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PROBLEMS OF COEXISTENCE OF SUPERCONDUCTIVITY AND MAGNETIC ORDERING OF COPPER SUBLATTICES IN YBa2Cu3-XFeXO7-X CERAMICS

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PROBLEMS OF COEXISTENCE OF SUPERCONDUCTIVITY AND MAGNETIC ORDERING OF COPPER SUBLATTICES IN $YBa_2Cu_{3-x}Fe_xO_{7-x}$ CERAMICS

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Abstract. *The article studies the problems of coexistence of superconductivity and magnetic ordering of copper sublattices in $YBa_2Cu_{3-x}Fe_xO_{7-x}$ ceramics.*

It is known that in superconducting ceramics $YBa_2Cu_3O_{7-z}$ with orthorhombic lattice as the oxygen content decreases, the transition temperature in superconducting state T_c decreases, and at $z>0.6$ superconductivity disappears, the lattice becomes tetragonal and at the same time, antiferromagnetic ordering of sublattices Cu (2) appears. The substitution in ceramics of $YBa_2Cu_3O_7$ part of copper atoms by iron atoms (i.e., the formation of a solid solution of $YBa_2Cu_{3-x}Fe_xO_{7+y}$) is accompanied by similar effects: as x increases, Tc decreases, at $x>0.05$ the orthorhombic lattice becomes tetragonal, at $x>0.45$ the superconductivity disappears. The most significant moment is the fact of coexistence in ceramics $YBa_2Cu_{3-x}Fe_xO_{7+y}$ in the region of compositions $0.03 < x < 0.45$ of superconductivity and magnetic ordering of iron atoms in copper nodes (the latter is established by Mossbauer spectroscopy on isotope ^{57}Fe in a large number of works. However, it remains unclear whether the magnetic ordering of iron atoms in the $YBa_2Cu_{3-x}Fe_xO_{7+y}$ lattice is related to the magnetic ordering of copper atoms.

Key words: *Mossbauer spectroscopy, electric field gradient, semiconductor ceramics, orthorhombic, copper node.*

It is known that in superconducting ceramics $YBa_2Cu_3O_{7-z}$ with orthorhombic lattice as oxygen content decreases, the temperature of transition in superconducting state T_c decreases, and at $z>0.6$ superconductivity disappears, the lattice becomes tetragonal and at the same time, antiferromagnetic ordering of the Cu (2) sublattices appears [1,2]. The substitution in ceramics of $YBa_2Cu_3O_7$ part of copper atoms by iron atoms (i.e., the formation of a solid solution of $YBa_2Cu_{3-x}Fe_xO_{7+y}$) is accompanied by similar effects: as x increases, T_c decreases, at $x>0.05$ the orthorhombic lattice becomes tetragonal, at $x>0.45$ the superconductivity disappears [3]. The most significant moment is the fact of coexistence in the ceramic $YBa_2Cu_{3-x}Fe_xO_{7+y}$ in the region of compositions $0.03 < x < 0.45$ of superconductivity and magnetic ordering of iron atoms in copper nodes (the latter was established by Mossbauer spectroscopy on isotope ^{57}Fe in a large number of papers [4-7]. However, it remains unclear whether the magnetic ordering of iron atoms in the $YBa_2Cu_{3-x}Fe_xO_{7+y}$ lattice is related to the magnetic ordering of copper atoms (see, for example, [8]).

To solve this problem, it seems promising to use emission Mossbauer spectroscopy on isotope ^{61}Cu (^{61}Ni): after the decay of the mother nucleus ^{61}Cu, the Mossbauer probe ^{61}Ni is formed in the copper node, whose nuclear parameters allow reliable recording of magnetic ordering in copper nodes [9]. Two pairs of samples were used for investigations: $YBa_2Cu_3O_{6.96}$ (orthorhombic modification, T_c = 92 K), $YBa_2Cu_3O_{6.1}$ (tetragonal modification, T_c<4.2K) and $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ (tetragonal modification, T_c= 50 K), $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ (tetragonal modification, T_c<4.2K)..

Samples of $YBa_2Cu_3O_{6.96}$ and $YBa_2Cu_{3-x}Fe_xO_{7+y}$ were prepared by high-temperature solid phase synthesis. Y_2O_3, CuO, Fe_2O_3 (enrichment by isotope ^{57}Fe was 92 %) and $BaCO_3$ were used as components. After sintering at 900˚C for 20h in the air, the samples were annealed in an oxygen current at 920˚C for 70 "s with subsequent cooling at a rate of 5 K/min. The annealing of the sample $YBa_2Cu_3O_{6.96}$ at 800˚C for 2 h with constant pumping resulted in $YBa_2Cu_3O_{6.1}$.

Samples were doped with ^{61}Cu by diffusion annealing at 450˚C for 30 min in oxygen current (except for sample $YBa_2Cu_3O_{6.1}$, which was doped by diffusion annealing at 650˚C for 30 min during pumping). No changes in structure, T_c value or oxygen content were observed for control samples. According to [10], the described procedure guarantees that the ^{61}Cu isotope enters the copper nodes of the lattice.

The ^{61}Cu (^{61}Ni) Mossbauer emission spectra were shot at 80 and 4.2 K on an industrial spectrometer, the standard absorber was $Ni_{0.86}V_{0.14}$ with a surface density of 1500 mg/cm^2. Typical spectra are shown in Fig. 1.

In the Y system, copper atoms occupy two crystallography non-equivalent positions Cu (1) and Cu (2), populated as 1:2. In accordance with this, we represented the experimental Mossbauer spectra of ^{61}Cu (^{61}Ni) of the above ceramics as an overlay of two multiplets corresponding to the $^{61}Ni^{2+}$ centers in Cu (1) and Cu (2) nodes. Each multiplet was described by a superposition of either five lines with relative intensities 10 : 4 : 1 : 6 : 9 (in the case of a pure quadrupole interaction), or twelve lines with relative intensities 10 : 4 : 1 : 6 : 6 : 3 : 3 : 6 : 6 : 1 : 4 : 10 (in the case of the combined quadrupole and Zeeman interactions), and the position of the multiplet lines was determined as the difference in the eigenvalues $E_m^I$of the Hamiltonian of the combined superfine interaction of the excited and the main states ^{61}Ni

$$E_m^I = mg\beta_N H + \{eQU_{zz}/4I(2I-1)\} \times \{3m^2 - I(I+1)\}\{(3cos^2\theta - 1)/2\},$$

where I is the spin of the nucleus, H is the magnetic field on the nucleus, $U_{zz}$is the main component of the electric field gradient (EFG) tensor on the nucleus, θ– is the angle between the main axis of the EFG tensor and the direction of the magnetic field, m is the magnetic quantum

number, Q is the quadrupole moment of the nucleus, g is the nuclear g is factor, β_N is the nuclear magneton. The above formula is valid for the axially symmetric EFG tensor both for $gH \gg eQU_{zz}$, and for $H = 0$ (but in the latter case we should take ($\theta = 0°$).

The computational spectrum was adjusted to the experimental method of least squares, and the fitting parameters were not the parameters of individual lines, but the Hamiltonian parameters $H$and$U_{zz}\{(3cos^2\theta - 1)/2\}$, as well as the positions of the centers of gravity of the multiplets. Since no isomer shift is observed in the Mossbauer ^{61}Ni spectra [11], we made sure that the center of gravity of the calculated multiplet does not deviate from zero speed by more than $\pm 0.05\ mm/s$.

Mossbauer spectrum ^{61}Cu (^{61}Ni) of superconducting ceramic $YBa_2Cu_3O_{6.96}$ is a superposition of two quadrupole multiplets corresponding to the centers of gravity of ^{61}Ni (1) and ^{61}Ni (2). Fig. 1 indicates the positions of the components of the corresponding multiplets and their relative intensities. The ratio of areas under the spectra ^{61}Ni (2) and ^{61}Ni (1) P=1.95 (5), which is close to the population ratio of nodes Cu (2) and Cu (1). The obtained parameters of the spectra are as follows: $eQU_{zz} = 32$ (2) MHz for centers ^{61}Ni (1) and $eQU_{zz} = -54$ (2) MHz for spectra ^{61}Ni (2) (here Q is the quadrupole moment of the ^{61}Ni core in the main state).

Mossbauer spectrum of ^{61}Cu (^{61}Ni) semiconductor ceramics $YBa_2Cu_3O_{6.1}$(Fig. 1, b) is a superposition of a quadrupole multiplets corresponding to the $^{61}Ni^{2+}$ nodes Cu (1) ($|eQU_{zz}| < 30MHz$), and a multiplate corresponding to the $^{61}Ni^{2+}$ nodes Cu (2), And the fine structure of the last spectrum is obliged by the origin to the combined superfine (Zeeman and electric quadrupole) interaction ($eQU_{zz} = -48$ (3) MHz, $H = 85$ (5) kOe, $\theta = 90$ (10)°). The ratio of areas under the spectra ^{61}Ni (2) and ^{61}Ni (1) remains equal to the ratio of the population of nodes Cu (2) and Cu (1) in the lattice $YBa_2Cu_3O_7$ (P=1.97 (5)). The spectra in Fig. 1 illustrate the possibilities of Mossbauer emission spectroscopy at the isotope ^{61}Cu (^{61}Ni) to observe the magnetic ordering of the copper sublattice of $YBa_2Cu_3O_{7-z}$ ceramics with a decrease in oxygen content.

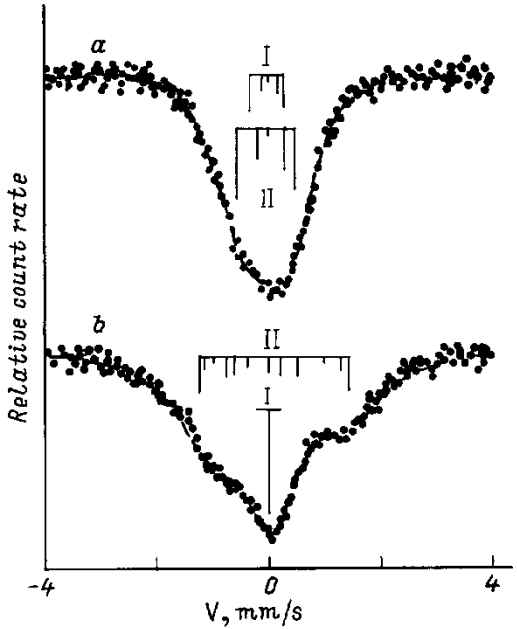

Fig.1. Mossbauer emission spectra of ^{61}Cu (^{61}Ni) at 80 K for $YBa_2Cu_3O_{6.96}$ (*a*) and $YBa_2Cu_3O_{6.1}$ (*b*) ceramics. The position of multiplets components corresponding to $^{61}Ni^{2+}$ centers in Cu (1) (I) and Cu (2) (II) nodes is indicated.

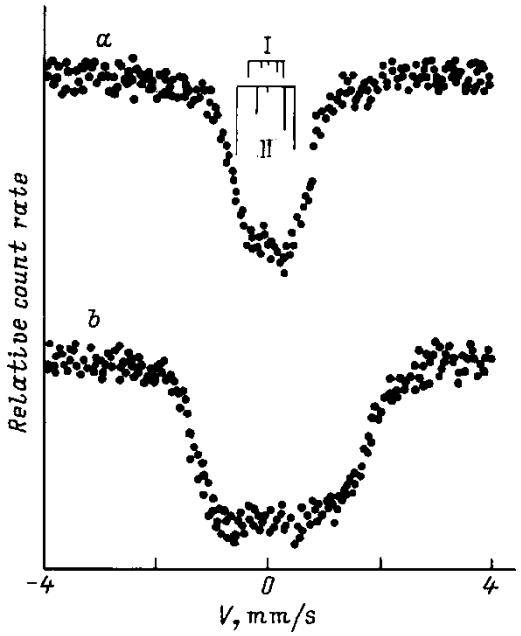

Fig.2. Mossbauer emission spectra of ^{61}Cu (^{61}Ni) at 4.2K for $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ (*a*) and $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ (*b*) ceramics. For spectrum *a*, the position of the multiplets components corresponding to centers $^{61}Ni^{2+}$ in nodes Cu (1) (I) and Cu (2) (II) is indicated.

The Mossbauer spectrum of ^{61}Cu (^{61}Ni) superconducting ceramics $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ is a superposition of two quadrupole multiplets (Fig. 2, a), whose parameters are close to the parameters of the corresponding spectra of the ceramics $YBa_2Cu_3O_{6.96}$, although the ratio of areas under the ^{61}Ni (2) and ^{61}Ni (1) spectra differs significantly from the expected value (P=4.0 (4)). This is obviously due both to a decrease in the proportion of Cu (1) centers (due to partial substitution of a part of Cu (1) nodes by iron impurity atoms) and to the influence of iron impurity atoms on the parameters of the Mossbauer ^{61}Ni spectra (which decreases the proportion of the undisturbed spectrum from ^{61}Ni (1) atoms).

For ceramics $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$, in which superconductivity is suppressed, a Zeeman splitting is observed in the ^{61}Cu (^{61}Ni) Mossbauer spectra (Fig. 2, b). Unfortunately, the resolution of the last ceramics spectra was insufficient for the extraction of components corresponding to $^{61}Ni^{2+}$ centers in Cu (1) and Cu (2) from experimental spectra. So, as in the case of ceramics $YBa_2Cu_3O_{7-z}$, for ceramics $YBa_2Cu_{3-x}Fe_xO_{7+y}$ there is an obvious correlation

between the appearance of magnetic ordering of one of the copper sublattice and disappearance of the superconductivity phenomenon.

For ceramics $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ the Mossbauer spectra ^{57}Fe were also measured (^{57}Co in palladium was used as a standard source). In agreement with the literature data at T<50 K, the spectra are poorly resolved Zeeman multiplets corresponding to impurity iron atoms in nodes Cu (1) in the "spin glass" state. Thus, we must state that there is no correlation between the magnetic ordering of impurity iron atoms in sublattice Cu (1) and the magnetic ordering of copper sublattice ceramics $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$. However, the increase in iron concentration (transition to $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$) is accompanied by both the complete suppression of superconductivity and the appearance of magnetic ordering of copper sublattice. Since in the latter ceramics a part of iron atoms is stabilized in the Cu (2) sublattice, it is obvious that the appearance of the magnetic ordering of the copper sublattice should be associated with these iron atoms.

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