Ferromagnetism and crystal fields in $YbInNi_4$

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We present transport and thermodynamic measurements as a function of temperature (0.1–300 K), pressure (1 bar–16 kbars), and magnetic field (0–600 kOe) on $YbInNi_4$. Ferromagnetism arises near 3 K out of a state with enhanced electronic specific-heat coefficient. Resistivity, specific-heat, and magnetization measurements imply that the ground state of $YbInNi_4$ is a crystal-field doublet, whereas earlier neutron-scattering results suggest a ground-state quartet. We also compare $YbInNi_4$ to $YbInCu_4$ and intermediate alloys. [S0163-1829(98)00613-4]

I. INTRODUCTION

Because of the near degeneracy of Yb's f^{13} and f^{14} configurations, many Yb compounds display intermediate-valence or heavy fermion behavior.[1] In this sense, Yb may be thought of as the ''f-hole'' analog of Ce, whose f^0 and f^1 configurations give rise to similar phenomena. The competition between localized and itinerant f-electron behavior gives rise to a wealth of ground states in intermetallic compounds that depend significantly on rather subtle differences in lattice constant and ligand ions.[2] As such, the systematic study of the evolution of physical properties with chemical composition has been a fruitful area of research in the study of f-electron materials.

$YbXM_4$ compounds (where X is a semimetal or late transition metal and M is a transition metal) have attracted attention because of the wide variety of ground states displayed in this isostructural series. In particular, $YbInCu_4$ exhibits an isostructural valence transition,[3–10] $YbAgCu_4$ displays heavy fermion behavior with no magnetic order to the lowest temperatures measured,[11–13] and $YbPdCu_4$ and $YbAuCu_4$ order magnetically below 1 K.[11,13–15] The Ni variants of these compounds are relatively unstudied.[16–18]

Here, we present the results of a detailed study of the physical properties of single-crystal $YbInNi_4$. $YbInNi_4$ orders ferromagnetically at low temperature, a relatively uncommon ground state for trivalent Yb compounds—only a few examples of such behavior are known: the dense Kondo compound YbNiSn,[19–21] Yb_2Ni_{17}, in which the Ni sublattice also orders ferromagnetically,[22,23] and $YbPt_2$.[24] However, as discussed below, crystal-field effects are dominant in $YbInNi_4$, making it more analogous to the antiferromagnetic compound $YbBe_{13}$ (Refs. 25–28) than to YbNiSn.

II. EXPERIMENTAL DETAILS

We have grown single crystals of $YbInNi_4$ using an In-Ni flux in a manner similar to that employed in Ref. 4. $YbInNi_4$ crystallizes in the cubic $AuBe_5$ ($C15b$) structure with a lattice constant of 6.98 Å, as determined by powder x-ray diffraction (see also Refs. 16–18). The Yb and In ions sit on distinct face-centered-cubic sublattices displaced by (1/4,1/4,1/4) along the body diagonal and are surrounded by space-filling Ni tetrahedra. The single crystals have a well-faceted, tetrahedral morphology with typical dimension 3–5 mm. Measured physical properties are quantitatively reproducible from batch to batch and are insensitive to the precise composition and temperature profiles used in the flux growths.

A variety of experimental techniques have been employed to span the range of parameter space for which we report data. A superconducting quantum interference device magnetometer was used to measure the temperature-dependent magnetic susceptibility and isothermal magnetization, as well as to provide an absolute calibration to high-field magnetization measurements. Electrical resistivity measurements as a function of pressure and temperature were performed in a He^3 cryostat using a Be/Cu clamp-type pressure cell with flourinert as the pressure transmitting medium. For temperatures above 1.5 K, a thermal relaxation technique was used to determine specific heat,[29] while for temperatures below 2 K, a quasiadiabatic method was used. The agreement between these techniques in the range of temperature overlap is excellent. Magnetoresistance measurements were performed in a 200 kOe superconducting magnet with the magnetic field applied perpendicular to the measurement current, and high-field magnetization measurements were performed in a 600 kOe pulsed magnet using a mutual inductance technique. Both high-field magnets are located at the National High Magnetic Field Laboratory—Los Alamos Pulsed Field Facility.

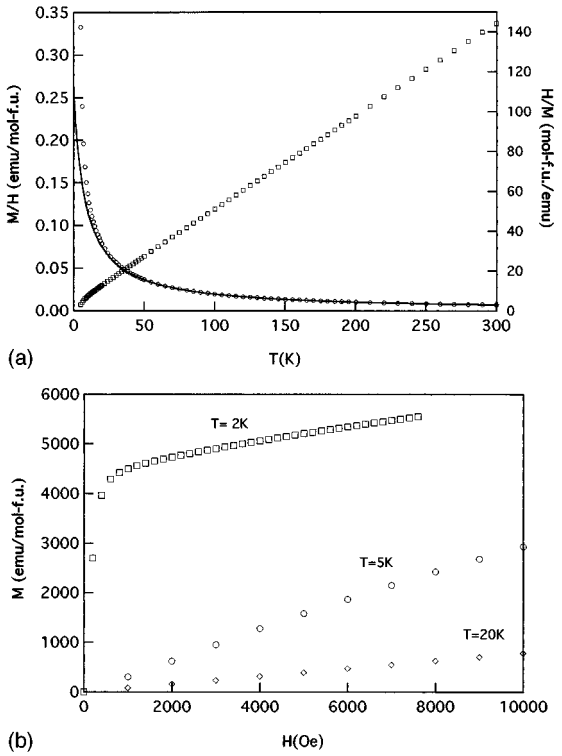

FIG. 1. (a) Magnetic susceptibility (circles) and inverse magnetic susceptibility (squares) for $YbInNi_4$. The solid line is a Curie-Weiss fit to the data which gives $\mu_{\text{eff}}=4.11\mu_B$ and $\theta=8.2$ K. (b) Isothermal magnetization as a function of field at various temperatures for $YbInNi_4$.

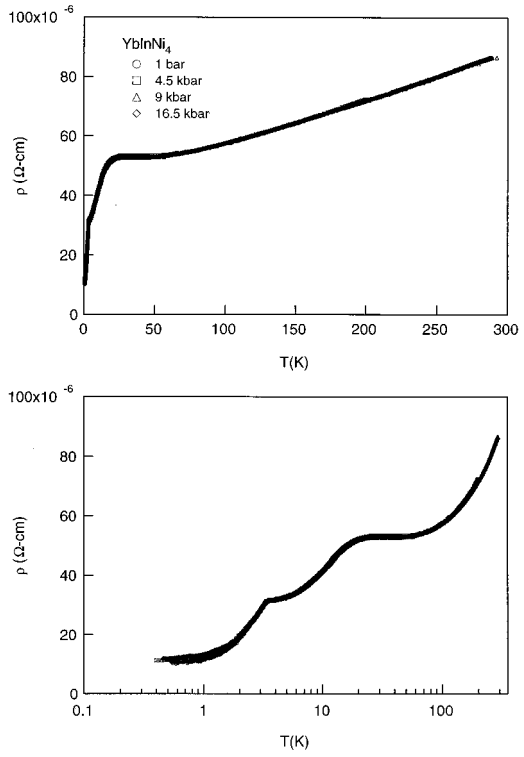

FIG. 2. Electrical resistivity as a function of temperature for $YbInNi_4$ (upper panel: linear T scale; lower panel: logarithmic T scale). The data at fixed pressures of 1 bar, 4.5, 9, and 16.5 kbars are indistinguishable.

III. RESULTS

In Fig. 1(a) we show the magnetic susceptibility of $YbInNi_4$ as a function of temperature. The high-temperature data are well-fit by a Curie-Weiss law with effective moment $4.11\mu_B$, nearly the full free-ion Yb value, and an antiferromagnetic Weiss temperature of 8.2 K. The deviation from Curie-Weiss behavior at low temperature is suggestive of crystal-field effects, a suggestion to be discussed in detail below, as well as the onset of short-range ferromagnetic correlations. Isothermal magnetization data in Fig. 1(b) indicate that $YbInNi_4$ orders ferromagnetically (consistent with previous observations[16]) at low temperature with nearly $1\mu_B$ of ordered moment. Note, however, that by 1 T, the magnetization is only weakly saturated. If one estimates the susceptibility at 2 K from $\Delta M/\Delta H$ in this region, one finds a susceptibility that is $\sim$50% of that expected from the high-T Curie-Weiss fit. This susceptibility is presumably due to a change in the population of excited crystal-field states with increasing magnetic field.

The electrical resistivity as a function of temperature and pressure is shown in Fig. 2. $YbInNi_4$ is a relatively good metal with a residual resistivity of only 10 $\mu\Omega$ cm. Two principal features are apparent in the 1-bar temperature-dependent resistivity: a shoulderlike feature near 20 K, suggestive of either the onset of Kondo coherence or the influence of crystal-electric fields, and a sharp drop at 3 K, consistent with the onset of magnetic order. The application of 16 kbars of hydrostatic pressure has no effect on either the magnetic ordering temperature or the position or magnitude of the 20-K feature. This argues strongly for a crystal-field-induced drop in resistivity.[30] Kondo coherence effects usually depend strongly on pressure due to the pressure dependence of the exchange coupling constant J.[31] For the case of crystal-field effects, Cornut and Coqblin[30] have shown that a drop in resistivity generally occurs at 0.7Δ, where Δ is the energy splitting between the ground state and the first excited crystal-field state. For $YbInNi_4$ this would imply a first excited state approximately 30 K above the ground state.

The temperature-dependent specific heat of $YbInNi_4$ is depicted in Fig. 3. Below 20 K the zero-field data are domi-

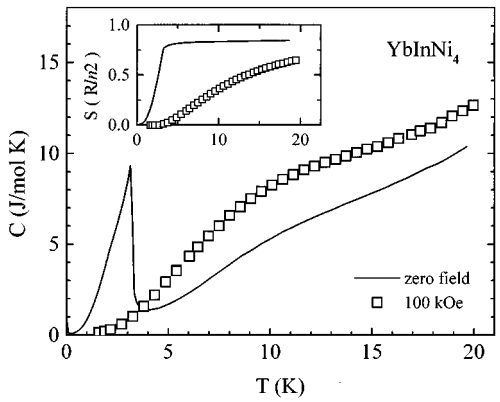

FIG. 3. Specific heat as a function of temperature for $YbInNi_4$ for $H=0$ (solid line) and $H=100$ kOe (squares). The corresponding magnetic entropy (in units of R ln2) is shown in the inset. See text for details.

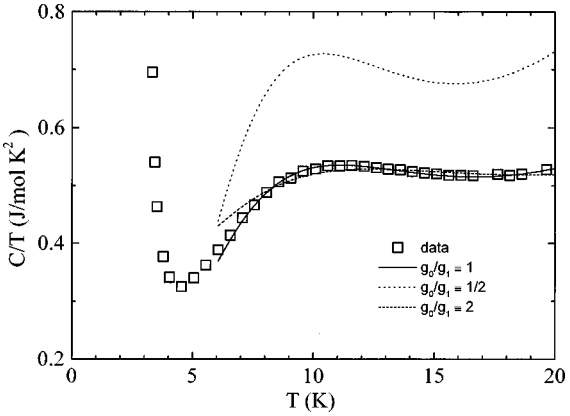

FIG. 4. Specific heat divided by temperature as a function of temperature for $YbInNi_4$. The squares represent the data, and the lines represent fits to the data assuming different crystal-field level degeneracies. See text for details.

nated both by the sharp magnetic ordering peak at 3 K as well as by a broad Schottky-like anomaly centered near 10–15 K. The data above 8 K are well described by a standard model that consists of an electronic contribution, a lattice contribution, and a Schottky contribution to C_p that is given by

$$C=\gamma T+\beta T^3+R\left(\frac{\delta}{T}\right)\frac{g_0}{g_1}\frac{\exp(\delta/T)}{[1+(g_0/g_1)\exp(\delta/T)]^2}, \quad (1)$$

with a Sommerfeld coefficient of $\gamma=150$ mJ/mole K^2, a T^3 phonon contribution with $\Theta_D=272$ K, and a Schottky contribution with an energy splitting of $\delta=32$ K. By subtracting these terms from the total measured C_p data it is possible to integrate the remaining specific heat to determine the magnetic entropy associated with the ferromagnetic transition. The observed entropy is very close to R ln 2 (see inset to Fig. 3), indicating that the ferromagnetic order arises from a ground-state doublet. The specific heat rises below 200 mK, presumably due to a nuclear Schottky contribution stemming from the influence of the internal field on nuclear hyperfine energy levels. The application of a 100-kOe magnetic field acts to smear the ferromagnetic transition considerably (open squares in Fig. 3), while the R ln 2 entropy is still recovered by 20 K. These in-field results are fully consistent with conventional ferromagnetic order.

The Schottky contribution to the specific heat is analyzed in greater detail in Fig. 4 where the data is plotted as C/T vs T. The near temperature-independence of C/T above 10 K is a clear indication that a low-lying Schottky anomaly is present in $YbInNi_4$. The eightfold degenerate ground state of Yb is split into two doublets and a quartet in the presence of a cubic crystal field.[32] As such, the ratio of the degeneracies of the ground state g_0 to the first excited state g_1 can be $g_0/g_1=1/2$, 1, or 2 for a doublet-quartet, doublet-doublet, or quartet-doublet configuration, respectively. These three level schemes can be tested by fitting the data in Fig. 4 with Eq. (1), and the resulting fits are shown in the figure. The fits clearly reveal that a doublet-doublet configuration with an energy splitting of $\delta=32$ K is most consistent with the data; this level splitting is also consistent with there being a broad feature in the resistivity data at roughly 20 K.

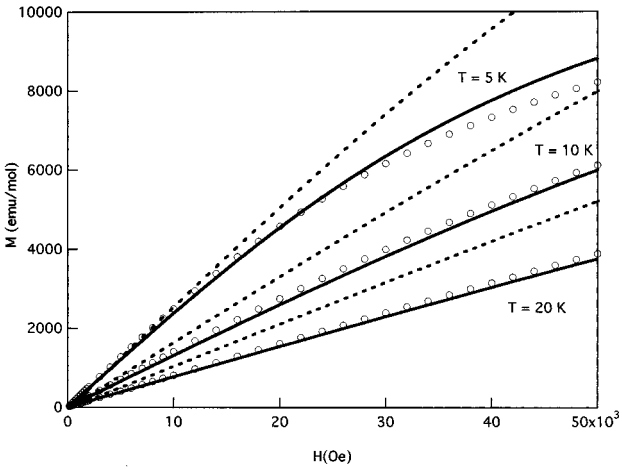

FIG. 5. Isothermal magnetization as a function of applied field at various fixed temperatures for $YbInNi_{3.25}Cu_{0.75}$. The circles are the experimental data and the solid lines represent a simultaneous fit to the 5, 10, and 20 K data. This fit corresponds to a ground-state doublet, an excited doublet at 64 K and a quartet at 122 K above the ground state [$x=0.53$, $W=0.48$ meV (Ref. 32)]. See text for details. For comparison, the dashed lines show the magnetization that would be expected for the crystal-field scheme [$x=0.38$, $W=-0.17$ meV (Ref. 32)] determined by Severing *et al.* for $YbInNi_4$ (Ref. 16).

Further insight into the crystal-field state of $YbInNi_4$ may be gained by more carefully examining the magnetization as a function of field at fixed temperature. Unfortunately, the presence of magnetic order complicates this analysis. Therefore, we have doped $YbInNi_4$ with sufficient Cu (approximate composition $YbInNi_{3.25}Cu_{0.75}$) to suppress long-range magnetic order. Despite the addition of this chemical disorder, it is reasonable to assume that the crystal-field state of Yb is, at least qualitatively, unchanged. The magnetization as a function of field for this sample is shown in Fig. 5. The magnetization is fit simultaneously using the 5, 10, and 20 K data. The crystal-field scheme that best fits these data is a ground-state doublet with an excited doublet 64 K above the ground state and with the quartet an additional 58 K above the excited doublet ($x=0.53$, $W=0.48$ meV in the notation of Lea, Leask, and Wolf[31]). Although the magnetization-derived crystal-field scheme has a somewhat larger splitting than that inferred from specific heat and resistivity, the relative level spacing and degeneracies are retained.

In Fig. 6 we show the magnetization of $YbInNi_4$ at 4 K as a function of magnetic field up to 600 kOe. The magnetization reaches a value of $3.25\mu_B$/Yb, close to the expected $g\cdot J$ value of $4\mu_B$ for the full multiplet of Yb ($g=8/7$, $J=7/2$), and no metamagnetic transitions are observed. Thus, despite the significant impact of crystal fields on the low-temperature, low-field properties of $YbInNi_4$, in sufficiently large magnetic fields, the behavior of the full Yb multiplet is recovered.

$YbInNi_4$ displays an appreciable negative magnetoresistance for temperatures much greater than $T_c=3$ K. This can be seen clearly in Figs. 7 and 8. In Fig. 7 we show the resistance as a function of temperature in zero-applied field and in an applied field of 180 kOe. The magnetic field was applied in a direction perpendicular to the measurement cur-

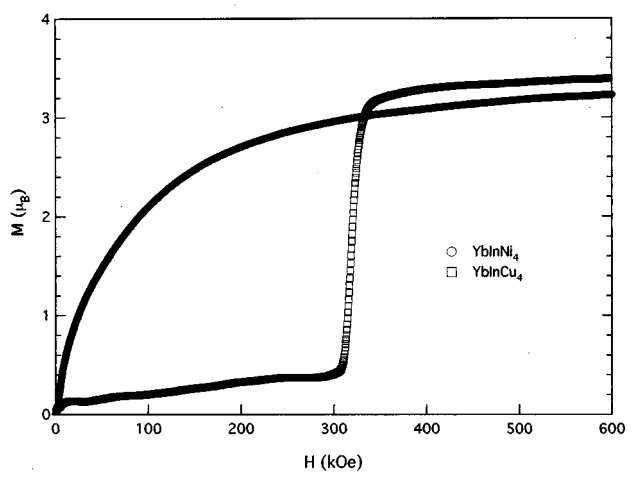

FIG. 6. Isothermal magnetization of $YbInNi_4$ at 4 K, measured using a 600-kOe pulsed magnet. For comparison data for $YbInCu_4$, which undergoes a field-induced, first-order valence transition (Ref. 6), are also shown.

rent. For $T \leqslant 100$ K, the applied field reduces the sample resistance, and the resulting resistance as a function of temperature is relatively featureless and typical of a metal. In Fig. 8(a) we show the transverse magnetoresistance $\{\Delta R \equiv [R(B)-R(B=0)]/R(B=0)\}$ as a function of magnetic field at various fixed temperatures. Negative magnetoresistances as large as 50% are observed. In the inset of Fig. 8(b), we show the magnetoresistance as a function of magnetic field for $T \leqslant T_c$ on an expanded scale. The saturated magnetoresistance decreases with decreasing temperature, suggesting that domain effects are dominant in the ferromagnetically ordered state.

In Fig. 8(b) we replot the magnetoresistance data for $T > 8$ K as a function of magnetic field divided by temperature (H/T). Remarkably, the data collapse onto a single curve. For $T < 8$ K, this scaling breaks down. It is interesting to note that this is the temperature at which ferromagnetic correlations begin to dominate the magnetic susceptibility—$\chi \cdot T$ passes through a minimum at 8 K (not shown). On the other hand, Kohler's rule is poorly obeyed: the data plotted as a function of $B/\rho(B=0,T)$ (not shown) are qualitatively similar to that of Fig. 8(a), indicating that the scattering rate is not independent of magnetic field (a necessary condition for the applicability of Kohler's rule).[33]

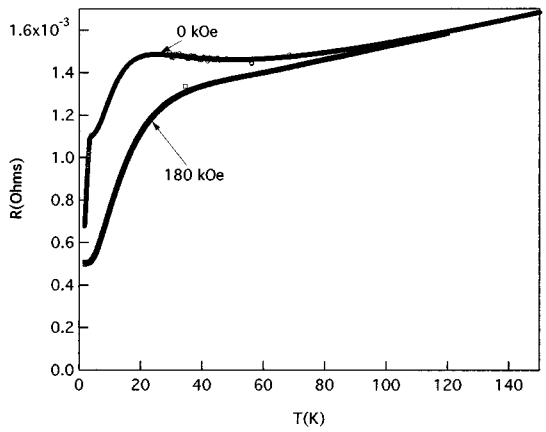

FIG. 7. Resistance as a function of temperature for $YbInNi_4$ in magnetic fields of 0 and 180 kOe. The magnetic field was applied perpendicular to the applied current. Note the appreciable magnetoresistance at temperatures as high as 120 K.

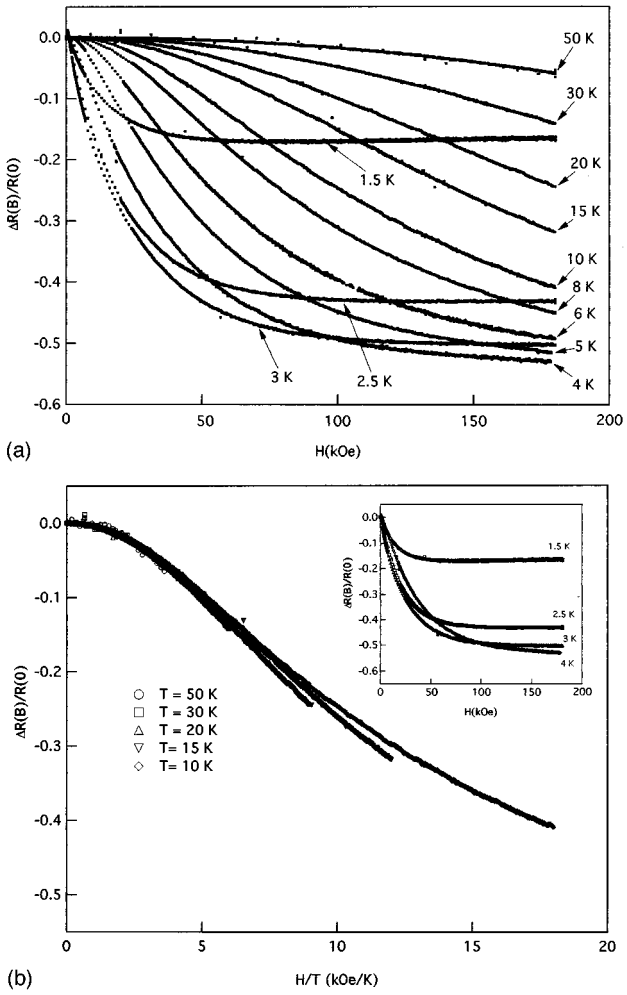

FIG. 8. (a) Transverse magnetoresistance $[R(B)-R(B=0)]/R(B=0)$ as a function of applied field at various fixed temperatures for $YbInNi_4$. (b) The data of Fig. 8(a) plotted as a function of H/T. For $T \geqslant 8$ K, the data collapse onto a single curve. The data for $T \leqslant T_c$ are shown in the inset. Note that the magnitude of the magnetoresistance at high field decreases with decreasing temperature.

Although magnetoresistance that is proportional to H/T is not atypical of f-electron materials, the range of temperature over which this scaling is observed is rather large. Two common mechanisms give rise to such behavior. Scattering from independent Kondo impurities produces magnetoresistance that is a universal function of B/B^*, where B^* is a characteristic field given by $B^*(T)=k_B(T_K+T)/(g\mu)$.[34] However, attempts to improve the scaling of our data by replotting as a function of $T+T_K$ give rise to values of T_K that do not vary monotonically with T—i.e., assuming $T_K=T_K(T)$ (Ref. 35)—and do not substantially improve the collapse of our data. On the other hand, Beal-Monod and Weiner have shown that $\Delta R \propto M^2$ should result from the suppression of normal spin-flip scattering due to the polarization of local

moments by the applied field.[36] In fact, our data, for small values of H/T, are well described by $\Delta R = -a(H/T)^2$, with a a positive constant. At large values of H/T, where $M = \chi H$ becomes a bad approximation due to saturation effects, our data deviate from this behavior. Unfortunately, we have not been able to measure isothermal magnetization in sufficiently high magnetic fields to test the extent to which $\Delta R \propto M^2$ experimentally.

IV. DISCUSSION

From the data presented in Sec. III, $YbInNi_4$ appears to be a rather conventional f-electron material in which crystal-field effects are dominant and ferromagnetism arises at low temperature out of a doublet ground state. These experimental observations are somewhat troubling for several reasons. In 1990 Severing *et al.*[16] performed inelastic neutron-scattering measurements on $YbInCu_4$ and $YbInNi_4$ and found evidence for a quartet ground state in both of these materials. This conclusion appears to be straightforward because direct doublet-doublet transitions within Yb's crystal-field multiplet in a cubic crystal field are forbidden by symmetry. Thus, the observation of two distinct crystal-field excitations—as observed by Severing *et al.* for both $YbInCu_4$ and $YbInNi_4$—is a clear signature of a quartet ground state. Intensity analysis of the neutron data is also consistent with a ground-state quartet, and the magnetization inferred from the neutron data is in good agreement with that measured by bulk susceptibility.[16] Furthermore, in the case of $YbInCu_4$, our own specific-heat data[37] show that approximately R ln6 of entropy is liberated at the first-order valence transition, strongly suggestive of a greater-than-doublet ground-state degeneracy for $YbInCu_4$. It is difficult to understand how relatively similar ligand ions can produce such a large change in the crystal electric field; however, $YbInNi_4$ is a substantially better metal than $YbInCu_4$, as deduced from Hall-effect measurements, and the increased conduction electron density may provide sufficiently enhanced hybridization to modify one's point-charge expectations. Thermodynamic[38] and electron paramagnetic resonance[39] experiments on $YbInNi_{4-x}Cu_x$ are currently underway to clarify these issues. However, if one calculates the magnetization as a function of field at fixed temperature that would be expected from the neutron-inferred crystal-field scheme for $YbInNi_4$ (see Fig. 5), the result is clearly inferior to the crystal-field scheme discussed above. It is perhaps possible that these differences could arise due to a disorder effect—Severing *et al.* studied polycrystalline material whereas all of our results were obtained with single-crystal samples; however, such a conclusion seems unlikely. In any event, inelastic neutron-scattering experiments on single crystals of $YbInNi_4$ would appear to be warranted.

The appearance of ferromagnetism in $YbInNi_4$ is also somewhat unexpected. Buschow *et al.*[40] have argued that, at least in pseudobinary compounds, Yb-Ni compounds favor a higher-valence state than do Yb-Cu compounds. It, therefore, may not be surprising that Kondo effects (which act to reduce the effective Yb moment) are dominant in $YbInCu_4$, to the extent that a first-order valence change is observed,[3–10] whereas local moment behavior is dominant in $YbInNi_4$. In fact, two of the three other compounds in which Yb orders ferromagnetically (YbNiSn and Yb_2Ni_{17}) are compounds in which Ni is present. However, Kondo physics plays a dominant role in YbNiSn,[19–21] and the ferromagnetic ordering of the Ni sublattices at higher temperature appears to influence the ordering of Yb in Yb_2Ni_{17}.[22,23]

If one applies a Friedel-type analysis[41] to the $YbXM_4$ compounds that are known to order magnetically ($YbInNi_4$, $YbAuCu_4$, and $YbPdCu_4$), the results are ambiguous. $F(k_Fa)$, where $F(x) = (x\cos x - \sin x)/x^4$, k_F is the Fermi momentum and a is the spacing between magnetic ions, changes sign between $YbInNi_4$ and $YbAuCu_4$ (which orders antiferromagnetically[14]), assuming a single-band picture in which the Hall constant $R_H = 1/ne$,[42] and taking a as the Yb-Yb distance. However, this simple analysis would predict antiferromagnetic order for $YbInNi_4$ and ferromagnetic order for $YbAuCu_4$. Because of sample purity problems, it has not proven possible to obtain a good estimate of the Hall coefficient of $YbPdCu_4$ (which appears to order ferromagnetically[14] rather than antiferromagnetically, as previously reported[11]). It is possible that a more careful analysis, including greater than nearest-neighbor interactions and a more realistic treatment of the electronic band structure, could explain the origin of ferromagnetism in $YbInNi_4$.

Finally, it is interesting to note that despite their drastically different low-temperature, low-field behavior, $YbInNi_4$ and $YbInCu_4$ are quite similar in high magnetic fields. Both materials have nearly identical magnetizations above 400 kOe (see Fig. 6)—the 5% difference at high field may well be within experimental uncertainty: the absolute calibration for $YbInCu_4$ is rather difficult given the small magnitude of the signal at low field (where comparison to magnetometer data is possible) and the enormous change in signal size at the valence transition. Apparently, the large Zeeman energy supplied by the magnetic field overwhelms the low-energy-scale effects of the Kondo interaction (in the case of $YbInCu_4$) and the effects of crystal-electric fields (in the case of $YbInNi_4$) to the extent that the magnetization of the full Yb multiplet is manifest at high field.

V. CONCLUSION

We have shown that ferromagnetism arises in $YbInNi_4$ at low temperature out of a crystal-field-dominated ground state. This ground state possesses both enhanced electronic specific heat and an appreciable and rather universal negative magnetoresistance. Further work is needed to resolve the differences between the crystal-field scheme inferred from our thermodynamic measurements and that inferred from earlier neutron-scattering results.[16] The detailed nature of the ferromagnetism in $YbInNi_4$ and the associated magnetoresistance also merit further study.

ACKNOWLEDGMENTS

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