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STRIPE MOTION IN CuO, PLANES OF $Y_{1-x}Pr_xBa_2Cu_3O_7$ AS SEEN FROM THE Cu(2) QUADRUPOLE RESONANCE

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Pulsed NQR at the frequencies of 28-33 MHz has been used to study copper NQR spectra in $YBa_2Cu_3O_7$, $TmBa_2Cu_3O_7$ and $Y_{0.9}Pr_{0.1}Ba_2Cu_3O_7$ compounds at temperatures of 4.2-200 K. Quantitative analysis of the spectra has shown that the "plane" Cu(2) spectra shape is well described by using a model of 1D correlations of charge and spin distribution in CuO_2 planes (stripe correlations). In the undoped superconductors the charge-spin stripe structure moves fast in the CuO_2 planes, but doping the $YBa_2Cu_3O_7$ lattice with praseodymium slows this motion down.

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The hypothesis of 1D ordering of charges and spins in a particular configuration of stripes in CuO_2 planes of $RBa_2Cu_3O_7$ superconductors has been suggested two years ago [1] and was used later to explain NMR and NQR data for $TmBa_2Cu_3O_{6+x}$ and $TmBa_2Cu_4O_8$ [2], but up to now it has not been corroborated directly in the shape of Cu(2) NQR spectra. In this paper we present such corroboration based on the analysis of the Cu(2) NQR spectra for $Y_{1-x}Pr_xBa_2Cu_3O_7$ ($x = 0, 0.1$) compounds. The $YBa_2Cu_3O_7$ (YBCO) and $Y_{0.9}Pr_{0.1}Ba_2Cu_3O_7$ (YPBCO) samples prepared by solid-state reaction method [3] were kindly placed at our disposal by Xu and Luetgemeier (Forschungszentrum Juelich, Germany). The critical temperatures found from susceptibility measurements at the frequency of 1 kHz appeared to be T_c (onset)=92.5 and 86 K, respectively. For comparison, besides YBCO and YPBCO, overdoped $TmBa_2Cu_3O_7$ (TmBCO) compound with the critical temperature of 91.5 K has been studied [2]. Home-built spin-echo coherent pulsed spectrometer was used for copper NQR spectra measurements.

The examples of YBCO and YBPCO spectra are shown in Fig.1. It is seen (Fig.1*a*, *c*) that both spectra, except the relatively narrow ^{63}Cu(2) and ^{65}Cu(2) NQR lines, have a broad "pedestal". Representing these spectra in a logarithmic scale (Fig.1*b*, *d*) one can clearly see the asymmetry of the narrow lines, i.e., complex composition of these lines also, in the sample doped with praseodymium.

We managed to get the best fit of the YBCO spectra to six Gaussian curves, three for each isotope: the narrow Gaussian line A', the broad one A'' and the pedestal P. The frequencies ν_i and mean square linewidths $\Delta\nu_i$ of each pair of isotope lines were supposed to be related by the ratio of nuclear quadrupole moments $\alpha = {}^{65}Q/{}^{63}Q$ ($^{65}\nu_i = \alpha\,{}^{63}\nu_i$, $^{65}\Delta\nu_i = \alpha\,{}^{63}\Delta\nu_i$, $i = A', A'', P$). Thus the fitting function involved 10 parameters: three line frequencies $^{63}\nu_i$, three linewidths $^{63}\Delta\nu_i$, three line intensities $^{63}a_i$ and the ratio $b = {}^{65}a_i/{}^{63}a_i$. While fitting the YBCO spectra taken at temperatures 200-4.2 K it was found that the pedestal linewidth (1.5 ± 0.1 MHz) did not depend on temperature, so its value was taken fixed at 1.5 MHz during the final fitting procedure (the results of which is shown in Fig.1*a*

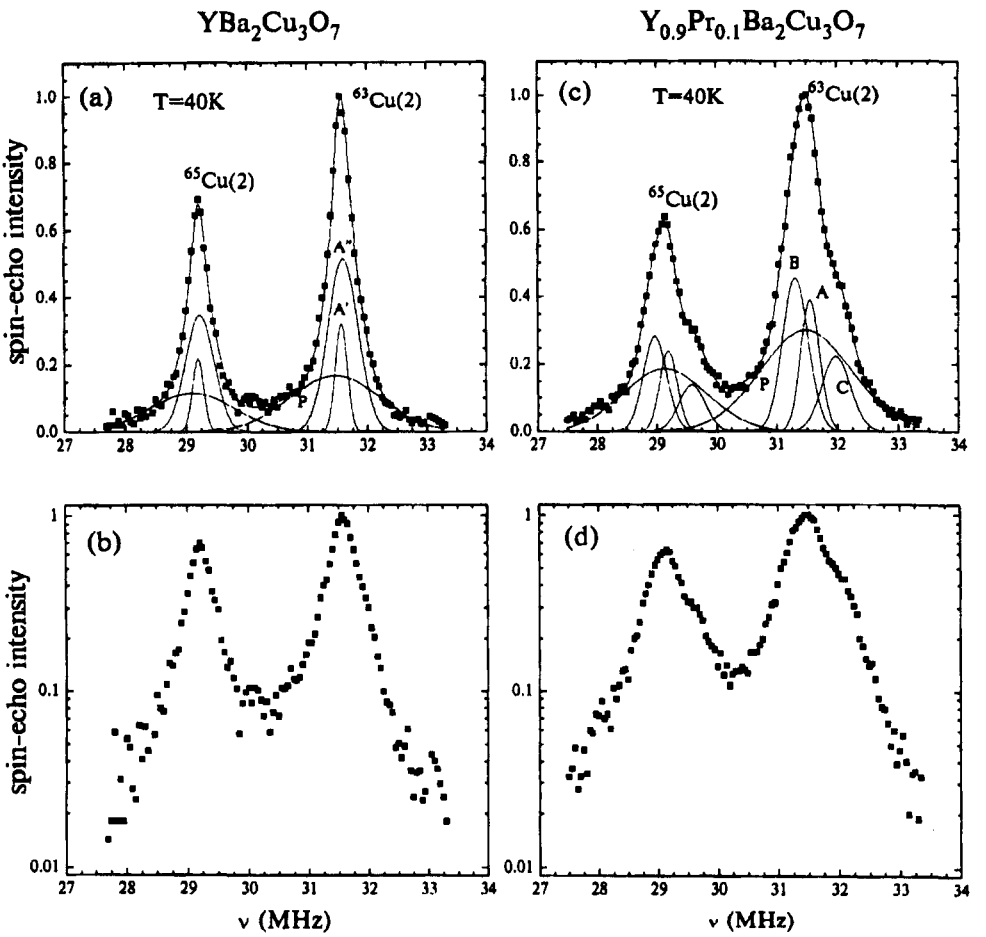

Fig.1. Copper NQR spectra for $YBa_2Cu_3O_7$ (a,b) and $Y_{0.9}Pr_{0.1}$ $Ba_2Cu_3O_7$ (c,d) taken at T=40K; solid lines - best fit (see text and Fig.2)

and Fig.2a,b,c) thus reducing the total number of fitting parameters to 9. It should be mentioned here that the presence of pedestal is typical for copper NQR spectra in 1-2-3-7 compounds (but not in 1-2-4-8 ones [2]). We observed it in the spectra of Tm1-2-3-7 and Y1-2-3-7 samples prepared in different laboratories [4]; in some of them the pedestal looked like several poorly resolved overlapping lines and, in particular, contained the broad NQR line of two-fold coordinated "chain" $^{63}Cu(1)_2$ copper at the frequency of 30.1MHz. We suppose the spectrum P to arise from the Cu(1) and Cu(2) centers located in the areas with partly disordered oxygen sublattice of CuO planes and, respectively, with reduced (and locally inhomogeneous) hole concentration in CuO_2 planes. Extracting the sum $(A'+A'')$ from the observed spectrum we obtain the spectrum of "good" superconductor, free (or almost free) of crystal structure defects. Actually this procedure allowed us to obtain (for the first time, to our knowledge) a quantitative description of Cu(2) NQR line shape for the superconductor with the 1-2-3-7 orthorhombic structure.

A comparative analysis of spectra for YBCO (Fig.1*a* and 2*a*, *b*, *c*) and TmBCO [4] has shown that the lineshape is intermediate between Gaussian and Lorentzian and for both samples the same relation holds between the parameters describing the lines A' and A''. In particular, in the temperature range of 100-150 K we have obtained $\nu_{A'} = \nu_{A''}$, $\Delta\nu_{A''}/\Delta\nu_{A'} = 2.4(1)$ and $a_{A''}/a_{A'} = 1.64(5)$, although the linewidths $\Delta\nu_i$ for TmBCO has appeared to be 25-30% larger than those for YBCO.

In order to describe the copper NQR spectra shape for YPBCO sample we used the fitting function containing four Gaussians for each isotope: besides the pedestal (its linewidth $^{63}\Delta\nu_P$ was again accepted to be equal to 1.5 MHz), the central line A (since the frequencies $\nu_{A'}$ and $\nu_{A''}$ are close in YBCO, we considered it possible to restrict the description of this line in YPBCO to one line) and the satellite C at the right slope of the spectrum, we have introduced the line B located at the frequency $\nu_B < \nu_A$ in the spectrum (the presence of this line is revealed by the bulging left slope of the ^{63}Cu line in Fig.1*d*). Thus the fitting function contained 12 parameters: four frequencies $^{63}\nu_i$, three linewidths $^{63}\Delta\nu_i$, four intensities $^{63}a_i$ and the intensity ratio $b = ^{65}a_i/^{63}a_i$. The results of fitting of YBCO spectra taken at 200-4.2 K are given in Fig.2*d*, *e*, *f*. Let us point out and discuss the main peculiarities of copper NQR spectra for YBCO, TmBCO and YPBCO samples.

1. The integral intensity of the pedestal P is the same for YBCO and TmBCO and equals approximately to 1/3, but for YPBCO it increases up to $\sim 1/2$; this confirms our assumption that the component P of the spectra belongs to partly disordered phase of the compounds studied.

Other remarks concern with "pure" spectra of Cu(2) NQR characterised by the components A', A'' (YBCO, TmBCO) and A, B, C (YPBCO).

2. In the spectra of YBCO (Fig.2*a*, *b*, *c*) and TmBCO [4] at temperatures 100-200 K the frequencies $\nu_{A'}$ and $\nu_{A''}$ are approximately equal to each other, and $\nu_{A'} < \nu_{A''}$ at $T < T_c$. In general the difference of these frequencies is small ($< 0.2\%$), so that for rough estimates it is possible to assume that the sum $(A' + A'')$ describes a single Cu(2) NQR line in YBCO and TmBCO samples.

3. The temperature dependence of the frequency of this line in the YBCO spectrum coincides with the temperature dependence of the line A in the spectrum of YPBCO sample (cf. Fig.2*a* and 2*d*). This allows us to ascribe the line A (Fig.2*d*) to the Cu(2) nuclei located far away from Pr atoms and not influenced by them. Let us call these nuclei as "remote".

4. The main result of this paper is represented by the temperature dependences of the parameters of the lines A, B, C for YPBCO samples (Fig.2*d*, *e*, *f*).

We ascribe the lines B and C which are absent in the YBCO spectrum (Fig.2*a*) to the "neighboring" nuclei, i.e., to those located not far from Pr atoms. Comparing the integral intensities a_i of the lines at temperatures above T_c (when the spectra are free of possible distortions due to different penetration depth of the RF field H_1 into areas with different local concentration of holes), we obtain the ratio of the mean intensities $< a_B > : < a_C > = 2 : 1$, just the same as that following from the model of quasi-1D ordering of charges and spins in the CuO_2 planes (stripe model) [1]. Furthermore, we have noticed that the lines B and C are located asymmetrically with respect to the line A, but in such a way that $(\nu_B a_B + \nu_C a_C)/(a_B + a_C) = \nu_A$ (open squares in Fig.2*d*). These two facts immediately suggest the idea that both spectra, that for "remote" nuclei (A) and "neighboring" ones (B+C), belong to the stripes moving in the CuO_2 planes, the

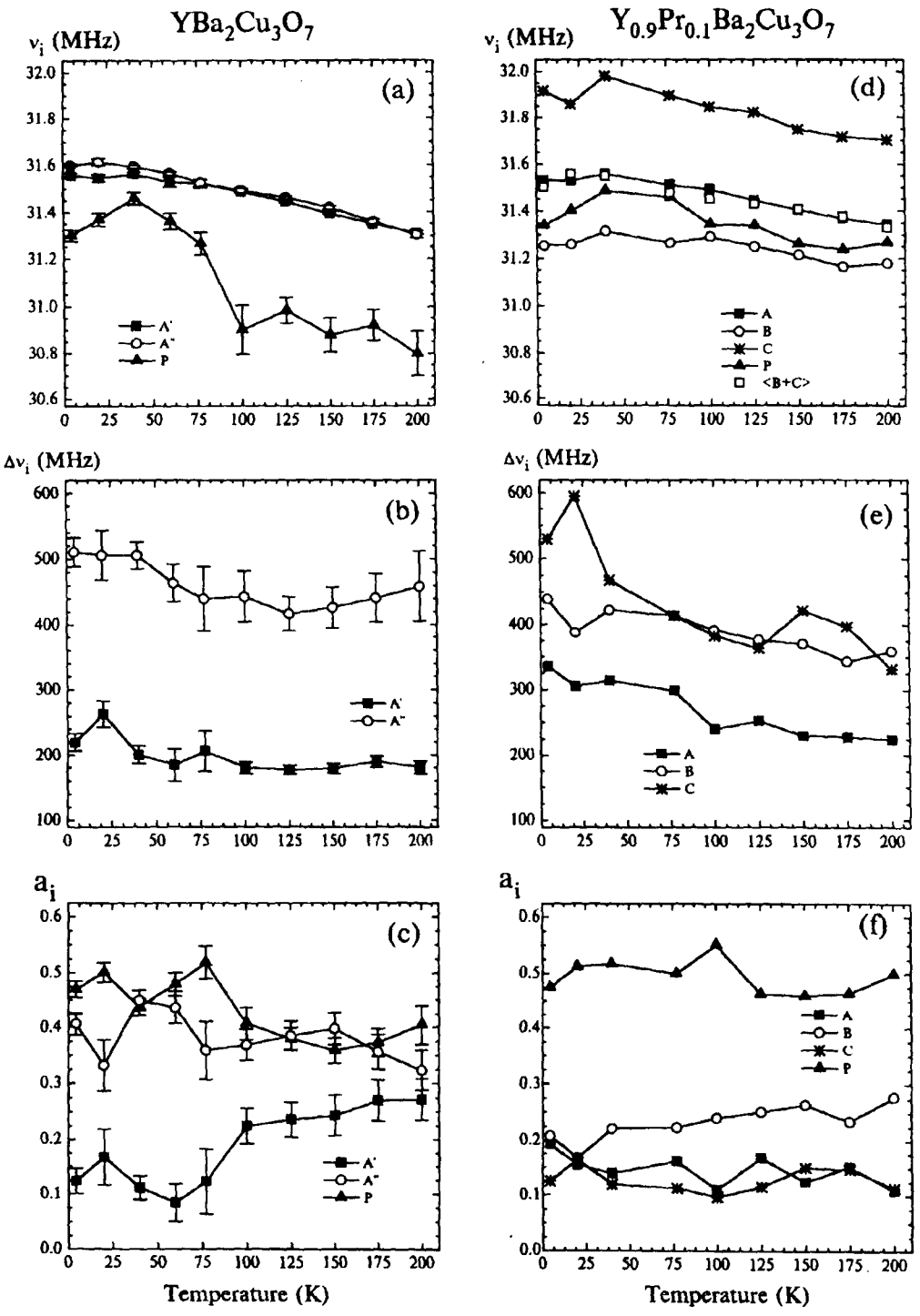

Fig.2. Temperature dependence of parameters of copper NQR spectra for $YBa_2Cu_3O_7$ (*a*, *b c*) and $Y_{0.9}Pr_{0.1}Ba_2Cu_3O_7$ (*d*, *e*, *f*); for details see text

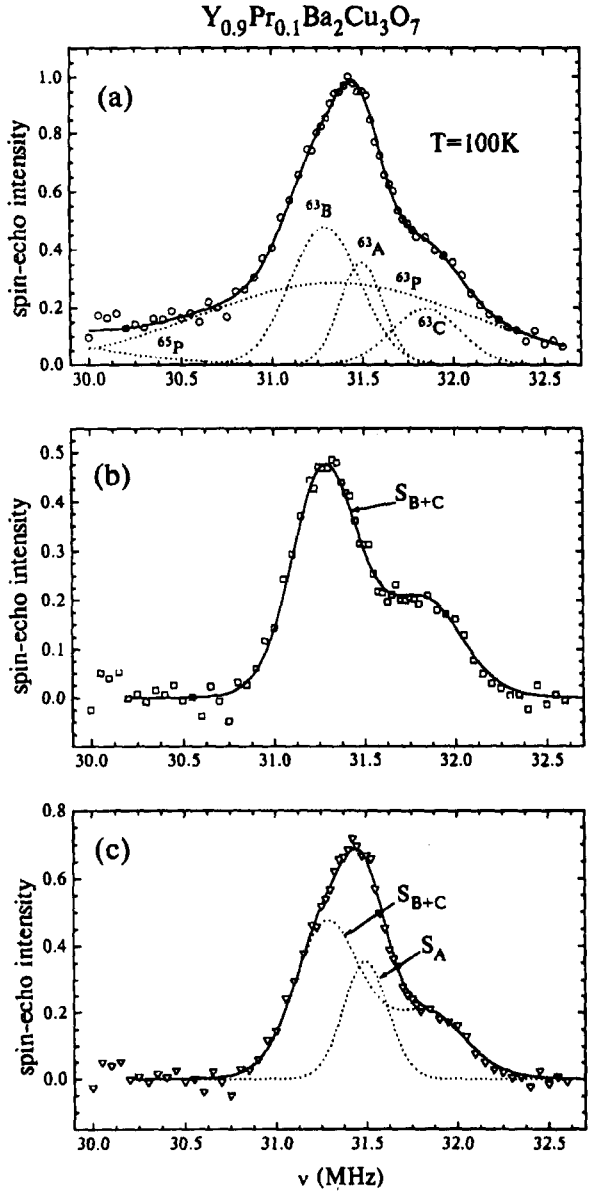

Fig.3. Fragments of ^{63}Cu(2) NQR spectra for $Y_{0.9}Pr_{0.1}Ba_2Cu_3O_7$ at $T = 100$K: (a) - best fit to the sum of five Gaussian curves $^{63}{}^{63}(B+C)$ contribution according to Eqs.(1),(2) with parameters $\Delta_B/2\pi = -217$ kHz, $\Delta_C/2\pi = 434$ kHz, $\tau_C = 2.9$ μs, $\tau_B = 2\tau_C$, $\omega_0/2\pi = 31.465$ MHz, $\sigma/2\pi = 159$ kHz; (c) - the same as (b) with addition of ^{63}A contribution with parameters $\Delta_B/2\pi = -217$ kHz, $\Delta_C/2\pi = 434$ kHz, $\tau_C < 10^{-7}$ s, $\tau_B = 2\tau_C$, $/2\pi = 119$kHz

only difference between them being the different rate of motion: for "remote" nuclei the case of fast motion is realised while for "neighboring" - that of slow motion. Two types of Cu(2) centers are distinguished in the model [1] - those located in the center of the stripe (type C, the hole density on the oxygen ligands is high) and at the stripe boundaries (type B, the hole density is low), so that at the optimal doping of the CuO_2 planes by holes and at the dense packing of the stripes the amount of centers B is twice as large as that of centers C. The shape of Cu(2) NQR spectrum in the system of moving stripes can be described

by the following function [5]:

$$I(\omega,\Omega) = \frac{W_B W_C(\omega_B-\omega_C)^2(\tau_B+\tau_C)\tau_B\tau_C}{[\tau_B\tau_C(\omega-\omega_B)(\omega-\omega_C)]^2+[\tau_B(\omega-\omega_B)+\tau_C(\omega-\omega_C)]^2}, \quad (1)$$

where W_i is the probability to find the nucleus in the i-th state, τ_i and $\omega_i = \Omega+\Delta_i$ are the lifetime of this state and the corresponding NQR frequency, respectively. Actually the frequency Ω is randomly distributed near the mean value $<\Omega>=\omega_0$ (the quadrupole broadening due to the lattice imperfections), thus the spectrum shape is obtained by averaging Eq.(1) with the Gaussian distribution of Ω:

$$S(\omega) \sim \int I(\omega,\Omega)\exp\left[-(\Omega-\omega_0)^2/2\sigma^2\right]d\Omega. \quad (2)$$

The ^{63}Cu(2) NQR spectrum taken at 100 K is shown by circles in Fig.3*a*, the squares in Fig.3*b* depict the spectrum $^{63}(B+C)$ for ^{63}Cu isotope obtained by subtraction of three lines (^{63}A, ^{63}P and the P component for ^{65}Cu isotope) from the experimental spectrum (Fig.3*a*), the triangles in Fig.3c display the $^{63}(A+B+C)$ spectrum. The solid line in Fig.3*b* represents the calculated spectrum $S_{B+C}(\omega)$ obtained at the following values of parameters: $\Delta_B = -\Delta = -2\pi\cdot 217\cdot 10^3$ s^{-1}, $\Delta_C = 2\Delta$, $\tau_C = \tau = 2.9\cdot 10^{-6}$ s, $\tau_B = 2\tau$, $\omega_0 = 2\pi\cdot 31.465\cdot 10^6$ s^{-1}, $\sigma = 2\pi\cdot 159\cdot 10^3$ s^{-1}. One can see that the experimental spectrum S_{B+C} of "neighboring" nuclei and total NQR spectrum $(S_A + S_{B+C})$ are described very well in the frame of our model of moving stripes (Fig.3*b*, *c*). The spectrum of "remote" nuclei (dashed line S_A in Fig.3c) is obtained at the same Δ_B, Δ_C values but when the short lifetime τ ($< 10^{-7}$ s) is assumed; the frequency of the line A appears to be 0.09% higher (this corresponds to higher mean concentration of the holes), and the quadrupolar linewidth - 25% less than these parameters for "neighboring" nuclei. Thus far away from impurity Pr ions the motion rate of the stripes is high, the corresponding lifetime τ is very small and cannot be evaluated in the present experiment.

Two conclusions can be inferred from above: 1) the fast motion of stripes in the CuO_2 planes seems to be necessary for cuprates to superconduct; 2) Pr doping leads to the pinning of the stripes resulting in a suppression of superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_7$. The 1D correlations in charge and spin distribution remain valid in the latter case too, but they become static in character.

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