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Pressure effects on magnetic properties and electronic structure of EuB_6 and GdB_6

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ABSTRACT

Experimental and theoretical studies of the pressure effects on magnetic properties and electronic structure of EuB_6 and GdB_6 compounds were carried out to shed light on the nature of their magnetic ordering. The magnetic susceptibility was measured under pressure up to 2 kbar at fixed temperatures of 78 and 300 K for the EuB_6 and GdB_6 compounds, and also for the carbon doped EuB_6, for comparison. The observed behavior of the paramagnetic Curie temperature Θ under pressure was analyzed within the RKKY-like approach based on the results of *ab initio* electronic structure calculations.

1. Introduction

The divalent EuB_6 and trivalent GdB_6 compounds with a cubic CaB_6-type crystal structure possess the same f-shell configuration (f^7), in which the rare earth (RE) ions are in the S state ($L = 0, S = 7/2$). However, these compounds demonstrate rather different magnetic and electronic properties that is presumably due to the difference in the number of conduction electrons. GdB_6 is known to have at least two successive magnetic transitions occurring at $T_1 \simeq 15$ K and $T_2 \simeq 10$ K, both believed to be into antiferromagnetic (AFM) phases [1,2]. It should be noted that $T_1 \simeq 15$ K is rather small compared to magnetic transitions temperatures of other heavy rare earth hexaborides ($T_N = 19.5$ K and 25.6 K for TbB_6 and DyB_6, respectively [3]). The magnetic structures of these phases, however, are still little studied by a conventional method of neutron scattering due to the high neutron absorption by both Gd and B. It makes difficult to understand the interplay between charge distribution and magnetic order. In addition, for the low temperature AFM phase, the magnetization and Hall-effect data have revealed the formation of magnetic domains [4].

In contrast to GdB_6, the semimetallic EuB_6 is the only RE hexaboride which orders ferromagnetically. It exhibits two consecutive magnetic transitions at $T_C = 12.5$ K and $T_M = 15.3$ K, where T_C is the bulk Curie temperature [5], and T_M was ascribed to a metallization temperature due to an increase in the number of itinerant electrons [5–7]. Also EuB_6 attracts considerable attention due to the "colossal" magnetoresistance effect [7–9], which presumably originates from a strong interaction of the itinerant charge carriers with the localized magnetic moments of Eu^{2+} ions.

The magnetic ground state is found to change from the ferromagnetic (FM) to AFM in the electron doped $Eu_{1-x}La_xB_6$ [6] and $EuB_{6-x}C_x$ [10,11] compounds with increasing of La (or carbon) concentration. A similar behavior was also observed in the isoelectronic $Eu_{1-x}Ca_xB_6$ alloys [12]. In all these cases, however, the decrease in T_C cannot be unambiguously attributed to behavior of the charge carrier density with doping. On the other hand, EuB_6 has shown a large growth of T_C under pressure P, which is saturated at $P \geq 70$ kbar [13] and ascribed to substantial increase of the conduction electron concentration [13,14].

The electronic structure calculations [15–17] and the de Haas–van Alphen (dHvA) effect measurements [18] have indicated a small overlap of conduction and valence bands in EuB_6 around the X-point of the Brillouin zone. This overlap provides nearly spherical pockets centered at the X-point with rather light effective masses, and corresponding to electron and hole concentrations of about 10^{20} cm^{-3}. It was suggested [13] that carriers in these very small

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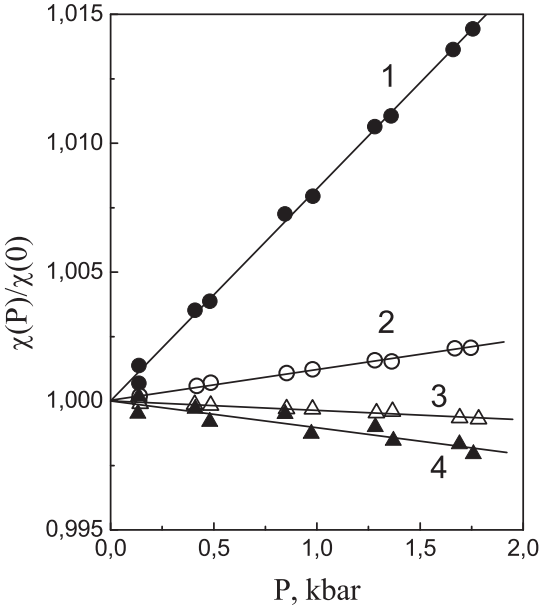

Fig. 1. Pressure dependencies of the magnetic susceptibility for EuB_6 (1,2) and $Eu_{0.97}B_{5.93}C_{0.07}$ (3,4) at $T = 78$ K (solid symbols) and 300 K (open symbols).

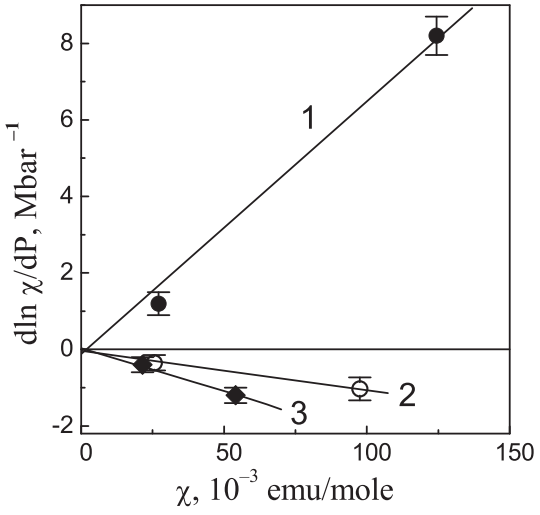

Fig. 2. dln χ/dP versus $\chi(P=0)$ for EuB_6 (1), $Eu_{0.97}B_{5.93}C_{0.07}$ (2) and GdB_6 (3), corresponding to Eq. (1).

pockets could mediate the interaction between magnetic moments of Eu. The peculiar band overlapping in EuB_6 is strongly affected by the non-stoichiometry and structural defects, which have often led to differing and contradictory experimental data and conclusions. Therefore, detailed experimental and theoretical studies are needed to elucidate intrinsic magnetic properties and peculiar electronic structure of the rare earth hexaborides. In this report we are mostly focused on theoretical and experimental studies of the pressure effect on electronic structure and magnetic properties of EuB_6 and GdB_6.

2. Experimental

We studied the temperature and pressure dependencies of the magnetic susceptibility χ for the $Eu_{0.99}B_6$ (further called EuB_6), GdB_6 and carbon doped $Eu_{0.97}B_{5.93}C_{0.07}$ single crystal samples, prepared by a floating zone method. The $\chi(T)$ studies were carried out in the range of 4.2–300 K by the Faraday method in a magnetic field of $H = 0.8$ T, and the Curie–Weiss behavior was revealed for all investigated samples. The pressure dependence of susceptibility was measured under helium gas pressure P up to 2 kbar at two fixed temperatures, $T = 78$ and 300 K, using a pendulum-type magnetometer placed directly into the pressure cell [19]. The relative errors of measurements under pressure did not exceed 0.05% for the employed magnetic field of $H = 1.7$ T. The experimental $\chi(P)$ dependencies were found to be linear (see Fig. 1), yielding the corresponding pressure derivatives dln χ/dP. Based on the Curie–Weiss character of $\chi(T)$, the dln χ/dP derivative is assumed to be predominantly governed by the pressure dependence of the paramagnetic Curie temperature Θ:

$$\frac{\mathrm{d}\ln\chi}{\mathrm{d}P} = \frac{\mathrm{d}\ln C}{\mathrm{d}P} + \frac{1}{(T-\Theta)}\frac{\mathrm{d}\Theta}{\mathrm{d}P} \simeq \frac{\chi}{C}\frac{\mathrm{d}\Theta}{\mathrm{d}P} \quad (1)$$

where the Curie constant C is close to that for the rare earth ion with the f^7 configuration and assumed to be pressure independent.

According to Eq. (1), the values of the pressure derivative dΘ/dP were evaluated from a slope of the dln χ/dP vs. χ dependence in Fig. 2, and presented in Table 1 together with other relevant magnetic parameters. As evident from the dΘ/dP data in Table 1, the EuB_6 compound exhibits the largest pressure effect in comparison with the carbon doped EuB_6 and GdB_6 compounds, where the dΘ/dP values are much lower in magnitude and opposite in sign.

3. Computational details and results

In order to analyze the experimental data on the pressure effects, the volume-dependent band structures of EuB_6 and GdB_6 were calculated *ab initio* for the paramagnetic (PM), FM and AFM phases of EuB_6 and GdB_6. For these calculations we employed a full-potential all-electron relativistic linear muffin-tin orbital method (FP-LMTO, code RSPt [24]). The exchange-correlation potential was treated within the local spin density approximation (LSDA) of the density functional theory (DFT), and the 4f states were considered as spin polarized outer core states with the spin occupation numbers to be fixed by applying the Russel–Saunders coupling scheme to the 4f shell (see Refs. [22,24] for details). Due to the half-filled 4f shells of EuB_6 and GdB_6, the Hund's rule ground state is $^8S7/2$ which provides an isotropic magnetic moment unaffected by the crystal fields. In fact, this approach appeared to be consistent with the observed effective magnetic moments of these compounds. Also, the calculated basic features of electronic structures of EuB_6 and GdB_6 are in a qualitative agreement with results of earlier DFT calculations [15–17,23].

The cubic crystal structure of the compounds is the CaB_6-type which can be described as a simple CsCl lattice, where RE atoms occupy the Cs sites, whereas B_6 octahedrons are positioned at the Cl sites. The lattice parameters a are 4.181 Åand 4.101 Åat $T \simeq 10$ K for EuB_6 and GdB_6, respectively [25]. The internal position parameter x, which determines the ratio between intra- and inter-octahedral B–B distances ($d_{\mathrm{intra}} = a\sqrt{2}/2(1-2x)$ and $d_{\mathrm{inter}} = 2xa$) is chosen to be $x = 0.207$ that corresponds to $d_{\mathrm{intra}} = d_{\mathrm{inter}}$ [15]. The volume dependent band structures and total energies were calculated for the fixed values of parameter x and the ratio of atomic sphere radii of Eu(Gd) and B.

Table 1
Magnetic susceptibility χ (10^{-3} emu/mol) and pressure derivative dln χ/dP(Mbar^{-1}) at $T = 78$ and 300 K, effective magnetic moment μ_{eff} (μ_B/Eu(Gd)), paramagnetic Curie temperature Θ (K) and its pressure derivative dΘ/dP (K/kbar) for hexaborides studied.

Parameter	$Eu_{0.99}B_6$	$Eu_{0.97}B_{5.93}C_{0.07}$	GdB_6
χ (78 K)	124.4	97.6	54.1
(300 K)	27.1	25.6	21.5
dln χ/dP (78 K)	8.2 ± 0.5	-1.0 ± 0.3	-1.2 ± 0.2
(300 K)	1.2 ± 0.3	-0.3 ± 0.2	-0.4 ± 0.2
μ_{eff}	7.85	7.88	7.98
Θ	16 ± 1	-4	-65
dΘ/dP	0.52 ± 0.04	-0.08 ± 0.03	-0.17 ± 0.03
dln Θ/dln V [a]	-55 ± 7	-34 ± 13	-4.4 ± 1

[a] To estimate dln Θ/dln $V = -B$dln Θ/dP, we used the experimental bulk modulus value, which is the very same for both EuB_6 and GdB_6 ($B \simeq 1.7$ Mbar, see Refs. [20–22]).

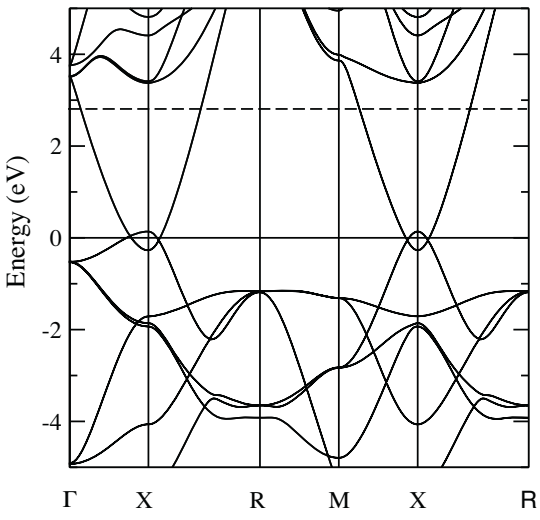

Fig. 3. Band structure of paramagnetic EuB_6. The Fermi level is set to zero and indicated by the horizontal solid line. The dashed line corresponds to the Fermi energy of PM GdB_6.

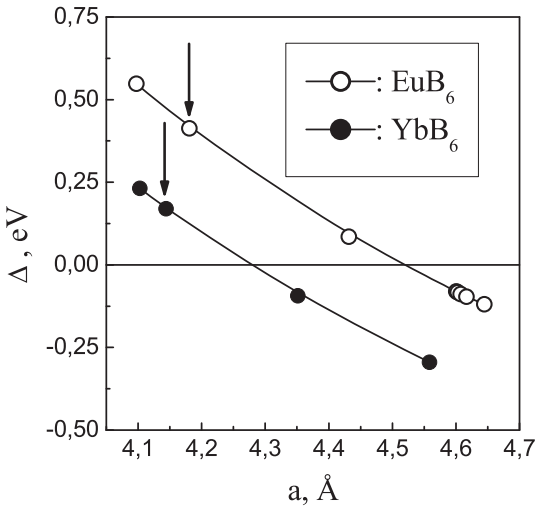

Fig. 4. Dependence of the bands overlap energy $\Delta = E_{top} - E_{bottom}$ on the lattice parameter a. The arrows denote the data for experimental a values at ambient pressure.

The calculations have confirmed the FM and semimetallic ground state for EuB_6. A comparison of the calculated total energies for the FM and PM states as a function of the atomic volume indicates an enhanced stability of the FM phase in EuB_6 under pressure. The PM band structures of EuB_6 and GdB_6 at zero pressure are quite similar and differ only in the Fermi energy positions due to a difference in the conduction electron number (see Fig. 3). There is a small overlap between the 5d–2p hybridized conduction and valence bands of EuB_6 at the X-point, which amounts to $\Delta = E_{top} - E_{bottom} \simeq 0.45$ eV at the theoretical value of the equilibrium lattice parameter $a = 4.143$ Å.

It can be noted that for the related divalent hexaboride YbB_6, which is isoelectronic to EuB_6, the analogous overlap is calculated to be $\Delta \simeq 0.17$ eV. These theoretical estimates of the overlap Δ exceed noticeably those evaluated from the dHvA effect [18], transport [12,26] and spectral properties [27,28]: $\Delta_{exp} \simeq 0.15$ eV and $\Delta_{exp} \sim 0$ for EuB_6 and YbB_6, respectively. This overestimation is presumably caused by the overbonding of the LSDA approach combined with a strong negative dependence of Δ on the lattice parameter (see Fig. 4).

As can be seen in Fig. 4, the calculated volume dependencies of the band overlap Δ appeared to be similar for EuB_6 and YbB_6 with the average volume derivative of $d\Delta/d\ln V \simeq -2$ eV. A close value, $d\Delta/d\ln V \simeq -1.6$ eV, was calculated also for isoelectronic SrB_6 compound [15] while a stronger volume dependence, $d\Delta/d\ln V \simeq -4$ eV, results from spin-polarized calculations of the EuB_6 band structure performed by FLAPW method within LDA+U approximation [16]. This suggests a crucial dependence of the calculated Δ and $d\Delta/d\ln V$ upon computational details. Also, there is a strong dependence of the band overlap on the position parameter x of the CaB_6 crystal structure. In particular, $d\Delta/dx$ amounts to -70 eV for SrB_6 [15] and around -100 eV for EuB_6 (this work). Therefore, a choice of the value of x and a possibility of its pressure dependence are of great importance.

4. Discussion

The obtained pressure derivative for the paramagnetic Curie temperature of $Eu_{0.99}B_6$, $d\Theta/dP = 0.52 \pm 0.04$ K/kbar, agrees reasonably with the reported experimental data on the initial pressure derivative of the Curie temperature for EuB_6, $dT_C/dP \sim 0.5$ K/kbar [7] and ~0.4 K/kbar [13]. It has been suggested [13], that the large pressure effect on T_C can be driven by the RKKY-type indirect exchange between 4f magnetic moments of Eu^{2+}, which is mediated by conduction electrons. This assumption is verified by our band structure calculations for EuB_6, which have revealed a free-electron like behavior of $E(k)$ dispersion curves (see Fig. 3). The Fermi surface consists of small pockets of electrons centred at the point X, and the pocket size is strongly dependent on volume.

The RKKY dependence of the paramagnetic Curie temperature on the charge carriers density n is given by [29]:

$$\Theta \propto J^2 n^{4/3} F(n), \tag{2}$$

where J is the effective exchange coupling between 4f moments and conduction electron spins, $F(n)$ the RKKY function. Since *ab initio* calculations of Θ represent a challenging task, it seems more feasible to estimate its volume derivative in the framework of the RKKY approach (2) as follows:

$$\frac{d\ln\Theta}{d\ln V} = 2\frac{d\ln J}{d\ln V} + \frac{d\ln n}{d\ln V}\left(\frac{4}{3} + n\frac{\partial \ln F}{\partial n}\right). \tag{3}$$

Based on the estimates of $d\ln J/d\ln V$ (~ -0.5 for RE compounds [31]) and $n(\partial \ln F/\partial n) < 1$ (for reasonable values of $\partial \ln F/\partial n$ [29,30] and $n \sim 0.01$/f.u. [18]), the corresponding terms in Eq. (3) can be neglected, in comparison with the dominant contribution to the volume dependence of Θ:

$$\frac{d\ln\Theta}{d\ln V} \approx \frac{4}{3}\frac{d\ln n}{d\ln V} \simeq 2\frac{1}{\Delta}\frac{d\Delta}{d\ln V}. \tag{4}$$

Here we used a free-electron like estimation $n \propto \Delta^{3/2}$, which relates the Fermi volume of quasi-spherical pockets with the band overlap Δ. The substitution of calculated $\Delta \simeq 0.15$ eV and $d\Delta/d\ln V = -3 \pm 1$ eV (which is the average of our value and that determined in Ref. [16]) in Eq. (4) gives $d\ln\Theta/d\ln V = -40 \pm 13$. The obtained value agrees reasonably with the experimental data (see Table 1), taking into account all estimations made above. This suggests that the electronic states at the X point and the variation of their density n with pressure are responsible for the magnetic ordering in EuB_6. It is worthy to note that the resulted from Eq. (4) value of the volume derivative, $d\ln n/d\ln V = -30 \pm 10$, is consistent with the corresponding estimates, obtained from available experimental data on the pressure effect in the resistivity ρ of EuB_6 at room temperatures, $d\ln n/d\ln V \simeq -d\ln\rho/d\ln V \simeq -18$ [13] and -50 [14], and for $EuB_{5.99}C_{0.01}$ at $T \simeq 20$ K, $-d\ln\rho/d\ln V \simeq -12$ [32].

As the charge carriers density increases (e.g. in the carbon doped EuB_6 and trivalent GdB_6), the pressure effect is expected to

diminish owing to both a gradual decrease of $d\ln n/d\ln V$ and an increase of the terms, which were omitted in Eq. (3) for EuB_6. This is in a qualitative agreement with our experimental data for the carbon doped EuB_6 and GdB_6 compounds, but a more detailed analysis of all contributions in Eq. (3) is needed for the quantitative comparison and assessment.

For GdB_6, the experimentally estimated pressure derivative of the paramagnetic Curie temperature, $d\Theta/dP$, can be verified by consideration of the spontaneous volume change due to the antiferromagnetic ordering $\Delta V/V \equiv \omega_m$, which is related to the squared molar magnetic moment $M^2(T)$ (see [33] and references therein):

$$\omega_m(T) = \frac{c}{B} M^2(T). \tag{5}$$

Here B is the bulk modulus, and c is the magnetoelastic coupling constant. The latter can be determined within the phenomenological relation [33,34]:

$$\frac{c}{B} = -\frac{1}{2\chi V}\frac{d\ln\chi}{dP}, \tag{6}$$

where χ and V are the molar susceptibility and volume, respectively. From Eq. (1) it follows

$$\frac{1}{\chi}\frac{d\ln\chi}{dP} = \frac{1}{C}\frac{d\Theta}{dP}. \tag{7}$$

By using the experimental values of $d\Theta/dP$, Curie constant C and $V \simeq 41.5\,cm^3$, one estimates c/B value to be temperature independent and equal

$$\frac{c}{B} = -\frac{1}{2CV}\frac{d\Theta}{dP} = (2.6 \pm 0.5) \times 10^{-13}(mol/emu)^2. \tag{8}$$

The substitution of the evaluated c/B value and the experimental molar magnetic moment of GdB_6 at $T = 0\,K$, $M(0) \simeq 3.91 \times 10^4$ emu/mol ($\simeq 7\mu_B$/Gd [35]) in Eq. (5), yields the volume change under AFM transition to be $\omega_m(0) = 0.04 \pm 0.01\%$. This estimate agrees reasonably with the value $\omega_m(0) \sim 0.03\%$, which resulted from the thermal expansion measurements for GdB_6 [25].

5. Summary

The measurements of the pressure effects on magnetic susceptibility of Eu and Gd hexaborides in their PM state have revealed a strong positive pressure dependence of the paramagnetic Curie temperature Θ in EuB_6 as compared with GdB_6, where the effect is markedly lower and opposite in sign. For EuB_6 the experimental $\Theta(P)$ dependence correlates positively with increase of the charge carriers density n under pressure. The latter was determined by *ab initio* calculations of the volume dependent band structure and resulted from the overlap of conduction and valence bands at X-point of the Brillouin zone. The reasonable description of the observed pressure effects on Θ was obtained within the RKKY-like approach. Nevertheless, an improvement of electronic structure calculations and further experimental studies are required to shed more light on the main mechanisms responsible for the peculiar behavior of magnetic properties of EuB_6 and other RE hexaborides under pressure and doping.

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