RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 13 1 OCTOBER 1993-I

Phenomenological theory of non-Fermi-liquid heavy-fermion alloys

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(Received 1 March 1993; revised manuscript received 15 June 1993)

We propose a phenomenological theory for heavy-fermion metallic alloys $U_{0.2}Y_{0.8}Pd_3$ and $UCu_{3.5}Pd_{1.5}$ whose behavior demonstrates strong deviations from the Landau Fermi-liquid theory. The theory implies that the alloys have a critical point at $T = 0$ and therefore their low-temperature thermodynamics is determined not by single-particle fermion excitations, as in the Fermi liquid, but by the collective modes corresponding to fluctuations of the order parameter in the vicinity of the critical point. The observed properties are consistent with the fluctuation spectrum $\omega \sim q^3$. Both quantum spin-glass transition and quadrupolar ordering are ruled out by the scaling analysis.

The experimental observation of non-Fermi-liquid behavior of some heavy-fermion metallic alloys[1–3] has not yet obtained a satisfactory theoretical explanation. Recall the facts: according to the measurements described in Refs. 1–3, the metallic alloys $U_{0.2}Y_{0.8}Pd_3$ and $UCu_{3.5}Pd_{1.5}$ have the following low-temperature properties:

(i) The specific heat c_v is proportional to $T \ln 1/T$ (see Refs. 1–3).

(ii) The electrical resistivity is linear in temperature:[1–3] $\rho(T) = \rho(0)(1 - \alpha T)$ and it is almost independent of the magnetic field[2,3] up to $H = 10^5$ Oe. $\rho(0)$ is about 250–400 $\mu\Omega$ cm, which is very large, but still well below the maximum metallic resistivity $\rho_{\max} \approx 3000$ $\mu\Omega$ cm; therefore, one still can consider these materials as metals.

(iii) A scaling of the magnetization

$$M = \frac{H}{T^{\gamma}} f\left(\frac{H}{T^{\beta+\gamma}}\right), \tag{1}$$

where $\gamma = 0.25 - 0.3$, $\beta + \gamma = 1.2 - 1.3$, and $f(x)$ is some nonsingular function.[2,3]

(iv) The same scaling holds for the specific heat:[2,3]

$$\frac{c_v(H,T)}{T} - \frac{c_v(0,T)}{T} = g\left(\frac{H}{T^{\beta+\gamma}}\right). \tag{2}$$

(v) The behavior of the thermodynamic quantities becomes even more singular in UCu_4Pd, which is closer to the antiferromagnetic part of the phase diagram.

We expect that the uranium compounds are not unique in exhibiting non-Fermi-liquid behavior. There is some evidence that for $Ce_{1-x}Th_xRhSb$ alloy for concentrations $x = 0.3, 0.4$ the low-temperature specific heat is $C_v/T \propto \ln T$, and for $x \geq 0.5$ the spin-glass transition takes place.[4]

All these properties hold below $T \approx 10$ K. Well above this temperature the materials behave like Kondo alloys with the logarithmic temperature dependence of the electrical resistivity originating from scattering of conducting electrons on uranium ions.

The described picture does not look like anything one would expect for a metal; even disordered Kondo alloys have $c_v \propto T$ and $R(T) = R(0)(1 - aT^2)$ in agreement with the Nozières theory of the localized Fermi liquid[5]. One of the explanations of the phenomenon takes into account the fact that the Nozières theory would fail to describe the Kondo effect in the overscreened case when the conduction band has an additional degeneracy. There are strong theoretical arguments[1,6] that such degeneracy appears for an isolated U ion placed in a cubic environment. Then scattering of conduction electrons is described by the two channel Kondo model[7–9] which does not have the Fermi-liquid fixed point at T = 0. The exact solution of this model[8,9] predicts $c_v \propto T \ln 1/T$, but the agreement with the available experimental data does not go beyond this point (see Ref. 2 for details). It is also known that the non-Fermi-liquid fixed point is very sensitive to perturbations which are undoubtedly present in such concentrated systems like the U alloys under consideration. It is known[1–3] that relatively small changes in concentration of uranium convert these alloys into spin glasses. Therefore the interaction between U ions is strong indeed and the ions cannot be described as isolated. Thus the magnetic subsystem in the strong-coupling limit at low temperatures is hybridized with conducting electrons and a narrow band is formed.

The Fermi-liquid theory is based on the assumption that the low-energy properties are dominated by fermionic excitations. This assumption via the Pauli principle, implies an appearance of a natural energy scale in the theory, the chemical potential ϵ_F. All thermodynamic properties depend on $T/\epsilon_F, H/\epsilon_F$. We emphasize again that the field and temperature dependences of the specific heat and the magnetization described in Eqs. (1) and (2), and especially their scaling clearly demonstrate the absence of such scale and therefore irrelevance of fermionic excitations.

Therefore we suggest that in the case under consideration the low-temperature thermodynamics is dominated

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by collective bosonic modes. These modes arise due to the fact that the systems are on the verge of a phase transition which occurs at zero temperature.[10] The nature of the $T = 0$ instability remains obscure. The observed divergence of the magnetic susceptibility demonstrates the enhancement of ferromagnetic fluctuations. We argue, however, that the essential physics comes from spin-glass fluctuations. This idea is compatible with the experiments. Thus, the specific-heat measurements for $U_xY_{1-x}Pd_3$ presented in Ref. 1 show that the temperature dependence of C_v changes at $x > 0.25$. The authors interpret the observed bump in $C_v(T)$ as the spin-glass transition. The measurements of magnetic susceptibility $\chi(T)$ for $UCu_{5-x}Pd_x$ show that the region of non-Fermi-liquid behavior belongs to the window of concentrations $1 < x < 1.5$. At $x < 1$ the system is apparently an antiferromagnet, and the data obtained for $x = 2.3$ and $x = 2$ (see Ref. 3) show that below a certain temperature T_f the zero-field-cooled susceptibility differs from the field-cooled one, which is believed to be an indication of spin-glass transition. The spin-glass temperature T_f decreases with concentration of Cu; the linear extrapolation of the data presented in Ref. 3 indicates that T_f vanishes at $x = 1.6$.

In this paper we describe the non-Fermi-liquid alloys as systems critical at $T = 0$ and apply to them the scaling analysis. The scaling arguments are similar to those employed by Fisher *et al.*[11] and Chubukov, Sachdev, and Ye.[12] Let us consider a system with critical point at $T = 0$. The most general expression for its free energy is

$$F = -Tf\left(\frac{T}{T_K}, \frac{h_i}{T^{\delta_i}}\right) \tag{3}$$

where T_K is the ultraviolet cutoff (the Kondo temperature in the present case) and h_i are external fields. The fields are relevant if their dimensions are positive $\delta_i > 0$. Strictly speaking, in the scaling region one must consider only the leading singularity of the free energy and then perform the limit $T_K \to \infty$. The limit should be taken, however, only after the most singular part of the free energy is found. Suppose that the excitation spectrum at the critical point is given by

$$\omega \sim q^z; \tag{4}$$

then the dimensional analysis gives the following estimate for the free energy:

$$F \approx -T\sum_n \int d^dq \ln D(i\omega_n, q) \sim T^{1+d/z} \tag{5}$$

where $D(i\omega_n, q)$ is the most singular propagator. Therefore in the limit $T_K \to \infty$ we have

$$F = -T^{1+d/z} g\left[\frac{h_i}{T^{\delta_i}}\right]. \tag{6}$$

Expanding this expression at small h_i, we find the indices of the singular susceptibilities:

$$F = -T^{1+d/z}\left[a_0 + \sum_i a_i \frac{h_i^2}{T^{2\delta_i}} + \cdots\right], \tag{7}$$

$$\chi_i(0) \sim T^{-\gamma_i} \equiv T^{-2\delta_i+1+d/z}. \tag{8}$$

Each external field is coupled to some operator, i.e., associated with the following change of the thermodynamic action:

$$S = S_0 + \sum_i h_i \int_0^{1/T} d\tau \int d^dx \Phi_i(\tau, x). \tag{9}$$

Therefore the susceptibilities $\chi_i(0)$ are related to the thermodynamic correlation functions of the operators Φ_i:

$$\chi_i(0) = \int_0^{1/T} d\tau \int d^dx \langle\langle \Phi_i(\tau, x)\Phi_i(0,0)\rangle\rangle. \tag{10}$$

At $T = 0$ the correlation length is infinite. This fact together with Eq. (4) guarantees the following form of the correlation functions at $T = 0$:

$$\langle\langle \Phi_i(\tau, x)\Phi_i(0,0)\rangle\rangle = \frac{1}{|x|^{\Delta_i}} G_i(\tau/|x|^z). \tag{11}$$

At finite temperatures we have a finite cutoff in the time direction: the correlation functions become periodic in τ with a period $1/T$. Therefore we expect that Eq. (11) holds until $\tau < 1/T, |x| < 1/T^{1/z}$. It means that at finite T there is a finite correlation length $\xi \sim T^{-1/z}$. Substituting expression (11) into Eq. (10), cutting the integration over x at ξ and comparing the result with (8) we get the following relation between the exponents:

$$\Delta_i = d + z(1 - \delta_i). \tag{12}$$

At $\tau = 0$ the correlations must decay at large distances, which means that $\Delta_i > 0$. It imposes the restriction on γ's:

$$\delta_i < 1 + d/z. \tag{13}$$

Now let us apply this general theory to the experimental data. The data on the specific heat show that $C \sim T \ln T$ which means $z = d = 3$ (the scaling arguments do not control logarithms). According to Eq. (13) it means that $\delta_i < 2$. This simple inequality is very important. In particular, it rules out any scenario related to a spin-glass transition or to quadrupolar fluctuations. In both these cases a magnetic field couples to a relevant field quadratically, i.e., $h = H^2$ in our notations. (Recall that the divergent susceptibility for the spin-glass transition is the nonlinear magnetic susceptibility, i.e., the singularity occurs in the fourth derivative of the free energy with respect to magnetic field). The data show that the magnetic field scales as T^δ with $\delta = 1.3 \pm 0.1$ which means that H^2 has the scaling di-

mension $\delta_{H^2} = 2\delta = 2.6 \pm 0.2 > 2$. Such an estimate contradicts the restriction (13). Therefore the magnetic field can couple to the relevant fields only linearly. From Eq. (8) we get

$$\beta_i = 1 - \tfrac{1}{2}\gamma_i. \tag{14}$$

The results of magnetic measurements are in apparent contradiction to this identity: the exponent of the magnetic susceptibility $\gamma = 0.25$–0.35 and the scaling dimension of the magnetic field is $\gamma + \beta = 1.3 \pm 0.1$ which gives the 15% discrepancy in Eq. (14). Recall, however, that the scaling form of the free energy (6) assumes the limit $T/T_K \to 0$. One has to expect deviations from the scaling at larger temperatures. For the systems under consideration the scaling breaks down around 10–15 K. The experimental points presented in Ref. 2 were taken in different intervals of temperature: the data for the specific heat at 0.35–2 K and the data for magnetization at 1.8–8 K. Another possibility is that the magnetic field is coupled not to one, but to two relevant fields with slightly different scaling dimensions. In the latter case the one parametric scaling fits of the data given by Eqs. (1) and (2), but with less accuracy at low temperatures. This problem requires more experimental data to be available.

We can try to guess what kind of theory would describe the observed scaling. Let us accept as a conjecture the following singular propagator at $T = 0$:

$$D^{(R)}(\omega, q) = \frac{1}{i\omega/q + aq^2}. \tag{15}$$

The propagator is diffusive which implies that there are no propagating gapless excitations even at $T = 0$. The conjectured propagator gives the correct specific heat:

$$C/T = \frac{\partial}{\partial T}\int d\omega \frac{\omega}{2T^2 \sinh^2 \omega/2T} \int d^3q \tan^{-1}\left(\frac{\Im m D^{(R)}(\omega, q)}{\Re e D^{(R)}(\omega, q)}\right) \propto \ln T. \tag{16}$$

It also gives the correct scaling dimension $\delta = 4/3$. One obvious way to test this conjecture is to measure the NMR relaxation rate. According to Eq. (15) it should be

$$1/T_1 \sim \lim_{\omega \to 0} \int d^3q \frac{\Im m \chi(\omega, q)}{\omega} \sim T^{1/3}. \tag{17}$$

In conclusion, we presented the scaling analysis of the low-temperature properties of the alloys $U_{0.2}Y_{0.8}Pd_3$ and $UCu_{3.5}Pd_{1.5}$ which shows the magnetic nature of this phenomena. The magnetic field is coupled linearly to the fields with singular correlation functions (relevant fields). Fluctuations of these fields dominate the thermodynamic properties; their spectrum scales as $\omega \sim q^3$.

We are grateful to B. Andraka and S. Sachdev for intensive and valuable discussions, to D. Cox for the constructive criticism, and to D. Sherrington, A. Bray, P. Coleman, and G. Lonzarich for interest in the work, valuable discussions, and illuminating remarks. This work was supported in part by the Oxford Department of Physics rolling grant (A.T.) and by U.S. DOE Contract No. DE-FG02-ER45347 (M.R.).

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