Magnetoresistance of Pr\textsubscript{6} and GdB\textsubscript{6}

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Abstract. The comprehensive study of transverse magnetoresistance (MR) has been carried out on the high quality single crystals of PrB\textsubscript{6} (\(T_N\approx6.7\)K) and GdB\textsubscript{6} (\(T_N\approx15.5\)K) in the wide range of temperatures 2-40K and magnetic fields up to 8T. The data obtained allow to establish the crossover of MR from negative (\(T>T_N\)) to positive (\(T<T_N\)) regime. The analysis of the curves \(\Delta \rho (H)/\rho\) allows to separate three contributions to MR of RB\textsubscript{6} (R=Pr, Gd). In addition to the (i) negative contribution (~\(H^2\)) interpreted in the framework of Yosida model, (ii) a linear (~\(H\)) and (iii) nonlinear saturated components were also observed. In the framework of approach developed the (ii), (iii) components should be ascribed to the ferromagnetic nanodomains (spin-polarized 5\textit{d}-states) embedded in the metallic matrix of RB\textsubscript{6} (R=Pr, Gd).

1. Introduction

The rare earth (RE) hexaborides (RB\textsubscript{6}) are typical correlated electron systems where the spin, charge and orbital degrees of freedom are at play. These compounds exhibit many interesting properties in their ordered states. It is generally believed that the formation of the ground state in the light hexaborides RB\textsubscript{6} (R-Ce, Pr, Nd) is governed by the multipole interactions resulting from the strong spin-orbit coupling (see [1] for details). In particular CeB\textsubscript{6} is known as proceeding of complicated magnetic phase diagram which is explained in terms of coexistence of \(O_{xy}\)-type antiferroquadrupolar (AFQ), \(T_{xy}\)-AFM-octupole and AFM interaction [2]. For PrB\textsubscript{6} the situation is simplified because there is no octupole moment due to \(J_3\) triplet ground state (see the inset in Fig.1a). Therefore it is believed that the only \(O_{xy}\)-type AFQ and AFM interaction responsible for very anisotropic view of H-T magnetic phase diagram of this compound [4]. Finally NdB\textsubscript{6} is also regarded as the system with \(O_{z\gamma}\)-type ferroquadrupolar interaction [1]. These speculations raise the question about the role of multipolar interaction for RB\textsubscript{6} series (see [5] for details). Recently neutron scattering experiments on GdB\textsubscript{6} allowed to obtain the magnetic structure with wave vector \(k=(1/4, 1/4, 1/2)\) below \(T_N\approx15\)K [6]. The same AFM structure was previously observed in CeB\textsubscript{6} below 2.4K (see [7] for details), PrB\textsubscript{6} below \(T_N\approx4.2\)K [8], and DyB\textsubscript{6} at \(T<T_N\approx26\)K [9]. However GdB\textsubscript{6} is considered as the system without orbital degrees of freedom (\(L=0, S=7/2\)) since the trivalent Gd\textsuperscript{3+} ion has half filled 4\textit{f}-shell. In this situation the comparison of the compounds in the series RB\textsubscript{6} should be promising both to testify the universality of magnetic structure and to shed a light on the mechanisms responsible for the magnetic ordering.

In present work Pr\textsubscript{6} and GdB\textsubscript{6} have been studied for comparison. In the absence of magnetic field these systems are characterized by two successive AFM transitions: incommensurate (IC1) and
Figure 1. Temperature behavior of resistivity $\rho(T)$ measured for (a) PrB$_6$ and (b) GdB$_6$ at magnetic field $\mu_0 H=0$T and 8T oriented along the directions $<111>$ and $<110>$ respectively. Panel (a) also represents the crystal field level scheme for PrB$_6$ [3]. The insets show the temperature dependences of the magnetoresistance $\Delta \rho(T)/\rho$ at $\mu_0 H=8$T.

Figure 2. Transverse magnetoresistance $\Delta \rho(H)/\rho$ as a function of external magnetic field for PrB$_6$ (a, b) and for GdB$_6$ (c, d) in C (a, c) and PM (b, d) states respectively. Solid lines on panels (a, c) represent the linear approximation (see the text). The curves $\Delta \rho(H)/\rho$ are shifted from each other for convenience.

2. Experimental details

The detailed investigation of transverse magnetoresistance has been carried out on the high quality single crystals of PrB$_6$ ($T_N$$\approx$6.7K) and GdB$_6$ ($T_N$~15.5K) in the wide range of temperatures 2-40K in magnetic fields up to 8T, oriented along the directions $<111>$ and $<110>$ respectively (with current configuration $I||<110>$). The single crystals RB$_6$ (R-Pr, Gd) were grown by the crucible-free inductive zone melting in the argon gas atmosphere. The experiments were carried out with the PrB$_6$ oriented single crystal annealed in vacuum at $T_{an}$$\approx1700$°C during 10 hours in charging of amorphous boron powder. The single crystal quality of the samples was controlled by Laue back patterns, purity – by optic spectral analysis. The original experimental setups presented previously in [11,12] allow to obtain the high accuracy of transport measurements $\sim$0.05-0.1%.
compounds: heavy-fermion system CeCoIn$_3$. The positive magnetoresistance with a noticeable amplitude (IC phase of chromium which is known to be a spin-density wave (SDW) antiferromagnet [16]. In our previous investigations [10,13]. The transition to AFM state at $T_N$ is clearly observed as a rapid drop of the resistivity $\rho(T)$. Another one transition to C phase for PrB$_6$ and AFM(II) state for GdB$_6$ is indicated below $T_{CIC1}=4.6K$ and $T^*≈4.7K$ respectively. The value of $T^*$ obtained from the complex investigation of magnetoresistance (see the inset on Fig.1b) is lower in comparison with the results $T^*≈5-10K$ [6, 10]. This fact could be explained by the better quality of investigated single crystals leading to the decrease of the temperature $T^*$.

The positive MR with maximal amplitude $\Delta\rho(PrB_6)/\rho≈58\%$, $\Delta\rho(GdB_6)/\rho≈12\%$ is observed in AFM phase of PrB$_6$ ($T<T_N$) and AFM(I) state ($T^*<T<T_N$) of GdB$_6$ at $\mu_0H=8T$ (see Fig.1 and insets). Taking into account that the magnetic scattering is described by de Gennes factor $G=(g−1)^2J(J+1)$ which is strongly renormalized with 4f-shell occupation ($G(PrB_6)$=0.8, $G(GdB_6)$=15.75) one can explain the drastic decrease of MR amplitude for GdB$_6$. The behavior of magnetoresistance in AFM (II) phase of GdB$_6$ will be analyzed elsewhere. The increasing of the temperature above $T_N$ is accompanied by the crossover to negative MR regime with the small amplitude $\Delta\rho/\rho≤3\%$ for both compounds (see insets on Fig.1). At the same time the rapid increase of magnetoresistance is detected for GdB$_6$ in the vicinity of Néel temperature ($15.3K< T<T_N$ see inset on Fig.1).

2. The field dependences of magnetoresistance $\Delta\rho(H)/\rho$ are presented for PrB$_6$ and GdB$_6$ on Fig.2a-b and Fig.2c-d correspondingly. We found that the character of the magnetoresistance is the similar in C and PM states of studied compounds. In particular in C phase the curves $\Delta\rho/\rho=f(H, T_\phi)$ may be analyzed by the positive linear component ($\Delta\rho/\rho=AH$) and by nonlinear saturated magnetic ($\Delta\rho/\rho=D$) contribution (Fig.2a, 2c). The procedure of MR analysis below $T_N$ is presented for convenience on Fig.3. On the contrary the negative magnetoresistance in PM phase of PrB$_6$ and GdB$_6$ is described by the quadratic term $\Delta\rho/\rho=BH^2$, $B<0$ (Fig.2b, 2d) which may be interpreted in the framework of Yosida theory [14]. This model describes the scattering of charge carries on localized magnetic moments (LMM) by the relationship between the MR and local magnetization $M_{loc}$.

$$\Delta\rho/\rho \sim M_{loc}^2$$

This phenomenological procedure was previously applied for MR analysis in antiferromagnetics CeB$_6$, PrB$_6$, NdB$_6$ [7,11]. The appearance of positive magnetoresistance was revealed in various compounds: heavy-fermion system CeCoIn$_5$, ferromagnetic CeRu$_2$Ge$_2$ [15] etc. Moreover the effect of positive magnetoresistance with a noticeable amplitude ($\Delta\rho/\rho=180\%$ at $\mu_0H=1.2T$) was also detected in IC phase of chromium which is known to be a spin-density wave (SDW) antiferromagnet [16]. In our opinion the origin of positive magnetoresistance in AFM states of RB$_6$ (R-Pr, Gd) may be attributed to the features of charge carrier scattering on the complex magnetic structure with LMM of (RE) ion (4f-component) and SDW (5d-component).

3. Results and discussion

1. The temperature dependences of electrical resistivity at $\mu_0H=0T$, 8T are presented for PrB$_6$ and GdB$_6$ on Fig.1. The data obtained demonstrate metallic behavior in agreement with the results of previous investigations [10,13]. The transition to AFM state at $T_N$ is clearly observed as a rapid drop of the resistivity $\rho(T)$. Another one transition to C phase for PrB$_6$ and AFM(II) state for GdB$_6$ is indicated below $T_{CIC1}=4.6K$ and $T^*≈4.7K$ respectively. The value of $T^*$ obtained from the complex investigation of magnetoresistance (see the inset on Fig.1b) is lower in comparison with the results $T^*≈5-10K$ [6, 10]. This fact could be explained by the better quality of investigated single crystals leading to the decrease of the temperature $T^*$.

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$$\Delta\rho/\rho \sim M_{loc}^2$$

Figure 3. The analysis of the contributions to MR in (a) C state of PrB$_6$, (b) AFM(I) phase of GdB$_6$.

The function $L(\alpha)$ is defined by Eq.2.
3. The appearance of the saturated magnetic term in MR may be explained by the formation of magnetic polarization in 5d-conduction band. In particular the analysis of the dependencies \( D(H, T_0) \) by Langeven function (see the solid lines on Fig.3)

\[
D(H, T_0) = -L(\alpha) = \text{cth}(\alpha) - 1/\alpha
\]

(2),

(where \( \alpha = \mu H/k_B T \)) allows to estimate the effective magnetic moment \( \mu(T) \). The relation (2) describes the curves \( D(H, T_0) \) more accurately in comparison with Brillouin function. The results obtained from Eq.2 are presented on Fig.4 for PrB6 and GdB6. It is seen from Fig.4 the values \( \mu(\text{PrB}_6) \approx 1.7\mu_B \) and \( \mu(\text{GdB}_6) \approx 5.5\mu_B \) determined at \( T_N \) do not coincide with magnetic moment of the ground state \( \mu_{\gamma} \approx 2\mu_B \) for Pr\(^{3+} \) and \( \mu = 8\mu_B \) for Gd\(^{3+} \). In our opinion such a discrepancy indicates on the formation of magnetic nanoclusters involving the LMM of RE ion and spin polarized 5d-states. The strongly correlated electron complexes including spin polarons and exciton polarons were previously detected in CeB\(_6\) [7], EuB\(_6\) [17] and SmB\(_6\) [18]. It was estimated that the characteristic spatial size of the many-body states or localization radius \( a_{sp}(\text{CeB}_6) \approx 5.4\text{Å}, a_{sp}(\text{SmB}_6) \approx 6\text{Å} \) is comparable with the lattice constant. 

The universal behavior of MR detected in C and PM phases of PrB\(_6\) and GdB\(_6\) allows to question the role of multipole interactions in the formation of ground state in RB\(_6\) (R= Ce, Pr, Nd, Gd). Taking into account similar magnetic ordering registered for C phase of CeB\(_6\), PrB\(_6\), GdB\(_6\), DyB\(_6\) we propose the alternative interpretation based on the LMM+SDW magnetic structure formation in PrB\(_6\) and GdB\(_6\).

In summary, the high precision measurements of magnetoresistance of PrB\(_6\) and GdB\(_6\) have been carried out at temperatures 2-40K in magnetic field up to 8T. The results and undertaken analysis allow to conclude in favour of the concurrence between AFM and FM interactions as the reason of magnetic structure formation in RB\(_6\) (R= Pr, Gd). Detailed analysis of MR for GdB\(_6\) will be presented elsewhere.

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