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NEUTRON POLARIZATION INVESTIGATIONS OF HIGH TEMPERATURE SUPERCONDUCTORS

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1. Introduction

The method of polarized neutron transmission through sample has been proved to be rather effective in revealing the origin of magnetic phenomena not only in magnetics but also in type II superconductors placed in an external magnetic field. Having high penetrating ability and oriented magnetic moment, the polarized neutrons are very sensitive to the microscopic distribution of the magnetic field inside a sample and, therefore, may serve as microscopic probe even in the case of a zero mean value of the macroscopic magnetic moment of a sample.

Two principal set-ups are conventionally used in staging polarized neutron transmission experiments [1]. Namely, the neutron beam polarization vector $\vec{P}$ is directed either parallel or normal to the external magnetic field direction. In the first set-up, the neutron beam depolarization is studied giving information about the value of the static fluctuations $\langle(\Delta B)^2\rangle$. In the second, the change in the Larmor precession frequency is measured, which is related to change of the mean value of the induction $\langle B\rangle$ of the magnetic field in a sample. The advantages provided by the neutron depolarization method in application to type-II superconductors were first demonstrated in [2]. This method as well as the neutron spin precession method were used in the investigation of the magnetic properties of high temperature superconductors (HTSC) [3-7]. However, they all were made at relatively low temperatures:

$T<0.5T_c$. At the same time, near T_c (at $T>0.7T_c$) HTSCs happen to exhibit numerous new properties that often find no unambigous interpretation [8]. In this respect, utilization of neutrons gives definitive advantages, which are examplified in [9].

The aim of the present paper is to summarize some representative results obtained in the polarized neutron experiments on HTSC. The paper is organized as follows. Section 2 contains the new data from the neutron depolarization measurements on two $YBa_2Cu_3O_{7-\delta}$ samples at temperatures from 78K to 125K and external magnetic fields up to 5 kOe. In Section 3 the experimental results on relaxation of a quenched magnetic flux in $YBa_2Cu_3O_{7-\delta}$ ceramics at T=78K obtained in neutron spin precession measurements are presented. The both measurements were carried out on the polarized neutron spectrometer SPN-1 on the high flux pulsed reactor IBR-2 of the Frank Laboratory of Neutron Physics, JINR, Dubna. The details of SPN-1 are given in [10,11]. This spectrometer has the advantage of analyzing the dependence of polarization on the neutron wavelength which, in turn, enables one to improve substantially the accuracy of the measurements. The paper is concluded with a Summary.

2. Neutron depolarization measurements

In depolarization experiments the neutron polarization on transmission through sample is measured [1] :

$$P(\lambda) = D(\lambda)P_0(\lambda), \quad (1)$$

where λ is the neutron wavelength, $P_0 = fP_1P_2$ the neutron beam polarization. Here P_1 and P_2 are the polarizing efficiencies of the neutron polarizer and analyzer, respectively, f is the spin-flipper efficiency, $f \approx 1$.

In turn, the neutron beam depolarization value in the sample is defined via the sample depolarization coefficient:

$$D(\lambda)=\frac{(I^+(\lambda)-I^-(\lambda))(I_0^+(\lambda)+I_0^-(\lambda))}{(I^+(\lambda)+I^-(\lambda))(I_0^+(\lambda)-I_0^-(\lambda))} \quad (2)$$

where I^- and I^+ are the neutron intensities on the detector with the spin-flipper on and off, respectively.

The investigations were carried out on two plate-like sintered samples of $YBa_2Cu_3O_{6.9}$, measuring $3.7x13.5x16mm^3$ and $4.1x19x52mm^3$, having the density $\rho=4.9g/cm^3$ and the superconducting temperature $T_c=89.2K$ (sample I) and $T_c=90.4K$ (sample II). The polarized neutron beam of the cross section $2x6mm^2$ (on sample I) and $2 \times 26\ mm^2$ (on sample II) was transmitted through the centers of the samples. The magnetic field on the sample was varied from zero to 5 kOe. The mean-field square deviation on the sample did not exceed 1%. The temperature of the sample was kept in the range from 77 to 125K with an accuracy of ±0.1 K. The measurements proceeded in three stages:

i) P_0 measured at 250K;

ii) the sample cooled down in the external magnetic field ≈20 Oe starting from the temperature of 250 K to the temperature, at which the measurements were performed;

iii) P(H) measurements in a fixed external magnetic field.

The P(H) measurements were performed at T=78.2K, 79.4K, 83.4K, 84.3K, 87.0K, 88.5K and 90.6K with the cooling rate of 4 K/min. Figure 1 examplifies the results of the depolarization measurements on sample I. The rate of the magnetic field variation was different, but no essentially different behavior has been observed. For example, Fig.1(a,c,d) illustrates the measurements with the magnetic field variation rate ca. 200 Oe/h, while Fig.1b with the rate ca. 400 Oe/h up to the field value H=1kOe, then increased up to 1kOe/h for H>1kOe.

It is clearly seen from the experimental P(H) curves, that with increasing magnetic field (forward) the polarization decreases and reaches minimum at H_1. The analogous polarization behaviour was observed in conventional superconductors [2] and and assigned to the establishment of the critical state in a bulk sample. In the case of ceramics this will include both the intergranular and intragranular space [3-6]. In the fields $H<H_1$ we observed a small decrease in polarization connected with the magnetic field penetration into the intergranular space (see [3]).

The P(H) curves obtained in a decreasing field mode (backward) behave like the forward ones but, starting from some $H_{irr}>H_1$, they go somewhat above them. This fact points to the existence of pinning at $H<H_{irr}$.

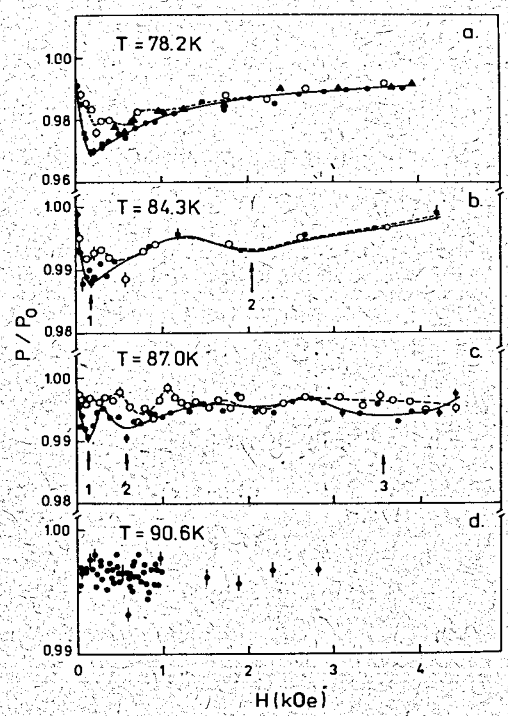

Fig.1. The experimental data on the magnetic field dependence of the depolarization P/P_0 of the sample at temperatures: a) 78.2 K, b) 84.3 K, c) 87.0 K, d) 90.6 K. o – the increasing magnetic field mode (forward); o – the decreasing magnetic field mode (backward); ∧ – the repeated forward mode after the backward curve has been measured and the field decreased down to 30 Oe. The solid curve (forward) and the dashed curve (backward) are the spline approximations of the experimental results. The arrows point to the positions of the minima at $H = H_1$ and H_2.

On some backward P(H) curves at $H = H_1 + \delta H$ a local maximum is seen, which, in accordance with the Bean model, can be related to the establishing of a more homogeneous induction distribution in the grains (see, e.g.[6]). However, at $H<H_1$ the backward curves show a tendency to P = 1, probably, due to weak pinning. The like behaviour of the forward and backward curves allows the conclusion that the pinning at these temperatures is rather weak.

Furthermore, as Fig.1a shows P(H) comes to saturation at H≅3kOe. At T = 84.3K (Fig.1b) the P(H) dependence measured in the increasing field mode has a second minimum, H_2, at 2kOe (in the figures the positions of the minima H_1 and H_2 are shown by arrows). The P(H) curves have the second minimum up to the temperature 88.5K. It is noticed that the backward curves in the vicinity of the second minimum manifest some local irreversibility (Fig.1b), that may be connected with the pinning effect selectivity with respect to the magnetic field.

The P(H) obtained in the region of the superconducting transition at T=90.6K (Fig.1d) shows the behaviour different from the polarization curves taken at lower temperatures . With increasing magnetic field the polarization remains constant with a mean value close to unity. Another peculiarity in the behaviour of the curve consists in the fact, if the measuring time is short (the initial part of the curve), the experimental data spread exceeds statistical errors and the curve itself reminds noise. The curve P(H) measured at 87.0K (Fig.1c) has a third minimum at H_3=3.8kOe (marked with the H_3

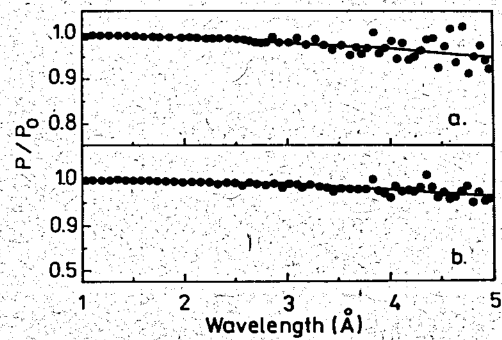

Fig.2. The neutron wavelength (λ) dependence of the neutron beam polarization at the sample temperature T = 87 K and the magnetic field for the first (a), H_1, and the second (b), H_2, minimum.

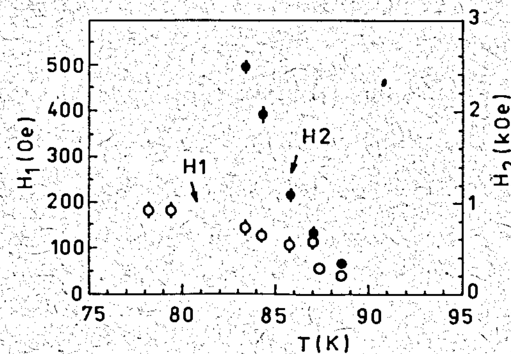

Fig.3. The positions of the minima H_1 and H_2 on the curve that represents the external magnetic field dependence of the depolarization P/P_0 as the function of the sample temperature.

arrow). The same behavior was observed at T=88.5K at H_3=1.85kOe.

Now, let us discuss in more detail the appearance of the second minimum in the behavior of P(H) near T_c. As is known, neutron depolarization periodicity in field may arise from neutron spin precession around the magnetic induction direction in a sample. The depolarization may also experience oscillations in the case of the so-called spatial resonance [12,13]. Note, that in all the above enumerated cases the presence of precession brings some periodic dependence of $P(\lambda)$ for every given value of the field H. However, the measurements of $P(\lambda)$ have revealed no periodicity (Fig.2). In this connection we assume that in our experiment the depolarization is caused by some random induction direction in some local region, as it has earlier been the case observed in conventional superconductors [2].

By assuming that the depolarization process goes in the interior of the grains with an average size of ca. 10 μm, we estimate the maximum polarization precession phase acquired on neutron flight through the local region with the induction B=1kG as

$$\Phi_{max} = \omega_L t \approx 0.1 \qquad (3)$$

where $\omega_L=\gamma B$ is the Larmor precession frequency, γ is the neutron gyromagnetic ratio, and $t=\ell/v$ is the time the neutron of the wavelength λ needs to go through the sample of length ℓ.

At $\Phi < 1$ the process of depolarization is defined by the dispersion $\langle B_\perp^2 \rangle$ of the perpendicular induction component [14,15] and the number N of the space regions along the neutron flight trajectory. In this case the spectral dependence of the depolarization has the form:

$$D(\lambda) \sim \exp\{-N\langle B_\perp^2\rangle\lambda^2\} \qquad (4)$$

The polarization in dependence on the magnetic field on the sample may behave as follows. At $H>H_1$ the pinned flux begins to flow and the polarization starts growing to its initial value. This polarization behaviour was observed in the conventional type-II superconductors [2] and in oxide superconductors at low temperatures [4,6]. At $78K<T<T_c$ the curves $H_1(T)$ and $H_2(T)$ are described by a parametrized function (Fig.3) :

$$F = F_0\left(1 - \frac{T}{T^*}\right)^m, \qquad (5)$$

Table I gives the values for the parameters T^* and m for two samples. The value obtained for T^* is near the temperature, at which the sample resistance turns into zero.

To uncover the reasons for the appearance of the second minimum on the P(H) curve one needs to clarify the character of the time dependence of polarization P(t).

Figure 4 shows this dependence taken at T = 87K and the

Table I

	H_1(sample I)	H_2(sample I)	H_2(sample II)
χ^2/n	1.21	1.17	1.2
m	0.5±0.1	3.6±1.5	2.6±0.5
T^*	89.0±0.4 K	95.2±3.9 K	89.1±1.1K

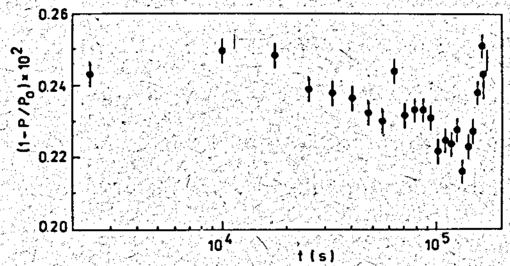

Fig.4. The time dependence of $P(t)/P_0$ taken at T=87K and H=300 Oe.

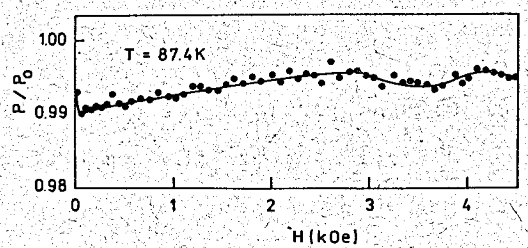

Fig.5. The experimental data on the magnetic field dependence of the depolarization P/P_0 at the sample temperature 87.4K taken under more adiabatic conditions (see the text).

field value H=300 Oe, which is the intermediate of those, corresponding to the first and second minimum. As is seen the depolarization $\delta P(t)$ decreases with time, reflecting the decrease in absolute magnetization in accord with the dependence $\delta P \sim \langle B_\perp^2 \rangle$. However, at some certain moment the depolarization starts increasing and tends to its initial value.

A possible and quite probable explanation of this behaviour of $\delta P(t)$ consists in the following. Change in t is equivalent to increase of the induction B, because of the magnetization M<0. As a consequence, the smallest change in magnetization causes change in $\delta P(t)$, which, in turn, reflects change in the P(H) behaviour. Then the conclusion follows that the depolarization growth of $\delta P(t)$ results in the appearance of the second minimum in P(H). As for the possible origin of the second minimum on the P(H) dependence, it can be related to the non-equilibrium behavior of the magnetic flux flow. This conclusion is confirmed by the fact of disapearance of this second minimum under more adiabatic (quasiequilibrium) conditions as depicted in Fig.5 showing P(H) measured after the slow cooling of the sample following the scheme: the sample cooling rate in the interval from 250 K down to 91 K was 4 K/min and in the interval 91-87.4K it was 1K/h. Up to the value H=2.4kOe the curve shows one minimum at H_1 followed by a monotonous rise in polarization. The similar results were obtained with sample II. This behavior reflects the well-known long-time character of magnetic relaxation, usually related to a rather complicated magnetic pattern in HTSC materails, which we are considering in the next Section.

3. Neutron spin precession measurements

To measure the time dependence of a captured magnetic flux, we use the method of the Larmor precession frequency of neutron spin inside a sample placed in a magnetic field [10]. If the direction of the incident polarization vector $\vec{P}_I(\lambda)$ is perpendicular to $\vec{B}$ of a superconductor, then at the sample outlet, the neutron polarization depending on the wavelength (or velocity) of the neutron is:

$$P_{\perp}(\lambda)/P_{\parallel}(\lambda) = A(\lambda)\mathrm{Cos}(\omega_{\lambda}\lambda) \quad , \qquad (6)$$

where $P_{\perp}(\lambda)$, $P_{\parallel}(\lambda)$ is the neutron polarization at the outlet of the sample corresponding to the incident neutron beam polarization perpendicular and parallel to $\vec{B}$, respectively

$$\omega_{\lambda} = (m/h) \int_{L} \omega_{L}(x)dx. \qquad (7)$$

L is the part of the neutron trajectory (including that in the sample) where the spin precession takes place.

Transition to the superconducting state leads to the change of the field B(x) inside the sample with the depth ℓ (the length of the sample along the neutron path is 18.3 mm). In this case the difference of the precession frequency measured above and below T_c is proportional to the average

Table II

H_{fc}(Oe)	$\tau^{-1}(s^{-1})$	β	χ^2/n
165	(1.00+/-0.45)10^{-5}	0.7+/-0.1	2.2
79	(2.65+/-0.15)10^{-5}	1.14+/-0.1	1.18
71	(5.68+/-0.15)10^{-5}	1.08+/-0.5	1.08
48	(4.33+/-3.45)10^{-5}	0.31+/-0.28	1.2

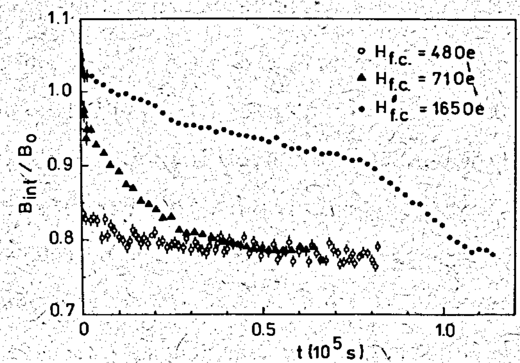

Fig.6. The time dependence of B/B_o at T=78K for the certain applied fields: 1) H_{fc}=165Oe, 2) H_{fc}=71Oe, 3) H_{fc}=48Oe, 4) H_{fc}=22Oe.

field in the superconductor. Consequently, the integral (7) changes by a value of

$$\Delta\omega_\lambda = \omega_\lambda(T) - \omega_\lambda(T>T_c) = b\ell[<B(x,T)> - B_o] \,, \qquad (8)$$

where $b = 0.04633\ G^{-1}cm^{-1}Å^{-1}$ and B(T) and B_o are related as

$$B(T) = B_o + \mu_o(1-N)M(T), \qquad (9)$$

Here, N is the demagnetization factor, M is the magnetization.

In order to reduce the demagnetization factor, the sample is mounted so that its longest side is parallel to the external field direction . Measurements were carried out on a plate-like sintered sample (3.7x13.5x18.3 mm^3) of $YBa_2Cu_3O_{6.9}$ having the superconducting temperature T_c=90.4K. In the experiment we did not exploit the whole of the sample volume. It was limited by the neutron beam cross section (2x8 mm^2). To study the time dependence of the trapped flux magnitude, the sample was cooled in the external field H_{fc} ("field-cooled" regime). At T=120K a homogeneous static magnetic field was switched on, and then the sample temperature was decreased down to T=78K. One hour after the start of the cooling, the field gradually (for 3 sec) decreased down to B_o=24G. The moment, when the field was switched off was considered as the start of the measurement of ω_λ versus time. In 10^4-10^5s the sample was heated up to T=120K. Then the frequency $\omega_\lambda(T>T_c)$ was measured for several hours.

Figure 6 presents the results of the T=78 K measurement of

the remnant induction of the magnetic field trapped in a sample versus observation time . The measurements were made for several values of H_{fc} (22Oe, 48Oe, 71Oe, 79Oe and 165Oe). In Fig.6 it is seen that the magnitude of the magnetic induction in a bulk sintered superconductor decreases with increasing observation time. The rate depends on the value of the remnant trapped field, in which the sample was cooled. In the course of the field, H_{fc}, increase in the interval of observation times $10^3<t<10^5$s, the decay rate grows and the time dependence of the induction starts to deviate from the logarithmic law.

The time dependences of the internal field amplitude are approximated by the LSQ-method using the following functions $f(t)=\ln(t/t_o)$ [16] and $f(t)=\exp[-(t/\tau)^\beta]$ [17]. The experimental values of $B(T)/B_o$ at T=78K for H_{fc}=48 Oe, 71 Oe, 79 Oe and 165 Oe for $t>10^3$s are best fitted by the parametric dependence $f(t) = A + B \exp[-t/\tau)^\beta]$ in the observation time interval up to 0.8×10^5 sec.

Table II summarizes the values of τ^{-1} and β (see [17]) for various values of H_{fc}. From Table II it is seen that the value of β in the limit of errors lies within the interval $0<\beta<1$.

Thus, in the presence of a static external magnetic field, B_o, change in the magnitude of a magnetic field, trapped in sintered $YBa_2Cu_3O_{6.9}$, as the function of the observation time exhibits the nonlogarithmic behaviour for $t<10^5$s and has the nonmonotonous character, which is seen quite well for H_{fc} = 165 Oe. The observed long-time relaxation behavior of the magnetization can be explained qualitatively in the framework of the mode-coupling theory of superconductive glass [18].

4. Summary

The neutron depolarization investigation of the inhomogeneous magnetic flux in a superconducting $YBa_2Cu_3O_{7-\delta}$ ceramics has shown, that, in general, the character of changes in the magnetic state of the HTSC confirms the observations for the low temperature superconductors. However, several new peculiarities were observed. In particular, from the sameness of the P(H) behaviour in the increasing and decreasing field with respect to the field direction it follows that pinning is rather weak above T = 79K. In the irreversibility region near T_c the P(H) has a second (non-equilibrium) minimum, which was assumed to appear due to the long-time character of the magnetic flux relaxation. Such behavior was confirmed by the observed nonmonotonous behaviour of the change in depolarization with time.

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