Fe-impurity-induced magnetic excitations in heavily over-doped La$_{1.7}$Sr$_{0.3}$Cu$_{0.95}$Fe$_{0.05}$O$_4$

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Abstract. Inelastic neutron scattering experiments were performed to investigate the effect of Fe substitution on the magnetic excitation spectra in a heavily hole-doped cuprate system La$_{1.7}$Sr$_{0.3}$Cu$_{0.95}$Fe$_{0.05}$O$_4$, with an effective concentration of holes of $x_{\text{eff}} = 0.25$. At $T = 7$ K, well-defined low-energy magnetic excitations, associated with a static magnetic order, were observed at the incommensurate positions $Q_{\text{en}} = (0.5 \pm \delta, 0.5)/0.5 \pm \delta$. Although the incommensurate structure resembles that observed in La$_2-x$Sr$_x$CuO$_4$ (LSCO) systems, the incommensurability ($\delta$) of 0.144 (r.l.u) was larger than that of ~0.125 (r.l.u) in LSCO with $x = 0.25$, suggesting an increase in $\delta$ by Fe substitution. In addition to the obvious effect on IC excitations, we show the emergence of ring-shaped magnetic excitations centered at $Q_{\text{en}} = (0.5, 0.5)$ in a constant energy spectrum by Fe-substitution. These ring-shaped excitations were observed for energies below ~8 meV, yielding a cylindrical continuum excitation in the energy and momentum spaces. Similar continuum excitations were recently observed in pristine LSCO with $x = 0.25$ at high temperatures, in our neutron experiments [1]. These results, which show the enhancement of $\delta$ (higher than $\delta = 1/8$) and the stabilization of continuum excitations by Fe substitution, are incompatible with a simple pinning scenario of the stripe order, which is characterized by local spin correlations. The metallic aspect of the low-energy part of the magnetic excitation, which underlies high-temperature superconductivity in over-doped cuprate systems, would be enhanced/induced by Fe substitution.

1. Introduction

To elucidate the mechanism of high-temperature superconductivity ($T_c$) in cuprate systems, it is important to understand the nature of spin correlations that couple with doped carriers in Mott insulators; whether localized and itinerant spin nature is necessary for realizing the ground state at a
particular hole concentration. One of the key aspects is that, in the archetypal cuprate superconductor \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (LSCO) system, antiferromagnetic low-energy signal splits into spots with wave vectors of \((0.5 \pm \delta, 0.5)/(0.5, 0.5 \pm \delta)\) in the tetragonal notation in the superconducting phase, where the incommensurability \(\delta\) is the half-distance between the pair of incommensurate (IC) magnetic peaks. The incommensurability increases linearly and almost saturates for the doping rates under and over \(x = 0.12\), respectively [2, 3], implying that magnetic correlations have different origins in the under- and over-doped regions.

Previous neutron scattering experiments reported that dilute doping of Fe into LSCO induces a static magnetic order in a wide superconducting phase [4, 5]. The IC wave vectors of the induced order resemble that observed in the static and/or low-energy channels in pristine systems. Angle-resolved photoemission spectroscopy (ARPES) measurements on over-hole-doped samples suggest that the induced magnetic peaks arise from the nesting of the Fermi surface, indicating that the magnetically ordered ground state is characterized by the spins of itinerant doped holes [5]. This stimulated the discussion on the long-studying issue in the high-\(T_c\) superconductivity: which of the two, localized or itinerant spin pictures, are more adequate for describing the magnetism in the superconducting phase?

To elucidate the nature of magnetic correlations related to impurity-induced magnetism, we performed inelastic neutron scattering (INS) experiments on Fe-doped LSCO samples, using a chopper spectrometer with wide-area detectors and time-of-flight (TOF) analysis. Because the TOF method yields the intensity maps of scattered neutrons in a wide space of momentum transfer \((Q)\) and energy transfer \((\omega)\), the overall dynamical susceptibility of continua excitations, which is often discussed in studies of metallic magnetism, and of the collective excitation associated with localized spins can be effectively measured.

2. Experiments

One single crystalline rod of Fe-substituted and heavily hole-doped sample \( \text{La}_{1.17}\text{Sr}_{0.3}\text{Cu}_{0.95}\text{Fe}_{0.05}\text{O}_4 \) was grown using the traveling-solvent floating-zone method. Because substituting \(\text{Fe}^{3+}\) for \(\text{Cu}^{2+}\) reduces the number of holes that are doped by the \(\text{Sr}^{2+}\) substitution for \(\text{La}^{3+}\) [6], we can estimate the concentration of holes \(x_{\text{eff}}\) in \( \text{La}_{1.17}\text{Sr}_{0.3}\text{Cu}_{0.95}\text{Fe}_{0.05}\text{O}_4 \) as \(x_{\text{eff}} = 0.30 - 0.05 = 0.25\). The weight of the crystal was 22 g. To minimize the background, we used an Al sample can-free from the exchange gas. The lattice constants of \( \text{La}_{1.17}\text{Sr}_{0.3}\text{Cu}_{0.95}\text{Fe}_{0.05}\text{O}_4 \) are \(a = b = 3.97\) Å and \(c = 13.2\) Å at room temperature, in the tetragonal notation. To describe the experimental results, we employed the orthorhombic notation, because by using this notation, the integration range can be easily described. In this notation, the magnetic \(\Gamma\) point \(Q_{\text{tot}} = (0.5, 0.5, L)\) for the parent antiferromagnet in the tetragonal notation corresponds to \(Q_{\text{tot}} = (1, 0, L)\), and the IC positions can be written as \((1+\delta, \pm \delta)/(1-\delta, \pm \delta)\).

The INS experiments were performed using BL14 AMATERAS[7], a cold-neutron chopper spectrometer installed at the Materials and Life Science Experimental Facility (MLF) of J-PARC. The Utsusemi software suite was used to analyze the spectral data obtained using the pulsed-neutron scattering technique [8]. The sample was mounted on a 4-K-temperature cryostat, and \(H_{\text{tot}}\) and \(L\) axes in \(Q_{\text{tot}} = (H_{\text{tot}}, K_{\text{tot}}, L)\) were aligned in the horizontal plane of the spectrometer. The \(L\) axis was at 30° to the direction of the incident neutron beam. This configuration allowed us to obtain the magnetic excitation effectively by projecting it onto the \(H_{\text{tot}}\)-\(K_{\text{tot}}\) plane, owing to its 2-dimensional nature. The parameters for monochromatizing choppers were tuned to gain neutrons with the incident energy of \(E_i = 10.52\) meV and to realize an energy resolution of \(\Delta \omega = 0.7\) meV at the elastic position. In these experimental setups, we measured magnetic excitations in the \(Q_{\text{tot}}\)-\(\omega\) space and their temperature dependence.
3. Results and Discussion

3.1. Incommensurate magnetic excitations

Figure 1 (a) shows INS intensity maps of the magnetic excitation spectra taken at \( T = 7 \) K, which is below the magnetic ordering temperature of 45 K. Excitations are clearly seen at IC positions around \( K_{\text{ort}} = 0 \) r.l.u., similar to the results for pristine LSCO with \( x = 0.25 \) [9]. At 300 K, the intensity below \(-5\) meV is much weaker than that at 7 K (Fig. 1(b)), suggesting that low-energy excitations below \(-5\) meV strengthen at low temperatures. In the present system, excitations arose from the elastic position, while there was no magnetic Bragg peak in the over-doped pristine LSCO. The incommensurability of \( \text{La}_{1.7}\text{Sr}_{0.3}\text{Cu}_{0.95}\text{Fe}_{0.05}\text{O}_4 \) is \( \delta = 0.144 \) (r.l.u) at \( T = 7 \) K, higher than \( \delta = 0.125 \) (r.l.u) for LSCO with \( x = 0.25 \) measured in an inelastic region [2]. As mentioned above, in the pristine LSCO system, \( \delta \) exhibits a linear dependence in the under-doped region and saturates with \( \delta = 0.125 \) (r.l.u) for \( x \geq 0.12 \). Therefore, \( \delta \) is increased by Fe substitution in the doping region while \( \delta \) is almost constant in the pristine system. Therefore, variations in the concentration of holes do not explain \( \delta \)-enhancement larger than 0.125, and the stabilization of the stripe fluctuation in the pristine LSCO is not directly related to the appearance of magnetic order. We speculate that the modification of the Fermi surface topology by Fe substitution induces magnetic response at different positions in the reciprocal space. The appearance of the IC magnetic order for quite high \( \delta \sim 0.2 \) has been also reported in \( \text{Bi}_{1.75}\text{Pb}_{0.25}\text{Sr}_{1.90}\text{Cu}_{0.91}\text{Fe}_{0.09}\text{O}_6 \), for the hole concentration \( p = 0.25 \) [10]. Fe impurities probably have the same effect on the correlations between spins in Cu sites, and stabilize static magnetic order in the high-density-carrier phase of cuprate superconductors.

![INS intensity maps in the \( K_{\text{ort}}-\omega \) plane, taken at \( T = 7 \) K and 300 K. Integration ranges over \( H_{\text{ort}} \) and \( L \) were from 0.8 to 1.2 r.l.u. and from -1.2 to 3.9 r.l.u., respectively. These intensities were divided by the Bose population factor at each temperature.](image-url)
3.2. Appearance of magnetic continuum excitations

Figure 2 shows the INS intensity maps in the $H_{\text{ort}}$-$K_{\text{ort}}$ plane, taken at $T = 7$ K and 300 K. As seen in this figure, at low temperatures there is an additional spectral weight between the IC positions with the ring-like intensity-distribution around $Q_{\text{ort}} = (1, 0)$. The radius of the ring-like signal was estimated as $r = 0.126$ (r.l.u) in the tetragonal notation, comparable to $\delta$ for pristine LSCO with $x = 0.25$. This ring-like signal would be the response of spins of itinerant electrons, because the distribution of the spectral weight cannot be simply reproduced by collective excitations from an ordered state of localized spins.

As shown in Fig. 1 and Fig. 2, the IC excitations disappeared at the high temperature $T = 300$ K, whereas the ring-like signal remained, although the intensity distribution became blurred. In the case of the YBa$_2$Cu$_3$O$_{6.95}$ system, a metallic nature has been discussed to understand the low energy component of magnetic excitations; excitations in the IC structure arose from the Fermi surface nesting and became continuum excitations confined at around the magnetic $\Gamma$ point, owing to the breakdown of the nesting condition in the high-temperature regime [11]. This consideration suggests that the magnetic excitation switches with temperature between the well-defined IC structure and continuum. In our case of Fe-doped LSCO, the difference in the incommensurability between the IC and ring-shaped signals and in their temperature dependence directly suggests that the low-energy magnetic excitations are composed of IC excitations and continua at the fixed energy transfer and at low temperatures. In the case of Fe 1% substituted LSCO with $x = 0.25$ and with $x = 0.29$, INS and ARPES experiments suggest the appearance of a spin density wave owing to the Fermi surface nesting [5]. This supports the notion of the metallic origin of the appearance of the magnetic order and related low-energy excitations. Although the evidence for continuum excitation has not been pointed out theoretically [5], it is interesting to determine whether continuum components can be reproduced from the viewpoint of Fermi surface topology.

Finally, we briefly report a comparison with the results for pristine LSCO with $x = 0.25$. Quite recently, we found similar ring-like continuum excitations in LSCO with $x = 0.25$ at high temperatures [1], whereas, as discussed previously [9], such excitation was not confirmed for low temperatures. Therefore, continuum excitations in the high-temperature region for pristine LSCO with $x = 0.25$ would be stabilized by the Fe substitution even at low temperatures. These results suggest that elucidating the roles of Fe substitution can provide further understanding of inherent magnetic correlations underlying the superconductivity of the LSCO system, taking a different approach in high-$T_c$ studies on the cuprates.

![Fig. 2. INS intensity maps in the $H_{\text{ort}}$-$K_{\text{ort}}$ plane, taken at $T = 7$ K and 300 K. Integration ranges over $L$ and $\omega$ were from -1.2 to 3.9 r.l.u. and from 2 to 3 meV, respectively. These intensities were divided by the Bose population factor at each temperature.](image-url)
4. Summary

Using INS, magnetic excitation spectra were measured for a single crystal of Fe-substituted and heavily hole-doped La$_{1.7}$Sr$_{0.3}$Cu$_{0.95}$Fe$_{0.05}$O$_4$. On the $K_{\text{ort}}$-$\omega$ and $H_{\text{ort}}$-$K_{\text{ort}}$ maps experimentally obtained at $T = 7$ K, we found that the excitation spectra are composed of incommensurate excitations and ring-like continua. The incommensurability of the induced magnetic peaks ($\delta = 0.144$ (r.l.u)) was different from the radius of ring-shaped signal $r = 0.126$ (r.l.u), and even larger than $\delta = 0.125$ (r.l.u), for pristine LSCO with the same concentration of holes. At a high temperature of $T = 300$ K, the incommensurate excitations disappeared, whereas the continuum excitations remained. Both excitations were intimately related to the inherent metallic nature of magnetic correlations in the heavily hole-doped LSCO system. Elucidating the effect of Fe substitution on spin correlations is an effective approach for obtaining key information on the coupling of electrons with strong spin correlation in high-$T_c$ cuprate superconductors.

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