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Transport and optical properties of epitaxial $\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_{4-\delta}$ thin films

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Abstract. Electrical transport measurements and transient optical pump-probe experiments have been performed on epitaxial films of the electron doped $\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_{4-\delta}$ compound for studying the non-equilibrium carrier dynamics in this material. Samples have been grown on (001)-oriented SrTiO_3 substrates by dc sputtering in a mixed atmosphere of both Ar and O_2 . X-ray diffraction analysis and scanning electron microscope equipped with a wavelength dispersive spectroscopy detector have been used to characterize the structure and the composition of the thin films. Time-resolved femtosecond pump-and-probe spectroscopy has been also carried out on our samples in the temperature range 4.2K-300 K.

1. Introduction

New superconductors have been discovered over the past century, nevertheless, electronic states and mechanism of superconductivity have not been clarified yet [1]. One of the most discussed issue regards the pseudogap phenomena, that are less explored in the case of the electron-doped cuprates in comparison to the hole-doped systems[2]. Recently, a crossed experimental and theoretical study on optimally doped $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ single crystal has shown the evidence of a pseudogap state with antiferromagnetic-like nature [3]. The results obtained by both methods supported competing order parameter fluctuations as the origin of the pseudogap instead of the superconducting scenario [4].

In hole-doped cuprates, time resolved optical spectroscopy has proven to be a powerful method for tracking the onset of both pseudogap and superconducting order with a single technique [5]. During the last year, Hilton and coworkers have reported time-resolved reflection experiments on the single crystal electron-doped superconductor $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$, measuring the change in reflectivity induced by an ultrashort optical pulse as a function of time [6]. They associate the positive signal that they observe at critical temperature T_c with superconductivity and the negative signal with the pseudogap. In fact, its onset at 75 K lies on the pseudogap temperature-cerium content line $T^*(x)$ of the doping phase diagram, as determined from the appearance of gaps in optical conductivity [7] and photoemission spectra [8]. In this work, the characterization from structural and compositional point of view of superconducting



$\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_{4-\delta}$ (NCCO) thin films will be presented. Moreover, the preliminary analysis of pump and probe measurements performed on them will be discussed. Nevertheless, further experiments should be performed in order to better understand the nature of the pseudogap state by correlating it to doping levels in thin films.

2. Experimental details

Dc sputtering technique was optimized to grow NCCO films on (100) SrTiO_3 (STO) substrates by using a single stoichiometric target as a sputtering source in an on-axis configuration with the substrate [9]. The sputtering depositions have been carried out in pure Argon (Ar) atmosphere at pressure of 1.7 mbar and substrate temperature of 900 °C. The as-grown samples do not show the superconductive transition. In order to observe the superconductivity, the films have to be thermal treated for several hours (it depends on their thickness) in flowing argon. Compositional and structural characterizations were performed to investigate the quality of the samples by wavelength dispersive spectroscopy (WDS) and x-ray diffraction (XRD), respectively. Standard four probe technique in a Cryogenic variable temperature cryogen-free system has been used to investigate the electrical transport properties of the films. Finally, the femtosecond pump-probe optical reflectivity experiments were performed using a mode-locked Ti:sapphire laser which produced 100-fs-wide, 790-nm-wavelength pulses at a repetition rate of 82 MHz. The pump and probe beams were cross-polarized to eliminate coherent artifacts arising from the interference between two beams. The samples were mounted on a cold finger in a temperature-controlled liquid helium continuous-flow optical cryostat, operating from 5 K to 300 K. Both pump and probe beams have spots close to 70 μm diameter onto the sample. The pump-probe average power ratio was set at 3:1, while the pump fluence ranged from 2.5 $\mu\text{J}/\text{cm}^2$ to 12.5 $\mu\text{J}/\text{cm}^2$.

3. Results and discussion

The compositional analysis confirms an over-doped cerium content equal to 0.17 in our samples. Neodymium and copper result in the right stoichiometry with respect this value, while it is not possible the oxygen content evaluation with our equipment. In fact, the use of the STO as substrate prevents the possibility to distinguish the contribution coming from the two oxides [10]. The left panel of Figure 1 shows the behaviour of the resistance versus temperature (r - T) of a film ex-situ thermal treated for 2 hours in flowing argon at 900 °C, with a $T_c = 9\text{K}$. The

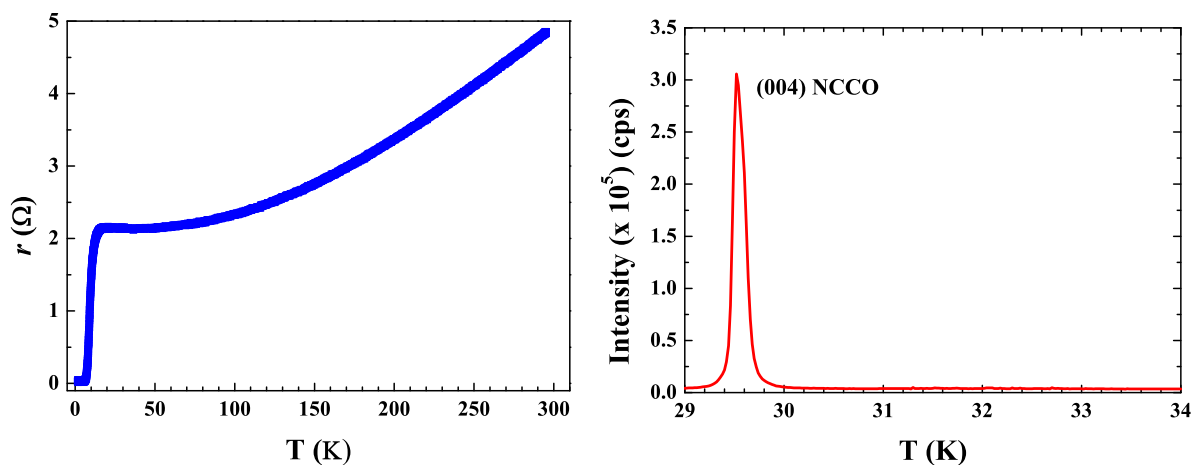


Figure 1. On the left, the r - T behavior of a superconducting NCCO sample. On the right, the corresponding $\theta - 2\theta$ scan around the (004) reflection of the NCCO phase are reported. No spurious phases are detected in the range investigated.

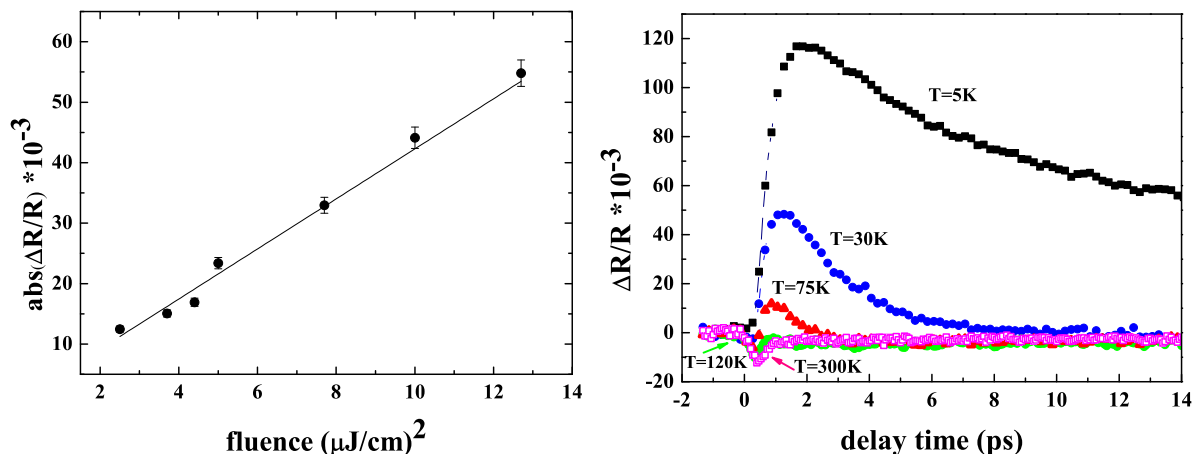


Figure 2. Left panel shows the $\Delta R/R$ as a function of pump fluence measured at 300K; the line is a guide for eyes. Right panel reports the $\Delta R/R$ as a function of the delay time between the pump and probe pulse at different temperatures.

right panel of Figure 1 reports the corresponding $\theta - 2\theta$ scan around the (004) reflection of the NCCO phase, from 29° to 34° . This particular range allows to detect the possible presence of spurious phases in the samples. The data show that our well oriented films are free from undesired features due to polycrystalline residuals or to the Cu-free phase, $(\text{Nd,Ce})_2\text{O}_4$ [9]. In our pump-probe experiments, the optical reflectivity change, ΔR , induced by the pump beam was measured by the probe beam, time delayed with respect to the pump. Thus, by studying the ΔR signal, normalized to the DC value, R , as a function of the time delay between the pump and the probe beams, time-resolved dynamics of the excited carrier relaxation processes can be measured directly. In the left panel of Figure 2, the $\Delta R/R$ as a function of pump fluence measured at 300K is plotted. In the range from $2.5 \mu\text{J}/\text{cm}^2$ to $12 \mu\text{J}/\text{cm}^2$, the behaviour results linear, then samples heating can be considered negligible. The minimum value we have chosen for the fluence in our experiment is comparable with that reported in ref. [6]. The right panel of Figure 2 reports the $\Delta R/R$ as a function of the delay time between the pump and probe pulse at different temperatures. At temperatures $T < 30\text{K}$, only a positive peak with a long tail for $T=5\text{K}$ is observed, probably due to a slow relaxation time related to the recombination process of excitations in Cooper pairs. At $T > 30\text{K}$ a small negative peak appears and by increasing the temperature the positive peak decreases and it disappears at a temperature $T > 75\text{K}$.

In spite of a sign inversion observed at temperatures close to 75K in the transient reflectivity data, our preliminary experiments do not allow us to clearly correlate such a feature to the occurring of a pseudogap component. Further experiments at different doping levels, and hence different strengths of the superconducting state, and different laser pump fluences are currently in progress aiming to better understand the nature of such physical features.

4. Conclusions

In conclusion, highly oriented over-doped NCCO films with $x_{\text{Ce}} = 0.17$ are grown on (100) STO substrates by dc sputtering deposition technique. No spurious phases have been detected after the high temperature ex-situ thermal treatment to obtain the superconductivity. Preliminary analysis of pump and probe measurements point out the decrease of the reflectivity amplitude by increasing the temperature up to its disappearance for $T > 75\text{K}$ in overdoped superconducting NCCO films. Further analysis are in progress in order to evaluate the time responses and to interpret the different behavior of the films with respect to the data on the single crystals of the literature.

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