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HEAVY-FERMION SYSTEMS IN MAGNETIC FIELDS:

THE METAMAGNETIC TRANSITION*

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ABSTRACT

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.

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Heavy-fermion systems have been an active area of research for both experimentalists\textsuperscript{1} and theorists\textsuperscript{2-4} since their discovery in the mid-1970's. Heavy-fermion systems are characterized by huge coefficients ($\gamma$) to the term linear in $T$ in the specific heat, quasi-elastic 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.\textsuperscript{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.\textsuperscript{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 $\gamma$, the elastic coefficients, and the magnetic properties. At the critical field $B_c$, the coefficient $\gamma$ 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 \((U > 10 \text{ eV})\). The physics of such an electronic system is described by the lattice (or periodic) Anderson impurity model\(^9\)

\[
H_A = \sum_{k\sigma} \epsilon_k a_{k\sigma}^+ a_{k\sigma} + \epsilon \sum_{i\sigma} f_{i\sigma}^+ f_{i\sigma} + U \sum_i f_{i\uparrow}^+ f_{i\uparrow} f_{i\downarrow}^+ f_{i\downarrow} + \sum_{ik\sigma} [V_{ik} f_{i\uparrow}^+ a_{k\sigma} + V_{ik}^* a_{k\sigma}^+ f_{i\sigma}] ,
\]

in the large-\( U \) \((U \to \infty)\) limit.\(^10\) The parameters and operators in Eq. (1) include the conduction-band creation (annihilation) operators \( a_{k\sigma}^+ \left( a_{k\sigma} \right) \) for a conduction electron in an extended state with wavevector \( k \), spin \( \sigma \), and energy \( \epsilon_k \); the localized electron\(^11\) creation (annihilation) operators \( f_{i\sigma}^+ \left( f_{i\sigma} \right) \) for localized electrons in an atomic orbital centered at lattice site \( i \) with energy \( \epsilon_i \); 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

\[
V_{ik} = \exp(i \mathbf{R}_i \cdot \mathbf{k}) V g(k) / \sqrt{N} ,
\]

with \( g(k) \), the form factor, a dimensionless function of order one, 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 so that \( E_F = 0 \). The conduction-band density of states per site at the Fermi level is
then defined to be $\rho$.

Heavy-fermionic behavior may occur in the region\(^1\) where $\epsilon_\rho = -V^2\rho^2 < 0$. The localized orbitals are almost singly occupied ($\langle f_i^{\uparrow} \bar{f}_i^{\uparrow} + f_i^{\downarrow} \bar{f}_i^{\downarrow} \rangle = 1 - \nu$, $\nu \ll 1$) and the conduction electron density of states at the Fermi level is small. The Anderson Hamiltonian (1) may be mapped onto the large-$U$ limit of the Hubbard\(^1\) Hamiltonian which, in turn, may be mapped onto a $t-J$ model\(^1\)

$$H_{t-J} = -\sum_{ij\sigma} t_{ij} (1 - f_i^{\uparrow} f_i^{\downarrow}) f_j^{\uparrow} f_j^{\downarrow} (1 - f_j^{\uparrow} f_j^{\downarrow}) + \sum_{ij} J_{ij} S_i \cdot S_j. \quad (3)$$

The hopping matrix $t_{ij}$ satisfies

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

and the antiferromagnetic superexchange is defined to be $J_{ij} \equiv 41 t_{ij}^2 / U$.

A heavy-fermion system is characterized\(^1\) 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) require 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.\(^1\) 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. 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.

The lattice Anderson impurity model [Eq. (1)] has been studied\textsuperscript{16-18} for various small clusters with at most four sites (for a review see Ref. 19). The results for the tetrahedral cluster\textsuperscript{17,18} (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 \( \varepsilon_k \) is such that the bottom of the band is at the \( \Gamma \) point of the face-centered-cubic Brillouin zone, a small range of values for \( \varepsilon \) are 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 \( \Gamma \) is the top of the conduction band, a magnetically ordered heavy-fermionic state is sometimes observed.

The small-cluster approach has also been applied to the \( t-J \) model\textsuperscript{20} which corresponds to the parameter regime of the lattice Anderson impurity model in between the Kondo lattice and the intermediate-valence state.\textsuperscript{12} A very good example of a heavy-fermion system lies in an eight-site face-centered cubic-lattice cluster with seven electrons.\textsuperscript{20} When the hopping parameters and antiferromagnetic superexchange parameters are chosen to be

\[
t_{ij} = \begin{cases} 
t > 0, & i, j = \text{first-nearest neighbors}, \\
0.1 t, & i, j = \text{second-nearest neighbors}, \\
0, & \text{otherwise},
\end{cases}
\]

\[
J_{ij} = \begin{cases} 
J, & i, j = \text{first-nearest neighbors}, \\
0, & \text{otherwise},
\end{cases}
\]  

(5)

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 include many different spin configurations (see Table 1). These many-body states are

\[
- 4 -
\]
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 degeneracy even more, since the many-body eigenstates with \( z \)-component of spin \( m_z \) have an energy

\[
E(B) = E(0) - m_z g \mu_B B = E(0) - m_z b J
\]

in a magnetic field \( B \). The symbols \( g \), \( \mu_B \), 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 \( b \).

The phenomena described above are all of 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

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

where \( k_B \) is Boltzmann’s constant, \( \beta \) is the inverse temperature (\( \beta = 1/k_B T \)) 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 1). Similarly the magnetization is expressed.
by

\[ M(b) = \frac{\sum_n m_z \exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)} \quad , \quad (8) \]

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 Figures 1 and 2, respectively, at the temperature where \( \beta J = 1 \) and in Figures 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 from a value of zero to a value of 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 lower temperatures.

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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References


10. The large-$U$ limit incorporated here implies both $U \to \infty$ and $\varepsilon + U \to \infty$, so that there is never more than one electron per $f$-orbital.
The degeneracy of the $f$-electrons is neglected in this model. Additional $f$-electron orbitals may easily be added without changing the qualitative nature of the model.


Table 1. 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. 20.

<table>
<thead>
<tr>
<th>Energy</th>
<th>Total Spin</th>
<th>Degeneracy</th>
<th>Spatial Symmetry Label</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-6t + 6t' - 3J$</td>
<td>$\frac{1}{2}$</td>
<td>14</td>
<td>$\Gamma_2 \oplus X_1 \oplus X_2$</td>
</tr>
<tr>
<td>$-6t + 6t' - 2J$</td>
<td>$\frac{1}{2}$</td>
<td>16</td>
<td>$L_3$</td>
</tr>
<tr>
<td>$-6t + 6t' - \frac{3}{2}J$</td>
<td>$\frac{3}{2}$</td>
<td>32</td>
<td>$\Gamma_{12} \oplus X_1 \oplus X_2$</td>
</tr>
<tr>
<td>$-6t + 6t' - \frac{1}{2}J$</td>
<td>$\frac{3}{2}$</td>
<td>16</td>
<td>$L_2$</td>
</tr>
<tr>
<td>$-6t + 6t' + J$</td>
<td>$\frac{5}{2}$</td>
<td>18</td>
<td>$X_2$</td>
</tr>
</tbody>
</table>
Figure Captions

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.

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.

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.
Figure 1
Figure 2
Figure 3