Title
Saturation magnetization in the anomalous ferromagnet, (Y, U)B4

Permalink
https://escholarship.org/uc/item/47p1d4pg

Authors
Wallash, A
Crow, JE
Fisk, Z

Publication Date
1986-02-01

DOI
10.1016/0304-8853(86)90703-1

License
CC BY 4.0

Peer reviewed
SATURATION MAGNETIZATION IN THE ANOMALOUS FERROMAGNET, (Y, U)B₄

A. WALLASH, J.E. CROW and Z. FISK

Physics Department, Temple University, Philadelphia, PA 19122, USA

For the (Y, U)B₄ 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 UB₄ is 3.7 Å which is slightly larger than Hill’s critical separation and UB₄ is weakly paramagnetic, presumably due to the delocalization of the f-electrons caused by f-f overlap. Upon dilution of UB₄ by YB₄, an anomalous magnetic phase diagram is obtained. Previously, it was reported that the (Y₁₋ₓ,Uₓ)B₄ 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 (Y₁₋ₓ,Uₓ)B₄. 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, Tₐ, 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 3θ/2θ diffractometer and the magnetization was measured using a commercial vibrating sample magnetometer.

Both YB₄ and UB₄ 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, (Y₁₋ₓ,Uₓ)B₄.

Fig. 2. Curie temperature and saturation magnet moment versus x for (Y₁₋ₓ,Uₓ)B₄.
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 Arrrott 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 Arrrott 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 $(J = 4)$ f 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 $\gamma$-Ce 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.