Complementary Response of Static Spin-Stripe Order and Superconductivity to Nonmagnetic Impurities in Cuprates

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We report muon-spin rotation and neutron-scattering experiments on nonmagnetic Zn impurity effects on the static spin-stripe order and superconductivity of the La214 cuprates. Remarkably, it was found that, for samples with hole doping $x \approx 1/8$, the spin-stripe ordering temperature $T_{\rm so}$ decreases linearly with Zn doping y and disappears at $y \approx 4\%$, demonstrating a high sensitivity of static spin-stripe order to impurities within a $\rm CuO_2$ plane. Moreover, $T_{\rm so}$ is suppressed by Zn in the same manner as the superconducting transition temperature T_c for samples near optimal hole doping. This surprisingly similar sensitivity suggests that the spin-stripe order is dependent on intertwining with superconducting correlations.

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One of the most astonishing manifestations of the competing ordered phases occurs in the system $\rm La_{2-x}Ba_xCuO_4$ (LBCO) [1], where the bulk superconducting (SC) transition temperature T_c exhibits a deep minimum at $x = 1/8$ [2–4]. At this doping level muon-spin rotation (μSR), neutron, and x-ray diffraction experiments revealed two-dimensional static charge and spin-stripe order [5–11]. The collected experimental data indicate that the tendency toward unidirectional stripelike ordering is common to cuprates [3,4,12–14]. However, the relevance of stripe correlations for high-temperature superconductivity remains a subject of controversy. On the theoretical front, the concept of a sinusoidally modulated pair-density wave (PDW) SC order, intimately intertwined with spatially modulated antiferromagnetism, has been introduced [15–17]. On the experimental front, quasi-two-dimensional superconducting correlations were observed in $\rm La_{1.875}Ba_{0.125}CuO_4$ (LBCO-1/8) and $\rm La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$, coexisting with the ordering of static spin stripes, but with frustrated phase order between the layers [18–23]. Recently, it was found that in $\rm La_{2-x}Ba_xCuO_4$ ($0.11 \le x \le 0.17$) the 2D SC transition temperature T_{c1} and the static spin-stripe order temperature $T_{\rm so}$ have very similar values throughout the phase diagram [24,25]. Moreover, a similar pressure evolution of T_c and $T_{\rm so}$ in the stripe phase of $x = 0.155$ and 0.17 samples was observed. These findings were discussed in terms of a spatially modulated and intertwined pair wave function [15–17,26]. There are also a few reports proposing the relevance of a PDW state in sufficiently underdoped $\rm La_{2-x}Sr_xCuO_4$ [27] and $\rm YBa_2Cu_3O_{6-x}$ [28,29]. At present it is still unclear to what extent PDW order is a common feature of cuprate systems where stripe order occurs.

To further explore the interplay between static stripe order and superconductivity in cuprates, we used nonmagnetic impurity substitution at the Cu site as an alternative way of tuning the physical properties. Since the discovery of cuprate High-temperature superconductor (HTS) much effort has been invested in the investigation of the effect of in-plane impurities. It is now well established that in cuprate HTSs nonmagnetic Zn ions suppress T_c even more strongly than magnetic ions [30–32]. Such behavior is in sharp contrast to that of conventional superconductors. This observation led to the formulation of an unconventional pairing mechanism and symmetry of the order parameter for cuprate HTSs. In addition, in several cases a ground state with static antiferromagnetic (AF) correlations is stabilized by Zn doping [33–38]. Up to now much less has been known about impurity effects on the static stripe phase in cuprates at 1/8 doping. From specific heat and neutron scattering measurements it was inferred that Zn doping leads to stripe destruction [39–41]. Such an effect is very interesting and it was not predicted theoretically. However, no systematic impurity effect studies on static stripe order have been carried out up to now. Moreover, specific heat is a very indirect method to characterize the stripe phase in cuprates. Therefore, it is very important to use experimental techniques which can directly probe stripe formation and its evolution with impurity doping.

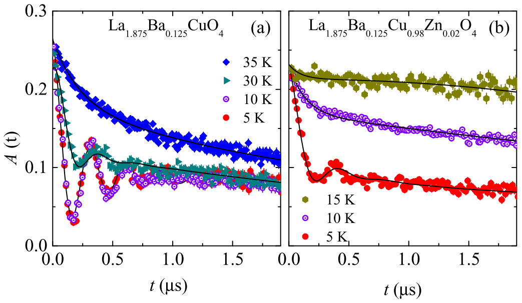

FIG. 1. ZF μSR time spectra $A(t)$ for $La_{1.875}Ba_{0.125}CuO_4$ (a) and $La_{1.875}Ba_{0.125}Cu_{0.98}Zn_{0.02}O_4$ (b) recorded at various temperatures. The solid lines represent fits to the data by means of Eq. 3 of Ref. [42].

In this Letter, we report on systematic muon-spin rotation μSR, neutron scattering, and magnetization studies of Zn impurity effects on the static spin-stripe order and superconductivity in the La214 cuprates. Remarkably, it was found that in these systems the spin-stripe ordering temperature T_{so} decreases linearly with Zn doping y and disappears at $y \simeq 4\%$. This means that T_{so} is suppressed in the same manner as the superconducting transition temperature T_c by Zn impurities. These results suggest that the stripe and SC orders may have a common physical mechanism and are intertwined.

In a μSR experiment, positive muons implanted into a sample serve as an extremely sensitive local probe to detect small internal magnetic fields and ordered magnetic volume fractions in the bulk of magnetic systems. Thus μSR is a particularly powerful tool to study inhomogeneous magnetism in materials [42]. Neutron diffraction experiments [46] allow us to directly probe the incommensurate spin structure of spin-stripe order and thus provide crucial complementary information to the μSR technique.

Figures 1(a) and 1(b) show representative zero-field (ZF) μSR time spectra for polycrystalline $La_{1.875}Ba_{0.125}Cu_{1-y}Zn_yO_4$ samples with $y = 0$ and 0.02, respectively, recorded at various temperatures. For $y = 0$, damped oscillations due to muon-spin precession in internal magnetic fields are observed below $T_{so} \approx 35$ K, indicating the formation of static spin order in the stripe phase [9,25,43,44,47]. It is seen in Fig. 1(b) that for the $y = 0.02$ sample the oscillating signal appears only below $T \approx 10$ K, showing strong suppression of the static spin-stripe order with Zn doping. We have studied this novel effect systematically as a function of Zn doping.

The temperature dependence of the average internal field B_μ, which is proportional to the ordered magnetic moment, is shown in Fig. 2(a) for various Zn dopings y. As evident from Fig. 2(a), $B_\mu(0)$, the internal magnetic field extrapolated to zero temperature, does not depend on the Zn content y, while T_{so} changes substantially with increasing y. Specifically, T_{so} decreases from $T_{so} \simeq 32.5$ K for $y = 0$ to $T_{so} \simeq 4$ K for $y = 0.04$. Figure 2(b) shows that a very similar behavior is observed for B_μ measured on polycrystalline samples of the related compound $La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$. Note that the low-temperature value of B_μ is enhanced by the ordering of the Nd moments. A similar suppression of T_{so} by Zn impurities was also observed in single-crystal samples of $La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.016). It seems that this effect is a generic feature of cuprates with static stripe order. We note that in all the above-mentioned systems, despite the suppression of T_{so} with Zn doping, the magnetic volume fraction V_m at the base temperature stays nearly 100%. The bulk Low-temperature tetragonal structural phase transition temperature also stays nearly unaffected by Zn doping (see Supplemental Note II and Supplemental Fig. S1 [42]).

The observed Zn impurity effects on T_{so} in $La_{1.875}Ba_{0.125}Cu_{1-y}Zn_yO_4$ and $La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ are summarized in Fig. 3. It is a remarkable finding that T_{so} linearly decreases with increasing Zn content y. Such a behavior is reminiscent of the well-known linear suppression of the SC transition temperature T_c in cuprates [30–32]. Since the superconducting volume fraction in 1/8-doped samples is tiny and the bulk T_c is also very low, it is difficult to follow the SC properties of these systems as a function of Zn content. Alternatively, in Fig. 3 we plot T_c

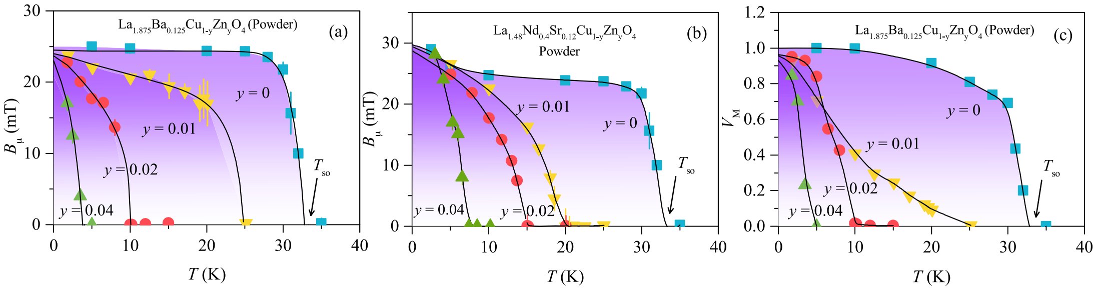

FIG. 2. The temperature dependence of (a) the internal magnetic field B_μ and (c) the magnetic fraction V_m for the polycrystalline samples of $La_{1.875}Ba_{0.125}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.02, 0.04), and (b) the temperature dependence of B_μ for the polycrystalline samples of $La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.01, 0.02, 0.04). The arrows mark the spin-stripe order temperature T_{so}. The solid curves are fits of the data to the power law $B_\mu(T) = B_\mu(0)[1-(T/T_{so})^\gamma]^\delta$, where $B_\mu(0)$ is the zero-temperature value of B_μ. γ and δ are phenomenological exponents.

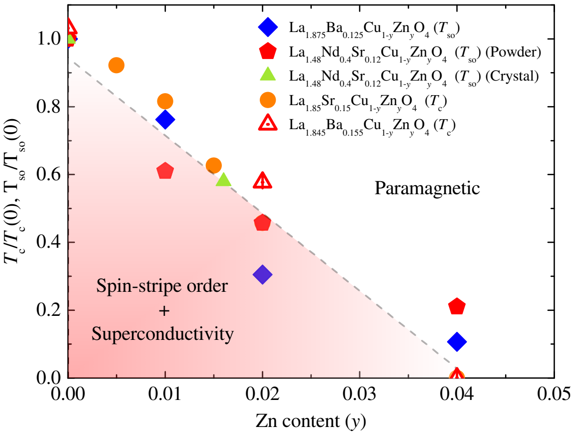

FIG. 3. The normalized static spin-stripe order temperature $T_{\rm so}/T_{\rm so}(0)$ for $\rm La_{1.875}Ba_{0.125}Cu_{1-y}Zn_yO_4$ and $\rm La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ as a function of Zn content y and the superconducting transition temperature $T_c/T_c(0)$ of $\rm La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ as a function of Zn content y. The dashed line is a guide to the eye.

values for optimally doped $\rm La_{1.85}Sr_{0.15}CuO_4$ [30–32] and $\rm La_{1.845}Ba_{0.155}CuO_4$ (see below) as a function of Zn content. Strikingly, suppression of $T_{\rm so}$ goes in a very similar manner as the well known impurity-induced T_c suppression.

We have confirmed the Zn-doping effect on the static spin-stripe order by neutron diffraction experiments on single-crystal samples of $\rm La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.016) [48]. The magnetic ordering wave vectors are $\mathbf{Q}_{\rm so} = (0.5 \pm h, 0.5, 0)$ and $(0.5, 0.5 \pm h, 0)$; i.e., they are displaced by δ from the position of the magnetic Bragg peak in the AF parent compound $\rm La_2CuO_4$ [5,6]. In Fig. 4(a) we show h scans through the $(0.5 + h, 0.5, 0)$ magnetic superlattice peaks, recorded at $T = 1.75$ K for the samples $y = 0$ and 0.016. It is clear that the intensity and incommensurability do not change with Zn doping. However, $T_{\rm so}$ is strongly suppressed from $T_{\rm so} \simeq 50$ K [5] for $y = 0$ to $T_{\rm so} \simeq 10$ K for $y = 0.016$, as demonstrated in Fig. 4(b), where the peak intensity is shown as a function of temperature.

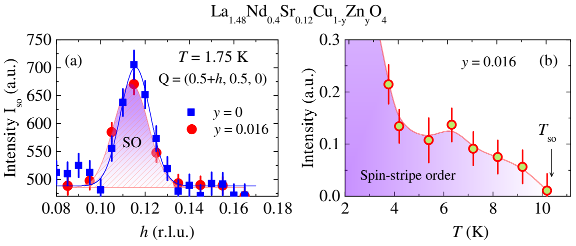

FIG. 4. (a) h scans through the spin order peak at $(0.5 + h, 0.5, 0)$ for the single crystals of $\rm La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.016), recorded at the base temperature $T = 1.75$ K. The intensities have been normalized to the crystal volume in the neutron beam. The solid lines represent the Gaussian fits to the data. (b) Peak intensity versus temperature of the $(0.5 + h, 0.5, 0)$ spin order peak, normalized to the crystal volume in the neutron beam.

Going further, we studied the Zn-impurity effects on $T_{\rm so}$ and T_c in $\rm La_{1.845}Ba_{0.155}CuO_4$. This compound ($x > 1/8$) exhibits a well-defined bulk SC transition with $T_c = 30$ K and at the same time shows static spin-stripe order $T_{\rm so} \simeq T_c = 30$ K [24]. This enables us to study impurity effects on $T_{\rm so}$ and T_c simultaneously in the same sample. Figure 5(a) shows the temperature dependence of the magnetic volume fraction V_m extracted from ZF-μSR data for $\rm La_{1.845}Ba_{0.155}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.02, and 0.04). The low-temperature value of V_m increases with increasing Zn content y and reaches 100% for the highest Zn content $y = 0.04$. On the other hand, $T_{\rm so}$ decreases with increasing y, similar to 1/8 doping. The values of $T_{\rm so}$ and T_c (see the Supplemental Note III and Supplemental Figs. S2 and S3 [42]) as a function of Zn content y are shown in Fig. 5(b). Again, with increasing y both T_c and $T_{\rm so}$ decrease linearly with the same slope, indicating that Zn impurities influence T_c and $T_{\rm so}$ in the same manner.

What is the significance of this surprising correlation? Let us start with the fact that it is unusual to have spin order occur in a hole-doped cuprate at a temperature of ∼35 K. Just a couple of percent of hole doping is generally sufficient to wipe out antiferromagnetic order [49]. One common point of view is that antiferromagnetism and superconductivity are competing orders [50]. From that

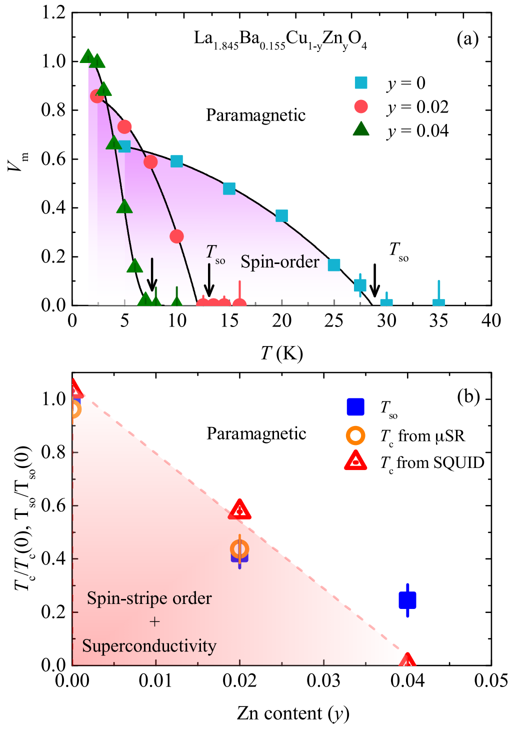

FIG. 5. (a) Temperature dependence of the magnetic volume fraction V_m for $\rm La_{1.845}Ba_{0.155}Cu_{1-y}Zn_yO_4$ ($y = 0$, 0.02, 0.04). (b) The normalized static spin-stripe order temperature $T_{\rm so}/T_{\rm so}(0)$ and the normalized superconducting transition temperature $T_c/T_c(0)$ for $\rm La_{1.845}Ba_{0.155}Cu_{1-y}Zn_yO_4$ as a function of Zn content y. The dashed line is a guide to the eye.

perspective, one might take the occurrence of spin-stripe order as evidence that hole pairing and superconductivity have been suppressed. In that case, we might expect the impact of Zn doping on T_{so} to be similar to its impact on the Néel temperature in $\mathrm{La_2CuO_4}$. That assumption leads to a problem, however, as experiment has demonstrated that it takes not 4%, but ~40% Zn to destroy Néel order [51]. One could also take account of the fact that the Zn tends to induce static Cu spin order in its immediate neighborhood [52,53], which, with random locations of the Zn sites, could lead, at higher Zn concentrations, to some disorder from neighboring pinned stripe domains being out of phase with one another; however, a shortening of the spin correlation length only becomes apparent with at least 3% Zn doping [41,54], while the drop in T_{so} is clear at much lower Zn concentrations.

Consider instead that previous experiments provide evidence that spin-stripe order coexists with two-dimensional superconducting correlations in $\mathrm{La_{2\text{-}x}Ba_xCuO_4}$ ($x = 1/8$) [19,20]. Here, the superconducting and spin orders are intertwined [15]. Superconducting correlations within charge stripes must establish Josephson coupling across the spin stripes, while the spins in neighboring stripes must establish an effective exchange coupling via the fluctuating pairs in the intervening charge stripe. A Zn ion will locally suppress hole motion, thus eliminating local superconducting coherence and weakening the superconductivity [55]. Local suppression of hole hopping will also disrupt the effective exchange coupling between spin stripes, leading to a reduction in T_{so}.

Previous μSR studies of Zn doping in LSCO and $\mathrm{YBa_2Cu_3O_7}$ have established the "swiss-cheese" model: a fixed carrier density per Zn atom is removed from the superfluid density, as if each Zn removes a fixed areal fraction of the superfluid [56]. The linear relationship between T_c and the average superfluid density, valid for underdoped through optimally doped cuprate HTSs, then explains the reduction of T_c with increasing Zn concentration [57]. For the stripe-ordered systems, it is plausible that both the superconducting and spin-stripe orders will respond in a similar fashion.

In conclusion, static spin-stripe order and superconductivity in cuprate systems $\mathrm{La_{2-x}Ba_xCu_{1-y}Zn_yO_4}$ ($x = 0.125$, 0.155) and $\mathrm{La_{1.48}Nd_{0.4}Sr_{0.12}Cu_{1-y}Zn_yO_4}$ were studied by means of magnetization, μSR, and neutron scattering experiments as a function of nonmagnetic Zn impurity concentration. High sensitivity of the static spin-stripe order temperature T_{so} to impurities in the $\mathrm{CuO_2}$ plane was demonstrated. Namely, the spin-stripe ordering temperature T_{so} strongly decreases linearly with Zn doping and disappears at about 4% Zn content. More strikingly, T_{so} is suppressed in the same fashion as is the superconducting transition temperature T_{c} by Zn impurities. These results strongly suggest that the existence of the stripe order requires intertwining with the SC pairing correlations, such as that which occurs in the proposed PDW state. The present findings should help us to better understand the complex interplay between stripe order and superconductivity in cuprates. More generally, since charge and spin orders are often observed in other transition-metal oxides, investigation of impurity effects and disorder on stripe formation may become an interesting research avenue in correlated electron systems.

The μSR experiments were carried out at the πM3 beam line of the Paul Scherrer Institute (Switzerland), using the general purpose instrument (GPS). The neutron scattering experiments were carried out with the three-axis spectrometer EIGER at the Swiss Spallation Neutron Source SINQ at the Paul Scherrer Institut, Switzerland. We are grateful to S. A. Kivelson for valuable discussions. Z. G. gratefully acknowledges the financial support by the Swiss National Science Foundation (SNF Early postdoc mobility fellowship No. P2ZHP2-161980 and SNF Grant No. 200021-149486). Z. G. thanks Martin Mansson for useful discussions. A. S. acknowledges support from the SCOPES Grant No. SCOPES IZ74Z0-160484. Work at Columbia University is supported by U.S. NSF Grant No. DMR-1436095 (DMREF) and NSF Grant No. DMR-1610633 as well as REIMEI project of Japan Atomic Energy Agency. J. M. T. is supported at Brookhaven by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-SC0012704.

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