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Liu, J.P.; de Boer, F.R.; de Chatel, P.F.; Buschow, K.H.J.

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Magnetic coupling and magnetic transitions in interstitial compounds of the series $\text{Er}_2\text{Fe}_{17}\text{C}_x$

J. P. Liu, F. R. de Boer, and P. F. de Châtel

Van der Waals-Zeeman Laboratory, University of Amsterdam, 1018 XE Amsterdam, The Netherlands

K. H. J. Buschow

Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands

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A detailed investigation of the structure and magnetic properties of $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds is presented in this paper. The interstitial carbides $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with $x=0.0, 0.5, 1.0, 1.5, 2.0, 2.5,$ and 3.0 have been synthesized by arc melting and by the gas-phase-interaction method. The unit-cell volume of the compounds is found to increase with increasing carbon concentration. The magnetic properties were studied in a Faraday balance and by measurements of the ac susceptibility and the high-field magnetization. The dependencies of Curie temperatures, spin-reorientation temperatures, and the Er-Fe magnetic-coupling strength on the carbon content were determined. The results have been discussed, including the difference of the effect of carbon and nitrogen atoms on the Er-Fe coupling.

I. INTRODUCTION

Among the rare-earth-transition-metal intermetallics, those of the type $R_2\text{Fe}_{17}$ (R = rare earth) are of interest, because they have the highest saturation magnetization. Unfortunately, the Curie temperatures of these compounds are very low, which prevents them from being applied as permanent magnets. Since it was discovered that compounds of this type can accommodate interstitial carbon atoms and that the Curie temperatures increase remarkably upon carbonation,^{1,2} these compounds have become interesting both from the fundamental and the applied point of view. Much work has been done,³⁻⁷ especially after Coey and co-workers^{8,9} discovered that carbon atoms can be introduced into the compounds also by the gas-phase-interaction (GPI) method by using hydrocarbon gases. With this method the carbon content in the $R_2\text{Fe}_{17}\text{C}_x$ compounds can be raised from $x=1$, the maximum value obtained by melting for most of the $R_2\text{Fe}_{17}$ compounds, to $x \approx 2.3$.⁹ Recently, Kong, Cao, and Shen¹⁰ reported results on $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with x up to 3.0, obtained by means of the melt-spinning method. Strong dependence of the magnetic properties of the compounds on the carbon content has been observed by all authors. In $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds, for example, it was reported that the Curie temperature and the saturation magnetization increase with increasing carbon content,^{2,9} and that introduction of interstitial carbon atoms results in a spin-reorientation transition.⁵ The effect of interstitial carbon atoms on the R - T exchange interaction in the $R_2\text{Fe}_{17}$ compounds was studied in Refs. 7 and 9, although with different results.

In order to understand more clearly the effect of interstitial carbon atoms on the structure and the magnetic properties of the compounds of $R_2\text{Fe}_{17}$ type, we carried out an investigation of the interstitial carbides of $\text{Er}_2\text{Fe}_{17}$. With the motivation to reach a better understanding of the dependence of the magnetic properties, especially the magnetic coupling, on the carbonation, we have syn-

thesized the series $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with $x=0.0, 0.5, 1.0, 1.5, 2.0, 2.5,$ and 3.0 by means of the arc melting and the GPI method. The Curie temperatures, the spin-reorientation temperatures, and the high-field magnetization have been determined to study the magnetic transitions and the magnetic coupling in the compounds.

II. EXPERIMENTS AND THE RESULTS

Ingots of $\text{Er}_2\text{Fe}_{17}$ and $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with $x=0.5, 1.0,$ and 1.5 were prepared by arc melting the starting materials with purity equal to or higher than 99.9%. After melting, the ingots were homogenized for two weeks at about 1000 °C, followed by a water quench to avoid the formation of the $\text{Er}_2\text{Fe}_{14}\text{C}$ phase. The samples obtained were checked by x-ray diffraction. $\text{Er}_2\text{Fe}_{17}$ was found to be single phase, but the carbides were not. Reflections of α -Fe appear in the diffractograms, that have higher intensities with increasing carbon contents. It appears to be virtually impossible to obtain carbide samples free of α -Fe when using standard casting as well as when using the GPI process. This is the result of nearly all studies in which the interstitial compounds were investigated thus far.

The carbides $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with $x=2.0, 2.5,$ and 3.0 were obtained by the GPI method. The carbon contents were determined by weighing the mass gain with an analytical balance. The carbonation was performed by exposing the fine-ground powder to flowing methane gas at about 500 °C for several hours. The average particle size of the powders was about 30 μm . The $\text{Er}_2\text{Fe}_{17}\text{C}_{3.0}$ sample was prepared by carbonating the $\text{Er}_2\text{Fe}_{17}$ compound sufficiently long. The realization of the idea of preparing a series of carbides with gradually increasing C content encountered difficulties when we attempted to synthesize $\text{Er}_2\text{Fe}_{17}\text{C}_{2.0}$ and $\text{Er}_2\text{Fe}_{17}\text{C}_{2.5}$. Under usual conditions it appears impossible to prepare single-phase $R_2\text{Fe}_{17}\text{Z}_x$ ($Z = \text{N}$ or C) samples with intermediate interstitial-atom content from the $R_2\text{Fe}_{17}$ compounds by the GPI method.

A detailed discussion of this aspect has been given elsewhere.¹¹ However, when starting from $\text{Er}_2\text{Fe}_{17}\text{C}_{1.5}$, it appears to be possible to prepare phases with intermediate carbon content. Careful processing was needed to establish the appropriate conditions for the synthesis. Starting from fine particles of $\text{Er}_2\text{Fe}_{17}\text{C}_{1.5}$ and using a shorter carbonation treatment than in the case of $\text{Er}_2\text{Fe}_{17}\text{C}_{3.0}$, we succeeded finally in obtaining quite homogeneous samples with the desired carbon contents. Figure 1 shows the diffraction patterns of the seven samples prepared in the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ series. It can be seen clearly that the reflections gradually shift to lower angles with increasing carbon content.

If one makes an explicit comparison, however, it can be found from the diffraction patterns that the widths of the peaks for the compounds with $x=2.0$ and 2.5 are slightly larger than those of the other compounds in the series, suggesting small compositional inhomogeneities in these two compounds. However, the difference is so small that we can practically consider these compounds as single phase. The measurements of the lattice parameters, Curie temperatures, and the spin-reorientation temperatures of the compounds with intermediate C contents, presented below, also indicate satisfactory homogeneity of the compounds.

The gradual shift of the reflections in the diffractograms reflects an increase of the unit-cell volume.

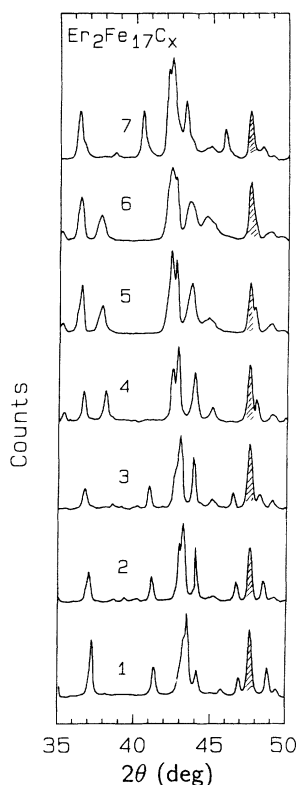


FIG. 1. X-ray-diffraction patterns taken with $\text{Cu } K\alpha$ radiation of $\text{Er}_2\text{Fe}_{17}\text{C}_x$ samples with $x=0.0, 0.5, 1.0, 1.5, 2.0, 2.5,$ and 3.0 corresponding to the patterns from 1 to 7, respectively. The dashed peaks are the reflections from the Si standard for calibration.

Figure 2 shows the dependence of the unit-cell volumes of the compounds on the carbon content. In this figure, the volumes of the compounds crystallizing in $\text{Th}_2\text{Ni}_{17}$ -type of structure have been multiplied by 1.5 in order to compare then with the unit-cell volumes of the compounds with the $\text{Th}_2\text{Zn}_{17}$ structure. The unit cell expands monotonically with increasing carbon content up to about 7.5% for $x=3.0$. A nearly linear relationship is found between the volume and the carbon content.

The expansion of the unit cells results in a change in the structure type. A change of the structure type of the compounds from the $\text{Th}_2\text{Ni}_{17}$ to $\text{Th}_2\text{Zn}_{17}$ takes place in cast $\text{Er}_2\text{Fe}_{17}\text{C}_x$ samples for carbon contents equal to or larger than $x=1.5$. However, during the GPI process the structure type of the starting compounds does not change. Therefore, $\text{Er}_2\text{Fe}_{17}\text{C}_{3.0}$ prepared from $\text{Er}_2\text{Fe}_{17}$ has kept the $\text{Th}_2\text{Ni}_{17}$ structure, even though its expansion is the largest. Another observation worth mentioning is that the reflection patterns of the compounds with $x=0.5, 1.0,$ and 3.0 are not completely similar to those for $x=0.0$, though no obvious changes in the structure took place in these carbides. One of the possible reasons for this difference may be the occurrence of different types and different amounts of stacking faults that may cause the disappearance of some of the smaller peaks in the diffraction patterns, as discussed in Ref. 3.

Figure 3 shows the results of the ac susceptibility measurements. The anomalies in the curves correspond to the spin-reorientation transitions occurring with decreasing temperature which may be attributed to the transition from planar to uniaxial anisotropy. In Fig. 3, one can see that the transitions take place at rather well-defined temperatures which supports our earlier conclusion that the samples are fairly homogeneous. In the curve for the sample with $x=1.0$, two transitions are recognizable, which has been explained by Kou *et al.*⁵ to be due to the coexistence of $\text{Er}_2\text{Fe}_{17}$ phases with the $\text{Th}_2\text{Ni}_{17}$ and the $\text{Th}_2\text{Zn}_{17}$ structures.

Figure 4 shows the results of Faraday-balance measurements on $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds with $x=0.5, 1.0, 2.0,$ and 3.0 . This figure illustrates how the Curie tem-

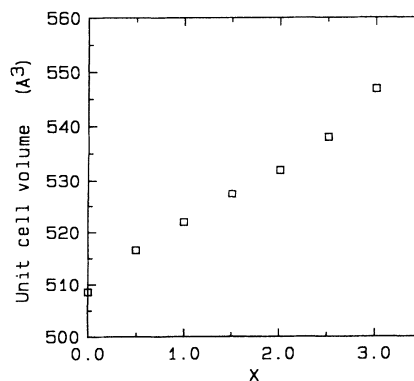


FIG. 2. Dependence of the unit-cell volume on the carbon content. The volume of the compounds with $\text{Th}_2\text{Zn}_{17}$ structure is calculated as $\frac{3}{2}$ of that of the $\text{Th}_2\text{Ni}_{17}$ structure.

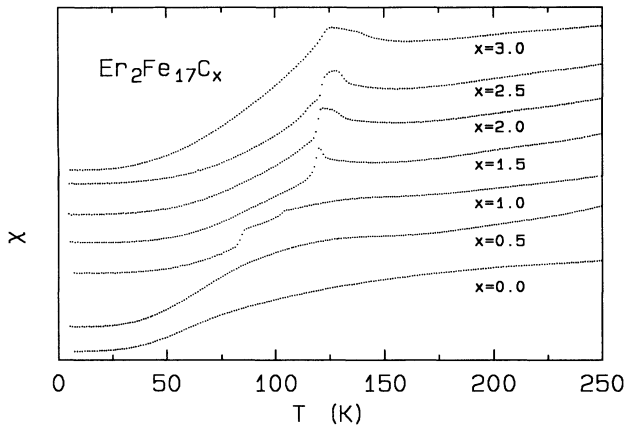


FIG. 3. Ac susceptibility vs temperature for $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds.

peratures of the carbides increase with x . In the compounds with $x = 2.0$ and 3.0 that were made by the GPI method, the Curie temperatures are not as pronounced as in the samples made by melting, which may be associated with the instability of the interstitial compounds at high temperatures.

High-field magnetization measurements on free powders of these compounds have been carried out in the Amsterdam High-Field Installation up to 35 T. Since the particle size of the powder is smaller than $40 \mu\text{m}$, the particles may be regarded as single crystals.¹² In the presence of an external field, the particles are free to orient themselves in the sample holder into the minimum-energy direction. Figure 5 shows the magnetic isotherms at 4.2 K of the seven compounds prepared in this series. It can be seen that there is a field-induced transition in every curve and that the transition fields (the crossing points of the low- and high-field branches of the isotherms) shift to lower-field values with increasing carbon content.

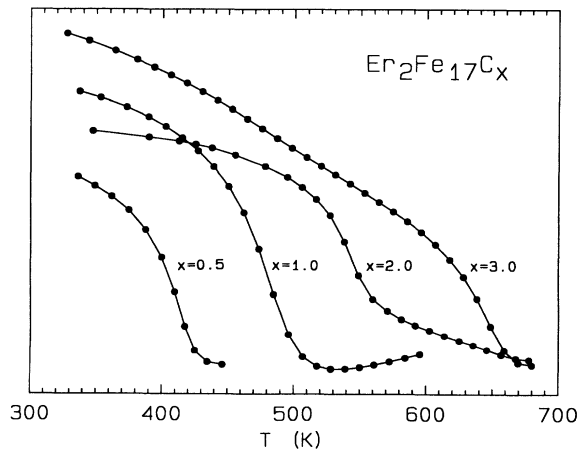


FIG. 4. M - T plots measured in the Faraday balance for $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds. The lines are guides to the eye.

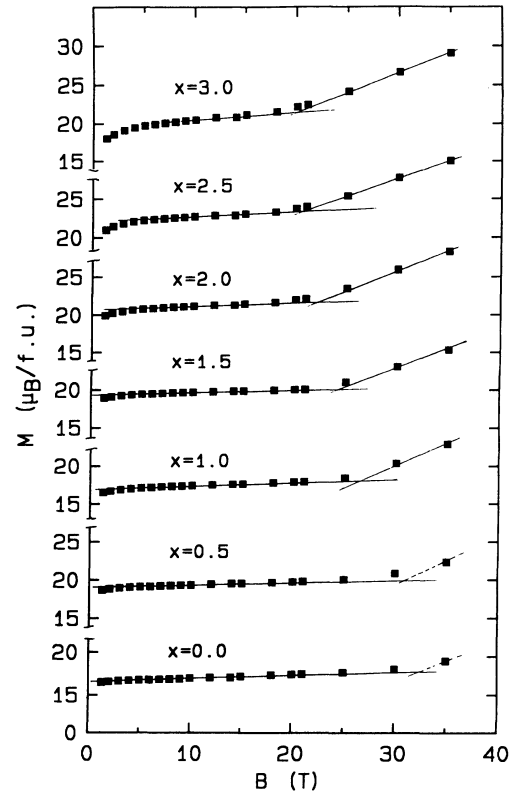


FIG. 5. Magnetic isotherms at 4.2 K for free-powder samples of $\text{Er}_2\text{Fe}_{17}\text{C}_x$. The lines are guides to the eye.

III. DISCUSSION

A. The magnetic phase diagram of the series

In Fig. 3, one can see the dependence of the spin-reorientation temperature (T_{sr}) on the carbon content. These results are in good agreement with the results reported already in the literature (see Table I).

TABLE I. Curie temperatures (T_c) and spin-reorientation temperatures (T_{sr}) of $\text{Er}_2\text{Fe}_{17}\text{C}_x$. Note: The data in brackets { } are the temperatures of the other peaks on the same curves.

Comp.	T_c (K)	T_{sr} (K)	Ref.
$x=0.0$	305		12
$x=0.5$	422		This work
$x=1.0$	488		2
	495	111{95}	5
	517	91	10
	489	105{88}	This work
$x=1.5$	509	125	5
	560	133	10
	529	120	This work
$x=2.0$	610	141	10
	549	121	This work
$x=2.5$	648	136	10
	594	127	This work
$x=3.0$	680	135	10
	645	125	This work

The spin configuration in $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds depends on the competition between the Er-sublattice anisotropy (favoring the x axis) and the Fe-sublattice anisotropy (favoring the basal plane). In the case of $\text{Er}_2\text{Fe}_{17}$, the Fe-sublattice anisotropy is dominant in the whole magnetically ordered range. After carbonation, the uniaxial Er-sublattice anisotropy is enhanced to such an extent that it dominates the planar Fe-sublattice anisotropy at low temperatures. This leads to a spin reorientation from the c direction (for $T < T_{\text{sr}}$) to the basal plane (for $T > T_{\text{sr}}$) or other directions. In the present investigation, it is found that the spin reorientation only takes place for $x \geq 1.0$ and that the spin-reorientation temperature reaches its maximum value for $x = 2.5$.

The Curie temperatures (T_c) of the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds have been determined by means of Faraday-balance measurements (see Fig. 4). The Curie temperatures reported earlier for $\text{Er}_2\text{Fe}_{17}$ are 292 K (Ref. 5) and 305 K.¹³ The introduction of carbon atoms increases the Curie temperature remarkably. The results are in good agreement with the data given by Kong, Cao, and Shen,¹⁰ Zhong *et al.*,² and Kou *et al.*⁵ (see Table I).

It is generally accepted that in R -Fe compounds the expansion of the unit-cell volumes leads to an enhancement of the Fe-Fe exchange interaction.¹⁴ Our results show that further expansion of the unit cells upon carbonation results in a further increase of Curie temperature, indicating an even more strong Fe-Fe exchange interaction. The jump of the Curie temperatures from about 300 K in $\text{Er}_2\text{Fe}_{17}$ to about 420 K in $\text{Er}_2\text{Fe}_{17}\text{C}_{0.5}$ indicates that the enhancement is particularly strong upon introduction of small amounts of carbon atoms. Similar results were reported by Kou *et al.*⁵ and Kong, Cao, and Shen.¹⁰

The transition points from the measurements shown in Figs. 3 and 4 have been used to construct a magnetic phase diagram of the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ series, which is shown in Fig. 6.

B. Effect of the interstitial carbon atoms on the magnetization

For ferrimagnetic compounds, a two-sublattice model based on the mean-field approximation has been

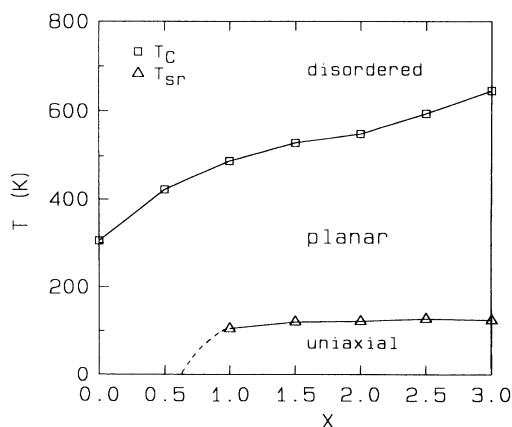


FIG. 6. Spin phase diagram of the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ series.

developed by Clark and Callen¹⁵ and applied to free single-crystalline powder particles of R - T (T =transition metal) compounds by Verhoef, Radwanski, and Franse.¹⁶ This model describes the magnetization process of single-crystalline particles of ferrimagnetic compounds that are free to rotate in the sample holder. The field-induced transitions observed in the high-field magnetic isotherms correspond with the starting points of a bending process of the antiparallel sublattice moments. With this model we have successfully analyzed the high-field magnetization processes in numerous R - T compounds.¹⁷ The observed field-induced transitions in the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds are shown in Fig. 5. In the low-field part of the magnetization curves, the ferrimagnetic orientation is not affected by the applied field and the magnetization corresponds to the difference of the two sublattice magnetizations, $M_{\text{Fe}} - M_{\text{Er}}$. If we assume that the Er moment is equal to the free-ion value, this value for the magnetization provides information about M_{Fe} . Information regarding the intersublattice molecular-field coefficient n_{ErFe} can be obtained from the value of the critical field ($B_{\text{cr}} = n_{\text{ErFe}} |M_{\text{Fe}} - M_{\text{Er}}|$), where bending of the two sublattice moments towards ferromagnetic alignment starts, and from the magnetization behavior above this critical field ($n_{\text{ErFe}} = B/M$).

In Fig. 5 it can be seen that for the compounds with $x = 3.0, 2.5,$ and 2.0 the magnetic isotherms have moderate values for B_{cr} , so that accurate values for n_{ErFe} can be obtained from the high-field magnetization behavior. However, in the case of the compounds with $x = 1.5, 1.0,$ and 0.5 , the high-field part of the magnetization curves where the bending process is too limited to allow for a precise determination of the n_{ErFe} values. For the cases of $x = 1.5$ and 1.0 , the values of n_{ErFe} have been derived from the transition fields B_{cr} . The determination n_{ErFe} via B_{cr} has also been done for the compounds with $x = 3.0, 2.5,$ and 2.0 . Agreement is achieved between the data derived in two ways for these three compounds. For the compound with $x = 0.5$, the value of B_{cr} is not sufficiently accurate to determine n_{ErFe} .

It should be kept in mind that, in deriving these parameters, we have to take into account the effect of magnetic impurity phases, mainly α -Fe. However, there is not yet an accurate way to detect the nature and to determine the amount of the impurity phases in the samples. Therefore it is difficult to determine precisely the saturation magnetization of the compounds. In this investigation, we estimated the amounts of α -Fe to be around 5% by analyzing the x-ray-diffraction patterns, and we ignore the effect of other impurity phases. With this estimation, the magnetization values of the carbides can be derived to be about $36 \mu_B/\text{f.u.}$, indicating that the magnetizations of the compounds have not changed much upon carbonation. This is consistent with a previous report about the magnetization of the interstitial carbides.⁶

The exchange constant J_{RT} that represents the exchange parameter appearing in a nearest-neighbor Heisenberg-type Hamiltonian can be derived from n_{RT} , as was described elsewhere.¹⁶ The values of J_{ErFe} for the $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds have been derived and are listed in Table II. In this table, we included the values of the ex-

TABLE II. Magnetization and the magnetic-coupling parameters for $\text{Er}_2\text{Fe}_{17}\text{C}_x$ compounds. Note: The data with * are quoted from Ref. 17. The bars--indicate that no data are available.

	n_{ErFe} (10^{23} T f.u./A m ²)	B_{exch} (T)	$-J_{\text{ErFe}}/k$ (K)
$\text{Er}_2\text{Fe}_{17}$	--	--	7.5*
$\text{Er}_2\text{Fe}_{17}\text{C}_{0.5}$	--	--	--
$\text{Er}_2\text{Fe}_{17}\text{C}_{1.0}$	2.03	220	7.3
$\text{Er}_2\text{Fe}_{17}\text{C}_{1.5}$	2.00	215	7.2
$\text{Er}_2\text{Fe}_{17}\text{C}_{2.0}$	1.95	210	7.0
$\text{Er}_2\text{Fe}_{17}\text{C}_{2.5}$	1.87	200	6.7
$\text{Er}_2\text{Fe}_{17}\text{C}_{3.0}$	1.86	195	6.6

change fields B_{exch} , which are also a measure for the strength of the exchange interaction.

In Table II, it is seen that the strength of the Er-Fe exchange interaction decreases monotonically with increasing carbon content. This result is visualized in Fig. 7. In fact, one can easily see from Fig. 5 that there is a tendency in the figure that the transition field shifts gradually to lower fields with increasing carbon contents. This is a direct result of the decrease of n_{ErFe} with increasing carbon contents. Although we argued above that the impurity phases may have an influence on the isotherms, this direct comparison can be made without risk because all the carbides contain roughly a similar amount of impurity phases.

As we mentioned at the beginning of this paper, different results have been reported about the effect of carbonation on the Er-Fe exchange interaction in $\text{Er}_2\text{Fe}_{17}$. Our experimental findings in this respect are clear (cf. Fig. 7); in what follows we outline a plausible interpretation of these results.

The R - T exchange interaction in $3d$ - $4f$ compounds is mediated by the interatomic $3d$ - $5d$ interaction and the intraatomic $4f$ - $5d$ interaction. For a given series of R - T compounds, the absolute value of J_{RT} usually decreases monotonically with increasing atomic number of the R component. This has experimentally been observed in most of the studied series¹⁷ and has theoretically been explained as being due to an increasing spatial separation

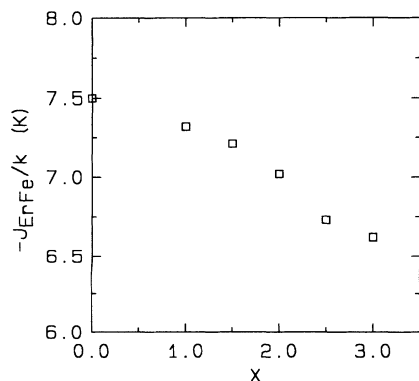


FIG. 7. Dependence of $-J_{\text{ErFe}}/k$ for $\text{Er}_2\text{Fe}_{17}\text{C}_x$ on the carbon content x .

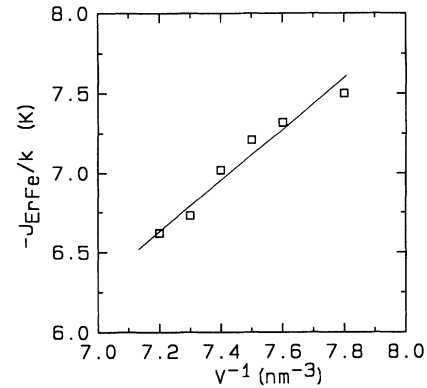


FIG. 8. Dependence of $-J_{\text{ErFe}}/k$ for $\text{Er}_2\text{Fe}_{17}\text{C}_x$ on the reciprocal unit-cell volume normalized to one R atom. The line is a guide to the eye.

between $4f$ and $5d$ shells in the R atoms.¹⁸

A reasonable inference we can make for the interstitial compounds is that, with increasing interatomic distance and thus a larger spatial separation between $3d$ and $5d$ shells, a reduction in hybridization between the $3d$ and $5d$ electron states will occur. If the lattice expands gradually, a monotonic decrease of $|J_{RT}|$ should be expected, which corresponds to the result we have obtained. In Fig. 8, $-J_{\text{ErFe}}$ is plotted vs the corresponding reciprocal volume (normalized unit-cell volume per R atom). A nearly linear relationship is found. In Ref. 17, $-J_{RT}$ is plotted vs the corresponding reciprocal volume for a large number of $\text{Er}-T$ compounds. Also there quite a good correlation was found. So the behavior of J_{RT} with regard to the corresponding volumes observed in the course of the present investigation and in Ref. 17 are consistent. This volume dependence may provide an explanation for the strong dependence of J_{RT} on the concentration of the T components which is still an unsolved problem.

Another feature worth mentioning here is that the effect of the interstitial atoms on the R - T exchange interaction also depends on the nature of the interstitial atoms. Figure 9 shows the magnetic isotherms of free-

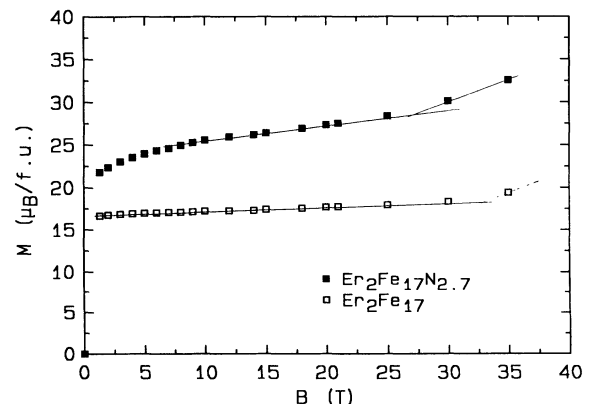


FIG. 9. Magnetic isotherms at 4.2 K for free-powder samples of $\text{Er}_2\text{Fe}_{17}\text{N}_{2.7}$ and $\text{Er}_2\text{Fe}_{17}$. The lines are guides to the eye.

powder $\text{Er}_2\text{Fe}_{17}\text{N}_{2.7}$ and $\text{Er}_2\text{Fe}_{17}$ compounds. It should be borne in mind that the unit-cell expansion of $\text{Er}_2\text{Fe}_{17}\text{N}_{2.7}$ is similar to that of $\text{Er}_2\text{Fe}_{17}\text{C}_{3.0}$. However, by comparing the curves in Figs. 5 and 9 one finds that the decrease of the critical field upon full nitrogenation is similar to that of $\text{Er}_2\text{Fe}_{17}\text{C}_{1.0}$, which means that the reduction of the exchange-interaction strength upon nitrogenation is less obvious than that of carbonation at the same expansion of the unit cell. A reasonable explanation for this behavior may be found in the difference in band structures between $R_2\text{Fe}_{17}\text{N}_x$ and $R_2\text{Fe}_{17}\text{C}_x$ and the concomitant higher hybridization of the $3d$ electron states with the valence-electron states of carbon than of nitrogen.¹⁹

IV. CONCLUSIONS

A series of the interstitial carbides $\text{Er}_2\text{Fe}_{17}\text{C}_x$ has been synthesized by means of normal arc casting and by means of the gas-phase-interaction method. The unit-cell volumes of the compounds are found to expand in a monotonic way with increasing carbon concentration. The structure type may change with x when the carbon atoms are introduced into the parent compounds by means of casting ($x \geq 1.5$), whereas carbonation with the GPI method does not change the structure.

The concentration dependence of the magnetic phase transitions and the magnetic coupling have been investigated systematically. A spin-reorientation transition is observed in the compounds with a carbon content larger than $x=0.5$, which is explained as resulting from the enhancement of the uniaxial R -sublattice anisotropy after carbonation.

A remarkable increase of the Curie temperatures upon carbonation is found, indicating a strong increase of the Fe-Fe exchange interactions in the carbides.

By means of high-field magnetization measurements on powder particles that were free to rotate in the sample holder, we have determined the Er-Fe exchange-interaction constants J_{ErFe} in the interstitial compounds and found that $|J_{\text{ErFe}}|$ decreases with increasing carbon content. It has also been found that $|J_{\text{ErFe}}|$ increases linearly with increasing reciprocal unit-cell volumes, i.e., with increasing overlap of the $3d$ and $5d$ wave functions. Furthermore, carbon atoms have a stronger effect on the Er-Fe coupling than that of nitrogen atoms.

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