CHANGE OF CURIE TEMPERATURES AND EFFECTIVE EXCHANGE FIELDS IN FERRIMAGNETIC $R_2Fe_{14}B$ COMPOUNDS UPON HYDROGENATION

Tereshina I.S.*, Tereshina E.A.[1], Nikitin S.A.[1], Chistyakov O.D., Burkhanov G.S., Drulis H.[2]

Baikov Institute of Metallurgy and Material Science RAS Leninski pr. 49, Moscow, 119991, Russia

[1] Department of Physics, Lomonosov Moscow State University, Leninskie Gory, Moscow,119992, Russia

[2] Institute of Low Temperatures and Structure Research PAS, ul. Okolna 2, Wroclaw, 50-950, Poland

* Fax: (095) 135 4438 E-mail: teresh@ultra.imet.ac.ru

Introduction

The insertion of light interstitial elements in the $R_2Fe_{14}B$ (R – rare-earth metal and Y) intermetallic compound's crystalline lattice results in an apparent change of their magnetic properties (the Curie temperature, saturation magnetization, magnetic anisotropy and spin-reorientation transitions) [1]. The purpose of the work is to study the influence of hydrogenation on exchange interactions (Fe – Fe, R – Fe) of $R_2Fe_{14}B$ compounds.

Results and discussion

High-purity rare-earth metals purified by vacuum distillation-sublimation (the content of basis metal is 99.956-99.983 at %), Armco iron, and Fe-(1.2-1.5 wt %)B alloying composition were used as starting components. The single-phase composition and directional structure of the synthesized $R_2Fe_{14}B$ (R = Gd, Tb, Dy, Ho, Er and Y) compounds were reached using threefold arc melting (nonconsumable tungsten electrode and copper water-cooled bottom) in a purified helium atmosphere and rather rapid cooling (that allows us to exclude the primary crystallization of α-Fe; at the same time, the solidification should not be very rapid to allow the occurrence of peritectic reaction for the formation of principal magnetic $Nd_2Fe_{14}B$-type phase). Samples placed in evacuated quartz ampoules were annealed for homogenization at 700ºC for 200 h. The samples were subjected to hydrogenation using the high-purity hydrogen (containing no more than 10^{-3}-10^{-4}% impurities) to the R/H proportion varied from 2.3 to 2.5. Such hydrogen content allowed us to hold the single-crystal structure of grains.

Thermomagnetic analysis was used to measure the temperature of magnetic ordering (T_C) of the host compounds and their hydrides. A substantial increase of the magnetic ordering temperatures upon hydrogenation was observed. For the host compounds $R_2Fe_{14}B$ and their hydrides the Curie temperatures decrease in the turn of R = Gd, Tb, Dy, Ho, Er, Lu (the investigations of the $Lu_2Fe_{14}B$ and $Lu_2Fe_{14}BH_{2.5}$ compounds has been described in the previous report [2]).

The Curie temperature is governed by three kinds of exchange interactions: Fe-Fe, R-Fe and R-R. It is known that the exchange interaction between iron atoms is stronger than the exchange interaction between the iron and rare-earth (R – Fe coupling) sublattices in these compounds. One of the ways to obtain information about the R-Fe intersublattice interaction is to compare the Curie temperatures of compounds containing magnetic and nonmagnetic R atoms and subsequently calculate the exchange field, induced to the rare-earth sublattice from the iron sublattice h_{21} by analyzing the Curie temperatures in terms of the molecular-field model [3].

The results of the calculations are summed up in Tables 1 and 2 for $R_2Fe_{14}B$ and their hydrides, respectively.

Table 1.

Compounds	T_C, K	h_{21}, 10^6 Oe	h_{11}, 10^6 Oe
$Gd_2Fe_{14}B$	661	1.14	-
$Tb_2Fe_{14}B$	620	1.11	-
$Dy_2Fe_{14}B$	598	1.14	-
$Ho_2Fe_{14}B$	573	1.09	-
$Er_2Fe_{14}B$	554	1.00	-
$Lu_2Fe_{14}B$	535	-	5.93
$Y_2Fe_{14}B$	565	-	6.26

The analysis of obtained results permits to conclude following:

(1) Introduction of hydrogen atoms into the crystal lattice of $R_2Fe_{14}B$ compounds leads to increase the Fe-Fe exchange interactions.

(2) Hydrogenation does not practically change the intersublattice exchange field h_{21}.

Table 2.

Compounds	T_C, K	h_{21}, 10^6 Oe	h_{11}, 10^6 Oe
$Gd_2Fe_{14}BH_{2.5}$	746	1.12	-
$Tb_2Fe_{14}BH_{2.5}$	705	1.10	-
$Dy_2Fe_{14}BH_{2.5}$	683	1.13	-
$Ho_2Fe_{14}BH_{2.5}$	658	1.09	-
$Er_2Fe_{14}BH_{2.5}$	639	1.01	-
$Lu_2Fe_{14}BH_{2.5}$	620	-	6.50
$Y_2Fe_{14}BH_{2.5}$	650	-	6.81

According to the theory, taking into account the spin fluctuations, the Curie temperature of band ferromagnets is inversely to the density of states in the subbands near the Fermi level, $N\uparrow(E_F)$ and $N\downarrow(E_F)$. The increase in the lattice parameters observed for incorporation of hydrogen atoms induces the decrease in the degree of wave-function hybridization of 3*d*-electrons of Fe atoms and 5*d*-electrons of rare-earth metal atoms and entails to decrease in densities of states in both zones $N\uparrow(E_F)$ and $N\downarrow(E_F)$. This decrease in the degree of hybridization apparently plays an important role in suppressing of the spin fluctuations and in the Curie temperature increase. The exchange interactions between ions in the iron sublattice (the exchange field h_{11}) make the main contribution in the Curie temperature. Furthermore, the insertion of hydrogen atoms doesn't change the intersublattice exchange interactions in $R_2Fe_{14}B$ intermetallic compounds of 4f- and 3d- transition metals. Interpretation of this result should take into account a number of factors: (i) the volume effect associated with the increase in the unit cell volume and in the Fe–Fe and *R*–Fe interatomic distances under incorporation of interstitial atoms, (ii) the enhanced magnetism of the iron sublattice, (iii) the elastic strains created by the incorporated hydrogen atom, (iv) the change in the local electron concentration around the interstitial atom (the chemical effect). It can be maintained that all these effects could compensate each other and lead to the intersublattice exchange interaction remain almost invariable.

Conclusions

Thus, we established that incorporation of hydrogen atoms into the $R_2Fe_{14}B$ compounds crystal lattice leads to a substantial increase in the Curie temperature T_C, an enhancement of the Fe–Fe exchange interaction and a conservation of R-Fe exchange interactions, which can be explained by the change in the unit cell volume and electronic structure of the compounds.

The work has been supported by the Federal Program on Support of Leading Scientific School Grant NSH -205.2003.2 and RFBR Grant № 04-03-32194.

References

1. Pourarian F. Review on the influence of hydrogen on the magnetism of alloys based on rare earth – transition metal systems. Physica B 2002;321:18-28.
2. Tereshina I.S., Andreev A.V., Tereshina E.A., Drulis H., Effect of hydrogen on magnetic properties of $Lu_2Fe_{14}B$ single crystal. J. Alloys Comp. 2005 (in press).
3. Nikitin S.A., Tereshina I.S., Effect of interstitial atoms on the effective exchange fields in ferromagnetic rare-earth and 3d transition metal compounds R_2Fe_{17} and $RFe_{11}Ti$. Fizika tverdogo tela 2003;45(10):1850-1856.