Temperature dependent striction effect in a single crystalline Nd$_2$Fe$_{14}$B revealed using a novel high temperature resistivity measurement technique

Kyuil Cho$^1$*, S L Bud’ko$^{1,2}$ and P C Canfield$^{1,2}$

$^1$ Ames Laboratory, Ames, IA 50011, United States of America
$^2$ Department of Physics & Astronomy, Iowa State University, Ames, IA 50011, United States of America

E-mail: kyuil.cho@mst.edu

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Abstract

We studied the temperature dependence of resistivity in a single crystalline Nd$_2$Fe$_{14}$B using a newly developed high temperature probe. This novel probe employs mechanical pin connectors instead of conducting glue/paste. From warming and cooling curves, the Curie temperature was consistently measured around $T_c = 580$ K. In addition, anomalous discrete jumps were found only in cooling curves between 400 and 500 K, but not shown in warming curves. More interestingly, when the jumps occurred during cooling, the resistivity was increased. This phenomenon could possibly be due to a temperature dependent striction effect induced by the re-orientation of magnetic domains well below the Curie temperature. Further microscopic study is needed to confirm this effect.

Keywords: Nd$_2$Fe$_{14}$B, magneto striction, Curie temperature, high temperature resistivity, permanent magnet, magnetic domain, NdFeB

(Some figures may appear in colour only in the online journal)

1. Introduction

Nd$_2$Fe$_{14}$B is the most widely used permanent magnet discovered in 1984 [1–3]. This intermetallic compound crystallizes in the tetragonal crystal structure, space group $P_4/nmm$ [4], and shows a ferromagnetic phase below $T_c = 586$ K [5]. Due to its high anisotropy field ($H_A \sim 7$ T) [5] as well as its high maximum energy product of $(BH)_{max} = 59$ MGOe [6], this compound is the strongest permanent magnet currently available and further effort has been put to enhance its performance by optimizing synthesis procedures [7–9]. In addition, Nd$_2$Fe$_{14}$B also shows an interesting spin-reorientation transition at $T_{SR} = 135$ K that has also drawn attention [10, 11].

* Author to whom any correspondence should be addressed.

In this article, we studied the temperature dependence of resistivity in a single crystalline Nd$_2$Fe$_{14}$B up to 700 K using a newly developed high temperature measurement technique, and found the multiple discrete jumps between 400 and 500 K which were only shown in cooling curves, not shown in warming curves (figure 1). These jumps were consistently found in multiple measurements with different sweep rates. The jumps could possibly be due to a temperature dependent striction effect caused by the reorientation of magnetic domains well below the Curie temperature. The magnetic field induced striction effect (magnetostriction) is a well known phenomenon in ferromagnetic materials that causes expansion or contraction in response to an applied magnetic field. Basically, upon application of a magnetic field, the underlying magnetic domains of a material are re-arranged, so this re-arrangement results in the change in materials’ dimensions. A similar striction effect can also occur when a sample is cooled...
below the Curie temperature. As the temperature decreases below the Curie temperature, magnetic domains start forming in arbitrary orientations. As the temperature further decreases, the size of each magnetic domain gets larger and the interaction among domains get stronger. As a result, the magnetic domains change their orientations to be aligned with nearby domains in order to reduce the magnetostatic energy associated with the domain boundaries. In the current study, we found that some of discrete jumps in resistivity only occur in the cooling curves. These jumps could possibly be due to a temperature dependent striction effect induced by the reorientation of magnetic domains. The jumps are not shown in the warming curves since the magnetic domains are already in a stable state from the lower temperature region. Thus, even though the temperature increases, the magnetic domains are not likely to change their orientations since they are already in a stable state. Further microscopic study is needed to uncover the origin of these jumps.

In Nd$_2$Fe$_{14}$B, the magnetostrictive effect was reported below the spin reorientation phase transition temperature $T_{SR} = 135$ K upon an application of pulsed magnetic field up to 15 T [12]. However, temperature-dependent striction effect has not been found from the previous studies [13, 14]. The possibility of temperature dependent striction effect suggested in this study needs to be further investigated.

2. Experiment

Single crystals of Nd$_2$Fe$_{14}$B were grown by using a solution growth method [15–17]. The sample for in-plane resistivity measurement has dimensions of 1.5 mm $\times$ 0.53 mm $\times$ 0.29 mm with accuracy of about 5%. To measure the resistivity at high temperatures up to 800 K, we developed a novel method (figure 2). First of all, four contacts of a sample were made by using a spot welding technique with long Pt-wires (50 $\mu$m in diameter) as shown in figure 2(a). Then the sample with long Pt wires are placed on top of a sapphire plate of 12 mm $\times$ 12 mm (panel (b)). After that, two small sapphire plates are placed on top of extended Pt-wire. In this way, the electrical shorting between Pt wires and bottom Copper plate is prevented. In panel (c), another top Copper plate is placed on top of both sapphire plates and securely pressed down by using two screws. Now, the sapphire plates are securely held between two Copper plates. At the same time, the Pt wires are held tightly between two sapphire plates without any shorting. In addition, the sample is securely held in contact with the bottom sapphire plate without any extra glue or paste. Then, the whole prepared unit in panel (c) is mounted on the heating stage of the high-temperature cryostat made by Cryo Industries of America, Inc. (panel (d)). Next procedure is to connect four Pt wires to the four thick Copper wires. As shown in panel (e), each wire is wound around a pin connector (P1) and the second pin connector (P2) is plugged into P1 connector. In this way, the Pt wire is mechanically held between two pin connectors (P1 and P2). Note that no extra conducting paste or glue is used. P1 connectors and thick Copper wires are permanently silver soldered. Since the melting point of silver solder is above 900 K, one can safely conduct measurements up to 800 K. The advantage of this novel method is that there is no need of conducting paste or glue. Once the experimental setup is ready, the cryostat is closed and pumped down to $1 \times 10^{-6}$ torr using a turbo pump. The resistivity was measured with about 1.5 mA current using a AC Resistance Bridge SIM921 by Standford Research Systems. The four probe resistivity measurement was conducted by using a Pt temperature sensor from Cryo Industries which was factory-calibrated up to 1123 K. LakeShore 335 Temperature controller was used.

3. Results and discussion

Using this novel method, we measured the temperature dependence of resistivity in a single crystalline Nd$_2$Fe$_{14}$B. Three different measurements were conducted with different sweep rates 2 K min$^{-1}$, 0.5 K min$^{-1}$ and 0.2 K min$^{-1}$. As shown in figure 1, all three measurements consistently show a ferromagnetic transition around $T_c = 580$ K. $T_c$’s of warming curves are slightly higher than those of cooling curves.
Figure 2. Experimental set up with a single crystalline Nd$_2$Fe$_{14}$B sample. (a) Four probe contacts with Pt wires were made using a spot welding technique. (b) The sample is placed on top of a sapphire plate (12 mm $\times$ 12 mm). Then, two other small sapphire plates are placed on top of the extension of Pt wires. (c) Another copper piece is placed on top and screwed down, so that two layers of sapphire plates are tightly held between top and bottom copper pieces. (d) The prepared unit in panel (c) is mounted to the high temperature heating stage with two other screws, and four extension of Pt wires are connected to the pin connectors. (e) This panel shows how a Pt wire is connected between two pin connectors. A Pt wire is wound around the first pin connector (P1) and then the second pin connector (P2) is plugged in with P1. Thus, the Pt wire is mechanically held between two pin connectors without any glue or paste. Furthermore, P1 connector is permanently silver soldered with a thick Cu wire.

And these differences get smaller when the sweeping rate decreases from $2 \text{ K min}^{-1}$ to $0.2 \text{ K min}^{-1}$. As the temperature decreases below $T_c$, the resistivity increased. This behaviour is opposite to common ferromagnetic materials such as Ni (Type-I) as shown in figure 3 since the loss of spin disorder scattering induces the decrease of resistivity below the Curie temperature. This anomalous increase in resistivity was discussed by Jen and Yao in 1987 [13, 14], explaining that Nd$_2$Fe$_{14}$B is not Type-I but Type-III ferromagnet. In Type-III ferromagnets, the anomalous increase in resistivity arises not from spin-disorder scattering but rather from the anomalous lattice contraction similar as c-axis resistivity of Gd [19, 20]. However, the authors commented that the Curie temperature of Nd$_2$Fe$_{14}$B is different from the expectation of Type-III model, so further investigation is needed.

In addition, multiple discrete jumps were found only from the cooling curves between 400 and 500 K. When the jumps occur, the resistivities always increase as shown in the insets of figure 1. These jumps show the tendency of the system to approach the state of the warming curve. We explain these jumps as a re-orientation of magnetic domains well below $T_c$. As the temperature decreases below the Curie temperature, magnetic domains start forming in arbitrary orientations. As the temperature further decreases, the interaction between domains gets stronger. Thus, some domains change their orientations by realigning with nearby domains. In this way, the total magnetostatic energy associated with the domain boundaries can be reduced. In addition, this realignment can induce the change in sample dimensions. The reason why the resistivity increases during the jumps is unclear, but it seems to be related to the characteristic of Type-III ferromagnet since the spin-lattice contraction is strong compared to Type-I ferromagnet. Further investigation is needed to understand the correlation between discrete jumps and reorientation of magnetic domains.

We also measured the resistivity of Ni single crystal (grown from Materials Preparation Center at Ames Laboratory, which is supported by the US DOE Basic Energy Sciences) to check the performance of the developed technique. As shown in figure 3, the $T_c$ of Ni single crystal was measured to be about 628 K, which is consistent with $T_c = 630 \text{ K}$ reported in literatures [18].

4. Conclusions

We investigated the temperature dependence of resistivity in a single crystalline Nd$_2$Fe$_{14}$B using a newly developed method. In addition to the Curie temperature around $T_c = 580 \text{ K}$, we identified anomalous discrete jumps between 400 and 500 K only from the cooling curves. These jumps occurred in a way to increase the resistivity. This phenomenon could possibly be due to a temperature dependent striction effect of magnetic
domains well below the Curie temperature. Further microscopic studies are needed to clarify this effect.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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ORCID IDs

Kyuil Cho https://orcid.org/0000-0003-2111-6355
S L Bud’ko https://orcid.org/0000-0002-3603-5585
P C Canfield https://orcid.org/0000-0002-7715-0643

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