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UNUSUAL MAGNETIC BEHAVIOR OF TmIr_2 and YbIr_2

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In contrast to the other magnetic Rare Earth (RE) ions for which the cubic Laves REIr_2 compound is ferromagnetic, YbIr_2 and TmIr_2 are antiferromagnetic below 0.40 and 0.05 K, respectively. Both are trivalent above 4 K, with a possible reduced moment for TmIr_2 below 0.3 K. In addition, CeIr_2 and LuIr_2 are superconductive below 0.20 and 0.23 K, respectively.

The predominant behavior of the magnetic Rare Earth (RE) 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 YbIr_2 were questionable as pointed out by the authors of ref. [1] because of unusual magnetic behavior for EuIr_2 and sample preparation difficulties for the other two compounds. The behavior of 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 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 YbIr_2 and 0.74736(2) nm for 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 TmIr_2 and $p_{\text{eff}} = 4.49\mu_B$ and $\Theta = -4$ K for 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 χ_{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 YbIr_2 this

resulted in a Θ of -0.3 K, much smaller than the high temperature extrapolated data. In addition, 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 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, χ_{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 CeIr_2 and 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 REIr_2 appears

Fig. 1. Inverse ac susceptibility vs. temperature for YbIr_2 . The line is only a guide to the eye.

Fig. 2. Inverse ac susceptibility vs. temperature for TmIr_2 . The solid lines are only a guide to the eye.

unfavorable for mixed valence formation, with the usual exception of CeIr_2 which is reported to have a valence of 3.21 from L_{III} X-ray absorption measurements [4]. EuIr_2 is superconducting at 0.2 K and thus must be in its trivalent $J = 0$ configuration. In contrast, EuRh_2 is not trivalent; rather it is mixed valent as determined from ^{151}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 REIr_2 series; the magnetic RE ions order magnetically. Ferromagnetic order is the rule except for Yb- and TmIr_2 which are antiferromagnetic with Néel temperatures much lower than the Θ values; this may be due to crystal field effects. 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 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 TmIr_2 as determined by X-ray diffraction. Susceptibility measurements on as-cast material yield $p_{\text{eff}} = 7.44\mu_B$ 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°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 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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