

High Resolution Study of Magnetic Ordering at Absolute Zero

M. Lee,¹ A. Husmann,² T. F. Rosenbaum,¹ and G. Aeppli³

¹*The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637, USA*

²*Toshiba Research Europe Ltd., 260 Cambridge Science Park, Cambridge CB4 0WE, United Kingdom*

³*London Centre for Nanotechnology and Department of Physics and Astronomy, UCL, London WC1E 6BT, United Kingdom*

(Received 12 January 2004; published 6 May 2004)

High resolution pressure measurements in the zero-temperature limit provide a unique opportunity to study the behavior of strongly interacting, itinerant electrons with coupled spin and charge degrees of freedom. Approaching the precision that has become the hallmark of experiments on classical critical phenomena, we characterize the quantum critical behavior of the model, elemental antiferromagnet chromium, lightly doped with vanadium. We resolve the sharp doubling of the Hall coefficient at the quantum critical point and trace the dominating effects of quantum fluctuations up to surprisingly high temperatures.

DOI: 10.1103/PhysRevLett.92.187201

PACS numbers: 75.40.Cx, 71.27.+a, 75.30.Fv

Phase transitions at the absolute zero of temperature are a result of Heisenberg's uncertainty principle rather than of a thermal exploration of states. Their ubiquity in materials of large technological interest, including transition metal oxides and sulfides, metal hydrides, superconducting cuprates, and colossal magnetoresistance manganites, combined with the intellectual challenge presented by many strongly interacting quantum degrees of freedom, places quantum phase transitions at the core of modern condensed matter physics. Quantum fluctuations inextricably intertwine the static and dynamical response of the material changing state, introducing new critical exponents, new scaling laws, and new relationships between the spin and charge degrees of freedom [1–3].

Varying the quantum fluctuations required for zero-temperature phase transitions is more difficult than changing temperature, with the result that the understanding of quantum phase transitions is far less detailed than that of their classical analogs. Typically, fluctuations are varied by scanning the composition of an alloy such that a new sample, each with unique disorder, must be fabricated for every zero-point fluctuation rate sampled. Tuning the transition with an external magnetic field—a quantity that is easily and precisely regulated—is a cleaner technique and provides correspondingly greater detail for both insulators [4] and metals [5]. However, magnetic fields also break time-reversal symmetry, which is particularly significant for quantum phase transitions because the dynamics responsible for zero-point fluctuations are altered profoundly. It is important, therefore, to examine a quantum phase transition for a simple material with high precision without applying a symmetry-breaking field. Hence, we have performed a high-resolution hydrostatic pressure study of a model quantum phase transition: elemental chromium diluted with its neighbor vanadium, small amounts of which can smoothly sup-

press Cr's spin-density-wave transition to $T = 0$. The Cr-V single crystals permit tuning with high fidelity, and we are able to characterize precisely the signatures of vanishing magnetic order in a system sufficiently simple to promise theoretical tractability.

Cr is the archetypical metallic antiferromagnet for which conduction electrons are lost as they order magnetically when the temperature passes below the Néel temperature T_N . The loss of carriers is most dramatically seen in the Hall effect, which measures the density of free, metallic carriers. The fundamental issue at a metallic quantum critical point (QCP) is what occurs to the free carriers that eventually order magnetically: should they be counted as free or as localized? In a previous experiment, we have examined alloys of $\text{Cr}_{1-x}\text{V}_x$ and tracked the loss of carriers as a function of x . We found a jump in the Hall number at the QCP even though other measures, such as the Néel temperature and the internal magnetization, vanished continuously [6]. The present work demonstrates that if we resolve the quantum critical point with 2 orders of magnitude greater precision than is possible by varying alloy composition, the Hall number does eventually undergo a continuous evolution at the $T = 0$ phase transition. Moreover, we are able to fix three critical exponents that characterize this quantum many-body problem and find that the diagonal and off-diagonal elements of the resistivity tensor behave differently through the QCP.

We applied hydrostatic pressure to single crystals of $\text{Cr}_{0.968}\text{V}_{0.032}$ of 1 mm³ volume with a BeCu piston-anvil cell with a WC insert. Tuning the $T = 0$ transition with pressure fixes the disorder from the V substitution and, in fact, we find that the $T \rightarrow 0$ disorder scattering (paramagnetic contribution to the longitudinal resistivity) is independent of applied pressure. At $x = 0.032$, the sample has a Néel temperature $T_N = 52$ K, significantly reduced from $T_N = 311$ K for pure Cr and carefully chosen to

sit on the leading edge of the jump in the Hall coefficient [6] at $x_c \sim 0.034$. The pressure cell was mounted in the bore of a superconducting magnet in a ^3He cryostat to reach sub-Kelvin temperatures.

The variation of the longitudinal resistivity as a function of pressure and temperature in the immediate vicinity of the QCP is captured in Fig. 1. The minimum in $\rho(T)$ at T_{\min} marks the opening of the spin-density-wave gap [7]. It collapses as a function of pressure, with the low-temperature rise in $\rho(T)$ disappearing at $P_c = 7.5$ kbar. The temperature dependence of ρ for $T > T_{\min}$ when $P < P_c$ and for all $T < 100$ K for $P > P_c$ follows a T^3 power law expected from phonon scattering in a disordered metal alloy [8]. Although $T_{\min}(P)$ tracks P_c , the full T dependence of the electrical resistivity at $P \sim P_c$ appears oblivious to the existence of the QCP, even under the magnifying glass of pressure tuning. This smooth variation through P_c contrasts sharply with the “non-Fermi liquid” behavior seen in some superconducting cuprates [9], heavy fermion compounds [10], and metamagnets [5], where a reduced power law in T emerges at the QCP. We note that we find no evidence for a coexisting superconducting state near P_c [11] for $T > 0.4$ K.

The approach of the $T \rightarrow 0$ longitudinal resistivity to P_c from below is one key measure of the quantum critical behavior. The opening of the spin-density-wave gap leads to an excess, normalized resistivity, $\Delta\rho/\rho(T) = [\rho(T) - \rho_P]/\rho$, where the baseline paramagnetic resistivity ρ_P is determined from the extrapolation of the T^3 fit for $T > T_{\min}$ to $T = 0$. We focus in Fig. 2(a) on $\Delta\rho/\rho$ ($T \rightarrow 0$), which falls continuously to zero as a function of

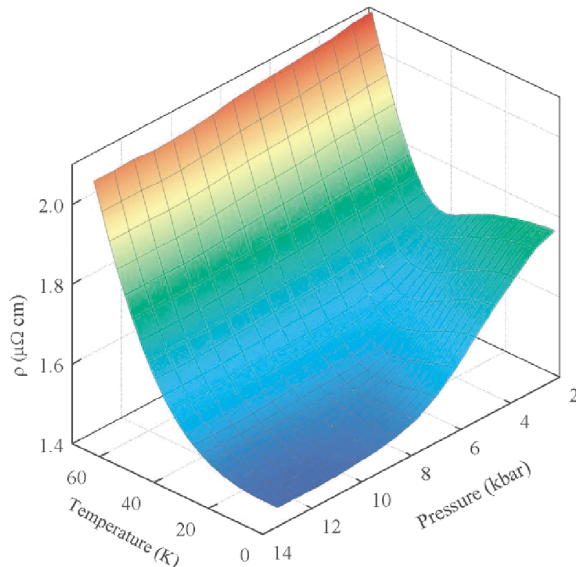


FIG. 1 (color). Variation of the longitudinal resistivity ρ with temperature and pressure (in descending order) in the immediate vicinity of the quantum critical point for single crystal $\text{Cr}_{0.968}\text{V}_{0.032}$. The minimum in the resistivity tracks the opening of the spin-density-wave gap, which is completely suppressed at the $T = 0$ critical pressure, $P_c = 7.5$ kbar.

pressure with a critical exponent less than 1. The rounding in the immediate vicinity of P_c can be attributed to small differences in V concentration across the sample. We fit the data to a critical form $\Delta\rho/\rho(T = 0.5\text{K}, P) \sim (P_c - P)^\beta$ convolved with a Gaussian distribution of critical pressures. The three-parameter fit [dotted line in Fig. 2(a)] yields a critical exponent $\beta = 0.68 \pm 0.03$ at the critical pressure $P_c = 7.5 \pm 0.1$ kbar. This P_c corresponds to a mean $x_0 = 3.189 \pm 0.001\%$, close to the nominal V concentration of 3.2% and set by the empirical formula $P_c = 31.042(3.430 - x)$, which is determined from a consideration of all available data reported in the literature

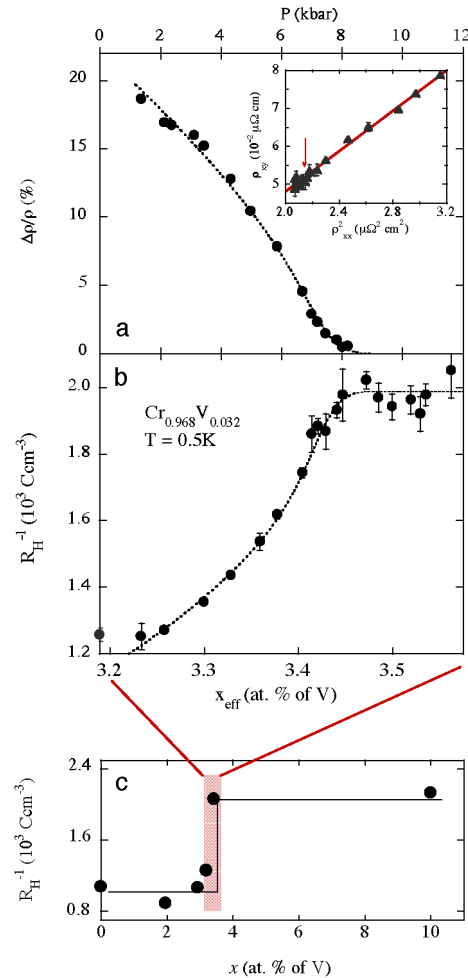


FIG. 2 (color). Critical behavior of the (a) normalized resistivity and (b) the inverse Hall coefficient in the $T \rightarrow 0$ limit as a function of pressure P . The resistivity and the inverse Hall coefficient approach the quantum phase transition with different critical exponents. The curves are fits to $(P - P_c)^{0.68 \pm 0.03}$ and $(P - P_c)^{0.50 \pm 0.02}$, respectively, convolved with a Gaussian spread in stoichiometry (see the text). (c) The exploded view at the bottom illustrates the high resolution afforded by pressure measurements as compared to results [6] from a series of crystals with different vanadium concentrations. The inset in (a) shows that $\rho_{xy} \sim \rho_{xx}^2$, implying that the transverse conductivity $\sigma_{xy}(T = 0)$ is actually constant through the QCP.

(discussed below). The observed tail in $\Delta\rho/\rho(P)$ gives a width $\delta x = (0.019 \pm 0.001)\%$.

Perhaps the most striking feature associated with the QCP is the 100% jump in the $T = 0$ carrier density over a very narrow range of x [Fig. 2(c)]. Fixing x and tuning the transition with P permits an investigation of whether this “jump” is sharp but continuous, and if so, whether there is an additional critical exponent that characterizes the transition. We plot in Fig. 2(b) the inverse Hall coefficient (R_H^{-1}) at $T = 0.5$ K in the immediate vicinity of P_c . $R_H^{-1}(T \rightarrow 0)$ assumes separate and fixed values in the antiferromagnet (< 2 kbar) and in the paramagnet ($P > 8$ kbar) but changes continuously between the two regimes. We fit the data by fixing δx from $\Delta\rho/\rho(P)$ and again convolving a critical form $R_H^{-1}(T = 0.5 \text{ K}, P) \sim (P_c - P)^\alpha$ with a Gaussian distribution of critical pressures in a two-parameter fit [dotted line in Fig. 2(a)]. We find a consistent $P_c = 7.5 \pm 0.1$ kbar, but a second critical exponent $\alpha = 0.50 \pm 0.02$. The finite width in $R_H^{-1}(P)$ cannot arise from the convolution of a step function (representing a discontinuous transition) with large inhomogeneities in x because (i) the fit would require an inconsistent, far larger δx and (ii) the value of P_c would be fixed at such a low pressure that it would be unphysical.

The connection of the $T = 0$ critical behavior to the finite temperature response is summarized in Fig. 3. We fix temperature, vary P , and find that $\Delta\rho/\rho(P)$ for all T up to 38 K, the highest temperature where we have sufficient data to draw meaningful conclusions, can be fit (dotted lines) with the same critical exponent ($\beta = 2/3$) and the identical constant of proportionality (0.056 ± 0.003). The mean field result, $\beta = 1/2$, is not

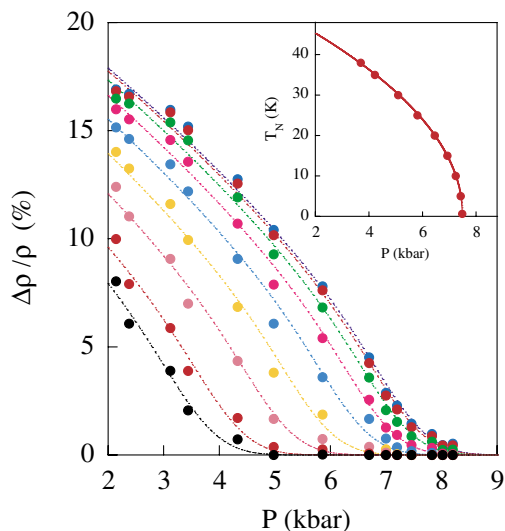


FIG. 3 (color). The normalized resistivity can be fit with a critical exponent $2/3$ at all temperatures from 0.5 to 38 K. Pressure curves shown are for 0.5, 5, 10, 15, 20, 25, 30, 35, and 38 K. In all cases, we fix the spread in stoichiometry to the $T = 0$ result, $\delta x = 0.019\%$. Inset: the Néel temperature approaches zero as $(P - P_c)^{0.49 \pm 0.02}$.

187201-3

recovered even for $P \ll P_c$, indicating the predominant effects of quantum fluctuations up to $T \sim T_N$. Strongly enhanced fluctuations have been identified up to 0.5 eV in inelastic neutron scattering studies of $\text{Cr}_{0.95}\text{V}_{0.05}$ [12] and may account as well for the unusual temperature dependence of the Hall resistivity for $T > T_N$ found in Cr-V alloys [6] and familiar from studies of the cuprate superconductors [13]. Finally, we can use the $P_c(T)$ from the fits in the main part of Fig. 3 to construct the T - P phase diagram for $\text{Cr}_{0.968}\text{V}_{0.032}$. The Néel temperature is suppressed with pressure as $T_N \sim (P_c - P)^\gamma$ with $\gamma = 0.49 \pm 0.02$ and $P_c = 7.5 \pm 0.1$ kbar (Fig. 3 inset). Isothermal rather than isobaric cuts of the data are essential to determine γ accurately given the almost vertical approach of the phase boundary to the pressure axis as $T_N \rightarrow 0$.

A close proximity to the QCP is an additional requirement for an accurate determination of γ . We collect in Fig. 4 the variation of T_N with x for a broad range of Cr-V alloys measured via electrical transport [6,14,15], neutron diffraction [16], nuclear magnetic resonance [17], and thermal expansion [18], as well as the suppression of spin-density-wave order with pressure for pure Cr [19], $\text{Cr}_{0.988}\text{V}_{0.012}$ [20], $\text{Cr}_{0.972}\text{V}_{0.028}$ [20], and $\text{Cr}_{0.968}\text{V}_{0.032}$ (our data). All the data can be collapsed onto a universal curve using an effective V concentration x_{eff} that assumes the simplest linear conversion between chemical doping and applied pressure: $P_c = 31.042(3.430 - x)$. By the congruence of the data for pure Cr and its alloys, it appears that disorder is not a dominant factor. T_N decreases linearly with x_{eff} across almost the entire composition range, only assuming its critical form with exponent $\gamma = 1/2$ very close to $x_c \sim 3.430\%$. The initial linear onset

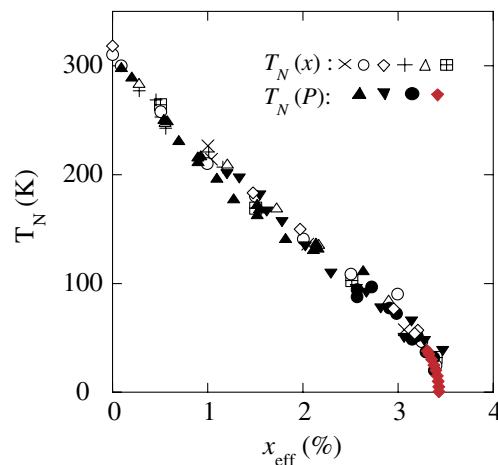


FIG. 4 (color). Collapse onto a universal curve of all available data on the pressure and doping dependence of the Néel temperature in $\text{Cr}_{1-x}\text{V}_x$, assuming a linear conversion between P and x . Data very close to the QCP are required to reveal the true critical behavior: T_N from various x 's: \square [14], \circ [15], \diamond [6], $+$ [16], \triangle [17], \boxplus [18]; from P measurements: pure Cr \square [19], V 1.2% \square [20], V 2.8% \square [20], V 3.2% \square (our data).

187201-3

mimics the suppression of T_N with x and P in the heavy fermion antiferromagnet $\text{CeCu}_{6-x}\text{Au}_x$ [21], where measurements approached $\delta x_{\text{eff}}/x_{\text{eff}} \sim 5 \times 10^{-3}$. For the $\text{Cr}_{1-x}\text{V}_x$ antiferromagnet, the curvature close to the QCP becomes apparent only because of our pressure experiment's resolving power at the critical point, $\delta x_{\text{eff}}/x_{\text{eff}} \sim 1 \times 10^{-4}$ [22].

The two-band itinerant model for antiferromagnetism [20,23] has been used to consider the pressure dependent Néel temperature of pure Cr and its alloys and predicts $\gamma = 1/2$. A scaling model [24] for a $z = 2$ antiferromagnetic QCP predicts $\gamma = z/(d - 2 + z) = 2/3$ in dimension $d = 3$ but does not apply strictly to a magnet with a partially nested Fermi surface. Recent calculations [25,26] consider explicitly the disappearance of itinerant spin density wave order at a QCP and predict confluent critical exponents for R_H^{-1} and T_N . Although we do find $\alpha = \gamma = 1/2$, there is no theoretical basis for an additional exponent to characterize the critical behavior of $\Delta\rho/\rho$, the experimentally observed $\beta = 2/3$. If the nesting is imperfect, then different regions of the Fermi surface, "hot spots" with low curvature and "cold spots" with high curvature, could dominate different aspects of the critical behavior, akin to theories for high- T_c and heavy fermion superconductors [27].

At $T = 0$, the ordinary Hall number is an intrinsic, nonzero quantity, but the longitudinal resistivity—which is characterized by the anomalous power law of $2/3$ —is nonzero only by virtue of scattering from impurities and other defects. Could the resistivity be tracking the transverse conductivity $\sigma_{xy} = \rho_{xy}/\rho_{xx}^2$? We find that $\rho_{xy} \sim \rho_{xx}^2$ (Fig. 2 inset), implying that $\sigma_{xy}(T = 0)$ is a constant, insensitive to P even as it crosses P_c . This result depends crucially on tuning the transition with P because varying the composition x alters the disorder and the scattering potential for each sample, precluding any simple relationship between ρ_{xy} and ρ_{xx} . A theory of our ρ_{xx} data near P_c in $\text{Cr}_{0.968}\text{V}_{0.032}$ can seek to explain either the apparently anomalous power law describing $\Delta\rho/\rho$ or the seeming pressure independence of σ_{xy} .

The fact that the asymptotic critical behavior manifests itself only very close to the quantum phase transition emphasizes the need for similar high precision work on other systems, as well as underlining the possibility that much of the behavior of $\text{Cr}_{1-x}\text{V}_x$ and Cr itself may be controlled by a more radical type of quantum criticality, namely, one where the Fermi surface undergoes sudden collapse [28]. The simplicity of the Cr-V system provides the means to test directly our fundamental notions of physics at a QCP, including the use of the Hall coefficient as a sensitive diagnostic of the underlying order [6], and the manifestation of these ideas in real-world materials.

We are grateful to P. Coleman, A.J. Millis, and M. Norman for illuminating discussions. The work at the University of Chicago was supported by the

National Science Foundation under Grant No. DMR-0114798.

-
- [1] S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999).
 - [2] S. L. Sondhi, S. M. Girvin, J. P. Carini, and D. Shahar, *Rev. Mod. Phys.* **69**, 315 (1997).
 - [3] S. Chakravarty, B. I. Halperin, and D. R. Nelson, *Phys. Rev. B* **39**, 2344 (1989).
 - [4] D. Bitko, T. F. Rosenbaum, and G. Aeppli, *Phys. Rev. Lett.* **77**, 940 (1996).
 - [5] S. A. Grigera *et al.*, *Science* **294**, 329 (2001); G. Aeppli and Y.-A. Soh, *Science* **294**, 315 (2001).
 - [6] A. Yeh *et al.*, *Nature (London)* **419**, 459 (2002).
 - [7] S. Alexander, J. S. Helman, and I. Balberg, *Phys. Rev. B* **13**, 304 (1976); S. A. Carter *et al.*, *Phys. Rev. Lett.* **67**, 3440 (1991).
 - [8] I. A. Campbell, A. D. Caplin, and C. Rizzuto, *Phys. Rev. Lett.* **26**, 239 (1971); D. L. Mills, *Phys. Rev. Lett.* **26**, 242 (1971).
 - [9] P. Coleman, in *Electron*, edited by M. Springford (Cambridge University Press, Cambridge, 1997), Chap. 7.
 - [10] G. R. Stewart, *Rev. Mod. Phys.* **73**, 797 (2001).
 - [11] E. Fawcett, *Rev. Mod. Phys.* **66**, 25 (1994).
 - [12] S. M. Hayden *et al.*, *Phys. Rev. Lett.* **84**, 999 (2000).
 - [13] T. R. Chien, Z. Z. Wang, and N. P. Ong, *Phys. Rev. Lett.* **67**, 2088 (1991).
 - [14] S. Arajis, *Can. J. Phys.* **47**, 1005 (1969).
 - [15] A. L. Trego and A. R. Mackintosh, *Phys. Rev.* **166**, 495 (1968).
 - [16] S. Komura, Y. Hamaguchi, and N. Kunitomi, *J. Phys. Soc. Jpn.* **23**, 171 (1967); *Phys. Lett.* **24A**, 299 (1967).
 - [17] R. G. Barnes and T. P. Graham, *J. Appl. Phys.* **36**, 938 (1965).
 - [18] G. K. White, R. B. Roberts, and E. Fawcett, *J. Phys. F* **16**, 449 (1986).
 - [19] D. B. McWhan and T. M. Rice, *Phys. Rev. Lett.* **19**, 846 (1967).
 - [20] T. M. Rice, A. S. Barker, Jr., B. I. Halperin, and D. B. McWhan, *J. Appl. Phys.* **40**, 1337 (1969).
 - [21] B. Bogenberger and H. v. Löhneysen, *Phys. Rev. Lett.* **74**, 1016 (1995).
 - [22] The increased sensitivity derives from both a factor of three smaller step size in P at the QCP as well as a different constitutive relationship between x and P for $\text{CeCu}_{6-x}\text{Au}_x$ and $\text{Cr}_{1-x}\text{V}_x$.
 - [23] P. A. Fedders and P. C. Martin, *Phys. Rev.* **143**, 245 (1966).
 - [24] J. A. Hertz, *Phys. Rev. B* **14**, 1165 (1976); A. J. Millis, *Phys. Rev. B* **48**, 7183 (1993).
 - [25] S. Chakravarty, C. Nayak, S. Tewari, and X. Yang, *Phys. Rev. Lett.* **89**, 277003 (2002).
 - [26] M. R. Norman, Q. Si, Ya. B. Bazaliy, and R. Ramazashvili, *Phys. Rev. Lett.* **90**, 116601 (2003).
 - [27] R. Hlubina and T. M. Rice, *Phys. Rev. B* **51**, 9253 (1995).
 - [28] A. Schroeder *et al.*, *Nature (London)* **407**, 351 (2000); P. Coleman, C. Pepin, Q. Si, and R. Ramazashvili, *J. Phys. Condens. Matter* **13**, R723 (2001).