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The normal states of magnetic d and f transition metals

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Abstract. The normal states of magnetic metals with vanishing Curie (T_c) or Néel (T_N) temperatures are investigated by means of measurements of the temperature and pressure dependence of the resistivity in the stoichiometric d and f compounds MnSi, ZrZn₂, CePd₂Si₂ and CeNi₂Ge₂. The results for the nearly ferromagnetic d metals may be described over a wide range in temperature and pressure in terms of a quantitative model of a marginal Fermi liquid based on dispersive spin-fluctuation spectra inferred from inelastic neutron scattering data. The behaviour of the antiferromagnetic f metals is also unconventional, but in a way which cannot yet be readily categorized. Near the critical pressure where $T_N \rightarrow 0$ K, CePd₂Si₂ displays a resistivity of the form $\rho \sim T^{1.2\pm0.1}$ over nearly two decades in temperature before condensing into a short-coherence-length superconducting state below 0.4 K. The isoelectronic and isostructural compound CeNi₂Ge₂, with a lattice cell volume slightly smaller than that of CePd₂Si₂, remains normal down to 20 mK and shows a resistivity of the form $\rho \sim T^{1.4\pm0.1}$ over a decade below several kelvin at ambient pressure. These findings for the Ce systems have not yet been described consistently in terms of an extension of the model developed for the ferromagnetic d metals.

1. Introduction

An increasing number of systems are coming to light which appear to have normal-state properties at odds with that usually expected for a simple Fermi liquid [1, 2]. If by the latter we mean a state in which the low-lying propagating modes have a finite overlap with the non-interacting one-particle excitations [1], then among the possible non-Fermi liquids it may also be reasonable to include the normal phases of some magnetic metals with vanishing Curie or Néel temperatures [3]. The study of such materials has a long history [4, 5] which parallels in some degree current investigations of the normal-state properties of the short-coherence-length superconductors. In all of these systems fluctuations of the order parameter or in some associated variable may strongly suppress the conventional mean-field transition temperature and leave behind an unusual normal-metallic state.

Here we focus attention on relatively simple examples of stoichiometric magnetic materials in which the normal state can be explored over a wide temperature range by suppressing the Curie or Néel temperatures towards absolute zero with the application of hydrostatic pressure. The chief goal will be to compare and contrast the behaviour observed in the better-understood d metals with that of the heavy-fermion systems in this critical regime. Superficially, they appear to be qualitatively similar, but on close analysis, some startling differences emerge which have not yet been shown to be consistent with current models for the role of spin fluctuations in the normal-metallic state.

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2. Effects of long-range interactions

To gain some qualitative insights on the possible breakdown of the Fermi-liquid description, it is helpful to consider the nature of the quasiparticle potential near a ferromagnetic phase transition. We may think of a quasiparticle as interacting with various effective fields set up by other quasiparticles. In analogy to Maxwell's electromagnetism, these renormalized fields couple in lowest order to the charge, current and spin magnetic moment of the quasiparticle. Of particular interest here is the coupling to the spin moment which arises in our case chiefly from the exchange or Weiss molecular field proportional, in the simplest case, to the spaceand time-dependent magnetization. If we think of this field acting on a given quasiparticle as a disturbance generated by another quasiparticle, we are led to an effective induced quasiparticle–quasiparticle interaction which, in a linear response approximation, is given by the space- and time-dependent magnetic susceptibility.

The spatial range of this interaction is then simply the magnetic correlation length which diverges at the Curie point T_c . In the standard picture the propagation frequency of the interaction is purely imaginary and described by a relaxation spectrum which varies with wavevector q as $\Gamma_q \propto q^z$, where z is the dynamical exponent. For an isotropic and homogeneous metal with $T_c \rightarrow 0$, we normally expect z = 3 at low temperatures. One factor of q in Γ_q arises from Landau damping and an additional factor of q^2 comes from the dependence of Γ_q on the inverse of the static susceptibility χ_q , which should vary as $1/q^2$ near a quantum critical point ($T_c \rightarrow 0$). This model for the dynamics of low-lying spin fluctuations is expected to hold for a sufficiently high spatial dimensional (i.e. for d > 4 - z [6]) and has considerable support from inelastic neutron scattering measurements for a number of cubic d metals [7].

The nature of the spin fluctuations in antiferromagnetic metals near quantum criticality is less well understood, but a naive extension of the above model (with q now measured from the antiferromagnetic ordering wavevector Q) would suggest that the corresponding dynamical exponent is z = 2 [6].

The divergence in the range of the quasiparticle–quasiparticle interaction suggested by the above might be expected to produce concomitant singularities in the effective quasiparticle 'mass' defined by the ratio of the heat capacity C and temperature T at low T and in the effective quasiparticle 'cross-section', which may be defined in analogy to the mass via the ratio of the resistivity ρ to the T^2 -factor associated with the constraints imposed by the Pauli principle. In this case the saturation of C/T and ρ/T^2 , which we usually expect for a Fermi-liquid state at low T, may arise only at very low temperatures or not at all. The starting state, in which singular interactions appear, may eventually give way to a non-Fermi-liquid fixed point [2].

3. Cubic d metals near ferromagnetic quantum criticality: MnSi and ZrZn₂

A relatively simple system in which to investigate the effects of long-range quasiparticle interactions is the cubic d metal MnSi which condenses below 30 K into a long-wavelength helical spin structure. Over distances of the order of a hundred ångströms it appears locally ferromagnetic, and over a wide range in q the spin-fluctuation spectrum is of the type discussed above [8].

The temperature dependence of the resistivity has been measured at various pressures for a stoichiometric sample with a low residual resistivity $\rho_0 \simeq 0.4 \ \mu\Omega$ cm (figure 1, [9]). The shoulder in the ρ versus T curves marks the transition temperature T_c which falls continuously to zero with increasing pressure. At the critical pressure $p_c \simeq 14.8$ kbar, ρ



Figure 1. The resistivity for MnSi versus temperature at different pressures (5.55 kbar, 8.35 kbar, 10.40 kbar, 11.40 kbar, 12.90 kbar, 13.55 kbar, 14.30 kbar and 15.50 kbar going down starting from the top curve at the far right). The magnetic ordering temperature $T\alpha_c$ (marked by the shoulder in ρ versus T) decreases towards absolute zero at $p_c \simeq 14.6$ kbar. For $p > p_c$ a non-Fermi-liquid form of ρ versus T (i.e. a variation T^{β} with $\beta < 2$) is seen to extend over a wide range down to mK temperatures (see the inset) [9].

varies more slowly than quadratically in temperature over the entire range explored. This is highlighted in a plot of the effective 'cross-section' $(\rho - \rho_0)/T^2$ shown in figure 2. Well away from p_c , $(\rho - \rho_0)/T^2$ saturates at low T as required in a Fermi-liquid picture, but near p_c it appears to grow without limit with decreasing temperature. As shown in the inset of figure 2, the temperature dependence of ρ is described by an asymptotic exponent of approximately 1.6. A similar result is also obtained for the ferromagnetic cubic metal ZrZn₂ over a range of pressures near the corresponding quantum critical point [10].

4. Comparison with predictions of a spin-fluctuation model

These findings are at least qualitatively consistent with a simple model in which quasiparticles scatter from thermally excited waves of the exchange molecular field described by the relaxation spectrum given in section 2 [4]. The resistivity can in this model be written in a form similar to that for phonon scattering with the crucial difference that the relevant thermal population factor n_q [5] involves now an $\omega > 0$ integral of the Bose function



Figure 2. The ratio of $\Delta \rho = \rho - \rho_0$ to T^2 versus temperature *T* for MnSi at different pressures (10.40 kbar, 12.90 kbar, 14.30 kbar and 15.50 kbar going up starting from the bottom curve at the far left), highlighting the breakdown of the Fermi-liquid form $\Delta \rho \propto T^2$ at low *T* as p_c is approached. Near p_c , $\Delta \rho \propto T^\beta$ with $\beta \simeq 1.6$ in the low-temperature limit (the curved line in the inset; the straight line gives the asymptotic low-temperature form) [9].

 $1/(e^{\omega/T}-1)$ over the function $(\pi\omega/2)/(\omega^2 + \Gamma_q^2)$, rather than the usual delta function centred on the phonon frequency. This leads us immediately to an asymptotic exponent near p_c of 5/3 when d = z = 3 [4], in reasonable agreement with observation. A complementary analysis leads to a contribution to the heat capacity of the form $C \sim T \ln(T^*/T)$ for d = z. These results are believed to be qualitatively correct when d + z, the effective dimension appropriate to a description of quantum criticality, exceeds the upper critical dimension of 4 [11]. For our case where d = z = 3, the asymptotic exponents are those expected for a marginal Fermi liquid such as that thought to arise in a charged plasma via the effects of transverse photons which produce long-range current–current interactions important in the relativistic limit [12–14].

For a more quantitative comparison between the above model and the measured temperature dependence of the resistivity, we consider the logarithmic derivative $\partial \ln \rho / \partial \ln T$ which may be viewed as a temperature-dependent resistivity exponent. In the present framework $\partial \ln \rho / \partial \ln T$ depends only on Γ_q . The exponent reduces, as we have noted, to 5/3 in the limit $T \rightarrow 0$ at the quantum critical point, and vanishes at high temperatures when ρ saturates. The temperature dependence of $\partial \ln \rho / \partial \ln T$ arises explicitly from the Bose function defining the thermal population n_q , and also implicitly via the temperature dependence of the static susceptibility χ which enters the relaxation spectrum. At low q in a cubic system this spectrum can be expressed in the form $\Gamma_q = \gamma q(\chi^{-1} + cq^2)$ which, as discussed in section 2, is third order in q at a quantum critical point defined by $\chi^{-1} \rightarrow 0$ as $T \rightarrow 0$. In this expression, γ and c are weakly temperature-dependent parameters which may be inferred directly from inelastic neutron

scattering data. $\partial \ln \rho / \partial \ln T$ is then fully determined by these parameters, $\chi^{-1}(T)$ which is taken from bulk susceptibility measurements, and a cut-off wavevector q_c set equal to the characteristic dimension (ΓX) of the Brillouin zone.



Figure 3. The calculated temperature dependence of the logarithmic derivative of the resistivity in the limit $T_c \rightarrow 0$ plotted versus $\log_{10}(T/T_{sf})$ for the ferromagnetic (FM) model (upper and middle curves) and antiferromagnetic (AFM) model (lower curve) as described in the text. T_{sf} is the effective bandwidth of the spin-fluctuation spectrum and κ is the correlation wavevector $1/\sqrt{c\chi}$.

The results of numerical evaluations of $\partial \ln \rho / \partial \ln T$ as a function of T/T_{sf} , where $T_{sf} = \gamma c q_c^3$, are shown in figure 3 for γ and c as measured in MnSi at ambient pressure [8] and for χ^{-1} in Γ_q set to zero at all T (upper curve) and for χ^{-1} proportional to T at all T (middle curve). The lower curve is that expected for a naive extension of the model for a nearly antiferromagnetic system. In this case the spin fluctuations are soft at small q measured relative to the ordering wavevector Q. The shift in origin $q \rightarrow Q + q$ leads to an expression for the relaxation rate at small q of the form $\Gamma_{Q+q} = \gamma(\chi_Q^{-1} + cq^2)$ for a cubic system, where χ_Q is the staggered static susceptibility and γ and c are appropriate new constants. This shift in origin implies that Landau damping does not affect the q-dependence of Γ_{Q+q} in leading order in q so we have z = 2 at the quantum critical point defined by $\chi_Q^{-1} \rightarrow 0$ as $T \rightarrow 0$. Further, we note that in contrast to the ferromagnetic limit the critical scattering of quasiparticles by spin fluctuations is now chiefly at large angle rather than small angle. These changes lead to an asymptotic exponent $\partial \ln \rho / \partial \ln T$ of 3/2, compared with 5/3 for the ferromagnetic case [4]. T_{sf} for the lower curve in figure 3 is defined as $T_{sf} = \gamma c q_c^2$. The parameters γ , c and q_c are numerically the same as those employed for the other curves (although now γ has different units), and χ_Q^{-1} is linear in T with the same slope as that used for the middle curve. It is found that the form of







Figure 4. A comparison of measured (a) and calculated (b) temperature dependence of the logarithmic derivative of the resistivity in MnSi plotted versus $\log_{10}(T \, (\text{K}))$ at the critical pressure. The calculation involves only the measured temperature dependence of the static susceptibility (the inset of (b)) [9], the parameters of the spin-fluctuation spectrum inferred from neutron scattering data [8] as discussed in the text, and a cut-off wavevector set equal to the Brillouin zone dimension (Γ X).

 $\partial \ln \rho / \partial \ln T$ as a function of T/T_{sf} is relatively insensitive to the precise values of the parameters γ , c and q_c , but may depend quite strongly on the temperature dependence of the relevant static susceptibility.

We note that the asymptotic powers (5/3 or 3/2) are reached only at temperatures far below (two or more orders of magnitude below) the effective spin-fluctuation band width T_{sf} . Further, a linear temperature dependence of ρ is seen to arise only in a relatively limited crossover range, except in the unrealistic case where the spin-fluctuation spectrum is independent of T (the upper curve in figure 3).

The experimental results for MnSi shown in figure 4(a) have some correspondence with the prediction for the ferromagnetic case (the middle curve in figure 3) except for an anomaly at low temperature. This feature is, however, largely accounted for when we replace in our expression for Γ_q the linear temperature variation of $\chi^{-1}(T)$ employed above with the functional form actually measured in MnSi at the critical pressure [9] (the inset of figure 4(b)). The static susceptibility is large at low *T*, but even at p_c it is not singular as naively expected. In particular, it exhibits a peak which is seen to account well for the experimental anomaly in $\partial \ln \rho / \partial \ln T$ (figures 4(a), 4(b)).

The calculations presented in figures 3 and 4(b) include only the effects of spin fluctuations in the simplest cases. In particular, they ignore scattering from phonons which may be expected to become progressively more important with increasing temperature. Further, they neglect effects due to residual frozen-in disorder and any drag phenomena which may lead to significant modifications in the low-temperature limit.

The spin-fluctuation model in its most elementary form also leads us to expect a linear dependence of $T_c^{4/3}$ with pressure [3, 5]. As shown in figure 5(a), however, this prediction breaks down near p_c . This behaviour and the non-singular form of $\chi(T)$ (which exhibits a peak near and above p_c) are related to each other and may be connected with the existence of a deep minimum in the one-particle density of states at the Fermi level [15]. The latter leads to a sharp initial upturn in the static susceptibility versus temperature and to the appearance of a weak first-order transition near the critical pressure [9]. We note that the related cubic ferromagnet ZrZn₂ (figure 5(b)) in which the magnetic transition appears to be essentially continuous [10].

Since $\chi(T)$ in MnSi is not strictly singular at p_c , we expect that the exponent $\partial \ln \rho / \partial \ln T$ will gradually increase towards 2 with decreasing temperature (figure 4(b)). There is evidence (figure 4(a) and the inset of figure 1), however, that the experimental exponent falls progressively below expectations as the temperature is decreased. A similar, but much more dramatic, discrepancy appears in the f-heavy-fermion systems CePd₂Si₂ and CeNi₂Ge₂ discussed in the next section.

5. f-heavy-fermion metals on the boundary of antiferromagnetic order: $CePd_2Si_2$ and $CeNi_2Ge_2$

CePd₂Si₂ and CeNi₂Ge₂ crystallize in a body-centred tetragonal structure (see the inset in figure 6) which characterizes a large family of Ce ternary compounds that includes the first of the heavy-fermion superconductors CeCu₂Si₂ [16]. At ambient pressure and below 10 K, CePd₂Si₂ orders in an antiferromagnetic structure with a relatively small static moment of approximately 0.6 μ_B at low temperatures [17]. As shown in the inset of figure 6, the spin configuration consists of ferromagnetic (110) planes with spins normal to the planes and alternating in directions along the spin axis. The isoelectronic and isostructural relative



Figure 5. The pressure dependence of the transition temperature for MnSi [9] (a) and ZrZn₂ [10] (b) plotted as $T_c^{4/3}$ versus pressure (and, in the inset, as T_c^2 versus pressure). In the vicinity of the critical pressure when T_c^2 is linear in pressure, the magnetic transition in MnSi is found to be weakly first order [9].



Figure 6. The temperature dependence of the resistivity along the *a*-axis of CePd₂Si₂ at different pressures [21]. The Néel temperatures T_N , marked by arrows, are visible as significant changes in the slope of ρ versus *T*. The ThCr₂Si₂ lattice structure of CePd₂Si and the spin configuration below T_N are illustrated in the insets. The Ce atoms are on the corners and at the centre of the tetragonal unit cell, and the Pd atoms are on the cell faces.

CeNi₂Ge₂ has a slightly smaller lattice constant and at ambient pressure exhibits no sharp phase transition down to the lowest temperature reached ($\sim 20 \text{ mK}$) [18–20]. It is reasonable to expect that its behaviour at ambient pressure is similar to that of CePd₂Si₂ at a pressure somewhat above that required to suppress antiferromagnetic order. CeNi₂Ge₂ therefore provides us with the opportunity to expand the effective range in pressure over which we may explore the behaviour of essentially the same stoichiometric heavy-fermion system close to the boundary of antiferromagnetic order.

As in the case of MnSi, we find that the transition temperature in CePd₂Si₂ falls continuously towards absolute zero (figure 6) and at the critical pressure $p_c \simeq 28$ kbar the temperature dependence of the resistivity is again found to be significantly slower than quadratic [21]. But in sharp contrast to the case for MnSi, not only T_N , but also the shoulder of ρ versus T, shifts rapidly with pressure (in a direction opposite to T_N [22]). At the critical pressure, the shoulder has shifted by nearly an order of magnitude above its position at ambient pressure.

In the wide range opened up between these characteristic temperatures near p_c , we observe a remarkably constant slope for $\log_{10}(\rho - \rho_0)$ versus $\log_{10}T$ (see the inset of figure 7(a)). As emphasized in figure 7(b), the resistivity is linear in T to a power close to 1.2 over nearly two decades in temperature down to approximately 0.4 K where our samples with the lowest residual resistivity become superconducting.

As shown in the phase diagram of figure 8, the superconducting regime extends over a relatively narrow pressure range following (and perhaps slightly overlapping with) a



Figure 7. The resistivity versus T (a) and versus $T^{1.2}$ at lower T (b) of CePd₂Si₂ near the critical pressure. The resistivity is essentially linear in $T^{1.2}$ over nearly two decades down to the onset of a superconducting transition near 0.4 K (the inset of (b)) [21].

regime where T_N falls towards absolute zero almost linearly with pressure. From the temperature variation of the superconducting upper critical field near p_c we infer a low-temperature BCS coherence length of approximately 150 Å, a magnitude characteristic of heavy-fermion superconductivity (see the inset of figure 7). Related high-pressure results have been reported for CeCu₂Ge₂ [23] and CeRh₂Si₂ [24]. What is unusual here, however, is that the normal state above T_s does not exhibit a temperature-dependent resistivity normally associated with a Fermi-liquid state. In some sense, this then represents a form of 'high-temperature' superconductivity; not high in absolute terms but in relation to some low-temperature scale apparently not yet reached on cooling to T_s . Among the heavy-fermion systems another extreme but qualitatively different example of such 'high-temperature' superconductivity is found in UBe₁₃ [25].

At sufficiently high pressures we expect to recover a Fermi-liquid (quadratic) form of ρ versus T which is ubiquitous in other paramagnetic heavy-fermion metals at low T. As



Figure 8. The temperature–pressure phase diagram of CePd₂Si₂. The Néel temperature T_N falls monotonically towards zero and is nearly linear in pressure before entering a relatively narrow region where superconductivity appears in the millikelvin range [21]. The magnetic field dependence of the superconducting transition temperature T_s (inset) exhibits a high slope characteristic of heavy-fermion superconductors.

stated earlier, CeNi₂Ge₂, with a slightly smaller cell volume than CePd₂Si₂ but otherwise with similar lattice and starting electronic structures, provides us with the opportunity to examine the crossover to the Fermi-liquid form of ρ versus T without the use of very high applied pressures. Initial studies in CeNi₂Ge₂ suggested a more or less unexceptional behaviour. In particular, ρ versus T was thought to have a conventional form characteristic of many normal heavy-fermion systems. But more detailed studies in samples with low residual resistivities have revealed that ρ does not exhibit a simple quadratic temperature variation except perhaps below one or two hundred mK and in fact varies as $\rho \sim T^{1.4\pm0.1}$ over a decade below a few K (see figure 9(a)) [26, 20]. This anomalous behaviour of the temperature dependence of the resistivity appears to extend up to 10 kbar or higher. A more conventional form of ρ versus T is observed, however, at 30 kbar and above the onset of a still unidentified phase transition which may emerge at a pressure intermediate between the above values. It is also noted that at ambient pressure ρ versus T has an unusual temperature dependence even in quite high applied magnetic fields (see figure 9(b)) and that, moreover, no quantum oscillatory effects have been observed in samples with residual resistivities as low as 1 $\mu\Omega$ cm under experimental conditions normally expected to be favourable to the detection of such effects.

The natures of the anisotropic spin-fluctuation spectra of CePd₂Si₂ and CeNi₂Ge₂ are not yet sufficiently well known to enable us to carry out a quantitative analysis analogous to that presented for the d-metal ferromagnets in section 4. Further, the validity of the naive extension of this model to antiferromagnetic systems is not clearly self-evident [27]. For the



Figure 9. The temperature variation of the basal plane resistivity of $CeNi_2Ge_2$ at ambient pressure in a small applied magnetic field (a) and at 15.8 T (b) along the *c*-axis [20].

very simplest case depicted by the lower curve in figure 3, the exponent $\partial \ln \rho / \partial \ln T$ falls monotonically from 3/2 (for $T \ll T_{sf}$) towards zero (for $T \gg T_{sf}$). The observation of a low-temperature exponent of less than 3/2 in CePd₂Si₂ at p_c is not necessarily inconsistent with this model since convergence of $\partial \ln \rho / \partial \ln T$ to 3/2 is seen to be very slow (figure 3). But the locking in of the experimental exponent to a fixed value over a wide temperature range (figure 7(a) inset, figure 7(b)) is not a feature of the present spin-fluctuation model. Also difficult to understand within this same framework is the non-Fermi-liquid form of ρ versus *T* at still higher pressures above p_c in CePd₂Si₂, or in the smaller-volume relative CeNi₂Ge₂ at ambient or low pressures and above one or two hundred mK.

It is not yet clear whether a consistent description of the above findings can be given in terms of a refined version of the model developed for the d-metal ferromagnets (section 4) or whether a radically different approach is required. In the f-heavy-fermion systems, in contrast to typical d metals, there may be an ambiguity in the spin-fluctuation theory as it is conventionally formulated. It is perhaps unclear in our systems whether the Fermi surface close to which the relevant quasiparticles are excited is that formed by the 'conduction electrons' together with the f electrons, as suggested by de Haas–van Alphen studies on a number of normal heavy-fermion compounds, or by the conduction electrons alone as is often assumed in 'intermediate-temperature' descriptions.

6. Summary

The idealized model for describing nearly ferromagnetic d metals, such as MnSi and ZrZn₂, near the critical point $T_c \rightarrow 0$, appears to be that of a marginal Fermi liquid which has also been invoked in theoretical treatments of the coupling of electrons to transverse photons and in the study of nuclear matter [28]. In both cases, the starting picture remains that of

fermion quasiparticles excited above a normal Fermi surface. In more extreme cases, an altogether different starting point may be required.

This is conceivably the case in some of the more strongly correlated electron systems among the heavy-fermion compounds (see also [29–35]). In particular, we have noted that a naive extension of the spin-fluctuation model used for the d-metal ferromagnets, but now with a dynamical exponent z = 2, cannot readily account for the curious locking into a fixed exponent $\partial \ln \rho / \partial \ln T$ over nearly two decades in temperature in CePd₂Si₂. Equally unusual is the appearance of a short-coherence-length superconducting state at temperatures which appear to be well above any effective 'Fermi' temperature.

Also difficult to reconcile with the simplest spin-fluctuation model is the slow return to a Fermi-liquid form of ρ versus *T* at pressures above p_c and in particular at ambient pressure in the related paramagnetic metal CeNi₂Ge₂, which has a unit-cell volume close to, but lower than, that of CePd₂Si₂. An analysis free of adjustable parameters, such as that carried out for MnSi and ZrZn₂, awaits a more complete determination of the spin-fluctuation spectrum for these f-electron metals.

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