Effects of Gd/Ba Nonstoichiometry in Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ on Superconducting and Magnetic Properties

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Effects of slight nonstoichiometry in Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ compounds on superconducting and magnetic properties were studied. The series of single-phase samples of Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ with composition deviation $x$ from the stoichiometric value of 0 to 0.1 and to $-0.1$ were synthesized by the solid-state reaction method from Gd$_2$O$_3$, BaCO$_3$ and CuO precursors, sintered at the temperature of $\approx 1000$ $^\circ$C in flowing oxygen and annealed at 450 $^\circ$C for 24 h.

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1. Introduction

The Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ compounds belong to e.g. LRE-123 superconductors, in which LRE - light rare earth cations (where LRE = Nd, Sm, Eu and Gd) can form solid solutions with Ba ions. Among the elements, gadolinium has the highest Gennes factor, which indicates on exchange interaction of Gd ions. Moreover, between them, only the total orbital magnetic moment is zero. Thus it is interesting to study the superconducting properties [12] and interactions between localized magnetic spins and conduction electrons [35] as functions of light nonstoichiometry and magnetic field.

2. Experimental

The critical temperature $T_c(R = 0)$ was determined by a standard resistance four-point method. The transition width, $\Delta T_c$, was characterized by the 10-90% criterion. The phase composition was studied by X-ray diffraction measurements (CuK$_\alpha$ radiation). AC low field magnetization at 77 K was measured by a compensation method using the second-order SQUID gradiometer [6]. The temperature dependences of the zero-field cooled (ZFC) and field cooled (FC) DC magnetic moment of samples were measured in the Quantum Design SQUID magnetometer MPMS XL-7 at field of 1.6 kA m$^{-1}$ and 5.6 MA m$^{-1}$. The corresponding temperature dependences of ZFC molar (per Gd atom) susceptibility $\chi$ at low and high field was determined and fitted to the Curie-Weiss law. The values of the Néel temperature $T_N$ were estimated from the maximum of molar susceptibility.

3. Results and discussion

From X-ray diffraction data, it can be concluded that all samples are single-phase, except the one with $x = -0.1$, where the observed weak peaks could be ascribed to an excess of Ba-Cu-O phase. Figure 1 shows a positive effect of Ba-excess on $T_c$ and $\Delta T_c$.

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Fig. 1. $T_c$ vs. $x$ for Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$. The inset shows $\Delta T_c$ vs. $x$.

Fig. 2. $M$ vs. $H$ dependences of Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ at 77 K, for $x \geq 0$.

The hysteresis curves of mass magnetization $M$ vs. applied field $H$ for Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ at 77 K and low applied field are shown in Fig. 2 and 3. Figures 4 and 5 show ZFC molar susceptibility $\chi$ vs. $T$ for the Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ samples at 5.6 MA m$^{-1}$ and 1.6 kA m$^{-1}$, respectively. The inset in Fig. 4 shows the
4. Conclusions

The nonstoichiometric Gd$_{1+x}$Ba$_{2-x}$Cu$_3$O$_{7-\delta}$ samples with 0.1 $\leq$ $x$ $\leq$ 0.07 show critical temperature $T_c$ over 92 K and $\Delta T_c$ about 1 K up to almost $|x|$ $\leq$ 0.04. The excess of Ba has a positive effect on superconducting properties at 77 K. We have found that the change of the applied magnetic field from 5.6 MA m$^{-1}$ to 1.6 kA m$^{-1}$ results in a weak change of magnetic moment $\mu_{eff}$ and an order of magnitude increase in negative Weiss temperature $\Theta$. The Gd (Ba)-excess increases (decreases) the $\mu_{eff}$. The results indicate the role of exchange interaction of Gd ions and effect of structure disorder with increasing $x$. The highest Néel temperature $T_N$ = 4 K was estimated for the stoichiometric sample.

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References


