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MAGNETIC SUSCEPTIBILITY AND SPECIFIC HEAT STUDY ON THE NONMAGNETIC TO MAGNETIC TRANSITION IN URh₃B_x ($0 \le x \le 1$) SYSTEM

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Abstract. – Lattice parameter, succeptibility, specific heat and resistivity measurements are presented on URh_3B_x ($0 \le x \le 1$). This alloy series exhibits a transition from an almost temperature independent paramagnet to a magnetic system. Results are interpreted in terms of U-5f Rh-4d dehybridization induced by boron addition.

One of the basic parameters dictating the properties of f-electron systems is the degree of hybridization of the f-electrons with the conduction band electrons [1]. The alloy system URh₃B_x with $0 \le x \le 1$ exhibits a transition from an itinerant paramagnet (x = 0) to an antiferromagnet (x = 1) in which the 5f hybridization effects can be studied in a continuous way as a function of x. URh₃ is a temperature independent paramagnet [2] where the 5f electrons are itinerant, while URh₃B exhibits a Curie-Weiss susceptibility and orders antiferromagnetically at 9.8 K [3] suggesting a more local nature of the 5f electrons.

URh₃ and URh₃B parents were prepared by arc melting in an argon atmosphre stoichiometric amounts of appropriate elements of higher than 99.9 % purity. X-ray patterns of both compounds showed single phase AuCu₃ structure with lattice parameters of 3.98 Å and 4.14 Å respectively in agreement with previous studies [2, 3]. Parents were then cut into few pieces, mixed in the appropriate ratios and remelted to form the final URh₃B_x alloys with $0 \le x \le 1$. No second phases were detected by X-ray diffraction.

Lattice parameter measurements are shown in figure 1a. The lattice parameter increases almost linearly up to $x \sim 0.8$ after which the lattice parameter stabilizes. As stated above, careful X-ray studies for x > 0.8 indicate that these samples are single phase. Thus, the nonlinearity of the x-dependence of the lattice constant reflects that boron addition not only expands the lattice but also changes the electronic structure of the matrix presimably by bonding with the Rh atoms [4] and by adding electrons to the conduction band. Similar behavior was found in all the LRh₃B_x where L =rare earth element [5].

Susceptibility measurements were done using a dc SQUID magnetometer in a external field of 10 kOe. The data (Fig. 2) was fitted to a Curie-Weiss susceptibility plus a constant, $\chi = \chi_0 + C/(T - \theta)$. In figure 1b we show the fitted values for the high temperature paramagnetic moment and the Curie-Weiss temperature θ . The susceptibility data shows a cusp (Fig. 2) for x = 1.0 and x = 0.9 at $T_N=9.8$ K and 6.8 K respectively. This magnetic anomaly is believed



Fig. 1. – a) Temperature of the magnetic anomaly $T_{\rm N}$ as determined from zero-field resistivity (circles) and susceptibility at 10 kOe (squares) and lattice parameter (triangles) as a function of boron concentration; b) high temperature paramagnetic moment $\mu_{\rm p}$ (circles) and Curie-Weiss paramagnetic temperature θ (triangles) as a function of boron concentration.

to be due to antiferromagnetic or spin glass-like ordering of the U ions. This speculation is supported by the fact that the high temperature paramagnetic moment, $\mu_{\rm p} = 3.2 \ \mu_{\rm B}$, is close to that of the free uranium $U^{2+,3+}$ ion ($\mu_{\rm p} = 3.6 \ \mu_{\rm B}$) and $T_{\rm N}$ decreases with field as expected for an antiferromagnet. The large negative values of θ point to strong hybridization of the f-electrons, which decreases with increasing x.

From figure 1 we may divide the alloy series into three different regions; $x \le 0.5$, 0.5 < x < 0.8 and $0.8 \le x$. In the first region, $x \le 0.5$, boron addition



Fig. 2. – Susceptibility as a function of temperature for various boron concentrations x = 0.0; 0.5; 0.6; 0.7; 0.8; 0.9 a,d 1.0.

expands the lattice but the 5f electrons remain essentially itinerant. In the second region, 0.5 < x < 0.8, boron addition expands the lattice even further but the 5f electrons start to dehybridize as indicated by the increase of $\mu_{\rm p}$ and θ . At x = 0.8, $\mu_{\rm p} = 3.2 \ \mu_{\rm B}$ is almost that of the free uranium atom $U^{2+,3+}$. In the third region these quantities stabilize and clear evidence for a magnetic transition is seen in $\chi(T)$.

A change in slope in the resistivity is seen at temperatures close to $T_{\rm N}$, as defined by the cusp in $\chi(T)$. This allows for an independent estimate of the magnetic ordering temperature shown in figure 1a. From there we see an increase in the transition temperature as a function of boron content with further stabilization at around x = 0.8. These results track the susceptibility ones and are further evidence of the dehybridization.

Finally, in figure 3 the specific heat C_p is plotted as C_p/T versus T^2 for URh₃B. The electronic contribution to C_p , i.e. γ , is enhanced by about one order of magnitude (from 14 mJ/mole-K² to 114 mJ/mole-K²). This increase in γ also points towards a dehybridization of the 5f electrons induced by boron addition. Surprisingly, there is no anomaly in the specific heat of URh₃B as would be expected for an antiferromagnetic system. This result shadows the interpretation of the magnetic anomaly as originated from an antiferromag-



Fig. 3. – Specific heat over temperature versus T^2 for URh₃ and URh₃B. γ values are 14 and 114 mJ/mole-K² for x = 0.0 and 1.0 respectively.

netic orderidng. Currently these studies are extended to include ac susceptibility, time-dependent magnetization and mössbauer measurements to check for spin glass-like behavior.

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