Pis'ma v ZhETF, vol.60, N6, pp.439 - 441 © 1994 September 25

SUPERCONDUCTIVITY AND MAGNETIC ORDER IN THE COPPER SUBLATTICES OF THE $YBa_2Cu_{3-x}Fe_xO_{7+y}$ CERAMICS

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Submitted 16 August, 1994

The correlation between the superconductivity suppression and the magnetic ordering of the copper atoms is determined in the $YBa_2Cu_{3-x}Fe_xO_{7+y}$ lattice by the $^{61}Cu(^{61}Ni)$ emission Mossbauer spectroscopy (EMS).

It is known that the superconducting transition temperature T_C decreases as oxygen content decreases in the $YBa_2Cu_3O_{7-z}$ superconductors with orthorhombic lattice as well as superconductivity disappears, the antiferromagnetic order appears in the Cu(2) sublattice and the lattice becomes tetragonal when $z > 0.6$ [1,2]. When the $YBa_2Cu_{3-x}Fe_xO_{7+y}$ solid solutions form, the similar effects are observed: T_C decreases as x increases, the orthorhombic lattice transforms to the tetragonal one when $x > 0.05$ and the superconductivity disappears when $x > 0.45$ [3]. It is important that the superconductivity and the magnetic order of the Fe ions at the Cu sites may coexist in the composition range $x = 0.03$ to 0.45 [4-7]. However, it is still unknown whether the iron magnetic ordering in $YBa_2Cu_{3-x}Fe_xO_{7+y}$ is resulted from the magnetic order of the copper atoms (see, e.g., [8]).

The $^{61}Cu(^{61}Ni)$ EMS accepted in this work should be useful to solve this problem. In the above technique a decay of the ^{61}Cu parent nucleus lead to the ^{61}Ni Mossbauer probe with nuclear parameters provided a reliable revealing of the magnetic order of copper sites [9]. Two pairs of compositions were investigated: $YBa_2Cu_3O_{6.96}$ (orthorhombic, $T_C = 92\,K$), $YBa_2Cu_3O_{6.1}$ (tetragonal, $T_C < 4.2\,K$) on the one hand and $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ (tetragonal, $T_C = 50\,K$) and $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ (tetragonal, $T_C < 4.2\,K$) on the other hand.

The samples were prepared by a ceramic technique. The doping with ^{61}Cu was carried out by diffusion during 30 min at 650°C under pumping-out for $YBa_2Cu_3O_{6.1}$ and at 450°C for the other samples. According [10] this treatment provides ^{61}Cu at the regular Cu sites and does not change the structure and T_C of the samples. The $^{61}Cu(^{61}Ni)$ emission Mossbauer spectra (MS) were recorded at 80 and 4.2 K. The spectra are shown in Fig.1 and 2.

We considered the $^{61}Cu(^{61}Ni)$ MS as superpositions of two multiplets corresponding to Ni at the Cu(1) and Cu(2) sites. The multiplets were described by 5 or 12 lines for the pure quadrupole interaction and for the combined quadrupole and Zeeman interactions, respectively.

The calculated spectra were fitted to the measured ones by the least square method. The fitting quantities were the Hamiltonian parameters H and $U_{zz}\{(3\cos^2\theta - 1)/2\}$ (where H is magnetic field and U_{zz} is the principal component of the electric field gradient (EFG) tensor, both on the nucleus, θ is the angle between the z axis of the EFG tensor and the magnetic field direction) as well as multiplet centroids positions rather than parameters of the separate lines. The fit

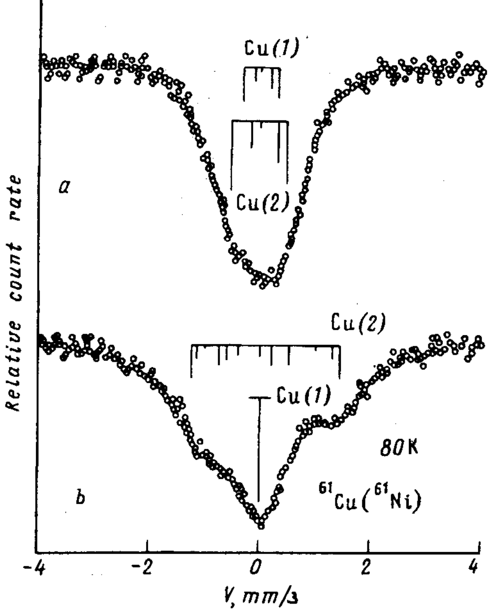

Fig.1

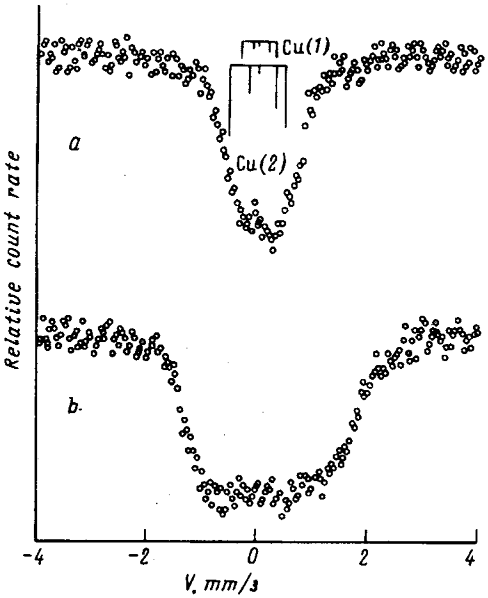

Fig.2

Fig.1. The $^{61}Cu(^{61}Ni)$ MS recorded at 80K for the $YBa_2Cu_3O_{6.96}$ (*a*) and $YBa_2Cu_3O_{6.1}$ (*b*) ceramic samples. The positions and the intensities of the components of the quadrupole multiplets are shown

Fig.2. The $^{61}Cu(^{61}Ni)$ MS recorded at 4.2K for the $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ (*a*) and $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ (*b*) ceramic samples. The positions and the intensities of the components of multiplets are shown

was tested by the χ^2-criterion. In addition we selected the multiplet centroids in the range ± 0.05 mm/s only because the isomer shift does not reveals in the ^{61}Ni MS [11].

The $^{61}Cu(^{61}Ni)$ MS of the $YBa_2Cu_3O_{6.96}$ superconducting sample (Fig.1) consists of two quadrupole multiplets corresponding to $^{61}Ni(2)$ and $^{61}Ni(1)$ centres. Their relative intensities $P = 1.95(5)$ is quite close to the above relative populations of the Cu(2) to Cu(1) sites. The quadrupole coupling constants eQU_{zz} as regarded to the ^{61}Ni ground state were found -32(2) MHz and -54(2) MHz for Ni(1) and Ni(2), respectively.

The $^{61}Cu(^{61}Ni)$ MS of the $YBa_2Cu_3O_{6.1}$ semiconducting sample (Fig.1) is a superposition of a narrow ($|eqU_{zz}| < 30$ MHz) quadrupole multiplet ascribed to $^{61}Ni(1)$ and a multiplet arising from the combined quadrupole and Zeeman interaction of $^{61}Ni(2)$. The obtained parameters are $H = 85(5)$ kOe and $eQU_{zz} = -48(3)$ MHz assuming $\theta = 90(10)^\circ$ [2]. The relative intensity of the multiplets $P = 1.97(5)$ is also close to the relative population of the Cu(2) to Cu(1) sites. The magnetic field for Ni(2) is in good agreement with the magnetic order in the Cu(2) sublattice of the tetragonal $YBa_2Cu_3O_{7-z}$ phase and, thus, supports the ability of the $^{61}Cu(^{61}Ni)$ EMS to discover a magnetic ordering of copper sublattices.

The $^{61}Cu(^{61}Ni)$ MS of the $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ superconducting sample (Fig.2) consists of two quadrupole multiplets. Their parameters are very close to the parameters of the corresponding multiplets in the $YBa_2Cu_3O_{6.96}$ MS, but their relative intensity $P = 4.0(4)$ is considerably differs from the expected value 2.0. This discrepance could be accounted for by a decreasing of a share of the regular Cu(1) sites when Fe occupies predominantly the Cu(1) sublattice. The intensity of the regular Ni(1) spectrum could be additionally decreased by the Ni(1) atoms with the Fe atoms in their nearest surrounding.

The $^{61}Cu(^{61}Ni)$ MS of the $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ sample with the suppressed superconductivity (Fig.2) reveals a Zeeman splitting. The resolution of the spectrum is not enough unfortunately to separate contributions from the Ni(1) and Ni(2) centers. Thus a clear correlation between the magnetic order in one of the copper sublattices and the superconductivity suppression is observed for both $YBa_2Cu_3Fe_xO_{7-z}$ and $YBa_2Cu_{3-x}O_{7+y}$ ceramics.

For the $YBa_2Cu_{3-x}Fe_xO_{7+y}$ samples the ^{57}Fe MS were recorded also. In accordance with [4-7] the spectra of $YBa_2Cu_{2.8}Fe_{0.2}O_{7.03}$ consist of four quadrupole doublets. At $T < 50\,K$ one of them transforms in a badly resolved Zeeman sixtet corresponding to a spin-glass-like Fe in Cu(1) sublattice. Together with our $^{61}Cu(^{61}Ni)$ data it shows that the magnetic field on ^{57}Fe does not arise from the magnetic order of copper. For the $YBa_2Cu_{2.5}Fe_{0.5}O_{7.18}$ sample the magnetic field on ^{57}Fe is observed in the both copper sublattices.

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