

ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

$E \rightarrow$ Д14-89-455

μ SR - INVESTIGATION
OF HIGH- T_c SUPERCONDUCTORS
AT THE LNP JINR PHASOTRON

Submitted to the International Seminar on High
Temperature Superconductivity, Dubna, June 28 -
July 1, 1989

1989

D.T.Bezhitadze¹, I.P.Borovinskaya², J.Burianek³,
V.N.Duginov, Yu.F.Eltzev⁴, V.G.Grebinnik, I.I.Gurevich¹,
S.Kapusta, V.R.Karasik⁴, B.F.Kirillov¹, E.P.Krasnoperov¹,
A.B.Lazarev, M.D.Nersesyan², B.A.Nikolsky¹, V.G.Olshevsky,
O.E.Omelyanovsky⁴, A.G.Peresada², A.V.Pirogov¹,
V.Yu.Pomjakushin, A.N.Ponomarev¹, S.Safrata⁵, J.Šebek⁵,
S.N.Shilov, V.A.Suetin¹, G.F.Tavadze¹, V.Valvoda³, V.A.Zhukov

¹I.V.Kurchatov Institute of Atomic Energy, Moscow, USSR

²Institute of Structural Macrokinetics, USSR Academy of Sciences, Chernogolovka, USSR

³Faculty of Mathematics and Physics, Charles University, Prague, Czechoslovakia

⁴Physical Institute, USSR Academy of Sciences, Moscow, USSR

⁵Physical Institute of Czechoslovak Academy of Sciences, Prague, Czechoslovakia

Introduction

The valuable information can be obtained about the values of the internal magnetic fields and their distributions in the study of superconductors by the μ SR-method /1/. In high- T_c superconductors these fields are connected, on the one hand, with the formation of the vortex lattice in the mixed state and, on the other hand, with the magnetic ordering of Cu-ions in the CuO-layers /2-10/ or with the presence of rare-earth elements with high atomic magnetic moments in the superconductor /11-14/.

The magnetic field distribution data in the mixed state allow the penetration depth of the magnetic field, magnetization to be determined and the pinning effects to be studied. The study of the phenomena of the magnetic ordering and coexistence of magnetism and superconductivity is of interest from the point of view of the explanation of the mechanism responsible for superconductivity /15/.

In the present work the results of the μ SR-investigation are presented for the following high- T_c superconductors: $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.00, 0.01, 0.05, 0.07, 0.10, 0.15, 0.25$), $\text{Ho(Er)Ba}_2\text{Cu}_3\text{O}_{7-y}$ and Bi-Sr-Ca-Cu-O .

Experimental conditions

The phasotron of the Laboratory of Nuclear Problems produces a (59 MeV proton beam /16/. Usually the extracted proton beam of the intensity $\sim 1 \mu\text{A}$ is used in the μ SR-experiments. The secondary beam of the positive muons has the following characteristics: momentum 130 MeV/c, intensity $\sim 10^5$ 1/s, polarization $\sim 80\%$, aperture $5 \times 5 \text{ cm}^2$. The experiments were carried out with one of the spectrometers of the μ SR-facility /17/, which ensured measurements in the magnetic field up to 0.5 T, directed perpendicular to the muon spin direction. The investigations were carried out in the temperature interval 4.2-300 K. The samples had a form of disc ~ 40 mm in diameter and ~ 10 mm thick. The discs were so placed in the beam that their faces were perpendicular to the beam direction and the external magnetic field was directed along these faces.

The experiments were carried out under three conditions:

a) measurements on zero-field-cooled samples (ZF), b) measurements on field-cooled samples (FC) and c) measurements on zero-field-cooled samples followed by an increase in the magnetic field at a fixed temperature or an increase in the temperature at a fixed magnetic field (ZFC).

The μ SR-signals are fitted to the function:

$$N(t) = N_0 e^{-t/\tau_\mu} (1 + P(t)) + N_B, \quad (1)$$

where N_0 is the count of the $\mu^+ \rightarrow e^+$ decay positrons at the initial moment of time, $\tau_\mu \approx 2.2 \mu\text{s}$ is the muon lifetime, N_B is the time-independent constant, $P(t)$ is the time-dependent function containing information about the muon spin evolution inside the sample. The form of $P(t)$ will be given below for each individual case.

Antiferromagnetism and spin-glassy state

The $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ samples with $x = 0.00, 0.01, 0.05$ and 0.07 were selected to study these phenomena.

For the samples with $x = 0.00$ and 0.01 at (ZF)-conditions the function $P(t)$ was taken in the form:

$$P(t) = a \left[\frac{1}{3} + \frac{2}{3} e^{-\Lambda t} \cos \omega_\mu (t+t_0) \right] + (a_\Sigma - a) e^{-\Lambda_0 t}, \quad (2)$$

where the first term describes the behaviour of the spins of muons stopped at the interstitial sites, where there is a magnetic field. The second term describes the same behaviour at the places free of the magnetic field. a is the asymmetry coefficient of the $\mu^+ \rightarrow e^+$ decay related to the muons affected by the magnetic field. $a_\Sigma \approx 0.18$ is the total asymmetry coefficient. Λ is the muon spin relaxation rate connected with the dispersion $\langle \Delta B_\mu^2 \rangle^{1/2}$ of the magnetic field on the muon by the equation $\Lambda^2 = \gamma_\mu^2 \langle \Delta B_\mu^2 \rangle$, where $\langle B_\mu \rangle$ is the average magnetic field on the muon. The coefficients $1/2$ and $1/3$ arise from the averaging over polycrystal.

The temperature dependences of the average magnetic field $\langle B_\mu \rangle$ on the muon, relaxation rate Λ and asymmetry a are represented in Fig. 1 a,b,c. As seen from Fig. 1a, the noticeable magnetic field is acting on the muons at low temperatures ($< T_N$). This demonstrates the presence of the antiferromagnetic ordering in the samples. The solid lines in this figure are the theoretical dependences of magnetization calculated in the approach of the molecular field for spin $1/2$.

The slow (except for the region near T_N) temperature dependence of Λ (Fig. 1b) shows that the relaxation rate Λ is determined by the static fields. Despite the fact that the average fields on the muon (Fig. 1a) do not so much differ for both the samples their relaxation rates differ by more than a factor of two: $\Lambda (x=0.00) \sim 1.8 \mu\text{s}^{-1}$ and $\Lambda (x=0.01) \sim 4.5 \mu\text{s}^{-1}$. It can be interpreted as the increasing deviations of the magnetic moments of Cu^{2+} from the antiferromagnetic ordering axis.

Fig. 1. Temperature dependence of the average magnetic field on the muon $\langle B_M \rangle$, relaxation rate Λ and asymmetry Q (see form. (2)) for the samples $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.00$ and 0.01) in a zero magnetic field. Solid lines are theoretical.

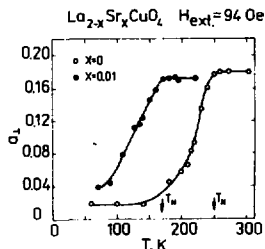
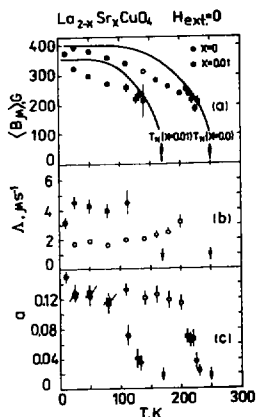


Fig. 2. Temperature dependence of the asymmetry coefficient Q_1 (see form. (3)) in the external magnetic field ~ 100 Oe for the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.00$ and 0.01). The curves are to guide the eye.

Fig. 1c shows the temperature dependence of the asymmetry coefficient Q . In the temperature interval significantly lower than T_N the ratio Q/Q_Z represents the part of the sample which is in the antiferromagnetic state. In the region near T_N the gradual reduction of Q can be connected both with inhomogeneity of the oxygen content over the sample and with an increase in the amplitude of the fluctuated part of the magnetic field.

It was difficult to evaluate T_N in the (ZF)-experiments because of a rapid increase in the muon spin relaxation rate leading to a decrease of the μSR -signal amplitude. To determine T_N the experiments were carried out at $H_L \sim 100$ Oe. In this case the function $P(t)$ was selected in the form:

$$P(t) = a_{\perp} e^{-\Lambda t} \cos \omega_M(t+t_0). \quad (3)$$

The Neel temperature T_N was determined as the one corresponding to the beginning of the decrease in the amplitude of the precession signal (Fig. 2). According to the evaluation, $T_N(x=0.00) \approx 250$ K and $T_N(x=0.01) \approx 170$ K. As seen from Fig. 2 the transition into the antiferromagnetic state is considerably delayed (by more than 50 K).

No precession frequency was found for the sample with the Sr-concentration $x = 0.05$ in the accessible temperature interval 4.8-300 K.

In this case the experimental spectrum was described by the function:

$$P(t) = a_{\Sigma} e^{-(\Lambda t)^{\alpha}} \quad (4)$$

Fig. 3 shows the temperature dependence of Λ and α . The value α was accepted equal to 1 at the temperatures higher than 7 K where the exponential function satisfactory describes the polarization function. The fluctuations of the internal magnetic fields are slowed down below this temperature at the approach to the phase transition point.

As follows from the (FC)-measurements for the sample with $x=0.07$, the transition into the magnetic ordering state starts at the temperatures lower than $T_f \sim 10$ K. We have not observed the muon spin precession for the sample with $x = 0.07$ (like the sample with $x = 0.05$) in the (ZF)-measurements at the accessible for us temperatures. However the relaxation function begins acquiring the form of the Kubo-Toyabe function $1/18$ as the temperature drops. In this connection it was assumed that the magnetic ordering state of the sample with $x = 0.07$ at low temperatures is the spin-glass state. Therefore

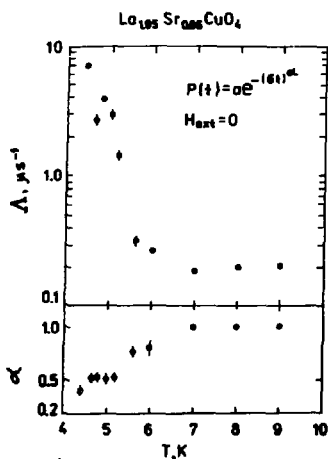


Fig. 3. Temperature dependence of the Λ and α -parameters (see form. (4)) for the $\text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4$ sample.

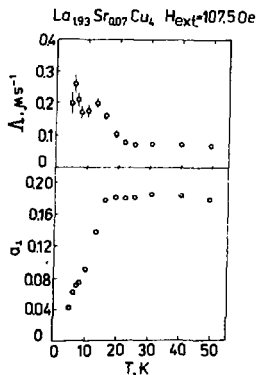


Fig. 4. Temperature dependences of the relaxation rate Λ and the asymmetry coefficient α_1 in the external magnetic field ~ 100 Oe for the $\text{La}_{1.93}\text{Sr}_{0.07}\text{CuO}_4$ sample.

for describing the μ SR-spectra under the (ZF)-conditions the function $P(t)$ may be expressed as

$$P(t) = a \left[\frac{1}{3} + \frac{2}{3} (1 - \Delta^2 t^2) \exp(-\frac{1}{2} \Delta^2 t^2) \right] + (a_\Sigma - a) e^{-\Lambda t} \quad (5)$$

where $\Delta^2 = \gamma_M^2 \langle (B_M - \langle B_M \rangle)^2 \rangle$. At the temperature 4.8 K the value of Δ corresponds to the width of the magnetic field distribution $\langle (B_M - \langle B_M \rangle)^2 \rangle^{1/2} \approx 210$ G.

Magnetic ordering and superconductivity in RE-Ba-Cu-O-compounds

The $\text{HoBa}_2\text{Cu}_3\text{O}_{7-y}$ and $\text{ErBa}_2\text{Cu}_3\text{O}_{7-y}$ samples were selected to study the influence of the magnetic moments of rare-earth elements in high- T_c superconductors. The experiments were carried out under the (ZF)-conditions. The function $P(t)$ was selected in the form:

$$P(t) = a \cdot e^{-\Lambda t} + (a_\Sigma - a) e^{-(\sigma \cdot t)^2} \quad (6)$$

where the first term corresponds to the case when muons are stopped at the sites nearest to the magnetic atoms (Ho,Er) and the second term describes the relaxation process for the muons stopped at the sites far from Ho(Er)-atoms.

The temperature dependences of Λ for the Ho(Er)-Ba-Cu-O samples are shown in figure 5. As seen, the Ho-Ba-Cu-O sample shows fast muon spin depolarization in the zero magnetic field in the temperature region 4.2 - 10 K which is ten times greater than that at high temperatures ($0.2 \mu\text{s}^{-1}$).

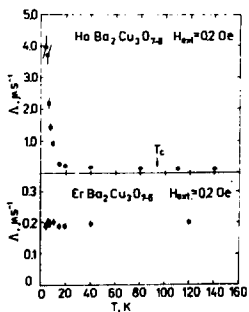


Fig. 5. Temperature dependence of the muon spin relaxation rate Λ (see form. (6)) for the Ho(Er)Ba₂Cu₃O_{7-y} in the zero external magnetic field.

In the case of Er-Ba-Cu-O the depolarization rate remains low ($0.2 \mu\text{s}^{-1}$) in the temperature region 4.6 - 270 K. It is necessary to point out that the behaviour of both samples is similar in ZFC-measurements at low temperatures.

When the magnetic field is increased, one can observe a sharp increase in the depolarization rate, which becomes greater than $8 \mu\text{s}^{-1}$ at the temperature < 10 K in the magnetic fields higher than 100 Oe.

Investigation of high- T_c superconductors in the mixed state

To obtain the data about the magnetic field penetration depth of superconductors and about the pinning effects the following ceramics were investigated: $La_{2-x}Sr_xCuO_4$ ($x = 0.10, 0.15, 0.25$), $ErBa_2Cu_3O_{7-y}$ and $Bi-Sr-Ca-Cu-O$. The measurements were performed under (FC) and (ZFC)-conditions in the magnetic fields 100 and 400 Oe. The function $P(t)$ was approximated by the equation:

$$P(t) = a_{\perp} e^{-(\sigma \cdot t)^2} \cos \omega_{\mu} (t + t_0), \quad (7)$$

where σ is the muon spin relaxation rate connected with the dispersion of the magnetic field distribution by the formula $2\sigma^2 = \gamma_{\mu}^2 \langle \Delta B_{\mu}^2 \rangle$

The magnetic field penetration depth was calculated from the experimental dependences $\sigma(T)$ obtained under the (FC)-conditions on the basis of the equation /19/ :

$$\langle \Delta B^2 \rangle = 0.0037 \varphi_0^2 \lambda_{eff}^{-4}, \quad (8)$$

where $\varphi_0 = 2.07 \cdot 10^{-7} \text{ G/cm}^2$ is the quantum of the magnetic flux, λ_{eff} is the effective penetration depth of the magnetic field for a polycrystalline superconductor.

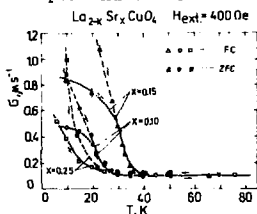


Fig. 6. Dependence of the relaxation rate σ (see form. (7)) on the temperature for the $La_{2-x}Sr_xCuO_4$ ($x=0.10, 0.15$ and 0.25) in the magnetic field 400 Oe. The curves are to guide the eye.

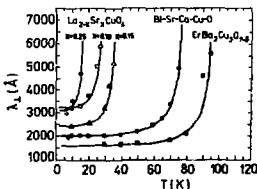


Fig. 7. Temperature dependences of the magnetic field penetration depth λ_{\perp} for the $La-Sr-Cu-O$, $Bi-Sr-Ca-Cu-O$ and $Er-Ba-Cu-O$ samples. Solid curves show the theoretical dependences calculated according to formula (10).

Relation (8) can be used for the perfect triangular vortex lattice if $\lambda_{eff} > L$ (where L is the average distance between vortices). The penetration depth values λ_{\perp} corresponded to the external magnetic field direction perpendicular to the basis face of the crystal was determined according to formula /20/ :

$$\lambda_{eff} = 1.23 \lambda_{\perp} \quad (\text{at } \lambda_{\parallel} / \lambda_{\perp} > 5). \quad (9)$$

Fig. 6 shows the dependences $\sigma(T)$ for the La-Sr-Cu-O samples obtained under (ZFC)-conditions in the external magnetic field 400 Oe. The dependences $\sigma(T)$ have the same form in the magnetic field 100 Oe. The fact that σ is independent of the magnetic field allows us to use formula (1) for calculation of λ_{eff} . The positive curvature in $\sigma(T)$ near T_c can be explained by the finite width of the transition into the superconducting state. The error connected with the demagnetization factor of the superconducting grain was not taken into account in the analysis of the experimental data.

The calculations of λ_1 for Er-Ba-Cu-O and Bi-Sr-Ca-Cu-O samples were carried out in a similar manner. The summary results for all samples are shown in Fig. 7.

Fig. 8. Dependence of the muon spin relaxation rate σ and difference $B_M - H_{ext}$ on the external magnetic field at an increase followed by a decrease of the magnetic field for the $La_{1.85}Sr_{0.15}CuO_4$. The lines are drawn only to guide the eye.

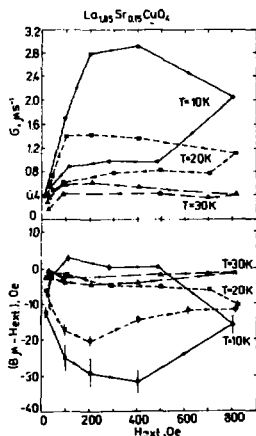
The obtained data were approximated by the formula :

$$\lambda_1(T) = \lambda_1(0) / \sqrt{1 - (T/T_c)^4}, \quad (10)$$

in which the values of $\lambda_1(0)$ were found by the least square method. For the samples for which the temperature

dependences of λ_1 were well described by this formula, the following values of $\lambda_1(0)$ were obtained: $3200 \pm 70 \text{ \AA}$ for $La_{1.9}Sr_{0.1}CuO_4$ ($T_c \approx 30 \text{ K}$), $2420 \pm 60 \text{ \AA}$ for $La_{1.85}Sr_{0.15}CuO_4$ ($T_c \approx 37 \text{ K}$), $2000 \pm 50 \text{ \AA}$ for Bi-Sr-Ca-Cu-O (phase 2212, $T_c \approx 80 \text{ K}$) and $1600 \pm 50 \text{ \AA}$ for $ErBa_2Cu_3O_{7-y}$.

The magnetic field penetration depths can be also calculated from the dependences $\sigma(H_{ext})$ obtained under (ZFC)-conditions in the magnetic field corresponding to the established vortex lattice. However the influence of the pinning in this procedure can introduce the errors in the evaluation of λ . One can get information about the pinning effects from Fig. 8 which shows the dependences of the relaxation rate σ and difference $B_M - H_{ext}$ on the external magnetic field at different temperatures under (ZFC)-conditions, for the



$\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. As seen from the figure, a noticeable hysteresis is observed at low temperatures and reduced at high temperatures.

Conclusion

In the given paper the essential results on the μSR -study of the high- T_c superconductors obtained in the recent time at the IEP JINR phasotron are presented. More detailed results will be published elsewhere. The detailed investigations of the most interesting phenomena will also be continued. The spin-glassy behaviour of the high- T_c superconductors, glassy superconductivity and the relevant long time relaxation processes should be carefully studied. It is also intended to extend the measurements of the penetration depths in various superconductors with different T_c .

References

1. Schenck A. Muon Spin Rotation Spectroscopy (Adam Hilger Ltd 1985)
2. Uemura Y.J. et al. Phys.Rev.Lett., 1987, v.58, p.1045
3. Vaknin D. et al. Phys.Rev.Lett., 1987, v.58, p.2802
4. Budnick J.I. et al. Phys.Lett., 1987, v.A124, p.103
5. Budnick J.I. et al. Europhys.Lett., 1988, v.5(7), p.651
6. Watanabe I. et al. J.Phys.Soc.Jpn., 1987, v.56, p.3028
7. Kitaoka Y. et al. J.Phys.Soc.Jpn., 1987, v.56, p.3024
8. Mishida N. et al. Jpn.J.Appl.Phys., 1987, v.26, p.L1056,
J.Phys.Soc.Jpn., 1988, v.57, p.599
9. Brewer J.H. et al. Phys.Rev.Lett., 1988, v.60, p.1073
10. Tranquada J.M. et al. Phys.Rev.Lett., 1988, v.60, p.156
11. Golnik A. et al. Phys.Lett., 1987, v.A125, p.71
12. Mishida N. et al. Jpn.J.Appl.Phys., 1988, v.27, p.L94
13. Duginov V.H. et al. JINR Rapid Commun., N.4(30), Dubna, 1988, p.63.
14. Kuno Y. et al. Phys.Rev., 1988, v.B38, p.9276
15. Anderson P.W. et al. Phys.Rev.Lett., 1987, v.58, p.2750
16. Abazov V.M. et al. JINR, 9-87-322, Dubna, 1987
17. Gaganov I.A. et al. in: Muons and Pions in Matter,
JINR, D14-87-799, Dubna, 1987, p.431
18. Kubo R. and Toyabe T. in: Magnetic Resonance and Relaxation,
ed. by Blinc R., 1967, p.810
19. Brandt E.H. Phys.Rev., 1988, v.B37, p.2349
20. Barford N. and Gunn J.M.E. Physica, 1988, v.C156, p.515

Received by Publishing Department
on June 26, 1989.

Исследование высокотемпературных сверхпроводников μ SR-методом на фазотроне ЛЯП ОИЯИ

μ SR-методом исследованы высокотемпературные сверхпроводники (ВТСП) $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.0, 0.25$), $\text{Ho}(\text{Er})\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ и Bi-Sr-Ca-Cu-O (фаза -2212 с небольшой примесью фазы 2223) в нулевом и поперечных внешних магнитных полях -100 и -400 Э в диапазоне температур 4,2+300 К. Для образцов La-Sr-Cu-O с $x=0$ и $x=0.01$ магнитное упорядочение наступает при температурах ниже $T = 250$ К и $T = 170$ К соответственно. Значительное возрастание ширины распределения магнитного поля на муоне при увеличении содержания стронция свидетельствует об увеличении отклонения магнитных моментов Cu^{2+} от оси магнитного упорядочения. При содержании стронция $x = 0,07$ результаты обработки μ SR-спектров указывают на формирование спин-стеклового состояния в образце. В противоположность керамике $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ не обнаружено магнитного упорядочения атомов Er в образце $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Определены глубины проникновения магнитного поля в сверхпроводник $\lambda_1(0)$: $(3200 \pm 79) \text{ \AA}$ - $\text{La}_{1,9}\text{Sr}_{0,1}\text{CuO}_4$; $(2420 \pm 60) \text{ \AA}$ - $\text{La}_{1,85}\text{Sr}_{0,15}\text{CuO}_4$; $(2000 \pm 50) \text{ \AA}$ - Bi-Sr-Ca-Cu-O (фаза 2212) и $(1600 \pm 50) \text{ \AA}$ - $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1989

μ SR - Investigation of High- T_c Superconductors at the LNP JINR Phasotron

High- T_c superconductors $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.0, 0.25$), $\text{Ho}(\text{Er})\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$ and Bi-Sr-Ca-Cu-O (the main phase 2212 with a small admixture of the 2223-phase) were investigated in the zero and 100+400 Oe transverse external magnetic fields in the temperature interval 4.2+300 K. The magnetic ordering for the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ samples with $x=0.00$ and $x=0.01$ occurs below $T_N = 250$ K and $T_N = 170$ K respectively. A considerable increase in the distribution width of the magnetic field on the muon with an increase in the Sr-content indicates an enhancement of the Cu^{2+} magnetic moment deviation from the magnetic ordering axis. The results of the analysis of the μ SR spectra indicate the formation of the spin glass state in the $\text{La}_{1,85}\text{Sr}_{0,15}\text{CuO}_4$ sample. In contrast to the behaviour of the $\text{HoBa}_2\text{Cu}_3\text{O}_{7-y}$ ceramic the magnetic ordering of the Er-atoms in the $\text{ErBa}_2\text{Cu}_3\text{O}_{7-y}$ sample is not observed in the comparable temperature interval. Our data has also allowed the magnetic field penetration depth $\lambda_1(0)$ to be determined: $3200 \pm 70 \text{ \AA}$ for $\text{La}_{1,9}\text{Sr}_{0,1}\text{CuO}_4$, $2420 \pm 60 \text{ \AA}$ for $\text{La}_{1,85}\text{Sr}_{0,15}\text{CuO}_4$, $2000 \pm 50 \text{ \AA}$ for Bi-Sr-Ca-Cu-O (phase 2212), $1600 \pm 50 \text{ \AA}$ for $\text{ErBa}_2\text{Cu}_3\text{O}_{7-y}$.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

Preprint of the Joint Institute for Nuclear Research, Dubna 1989

13 коп.

Редактор Э.В.Ивашкевич. Макет Р.Д.Фоминой.

Подписано в печать 12.07.89.

Формат 60x90/16. Офсетная печать. Уч.-изд.листов 0,85.

Тираж 200. Заказ 42317.

**Издательский отдел Объединенного института ядерных исследований.
Дубна Московской области.**