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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[1] and theorists[2–4] since their discovery in the mid-1970's. Heavy-fermion systems are characterized by huge coefficients (γ) 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.[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]

$$H_A = \sum_{k\sigma} \varepsilon_k a_{k\sigma}^{\dagger} a_{k\sigma} + \varepsilon \sum_{i\sigma} f_{i\sigma}^{\dagger} f_{i\sigma} + U \sum_{i} f_{i\uparrow}^{\dagger} f_{i\uparrow} f_{i\downarrow}^{\dagger} f_{i\downarrow}$$

$$+ \sum_{ik\sigma} [V_{ik} f_{i\sigma}^{\dagger} a_{k\sigma} + V_{ik}^{*} a_{k\sigma}^{\dagger} f_{i\sigma}] \quad , \qquad (1)$$

in the large-U ($U \rightarrow \infty$) 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 wavevector 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

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

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 $V \rightarrow 0$ 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 ρ.

Heavy-fermionic behavior may occur in the region[12] where $\varepsilon\rho \approx -V^2\rho^2 < 0$. The localized orbitals are almost singly occupied ($<f_{i\uparrow}^{\dagger} f_{i\uparrow} + f_{i\downarrow}^{\dagger} f_{i\downarrow}> = 1-\nu$, $\nu \ll 1$) 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[13] hamiltonian which, in turn, may be mapped onto a $t-J$ model[14]

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

The hopping matrix t_{ij} satisfies

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

and the antiferromagnetic superexchange is defined to be $J_{ij} \equiv 4\,|t_{ij}|^2/U$.

A heavy-fermion system is characterized[12] 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.[15] 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 ($N \rightarrow \infty$) has not yet been found.

The lattice Anderson impurity model [Eq. (1)] has been studied[16–18] for various small clusters with at most four sites (for a review see Ref. 19). The results for the tetrahedral cluster[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 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 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 Γ 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[20] which corresponds to the parameter regime of the lattice Anderson impurity model in between the Kondo lattice and the intermediate-valence state.[12] A very good example of a heavy-fermion system lies in an eight-site face-centered cubic-lattice cluster with seven electrons.[20] When the hopping parameters and antiferromagnetic superexchange parameters are chosen to be

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

$$J_{ij} = \begin{cases} J \,, & i,j = \text{first–nearest neighbors} \,, \\ 0 \,, & \text{otherwise} \,, \end{cases} \tag{5}$$

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 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

$$E(B) = E(0) - m_z g \mu_B B \equiv E(0) - m_z bJ \quad , \tag{6}$$

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.

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

$$\frac{C_V(b)}{k_B} = \beta^2 \left[\frac{\sum_n E_n^2 \exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)} - \left\{ \frac{\sum_n E_n \exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)} \right\}^2 \right] \quad , \tag{7}$$

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 1). Similarly the magnetization is expressed by

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

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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[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.

11 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.

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Table

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

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.

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.

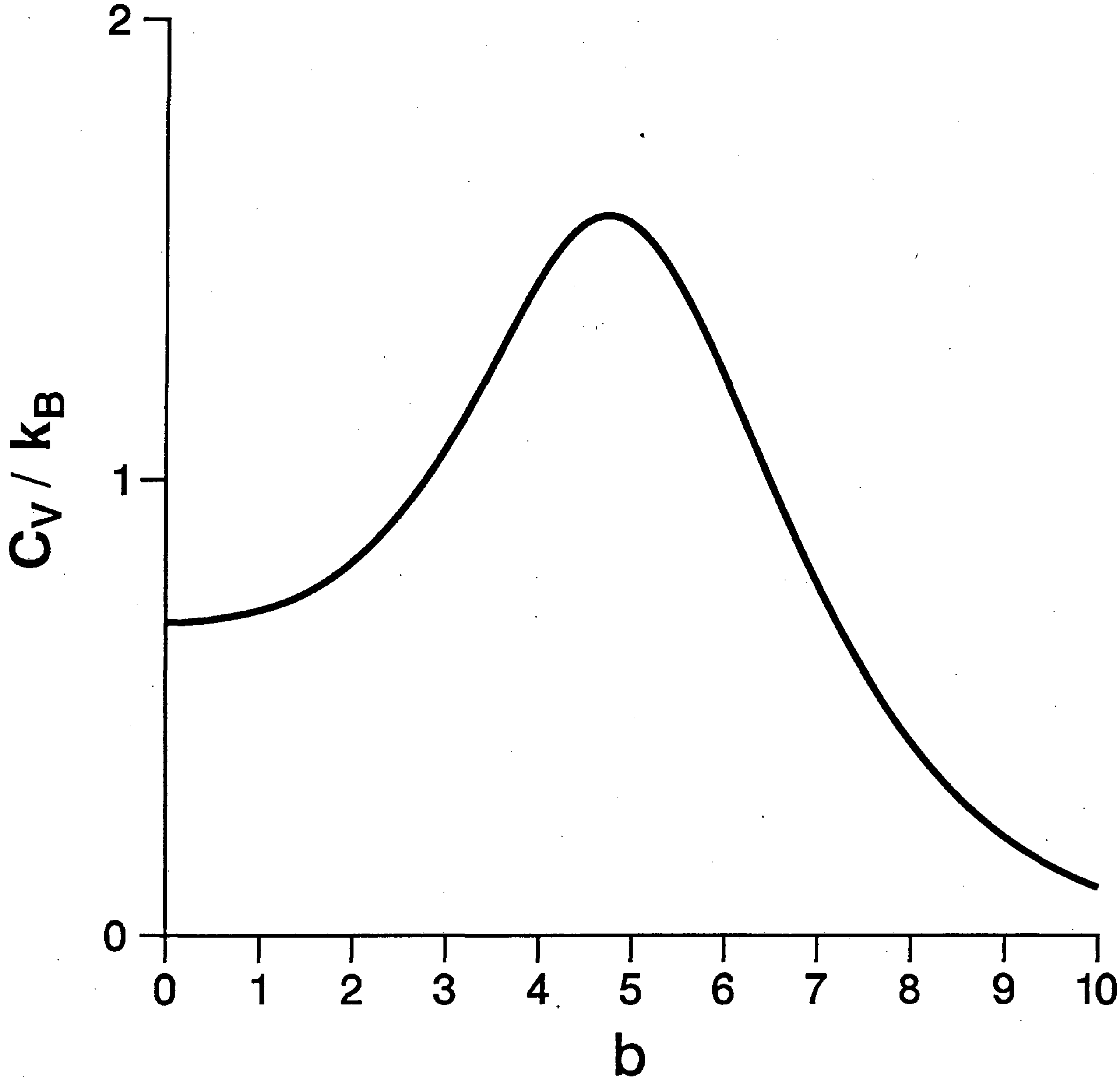

Figure 1

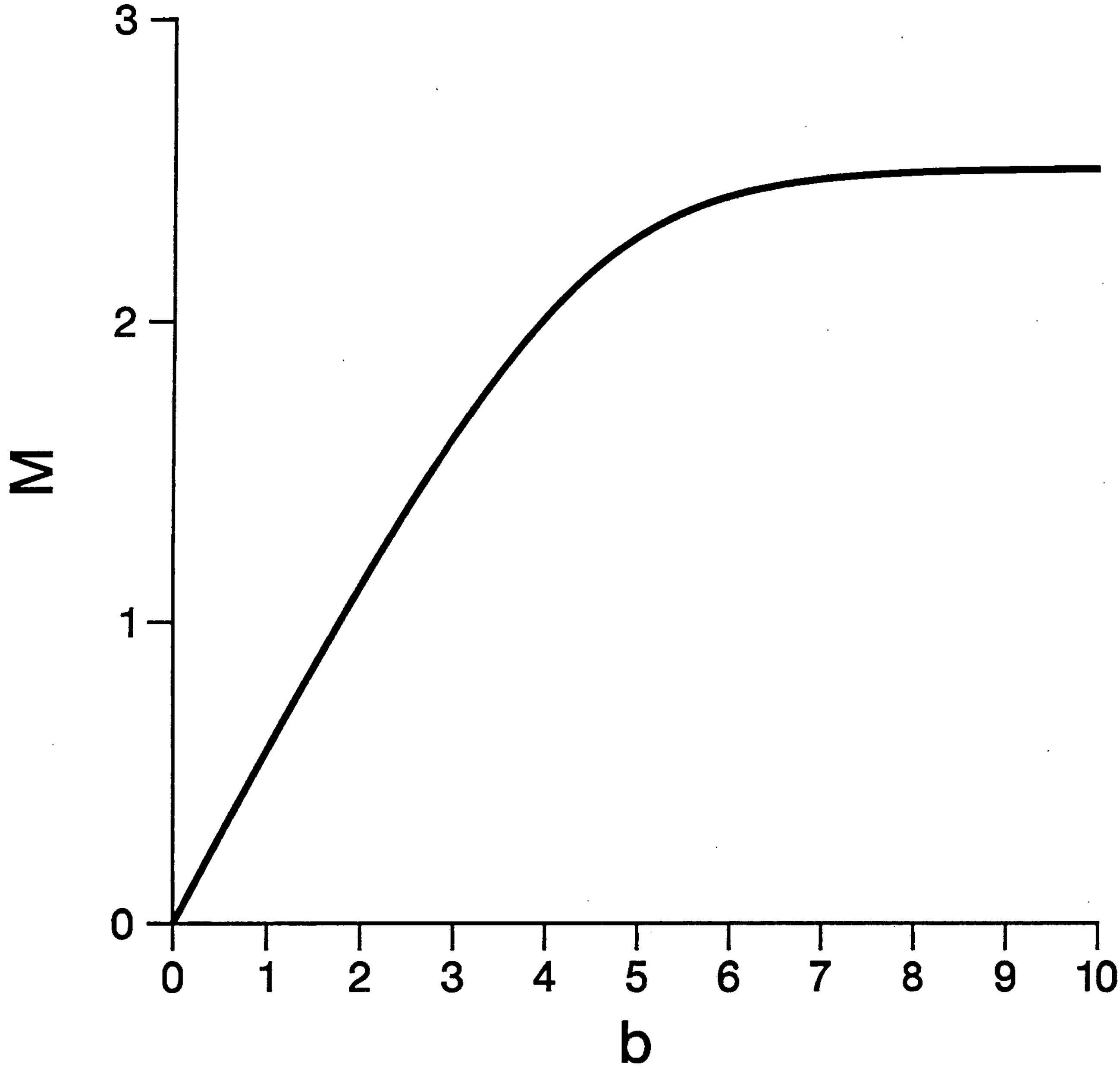

Figure 2

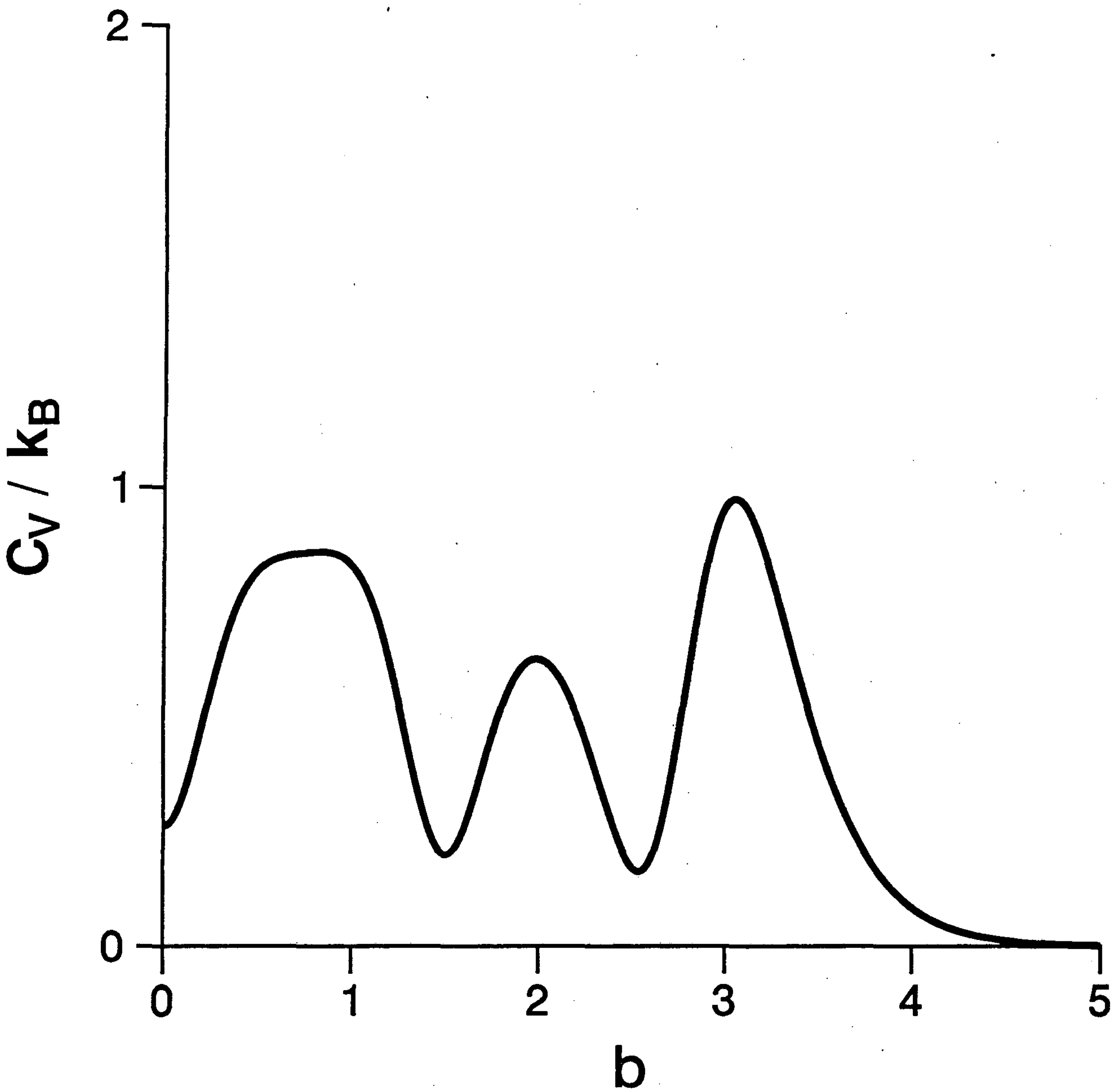

Figure 3

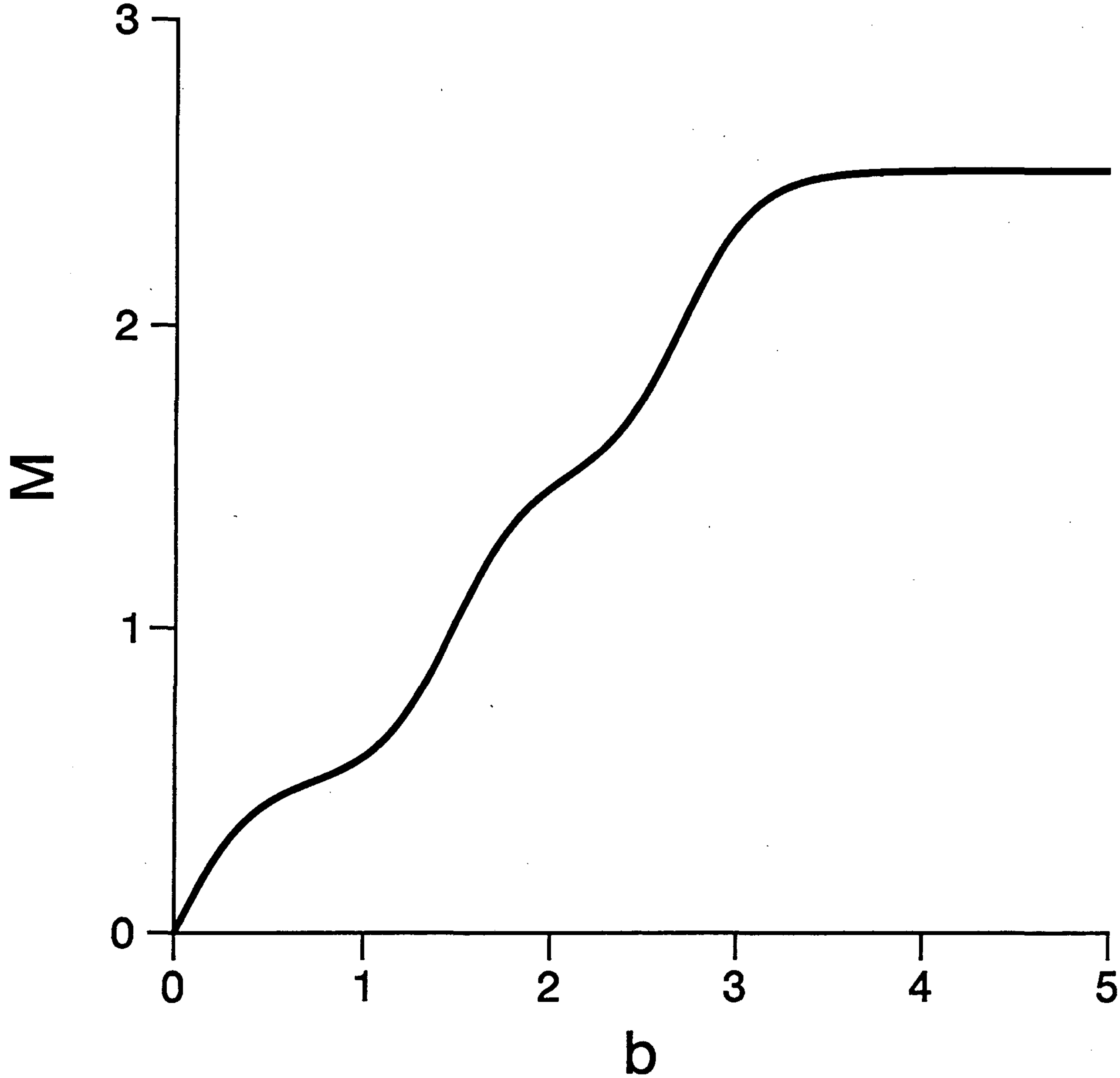

Figure 4

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