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Magnetic and electrical properties of the $\text{Ho}(\text{Co}, \text{Si})_2$ compounds

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Abstract. The magnetization, a.c. susceptibility, resistivity and magnetoresistance have been measured for the invariable-unit-cell-parameter $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds. The Si substitution for Co in HoCo_2 yields a dramatic initial increase of T_C , whereas the 3d magnetic moment is reduced. These findings may be attributed to the strong involvement of Si p states in the hierarchy of exchange interactions. The magnetic phase transition at T_C is of first order for the compounds with $x < 0.075$. For these compounds, the coexistence of metamagnetism and the quenching of spin fluctuations at $T > T_C$ are provided.

1. Introduction

The RCO_2 (R being Y and rare earths) compounds have been intensively studied as a typical example to investigate itinerant electron metamagnetism (Duc *et al* 1992a, Goto *et al* 1994), spin fluctuations (Pillmayr *et al* 1990, Goto *et al* 1994, Duc *et al* 1995) and magnetovolume effects (Armitage *et al* 1990, Duc *et al* 1992a, 1993c). In these compounds, the Co subsystem itself is in the exchange-enhanced paramagnetic state due to the location of the Fermi level at the (low) minimum position of the density of states. Co magnetism may be formed through a metamagnetic transition (MT) with an external magnetic field of the megagauss range in $\text{Lu}(\text{Y})\text{Co}_2$ (Goto *et al* 1990) or by employing the large molecular field due to ferromagnetically ordered 4f moments in the magnetic rare-earth– Co_2 compounds (Duc *et al* 1992a and references therein). As a consequence, the magnetic phase transition of first-order type at the Curie temperature (T_C) was observed for the compounds with $\text{R} = \text{Dy}, \text{Ho}, \text{Er}$ and Tm . The field induced MT can occur in a limited temperature range above T_C (Steiner *et al* 1978).

Co-magnetism can also be induced in RCO_2 if one can increase the density of state at the Fermi level $N(E_F)$. The nature of increasing $N(E_F)$ may be attributed to the volume effects or the effects of the decrease of the d-electron concentration (Cyrot and Lavagna 1979). Al substitutions are especially effective in producing these effects. Indeed, the Al substitution leads to the enhancement of the Co susceptibility, to the reduction of the critical magnetic field (B_c) of the MT and, finally, to the formation of magnetic ordering in the $\text{Y}(\text{Lu})(\text{Co}_{1-x}\text{Al}_x)_2$ compounds. Moreover, a giant increase of T_C was observed in $\text{R}(\text{Co}_{1-x}\text{Al}_x)_2$ with $\text{R} = \text{Dy}, \text{Ho}, \text{Er}$ and Tm (Duc *et al* 1992a).

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Special compositions were invented in order to fix one of the values of these variables and to study systematically the involvement of the other parameters. Such studies were performed with the invariable-crystal-unit-cell-parameter $(Y_{1-t}Lu_t)(Co_{1-x}Al_x)_2$ compounds studied at ambient (Dubenko *et al* 1991) and high pressure (Armitage *et al* 1990, Duc *et al* 1993c). In this context, the $R(Co_{1-x}Si_x)_2$ compounds seem to be particularly suitable for such studies because they allow a nearly constant lattice parameter in a wide concentration range. Like Al-adopted materials, a strong enhancement of the magnetic susceptibility and a drastic reduction of critical fields for metamagnetism with increasing x up to 0.1 were observed also in $Y(Lu)(Co_{1-x}Si_x)_2$ systems at low temperatures (Michels *et al* 1990, Murata *et al* 1993). However, no ferromagnetic ground state can be stabilized. The magnetic $R(Co_{1-x}Si_x)_2$ compounds with $R = Gd, Tb, Dy$ and Er have been studied so far (Duc 1996, Duc *et al* 1997). In these compounds, a strong enhancement of the ordering temperature was observed for the $Er(Co_{1-x}Si_x)_2$ compounds. However, this increase is only about half of that observed in the $Er(Co_{1-x}Al_x)_2$ system. The slight modification of the magnetic ordering behaviours observed in the $R(Co, Al)_2$ and $R(Co, Si)_2$ systems have been associated with the difference of the degree of the 3d-p hybridization due to the Si atom having one more 2p electron than Al.

In attempt to complete studies on the heavy-rare-earth-(Co, Si)₂ compounds, we investigate in this work magnetization, a.c. susceptibility and electrical resistivity for the $Ho(Co_{1-x}Si_x)_2$ compounds. Emphasis is put on metamagnetism and quenching of spin fluctuation scattering in the vicinity of T_C . The effects of the 2p(Si)-3d(Co) hybridization on the 4f-3d exchange interactions are also discussed.

2. Experimental details

The polycrystalline samples of the $Ho(Co, Si)_2$ compounds were prepared by arc melting a stoichiometric mixture of rare earths (4N8), Co (5N) and Si (6N) under argon atmosphere. The melted buttons were wrapped in tantalum foil, sealed under argon in silica tubes and annealed at 950 °C for 60 h. The x-ray analysis revealed only the expected C15 phase with an almost invariable lattice parameter (of 7.171 ± 0.005 Å).

Magnetization was measured using the induction method. A.c. susceptibility measurements were carried out at frequency of 318 Hz using a phase sensitive detector. The electrical resistivity data and magnetoresistance were obtained by means of a four-terminal measuring technique on bar-shaped samples (size about $1 \times 1 \times 7$ mm³).

3. Experimental results and analysis

3.1. Magnetization

The temperature dependence of the magnetization in an applied magnetic field of 0.2 T is presented in figure 1 for the $Ho(Co_{1-x}Si_x)_2$ compounds. The Curie temperatures were determined from a.c. susceptibility data (see the next subsection) and are listed in table 1. Note that the ordering temperature initially increases from $T_C(0) = 78$ K for $x = 0$ and reaches the maximal value of $T_C^{max} = 115$ K at a sample with $x = 0.075$, then slightly decreases and finally shows a tendency to saturate to the value of 112 K at $x \geq 0.1$. For this system, the ratio of $T_C^{max}/T_C(0)$ is about 1.5. This enhancement of T_C is rather large compared to that of the $R(Co_{1-x}Si_x)_2$ compounds with $R = Dy, Tb$ and Gd , but is smaller compared to the $Er(Co_{1-x}Si_x)_2$ compounds (see figure 2) and the $Ho(Co_{1-x}Al_x)_2$ compounds; we will come back to this problem in section 4. For the

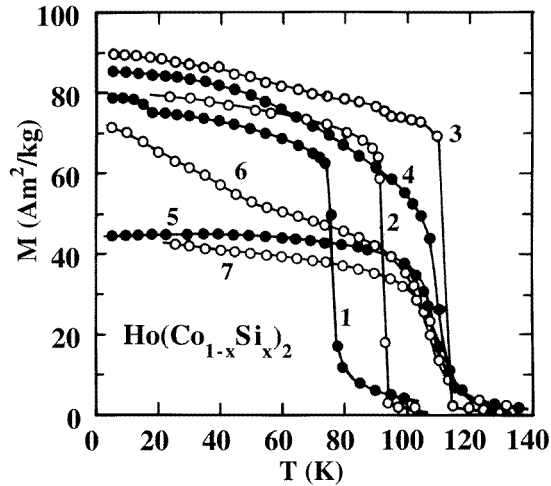


Figure 1. The temperature dependence of the magnetization for the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds: 1, $x = 0.0$; 2, $x = 0.025$; 3, $x = 0.05$; 4, $x = 0.075$; 5, $x = 0.10$; 6, $x = 0.15$; 7, $x = 0.2$.

Table 1. The values of T_C , spontaneous magnetization (M_s) and 3d magnetic moment (M_{3d}) in the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds.

x	T_C (K)	M_s ($\mu_B \text{ fu}^{-1}$)	M_{3d} (μ_B/atom)
0.0	78	7.80	1.00
0.025	98	7.80	1.03
0.05	112	7.80	1.06
0.075	115	7.85	1.05
0.10	112	8.20	0.88
0.15	112	8.40	0.82
0.20	112	8.75	0.65

compounds investigated, the first-order transition (FOT) is characterized by a sharp change of the magnetization at T_C . Upon substitution of Co by Si the sharp transition exists up to $x \leq 0.05$. For the remaining samples a gradual change of the magnetization is observed around T_C . This difference in the behaviour is ascribed to the change-over from a first-order transition to a second-order one.

At 4.2 K, the magnetization is almost saturated in the applied magnetic field of 10 T for all the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds. The magnetization curves are illustrated in figure 3 for three compounds. The saturation magnetizations (M_s) are listed in table 1. Note that M_s is almost constant for $x \leq 0.075$, then starts to increase at $x > 0.075$. Taking the value of $9.8 \mu_B/\text{atom}$ as reported for the Ho magnetic moment in HoCo_2 (Voiron 1973) and assuming that this value is independent of Si substitution, we then obtain the values of the magnetic moment of the 3d subsystem as listed in table 1. For comparison, we present in figure 4 the 3d magnetic moment for the $\text{R}(\text{Co}_{1-x}\text{Si}_x)_2$ ($\text{R} = \text{Dy}, \text{Ho}, \text{Lu}$) and $\text{Ho}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds. It is seen that the variation of the 3d magnetic moments with respect to x is rather similar. The larger 3d moment observed in $\text{R}(\text{Co}, \text{Si})_2$ than that in $\text{Lu}(\text{Co}, \text{Si})_2$ may be accounted for by the influence of the R–Co exchange interactions (Duc *et al* 1992b).

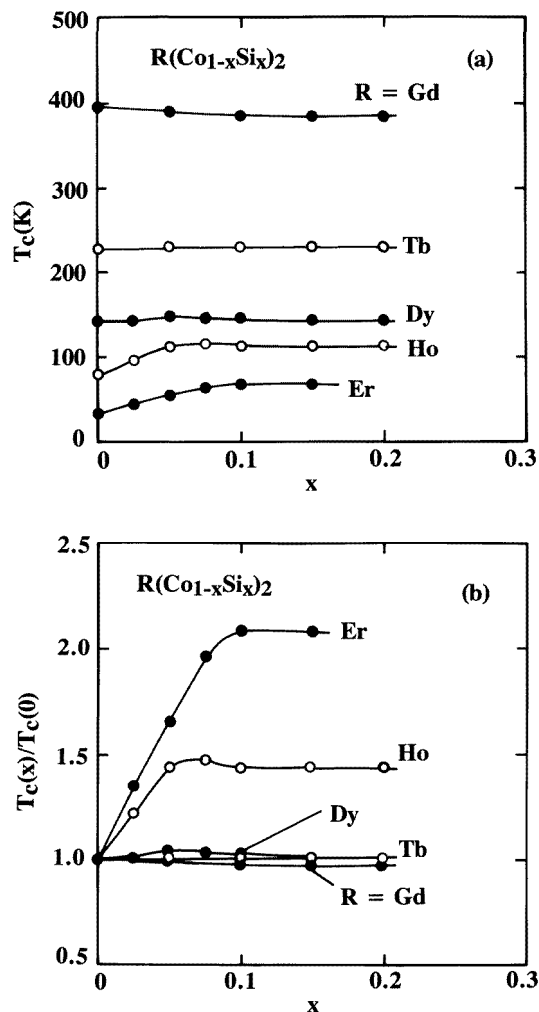


Figure 2. The Si-concentration dependence of (a) T_C and (b) $T_C(x)/T_C(0)$ for the $R(\text{Co}_{1-x}\text{Si}_x)_2$ compounds.

Figure 5 presents the magnetization isotherms for the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ ($x = 0, 0.025, 0.050$ and 0.075) compounds at temperatures above T_C . The results are very similar to each other for the first three compounds and are in good agreement with those reported previously for HoCo_2 (Steiner *et al* 1978) concerning (i) the metamagnetic behaviour in a limited temperature range above T_C and (ii) the increase of B_c and (iii) the decrease of the magnetization step (ΔM) at the MT with increasing temperature. This metamagnetic behaviour is more clearly demonstrated in the Arrott plots (see figure 6). In this figure, the experimental points show the S-shaped curves with two linear parts corresponding to the paramagnetic state (with $M_s = 0$) and the ferromagnetic state (with $M_s \neq 0$) at low and high magnetic fields, respectively. This implies that the c_3 -coefficient in the description of the equation of state,

$$B/M = c_1 + c_3 M^2 + c_5 M^4 + \dots \quad (1)$$

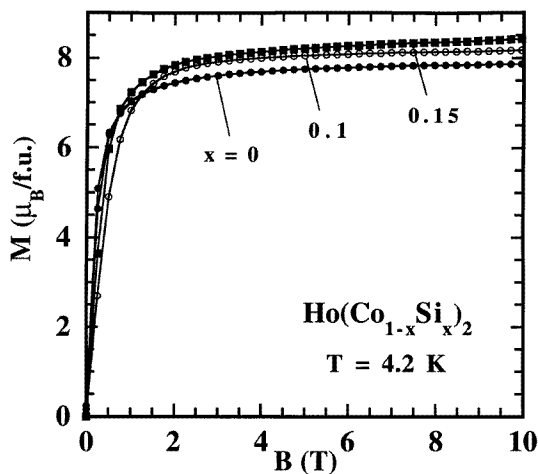


Figure 3. The magnetization curves at 4.2 K for three $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds.

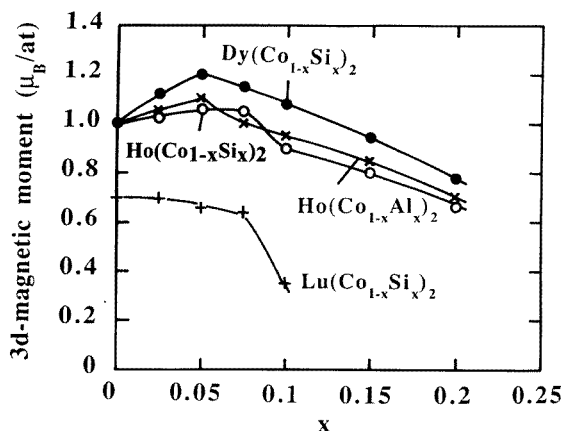


Figure 4. The Si-concentration dependence of 3d magnetic moments in the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds. The corresponding data in $\text{Dy}(\text{Co}_{1-x}\text{Si}_x)_2$, $\text{Lu}(\text{Co}_{1-x}\text{Si}_x)_2$ and $\text{Ho}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds are also included.

is negative (see also section 4). The value of the critical field for the MT can be determined from the inflection points of these curves. The results show an almost linear decrease of B_c with increasing temperature. For the compound with $x = 0.075$, where the second-order transition (SOT) at T_c occurs, the Arrott plots are rather monotonic.

3.2. A.c. susceptibility

In figure 7, a.c. susceptibility data of the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds are presented as a plot of the reduced χ/χ_{max} against T , where χ_{max} is the highest maximum value of χ . For the samples with $x = 0$ and $x = 0.05$, for which compounds the para–ferrimagnetic transition is of first order, $\chi_{ac}(T)$ curves are characterized by a double peak χ_k (on the low-temperature

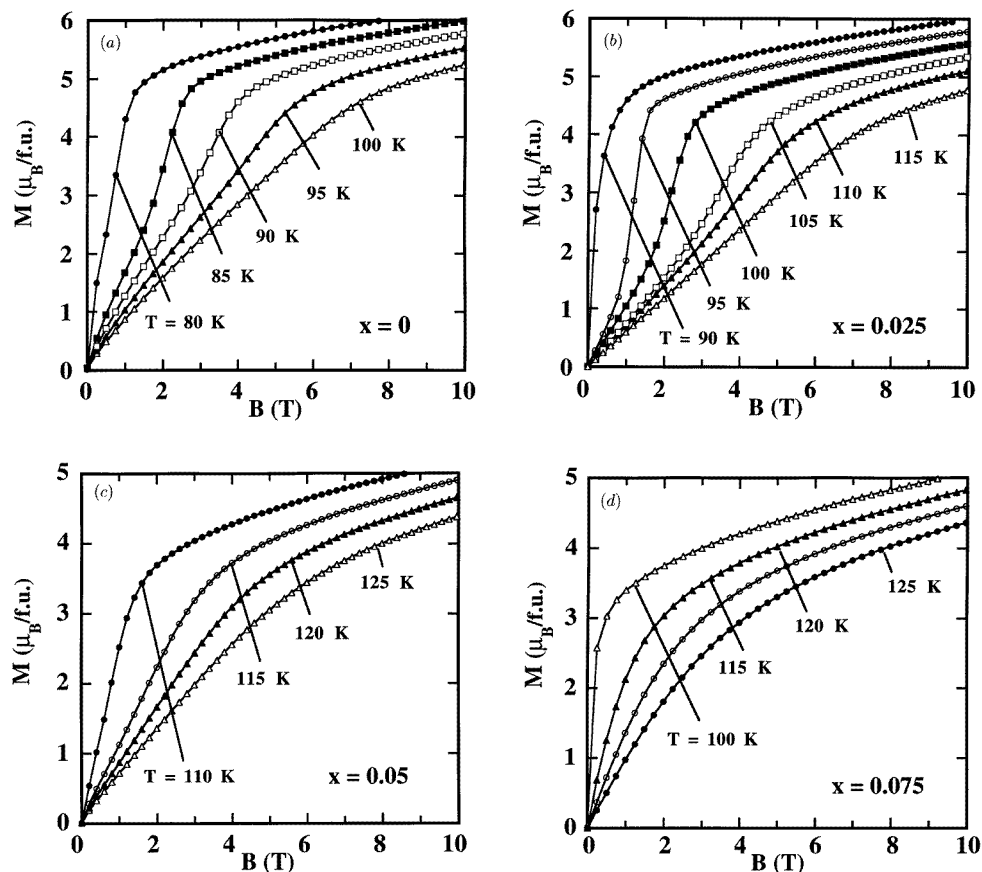


Figure 5. Magnetization isotherms for the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds at temperatures above T_C .

side) and χ_m (on the high-temperature side) in the vicinity of the magnetic phase transition. We note that χ_m appear at the temperature at which the discontinuity in magnetization occurs. In table 1, this temperature is given as T_C . With increasing Si concentration the high-temperature peak (i.e. χ_m) becomes an inflection point. The temperature of this point is a few degrees higher than the temperature at which χ_k appears. Thus in the compounds having the SOT $\chi_{ac}(T)$ shows a single peak as usual. Duc *et al* (1988a,b) have found that χ_k becomes broader and shifts to lower temperature in applied magnetic fields. It originates from the details of the magnetization process and is named the Hopkinson peak. χ_m , however, shifts to higher temperature in applied field. This is the critical peak which is associated with the para-process.

3.3. Resistivity and magnetoresistance

The temperature dependence of the electrical resistivity of the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds is presented in figure 8. For all samples, the electrical resistivity tends to saturate above T_C . In the compounds with $x \leq 0.05$, the FOT shows up in the almost discontinuous change in the resistivity at T_C . The increase of the resistivity with decreasing temperature

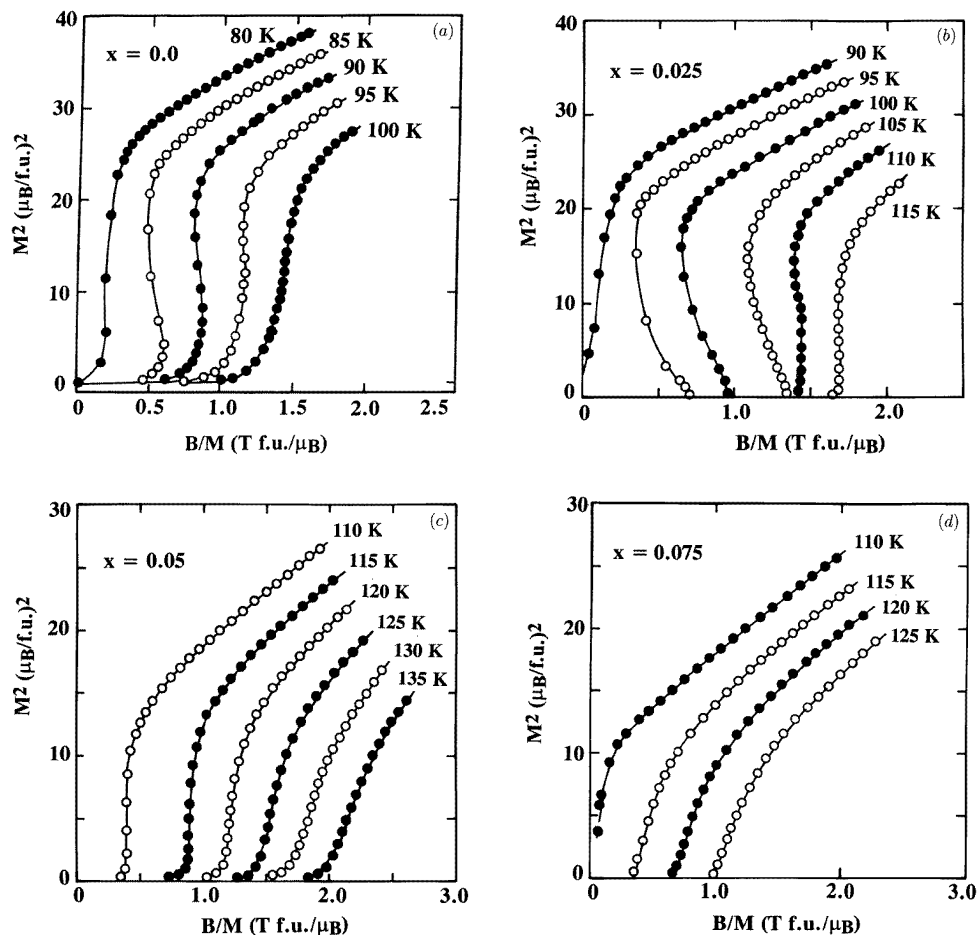


Figure 6. The Arrott plots for the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds at $T > T_C$.

in the magnetic phase (below T_C) starts to appear at $x = 0.075$. For the compounds with $x \geq 0.075$, a resistivity minimum, thus, occurs at T_C . A similar resistivity behaviour was reported for the $\text{Dy}(\text{Co}_{1-x}\text{Si}_x)_2$, $\text{Er}(\text{Co}_{1-x}\text{Si}_x)_2$ and $\text{Ho}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds (Duc *et al* 1992c, Duc 1996, Duc *et al* 1997). In these works, the resistivity enhancement was related to the magnetic enhancement (or, even, the appearance of the 3d magnetism) due to Si substitution.

Magneto-resistance was measured for a sample with $x = 0.025$. The result is presented in figure 9 in a plot of $\Delta R/R(0) = [R(B) - R(0)]/R(0)$ versus B , where $R(0)$ and $R(B)$ are the resistance in zero magnetic field and applied magnetic field, respectively. This figure indicates that at $T > T_C$ a rise in B to B_c initially causes an insignificant decrease in $\Delta R/R(0)$, whereas the magnetization of the compound, which is determined mainly by the R subsystem in these fields, reaches a rather large value (more than of approximately equal to one-third of the maximum value at several temperatures). The metamagnetic nature of the magnetization process at $T > T_C$, however, becomes more clearly manifested in magneto-resistance analysis: a sharp drop in the electrical resistance has been found at $B \sim B_c$, where there is a jump in the magnetization for the $\text{Ho}(\text{Co}_{0.975}\text{Si}_{0.025})_2$ compound.

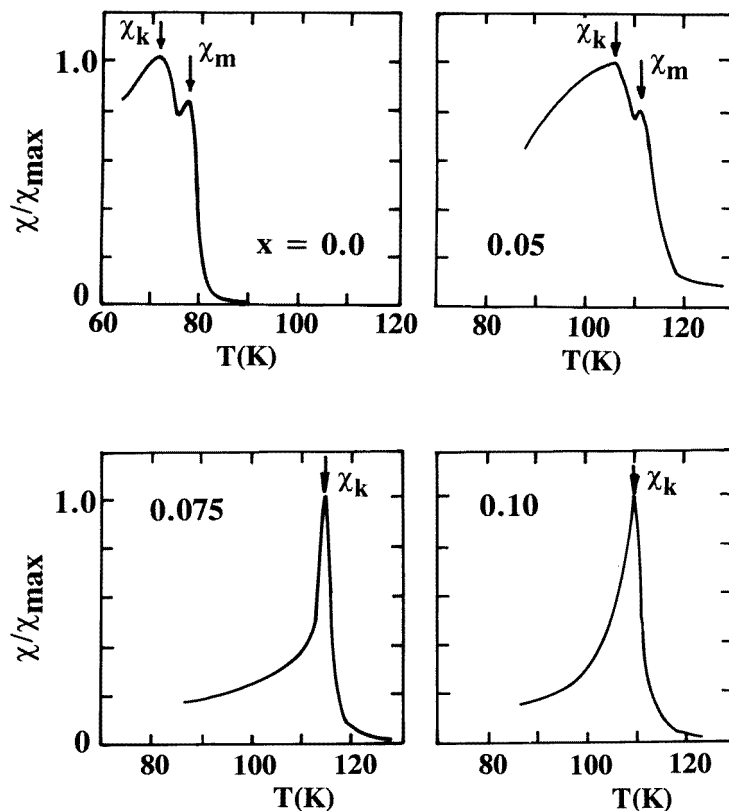


Figure 7. A.c. susceptibility of the $\text{Ho}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds.

To explain the resistance results we propose the following scenario. Above T_C , in RCo_2 , the resistivity is governed by spin-disorder scattering of conduction electrons on the paramagnetic rare-earth moments and on spin fluctuations arising mainly from local splitting of the cobalt 3d band (Duc *et al* 1991, Gratz *et al* 1995). When the temperature is lowered, the 4f moments which order ferromagnetically at T_C yield a strong molecular field in the material. In a strong enough field, the spin fluctuations at the Co sites are quenched and the 3d-band metamagnetic state is induced. As a consequence, the spin-disorder scattering is drastically suppressed, which yields the resistivity drop. This process may occur in zero magnetic field with the magnetic phase transition at T_C (as already indicated in figure 8) or at higher temperatures with the field-induced metamagnetic transition.

According to Ikeda *et al* (1991) the magnetoresistance of an itinerant magnet is written as

$$\Delta R(B, T) = \Delta R_c(B, T) + \Delta R_{sf}(B, T) \quad (2)$$

where $\Delta R_c(B, T)$ is a positive contribution, ascribed to the cyclotron motion of the conduction electrons, and the term $\Delta R_{sf}(B, T)$, which is a consequence of spin fluctuations, is negative.

We note that, for the $\text{Y}(\text{Co}, \text{Al})_2$ system, where only 3d magnetism is present, the positive contribution in (2) was found to be consistent with a quadratic dependence on

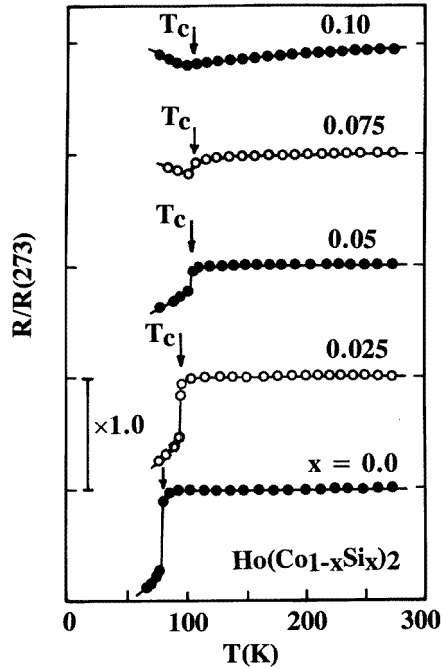


Figure 8. The temperature dependence of the electrical resistivity for the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds.

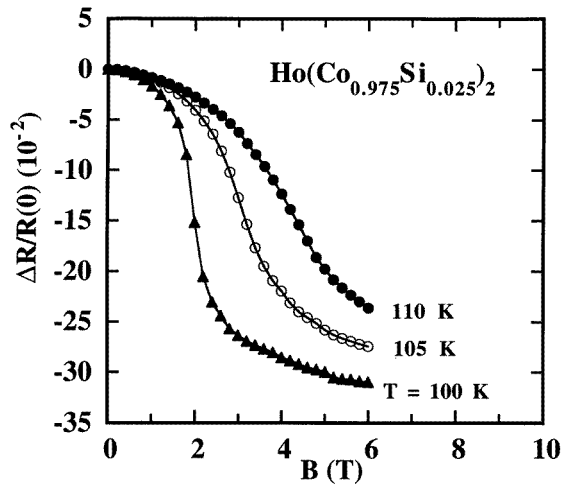


Figure 9. Magnetoresistance of the $\text{Ho}(\text{Co}_{0.975}\text{Si}_{0.025})_2$ compound.

the magnetization as expected for a pure volume effect. Denoting this relationship as $\Delta R/R(0) = \alpha M^2$, it emerges that $\alpha = 0.2 \mu_B^{-2}$ (Duc *et al* 1995). For the $\text{Er}(\text{Co}_{0.85}\text{Si}_{0.15})_2$ compound, where spin fluctuations are absent (Duc *et al* 1996), magnetoresistance simply increases with increasing field. In addition, the relationship between $\Delta R/R(0)$ and M^2 also holds, but with the α -value of $1.95 \times 10^{-3} \mu_B^{-2}$, i.e. two order of magnitude smaller

than that found for the $Y(\text{Co}, \text{Al})_2$ compounds. The weak positive magnetoresistance in $\text{Er}(\text{Co}_{0.85}\text{Si}_{0.15})_2$ might be due to the compensation between the contribution from the mentioned volume effect resulting from the 3d subsystem and that from the spin-disorder scattering on the paramagnetic rare-earth moments. In this way, instead of (2) we propose an expression of the magnetoresistance for a system consisting of localized spins and itinerant electrons as

$$\Delta R(B, T) = \Delta R_{mv}(B, T) + \Delta R_{spd}(B, T) + \Delta R_{sf}(B, T). \quad (3)$$

Here, the first term is a positive contribution caused by the appearance of the (stable) 3d magnetism; the second and third terms are the negative contributions from 4f spins and spin fluctuations.

It is allowed from the above discussion to consider that the observed suppression of the magnetoresistance is predominantly characterized by the quenching of spin-fluctuation scattering. This suggestion was confirmed in a systematic study of the effect of Si substitution on the quenching of spin fluctuations in the $R(\text{Co}, \text{Si})_2$ compounds (Duc *et al* 1997).

4. Discussions

4.1. R–Co intersublattice exchange interactions

As already mentioned above, strong enhancement of the ordering temperature was observed in the $R(\text{Co}_{1-x}\text{Si}_x)_2$ compounds with $R = \text{Ho}$ and Er . Note that such an enhancement is much stronger in the $R(\text{Co}_{1-x}\text{Al}_x)_2$ system. For the latter compounds, the variation of T_C as a function of Al concentration was associated with the enhancement of the Co susceptibility (Inoue and Shimizu 1988), but the strength of the 4f–3d exchange interactions was found to be constant (Ballou *et al* 1992, Brommer *et al* 1993). For the compounds under consideration, the variation of the Co susceptibility itself is not enough to describe the Si-concentration dependence of T_C . We, thus, consider the effect of the intersublattice exchange interactions.

In the mean-field approximation, the strength of the R–Co exchange coupling parameter A_{RCo} can be estimated from the ordering temperature. The corresponding expression for A_{RCo} reads as (Duc *et al* 1993b)

$$A_{RCo}^2 = \frac{3k_B(T_C - T_R)}{Z_{RCo}Z_{CoR}G_R\chi_{Co}(T_C)} \quad (4)$$

where G_R is the de Gennes factor for the rare-earth atom $G_R = (g - 1)^2 J(J + 1)$. T_R is the contribution to T_C due to the interactions between the rare-earth moments, which is determined by the ordering temperature of the $R\text{Ni}_2$ compounds (Duc *et al* 1993b). $\chi_{Co}(T_C)$ is the 3d susceptibility at T_C , which can be deduced from the temperature dependence of the magnetic susceptibility in non-magnetic $Y(\text{Lu})(\text{Co}, \text{Si})_2$ compounds. Z_{RCo} and Z_{CoR} are the number of Co neighbours to an R atom and the number of R neighbours to an Co atom, respectively.

By means of expression (4), the values of the microscopic R–Co exchange coupling parameter A_{RCo} are estimated and are listed in table 2 for the $R(\text{Co}_{1-x}\text{Si}_x)_2$ compounds ($R = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}$ and Er). The results show that with increasing Si concentration, A_{RCo} decreases. The effect of 3d–p hybridization on the 4f–3d exchange interactions was discussed by Duc and Givord (1995) for the Gd–Co–B system. These authors found that the presence of the B atom causes the 3d magnetic moment and then A_{RCo} to decrease. This is due to the transfer of the p electrons into the 3d band. This leads to an increase of the 3d–p

hybridization but to a decrease of the degree of 3d–5d hybridization. As a consequence, the strength of 4f–3d exchange interactions is reduced in the Gd–Co–B system. Now, the reduction of A_{RCo} in the $\text{R}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds can also be understandable on the same grounds. This finding is different to that observed for the $\text{R}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds. This may relate to the fact that Si atoms have one more 2p electron than to Al.

Table 2. Values of T_C (K) and the exchange–coupling parameter A_{RCo} (10^{-23} J) for a number of $\text{R}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds.

x	Gd		Tb		Dy		Ho		Er	
	T_C	A_{RCo}	T_C	A_{RCo}	T_C	A_{RCo}	T_C	A_{RCo}	T_C	A_{RCo}
0	395	18.8	227	18.1	142	18.0	78	17.8	32.6	17.6
0.025	—	—	—	—	142	15.6	95	16.4	44	18.0
0.050	390	16.7	230	14.7	148	14.5	112	15.4	54	15.6
0.075	—	—	—	—	146	12.4	115	12.0	64	15.1
0.100	385	17.6	230	13.9	146	10.9	112	10.7	68	13.4

4.2. The change in the type of magnetic phase transition

In principle, the FOT in RCO_2 must be discussed considering not only the sign of the c_3 -coefficient of the M^4 term in the expansion of the total free energy, but also the ratio between c_1 , c_3 and c_5 . Shimizu (1982) has already reported that in the case of $c_1 > 0$, $c_3 < 0$ and $c_5 > 0$ and the ratio of $\alpha = c_1 c_5 / c_3^2 > 9/20$ the magnetization curve $M(B)$ increases monotonically with increasing B . On the other hand, when $3/16 < \alpha = c_1 c_5 / c_3^2 < 9/20$ $M(B)$ is S-shaped, that is, the MT from the paramagnetic to ferromagnetic state with a hysteresis in the magnetization curve. Then the generalized condition for the appearance of the FOT is given as

$$c_1 > 0 \quad c_3 < 0 \quad c_5 > 0 \quad 3/16 < \alpha = c_1 c_5 / c_3^2 < 9/20. \quad (5)$$

When $\alpha < 3/16$, the system becomes ferromagnetic even at $B = 0$. Such an approximation, however, is not completely developed yet for the system containing both localized spins and itinerant electrons. The magnetic phase transitions in RCO_2 , until now, have been explained in terms of the Inoue–Shimizu model (Inoue and Shimizu 1982) by considering the sign of the c_3 -coefficient only. The value of c_3 at T_C is given as

$$c_3(T_C) = \{b_3 + a_3[n_{RCo}\chi_d(T_C)]^4\} / \{1 + n_{RCo}\chi_{Co}(T_C)\}^4. \quad (6)$$

In this expression, b_3 and a_3 are the coefficients of the M_f^4 and M_d^4 terms, respectively, in the expansion of the free energy of the R and Co sublattices. n_{RCo} is the molecular field coefficient which relates to A_{RCo} as follows:

$$n_{RCo} = Z_{RCo} A_{RCo} (g - 1) / g N_{Co} \mu_B^2. \quad (7)$$

According to Bloch *et al* (1975), the approximation of temperature dependence of a_3 is given by

$$a_3(T) = a_3(0) \{1 - (T/T_3)^2\}. \quad (8)$$

Here, $a_3(0)$ is negative and usually takes the value of $-15 \text{ T} (\text{mol A}^{-1} \text{ m}^{-2})^3$ (Duc *et al* 1988a, 1993a). T_3 is a characteristic temperature depending on the detailed band structure, but it is more suitable to regard T_3 as the temperature at which $a_3(T)$ changes sign.

The sign of $c_3(T_C)$ is practically governed by that of $a_3(T_C)$. Thus, the possibility of the appearance of an FOT in RCo_2 is, first of all, determined by the metamagnetic behaviour of the 3d subsystem. For the compounds under investigation, the magnetic character of the 3d subsystem can be considered as that in $\text{Y}(\text{Lu})(\text{Co}, \text{Si})_2$. In the work of Murata *et al* (1993), the metamagnetic transition was observed at low temperatures for $\text{Lu}(\text{Co}_{1-x}\text{Si}_x)_2$ with $x < 0.09$. The high-field magnetization data at high temperatures, however, are still absent for these compounds. For the $\text{R}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds with $\text{R} = \text{Dy}, \text{Ho}$ and Er , the FOT at T_C is observed with $x \leq 0.025$, $x \leq 0.075$ and $x \leq 0.01$, respectively (Duc 1996, Duc *et al* 1996). This finding is similar to that in $\text{R}(\text{Co}, \text{Al})_2$ compounds (Duc *et al* 1992b). Experimentally, guided by the linear relationship between the critical field of metamagnetism B_C and the temperature T_m —the temperature at which the 3d susceptibility exhibits a maximum, a scaling between T_m and T_3 has been made for the $\text{R}(\text{Co}, \text{Al})_2$ compounds (Duc *et al* 1992b). This assumption gives rise to rather large difference between T_3 and T_m compared to that deduced from the theory of Yamada (1993). However, it shows the possibility of explaining that the cross-over of the decrease in $T_3(x)$ and increase in $T_C(x)$ agrees with the observed change of the type of phase transition in $\text{R}(\text{Co}, \text{Al})_2$. For the compounds under investigation, the same procedure was applied. In table 3, the values of T_3 deduced from $\text{Y}(\text{Co}, \text{Si})_2$ (Michels *et al* 1990) and the values of $c_3(T_C)$ are listed for $\text{R}(\text{Co}, \text{Si})_2$ with $\text{R} = \text{Dy}, \text{Ho}$ and Er . Clearly, the calculated results agree well with the experimental observation of the type of magnetic phase transition in these compounds. Physically, the change from the FOT to the SOT on substitution of Co by Si, on one hand, may relate to the enhancement or even formation of 3d magnetism in these compounds. This may be consistent with the fact that no susceptibility maximum but only an enhancement of the magnetic susceptibility values with decreasing temperature was found for $(\text{Lu}, \text{Y})(\text{Co}_{1-x}\text{Si}_x)_2$ with $x > 0.1$ (Michels *et al* 1990, Murata *et al* 1993). Another mechanism might be speculated when considering the possibility of decoupling of critical temperatures of ordering of the 4f sublattice and of the Co metamagnetism (the latter might be pushed to lower temperature (Duc *et al* 1997)). Also, we should take into account the fact that Si atoms introduce in the compounds an additional type of exchange interaction and the statistical distribution of Si in the Co sublattice may lead for higher x to kinds of clustering effect and broadening of the critical temperature interval. The annealing process, however, is hard to improve this state.

Table 3. T_3 , the temperature at which $a_3(T)$ changes sign, for $\text{Y}(\text{Co}_{1-x}\text{Si}_x)_2$. T_C , the observed transition temperature, and $c_3(T_C)$ ($10^{-2} \text{ T (mol A}^{-1} \text{ m}^{-2})^3$), calculated on the basis of the corresponding T_3 -values.

x	T_3 (K)	$\text{Dy}(\text{Co}_{1-x}\text{Si}_x)_2$		$\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$		$\text{Er}(\text{Co}_{1-x}\text{Si}_x)_2$	
		T_C (K)	$c_3(T_C)$	T_C (K)	$c_3(T_C)$	T_C (K)	$c_3(T_C)$
0.0	178	142	-0.850	78	-0.089	32.6	-0.039
0.025	145	142	-0.086	95	-0.41	44	-0.085
0.050	100	148	6.8	112	-0.64	54	-0.41
0.075	80	146	25.2	115	13.9	64	-0.57
0.100	80	146	46.5	112	15.6	68	-1.48

The FOT at T_C as well as the metamagnetism at higher temperatures can be ascribed to the same origin, connected with the negative value of the c_3 -coefficient. The question, however, of why the metamagnetism exists only in a narrow temperature range (of about 15 K) above T_C is still open. It invites more detailed theoretical studies.

5. Concluding remarks

In order to obtain more information concerning the role of substituting atoms at Co sites, the $\text{R}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds are of special interest. Compared to the $\text{R}(\text{Co}_{1-x}\text{Al}_x)_2$ compounds, the effects of the 3d-electron concentration in the investigated compounds are the same; moreover, the influence of the volume effects is negligible. The special feature is only related to the fact that Si has one p electron more than Al. This leads to the difference in the degree of the 3d–p hybridization and then is the main reason for the modification of the magnetic behaviour. The effect of p electrons, first of all, causes the 4f–3d exchange interaction to decrease in the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds. The FOT in these compounds can be understood in terms of the metamagnetic behaviour of the 3d subsystem below the susceptibility maximum of the corresponding $\text{Y}(\text{Lu})(\text{Co}_{1-x}\text{Si}_x)_2$ compounds, whereas the SOT in the compounds with higher Si content (i.e. with $x \geq 0.075$) may relate to the enhancement or even formation of 3d magnetism in these compounds.

The MT above the FOT was observed not only by the magnetization but also by the magnetoresistance measurements for a number of the $\text{Ho}(\text{Co}_{1-x}\text{Si}_x)_2$ compounds. The effects of spin fluctuations play a very important role in the near-ferromagnets. The resistance in RCo_2 is influenced by several parameters; however, in our analysis, the suppression of the magnetoresistance at the MT can be considered as a main contribution from the quenching of spin-fluctuation scattering. The magnetoresistance at the MT, thus, can be considered as a useful way to measure the effects of spin fluctuations in the compounds investigated.

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