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Magnetism and heavy fermion-like behavior in the RBiPt series

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Members of the RBiPt (R = Ce–Lu with the exceptions of Pm and Eu) series have been grown as single crystals. Magnetic susceptibility and electrical resistance have been measured on all members of the series, and specific heat measurements have been performed on rcprcscntatives. The high temperature resistance uniformly changes from that of a small-gap semiconductor or semimetal seen in NdBiPt to that of a heavy-fermion metal seen in YbBiPt, which shows a linear coefficient of specific heat at low temperatures of 8 J/K^2 mole. Further, the lighter rare earth members show an unusually sharp increase in their resistance associated with antiferromagnetic ordering at low temperatures.

Recently, during attempts to grow single crystals of the $R_3Bi_4Pt_3$ series past the La, Ce, and Pr members that have been reported elsewhere,[1,2] we discovered that the synthesis technique being used favored the growth of RBiPt (R = rare earth) for R members beyond Pr. X-ray diffraction of powdered single crystals show that the RBiPt series forms in the cubic, AgAsMg structure, which can be viewed as three face-centered-cubic sublattices placed at (0,0,0), (1/4,1/4,1/4), and (3/4,3/4,3/4) along the body diagonal. Until now, all that has been reported about these compounds is an incomplete set of lattice parameters.[3–5] However, two other systems, UNiSn and MnNiSb (MnPtSb), in the same structure have been studied[6,7] in some detail because of their interesting "half-metallic" ferromagnetic properties.

Single crystals of RBiPt (R = Nd–Lu and Y with the exceptions of Pm and Eu) were grown out of Bi flux[8] and characterized by powder x-ray diffraction, four-probe electrical resistance and dc magnetic susceptibility measurements. In some cases the low temperature ($T < 20$ K) specific heat was also determined.

Figure 1 shows the temperature variation of the electrical resistance for representative members of the series normalized at room temperature to the resistance of NdBiPt. Above approximately 150 K, all members, except Yb and Lu, have a negative dR/dT, suggesting semiconductor or semimetal-like behavior. Indeed, a plot of ln R versus $1/T$ for NdBiPt is linear in the range of $130 < T < 300$ with a slope of $\Delta = 175$ K. Interestingly, the magnitude of dR/dT as seen in the high temperature resistance of these compounds (Fig. 1 inset) decreases monotonically as the rare earth series is traversed. This trend is reflected as well in the absolute magnitude of ρ(290 K) which ranges from 0.95 ± 0.05 mΩ cm for Nd, and 0.9 ± 0.2 mΩ cm for Gd to 0.35 ± 0.05 mΩ cm for Yb. These systematics suggest that the resistance may be dominated by variations in the unit cell volume. Lattice parameters a_0 for members of the series are shown in Fig. 2 and agree reasonably well with previously reported values.[3,4] As expected from the well-known lanthanide contraction, we see a monotonic decrease in a_0 in going from Ce to Lu. However, we note an apparently discontinuous drop in a_0 between Gd and Tb for which we have no explanation.[9] To test the assumption that a_0 dominates changes in the high temperature resistance, alloys $Nd_{0.5}R_{0.5}BiPt$ and YBiPt were studied as well. Their lattice parameters and temperature dependent resistances follow the systematics observed in the RBiPt compounds, as shown in Figs. 1 and 2.

At low temperatures anomalies appear in $R(T)$ for R = Nd, Sm (not shown in Fig. 1), Gd, Tb, and Dy. In all cases, distinct features in the magnetic susceptibility and specific heat coincide with the temperature at which these anomalies occur. As an example, we show in Fig. 3, the resistance, susceptibility and specific heat divided by temperature for GdBiPt. The temperature variation of χ and C/T near the anomaly at 9 K is characteristic of antiferromagnetic order.[10] The Neel temperatures, defined by these measurements, for the RBiPt compounds are given in Table I. Assuming a linear extrapolation of C/T from the lowest measured temperature (1.5 K) to $T = 0$ allows an estimate of the entropy S associated with the magnetic transition. In the case of Gd, we find $S \sim 0.8$R ln 8 between $T = 0$ and the peak in C/T, which is close to the expected R ln 8 entropy for ordering in the $J = 7/2$ multiplet. Entropy below T_N for Nd, Tb, and Dy (Table I) is considerably less than the R ln$(2J + 1)$ expected for full Hund's rule J multiplet, implicating partial lifting of the f-ground state degeneracy.

Above approximately 100 K, the inverse magnetic susceptibility $1/\chi$ is linear in temperature for all members of the series except Sm which probably has a nontrivial Van Vleck contribution to its susceptibility. From these data, values of the effective moment μ_{eff} and paramagnetic θ may be determined. Results are summarized in Table I where we see that μ_{eff} is very close to that expected from Hund's rules for the trivalent rare-earth ions. In alloyed crystals $Nd_{0.5}R_{0.5}BiPt$, the effective moment per mole of rare earth falls between that of Nd and the R ion. The negative paramagnetic θ's argue for antiferromagnetic coupling between the rare-earth ions and tend to decrease across the series. At a temperature below 100 K, which depends on the R ion, $1/\chi$ deviates below the high-temperature extrapola-

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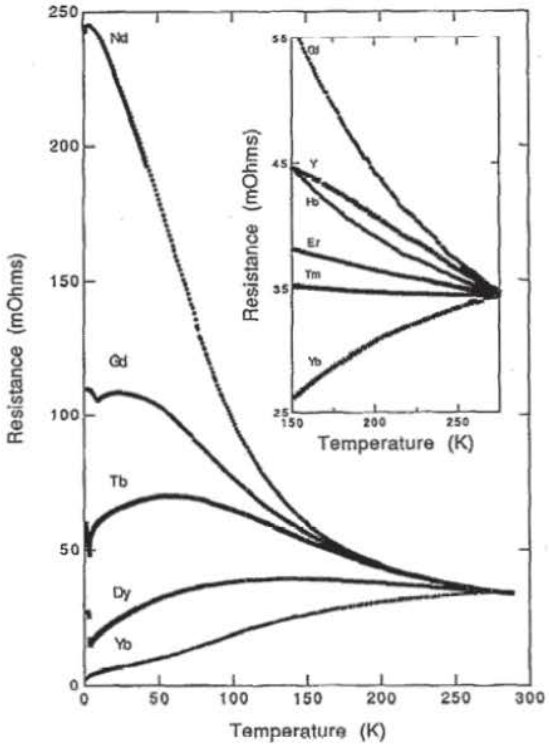

FIG. 1. Resistance vs temperature for representative members of the RBiPt series. Inset: High temperature resistance vs temperature for RBiPt members and YBiPt.

tion, probably suggesting crystal-field effects.

YbBiPt is particularly interesting among the RBiPt scrics. Mcasurements of its specific heat C and electrical resistance to 40 mK reveal a very weak anomaly in C centered at 0.35 K at which temperature R abruptly changes slope and becomes temperature independent down to 40 mK. Remarkably, C/T is huge, 8 J/K^2 mole, and is constant below 0.35 K. An entropy of R ln 2 is recovered only upon warming to approximately 1 K, well above the

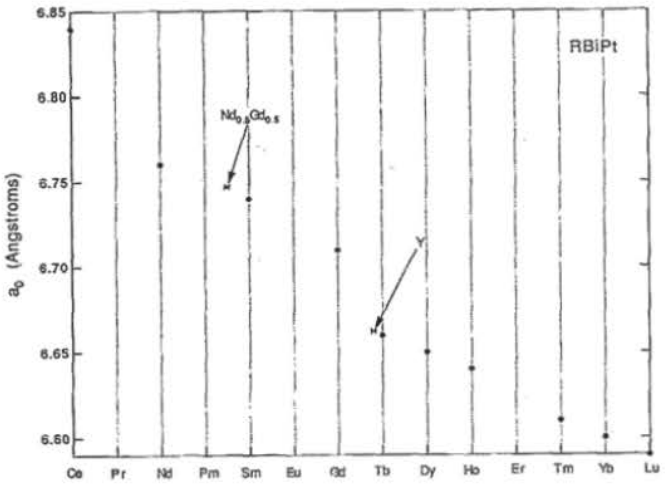

FIG. 2. Lattice parameter for RBiPt members including YBiPt and $Nd_{0.5}Gd_{0.5}BiPt$.

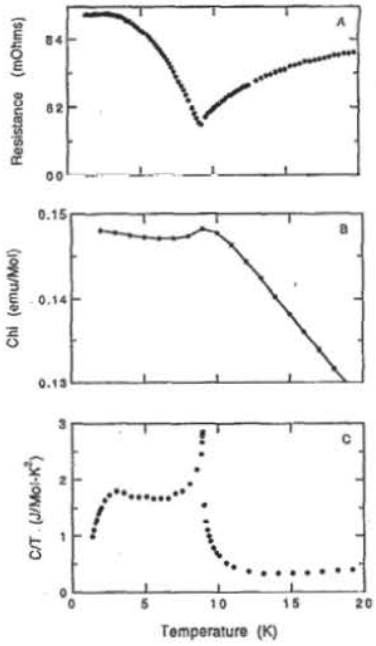

FIG. 3. Low temperature data on GdBiPt: (a) electrical resistance, (b) magnetic susceptibility, (c) specific heat divided by temperature.

anomalous region in R(T) and $C(T)$. A possible interpretation of this behavior is that this is a manifestation of strong electronic correlations that produce a low-temperature heavy-electron state out of which, for example, small moment ordering might occur. Clearly, additional measurements, e.g., nuclear magnetic resonance or muon spin resonance, are called for to test for the existence of long range, small moment ordering. If our interpretation withstands additional investigation, YbBiPt represents by far the most strongly renormalized effective mass state yet reported.

Our observations on RBiPt compounds raise a number of unanswered questions. First, the semiconductor-like resistance of NdBiPt seems quite unusual for an intermetallic compound. Whatever mechanism is responsible for this behavior appears to be dominated by volumetric considerations, given the systematics of Figs. 1 and 2. Further, the $4f$ electrons play no role in this, except for their lanthanide

TABLE I. High temperature effective moment μ_{eff}, paramagnetic θ, Neel temperature T_N, and entropy up to T_N for the RBiPt series.

Lanthanide	μ_{eff} (μ_B)	θ (K)	T_N (K)	S (J/mole K)
Ce[a]	2.50	−44.3	2.5	
Pr[a]				
Nd	3.59	−23.4	2.2	4.0
Sm			2.1	
Gd	7.83	−40.4	9.0	13.1
Tb	9.41	−23.1	3.4	8.6
Dy	10.22	−15.6	3.3	8.4
Ho	10.53	−12.4	<1.2	
Er	9.27	−6.9	<1.2	
Tm	7.24	−4.6	<1.2	
Yb	4.22	−1.4	$\leqslant 0.3$	
$Nd_{0.5}Gd_{0.5}$	6.00	−12.6		
$Nd_{0.5}Dy_{0.5}$	7.35	−7.4	2.1	
$Nd_{0.5}Yb_{0.5}$	3.88	−9.0		

[a]Grow out of Pb flux.

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contraction, since YBiPt falls within the progression. Second, there is the question of the mechanism responsible for the antiferromagnetic order. With the exception of GdBiPt, in which crystal-field effects are absent because $L = 0$ for a $4f^7$ configuration, T_N is relatively constant for R = Ce through Dy. A Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction may be responsible for the ordering but it would have to be mediated by a possibly small number of conduction electrons, given the high resistivity of the lighter rare-earth compounds, particularly NdBiPt. (This would require a process in which the conduction band is populated by electrons excited across the small gap; i.e., virtual excitations.) Obviously, T_N does not follow de-Gennes scaling $T_N = (g_J - 1)^2 J(J + 1)$, if Hund's rules J values are used; although, crystal-field effects may explain this discrepancy. Further, the resistive anomalies seen in several of the lighter RBiPt members at T_N suggest the development of a superzone gap, implying the existence of an s–f interaction necessary for the RKKY mechanism. Finally, there is the possibility of a very heavy-electron state in YbBiPt that coexists with small-moment ordering. If this is indeed true, the systematics displayed by the RBiPt series should provide important new insight into the origin of heavy-electron behavior in this material.

ACKNOWLEDGMENT

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[5] The Nd, Sm, Tb, Tm, Lu, and Y compounds have not been reported prior to this and this is the first time any of the RBiPt series have been made in single crystal form.

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[7] R. C. Albers, A. M. Boring, G. H. O. Daalderop, and F. M. Mueller, Phys. Rev. B **36**, 3661 (1987).

[8] CeBiPt and PrBiPt were grown out of Pb flux since the $R_3Bi_4Pt_3$ structure is favored for La, Ce and Pr growths from Bi flux.

[9] In the MgAsAg structure the (0,0,0) site is unique since it has nearest neighbors on both sides: (1/4,1/4,1/4) and (3/4,3/4,3/4). We have determined via intensity analysis that the Bi is on this unique site for YbBiPt (as is As for MgAsAg). It is possible, though, that for the lighter rare earths the Bi moves off the unique site, possibly causing the discontinuity in lattice parameter seen between Gd and Tb.

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