Heavy-fermion systems in magnetic fields: The metamagnetic transition

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Heavy fermions have a large number of low-lying excitations. Antiferromagnetic superexchange typically favors low-spin arrangements for the ground state. A magnetic field favors high-spin arrangements over low-spin arrangements. The transition from a low-spin ground state to a high-spin ground state, as a function of magnetic field, passes through a range where there is a peak in the many-body density of states. This range qualitatively describes the metamagnetic transition.

I. INTRODUCTION

Heavy-fermion systems have been an active area of research for both experimentalists[1] and theorists[2–4] since their discovery in the mid 1970s. Heavy-fermion systems are characterized by huge coefficients (γ) to the term linear in T in the specific-heat, quasielastic spin excitations (large magnetic susceptibility), and poor metallic conductivity. These features may be qualitatively described by a Fermi liquid with a very large density of states at the Fermi level.[2–4] Heavy-fermion systems may become superconductors (UPt_3, UBe_{13}, $CeCu_2Si_2$, URu_2Si_2, etc.), possess long-range magnetic order (UPt_3, URu_2Si_2, $NpBe_{13}$, U_2Zn_{17}, etc.), or remain paramagnetic metals ($CeRu_2Si_2$, $CeAl_3$, $CeCu_6$, etc.) at low temperatures.

Recent experimental work has concentrated on the properties of heavy-fermion systems in high magnetic fields.[5–8] A "transition" is observed (the so-called metamagnetic transition) at a characteristic magnetic field B_c in $CeRu_2Si_2$ ($B_c=7.8$ T), UPt_3 ($B_c=21$ T), and URu_2Si_2 ($B_c=36$ T). The transition is characterized by a magnetic-field dependence of the coefficient γ, the elastic coefficients, and the magnetic properties. At the critical field B_c, the coefficient γ has a single peak, the elastic coefficients are softened, and the magnetic fluctuations change character. The magnetization shows a steplike structure as a function of magnetic-field strength. This contribution presents a many-body theory (*without* the assumptions of Fermi-liquid theory) that describes all of the above electronic properties of heavy-fermion systems (except superconductivity) and their field dependence.

Every heavy-fermion system is composed of ions with localized f orbitals (lanthanides and actinides) that do not overlap with the corresponding f orbitals on neighboring ions, but do hybridize with the extended states of the conduction-band electrons. The f electrons interact very strongly with each other via a screened (on-site) Coulomb interaction U that acts only between two f electrons that are localized about the same lattice site. Double-occupied f orbitals are effectively forbidden, since the Coulomb energy is larger than any other energy in the problem ($U > 10$ eV). The physics of such an electronic system is described by the lattice (or periodic) Anderson impurity model[9]

$$H_A=\sum_{k,\sigma}\varepsilon_k a_{k\sigma}^\dagger a_{k\sigma}+\varepsilon\sum_{i,\sigma}f_{i\sigma}^\dagger f_{i\sigma}+U\sum_i f_{i\uparrow}^\dagger f_{i\uparrow}f_{i\downarrow}^\dagger f_{i\downarrow}+\sum_{i,k,\sigma}[V_{ik}f_{i\sigma}^\dagger a_{k\sigma}+V_{ik}^* a_{k\sigma}^\dagger f_{i\sigma}] \quad (1)$$

in the large-U ($U\to\infty$) limit.[10] The parameters and operators in Eq. (1) include the conduction-band creation (annihilation) operators $a_{k\sigma}^\dagger$ ($a_{k\sigma}$) for a conduction electron in an extended state with wave vector k, spin σ, and energy ε_k; the localized electron[11] creation (annihilation) operators $f_{i\sigma}^\dagger$ ($f_{i\sigma}$) for localized electrons in an atomic orbital centered at lattice site i with energy ε; the on-site Coulomb interaction U; and the hybridization integral V_{ik} that mixes together the localized and extended states. The hybridization matrix elements are assumed to be of the form

$$V_{ik}=\exp(i\mathbf{R}_i\cdot\mathbf{k})Vg(k)/\sqrt{N}\ , \quad (2)$$

with $g(k)$, the form factor, a dimensionless function of order 1 and N the number of lattice sites. The Fermi level E_F is defined to be the maximum energy of the filled conduction-band states, in the limit $V\to 0$, and the origin of the energy scale is chosen at the Fermi level, $E_F=0$. The conduction-band density of states per site at the Fermi level is defined to be ρ.

Heavy-fermionic behavior may occur in the restricted region[12] of parameter space where $-V\rho \ll \varepsilon\rho \ll -V^2\rho^2<0$. The localized orbitals are *almost* singly occupied ($\langle f_{i\uparrow}^\dagger f_{i\uparrow}+f_{i\downarrow}^\dagger f_{i\downarrow}\rangle=1-\nu$; $\nu\ll 1$) and the conduction electron density of states at the Fermi level is small. In this case, the kinetic energy of the holes that hop within a narrow "effective" band dominates over the magnetic spin-spin interactions and the spin-flipping terms of the Kondo effect. The Anderson Hamiltonian (1) may be mapped onto the large-U limit of the Hubbard[13] Hamiltonian which, in turn, may be mapped onto a t-J model[14]

$$H_{t\text{-}J}=-\sum_{i,j,\sigma} t_{ij}(1-f^\dagger_{i-\sigma}f_{i-\sigma})f^\dagger_{i\sigma}f_{j\sigma}(1-f^\dagger_{j-\sigma}f_{j-\sigma}) +\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \; . \tag{3}$$

The unrenormalized (bare) hopping matrix t_{ij} satisfies

$$t_{ij}=\sum_k \frac{V^*_{ik}V_{jk}}{\varepsilon_k-\varepsilon}=\frac{V^2}{N}\sum_k \frac{g^2(k)}{\varepsilon_k-\varepsilon}e^{-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)} , \tag{4}$$

and the antiferromagnetic superexchange is defined to be $J_{ij}\equiv 4|t_{ij}|^2/U$. It should be noted that the canonical transformation that maps the Anderson model onto the t-J model is valid only within a narrow region of parameter space (for details see Sec. II).

In Sec. II the Schrieffer-Wolff transformation[12] is employed to establish the mapping of the Anderson model onto the t-J model. In Sec. III the exact solution of the t-J model on a finite cluster is analyzed in a magnetic field illustrating both heavy-fermionic and metamagnetic behavior. A brief discussion of these results follows in Sec. IV.

II. SCHRIEFFER-WOLFF TRANSFORMATION AND THE t-J MODEL

The relationship between the lattice Anderson impurity model (1) and the t-J model (3) is not widely known, and frequently misunderstood. The equivalence may be established by employing a Schrieffer-Wolff[12] canonical transformation to the lattice Anderson impurity model. Since the details of this transformation are well known,[12] only an outline is given here.

The Anderson Hamiltonian H_A is divided into two terms $H_A=H_0+H_{\rm hyb}$, where $H_{\rm hyb}$ is the last term in Eq. (1). A canonical transformation $H'=\exp(S)H_A\exp(-S)$ is performed with S chosen to satisfy $[H_0,S]=H_{\rm hyb}$. One finds (to lowest order in V) that $H'=H_0+\frac{1}{2}[S,H_{\rm hyb}]$ or

$$H'-H_0=-\sum_{i,k,k'} J_{iikk'}\psi^\dagger_{fi}\boldsymbol{\sigma}\psi_{fi}\cdot\psi^\dagger_k\boldsymbol{\sigma}\psi_{k'} \tag{5a}$$

$$-\sum_{i,i',k,\sigma}[W_{ii'kk}+\tfrac{1}{4}J_{ii'kk}(f^\dagger_{i-\sigma}f_{i-\sigma}+f^\dagger_{i'-\sigma}f_{i'-\sigma})]f^\dagger_{i\sigma}f_{i'\sigma} \tag{5b}$$

$$+\sum_{i,k,k'}[W_{iikk'}+\tfrac{1}{4}J_{iikk'}\psi^\dagger_{fi}\psi_{fi}]\psi^\dagger_k\psi_{k'} \tag{5c}$$

$$+\tfrac{1}{4}\sum_{i,k,k',\sigma}[K_{iikk'}f^\dagger_{i\sigma}f^\dagger_{i-\sigma}a_{k\sigma}a_{k'-\sigma}+\text{H.c.}] , \tag{5d}$$

where the spinors are defined to be

$$\psi_k\equiv\begin{bmatrix} a_k\uparrow \\ a_k\downarrow\end{bmatrix},\quad \psi_{fi}\equiv\begin{bmatrix} f_i\uparrow \\ f_i\downarrow\end{bmatrix}, \tag{6}$$

$\boldsymbol{\sigma}$ denotes the Pauli spin matrices, and the coefficients are

$$J_{ii'kk'}\equiv V_{ik}V^*_{i'k'}\left[\frac{1}{\varepsilon_k-\varepsilon-U}+\frac{1}{\varepsilon_{k'}-\varepsilon-U}-\frac{1}{\varepsilon_k-\varepsilon}-\frac{1}{\varepsilon_{k'}-\varepsilon}\right], \tag{7a}$$

$$K_{ii'kk'}\equiv V_{ik}V_{i'k'}\left[\frac{1}{\varepsilon_k-\varepsilon-U}+\frac{1}{\varepsilon_{k'}-\varepsilon-U}-\frac{1}{\varepsilon_k-\varepsilon}-\frac{1}{\varepsilon_{k'}-\varepsilon}\right], \tag{7b}$$

$$W_{ii'kk'}\equiv\tfrac{1}{2}V_{ik}V^*_{i'k'}\left[\frac{1}{\varepsilon_k-\varepsilon}+\frac{1}{\varepsilon_{k'}-\varepsilon}\right]. \tag{7c}$$

The last term (5d) can always be neglected (in the large-U limit) because it only connects configurations that have zero electrons at site i to configurations with two electrons at site i, which are explicitly forbidden.

The canonical transformation of the lattice Anderson impurity model is approximated well by the lowest-order term in V, Eq. (5), when $|V^2\rho/\varepsilon|\ll 1$. (Higher-order terms both renormalize the parameters J, K, and W and introduce new interactions.) In this region of parameter space the localized orbitals have an occupancy per site (a) close to one ($\varepsilon<0$) or (b) close to zero ($\varepsilon>0$).

(a) When the occupancy is close to one, the operator $\psi^\dagger_{fi}\psi_{fi}$ may be replaced by unity and both the term (5c) and the diagonal ($i=i'$) terms in (5b) may be absorbed into a renormalized H_0. The remaining terms in Eq. (5) describe spin scattering of the conduction electrons at the Fermi level by the localized moments (5a), and direct hopping terms within the (narrow) f band (5b).

(b) When the occupancy is close to zero, the operators $\psi^\dagger_{fi}\psi_{fi}$, $\psi^\dagger_{fi}\boldsymbol{\sigma}\psi_{fi}$, and $f^\dagger_i f_{i'}$ may be all replaced by zero. The only important terms remaining in Eq. (5) are the changes in the one-particle band structure arising from (5c).

Only regime (a) is of interest for the current purposes.

The magnitude of each of the interaction terms in Eq. (5) may be estimated by using the bare values of the interaction parameters in (7). The magnetic interactions (mediated through the conduction-band electrons) fall into two categories,[15] a superexchange interaction of order V^4/ε^3 and a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction[16] of order $V^4\rho/\varepsilon^2$. The magnitude of the kinetic energy may be estimated to be also of order $V^4\rho/\varepsilon^2$ determined by multiplying the effective hopping integral (4) by the hole concentration ($\nu \approx V^2\rho/\varepsilon$). Therefore, in the bare theory (truncating the Schrieffer-Wolff transformation to lowest order) the magnetic interactions and the hole kinetic energy are the *same order of magnitude.*

The Kondo effect[17] arises from a resonance of the localized electrons with the conduction electrons that quenches the localized magnetic moments. It is a nonperturbative effect which is small in magnitude and is not directly present in any model (such as the *t-J* model) that projects out the conduction-electron degrees of freedom. The physics of the Kondo effect in the presence of a single impurity is well known.[17] The case of a lattice of impurities is still controversial.[2-4] Large-N expansions show a quenching of magnetic moments everywhere[18] but are always in the atomic limit[2] and do not illustrate what happens in the dense "impurity" limit. The Kondo effect can be treated (in a mean-field sense) directly in the *t-J* model by renormalizing the magnetic interactions J, energetically taking into account a partial quenching of the local moments.

The renormalized Schrieffer-Wolff transformation of the lattice Anderson model is then well described by a *t-J* model in the limit $|V^2\rho/\varepsilon| \ll 1$. When $\rho\varepsilon \ll -V\rho < 0$, the renormalized magnetic interactions J between the local moments of the f electrons dominate. The local moments interact with each other via all forms of exchange interactions, which determine, at low enough temperatures, the long-range magnetic order. As $\rho\varepsilon$ increases, two effects occur: (i) the kinetic energy of the holes in the narrow f band become important; and (ii) a residual Kondo effect begins to quench the local magnetic moments. In this regime,

$$-V\rho \ll \varepsilon\rho \ll -V^2\rho^2 , \tag{8}$$

the Anderson model is approximated well by the full *t-J* model. The conduction electrons are decoupled from the f band and act only as a buffer that determines the filling of the f band. This picture is supported by numerical evidence found in exact solutions[19-21] of the lattice Anderson model on four-site clusters (see the next section).

III. HEAVY-FERMIONIC BEHAVIOR IN THE *t-J* MODEL

A heavy-fermion system is characterized by a many-body ground state with a very large number of low-lying excited states that have many different spin configurations (a partial decoupling of spatial and spin degrees of freedom). The localized states broaden into a strongly correlated narrow band in which all electronic transport takes place; the conduction band is (effectively) decoupled and acts only as a buffer that determines the concentration of electrons in the narrow band. The formation of a heavy-fermion ground state (and its low-lying excitations) requires a fine tuning of the parameters in the (effective) *t-J* model and depends strongly upon the geometry and connectivity of the lattice.

One way to study the formation of a many-body ground state that possesses the properties of a heavy-fermion system (without any *a priori* assumptions of Fermi-liquid behavior) is to diagonalize exactly the many-body problem for small systems—the so-called small-cluster approach.[22] This approach to the many-body problem begins with the periodic crystal approximation (replacing an infinite lattice by a lattice with N sites and periodic boundary conditions) with a *small* number of inequivalent sites. The cluster is chosen to be small enough that the many-body Hamiltonian may be exactly diagonalized but (hopefully) large enough that the physics of the infinite lattice is captured. The mapping of the Anderson model (1) onto the *t-J* model (3) reduces the size of the Hilbert space by a factor of $(3/16)^N$, which allows larger clusters to be studied. An understanding of exactly how to extrapolate the results for a small-cluster calculation to the thermodynamic limit ($N \to \infty$) has not yet been found.

A. Mapping of the lattice Anderson model to the *t-J* model

The lattice Anderson impurity model [Eq. (1)] has been studied[19-21] for various small clusters with at most four sites (for a review see Ref. 23). The results for the tetrahedral cluster[20,21] (with one electron per site) illustrate the formation of the heavy-fermionic state and how sensitive it is to variations in the parameters. When the band structure ε_k is such that the bottom of the band is at the Γ point of the face-centered-cubic Brillouin zone, a small range of values for ε is found where the ground state is a spin singlet with (nearly degenerate) triplet and quintet excitations. The specific heat has a huge low-temperature peak and the magnetic susceptibility is large. When Γ is the top of the conduction band, a magnetically ordered heavy-fermionic state is sometimes observed.

B. A heavy-fermion case in a small-cluster *t-J* model

The small-cluster approach has also been applied to the *t-J* model.[24] A very good example of a heavy-fermion system lies in an eight-site face-centered cubic-lattice cluster with seven electrons.[24] When the hopping parameters and antiferromagnetic superexchange parameters are chosen to be

$$t_{ij} = \begin{cases} t > 0, & i,j = \text{first-nearest neighbors} \\ t' = 0.1t, & i,j = \text{second-nearest neighbors} \\ 0 & \text{otherwise,} \end{cases}$$
$$J_{ij} = \begin{cases} J, & i,j = \text{first-nearest neighbors} \\ 0 & \text{otherwise,} \end{cases} \tag{9}$$

then the many-body eigenstates possess a low-energy manifold of 96 states (out of a total of 1024 states) that is split off from the higher-energy excitations and which includes many different spin configurations (see Table I). These many-body states are degenerate at $J=0$ but the degeneracy is partially lifted for finite J, with low-spin configurations favored (energetically) over high-spin configurations.

A magnetic field (in the z direction) partially lifts the

TABLE I. Low-energy manifold of many-body eigenstates, at zero magnetic field, for the model heavy-fermion system discussed in the text. The notation is that of Ref. 24.

Energy	Total spin	Degeneracy	Spatial symmetry label
$-6t+6t'-3J$	$\frac{1}{2}$	14	$\Gamma_2 \oplus X_1 \oplus X_2$
$-6t+6t'-2J$	$\frac{1}{2}$	16	L_3
$-6t+6t'-\frac{3}{2}J$	$\frac{3}{2}$	32	$\Gamma_{12} \oplus X_1 \oplus X_2$
$-6t+6t'-\frac{1}{2}J$	$\frac{3}{2}$	16	L_2
$-6t+6t'+J$	$\frac{5}{2}$	18	X_2

degeneracy even more, since the many-body eigenstates with z component of spin m_z have an energy

$$E(B)=E(0)-m_z g\mu_B B \equiv E(0)-m_z bJ \tag{10}$$

in a magnetic field B. The symbols g, μ_B, and b denote the Landé g factor, Bohr magneton, and dimensionless magnetic field, respectively. The high-spin eigenstates are energetically favored in a strong magnetic field and level crossings occur as a function of b.

C. The metamagnetic transition

The phenomena described above are all the necessary ingredients for a metamagnetic transition. The heavy-fermion system is described by a ground state with nearly degenerate low-lying excitations of many different spin configurations. The antiferromagnetic superexchange pushes high-spin states up in energy with splittings on the order of J. The magnetic field pulls down these high-spin states (with maximal m_z) and generates level crossings in the ground state. In the region near the level crossings, there is an increase in the density of low-lying excitations that produces a peak in the specific heat as a function of b. The magnetization and spin-spin correlation functions both change abruptly at the level crossings.

To illustrate the metamagnetic transition for the simple model above, the specific heat and magnetization are calculated as a function of the magnetic field (at a fixed low temperature). The specific heat satisfies

$$\frac{C_V(b)}{k_B}=\beta^2\left[\frac{\sum_n E_n^2\exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)}-\left\{\frac{\sum_n E_n\exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)}\right\}^2\right], \tag{11}$$

where k_B is Boltzmann's constant, β is the inverse temperature ($\beta \equiv 1/k_B T$), and E_n is the energy of the nth many-body eigenstate in a magnetic field b (the summations are restricted to the 96 eigenstates in Table I). Similarly the magnetization is expressed by

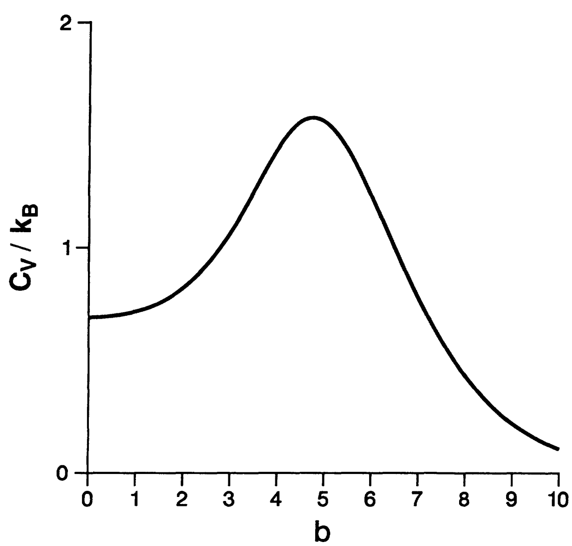

FIG. 1. Calculated specific heat as a function of magnetic field for the heavy-fermion model discussed in the text. The temperature is fixed at $T=J/k_B$. The horizontal axis contains the dimensionless magnetic field and the vertical axis contains the dimensionless specific heat C_V/k_B. Note the single peak in the specific heat, characteristic of the high-temperature regime (temperature larger than the energy-level spacings).

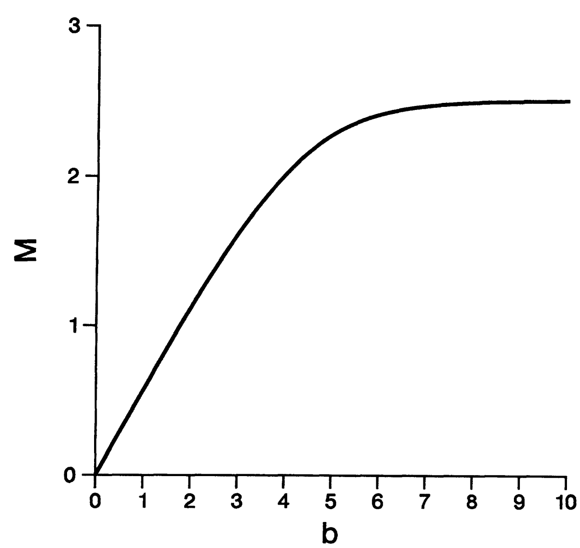

FIG. 2. Calculated magnetization as a function of magnetic field at a temperature $T=J/k_B$. Note the smooth transition in the magnetization, characteristic of the high-temperature regime.

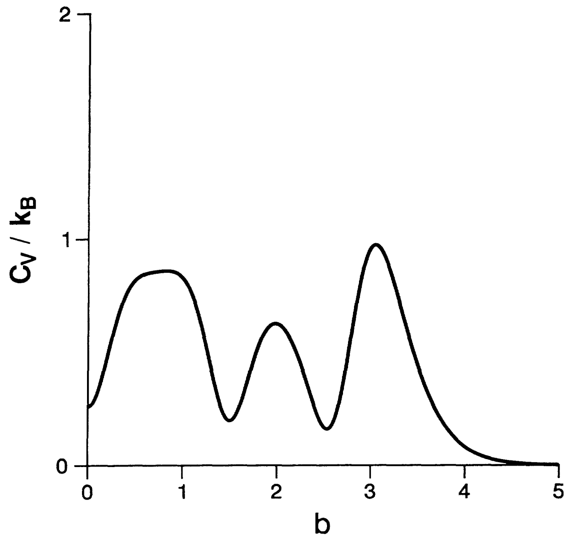

FIG. 3. Calculated specific heat as a function of magnetic field at a temperature $T=J/5k_B$. Note the multipeak structure in the specific heat, characteristic of the low-temperature regime (temperature smaller than the energy-level spacings).

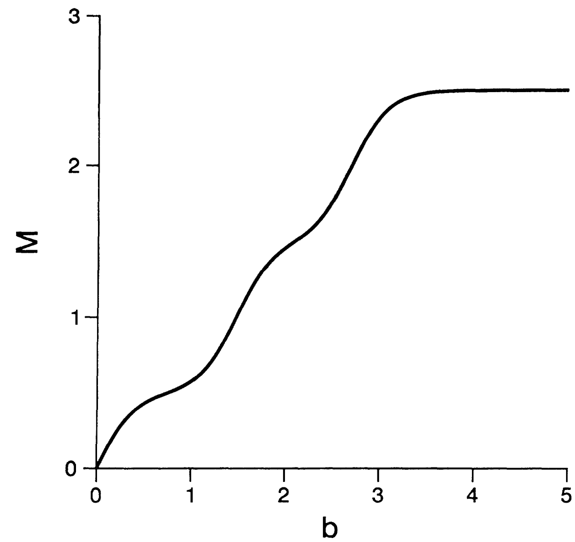

FIG. 4. Calculated magnetization as a function of magnetic field at a temperature $T=J/5k_B$. Note the steplike transitions in the magnetization at each level crossing, characteristic of the low-temperature regime.

$$M(b)=\frac{\sum_n m_z \exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)}, \tag{12}$$

where m_z is the z component of spin for the nth many-body eigenstate. The results for the specific heat and magnetization are given in Figs. 1 and 2, respectively, at the temperature where $\beta J=1$ and in Figs. 3 and 4, respectively, at the temperature where $\beta J=5$.

The results for $\beta J=1$ are representative of the high-temperature regime $\beta J<2$ where the temperature is larger than the energy-level spacing. The specific heat has a single broad peak as a function of magnetic field with the center of the peak moving to larger values of b and the zero-field intercept becoming smaller as the temperature increases. The magnetization smoothly changes form a value of zero to a value of $\frac{5}{2}$ as a function of magnetic field, showing little structure.

The results for $\beta J=5$ are representative of the low-temperature regime $\beta J>2$ where the temperature is smaller than the energy-level spacing. The specific heat has a multiple-peak structure arising from each level crossing in the ground state and the magnetization shows steps at the various level crossings.

The results fit the experimental data[5-8] extremely well. The specific-heat measurements resemble the "high-temperature" result (Fig. 1) with a single-peak structure and the magnetization measurements resemble the "low-temperature" result (Fig. 4) with noticeable steps. This is to be expected since magnetization measurements take place at a *constant* low temperature while specific-heat measurements require measurements over a temperature range. Figure 3 suggests that specific-heat measurements may show additional structure if they can be made at temperatures low enough to probe any features in the many-body density of states. Note that the low-field region ($b<1$) is not faithfully represented by a small-cluster calculation, since the discreteness of the energy levels will always produce a linear magnetization.

IV. DISCUSSION

In summary, the physics of the metamagnetic transition can be described as follows: a heavy-fermion system is composed of a ground state with nearly degenerate low-lying excitations of many different spin configurations; the weak antiferromagnetic superexchange interaction slightly favors low-spin arrangements over high spins (at zero magnetic field); a magnetic field pulls down the high-spin configurations causing (multiple) level crossing(s) in the ground state and producing a peak in the many-body density of states. The result is a peak in the specific heat (and possibly a richer structure at lower temperatures), steplike transitions in the magnetization, and abrupt changes in ground-state correlation functions.

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