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MAGNETIC AND SUPERCONDUCTING PROPERTIES OF SUBSTITUTED YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_7$ COMPOUNDS (M = 3d METAL)

Ying-Chang Yang, Yuan-Bo Zha, Wei-Chun Yuan, Jian Lan, Zun-Xiao Liu, Guo-Zhong Li and Yun-Xi Sun

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Abstract. — The structural, magnetic and superconducting properties of the mixed compounds YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_7$, where M represents 3d metals, have been investigated by X-ray diffraction, electrical resistivity and magnetic measurements. The magnetic properties in the normal and superconducting states have been measured. The substituting effects for copper and their relationship to the structure are discussed.

1. Introduction

In the passing year, many works on the high transition temperature superconductors of YBa$_2$Cu$_3$O$_7$ system have been done. But the origin and the mechanism of the high-$T_c$ are still obscure. It has been found that the substitution of Y with rare earth elements has little effect on the superconducting properties [1], while Cu with 3d metals has a considerable effect [2]. Thus aroused a keen interest in the role of Cu in the superconductivity. In this paper we report a systematic experiment with 5% of Cu being substituted by 3d metal, which leads to measurable changes in their properties. With the substituted materials, we examined their structure, superconductivity and magnetic behaviours, and some interesting results were obtained.

2. Experimental

A series of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$, where M = 3d metal Ti, V, Cr, Mn, Fe, Co, Ni, Cu and Zn, were made by the conventional solid sintering procedure from Y$_2$O$_3$, BaCO$_3$, CuO and corresponding 3d metal oxides or carbonates. We also prepared a YBa$_2$(Cu$_{1-x}$Mn$_x$)$_3$O$_7$ series, where $x = 0$, 0.01, 0.03, 0.05. The ready made samples then underwent powder X-ray diffraction, electric and magnetic measurements.

3. Results and discussions

As 3d metal elements have the similar size and electrical structure to Cu, it is expected that the M atoms substitute Cu atoms in YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_7$ compounds. This assumption is supported by our results of the powder X-ray diffraction, which shows all the samples, including the original one YBa$_2$Cu$_3$O$_7$, are of the same structure, and all single phased. Furthermore, the monotonic variation of the lattice constants with $x$ in the YBa$_2$(Cu$_{1-x}$Mn$_x$)$_3$O$_7$ series also demonstrates the predicted substitution (Tab. I).

The transition temperature $T_c$ was determined by two methods. One is the conventional four probe method to measure the zero resistance point $T_c$ ($\rho_0$), the other depending on the Meissner effect defines the temperature where the susceptibility $\chi$ changes from positive to negative as $T_c$ ($\chi_0$). From table II and figure 1, we can see that although the details of these two results do not correspond completely, they clearly have the same trend: the early 3d transition elements, from Ti to Mn, effect $T_c$ slightly, while the later ones Fe, Co, Ni and Zn decrease $T_c$ dramatically. Figure 2a shows some $R(T)$ of the early 3d substituted compounds while figure 2b shows the Meissner effect below $T_c$.

The susceptibilities $\chi$ at $T = 150$ K were also drawn in figure 1, and it can be easily seen from the figure the existence of a converse relation between $T_c$ and $\chi$, except Zn which has little $\chi$ but also very low $T_c$. In figure 1, we can see that samples doped by Fe, Co, Ni with a relatively larger magnetic susceptibility have lower $T_c$ than those doped by Mn, Cr, V, Ti, Zn.

![Fig. 1. Susceptibility at 150 K and the $T_c$ defined from the Meissner effect of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$.](image-url)
Table II. - $T_c(\rho_0)$, $T_c(x_0)$, and $\chi$ at 150 K of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$.

<table>
<thead>
<tr>
<th></th>
<th>Ti</th>
<th>V</th>
<th>Cr</th>
<th>Mn</th>
<th>Fe</th>
<th>Co</th>
<th>Ni</th>
<th>Cu</th>
<th>Zn</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T(\rho_0)$ (K)</td>
<td>91.5</td>
<td>90.9</td>
<td>90.9</td>
<td>85.5</td>
<td>66.3</td>
<td>62.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T(x_0)$ (K)</td>
<td>93.5</td>
<td>93.5</td>
<td>90.9</td>
<td>89.8</td>
<td>73.7</td>
<td>74.3</td>
<td>75.7</td>
<td>96.2</td>
<td>56.0</td>
</tr>
<tr>
<td>$\chi (10^{-2}$ emu/g)</td>
<td>4.01</td>
<td>3.46</td>
<td>2.56</td>
<td>3.59</td>
<td>8.62</td>
<td>7.80</td>
<td>4.12</td>
<td>1.57</td>
<td>3.00</td>
</tr>
</tbody>
</table>

Table III. - $\chi$ at 150 K, fitted $x_0$ and $T_0$, deduced $P_M$, and the magnetic moment of free $M^{2+}$ ion of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$.

<table>
<thead>
<tr>
<th>M</th>
<th>$(10^{-2} \chi$ emu/g)</th>
<th>$(10^{-2} x_0$ emu/g)</th>
<th>$T_0$ (K)</th>
<th>$P_M$ ($\mu_B$)</th>
<th>$P_{M^{2+}}$ ($\mu_B$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>1.57</td>
<td>2.52</td>
<td>-0.614</td>
<td>0.4003</td>
<td>1.9</td>
</tr>
<tr>
<td>Fe</td>
<td>8.62</td>
<td>0.22</td>
<td>-43.9</td>
<td>5.080</td>
<td>5.4</td>
</tr>
</tbody>
</table>

Fig. 2. - (a) Temperature dependence of resistance of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$ (M = Ti, V, Cr). (b) Meissner effect of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$ ($H = 60$ Oe).

Fig. 3. - Temperature dependence of susceptibility of YBa$_2$Cu$_3$O$_7$ and YBa$_2$(Cu$_{0.95}$Fe$_{0.05}$)$_3$O$_7$.

The susceptibility suppresses $T_c$ severely while those doped by the 3d metals of Ti, V, Cr, and Mn with less magnetic susceptibility do not decrease it. As susceptibilities are directly connected with the intrinsic magnetism, it strongly suggests a close relationship between magnetism and superconductivity. Moreover, the converse relation also suggests that the high $T_c$ superconductivity come from the Cooper pair, and that after doping some elements causing larger magnetism, the stronger coupling of magnetic moment with superconductive electrons break the Cooper pair and hence decrease $T_c$. In order to study the role of magnetic moment, susceptibilities of both pure and Fe doped samples were measured from 120 K to 300 K, and it turned out to abide perfectly the reformed Curie-Weiss law (Fig. 3):

$$\chi = x_0 + NP_{\text{eff}}^2\mu_B^2/3K_B(T - T_0)$$

where $x_0$ is temperature independent (resulting from diamagnetism and Pauli paramagnetism), $N$ is the number of magnetic ions, $P_{\text{eff}}$ is the effective magnetic moment in unit of $\mu_B$, and $T_0$ is the Curie-Weiss temperature. In order to calculate the moment of Fe in YBa$_2$(Cu$_{0.95}$Fe$_{0.05}$)$_3$O$_7$, the following relation is used.

$$0.05 P_M^2 + 0.95 P_{\text{Cu}}^2 = P_{\text{eff}}^2$$

where $P_{\text{eff}}$ is the measured magnetic moment from the Curie-Weiss law, $P_M$ and $P_{\text{Cu}}$ are the moments of the doped atoms and that of Cu, and 0.05 and 0.95 are the relative concentrations of the Fe and Cu. By assuming that the $P_{\text{Cu}}$ is the same as in the pure sample, $P_{\text{Fe}}$ is determined from the Fe doped sample. Values of $\chi$ at 150 K, $x_0$, $T_0$ and $P_M$ of YBa$_2$(Cu$_{0.95}$M$_{0.05}$)$_3$O$_7$ are shown in table III.

It can be drawn that the susceptibility of the high $T_c$ ceramics contains two parts: one from the localized moment, represented by $P_{\text{eff}}$, the other from the energy band structure (e.g. Pauli paramagnetism) represented by $x_0$. $P_{\text{Cu}}$ in table III is much smaller than the moment of free Cu$^{2+}$ and Cu$^{3+}$. This suggests that the electrons of Cu$^{2+}$ in the YBa$_2$Cu$_3$O$_7$ can not be handled as localized ones, and that the local magnetic moment have an negative effect on superconductivity. This agrees with our experiment showing that the $T_c$ of Fe substituted sample which has smaller $x_0$ and larger localized moment is much lower than that of the pure sample.