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Effects of Gd/Ba Nonstoichiometry in $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ on Superconducting and Magnetic Properties

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Effects of slight nonstoichiometry in $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ compounds on superconducting and magnetic properties were studied. The series of single-phase samples of $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ 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_2O_3, $BaCO_3$ and CuO precursors, sintered at the temperature of ∼ 1000 °C in flowing oxygen and annealed at 450 °C for 24 h.

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1. Introduction

The $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ 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 [1–2] and interactions between localized magnetic spins and conduction electrons [3–5] 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, ΔT_c, was characterized by the 10-90% criterion. The phase composition was studied by X-ray diffraction measurements (CuK_α 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 χ 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 ΔT_c.

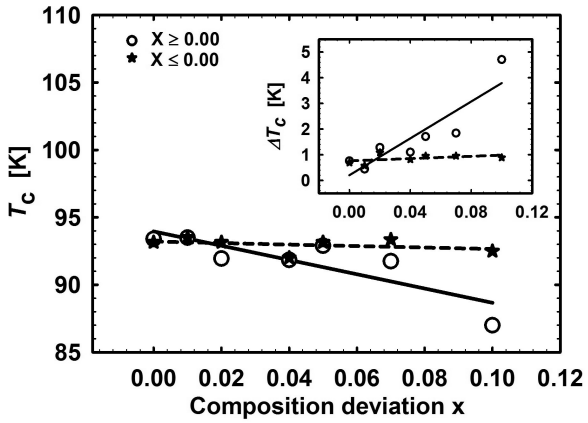

Fig. 1. T_c vs. x for $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$. The inset shows ΔT_c vs. x.

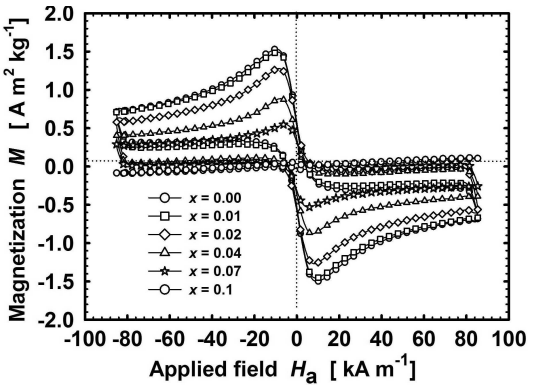

Fig. 2. M vs. H dependences of $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ at 77 K, for $x \geq 0$.

The hysteresis curves of mass magnetization M vs. applied field H for $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ at 77 K and low applied field are shown in Fig. 2 and 3. Figures 4 and 5 show ZFC molar susceptibility χ vs. T for the $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ samples at 5.6 MA·m^{-1} and 1.6 kA·m^{-1}, respectively. The inset in Fig. 4 shows the

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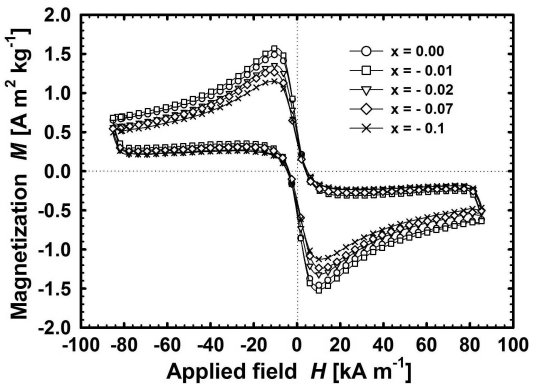

Fig. 3. M vs. H dependences of $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ at 77 K, for $x \leq 0$.

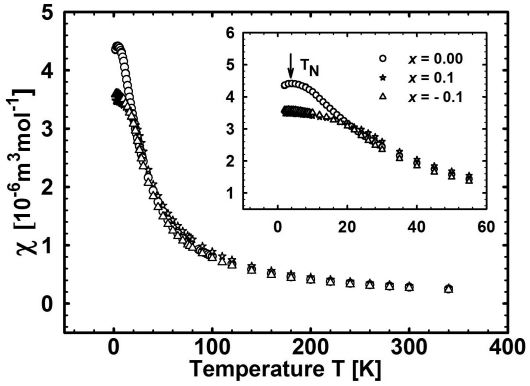

Fig. 4. ZFC χ vs. T of the $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ at 5.6 MA·m^{-1}. The inset shows the enlarged view on magnetic phase transition.

enlarged view of magnetic phase transition for $x = 0.00$ and the one in Fig. 5 shows χ^{-1} vs. T. The solid line represents the fit of the Curie-Weiss law. The effective magnetic moment μ_{eff} for $x \geq 0$ increases from 7.33 to 7.72 μ_B with increasing x at applied field of 5.6 MA m^{-1}. The linear fit starts from the highest T and ends at about 20 K. The Weiss temperature Θ ranges from –5.76 to –4.27 K. For $x < 0$, μ_{eff} shows an opposite tendency of the decrease, with increasing x, from 7.40 to 7.11 μ_B and Θ changes between –5.76 and –5.26 K. At low applied field μ_{eff} shows the same behavior, it increases with increasing x from 7.49 to 7.89 μ_B for $x \geq 0$ and decreases with x, from 7.50 to 7.27 K, for $x < 0$, while negative Θ is by more than an order higher, Θ changes from –56 K up to –99 K. However, the linear fit of the inverse susceptibility could not be used for $T < T_c$.

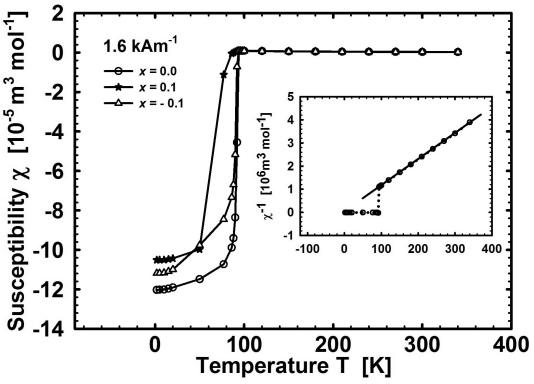

Fig. 5. ZFC χ vs. T of the $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ at 1.6 kA·m^{-1}. The inset shows χ^{-1} vs. T for $x = 0.00$. The solid line shows the fit of the Curie-Weiss law.

4. Conclusions

The nonstoichiometric $Gd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ samples with $0.1 \leq x \leq 0.07$ show critical temperature T_c over 92 K and ΔT_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 μ_{eff} and over an order of magnitude increase in negative Weiss temperature Θ. The Gd (Ba)-excess increases (decreases) the μ_{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.

Acknowledgments

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