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

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Abstract. The article studies the problems of coexistence of superconductivity and magnetic ordering of copper sublattices in YBa$_2$Cu$_{3-x}$Fe$_x$O$_{7-x}$ ceramics.

It is known that in superconducting ceramics YBa$_2$Cu$_3$O$_{7-z}$ 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$_2$Cu$_3$O$_7$ part of copper atoms by iron atoms (i.e., the formation of a solid solution of YBa$_2$Cu$_{3-x}$Fe$_x$O$_{7+y}$) 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. The most significant moment is the fact of coexistence in ceramics YBa$_2$Cu$_{3-x}$Fe$_x$O$_{7+y}$ 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$_2$Cu$_{3-x}$Fe$_x$O$_{7+y}$ lattice is related to the magnetic ordering of copper atoms.

Key words: Mossbauer spectroscopy, electric field gradient, semiconductor ceramics, orthorhombic, copper node.
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$_2$Cu$_3$O$_{6.96}$ (orthorhombic modification, $T_c = 92$ K), YBa$_2$Cu$_3$O$_{6.1}$ (tetragonal modification, $T_c < 4.2$K) and YBa$_2$Cu$_2$Fe$_{0.2}$O$_{7.03}$ (tetragonal modification, $T_c = 50$ K), YBa$_2$Cu$_{2.5}$Fe$_{0.5}$O$_{7.18}$ (tetragonal modification, $T_c < 4.2$K).

Samples of YBa$_2$Cu$_3$O$_{6.96}$ and YBa$_2$Cu$_{3-x}$Fe$_x$O$_{7+y}$ were prepared by high-temperature solid phase synthesis. Y$_2$O$_3$, CuO, Fe$_2$O$_3$ (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$_2$Cu$_3$O$_{6.96}$ at 800°C for 2 h with constant pumping resulted in YBa$_2$Cu$_3$O$_{6.1}$.

Samples were doped with $^{61}$Cu by diffusion annealing at 450°C for 30 min in oxygen current (except for sample YBa$_2$Cu$_3$O$_{6.1}$, 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 : 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 $E_m$ of the Hamiltonian of the combined superfine interaction of the excited and the main states $^{61}$Ni

$$E_m = m g \beta_N H + \left\{eQ U_{zz}/4I(2I - 1)\right\} \times \left\{3m^2 - I(I + 1)\right\}((3\cos^2 \theta - 1)/2),$$

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, $\theta$ – 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, $\beta_N$ 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 $\theta = 0^\circ$).

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 $H_{\text{and}}U_{zz} \{ (3\cos^2\theta - 1)/2 \}$, as well as the positions of the centers of gravity of the multiplets. Since no isomer shift is observed in the Mossbauer $^{61}\text{Ni}$ spectra [11], we made sure that the center of gravity of the calculated multiplet does not deviate from zero speed by more than $\pm 0.05 \text{ mm/s}$.

Mossbauer spectrum $^{61}\text{Cu} (^{61}\text{Ni})$ of superconducting ceramic $\text{YBa}_2\text{Cu}_3\text{O}_{6.96}$ is a superposition of two quadrupole multiplets corresponding to the centers of gravity of $^{61}\text{Ni}$ (1) and $^{61}\text{Ni}$ (2). Fig. 1 indicates the positions of the components of the corresponding multiplets and their relative intensities. The ratio of areas under the spectra $^{61}\text{Ni}$ (2) and $^{61}\text{Ni}$ (1) $P=1.95$ (5), which is close to the population ratio of nodes $\text{Cu}$ (2) and $\text{Cu}$ (1). The obtained parameters of the spectra are as follows: $eQU_{zz} = 32$ (2) MHz for centers $^{61}\text{Ni}$ (1) and $eQU_{zz} = -54$ (2) MHz for spectra $^{61}\text{Ni}$ (2) (here $Q$ is the quadrupole moment of the $^{61}\text{Ni}$ core in the main state).

Mossbauer spectrum of $^{61}\text{Cu} (^{61}\text{Ni})$ semiconductor ceramics $\text{YBa}_2\text{Cu}_3\text{O}_{6.1}$ (Fig. 1, b) is a superposition of a quadrupole multiplets corresponding to the $^{61}\text{Ni}^{2+}$ nodes $\text{Cu}$ (1) ($|eQU_{zz}| < 30 \text{MHz}$), and a multiplet corresponding to the $^{61}\text{Ni}^{2+}$ nodes $\text{Cu}$ (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)$^\circ$). The ratio of areas under the spectra $^{61}\text{Ni}$ (2) and $^{61}\text{Ni}$ (1) remains equal to the ratio of the population of nodes $\text{Cu}$ (2) and $\text{Cu}$ (1) in the lattice $\text{YBa}_2\text{Cu}_3\text{O}_7$ ($P=1.97$ (5)). The spectra in Fig. 1 illustrate the possibilities of Mossbauer emission spectroscopy at the isotope $^{61}\text{Cu} (^{61}\text{Ni})$ to observe the magnetic ordering of the copper sublattice of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ceramics with a decrease in oxygen content.
Fig. 1. Mossbauer emission spectra of $^{61}$Cu ($^{61}$Ni) at 80 K for YBa$_2$Cu$_3$O$_{6.96}$ (a) and YBa$_2$Cu$_3$O$_{6.1}$ (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$_2$Cu$_{2.8}$Fe$_{0.2}$O$_{7.03}$ (a) and YBa$_2$Cu$_{2.5}$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$_2$Cu$_{2.8}$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$_2$Cu$_3$O$_{6.96}$, 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$_2$Cu$_{2.5}$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$_2$Cu$_{3}$O$_{7-z}$, for ceramics YBa$_2$Cu$_{3-z}$Fe$_{z}$O$_{7+y}$ 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 $\text{YBa}_2\text{Cu}_{2.8}\text{Fe}_{0.2}\text{O}_{7.03}$ the Mossbauer spectra $^{57}\text{Fe}$ were also measured ($^{57}\text{Co}$ 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 $\text{Cu (1)}$ in the "spin glass" state. Thus, we must state that there is no correlation between the magnetic ordering of impurity iron atoms in sublattice $\text{Cu (1)}$ and the magnetic ordering of copper sublattice ceramics $\text{YBa}_2\text{Cu}_{2.8}\text{Fe}_{0.2}\text{O}_{7.03}$. However, the increase in iron concentration (transition to $\text{YBa}_2\text{Cu}_{2.5}\text{Fe}_{0.5}\text{O}_{7.18}$) 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 $\text{Cu (2)}$ sublattice, it is obvious that the appearance of the magnetic ordering of the copper sublattice should be associated with these iron atoms.

Reference