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

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For the $(Y_{1-x}U_x)B_4$ 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 obained. Previously, it was reported that the $(Y_{1-x}U_x)B_4$ 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 $(Y_{1-x}U_x)B_4$. 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 occured during melting. The lattice constants were measured using a Siemens $2\theta/\theta$ diffractometer and the magnetization was measured using a commercial vibrating sample magnetometer.

Both YB_4 and UB_4 crystallize in the tetragonal ThB_4 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].

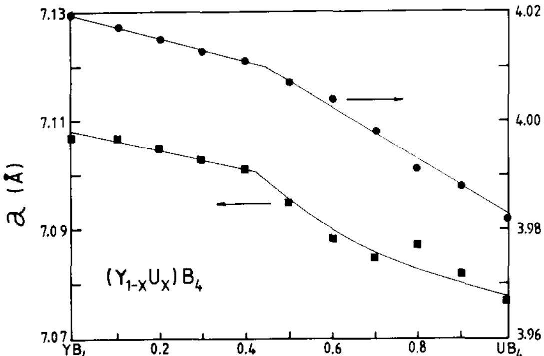

Fig. 1. Lattice constants versus x for the tetragonal system. $(Y_{1-x}U_x)B_4$.

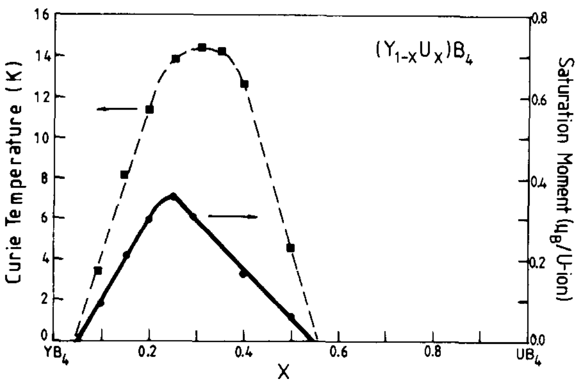

Fig. 2. Curie temperature and saturation magnet moment versus x for $(Y_{1-x}U_x)B_4$

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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, μ_0, versus x, as determined from the extrapolation of the temperature dependence of the saturation magnetization. Note, the U-concentration dependence of μ_0 qualitatively resembles that seen for T_c versus x. For a local moment model without crystalline electric field (CEF) effects, μ_0 should be nearly independent of x. Qualitatively, the observed dependence of μ_0 and T_c can be explained with a local moment model assuming CEF effects with a

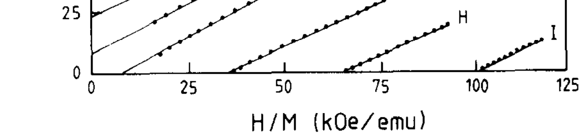

Fig. 3. M^2 versus H/M (Arrott Plot) at various temperatures for $(Y_{0.75}U_{0.25})B_4$

($J = 4$) 5f configuration and a nomagnetic 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 $x \approx 0.1$ and $x \approx 0.8$ with the maximum occurring near $x \approx 0.5$. 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 γ-α 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 μ_0 with T_c and the reduced size of μ_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 μ_0 are presently being pursued with the hope of establishing which model is more appropriate.

[1] H.H. Hill, Plutonium 1970 and other Actinides ed. W.N. Miner (The Metallurgical Society of the AIME, New York, 1970) p. 2.

[2] A.L. Giorgi, E.G. Szklarz, R.W. White and H.H. Hill, J. Less-Common Metals 34 (1974) 348.

[3] H.H. Hill, A.L. Giorgi, E.G. Szklarz and J.L. Smith, J. Less-Common Metals 38 (1978) 239.

[4] E. Fukushima, V.O. Struebing and H.H. Hill, J. Phys. Soc. Japan 39 (1980) 921.

[5] A. Wallash, J.E. Crow and Z. Fisk, J. Appl. Phys. 57 (1985) 3143.

[6] See, for example, Valence Instabilities, eds. P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982) J. Magn. Magn. Mat. 47&48 (1985).

[7] B. Frick, J. Schoenes, F. Hulliger and O. Vogt, Solid State Commun. 49 (1984) 1133.

[8] A. Arrott, Phys. Rev. 108 (1957) 1394.

[9] W.E. Pickett, A.J. Freeman and D. Koelling, Phys. Rev. B23 (1981) 1266.

[10] See for example: M. Shimizu, Rep. Prog. Phys. 44 (1981) 21.