The magnetic environment of the rare earth site in RT_2Si_2 compounds (R = rare earth, T = Cr & Mn)

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Specific heat measurements and ^{169}Tm Mössbauer spectroscopy demonstrate that the R sub-lattice in $ErMn_2Si_2$, $ErCr_2Si_2$, $TmMn_2Si_2$ and $TmCr_2Si_2$ is not influenced by the Mn or Cr sub-lattice magnetisation and orders independently close to liquid helium temperature with T_C = 4.6 K, 1.9 K, 6.8 K and <2.7 K respectively.

1. Introduction

The intermetallic compounds RT_2Si_2 (R = rare earth, T = extensive set of 3d transition metal elements) form with the body centred tetragonal, $ThCr_2Si_2$ prototype structure (space group #139, I4/mmm). A systematic study of the crystal field (CF) acting at the R sites in this series has so far been directed at T = Cu, Ni, Co and Fe ([1] and references therein). It was planned to extend the study to include T = Cr and Mn. However, for these compounds the T sub-lattice is reported to order antiferromagnetically well above room temperature [2, 3] and the R sub-lattice ordering temperatures are not well known. It is important that the CF study is not confused by any magnetic fields acting at the R site. In this work specific heat measurements, which are very sensitive to low temperature magnetic transitions, and ^{169}Tm Mössbauer spectroscopy have been employed to verify that the R sub-lattice orders independently close to liquid helium temperature and that the R-site is not perturbed by the Mn or Cr sub-lattice magnetisation.

2. Specimen preparation

Polycrystalline specimens of $ErMn_2Si_2$, $ErCr_2Si_2$, $TmMn_2Si_2$ and $TmCr_2Si_2$ were prepared from the elements by argon arc melting at least 6 times to achieve homogeneity. The starting metal impurities were 99.99 % for Mn, Cr and Si and 99.9 % for Tm and Er. The specimens with T = Cr were also vacuum-annealed in Ta foil at 800 °C for 6 days.

The x-ray powder diffraction patterns were recorded using Cu K_α radiation and

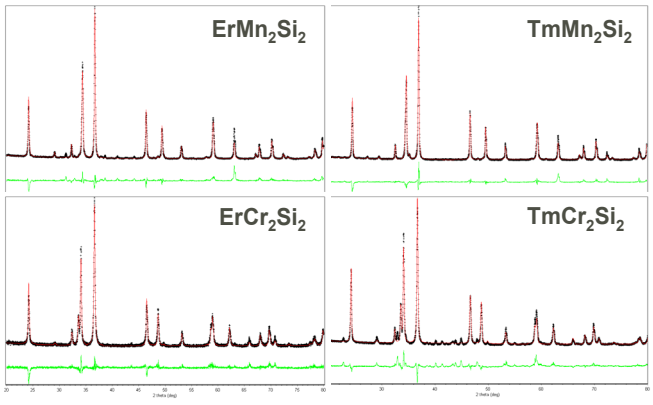

Fig. 1. X-ray diffraction patterns for $ErMn_2Si_2$, $ErCr_2Si_2$, $TmMn_2Si_2$ and $TmCr_2Si_2$

calibrated against silicon powder. Rietveld analyses were performed using *Rietica* [4]. All four specimens were predominantly single phase with lattice parameters in good agreement with literature values (see Table 1). Small concentrations of R_2O_3 were found in all specimens and $TmSi_2$ was identified only in $TmMn_2Si_2$. Some further unidentified diffraction lines were present in the pattern for $TmCr_2Si_2$. The presence of these impurities in such small amounts (e.g. < 0.5 wt% Tm associated with $TmSi_2$ in $TmCr_2Si_2$) is unlikely to contribute noticeably to the specific heat signal or the Mössbauer spectra.

3. Results

3.1 Specific heat

The specific heat measurements (Fig. 2) were carried out at Toyoma University, Japan using the relaxation method on a Quantum Design PPMS. The Curie temperatures shown in Fig. 2 have been taken as the points of greatest rate of change in specific heat. The more complex nature of the data for the Tm-based intermetallics may be associated with low-lying singlet states of the CF scheme for the non-Kramers Tm^{3+} ion. It is not clear if the broad low temperature feature observed for $TmCr_2Si_2$ corresponds to a magnetic transition. For this reason, preliminary ^{169}Tm Mössbauer spectra were recorded for this compound.

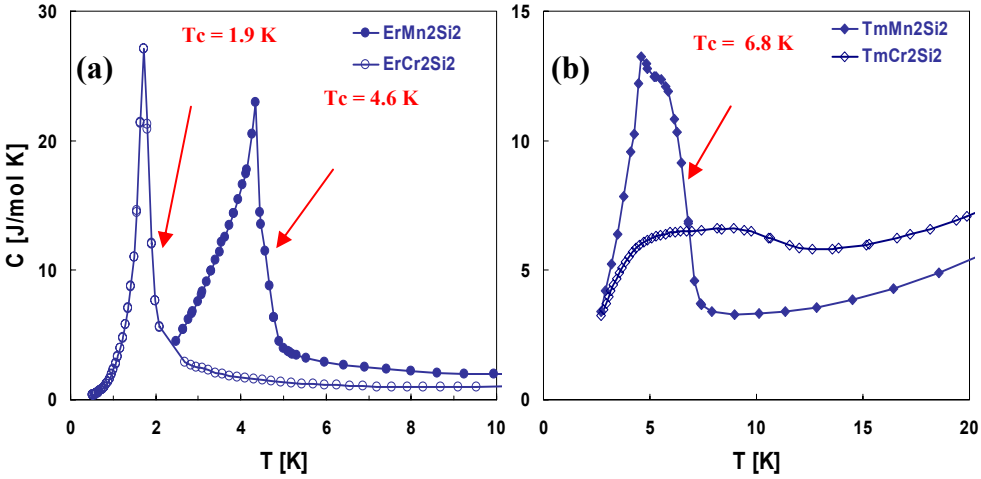

Fig. 2. Specific heat data for (a) $ErMn_2Si2$ & $ErCr_2Si_2$ and (b) $TmMn_2Si_2$ & $TmCr_2Si_2$.

3.2 ^{169}Tm Mössbauer spectroscopy

The ^{169}Tm Mössbauer spectra (Fig. 3) were recorded with the $TmCr_2Si_2$ absorber (≈ 8 mg cm^{-2} specimen material) mounted in a transmission geometry cryostat. The source was a

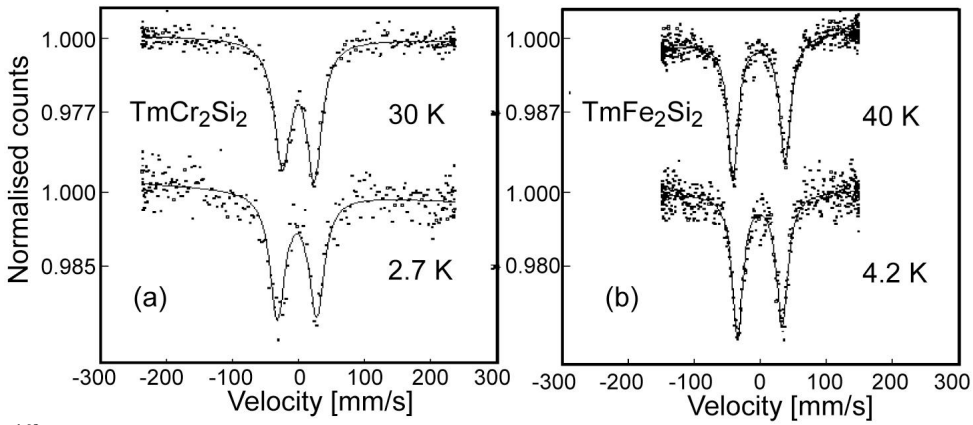

Fig. 3. ^{169}Tm Mössbauer spectra for (a) $TmCr_2Si_2$ at 2.7 K and 30 K and (b) $TmFe_2Si_2$ at 4.2 K and 40 K

neutron activated foil of ^{169}Er:$^{168}ErAl_9$ mounted outside the cryostat on a transducer which was moved with sinusoidal motion and calibrated against a standard TmF_3 absorber at 4.2 K. The spectra for $TmCr_2Si_2$ exhibit no magnetic splitting down to 2.7 K and the line half-widths at 2.7 K (≈ 14.0 mm/s) and 30 K (≈ 13.9 mm/s) are comparable. This indicates that $T_C < 2.7$ K and rules out magnetic line broadening at these temperatures. The line-widths for the $TmCr_2Si_2$ spectra are also comparable to those for $TmFe_2Si_2$ spectra where the Fe sub-lattice is known not to order magnetically, at least down to liquid helium temperature.

Table 1. Lattice parameters and T_C for Er Mn_2Si_2, $ErCr_2Si_2$, $TmMn_2Si_2$ and $TmCr_2Si_2$

Compound	a [nm]	c [nm]	Ref	T_N [K]	Method	Ref
$ErMn_2Si_2$	0.3895(1)	1.0410(1)	This work	4.6	Specific heat	This work
	0.3920(3)	1.0456(1)	[5]	5	Magnetic susceptibility	[6]
$ErCr_2Si_2$	0.3888(3)	1.0617(5)	This work	1.9	Specific heat	This work
	0.3889(1)	1.0614(1)	[7]	2.4	Neutron diffraction	[3]
$TmMn_2Si_2$	0.3886(1)	1.0404(1)	This work	6.8	Specific heat	This work
	0.3887(1)	1.0398(1)	[6]	6.5	Magnetic susceptibility	[6]
$TmCr_2Si_2$	0.3871(1)	1.0617(5)	This work	<2.7	^{169}Tm Mössbauer	This work

Conclusion

For three of the compounds, the R sub-lattice orders magnetically close to liquid helium temperature. Preliminary Mössbauer spectroscopy measurements for the fourth compound, $TmCr_2Si_2$, demonstrate that the ^{169}Tm nucleus is not subject to significant magnetic broadening or transferred hyperfine fields down to 2.7 K. On the basis of these results, we will proceed with the planned ^{169}Tm Mössbauer spectroscopy and inelastic neutron scattering studies of the CF interaction at the R site in RT_2Si_2 (T = Mn, Cr).

Acknowledgements

This project was supported by AINSE Grant 06/165. Banchachit Saensunon gratefully acknowledges the Royal Thai Army for his scholarship. Vernon Edge and Wayne Hutchison are thanked for their assistance with specimen preparation.

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