Competition between Charge- and Spin-Density-Wave Order and Superconductivity in $La_{1.875}Ba_{0.125-x}Sr_xCuO_4$

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We have performed a series of elastic neutron scattering measurements on 1/8-hole doped $La_{1.875}Ba_{0.125-x}Sr_xCuO_4$ single crystals with $x = 0.05$, 0.06, 0.075, and 0.085. Both charge-density-wave (CDW) and spin-density-wave orders are found to develop simultaneously below the structural transition temperature between the low-temperature orthorhombic (LTO) and low-temperature tetragonal (LTT) or low-temperature less-orthorhombic (LTLO) phases. In the ground state the CDW order is observed only in the LTT/LTLO phase and drastically degrades towards the LTO boundary. The x dependence of T_c strongly suggests a direct effect of the CDW order on the suppression of superconductivity.

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The interplay between magnetism and superconductivity is a central issue in high-T_c superconductivity [1]. Neutron scattering measurements on the superconducting $La_{2-x}Sr_xCuO_4$ (LSCO) system have shown dynamical incommensurate (IC) magnetic correlation [2–4]. The linear doping dependence of incommensurability with the superconducting transition temperature (T_c) in the underdoped region suggests a relationship between magnetic correlation and superconductivity [5].

On the other hand, it is well known that superconductivity in the $La_{2-x}M_xCuO_4$ ($M = \mathrm{Ba, Sr}$) system is anomalously suppressed at the specific hole concentration of $x \sim 1/8$. For the $La_{2-x}Ba_xCuO_4$ (LBCO) system, this 1/8 anomaly is accompanied by an occurrence of the LTT phase with $P4_2/ncm$ symmetry [6–8]. Superconductivity in the LTO phase of LSCO with $Bmab$ symmetry is weakly suppressed in comparison with the LBCO system [9,10]. Recently, the IC spin-density-wave (SDW) and the charge-density-wave (CDW) orders were discovered in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ (LNSCO) with $x \sim 1/8$. These provide important clues for understanding the mechanism of the 1/8 anomaly based on the stripe model [11,12]. In the framework of the stripe model it is recognized that the orders are a manifestation of dynamical spin/charge correlation and that stabilization of the orders competitively induces instability of superconductivity [13]. In the low-temperature tetragonal (LTT) phase stripe-shaped orders parallel or perpendicular to Cu-O bonding are favorably stabilized by the corrugated pattern of the in-plane lattice potential. Hence, T_c is suppressed to a greater extent in the LTT phase than in the low-temperature orthorhombic (LTO) phase. Whether visible signs, such as the anomalous suppression of T_c, appear depends on the stability of CDW/SDW orders. Therefore investigation of the relationships between the crystal structure, CDW/SDW orders, and superconductivity provides the insight necessary to clarify the mechanism of T_c suppression.

In order to determine the nature of the aforementioned relationships, we performed systematic elastic neutron scattering measurements on a series of $La_{1.875}Ba_{0.125-x}Sr_xCuO_4$ (LBSCO) single crystals. As an experimental advantage in this system variation of the Ba/Sr ratio can modify the crystal structure from the LTT to the LTO phase while maintaining a constant carrier concentration [14]. Therefore, LBSCO is a favorable compound for investigating the physical properties of a 1/8-doped system in different crystal structures. Furthermore, the LBSCO system is free from large rare-earth magnetic moments such as Nd spins in LNSCO. Our measurements yield two important results: (i) a direct relation between CDW order and suppression of superconductivity and (ii) the coincident appearance of CDW and SDW orders below the structural transition temperature.

Single crystals with $x = 0.05$, 0.06, 0.075, and 0.085 were grown through a standard traveling-solvent floating-zone method and annealed under oxygen gas flow to minimize oxygen deficiencies. Neutron scattering measurements were carried out on the triple-axis spectrometers TOPAN and TAS-1 installed in the JRR-3M reactor at the Japan Atomic Energy Research Institute (JAERI) in Tokai. We selected incident neutron energies of $E_i = 14.7$ meV for TOPAN and 13.7 meV for TAS-1 using the (0 0 2) reflection of a pyrolytic graphite monochromator. Typical horizontal collimator sequences used in detecting superlattice peaks were $15'(30')$-$30'$-$30'$-$80'$ and $80'$-$40'$-$40'$-$80'$ for TOPAN and TAS-1, respectively. Additionally, pyrolytic graphite filters were inserted both up- and downstream of the sample position in order to eliminate the higher-order reflected beams. Each sample was mounted with the (h k 0) plane parallel to the scattering plane. Followed by a previous work [15], we denote the reciprocal space by the high-temperature tetragonal phase with $I4/mmm$ symmetry.

To characterize the crystal structure we first examined nuclear Bragg peaks for each sample. In all samples, the (0 1 0) reflection, which is not allowed in the LTO phase, was observed at lowest temperature corresponding to either the LTT or low-temperature less-orthorhombic (LTLO) phase with *Pccn* symmetry. The double (0 1 0) peak from twinned orthorhombic domains in $x = 0.06$, 0.075, and 0.085 samples indicates the LTLO phase, while the single (0 1 0) peak in the $x = 0.05$ sample is consistent with the LTT phase.

As seen in Figs. 1(a) and 1(b), both IC superlattice peaks from CDW and SDW orders are clearly observed for the $x = 0.05$ sample in the LTT phase consistent with results for the LNSCO system [11]. However, in the case of the $x = 0.085$ sample in the LTLO phase peak intensity from the CDW order is dramatically reduced while a well-defined peak from SDW order is observed [see Figs. 1(c) and 1(d)]. Note that the vertical scales in Fig. 1 are normalized by taking the sample volume estimated from phonon intensities into account. Hence relative peak intensities between the two samples can be compared quantitatively. It is also noted that the magnetic IC peak intensity in $La_{1.875}Ba_{0.075}Sr_{0.05}CuO_4$ [16,17] is approximately 6 times stronger than that of $La_{1.88}Sr_{0.12}CuO_4$ ($T_c = 31.5$ K) [18]. Therefore both static CDW and SDW are well stabilized in the LTT phase where superconductivity is strongly suppressed.

In Fig. 2, volume-corrected peak intensities integrated within the planes are shown for the (0 1 0) reflection and the CDW and SDW peaks as a function of Sr concentration. We observed the same shift of both static SDW and CDW peaks to low symmetric positions in the LTLO system [19] as previously observed for SDW peaks in LTO systems [20,21]. We note these peak shifts do not affect the integrated values for the peak intensities. As shown in Figs. 2(a) and 2(b), the Sr substitution similarly reduces both intensities of the (0 1 0) reflection and the CDW order. In fact, no well-defined CDW peak is observed in the LTO phase where (0 1 0) reflection is not allowed. On the other hand, the substantial intensity from the SDW order remains even in the LTO phase. These results strongly suggest the stability of CDW order more strongly depends on the crystal structure than the SDW order. Furthermore, a direct relation of the CDW order with the suppression of T_c becomes clear when the x dependence of T_c in Fig. 4 is mapped in Fig. 2(b) [22,23]. In other words, the strong instability of superconductivity is considered to be triggered by the charge localization rather than by magnetic

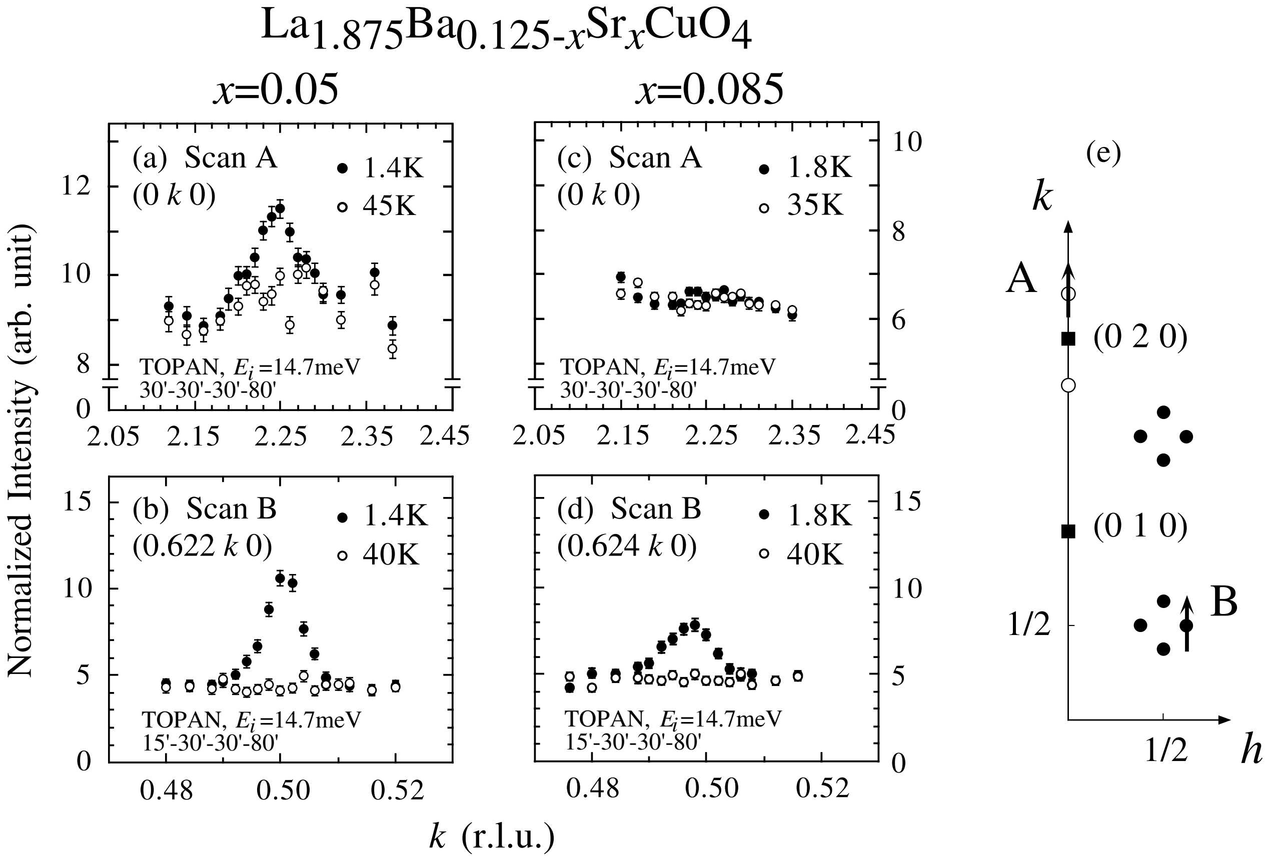

FIG. 1. IC peaks from CDW and SDW orders in $La_{1.875}Ba_{0.125-x}Sr_xCuO_4$ with $x = 0.05$ ([(a) and (b)] and 0.085 [(c) and (d)] measured below T_{d2} (closed circles) and above T_{d2} (open circles). Vertical scales are normalized through the sample volumes and counting time. (e) Scan geometry in the (h k 0) tetragonal plane. Closed squares show nuclear Bragg peaks; open and closed circles denote nuclear and magnetic IC superlattice peaks, respectively.

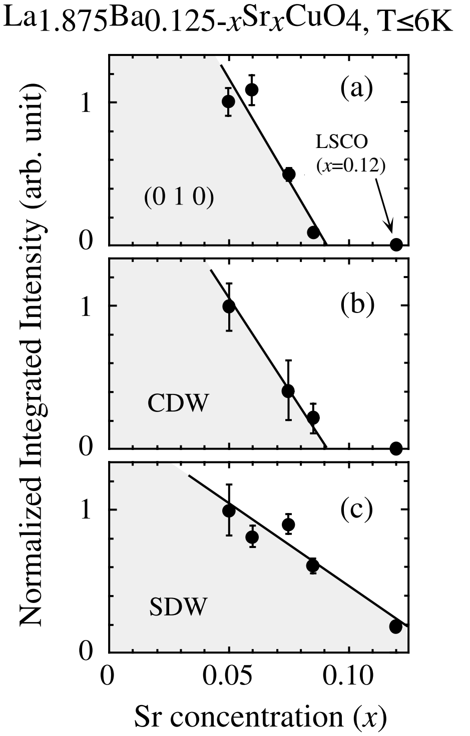

FIG. 2. Peak intensity integrated within the plane for (a) (0 1 0), (b) CDW, and (c) SDW peaks as a function of Sr concentration. The result for $La_{1.88}Sr_{0.12}CuO_4$ is plotted as a reference [16,17]. Solid lines are guides to the eye.

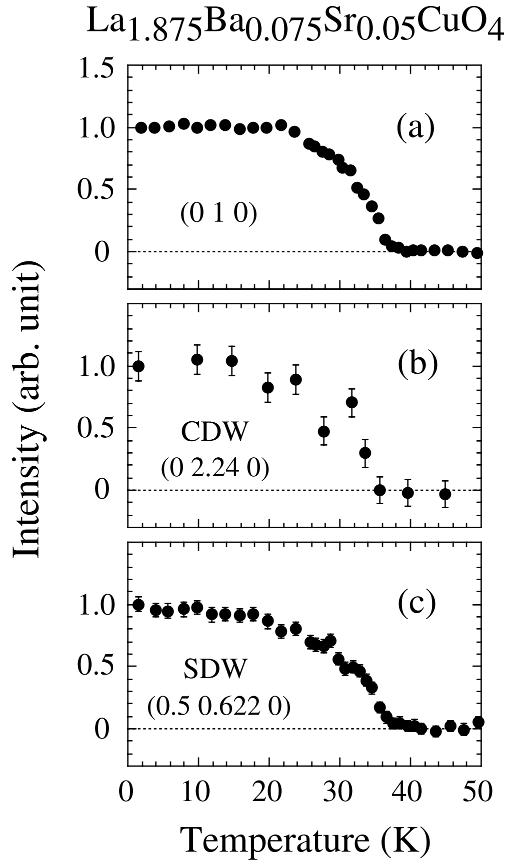

FIG. 3. Temperature dependencies of (a) (0 1 0), (b) CDW, and (c) SDW superlattice peak intensities.

pair breaking. Note that the intensity from CDW order in the $x = 0.085$ sample in Fig. 2(b) is evaluated from a measurement with a higher counting rate compared to the case shown in Fig. 1(c).

Next, we introduce the second new result, that is, the coincident appearance of both CDW and SDW orders. In Fig. 3, the temperature dependence of the order parameter is shown for the (0 1 0), CDW, and SDW peak intensities with the $x = 0.05$ sample. Intensities are normalized by the data at 1.4 K after subtracting the background taken at a higher temperature. Both the CDW and SDW peak intensities were found to begin appearing at the structural transition temperature T_{d2} ($= 37$ K) with a similar temperature dependence to that of the order parameter of the (0 1 0). The coincident appearance of the CDW and SDW orders below T_{d2} is also observed for the $x = 0.075$ ($T_{d2} = 32$ K) and 0.085 ($T_{d2} = 30$ K) samples. This behavior, however, is qualitatively different from that found in the LNSCO system, in which the SDW ordering temperature (T_m) is lower than T_{d2} or the CDW ordering temperature. One possible explanation for such distinct behavior between the two systems is addressed later in this Letter.

In Fig. 4, we present the superconducting [22,23] and structural phase diagram as a function of Sr concentration. T_c of $La_{1.88}Sr_{0.12}CuO_4$ is also plotted as a reference [18]. The shaded area corresponds to the LTT or LTLO phase. In Fig. 4, we set the ground state LTLO-LTO phase boundary at $x_c \sim 0.09$ with the extrapolation of (0 1 0) peak intensity shown in Fig. 2(a). An abrupt drop in T_c with decreasing x is then found around the phase boundary. That is, T_c in the LTO phase is independent of x, while in the

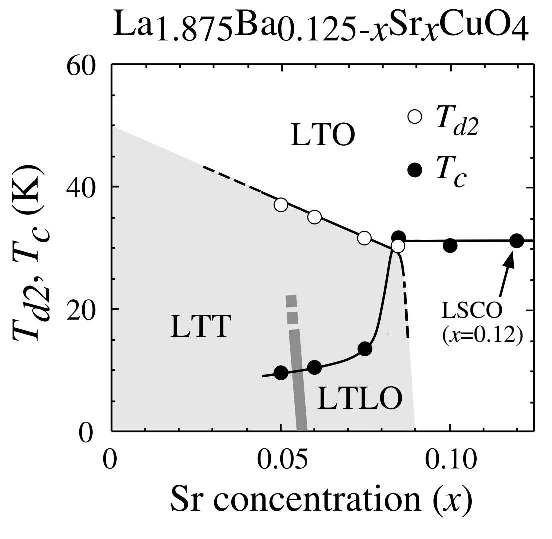

FIG. 4. Phase diagram of T_{d2} (open circles) and T_c (closed circles) for the $La_{1.875}Ba_{0.125-x}Sr_xCuO_4$ system. T_c's are taken from Refs. [18,22,23]. Thick lines correspond to LTT-LTLO phase boundary at low temperature. Solid and dashed lines are guides to the eye.

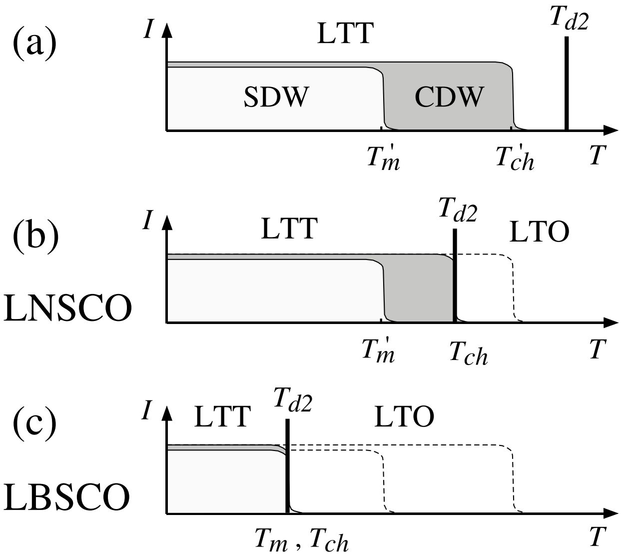

FIG. 5. Schematic ordering sequences for the cases of (a) $T_{d2} > T'_{ch}$, (b) $T'_m \leq T_{d2} \leq T'_{ch}$, and (c) $T_{d2} < T'_m$.

LTT/LTLO phase T_c varies for $0.05 \leq x \leq 0.085$, where T_{d2} changes. This result also suggests the close interplay between superconductivity and the crystal structure.

Previously other groups already reported a close relation between the superconductivity and the crystal structure. Dabrowski *et al.* found the strong correlation between T_c and the tilt angle of the CuO_6 octahedron or the amplitude of the corrugated lattice potential of CuO_2 planes [24]. Furthermore, in both thin films and pressure-applied 2-1-4 compounds with flat CuO_2 planes the disappearance of the $1/8$ anomaly and the substantial increment of T_c are reported [25–28]. Such interplay is easily interpreted through the stripe model or from the viewpoint of the stability of CDW order. In terms of the stripe model, as CDW order on the flat CuO_2 plane is free from the pinning potential, T_c of the orthorhombic phase also should be restored.

Finally, we discuss the possible reason for the different ordering sequence between the LNSCO and LBSCO systems. When T_{d2} is high enough, CDW order is expected to appear at T'_{ch} with decreasing temperature followed by SDW order at T'_m as the schematic ordering sequence is shown in Fig. 5(a). Then, if T_{d2} is lowered, since static CDW and SDW are more stabilized in the LTT than in the LTO phase, these orders can be controlled by the structural transition at T_{d2}. LNSCO and LBSCO systems correspond to the cases shown in Figs. 5(b) and 5(c), respectively: only the CDW ordering for the former and both CDW and SDW orderings for the latter are triggered by the phase transition to the LTT phase. Therefore the coincident appearance of both orders in the LBSCO system is originated from the lower T_{d2} than that in the LNSCO system.

In conclusion, we found in the 1/8-hole doped LBSCO system the strong structural effect on the CDW order. The order is stabilized in the LTT/LTLO phase and thereby severely suppresses the superconductivity, while no well-defined CDW order is observed in the LTO phase where the suppression of T_c is small. Most of these results are explained through the stripe model. However, the absence of the CDW order in the LTO phase with remaining static/quasistatic SDW is difficult to understand by the simple stripe model. More comprehensive study on the charge ordering in the LTO phase is highly required.

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[1] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).
[2] H. Yoshizawa, S. Mitsuda, H. Kitazawa, and K. Katsumata, J. Phys. Soc. Jpn. **57**, 3686 (1988).
[3] R. J. Birgeneau *et al.,* Phys. Rev. B **39**, 2868 (1989).
[4] S.-W. Cheong *et al.,* Phys. Rev. Lett. **67**, 1791 (1991).
[5] K. Yamada *et al.,* Phys. Rev. B **57**, 6165 (1998).
[6] A. R. Moodenbaugh *et al.,* Phys. Rev. B **38**, 4596 (1988).
[7] K. Kumagai *et al.,* J. Magn. Magn. Mater. **76–77**, 601 (1988).
[8] J. D. Axe *et al.,* Phys. Rev. Lett. **62**, 2751 (1989).
[9] H. Takagi *et al.,* Phys. Rev. B **40**, 2254 (1989).
[10] K. Kumagai *et al.,* J. Supercond. **7**, 63 (1994).
[11] J. M. Tranquada *et al.,* Nature (London) **375**, 561 (1995).
[12] J. M. Tranquada *et al.,* Phys. Rev. Lett. **78**, 338 (1997); **78**, 338 (1997).
[13] G. Baskaran, Mod. Phys. Lett. B **14**, 377 (2000).
[14] Y. Maeno *et al.,* Physica (Amsterdam) **173C**, 322 (1991).
[15] M. Fujita, H. Goka, and K. Yamada, J. Phys. Soc. Jpn. Suppl. A **70**, 49 (2001).
[16] H. Kimura *et al.,* J. Phys. Chem. **60**, 1067 (1999).
[17] H. Kimura (private communication).
[18] H. Kimura *et al.,* Phys. Rev. B **59**, 6517 (1999).
[19] M. Fujita (unpublished).
[20] Y. S. Lee *et al.,* Phys. Rev. B **60**, 3643 (1999).
[21] H. Kimura *et al.,* Phys. Rev. B **61**, 14 366 (2000).
[22] M. Fujita, H. Goka, and K. Yamada, Int. J. Mod. Phys. B **14**, 3466 (2000).
[23] H. Goka, M. Fujita, Y. Ikeda, and K. Yamada, Physica (Amsterdam) **357C–360C**, 256 (2001).
[24] B. Dabrowski *et al.,* Phys. Rev. Lett. **76**, 1348 (1996).
[25] H. Sato, A. Tsukada, M. Naito, and A. Matsuda, Phys. Rev. B **62**, R799 (2000).
[26] H. Sato, A. Tsukada, M. Naito, and A. Matsuda, Phys. Rev. B **61**, 12 447 (2000).
[27] T. Goko, F. Nakamura, and T. Fujita, J. Phys. Soc. Jpn. **68**, 3074 (1999).
[28] F. Nakamura *et al.,* Phys. Rev. B **61**, 107 (2000).