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## UNUSUAL MAGNETIC BEHAVIOR OF $\text{TmIr}_2$ and $\text{YbIr}_2$

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In contrast to the other magnetic Rare Earth (RE) ions for which the cubic Laves  $\text{REIr}_2$  compound is ferromagnetic,  $\text{YbIr}_2$  and  $\text{TmIr}_2$  are antiferromagnetic below 0.40 and 0.05 K, respectively. Both are trivalent above 4 K, with a possible reduced moment for  $\text{TmIr}_2$  below 0.3 K. In addition,  $\text{CeIr}_2$  and  $\text{LuIr}_2$  are superconductive below 0.20 and 0.23 K, respectively.

The predominant behavior of the magnetic Rare Earth (RE) $\text{Ir}_2$  (C15, cubic Laves phase) intermetallic compounds is local moment paramagnetism at high temperature and ferromagnetic order at low temperature, as reported by Bozorth et al. [1]. Curie points  $T_C$  were found to vary roughly as  $(g-1)^2J(J+1)$ , the De Gennes factor [2], and ordered moment values to fall between  $2S$  and  $gJ$ . Here  $S$  is the spin and  $J$  is the total quantum number, and  $g$  is the Landé  $g$  factor. The data for Eu-, Tm- and  $\text{YbIr}_2$  were questionable as pointed out by the authors of ref. [1] because of unusual magnetic behavior for  $\text{EuIr}_2$  and sample preparation difficulties for the other two compounds. The behavior of  $\text{EuIr}_2$  was resolved when Matthias, Fisk and Smith [3] reported superconductivity at 0.2 K, proving that here Eu is in its purely trivalent, nonmagnetic state.

To reexamine the properties of the often mixed-valent elements Yb and Tm in the  $\text{REIr}_2$  system, we have prepared single crystals by the flux growth technique using a Cu solvent. The crystals were all cubic Laves phase; lattice parameters of 0.74638(1) nm for  $\text{YbIr}_2$  and 0.74736(2) nm for  $\text{TmIr}_2$  were measured. Susceptibility measurements were made in a Faraday magnetometer over the range 230–4 K. A Curie-Weiss fit to the susceptibility  $\chi = (p_{\text{eff}}^2/8)/(T - \Theta)$  yields  $p_{\text{eff}} = 7.57\mu_B$  and  $\Theta = -4$  K for  $\text{TmIr}_2$  and  $p_{\text{eff}} = 4.49\mu_B$  and  $\Theta = -4$  K for  $\text{YbIr}_2$ , in good agreement with the trivalent free-ion values for the effective moments. The ac susceptibilities were measured from 4 to 0.012 K with arbitrary sensitivity, and therefore no low temperature effective moment values could be obtained. A constant was subtracted from the ac susceptibility to correct for a temperature-independent paramagnetic background. The constant  $\chi_{\text{ac}}$  (less than about 30% of the peak susceptibility) which gave the best straight line fit to  $1/\chi$  was employed in the analysis. For  $\text{YbIr}_2$  this

resulted in a  $\Theta$  of  $-0.3$  K, much smaller than the high temperature extrapolated data. In addition,  $\text{YbIr}_2$  has a cusp in the ac susceptibility at 0.42 K, indicating antiferromagnetic ordering; see fig. 1. Helping to confirm this assertion, we find that the cusp moves to lower temperatures in an applied magnetic field at the rate of  $-1.5$  K/T. The situation for  $\text{TmIr}_2$  is more complex. Below 4 K, a linear fit to  $1/\chi_{\text{ac}}$  extrapolates to  $\Theta \approx -1.4$  K, compared to  $\Theta = -4$  K from the high temperature data. In addition, at 0.3 K,  $\chi_{\text{ac}}$  has a discontinuity in the slope (steeper at lower temperature) followed by a cusp at 0.05 K, seen in fig. 2. The cusp moves to lower temperatures in a field at the rate of  $-2.5$  K/T. We have also grown single-crystal  $\text{CeIr}_2$  and  $\text{LuIr}_2$ ; these compounds are superconductive below 0.20 and 0.23 K with upper critical fields of 0.11 and 0.10 T, respectively.

The cubic Laves C15 structure with  $\text{REIr}_2$  appears

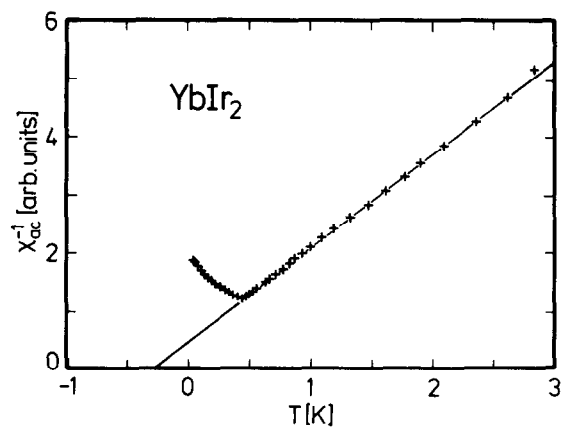


Fig. 1. Inverse ac susceptibility vs. temperature for  $\text{YbIr}_2$ . The line is only a guide to the eye.

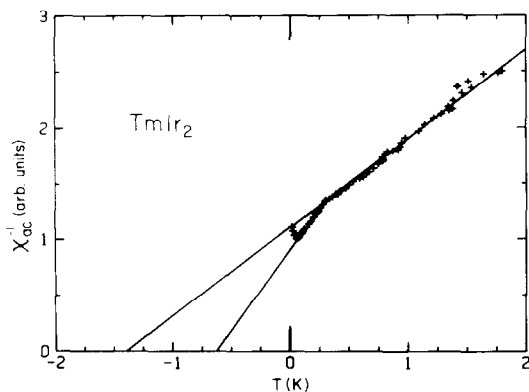


Fig. 2. Inverse ac susceptibility vs. temperature for  $\text{TmIr}_2$ . The solid lines are only a guide to the eye.

unfavorable for mixed valence formation, with the usual exception of  $\text{CeIr}_2$  which is reported to have a valence of 3.21 from  $L_{\text{III}}$  X-ray absorption measurements [4].  $\text{EuIr}_2$  is superconducting at 0.2 K and thus must be in its trivalent  $J = 0$  configuration. In contrast,  $\text{EuRh}_2$  is not trivalent; rather it is mixed valent as determined from  $^{151}\text{Eu}$  Mössbauer effect measurements [5] and is strongly paramagnetic at low temperatures, indicating a probable divalent state. Generally then, the nonmagnetic RE ions form superconductors in the  $\text{REIr}_2$  series; the magnetic RE ions order magnetically. Ferromagnetic order is the rule except for Yb- and  $\text{TmIr}_2$  which are antiferromagnetic with Néel temperatures much lower than the  $\Theta$  values; this may be due to crystal field effects.  $\text{TmIr}_2$  has an additional susceptibility feature below 0.3 K which may be interpreted as a moment reduction. This may be due to crystal-field effects, the onset of mixed valency or quadrupolar ordering as seen in  $\text{TmZn}$  slightly above a magnetic ordering temperature [6]. The very low temperatures at which these effects occur make further investigation most difficult; they also indicate that extremely small energies are responsible for this unusual behavior.

As a further note on the unusual properties of these materials, we comment on arc melted  $\text{Tm}_{0.2}\text{Ir}_{0.8}$ . This

composition forms a eutectic of Ir and  $\text{TmIr}_2$  as determined by X-ray diffraction. Susceptibility measurements on as-cast material yield  $p_{\text{eff}} = 7.44\mu_{\text{B}}$  and  $\Theta = -2.6$  K, consistent with a full, trivalent moment on the Tm ion. Superconductivity is observed by ac susceptibility techniques at 1.5–1.8 K. Annealing this sample (16 days,  $1000^\circ\text{C}$ ) destroys superconductivity above 0.1 K, the transition temperature of pure Ir. We speculate that one of two phenomena may be occurring: enhanced superconductivity [7] in Ir due to lattice mismatch with  $\text{TmIr}_2$  giving rise to a lattice expansion and softening in Ir, which has been shown to lack credibility [8]; or the first case of superconductivity in a binary compound of undetermined crystal structure containing a rare earth element carrying its full local moment at low temperatures. At present, more experimental work needs to be done to confirm either of these hypotheses.

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