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TEMPERATURE DEPENDENCE OF LATTICE DYNAMICS AND STRUCTURE OF SUPERCONDUCTING CERAMICS $La_{2-x}Sr_xCuO_{4-y}$ BY NEUTRON SCATTERING

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Almost immediately upon the discovery of new superconducting ceramics the first neutron works have been performed[1]. The main peculiarities of the phonon density of states have been obtained and the results for nonsuperconducting La_2CuO_4 and superconducting $La_{1.85}Sr_{0.15}CuO_4$ have been compared. Now on the single crystal of La_2CuO_4 the soft mode was measured, which is responsible for phase transition from the tetragonal into orthorhombic phase at about 500K[2]. But the detailed investigations of the low frequency part of spectra at low temperatures are absent so far. We have performed the experiments on neutron scattering in $La_{2-x}Sr_xCuO_4$ compounds for two x values and in a temperature range from 290K to 10K. The measurements have been made with the KDSOG-M spectrometer[3] installed on the high flux pulsed reactor IBR-2[4]. The main preferences of this spectrometer consist in the possibility of measuring neutron diffraction (ND) and inelastic neutron scattering (INS) simultaneously and of measuring INS in the neutron energy loss regime which allows one to investigate lattice dynamics at low temperatures.

Our samples were prepared from La_2O_3, $SrCO_3$ and CuO by the well-known method (see for example [5]). To control the chemical composition, we used the neutron activation analysis[6]. The amount of impurities were less than 0.5 at.% and strontium concentration in $La_{2-x}Sr_xCuO_{4-y}$ was $x = 0.2 \pm 0.02$. The structure of the samples at room temperatures have been tested with the DRON-3M diffractometer. The structure of La_2CuO_{4-y} completely agrees with the existing data[7] - the sample revealed the orthorhombic phase (Bmab) which could be found from the (200) and (020) reflections splitting. The $La_{1.8}Sr_{0.2}CuO_{4-y}$ had a tetragonal structure I4/mmm with lattice parameters completely the same as published earlier.[8] The measurements of the superconducting transition temperature by resistive and magnetic methods revealed that the superconducting transition began at about 20K. This fact proves the oxygen deficiency in the sample.[9]

For neutron measurements about 100g of the samples were put into a helium cryostat. ND and INS spectra were measured simultaneously. Experimental data were normalized to the monitor. Diffraction spectra measured for the scattering angles 2θ= 28°, 48°, 68° and 88° were normalized to the spectra of incident neutrons. Inelastic scattering spectra were measured for the scattering angles 30°, 50°, 70° and 90° in transmission geometry and for 80°, 100°, 120° and 140° in reflection geometry. The background from the cryostat without sample was subtracted and data from different angles were summed up. The average time to measure one spectrum was about one day.

As one can see from fig. 1, the diffraction spectrum at T = 10K for the orthorhombic phase of La_2CuO_{4-y} differs from the spectrum for the tetragonal phase of $La_{1.8}Sr_{0.2}CuO_{4-y}$ in the presence of (012) and (014) reflections which are forbidden for the tetragonal phase and in the splitting of (020), (200) and (022), (202) reflections.

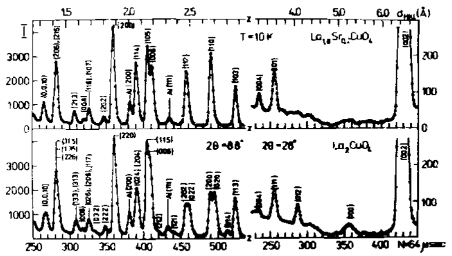

Fig.1. Neutron diffraction spectra at T = 10K from the tetragonal phase of $La_{1.8}Sr_{0.2}CuO_{4-y}$ and from the orthorhombic phase of La_2CuO_{4-y} for the scattering angles 2θ = 28° and 88°. N - channel number with the width 64μsec.

At T = 77K the diffraction spectra are practically the same and at 290K we cannot see the splitting of (020), (200) and (022), (202) reflections due to the resolution limitation of our spectrometer. But also at a room temperature one can easily distinguish the orthorhombic phase of La_2CuO_{4-y} by the (012) and (014) reflections. At 77K the (100) reflection appears in diffraction spectrum of La_2CuO_{4-y} and its intensity increases when temperature decreases down to 10K (fig.1). The existence of this reflection proves the antiferromagnetic ordering/10/ and oxygen deficiency in the sample /9/. The $La_{1.8}Sr_{0.2}CuO_{4-y}$ sample was in the tetragonal phase at all temperatures in good agreement with the published data/11/. The temperature decreasing from 290K to 10K causes only shifting of the peaks, which proves the change of interplane distances less than on 0.006Å.

Inelastic neutron scattering spectra from La_2CuO_{4-y} and $La_{1.8}Sr_{0.2}CuO_{4-y}$ at temperatures 10K, 77K and 290K are shown in figs.2a, b. The main features in these spectra well agree with the data published earlier/1/. But at 77K and 10K one can see an inelastic line near the energy transfer 6meV which was not observed before.

The raising of its intensity is obvious when temperature decreases. The transformation of experimental data into the generalised frequency distribution by using the one - phonon formula gives us a good coincidence of data obtained at different temperatures. These results are also in good agreement with the literature/1/. Exception is the above - mentioned peculiarity at 6meV. After the transformation their intensity increases when the temperature decreases. This fact and an angular dependence of the intensity of this line prove their nonphonon character.

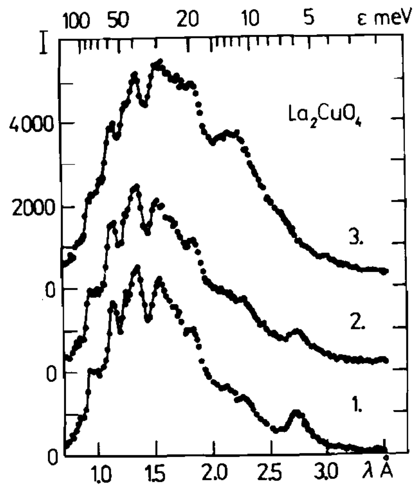

Fig.2a. INS spectra from La_2CuO_{4-y}. 1 - T = 10K, 2 - T = 77K, 3 - T = 290K, the vertical axis - intensity normalised to 10^7 monitor counts, ε - energy transfer in meV.

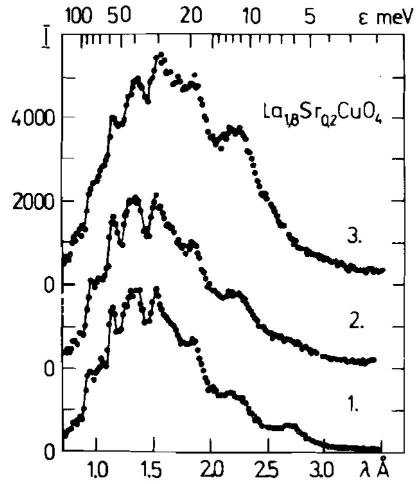

Fig.2b. INS spectra from $La_{1.8}Sr_{0.2}CuO_{4-y}$. 1 - T = 10K, 2 - T = 77K, 3 - T = 290K

For the quantitative analysis of the 6meV feature a simple model was used. The scattering law was chosen as a Gauss function plus the Debye distribution ($\sim \varepsilon^2$) and convolution with the spectrometer resolution was performed. The results of fitting of the experimental data by that model using a least square method are shown in fig.3. The parameters of the Gauss function are shown in the table. It is clearly seen that the intensity of the line at 6meV increases when temperature decreases and its width is practically invariable for both the samples under investigation. This fact and the decrease of the intensity of this line with increasing scattering angle and therefore momentum transfer prove that the corresponding excitation has a magnetic nature. This circumstance is rather strange for such - type compounds. Nominally magnetoactive are $3d^9$ electrons of copper whose orbital moment is practically fully "frozen" by the crystal field. If the observed excitation is due

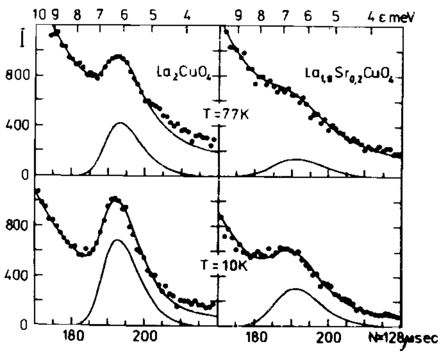

Fig. 3. The results of model fitting of the low - frequency part of the experimental INS spectra at 77K and 10K Lowest solid curve - peculiarity at 6meV without Debye background N - channel number with the width 128 μsec, ε - energy transfer in meV.

Table. The parameters of Gauss function which have been obtained from the fitting of low frequency part of the spectra.

Sample	Temperature (K)	Peak position (meV)	Integral peak intensity (arb. units)	FWHM (meV)
La_2CuO_{4-y}	77	6.2 ± 0.1	11 ± 1	0.9 ± 0.1
	10	6.27±0.03	18.1±0.5	0.90±0.05
$La_{1.8}Sr_{0.2}CuO_{4-y}$	77	6.6 ± 0.2	5 ± 2	1.5 ± 0.5
	10	6.57±0.05	9.8 ± 0.5	1.5 ± 0.1

to the antiferromagnetic spin waves, then there exists a gap of about 6meV and a low - frequency part of the magnetic excitation spectrum has a small dispersion, because the position of the peak is practically independent of the momentum transfer and this peak has a small width. The gap may exist due to anisotropic exchange interactions.

The correspounding strength of the internal molecular field influencing on S = 1/2 state of 3d electrons of copper will be (g = 2, ΔE = 6.2meV) of about 54.4T. In that interpretation, the Sr doping may break the long - range magnetic order but conserve the small range one. Due to this conservation one can observe the above mentioned excitation in $La_{1.8}Sr_{0.2}CuO_{4-y}$ with a less intensity and a larger width.

This is only a preliminary analysis of the results. Now it is difficult to make a final conclusion about the physical nature of the observed peculiarity and about their connection with the superconducting properties. But it is interesting to point out that the excitation energy by the order of magnitude corresponds to the superconducting transition temperature in La - Sr - Cu - O systems. In connection with this fact we are planning to continue the investigations of such systems for different Sr concentrations in the samples with oxygen deficiency as well in the samples annealed in the oxygen atmosphere and with good superconducting properties.

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Нейтронные исследования температурной зависимости динамики и структуры сверхпроводящих керамик $La_{2-x}Sr_xCuO_{4-y}$

На спектрометре КДСОГ-М на импульсном реакторе ИБР-2 одновременно исследованы спектры неупругого рассеяния и дифракции нейтронов в соединениях La_2CuO_{4-y} и $La_{1,8}Sr_{0,2}CuO_{4-y}$ при температурах 290, 77 и 10 К. В спектрах неупругого рассеяния при температурах 77 и 10 К обнаружена линия с энергией около 6 мэВ, температурная зависимость интенсивности которой указывает на магнитный характер соответствующего ей возбуждения. По результатам дифракции нейтронов исследуемые образцы во всем интервале температур имели орторомбическую и тетрагональную симметрию соответственно. Проявление рефлекса (100) в орторомбической фазе La_2CuO_{4-y} свидетельствует о наличии антиферромагнитного упорядочения при низких температурах.

Работа выполнена в Лаборатории нейтронной физики ОИЯИ.

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Temperature Dependence of Lattice Dynamics and Structure of Superconducting Ceramics $La_{2-x}Sr_xCuO_{4-y}$ by Neutron Scattering

The spectra of inelastic neutron scattering and neutron diffraction from La_2CuO_{4-y} and $La_{1.8}Sr_{0.2}CuO_{4-y}$ at temperatures 290, 77 and 10 K are investigated simultaneously with the KDSOG-M spectrometer on the IBR-2 pulsed reactor. A new line at 6 meV has been observed in inelastic spectra at 77 K and 10 K. The temperature dependence of the intensity of this line points to the magnetic nature of the corresponding excitation. According to diffraction data the samples under investigation had orthorhombic and tetragonal symmetry respectively in the whole temperature range. The (100) reflection for the orthorhombic phase of La_2CuO_{4-y} reveals the antiferromagnetic ordering at low temperatures.

The investigation has been performed at the Laboratory of Neutron Physics, JINR.

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