous studies on high- T_c superconductors have demonstrated that the electronic properties of the CuO_2 layers are extremely sensitive to impurity doping, such as the substitution of Cu with 3d-transition metals. The most obvious effect is the suppression of the superconductivity (SC) [1]. In

addition, in several cases a ground state with static or slowly fluctuating antiferromagnetic (AF) correlations is stabilized or enhanced [2–4]. In Zn-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) the competing AF order is incommensurate, and similar to the magnetism in charge (CO) and spin (SO) stripe ordered $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ (LBCO) [5–8]. For many years it was unclear whether the two ground states are indeed identical, due to lack of evidence of CO in Zn-doped LSCO. Although the presence of CO has been expected [9], only recently it was confirmed when weak CO-reflections were identified in Zn-doped LSCO with $x \sim 1/8$ [10].

However, numerous experiments suggest that for symmetry reasons stripes are pinned more effectively in the low-temperature tetragonal (LTT) phase (space group $P4_2/ncm$) of LBCO and related compounds such as $\text{La}_{2-x-y}\text{Nd}_y\text{Sr}_x\text{CuO}_4$ [7] than in the low-temperature orthorhombic (LTO) phase of LSCO ($Bmab$). The LTT phase lifts the degeneracy of the two orthogonal directions parallel to the CuO_2 square lattice which promotes the pinning of stripes parallel to the Cu–O–Cu bonds, while the LTO phase lifts the degeneracy of the diagonal lattice directions which leaves all Cu–O–Cu bonds equivalent. However, high pressure experiments revealed that exactly at $x = 1/8$, which is the hole content most favorable for stripe order [11], CO can spontaneously break the in-plane fourfold rotational symmetry even in the high-temperature tetragonal (HTT) phase ($I4/mmm$) with average flat CuO_2 planes [12], possibly with help of local lattice distortions. Whether these distortions are LTT or LTO-like is unknown. In [13] it is reported that high (Zn, Mg)-concentrations in La_2CuO_4 ultimately stabilize the LTT phase. Thus, beside the effect of depressed SC around Zn, small amounts of Zn could locally promote LTT-like distortions that further support the pinning of stripes in the LTO phase [14].

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An interesting discussion of stripe pinning mechanisms in (Zn, Ga, Fe)-doped LSCO at $x \sim 1/8$ is given in [10]. Although any of these defects could stabilize CO, especially large effects on both CO and SO are observed for Fe-doping which introduces large magnetic spin defects ($S = 5/2$) [10]. An interesting feature of [10] is that Zn stabilizes a weak CO, but does not enhance the weak SO already present in pure LSCO. Earlier observations near optimum doping indicated a Zn-induced stabilization, in particular for the SO [5, 6].

To further elucidate the possible mechanisms by which stripes can be stabilized, we decided to study the effect of Zn-doping on LBCO, i.e., a system where CO and SO forms naturally without impurity doping [8]. Two crystals with $x = 0.095$ were studied, one with 0% Zn (LBCO95) and the other with 1% Zn (LBCZO95). Note that the crystals low- T structure is not perfectly LTT, but assumes the so-called less-orthorhombic low-temperature (LTLO) phase ($Pccn$), which is an intermediate phase between LTO and LTT with reduced orthorhombic strain. However, at base temperature the remaining strain is extremely small, and the crystals are very close to LTT [11]. In LBCO95 stripe order is much weaker than at $x = 1/8$ [11], which enables us to identify both possible scenarios: The enhancement or the degradation of CO and SO order by Zn-doping. Indeed we found a Zn-induced increase of the SO, which led us to expect that the CO order will increase as well. To our surprise, however, the opposite is true in our LBCZO95 crystal. Possible reasons will be discussed.

2 Experimental

The LBCO95 and LBCZO95 crystals were grown with the traveling-solvent floating-zone technique. The stripe order, structure and resistivity of the LBCO95 crystal were subject of two further studies [11, 15]. The x-ray diffraction (XRD) experiments were performed with a triple-axis diffractometer at wiggler beamline BW5 at DESY, using 100 keV photons in transmission geometry; the neutron diffraction (ND) experiments with triple-axis spectrometer HB1A at the High Flux Isotope Reactor, Oak Ridge National Laboratory, using beam collimations of $48' - 48' - S - 40' - 136'$ with a fixed incident energy $E_i = 14.6$ meV. Scattering vectors $\mathbf{Q} = (h, k, \ell)$ are specified in units of $(2\pi/a, 2\pi/a, 2\pi/c)$, where $a \simeq 3.78$ Å and $c \simeq 13.2$ Å are the lattice parameters of the HTT phase. The DC magnetic susceptibility $\chi = M/H$ in the SC state as well as in the normal state has been analyzed with a SQUID-magnetometer for H parallel and perpendicular to the CuO_2 planes.

Fig. 1 (Color online) Normalized field cooled susceptibility vs. T of pure and Zn-doped $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$ at $H = 100$ G applied parallel ab -plane and c -axis

3 Results

3.1 Superconductivity

First we look at the SC properties which have been characterized with $\chi(T)$ measurements at $H = 100$ G, and are displayed in Fig. 1 on normalized χ scale. Obviously, LBCO95 shows a peculiar sequence of transitions, discussed in [11]. At $T_c \simeq 32$ K the sample turns bulk SC in the LTO phase. Just slightly below that temperature, at $T \simeq 30$ K, the diamagnetic signal collapses when the transitions to CO and to the LTLO phase occur; see next sections. At $T_c^* \simeq 27$ K occurs a second SC transition that coincides with the completion of the LTO $\rightarrow$ LTLO transition [11]. The LBCZO95 crystal shows a much simpler behavior. There is only one SC transition at $T_c \simeq 18$ K, which is pushed below those in LBCO95 by the Zn. Moreover, there is no clear signature of the LTO $\leftrightarrow$ LTLO transition, except for a small hump at ~ 15 K. As will be discussed in Sect. 3.2 this hump may coincide with the completion of the LTO $\rightarrow$ LTLO transition in LBCZO95.

3.2 Crystal Structure

Next we focus on the crystal structure. For the present work it is sufficient to analyze the LTO $\leftrightarrow$ LTLO transition, by following the T dependence of the $(3, 0, 0)$ peak intensity; see the reciprocal lattice in Fig. 2(b). The XRD data in Fig. 2(a) show a significant change with Zn. While the transition is sharp in LBCO95, it is very broad in LBCZO95, even though the onset temperatures are about the same. How does this affect the CO? (The results for LBCO95 were measured on the identical sample as in [11], but with different cryostat and scattering geometry, explaining the slightly different T_{LT} and width of the transition.)

Fig. 2 (Color online) Crystal structure and stripe order of pure and Zn-doped $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$. **(a)** Normalized integrated intensity from k -scans through $(3, 0, 0)$ LTLO-peak. **(b)** Reciprocal lattice of the $(h, 0, l)$ -plane in the LTLO phase. (●) Bragg, (◇) CO, and (▲) LTLO-reflections. Red arrows indicate typical scans, and blue rectangle a studied mesh. q_{CO} indicates CO ordering wave vector $(2\delta, 0, 0.5)$. **(c)** h -scans through $(2\delta, 0, 8.5)$ CO-peak at $T = 3$ K and $H = 0$ T and 10 T applied parallel c -axis. Data for 0% Zn are shifted by 30 counts/s. **(d)** k -scans through $(0.5, 0.5 + \delta, 0)$ SO-peak. Data show the difference between 10 K and 50 K. Inset: SO-peaks in $(h, k, 0)$ -plane

3.3 Spin and Charge Stripe Order

The CO leads to weak reflections with ordering wave vector q_{CO} , as indicated in Fig. 2(b). In Fig. 2(c) we present h -scans at 3 K through $q = (2\delta, 0, 8.5)$, which is among the most intense CO reflections. While for LBCO95 a CO-peak is clearly observed, in LBCZO95 it is not identifiable. This is in stark contrast to the SO-peak in Fig. 2(d), which increases with Zn-doping. When applying a field of 10 Tesla parallel to the c -axis, the CO-peak in LBCO95 grows while it has no effect on LBCZO95. To check whether Zn-doping caused the CO-peak position to shift, we performed scans in a wider range, and indeed found some broad peaks that are displayed in Fig. 3(a). However, these weakly T depen-

dent features turned out to be the tails of the butterfly like diffuse scattering around the nearby $(0, 0, 8)$ Bragg reflection; see Fig. 3(c). LBCO95 in Fig. 3(b), (d) shows a similar diffuse scattering, but in addition two sharp superimposed CO-peaks.

3.4 Normal State Magnetic Susceptibility

The magnetic susceptibility χ of the normal state has proven a valuable tool for the analysis of the stripe phase in LBCO [11, 16, 17]. This is particularly true for $x = 1/8$ where bulk SC at $T_c \sim 3$ K occurs much below the CO and SO transitions at $T_{CO} = T_{LT} = 54$ K and $T_{SO} = 42$ K, thus presenting a broad T -window for χ -studies of the stripe phase above the bulk T_c . For LBCO95 the situation is less favorable, because at low fields $T_c = 32$ K is even slightly higher than $T_{CO} = T_{LT} \simeq 30$ K [11]. However, T_c can be pushed below T_{CO} and T_{LT} when a high magnetic field is applied. In addition, T_{SO} will shift to higher T [16]. Corresponding results for LBCO95 and LBCZO95 for both $H \parallel c$ and $H \parallel ab$ are shown in Fig. 4. The 1 Tesla low-field data in Fig. 4(a) display an overview up to 375 K. The 7 Tesla high-field data in Fig. 4(b) zoom in on the relevant low- T region. There one can see that LBCO95 shows small anomalies at ~ 33 K in both field directions. These anomalies are certainly connected to the stripe transition. But it remains unclear if they represent T_{SO} or T_{CO} , or both, and if they have indeed shifted by ~ 3 K in 7 Tesla, cf. Fig. 1. In contrast, LBCZO95 lacks any anomaly that would indicate a sharp transition, and this is consistent with the broad LTO $\leftrightarrow$ LTLO transition in Fig. 2(a). At low T LBCZO95 shows a Curie-type upturn caused by the effective paramagnetic moments induced by Zn [1, 18].

4 Discussion and Conclusions

The key result that in LBCZO95 CO-peaks are not identifiable, while SO-peaks are enhanced, can either reflect an intrinsic effect of Zn, or indicate sample specific properties such as a lower quality or a difference in the Ba content. With a mosaic of 0.01° for LBCO95 and 0.018° for LBCZO95, measured with XRD at the $(0, 0, 8)$ Bragg reflection at base temperature, both crystals demonstrate an excellent quality. Also the bulk SC transition of LBCZO95 is nearly as sharp as for LBCO95; see Fig. 1. To estimate the actual Ba content x' of LBCZO95, we apply the same strategy as in [11], which exploits the doping dependence of the HTT $\leftrightarrow$ LTO transition temperature T_{HT} . As indicated in the inset of Fig. 4(a) the crystals T_{HT} differ by ~ 18 K. With $dT_{HT}/dx \simeq -23$ K/0.01 Ba and $dT_{HT}/dz \simeq 8$ K/0.01 Zn, we calculate that LBCZO95 has a $\Delta x \simeq 0.004$ lower Ba content than LBCO95, consistent with the shift of

Fig. 3 (Color online) Diffuse scattering around $(0, 0, 8)$ Bragg reflection and $(\pm 2\delta, 0, 8.5)$ CO-peak for (c) Zn-doped and (d) pure $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$. (a) and (b) show h -scans along $(h, 0, 8.5)$ as indicated by white arrows in (c) and (d)

Fig. 4 (Color online) Normal state $\chi(T)$ of pure and Zn-doped $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$ for (a) $H = 1$ T and (b) 7 T applied parallel ab -plane and c -axis. Inset: $d\chi/dT$ for $H \parallel ab$

the SO-peak in Fig. 2(d). Absolute values were calculated to $x' \simeq 0.099$ for LBCO95 and $x' \simeq 0.095$ for LBCZO95 [11]. Does the absence of CO-peaks in LBCZO95 reflect that slightly lower Ba content, since CO rapidly decreases for

$x < 1/8$ [11]? Clearly not, since it is in discord with the observed increase of the SO. Hence, is it possible that CO is more sensitive to Zn impurities than SO? Zn introduces spin $S = 0$ vacancies to the $S = 1/2$ spin lattice, but these do

not cause strong perturbations of the AF correlations [18]. A sensible broadening of the SO-peaks only sets in for several percent Zn [19]. For charge carriers, Zn impurities act as localization centers [20], and because of their random distribution may cause significant disorder in the CO pattern. If the associated broadening of the CO-peaks is strong they may become undetectable, because at $x = 0.095$, even without Zn, CO-peaks are much smaller than at $x = 0.125$ [11]. According to the stripe model, SO and CO are intimately connected [7, 21]. Without charge stripes, which act as antiphase domain boundaries between spin stripes, the incommensurate SO would disintegrate, or become commensurate. Thus the question regarding LBCZO95 is: how disordered can the CO be until the SO collapses? Additional studies on LBCZO with $z < 0.01$ and a more pronounced initial ($z = 0$) CO, such as at $x = 1/8$, may tell.

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