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# The paramagnetic contribution in the magnetization behavior of $Y_{1-x}Gd_xBa_2Cu_3O_7$

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#### Abstract

Polycrystalline samples of  $Y_{1-x}Gd_xBa_2Cu_3O_7$  with different Gd contents (x = 1, 0.75, 0.5, 0.25) were prepared using the usual solid state reaction method. X-ray characterization of these samples showed an orthorhombic phase. The lattice parameters *a*, *b*, and *c* were found to increase with increasing Gd concentration which is due to the larger volume of Gd atoms. Magnetization and transport measurements show the same transition temperature of 91 K for all samples studied. Initial and magnetization hysteresis loop measurements showed that the paramagnetic behavior is dominant at high fields. © 2002 Elsevier Science B.V. All rights reserved.

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### 1. Introduction:

Several attempts have been made to study the effects of substituting other metals for Cu [1–5] in different superconducting high  $T_c$  materials such as YBCO. It was found that the effect of transition metals will generally reduce the transition temperature. Xiao et al. [1] found that there is a strong correlation between the superconducting transition temperature and the magnetic and electronic characteristics of the 3d element alloyed with

Ba<sub>2</sub>Cu<sub>3</sub>O<sub>6+y</sub> system. Tarascon et al. [2] studied the YBa<sub>2</sub>Cu<sub>3-x</sub>M<sub>x</sub>O<sub>z-y</sub>, where M = Ni, Zn, Fe, Co and Al and showed that there is a depression of  $T_c$ with increasing doping element concentration x irrespective of the doping ion is magnetic (Co, Fe, or Ni) or diamagnetic (Al or Zn). Others studied the effects of substituting rare earth elements such as Gd in YBCO system [6–7]. In the rare earth substitution, the transition temperature remains either the same or moves to lower value. Usually a magnetic transition to ferromagnetic or antiferromagnetic phase will occur. This leads to the conclusion that the magnetic atoms act against superconducting state [5].

In this work we will focus on the effect of increasing the content of Gd atoms in the YBCO

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superconductors by studying the crystal parameters and the different magnetic properties of these samples below  $T_c$ .

#### 2. Samples preparation and experimental results

For this study we have prepared samples with the form  $Y_{1-x}Gd_xBa_2Cu_3O_7$  using the usual solid state reaction method. We used high purity (~99.99%) powders of  $Y_2O_3$ , BaCO<sub>3</sub>, CuO and  $Gd_2O_3$ . The powders were mixed in agate mortar for about 20 min, pressed, and then heated to 900°C for about 24 h under atmosphere in an oven that allows the ventilation of CO<sub>2</sub>. The samples were furnace cooled to room temperature, pressed, then reannealed at 900°C for other 24 h in oxygen gas. They were cooled down to 400°C in oxygen flow where they kept for about 24 h. Finally, the samples were furnace cooled to room temperature.

Four samples were prepared using this procedure with different amounts of Gd content (x), where x = 1, 0.75, 0.50, and 0.25. We call these samples, respectively, as Gd(1), Gd(2), Gd(3), and Gd(4). Parts of these samples were cut into a rectangular shape to perform the magnetization measurements.

Our susceptibility measurements show the same transition temperature for the four samples, viz.  $T_c = 91 \text{ K}$ . A confirmation of this also has been done using a four-probe technique by using a sensitive nanovoltmeter (Keithley 182).

The X-ray diffraction patterns of these samples were obtained using a JEOL JDX-3530 X-ray diffractometer employing a Cu K<sub> $\alpha$ </sub> X-ray tube operating at 40 kV and 30 mA. Analysis of these patterns showed that all samples have the orthorhombic phase. The calculated lattice parameters for the four samples are shown in Table 1. The

Table 1 Lattice parameters for the four  $Y_{1-x}Gd_xBa_2Cu_3O_7$  samples

Gd concentration	a (Å)	b (Å)	c (Å)	V (Å <sup>3</sup> )
1	3.8958	3.8466	11.6794	175.02
0.75	3.8947	3.8350	11.6992	174.74
0.50	3.8885	3.8243	11.687	173.79
0.25	3.8856	3.8185	11.6804	173.30

data show gradual increase in the lattice parameters with increasing Gd content. This increase is due to the larger size of atoms of Gd that substitute Y atoms.

The four rectangular shape samples were used for the magnetization measurements. We used an Oxford vibrating sample magnetometer (VSM). As an example, a typical hysteresis loop measurement is shown in Fig. 1 for x = 1. Results for x =0.25, 0.5 and 0.75 show similar behavior. It is evident from the plots that at high fields no typical behavior exists where the paramagnetic contribution becomes dominant. This paramagnetic effect is due to the Gd atoms introduced instead of Y atoms. In order to investigate and take care of the paramagnetic contribution, data were obtained at higher temperatures above  $T_c$ . In Fig. 2 we plot M versus H for x = 1 at T = 95 K which is just above  $T_{\rm c}$ . The data is a straight line and its slope can be used to correct the data below  $T_{\rm c}$  for this sample. Similar behavior was obtained for the other samples but with different slopes. Since the susceptibility has a gradual temperature dependence between T = 80 and 95 K, we can correct the data at 80 K using the paramagnetic contribution at 95 K [7].

In Fig. 3 we have replotted the data of Fig. 1 after subtracting the paramagnetic correction. It is clear from Fig. 3 that the behavior of M versus H is typical of the type II superconductors.

The temperature dependence of the paramagnetic contribution has been investigated at other



Fig. 1. Magnetization (*M*) versus the applied field (*H*) for x = 1 at 80 K.



Fig. 2. Magnetization (*M*) versus the applied field (*H*) for x = 1 at T = 95 K.



Fig. 3. Corrected magnetization (*M*) versus the applied field for x = 1 at T = 80 K.

temperatures. Therefore, in Fig. 4 we represent our results for x = 1 of M versus H at different temperatures. The behavior of M versus H is linear for all temperatures. The slope gives the susceptibility  $\chi(T)$  at each temperature. Similar results were obtained for the other samples under investigation.

Plotting the inverse of the susceptibility versus H shows the paramagnetic behavior of these materials, as shown in Fig. 5. The plots show a Curie law behavior since  $\chi^{-1}(T)$  passes through the origin. From these data we cannot detect any antiferromagnetic phase by extending  $\chi^{-1}(T)$  to



Fig. 4. Magnetization (M) versus the applied field (H) at different temperatures for x = 1.0.



Fig. 5. The inverse susceptibility versus the temperature (T) for x = 1.0.

low *T*. However, the existence of anti-ferromagnetic phase has been reported when one substitutes a rare-earth metal in YBCO [8]. Therefore, in these materials only paramagnetic and superconductiong phase exists below  $T_c$  with paramagnetic contribution showing gradual temperature dependence as seen from Fig. 5.

## 3. Conclusions

The effect of Gd substitution in YBCO polycrystalline stabilizes an orthorhombic phase while the lattice parameters are found to increase due to a larger size of the Gd atoms than Y atoms. There is no effect in the transition temperature regardless of the concentration of Gd atoms introduced in YBCO. The magnetization measurements of M versus H show a paramagnetic behavior existing at high fields. Data were corrected to eliminate the paramagnetic contribution below  $T_c$ . More investigation of the paramagnetic phase has been carried out at temperatures above  $T_c$  where all samples show a Curie type behavior. The susceptibility is found to increase with increasing Gd concentration.

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