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MÖSSBAUER EFFECT STUDIES OF $Nd_2Fe_{14-x}Si_xB$ AND $Y_2Fe_{14-x}Si_xB$

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$R_2Fe_{14-x}T_xB$ magnets, where R is yttrium or a rare earth and T is a transition metal or metalloid, are of interest because real-world magnets may be made using such substitutions, and because systematic investigation of their effects may lead to a better understanding of the origins of the desirable $R_2Fe_{14}B$ magnetic properties. There are six different iron sites in $Nd_2Fe_{14}B$, each having different local environments. The iron-iron distances cover a wide range of values. It is believed that short iron-iron distances ($d_{Fe\text{-}Fe} < 2.45$Å) and their resulting negative exchange interactions are a cause of the relatively low Curie temperature of $Nd_2Fe_{14}B$. In particular, the interactions between $8j_1$-$16k_2$ iron atoms are strongly negative, and the interactions between $8j_1$-$8j_2$ and $8j_2$-$16k_1$ are weakly negative. The Curie temperature of $Nd_2Fe_{14}B$ is little more than half that of pure iron. Replacement of iron by silicon in $Nd_2Fe_{14}B$, $Pr_2Fe_{14}B$, and $Er_2Fe_{14}B$ has the surprising effect of raising the Curie temperature of these compounds.[1] One might expect that the substitution of iron by nonmagnetic silicon would lower the Curie temperature. Thus, it was suggested that silicon must preferentially occupy iron sites which contribute predominately to the negative iron-iron exchange interactions, namely the $16k_2$ and $8j_1$ sites.[1]

We have carried out a Mössbauer effect investigation of $Nd_2Fe_{14-x}Si_xB$ for x equal to 0.0, 0.25, 0.50, 0.75, 1.0, 1.5, and 2.0, and of $Y_2Fe_{14-x}Si_xB$ for x equal to 0.0, 1.0, 1.5, and 2.0. The $R_2Fe_{14}B$ Mössbauer spectra are composed of six overlapping sextets. Each sextet corresponds to one of the six different iron sites, and the relative areas of the sextets are proportional to the occupancies of iron on the corresponding site. Thus, Mössbauer effect measurements can be useful for investigating preferential site occupancies in these compounds. Our initial fits of the $Nd_2Fe_{14-x}Si_xB$ and $Y_2Fe_{14-x}Si_xB$ Mössbauer spectra, made by assuming random occupancy of silicon on the different iron sites, clearly indicated a non-random occupancy. A series of trial fits with different silicon occupancies eventally led to good results with silicon primarily substituted on the $16k_2$ and $8j_1$ sites, and also partially on the $16k_1$ sites. Figure 1 shows the room temperature Mössbauer spectra and fits for $Nd_2Fe_{14}B$ and $Nd_2Fe_{12}Si_2B$. Thus, our Mössbauer effect measurements appear to confirm the silicon occupancy proposed by Pedziwiatr and Wallace.[1] The spectra shown in Figure 1 and those for the other silicon substituted compounds indicate a distinct increase in linewidths and a decrease in the internal hyperfine fields with increasing silicon content. Both of these changes indicate a distribution of the hyperfine field on a given site which is related to the number of near neighbor silicon atoms. In an alternative approach we have fit the observed spectra with a model in which a binomial distribution of silicon near neighbor atoms, in conjunction with a reduced transferred hyperfine field in the presence of the silicon, defines the distribution of hyperfine fields for each iron site.

Our fits for the $Nd_2Fe_{14-x}Si_xB$ and $Y_2Fe_{14-x}Si_xB$ spectra are consistent with a recently-developed model for the Mössbauer spectra of $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$.[2] In this model, the principal axis of the electric field gradient tensor is defined by the location of the near neighbor neodymium atoms. The composition dependence of the hyperfine parameters obtained from our fits are normal and consistent with this model. The $8j_2$ site has the largest volume of all the iron sites in $Nd_2Fe_{14}B$, and the $16k_2$ site has the smallest volume. The $8j_1$ and $16k_1$ sites are intermediate volume sites. Our analysis suggests that silicon prefers to occupy lower-volume sites in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$.

Caution is required when using Mössbauer spectral fits in assigning preferential site occupancies. Different models can produce calculated spectra which fit the observed spectra equally well. A series of fits in which we assigned silicon occupancy exclusively to the $16k_1$ site, and another series of fits in which we assigned silicon occupancy equally to the $16k_1$ and $16k_2$ sites, both produced fits as satisfactory as those shown in Figure 1. Further, a series of fits in which silicon was assigned exclusively to the $8j_2$ site was clearly wrong only for higher silicon concentrantions. In a case such as this, it may be better to use the model with a distribution of hyperfine fields to evaluate the different possible silicon site occupancies. If the silicon occupancy is known, such as from neutron diffraction, it may then be possible to determine the exact extent to which the silicon reduces the transferred hyperfine field at a specific iron site.

1. A. T. Pedziwiatr and W. E. Wallace, J. Less-Common Met. 126, 41 (1986).
2. F. Grandjean, G. J. Long, D. E. Tharp, O. A. Pringle, and W. J. James, J. Phys. (Paris) Colloq., in press.

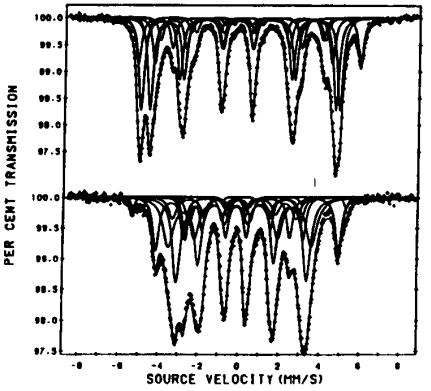

Figure 1. Room temperature Mössbauer spectra and fits for $Nd_2Fe_{14}B$ (upper) and $Nd_2Fe_{12}Si_2B$ (lower).