Using Cu NQR in Eu-doped La$_{2-x}$Sr$_x$CuO$_4$ we find the evidence of the pinned stripe phase at 1.3K for $0.08 \leq x \leq 0.18$. The pinned fraction increases by one order of magnitude near hole doping $x = 1/8$. The NQR lineshape reveals three inequivalent Cu positions: i) sites in the charged stripe; ii) nonmagnetic sites outside the stripes; iii) sites with a magnetic moment of 0.29$\mu_B$ in the AF correlated regions. A dramatic change of the NQR signal for $x > 0.18$ correlating with the onset of bulk superconductivity corresponds to the depinning of the stripe phase.

PACS 74.25.Nf, 74.72.Dn
The recognition is growing that doping of the antiferromagnetic (AF) insulating phase of a high-$T_c$ superconductor by holes has an explicit topological character. In fact, according to the time reversal symmetry the segregation of charges to periodical domain walls (stripes) requires an antiphase arrangement of the created AF domains [1–3]. The first evidence for such stripe phase has been provided by neutron studies of the low temperature tetragonal (LTT) phase of Nd-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ [4]. A number of recent papers confirmed the presence of stripe correlations in the other cuprates as well [5–7]. But in spite of the hot interest to the problem surprisingly little is known about the local properties of the stripe structure.

In this Letter we report the results of the direct study of the stripe phase local structure by means of Cu NQR. The application of NMR and NQR for study of stripes meets serious difficulties due to the slowing of the charge fluctuations down to MHz frequency range which wipes out a large part of the nuclei from the resonance. An important breakthrough [8] based on the quantitative analysis of the fraction of the nuclei wiped out from the NQR brought insight in the behaviour of the stripe phase order parameter both in cuprates [8] and nickelates [9].

Unfortunately, based on wipeout effects alone, it is impossible to determine the local structure of the stripes, i.e. the charge and internal magnetic field distribution, and the values of the typical local parameters. Such information can be obtained only from the NQR analysis of the stripe phase itself, which is possible after reappearance of the signal in the slow fluctuation limit. The pinning of stripes at low temperatures enables us to take the advantages of the extreme sensitivity of Cu NQR to the local charge and magnetic field distribution. In addition to the measurements at temperatures of 1.3 K, this program could be realized easier for the LTT structure, which is helpful for pinning of the stripe structure. This structure was induced by doping with non-magnetic Eu rare-earth ions instead of magnetic Nd ones (the ordering of Nd moments causes fast Cu nuclear relaxation hindering the observation of Cu NQR). We expect that in the stripe structure the different Cu sites will be inequivalent with respect to the NQR, providing information on the local
properties at given points of the structure.

For our experiments we have chosen fine powders of the series La$_{2-x-y}$Eu$_y$Sr$_x$CuO$_4$ with variable Sr content $x$ and fixed Eu content $y = 0.17$. The preparation of single phase samples was described in [10]. It was found [10] that for such Eu content the LTT phase is realized for $x > 0.07$. For Sr concentrations $x > 0.12$ the ac-susceptibility and microwave absorption measurements reveals the presence of superconductivity with $T_c = 6; 9; 14; 19; 18; 16; 13K$ for resp. $x = 0.12; 0.13; 0.15; 0.18; 0.20; 0.22; 0.24$. The superconducting fraction is small for $x \leq 0.18$ and starting from $x > 0.18$ a transition to bulk superconductivity take place. The NQR measurements are performed with the standard spectrometer in the range 20 - 100 MHz. By lowering the temperature down to 1.3K we succeed to observe the Cu-NQR spectra at all Sr concentrations. Regarding their NQR properties the samples should be separated into two groups.

The first one corresponds to Sr concentrations $x \leq 0.18$. The superconducting fraction of these samples, if any, was rather small. Each of the spectra, which are very similar for $0.08 \leq x \leq 0.18$, consists of a broad line in the region from 20 MHz up to 80 MHz with an unresolved peak between 30 and 40 MHz (examples of some spectra are shown in Fig. 1). The main distinctions of the spectra for different $x$ are i) the integral intensity of the spectra, which is peaked near $x = 0.12$ (Fig. 2a), and ii) the temperatures below which it is possible to observe them (for $x = 0.12; 0.13$ they are observable even for the temperatures higher than 4.2K).

The second group of samples with $x > 0.18$ showing bulk superconductivity possesses completely different and much narrower NQR spectra (Inset to Fig.1), which can also be observed at much higher temperatures. The intensity grows with increasing $x$ from 0.18 (Fig. 2b).

Beginning the discussion with the $x \leq 0.18$ group, we first consider the above mentioned complicated peak in the lineshapes. Since the natural abundance ratio of $^{63}$Cu and $^{65}$Cu is 2.235 and the ratio of their quadrupole moments is 1.081 it is clear that this peak contains more than one pair of $^{63}$Cu and $^{65}$Cu signals. The possibility that such a picture arises from
one site due to the splitting of the signal by the hyperfine field can be ruled out by the
different behaviour of the relative intensities of both components upon variation of $x$ and by
their different echo decay times. The gaussian fit to these peaks reveals the existence of two
independent copper sites 1 and 2 (we use this notation in order to distinguish them from
the sites A and B known for the superconductors in the low temperature orthogonal (LTO)
phase $[11]$), having different NQR frequencies (Fig. 3).

To make the site assignment, we note that the NQR frequency is sensitive to the the
local hole concentration changing between 0.5 and 0 hole per Cu atom $[4]$. In a linear
approximation we obtain that for the given $x$ the resonant frequency $\nu_Q$ is connected with
the local hole density $n(r)$ via the relation $\nu_Q(x, n) = \nu_Q^0 - \alpha x + \beta n$ with the empirical
constants $\alpha$ and $\beta$. The first term here is the NQR frequency for the compound with zero
Sr content, the second one is due to the negative shift caused by the contraction of Cu-O
bond length induced by the internal pressure appearing upon substitution of La with Sr,
the third corresponds to the positive shift due to the local increase of the effective fractional
charge on Cu. This expression agrees both with the calculations in the frames of the ionic
$[12]$ as well as of the cluster $[13]$ models (in the uniform case $n = x$).

It follows from our results (Fig.3) that the resonance frequencies $63\nu_Q^{(1)}(x)$ for line 1
are shifted to lower values from the reference value $63\nu_Q(0, 0) = 63\nu_Q^0$ (we use here $63\nu_Q^0 =
31.9$ MHz estimated for $La_2CuO_4$ $[14]$). This indicates that the positive contribution to
$63\nu_Q(x, n)$ is small and that the effective fractional charge on sites 1 is near zero. It means
that they are located in the regions free of doped holes. In contrast line 2 is due to the
sites which in addition to the negative shift exhibit a positive one. It means that these sites
belong to the regions with an increased average charge (hole density) on the Cu ions.

The high frequency part of the spectrum can be analyzed by subtraction of the 1 and
2 contributions from the entire signal. The resulting spectra are shown in Fig.1. The
frequencies corresponding to their maxima are plotted in Fig.3. We assume that this line
corresponds to the broadened $(\pm1/2) \leftrightarrow (\mp1/2)$ transitions of nuclei located in sites 3 expe-
riencing an internal magnetic field (note the broad high-frequency shoulder). The satellites
are unresolved due to inhomogeneities of the internal magnetic field and of the NQR frequencies. If the orientation of the internal field with respect to the electric field gradient is identical to that observed for \( \text{La}_2\text{CuO}_4 \) \[14\] the frequency of this transition enables us to estimate the quadrupole shift and to determine the Larmor frequency for this Cu site to be 45.2 MHz for \( x = 0.12 \). It corresponds to an internal field of 40.1 kOe. Using the value of the hyperfine constant \( |Q| = 139 \pm 10 \text{kOe/}\mu_B \) \[15\] we estimate that in order to create such a field the effective magnetic moment of Cu at site 3 has to be equal to 0.29 \( \pm 0.02 \mu_B \), coinciding with the value obtained from neutron and muon experiments \[5,16\].

Since quantitatively similar spectra were observed for each compound of the first group we believe that they contain the same elementary "bricks" of the phase under study. Discussing the relative weight of the different contributions, we note that the echo decay can be described in terms of stretched exponents \( \exp[-(2t/T_2)^a] \) with different \( T_2 \) for each site. For \( x = 0.12 \) the numerical fit of the measured echo decay at the frequencies of different sites gives the same \( a \simeq 0.5 \) and \( T_2^{(1)} = 11 \mu\text{sec}; \ T_2^{(2)} = 8.8 \mu\text{sec}; \ T_2^{(3)} = 5.5 \mu\text{sec} \). Such a relaxation law is typical for the relaxation via randomly distributed magnetic moments \[17\] whereas the values of the relaxation rates depend on the location of these moments with respect to different Cu sites. Extrapolating the corresponding signal intensities to \( t = 0 \) we find the contributions of sites 1, 2, 3 to be given by the ratio (1:6:13).

As for the origin of the sites 1 it is possible to conclude that on one hand they do not belong to the AF domains, and on the other hand they are outside of stripes since their effective charge is equal to zero. We assume that they correspond to defects terminating the stripes. From their relative number we estimate the average length of the stripe to equal at least 6 lattice constants.

It is important that the NQR frequencies for the site 2 (See Fig.4) are almost the same for any \( x \) thus indicating that for all Sr concentrations the stripes are equally charged. The effective charge in a stripe is near 0.18-0.19. This is larger than the average hole concentration (x) but less than 0.5 expected for the ideal stripe picture \[4\]. It means that the charge is distributed over the domain wall of a finite thickness. Together with the
above-mentioned intensity ratio this indicates that the real stripe picture differs from the
ideal one. Another sign for this is the broadening of line 2 due to a distribution of NQR
frequencies. Its linewidth (Fig. 3) reflects the behaviour of the pinning: at $x = 0.12$ where
pinning is stronger, the narrowing due to the motion of stripes is weaker and the linewidth
is larger. The decrease of the internal magnetic field with the deviation $x$ from 0.12 reflects
the suppression of magnetic order by holes penetrating into AF domains.

The changes in intensity of the NQR spectra are due to variation of the number of
"bricks" for the compounds with different Sr content, which depends on the pinning strength.
Our results indicate, that the stripe phase is pinned at least for the time scales shorter than
$10^{-6}$ sec (the same conclusion was reached also by La NQR [18]).

The pinning for $0.08 \leq x \leq 1.8$ is due to the buckling of the CuO$_2$ plane. It is connected
with the CuO$_6$ octaedra tilts around the [100] and [010] axis by the angle $\Phi$, which for given
Eu substitution is governed by the Sr content. It follows from Fig. 2a, that the quantity of
the pinned phase, which is proportional to the NQR signal intensity, is peaked at $x = 0.12$.
This indicates additional strong pinning due to the commensurability effect. Such pinning
is not unique for the LTT phase (as buckling is). It is a manifestation of the plane character
of the inhomogeneities of the charge and spin distributions. Together with the existence
of three different Cu sites this gives an independent justification of the conventional stripe
picture [1], where the charges are uniformly distributed in rivers of holes across one Cu-
chain separated by the bare three leg ladders (we do not discuss here the possibility of two
magnetic sites which may be deduced from the wide distribution of the internal field seen
in Fig. 1).

Upon increasing $x$ over $x = 0.18$ the tilt angle is decreasing below the critical value
$\Phi_c \approx 3.6^\circ$ [19] and depinning of the stripe phase takes place. Such behaviour occurs for
the compounds with Sr concentrations $x > 0.18$ belonging to the second group for which
the broad signals, typical for the pinned phase, disappear. The corresponding NQR spectra
gradually transform to the narrow signal at higher frequencies, which for $x = 0.24$ is shown
in the inset to Fig. 1. The intensity of this line (proportional to the quantity of the unpinned
stripe phase) is shown in Fig. 2b.

The analysis of this relatively narrow signal reveals only two different sites with $^{63}$Cu-NQR frequencies of 37.60 MHz and 39.82 MHz. Within 1% accuracy these frequencies coincide with those known at the same $x$ for the $A$ and $B$ sites in the LTO superconducting phase [20] confirming that the LTT structure differs only in the directions of CuO$_6$ octahedra tilts. The satellite $B$ is due to Cu having a localized hole in the nearest surrounding since, according to [13,21], its NQR frequency has the additional positive shift $\delta \nu_Q \simeq 2.5$ MHz. The observed transformation of the NQR spectra (in comparison with those for $0.08 \leq x \leq 0.18$) is due to the fast transverse motion of stripes in the depinned phase. As a result the internal magnetic field on Cu nuclei is averaged out, and the effective fractional charge is homogeneously distributed over all Cu nuclei giving the usual NQR frequencies. Such depinning leads to the drastic changes in the magnetic properties. The echo signals decay for samples with $x > 0.18$ becomes purely exponential ($T_2^{(2)} = 35.4 \mu$sec for sample with $x = 0.24$).

An important feature of the compounds with $0.08 \leq x \leq 0.18$ is the possibility to observe the NQR line in the state without bulk superconductivity. Usually for La$_{2-x}$Sr$_x$CuO$_4$ compounds for moderate doping within the so-called spin glass phase between $x \simeq 0.02$ and $x \simeq 0.06$ (the bulk superconductivity threshold) the fast relaxation via the localized moments [17] hinders the observation of the Cu NQR. In our case the Cu NQR of compounds moderately doped with Sr is observable even in the absence of bulk superconductivity. It is an indication that we are dealing with the unusual correlated state where the magnetic moments created upon Sr doping are not effective in relaxation. Note, that at 1.3K for $x = 0.12$ compound the entire stripe phase is pinned. This follows from the comparison of the number of Cu nuclei, responsible for the NQR, with that for $x = 0.24$ compound (Inset to Fig.1), which is due to 100% of the Cu nuclei. Both these quantities were obtained by extrapolation the signals to $t = 0$ and calculation of the integrated intensities.

It is also possible to make some remarks about the superconducting properties. The main is that the depinning point separates two different types of superconductivity. For $x \leq 0.18$
we are dealing with a weak Meissner effect, an increased London penetration length and with \( T_c \) increasing with \( x \) growing up to 0.18. Combining these facts with the absence of a narrow signal typical for the bulk superconducting phase, indicating that the impure LTO phase is absent, and with the suppression of the relaxation via magnetic moments of doped holes, one has arguments in favor of possible one-dimensional superconductivity along the charged rivers of stripes - the issue which is widely discussed now \[22\]. For \( x > 0.18 \) we have bulk superconductivity with conventional London length, typical NQR signal and decreasing \( T_c(x) \). Such crossover may be caused by the transverse motion of the stripes carrying superconducting currents which gives rise to the conventional superconductivity in CuO\(_2\) planes. Although possibly a simple coincidence, it happens when the doping \( x \) is equal to the effective charge \((n)\) in a stripe.

In conclusion we carried out Cu NQR studies of the Eu doped La\(_{2-x}\)Sr\(_x\)CuO\(_4\). We demonstrated that at 1.3K the ground state for moderate Sr content corresponds to the pinned stripe-phase and that the pinning is enhanced at the commensurability. Three nonequivalent copper positions in the CuO\(_2\) planes were found. One of them with a magnetic moment of 0.29\(\mu_B\) is related to the AF correlated antiphase domains. From the behaviour of the NQR frequencies it follows that the effective charge of the domain walls separating these domains is almost independent on the Sr content \( x \). The onset of the bulk superconductivity at larger \( x \) correlates with the dramatic transformation of the NQR spectra, indicating the depinning of the stripe phase.

The authors are grateful to H.Brom, A.Egorov and N.Garifyanov for valuable help and discussions. This research was supported by the Deutsche Forschungsgemeinschaft. The work of G.T. was supported in part by the State HTSC Program of the Russian Ministry of Sciences (Grant No. 98001) and by the Russian Foundation for Basic Research (Grant No. 98-02-16582).
REFERENCES

[1] V.J. Emery and S.A. Kivelson, Physica 209C, 597 (1993); J.Phys. Chem. Solids, Vol. 59, 1705 (1998)

[2] D. Poilblanc and T.M. Rice, Phys. Rev., B 39, 9749 (1989); J. Zaanen and O. Gunnarson, Phys. Rev., B 40, 7391 (1989); J. Zaanen, J. Phys. Chem. Solids, 59, 1769 (1998)

[3] A.H. Castro Neto and D. Hone, Phys. Rev. Lett. 76, 2165 (1996)

[4] J.M. Tranquada et al., Nature 375, 561, (1995)

[5] J.M. Tranquada, J. Phys. Chem. Solids, 59, 2150 (1998)

[6] H.A. Mook et al., Nature 395, 580, (1998)

[7] H.P. Fong et al., cond-mat/9902262

[8] A.W. Hunt et al., Phys. Rev. Lett. 82, 4300 (1999)

[9] I.M. Abu-Shikah et al., cond-mat/9906310

[10] B. Büchner et al., Physica 185 – 189C, 903 (1991); Europhys. Lett., 21, 953 (1993)

[11] K. Yoshimura et al., J. Phys. Soc. Jpn., 58, 3057 (1989)

[12] T. Shimizu, J. Phys. Soc. Jpn., 62, 772 (1993)

[13] R.L. Martin, Phys. Rev. Lett., 75, 744 (1995)

[14] T. Tsuda et al., J. Phys. Soc. Jpn., 57, 2908 (1988)

[15] T. Imai et al., Phys. Rev. Lett., 70, 1002 (1993)

[16] G.M. Luke et al., Hyp. Int., 105, 113 (1997)

[17] M.R. McHenry, B.G. Silbernagel and J.H. Wernick, Phys. Rev. B 5, 2958, (1972)

[18] G.B. Teitelbaum et al., JETP Letters, 67, 363 (1998)
[19] B. Büchner et al., Phys. Rev. Lett., 73, 1841 (1994)

[20] K. Yoshimura et al., Hyp. Int., 79, 867 (1993); S. Oshugi et al., J. Phys. Soc. Jpn., 63, 2057 (1994)

[21] P. C. Hammel et al., Phys. Rev. B 57, R712 (1998)

[22] J. M. Tranquada et al., Phys. Rev. Lett., 78, 338 (1997)
FIGURES

FIG. 1. Representative Cu-NQR lineshapes at 1.3K of La$_{2-x-y}$Eu$_y$Sr$_x$CuO$_4$ with $y = 0.17$. The value of $x$ is shown for each line. All lineshapes include standard frequency corrections of $1/\nu^2$ and are normalized to equal heights. The raw data points are shown together with their fits and the decomposition to different contributions is also shown. The continuous line corresponds to the contribution of sites 1 and 2. Filled circles show the contribution of antiferromagnetic site 3. Inset: A typical signal for $x > 0.18$ decomposed into two contributions (T=4.2K).

FIG. 2. The integrated intensity (normalized to maximal values) of the Cu NQR signals for La$_{2-x-y}$Eu$_y$Sr$_x$CuO$_4$: for $0.08 \leq x \leq 0.18$ at 1.3K (a); for $x \geq 0.18$) at 4.2K (b).

FIG. 3. The different contributions to the Cu NQR signal for the pinned stripe phase in La$_{2-x-y}$Eu$_y$Sr$_x$CuO$_4$ as function of Sr content $x$ (T=1.3K): $^{63}$Cu NQR frequencies of sites 1 - open circles and 2 - filled circles; the corresponding half widths at half maximum (HWHM) - open and filled triangles resp.; the frequencies corresponding to the maxima of the magnetic contribution 3 are shown by squares.
Int. intensity (a.u.)

Sr content (x)

(a)  

(b)
