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On the 4f–3d exchange interaction in intermetallic compounds

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Abstract

A review is given of the experimental values of the magnetic intersublattice-coupling constants in rare-earth (R) – transition-metal (T) intermetallics, derived from high-field magnetization measurements on single-crystalline powder particles that are free to be oriented by the applied magnetic field. The results are compared with values for the intersublattice-coupling constant obtained by other experimental methods and with results of electronic-band-structure calculations made for a few of the compounds investigated. An empirical relationship is found between the intersublattice-coupling constants and the reciprocal normalized molar volumes of the various R–T compounds, including T = Ni, Co and Fe.

1. Introduction

Alloys and intermetallic compounds obtained by combining rare-earth metals (R) with 3d metals (T) form an important class of materials that find applications in permanent magnets, magnetostrictive devices and magneto-optical recording. In general it can be said that the 3d sublattice is responsible for a sufficiently high magnetization and magnetic-ordering temperature while the 4f sublattice (R sublattice) provides a high enough magnetic anisotropy. Optimal properties can be expected only when there is a sufficiently strong magnetic coupling between the two sublattices.

The intersublattice-coupling strength can be determined experimentally by several different

methods. Neutron scattering is the least generally accessible method and has been used up to now only in a few compounds of the RFe_2 and $R_2Fe_{14}B$ type [1,2]. When single crystals are available, the intersublattice-coupling strength can be derived by analyzing the low-temperature magnetic isotherms measured along the main crystallographic directions. This method has been used primarily for compounds of the type R_2Co_{17} , R_2Fe_{17} and $R_2Fe_{14}B$ [3]. Rare-earth elements that have a suitable Mossbauer nucleus can also be used to determine the magnetic-coupling constant in the compounds of the corresponding 4f element with 3d elements [4]. It is also possible to determine the intersublattice coupling strength from the difference in magnetic-ordering temperature of compounds in which R has a magnetic moment and in which R is non-magnetic, see for instance Refs. [3,5,6]. More recently, the high-field free-powder method (HFFP) has become

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applicable and has been used to determine the intersublattice-coupling strength in a fairly large number of different compounds [7,8]. A more detailed description of this method and a comparison between this method and the other methods to determine the intersublattice-coupling constants will be given later in this review.

From the theoretical point of view, several models exist which have been able to explain why the heavy rare-earth spin invariably couples antiparallel with the transition-metal spin [9] and have predicted that there is a general decrease in the 4f–3d coupling constant when proceeding from La to Lu across the lanthanide series [10]. In this article it will be discussed in how far the latter predictions are correct. The problem of the 4f–3d exchange coupling has also been addressed in various types of electronic-band-structure calculations [11,12] and some of these calculations have offered more details about the general behavior mentioned above.

Neither from the theoretical models nor from the limited number band-structure calculations that have been made so far, has a description emerged of the variation of the interatomic-exchange coupling in different types of compounds. For this reason, it is not yet possible to describe the variation of the interatomic exchange coupling in this class of materials in terms of other physical properties. Such a description would be very desirable since it would allow to make a priori predictions of the intersublattice-coupling strength. It is a purpose of the present study to investigate whether there exists a correlation between the intersublattice-coupling strength and some other well established physical parameters. It will be shown that there is a strong correlation between this coupling strength and the reciprocal molar volume.

This paper is organized as follows. In Section 2, we will give a theoretical outline of the HFFP method. In Section 3, we will present a digest of the numerous data on the intersublattice-exchange-coupling constant obtained by means of the HFFP method and discuss the systematics of the results. These results will be compared with data obtained by other methods in Section 4, and with results of band-structure calculations in Sec-

tion 5. Finally, in Section 6, we will discuss the possibility to correlate the intersublattice-coupling strength with other physical parameters.

2. Fundamentals of the HFFP method

In previous investigations [7,8], the magnetic isotherms measured at 4.2 K in fields up to 40 T were analyzed by means of a two-sublattice mean-field model using the fact that the powder particles employed for the magnetization measurements are fairly small so that they may be regarded as monocrystalline particles (particle size $\leq 40 \mu\text{m}$). These particles will orient their magnetic moment parallel to the external field, since they can rotate freely in the sample holder, the field strengths used in the experiments being much larger than the stray fields of adjacent particles.

Previous investigations [3] have shown that the anisotropy energy of the 3d sublattice at 4.2 K is generally much smaller than the 4f-sublattice anisotropy, so that the former can be ignored and the R-sublattice magnetization can be taken parallel to its easy magnetization direction for all possible moment configurations of the two sublattices. When minimizing the free energy E in the presence of an external field, we need for this reason only take into consideration the exchange energy and the Zeeman energy. The free energy then has the form [7]

$$E = M_T M_R n_{RT} \cos \alpha - B [M_T^2 + M_R^2 + 2 M_T M_R \cos \alpha]^{1/2}, \quad (1)$$

where α is the angle between the two sublattice magnetizations ($\alpha \leq 180^\circ$) and n_{RT} the intersublattice-molecular-field coefficient. This coefficient is related to the R–T exchange-coupling constant J_{RT} appearing in the nearest-neighbor Heisenberg-type Hamiltonian $H_{\text{ex}} = -\sum 2J_{RT} S_R S_T$ via the expression

$$n_{RT} = \frac{J_{RT} Z_{RT} (g_R - 1)}{N_T \mu_B^2 g_R} \quad (2)$$

where Z_{RT} represents the number of nearest transition-metal-atom neighbors to an R atom and N_T is the number of transition-metal atoms per formula unit and we have assumed that $g_T = 2$. It should be borne in mind that only nearest-neighbor interactions have been taken into account in Eq (2)

The equilibrium state is found by minimizing the free energy (Eq (1)) with respect to α . The behavior expected as a function of the applied field B can then be characterized as follows: for relatively low fields, the moment configuration is strictly antiparallel and the magnetization is equal to $M_s = |M_T - M_R|$. Beyond a critical field strength ($B_{crit} = |M_T - M_R|/n_{RT}$), the exact an-

tiparallel moment configuration is broken and the two sublattice moments commence to bend towards each other. From there on, the magnetic moment is described by $M = B/n_{RT}$ and thus

$$n_{RT} = (dB/dM)^{-1} \quad (3)$$

This means that n_{RT} can be derived straightforwardly from the slope of the magnetic isotherm at 4.2 K, provided that the sloping range falls within the field range available. The method is illustrated by means of Fig 1 (top part), where also examples of experimental curves are shown (bottom part).

It should be mentioned that the values of J_{RT} derived directly from the experimental curves via Eq (2) are actually far from unique due to the difficulties in determining the values of Z_{RT} . Despite some attempts to introduce a rational restriction rule [13], the choices for the latter in compounds with complex structures remain ambiguous.

The molecular field experienced by the R moments can be given in an obvious notation as

$$B_{mol} = n_{RT}M_T + n_{RR}M_R = B_{mol}^{RT} + B_{mol}^{RR} \quad (4)$$

In most cases the R–R interaction can be neglected, being more than an order of magnitude smaller than the R–T interaction. In that case, one has $B_{mol} = B_{mol}^{RT} = n_{RT}M_T$. Frequently, the exchange field B_{exch} (acting on the spins) is considered instead of the molecular field (acting on the moments). Both quantities can be transformed into each other via the expression

$$B_{mol} = \frac{2(g_R - 1)}{g_R} B_{exch} = \gamma B_{exch} \quad (5)$$

and

$$B_{exch} = n_{RT}M_T/\gamma \quad (6)$$

It should be kept in mind that B_{exch} (or B_{mol}) as expressed in Eq (6) is a purely experimental quantity, if n_{RT} and M_T can be derived from the high-field measurements. Unlike J_{RT} , no assumption regarding the number of neighbors has to be taken into account. For this reason, we have listed also B_{exch} in the tables presented below.

Fig 1 Schematic representation of the low-, medium- and high-field behaviour of magnetic isotherms obtained from the HFFP method (top part). Experimental magnetic isotherms obtained for two compounds of the $\text{EuNi}_{12}\text{B}_6$ type by means of the HFFP method.

Table 1

R–T compounds investigated to study the magnetic coupling by means of the HFFP method. Included are the coupling constants $-J_{\text{RT}}/k$ (in K), the magnetic moments of the T sublattice M_{T} (in μ_{B} per formula unit), the exchange fields B_{exch} (in T) and numbers of the references to the original papers. The bold data are assessed values for the compounds without substitutions.

RFe ₂ -based compounds ($Z_{\text{RFe}} = 12$)			M_{Fe}	B_{exch}	$-J_{\text{RFe}}/k$	Ref
GdFe₂			3 2	315	22	
Gd _{0.40} Y _{0.60} Fe ₂	free powder				22 0	[17]
TbFe₂			3 2	300	21	
Tb _{0.30} Y _{0.70} Fe ₂	free powder				21 0	[17]
DyFe₂			3 2	300	21	
Dy _{0.30} Y _{0.70} Fe ₂	free powder				21 0	[17]
HoFe₂			3 2	300	21	
Ho _{0.30} Y _{0.70} Fe ₂	free powder				21 0	[17]
ErFe₂			3 2	270	19	
ErFe ₂	free powder		3 2			[17]
Er _{0.40} Y _{0.60} Fe ₂	free powder		2 8		19 0	[17]
Er _{0.35} Y _{0.65} Fe ₂	free powder		2 7		18 3	[17]
Er _{0.30} Y _{0.70} Fe ₂	free powder		2 7		19 3	[17]
Er _{0.25} Y _{0.75} Fe ₂	free powder		2 7		20 6	[17]
RFe ₃ -based compounds ($Z_{\text{RFe}} = 14$)			M_{Fe}	B_{exch}	$-J_{\text{RFe}}/k$	Ref
GdFe₃			5 2	220	14	
Gd _{0.75} Y _{0.25} Fe ₃	free powder		5 2		14 4	[17]
TbFe₃			5 2	220	14	
Tb _{0.60} Y _{0.40} Fe ₃	free powder				14 1	[17]
DyFe₃			5 2	240	16	
Dy _{0.50} Y _{0.50} Fe ₃	free powder				15 5	[17]
HoFe₃			5 2	260	17	
Ho _{0.50} Y _{0.50} Fe ₃	free powder				16 6	[17]
ErFe₃			5 2	260	17	
Er _{0.45} Y _{0.55} Fe ₃	free powder				16 7	[17]
RCO ₃ -based compounds ($Z_{\text{RCO}} = 14$)			M_{Co}	B_{exch}	$-J_{\text{RCO}}/k$	Ref
TbCo₃			2 5	110	13	
Tb _{0.30} Y _{0.70} Co ₃	free powder		2 7		12 8	[18]
DyCo₃			2 5	120	14	
Dy _{0.25} Y _{0.75} Co ₃	free powder		2 5		13 8	[18]
HoCo₃			2 5	120	14	
Ho _{0.20} Y _{0.80} Co ₃	free powder		2 5		13 6	[18]
ErCo₃			2 5	110	13	
Er _{0.25} Y _{0.75} Co ₃	free powder		3 3		13 0	[18]
R ₂ Co ₇ -based compounds ($Z_{\text{RCO}} = 15$)			M_{Co}	B_{exch}	$-J_{\text{RCO}}/k$	Ref
Gd₂Co₇			10 4	200	12	
Gd ₂ Co ₇	free powder		10 4			[19]
Gd _{1.5} Y _{0.5} Co ₇	free powder		10 2		11 2	[19]
Gd _{1.4} Y _{0.6} Co ₇	free powder		10 3		11 6	[19]
Gd _{1.3} Y _{0.7} Co ₇	free powder		10 3		12 7	[19]
Gd _{1.2} Y _{0.8} Co ₇	free powder		10 3			[19]
Gd _{1.1} Y _{0.9} Co ₇	free powder		10 3			[19]
Gd _{0.9} Y _{1.1} Co ₇	free powder		10 1			[19]
Tb₂Co₇			10 7	190	11	
Tb ₂ Co ₇	free powder		10 7			[19]
Tb _{1.3} Y _{0.7} Co ₇	free powder		10 0		10 9	[19]
Tb _{1.2} Y _{0.8} Co ₇	free powder		9 5		10 5	[19]
Tb _{1.0} Y _{1.0} Co ₇	free powder		10 4		11 1	[19]
Tb _{0.8} Y _{1.2} Co ₇	free powder		10 0			[19]

Table 1 (continued)

R ₂ Co ₇ -based compounds ($Z_{\text{RCo}} = 15$)		M_{Co}	B_{exch}	$-J_{\text{RCo}}/k$	Ref
Dy₂Co₇		10 7	190	11	
Dy ₂ Co ₇	free powder	10 7			[19]
Dy _{1 0} Y _{1 0} Co ₇	free powder	10 9		10 2	[19]
Dy _{0 9} Y _{1 1} Co ₇	free powder	10 4		10 8	[19]
Dy _{0 8} Y _{1 2} Co ₇	free powder	10 3		13 3	[19]
Ho₂Co₇		10 1	190	12	
Ho ₂ Co ₇	free powder	10 1		12 0	[19]
Ho _{1 0} Y _{1 0} Co ₇	free powder	11 7		12 0	[19]
Ho _{0 8} Y _{1 2} Co ₇	free powder	10 8		12 0	[19]
Er₂Co₇		11 1	210	12	
Er ₂ Co ₇	free powder	11 1			[19]
Er _{1 2} Y _{0 8} Co ₇	free powder	10 2		11 7	[19]
Er _{1 1} Y _{0 9} Co ₇	free powder	10 3		12 0	[19]
Er _{1 0} Y _{1 0} Co ₇	free powder	10 1			[19]
Er _{0 9} Y _{1 1} Co ₇	free powder	10 3			[19]
Er _{0 8} Y _{1 2} Co ₇	free powder	9 7			[19]
Tm₂Co₇		11 2	215	12	
Tm ₂ Co ₇	free powder	11 2		10 8	[19]
Tm _{1 1} Y _{0 9} Co ₇	free powder	11 3		12 6	[19]
R ₂ Ni ₇ -based compounds ($Z_{\text{RNi}} = 15$)		M_{Ni}	B_{exch}	$-J_{\text{RNi}}/k$	Ref
Gd₂Ni₇		1 2	25	13	
Gd ₂ Ni ₇	free powder	1 2			[20]
Gd _{0 20} Y _{1 80} Ni ₇	free powder	0 56		15 4	[20]
Gd _{0 15} Y _{1 85} Ni ₇	free powder	0 69		10 8	[20]
Gd _{0 10} Y _{1 90} Ni ₇	free powder	0 42		11 2	[20]
Gd _{0 08} Y _{1 92} Ni ₇	free powder	0 83		15 1	[20]
Tb₂Ni₇		1 2	21	11	
Tb _{0 15} Y _{1 85} Ni ₇	free powder	0 55		9 7	[20]
Tb _{0 10} Y _{1 90} Ni ₇	free powder	0 45		11 4	[20]
Dy₂Ni₇		1 2	21	11	
Dy _{0 14} Y _{1 86} Ni ₇	free powder	0 52		10 6	[20]
Dy _{0 10} Y _{1 90} Ni ₇	free powder	0 09			[20]
Ho₂Ni₇		1 2	15	8	
Ho _{1 0} Y _{1 0} Ni ₇	free powder	0 62		7 6	[20]
Ho _{0 8} Y _{1 2} Ni ₇	free powder	0 50		8 4	[20]
Er₂Ni₇		1 2	17	9	
Er _{0 15} Y _{1 85} Ni ₇	free powder	0 47		9 1	[20]
Er _{0 10} Y _{1 90} Ni ₇	free powder	0 63		8 8	[20]
R ₂ Co ₇ B ₃ -based compounds ($Z_{\text{RCo}} = 10$)		M_{Co}	B_{exch}	$-J_{\text{RCo}}/k$	Ref
Er₂Co₇B₃		3 2	30	9	
Er _{0 6} Y _{1 4} Co ₇ B ₃	free powder	3 2		19 8	[21]
Er _{0 5} Y _{1 5} Co ₇ B ₃	free powder	3 3		20 7	[21]
Er _{0 3} Y _{1 7} Co ₇ B ₃	free powder	3 2		23 8	[21]
Er _{0 1} Y _{1 9} Co ₇ B ₃	free powder	3 3		22 1	[21]
Note An extended model has been applied in the analysis of this series, see the reference The value of Z_{RCo} is rather ambiguous					
R ₆ Fe ₂₃ -based compounds ($Z_{\text{RFe}} = 13$)		M_{Fe}	B_{exch}	$-J_{\text{RFe}}/k$	Ref
Er₆Fe₂₃		46 5	240	12	
Er ₆ Fe ₂₃	free powder	46 5		12 1	[22]
Er ₅ YFe ₂₃	free powder	49 1		11 9	[22]
Note The value of Z_{RFe} is rather ambiguous					

Table 1 (continued)

RFe ₄ B-based compounds ($Z_{\text{RFe}} = 15$)		M_{Fe}	B_{exch}	$-J_{\text{RFe}}/k$	Ref
HoFe₄B		68	185	97	
Ho _{0.6} Lu _{0.4} Fe ₄ B	free powder	68		97	[23]
Ho _{0.4} Lu _{0.6} Fe ₄ B	free powder	68		97	[23]
ErFe₄B		68	210	11	
ErFe ₄ B	free powder	68		110	[23]
Er _{0.8} Lu _{0.2} Fe ₄ B	free powder	68		110	[23]
Er _{0.7} Lu _{0.3} Fe ₄ B	free powder	68		110	[23]
Er _{0.5} Lu _{0.5} Fe ₄ B	free powder	68		110	[23]
TmFe₄B		68	185	97	
TmFe ₄ B	free powder	68		97	[23]
Tm _{0.7} Lu _{0.3} Fe ₄ B	free powder	68		97	[23]
RCO ₄ B-based compounds ($Z_{\text{RCO}} = 15$)		M_{Co}	B_{exch}	$-J_{\text{RCO}}/k$	Ref
GdCo₄B		37	105	10	
GdCo ₄ B	free powder	37			[24]
Gd _{0.5} Y _{0.5} Co ₄ B	free powder	31		101	[24]
TbCo₄B		38	95	91	
TbCo ₄ B	free powder	38			[24]
Tb _{0.45} Y _{0.55} Co ₄ B	free powder	32		91	[24]
DyCo₄B		37	90	84	
DyCo ₄ B	free powder	37			[24]
Dy _{0.4} Y _{0.6} Co ₄ B	free powder	31		84	[24]
HoCo₄B		43	80	67	
HoCo ₄ B	free powder	43			[24]
Ho _{0.4} Y _{0.6} Co ₄ B	free powder	32		67	[24]
ErCo₄B		38	80	75	
ErCo ₄ B	free powder	38			[24]
Er _{0.45} Y _{0.55} Co ₄ B	free powder	28		75	[24]
RCO ₅ -type compounds ($Z_{\text{RT}} = 18 + \epsilon$)		M_{T}	B_{exch}	$-J_{\text{RT}}/k$	Ref
GdCo₅		89	240	10	
GdCo ₅	free powder	89			[25]
GdCo _{4.50} Ni _{0.50}	free powder	79		97	[25]
GdCo _{4.20} Ni _{0.80}	free powder	76		92	[25]
GdCo _{4.05} Ni _{0.95}	free powder	73		91	[25]
GdCo _{3.95} Ni _{1.05}	free powder	70		89	[25]
TbCo_{5.1}		90	190	81	
TbCo _{5.1}	free powder	90		81	[25]
Tb _{0.9} Y _{0.1} Co _{5.1}	free powder	87		74	[25]
DyCo_{5.2}		89	160	71	
DyCo _{5.2}	free powder	88		71	[25]
Dy _{0.9} Y _{0.1} Co _{5.2}	free powder	89		70	[25]
HoCo_{5.4}		100	180	75	
HoCo _{5.4}	free powder	100		75	[25]
Ho _{0.9} Y _{0.1} Co _{5.4}	free powder	90		71	[25]
ErCo_{5.8}		103	190	80	
ErCo _{5.8}	free powder	103		80	[25]
TmCo_{6.1}		112	180	74	
TmCo _{6.1}	free powder	112		74	[25]
R ₂ Fe ₁₄ B-based compounds ($Z_{\text{RT}} = 16$)		M_{T}	B_{exch}	$-J_{\text{RT}}/k$	Ref
Gd₂Fe₁₄B		316	270	10	
Gd ₂ Fe ₁₄ B	single crystal	316		102	[26]
Gd ₂ (Fe,Mn) ₁₄ B	free powder			102	[27]

Table 1 (continued)

R ₂ Fe ₁₄ B-based compounds (Z _{RT} = 16)		<i>M_T</i>	<i>B_{exch}</i>	− <i>J_{RT}</i> / <i>k</i>	Ref
Tb₂Fe₁₄B		31	275	11	
Tb ₂ (Fe,Mn) ₁₄ B	free powder			10.5	[27]
Dy₂Fe₁₄B		31.4	250	9.7	
Dy ₂ Fe ₁₄ B	single crystal	31.4		9.1	[26]
Dy ₂ (Fe,Mn) ₁₄ B	free powder			9.7	[27]
Ho₂Fe₁₄B		31.2	235	9.0	
Ho ₂ Fe ₁₄ B	single crystal	31.2		8.1	[28]
Ho ₂ (Fe,Mn) ₁₄ B	free powder			9.0	[27]
Er₂Fe₁₄B		31.3	230	8.8	
Er ₂ Fe ₁₄ B	free powder	31.3			[29]
Er ₂ Fe _{13.5} Al _{0.5} B	free powder	28.0		9.1	[29]
Er ₂ Fe _{13.0} Al _{1.0} B	free powder	26.2		8.8	[29]
Er ₂ Fe _{12.7} Al _{1.3} B	free powder	24.7		8.8	[29]
Er ₂ Fe _{12.4} Al _{1.6} B	free powder	23.0		8.8	[29]
Er ₂ Fe _{12.0} Al _{2.0} B	free powder	21.4		9.1	[29]
Er ₂ (Fe,Mn) ₁₄ B	free powder			7.8	[27]
Tm₂Fe₁₄B				8.4	
Tm ₂ (Fe,Mn) ₁₄ B	free powder			8.4	[27]
R ₂ Co ₁₄ B-based compounds (Z _{RT} = 16)		<i>M_T</i>	<i>B_{exch}</i>	− <i>J_{RT}</i> / <i>k</i>	Ref
Gd₂Co₁₄B		19.7	195	11	
Gd ₂ Co ₁₄ B	aligned powder	19.7		10.5	[30]
Gd ₂ (Co,Al) ₁₄ B	aligned powder	19.4		12.6	[31]
Tb₂Co₁₄B		20.2	180	9.7	
Tb ₂ Co ₁₄ B	free powder	20.2		9.7	[30]
Dy₂Co₁₄B		20.0	170	9.5	
Dy _{1.8} La _{1.2} Co ₁₄ B	free powder	20.0		9.5	[30]
Ho₂Co₁₄B		20.0	160	9.1	
Ho _{1.8} La _{1.2} Co ₁₄ B	free powder	20.0		9.1	[30]
R ₂ Fe ₁₄ C-based compounds (Z _{RT} = 16)		<i>M_T</i>	<i>B_{exch}</i>	− <i>J_{RT}</i> / <i>k</i>	Ref
Gd₂Fe₁₄C		30.0	270	11	
Gd ₂ (Fe,Mn) ₁₄ C	free powder			10.5	[32]
Ho₂Fe₁₄C		31.6	240	9.0	
Ho ₂ (Fe,Mn) ₁₄ C	free powder			9.0	[33]
Er₂Fe₁₄C		29.5	215	8.6	
Er ₂ (Fe,Mn) ₁₄ C	free powder			8.6	[34]
R ₂ Fe ₁₇ -based compounds (Z _{RT} = 19)		<i>M_T</i>	<i>B_{exch}</i>	− <i>J_{RT}</i> / <i>k</i>	Ref
Gd₂Fe₁₇		36.9	305	10	
Gd ₂ Fe ₁₇	single crystal	36.9	260	8.5	[26]
Gd ₂ Fe ₁₂ Al ₅	free powder	20.4			[35]
Gd ₂ Fe ₁₁ Al ₆	free powder	17.6			[35]
Gd ₂ Fe ₁₀ Al ₇	free powder	15.6		10.1	[35]
Gd ₂ Fe ₉ Al ₈	free powder	14.0		10.3	[35]
Gd ₂ Fe _{8.5} Al _{8.5}	free powder	12.2		10.5	[35]
Gd ₂ Fe ₁₆ Mn ₁	free powder	29.7			[35]
Gd ₂ Fe ₁₅ Mn ₂	free powder	24.9			[35]
Gd ₂ Fe ₁₃ Mn ₄	free powder	17.4			[35]
Gd ₂ Fe ₁₂ Mn ₅	free powder	15.7			[35]
Gd ₂ Fe ₁₁ Mn ₆	free powder	10.7		10.4	[35]
Tb₂Fe₁₇		36.6	275	9	
Tb ₂ Fe ₁₇	single crystal	36.6	215	7.0	[26]

Note: The data for the aligned samples have been obtained by fitting magnetization curves.

Table 1 (continued)

R ₂ Fe ₁₇ -based compounds ($Z_{RT} \approx 19$)		M_T	B_{exch}	$-J_{RT}/k$	Ref
Tb ₂ Fe ₁₀ Al ₇	free powder	19.1		10.4	[35]
Tb ₂ Fe ₉ Al ₈	free powder	14.8		10.4	[35]
Dy₂Fe₁₇		36.9	215	7.0	
Dy ₂ Fe ₁₇	single crystal	36.9	215	7.0	[36]
Dy ₂ Fe ₁₇	free powder	36.0			[37]
Dy ₂ Fe ₁₆ Al ₁	free powder	32.9			[37]
Dy ₂ Fe ₁₅ Al ₂	free powder	29.5			[37]
Dy ₂ Fe ₁₄ Al ₃	free powder	27.0		(10.6)	[37]
Dy ₂ Fe ₁₃ Al ₄	free powder	24.0		9.0	[37]
Dy ₂ Fe ₁₂ Al ₅	free powder	20.7		8.8	[37]
Dy ₂ Fe ₁₁ Al ₆	free powder	19.2		9.2	[37]
Dy ₂ Fe ₁₀ Al ₇	free powder	17.7		10.1	[37]
Dy ₂ Fe ₉ Al ₈	free powder	16.3		10.9	[37]
Ho₂Fe₁₇		38.7	220	7.0	
Ho ₂ Fe ₁₇	single crystal	38.7	220	6.8	[36]
Ho ₂ Fe ₁₄ Al ₃	free powder	26.9		7.8	[38]
Ho ₂ Fe ₁₃ Al ₄	free powder	24.2		8.3	[38]
Ho ₂ Fe ₁₂ Al ₅	free powder	20.0		8.5	[38]
Ho ₂ Fe ₁₁ Al ₆	free powder	15.8		8.3	[38]
Ho ₂ Fe ₁₀ Al ₇	free powder	14.0		9.0	[38]
Ho ₂ Fe ₉ Al ₈	free powder	12.7		8.4	[38]
Ho ₂ Fe ₈ Al ₉	free powder	11.9		(10.0)	[38]
Ho ₂ Fe ₇ Al ₁₀	free powder	11.1		(11.9)	[38]
Er₂Fe₁₇		35.5	200	7.5	
Er ₂ Fe ₁₇	single crystal	35.5	190	6.4	[36]
Er ₂ Fe ₁₄ Al ₃	free powder	25.9		(8.3)	[18]
Er ₂ Fe ₁₃ Al ₄	free powder	23.3		7.5	[18]
Er ₂ Fe ₁₂ Al ₅	free powder	20.8		7.5	[18]
Er ₂ Fe ₁₁ Al ₆	free powder	18.0		7.5	[18]
Er ₂ Fe ₁₀ Al ₇	free powder	19.8		8.2	[37]
Er ₂ Fe ₉ Al ₈	free powder	15.0		9.2	[37]
Er ₂ Fe ₈ Al ₉	free powder	13.3		8.9	[18]
Tm₂Fe₁₇					
Tm ₂ Fe ₉ Al ₈	free powder			9.4	[37]

Note: The data in parentheses may be considered as less accurate, since the critical fields are very high.

R ₂ Co ₁₇ -based compounds ($Z_{RT} \approx 19$)		M_T	B_{exch}	$-J_{RT}/k$	Ref
Pr₂Co₁₇		27.0	185	8.3	
Pr ₂ Co ₁₇	single crystal	27.0	185	8.3	[36]
Nd₂Co₁₇		27.4	165	7.2	
Nd ₂ Co ₁₇	single crystal	27.4	165	7.2	[36]
Gd₂Co₁₇		27	200	9.6	
Gd ₂ Co ₁₄ Al ₃	free powder	17.3		9.6	[18]
Tb₂Co₁₇		28.1	185	8.0	
Tb ₂ Co ₁₇	single crystal	28.1	185	8.0	[36]
Tb ₂ Co ₁₄ Al ₃	free powder	17.9		8.3	[18]
Dy₂Co₁₇		27.4	165	7.3	
Dy ₂ Co ₁₇	single crystal	27.4	165	7.3	[36]
	free single crystal from [36]	27.4	170	7.4	[26]
	single crystal	27.7	170	7.3	[26]
	free powder	28.3	176	7.5	[39]
Dy ₂ Co ₁₅ Al ₂	free powder	18.2		7.7	[18]
Dy ₂ Co ₁₄ Al ₃	free powder	18.7		7.8	[18]

Table 1 (continued)

R ₂ Co ₁₇ -based compounds (Z _{RT} = 19)		<i>M_T</i>	<i>B_{exch}</i>	$-J_{RT}/k$	Ref
Ho₂Co₁₇		28 1	165	7.0	
Ho ₂ Co ₁₇	single crystal	28 1	160	6 8	[36]
	free single crystal from [36]	28 1	165	7 1	[26]
	single crystal (Ho rich)	27 9	155	6 7	[26]
	free single cryst (Ho rich)	27 9	160	6 8	[26]
	free powder	29 5	179	7 3	[39]
Ho ₂ Co ₁₄ Fe ₃	single crystal	30 8	175	6 8	[36]
	free single crystal from [36]	30 8	175	6 9	[26]
Ho ₂ Co ₁₅ Al ₂	free powder	18 9		7 3	[18]
Ho ₂ Co ₁₄ Al ₃	free powder	18 0		7 2	[18]
Er₂Co₁₇		29.4	165	6.9	
Er ₂ Co ₁₇	single crystal	29 4	145	6 0	[36]
	free single crystal from [36]	29 4	170	6 9	[26]
	free powder	28 3	162	6 9	[39]
Er ₂ Co ₁₄ Al ₃	free powder	17 8		7 2	[36]
Tm₂Co₁₇		27 2	150	6.5	
Tm ₂ Co ₁₇	free powder	27 2	152	6 7	[39]
R ₂ Fe ₁₇ N _{2.7} -type compounds (Z _{RT} = 19)		<i>M_T</i>	<i>B_{exch}</i>	$-J_{HoT}/k$	Ref
Ho₂Fe₁₇N_{2.7}		38	235	7.5	
Ho ₂ Fe ₁₇ N _{2.7}	free powder	45 0			[40]
Ho ₂ Fe ₁₅ Mn ₂ N _{2.7}	free powder	27 7		8 1	[40]
Ho ₂ Fe ₁₄ Mn ₃ N _{2.7}	free powder	29 5		6 9	[40]
Ho ₂ Fe ₁₃ Mn ₄ N _{2.7}	free powder	27 4		7 5	[40]
R ₂ Fe ₁₇ C-based compounds (Z _{RT} = 19)		<i>M_T</i>	<i>B_{exch}</i>	$-J_{RT}/k$	Ref
Dy₂Fe₁₇C		37 0	220	7.2	
Dy ₂ Fe ₁₇ C	aligned powder	37 0		7 2	[41]
Er₂Fe₁₇C		36 6	260	6.8	
Er ₂ Fe ₁₇ C	free powder	36 6		(9 0)	[42]
Er ₂ Fe ₁₆ MnC	free powder	28 8		6 7	[22]
Er ₂ Fe ₁₅ Mn ₂ C	free powder	25 9		6 6	[22]
Er ₂ Fe ₁₄ Mn ₃ C	free powder	23 8		6 9	[22]
Er ₂ Fe ₁₃ Mn ₄ C	free powder	18 0		7 8	[22]
Tm₂Fe₁₇C		38 4	290	10.8	
Tm ₂ Fe ₁₇ C	free powder	38 4		10 8	[42]
Note The values in parentheses may be considered as less accurate results The data for the aligned samples are obtained by the fitting, see the references					
Er ₂ Fe ₁₇ C _x compounds (Z _{RFe} = 19)		<i>M_{Fe}</i>	<i>B_{exch}</i>	$-J_{RFe}/k$	Ref
Er ₂ Fe ₁₇ C _{0.5}	free powder	(36)			[43]
Er ₂ Fe ₁₇ C _{1.0}	free powder	(36)	220	7 3	[43]
Er ₂ Fe ₁₇ C _{1.5}	free powder	(36)	215	7 2	[43]
Er ₂ Fe ₁₇ C _{2.0}	free powder	(36)	210	7 0	[43]
Er ₂ Fe ₁₇ C _{2.5}	free powder	(36)	200	6 7	[43]
Er ₂ Fe ₁₇ C _{3.0}	free powder	(36)	195	6 6	[43]
Note The values in parentheses may be considered as less accurate results					
R ₂ Ni ₁₇ -based compounds (Z _{RT} = 19)		<i>M_{Ni}</i>	<i>B_{exch}</i>	$-J_{RNi}/k$	Ref
Gd₂Ni_{16.8}		5.6	35	7.0	
Gd _{0.8} Y _{1.2} Ni _{16.8}	free powder	4 9		6 9	[44]
Gd _{0.7} Y _{1.3} Ni _{16.8}	free powder	4 9		6 2	[44]
Gd _{0.6} Y _{1.4} Ni _{16.8}	free powder	5 0		5 9	[44]
Gd _{0.5} Y _{1.5} Ni _{16.8}	free powder	4 6		5 6	[44]

Table 1 (continued)

R ₂ Ni ₁₇ -based compounds (Z _{RT} = 19)		<i>M</i> _{Ni}	<i>B</i> _{exch}	− <i>J</i> _{RNi} / <i>k</i>	Ref
Tb₂Ni₁₇				5 1	
Tb _{0.60} Y _{1.40} Ni ₁₇	free powder	5 4		5 1	[44]
Dy₂Ni₁₇		5 3	20	4 5	
Dy ₂ Ni ₁₇	free powder	5 3	20	4 5	[45]
Dy _{0.55} Y _{1.45} Ni ₁₇	free powder	5 5		4 8	[44]
Ho₂Ni₁₇		5 3	20	4 1	
Ho ₂ Ni ₁₇	free powder	5 3	18	4 1	[45]
Ho _{0.55} Y _{1.45} Ni ₁₇	free powder	5 5		4 3	[44]
Er₂Ni₁₇		5 2	15	3 9	
Er ₂ Ni ₁₇	free powder	5 2	17	3 9	[45]
Er _{0.60} Y _{1.40} Ni ₁₇	free powder	5 4		4 1	[44]
Tm₂Ni₁₇				3 1	
Tm _{0.80} Y _{1.2} Ni ₁₇	free powder	5 6		3 1	[44]
'RFe ₁₂ '-type of compounds (Z _{RT} = 20)		<i>M</i> _T	<i>B</i> _{exch}	− <i>J</i> _{RT} / <i>k</i>	Ref
'HoFe₁₂'		14	140	7	
HoFe _{9.0} V _{3.0}	free powder	12 5		10 1	[46]
HoFe _{8.5} V _{3.5}	free powder	11 6		12 1	[46]
HoFe _{8.0} V _{4.0}	free powder	7 9		13 1	[46]
'ErFe₁₂'		15	130	6	
ErFe _{9.0} V _{3.0}	free powder	12 6		8 5	[46]
ErFe _{8.5} V _{3.5}	free powder	10 9		9 9	[46]
ErFe _{8.0} V _{4.0}	free powder	7 7		12 3	[46]
RFe₁₀Cr₂					
HoFe ₁₀ Cr ₂	free powder	16 0	(180)	(9)	[47]
ErFe ₁₀ Cr ₂	free powder	16 0	(170)	(9)	[47]
TmFe ₁₀ Cr ₂	free powder	14 4	165	9 0	[47]
RFe₁₀Mo₂					
DyFe ₁₀ Mo ₂	free powder	16 5	(226)	(9)	[48]
HoFe ₁₀ Mo ₂	free powder	16 3	179	7 4	[48]
ErFe ₁₀ Mo ₂	free powder	17 1	153	6 0	[48]
Note The compounds RFe ₁₂ do not exist The data in parentheses should be considered as less accurate					
'RCo ₁₂ '-type of compounds (Z _{RT} = 20)		<i>M</i> _T	<i>B</i> _{exch}	− <i>J</i> _{RT} / <i>k</i>	Ref
RCo₁₀V₂					
GdCo ₁₀ V ₂	free powder	7 7	100	10 5	[49]
TbCo ₁₀ V ₂	free powder	9 0	112	10 0	[49]
DyCo ₁₀ V ₂	free powder	8 9	97	8 8	[49]
HoCo ₁₀ V ₂	free powder	8 3	81	7 9	[49]
ErCo ₁₀ V ₂	free powder	8 3	82	8 0	[49]
RCo₁₁Ti					
DyCo ₁₁ Ti	free powder	15 7	208	9 8	[50]
HoCo ₁₁ Ti	free powder	15 4	193	9 3	[50]
ErCo ₁₁ Ti	free powder	15 3	164	7 9	[50]
Note The compounds RCo ₁₂ do not exist					
RCo ₁₂ B ₆ -based compounds (Z _{RCo} = 18)		<i>M</i> _{Co}	<i>B</i> _{exch}	− <i>J</i> _{RCo} / <i>k</i>	Ref
GdCo₁₂B₆		5 2	30	5 1	
GdCo ₁₂ B ₆	free powder	5 2		5 1	[51]
Gd _{0.8} Y _{0.2} Co ₁₂ B ₆	free powder	5 1			[51]
Gd _{0.2} Y _{0.8} Co ₁₂ B ₆	free powder	5 3			[51]
Gd _{0.1} Y _{0.9} Co ₁₂ B ₆	free powder	5 3			[51]

Table 1 (continued)

RCo ₁₂ B ₆ -based compounds ($Z_{\text{RCo}} = 18$)		M_{Co}	B_{exch}	$-J_{\text{RCo}}/k$	Ref
TbCo₁₂B₆		5 2	28	4 9	
TbCo ₁₂ B ₆	free powder	5 2		4 9	[51]
Tb _{0.6} Y _{0.4} Co ₁₂ B ₆	free powder	4 7			[51]
Tb _{0.4} Y _{0.6} Co ₁₂ B ₆	free powder	5 6		4 8	[51]
Tb _{0.2} Y _{0.8} Co ₁₂ B ₆	free powder	5 6			[51]
DyCo₁₂B₆		5 0	24	4 1	
DyCo ₁₂ B ₆	free powder	5 0		6 9	[51]
Dy _{0.9} Y _{0.1} Co ₁₂ B ₆	free powder	5 2		6 2	[51]
Dy _{0.8} Y _{0.2} Co ₁₂ B ₆	free powder	5 2		5 9	[51]
Dy _{0.7} Y _{0.3} Co ₁₂ B ₆	free powder	5 1		5 6	[51]
Dy _{0.55} Y _{0.45} Co ₁₂ B ₆	free powder	4 8		4 8	[51]
HoCo₁₂B₆		5 1	21	3 7	
HoCo ₁₂ B ₆	free powder	5 1		3 6	[51]
Ho _{0.55} Y _{0.45} Co ₁₂ B ₆	free powder	5 2		3 2	[51]
ErCo₁₂B₆		5 0	21	3 7	
ErCo ₁₂ B ₆	free powder	5 0		3 7	[51]
Er _{0.6} Y _{0.4} Co ₁₂ B ₆	free powder	5 0		3 5	[51]
TmCo₁₂B₆		5 2	20	3 5	
TmCo ₁₂ B ₆	free powder	5 2		3 5	[51]

3. Results obtained with the HFFP method and conclusions

Since 1988, more than 20 series of 3d–4f compounds have been investigated by the HFFP method to determine the magnetic coupling. The results are collected in Table 1.

For comparison, also several results obtained from high-field magnetization measurements on single crystals are included in Table 1. The difference between the high-field magnetic isotherms of single crystals measured in various crystallographic directions reflects mainly the magnetic anisotropy of a given compound [3]. Since the intersublattice exchange couples the strongly anisotropic R-sublattice magnetization to the weakly anisotropic T-sublattice magnetization, the intersublattice-coupling strength enters as a parameter in quantitative fits of the magnetic isotherms. It can be inferred from Table 1 that there is generally good agreement between single-crystal data and data obtained by means of the HFFP method.

In many of the binary and ternary compounds, the difference $|M_{\text{T}} - M_{\text{R}}|$ is fairly large so that the sloping range is inaccessible even with an available field strength as high as 40 T. For this

reason, it is usually necessary to apply magnetic dilution of the larger of the two sublattice magnetizations M_{R} or M_{T} . This is why many pseudobinary or pseudoternary compounds are listed in Table 1. A difficulty exists for compounds of the type $\text{RFe}_{12-x}\text{V}_x$ since here the V atoms were shown to have small magnetic moments coupled parallel to the R spin moment [14,15]. Fortunately, the V concentrations needed were not too large and the intersublattice-coupling constants for several of these series have also been determined from measurements of the Curie temperature [16]. Although this method is less reliable than the HFFP method, one may use the concentration dependence of J_{RT} found from these measurements as a guide to extrapolate the more limited number of J_{RT} values derived from high-field measurements to $x = 0$. These extrapolated values have been listed under 'RFe₁₂' in Table 1.

In Table 1, the data printed in bold are the values assessed for the compounds without the substitutions. The B_{exch} values are only given for these unsubstituted compounds, because this intrinsic magnetic property is meaningful only when describing the interaction between a magnetic ion and its magnetic neighbors.

The availability of so many results has made it

Fig 2 T-concentration dependence of the magnetic-coupling constants J_{ErT} of various Er-T compounds derived by means of the HFFP method

possible to find systematics in the magnetic-coupling behavior in the R-T compounds, which will be discussed below

3.1 Dependence of J_{RT} on the transition-metal concentration

In Table 1, we have listed the values of J_{RT} obtained for various series of compounds in the sequence of increasing transition-metal concen-

tration. Inspection of the data listed shows that there is a tendency of $|J_{\text{RT}}|$ to decrease with increasing T concentration. The data collection in Table 1 is most complete for R = Er. In order to demonstrate the dependence of J_{RT} on the T concentration, we have therefore selected the Er compounds for plotting J_{ErT} versus T concentration. Fig 2 shows this plot for T = Fe, Co and Ni. It is seen that the trend of $|J_{\text{ErT}}|$ to decrease with T concentration is very pronounced in the Fe compounds and somewhat less pronounced

Fig 3 The dependence of $-J_{\text{RT}}/k$ (K) on the atomic number of R components in the compounds of the type $\text{R}_2\text{T}_{14}\text{B}$ (a), R_2T_{17} type (b) (T = Fe, Co and Ni) and the four types of the Co compounds (c). In parts (a) and (b), the open circles and triangles represent single-crystal and free-powder samples of the Fe compounds, respectively. The dashed triangles represent free-powder samples of the Co compounds. In part (b), the full triangles represent free-powder samples of the Ni compounds. The dashed circles represent single-crystal samples of the Co compounds.

for $T = \text{Co}$ and Ni . The results shown in Fig. 2 closely resemble the results of Duc et al. [52] that for $T = \text{Fe}$ and to some extent also for $T = \text{Co}$ the intersublattice-coupling constant $|J_{\text{RT}}|$ decreases almost linearly with the value of the atomic transition-metal moment μ_{T} in the compounds. This agrees closely with the behavior of J_{RT} shown in Fig. 2, if one bears in mind that the transition-metal moment per T atom in $R-T$ compounds increases with increasing T concentration. As already proposed earlier by Duc [5], the origin of this behavior may be found in the variable degree of 3d–5d hybridization in these compounds. In fact, it is well known from band-structure calculations for Y-Fe compounds that the induced moment on the Y atoms increases with decreasing Fe concentration in spite of the decreasing Fe moments [53]. Indeed, further work by Brooks and Johansson [11] showed that increasing 3d–5d hybridization can be taken to lead to lower 3d moments but to higher $|J_{\text{RT}}|$ values. The tendency of $|J_{\text{RT}}|$ to decrease with increasing T concentration and the tendency of μ_{T} to increase with increasing T concentration can then satisfactorily be understood if one assumes that the 3d–5d hybridization decreases with increasing T concentration. We will return to this in Section 6.

3.2 Dependence of J_{RT} on the nature of the rare-earth component

For most of the series of $R-T$ compounds tabulated in Table 1, a decrease of $|J_{\text{RT}}|$ with increasing atomic number Z of the R ion can clearly be recognized. To illustrate this phenomenon, the R dependence of J_{RT} is shown in Fig. 3 for a number of series. For $\text{R}_2\text{Fe}_{14}\text{B}$ compounds, for example, the value of $|J_{\text{RT}}|$ decreases by about 20% when the R component changes from Gd to Tm . A similar trend is observed in other series.

As mentioned in the Introduction, an explanation for the observation that the intersublattice-magnetic-coupling strength becomes weaker with increasing Z of the R component has been given by Belorizky et al. [10]. Besides the lanthanide contraction, i.e. the reduction of the atomic ra-

dius of the R ions with increasing Z , an even stronger decrease of the radius of the 4f-shell occurs with increasing Z . This leads to a smaller overlap of the 4f and 5d shells. The 4f–5d exchange interaction is determined by the 4f–5d overlap and since the 4f–3d inter-atomic exchange interactions are mediated by the intra-atomic 4f–5d interaction, this larger spatial separation of 4f and 5d shells results in a reduction of the $R-T$ interaction.

However, there is a second mechanism which affects the $R-T$ interaction when passing through the lanthanide series. Band-structure calculations (see Section 5) show that the decreasing lattice constants alone would lead to an increase in the $R-T$ interaction due to enhanced 5d–3d hybridization. Therefore, depending on which of the two counteracting effects prevails, the $R-T$ interaction may decrease as well as increase with increasing atomic number of the R component. Inspection of the results presented in Table 1 suggests that the 4f–5d effect prevails in most of the series studied. The series RFe_3 may be an example where the 5d–3d effect prevails. Both effects nearly cancel in the RFe_2 , R_2Co_7 and RFe_4B series.

3.3 Dependence of J_{RT} on the nature of the 3d component

In order to see whether the intersublattice-coupling constants depend on the nature of the 3d component, we will use again the results presented in Fig. 2 for the Er compounds. Insofar as a comparison of compounds of similar composition is possible, one may infer from the data shown in Fig. 2 that there is a tendency of $|J_{\text{RT}}|$ to decrease in the sequence $T = \text{Fe}, \text{Co}, \text{Ni}$. Most likely, this behaviour is associated with the decreasing value of $(r_{3d}/r_{\text{atom}})^3$ which means that the 3d electrons become more localized in the sequence $\text{Fe}, \text{Co}, \text{Ni}$. As a consequence, one may expect that the 5d–3d hybridization decreases in the same sense, and so does $|J_{\text{RT}}|$.

3.4 Effect of interstitial atoms on J_{RT}

The effect of interstitial nitrogen on the intersublattice coupling has been investigated by

means of the HFFP method on the series of $\text{Ho}_2\text{Fe}_{17-x}\text{Mn}_x\text{N}_{2.7}$ [40] with the conclusion that there is no significant effect of the interstitial atoms on the coupling constant. Later experiments, however, have pointed out that there is a tendency of $|J_{\text{RT}}|$ to decrease with increasing content of the interstitial atoms in the series of $\text{Er}_2\text{Fe}_{17}\text{C}_x$ with $x = 0.5, 1.0, 1.5, 2.0, 2.5$ and 3.0 [43] (see Table 1). A similar effect was also observed in the compounds of $\text{Er}_2\text{Fe}_{17}\text{N}_{2.7}$ [54], $\text{Dy}_2\text{Fe}_{17-x}\text{Al}_x\text{Z}_y$ ($Z = \text{N}$ or H) [55] and the series $\text{R}_2\text{Co}_{17}\text{N}_x$ [39].

A possible explanation for this effect is that the expansion of the unit cells associated with the uptake of interstitial atoms increases the interatomic distance and hence reduces the 3d–5d hybridization. In addition, the effect of interstitial atoms on the d-band filling should also be taken into account [43].

4. Comparison with other methods

It has been mentioned already in Section 3 that there is good agreement between the J_{RT} values derived from magnetization measurements on single crystals and those obtained by the HFFP method.

Inelastic-neutron scattering (INS) was used by Loewenhaupt et al [1,2] to determine the molecular field acting on the Gd moments in $\text{Gd}_2\text{Fe}_{14}\text{B}$ and GdFe_2 . Their values for B_{exch} (denoted by B'_{exch}) are listed in Table 2 where they can be compared with the values of B_{exch} derived by means of the HFFP method using Eq. (6). Whereas the latter method samples only the R–T interaction, the molecular field derived from in-

elastic-neutron scattering represents the total exchange field experienced by the Gd moments and hence contains also the contribution due to the Gd sublattice ($B_{\text{exch}}^{\text{RR}}$). This may explain that the values derived from INS experiments are higher than those determined from HFFP experiments. Assuming that this is the only source of the difference, one may take advantage of the fact that both methods are parameter-free direct experimental determinations of the molecular field and use these values to determine the contributions due to the Gd sublattice via the relation $B'_{\text{exch}} = B_{\text{exch}} + B_{\text{exch}}^{\text{RR}}$. The values for $B_{\text{exch}}^{\text{RR}}$ obtained in this way have been included in Table 2.

An impression as to whether the numbers listed for $B_{\text{exch}}^{\text{RR}}$ are realistic ones may be obtained by means of the mean-field expression

$$B_{\text{mol}} = \frac{3kT_C}{g_R(J+1)\mu_B} \quad (7)$$

using the experimental T_C values of isotypic compounds in which the T component does not carry a magnetic moment.

Assuming that the Gd–Gd exchange in GdFe_2 is roughly the same as in GdNi_2 , one finds with $T_C = 85$ K for GdNi_2 the value $B_{\text{exch}}^{\text{RR}} = 42$ T. In $\text{Gd}_2\text{Fe}_{14-x}\text{Mn}_x\text{C}$, one finds that substitution of Mn for Fe leads to a disappearance of the 3d-sublattice magnetization for $x \geq 6$. Simultaneously, the Curie temperature has dropped to $T_C = 125$ K [56]. If this value is taken to reflect the Gd–Gd exchange interaction in $\text{Gd}_2\text{Fe}_{14}\text{C}$ (and $\text{Gd}_2\text{Fe}_{14}\text{B}$), one finds $B_{\text{exch}}^{\text{RR}} = 62$ T. These two values of $B_{\text{exch}}^{\text{RR}}$ are very rough estimates but they may serve to show that the values listed for $B_{\text{exch}}^{\text{RR}}$ in $\text{Gd}_2\text{Fe}_{14}\text{B}$ and GdFe_2 in Table 2 have the correct order of magnitude. This may justify the interpretation of the difference between the exchange fields derived from INS and HFFP as being due to the Gd-sublattice contribution.

The intersublattice-coupling constant can also be derived from rare-earth Mossbauer spectroscopy [4] by measuring the temperature dependence of the rare-earth hyperfine field $B_{\text{eff}}(T)$.

$$B_{\text{eff}}(T) = B_{\text{eff}}(0) \langle |J_z| \rangle / J \quad (8)$$

The expectation value of J_z in Eq. (8) is derived at each temperature from the thermal pop-

Table 2

Exchange fields B'_{exch} derived from the inelastic-neutron-scattering energy transfer Δ by Loewenhaupt et al [1,2]. The exchange fields B_{exch} are experimental values derived from HFFP measurements. The values listed for $B_{\text{exch}}^{\text{RR}}$ represent the difference between B'_{exch} and B_{exch} .

Compound	Δ (meV)	B'_{exch} (T)	B_{exch} (T)	$B_{\text{exch}}^{\text{RR}}$ (T)
$\text{Gd}_2\text{Fe}_{14}\text{B}$	37.5	324	270	54
GdFe_2	44.7	389	315	72

Table 3

Intersublattice-coupling parameters (J'_{DyT}) derived from ^{161}Dy Mossbauer spectroscopy by means of a mean-field approach using average transition-metal spin moments listed as μ_{T} . The coupling constants listed in the last column are those obtained by the HFFP method

Compound	J'_{DyT} (10^{-22} J)	μ_{T} (μ_{B})	$-J'_{\text{DyT}}/k$ (K)	$-J_{\text{DyT}}/k$ (K)
$\text{Dy}_2\text{Fe}_{17}$	−1.2	2.0	8.7	7.0
$\text{Dy}_2\text{Fe}_{14}\text{B}$	−1.8	2.2	13.0	9.7
$\text{Dy}_2\text{Fe}_{14}\text{C}$	−1.8	2.1	13.0	9.0
$\text{Dy}_2\text{Co}_{17}$	−1.4	1.6	10.1	7.3
DyCo_5	−2.1	1.5	15.2	7.1

ulation of the $2J+1$ energy levels determined after diagonalization of the 4f-Hamiltonian $H = H_{\text{cr}} - g_{\text{R}}\mu_{\text{B}}B_{\text{mol}}J$ (H_{cr} is the crystal-field Hamiltonian). Inspection of Eq. (4) shows that this procedure requires the knowledge of $M_{\text{T}}(T)$ as well as $M_{\text{R}}(T)$ and that n_{RT} and n_{RR} can then be derived from the fitting of $B_{\text{eff}}(T)$ by means of Eq. (8). To make this procedure more tractable, one may neglect the R–R contribution and approximate the temperature dependence of the T-sublattice magnetization by

$$M_{\text{T}}(T) = M_{\text{T}}(0)[1 - bT^2] \quad (9)$$

For Fe-based compounds it was shown by ^{57}Fe Mossbauer spectroscopy that the latter approximation does not introduce a large error [57]. Results obtained by Gubbens et al. [57–59] by means of ^{161}Dy Mossbauer spectroscopy are listed in Table 3 under J'_{DyT} together with the values of the zero-temperature 3d moments μ_{T} used to derive the molecular field B_{mol} mentioned above. With the exception of DyCo_5 , these μ_{T} values are very close to the values derived from the corresponding M_{T} values listed in Table 1. When expressed in the same units, the J'_{DyT} values derived from fitting of the temperature dependence of the ^{161}Dy hyperfine fields may be compared with the J_{DyT} values obtained by means of the HFFP method. As may be seen from the last two columns in Table 3, satisfactory agreement is found only for $\text{Dy}_2\text{Fe}_{17}$, whereas J'_{DyT} is substantially larger than J_{DyT} for the other compounds. One reason for this discrepancy may be the neglect of the Dy–Dy interaction in the diagonaliza-

tion procedure, meaning that the Dy–Fe interaction strength is overestimated. Another source for the discrepancy between the two sets of coupling parameters listed in Table 3 may be the neglect of crystal-field interaction (H_{cr}) relative to the exchange interaction in the Hamiltonian used for the diagonalization and subsequent fitting of $B_{\text{eff}}(T)$ [4]. In a more recent investigation on other similar compounds, the diagonalization procedure was performed with inclusion of the crystal-field splitting [60]. This led to lower values of J_{RFe} , removing the discrepancy between the HFFP results and the results derived from rare-earth Mossbauer spectroscopy.

Another approach to determine the intersublattice-coupling strength from experimental data uses a mean-field fitting of the temperature dependence of the total magnetization by means of Brillouin functions via the following equations

$$M_{\text{R}}(T) = M_{\text{R}}(0)B_J(x_{\text{R}}) \quad (10)$$

with $x_{\text{R}} = \mu_{\text{R}}(n_{\text{RT}}M_{\text{T}})/kT$, and

$$M_{\text{T}}(T) = M_{\text{T}}(0)B_J(x_{\text{T}}) \quad (11)$$

with $x_{\text{T}} = \mu_{\text{T}}(n_{\text{RT}}M_{\text{R}} + n_{\text{TT}}M_{\text{T}})/kT$, where μ_{R} and μ_{T} are the magnetic moments of the R and T atoms at 0 K, respectively. The quantities n_{RT} and n_{TT} represent the molecular-field coefficients corresponding to the R–T and T–T interactions, respectively. Values of n_{TT} can be determined from the ordering temperatures of the isostructural compounds with non-magnetic rare earths (La, Y and Lu).

Compounds showing a compensation temperature in the temperature dependence of their magnetization are particularly suitable for such fitting procedures and there is generally good agreement with the data derived from the HFFP method [38]. This may be illustrated by means of the results shown in Fig. 4.

Finally, we wish to compare the results of the HFFP method with the results of the method based on magnetic-ordering temperatures. In the high-temperature limit of the mean-field approach, one may derive J_{RT} by comparing the Curie temperature (T_{C}) of a given compound with that of the corresponding compound in which R is non-magnetic (T'_{C}) and with that of a further

Fig. 4 Magnetic coupling constants J_{RN1} of R_2Ni_{17} compounds. The triangles and the squares represent the data derived from the HFFP method and compensation point analysis, respectively.

compound in which T is non-magnetic (T_C'') by using the following expression

$$(J_{RT}/k)^2 = \frac{9(T_C - T_C')(T_C - T_C'')}{4Z_{RT}Z_{TR}S_T(S_T + 1)(g_R - 1)^2J(J + 1)} \quad (12)$$

where the number of nearest neighbors Z_{RT} and Z_{TR} of the R and T atoms have to obey the relation [13]

$$N_R Z_{RT} = N_T Z_{TR} \quad (13)$$

Since isotopic compounds in which the T moment is zero are usually not available and since T_C'' can be expected to be much smaller than T_C , one frequently neglects the R–R interaction by setting T_C'' equal to zero. It will become clear from inspection of Eq. (12) that this method leads to accurate results when the experimental error for the comparatively small differences between T_C and T_C' can be kept low. Since the Gd compounds have the highest Curie temperatures, the magnetic coupling can most accurately be determined for these compounds. Duc et al. [52], using experimental values for T_C and T_C' and making reasonable estimates for T_C'' , have derived the

intersublattice-coupling constants for quite a number of Gd–T compounds and have compared these values with the corresponding values obtained by the HFFP method. With the exception of RFe_2 and RFe_3 , there is a good agreement between both sets of data.

5. Results of band-structure calculations

Band-structure calculations have been performed by Brooks et al. [11] and Liebs et al. [12]. Following Campbell [9], Brooks et al. argue that the key role in the R–T coupling is played by the 5d electrons of the R component. The spin-up and spin-down rare-earth 5d bands hybridize with the transition-metal 3d bands but do so to a different degree, depending on the degree of exchange splitting between the spin-up and spin-down 3d bands. The 5d(R)–3d(T) hybridization leads to a larger occupation of the 5d spin-down band than the spin-up band, and the 5d(R) moment is therefore antiparallel to the 3d(T) moment. The antiferromagnetic coupling between the 3d and 4f spins then follows from the ferromagnetic intra-atomic exchange interaction between the 4f-spin moment and the 5d-spin density. Brooks et al. [11] calculate the effective exchange field in a perturbative way: they calculate the rare-earth-valence-electron and 4f-spin density for the ground state, and derive the interaction energy under the assumption that upon reversal of the 4f-spin moment the valence-electron-spin density remains the same. The exchange field, B_{exch} , follows from the energy difference ΔE (per R atom) between the ground state and the reversed-spin state by means of the relation $B_{\text{exch}} = \Delta E / 4\mu_B S_R$. In the case of Gd, one has, for instance, $S_R = 7/2$, and $B_{\text{exch}}(T) = 1233 \times \Delta E$ (eV). In Table 4, the result of this calculation for $GdFe_2$ is compared with experimental HFFP data. The calculated value of the exchange field is more than 50% larger than the experimental value. The intersublattice-coupling constants were calculated by Brooks et al. also for the remainder of the heavy RFe_2 compounds and the trend predicted roughly follows the experimental data listed for RFe_2 in Table 1.

Table 4

Comparison of exchange fields B_{exch} obtained by electronic-band-structure calculations in Refs. [11,12], and as presented in this study, with experimental values derived from HFFP experiments

Compound	B_{exch} (T)	
	theory	HFFP experiments
GdFe ₂	519 ^[11] , 274 ^a	315
GdCo ₅	237 ^a	240
Gd ₂ Fe ₁₄ B	323 ^[12]	270
Tb ₂ Fe ₁₄ B	289 ^[12]	275
Dy ₂ Fe ₁₄ B	254 ^[12]	250
Gd ₂ Fe ₁₇	302 ^a	305
Ho ₂ Fe ₁₇	247 ^[12]	220

Note: The superscript ^a indicates that the results are from the present work

Liebs et al. [12] use a different approach, avoiding any specific model regarding the 4f–3d exchange mechanism. By performing selfconsistent *ab initio* Linearized Muffin Tin Orbitals (LMTO) band-structure calculations they calculate the change in total energy E , achieved by rotating the 4f moments 180° out of the ground state. Results of their calculations for $R_2\text{Fe}_{14}\text{B}$ ($R = \text{Gd}, \text{Tb}$ and Dy) and $\text{Ho}_2\text{Fe}_{17}$ are compared in Table 4 with results from the HFFP method. For all cases, theory and experiment agree within 20%.

The band-structure calculations by Liebs et al. [12] reveal in more detail what happens to the electronic structure and magnetism upon the reversal of all 4f spins. They find for $\text{Gd}_2\text{Fe}_{14}\text{B}$ that the Fe moments are only weakly affected, being at most 4% smaller in the reversed state. In contrast, the valence-electron contributions (6s, 6p and 5d orbitals) to the total moment of the R sites were found to decrease by a factor 2–3 upon the reversal of the 4f moment. This implies that the 4f–3d exchange interaction cannot be treated in a perturbative way, as done by Brooks et al. [11], because the valence-electron-spin density does not remain constant. Liebs et al. [12] agree that the rare-earth valence electrons (mainly 5d) mediate the 4f–3d exchange interaction, but emphasize that calculations of the interaction constant should be selfconsistent. Non-selfconsistent calculations, based on the change of the ex-

change-correlation energy only, yield a much too large value for the coupling parameter. This may qualitatively explain the discrepancy for GdFe_2 between calculations of Brooks et al. and the results of the HFFP experiment.

We have investigated to what extent the results of Liebs et al. [12] have a more general validity, by performing similar selfconsistent band-structure calculations of ΔE for GdFe_2 , GdCo_5 and $\text{Gd}_2\text{Fe}_{17}$. The calculations were scalar relativistic, and based on the local-spin-density approximation, using the form of the exchange-correlation potential as proposed by von Barth and Hedin [61] and with the parameters as given by Janak [62]. We used the augmented spherical wave (ASW) method, which employs the atomic spheres approximation. Wigner–Seitz sphere radii were chosen according to the ratio $r_{\text{Gd}}/r_{\text{Fe}}/r_{\text{Co}} = 1.35/1.00/1.00$, resulting in $r_{\text{Gd}} \approx 1.9\text{Å}$, $r_{\text{Fe}} \approx 1.4\text{Å}$ and $r_{\text{Co}} \approx 1.4\text{Å}$. The calculations were performed for the experimental lattice parameters, and for $\text{Gd}_2\text{Fe}_{17}$ in the rhombohedral crystal structure. The total energy difference was obtained from calculations with 280, 216 and 10 k-points in the irreducible parts of the Brillouin zones of GdFe_2 , GdCo_5 and $\text{Gd}_2\text{Fe}_{17}$, respectively. The 4f states were treated as band states. In earlier investigations, this procedure was found for a large number of compounds to lead to excellent theoretical predictions of 4f-crystal-field parameters, electric-field gradients at rare-earth nuclei [63], and hyperfine fields at Gd nuclei [64]. The majority-spin 4f-electron states are completely occupied, leading to a (localized) moment of $7.0\mu_B$. There is a very small population of minority-spin 4f electrons. These states are delocalized, and contribute to the valence-electron-spin density. In Table 5, which gives a decomposition of the total magnetization into local contributions, these Gd 4f contributions are indicated as 4f(maj) and 4f(min), respectively. It follows from Table 5 that for these compounds too, the rare-earth-valence-electron spin density alters significantly upon reversal of the 4f moments, and that also the Fe or Co moments are slightly affected. The calculated total energy differences ΔE are 0.221 eV, 0.192 eV and 0.245 eV (per Gd atom) for GdFe_2 , GdCo_5 and $\text{Gd}_2\text{Fe}_{17}$, respectively. The corre-

Table 5

Local spin magnetic moments for Gd, Fe and Co sites in GdFe_2 , GdCo_5 and $\text{Gd}_2\text{Fe}_{17}$, given in μ_B/atom . The Gd moments are further subdivided into contributions from s, p, d and f states

Compound	Site		In ground state moment	In state with reversed 4f spin
GdFe_2	Gd	4f (maj)	−7.00	7.00
		5s	−0.09	−0.02
		6p	−0.15	−0.02
		5d	−0.54	−0.11
		4f (min)	0.14	−0.17
	Fe	(total)	1.86	1.84
GdCo_5	Gd	4f (maj)	−7.00	7.00
		6s	−0.04	0.00
		6p	−0.08	−0.03
		5d	−0.40	−0.07
		4f (min)	0.16	−0.27
	Co	(2(a))	1.48	1.50
	Co	(3(g))	1.44	1.42
$\text{Gd}_2\text{Fe}_{17}$	Gd	4f (maj)	−7.00	7.00
		6s	−0.05	−0.02
		6p	−0.10	−0.04
		5d	−0.37	−0.08
		4f (min)	0.19	−0.18
	Fe	(6(c))	2.34	2.33
	Fe	(9(d))	2.02	1.97
	Fe	(18(f))	2.27	2.30
	Fe	(18(h))	2.06	2.04

sponding values for the exchange field are given in Table 4. They agree very well with the experimental results. For GdFe_2 , theory and experiment agree within 15%, which is much better than for the non-selfconsistent calculations by Brooks et al. [11]. These results further stress the importance of performing selfconsistent calculations.

In addition, we have calculated ΔE for hypothetical GdFe_2 with the lattice parameters of ErFe_2 . This corresponds to a reduction of the cubic lattice parameter by 1.5%. The calculated total-energy difference is 0.242 eV per formula unit, almost 10% larger than for the uncompressed lattice. This result shows that for a given series of compounds there is an increase in the exchange field $B_{\text{exch}}^{\text{RT}}$ associated with the decrease in lattice constant when passing through the lanthanide series. In terms of Eq. (6), with M_T

remaining constant or decreasing slightly, this means that the intersublattice-coupling strength increases when passing through the lanthanide series. This effect reflects the increasing 5d–3d hybridization and is opposite to the effect of the intra-atomic 4f–5d interaction discussed in Section 3.2. The latter interaction decreases when passing through the lanthanide series and reduces the concomitant R–T coupling strength.

6. Discussion

We have shown in the previous sections that the results obtained by the HFFP method are in good agreement with results of other experimental techniques provided that the effect of the R–R exchange coupling is accounted for in a reasonable way in the latter techniques. Here, we recall that the HFFP method is free of any assumptions regarding the R–R coupling and the size of the participating moments.

It is gratifying that there is satisfactory agreement between the experimental and calculated intersublattice-coupling constants. However, with the limited number of band-structure calculations available at present it is not yet possible to describe the variation of the intersublattice-coupling constants across the various series of compounds.

Experimentally, one observes that the intersublattice coupling constant tends to decrease when going from low to high 3d-atom concentrations. Duc [5] has made attempts to find an empirical relationship between J_{RT} and the corresponding value of the 3d moment. However, a linear relationship between both quantities was found to hold only for a limited number of compounds and the assumptions made as to the size of the 3d moment are not always very clear (for instance the Co moment in $\text{Gd}_2\text{Co}_{14}\text{B}$ was taken close to $1.7\mu_B/\text{Co}$ whereas the experimental value is only about $1.5\mu_B/\text{Co}$). In Fig. 5, we have employed the experimental data given in Section 3 to investigate the mentioned relationship in a more extended way by including the data presently available for more extreme cases (see Table 6). Since data for the Er compounds are the most

Fig. 5 Experimental values of the intersublattice-coupling constant obtained from HFFP measurements on Er-3d compounds plotted versus the corresponding 3d moments

Fig. 6 Experimental values of the intersublattice-coupling constant obtained from HFFP measurements on Er-3d compounds plotted versus the corresponding reciprocal values of the normalized volume per formula unit

Table 6

Experimental data used in the plots of J_{ErT} vs μ_{3d} and J_{ErT} vs V^{-1} shown in Figs 5 and 6, respectively

Compound	$-J_{\text{ErT}}/k$ (K)	μ_{3d} (μ_B /atom)	Lattice constants		V^{-1} (nm^{-3})
			a (pm)	c (pm)	
ErFe ₂	19	1.6	728.3	—	20.8
ErFe ₃	17	1.7	508.6	2446	16.6
Er ₆ Fe ₂₃	12	1.9	1200.4	—	13.8
ErFe ₄ B	11	1.87	503.2	698.6	13.1
Er ₂ Fe ₁₄ B	8.8	2.2	873.4	1194.2	8.8
Er ₂ Fe ₁₄ C	8.6	1.95	873.0	1177.5	8.9
ErFe ₁₂	6	2.1	845	474	5.8
Er ₂ Fe ₁₇	7.5	2.1	841.6	829.1	7.8
Er ₂ Fe ₁₇ C ₁₀	7.3	1.8	853.8	833.1	7.6
Er ₂ Fe ₁₇ C ₁₅	7.2	1.7	858.8	1238.5	7.5
Er ₂ Fe ₁₇ C ₂₀	7.0	1.8	864.3	1234.1	7.4
Er ₂ Fe ₁₇ C ₂₅	6.7	1.8	867.9	1238.4	7.3
Er ₂ Fe ₁₇ C ₃₀	6.6	1.8	862.3	848.1	7.2
ErCo ₃	13	1.1	479.2	2418	17.4
Er ₂ Co ₇	12	1.5	496.0	3607	15.6
ErCo ₄ B	7.5	0.95	496.8	685.8	13.7
ErCo ₅ B	8.0	1.77	487.0	400.2	12.2
Er ₂ Co ₁₄ B	9	1.42	860	1160	9.2
Er ₂ Co ₁₇	6.9	1.72	831.0	811.3	8.3
ErCo ₁₂ B ₆	3.7	0.43	943.8	743.1	5.3
Er ₂ Ni ₇	9	0.55	490.9	3607	15.9
Er ₂ Ni ₁₇	3.9	0.31	828.7	801.7	8.4

abundant, we have restricted this plot to Er compounds. It may be seen that there is a considerable scatter of the data, precluding any correlation to be made between the two experimental quantities considered.

An interesting correlation is found, though, by plotting J_{RT} versus the reciprocal of the volume adopted by the various types of compounds. In view of the different types of unit cells associated with the corresponding crystal structure, we have rewritten the formula composition as RT_nM_m (T = transition-metal component, M = non-magnetic component) and used the lattice constants to calculate the volume (V) occupied by one formula unit RT_nM_m . It may be seen from Fig. 6 that there is a fairly strong correlation between J_{RT} and V^{-1} , $|J_{\text{RT}}|$ increasing strongly with increasing V^{-1} . In view of the important role attributed by Brooks and Johansson [11] to the 5d-3d hybridization, the strong volume dependence shown in Fig. 6 may reflect the strong volume dependence of the 5d-3d hybridization. The 5d-3d hybridization will increase with decreasing volume per unit RT_nM_m , meaning that J_{RT} , unlike μ_{T} , will increase in the same sense. It

is clear that the average 3d–3d separation does not increase when going from left to right in Fig 6, suggesting that the role of the 3d electrons alone is less important in the empirical relationship between J_{RT} and V^{-1} shown in this figure

The strong dependence of J_{RT} on the volume per unit RT_nM_m has also been confirmed by the experimental results on the interstitial compounds [43], as discussed in Section 3.4. Though the volume increase upon introduction of the interstitial atoms is small (less than 7%), the correlation can be clearly observed. This lends further credence to the fact that the correlation between J_{RT} and the reciprocal volume is not simply a reflection of the dependence of J_{RT} on the 3d concentration.

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