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Valence and magnetic ordering in $YbMn_2Si_{2-x}Ge_x$

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Abstract

^{170}Yb Mössbauer spectroscopy has been used to study $YbMn_2Si_{2-x}Ge_x$ for $0 \leqslant x \leqslant 2$. $YbMn_2Si_2$ contains only trivalent Yb. A Yb^{2+} component appears at $x = 1.15(5)$ and the Yb in $YbMn_2Ge_2$ is fully divalent. Transferred hyperfine fields at the Yb site for $x < 1.2$ show that a canting of the Mn moments occurs between 35 and 65 K, while a second event is seen at ∼6 K for $0.4 \leqslant x \leqslant 1.0$ which probably reflects a further reorganization of the Mn order rather than ordering of the Yb^{3+} moments.

1. Introduction

The RT_2X_2 system (where R is a rare earth, T is a 3d transition metal and X is Si or Ge) exhibits a wide variety of interesting magnetic and physical properties associated with the interplay of rare earth exchange interactions and anisotropies [1]. Mn is the only transition metal to carry a moment in these alloys, and the strong Mn–Mn exchange leads to relatively high ordering temperatures. The $YbMn_2Si_{2-x}Ge_x$ system is especially interesting as the Yb ion can exist in two valence states, being trivalent (Yb^{3+}) for $x = 0$ and divalent (Yb^{2+}) for $x = 2$. The entire series adopts the tetragonal $ThCr_2Si_2$-type (bct) structure (space group $I4/mmm$, No 139) with Yb on the 2a site, Mn on the 4d site and $Si_{2-x}Ge_x$ occupying the 4e site. Early work by Nowik *et al* [2] showed that the trivalent silicide ($x = 0$) behaved much as other RMn_2Si_2 alloys with the heavy rare earths, with a comparable Néel temperature and similar ordering; however, a second transition at ∼35 K was observed and attributed to ordering of the Yb^{3+} moments. By contrast, the divalent germanide exhibited multiple magnetic transitions reminiscent of alloys with the light rare earths, with four magnetic events being identified in $YbMn_2Ge_2$ [2].

More recent neutron diffraction work has led to a better understanding of the magnetic ordering of the end-members ($x = 0, 2$) of this series. Only two of the reported four transitions in the germanide have been confirmed. $YbMn_2Ge_2$ is a planar antiferromagnet below $T_N = 510$ K and undergoes a spin canting starting at $T_{N2} \sim 185$ K [3]. While no ordering of the Yb sublattice was detected, an unusual temperature dependence of the a lattice

parameter was attributed to changes in the Yb valence, opening the possibility that the Yb might not be purely divalent in the germanide. Analysis of the cell volume has also led to the suggestion that the Yb atoms may depart from a pure Yb^{2+} charge state, and a valence of ∼2.35 [4] was deduced.

$YbMn_2Si_2$ follows a similar pattern except that when the Mn moments order (with a slightly higher T_N of 526 K), they adopt an axial antiferromagnetic state with the Mn moments oriented parallel to the c axis [5]. Previous work attributed a second transition at ∼35 K to ordering of trivalent Yb moments [2]; however, this was not confirmed by neutron diffraction investigation which showed rather that this event corresponds to a rearrangement of the Mn moments into a cell-doubled antiferromagnetic state [5]. Ordering of the Yb moments was not seen to occur until somewhere between 10 and 1.5 K.

Only limited data exist on the composition dependence of magnetic ordering across the complete $YbMn_2Si_{2-x}Ge_x$ system. The evolution of the room temperature a lattice parameter with x shows a break near $x = 1.5$ suggesting that there is a change-over from Yb^{3+} to Yb^{2+} with increasing x [4]. More recent work reveals that there is also a switch from axial to planar Mn ordering around $x = 1.6$ [6]. For $x \leqslant 1.4$, magnetic scattering attributed to ordering of the Yb^{3+} moments was observed at 1.5 K. Samples with $x > 1.6$ showed a splitting of the (110) reflection which was attributed to coexisting Yb^{3+} and Yb^{2+} forms. The peak intensities were temperature dependent for $x = 1.6$, as the Yb^{3+} form replaces the Yb^{2+} form on heating, leading to an entirely trivalent compound by ∼150 K.

Two issues have clearly emerged as a result of the previous measurements on the $YbMn_2Si_{2-x}Ge_x$ system, and both concern the behaviour of the Yb ions. First, there is the actual valence of the Yb. Indirect information from neutron scattering suggests that it is trivalent at $x = 0$ with a crossover to almost divalent near $x = 1.5$. Second, there are claims of magnetic ordering of the Yb^{3+} moments for $x \leqslant 1.4$; however, the deduced ordering temperature ($\geqslant$1.5 K) is somewhat high in view of the other rare earth alloys in this series. Indeed, scaling the transition temperatures of the alloys with Tb–Er using the de Gennes factor suggests that the transition temperature should pass through zero before reaching Tm. This scaling works for the RCo_2Si_2 and RNi_2Si_2 systems and does yield Yb ordering temperatures that are consistent with the experimental values (1.7 K in $YbCo_2Si_2$ and 0.75 K in $YbFe_2Si_2$ [7]).

^{170}Yb Mössbauer provides an independent and direct method for investigating both issues. It is extremely sensitive to valence, it can probe the magnetic environment of the Yb atoms through transferred hyperfine fields and, finally, it can provide a direct indication of Yb ordering.

2. Experimental methods

Samples were prepared from high-purity elements (Yb 99.9%, Mn 99.99%, Si 99.999% and Ge 99.999%). The germanide ($x = 2$) was prepared by conventional argon-arc melting. For the other alloys, the metals were first powdered, mixed and then pressed into a pellet before arc melting. In order to compensate for evaporative losses, 17% excess Yb and 3% excess Mn were added for Si-rich samples, decreasing to 11% excess Yb and 2% excess Mn for Ge-rich samples. Many of the samples used here were prepared specifically for this ^{170}Yb Mössbauer study; however, others ($x = 0, 0.6, 1.0$ and 1.6) were also used previously for neutron diffraction studies [3, 5].

The 10 mCi ^{170}Tm sources were prepared by neutron activation of ∼25 mg of Tm as a 10 wt% alloy in aluminium. The source and sample were mounted vertically in a helium-flow cryostat and the drive was operated in sine mode. The 84.25 keV γ-photons used for the ^{170}Yb Mössbauer study were isolated from the various x-rays emitted by the source using a high-purity

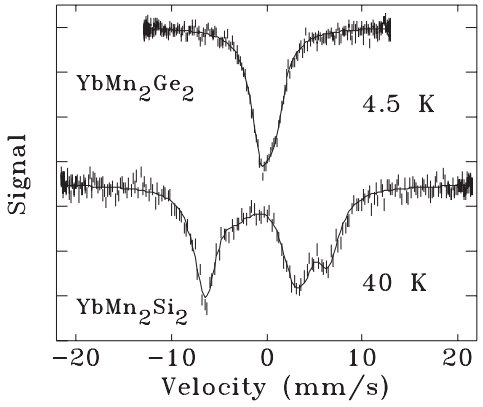

Figure 1. ^{170}Yb Mössbauer spectra of $YbMn_2Ge_2$ at 4.5 K (top) and $YbMn_2Si_2$ at 40 K (bottom) showing the clear difference in appearance between divalent Yb in the germanide and the trivalent Yb in the silicide.

Ge detector. Calibration of the spectrometer was achieved using a laser interferometer mounted on the back of the drive. Velocities were cross-checked against ^{57}Co/α-Fe at room temperature. This procedure has also been checked at velocities higher than those employed here by recording and fitting the ^{166}Er Mössbauer spectrum of $ErFe_2$ at 5 K [8]. We observed a ^{170}Yb linewidth of 1.16(5) mm s^{-1} with the source and a standard sample of YbB_6 at 4.5 K. Spectra were fitted using a non-linear least-squares minimization routine. Line positions and intensities were derived from an exact solution to the full Hamiltonian. Isomer shifts are quoted relative to ^{170}Yb in the $TmAl_2$ source. For $x \leqslant 0.8$ approximately 10% of the Yb was found to be present as Yb_2O_3 probably produced during the powdering of the ytterbium metal. Lesser amounts of this oxide have also been detected in these samples by neutron diffraction [4–6]. As the ordering temperature of Yb_2O_3 is 2.3 K [9], its magnetic behaviour is not expected to affect any of the conclusions reached here. The Yb_2O_3 was included in the fits with parameters constrained to those derived from measurements on a pure Yb_2O_3 reference sample.

The 84.25 keV Mössbauer transition in ^{170}Yb involves a 2+ $\rightarrow$ 0+ transition [10]. The 0+ ground state has neither a quadrupole moment nor a magnetic moment, so all hyperfine interactions involve the excited state. Furthermore, as the 2+ state is a rotational excited state, the nuclear size change is minimal and isomer shifts are small. The presence of an axially symmetric electric field gradient (efg) partially lifts the degeneracy of the excited state ($\pm 2, \pm 1$ and 0) yielding three hyperfine transitions with intensity ratios 2:2:1 (see $YbMn_2Si_2$ at 40 K in figure 1), while departures from axial symmetry ($\eta \neq 0$) fully lift the degeneracy and lead to a five-line pattern [10]. Similarly, a pure magnetic hyperfine field (B_{hf}) splits the excited state into five, equally spaced, levels and the Mössbauer spectrum consists of five equal-intensity lines with a uniform spacing. Including the effects of an efg and $\eta \neq 0$ demands a full solution of the nuclear Hamiltonian and leads to complex changes in both line positions and intensities that also depend on the relative orientations of the efg and B_{hf} axis systems. For most of the spectra presented here, B_{hf} and the efg are of comparable magnitude and no simple description of the observed pattern is possible.

3. Results

3.1. Valence

The first issue to be dealt with is the valence of the Yb ions in the two end-member compounds ($x = 0, 2$). While the magnetic structure of $YbMn_2Ge_2$ changes from layered planar to a canted structure below $\sim$185 K [3], the magnetic environment of the Yb atoms is such that no

net transferred field is expected at any temperature. By contrast, the change in the Mn moment arrangement in $YbMn_2Si_2$ at ∼35 K [5] should lead to a substantial transferred hyperfine field at the Yb site. We therefore start by examining the valence by comparing spectra in magnetically simple states.

The ^{170}Yb Mössbauer spectrum of $YbMn_2Ge_2$ at 4.5 K shown in figure 1 is characteristic of divalent Yb. It exhibits a slightly asymmetric line reflecting a small ($+3.5 \pm 0.1$ mm s^{-1}) quadrupole splitting (eQV_{zz}), essentially zero isomer shift (-0.04 ± 0.02 mm s^{-1}) and no hyperfine magnetic field. While the isomer shifts are always small for ^{170}Yb, the value found here in $YbMn_2Ge_2$ is typical of divalent Yb in a metallic environment [11]. Furthermore, the small quadrupole splitting is similar to that seen in other divalent Yb alloys such as the Yb–Cd system [12] and suggests that the 4f shell is full. There is no indication of additional quadrupole splitting that would be associated with even a partial hole in the 4f shell (removing one electron from the 4f shell without changing the crystal structure leads to a ∼28 mm s^{-1} change in the quadrupole splitting—see $YbMn_2Si_2$ below); we therefore find no evidence to support the valence of 2.35 inferred from the cell volume in recent neutron diffraction work [4] and conclude that the Yb ions are fully divalent in $YbMn_2Ge_2$ at 4.5 K.

By contrast, $YbMn_2Si_2$ at 40 K (figure 1) has a substantial and well-resolved quadrupole splitting of -25.6 ± 0.2 mm s^{-1}, and a small, positive, isomer shift (0.26 ± 0.04 mm s^{-1}), typical of trivalent Yb in a metallic environment [11]. No transferred hyperfine field from the Mn order is expected at the Yb sites as they lie between oppositely directed sheets of Mn moments and their contributions should cancel. Inclusion of a small asymmetry parameter ($\eta = 0.19 \pm 0.03$) leads to a slight improvement in the fit quality without changing the derived quadrupole splitting (within error). While $\eta \neq 0$ cannot be justified on the basis of the point symmetry of the Yb site in the $I4/mmm$ structure, a similar observation has been reported for the isostructural compound $YbFe_2Si_2$ [7].

Yb is clearly trivalent in $YbMn_2Si_2$ and divalent in $YbMn_2Ge_2$. These conclusions, reached here on the basis of ^{170}Yb Mössbauer data, are fully consistent with recent results of both electronic structure calculations and XPS measurements on these same compounds [13].

The boundary between the trivalent and divalent Yb forms is not expected to be sharp, as the neutron scattering suggested that the Yb^{3+} and Yb^{2+} forms coexist for $x > 1.6$ [6]. The spectra of $YbMn_2Si_{2-x}Ge_x$ taken at 4.5 K (figure 2) exhibit two clear trends with increasing x: (i) the hyperfine field increases up to $x = 1.2$ before disappearing (this is dealt with below); and (ii) a central unsplit component develops for $x \geqslant 1.4$. This central component is due to divalent Yb and it clearly coexists with the trivalent form over a wide composition range at 4.5 K. Mapping spectral areas onto the two valence fractions is not necessarily automatic. Europium also changes from trivalent to divalent in $EuMn_2Si_{2-x}Ge_x$ with increasing x [14], but the two forms exhibit very different recoil-free fractions in the ^{151}Eu Mössbauer spectra as a result of differing binding. The temperature dependence of the spectral areas in $EuCo_2Si_{2-x}Ge_x$ was used to deduce very different Debye temperatures for the two valence forms ($\Theta_D = 175$ K for Eu^{2+} while it is 250 K for Eu^{3+}) [15]. We have found that this difference in Debye temperature leads to a factor of two difference between the Eu-mass normalized absorption areas at room temperature in $EuMn_2Si_2$ and $EuMn_2Ge_2$. Examination of the composition dependence of the Yb-mass normalized spectral areas at 4.5 K in the ^{170}Yb Mössbauer spectra of $YbMn_2Si_{2-x}Ge_x$ revealed no statistically significant trend; we therefore conclude that mapping fractional spectral areas onto the fractions of the two Yb valence states is a valid procedure in this system. Figure 3 shows the results of this analysis. The Yb^{2+} component clearly develops above a threshold of $x = 1.15(5)$, and coexists with the trivalent form. By $x = 2$, only divalent Yb is present. We found some evidence for a decline in the Yb^{3+} component on heating for $x = 1.6$. This is consistent with neutron diffraction data [6]; however, our

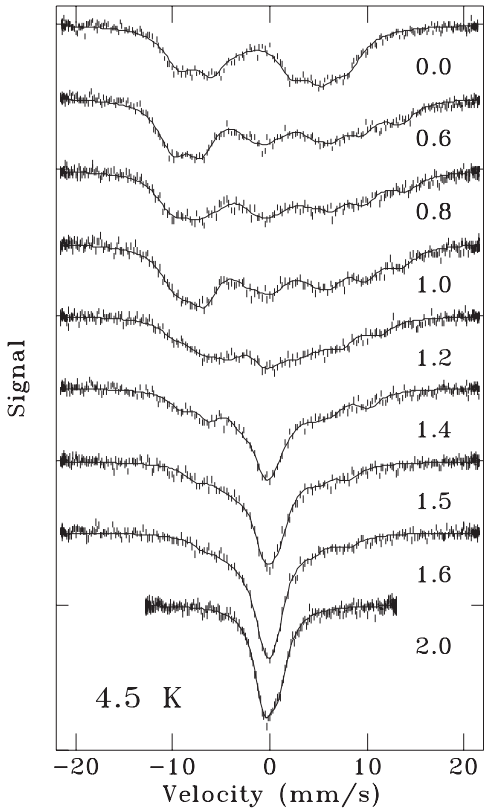

Figure 2. ^{170}Yb Mössbauer spectra of $YbMn_2Si_{2-x}Ge_x$ at 4.5 K showing the evolution of magnetic order with increasing x and the appearance of a Yb^{2+} component for $x \geqslant 1.4$.

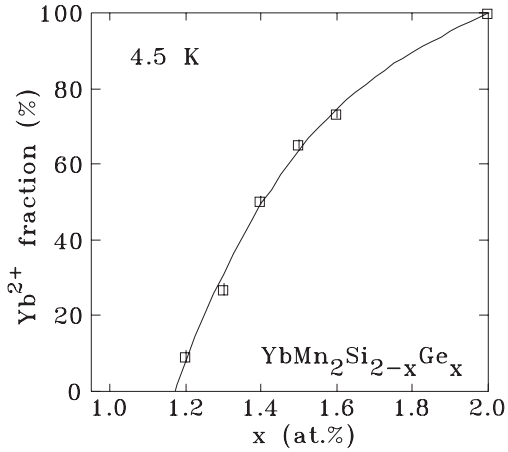

Figure 3. The composition dependence of the Yb^{2+} component in the ^{170}Yb Mössbauer spectra of $YbMn_2Si_{2-x}Ge_x$ at 4.5 K showing the onset of divalent Yb at $x = 1.15(5)$. The solid curve is a guide to the eye.

maximum temperature is limited to 60 K by the loss of a recoil-free fraction and no stronger confirmation was possible.

3.2. Magnetic order

The 35 K transition in $YbMn_2Si_2$ was originally attributed to ordering of Yb^{3+} moments [2], but more recent neutron diffraction work identified it with a doubling of the Mn magnetic cell

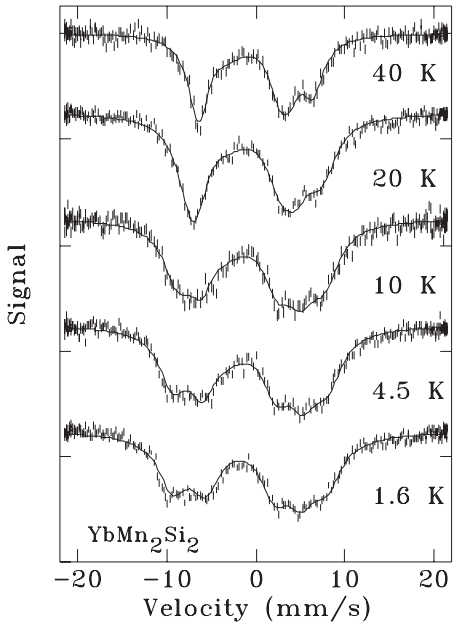

Figure 4. ^{170}Yb Mössbauer spectra of $YbMn_2Si_2$ at various temperatures.

and suggested that Yb^{3+} ordering occurred somewhere below 10 K [5]. However, the situation remains confused, with Szytuła *et al* continuing to attribute the 35 K event to ordering of Yb^{3+} moments [13]. The cell-doubled $+\ -\ -\ +$ magnetic structure for the Mn moments below 35 K proposed by Hofmann *et al* on the basis of neutron diffraction data [5] leads to two equal-population *magnetically* inequivalent Yb sites. One Yb site lies between the antiferromagnetically coupled $+-$ Mn layers and should continue to experience zero transferred hyperfine field, while the other Yb site is surrounded by eight ferromagnetically coupled $\sim 2\ \mu_B$ Mn moments in the $--$ layers and should exhibit a substantial magnetic splitting. Figure 4 shows the temperature dependence of the ^{170}Yb Mössbauer spectra of $YbMn_2Si_2$. The quadrupole triplet broadens and develops a significant hyperfine field by 1.6 K. It is thus clear that some magnetic splitting develops on cooling below 40 K, but there is no obvious change that could be attributed to ordering of Yb^{3+} moments below 10 K. Furthermore, the spectra are well fitted using only a *single* magnetic pentet and cannot be fitted as a sum of two equal-area components, one with the parameters derived from the 40 K spectrum and one with an additional magnetic splitting. The best fit for each spectrum (shown as solid curves in figure 4) was obtained with eQV_{zz} and η constrained to their 40 K values and the angle (θ) between the principal axis of the efg and B_{hf} at 45(5)°. The deduced hyperfine magnetic field is plotted versus temperature for two different samples of $YbMn_2Si_2$ in figure 5. The onset of magnetic splitting is clearly consistent with the transition at 35 K; however, there is no break in slope that could be attributed to an additional Yb ordering at a lower temperature. The ^{170}Yb Mössbauer results in figures 4 and 5 appear therefore to be inconsistent with both the proposed cell-doubled $+\ -\ -\ +$ magnetic structure for the Mn moments below 35 K, and with ordering of the Yb^{3+} moments below 10 K [5].

Neutron diffraction data suggested that the Yb moments order somewhere below 10 K, although only a small ($0.6 \pm 0.1\ \mu_B$) moment was observed by 1.5 K [5]. The maximum observed hyperfine field in figure 5 is only 35(2) T, well below the free-ion value of 416 T [16], and would be consistent with a Yb moment of about $0.3\ \mu_B$, half that inferred from neutron diffraction [5] at 1.5 K. Furthermore, for this field to be due to ordered Yb moments, the onset

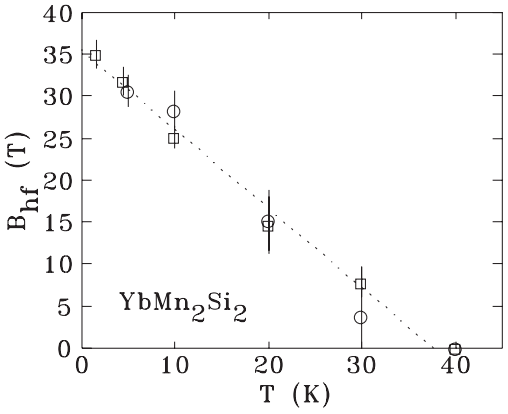

Figure 5. The temperature dependence of the hyperfine field (B_{hf}) for two samples of $YbMn_2Si_2$: □, a sample made for this study; ○, data from the same sample as was used by Hofmann *et al* [5].

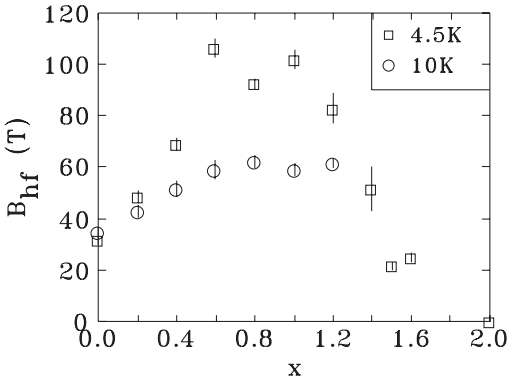

Figure 6. The composition dependence of the hyperfine field (B_{hf}) in $YbMn_2Si_{2-x}Ge_x$: ○, data at 10 K; □, data at 4.5 K.

of this order would have to be about 35 K (figure 5) which would be inconsistent with this transition being due to the Mn reorientation. As noted above, ordering of Yb^{3+} moments in these compounds is not likely to occur above a few kelvins at most, so Yb ordering at 35 K can be ruled out as the possible origin of the hyperfine field. With no apparent change in the spectral linewidth on cooling, we can also rule out contributions from slow electronic relaxation effects. The magnetic splitting therefore comes from changes in the Mn order alone. Unfortunately, our observation of a single Yb site with a modest transferred hyperfine field is inconsistent with the proposed cell-doubled $+\ -\ -\ +$ Mn structure [5], as is the slow increase in hyperfine field observed on cooling: the formation of a cell-doubled structure should cause an abrupt change in the ^{170}Yb Mössbauer spectrum as half of the Yb atoms suddenly lie between two layers of ferromagnetically coupled Mn moments. The trends in the hyperfine field would appear more consistent with a gradual canting of the Mn moments on cooling through 35 K and it would be interesting to see whether the neutron diffraction data could be reinterpreted so as to remove the conflict with the Mössbauer results.

Adding Ge to the alloy initially leads to an increase in the hyperfine field at the Yb site (figure 6). This trend reverses beyond $x = 1.2$ as divalent Yb starts to appear. Only a modest increase in transferred field is observed at 10 K and this is most probably associated with an increase in the canting of the Mn moments with increasing x. However, there is also a striking increase in the Yb hyperfine field on further cooling for $0.4 \leqslant x \leqslant 1.2$ and fields of order 100 T develop by 4.5 K. This increase is shown for $x = 0.6$ in figure 7 where both the hyperfine field ($B_{hf}(T)$) and the ac susceptibility ($\chi'(T)$) are plotted. It is clear that two distinct ordering

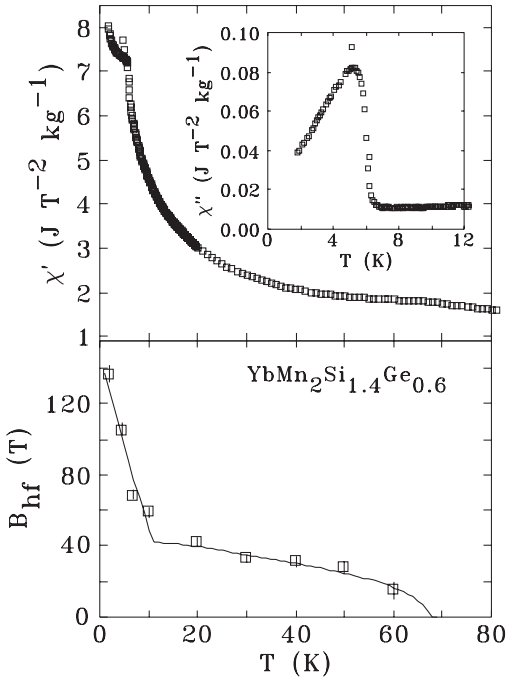

Figure 7. Top: susceptibility (χ') versus temperature (the inset shows the out-of-phase signal (χ'')); and bottom: the temperature dependence of the hyperfine field ($B_{\rm hf}$) for $YbMn_2Si_{1.4}Ge_{0.6}$. The onset of magnetic splitting at ∼65 K is associated with a weak feature in the $\chi_{\rm ac}$ data, while the marked increase below 10 K corresponds to peaks in both χ' and χ'' data.

events occur and that they are noted by both techniques. The upper one at 65 K appears to be a continuation of the 35 K transition in $YbMn_2Si_2$ and leads to a similar extrapolated ^{170}Yb field of about 40 T. The lower transition causes a further 100 T increase in the hyperfine field by 1.6 K which would correspond to a Yb^{3+} moment of about 1 $\mu_{\rm B}$ if it were due to ordering of the ytterbium. Alternatively, the lower transition could reflect a substantial change in the Mn ordering. While the derived Yb moment is quite reasonable, the transition temperature (6.0(3) K) appears to be too high to be due to Yb–Yb exchange alone.

An Arrott analysis of magnetization curves yielded no evidence of a significant spontaneous ferromagnetic moment in any of these materials—the magnetic response being dominated by that of the large, antiferromagnetically coupled, Mn moments. However, we did observe a small remanence at low temperatures in the Si-rich alloys. For $0.4 \leqslant x \leqslant 1.4$ there was a marked increase in the remanence below ∼6 K (figure 8), and while the onset temperature appeared to be essentially composition independent, the magnitude of the remanence exhibited a clear maximum for $x = 0.8$ (figure 8). The maximum moment associated with this remanence is only ∼0.08 $\mu_{\rm B}$/fu at $x = 0.8$, and could easily be dismissed as being due to a small concentration of a magnetic impurity. However, the remanence peak in the right panel of figure 8 corresponds well with the excess hyperfine field that is found to develop below 10 K (figure 6). Furthermore, the onset of this remanence on cooling matches the temperature at which the marked increase in $B_{\rm hf}$ occurs in the ^{170}Yb Mössbauer data (see for example figure 7). We therefore conclude that the increased remanence does indeed reflect a change in the bulk magnetic order in these alloys.

The magnetic transitions in the $YbMn_2Si_{2-x}Ge_x$ system are summarized in figure 9. We were able to track the upper transition both via the onset of magnetic splitting in the ^{170}Yb

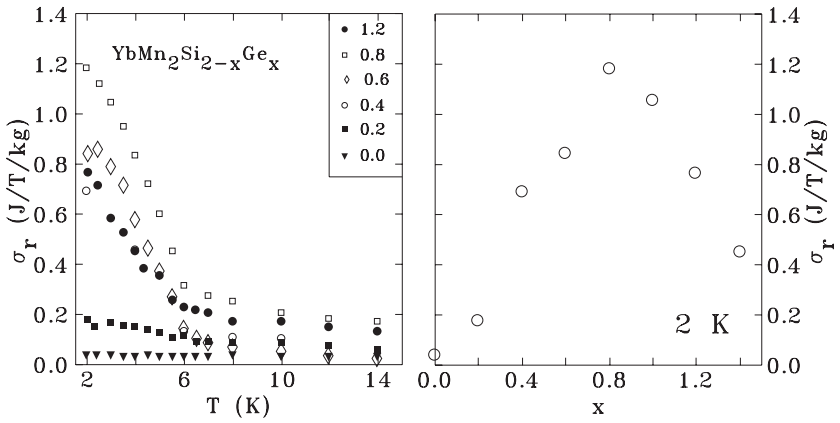

Figure 8. Left: remanence versus temperature for $YbMn_2Si_{2-x}Ge_x$ following magnetization in a field of 5 T. Right: the composition dependence of the remanence at 2 K.

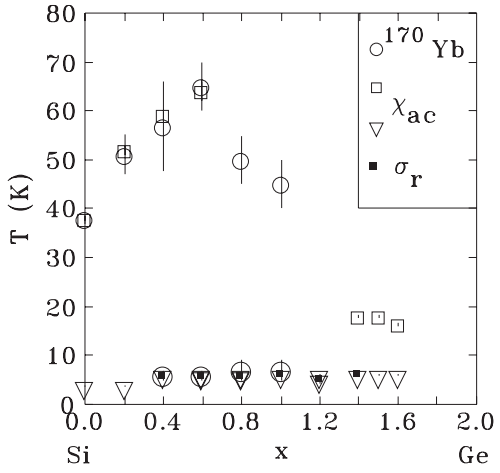

Figure 9. Magnetic transition temperatures for $YbMn_2Si_{2-x}Ge_x$ derived from ^{170}Yb Mössbauer spectroscopy, the ac susceptibility and the remanence.

Mössbauer spectrum and via the feature in $\chi'(T)$ only as far as $x = 0.6$. As can be seen in figure 7, the susceptibility signature is only a weak inflection and we could not locate it reliably for $x > 0.6$. Beyond this composition ($0.6 \leqslant x \leqslant 1.0$) only the onset of magnetic splitting in the ^{170}Yb Mössbauer data could be used. However, where both the signatures could be seen, they were in perfect agreement. The lower transition appears to be largely independent of composition and was seen in both $\chi'(T)$ and $\chi''(T)$ for $0 \leqslant x \leqslant 1.6$. While it is possible that for some compositions this susceptibility signal could reflect the presence of a minor impurity phase, ^{170}Yb Mössbauer spectroscopy shows that for $0.4 \leqslant x \leqslant 1.0$ the transition is definitely a bulk event, involving all of the Yb present in the sample. Furthermore, ^{170}Yb Mössbauer, remanence and χ' studies all yield consistent values for the transition temperature. The upper transition clearly occurs for $x \leqslant 1.0$, while the lower event has been detected by two or more techniques for $0.4 \leqslant x \leqslant 1.4$. Neither bulk magnetization nor ^{170}Yb Mössbauer data provide any support for the presence of the ∼5 K event detected via χ' for $x < 0.4$. This could reflect the greater sensitivity of χ', or an undetected impurity.

Given the inconsistency between the Mn magnetic structure derived from neutron scattering [5] and the single magnetic Yb site observed by ^{170}Yb Mössbauer spectroscopy

below the upper (35–65 K) transition, it is difficult to proceed to a clear identification of the lower event at ∼6 K. Not only is the observed hyperfine field at 1.6 K well below the free-ion value, it is also significantly less than that observed in the structurally identical $YbCo_2Si_2$ and $YbFe_2Si_2$ alloys [7]. At 6.0(3) K, the transition temperature is far higher than expected from either a simple de Gennes scaling or comparison with the isostructural Fe and Co based alloys. It therefore seems likely that this lower event reflects a further change in the Mn ordering, and that any possible ordering of the Yb moments takes place at a still lower temperature.

4. Conclusions

^{170}Yb Mössbauer spectroscopy confirms that Yb is divalent and non-magnetic in $YbMn_2Ge_2$, with no indication of a departure from a pure Yb^{2+} charge state. By contrast, only trivalent Yb is seen in $YbMn_2Si_2$. Divalent Yb appears in $YbMn_2Si_{2-x}Ge_x$ at $x = 1.15(5)$ and coexists with trivalent Yb as it gradually replaces it with increasing x.

Two bulk magnetic transitions have been identified. The upper one is strongly composition dependent and ranges from 35–65 K for $x \leqslant 1.0$ and reflects a canting of the Mn moments. The lower transition is seen in susceptibility for all $x \leqslant 1.2$, but is only confirmed as a bulk event for $0.4 \leqslant x \leqslant 1.2$. The observed hyperfine field and transition temperature suggests that both events reflect reorganization of the Mn order rather than ordering of Yb^{3+} moments.

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