Resistivity, magnetization, and specific heat of $YbAgCu_4$ in high magnetic fields

T. Graf, J. M. Lawrence,* M. F. Hundley, and J. D. Thompson
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

A. Lacerda, E. Haanappel, and M. S. Torikachvili†
National High Magnetic Field Laboratory, Pulse Facility, Los Alamos, New Mexico 87545

Z. Fisk
Florida State University and National High Magnetic Field Laboratory, Tallahassee, Florida 32306

P. C. Canfield
Ames Laboratory, Ames, Iowa 50011

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We report measurements of the resistivity ρ, Sommerfeld coefficient of specific heat γ, and magnetization M of polycrystalline $YbAgCu_4$ in high magnetic fields $0 \leq B \leq 18$ T [in the case of $M(B)$ to 50 T]. A comparison of the temperature-dependent susceptibility $\chi(T)$ as well as field-dependent Sommerfeld coefficient γ and magnetization to Kondo theory for a $J=7/2$ impurity shows that theory correctly predicts the functional dependence of these quantities on T and B, but the characteristic temperatures determined from the various measurements (120, 98, 77, and 63 K) differ by nearly a factor of 2, which is difficult to understand within the context of Kondo theory even when other possible contributions are considered. In addition the normalized (Wilson) ratio of χ to γ is 1.00 at zero field (compared to 1.14 in Coqblin-Schrieffer theory) and decreases with increasing magnetic field. The magnetoresistance is positive at all temperatures, reaching a value $\Delta\rho(B)/\rho(B=0)=0.6$ at 25 mK and 18 T. The low-temperature magnetoresistivity $\Delta\rho(B)$ varies as $B^{1.5}$. We argue that this is dominated by an ordinary impurity effect. Kohler's rule is clearly violated as the temperature is raised; the scattering rate appears to increase with field below 40 K and decrease with field above 40 K. This behavior is expected for an Anderson lattice when a pseudogap is present. At low temperature the resistivity increases as AT^2. The coefficient A (corrected for cyclotron-orbit effects) increases with field such that the ratio $A(B)/\gamma(B)^2$ is a constant. Doping with Lu onto the Yb site, or with Ni onto the Cu site, changes the magnitude of the low-temperature resistivity in a manner consistent with the predictions of the theory of ligand-induced disorder in an Anderson lattice.

I. INTRODUCTION

Magnetic fields have proven to be a useful probe of the ground state of strongly correlated electron systems[1] and have revealed unexpected phenomena in heavy-fermion compounds, such as the metamagnetic transitions that occur in $CeRu_2Si_2$ (Refs. 2 and 3) and UPt_3 (Refs. 4 and 5) at fields of 8 and 20 T, respectively. In addition, magnetic field as a thermodynamic variable can be used to test theoretical models commonly invoked to explain zero-field properties of these materials. In particular, we are interested in addressing two specific issues: (1) How well can the thermodynamic properties of periodic mixed-valent compounds be described in terms of the Kondo-Anderson impurity model, and (2) how well do existing theories of the field-dependent scattering rate of heavy-fermion compounds describe the measured magnetoresistivity? To address these issues, we have measured the field dependence of the specific heat $C(T)$ and electrical resistivity $\rho(T)$, and the magnetization $M(B)$ of the weakly mixed-valence compound $YbAgCu_4$.[6] Zero-field studies of this material show that the $4f$-occupation number is $n_f=0.88$ (Ref. 7) and that the linear-in-temperature (Sommerfeld) coefficient of specific heat has the value $\gamma=243$ mJ/mole K^2.[6,8] Neutron scattering finds[9] only a quasielectric component at temperatures above approximately 75 K and no evidence for well-defined crystal-field excitations. The quasielastic linewidth ($\Gamma/2\approx 80$ K) gives a characteristic energy scale for Kondo-like spin fluctuations that is consistent with the values for n_f and γ. Even at low temperatures hybridization is sufficiently strong that crystal-field splitting of the Hund's rule ground state, in the classical sense, is not well defined. These observations, together with high-pressure studies,[10] favor the interpretation that the ground state of $YbAgCu_4$ at ambient pressure is eightfold degenerate, i.e., that the total angular momentum of the Yb ion is $J=7/2$. Thus, $YbAgCu_4$ appears to be a very good material with which the issues of interest can be investigated. In the following, we briefly review theoretical issues that will be used to interpret the measurements.

There is substantial evidence that the thermodynamic

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properties of periodic heavy-fermion–mixed-valent compounds can be described over a broad temperature range by the single-impurity Anderson model.[11–13] In principle, the field and temperature dependence of specific heat and magnetization of a degenerate ($N_J=2J+1$) Anderson impurity in the Kondo limit $n_f \rightarrow 1$ (which for $N_J>2$ is often called the Coqblin-Schrieffer limit) should be universal functions of T/T_K, where T_K is the Kondo temperature.[11,14] For this limit, calculations by the Bethe ansatz exist for the zero-field susceptibility $\chi(T)$ and specific heat divided by temperature $C(T)/T$ for $J=1/2-7/2$,[15] for the low-temperature magnetization $M(B)$ for $J=1/2-7/2$ (Ref. 16) and for the field dependence of $C(T)$ for $J=1/2$ (Ref. 17) and 5/2 (Ref. 18). For each calculation, the characteristic temperature is defined in a different manner; but well-defined relationships between the variously defined characteristic temperatures exist.[16] In what follows, we primarily use the characteristic temperature T_L obtained from the low-temperature susceptibility by the relation $\chi(0)=C_J/T_L$, where C_J is the free-ion Curie constant $C_J=(g\mu_B)^2J(J+1)/3k_B$. This can be related to the characteristic temperature T_0 used by Rajan [$T_0=(2J+1)T_L/2\pi$, Ref. 15] and to the characteristic temperature T_1 defined in calculations[16] of the field-dependent magnetization (for $J=7/2$, $T_1=0.321T_L$). The quantity called H/T_K in the papers of Schlottmann[12,14] and of Okijj and Kawakami[18] is $g\mu_B B/k_B T_L$. The Kondo temperature T_K, as defined from a high-temperature expansion of the susceptibility, similarly can be related to T_L, e.g., $T_K=0.6745T_L$ for $J=7/2$.[16]

In solutions to the degenerate Anderson model by use of the NCA (noncrossing approximation or finite temperature $1/N_J$ expansion), it has been shown[9,11] for the $J=5/2$ case that the Coqblin-Schrieffer limit is valid for $n_f>0.85$. Assuming this is true for $J=7/2$, we should be able to compare the $\chi(T)$, $M(B)$, and $\gamma(B)$ data for $YbAgCu_4$ to the Bethe-ansatz calculations in the Coqblin-Schrieffer limit by finding a *single* value of T_L that is valid for all measurements. Unfortunately, calculations of $\gamma(B)$ do not exist for the $J=7/2$ case appropriate to Yb. However, for $J=1/2$ it has been shown explicitly[17] that the Wilson ratio $\Re=\pi^2R\chi(0)/3\gamma C_J$ (where R is the gas constant) is independent of magnetic field. By numerical analysis of the theoretical results of Okiji and Kawakami[18] we have convinced ourselves that this is true for $J=5/2$. In what follows, we assume that this is also true for $J=7/2$, so that $\gamma(B)$ should be proportional to the differential susceptibility $\chi(B)=\partial M/\partial B$. Finally, we note that Schlottmann[14] has shown that discrepancies between the Coqblin-Schrieffer predictions and the zero-field susceptibility and specific-heat data can be removed partially by calculations for the degenerate Anderson model for weak mixed valence (as opposed to the Kondo limit). In this case, the above-mentioned universal behavior is no longer strictly valid, and $C(B,T)$ and $M(B,T)$ depend on n_f as well as T_K. No calculations of the response to a magnetic field exist for the mixed-valence case, however. Hence, we restrict our comparison of data to the theory in the Kondo limit.

The theory of the magnetoresistance of the Anderson lattice is meager. Kawakami and Okiji[20] used the single-site approximation, where the basic idea is that in the pure lattice at $T=0$ the phase shifts δ on all sites are equal to the unitarity limit so that the resistance and magnetoresistance (which arise from fluctuations in the phase shift) vanish. At high temperatures thermal disorder in the phase shifts causes the scattering rate to increase with T but in a magnetic field it decreases, as for a Kondo impurity. At the lowest temperatures a magnetic field induces only small changes in the scattering rate, which can be positive in the situation that the Fermi level lies below a pseudo-gap in the $4f$-density of states. Ohkawa[21,22] has identified two types of residual disorder that can give a field-dependent *residual* resistivity in the single-site approximation. Kondo holes, where the rare-earth atom is replaced with a nonmagnetic counterpart, give a zero-phase shift at the impurity site, equivalent to a large local fluctuation in phase shift. The scattering rate then decreases in a magnetic field, proprotional to $x\rho_u[\rho_K(B)/\rho_K(0)]$, where x is the concentration of Kondo holes, ρ_u is the unitarity limit, and $\rho_K(B)$ is the theoretical scattering rate of a Kondo impurity in a magnetic field. Ligand disorder occurs when disorder on neighbors to the rare-earth atom induces local variations in hybridization with the nearby rare-earth site and consequently a spatial variation of the Kondo temperature. The application of a magnetic field causes the phase shifts on the affected sites to vary from the average value, and the scattering rate increases with increasing field. At low fields the effect is proportional to $x\rho_u(\delta T_K/T_K)^2(gJ\mu_B B/k_B T_K)^2$, where x is the concentration of rare-earth sites neighboring disordered ligands and δT_K is a measure of the magnitude of the disorder in T_K. These theories concern the field-dependent scattering rate that must be distinguished from the magnetoresistance arising from the effect of field-induced cyclotron orbits on the measured resistance at fixed scattering rate,[23,24] which effect is necessarily positive. Hong and Gehring[25] have calculated the magnetoresistance of an Anderson lattice due to a field-independent scattering rate induced by impurities. They used the slave boson approximation and a formula for the magnetoresistance appropriate to a two-band situation (one band for each spin direction). They argued that the magnetoresistance of $CeRu_2Si_2$ (Ref. 2) could be understood in this way.

In what follows, we first give in Sec. II the experimental details. Then, we treat in turn the high-field magnetization and specific heat in Sec. III, and the magnetotransport in Sec. IV, giving the data, analysis, and discussion separately in each section. Results are summarized in Sec. V.

II. EXPERIMENTAL DETAILS

Polycrystalline samples $YbAgCu_4$, $YbAgCu_{4-x}Ni_x$, and $Yb_{1-x}Lu_xAgCu_4$ were prepared by melting the starting elements in sealed, evacuated tantalum tubes. As shown in Fig. 1, the low-field (0.1-T) magnetic susceptibility of $YbAgCu_4$, measured with a superconducting quantum interference device (SQUID) magnetometer,

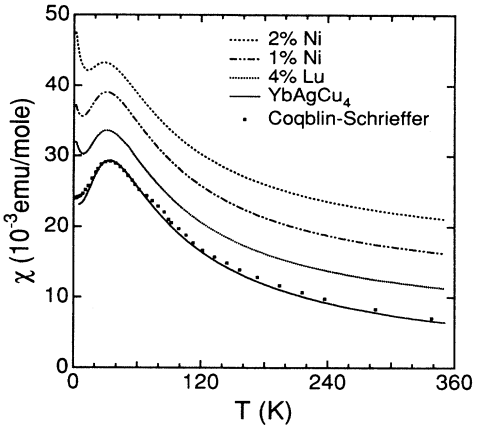

FIG. 1. Susceptibility of $YbAgCu_4$, $YbAgCu_{3.99}Ni_{0.01}$, $YbAgCu_{3.98}Ni_{0.02}$, and $Yb_{0.96}Lu_{0.04}AgCu_4$ as a function of temperature at 0.1 T. The zero of the ordinate is offset successively by 5×10^{-3} emu/mole starting from the bottom. The solid points are the predictions of the Coqblin-Schrieffer model for $J=7/2$ and $T_L=117$ K but increased in magnitude by a multiplicative factor of 1.09 (see text for details).

shows a vanishingly small "Curie tail" at low temperatures. Comparison to the low-temperature susceptibility of $YbAgCu_{3.99}Ni_{0.01}$ indicates that the level of magnetic impurities is much less than 1%. The zero-field residual resistance ratio $R(300K)/R(4\text{ K})=6.3$ is typical of the best values reported to data for $YbAgCu_4$.[6,26]

The field dependence of the electrical resistivity and magnetization was measured at the National High Magnetic Field Laboratory (Pulsed Field Facility, Los Alamos). Magnetoresitivity $\rho(B,T)$ was determined in fields $0\le B\le 18$ T generated in a superconducting magnet for temperatures in the range $0.025\le T\le 120$ K. Absolute values of ρ are estimated to be accurate to $\pm20\%$ because of uncertainty in determining precise dimensions of the approximately bar-shaped samples. Magnetization experiments were performed in both the 18-T superconducting magnet and in a 50-T pulsed magnet. M versus B to 18 T was measured on a 150-mg piece of $YbAgCu_4$ in the temperature interval $2\le T\le 200$ K with a calibrated vibrating sample magnetometer. Variable temperature was provided by helium-gas flow in the sample space. Pulsed fields to 50 T were generated in a capacitively driven copper solenoid such that the total pulse length was less than 30 msec. Measurements were performed with sample and pick-up coils immersed in liquid ^{3}He at 0.40 K. The signal, which is proportional to the magnetization (but which is not calibrated in absolute units), was determined by integrating the voltage output from a pair of nominally compensated pick-up coils, each having 1000 turns and wound in series opposition. A background signal, obtained with the sample removed from one of the coils, was subtracted before performing the integration. Because the coils were not exactly compensated, a small difference voltage develops that varies from pulse to pulse, and its integration with the desired signal leads to an additional, apparent linear-in-field contribution to the magnetization. This undesired signal was suppressed by subtracting a variable fraction of the voltage induced across a third, few-turn coil located next to the opposed coil pair. "Fine tuning" the system in this way was done before each measurement with the sample removed from the pick-up coils. Finally, a thermal-relaxation method was used to measure the specific heat of a small ($\approx$10-mg) piece of $YbAgCu_4$. Absolute accuracy in $C(T)$ is better than $\pm1\%$. For C/T in the highest magnetic field, this increases to $\pm2\%$.

III. MAGNETIZATION AND SPECIFIC HEAT

The temperature-dependent susceptibility of $YbAgCu_4$ (as well as of several alloys, to be discussed in Sec. IV) is shown as the solid line in Fig. 1. The magnetization at 400 mK for field up to 50 T is given in Fig. 2(a) (solid line). Because of the above-mentioned lack of calibration

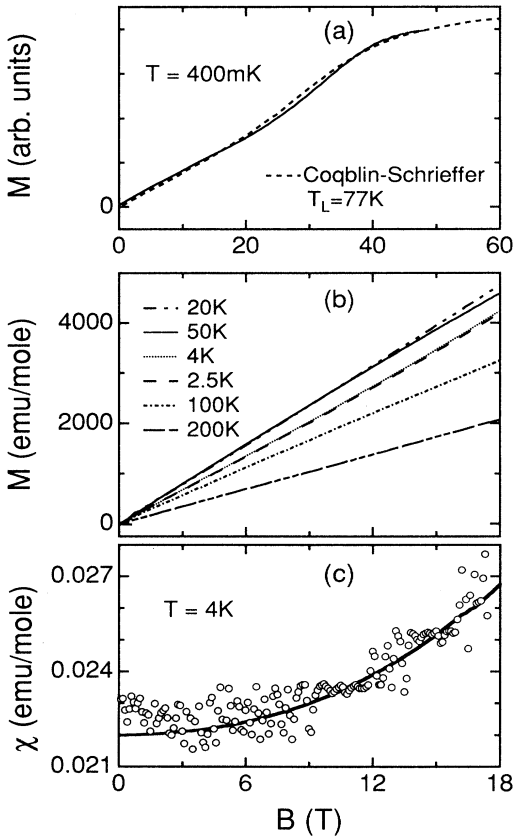

FIG. 2. (a) Low-temperature magnetization of $YbAgCu_4$ (solid line, arbitrary units) as a function of magnetic field. The dashed line is the prediction of the Coqblin-Schrieffer model for $M(B)$ (ordinate arbitrary) for a characteristic temperature $T_L=77$ K (see text). (b) Magnetization at six different temperatures vs field to 18 T generated in a superconducting magnet. For these data the ordinate is calibrated. (c) The differential susceptibility, obtained by differentiating the data of Fig. 2(b) at 4 K (open circles). The solid line is the prediction of the Coqblin-Schrieffer model for $\chi(B)/\chi(0)$ with a value $T_L=77$ K (see text).

of the absolute magnetization, the ordinate is given in arbitrary units. The (calibrated) magnetization for fields to 18 T and for six temperatures between 2.5 and 200 K is given in Fig. 2(b). Derivatives of these data obtained at low fields agree very well with the low-field susceptibility results shown in Fig. 1, at all six temperatures. In Fig. 3(a) we show the specific heat divided by temperature $C(T)/T$ obtained in applied fields of $B=0$, 2, 4, 6, 8, and 10 T. The specific heat varies as $\gamma T+\beta T^3$ for all fields in this temperature range. The Debye temperature obtained from the T^3 term is essentially field independent, with value $\Theta_D=245$ K. The linear coefficients, plotted in Fig. 3(b), vary as B^2 (solid line) and increase by 10% at $B=10$ T.

We compare these data to predictions of the $J=7/2$ Coqblin-Schrieffer model, obtained through the Bethe ansatz and convert the characteristic temperatures utilized in the theoretical papers to a common temperature T_L, as discussed in the Introduction. Before fitting the susceptibility, we first determined the non-4f background by measuring the susceptibility of $LuAgCu_4$ (not shown). This has a broad maximum at 140 K with a value

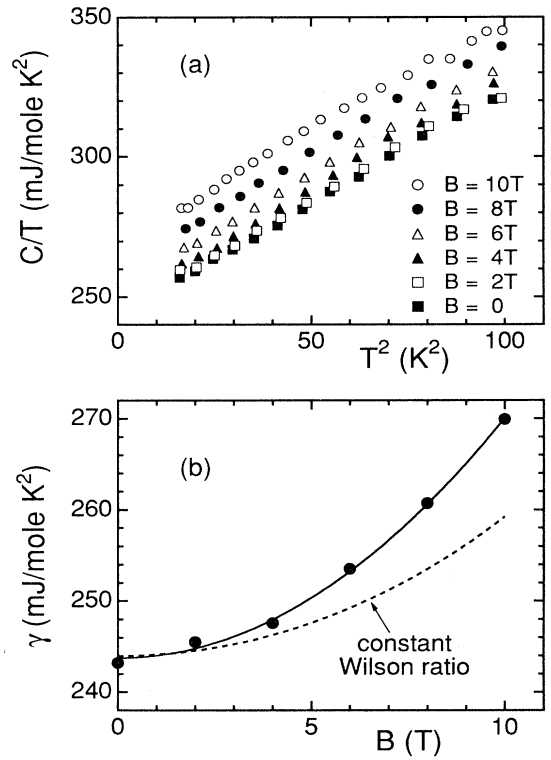

FIG. 3. (a) Specific heat $C(T)/T$ of $YbAgCu_4$ at various fields to 10 T plotted as a function of the square of the temperature. The Debye temperature deduced from the T^3 term is 245 K, independent of field. (b) Linear coefficient of specific heat γ determined from (a) vs the magnetic field. The estimated experimental uncertainty in γ is ± 2 mJ/mole K^2. The dashed line, marked "constant Wilson ratio" is determined assuming proportionality of $\gamma(B)/\gamma(0)$ to the theoretical susceptibility $\chi(B)/\chi(0)$ shown in Fig. 2(c), where $T_L=77$ K (see text). The solid line fits the data with $T_L=63$ K.

0.143×10^{-3} emu/mole and falls to 0.140×10^{-3} (0.120×10^{-3}) emu/mole at $T=0$ K (300 K). Because this is negligibly small on the scale of Fig. 1, we ignore it in the following. We use the theoretical results of Rajan[15] for $\chi(T)$; to compare to the data requires choice of T_L. If we evaluate this directly using the measured value of $C_J/\chi(0)$, we obtain $T_L=110$ K. Alternatively, T_L can be chosen so that the temperature T_m of the maximum in the data and theory coincide; this gives a similar value $T_L=117$ K. However, for this value of T_L the magnitude at the maximum should be $\chi(T_m)=26.9\times10^{-3}$ emu/mole, but the actual value at the maximum is 29.4×10^{-3} emu/mole. In Fig. 1 we have plotted the theory curve (solid squares) using $T_L=117$ K (so that the temperature of the maximum coincides with that of the data) but have multiplied the theoretical values of χ by a factor of 1.09. This should allow the reader to see that the theory reproduces the shape of the data very well; however, it also implies that theory underestimates the magnitude of χ by as much as 10%. We have measured χ in three samples of $YbAgCu_4$ and find that, although T_m has the same value (34 K) in all samples, there are $\pm10\%$ deviations from the average values of $\chi(0)=20.1\times10^{-3}$ emu/mole and of $\chi(T_m)=27.7\times10^{-3}$ emu/mole, and in each case, there are 10% deviations between theory and data over a significant temperature range. The origin of these differences is unclear, it possibly reflects variations in the site occupancies, in part due to the volatility of Yb. The apparent 10% disagreement with theory is thus of the same order as the experimental uncertainties. The average value of T_L deduced from these studies is $T_L=120\pm$ K.

In Fig. 2(a) we plot the prediction[16] for the low-temperature, high-field magnetization as the dashed line. In this plot, T_L was chosen to give best overall agreement. Because the ordinate has arbitrary units, there are actually two fit parameters: T_L, which is primarily determined by the shape [i.e., by the characteristic field B_0 at which the inflection in $M(B)$ occurs], and the overall amplitude. The value obtained, $T_L=77$ K, is substantially smaller than that expected on the basis of the susceptibility. Hence, while the S shape of the $M(B)$ curve determined experimentally is well reproduced by the Coqblin-Schrieffer theory, given T_L obtained from $\chi(T)$, the theory incorrectly overestimates the characteristic field B_0 by a factor of 1.6.

That this is not an artifact of the uncalibrated ordinate is demonstrated in Fig. 2(c). Hewson and Rasul[16] give a low-field expansion for the zero-temperature magnetization $M(B)$, which can be differentiated to give an expression for the susceptibility of the form $\chi(B)/\chi(0)=1+a_2x^2+a_4x^4$, where $x=g\mu_B B/k_B T_L$. The values of the coefficients can be determined from those tabulated in the theoretical paper.[16] In Fig. 2(c) we show the resulting curve for $J=7/2$ plotted as $\chi(0)(1+a_2x^2+a_4x^4)$ for $\chi(0)=0.022$ emu/mole and $T_L=77$ K. This is compared to the differential susceptibility $\chi(B)=\partial M/\partial B$ obtained by differentiating the data of Fig. 2(b) taken at 4 K. Note that for these data the ordinate *is* calibrated. We see that the fit is respectable. Were we to use a value

for $T_L=120$ K, the theory curve would underestimate the measured ratio of $\chi(18T)/\chi(0)$ by a factor of 1.14. Note that the shape of the theory curve is good for the choice $T_L=77$ K, but the value of $\chi(0)$ utilized is that appropriate to a characteristic temperature $T_L=117$ K. Again, this means that the characteristic *field* B_0 is overestimated by the theory.

The value of characteristic temperature determined from the specific-heat coefficient at zero field [Fig. 3(a)] using the Bethe-ansatz formula for $J=7/2$ is $T_L=7\pi^2R/24\gamma=98$ K; hence, at zero field there is a disagreement of order 20% between the values of T_L determined from the specific heat and from the overall shape of the susceptibility. The Wilson ratio [normalized ratio of $\chi(0)$ to γ; see the Introduction] is $\Re=1.00$ compared to the value $(2J+1)/2J=1.14$ expected on the basis of the Coqblin-Schieffer model.

In Fig. 3(b) we analyze the variation of the specific-heat coefficient with the field. As mentioned in the Introduction, there is no existing calculation of $\gamma(B)$ for $J=7/2$. However, the Wilson ratio has been shown[17] to be constant as a function of field for $J=1/2$. By numerically analyzing the calculated data[18] for $J=5/2$, we have determined that $\Re$ should be constant for this degeneracy. We assume that the Coqblin-Schrieffer theory for $J=7/2$ also will predict a constant $\Re(B)$. At low fields, then, we expect to have the relation $\gamma(B)/\gamma(0)=1+a_2x^2+a_4x^4$. The dashed curve, marked "constant Wilson ratio" in Fig. 3(b) corresponds to this expression and was obtained from $\gamma(0)(1+a_2x^2+a_4x^4)$ with $\gamma(0)=243$ mJ/mol K^2) and $T_L=77$ K. The experimental data increase more rapidly than predicted. This has two important implications: (1) The Wilson ratio is *not* constant but decreases by 4% over the interval 0–10 T. (2) The characteristic field as measured by the variation of $\gamma(B)/\gamma(0)$ is yet smaller than that determined from $M(B)$. The value $T_L=63$ K must be used to fit the data [solid line in Fig. 3(b)]. Hence, again, the theory overestimates the characteristic field.

We have the situation that the Bethe-ansatz solutions of the Coqblin-Schrieffer model give excellent agreement for the *shape* of the several measurements but incorrectly predict the relationship between the characteristic temperatures and fields. To summarize, the characteristic temperatures are 120, 98, 77, and 63 K as determined from $\chi(T)$, $\gamma(B=0)$, the low-temperature $\chi(B)/\chi(0)$, and $\gamma(B)/\gamma(0)$, respectively. In addition the Wilson ratio decreases in an applied field (by 4% at 10 T).

As mentioned in the Introduction, Schlottmann[14] was able to remove part of the discrepancy between values of T_L obtained from the measured values of $\gamma(B=0)$ and $\chi(T)$ by comparing the data to the predictions for the degenerate ($J=7/2$) Anderson model. In this treatment, the calculated ground-state valence, approximately 2.90, corresponds well to the measured value.[7] It is also possible that inclusion of crystal-field (CF) effects will alter the relationship between the characteristic temperature T_L and the characteristic field B_0. Polatsek and Bonville[27] have shown that the susceptibility and neutron-scattering line shape of $YbAgCu_4$ can be fit assuming a smaller hybridization (than for no CF splitting) and an overall crystal-field splitting of 80 K. It remains to be seen whether such treatments can also correctly predict the characteristic magnetic fields. Given the large discrepancy between the measured and calculated characteristic fields, we are skeptical that they can be so reconciled.

IV. MAGNETOTRANSPORT

The resistivity measured in five fields ranging from 0 to 18 T is plotted versus temperature in Fig. 4(a). The magnetoresistance is positive over these temperature and field intervals and becomes vanishingly small at higher temperatures as shown explicitly in Fig. 4(b).

In Fig. 5(a) we plot on a log-log scale the ratio

$$[\rho(B,T)-\rho(0,T)]/\rho(0,T)=\Delta\rho(B,T)/\rho(0,T)$$

versus the quantity Br_0 for $YbAgCu_4$ at several temperatures and compare these to data[28] obtained on polycrystalline indium at low temperatures. For the case of indium, the quantity r_0 is defined as the ratio $\rho(\Theta_D)/\rho_0$, where Θ_D is the Debye temperature and $\rho_0=\rho\ (T=0)$ is the residual resistivity. For $YbAgCu_4$ we set

$$r_0=[\rho(300\text{ K})-\rho(4\text{ K})]/\rho(T)$$

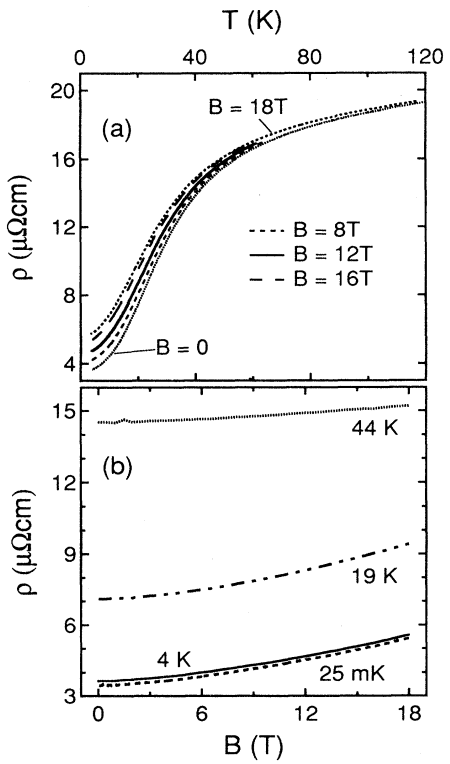

FIG. 4. (a) Resistivity of $YbAgCu_4$ as a function of temperature measured in several magnetic fields to 18 T. The magnetoresistance is positive at all temperatures and decreases substantially above 100 K. (b) Resistivity as a function of magnetic field at four different temperatures.

This formula assumes that $\rho(T)=\rho_t(T)+\rho_0$, i.e., $\rho_t(T)$ is the "true" resistivity in the absence of impurity effects, so that at 4 K $1/r_0$ measures the impurity residual resistivity in units of $\rho_t(300)$. From this plot [Fig. 5(a)] we conclude that below 60 K the magnetoresistance of $YbAgCu_4$ is comparable in magnitude and field dependence to that of indium. The data for 25 mK fall on top of that for In if r_0 for the former is multiplied by 1.4 This suggests that the positive magnetoresistance observed in Fig. 4 is primarily an ordinary impurity effect. We note that the low-temperature magnetoresistance varies with field as $B^{1.5}$ [dashed line in Fig. 5(a)]. If we assume that each atom contributes one conduction electron, then given 24 atoms per unit cell with a lattice constant of 7.07 Å,[6] the value of $\omega_c\tau$ (where ω_c is the cyclotron frequency and $1/\tau$ the scattering rate) is $B/ne\rho_0=0.05$ at low temperatures and 18 T. Hence, the system is in the small $\omega_c\tau$ limit, and this power law does not represent the effect of saturation.[28]

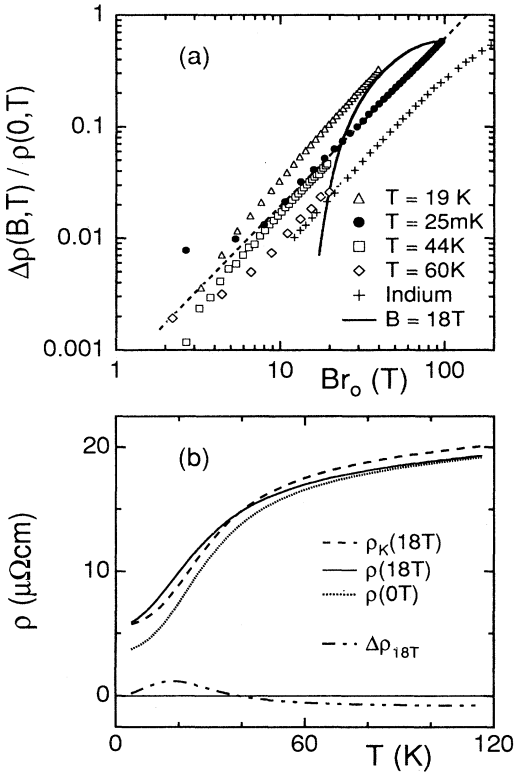

FIG. 5. (a) Magnetoresistance $\Delta\rho(B,T)/\rho(0,T)$ of $YbAgCu_4$ as a function Br_0 at constant temperature or at constant $B=18$ T. Data for polycrystalline indium at 4 K taken from Ref. 28 is also shown. For indium, r_0 stands for the resistivity ratio $\rho(\Theta_D)/\rho_0$, where Θ_D is the Debye temperature. For $YbAgCu_4$, $r_0=[R(300\text{ K})-R(4\text{ K})]/R(T)$, where R is the resistance (see text). The dashed line represents the behavior $\alpha(B/r_0)^{1.5}$ with $\alpha=6.5\times10^{-4}(T^{-1.5})$. (b) Measured resistivity for 0 and 18 T (dotted and solid lines). ρ_K (18 T) is the resistivity anticipated on the basis of Kohler's rule at $B=18$ T, if the scattering rate did not depend on field (dashed curve). $\Delta\rho_{18\text{ T}}$ is the extra resistivity taking into account the effect of a field-dependent scattering rate (dashed line, see text). $\Delta\rho_{18\text{ T}}$ has the magnitude and sign expected for the magnetoresistivity of the Anderson lattice.

We test next whether or not the magnetoresistance obeys Kohler's rule

$$\Delta\rho(B,T)/\rho(0,T)=f(B/\rho(0,T)) .$$

A necessary (though not sufficient) condition for this is that the scattering rate is not affected by the field, whose role then is simply to create cyclotron orbits.[23,24] In Fig. 5(a) the data do not collapse onto a single line. Therefore, Kohler's rule does not apply as the temperature is raised. In many metals, phonon scattering causes deviations from Kohler's rule, which, in general, are monotonic as a function of temperature.[28] However, a plot of $\Delta\rho(18,T)/\rho(0,T)$ [line in Fig. 5(a)] shows that the measured resistance at 18 T is larger than expected on the basis of Kohler's rule at low temperatures and smaller at high temperatures. As discussed in the Introduction, this is predicted by the theory of magnetoresistance in a Kondo lattice when a pseudogap is present.[20]

To determine the magnitude of the effect, we must separate out the magnetoresistance due to normal processes. This is an extremely difficult problem even for ordinary metals[24] because it depends in detail on the band structure, any momentum (Q) dependence of the impurity scattering, and on the Q and frequency dependence of the phonon scattering. For this reason, we attempt only a qualitative analysis. We assume that deviations from Kohler's rule arise entirely from the field dependence of the scattering rate. In the absence of these deviations, if $\rho(T)=\rho_t(T)+\rho_0$ is the zero-field resistivity, then by Kohler's rule the measured resistivity at finite field should be

$$\rho_K(B,T)=[\rho_t(T)+\rho_0]\left[1+\left[\frac{\alpha B}{\rho_t(T)+\rho_0}\right]^{1.5}\right] . \quad (1)$$

This quantity is plotted as the dashed line in Figs. 5(a) and 5(b). The coefficient α is determined from the 25-mK data in Fig. 5(a). Figure 5(b) again emphasizes that the deviations from the expected Kohler resistivity are positive at low temperature and negative at higher temperature. The effect of the field on the scattering rate is to change $\rho(T)=\rho_t(T)+\rho_0$ by an amount $\Delta\rho_B(T)$ so that the measured resistance in field is

$$\rho(B,T)=[\rho_t(T)+\rho_0+\Delta\rho_B(T)]\times\left[1+\left[\frac{\alpha B}{\rho_t(T)+\rho_0+\Delta\rho_B(T)}\right]^{1.5}\right] , \quad (2)$$

i.e., $\rho_t(T)+\rho_0+\Delta\rho_B(T)$ is the resistivity that would be measured in absence of field (and hence, no cyclotron orbits) but with the scattering rate being the same as in presence of a field. The correction $\Delta\rho_B(T)$ for field-dependent scattering was determined by solving Eq. (2) iteratively. The result is shown as the dashed-dotted line in Fig. 5(b). For comparison to theory, we note that, for 18 T and $T_L=120$ K, $g\mu_B B/k_B T_L=0.1$. The experimental $\Delta\rho_B(T)$ is very similar to the predicted curve for this value of B/T_L shown in Fig. 3 of the paper by

Kawakami and Okiji[20] for the case that the Fermi level lies below a pseudogap in the f density of states. The measured $\Delta\rho_B(T)$ changes sign near $T=0.3T_L$ and reaches a maximum value $\Delta\rho_B(T)/\rho_0\approx 0.17$, compared to the values $0.2T_L$ and 0.05 in theory. Given the qualitative nature of our result and of the theory (which treats a spin-$\frac{1}{2}$ Anderson lattice in the symmetric case $U=-2\varepsilon_f$ and in the single-site approximation), this order of magnitude agreement is quite encouraging.

The field-dependent low-temperature resistivity of many heavy-fermion compounds varies as $\rho(B,T)=\rho_0(B)+A(B)T^2$.[29] That $YbAgCu_4$ exhibits this behavior can be seen from Fig. 6(a). Kadowaki and Woods[30] argue that the ratio A/γ^2 has a common value of 1.0×10^{-5} $\mu\Omega$ cm(mole K/mJ)^2 in heavy fermions.

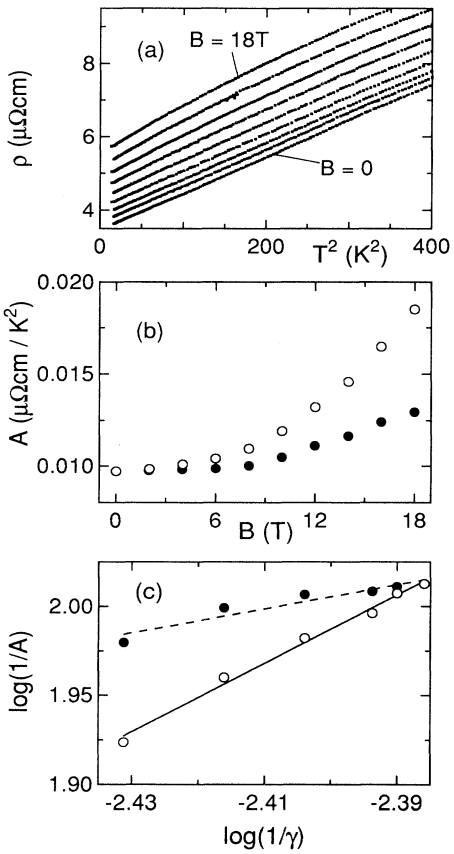

FIG. 6. (a) Low-temperature resistivity of $YbAgCu_4$ as a function of the square of the temperature at magnetic fields of 0,4,6, . . . , 18 T increasing from bottom to top. (b) Measured T^2 coefficient of resistivity A (solid symbols) and corrected value A_t (open symbols) as a function of magnetic field. A_t depends only on the field-dependent scattering rate, but not on the effect of cyclotron orbits. (c) Comparison of the measured $A(B)$ (solid symbols) and corrected $A_t(B)$ (open symbols) to the T-linear coefficient of specific heat $\gamma(B)$. The lines represent the relations $A(B)=2.3\times10^{-4}\gamma^{0.67}$ and $A_t(B)=2.5\times10^{-7}\gamma^{1.925}$, which is essentially $A_t/\gamma^2\approx$constant. The units of A are $\mu\Omega$ cm/K^2 and of γ are mJ/(mole K^2), and the logarithms are base 10.

This implies that $A\approx 1/T_K^2$, which is expected to be true on very general grounds for heavy fermions. It also implies that the prefactor ρ_s in the expression $\rho(T)=\rho_s(T/T_L)^2=AT^2$ should have a universal value for heavy fermions, which is an assumption that seems extremely unlikely, since ρ_s should depend on nonuniversal details of the band structure. Indeed, we find a value $A(B=0)=0.01$ $\mu\Omega$ cm/K^2 from the data of Fig. 6(a). Given $\gamma=243$ mJ/mole K^2, we obtain $A/\gamma^2=1.67\times10^{-7}$ $\mu\Omega$ cm(mole K/mJ)^2, which is a factor of 60 smaller than predicted by Kadowaki and Woods.[30] An obvious source of error in determining this ratio is the geometric factors needed to convert the resistance to resistivity. For our sample this should be a 20% effect, but not a factor of 60. Hence, we do not believe that this ratio is universal. However, we do believe that it can be universal for a given compound as T_L is varied, e.g., by application of pressure, and we have shown[31] that this is the case for $CeCu_6$. This raises the issue whether the ratio A/γ^2 is constant as a function of magnetic field in $YbAgCu_4$.

Figure 6(b) shows the coefficient A as a function of B (solid symbols). Because there is considerable curvature in plots of ρ versus T^2 at the higher fields [Fig. 6(a)], we have fit only to the lowest-temperature data (4–11 K) to determine these coefficients. Although A increases with the field, it clearly does not do so as rapidly as γ. Indeed, it can be seen from Fig. 6(c) (solid symbols) that A increases as $\gamma^{2/3}$. However, these data have not been corrected for the effect of cyclotron orbits. To accomplish this, we again assume that deviations from Kohler's rule arise only from the change in scattering rate in field, i.e., from the effect on A in field. Since the coefficient is derived in the $T\rightarrow0$ limit, this is a reasonable assumption. To determine the true value $A_t(B)$, we solve the formula

$$\rho(B,T)=[\rho_0(B)+A_t(B)T^2]\times\left[1+\left[\frac{\alpha B}{\rho_0(B)+A_t(B)T^2}\right]^{1/5}\right]. \quad (3)$$

As $T\rightarrow0$ the true $A_t(B)$ [open circles in Fig. 6(b) and 6(c)] is related to the measured value A (closed symbols) by the relation

$$A_t=A\Big/\left[1-\frac{\Delta\rho(B,0)}{2\rho_0}\right].$$

The solid line in Fig. 6(c) represents the relation $A_t=2.5\times10^{-7}\gamma^{1.925}$, which is in much better agreement with the Kadowski-Woods relation[30] $A\propto\gamma^2$ than for the case of the as-measured data. This increases our confidence that the use of Kohler's rule to obtain the field-dependent scattering rate [i.e., Eq. (3)] is sound. To the best of our knowledge, it has not been previously established that $A(B)/\gamma(B)^2$ should be constant for heavy fermions, neither experimentally nor theoretically.

We next consider the effect of impurities on the magnetoresistance. In particular, we examine whether the concepts of a Kondo hole or of ligand disorder, discussed in the Introduction, are applicable. To this end, we examine

the cases of $YbAgCu_4$ alloys $YbAgCu_{3.99}Ni_{0.01}$, $YbAgCu_{3.98}Ni_{0.02}$, and $Yb_{0.96}Lu_{0.04}AgCu_4$. The susceptibilities of these alloys are shown in Fig. 1. We note first that alloying results in a Curie tail; the effective moment per mole Lu calculated from this Curie tail gives $0.8\mu_B$/Lu, which is equivalent to 0.2 times the effective moment of a free-Yb ion. This moment could arise either from a Kondo-hole effect, whereby the Kondo hole has an effective moment comparable to a crystal-field-split Yb atom, or from a ligand-disorder effect whereby the Lu atom causes a smaller Kondo temperature (and hence enhanced susceptibility) on the 12 neighboring Yb sites. Doping with 1% Ni gives a Curie tail comparable to that for 4% Lu. The tail is proportional to the Ni content, and its magnitude is comparable to that expected for local moments on the nickel sites. Alternatively, if there are no local moments on the nickel sites, it could arise again from a ligand-disorder effect.

In Fig. 7(a) we compare the magnetoresistance at 4 K for these alloys to that of $YbAgCu_4$. Alloying decreases the magnetoresistance, but the effect increases uniformly as the resistance ratio r_0 decreases, which suggests that the decrease is primarily due to a decrease in $\omega_c\tau \approx Br_0$. In Fig. 7(b) we plot $\Delta\rho/\rho_0$ versus Br_0, which shows that the deviations from Kohler's rule are *positive* at 18 T. Because Kohler's rule works best for comparing the low-temperature magnetoresistance of a system as impurities are added,[28] and because at these temperatures phonons do not complicate the picture, it seems very likely that the impurities give rise to a field-dependent scattering rate, as expected on the basis of the ligand-disorder effect.[22] The effect should be proportional to $xn\rho_u(gJ\mu_B B/k_B T_L)^2(\delta T_L/T_L)^2$, where x is the impurity concentration, ρ_u the unitary limit resistivity, n the number of Yb atoms whose Kondo temperatures are affected by the ligand impurity, and δT_L the change in Kondo temperature at the affected sites. To quantify this, we extract the change in resistivity due to the field-dependent scattering rate by solving the equation

$$\rho(B,0)=[\Delta\rho_l(B)+\rho_0]\left[1+\left[\frac{\alpha B}{\Delta\rho_l(B)+\rho_0}\right]^{1.5}\right] \quad (4)$$

for $\Delta\rho_l(B)$. Here, $\rho(B,0)$ is the measured value and $\Delta\rho_l(B)+\rho_0$ is the resistivity of the alloy with a field-dependent scattering rate but without cyclotron orbits. We plot $\Delta\rho_l(18\text{ T})/\rho_t(300\text{ K})$ against the alloy parameter x in the inset to Fig. 7(b). The solid line represents the prediction

$$\Delta\rho_l(B)/\rho_t(300)=xn\frac{\rho_u}{\rho_t(300)}\left[\frac{\delta T_L}{T_L}\right]^2\left[\frac{gJ\mu_B B}{k_B T_L}\right]^2 \quad (5)$$

for $B=18$ T, $T_L=120$ K, and $n[\rho_u/\rho_t(300)](\delta T_L/T_L)^2 = 18$. This is a reasonable value for the last quantity: $\delta T_L/T_L$ should be of order unity if the Yb atoms neighboring the ligand have their Kondo temperatures reduced to very small values, $\rho_u/\rho(300)$ should be of order 2–3, and it is likely that ≈ 10 Yb atoms are affected by the ligand impurity. Hence, while the magnitude of the measured magnetoresistance in these alloys is that expected on the basis of ordinary impurity scattering, deviations from Kohler's rule have the right magnitude to be explained as a ligand-disorder effect. This holds even for $Yb_{0.96}Lu_{0.04}AgCu_4$, where we might expect a *negative* magnetoresistance due to a Kondo-hole mechanism.

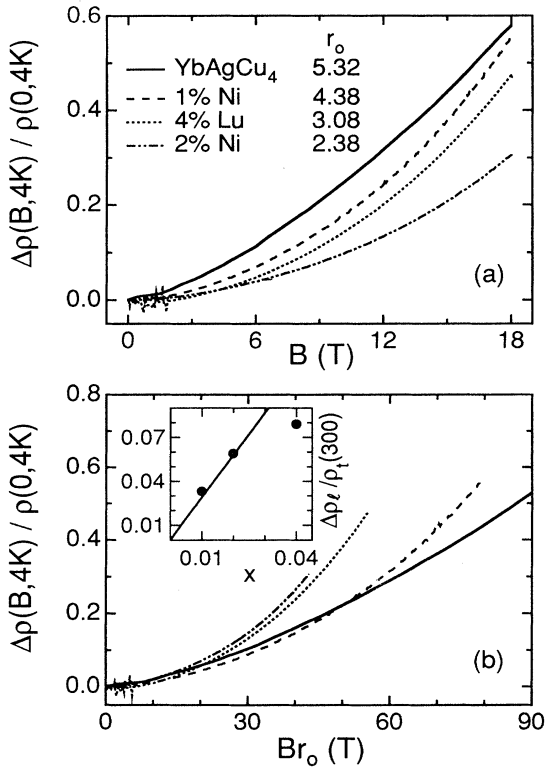

FIG. 7. (a) Magnetoresistance of $YbAgCu_4$, $YbAgCu_{3.99}Ni_{0.01}$, $YbAgCu_{3.98}Ni_{0.02}$, and $Yb_{0.96}Lu_{0.04}AgCu_4$ at 4 K. The values of the normalized resistivity ratio r_0 are given for each compound. The magnitude of the magnetoresistance at 18 T decreases uniformly as r_0 decreases. (b) Magnetoresistance plotted vs Br_0. Under the assumption that positive deviations from the curve for $YbAgCu_4$ represent a ligand-disorder effect on the field-dependent scattering rate, we have determined the magnitude of the extra magnetoresistance $\Delta\rho_l$ (18 T) induced by the ligand effect (see text). The quantity $\Delta\rho_l(18\text{ T})/\rho_t(300\text{ K})$ is plotted vs alloy parameter x in the inset. In the text we argue that the magnitude of the linear increase (solid line) is as expected for a ligand-disorder effect.

V. SUMMARY

Perhaps the most significant (and disconcerting) conclusion from this work is that the characteristic temperature T_L differs by nearly a factor of 2, depending on which measurement was used to extract T_L. Though theory appears to describe well the functional dependence of the magnetization and Sommerfeld coefficient on applied field, this discrepancy in values of T_L is well outside experimental uncertainty even considering that "background" contributions to $C(B,T)$ and $M(B,T)$ have not been subtracted, and emphasizes the need for extreme caution in drawing quantitative conclusions from fitting a given measurement to theory. The origin of these

discrepancies is not understood. Throughout we have assumed that the $4f$ occupancy is field independent and that impurity models are applicable to the lattice case. In the absence of direct measurements of $n_f(B)$, we cannot state unambiguously that our assumption about n_f is valid, but it is very unlikely that the $4f$ occupancy changes enough to account for the large difference in values of T_L. The possibility that a periodic Anderson lattice responds differently to field than a simple impurity is real. Clearly, additional theoretical and experimental work is needed. In particular, useful calculations should include the mixed-valence nature of the $4f$ ion and the effect of crystal fields and should be extended to the Anderson lattice case to allow a more direct comparison to experiment. Experimentally, a systematic study of the field dependence of thermodynamic properties of heavy-fermion–mixed-valent compounds is needed to establish the generality of effects we have observed as well as to determine $n_f(B)$ directly. Particularly useful would be measurements of the field-dependent spin dynamics, e.g., by performing inelastic neutron-scattering measurements in magnetic fields comparable to those used in this study.

Because electrical transport is not a thermodynamic quantity, it is much more difficult to calculate and depends most definitely on lattice periodicity. We have shown that in $YbAgCu_4$, which has a relatively large characteristic temperature compared to very heavy-fermion compounds, the magnetoresistance at low temperatures is dominated by simple potential scattering from defects, such as grain boundaries, etc. However, deviations from Kohler's rule suggest the existence of scattering processes expected for an Anderson lattice. A detailed, though qualitative, analysis of the magnetoresistance of $YbAgCu_4$ shows relatively good agreement with predictions for a spin-$\frac{1}{2}$ Anderson lattice, once conventional contributions are taken into account. We would expect these "unconventional" contributions to be significantly more pronounced in materials with a much smaller T_L. We also have established, at least for $YbAgCu_4$, that the field dependence of the T^2 coefficient of resistivity tracks field-induced changes in γ, i.e., that $A(B) \propto \gamma^2(B)$. Introduction of disorder, either through substitution on the $4f$ or ligand sites, accentuates deviations from Kohler's rule, scaling approximately with the degree of disorder. A theory of ligand-disorder effects[22] accounts reasonably well for the experimental observations.

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*Permanent address: Department of Physics, University of California, Irvine, CA 92717.

†Permanent address: San Diego State University, San Diego, CA.

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