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MÖSSBAUER STUDIES OF $(La_{1-x}Ba_x)_2CuO_4$ SUPERCONDUCTING OXIDES

by

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Samples with Cu substituted by small amount of iron (less than 1%) were prepared for x = 0.15, 0.25 and 0.35 mixtures. Despite the fact that there is only one Cu site in the $(La_{1-x}Ba_x)_2CuO_4$ compounds the Mössbauer spectra at room temperature indicated the presence of two quadrupole doublets whose behaviour were followed in different steps of samples preparation. They are mostly characteristic of Fe^{+3} and one of them may be due to the presence of Oxygen or Lanthanium vacancies. The existence of magnetic ordering is investigated by ^{57}Fe Mössbauer measurements at 4.2K for samples with 0.5% Fe.

Key-words: Superconductivity; Mössbauer spectroscopy; Iron in ceramics; Oxides; Magnetic ordering.

1. INTRODUCTION

The $La_{1.85}Ba_{0.15}CuO_4$ superconductor oxide has lower T_c as compared with $YBa_2Cu_3O_7$ oxides, however both are antiferromagnetically ordered in the corresponding insulator phase; i.e. La_2CuO_4 and $YBa_2Cu_3O_6$. The effect of several substituents in Cu sites is different in the two systems; for example 3% Fe in the $La_{1.85}Ba_{0.15}CuO_4$ destroys completely the superconductive transition[1], while in the $YBa_2Cu_3O_7$ even with 10% of Cu substituted by Fe it is possible to measure, by DC magnetization, a T_c onset of $\sim 54K$[2]. The local studies based on the Mössbauer Spectroscopy of ^{57}Fe: $YBa_2Cu_3O_y$ oxides allowed us to establish the preferential occupation of the Cu1 site by Fe with different oxygen coordination. We report here ^{57}Fe Mössbauer studies in $La_{2-x}Ba_xCuO_4$ oxides (x = 0, 0.15, 0.25 and 0.35) focusing on the quality of sample preparation and the presence of a magnetic ordered state mainly for 0.5% Fe concentration.

2. EXPERIMENTAL

The samples of ^{57}Fe: $La_{2-x}Ba_xCuO_4$, with x=0, 0.15, 0.25 and 0.35, were prepared by sintering in air pellets of appropriated amounts of La_2O_3, $BaCO_3$, CuO and $^{57}Fe_2O_3$ at 900C for 20h followed by regrinding and sintering two times at the same conditions. The next step involves the regrinding and heating at 1100C for 20h and cooling down to 600C at rate of 50C/h. The samples were kept at this temperature for 50h and then cooled down to room temperature (RT) at the same rate[3].

The X-ray analysis and Mössbauer spectra at RT were taken after the first sintetization and after the final preparation.

3. RESULTS AND DISCUSSION

The $La_{2-x}Ba_xCuO_y$ oxides have a single site for Cu with a distorted octahedral oxygen coordination. The Fe ions are expected to occupy the Cu site and in the case of La_2CuO_4 it is possible to introduce 10% Fe without disturbing the orthorhombic structure. The substitution of Cu by ^{57}Fe in very low concentration as 0.5% can be used as a probe to detect by means of ^{57}Fe Mössbauer Spectroscopy how the Ba and oxygen content influence the electronic charge distribution in Cu site.

The Mössbauer spectra at RT obtained after two treatment at 900C for 20h (sample I) are shown in Fig. 1, for x = 0.15, 0.25 and 0.35. The corresponding Mössbauer parameters are listed in Table I. The doublet D1 can be associated with Fe^{+3} at Cu site, while doublet D2 responsible for the asymmetry of the spectra, can be attributed to an impurity phase always present with broad lines and low relative intensity.

After the final treatment the sample II exhibited a better resolved Mössbauer spectrum (Fig. 2) with symmetrical lines, since D2 is no more present. The X-ray diffraction studies revealed also the improvement of the tetragonal structure.

The comparison of Mössbauer parameters between La_2CuO_4 (IS = 0.30mm/s, QS = 1.60mm/s) and $La_{2-x}Ba_xCuO_4$ (Table I) indicated that

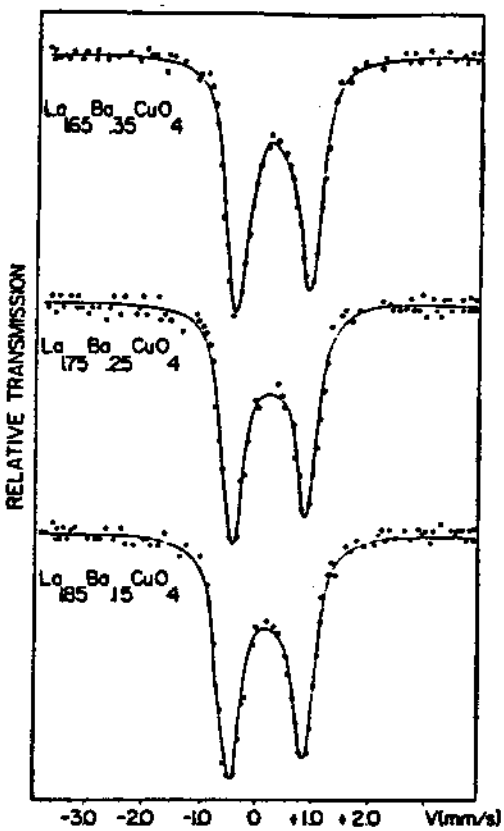

FIG. 1 - Mössbauer spectra at RT after two treatment at 900C for 20h.

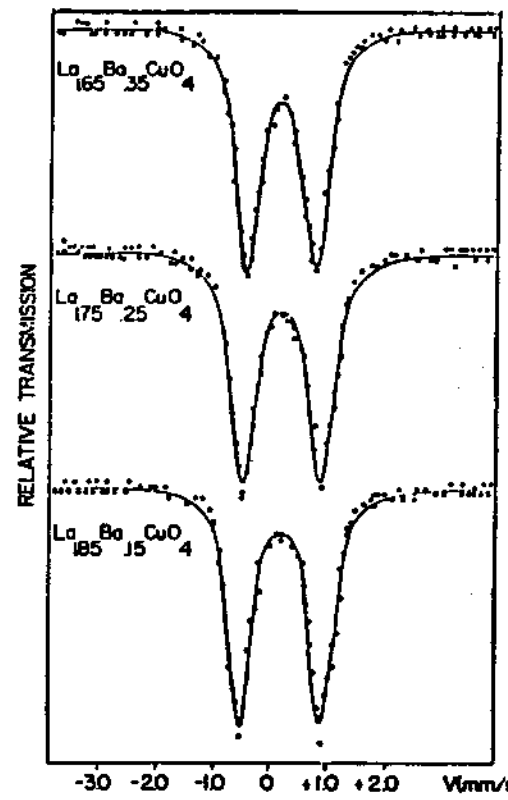

FIG. 2 - Mössbauer spectra at RT after the final treatment at 1100C for 20h.

$La_{2-x}Ba_xCuO_4$		IS1 mm/s	QS1 mm/s	IS2 mm/s	QS2 mm/s
x =0.15	I	0.28	1.32	0.15	0.56
	II	0.30	1.43	-	-
x =0.25	I	0.28	1.30	0.11	0.49
	II	0.29	1.37	-	-
x =0.35	I	0.28	1.30	0.18	0.62
	II	0.25	1.19	-	-

TABLE I: Fitted hyperfine parameters IS: isomer shift (ref. Fe-metal). QS: quadrupole splitting.

there is a clear decrease of the quadrupole interaction with the Ba content which may reflect the more symmetric electronic distribution at Cu site in the tetragonal symmetry.

The phase diagram of $La_{2-x}Ba_xCuO_4$ [3] is known to go from an antiferromagnetic ($x = 0$) through a spin-glass-like region ($0 \leq x \leq 0.15$) to the superconducting non magnetic region ($0.15 \leq x \leq 0.35$). The Mössbauer measurements performed at 4.2K with samples containing 0.5% Fe may reflect the ordered state of the Cu magnetic moments. Fig. 3a presents the spectra corresponding to 0.5% Fe in La_2CuO_4 displaying a magnetic pattern with internal field of ~ 48T which is perpendicular to the principal component of the electric field gradient (EFG) beeing parallel to the c direction at the iron site. This result is in agreement with neutron diffraction data [4] for pure La_2CuO_4 and is an indication that iron probe can provide important informations about the ordered state of Cu moments.

The broad absorption lines of the magnetically splitted Mössbauer spectra at 4.2K obtained for 0.5% Fe in $La_{1.85}Ba_{0.15}CuO_4$ (Fig. 3b) are typical of a distribution of H_{eff} at the iron site with the mean value very close to the one obtained for the ^{57}Fe: La_2CuO_4. This distribution of H_{eff} may be due to different orientation of the principal axis of EFG and the iron magnetic moments induced by the Ba atoms. Mössbauer measurements as a function of temperature may give information about the freezing mechanism of the iron magnetic moments.

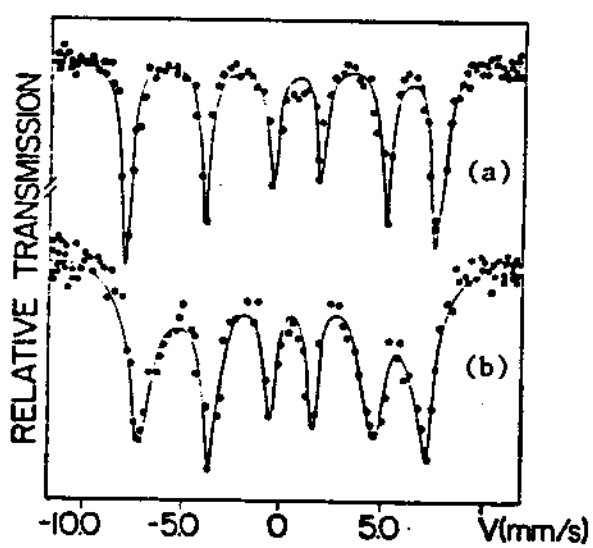

FIG. 3 - Mössbauer spectra at 4.2K:
a) La_2CuO_4 with 0.5% Fe.
b) $La_{1.85}Ba_{0.15}CuO_4$ with 0.5% Fe.

The origin of this magnetic ordering of iron moments is not clear

since it may arise from long range correlation of iron moments or can be established through magnetic moments induced in the Cu ions. Anyhow, this result implies again the coexistence of magnetic and superconducting states in the same sample of superconducting oxide, similar to what was observed for $x \geq 0.03$ Fe in $YBa_2Cu_3O_7$ compounds.

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