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POSITRON ANNIHILATION IN A HIGH-TEMPERATURE SUPERCONDUCTOR $YBa_2Cu_3O_{7-\delta}$

J.Wawryszczuk, T.Goworek[1], A.I.Ivanov[2], M.Lewandowski, P.Mazurek[1], V.N.Rybakov, I.F.Uchevatkin[2], I.A.Yutlandov

Positron annihilation as a function of temperature in high-temperature superconductors $YBa_2Cu_3O_{7-\delta}$ has been investigated. It is shown that a change in the annihilation character at the transition into superconducting state is relatively small. The change of τ_1 and τ_2 positron lifetimes as well as of the intensity of the component with $\tau_2 - J_2$ and Doppler broadening S parameter allows one to assume that transition into superconducting state is accompanied with a certain decrease in electron density and with decreasing number of defects or increasing their size.

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

Аннигиляция позитронов в высокотемпературном сверхпроводнике $YBa_2Cu_3O_{7-\delta}$

Я.Заврыщук и др.

Изучена аннигиляция позитронов в образцах высокотемпературной сверхпроводящей керамики $YBa_2Cu_3O_{7-\delta}$. Показано, что изменение характера аннигиляции при переходе в сверхпроводящее состояние относительно невелико. Изменения времен жизни позитронов τ_1 и τ_2, а также интенсивности компоненты J_2 и допплеровского уширения аннигиляционной γ-линии (параметра S) позволяют предполагать, что переход в сверхпроводящее состояние сопровождается некоторым уменьшением электронной плотности и уменьшением числа или увеличением размеров дефектов кристаллической решетки.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Nowadays superconductors like La-Ba-Cu-O/1/ have been intensely studied by all available methods, including the positron annihilation method which is especially sensitive to the structure of a sub-

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[1] *Institute of Physics of Curie-Sklodowska University, Lublin*
[2] *All-Union Mendeleev Research Institute of Metrology, Leningrad*

stance. Usually, the parameters describing the annihilation process (positron lifetime, electron momentum distribution width[2]) significantly change in the phase transition point. Despite the disappointing results obtained with common metal superconductors in the 50-s[3-5], it seems reasonable to follow the behaviour of the annihilation parameters in the transition region (T_c) of high-temperature superconductors. In the first paper[6] dealing with this problem the Doppler broadening of the 511 keV annihilation γ-line was measured for La-Sr-Cu-O and Y-Ba-Cu-O systems. In further experiments[7,8] positron lifetimes were also measured for Y-Ba-Cu-O system. Ambiguity and sometimes discrepancy of the results obtained make it necessary to continue the investigations.

This paper presents the results of positron lifetime measurements for $YBa_2Cu_3O_{7-\delta}$ and Doppler broadening measurements for the annihilation γ-line in the temperature interval 80-130 K.

Experimental Technique

Positron lifetimes were measured by means of a $\gamma\gamma$-coincidence time spectrometer with two BaF_2 crystalls 38x25 mm in size. The energy resolution of both scintillators with photomultiplyers XP2020Q was 7% for the ^{60}Co 1333 keV line. To eliminate distortions of the time spectrum shape at large loads in the coincidence selection circuits, blocks were used to reject overlapping pulses. Under the experimental conditions (for 1274 and 511 keV γ-quanta) the time resolution of the spectrometer was $2\tau_0$ = 220 ps. The shape of the instantaneous coincidence curve for ^{60}Co corresponded to one Gaussian distribution up to 0.001-th of its full maximum. The time scale was graduated to 22.0(1) ps/channel.

The Doppler broadening of the 511 keV annihilation γ-line (S-parameter) was measured by an X-ray Ge(Li)-detector of volume 1 sm^3 and energy resolution 1.02 keV for the ^{106}Ru 512 keV line. The energy value of the channel was 0.080 keV. Instability of the 511 keV line position during measurements did not exceed one channel.

For measurements at different temperatures the $YBa_2Cu_3O_{7-\delta}$ samples were placed in a cylinder-shaped liquid-nitrogen-cooled vacuum cryostat ($p \simeq 10^{-3}$ Torr), diameter 18 mm. Temperature was changed by heating the intermediate hollow copper cylinder by current flowing through a double-wound winding around this cylinder. Inside it there was a small copper cylinder with the sample tightly inserted in a slot. The temperature of the sample was measured in relation to liquid nitrogen temperature by means of a copper-constantan thermocouple. The voltage from the thermocouple was also used for temperature stabilisation (winding current correction). The stabilisation system we had developed allowed a constant temperature of the sample to an accuracy better than 0.3 K in the interval 79-200 K.

The positron source of activity ~ 30 μK was prepared by evaporating the aqueous solution of ^{22}NaCl on nikel foil 1.2 μm thick coated with a gold layer 50 Å thick. The source area was ~ 8 mm^2.

The time spectra were processed by the programme POSITRONFIT [9] in a microcomputer of the type IBM XT which is part of the measurement apparatus. Correction for positron annihilation in the nickel foil (~ 8%) was not taken into account. The time resolution of spectrometer $2\tau_0$ was also regarded as a fitting parameter. The values of $2\tau_0$ obtained in the fitting were within 222-225 ps. There were $\gtrsim 1.2 \cdot 10^6$ coincidences registered for each time spectrum.

To follow the annihilation γ-line shape varying with the sample temperature, the S parameter was calculated, which is the ratio of the number of counts in 14 channels of the central part of the 511 keV peak to the sum of counts in two windows (18 channels each) on the peak's left and right slopes.

$YBa_2Cu_3O_{7-\delta}$ Samples

The samples to be investigated were prepared by sintering Y_2O_3, BaO_2, CuO in the Laboratory of Nuclear Problems, JINR (sample 1) and by sintering Y_2O_3, $BaCO_3$, CuO in the Institute of Physics of the Curie-Sklodowska Lublin University (samples 2, 3). The sintering temperature was 950°C, the superconducting transition temperatures T_c were 96, 86, 95 K, respectively. The behaviour of the function R(T) allowed an assumption that all three samples were not single-phase ones.

Results of Measurements and Discussion

The components with $\tau_1 \simeq 180$ ps, $\tau_2 \simeq 350$ ps and $\tau_3 \simeq 1.9$ ns can be singled out in the positron lifetime spectra of the samples under investigation. The intensity of the longest-lived component τ_3 did not exceed 0.55 % and J_2 varied from a sample to a sample within the range of 8%-17 %. The attempts to single out only two components lead to a significantly worse reduced χ^2 (~ 1.3 at two components instead of ~1.1 at three components) and to $\tau_1 \simeq 190$ ps,

$\tau_2 \simeq 480$ ps. Since no variations of τ_3 were found in the temperature range investigated, the final analysis of all time spectra was performed at a fixed averaged value of this parameter.

Our measurements in the temperature range of 80-130 K and at room temperature showed that the change in the positron annihilation character in our samples at their transition to the superconducting state is relatively small. The transition to the superconducting state is seen to lead to larger τ_1 and τ_2, to a small intensity of J_2 and parameter S (Figure).

If one considers that the component with $\tau_1 = 180$ ps is related to the free positron annihilation in the space between lattice points, the small increase in τ_1 observed at the supperconducting transition of the sample may indicate a change in the electron structure which leads to a lower electron density. The component with $\tau_2 \simeq 350$ ps typical of annihilation of positrons captured by lattice defects should be perhaps associated with oxygen vacancies. A decrease in intensity of J_2 and parameter S at $T < T_c$ allows an assumption that the number of these defects reduces in the superconducting state. A larger τ_2 can be associated with the decreasing electron density or the increasing size of the defects. The weak component with $\tau_3 \simeq 1.9$ ns is probable due to positronium production in the porous structure of the metal-oxide ceramics.

It is state in Ref. /7/ that the lifetime τ_1 (139 ± 7 ps) does not depend on the sample temperature, the lifetime τ_2 (~210 ps) noticeably decreases at the superconducting transition while J_2 (~30%) increases. It is strange, however, that the parameter S in the superconducting state decreases as in our experiments. A possible reason for discrepancy between our results and those of Ref. /7/ is a difference in the composition of the samples investigated.

In the experiments /6/ only the Doppler broadening of the annihilation line was studied. The results for the Y-Ba-Cu-O system do not contradict our data.

An abnormal behaviour of τ_1, τ_2 and J_2 aroung T_c was observed in Ref. /8/. The lifetimes τ_1 and τ_2 have a sharp maximum of half-width ~1 K while J_2 has a deep minimum. However, the values of τ_1 and τ_2 below and above T_c agree with our results for the case of resolving the time spectrum into 2 components. Besides, an unusual increase in the positron thermalisation time t_0 (by ~130 ps) was observed in Ref. /8/ at T_c. An anomaly like this was found neither in our paper, nor in Ref. /7/.

Having compared the above results one may say that: (a) the superconducting transition of Y-Ba-Cu-O systems affects positron annihilation character; (b) quite probably, annihilation process is

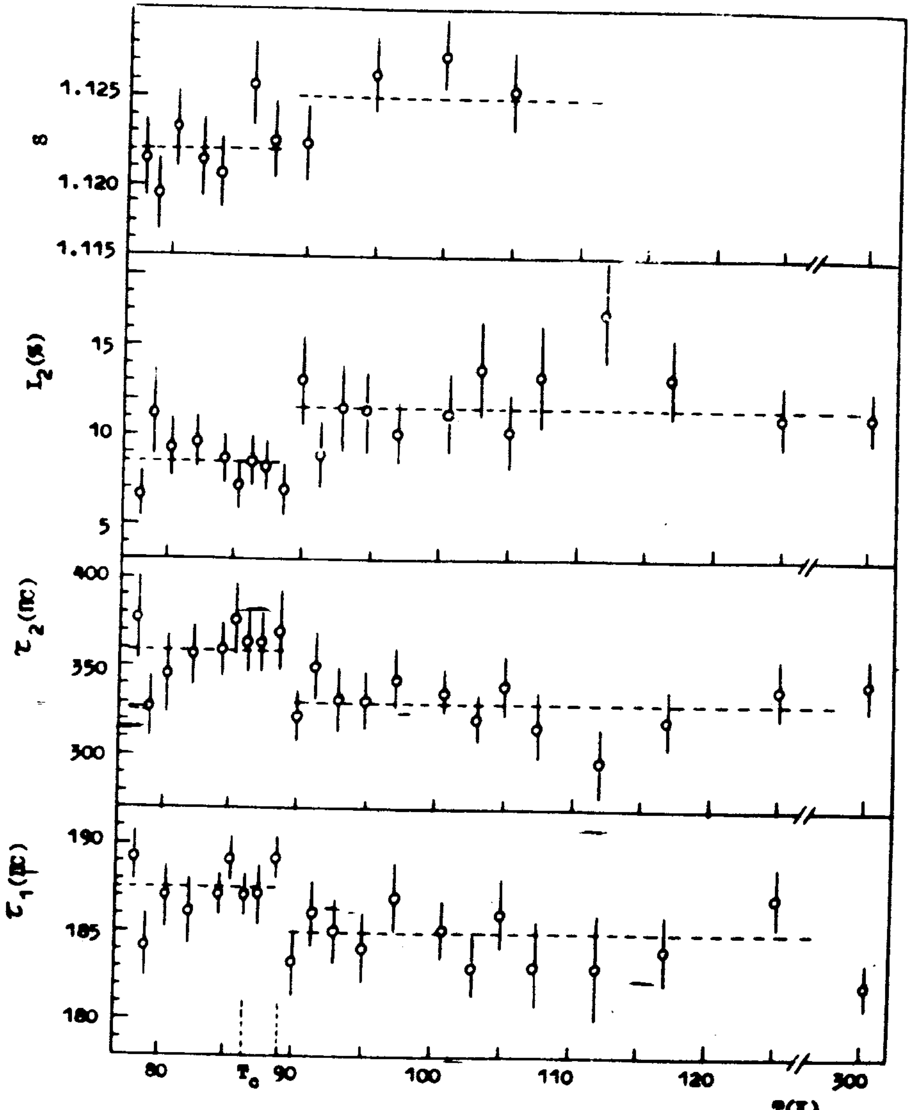

Figure. Temperature dependence of the parameters τ_1, τ_2, J_2 and S. Dashed lines denote the mean parameter values in regions below and above T_c.

very sensitive to the internal structure details of the samples and to their preparation technique. It is proved by the opposite temperature dependence of τ_2 and J_2 in this paper and Ref. /7/, and by their different absolute values.

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