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Year 1996

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(pagedate: 2014-11-14)

ac susceptibility study on $R_2Fe_{14}B$ single crystals (R=Y, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm)

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(Received 6 March 1996)

The temperature dependence of the ac susceptibility $\chi=\chi'+i\chi''$ has been measured on $R_2Fe_{14}B$ single crystals with R=Y, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm as a function of temperature in the temperature interval from 4.2 to 300 K, as a function of the strength of the applied ac field (up to 800 A/m), as a function of frequency (ranging from 5 to 1000 Hz), as well as the dependence on the crystallographic orientation. A reversible rotation of magnetic moments, excited by applying the field perpendicular to the easy magnetization direction, is found to be the principal contribution to the susceptibility (χ'). The energy loss is fairly small in this case ($\chi''\approx 0$). When the external field is applied parallel to the easy magnetization direction, domain-wall movement is the main contribution to the susceptibility (χ') and to the energy loss (χ''). For $Nd_2Fe_{14}B$, a peak in $\chi''(T)$ is detected at 135 K, the spin-reorientation temperature, in a 5 Hz field of 40 A/m applied along the [001] direction. This peak disappears upon increasing the frequency or upon changing the crystallographic orientation of the crystal. In addition, an anomaly in the temperature dependence of the susceptibility is detected around 220 K for $R_2Fe_{14}B$ single crystals with R=Pr, Nd, Sm, Tb, and Dy. The presence of this anomaly depends on the crystallographic orientation. It can only be detected when the external field is applied parallel to the [001] direction. For all frequencies used, the temperature dependence of χ' is completely different from that of χ''. The χ' values are nearly independent of the frequency in our measuring range from 5 to 1000 Hz. However, the χ'' values increase with increasing frequency when the field is applied along the easy magnetization direction and decrease when the field is applied perpendicular to the easy magnetization direction. [S0163-1829(96)06233-9]

I. INTRODUCTION

The intrinsic magnetic properties of tetragonal $R_2Fe_{14}B$ compounds (space group $P4_2/mnm$) (Refs. 1 and 2) have been intensively investigated after the discovery of excellent permanent magnets based on $Nd_2Fe_{14}B$.[3,4] Concerning magnetic phase transitions and the magnetocrystalline anisotropy in $R_2Fe_{14}B$ compounds, the following aspects can serve as a brief summary. (1) The Fe-sublattice anisotropy in $R_2Fe_{14}B$ favors the c axis. However, the R-sublattice anisotropy depends strongly on the combined interactions of the crystalline electric field (CEF) and the exchange field acting on the R ions, which leads to temperature-induced spin-reorientation transitions in some cases. These transitions are either due to the competition between the strongly temperature dependent planar R-sublattice anisotropy and the less temperature-dependent uniaxial Fe-sublattice anisotropy [e.g., in $Er_2Fe_{14}B$ (T_{sr}=323 K) and $Tm_2Fe_{14}B$ (T_{sr}=315 K) (Ref. 5)] or due to the R-sublattice anisotropy only [e.g., in $Nd_2Fe_{14}B$ (T_{sr}=135 K) (Ref. 6) and $Ho_2Fe_{14}B$ (T_{sr}=57.6 K) (Ref. 5)]. In the latter case, the temperature-induced competition between various CEF terms is the intrinsic origin of the spin reorientation. (2) Field-induced first-order magnetization processes (FOMP's) have been observed in $Pr_2Fe_{14}B$ and $Nd_2Fe_{14}B$ at low temperatures [$<$220 K (Ref. 7)] when the field is applied in specific crystallographic directions.[8,9]

ac-susceptibility measurements are very sensitive in detecting magnetic phase transitions initiated by a change of the magnetocrystalline anisotropy energy. It is therefore expected that any change involving the spin configuration, the domain configuration as well as the magnetic anisotropy can be detected by measuring the ac susceptibility. For $Nd_2Fe_{14}B$, an anomaly was found at about 220 K in the temperature dependence of ac susceptibility, in sintered magnets,[10] in polycrystalline material,[7,11] and in magnetically aligned fine-powder samples.[12] It was found that this anomaly is most pronounced if the field is applied parallel to the alignment direction. The anomaly vanishes if the field is applied perpendicular to the alignment direction. It is worth noting that anomalies in the temperature dependence of the ac susceptibility have been found in nearly all R-T intermetallic compounds, e.g., $R_2Fe_{14}C$,[7,12] $Ho_2Fe_{14}BN_x$,[13,14] $R_2Co_{14}B$,[7,15] R_2Fe_{17},[16] $R_2Fe_{17}N_x$,[17] $R_2Fe_{17}C_x$,[18] $Sm_2Fe_{17}H_x$,[19,20] $R_2Fe_{17}C_xN_y$,[17] R_2Co_{17},[21] $RFe_{11}Ti$,[22] $RFe_{10}Mo_2$.[23] Chen, Skumryev, and Kronmüller[24] have observed the anomaly in the temperature dependence of the ac susceptibility in a measurement on single-crystalline $Nd_2Fe_{14}B$. The temperature dependence of the ac susceptibility of a $Ho_2Fe_{14}B$ single crystal has been measured by Rillo *et al.*[25] for the [001], [100], and [110] directions.

In the present paper, we present a systematic investigation of the ac susceptibility of $R_2Fe_{14}B$ single crystals with R=Y, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm as a function of temperature in the temperature interval from 4.2 to 300 K, as a function of the strength of the applied ac field (up to 800 A/m), as a function of frequency (ranging from 5 to 1000 Hz), as well as the dependence on the crystallographic ori-

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entation. The present paper is organized as follows: After the introduction in Sec. I, the experimental procedures and facilities will be presented in Sec. II. The experimental results are discussed in Sec. III. In Sec. IV, a summary is given.

II. EXPERIMENTAL DETAILS

The single crystals of $R_2Fe_{14}B$ with R=Pr and Nd were grown at the FOM-ALMOS Centre at the University of Amsterdam by means of the tri-arc Czochralski method.[26] The single crystals of $R_2Fe_{14}B$ with R=Y, Gd, Tb, Dy, Er, and Tm were produced in an infrared imaging furnace by means of the floating-zone melting technique. The $Sm_2Fe_{14}B$ single crystal was taken out of large grains present in an ingot that had been cooled very slowly in a BN crucible. The details concerning the preparation of the single crystals are presented elsewhere.[5,26] The single crystals with well-defined [001], [100], and [110] directions and varying in shape from spherical to cubic were embedded in an epoxy-resin cube. The typical mass of the single crystals was 0.1–0.2 g. The quality of the single crystals was checked by various magnetization measurements.[5,9,27,28]

The temperature dependence of the ac susceptibility of the single crystals was measured in an ac susceptometer (Lake Shore Cryotronic, Model 7000) equipped with a lock-in amplifier. This system can be operated from 1.8 to 300 K with a frequency from 5 to 1000 Hz and an ac field up to 800 A/m. The temperature is stabilized within 0.5 K below 100 K, 1.0 K between 100 and 250 K and 1.5 K above 250 K. By using this susceptometer, the absolute values of the real component (χ') and imaginary component (χ'') of the ac susceptibility can be determined simultaneously. In most of the measurements, an external ac field of 40 A/m and a frequency of 1000 Hz were employed in order to obtain data that can be systematically compared. However, for $Nd_2Fe_{14}B$ the ac susceptibility was measured in much more detail as a function of temperature, ac field, frequency, and crystallographic orientation.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Before we come to the presentation and discussion of the ac susceptibility of the individual compounds, it is helpful to have a general understanding of the information that can be derived from ac-susceptibility measurements.

A. Fundamental understanding of the ac susceptibility in *R-T* intermetallic compounds

The complex ac susceptibility $\chi=\chi'+i\chi''$ is determined by measuring its two components, i.e., the real (χ') and the imaginary (χ'') part. The real component of the ac susceptibility is the initial susceptibility with the same phase as the external ac field and is correlated with the reversible initial magnetization process. The value of χ' gives a measure of how difficult it is to initiate the magnetization process. On the other hand, the imaginary part χ'' of the ac susceptibility represents the part of the susceptibility having a phase shift with respect to the external field and is connected with an irreversible magnetization process showing a hysteresis loop. The value of χ'' represents the energy loss during the initial magnetization process mainly caused by the motion of domain walls. Chen, Skumryev, and Kronmüller[24] have discussed the energy loss caused by eddy currents in a $Nd_2Fe_{14}B$ single crystal.

In magnetic materials, there are, in principle, two main contributions to the ac susceptibility, namely domain-wall movement and rotation of magnetic moments. In a uniaxial material, the domain walls are 180° domain walls. In a low external ac field parallel to the easy magnetization direction (EMD), domain-wall movement excited by the ac field is the only contribution to the ac susceptibility. Since domain-wall movement is involved, a magnetic hysteresis loop causing energy loss will be encountered. Therefore, in this case, a nonzero χ'' is expected. Domain-wall movement is associated with a change of the domain configuration and is determined not only by the magnetocrystalline anisotropy but also by the actual pinning mechanism of the domain walls. In general, the uniaxial anisotropy is stronger than the pinning force of domain walls. Therefore, the contribution to χ' due to the domain-wall movement is usually larger than that due to the rotation of the magnetic moments. The exception can be the case that the domain wall is extremely thin, due to very large anisotropy. The theoretical treatment of the ac susceptibility (both χ' and χ'') caused by the domain-wall movement is quite complex and up until present no good model is available that correctly accounts for the experiments. On the other hand, if an external field is applied perpendicular to the EMD, only the rotation of the magnetic moments contributes to the susceptibility. In a low field, this rotation of the magnetic moments within a domain is extremely small due to the strong uniaxial magnetic anisotropy. The rotation will take place very near to the EMD. We can conclude that the contribution to the susceptibility by the rotation of the magnetic moments is determined only by the magnetic anisotropy. For a single crystal with the EMD parallel to the [001] direction, the real part χ' along the [100] or [110] direction is represented by[7,24,25]

$$\chi'[100]=\frac{\mu_0 M_s^2}{2K_1}, \tag{1}$$

where M_S is the spontaneous magnetization and K_1 the second-order anisotropy constant if the anisotropy energy E_A is expressed as

$$E_A=K_1\sin^2\theta+K_2\sin^4\theta+K_3\sin^4\theta\,\cos 4\phi+\cdots. \tag{2}$$

In Eq. (2), θ represents the angle between the [001] direction and the direction of the magnetization M_S, ϕ is the angle between the projection of M_S on the basal plane and the [100] direction, K_1, K_2, and K_3 are the second-, the fourth-, and the sixth-order anisotropy constants, respectively. Since the Curie temperatures of the compounds studied are well above room temperature, the value of the spontaneous magnetization will not change much in the temperature range from 4.2 to 300 K. Therefore, the temperature variation of χ' is mainly determined by the temperature dependence of the magnetic anisotropy.

For compounds having planar anisotropy, the calculation of the contribution of the domain-wall movement to the susceptibility is even more complicated. This is because the domain-wall configuration in materials with easy-planar anisotropy is not well-defined. There exists magnetic anisot-

ropy within the basal plane, although it is rather small and vanishes at temperatures far below room temperature. The susceptibility measured along [100] will be different from that measured along the [110] direction. However, the above general discussion concerning the perpendicular susceptility in easy-axis materials is still valid for materials with in-plane anisotropy. Particularly, the theoretical treatment of the perpendicular susceptibility for materials with easy c-axis anisotropy will be the same as for the easy-plane materials.

If a temperature-induced change of the EMD or a change of the domain configuration occurs, this should be detectable by measuring the temperature dependence of the ac susceptibility. As far as the EMD changes are concerned, the domain configuration of the single crystals will change accordingly. Chen, Skumryev, and Kronmüller[24] have calculated χ' for a conical spin configuration (like in $Nd_2Fe_{14}B$) below the spin-reorientation temperature and obtained

$$\chi'[001]=\frac{\mu_0 M_s^2}{4K_1+8(K_2-K_3)}, \tag{3}$$

$$\chi'[100]=-\frac{\mu_0 M_s^2[(K_2/K_3)+1]}{16K_1}. \tag{4}$$

In this case, contributions to the ac susceptibility from the rotation of the magnetic moments and from the domain-wall movement will exist along both the [001] and the [100] directions.

In the following paragraphs, the ac-susceptibility results for single crystals with similar anisotropy behavior will be discussed together.

B. ac susceptibility of $R_2Fe_{14}B$ single crystals with R=Y, Pr, Nd, Gd, Tb, Dy, Ho, Er, or Tm

1. $Y_2Fe_{14}B$ and $Gd_2Fe_{14}B$

Since Y is a nonmagnetic element, the magnetic anisotropy in $Y_2Fe_{14}B$ originates only from the Fe-sublattice anisotropy. However, possibly because of an anisotropic exchange interaction between the Fe and Gd moments, the magnetic anisotropy field of Gd compounds shows a deviation from that of the Y compounds. It is noted that such a deviation is only pronounced in $GdCo_5$ (Refs. 29 and 30) compound and exists very slightly in, e.g., $Gd_2Fe_{14}B$,[5,31] $GdFe_{11}Ti$,[22] and Gd_2Fe_{17}.[32] It was found that the magnetic anisotropy in both $Y_2Fe_{14}B$ and $Gd_2Fe_{14}B$ shows an anomalous temperature dependence.[6,31] Initially, the anisotropy constant K_1 increases with increasing temperature and reaches a maximum at 150 K for $Y_2Fe_{14}B$ and at 380 K for $Gd_2Fe_{14}B$ and with further increasing temperature it decreases again. The EMD in $Y_2Fe_{14}B$ and $Gd_2Fe_{14}B$ is the [001] direction in the whole temperature range of magnetic order. Figures 1 and 2 show the temperature dependences of χ' and χ'' of $Y_2Fe_{14}B$ and $Gd_2Fe_{14}B$ measured with an ac field of 40 A/m parallel to the [001] or the [100] direction. The values of χ' measured with the field parallel to the [001] direction are roughly three times larger than the values measured with the field parallel to the [100] direction. As has been discussed previously, the main contribution to the ac susceptibility is the domain-wall movement when a field is applied parallel to the [001] direction. On the other hand, the rotation of the magnetic moment is the only contribution to the ac susceptibility when the field is applied parallel to the [100] direction. The higher values of χ' measured in the [001] direction suggest that the domain-wall movement is much easier than the rotation of the magnetic moments at the same field strength. This indicates that the anisotropy field is much stronger than the pinning force of the domain walls in $Y_2Fe_{14}B$ and $Gd_2Fe_{14}B$. In addition, it is noticed that the χ' values are nearly independent of temperature when the measurement is performed along the [001] direction. However, the values of χ' decrease slightly with increasing tem-

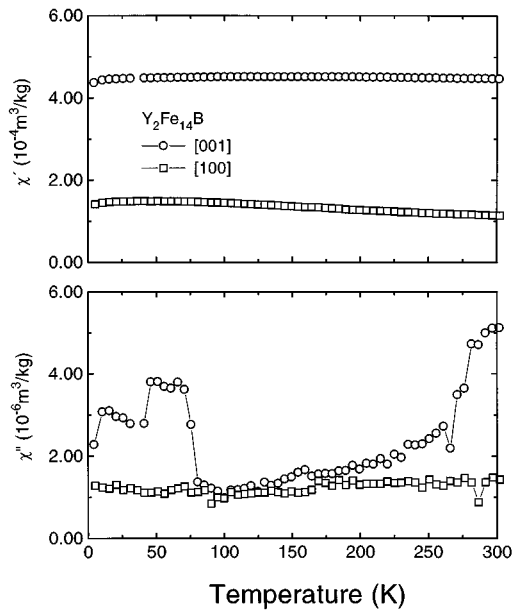

FIG. 1. The temperature dependence of the ac susceptibility measured for a $Y_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel to the [001] or [100] direction.

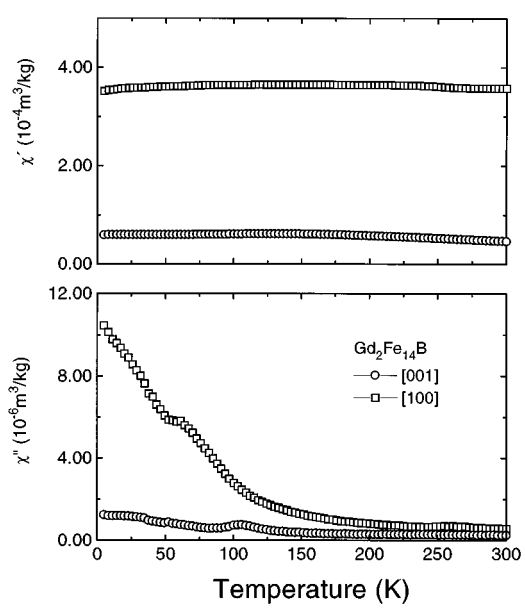

FIG. 2. The temperature dependence of the ac susceptibility measured for a $Gd_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel to the [001] or [100] direction.

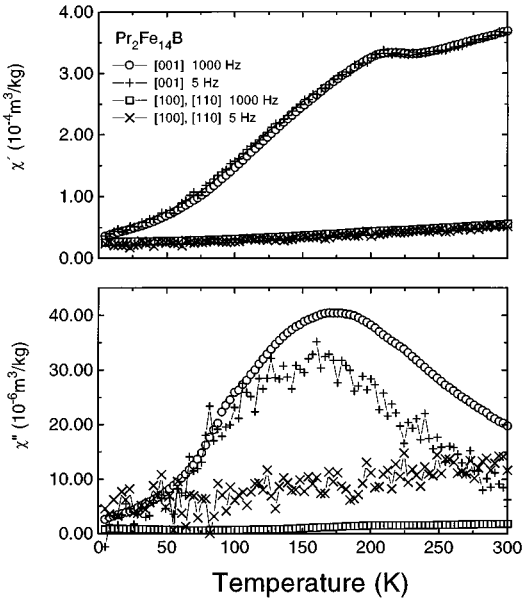

FIG. 3. The temperature dependence of the ac susceptibility measured for a $Pr_2Fe_{14}B$ single crystal with a field 40 A/m of 1000 or 5 Hz applied parallel to the [001] or [100] direction.

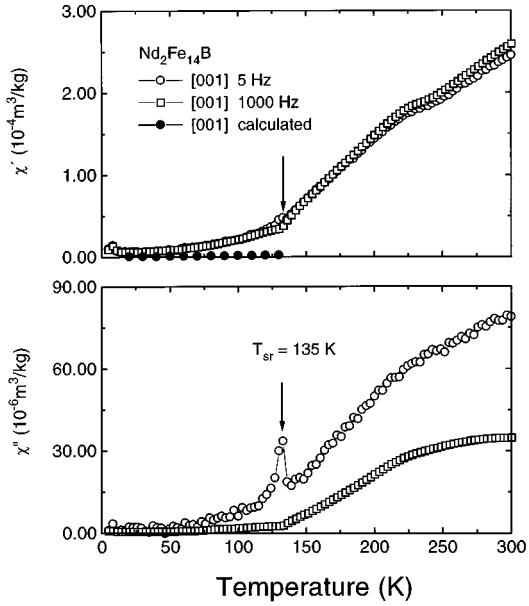

FIG. 4. The temperature dependence of the ac susceptibility measured for a $Nd_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 or 5 Hz applied parallel to the [001] direction.

perature when the measurement is made parallel to the [100] direction. The temperature dependence of χ'' is found to be very pronounced in the measurement along the [001] direction, whereas a nearly zero value of χ'' is detected along the [100] direction. From these experiments it follows that the rotation of the magnetic moments in a low ac field is a reversible process (indicated by a nearly zero energy loss), while the domain-wall movement in a low ac field is irreversible (indicated by the large value of χ'').

2. $Pr_2Fe_{14}B$

The EMD in $Pr_2Fe_{14}B$ is the [001] direction in the whole temperature range of magnetic order. Figure 3 shows the temperature dependences of χ' and χ'' for a $Pr_2Fe_{14}B$ single crystal measured in external fields of 40 A/m parallel to the [001], [100], and [110] directions with frequencies of 5 and 1000 Hz. The values of χ' and χ'' are identical for the [100] and [110] directions. Furthermore, the values of χ' and χ'' measured in these two directions are much smaller than in the [001] direction, indicating a large magnetic anisotropy. A slight increase of χ' and χ'' with increasing temperature is detected in the [100] and the [110] directions, reflecting a decrease of the anisotropy with increasing temperature. In the [001] direction, the value of χ' at 300 K is almost an order of magnitude larger than in the [100] and [110] directions suggesting that in this case the contribution to the susceptibility from the domain-wall movement is much larger than from the rotation of the magnetic moments. A strong temperature dependence of χ' and χ'' is detected along the [001] direction. The χ' values decrease with decreasing temperature. At 4.2 K, the value of χ' in the [001] direction is only slightly higher than for the [100] and [110] directions. In the measurement along the [001] direction, a clear anomaly is visible at about 220 K. No anomaly is detectable in the measurements along the [100] and [110] directions. It is worth noting that even in polycrystalline $Pr_2Fe_{14}B$ a pronounced anomaly at about 220 K has been observed.[7] As far as the frequency dependence of the ac susceptibility is concerned, χ' values of $Pr_2Fe_{14}B$ are nearly independent on the frequency in the range from 5 to 1000 Hz. However, the χ'' values show a strong frequency dependence (Fig. 3). It is interesting to note that the changes of the χ'' values also depend on the crystallographic orientation. The χ'' values are higher at low frequencies when the field is applied along the [100] direction. In contrast, the χ'' values are lower at low frequencies when the field is applied along the [001] direction. In general, the χ'' values are higher for the [001] direction than for the [100] direction, indicating that the domain-wall movement gives rise to a high-energy loss at low frequencies.

3. $Nd_2Fe_{14}B$ and $Ho_2Fe_{14}B$

The [001] direction is the EMD in $Nd_2Fe_{14}B$ from 135 K up to 585 K, the Curie temperature. Below 135 K, the EMD tilts away from the [001] direction. The tilting angle increases with lowering temperature and reaches about 30 degrees at 4.2 K. The $Nd_2Fe_{14}B$ single crystal used in the present investigation has a spherical shape and therefore a demagnetizing factor of 1/3. The temperature dependence of the susceptibility of $Nd_2Fe_{14}B$ has a peculiar character. In a field of 40 A/m of 1000 Hz, applied parallel to the [001] direction, χ' decreases continuously from 2.59×10^{-4} m^3/kg at 300 to about 3.79×10^{-5} m^3/kg at 135 K, the onset temperature of the spin reorientation. A clear anomaly, in the form of a change of slope, appears at 135 K (Fig. 4). Below 135 K, χ' continuously decreases with decreasing temperature, however only very slightly. Around 220 K, another anomaly, much less pronounced than in the measurement on polycrystalline $Nd_2Fe_{14}B$,[7] is visible. Very similar to χ', the temperature dependence of χ'' shows an anomaly exactly at $T_{sr}=135$ K. However, around 220 K no anomaly is visible in χ''. The decrease of χ' and χ'' with decreasing tempera-

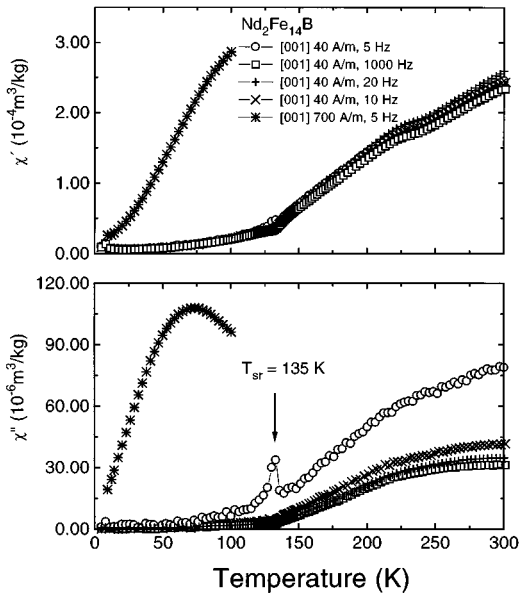

FIG. 5. The temperature dependence of the ac susceptibility measured for a $Nd_2Fe_{14}B$ single crystal with different field strengths (from 40 to 700 A/m) and frequencies (from 5 to 1000 Hz) applied parallel to the [001] direction.

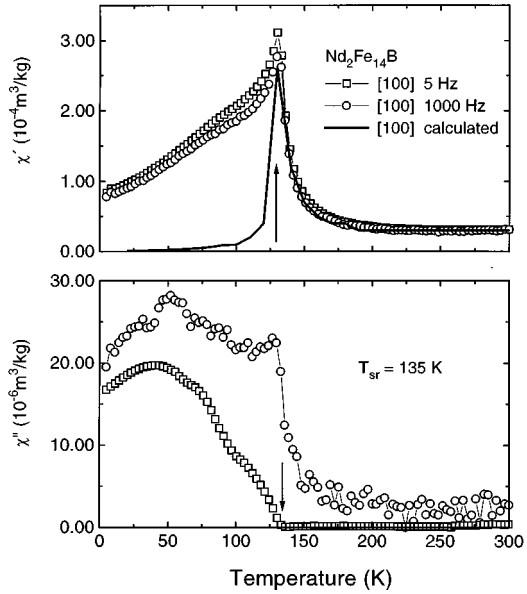

FIG. 6. The temperature dependence of the ac susceptibility measured for a $Nd_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 or 5 Hz applied parallel to the [100] direction. The solid line represents the calculated χ' due to the rotation of the magnetic moments.

ture in the temperature range of easy c-axis anisotropy (above 135 K) is caused by an increase of the magnetic anisotropy which makes the domain walls narrow and more difficult to move in an external field. Below 135 K, the EMD deviates from the [001] direction. The domain structure will change accordingly. The actual domain configuration is very complex as observed in a Kerr effect study of the domain patterns in a $Nd_2Fe_{14}B$ single crystal between 4.2 K and room temperature.[33] This investigation has pointed out that below 135 K the domain walls in $Nd_2Fe_{14}B$ are not 180° domain walls. In this temperature range, the domain-wall movement is practically frozen when the field is applied along the [001] direction which leads to very low values of both χ' and χ'' (see Fig. 4). In addition, it is of interest to note that the values of χ' as well as the temperature dependence of χ' change only slightly when the frequency of the applied field is changed from 5 to 1000 Hz (Fig. 5). The values of χ' and χ'' strongly depend on the strength of the external field. Above 100 K, measurements in a field of 700 A/m at 5 Hz were not possible since χ' becomes too large. The imaginary part χ'' was found to increase with decreasing frequency. At 135 K, exactly at the spin-reorientation temperature, a peak is observed (see Fig. 4). Chen, Skumryev, and Kronmüller[24] also found a peak at T_{sr} in $\chi''(T)$ at low frequencies, but they detected a peak at T_{sr} in $\chi'(T)$ as well, which is not expected for a second-order phase transition.

When an ac field of 40 A/m and 1000 Hz is applied parallel to the [100] direction, a peak in the temperature dependence of χ' is observed at 135 K, associated with the spin-reorientation (Fig. 6). Also in the temperature dependence of χ'' the occurrence of the spin reorientation is very clearly visible. From 135 to 300 K, χ'' is almost zero which suggests that in this temperature range the reversible rotation of the magnetic moments is the only contribution to the ac susceptibility. Exactly at 135 K, where χ' shows a peak, χ'' starts to increase. Below 135 K, the contribution due to the domain-wall movement appears. In order to calculate contribution to χ' from the rotation of the magnetic moments, the anisotropy constants (K_1, K_2, and K_3) as well as the spontaneous magnetization (M_S) are needed. Of all $R_2Fe_{14}B$ compounds, these data are available only for $Nd_2Fe_{14}B$ over a wide temperature range. In the present calculation, K_1, K_2, K_3, and M_S (Fig. 7, Ref. 34) derived from the magneti-

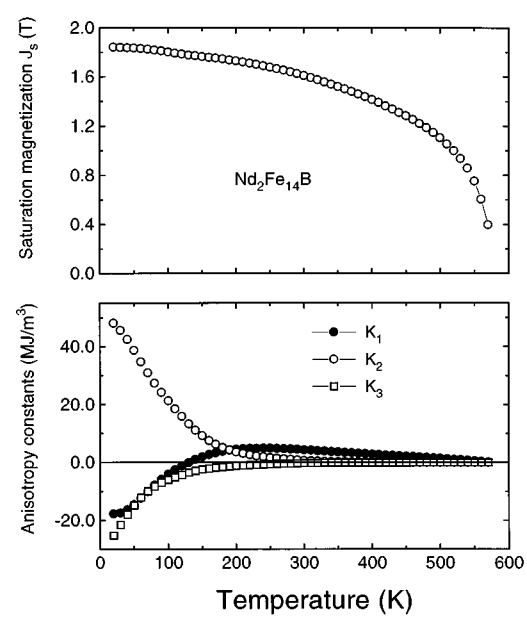

FIG. 7. The anisotropy constants K_1, K_2, and K_3 and the spontaneous magnetization M_S of $Nd_2Fe_{14}B$, obtained by Hock (Ref. 34) from the magnetization measurements on a $Nd_2Fe_{14}B$ single crystal.

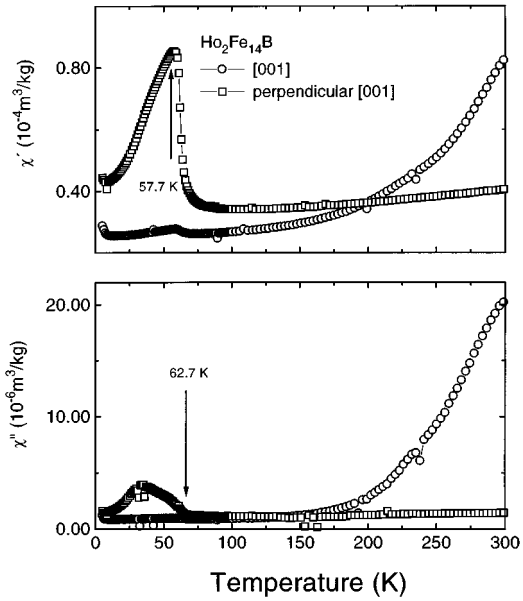

FIG. 8. The temperature dependence of the ac susceptibility measured for a $Ho_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel or perpendicular to the [001] direction.

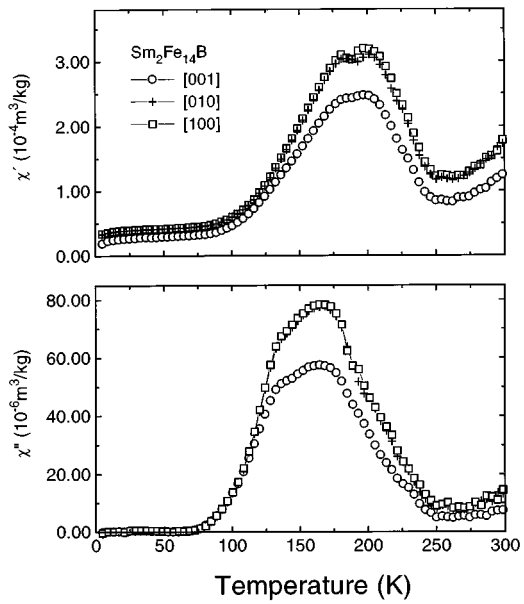

FIG. 9. The temperature dependence of the ac susceptibility measured for a $Sm_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel to the [001], the [010], and the [100] direction.

zation measurements on single-crystalline $Nd_2Fe_{14}B$ were used. By using Eq. (1) (135 K$<T<$300 K) and Eq. (4) ($<$135 K), the values of χ' of $Nd_2Fe_{14}B$, due to the rotation of the magnetic moments, can be calculated. In order to be compared with the experimental data, the calculated data have to be conserved by taking into account the demagnetizing factor (1/3) by $3\chi'/(3+\chi')$. The density of $Nd_2Fe_{14}B$ taken in converting the unit to m^3/kg is 7.6 g/cm^3. The calculated data of χ' are also shown in Fig. 6. In the easy c-axis temperature range from 135 to 300 K, the agreement between the calculation and experiments is perfect. Below 135 K, the calculated data are much smaller than experimental data, suggesting that below this temperature the main contribution to χ' is no more due to the rotation of the magnetic moments, but to domain-wall movement. A similar calculation, using Eq. (3), was made below 135 K for the case that the external field is applied parallel to the [001] direction. The results are shown in Fig. 4. The calculated data are much lower than the experimental data which implies that also in this case the rotation of the magnetic moments is not the main contribution to χ'.

The frequency dependences of χ'' and χ' are different. χ' increases with increasing frequency, whereas χ'' decreases with increasing frequency. For both, the frequency dependence is the strongest below T_{sr}. Since all measurements of the present investigation were performed in a low and relative narrow range of frequency, from 5 to 1000 Hz, the quantitative study of the relationship between χ'' and χ' are not made. However, it is evident that both, χ'' and χ', are functions of frequencies of applied ac fields.

Similar to $Nd_2Fe_{14}B$, the [001] direction is the EMD in $Ho_2Fe_{14}B$ from 57.6 K up to the Curie temperature. Figure 8 shows the temperature dependence of χ' and χ'' of $Ho_2Fe_{14}B$ with an ac field 40 A/m of 1000 Hz applied parallel or perpendicular to the [001] direction. The spin-reorientation transition is evident when the measurements are performed perpendicular to the [001] direction. However, the spin-reorientation transition is not so pronounced when the measurement is performed along the [001] direction. In the temperature range close to the room temperature, a strong temperature dependence of χ' and χ'' is detectable when measured along the [001] direction. No peak, as determined in $Nd_2Fe_{14}B$ at 135 K, is detectable for $Ho_2Fe_{14}B$ in the temperature dependence of χ'' at T_{sr} even in measurements in an ac field of 40 A/m and 5 Hz along the [001] direction.

4. $Sm_2Fe_{14}B$, $Er_2Fe_{14}B$, and $Tm_2Fe_{14}B$

The common feature of the three compounds $Sm_2Fe_{14}B$, $Er_2Fe_{14}B$, and $Tm_2Fe_{14}B$ is that their EMD lies within the basal plane in the temperature range from 4.2 to 300 K. No changes of spin configuration are reported below 300 K in these three compounds. The temperature dependences of χ' and χ'' measured for single-crystalline $Sm_2Fe_{14}B$ with the field applied parallel to the [001], the [010], and the [100] directions are shown in Fig. 9. It is evident that χ' and χ'' are identical for the [010] and the [100] directions, the easy magnetization directions of $Sm_2Fe_{14}B$. Three temperature ranges can be clearly distinguished. Below about 80 K, χ' is small and temperature independent whereas χ'' is zero. In the temperature range from 80 to 250 K, χ' and χ'' show a broad maximum. Above 250 K, χ' and χ'' exhibit an increase.

We will now try to understand the temperature dependences of χ' and χ'' of $Sm_2Fe_{14}B$ measured along the [100] or the [010] direction where the domain-wall movement is the only contribution to the susceptibility. The EMD of $Sm_2Fe_{14}B$ is within the basal plane over the whole magnetic-ordering temperatures range. The domain configuration of this compound is unknown. Irrespective of the domain configuration, the effective domain-wall width is pro-

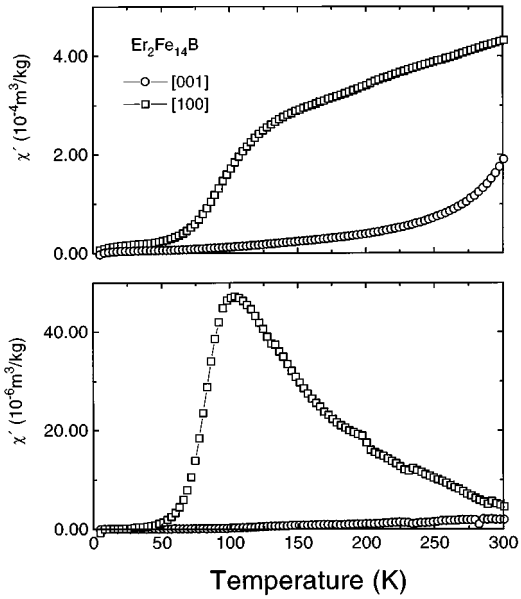

FIG. 10. The temperature dependence of the ac susceptibility measured for a $Er_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel or perpendicular to the [001] or [100] direction.

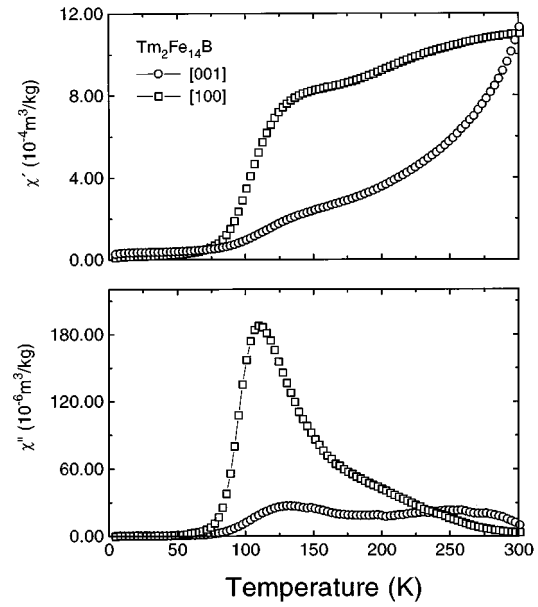

FIG. 11. The temperature dependence of the ac susceptibility measured for a $Tm_2Fe_{14}B$ single crystal with the field 40 A/m of 1000 Hz applied parallel to the [001] or [100] direction.

portional to $(K_A)^{-1/2}$, where K_A is the effective anisotropy constant.[35] If the anisotropy is very large, the domain walls become narrow and they are difficult to be moved by an external field. On the other hand, if the anisotropy is very small, the domain walls become so wide that in a single crystal they can be pinned by the sample surface. Such wide domain walls are also difficult to be moved by an external field. Therefore, only medium-size domain walls contribute to the ac susceptibility. Since the effective anisotropy constant K_A decreases with increasing temperature, the domain-wall width will change with temperature. Within this picture, the experimental results on $Sm_2Fe_{14}B$ can be interpreted as follows. At low temperatures, the magnetic anisotropy of $Sm_2Fe_{14}B$ is huge which can be seen from the magnetization measurements on a single crystal at 4.2 K.[36] The domain walls are so thin that the domain-wall movement is almost totally frozen, resulting in a very small χ' and zero χ''. With increasing temperature, the anisotropy of $Sm_2Fe_{14}B$ will decrease drastically, as is the case in practically all Sm compounds,[37] and the domain-wall width will increase accordingly. Around 100 K, considerable domain-wall movement will start to take place which will contribute to the susceptibility. With further increasing temperature to above 250 K, the domain walls become so wide they are pinned by the sample surface, resulting in low values of both χ' and χ''. The measurements in the [001] direction, the hard magnetization direction, reflect only the contribution from rotation of the magnetic moments.

In Figs. 10 and 11, the temperature dependences of χ' and χ'' are shown for $Er_2Fe_{14}B$ and $Tm_2Fe_{14}B$, measured in ac fields applied parallel to the [001] or the [100] direction. When the field is applied parallel to the [001] direction, the hard direction of the magnetization, χ' increases with increasing temperature, in particular at higher temperatures. This increase implies a drastic decrease of anisotropy which has indeed been found in singular point detection (SPD) experiments on $Er_2Fe_{14}B$.[38] The χ' values of $Tm_2Fe_{14}B$ are higher than those of $Er_2Fe_{14}B$ which indicates that the anisotropy of $Tm_2Fe_{14}B$ is lower than that of $Er_2Fe_{14}B$. The lower anisotropy of $Tm_2Fe_{14}B$ is corroborated by the lower spin-reorientation temperature of $Tm_2Fe_{14}B$ ($T_{sr}=315$ K) compared with $Er_2Fe_{14}B$ ($T_{sr}=323$ K).[5] The χ'' values are very small in this case, indicating that the movement of magnetic moments is a nearly reversible process. If the field is applied parallel to the [100] direction, the EMD of $Er_2Fe_{14}B$ and $Tm_2Fe_{14}B$, a drastic increase of χ' and χ'' starts at about 50 K for $Er_2Fe_{14}B$ and at about 80 K for $Tm_2Fe_{14}B$. In this case, the domain-wall movement will be the only contribution to the susceptibility. Following the discussion for $Sm_2Fe_{14}B$, it is suggested that the domain wall is almost frozen at low temperatures ($\chi'\approx 0$ and $\chi''\approx 0$). The extremely large magnetic anisotropy at low temperature leads to very narrow domain walls which cannot move in a small ac field. In addition, the noncollinear configuration of the Er (or Tm) and Fe moments at low temperatures[39,40] may cause a complex domain configuration which in turn impedes the domain-wall movement. The observed drastic increase of χ' and χ'' is the result of the domain-wall movement due to the reduction of the magnetic anisotropy with increasing temperature.

5. $Tb_2Fe_{14}B$ and $Dy_2Fe_{14}B$

The common feature of the compounds $Tb_2Fe_{14}B$ ($H_A\approx 18$ MA/m at 300 K) and $Dy_2Fe_{14}B$ ($H_A\approx 12$ MA/m at 300 K) is their huge uniaxial anisotropy[9] which results in extremely thin domain walls. According to the discussion in the previous paragraph, the movement of a thin domain wall in an ac field is difficult. Therefore, it is possible that the contribution to the ac susceptibility due to the domain-wall movement is smaller than that due to the rotation of the magnetic moments in the same ac field. The temperature

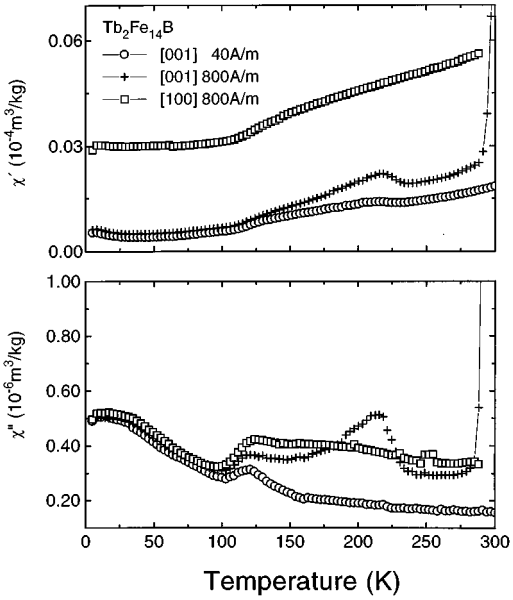

FIG. 12. The temperature dependence of the ac susceptibility measured for a $Tb_2Fe_{14}B$ single crystal with the field 40 or 800 A/m of 1000 Hz applied parallel to the [001] or [100] direction.

dependences of χ' and χ'' measured for $Tb_2Fe_{14}B$ and $Dy_2Fe_{14}B$ with the field applied parallel to [001] or the [100] direction are shown in Figs. 12 and 13, respectively. We do find that the ac susceptibility (χ' and χ'') measured along the [001] direction, originating from the domain-wall movement, is smaller than along the [100] direction, originating from the rotation of the magnetic moments. In a larger field of 800 A/m, the domain-wall movement starts at about 280 K for $Tb_2Fe_{14}B$ and at about 100 K for $Dy_2Fe_{14}B$. Compared to other compounds, $Tb_2Fe_{14}B$ and $Dy_2Fe_{14}B$ have the lowest ac susceptibility. This is due to their huge

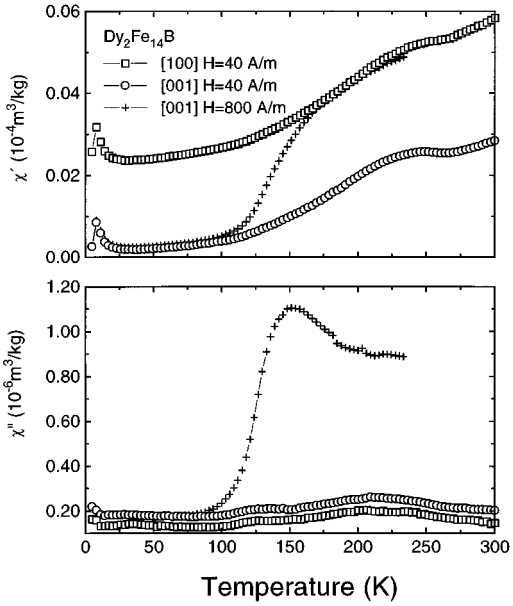

FIG. 13. The temperature dependence of the ac susceptibility measured for a $Tb_2Fe_{14}B$ single crystal with the field 40 or 800 A/m of 1000 Hz applied parallel to the [001] or [100] direction.

magnetic anisotropy. Also in contrast to the other measured compounds, the susceptibility of $Tb_2Fe_{14}B$ and $Dy_2Fe_{14}B$ is lower when the measurement is performed parallel to the EMD (the [001] direction) than perpendicular to the EMD (e.g., the [100] direction). The very low χ'' values in the measurements in 40 A/m suggest that both processes are reversible.

IV. CONCLUSIONS

In recent years, the technique of measuring ac susceptibility has undergone an impressive development. In particular, the need for accurate measurements on small crystals of the high-T_c superconductors has stimulated the development of very sensitive equipment for ac susceptibility measurements, so that now also the precise determination of the ac susceptibility of R-T compounds has become possible. In the present paper, we have reported a systematic study of the $R_2Fe_{14}B$ series of compounds by means of the ac susceptibility. In a small ac field, the reversible rotation of the magnetic moments is the principal contribution to the ac susceptibility when the measurement is performed perpendicular to the EMD. In this case, the energy loss is nearly zero. On the other hand, if the measurement is performed parallel to the EMD, irreversible domain-wall movement is the main contribution to the ac susceptibility. In this case, the energy loss is nonzero. In $Nd_2Fe_{14}B$, a singularity is detected in $\chi''(T)$ at 135 K, the spin-reorientation temperature, when the measurement is performed along the [001] direction. In the temperature range where the c axis is the EMD, the calculated χ' of $Nd_2Fe_{14}B$, due to the rotation of the magnetic moments, agrees perfectly with the experimental χ'. Peaklike anomalies in $\chi'(T)$ and $\chi''(T)$, are observed in $Pr_2Fe_{14}B$, $Nd_2Fe_{14}B$, $Tb_2Fe_{14}B$, and $Dy_2Fe_{14}B$, in which the EMD lies parallel to the c axis. For the compounds with the EMD in the basal plane, like $Sm_2Fe_{14}B$, $Er_2Fe_{14}B$, and $Tm_2Fe_{14}B$, it is found that the domain-wall movement is frozen at low temperature. This phenomenon is associated with the extremely large magnetic anisotropy of these compounds at low temperature. Generally, it was found that the dependence of χ' on temperature and frequency is completely different from that of χ''. The χ' values are nearly independent of the frequency in our measuring range from 5 to 1000 Hz. For all compounds investigated, it is found that χ'' increases with increasing frequency when the field is applied along the EMD and decreases with increasing frequency when the field is applied perpendicular to the EMD. This phenomenon, which is connected with the magnetic relaxation processes in the materials, will be investigated in more detail.

ACKNOWLEDGMENTS

The work was supported by the Austrian Science Foundation under Project Nos. S5604 and S5605. The authors thank Professor G. Kido, Tsukuba Magnet Laboratories, National Research Institute for Metals, Tsukuba, Japan, and Professor H. Kato, Tohoku University, Sendai, Japan, for providing the $Y_2Fe_{14}B$ single crystal and Dr. S. Hirosawa from Sumitomo Company, Japan, for providing the single crystals of $R_2Fe_{14}B$ with R=Sm, Gd, Tb, Dy, Ho, Er, and Tm.

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