

## UC Irvine

### UC Irvine Previously Published Works

**Title**

Magnetotransport measurements on Nd<sub>1-x</sub>CaxB<sub>6</sub>

**Permalink**

<https://escholarship.org/uc/item/8x85497t>

**Journal**

Journal of Physics Conference Series, 200(1)

**ISSN**

1742-6588

**Authors**

Stankiewicz, Jolanta  
Bianchi, Andrea D  
Fisk, Zachary

**Publication Date**

2010

**DOI**

10.1088/1742-6596/200/1/012192

**Copyright Information**

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

# Magnetotransport measurements on $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$

Jolanta Stankiewicz<sup>1</sup>, Andrea D Bianchi<sup>2</sup> and Zachary Fisk<sup>3</sup>

<sup>1</sup> Instituto de Ciencia de Materiales de Aragón and Departamento de Física de la Materia Condensada, CSIC–Universidad de Zaragoza, 50009-Zaragoza, Spain

<sup>2</sup> Département de Physique, Université de Montréal, Montréal, Quebec H3C 3J7, Canada

<sup>3</sup> Department of Physics and Astronomy, University of California, Irvine, CA 92697, USA

E-mail: jolanta@unizar.es

**Abstract.** We performed measurements of the Hall effect, electrical resistivity, and magnetization on random alloys of  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  ( $x < 0.4$ ) in the 2 to 300 K temperature range, and in magnetic fields of up to 5 T. We find that a large anomalous contribution to the Hall effect, observed in  $\text{NdB}_6$  single crystals in the paramagnetic regime, decreases sharply with Ca content. In addition, the electron concentration decreases as expected for a divalent substitution for trivalent Nd. We tentatively relate these observations to changes in the Fermi surface volume upon doping.

## 1. Introduction

Hexaboride compounds have a simple chemical and structural configuration. However, these compounds show unusual physical properties, more so if they are synthesized with divalent rare-earth or alkaline-earth metal cations. In this paper we report results of magnetic and electrical transport measurements on  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  alloys, where some of the rare-earth atoms are substituted with divalent Ca atoms. Our aim is to see how a decreasing valence, and the variations in the Fermi surface (FS) that follow from it, affect the properties of these alloys.

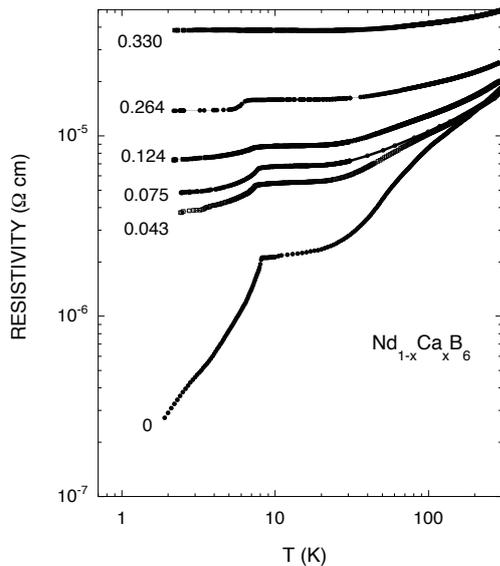
The rare-earth hexaboride compound  $\text{NdB}_6$  crystallizes in a cubic lattice with CsCl arrangement of Nd and  $\text{B}_6$  octahedra. Its magnetic and electronic properties have been the subject of quite intensive research, both experimental and theoretical, for the last few decades.  $\text{NdB}_6$  orders in an *A*-type collinear antiferromagnetic (AF) structure below  $T_N \approx 8$  K [1]. The ground state of the  $\text{Nd}^{+3}$  ions ( $J=9/2$ ) is split in a cubic crystal field into two  $\Gamma_8$  quartets and a  $\Gamma_6$  doublet [2,3]. The first excited energy is approximately 135 K above the ground  $\Gamma_8^2$  state. A competition between crystalline–electric field (CEF) and ferro–quadrupolar interactions gives rise to the low–field magnetic anisotropy in  $\text{NdB}_6$  that is much weaker than isotropic magnetic exchange interaction. Band structure calculations indicate that *4f* levels are rather deep in  $\text{NdB}_6$  [4]. The experimentally found frequency branches of the de Haas–van Alphen effect [5] in the AF phase can be well reproduced by calculated Fermi surfaces.

Recently, the topology of FS within the paramagnetic phase of  $\text{NdB}_6$  has been explored [6]. It resembles the FS observed in  $\text{LaB}_6$  [7], but with additional weak correlations. The FS consists of six large ellipsoids, centered at the *X* points of the Brillouin zone, which slightly overlap in the  $\Gamma\text{MX}$  plane. Upon alloying with divalent atoms, we expect the FS ellipsoids to shrink and eventually not to overlap as the number of conduction electrons decreases. Indeed, we find experimentally that the electron concentration becomes smaller as we substitute Ca for Nd in

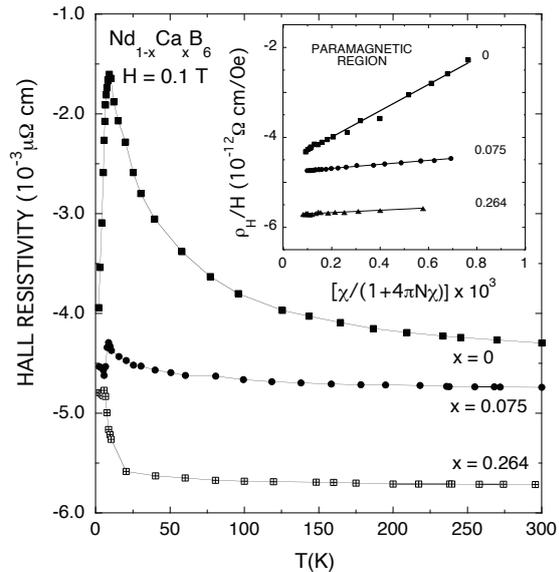
NdB<sub>6</sub>. We also find that the anomalous part of the Hall coefficient drops nearly to zero with alloying for  $x \lesssim 0.1$ . In our previous studies we encountered that the anomalous Hall coefficient in paramagnetic NdB<sub>6</sub> is much larger than the ordinary one and is independent of temperature [8].

## 2. Experiment

Single crystals of Nd<sub>1-x</sub>Ca<sub>x</sub>B<sub>6</sub> were grown from stoichiometric amounts of hexaboride components in Al flux. We checked its composition by electron probe microanalysis. All measurements were performed on small single crystals with approximate dimensions  $0.3 \times 1 \times 5.0$  mm<sup>3</sup>. These lengths are along the axes of the cubic structure. We have used the same crystals in electrical transport and magnetic measurements. In this way, we expect to avoid domain and sample-shape related effects when comparing results of different experiments. We measured the electrical resistivity and Hall effect with a six probe method. Contact leads (25 μm gold wire) were soldered to the sample using pure indium. The Hall resistivity  $\rho_H$  was measured as a function of magnetic field, from -2 T up to 2 T, for all experimental points. In addition, the variation of  $\rho_H$  with magnetic field, up to 5 T, was checked at several temperatures. The magnetization measurements in the temperature range from 2 to 300 K and in magnetic fields of up to 5 T were made using the same geometry with a commercial SQUID magnetometer.



**Figure 1.** Resistivity as a function of temperature for Nd<sub>1-x</sub>Ca<sub>x</sub>B<sub>6</sub> single crystals. An abrupt change in the slope for  $T < 10$  K corresponds to the Néel temperature. The solid lines are guides to the eye.



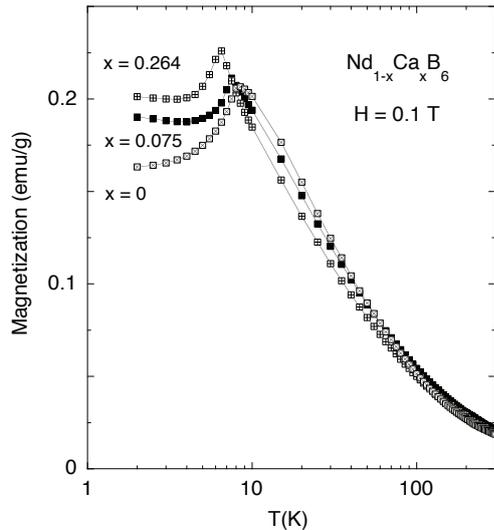
**Figure 2.** Hall resistivity vs. temperature for various Nd<sub>1-x</sub>Ca<sub>x</sub>B<sub>6</sub> single crystals. The inset shows  $\rho_H/H$  in the paramagnetic region plotted against the effective susceptibility for the same samples. The solid lines are linear fits to experimental points.

## 3. Results and Discussion

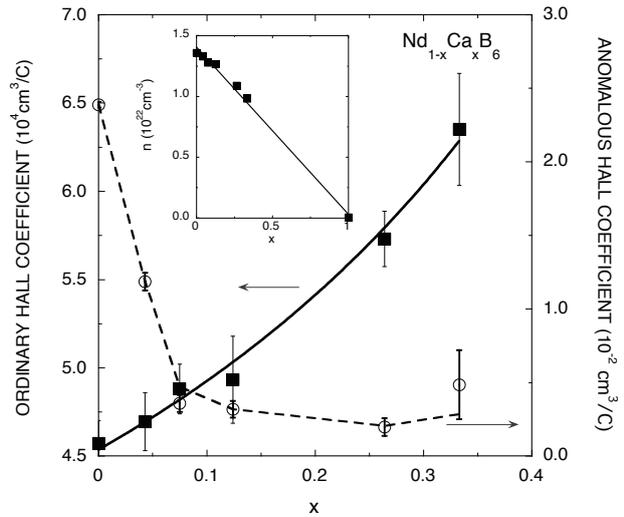
How the resistivity  $\rho$  of Nd<sub>1-x</sub>Ca<sub>x</sub>B<sub>6</sub> single crystals varies with temperature in the 2 to 300 K range is shown in Figure 1. The resistivity drops sharply below  $T_N$  as magnetic order sets in. CEF effects give rise to a broad shoulder, observed around 70 K [9]. Temperature-independent residual resistivity becomes larger with increasing content of Ca as a consequence of random

substitution of Nd by Ca atoms. We note that the magnetic part of the resistivity decreases with increasing  $x$ .

We now turn to the Hall effect results. Data from some samples are given in Hall resistivity  $\rho_H$  versus temperature plots in Fig. 2. At low fields,  $\rho_H$  increases as  $T$  decreases down to  $T_N$ ;  $\rho_H$  drops as  $T$  decreases below  $T_N$ . The Hall resistivity follows quite closely the magnetization of the samples. The low-field magnetization curves, for the same  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  single crystals as in Fig. 2, are plotted versus temperature in Fig. 3. To interpret these results, we use for the Hall resistivity the phenomenological expression:  $\rho_H = R_o B + R_s 4\pi M_s$ , where  $R_o$  is the normal Hall coefficient,  $R_s$  is the anomalous Hall effect (AHE) coefficient,  $B$  is the applied magnetic induction, and  $M_s$  is the spontaneous magnetization. In the paramagnetic region  $M_s = \chi H$ , where  $\chi$  is the magnetic susceptibility and  $H$  is the applied magnetic field. Using this relation we obtain:  $\rho_H/H = R_o + 4\pi\chi^*[R_s + R_o(1 - N)]$ , where  $\chi^* = \chi/(1 + 4\pi N\chi)$ , which includes the effects of the demagnetization field, is the effective susceptibility, and  $N$  is the demagnetization factor. The total Hall resistivity is linear in  $H$ ; however it has a normal part and a part which depends on the magnetic susceptibility. Plotting  $\rho_H/H$  versus  $\chi^*$ , we obtain the linear behavior shown in the inset of Fig. 2. This implies that both Hall coefficients are independent of  $T$ . The y-axis intercepts yield the values of  $R_o$  and the slopes of the straight lines give values which are proportional to  $R_s$ . Their variation with Ca content is plotted in Fig. 4.



**Figure 3.** Low-field magnetization as a function of temperature for the same  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  single crystals whose Hall resistivity is shown in Fig. 2. The solid lines are guides to the eye.



**Figure 4.** Ordinary (left axis) and anomalous (right axis) Hall coefficients in the paramagnetic region for  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  single crystals. The solid and dashed lines are to guide the eye. The inset shows a linear fit to the variation of the electron concentration with Ca content.

As expected for a substitution of trivalent ions by divalent ones, the effective electron concentration  $n$  in  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  decrease as the content of Ca increases. We find that this decrease is linear in  $x$  for  $x < 0.4$ . This is shown in the inset of Fig. 4. The linear fit to  $n(x)$  reproduces well the value of the electron concentration reported for  $\text{CaB}_6$  [10]. Unpublished results of high-magnetic field de Haas van Alphen measurements on  $\text{La}_{1-x}\text{Sm}_x\text{B}_6$  single crystals show that the FS volume decreases upon substitution of La by divalent Sm atoms [11]. In addition, this decrease in volume was found to be proportional to the decrease in the number of conduction electrons. For  $x = 0.10$ , a multi-connected FS of  $\text{La}_{1-x}\text{Sm}_x\text{B}_6$  changes into isolated

pockets. One might expect a similar effect in  $\text{Nd}_{1-x}\text{Ca}_x\text{B}_6$  alloys. Therefore, for some (small) Ca content in  $\text{NdB}_6$ , conduction electron ellipsoids would no longer overlap. This should affect transport properties since the Fermi surface changes radically then. Indeed, as seen in Fig. 4, the AHE coefficient decreases with alloying by more than one order of magnitude for  $x \lesssim 0.1$ . The AHE values for  $x > 0.10$  are comparable to those reported for other paramagnetic rare-earth compounds [12]. However, a large anomalous contribution to the Hall effect for  $x \lesssim 0.1$  in the paramagnetic phase and its strong variation upon alloying are puzzling. The intrinsic AHE in magnetic materials is very sensitive to details of the electronic band structure and gets its significant contributions from band crossing. A calculation of the anomalous velocity for  $\text{NdB}_6$ , evaluating the Berry curvature tensor over the Fermi surface, would be desirable. In fact,  $\text{NdB}_6$  might provide a clean test for some of the current theories of the anomalous Hall effect.

### Acknowledgments

This work was supported in part by grant MAT2008/03074 from the Spanish Ministerio de Ciencia e Innovación and by NSF grant DMR-0854781. Additional support from Diputación General de Aragón (DGA-IMANA) is also acknowledged.

### References

- [1] McCarthy C M and Tompson C W 1980 *J. Phys. Chem. Solids* **41** 1319
- [2] Loewenhaupt M and Prager M 1986 *Z. Phys. B: Condens. Matter* **62** 195
- [3] Uimin G. and Brenig W 2000 *Phys. Rev. B* **61** 60
- [4] Kitamura M 1994 *Phys. Rev. B* **49** 1564
- [5] Onuki Y, Umezawa A, Kwok W K, Crabtree G W, Nishihara M, Yamazaki T, Omi T and Komatsubara T 1989 *Phys. Rev. B* **40** 11195
- [6] Goodrich R G, Harrison N and Fisk Z 2006 *Phys. Rev. Lett.* **97** 146404
- [7] Arko A P J, Crabtree G, Karim D, Mueller F M and Windmiller L R 1976 *Phys. Rev. B* **13** 5240; Ishizawa Y, Tanaka T, Bannai E and Kawai S 1977 *J. Phys. Soc. Japan* **42** 112
- [8] Stankiewicz J, Nakatsuji S and Fisk Z 2005 *Phys. Rev. B* **71** 134426
- [9] Fisk Z 1976 *Solid State Commun.* **18** 221
- [10] Souma S, Komatsu H, Takahashi T, Kaji R, Sasaki T, Yokoo Y and Akimitsu J 2003 *Phys. Rev. Lett.* **90** 027202
- [11] Goodrich R G, Alver U, Young D P, Harrison N, and Fisk Z *unpublished*
- [12] Hurd C M 1972 *The Hall Effect in Metals and Alloys* (New York-London: Plenum Press)