High Resolution Study of Magnetic Ordering at Absolute Zero

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Abstract

High fidelity 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 exactitude 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.

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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. 

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 analogues. Typically, fluctuations are varied by scanning the composition of an alloy, such as the doped cuprates that host high-temperature superconductivity. The result is 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 and metals. 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. In response to this challenge, 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 suppress 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 become localized due to magnetic order: should they be counted as free or as localized? In a previous experiment, we have examined alloys of Cr$_{1-x}$V$_x$ and tracked the loss of carriers as a function of $x$. We found a jump in the Hall number at the QCP, indicating giant fluctuations in the number of free carriers, even though other measures, such as the Néel temperature and the internal magnetization vanished continuously. The present work demonstrates that if we resolve the quantum critical point with two 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 Cr$_{0.968}$V$_{0.032}$ of 1 mm$^3$ 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 $T_N = 52K$, significantly reduced from $T_N = 311K$ for pure Cr and carefully chosen to sit on the leading edge of the jump in the Hall coefficient 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 single crystals of Cr$_{1-x}$V$_x$ were grown by Ames Laboratory in an arc zone refining furnace and annealed for 72 hours at 1600°C to relieve internal strain. Laue backscattering was used to determine the orientation of the bcc lattice, and several pieces aligned with the (100) crystallographic direction were cut from the center of each boule using spark erosion. Proper etching to remove the surface oxide (with a 3:1:HCl:H2O2 solution quenched with THF) was found to be essential for reproducible results. The long axis of the crystal was mounted in the pressure cell perpendicular to the applied field with Fluorinert as the pressure medium and a chip of (V$_{0.99}$Ti$_{0.01}$)$_2$O$_3$ serving as the manometer. We could measure relative values of P to better than 0.1 kbar. The diagonal and off-diagonal components of the resistivity were measured using a conventional five-probe ac-bridge technique in the Ohmic and frequency-independent limits. The Hall coefficient was determined from the extrapolation of the $\frac{\Delta \rho}{\rho}$ limit of Eq. (1). ∆$\rho/\rho$ falls continuously to zero as a function of pressure with a critical exponent less than one. 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 $\frac{\Delta \rho}{\rho}(T=0.5 K, P) \sim (P_c - P)^3$, convolved with a Gaussian distribution of critical pressures. The three-parameter fit (solid line in Fig. 2a) 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 (discussed below). The observed tail in $\frac{\Delta \rho}{\rho}(P)$ 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 zero temperature carrier density over a very narrow range of $x$ (Fig. 2a). 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. 2b: 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 ($P < 2$ kbar) and in the paramagnet ($P > 8$ kbar), but changes continuously between the two regimes. We fit the data by fixing $\delta x$ from $\frac{\Delta \rho}{\rho}(P)$ and again convolving a critical form $R_H^{-1}(T=0.5 K, P) \sim (P_c - P)^3$ with a Gaussian distribution of critical pressures in a two-parameter fit (solid line in Fig. 2b). 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 $\delta x$ 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 ($0.056 \pm 0.003$). The mean field result, $\beta = 1/2$, is not 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 $\text{Cr}_0.968\text{V}_{0.032}$ and may account as well for the unusual temperature dependence of the Hall resistivity for $T > T_N$ found in Cr-V alloys and familiar from studies of the cuprate superconductors. Finally, we can use the $P_c(T)$ from the fits in the main part of Fig. 4 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. 6 inset). Isothermal rather than isobaric cuts of the data are essential to determine $\gamma$ accurately given the almost vertical approach of the phase boundary to the pressure axis as $T_N \to 0$.

Close proximity to the QCP is an additional requirement for an accurate determination of $\gamma$. We collect in Fig. 4 the variation of $T_N$ with $x$ for a broad range of Cr-V alloys measured via electrical transport, neutron diffraction, nuclear magnetic resonance, and thermal expansion, as well as the suppression of spin-density-wave order with pressure for pure Cr, $\text{Cr}_{0.988}\text{V}_{0.012}$, $\text{Cr}_{0.972}\text{V}_{0.028}$, 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 $x_{\text{eff}}$ that assumes the simplest linear conversion between chemical doping and applied pressure: $P_c = 31.042 \cdot (3.430 - x)$. 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 $x_{\text{eff}}$ 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 mimics the suppression of $T_N$ with $x$ and $P$ in the heavy fermion antiferromagnet $\text{CeCu}_6-x\text{Au}_x$, where measurements approached $\delta x_{\text{eff}}/x_{\text{eff}} \sim 5 \times 10^{-3}$.
For the Cr$_{1-x}$V$_x$ antiferromagnet, the curvature close to the QCP becomes apparent only because of our pressure experiments resolving power at the critical point, $\delta x_{\text{eff}}/x_{\text{eff}} \sim 1 \times 10^{-4}$ [22]. Although Cr$_{0.968}$V$_{0.032}$ appears to behave critically over almost its entire range of $x_{\text{eff}}$, $T_N(P = 0) = 52$ K is far reduced from that of pure Cr (311K), which in turn is far below the bare magnetic transition $T$ of Cr($\sim 0.5$ eV).

The two-band itinerant model for antiferromagnetism [20, 23] has been used to consider the pressure dependent fundamental quantity, such as the transverse conductivity $\sigma_{xy}$, in the same manner that the scaling model [19] for a non-zero only by virtue of scattering from impurities and other defects. The two-band itinerant model for antiferromagnetism [20, 23] has been used to consider the pressure dependent fundamental quantity, such as the transverse conductivity $\sigma_{xy}$, in the same manner that the scaling model [19] for a non-zero only by virtue of scattering from impurities and other defects.

The two-band itinerant model predicts $\Delta_T = 1/2$, implying that $\Delta_T$ is a constant, insensitive to P even as it crosses $P_c$. This result depends crucially on the tuning complex order parameter $x$, which characterizes the underlying order [6], and the manifesting of the underlying order $x$ alters the disorder and the scattering potential for each sample, precluding any simple relationship between $\Delta_T$ and $\rho_{xx}$. A theory of $\rho_{xx}$ data near $P_c$ in Cr$_{0.968}$V$_{0.032}$ can seek to explain either the apparently anomalous power law describing $\Delta_T/\rho$ or the seeming pressure independence of $\sigma_{xy}$. 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 Cr$_{1-x}$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.

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