Anisotropic magnetic behavior of GdBa$_2$Cu$_3$O$_{6+y}$ single crystals

Vladimir N. Narozhnyi$^{a, b, 1}$, Dieter Eckert$^a$, Günter Fuchs$^a$, Vladimir Nekvasil$^c$, Karl-Hartmut Müller$^a$

$^a$Leibniz-Institut für Festkörper- und Werkstoffforschung Dresden, PO Box 270116, D-01171 Dresden, Germany
$^b$Institute for High Pressure Physics, Russian Academy of Sciences, Troitsk, Moscow Region, 142190, Russia
$^c$Institute of Physics, Czech Academy of Sciences, Cukrovarnická 16, 16253 Praha 6, Czech Republic

Abstract

Magnetic properties of high-quality Al-free nonsuperconducting GdBa$_2$Cu$_3$O$_{6+y}$ single crystals grown by flux method have been studied. The magnetic anisotropy below the Néel temperature $T_N \approx 2.3$ K corresponds to the direction of Gd$^{3+}$ magnetic moments along the tetragonal $c$-axis. At $T < T_N$ clear indications of spin-flop transitions for $H \parallel c$ have been observed on magnetization curves at $H_{sf} \approx 10$ kOe. Magnetic phase diagrams have been obtained for $H \parallel c$ as well as for $H \perp c$. A pronounced anisotropy in the magnetic susceptibility (unexpected for Gd-based compounds) has been found above $T_N$.

Key words: GdBa$_2$Cu$_3$O$_{6+y}$; magnetic anisotropy; spin-flop transition; single crystal

Collinear antiferromagnetic ordering was found for GdBa$_2$Cu$_3$O$_{6+y}$ (Gd1236) with the Gd magnetic moments directed along the $c$-axis [1,2]. So far magnetism in Gd1236 single crystals were studied on Al-containing samples [3] or by indirect methods (e.g., NMR [4]). However, the reported results are controversial.

In this work we report on the magnetic properties of high quality Al-free nonsuperconducting Gd1236 single crystals grown in Pt crucibles by the flux method [5]. Atomic absorption spectroscopy has shown that the Pt contamination does not exceed $3 \cdot 10^{-3}$ at. % [6]. To reduce the oxygen concentration the samples were annealed at $T = 600$ C under high vacuum during 4 days. The magnetization $M$ was measured by SQUID magnetometer. The data are compared with the results obtained earlier for PrBa$_2$Cu$_3$O$_{7-y}$ (Pr123) [7] as well as for the Gd-sublattice of mixed Gd$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-y}$ [(Gd-Pr)123] single crystals [6].

The temperature dependence of susceptibility $\chi = M/H$, shown in Fig. 1 for two directions of the magnetic field $H$, has a maximum at $T \approx T_N$. The position of the maximum is field dependent. The anomaly is more pronounced for $H \parallel c$ (at least for small fields), which corresponds to the $c$-axis direction of the Gd$^{3+}$ magnetic moments (in accord with neutron diffraction and Mössbauer spectrometry [1,2]). The maximum in
\( \chi(T) \) disappears for \( H \geq 15 \text{ kOe} \) at \( T \geq 1.8 \text{ K} \).

The \( \chi^{-1}(T) \) curves can be well fitted to the Curie-Weiss law \( \chi^{-1}(T) = (T - \theta)/C \) for \( 5 \leq T \leq 300 \text{ K} \). The values of \( \theta \) and the effective magnetic moment \( m_{\text{eff}} \) (determined from the Curie constants \( C \)) are -4.6 K and 8.90 \( m_B \) and -1.8 K and 8.88 \( m_B \) for \( H \parallel c \) and \( H \perp c \), respectively. \( m_{\text{eff}} \) and \( m_{\text{ab}} \) are very close to each other as expected for the spin-only magnetic moment of Gd. The values of \( m_{\text{eff}} \) are little higher than the value of the free Gd\(^{3+} \) ion. This difference may be connected with some error in determination of the small mass of the sample (\( m = 0.50 \text{ mg} \); the accuracy of mass determination in this case is about 10%).

The anisotropy in \( \chi \) in paramagnetic state is clearly seen in this figure. Phenomenologically the observed anisotropy can be connected with the difference between \( \theta_c \) and \( \theta_{ab} \). (From the fit the accuracy in \( \theta \) determination is better than 0.1 K.) It is found that \( \theta_c > |\theta_{ab}| \). The sign of magnetic anisotropy for Gd1236 is the opposite to the observed for Pr123 [6,7]. The different signs of magnetic anisotropy for Gd- and Pr-sublattices explain the crossover of magnetic anisotropy reported for (Gd-Pr)123 single crystals [6].

\( M(H) \) curves clearly show an anisotropy below as well as above \( T_N \), see Fig. 2. At lowest \( T \) there is a clear tendency for isotropization in high fields. \( M_{ab} > M_c \) is in accord with our data obtained earlier from subtraction of (Y-Pr)123 data from (Gd-Pr)123 results [6]. The general agreement with these previously indirectly obtained results on the magnetization of the Gd-sublattice is fair. Even the values and the anisotropy of \( \theta \) correspond well to the directly measured for Gd1236.

Below \( T_N \), a distinct indication of a spin-flop transition can be seen for \( H \parallel c \), see Fig. 2. The spin-flop field \( H_{sf} \) was determined as an inflection point of the \( M(H) \) dependence at \( T < T_N \). \( H_{sf} \) is practically temperature independent in the studied temperature interval, see Fig. 3. As expected, the spin-flop transition disappears above \( T_N \). No anomaly has been detected for \( H \perp c \) at all \( T \).

A magnetic phase diagram is constructed from the field dependence of \( T_N \), determined for two directions of \( H \) (see Fig. 3). The field dependence of \( T_N \) for \( H \perp c \) is described by a quadratic dependence similar to that observed by us earlier for Pr123 [7]. Below \( H_{sf} \) the \( T_N(H) \) dependence for \( H \parallel c \) is weaker than for \( H \perp c \). At the same time above \( H_{sf} \) the \( T_N(H) \) dependencies are close for both directions of \( H \).

The pronounced magnetic anisotropy found for Gd1236 may be connected with several mechanisms including: (i) dipole-dipole interaction of the Gd ions; (ii) interaction between Gd and Cu sublattices; (iii) anisotropic exchange interaction; (iv) crystal-field effects on the excited Gd\(^{3+} \) states. Further investigations are necessary to clarify the situation.

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