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## Investigation of ultrathin magnetic fields on $^{57}\text{Fe}$ nuclei in $\text{Nd}_2\text{Fe}_{14}\text{B}$ and Fe compounds with metals

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Gamma-resonance studies of ultrathin magnetic fields on  $^{57}\text{Fe}$  nuclei in multicomponent compounds of iron with metals and a neodymium magnet of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  composition have been carried out. It is shown that ultrathin magnetic fields  $H_n$  are observed on  $^{57}\text{Fe}$  iron nuclei in all studied compounds, the largest of which falls on  $^{57}\text{Fe}$  nuclei in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , which significantly exceeds the  $^{57}\text{Fe}$  field in a neodymium magnet, which also has the highest magnetic induction among known permanent magnets. A qualitative analysis of the mechanism of formation of ultrathin magnetic fields on iron nuclei in the studied compounds is proposed.

**Keywords:** magnetism, permanent magnets, metals, gamma resonance, neodymium magnet, nuclear spectrum, ultrathin magnetic fields, isomer shift.

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### 1. Introduction

Magnetism, like natural radioactivity stays till now one of the mysterious and little understood natural phenomena. And it is remarkable that, along with other methods, magnetism is studied by radioactivity of iron nuclei  $^{57}\text{Fe}$ , successfully used in nuclear gamma resonance spectroscopy (NGRS), and which is main carrier of magnetism in nature. Having extra high sensitivity to short-range effects NGRS is the powerful experiment method to study ultrathin interactions in substances, in their nuclei of atoms the Mössbauer effect is observed. Such objects are also permanent magnets based on iron.

The permanent magnets of different shapes and size are widely used in modern technique, industry and household activities. Naturally, with further scientific and technical development the need in them will rise. So, search of material, technology development and production of permanent magnets with different shape, size and required technical characteristics stay the actual scientific and technical objective. At that the main requirements for the permanent magnets are their high magnetic induction, variety of dimensions and shapes, resistance to temperature changes, corrosion environment and electromagnetic radiation.

### 2. Permanent magnets (brief review)

Four classes of permanent magnets are widely used in modern technology and everyday life, for their production four basic compositions of materials are used: neodymium-iron-boron ( $\text{Nd}_2\text{Fe}_{14}\text{B}$ ), samarium-cobalt (SmCo), aluminium-Nickel-cobalt (Alnico), ferrites (ceramic magnets), each of them has own benefits and drawbacks. In terms of magnetic induction the neodymium

magnets stay the most powerful. They are resistant to demagnetization, have high magnetic characteristics and retain their original values for many years [1].

Alnico magnets, based on aluminum-nickel-cobalt alloy, have high stability of magnetic parameters at various mechanical and climatic conditions, high work temperature (up to  $800^\circ\text{C}$ ), parameters stability with time, they are resistant to demagnetization, corrosion, structural ageing and radiation effect.

Ceramic magnets (ferrites) are ferromagnetic compounds obtained by mixing iron oxide with Mn, Ba, Zn.

Samarium-cobalt magnets are resistant to demagnetization, corrosion and oxidation, and their coercitive force exceeds the same of neodymium magnets. They have magnetic anisotropy. So, they can be magnetized along axis of their magnetic orientation only.

Magnets  $\text{SmCo}_5$  or  $\text{Sm}_2\text{Co}_{17}$  as per combination of the magnetic properties are positioned between neodymium and ferritic magnets. Due to high magnetic induction, high resistance to demagnetization, small size and various shapes they are also used in space vehicles, aviation and computer equipment, mobile phones. Their drawbacks are fragility and tendency to chip.

Basic reason of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  high magnetism is tetragonal crystal structure which has high uniaxial magnetocrystalline anisotropy and preferable magnetization along definite axis of lattice comprising of alternating layers of atoms of iron and compound Nd-B [2]. The magnetism of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  is also promoted by the geometric shape and grain size of material within range  $45\text{--}100\mu\text{m}$ , their better alignment and sintering temperature [3,4].

Their drawback is low work temperature and instability to corrosive environment [5–7]. But due to better magnetic characteristics, various shapes and dimensions the

neodymium magnets are widely used in technique — in hard discs of computers, mobile phones and other technical devices.

There are also specific magnets, one of them ferritin–organometallic monomolecular compound having superparamagnetic behavior in the temperature range 4–60 K, able to store information and ensure much denser magnetic media than traditional magnets [8].

Data provided on permanent magnets show that to date different types of permanent magnets were developed, each of them can be successfully used in specific conditions of temperature, external magnetic fields, corrosive environment and radiation.

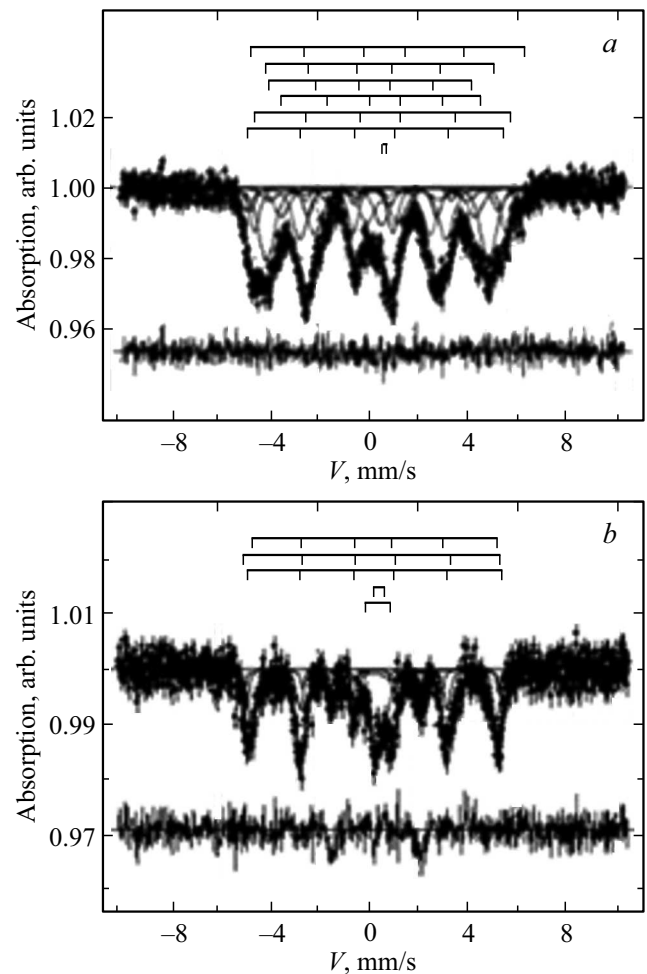
Further technical progress associated with permanent magnets use will substantially depend on development of universal permanent magnet having better summary technical characteristics required for their use under any environmental conditions and strong electromagnetic fields. However, there may be fundamental difficulties on the way of such magnet development, they are associated with restriction of both limit value of the magnetic induction of the actual system of atom electrons, and restriction of natural types of crystal lattices of the magnet material. However, we can not agree that the saturation magnetization  $J_s \approx 1.6$  T and residual magnetization — 1.3 T are limit for permanent magnets, and we need to find new materials and new technologies of magnets production.

It is known that main segment in power generation system — important for modern world of energy, is magnet specifically [9], due to this the search of material composition for strong magnet is actual objective for physicists, technologists, engineers. Studies show that strong magnet production requires combination of material composition, its crystal structure and practical technology. The unique character of the neodymium magnet is apparently somewhat random, although, as follows from the history of its development, reasonable selection by authors of the elementary composition, that in combination with crystal structure and technology is rather favorable for its magnetism.

And as the neodymium magnets are currently the best in terms of the magnetism, improvement of all characteristics of Nd-Fe-B and search of similar materials relate to many papers that study ultrathin interactions in various magnetic systems [10–19]. But authors do not focus attention to relation of intra-atomic ultrathin magnetic fields (UTMF) and substance magnetization, although such fields are observed even on non-magnetic atoms alloyed with iron [20].

### 3. Experiment and results

Measurements were made using spectrometer MC-1104Em in compressed transmission geometry in the Doppler velocity range  $\pm 10$  mm/s at  $T = 300$  K. The gamma-beam source is  $^{57}\text{Co}(\text{Cr})$  isotope, spectra were interpreted using program UnivemMS. Samples were fine powders of iron compounds obtained by a method of multi-stage cold rolling on four-high mill with force 50 kN



**Figure 1.** Mössbauer spectrum of  $^{57}\text{Fe}$  nuclei in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (a) and  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  (b) at  $T = 300$  K.

and same powder of neodymium flat magnet from hard disc of the computer.

Due to UTMFs observed in iron compounds on  $^{57}\text{Fe}$  nuclei in [16,18–20] we were interested in role of magnetization (magnetic moment) in formation of ultrathin magnetic fields  $H_n$  on  $^{57}\text{Fe}$  nuclei in permanent magnets. We also are interested in the observed in magnetic and Mössbauer measurements differences in temperature determination of phase transition ( $T_c$ ).

In present paper we studies some compounds of iron with metals of various groups of Periodic system (Co, Ni, Ag, Zn, B), and performed own studies of the neodymium magnet, in which high values of UTMF were also detected on  $^{57}\text{Fe}$  nuclei.

Figure 1 shows spectra of  $^{57}\text{Fe}$  nuclei in samples studied by us. Spectrum of  $^{57}\text{Fe}$  nuclei in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , obtained in paper [15] is described by the authors in model of six Zeeman sextets corresponding to six positions of Fe atoms in its lattice cell [2]. Spectrum of  $^{57}\text{Fe}$  in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  studied by us also comprises six unresolved Zeeman sextets and p[aramagnetic line, and spectrum in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  — is of similar in  $H_n$  three unresolved sextets and two doublets,

the origin of which is probably related to the non-cubic symmetry of the lattice of their environment. We do not know the origin of the paramagnetic line in spectrum of  $^{57}\text{Fe}$  in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , may be it is associated with local inhomogeneity of the sample made of powder of flat magnet from hard disc of computer. Fields  $H_n$  were detected in  $^{57}\text{Fe}$  nuclei in all studied samples, and all of them have ferromagnetic nature.

As it is known, the specific nature and individuality of a chemical element are determined by the charge of the nucleus [21]. In studied two and three component compounds ( $\text{Fe}_7\text{Ag}$ ,  $\text{Fe}_7\text{Zn}$ ,  $\text{Fe}_{50}\text{Zn}_{50}$ ,  $\text{Fe}_{50}\text{Ni}_{50}$ ,  $\text{Fe}_{95}\text{Zn}_5$ ,  $\text{Fe}_{50}\text{Co}_{45}\text{Zn}_5$ ,  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ ), the iron ions have in the immediate vicinity atoms of different periods and groups of Periodic system of elements — Co, Ni (VIII group of IV period, with electronic configuration  $3d^74s^2$  and  $3d^84s^2$  respectively), Ag (I group of V period, with configuration  $4d^95s^2$ ), Zn (II group of IV period with configuration  $3d^{10}4s^2$ ) and B (III group of II period with configuration  $2s^22p^1$ ). In formation of lattice type and chemical bond nature each of these elements plays its own specific role associated with their electronic configuration. We can assume that UTFM observed on nuclei of atoms of „non-magnetic“ elements alloyed with iron [20] are result of presence of orbital and spin magnetic elements of valence electrons of magnetic matrix atoms. That is, if the substance has magnetization then on iron nuclei the ultrathin magnetic fields should be observed, probably. As the physical and chemical properties of the substance depend on the crystal structure and configuration of electron shells of atoms, they can be different depending on their nature and chemical bond between them. As the iron is the basic component of composition of almost all materials for magnets, then we need to know in what exact composition the iron ions can show the maximum possible magnetic properties. And to answer the question — why exactly the neodymium magnets have high magnetic induction values, it is necessary to study in detail not only the structure of the crystal lattice, but also the electronic configuration of the  $\text{NdFeB}$  compound as a whole and specifically the iron in it. During compounds formation the electronic configurations of atoms in accordance with their valency and number of actually formed chemical bonds change such that as a result the total spin system of the magnet, depending on type of crystal structure, can be favorable for both increasing and decreasing of sample magnetization. At that we expected that for the studied samples the values of ultrathin magnetic fields on  $^{57}\text{Fe}$  nuclei in compositions  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ ,  $\text{Fe}_{50}\text{Co}_{45}\text{Zn}_5$ ,  $\text{Fe}_{50}\text{Ni}_{50}$ , where ferromagnetic Co and Ni are contained, will be such higher than in  $\text{Fe}_7\text{Zn}$ ,  $\text{Fe}_7\text{Ag}$ ,  $\text{Fe}_{50}\text{Zn}_{50}$  with diamagnetic Ag and Zn. Besides, fields on  $^{57}\text{Fe}$  nuclei in compound  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , similar to  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (but with Ni instead of Nd), due to mainly ferromagnetic composition, shall be much higher than in other compositions. If Ni and Co are ferromagnetics and can increase magnetization of the iron compounds with them, then diamagnetic zinc, argon and boron can increase or decrease magnetism of iron compounds with

them depending on how the total magnetic moment will be aligned, and what the type of lattice of the sample will be. Experiment, in some extent, confirms this: nuclei of iron compounds with these elements have lower values of fields  $H_n$  as compared to fields in their compounds with transition and rare earth elements (Figure 2). Iron atom in ground state has the electronic configuration of the valence band  $3d^64s^24p^0$  with four uncompensated electrons on  $d$ -shell. If type of crystal lattice  $\text{Nd}_2\text{Fe}_{14}\text{B}$  is known, and promotes the strong magnetism of the compound, and field  $H_n$  observed on iron nuclei than we can assume that exactly the total spin configuration of the compound promotes this resulting in presence of the direct relation between the magnetic moment  $\text{Nd}_2\text{Fe}_{14}\text{B}$  and  $H_n$  on  $^{57}\text{Fe}$  nucleus in it. As it is known, the contribution to  $H_n$  is provided by several types of magnetic fields, their competition results in total effective field  $H_n$  in area of nucleus. At that the main contribution to  $H_n$  is made by the field of contact interaction of  $s$ -electrons localized in area of nucleus and nucleus (Fermi field) with insignificant contribution of  $p$ -electrons of own atom [22]:

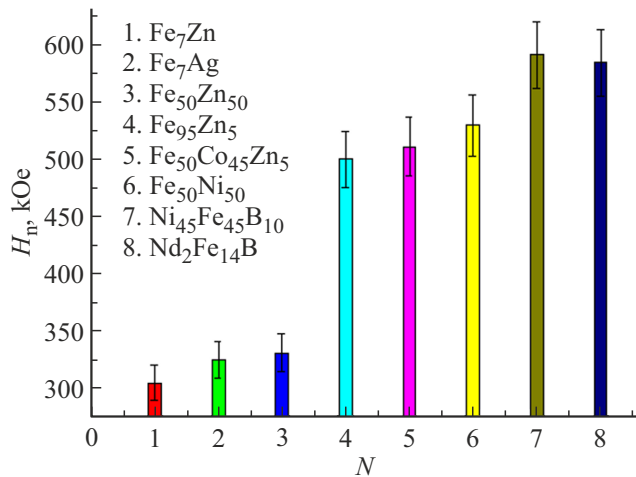
$$H_{\text{fermi}} = -\frac{8\pi}{3} \mu_B \sum_n V \rho_{ns}^{\uparrow\downarrow}(0) i, \quad (1)$$

where

$$\sum_n \rho_{ns}^{\uparrow\downarrow}(0) = \sum_n (\rho_{ns}^{\uparrow}(0) - \rho_{ns}^{\downarrow}(0))$$

— spin density in area of location of nucleus,  $\rho_{ns}^{\downarrow}(0)$  — densities of  $s$ -electrons with antiparallel spins on  $ns$ -shell of atom,  $\mu_B$  — Bohr magneton. Paired electrons with antiparallel spins have no contribution to the field, by resulting spin of  $d$ -electrons polarizes internal  $s$ -electrons of atom, and as a result of their exchange interaction there is deformation of electron cloud of internal  $s$ -electrons, which form the field  $H_n$  in area of the nucleus. So, according to the theoretical calculations [22],  $H_{\text{fermi}}$  contribution is proportional to the magnetic moment of  $3d$ -shell of iron atom, and in some compounds its contribution to the field  $H_n$  is determinative:  $H_n = -k\mu_{\text{Fe}}$ .

In the high-spin state iron has the configuration  $3d^64s^24p^2$  with eight unpaired electrons on  $d$ - and  $p$ -shells. Neodymium — element of III group of VI period, relates to group of lanthanides and in normal state has configuration  $4d^{10}5p^66s^24f^4$  with four uncompensated spins on  $f$ -shell. For chemical bond with neighbours the neodymium shall transit to high-spin state connecting for this one electron from  $6s$ -shell to free cell of  $f$ -orbital, and increasing to 6 the number of uncompensated spins ( $5s^25p^66s^14f^5$ ). The electronic configuration of the boron atom in ground state  $1s^22s^22p^1$ . In excited state its one electron from  $s$ -orbital can go to the vacant  $p$ -orbital. and form configuration of three uncompensated spins. Such rare combination of 17 spins oriented in the selected direction of the lattice, of course, can ensure high magnetic induction of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . But we do not know in what electronic configuration bonds of Fe, Nd and B between each other are formed. So, we do not know value of the total magnetic moment of the



**Figure 2.** Ultrathin magnetic fields on  $^{57}\text{Fe}$  nuclei in studied samples at  $T = 300\text{K}$ .

magnet. But considering the known crystal structure we can suppose the most probable orientation of the magnetic moment of sample along known light axis of the crystal. However, it can be assumed that exactly  $\text{Nd}_2\text{Fe}_{14}\text{B}$  the total electronic configuration is implemented that is favourable for creation of the high magnetic induction, maximum of uncompensated spins of the compound as a whole is at such electronic configuration.

Neodymium is weakly antiferromagnetic, and in its pure form its magnetism manifests itself only at low temperatures, but the compound of neodymium with iron has a rather high Curie temperature. Due to boron relation to the accessory content there are different opinions relating boron role in magnetism of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . We assume that diamagnetic boron atoms do not contribute directly to the magnetism, but improve ions adhesion due to strong covalent bond. Paper [23] shows that the dependence of coercive force of compound Nd-Fe-Al-Ti-B on the boron content has extreme nature. Value and position of maximum depend on boron content — with formation of boron-containing phases Ti-B and supersaturation of the main magnetic phase with boron. But, it is known that admixture can drastically change property of substance, and boron, due to its required minimum of content in the compound and interaction with Nd atoms, may be form the structure  $\text{Nd}_2\text{Fe}_{14}\text{B}$  suitable for high magnetic induction. Figure 2 shows that fields in iron nuclei diluted by diamagnetics Ag and Zn actually have low values as compared with the fields in compounds of iron with 3d-metals — cobalt and nickel. At the same time the maximum value of field is observed in compound  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , where ferromagnetic Ni and diamagnetic boron, forming, probably, common magnetic lattice parallel to lattice of iron Ni-B, promote the formation of strong field on  $^{57}\text{Fe}$ . In compound  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , similar by composition in percent of components to compound  $\text{Fe}_{50}\text{Co}_{45}\text{Zn}_5$ , but with diamagnetic boron instead of zinc, the field on iron nuclei is large by tens of kilooersted then in compounds with Ag and Zn. I.e., the diamagnetic

Parameters of sextets of Mössbauer spectra of samples

№	Sample	$\delta_s$ , mm/s	$Q_s$ , mm/s	$H_n$ , kOe	$G$ , mm/s	$S$ , %
1	$\text{Fe}_7\text{Zn}$	0.2192	5.7842	<b>304.48</b>	0.3480	7.46
2	$\text{Fe}_7\text{Ag}$	-0.0028	1.5231	<b>324.23</b>	0.1794	1.87
3	$\text{Fe}_{50}\text{Zn}_{50}$	-0.1278	0.7501	<b>330.43</b>	0.2870	5.72
4	$\text{Fe}_{95}\text{Zn}_5$	-0.0738	1.5179	<b>499.33</b>	0.2009	0.49
5	$\text{Fe}_{50}\text{Co}_{45}\text{Zn}_5$	0.1559	3.7580	<b>510.60</b>	0.5899	7.98
6	$\text{Fe}_{50}\text{Ni}_{50}$	-0.2166	5.0564	<b>529.04</b>	0.7759	8.51
7	$\text{Nd}_2\text{Fe}_{14}\text{B}$	0.3163	0.6132	<b>563.21</b>	0.776	16.40
8	$\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$	1.0182	1.0922	<b>590.32</b>	0.7760	11.58

boron and zinc in these compounds have different effect on the electronic structure of Fe, this is shown different in its energy spectra. Exactly bond Ni-B, may be, is the main agent of forming high-spin state of iron in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , as Nd-B in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ . Based on the fact that  $H_n$  on  $^{57}\text{Fe}$  nuclei in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  is significantly higher than field  $H_n$  in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , we assume that magnetic induction of  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ , manufactured as per technology of sintering and magnetization of neodymium magnet can be higher than the magnetic induction of  $\text{Ni}_2\text{Fe}_{14}\text{B}$ . From electronic configurations of Ni ( $4s^23d^8$ ) and Nd ( $4f^56s^1$ ) in excited state it also follows that Ni has 8, and Nd 6 unpaired electrons on external shell, this indicates Ni advantage in formation of magnetism in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  over Nd in magnetism  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , but only is Ni is part of the compound exactly in high-spin state. Issue of mechanism of interrelation of the magnetic fields on  $^{57}\text{Fe}$  nuclei and magnetic induction of substance requires more detailed theoretical analysis. So far, we can only assume that such relationship exists, since all the samples of compounds we studied have ferromagnetic properties, and in each of them on  $^{57}\text{Fe}$  nuclei there are strong ultrathin magnetic fields. Table, for this paper, shows parameters only of the widest sextet of Mössbauer spectra of the studied samples.

## 4. Conclusion

Till now we do not have theoretical justification if the direct relationship presence between the substance magnetization and ultrathin magnetic fields on iron nuclei in it, but the observed features on  $^{57}\text{Fe}$  nuclei in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (having also high magnetic induction), in compound  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  and other intermetallic compounds can be assumed as justification of such statement. In other words, if the sample (permanent magnet) is magnetized, ultrathin magnetic fields shall also be observed on the iron nuclei in it. And as fields on  $^{57}\text{Fe}$  nuclei in  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  are significantly higher than in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , we assume that the magnetic induction of the magnet made of  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$  as per appropriate technology will be higher than in neodymium magnets. And it is boron that, probably, plays the basic role in formation of the high-spin state of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  and  $\text{Ni}_{45}\text{Fe}_{45}\text{B}_{10}$ .

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