SATURATION MAGNETIZATION IN THE ANOMALOUS FERROMAGNET, \((Y, U)B_4\)

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For the \((Y, U)B_4\) system, long-range ferromagnetism only occurs for \(0.05 < x < 0.55\). The anomalous magnetic phase diagram has been attributed to a delocalization of the \(U\) 5f-electrons due to increasing f-f overlap as the average \(U-U\) separation is varied. Measurements of the saturation magnetic moment versus \(x\) in the ferromagnetic region and measurements of the lattice constants versus \(x\) are presented.

The magnetic to nonmagnetic transition seen in most \(U\)-based alloys and intermetallic compounds is a result of the delocalization of the f-electrons due to f-f overlap and/or f-spd hybridization. H.H. Hill established that f-f overlap significantly contributes to this delocalization for \(U-U\) separations less than 3.4–3.6 Å, whereas f-spd hybridization tends to dominate for larger \(U\)-separations [1]. The \(U-U\) separation in \(UB_4\) is 3.7 Å which is slightly larger than Hill’s critical separation and \(UB_4\) is weakly paramagnetic, presumably due to the delocalization of the f-electrons caused by f-f overlap. Upon dilution of \(UB_4\) by \(YB_4\), an anomalous magnetic phase diagram is obtained. Previously, it was reported that the \((Y, U)B_4\) system was paramagnetic for \(x > 0.6\), ferromagnetic for \(0.1 < x < 0.6\) and paramagnetic for \(x < 0.1\) [2]. Also, it has been shown that the variations of the lattice constants [3], hyperfine field [4], and paramagnetic susceptibility [5] versus \(x\) are consistent with a two-site model. This model assumes that the 5f electrons associated with \(U\) ions having 4 or less \(U\) nearest neighbours (nn) become localized and develop a local magnetic moment, whereas those with more than 4 nn remain weakly paramagnetic. We have measured the lattice constants versus \(x\) and the saturation magnetization versus \(x\) and \(T\) for \((Y, U)B_4\). The variation of the lattice constants with \(x\) is consistent with those previously published [3] and the saturation magnetization dependence on \(x\) mirrors the variation of the Curie temperature, \(T_c\), with \(x\).

The samples were prepared in a conventional inert atmosphere arc furnace. Appropriate amounts of \(Y\) and \(U\) were added to compensate for the slight evaporation of these more volatile constituents which occurred during melting. The lattice constants were measured using a Siemens D/θ diffractometer and the magnetization was measured using a commercial vibrating sample magnetometer.

Both \(YB_4\) and \(UB_4\) crystallize in the tetragonal ThB₄ structure [2]. Shown in fig. 1 is the variation of the lattice constants, \(a\) and \(c\), versus \(x\). These results are very similar to those previously reported by Hill et al. [3]. Note the clear departure in the vicinity of \(x = 0.45\) from a linear Vegard’s law for both \(a\) and \(c\). This departure from the initial linear dependence of \(a\) and \(c\) for \(x > 0.45\) has been attributed to a delocalization of the 5f electrons due to increasing f-f overlap as the average \(U-U\) separation is reduced with increasing \(x\). Such behavior is commonly seen in Ce-based alloys and intermetallic compounds [6] and was also reported for \((U, Y)Sb\) [7].

Fig. 1. Lattice constants versus \(x\) for the tetragonal system \((Y_{1-x}U_x)B_4\).

Fig. 2. Curie temperature and saturation magnet moment versus \(x\) for \((Y_{1-x}U_x)B_4\).
Shown in fig. 2 by the dashed curve is the U-concentration dependence of $T_c$, the ferromagnetic Curie temperature. The $T_c$ versus $x$ behavior shown in fig. 2 was determined from an Arrott plot analysis of the field and temperature dependence of the magnetization. The $T_c$ versus $x$ behavior shown in fig. 2 is similar to the behavior previously reported by Giorgi et al. [2] with the exception that our ferromagnetic-paramagnetic phase boundary is shifted slightly to lower $x$-values. The maximum $T_c$ of 14.5 K is consistent with the previous measurements. For $x > 0.3$ the rapid depression of $T_c$ with increasing $x$ has been attributed to a quenching of the local moments due to the delocalization of the 5f electrons caused by increasing f-f overlap. Note that this rapid depression of $T_c$ with increasing $x$ occurs in the region where the delocalization as seen in the lattice constants becomes apparent.

Shown in fig. 3 is an Arrott plot [8] of the field and temperature dependence of the magnetization for $x = 0.25$. From such a plot both the temperature dependence of the saturation magnetization and $T_c$ can be determined. Shown in fig. 2 by the solid curve is the zero temperature saturation moment/U-ion, $\mu_0$, versus $x$, as determined from the extrapolation of the temperature dependence of the saturation magnetization. Note, the U-concentration dependence of $\mu_0$ qualitatively resembles that seen for $T_c$ versus $x$. For a local moment model without crystalline electric field (CEF) effects, $\mu_0$ should be nearly independent of $x$. Qualitatively, the observed dependence of $\mu_0$ and $T_c$ can be explained with a local moment model assuming CEF effects with a $J = 4$ 5f configuration and a nonmagnetic singlet ground state. Using a two site model and assuming the exchange and CEF parameters are independent of $x$, then an appropriate set of parameters can be selected such that the mean field $T_c$ goes to zero at $x = 0.1$ and $x = 0.8$ with the maximum occurring near $x = 0.5$. Such behavior only qualitatively reproduces the observed behavior of $T_c$ versus $x$.

An alternate explanation of these results may be available in an itinerant model with the variation in the lattice constants reflecting the delocalization of the f-electrons in much the same way as occurs in the $\gamma$-\alpha transition in Ce [9]. As shown by Pickett et al., a slight increase of the f-f overlap can account for the isostructural transitions and lattice collapse in Ce. Similarly, the increase of f-f overlap and lattice pressure with increasing U-concentration could result in a localized–itinerant transition in (Y, U)B$_4$. With an itinerant model, the $T_c$ versus $x$ could be qualitatively accounted for using a Stoner model [10]. Furthermore, the approximate scaling of $\mu_0$ with $T_c$ and the reduced size of $\mu_0$ as compared to that expected for a well localized magnetic system can be easily obtained with an itinerant theory of magnetism.

Measurements of the magnetization versus temperature and magnetic field up to 9 T, along with measurements of the pressure dependence of $T_c$ and $\mu_0$ are presently being pursued with the hope of establishing which model is more appropriate.

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