Response of the heavy-fermion superconductor $CeCoIn_5$ to pressure: roles of dimensionality and proximity to a quantum-critical point

2001 J. Phys.: Condens. Matter 13 L905

(http://iopscience.iop.org/0953-8984/13/44/104)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) L905–L912 PII: S0953-8984(01)28178-8

LETTER TO THE EDITOR

Response of the heavy-fermion superconductor $CeCoIn_5$ to pressure: roles of dimensionality and proximity to a quantum-critical point

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Received 23 August 2001, in final form 18 September 2001
Published 19 October 2001

Abstract

We report measurements of the pressure-dependent superconducting transition temperature T_c and electrical resistivity of the heavy-fermion compound $CeCoIn_5$. Pressure moves $CeCoIn_5$ away from its proximity to a quantum-critical point at atmospheric pressure. Experimental results are qualitatively consistent with theoretical predictions for strong-coupled, d-wave superconductivity in an anisotropic three-dimensional superconductor.

Although heavy-fermion superconductivity has been known of for over two decades, a microscopic theory of this broken-symmetry state has remained elusive. Experiments have established, however, that the unconventional superconductivity in heavy-fermion compounds is most probably mediated by magnetic fluctuations, which also are responsible for enhancing the effective mass of itinerant quasiparticles by two to three orders of magnitude [1–3]. In spite of 20 years of searching, there has been, until very recently, only one example known of a Ce-based heavy-fermion compound that is superconducting at atmospheric pressure, namely $CeCu_2Si_2$ [4]. In contrast, several examples of Ce-based heavy-fermion antiferromagnets have been found in which superconductivity appears as their Néel temperature is tuned toward $T = 0$ with the application of pressure [5–8]. This recent series of discoveries reinforces the belief not only that a magnetic interaction is responsible for superconductivity in these materials but also that heavy-fermion superconductivity may be favoured at a particular 'soft spot' in phase space, i.e., near a quantum-critical point [9].

In heavy-fermion superconductors, the characteristic spin-fluctuation temperature T_{sf} appears to set the scale for T_c, in much the same way that the characteristic phonon frequency does in the BCS (Bardeen–Cooper–Schrieffer) theory of conventional superconductors. The most direct measure of T_{sf} comes from the linewidth of neutron quasi-elastic scattering, but

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it is frequently estimated from the magnitude of the specific heat Sommerfeld coefficient $\gamma \propto 1/T_{sf}$. A very large Sommerfeld coefficient, the hallmark of heavy-fermion behaviour, implies, then, a small T_{sf} and consequently low T_c. As might be expected, the T_cs of all pressure-induced Ce superconductors are less than 0.5 K and that of $CeCu_2Si_2$ is about 0.7 K. At the other extreme are the cuprates, in which T_{sf} and T_c are both larger by roughly two orders of magnitude than in heavy-fermion systems. Monthoux and Lonzarich have argued, specifically in the context of cuprates, that, in addition to T_{sf}, dimensionality is a factor in determining the transition temperature of magnetically mediated superconductors [10]. Their solution of Eliashberg equations shows that, with all other factors being the same, the mean-field T_c of a two-dimensional system will be higher than that of its 3D analogue. There have been no examples, however, of families of magnetically mediated superconductors that crystallize in related 3D and 2D structures available to test these suggestions.

The very recent observation of heavy-fermion superconductivity in $CeMIn_5$ (M = Co, Ir, and Rh) offers the opportunity for at least a qualitative inspection of the role of dimensionality. These compounds form in the tetragonal $HoCoGa_5$ crystal structure that can be viewed as layers of $CeIn_3$ and 'MIn_2' stacked sequentially along their c-axis. $CeCoIn_5$ [11] and $CeIrIn_5$ [12] superconduct at atmospheric pressure with bulk T_cs of 2.3 and 0.4 K, respectively, while $CeRhIn_5$ [13] is an incommensurate antiferromagnet that transforms to a superconductor with $T_c = 2.1$ K at pressures greater than about 1.6 GPa. For comparison, cubic $CeIn_3$, which is the infinite-layer variant of these compounds, becomes a superconductor at pressures near 2.5 GPa with a maximum T_c of about 0.25 K [8]. A very crude approximation of the role of T_{sf} in determining the magnitude of T_c in these materials comes from taking $T_c \propto T_{sf} \propto 1/\gamma$. For $CeMIn_5$, $T_c = 2.3$, 0.4, and 2.1 K for M = Co, Ir, and Rh, respectively. Their corresponding values of γ are 250 [11], 750 [12], and 380 [14] mJ mol^{-1} K^{-2} for $T \geqslant T_c$. Within the $CeMIn_5$ family, then, T_c scales approximately with $1/\gamma \propto T_{sf}$. The Sommerfeld coefficient of $CeIn_3$ at 2.5 GPa is not known, but an upper limit should be its $P = 0$ value of 130 mJ mol^{-1} K^{-2} [15]. With all other factors assumed equal, we would expect from this comparison that T_c for $CeIn_3$ should be over 3 K at 2.5 GPa, a factor of 10 higher than observed experimentally. In view of the calculations by Monthoux and Lonzarich, the high transition temperatures of $CeMIn_5$ members, relative to that of $CeIn_3$, may be due in part to their layered crystal structure.

In the following, we present results of pressure-dependent measurements on $CeCoIn_5$ and discuss the relationship of its layered structure, particularly in the context of predictions by Monthoux and Lonzarich [10], and apparent proximity to a quantum-critical point to an interpretation of its pressure response.

Four-probe AC resistivity measurements, with current flow in the tetragonal basal plane, were performed on a single crystal of $CeCoIn_5$ grown from excess-In flux. Pressure was produced in a conventional Be–Cu clamp-type cell using pentene as the pressure medium, and the clamped pressure at low temperatures was determined from the shift in the inductively measured T_c of a small piece of Pb located in close proximity to the sample. A comparison of the T_cs of Pb and $CeCoIn_5$ with literature values at atmospheric pressure showed that there was a small temperature gradient, at most 0.7% of the thermometer temperature, between the sample volume and the calibrated Cernox thermometer that was thermally anchored in the wall of the pressure cell. Independent measurements of the pressure dependence of T_c of $CeCoIn_5$ were carried out over a more limited pressure range using a small clamp cell in a Quantum Design Superconducting Quantum Interference Device magnetometer, with Flourinert FC 75 (3M) as the pressure medium and a Pb manometer.

The pressure dependence of the resistivity of $CeCoIn_5$ over a broad temperature range is plotted in figure 1. At atmospheric pressure, $\rho(T)$ reaches a maximum at $T_M = 50$ K that is generally associated with a crossover from incoherent Kondo-like scattering at high

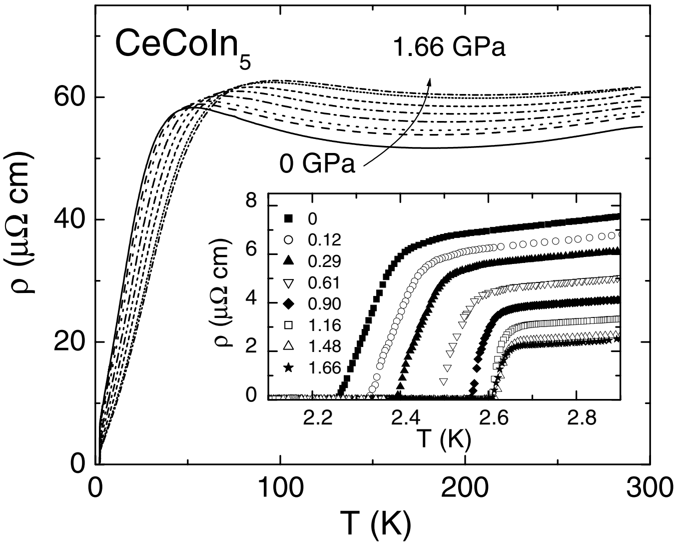

Figure 1. The temperature dependence of the in-plane resistivity of CeCoIn$_5$ at various fixed pressures noted in the inset. With increasing pressure, the resistivity increases at high temperatures and decreases at low temperatures. The inset shows the pressure response of the low-temperature resistivity and superconducting transition temperature.

temperatures to a heavy-fermion band state at low temperatures. With increasing P, T_M moves to higher temperatures, and the resistivity above T_M increases monotonically, whereas the resistivity below T_M decreases. This overall pressure response is typical of Ce-based heavy-fermion materials and is consistent with a P-induced shift of the characteristic spin-fluctuation temperature T_{sf} and spin-fluctuation scattering to higher temperatures [16]. A more detailed view of the low-temperature behaviour, given in the inset, shows that the resistivity just above T_c decreases from slightly over 0.06 to about 0.02 $\mu\Omega$ m and T_c increases from 2.25 to 2.6 K as the pressure is raised.

Explicit pressure dependencies of T_M and T_c are shown in figure 2. T_M is linear in P over the entire pressure range, with $\mathrm{d}T_M/\mathrm{d}P = 28$ K GPa^{-1}. Assuming that $T_M \propto T_{sf} \propto 1/\gamma$, such a large rate of increase implies a rapid decrease in the electronic contribution to the specific heat. Indeed, direct measurements of $\gamma(P)$ show [17] this expected behaviour, and we find that $1/T_M(P)$ is linearly proportional to the normal-state $\gamma(P)$. The relative change $\partial \ln \gamma / \partial \ln(1/T_M) = 1.2$ falls well within the range of values where this comparison has been possible for other heavy-fermion systems [16]. Over this same pressure interval, T_c increases initially at a rate $\mathrm{d}T_c/\mathrm{d}P = 0.5$ K GPa^{-1} and reaches a maximum value of 2.61 K near 1.5 GPa before decreasing. The inset shows that the resistive transition width sharpens substantially from $\Delta T_c \approx 80$ mK at $P = 0$ to around 20 mK for $P > 0.9$ GPa.

It is interesting to compare these trends with the pressure response of CeRhIn$_5$. In that case, pressure induces a rather abrupt (as measured by the Néel temperature) change from antiferromagnetic to superconducting states near 1.6 GPa. Just beyond the critical pressure required for superconductivity, the resistivity at $T \geqslant T_c$ is large and ΔT_c is relatively broad, but with increasing P, $\rho(T \geqslant T_c)$ drops, ΔT_c sharpens, $\mathrm{d}T_M/\mathrm{d}P$ is comparable to that found in CeCoIn$_5$ [13], and T_c passes over a maximum near 3.0 GPa [18]. From this comparison, it appears that CeCoIn$_5$ at atmospheric pressure is very similar to CeRhIn$_5$ at a pressure of about 1.6 GPa, and one might infer that CeCoIn$_5$ is near a quantum-critical point at

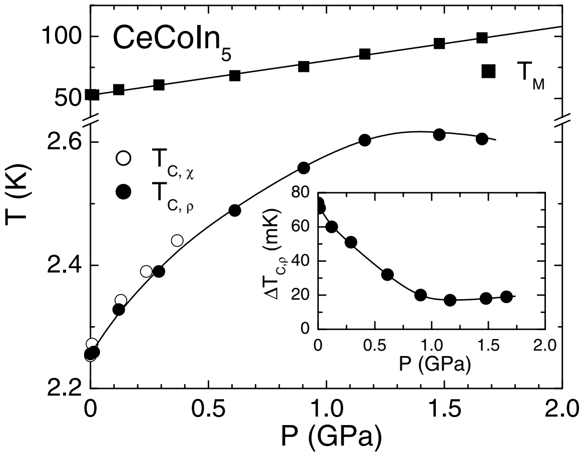

Figure 2. Pressure dependences of the temperature T_M at which the resistivity is a maximum (solid squares) and the superconducting transition temperature T_c, determined where the resistivity is zero (solid circles) and by the mid-point of the magnetic transition (open circles). The inset plots the resistive transition width as a function of pressure. ΔT_c was obtained from the temperatures at which the resistivity reached 90% and 10% of its value just above onset of superconductivity.

atmospheric pressure. Indeed, specific heat measurements on CeCoIn$_5$ in a magnetic field sufficiently large to suppress T_c to zero find $C/T \propto -\ln T$ over more than a decade in temperature above 100 mK, behaviour that is accompanied by a resistivity $\rho - \rho_0 \propto T^{\alpha}$, with $\alpha \approx 0.95$ [11, 17, 19]. These temperature dependencies of C/T and ρ are clear indications of a non-Fermi liquid state [20].

A variation in T_c with tuning parameter, similar to $T_c(P)$ in figure 2, is also predicted in the calculations of Monthoux and Lonzarich [10] for a p-wave or d-wave superconductor in proximity to ferromagnetic and commensurate antiferromagnetic instabilities, respectively. There is substantial evidence pointing to the conclusion that CeCoIn$_5$ is a d-wave superconductor [21, 22] and, thus, we focus on that case. Before comparing our measurements to these calculations, we briefly summarize relevant assumptions and results of those calculations. In reference [10], the authors consider a square (2D) or cubic (3D) lattice in which the dominant scattering of quasiparticles is magnetic, with a constant spin-fluctuation temperature T_{sf}. From a mean-field solution of Eliashberg equations, they obtain T_c/T_{sf} as a function of two dimensionless parameters, the square of the inverse magnetic correlation length κ^2, and an effective interaction parameter $g^2\chi_0/t$ arising from the exchange of magnetic fluctuations. $\kappa^2 \to 0$ corresponds to the quantum-critical point where the magnetic correlation length diverges. For fixed values of $\kappa^2 > 0$, T_c/T_{sf} increases monotonically from zero as a function of $g^2\chi_0/t$ and saturates in the strong-coupling limit; whereas, for fixed $g^2\chi_0/t$, T_c/T_{sf} decreases monotonically with κ^2 for a 3D superconductor but passes over a maximum at small κ^2 for the 2D case. In the absence of direct measurements of either κ^2 or $g^2\chi_0/t$, we follow the suggestions in reference [10] and take pressure as a qualitative measure of κ^2 and the resistivity ρ_0 just above T_c as a comparable measure of $g^2\chi_0/t$ for a comparison of the theoretical calculations to experimental quantities.

The theoretical prediction of T_c/T_{sf} versus κ^2 for a relatively strong-coupled, 2D, d-wave superconductor near an antiferromagnetic quantum-critical point is qualitatively like $T_c(P)$ for CeCoIn$_5$, particularly in the sense that they each exhibit a maximum. Because CeCoIn$_5$ is strongly coupled [11] and appears to be a d-wave superconductor near a quantum-critical point, we might conclude from this comparison that it is also in the 2D limit. The model calculations, however, take T_{sf} to be a rather large energy scale, a substantial fraction of the tight-binding hopping integral, and thus to be independent of κ^2 or pressure. Though this assumption may be quite reasonable for cuprate superconductors, T_{sf} in heavy-fermion systems is typically much smaller, by one to two orders of magnitude, than in the cuprates and is pressure dependent [16]. Taking $T_M \propto T_{sf}$, we find that the ratio $T_c(P)/T_M(P)$ is a monotonically decreasing function of pressure, as shown in figure 3. Explicitly including this pressure dependence of T_M makes a definitive comparison between our experimental results and the theoretical expectation for a 2D superconductor ambiguous.

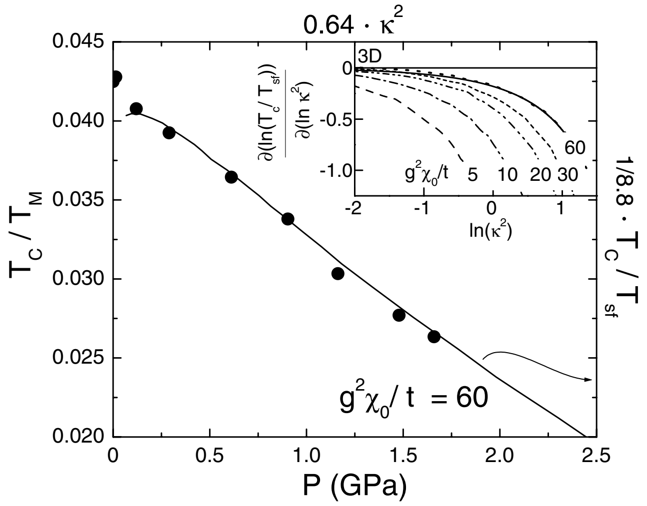

Figure 3. $T_c(P)/T_M(P)$, left-hand ordinate, versus pressure, bottom abscissa. Experimental values plotted as solid circles were taken from data in figure 2. The solid line is a theoretical prediction (right-hand ordinate, top abscissa) of T_c/T_{sf} versus κ^2 for a 3D superconductor with coupling constant $g^2\chi_0/t = 60$. See the text for details. In this comparison, we have taken $0.64\kappa^2 = P$ and $8.8T_{sf} = T_M$. A value of $g^2\chi_0/t = 60$ was chosen for the comparison on the basis of results shown in the inset, where theoretical values (broken curves) of $\partial \ln(T_c/T_{sf})/\partial \ln(\kappa^2)$ versus $\ln \kappa^2$ for fixed $g^2\chi_0/t$ are plotted on the same scale as experimental values (solid curve) of $\partial \ln(T_c/T_M)/\partial \ln P$ versus $\ln P$.

We emphasize this ambiguity by comparing theoretical predictions for a 3D superconductor to $T_c(P)/T_M(P)$ versus pressure in figure 3 and in figure 4 to T_c/T_M as a function of the electrical resistivity $\rho_0(P)$. We have narrowed the phase space of possible theoretical parameters by comparing logarithmic derivatives of the theoretical curves and the experimental data. This type of comparison is independent of proportionality factors between experimental observables and theoretical parameters. As shown in the inset to figure 3, the experimental data $\partial \ln(T_c/T_M)/\partial \ln P$ versus $\ln P$ are described best by the theoretical prediction for $\partial \ln(T_c/T_{sf})/\partial \ln \kappa^2$ versus $\ln \kappa^2$ when the coupling constant $g^2\chi_0/t = 60$. The only free parameters in this comparison are a proportionality factor between T_{sf} and T_M and a factor relating P to κ^2. We plot in the main body of figure 3 the theoretical curve from reference [10] for T_c/T_{sf} as a function of κ^2 at fixed $g^2\chi_0/t = 60$. The value of T_c/T_{sf} has been multiplied by 1/8.8 and κ^2 by 0.64. These scaling factors produce semiquantitative agreement with the experimental data, T_c/T_M versus P, shown by solid circles. Likewise, in the inset of figure 4, the experimentally determined $\partial \ln(T_c/T_M)/\partial \ln \rho_0$ versus $\ln \rho_0$ is compared to the theoretical predictions for $\partial \ln(T_c/T_{sf})/\partial \ln(g^2\chi_0/t)$ versus $\ln(g^2\chi_0/t)$. Best agreement

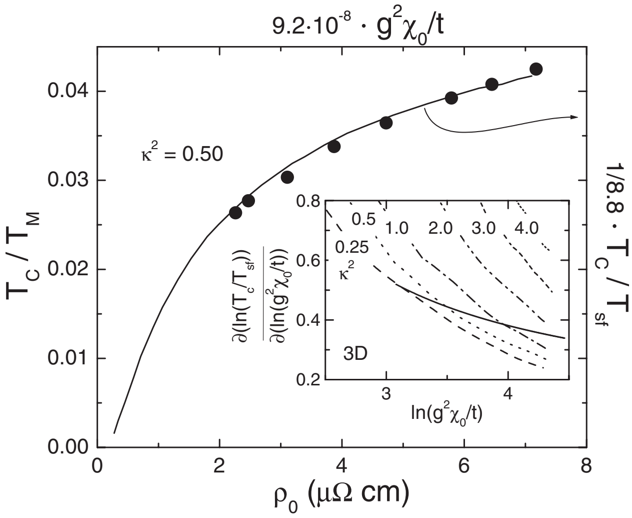

Figure 4. $T_c(P)/T_M(P)$, left-hand ordinate, versus electrical resistivity ρ_0 determined just above the onset of superconductivity, bottom abscissa. Solid circles are experimental data taken from figures 1 and 2. The solid line is a theoretical prediction (right-hand ordinate, top abscissa) of T_c/T_{sf} versus $g^2\chi_0/t$ for a 3D superconductor with $\kappa^2 = 0.5$. For the theoretical curve, we have taken $9.2 \times 10^{-8} g^2\chi_0/t = \rho_0$ and $8.8T_{sf} = T_M$. The inset shows $\partial \ln(T_c/T_{sf})/\partial \ln(g^2\chi_0/t)$ versus $\ln(g^2\chi_0/t)$ for fixed κ^2 (broken curves) and the experimentally determined $\partial \ln(T_c/T_M)/\partial \ln \rho_0$ versus $\ln \rho_0$ (solid curve) on the same scales.

between experiment and theory is found for values of κ^2 less than ~1.0. The main body of figure 4 shows experimentally determined T_c/T_M versus ρ_0 and the theoretical prediction of T_c/T_{sf} as a function of $g^2\chi_0/t$ for fixed $\kappa^2 = 0.5$. T_c/T_{sf} has been multiplied by 1/8.8, as in figure 3, and $g^2\chi_0/t$ has been multiplied by 9.2×10^{-8}. Again, agreement between experiment and theory is satisfactory. Overall, the theoretical predictions shown in figures 3 and 4 describe the experimental results reasonably well. The value $\kappa^2 = 0.50$, which provides a reasonable approximation of $T_c(P)/T_M(P)$ versus ρ_0, implies that $CeCoIn_5$ is close to a quantum-critical point. Likewise, the theoretical curve for $g^2\chi_0/t = 60$, which corresponds to very strong coupling[4], qualitatively reproduces the $T_c(P)/T_M(P)$ versus pressure data. (For simplicity in this comparison, in figure 3 we have taken $\kappa^2 = 0$ at $P = 0$, which implies that $CeCoIn_5$ is precisely at a quantum-critical point at atmospheric pressure. Somewhat better agreement between experimental values and the theoretical prediction would result if we had chosen $P \propto (\kappa_0^2 + \kappa^2)$, where κ_0^2 corresponds to a large but finite magnetic correlation length at $P = 0$.) If these parameters have been chosen correctly, they also should 'predict' $\rho_0(P)$ with no other adjustments. From theoretical curves [10] of T_c/T_{sf} versus κ^2, for fixed $g^2\chi_0/t = 60$, and of T_c/T_{sf} versus $g^2\chi_0/t$, for fixed $\kappa^2 = 0.5$, we can obtain $g^2\chi_0/t$ ($\propto\rho_0$) as a function of κ^2 ($\propto P$), using T_c/T_{sf} as the implicit variable. The rather good agreement found in figure 5 between direct measurements of $\rho_0(P)$ and the solid theoretical curve for $g^2\chi_0/t$ versus κ^2 indicates that our choice of $g^2\chi_0/t = 60$ for varying κ^2 in figure 3 and $\kappa^2 = 0.5$ for varying $g^2\chi_0/t$ in figure 4 are reasonable. Strong coupling, d-wave symmetry, and proximity to a quantum-critical point are also conclusions drawn in the preceding paragraph. Unlike

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[4] We note that RPA estimates of $g^2\chi_0/t$ given in reference [10] find expected values of the coupling parameter to be in the range 5 to 20. The large jump in specific heat $\Delta C/\gamma T_c \approx 4.5$ found for $CeCoIn_5$ [12] indicates very strong coupling that could exceed the RPA estimate.

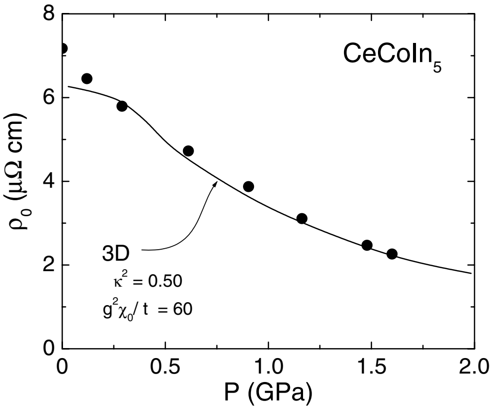

Figure 5. Resistivity ρ_0, determined just above the onset of superconductivity, as a function of pressure. Solid circles are experimental results and the solid curve is a theoretical prediction for a 3D superconductor with $\kappa^2 = 0.5$ and $g^2\chi_0/t = 60$. For the theoretical curve, we have used, as in figures 3 and 4, $P = 0.64\kappa^2$ and $\rho_0 = 9.2 \times 10^{-8} g^2\chi_0/t$.

the previous conclusions, however, those drawn from solid curves in figures 3–5 are based on theoretical curves for a 3D superconductor. Comparable agreement between experiment and theory for the 2D limit was not possible.

The semiquantitative agreement between experiment and theory must be considered somewhat fortuitous. As noted earlier, the calculations of reference [10] assume that T_{sf} is independent of pressure, and, further, that the coupling constant $g^2\chi_0/t$ and κ^2 are independent parameters. The substantial pressure dependence of T_M and specific heat Sommerfeld coefficient of CeCoIn$_5$ point to an intrinsic pressure dependence of its characteristic spin-fluctuation temperature. Though we have used ρ_0 as an approximation to $g^2\chi_0/t$, more direct measurements of the coupling, from measurements of the specific heat of CeCoIn$_5$ as a function of pressure [17], find that $\Delta C/\gamma T_c|_{T_c}$ is a monotonically decreasing function of P, qualitatively similar to $\rho_0(P)$. Therefore, it appears that T_c, T_{sf}, $g^2\chi_0/t$, and κ^2 are intimately coupled in CeCoIn$_5$ in a much more complex way than was assumed in reference [10]. In spite of these difficulties, the general trends found experimentally and in the theoretical calculations are those that might be expected from what is known about magnetically mediated superconductivity and CeCoIn$_5$ specifically, and we believe that, at least qualitatively, the two are consistent. Further refinement of theory to account for the interdependence of parameters would allow a more straightforward comparison to experiment. Beyond this is the issue of dimensionality that has not been resolved. However, very recent calculations by Monthoux and Lonzarich [23] show that any amount of anisotropy enhances T_c above a strictly 3D value for any κ^2 and $g^2\chi_0/t$. A comparison of CeCoIn$_5$ to cubic CeIn$_3$ supports this conclusion and suggests that CeCoIn$_5$ should be considered to be an anisotropic 3D system.

In summary, a comparison to CeRhIn$_5$ indicates that the electronic state of CeCoIn$_5$ at atmospheric pressure is analogous to that of CeRhIn$_5$ at a pressure of about 1.6 GPa, where superconductivity develops as its Néel transition disappears. Thus, from its pressure response as well as its properties at atmospheric pressure, CeCoIn$_5$ appears to be close to a quantum-critical point. We have analysed data in terms of theoretical predictions for a d-wave superconductor in both 2D and 3D limits and find that, though the 3D predictions semiquantitatively reproduce experimental observations, this agreement must be treated with caution. The analysis, however, does suggest avenues for extending the calculations to make a comparison to experiment more meaningful. It is unlikely that $CeCoIn_5$ should be considered a formally 2D system, but its clear electronic [24, 25] and crystallographic anisotropy plays a role in determining T_c at both atmospheric and applied pressures.

We thank P Monthoux for helpful discussions. Work at Los Alamos was performed under the auspices of the US Department of Energy. Work at the MPI-CPfS was conducted within the FERLIN programme of the ESF.

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