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LETTER TO THE EDITOR

Electrical quadrupole interaction at the Ni site in RNiBC compounds

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Abstract. ^{57}Fe Mössbauer spectra of RNiBC (R = Gd, Tb, Dy, Ho, and Er) compounds doped with ^{57}Fe (0.5 at.%) taken above their magnetic ordering temperatures reveal a quadrupole doublet with a splitting $|\Delta E_Q|$ which decreases on going from Gd to Er. The values of $|\Delta E_Q|$ depend linearly on the parameter c'/a , with c' being the distance apart of the R–C layers between which the $\text{Ni}_2\text{--B}_2$ layer is sandwiched and a being the lattice parameter in the basal plane. The $|\Delta E_Q|$ versus c'/a relation is identical with that previously found for $\text{RNi}_2\text{B}_2\text{C}$ compounds.

Soon after the first observation of superconductivity in a new family of intermetallic compounds, $\text{RNi}_2\text{B}_2\text{C} = (\text{RC})_1(\text{NiB})_2$, with R being a rare-earth element [1–8], another similar family, namely $\text{RNiBC} = (\text{RC})_2(\text{NiB})_2$ was discovered [9–14]. The coexistence or interplay between superconductivity and magnetically ordered structures in $\text{RNi}_2\text{B}_2\text{C}$ compounds was the main reason for the great interest in this class of materials. Therefore, it has been very disappointing that so far in the whole family of RNiBC compounds only LuNiBC has been reported to be superconducting [12]. The structures of both classes of compounds, on the other hand, are quite similar: while in $(\text{RC})_1(\text{NiB})_2$ there are alternating R–C and $\text{Ni}_2\text{--B}_2$ layers, an additional R–C layer exists between the $\text{Ni}_2\text{--B}_2$ layers in $(\text{RC})_2(\text{NiB})_2$; thus, in both classes of compounds the $\text{Ni}_2\text{--B}_2$ layers which are responsible for superconductivity are sandwiched between two R–C layers. For that reason, one has to question why superconductivity does not occur in $(\text{RC})_2(\text{NiB})_2$ compounds (with the exception of LuNiBC alone). Band-structure calculations [15] seem to give the answer: the density of states (DOS) at the Fermi energy (E_F) is much lower in LuNiBC than in $\text{LuNi}_2\text{B}_2\text{C}$. Mattheiss *et al* [15] argue that this change in the DOS at E_F is caused by a change in the B–Ni–B bonding angle of the NiB_4 tetrahedra forming the $\text{Ni}_2\text{--B}_2$ layer.

As was shown in previous papers, ^{57}Fe Mössbauer spectroscopy of ^{57}Fe sitting at the Ni site in $\text{RNi}_2\text{B}_2\text{C}$ compounds can provide information on the local structure via the electrical quadrupole splitting ΔE_Q [16] as well as on the magnetic structure via the transferred magnetic hyperfine field [17–19]. It has been found that ΔE_Q varies linearly with the c/a ratio (c and a being the lattice parameters in the c - and a -directions, respectively). It was the intention of this work to study the electrical quadrupole interaction at the Fe(Ni) site in RNiBC compounds and to see whether it can be correlated with that previously observed in the $\text{RNi}_2\text{B}_2\text{C}$ compounds. If such a correlation exists it will give us information on connections between the local structures at the Ni site in RNiBC and $\text{RNi}_2\text{B}_2\text{C}$.

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The $\text{RNi}_2\text{B}_2\text{C}$ and RNiBC samples were doped with 0.5 at.% ^{57}Fe , located at the Ni sites. The RNiBC compounds are more difficult to prepare than the corresponding $\text{RNi}_2\text{B}_2\text{C}$ series. Making several batches was necessary to optimize the preparation conditions and to obtain good quality samples, as was confirmed by x-ray and magnetic measurements [14]. The optimum preparation conditions have been found to be as follows: the samples were prepared by arc melting stoichiometric amounts of the pure elements in a partial argon atmosphere in a sealed quartz tube at 1000°C for two days and then quenched in liquid nitrogen. ^{57}Fe Mössbauer effect (ME) spectra were taken with the RNiBC samples and the $^{57}\text{Co}:\text{Rh}$ source at room temperature, moving in a sinusoidal mode outside the cryostat.

Figure 1 shows the ^{57}Fe ME spectra for all RNiBC compounds studied ($R = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{and Er}$) taken at room temperature, i.e. well above their magnetic ordering temperatures [14]. The asymmetry of the quadrupole doublets results from a weak impurity line (about 10% of the absorption area) located at about 0.1 mm s^{-1} . The impurity phase is also seen in the x-ray diffraction data, and may be related to a Ni-B phase because it does not depend on the R atoms. Since in our preparation procedure a mixture with stoichiometric amounts of Ni, ^{57}Fe , and B is melted, some fraction may not react properly, leading to the spurious phase.

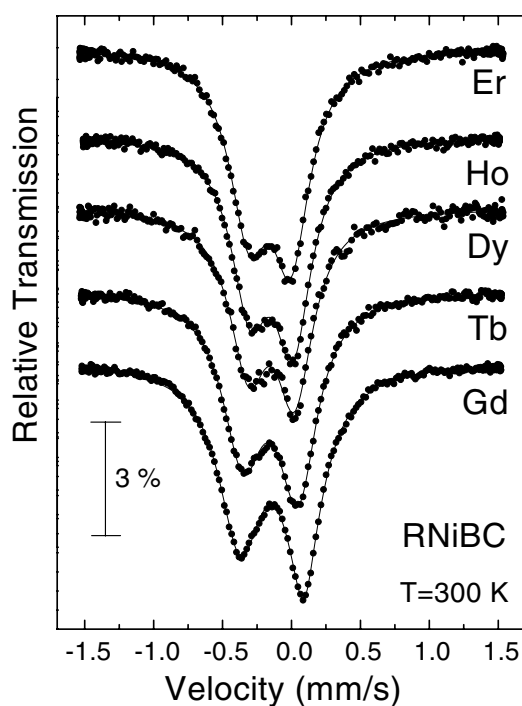


Figure 1. ^{57}Fe Mössbauer spectra of RNiBC ($R = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}$) compounds doped with 0.5 at.% ^{57}Fe taken at room temperature. The lines through the data points are least-squares fittings with a quadrupole doublet and a weak impurity line located at a Doppler velocity of about 0.1 mm s^{-1} .

From figure 1 one clearly sees an increase in the quadrupole splitting $|\Delta E_Q|$ going from Er to Gd. In order to compare the experimentally observed $|\Delta E_Q|$ values for RNiBC compounds with those obtained for the $\text{RNi}_2\text{B}_2\text{C}$ compounds we have to find the relevant lattice parameters which determine ΔE_Q . In figures 2(a) and 2(b) we show the crystal structures for the RNiBC and $\text{RNi}_2\text{B}_2\text{C}$ compounds, respectively, together with the relevant lattice parameters. In the

case of $\text{RNi}_2\text{B}_2\text{C}$ compounds it has been found that $|\Delta E_Q|$ depends linearly on the c/a ratio. In these compounds c is twice the separation c' of the R–C layers between which the $\text{Ni}_2\text{–B}_2$ layer is sandwiched, i.e. $c' = 0.5c$ (see figure 2(b)). The corresponding distance c' for RNiBC compounds, on the other hand, is $c' = c - d$, with d being the distance between the two adjacent R–C layers (see figure 2(a)). If we consider that the local structure at the Ni site is essentially given by the B–Ni–B bonding angles φ within the NiB_4 tetrahedra forming the $\text{Ni}_2\text{–B}_2$ layers (see figures 2(a) and 2(b)), it becomes obvious that it is not c/a but c'/a that is the relevant parameter which determines $|\Delta E_Q|$.

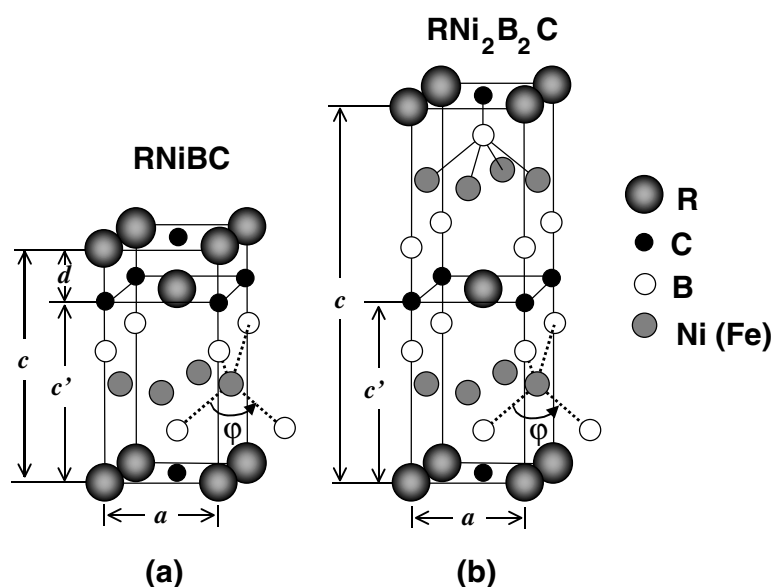


Figure 2. Crystal structures of the unit cell of (a) RNiBC and (b) $\text{RNi}_2\text{B}_2\text{C}$ compounds. The relevant lattice parameters used in the text are indicated.

We have plotted in figure 3 all experimentally observed $|\Delta E_Q|$ values for the RNiBC compounds together with the already published values for $|\Delta E_Q|$ for $\text{RNi}_2\text{B}_2\text{C}$ ($\text{R} = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{and Er}$) [16] and two additional measured but not yet published values of $|\Delta E_Q|$ for $\text{PrNi}_2\text{B}_2\text{C}$ and $\text{NdNi}_2\text{B}_2\text{C}$ as a function of the parameter c'/a . The $|\Delta E_Q|$ values are those taken at room temperature. A value of $c' = 5.14 \text{ \AA}$ has been used for all RNiBC compounds studied since the lattice parameters c and d are essentially the same ($c = 7.55 \text{ \AA}$ and $d = 2.41 \text{ \AA}$ [14]) for these compounds. As can be seen in figure 3, all $|\Delta E_Q|$ data points fall on the same straight line, i.e. all $|\Delta E_Q|$ values show the same linear correlation between $|\Delta E_Q|$ and c'/a . This is a clear indication that there is no difference in the local structure at the Fe(Ni) site between RNiBC and $\text{RNi}_2\text{B}_2\text{C}$ in the case where their c'/a ratios are the same. For example, $\text{GdNi}_2\text{B}_2\text{C}$ and ErNiBC which have the same c'/a ratio also show the same quadrupole splitting.

First-principles density-functional calculations of the electric field gradient V_{zz} for ^{57}Fe at the Ni site in $\text{RNi}_2\text{B}_2\text{C}$ compounds have been performed [16]. These calculations show that V_{zz} is composed of two contributions having the same order of magnitude but different signs: the so-called ‘valence’ contribution, essentially arising from the ^{57}Fe valence electrons, and the contribution resulting from the nuclei of the atoms surrounding Fe (usually called the ‘lattice’ contribution). There is a very good agreement as regards sign and magnitude between the measured value of ΔE_Q and the calculated value $\Delta E_Q = (1/2)eV_{zz}Q$ (Q is the ^{57}Fe

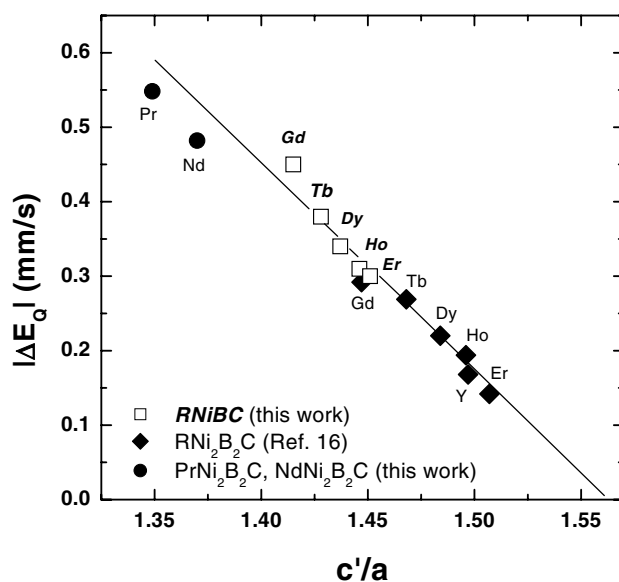


Figure 3. Quadrupole splittings $|\Delta E_Q|$ observed in RNiBC ($R = \text{Gd, Tb, Dy, Ho, and Er}$) compounds as a function of the c'/a ratio. The previously published $|\Delta E_Q|$ values for $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Gd, Tb, Dy, Ho, Er}$) compounds (see reference [16]) and unpublished $|\Delta E_Q|$ values for $\text{NdNi}_2\text{B}_2\text{C}$ and $\text{TmNi}_2\text{B}_2\text{C}$ have been added. All $|\Delta E_Q|$ values given are room temperature values.

nuclear quadrupole moment). Our new experimental finding—namely, that the ΔE_Q values for RNiBC and $\text{RNi}_2\text{B}_2\text{C}$ compounds do not differ if their c'/a ratios are the same—therefore indicates that not only the local structure (symmetry) but also the electronic structure of the $^{57}\text{Fe}(\text{Ni})$ atoms in RNiBC and $\text{RNi}_2\text{B}_2\text{C}$ compounds are the same for a given c'/a .

For an ideal local tetrahedral symmetry the electric field gradient is zero and thus $\Delta E_Q = 0$. Large $|\Delta E_Q|$ values at the Ni site, therefore, mean large deviations from the ideal tetrahedral symmetry of the NiB_4 tetrahedra. With the RNiBC compounds all having much larger $|\Delta E_Q|$ values than the corresponding $\text{RNi}_2\text{B}_2\text{C}$ compounds (see figure 2), it follows that the NiB_4 tetrahedra in RNiBC are further away from the ideal tetrahedral symmetry than those in the corresponding $\text{RNi}_2\text{B}_2\text{C}$ compounds. Since for an ideal tetrahedral symmetry of the NiB_4 tetrahedra there is a large DOS at E_F [15] which is favourable for superconductivity, it becomes quite evident that superconductivity is less favourable in RNiBC compounds than in the corresponding $\text{RNi}_2\text{B}_2\text{C}$ compounds.

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