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Measurement of pressure effects on the magnetic and the magnetocaloric properties of the intermetallic compounds DyCo$_2$ and Er(Co$_{1-x}$Si$_x$)$_2$

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Abstract

The effect of external pressure on the magnetic properties and magnetocaloric effect of polycrystalline compounds DyCo$_2$ and Er(Co$_{1-x}$Si$_x$)$_2$ ($x = 0, 0.025$ and 0.05) has been studied. The ordering temperatures of both the parent and the Si-substituted compounds are found to decrease with pressure. In all the compounds, the critical field for metamagnetic transition increases with pressure. It is seen that the magnetocaloric effect in the parent compounds is almost insensitive to pressure, while there is considerable enhancement in the case of Si-substituted compounds. Spin fluctuations arising from the magnetovolume effect play a crucial role in determining the pressure dependence of the magnetocaloric effect in these compounds. The variation of the magnetocaloric effect is explained on the basis of the Landau theory of magnetic phase transitions.

1. Introduction

The magnetocaloric effect (MCE) is an intrinsic property of all magnetic materials and is induced via the coupling of the magnetic sublattice with an applied magnetic field. The MCE is measured in terms of isothermal magnetic entropy change and/or adiabatic temperature change [1]. The MCE is being exploited in magnetic refrigeration technology and there is a strong demand for materials with giant MCE to be used as magnetic refrigerants. The possibility of use of magnetic refrigeration in both 'near room temperature' as well as in the cryogenic temperature regime has led to intense research in the field of MCE [2–8].

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The variety of the magnetic properties exhibited by the rare earth–transition metal intermetallics in general and the occurrence of giant MCE in materials such as $Gd_5(Si, Ge)_4$ in particular have made these materials the natural probe for fundamental studies as well as for applications based on MCE [2, 5–9]. It has been established that giant MCE is possible only in materials which exhibit first order transitions, metamagnetic transitions or field-induced magnetostructural transitions. Several studies have shown that the giant MCE in $Gd_5(Si, Ge)_4$ compounds is a result of magnetostructural transitions [2, 3]. Among the various intermetallics, $La(Fe, Si)_{13}$ compounds are known to possess large MCE near room temperature by virtue of the first order transition caused by the occurrence of itinerant electron metamagnetism in the Fe sublattice [7]. Another class of compounds which exhibits itinerant electron metamagnetism and therefore first order transition is RCo_2 with R = Er, Ho and Dy [10–12]. The creation of the Co moment by the molecular field of the rare earth, as these compounds are cooled through their ordering temperatures (T_C), is termed itinerant electron metamagnetism, which leads to first order transition at T_C. The presence of itinerant electron metamagnetism in these compounds leads to significant MCE and has made these materials attractive from the point of view of magnetic refrigeration applications. Considerable effort, both on the experimental as well as theoretical fronts, has been put into understanding the origin of itinerant electron metamagnetism and giant MCE in these compounds [10, 13–18]. According to previous theories, the critical parameter that governs the itinerant electron metamagnetism and the first order nature of the magnetic transition in the RCo_2 compounds is the molecular field or the ordering temperature [14]. Though these theories were successful in the case of compounds with Er, Ho, and Dy, the second order transition observed in other compounds of this series could not be explained satisfactorily. Recently, these models have been modified by incorporating the contributions from the magnetovolume effect and spin fluctuations [10]. The most important feature in the modified model is the role of the lattice parameter in determining the order of magnetic transition. Therefore, it is of great interest to study the magnetic and magnetocaloric properties of these compounds as a function of substitutions as well as applied pressure. The compounds selected for the study are $DyCo_2$ and $Er(Co_{1-x}Si_x)_2$ ($x = 0, 0.025$ and 0.05). Though the pressure dependence of magnetic and electrical properties has been studied in RCo_2 compounds [19–23], to the best of our knowledge this is the first report on the influence of pressure on the MCE in parent as well as substituted compounds.

2. Experimental details

The preparation and the characterization techniques for all the compounds have been reported elsewhere [12, 24]. The magnetization (M) measurements under various applied pressures (P) have been performed using a Cu–Be clamp type cell, which can work up to 12 kbar, attached to a SQUID magnetometer. The magnetic ordering temperatures have been calculated from the (dM/dT) plots obtained from the 'field-cooled' magnetization variation as a function of temperature (T). In the 'field-cooled' mode the samples were cooled in the presence of a field (H) of 200 Oe and the magnetization was measured under the same field in the warming cycle. The MCEs of all the compounds have been determined from the $M–H$ isotherms collected at intervals of 4 K close to the ordering temperatures.

3. Results and discussion

The Rietveld refinement of the room temperature powder x-ray diffractograms confirms that $DyCo_2$ and $Er(Co_{1-x}Si_x)_2$ ($x = 0, 0.025$ and 0.05) compounds are single phase and crystallize in $MgCu_2$-type cubic Laves phase structure. The lattice parameter (a) of $Er(Co_{1-x}Si_x)_2$
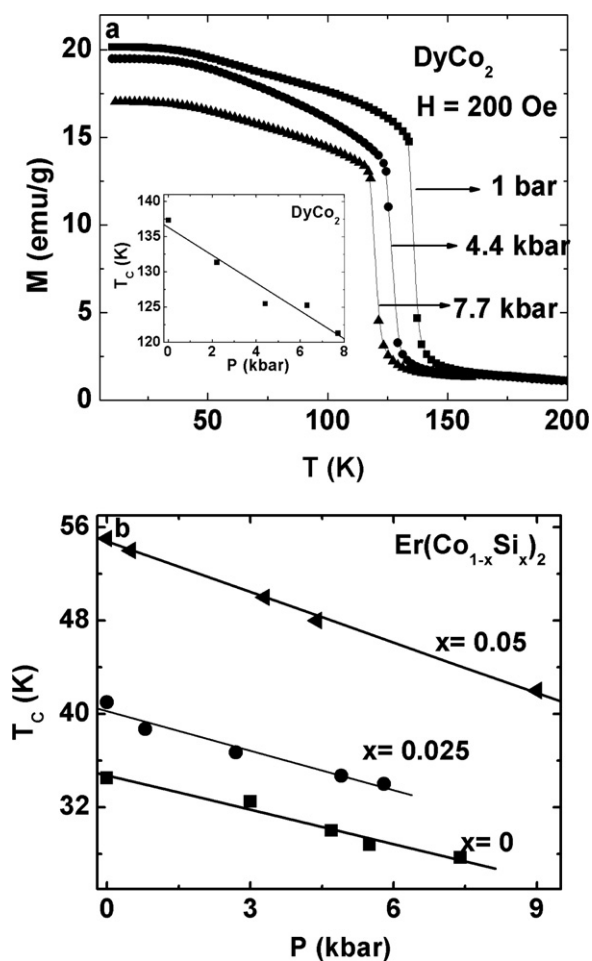
Figure 1. (a) Temperature variation of magnetization data of DyCo$_2$ obtained under different pressure conditions and in an applied field of 200 Oe. The inset of (a) shows the pressure dependence of ordering temperatures of DyCo$_2$. (b) T_C versus P plot of Er(Co$_{1-x}$Si$_x$)$_2$ ($x = 0, 0.025$ and 0.05) compounds.

compounds was found to increase from 7.135 ± 0.004 Å to 7.148 ± 0.005 Å as the Si concentration increases from 0 to 0.05, whereas the a value for DyCo$_2$ was estimated to be 7.179 ± 0.003 Å. Figure 1 shows the temperature dependence of the 'field-cooled' magnetization data of DyCo$_2$ obtained under different pressures and in an applied field of 200 Oe. It can be seen from the figure that the first order transition in DyCo$_2$ is characterized by a sharp change in the magnetization near T_C. The variation of T_C as a function of pressure is shown in the inset of figure 1(a). The M–T plots in the same field and under various pressures have been obtained in Er(Co$_{1-x}$Si$_x$)$_2$ compounds as well. Figure 1(b) shows the T_C versus P plot of Er(Co$_{1-x}$Si$_x$)$_2$ compounds. It may be noticed from figure 1(b) that the T_C values of Er(Co$_{1-x}$Si$_x$)$_2$ increase with Si concentration, whereas they decrease with pressure. The dT_C/dP value obtained for ErCo$_2$ in the present case is in good agreement with the value reported for single crystals of ErCo$_2$ [19].

The increase in the T_C with Si is attributed to the magnetovolume effect resulting from the increased lattice parameter in the Si-substituted compounds. In this context, it may be

Table 1. The ordering temperature (T_C) and the maximum value of the isothermal magnetic entropy change (ΔS_M^{max}) of $DyCo_2$ and $Er(Co_{1-x}Si_x)_2$ ($x = 0$, 0.025 and 0.05) compounds under various pressures (P).

DyCo₂			ErCo₂			Er(Co₀.₉₇₅Si₀.₀₂₅)₂			Er(Co₀.₉₅Si₀.₀₅)₂		
P	T_C	ΔS_M^{max}	P	T_C	ΔS_M^{max}	P	T_C	ΔS_M^{max}	P	T_C	ΔS_M^{max}
(kbar)	(K)	(J kg⁻¹ K⁻¹)	(kbar)	(K)	(J kg⁻¹ K⁻¹)	(kbar)	(K)	(J kg⁻¹ K⁻¹)	(kbar)	(K)	(J kg⁻¹ K⁻¹)
0[a]	137	12	0[a]	35	33	0[a]	41	27.4	0[a]	55	22.7
4.4	126	12.3	4.7	30	32.8	2.7	37	28.8	3.3	50	24.8
7.7	121	12.4	7.4	28	32.5	5.8	34	29.9	9	42	26.6

[a] Ambient pressure.

mentioned here that, according to Khmelevskyi and Mohn [10], the condition of itinerant electron metamagnetism in the RCo_2 compounds is satisfied only for the compounds having the lattice parameter (a) in the range of 7.05 to 7.22 Å. The compounds with $a < 7.05$ Å possess a nonmagnetic Co sublattice, whereas in compounds with $a > 7.22$ Å the Co sublattice is magnetic. Therefore, the increase in the lattice parameter as a result of Si substitution takes the system closer to the critical lattice parameter required for the stable moment formation in the Co sublattice, thereby contributing to the enhancement of T_C. A similar effect has been observed in Si-substituted $DyCo_2$ and $HoCo_2$ compounds as well [12, 25]. The lattice expansion brought about by Si causes the 3d band narrowing and consequently an enhancement of the 3d magnetic character. Based on the studies of the magnetic properties of $R(Co_{1-x}Al_x)_2$, Duc *et al* [13] have also pointed out a strong volume dependence of the Co magnetic state.

On the other hand, application of external pressure would cause a reduction in the lattice parameter and therefore would lead to a decrease in the T_C of both the parent as well as the substituted compounds, as seen in figure 1. This is because of the 3d band broadening resulting from the lattice contraction, caused by the pressure. It is also found that the rate of decrease of T_C (i.e. dT_C/dP) increases with Si concentration. This implies that the applied pressure has a larger role in the Si substituted compounds. Since the Si substituted compounds have enhanced spin fluctuations as compared to the parent compounds, the pressure has a larger role in the former and therefore the dT_C/dP values are consequently larger. Therefore, the application of pressure is found to produce a negative magnetovolume effect, in contrast to the positive magnetovolume effect resulting from the Si substitution.

It may be mentioned here that the role of magnetovolume effect on the Co magnetic state of the RCo_2 (R = Er, Ho) compounds has been illustrated by the pressure dependent study of magnetic properties and electrical resistivity [19, 22]. Based on the resistivity studies as a function of pressure, Syshchenko *et al* [22] have arrived at a critical pressure of ∼40 kbar for the disappearance of magnetic moment in Co sublattice in $ErCo_2$. Above this critical pressure the Co sublattice becomes similar to the Ni sublattice (which is nonmagnetic) in $ErNi_2$ and the ordering temperature becomes almost insensitive to pressure. The variation of T_C in $DyCo_2$ and $Er(Co_{1-x}Si_x)_2$ compounds with pressure is shown in table 1.

Figure 2 shows the field dependence of magnetization isotherms of $ErCo_2$ (T_C under ambient conditions is 35 K) collected at 34 K and under various pressures. The M–H plots of $Er(Co_{0.95}Si_{0.05})_2$ (T_C under ambient conditions is 55 K) obtained at 58 K and at different pressures are given as an inset in this figure. It can be seen from the figure that the M–H isotherm at ambient pressure in $ErCo_2$ does not show any metamagnetic transition whereas a clear metamagnetic transition is seen in the M–H data at higher pressures. Furthermore, the critical field required for metamagnetism increases with pressure and eventually the metamagnetism vanishes completely for pressures as high as 7.4 kbar. A similar effect of
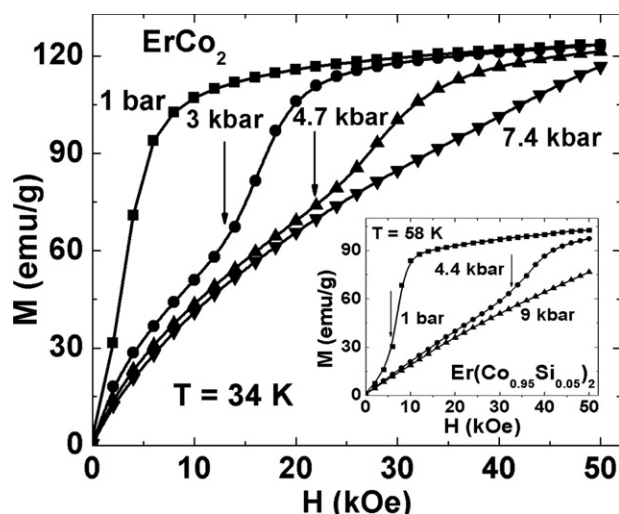
Figure 2. Field dependence of the magnetization isotherm of ErCo$_2$, obtained at $T = 34$ K under various applied pressures. The inset of the figure shows the M versus H plot of Er(Co$_{0.95}$Si$_{0.05}$)$_2$ collected at $T = 58$ K under different pressures. The arrows in the figure indicate the onset of metamagnetic transition.
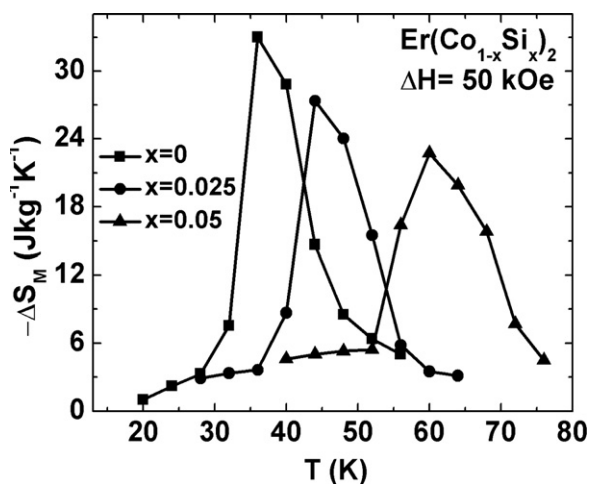

Figure 3. Temperature variation of isothermal entropy change of Er(Co$_{1-x}$Si$_x$)$_2$ compounds at ambient pressure for a field change of 50 kOe.

pressure on the critical field is observed in the Si-substituted compounds as well (inset of figure 2). These observations also suggest that the Co sublattice is driven away from the magnetic state by the applied pressure. Yamada has also reported a similar effect of pressure on the critical field required for the metamagnetism in the YCo$_2$ compound [26].

The magnetocaloric effect, in terms of isothermal entropy change ($-\Delta S_M$), for all the compounds has been calculated using the Maxwell's relation [27]. Figure 3 shows the ΔS_M versus T plot of Er(Co$_{1-x}$Si$_x$)$_2$ compounds at ambient pressure. It can be seen from the figure that the ΔS_M versus T plot of all the compounds show a maximum near the T_C. The maximum value of ΔS_M (ΔS_M^{max}) for ErCo$_2$ for $\Delta H = 50$ kOe is 33 J kg^{-1} K^{-1}. The
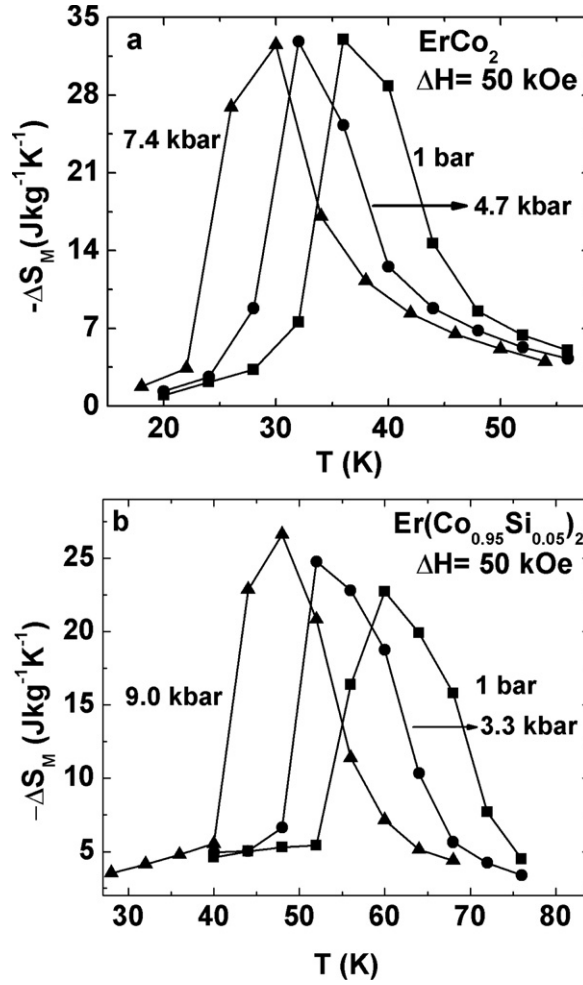
Figure 4. (a), (b) ΔS_M versus T plots of ErCo$_2$ and Er(Co$_{0.95}$Si$_{0.05}$)$_2$ under various pressures. All the ΔS_M values have been calculated for a field change of 50 kOe.

$\Delta S_M^{\max}$ of Er(Co$_{0.95}$Si$_{0.05}$)$_2$, for the same ΔH, is found to be 22.7 J kg^{-1} K^{-1}. The decrease in $\Delta S_M^{\max}$ with increase in Si concentration is consistent with previous reports [11, 24, 28]. A similar reduction in the MCE values has also been observed in Dy(Co, Si)$_2$ compounds [12]. Enhanced spin fluctuations resulting from the Si substitution decreases the strength of itinerant electron metamagnetism and consequently weakens the first order nature of the transition at T_C. Therefore, the MCE decreases with increase in Si concentration. The detrimental role of the spin fluctuations on the strength of itinerant electron metamagnetism and MCE has been reported by Han *et al* as well [29].

Figures 4(a) and (b) show the ΔS_M versus T plots of ErCo$_2$ and Er(Co$_{0.95}$Si$_{0.05}$)$_2$, calculated for various applied pressures. Table 1 gives the summary of the pressure dependence of T_C and $\Delta S_M^{\max}$ in DyCo$_2$ and Er(Co$_{1-x}$Si$_x$)$_2$ compounds. It can be seen from figure 4 (also from table 1) that, with increase in pressure, the peak in the ΔS_M versus T plot moves towards low temperatures in both the parent as well as the substituted compounds. However, in the former case, the $\Delta S_M^{\max}$ value almost remains insensitive to pressure while in the latter case

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ΔS_M^{max} is found to increase. It is evident from table 1 that the pressure dependence of MCE in DyCo$_2$ is similar to that of ErCo$_2$, while the dependence in Er(Co$_{0.975}$Si$_{0.025}$)$_2$ is similar to that of Er(Co$_{0.95}$Si$_{0.05}$)$_2$. The insensitivity of MCE to applied pressure in the case of ErCo$_2$ and DyCo$_2$ may be due to the fact that the strength of itinerant electron metamagnetism has diminished only nominally even at a pressure of about 7.7 kbar. In fact, Hauser *et al* [21] have reported that the discontinuity (at T_C) in the magnetic contribution to the electrical resistivity in a polycrystalline sample of ErCo$_2$ decreases to about 60% as the pressure is increased from 1 bar to ∼16 kbar, which is attributed to the reduction in the strength of metamagnetism. In view of this, it is reasonable to assume that for a pressure of 7.7 kbar the reduction in the strength of metamagnetism is not very much and therefore would contribute only to a nominal reduction in ΔS_M^{max}. However, the reduction in T_C brought about by pressure would try to increase ΔS_M^{max} due to the reduction in the thermal spin fluctuations. Therefore, it is quite possible that the reduction in MCE caused by the weakening of the metamagnetism is just compensated by the increase in MCE arising out of the reduction in T_C. Though the pressure dependence of MCE in DyCo$_2$ is similar to that of ErCo$_2$, the scenario may be slightly different in the former. By virtue of the larger lattice parameter, the pressure dependence of the strength of itinerant electron metamagnetism would be weaker in DyCo$_2$ [21]. The insensitivity of MCE to pressure seen in the case of DyCo$_2$ is consistent with the observations made by Hauser *et al* [21].

On the other hand, the pressure dependence of ΔS_M^{max} in the case of Si-substituted compounds is considerable. In this context, it is of importance to note that the dT_C/dP value also showed an increase upon Si substitution in Er(Co, Si)$_2$ compounds. The increase seen in ΔS_M^{max} with pressure may be attributed to the suppression of spin fluctuations by the pressure. The negative magnetovolume effect caused by the pressure tries to compete with the positive magnetovolume effect resulting from the Si substitution. In order to understand the effect of pressure on the Si-substituted compounds, we have studied the nature of magnetic transition occurring in these compounds as a function of pressure. This has been done by calculating the temperature variation of the Landau coefficients. It is well known that the magnetic free energy, $F(M, T)$, in general can be expressed as a Landau expansion in the magnetization as

$$F(M, T) = \frac{C_1}{2}M^2 + \frac{C_3}{4}M^4 + \frac{C_5}{6}M^6 + \cdots - \mu_0 H M \tag{1}$$

where C_1, C_3 and C_5 are the Landau coefficients which are temperature dependent. The temperature and magnetic field dependences of $F(M, T)$ determine the nature of the magnetic transition. The Landau coefficients can be calculated using the equation of state, given by

$$\mu_0 H = C_1 M + C_3 M^3 + C_5 M^5. \tag{2}$$

It may be noted from equation (2) that the magnetization isotherms obtained at various temperatures allow one to determine the temperature variation of the Landau coefficients. It is well known that the temperature dependence of the Landau coefficients can be utilized to distinguish between the first and second order transitions of magnetic materials [7, 15, 30]. Generally, the compounds with first order transition possess positive values for $C_1(T_C)$, $C_5(T_C)$ and negative value for $C_3(T_C)$. Furthermore it has been reported that the magnitude of the C_3 at temperatures well below T_C determines the magnitude of MCE in giant magnetocaloric materials [16].

The Landau coefficients of all the compounds have been determined from the $M–H$ isotherms obtained at various temperatures. The temperature variation of C_1 of all the compounds exhibits a minimum near their T_C. Figure 5 shows the temperature variation of the coefficient C_3 of Er(Co$_{1-x}$Si$_x$)$_2$ (with $x = 0$ and 0.05) under different pressures. It may be noted from the figure that for both the compounds the sign of C_3 near T_C is negative and that its magnitude decreases with increase in temperature. A similar trend has been obtained
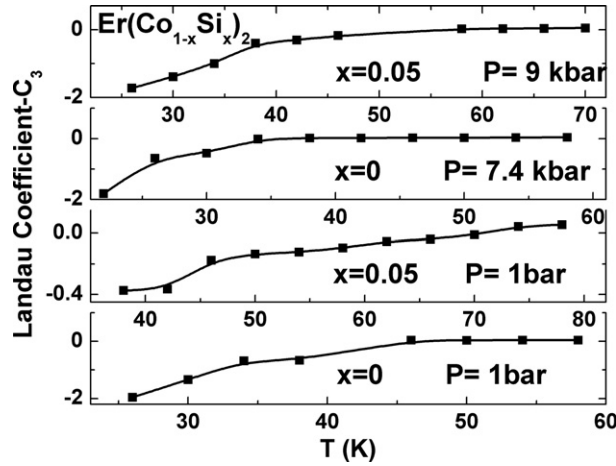
Figure 5. Temperature dependence of the Landau coefficient C_3, obtained under various external pressures, in $Er(Co_{1-x}Si_x)_2$ compounds with $x = 0, 0.05$. (C_3 values have been calculated in cgs units.)

for $Er(Co_{0.975}Si_{0.025})_2$ and $DyCo_2$ as well. Therefore, the temperature variation of C_3 of all the compounds indicates the presence of a first order transition at T_C. It may also be seen from figure 5 that at ambient pressure the magnitude of C_3 in $Er(Co_{0.95}Si_{0.05})_2$ at temperatures well below T_C is lower than that of $ErCo_2$. The low temperature C_3 value of $Er(Co_{0.95}Si_{0.05})_2$ increases with increasing pressure whereas no significant change in the C_3 value has been observed in $ErCo_2$. In the case of $DyCo_2$, the variation was similar to that of $ErCo_2$. This implies that with the application of pressure, the strength of itinerant electron metamagnetism increases considerably in the Si-substituted compounds whereas there is no significant change in the parent compounds. It may also be noticed from figure 5 that the difference between the low temperature C_3 values of $ErCo_2$ and the Si-substituted compound almost vanishes with increase in pressure. At this point it is worth noting the fact that the ΔS_M^{max} value in the case of $ErCo_2$ and $DyCo_2$ is almost insensitive to the pressure change, but ΔS_M^{max} is quite dependent on pressure in the case of Si-substituted $ErCo_2$ compounds. Therefore, the variations of the magnitude of C_3 with Si concentration (at ambient pressure) as well as with pressure are consistent with the MCE variation, which implies that there is a strong correlation between the C_3 value and the MCE in an RCo_2-based itinerant electron metamagnetic system. In this context, it is of importance to mention that Yamada *et al* [16] have indeed shown that the MCE in itinerant electron metamagnetic systems is primarily governed by the magnitude of C_3. Fujita *et al* have also reported a similar dependence of MCE on C_3 in $La(Fe, Si)_{13}$ compounds, which is also a well known itinerant electron metamagnetic system [31].

It has been mentioned above that the partial substitution of Si for Co in $ErCo_2$ decreases the magnitude of C_3 at ambient pressure. We attribute this decrease to the reduction in the strength of itinerant electron metamagnetism, which, in turn, may be assumed to arise from the magnetovolume effect. Local magnetic moments developed as a result of increased lattice parameter after Si substitution (magnetovolume effect) are not properly exchange coupled and therefore they act as spin fluctuations. The effect of these fluctuations on the magnetic and electrical resistivity behaviour has already been reported in $R(Co, Al)_2$ compounds [32]. These fluctuations may suppress the metamagnetism and hence the C_3 values. On the other hand, the effect of applied pressure (in the Si-substituted compounds) is to compete with the positive

magnetovolume effect and to reduce it. In other words, pressure tries to make the magnetic nature of the Co sublattice in the Si-substituted compounds similar to that of $ErCo_2$. This is exactly seen in figure 5, which shows that at high pressures, the low temperature C_3 value of $Er(Co_{0.95}Si_{0.05})_2$ is almost equal to that of $ErCo_2$.

In short, we find that the pressure dependence of T_C and the MCE in the substituted compounds is larger, as compared to that of the parent compounds. The reason for this is that the pressure causes a negative magnetovolume effect, which is more predominant in the case of Si-substituted compounds. In the case of parent compounds, the pressure dependence of magnetic and magnetocaloric properties seen in the present work seems to be in agreement with the pressure dependent electrical resistivity data reported recently. Unfortunately, there are no reports available on the pressure dependent resistivity studies on substituted RCo_2 compounds, which would have been quite useful in this context.

4. Conclusions

In conclusion, we have studied the effect of pressure on the magnetic and magnetocaloric properties of $DyCo_2$ and $Er(Co_{1-x}Si_x)_2$ compounds which show itinerant electron metamagnetism. The application of pressure results in the reduction of the ordering temperature, both in the parent as well as in the Si-substituted compounds. The external pressure causes an enhancement in the critical field required for the metamagnetism. The magnetocaloric effect in the parent compounds is found to be almost insensitive to pressure, while there is considerable enhancement in the case of Si substituted compounds. Due to the presence of enhanced spin fluctuations (arising due to the magnetovolume effect) in the Si substituted compounds, the influence of pressure is more visible. With increase in pressure, the negative effect of pressure is found to compete with the magnetovolume effect, thereby enhancing the MCE. The isothermal magnetic entropy change is found to behave in the same manner as C_3, both with Si concentration (at ambient pressure) and with the applied pressure.

Acknowledgment

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