

Why the iron magnetization in $\text{Gd}_2\text{Fe}_{14}\text{B}$ and the spontaneous magnetization of $\text{Y}_2\text{Fe}_{14}\text{B}$ depend on temperature differently

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It is demonstrated that the temperature dependence of the iron sublattice magnetization in $\text{Gd}_2\text{Fe}_{14}\text{B}$ is affected significantly by the Gd–Fe exchange interaction. This is at variance with the common perception that $M_{\text{Fe}}(T)$ in iron-rich rare-earth intermetallics is determined predominantly by the Fe–Fe exchange. This phenomenon is discussed by considering the modification of the low-energy spin-wave spectrum of $\text{Gd}_2\text{Fe}_{14}\text{B}$, as compared to that of $\text{Y}_2\text{Fe}_{14}\text{B}$, under the influence of the Gd–Fe interaction. The result is of particular significance for evaluating the temperature dependence of the magnetocrystalline anisotropy of iron or cobalt compounds with anisotropic rare earths (e.g., $\text{Nd}_2\text{Fe}_{14}\text{B}$) and in turn, of the hard magnetic properties of such compounds. © 2010 American Institute of Physics. [doi:10.1063/1.3443576]

I. INTRODUCTION

In permanent magnet applications it is important that the material maintains its strong magnetic anisotropy at elevated temperatures. As far as modern, $3d$ - $4f$ intermetallic-based hard magnetic materials are concerned, the temperature endurance of their anisotropy is determined by two principal factors—the leading coefficient of the crystal field on the rare earth in these uniaxial compounds, A_{20} , and the $3d$ - $4f$ exchange field seen by the rare earth, B_{ex} . Namely, the main contribution to the anisotropy at or above room temperature is proportional to the product $A_{20}B_{\text{ex}}^2$.¹ A_{20} is an essentially temperature-independent material constant, whereas, B_{ex} does depend on temperature in an important way. The fact that the form of this dependence is not exactly known is a major obstacle to calculations of the magnetic properties in the room-temperature range.

The intensities of the main exchange interactions in iron-rich and cobalt-rich intermetallics are so related that the $3d$ - $3d$ exchange is the strongest. This interaction is generally thought to determine, practically alone, the temperature evolution of the $3d$ magnetization, $M_{3d}(T)$, including the point where the latter vanishes, T_C . Second comes the $3d$ - $4f$ exchange; it plays a decisive role in polarizing the lanthanide sublattice, while having only a minor effect on $M_{3d}(T)$. The $4f$ - $4f$ interaction is the weakest of all and can be safely neglected.² In this situation the influence of the $3d$ - $4f$ exchange on the rare earth sublattice can be described by means of an effective exchange field, $B_{\text{ex}} = n_{3d-4f} M_{3d}(T)$, where n_{3d-4f} is a constant. In the simplest approximation one neglects the repercussion of the $3d$ - $4f$ exchange on $M_{3d}(T)$; the latter should then equal the same dependence in an isomorphous compound with a nonmagnetic rare earth, where it

can be determined experimentally. The main objective of the present work is to find out to what extent this neglect is justified.

$\text{Gd}_2\text{Fe}_{14}\text{B}$ has been chosen as an object of study for an obvious reason that it has the same structure as the celebrated permanent-magnet materials $\text{Nd}_2\text{Fe}_{14}\text{B}$ and $\text{Pr}_2\text{Fe}_{14}\text{B}$. An earlier work³ had found the difference of the spontaneous magnetizations of $\text{Gd}_2\text{Fe}_{14}\text{B}$ (thought to equal $M_{\text{Fe}} - M_{\text{Gd}}$) and $\text{Y}_2\text{Fe}_{14}\text{B}$ ($\equiv M_{\text{Fe}}$)—both regarded as functions of reduced temperature, T/T_C , with their respective T_C 's—to follow the Brillouin function, $B_{7/2}(7\mu_B B_{\text{ex}}/kT)$, the exchange field on Gd B_{ex} being proportional to M_{Fe} . This seemed to have confirmed the often professed view that, apart from a slight rescaling, the iron sublattice magnetization $M_{\text{Fe}}(T/T_C)$ in both compounds is basically the same. The maximum value of B_{ex} , corresponding to $T \rightarrow 0$, was determined to be 276 T.³ A problem with this interpretation arose a decade later, when an inelastic neutron scattering experiment found in $\text{Gd}_2\text{Fe}_{14}\text{B}$ a significantly larger exchange field, $B_{\text{ex}} = 318$ T at $T = 14$ K.⁴ (The same experiment⁴ demonstrated that the exchange fields seen by Gd atoms situated on the two nonequivalent sites in $\text{Gd}_2\text{Fe}_{14}\text{B}$ differ by no more than a few percent so that it is a valid approximation to consider just one exchange field acting on all Gd atoms.) It will be demonstrated below that the discrepancy is a manifestation of the falsity of the assumption made in Ref. 3; in fact, $M_{\text{Fe}}(T/T_C)$ in $\text{Y}_2\text{Fe}_{14}\text{B}$ and in $\text{Gd}_2\text{Fe}_{14}\text{B}$ is not the same.

The quantity under study herein is spontaneous magnetization. As a new tool for data analysis we intend to employ the following expression for $M_{\text{Fe}}(T)$ (Ref. 5):

$$M_{\text{Fe}}(T) = M_{\text{Fe}}(0) \left[1 - s \left(\frac{T}{T_C} \right)^{3/2} - (1-s) \left(\frac{T}{T_C} \right)^{5/2} \right]^{1/3}. \quad (1)$$

This formula has been found to describe well the temperature dependence of spontaneous magnetization of several ferro-

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magnetic intermetallics with nonmagnetic rare earths: YCo_5 and Y_2Fe_{17} ,⁵ $\text{Y}_2\text{Fe}_{14}\text{Si}_3$,⁶ $\text{Lu}_2\text{Co}_{17-x}\text{Si}_x$, including pure $\text{Lu}_2\text{Co}_{17}$.⁷ Two goals are achieved by applying Eq. (1) to $\text{Gd}_2\text{Fe}_{14}\text{B}$. First, the formula is subjected to a more stringent test than in the case of ferromagnets because the spontaneous magnetization of the ferrimagnet $\text{Gd}_2\text{Fe}_{14}\text{B}$ is a difference of the sublattice magnetizations, $M_{\text{Fe}} - M_{\text{Gd}}$, Eq. (1) entering both in the minuend M_{Fe} and in the subtrahend M_{Gd} (through the exchange field in the argument of the Brillouin function, $B_{\text{ex}} = n_{\text{GdFe}} M_{\text{Fe}}$). Second, Eq. (1) enables us to couch in simple mathematical terms the question stated above, namely, whether the functions $M_{\text{Fe}}(T)$ in $\text{Y}_2\text{Fe}_{14}\text{B}$ and in $\text{Gd}_2\text{Fe}_{14}\text{B}$ differ in anything other than scale. Now we wish to know if the two compounds are described by Eq. (1) with the same shape parameter s or with two different s .

II. EXPERIMENTAL DETAILS

The single crystals of $\text{Gd}_2\text{Fe}_{14}\text{B}$ and $\text{Y}_2\text{Fe}_{14}\text{B}$ used in this study were grown at Laboratoire Louis Néel, Grenoble (today Institut Néel) by the Czochralski technique from a levitating melt in a so called cold crucible.³ The crystals were oriented by Laue x-ray diffraction and shaped using spark erosion. The $\text{Y}_2\text{Fe}_{14}\text{B}$ single crystal was a 2 mm diameter sphere whereas the $\text{Gd}_2\text{Fe}_{14}\text{B}$ single crystal was a 3.5 mm cube with edges along the [001], [100], and [010] crystallographic directions. Magnetization measurements were performed in magnetic fields up to 5 T applied along the easy magnetization axis using a vibrating-sample magnetometer (VSM) (PPMS Quantum Design) ($5 \text{ K} < T < 400 \text{ K}$) and using a home made extraction magnetometer ($300 \text{ K} < T < 700 \text{ K}$). For better temperature control the crystals were mounted on a Cu sample holder. The difference between the set temperature and the temperature of the sample was estimated to be of the order of 0.1 K below room temperature and no more than 2 K at the highest measurement temperature. The excellent overlap between both sets of data in the 300 to 400 K temperature range, together with the reproducibility in the Curie point found for Ni (633 K), gave us confidence in the accuracy of the thermometry. The error in the measured magnetization value was estimated to be below 2%.

III. DATA TREATMENT

The spontaneous magnetization M_s was determined using the technique due to Belov and Goryaga⁸ (in Russian, see also Belov's monograph⁹), often referred to as Arrott's plot. Figures 1 and 2 illustrate the method for $\text{Y}_2\text{Fe}_{14}\text{B}$ at three representative temperatures: Fig. 1 displays the original experimental magnetization curves, while Fig. 2 presents the same data as H/M versus M^2 . The upper (higher-field) portions of the latter curves were extrapolated to zero internal field to yield M_s^2 . In Figs. 1 and 2, H stands for external magnetic field. In order to convert Fig. 2 to internal magnetic field, a demagnetization correction (for a sphere), $(4\pi/3) \times (4 \text{ f.u.}/V_{\text{cell}})$, should be deducted from all the ordinates. Alternatively, the abscissa axis could be shifted upward by the same amount; its new position is indicated with a dashed line in Fig. 2. Thus, the values of M_s^2 were determined as the

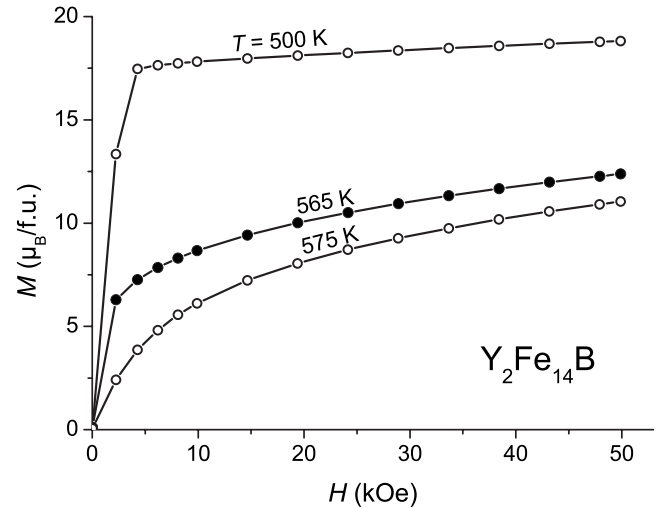


FIG. 1. Selected magnetization curves of $\text{Y}_2\text{Fe}_{14}\text{B}$. The lines are guides for the eye.

abscissas of the crossing-points of the dashed line with the fits to the data. This is marked with an arrow on the middle curve.

The left-most curve in Fig. 2 yields a negative intercept, which corresponds to $T > T_C$. One readily recognizes that the T_C of $\text{Y}_2\text{Fe}_{14}\text{B}$ lies between 565 and 575 K.

The low-field parts of the data sets in Fig. 2, strongly curved on account of the domain phenomena, were not included in the fits. More precisely, all data points whose ordinates were less than three times the demagnetization correction (i.e., three times the ordinate of the dashed line in Fig. 2) were ignored by the fitting routine. The weak intrinsic curvature of the upper portions of the Belov–Goryaga plots was allowed for by using quadratic polynomial fits. This was only necessary at high temperatures near T_C (above $T = 400 \text{ K}$ for $\text{Y}_2\text{Fe}_{14}\text{B}$ and above $T = 600 \text{ K}$ for $\text{Gd}_2\text{Fe}_{14}\text{B}$). At lower temperatures the intrinsic curvature was negligible and the more stable linear extrapolation was used.

IV. RESULTS AND DISCUSSION

Figure 3 presents the spontaneous magnetization of $\text{Y}_2\text{Fe}_{14}\text{B}$ plotted against temperature. The continuous line is

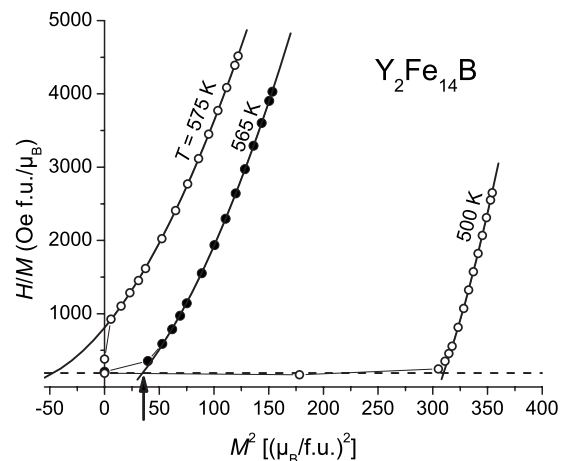


FIG. 2. The same data as in Fig. 1 but presented as H/M vs M^2 . Solid lines are quadratic polynomial fits to the data points. The arrow shows the square of the spontaneous magnetization at $T = 565 \text{ K}$. The dashed horizontal line represents the demagnetization correction (see text).

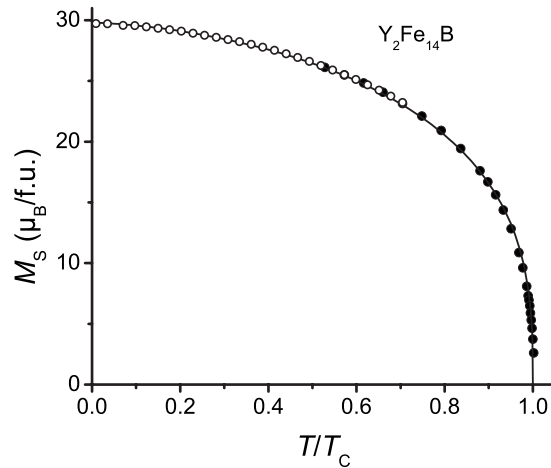


FIG. 3. Spontaneous magnetization of $\text{Y}_2\text{Fe}_{14}\text{B}$ vs reduced temperature. Open circles correspond to data measured with the low temperature VSM, and black dots to data with the high temperature extraction magnetometer. Solid line is a fit to Eq. (1) with $M_{\text{Fe}}(0)=29.8 \mu_{\text{B}}/\text{f.u.}$ and $s=0.7$.

a fit to Eq. (1) with $M_{\text{Fe}}(0)=29.8 \mu_{\text{B}}/\text{f.u.}$, $T_{\text{C}}=568 \text{ K}$, and $s=0.7$. In practice the fitting involved only a minor adjustment of $M_{\text{Fe}}(0)$ and T_{C} , our best-fit values deviating only slightly from those known from the literature, $M_{\text{Fe}}(0)=29.6 \mu_{\text{B}}/\text{f.u.}$ (Ref. 3) and $T_{\text{C}}=571 \text{ K}$.¹⁰ The shape parameter s was varied rather more freely, but the final result is not unlike the values encountered in other similar intermetallics [cf. $s=0.7$ for YCo_5 or $s=0.6$ for Y_2Fe_{17} (Ref. 5)].

The spontaneous magnetization of $\text{Gd}_2\text{Fe}_{14}\text{B}$ is presented in Fig. 4. The solid line is a fit to the following expression:

$$M_s(T) = M_{\text{Fe}}(T) - 14(\mu_{\text{B}}/\text{f.u.})\text{B}_{7/2}(x). \quad (2)$$

Here $M_{\text{Fe}}(T)$ is given by Eq. (1) and $\text{B}_{7/2}(x)$ stands for the Brillouin function,

$$\text{B}_{7/2}(x) = \frac{8}{7} \coth\left(\frac{8}{7}x\right) - \frac{1}{7} \coth\left(\frac{1}{7}x\right), \quad (3)$$

with

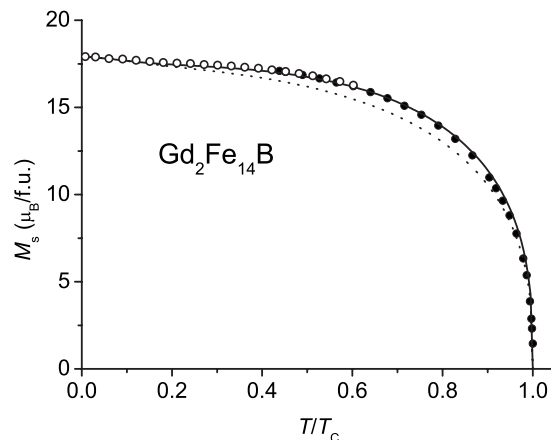


FIG. 4. Spontaneous magnetization of $\text{Gd}_2\text{Fe}_{14}\text{B}$ vs reduced temperature. Open circles correspond to data measured with the low-temperature VSM, and black dots to data with the high-temperature extraction magnetometer. Solid line is a fit to Eqs. (1)–(4) with $M_{\text{Fe}}(0)=31.9 \mu_{\text{B}}/\text{f.u.}$, $s=0.7$, and $B_{\text{ex}}(0)=270 \text{ T}$. Dotted line: the same as previous but with $B_{\text{ex}}(0)=318 \text{ T}$.

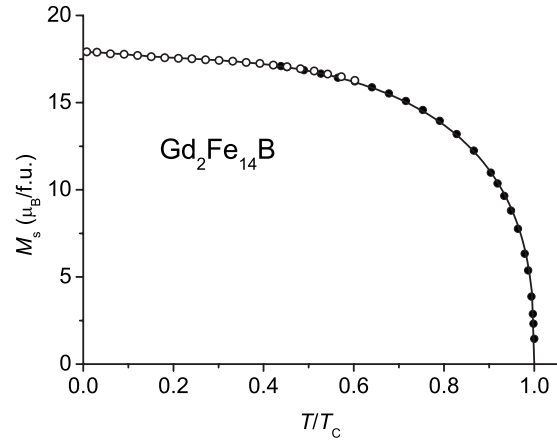


FIG. 5. The same as Fig. 4 but the fit is with $s=0.4$ and $B_{\text{ex}}(0)=318 \text{ T}$.

$$x = \frac{7\mu_{\text{B}}B_{\text{ex}}(0)M_{\text{Fe}}(T)}{kT M_{\text{Fe}}(0)}. \quad (4)$$

These expressions adopt for the Gd 0 K moment a free-ion value of $7 \mu_{\text{B}}$. The shape parameter used in the calculation was the same as that found for $\text{Y}_2\text{Fe}_{14}\text{B}$, $s=0.7$. The Curie point was set to 664 K, which is reasonably close to the handbook value, $T_{\text{C}}=660 \text{ K}$,¹⁰ representing the average of data from several independent sources. The best-fit values of the other adjustable parameters, $B_{\text{ex}}(0)=270 \text{ T}$ and $M_{\text{Fe}}(0)=31.9 \mu_{\text{B}}/\text{f.u.}$, are rather close to those found in Ref. 3, $B_{\text{ex}}(0)=276 \text{ T}$ and $M_{\text{Fe}}(0)=32 \mu_{\text{B}}/\text{f.u.}$

One observes in Fig. 4 satisfactory agreement between the experiment (symbols) and the fit (solid line). At this stage any numerical mistake in the analysis of Ref. 3 can be ruled out. Nor was it seriously affected by the limitation of the temperature range. All the more striking is the discrepancy between the result of the fitting, $B_{\text{ex}}(0)=270 \text{ T}$, and the trustworthy neutron-spectroscopic value, $B_{\text{ex}}=318 \text{ T}$.⁴ The difference is not insignificant—it will be recalled that room-temperature anisotropy scales as B_{ex}^2 .¹ A straightforward setting $B_{\text{ex}}(0)=318 \text{ T}$ without changing anything else in the above algorithm produces a prediction that lies some way off the data points (dotted line in Fig. 4). Clearly, if the spectroscopic exchange field is to be used in conjunction with the above expressions, some other parameter therein has to be changed in order to bring the output in line with experiment. This result thus shows, that the question raised at the outset of this letter was not in vain and that the presence of Gd–Fe interactions in $\text{Gd}_2\text{Fe}_{14}\text{B}$ does have an effect on the temperature dependence of the Fe magnetization.

The only disposable parameter in Eq. (1) is s . So we set $B_{\text{ex}}(0)=318 \text{ T}$ and let s vary. The best fit is found for $s=0.4$ (the other parameters as above), see Fig. 5, solid line. The agreement with experiment is better than in Fig. 4 and the parameters now have physically meaningful values. The fact that s has a smaller value in $\text{Gd}_2\text{Fe}_{14}\text{B}$ than in $\text{Y}_2\text{Fe}_{14}\text{B}$ implies that $M_{\text{Fe}}(T)$ in the former has a more rectangular shape (falling off more slowly at $T \ll T_{\text{C}}$ but more steeply at $T \approx T_{\text{C}}$) than in the latter. In very simple terms, the presence of magnetized Gd spins effectively prolongs the state of saturation in the Fe sublattice to higher temperatures.

At a more fundamental level, the shape of $M_{\text{Fe}}(T)$ at $T \ll T_C$ is governed by spin-wave excitations. These excitations are characterized by magnon dispersion curves $E(\mathbf{q})$ where E is the magnon energy and \mathbf{q} the wave vector. For an isotropic ferromagnet without applied magnetic field $E(\mathbf{q})$ is quadratic for q small: $E(\mathbf{q})=Dq^2$. The coefficient D , known as spin-wave stiffness, determines the prefactor of $T^{3/2}$ in Bloch's law,¹¹ which describes the reduction in spontaneous magnetization with temperature. By mapping Eq. (1) in the low-temperature limit onto Bloch's law, one arrives at the following expression for the shape parameter:¹²

$$s = 0.352 \frac{\mu_B}{M_{\text{Fe}}(0)} \left(\frac{kT_C}{D} \right)^{3/2}. \quad (5)$$

Here $M_{\text{Fe}}(0)$ stands for volume saturation magnetization. Equation (5) is just Eq. (9) of Ref. 12 with $g=2$ and $\beta = 1/3$. (Note that the g -factor is missing in Eq. (2) of Ref. 5.)

The spin-wave stiffness of $\text{Y}_2\text{Fe}_{14}\text{B}$ was determined from inelastic neutron experiments by Mayer *et al.*¹³ and communicated privately to the authors of Ref. 14 (Ref. 37 therein). From the data cited by Ried *et al.*¹⁴ by using their Eq. (35) we find

$$D_{\text{Fe}} = 2.13 \times 10^{-29} \text{ erg cm}^2. \quad (6)$$

Strictly speaking, spin-wave stiffness is a tensor quantity, the above value (6) referring to the fourfold axis of the tetragonal crystal (D_{\parallel}). By the symmetry, the other two tensor components, referring to the basal plane (D_{\perp}), equal each other but are generally distinct from the longitudinal component. Equation (5) must contain the mean geometric of all three principal values, $(D_{\parallel}D_{\perp}^2)^{1/3}$. Now the transversal spin-wave stiffness of $\text{Y}_2\text{Fe}_{14}\text{B}$ is unknown, but it is likely to be close to the longitudinal one. So in our estimations we simply set $D_{\perp}=D_{\parallel}$, as if the crystal symmetry were cubic. Then, substituting the value (6) for D and $1.2 \times 10^3 \text{ emu/cm}^3 \text{ kG}$ for $M_{\text{Fe}}(0)$ in Eq. (5), we obtain for $\text{Y}_2\text{Fe}_{14}\text{B}$ $s=0.6$. In view of the crudity of our calculations, the agreement with the value found from the fits, $s=0.7$, should be regarded as good.

Let us now turn to the ferrimagnet $\text{Gd}_2\text{Fe}_{14}\text{B}$. As well known,^{11,14,15} the spin-wave spectrum of ferrimagnets consists of an acoustical and an optical branches. At low temperatures only the bottom part of the acoustical branch is of relevance. If the applied field is nil and the magnetic anisotropy negligible, the (isotropic) dispersion relation is just $E = D_{\text{GdFe}}q^2$, the spin-wave stiffness being¹⁶

$$D_{\text{GdFe}} = \frac{M_{\text{Fe}}(0)}{M_s(0)} D_{\text{Fe}}, \quad (7)$$

with $M_s(0)=M_{\text{Fe}}(0)-M_{\text{Gd}}(0)$. Thus, even if the Fe–Fe exchange interaction in $\text{Gd}_2\text{Fe}_{14}\text{B}$ is exactly the same as in $\text{Y}_2\text{Fe}_{14}\text{B}$, the magnons responsible for the reduction in the spontaneous magnetization at $T \ll T_C$ are a factor of $M_{\text{Fe}}(0)/M_s(0)=1.78$ stiffer and therefore harder to excite. Note that in this temperature range all thermally excited magnons and the associated magnetization change $\propto T^{3/2}$ can be unambiguously ascribed to the iron sublattice, because the deviation of the gadolinium magnetization from its saturation

value is exponentially small. According to Eq. (5), the higher D means a smaller s for $\text{Gd}_2\text{Fe}_{14}\text{B}$. This effect is only slightly mitigated by the 1.17 times higher T_C , and yet more weakly enhanced by the 1.07 times higher $M_{\text{Fe}}(0)$ of $\text{Gd}_2\text{Fe}_{14}\text{B}$. All in all, Eq. (5) predicts that the shape parameter of $\text{Gd}_2\text{Fe}_{14}\text{B}$ should equal one-half of that of $\text{Y}_2\text{Fe}_{14}\text{B}$, i.e., 0.3. This is in reasonable agreement with $s=0.4$, obtained by fitting the magnetization data.

We have demonstrated in this paper that $M_{\text{Fe}}(T/T_C)$ in $\text{Gd}_2\text{Fe}_{14}\text{B}$ differs from $M_{\text{Fe}}(T/T_C)$ in $\text{Y}_2\text{Fe}_{14}\text{B}$. In both compounds the form of $M_{\text{Fe}}(T)$ is well reproduced by Eq. (1), but the shape parameters s differ by almost a factor of two. By means of Eqs. (5) and (7) the difference is traced back to the enhanced spin-wave stiffness at the bottom of the acoustical branch in $\text{Gd}_2\text{Fe}_{14}\text{B}$. A similar shift in s can be expected in other rare-earth-iron (or -cobalt) intermetallics, the sign and magnitude of this shift depending on the kind of compound and on whether the rare-earth moments are parallel or antiparallel to those of Fe or Co. This result implies that, unlike usually assumed, the R magnetization in a given M-rich R–M compound ($\text{Gd}_2\text{Fe}_{14}\text{B}$ in the present case) cannot be obtained by simple subtraction of the spontaneous magnetization in an isomorphous compound with a nonmagnetic R (Y or Lu) from the measured magnetization in the considered compound.

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