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PERSPECTIVE ON HEAVY-ELECTRON AND KONDO-LATTICE SYSTEMS FROM HIGH PRESSURE STUDIES

J.D. THOMPSON and Z. FISK
Los Alamos National Laboratory, Los Alamos, NM 87545, USA

G.G. LONZARICH
Cavendish Laboratory, Cambridge CB3 0H3, UK

The application of modest pressures p to rare-earth and actinide-based heavy-electron / Kondo-lattice materials produces significant changes in both their temperature dependent electrical resistivity ρ and electronic specific heat γ. For a given compound, $\gamma(p, T=0)$ appears to scale inversely with a characteristic temperature associated with features in $\rho(p, T)$. These changes can be understood as arising from the strongly volume-dependent competition of interactions giving rise to the heavy-mass ground state. Similar behavior also may be found in transition-metal compounds, e.g., MnSi.

1. Introduction

De Haas–van Alphen experiments on heavy-electron compounds UPt_3 [1] and $CeCu_6$ [2] establish the strong mass renormalization of all conduction electrons in these materials, with the extent of mass enhancement generally consistent with band structure calculations and specific heat measurements. These fundamental experiments are, however, insensitive to the mechanism responsible for the large renormalizations characteristic of heavy-electron systems. Some evidence for the mechanism comes from electrical resistivity ρ measurements [3] that find a temperature region in many cases where $\partial\rho/\partial T < 0$, reminiscent of single-impurity Kondo behavior which is known to produce a resonance in the density of states near the Fermi energy. However, at sufficiently low temperatures, the resistivity no longer resembles that of a single Kondo impurity but instead increases with temperature as $\rho \propto \rho_0 + AT^2$. This "transition" from impurity-like behavior at moderately high temperatures to Fermi-liquid-like behavior at low temperatures is generally ascribed to the development of coherence in Kondo scattering at Kondo sites distributed periodically throughout the lattice [3, 4]. Hence, heavy-electron materials are sometimes called Kondo-lattice systems. The "transition" is expected to produce a maximum in the resistivity at some temperature T_{max} which reflects the cross-over from predominantly inelastic to elastic scattering. From this perspective the relevant energy scale is the Kondo temperature T_K. Indeed, one can argue qualitatively that the large γ in heavy-electron compounds is given approximately by $\gamma \approx (k_B \ln D)/T_K$, where D is the degeneracy of the f-moment [5]. This gives values of T_K on the order of 1 to 10 K for Ce- and U-based heavy-electron materials, respectively. However, neutron-scattering experiments [6] clearly reveal the presence of momentum q and temperature-dependent Ruderman–Kittel–Kasuya–Yoshida (RKKY) interactions in addition to Kondo interactions and that they are of comparable magnitude. It is the competition between these interactions that determines the ultimate ground state and, as will be discussed, possibly the relevant framework for a description of heavy-electron behavior.

Pressure is a particularly valuable technique for studying this competition because it is a "clean" variable in the sense that it does not disrupt the lattice periodicity (unlike substitutional studies) but does produce significant changes in measurable quantities as the conduction electron-f-moment interaction is tuned by decreasing volume. Most attention has focused

on establishing the pressure dependence of γ and $\rho(T)$ in a variety of heavy-electron/Kondo-lattice systems; although, other probes, e.g. NMR [7], thermoelectric power [8], magnetostriction [9] and magnetic susceptibility [10], have provided useful information on heavy-electron behavior. Here we briefly review what has been learned from studying the pressure response of these materials.

2. Results and discussion

2.1. Rare-earth and actinide systems

An interesting property of Kondo-lattice systems is that their pressure dependent electrical resistivity scales over a wide low-temperature interval about T_{max} [11] provided that the ground state degeneracy is unchanged, as could happen, for example, should T_{max} become comparable to crystal-field splittings [12]. In Ce- and U-based systems, T_{max} generally increases with decreasing volume [13], whereas in Yb-based compounds, the opposite is observed [14]. (The sign difference in dT_{max}/dp among these compounds is also found in the pressure dependence of the susceptibility and γ [14], as well as in magnetostriction experiments [9]). Resistance scaling in the form $\rho(T, p)/\rho_{max}(p)$ versus $T/T_{max}(p)$, where $\rho_{max} = \rho(T_{max})$, seems to hold generally also in the low temperature regime where $\rho \propto \rho_0 + AT^2$, implying that the inverse square root of the T^2-coefficient of resistivity $A^{-1/2}$ is a comparably valid measure of the scaling temperature. That is, $T_{max} \propto A^{-1/2}$, which is found experimentally [13, 14].

The electronic contribution to the specific heat of heavy-electron compounds is extremely sensitive to volume changes [15]. For example, in $CeCu_6$ at ambient pressure, $\gamma \sim 1600$ mJ/mole K^2, but at 8.8 kbar is depressed to about half this value. With a bulk modulus of ~ 0.8 Mbar for $CeCu_6$, this corresponds to an electronic Grüneisen parameter $\Omega_e = -\partial \ln \gamma / \partial \ln V \sim -50$ [15], reminiscent of the large negative Ω_e found [16] in dilute Kondo impurity systems. Similar Ω_e's are found [15] in other heavy-electron compounds UPt_3, UBe_{13} and $CeAl_3$.

To establish a connection between specific heat and resistivity measurements, it is instructive to compare the pressure dependence of T_{max} and $\gamma_0 = C/T|_{T=0}$. Figures 1 and 2 show plots of γ_0 versus $1/T_{max}$ for $CeCu_6$ and UBe_{13}, respectively, in which pressure is the implicit variable [17, 18]. In both cases an approximately linear relationship is found, with a logarithmic derivative $\Gamma_e = \partial \ln \gamma_0 / \partial \ln (1/T_{max}) = 0.99$ for $CeCu_6$ and 0.73 for UBe_{13}. In the case of UPt_3 for which there is no resistivity maximum below room temperature [19], we plot in fig. 3 γ_0 versus $1/T_{sf}$, where $T_{sf} \propto A^{-1/2}$ is an easily derived temperature scale by which the pressure dependent resistivity can be scaled [19, 20]. Here, again, we find a nearly linear relationship and $\Gamma_e = 1.21$. Therefore, in instances where comparisons can be made ($CeCu_2Si_2$ is excluded because its analysis is complicated by low-lying crystal field levels), Γ_e is near unity even though zero pressure values of the scaling temperature (T_{max} or T_{sf}) and γ_0 vary substantially among these compounds. In the single impurity Kondo problem, Γ_e is expected to be identically unity because $\gamma_0 T_K = 0.68\,R$, where R is the gas con-

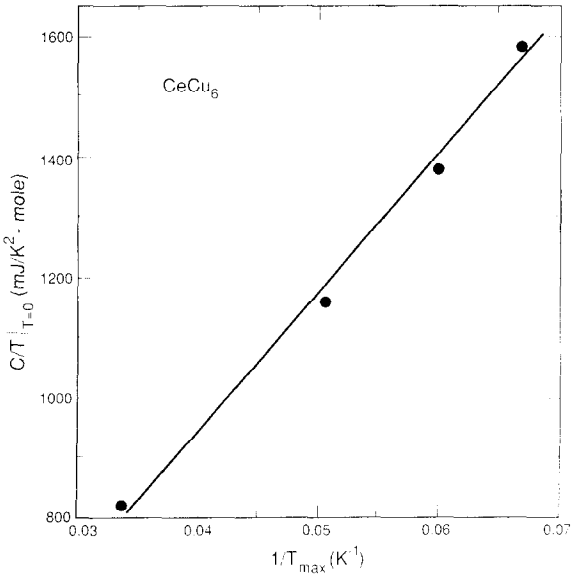

Fig. 1. Specific heat C divided by temperature T, extrapolated to $T = 0$, $(C/T|_{T=0})$ as a function of $1/T_{max}$ for $CeCu_6$. Pressure is the implicit variable. Values of C/T from ref. [15] and $1/T_{max}$ from ref. [11].

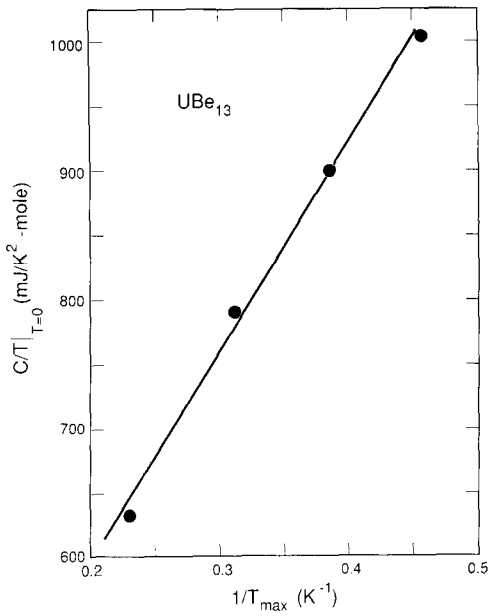

Fig. 2. Specific heat C divided by temperature T, extrapolated from $T_c = 0.9$ K to $T = 0$, as a function of $1/T_{max}$ for UBe_{13}. Pressure is the implicit variable. Pressure dependence of C/T from ref. [17] and $1/T_{max}$ from ref. [18].

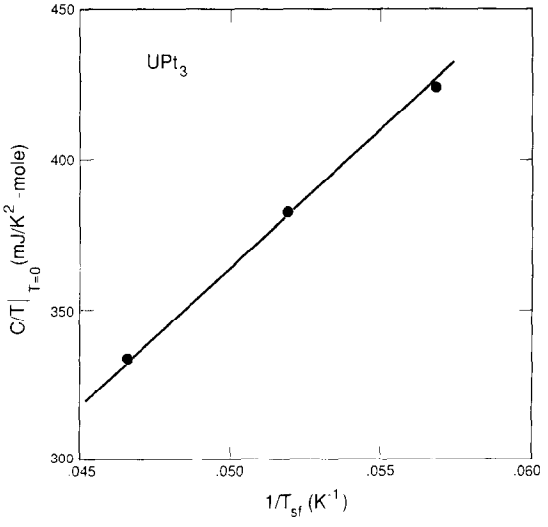

Fig. 3. Specific heat C divided by temperature T, extrapolated from $T_c \simeq 0.5$ K to $T = 0$, as a function of $1/T_{sf}$ for UPt_3. See text for definition of T_{sf}. Pressure is the implicit variable. $C/T(p)$ from ref. [20] and $T_{sf}(p)$ from ref. [19].

stant [21]. Given uncertainties in extrapolating C/T to $T = 0$ and that specific heat and resistivity measurements were performed on different samples, agreement with the Kondo-impurity prediction is rather good. To what extent this agreement is quantitative remains an open question. However, to leading order it appears that the pressure response of the specific heat and resistivity of heavy-electron/Kondo-lattice materials is determined primarily by the Kondo effect.

Schilling [22] has argued that the resistivity maximum results from the competition between Kondo and RKKY interactions. This point is well-taken especially since recent neutron scattering [6], μSR [23] and NMR [24] experiments are revealing at very low temperatures evidence for antiferromagnetic correlations among f-moments in virtually all heavy-electron compounds, even those that become superconducting. The observed moments are reduced substantially from their high temperature values found in magnetic susceptibility measurements. Such moment reduction is expected due to Kondo interactions. Further, pressure experiments [25–27] also clearly indicate a competition between Kondo and RKKY interactions for the ground state of Kondo-lattice systems. Therefore, even though RKKY interactions may be responsible for producing a resistivity maximum and the appearance at low temperatures of a coherent heavy band-like state, they appear not to alter substantially the pressure response of T_{max} and γ_0.

A possible explanation comes from a model of the spin–spin correlation function in which moment fluctuations at one f-site are coupled to those at other sites by an effective exchange interaction [28]. Because of this coupling, a small energy scale T_K^* arises in the limit of low frequencies that is related to the q-dependent exchange J and Kondo susceptibility χ_0 by $T_K^*(q, T) \simeq T_K[1 - J(q, T)\chi_0(T)]$, where $\chi_0 \propto C/(T + T_K)$. For $T_K \gg T$, this expression reduces to $T_K^*(q, T) \simeq T_K[1 - T_R(q, T)/T_K]$ where T_R is the q and T dependent intersite scale given by $J(q, T)C$. Qualitatively, this simple result appears to embody much of the essential physics: T_K^* reflects the competition between intrasite Kondo and intersite RKKY interactions and has

a pressure dependence dominated by T_K. Phenomenologically, it suggests an "effective q-dependent Kondo temperature", consistent with weakly anisotropic mass enhancements found in dH–vA experiments [1, 2] and a directional dependence of T_{max} and A observed [29, 19] in non-cubic heavy-electron single crystals. It also allows for an effectively temperature dependent Kondo temperature, as implied from analysis of the magnetoresistance of UBe_{13} at ambient [30] and elevated pressures [13, 31]. Of course, if for some q, $T_R(q, T)/T_K \geq 1$, the electronic system should order, producing heavy-electron antiferromagnets like U_2Zn_{17}, UCu_5 and UCd_{11}.

2.2. MnSi

Thus far the discussion has focused on systems in which electrons responsible for magnetism are relatively localized. One must wonder if there is not a continuum in the mass renormalization as these electrons become progressively delocalized, e.g., as in transition metal compounds. MnSi may represent a case in which reasonably strong (for a transition metal) mass renormalization appears because of strong spin fluctuations within the conduction electron sea itself. At ambient pressure MnSi orders below $T_c \sim 29$ K to a long wavelength helical structure [32]. Interestingly, MnSi differs from most other transition metal compounds characterized as weak or incipient ferromagnets in exhibiting magnetic fluctuation modes (Fourier components of the magnetization density) that have low characteristic frequencies over large portions of the Brillouin zone [33]. Such modes may be very sensitive to volume change and could lead to renormalization of the fermion mass, the quasiparticle interactions and T_c as a function of pressure.

The pressure dependence of the electrical resistivity and T_c of MnSi is given in fig. 4. The inset shows that T_c is depressed rapidly to zero at a critical pressure $p_c \simeq 15$ kbar. The initial rate of decrease $dT_c/dp = -1.13$ K/kbar agrees quantitatively with results of magnetization measurements to 5.2 kbar [34] and with that calculated (−1.2 K/kbar) from Ehrenfest's relation

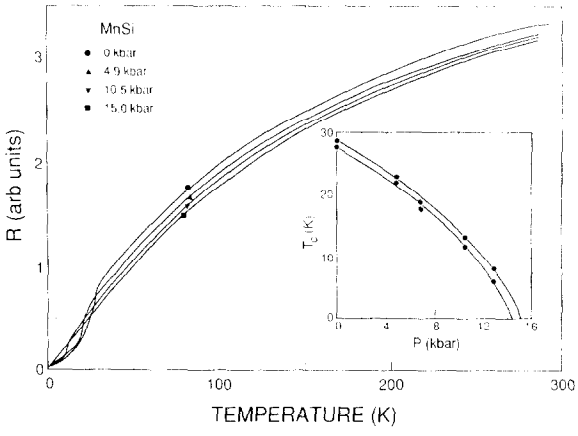

Fig. 4. Electrical resistance R versus temperature for MnSi subjected to various pressures. The inset shows T_c vs. pressure determined from maxima in dR/dT. At each pressure two peaks in dR/dT were found corresponding to possibly two phase transitions also found in ultrasound studies [S. Kusaka et al., Solid State Commun. 20 (1976) 925.]

for a second order phase transition. Already at ambient pressure γ_0 is enhanced ($\gamma_0 \sim 50$ mJ/mol K^2) and the cyclotron masses are typically five times larger than calculated band masses [35]. This enhancement may be associated primarily with the nearly critical fluctuations of the spin density discussed above. However, near p_c the degree of renormalization is expected to be even larger as the long wavelength modes become critical at low temperatures. In the following, we present a theoretical model for the specific heat of MnSi that supports this speculation.

Within the conventional paramagnon approximation the spin fluctuation correction to the free energy F_0 of non-interacting carriers of the starting band model can be expressed essentially as

$$\Delta F = \sum_{\nu,\boldsymbol{q}}^{BZ} \int_0^\infty d\omega F(\omega) \times \left\{ -\mathrm{Im} \frac{\partial}{\pi \partial \omega} \ln [1 - \lambda(\boldsymbol{q}) \chi_{0\nu}(\boldsymbol{q}, \omega)] \right\}, \quad (1)$$

where $F(\omega)$ is the free energy of an oscillator of frequency ω, $\chi_{0\nu}(\boldsymbol{q}, \omega)$ is a component of the generalized spin susceptibility of the non-interacting carriers in a *diagonal* representation, and

$\lambda(q)$ is a molecular field or interaction parameter defining the renormalised susceptibility (see, e.g. ref. [36]).

The average mass renormalization parameter $(m^*/m - 1)$ is determined from the ratio of the coefficient of the T^2 term in ΔF to that in F_0. From (1) we find

$$\frac{m^*}{m} = 1 + \frac{1}{g(\varepsilon_F)} \sum_{\nu,q} \frac{\bar{\lambda}_\nu^2(q)}{2\pi\hbar\Gamma_\nu(q)}, \qquad (2)$$

where $g(\varepsilon_F)$ is the density of states per spin at ε_F for the non-interacting carriers, $\bar{\lambda}_\nu(q)$ is $\lambda(q)\chi_{0\nu}(q, \omega = 0)$ and $\Gamma_\nu(q) = \Gamma_{0\nu}(q)(1-\bar{\lambda}_\nu(q))$ where $\Gamma_{0\nu}(q)$ is defined by the condition $\chi_{0\nu}(q, \omega = 0)/\Gamma_{0\nu}(q) = \mathrm{Im}\,\partial\chi_{0\nu}(q, \omega = 0)/\partial\omega$. In arriving at (2) the Hartree–Fock component of (1), i.e., that part linear in $\lambda(q)$ (assumed to be included already in F_0), has been subtracted. In the ferromagnetic state $\mathrm{Im}\,\partial\chi_{0\nu}(q, \omega = 0)/\partial\omega$ vanishes for components transverse to the spontaneous magnetization at small q where well-defined spin waves are expected to exist. The transverse components in this portion of the q-space hence do not, in this model, contribute to the sum in (2). Within the remaining portion of q space where the Fourier components of the spin density exhibit a broad power spectrum, or in the paramagnetic state in general, it is helpful to think of $\Gamma_\nu(q)$ as a characteristic relaxation frequency of a spin fluctuation of wavevector q and of polarization ν. For a more precise meaning we return to the definition given under (2).

When the factor $\bar{\lambda}_\nu^2(q)$ on the right hand side of (2) can be approximated by unity, i.e., when $\Gamma_\nu(q)$ is well below $\Gamma_{0\nu}(q)$ in major portions of the Brillouin zone, then e.g. (2) reduces to the mass enhancement factor discussed in ref. [37].

It is possible that the relationship between the mass and the fluctuation spectrum implied by (2) is somewhat more general than the above elementary analysis would suggest. Indeed, in terms of a spectrum $\Gamma_\nu(q)$ *fitted* to inelastic neutron data, it was shown that (2) with $\bar{\lambda}_\nu^2(q) = 1$ yields m^*/m of approximately 6, a value close to the ratio of the observed and the band calculated linear heat capacity (and far above that expected from the electron–phonon interaction alone) [37].

ΔF also can be used to arrive at a magnetic equation of state that yields, when it is made self-consistent in the bulk susceptibilities, an expression for a renormalized Curie temperature [37, 38]. The value of T_c predicted by this model, in terms of the empirically derived spectrum $\Gamma_\nu(q)$, is far below that expected for the starting band theory and is within 10% of experiment.

The renormalization of the susceptibility achieved by the self-consistent model implies a modification of the original free energy ΔF. This modification leads to a renormalization of $\Gamma_\nu(q)$ in eq. (2) within our approximation, and also changes the form of ΔF beyond second order in T. Calculations within this improved model for ΔF [39] permit a good account to be given of the overall heat capacity $C(T)$ from low temperatures through T_c in terms of the empirical spin fluctuation spectrum at ambient pressure (fig. 5).

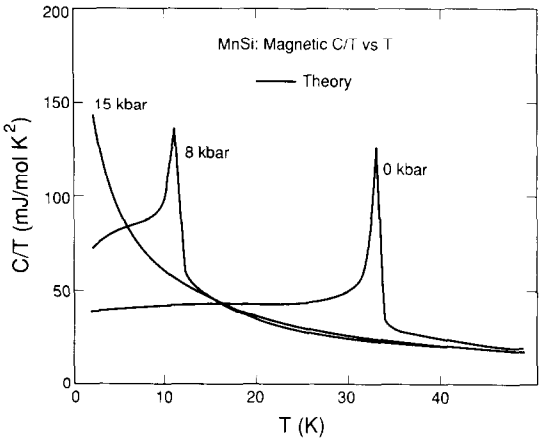

Fig. 5. The magnetic fluctuation contribution to the heat capacity divided by temperature for MnSi at different pressures calculated in terms of the self-consistent model described in the text. The parameters defining the relaxation frequency spectrum [37] are chosen to be consistent with neutron scattering data at ambient pressure [32] and with the pressure dependence of the transition temperature (fig. 4) and the spontaneous magnetization (see ref. [39] for fuller details). Consistent with experiment at ambient pressure [40] to a precision of better than 20% are the quantities C/T at low T, the entropy change upon crossing T_c, C/T above T_c, and T_c itself.

For a spectrum $\Gamma_\nu(q)$ consistent with neutron scattering data at ambient pressure and with the known pressure variation of the transition temperature (fig. 4) and the spontaneous magnetization [34], this spin fluctuation model predicts that the mass enhancement for MnSi could approach that of the rare earth and actinide heavy-electron systems for pressures close to p_c (fig. 5).

It is stressed, however, that very close to p_c when $\Gamma_\nu(q)$ is very strongly reduced below $\Gamma_{0\nu}(q)$, the model for ΔF, even as modified in the manner described above, is not expected to hold. In this interesting regime a treatment analogous to that employed to describe quantum critical phenomena is required.

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