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We report a chemical substitution-induced ferromagnetic quantum critical point in polycrystalline Ni$_{1-x}$Rh$_x$ alloys. Through magnetization and muon spin relaxation measurements, we show that the ferromagnetic ordering temperature is suppressed continuously to zero at $x_{\text{crit}} = 0.375$ while the magnetic volume fraction remains 100% up to $x_{\text{crit}}$, pointing to a second order transition. Non-Fermi liquid behavior is observed close to $x_{\text{crit}}$, where the electronic specific heat $C_d/T$ diverges logarithmically, while immediately above $x_{\text{crit}}$ the volume thermal expansion coefficient $\alpha_V/T$ and the Grüneisen ratio $\Gamma = \alpha_V/C_d$ both diverge logarithmically in the low temperature limit, further indication of a ferromagnetic quantum critical point in Ni$_{1-x}$Rh$_x$.

A quantum critical point (QCP) occurs when a phase transition is continuously suppressed to zero temperature. The intense quantum fluctuations in the vicinity of a QCP profoundly alter a material’s electronic properties, resulting in non-Fermi liquid behavior and, in some cases, unconventional superconductivity [1, 2]. The most ubiquitous QCP separates an antiferromagnetically ordered state from one in which quantum fluctuations disrupt the order. Notable examples are found among heavy fermion systems [1, 3, 4]. QCPs in ferromagnetic (FM) metals have proven far more elusive [5]. It is now understood that a FM QCP is inherently unstable and can survive only in rare circumstances [6]. In this work, we report the discovery of a FM QCP in Ni$_{1-x}$Rh$_x$, as evidenced by (i) a second-order phase transition up to the critical concentration $x_{\text{crit}}$, and (ii) divergence of the electronic specific heat coefficient $C_d/T$, the volume thermal expansion $\alpha_V/T$, and the Grüneisen ratio $\Gamma = \alpha_V/C_d$. The dilution of the $d$-electron magnetic sublattice as the tuning parameter to induce a FM QCP opens a new route for exploring FM quantun criticality and possible new collective phases near the QCP, such as unconventional superconductivity [7].

FM QCPs are revealed via chemical substitution in Zr$_{1-x}$Nb$_x$Zn$_2$ [8], SrCo$_2$(Ge$_1-x$P$_x$)$_2$ [9], YbNi$_4$($\text{P}_{1-x}$As$_x$)$_2$ [10], and (Sc$_{1-x}$Lu$_x$)$_{3,1}$In [11]. The disorder effect is minimal or negligible in these systems. For SrCo$_2$(Ge$_1-x$P$_x$)$_2$, the QCP is induced by the breaking of dimers [9]. However, the exact mechanism responsible for the FM QCP in the other three systems remains unclear. In most other FM metals, the QCP is preempted when the continuous (second-order) transition as a function of non-thermal control parameter either becomes discontinuous (first-order), or the ferromagnetism is replaced by a spatially-modulated ordered state [5, 12–15]. Theoretical work by Belitz, Kirtpatrick, and Vojta (BKV) has proposed a route towards a FM QCP by long-range effective spin interactions that occur in the presence of quenched disorder [6, 16, 17]. A handful of FM QCPs have been identified as candidates for this phenomenology, including UCo$_{1-x}$Fe$_x$Ge [18], (Mn$_{1-x}$Fe$_x$)Si [19], NiCoCr$_2$ [20], and Ce($\text{Pd}_{1-x}\text{Ni}_x$)$_2$P$_2$ [21], where disorder is inherently introduced by the chemical substitution. In most of these systems, the proposed existence of a QCP is based on either divergence of some thermodynamic parameters [18, 20, 21] or the second order nature of the transition [19]. However, the unambiguous identification of a QCP requires that both these criteria be fulfilled. This point is exemplified by disordered Sr$_{1-x}$Ca$_x$RuO$_3$, for which a QCP can be ruled out because the transition at $T = 0$ is first order [22], and yet, quantum critical scaling is still observed [23]. Thus, in order to unambiguously identify a FM QCP it is essential that both thermodynamic signatures of quantum fluctuations and second-order behavior be observed simultaneously. Our observation of both these requisite signatures in a chemically simple material where the FM QCP is induced via direct dilution of its $d$-electrons elevate Ni$_{1-x}$Rh$_x$ to a top tier of candidates.

Elemental Ni, which has a simple face-centered cubic structure, is known to order ferromagnetically below its Curie temperature $T_C = 627$ K [24]. Upon alloying with Rh, the $T_C$ of Ni$_{1-x}$Rh$_x$ is quickly suppressed [25].
Ni\textsubscript{1−x}Rh\textsubscript{x} has more configuration entropy than pure Ni [26]. Also, the metallic radii of Ni (124 pm) and Rh (134 pm) differ by ~8%. Naturally, one would expect that, compared to pure Ni, there is more disorder in Ni\textsubscript{1−x}Rh\textsubscript{x} alloy, making it a good candidate to test for the existence of a disorder-driven FM QCP. Polycrystalline Ni\textsubscript{1−x}Rh\textsubscript{x} samples with 0.3 ≤ x ≤ 0.42 were prepared by arc-melting the constituents Ni and Rh and annealed at 1000°C. Magnetization measurements were carried out using a Quantum Design (QD) magnetic property measurement system. Zero-field muon spin relaxation measurements were performed at the M20 surface muon channel at TRIUMF. Specific heat was measured using a QD Dynacool physical property measurement system equipped with a dilution refrigerator. Thermal expansion was measured with a homemade capacitance dilatometer. More details about the sample characterizations and experimental methods are provided in the Supplemental Material [27–34].

Figure 1(a) shows the $\mu_0 H = 0.01$ T magnetic susceptibility $\Delta M(T)/H$ of Ni\textsubscript{1−x}Rh\textsubscript{x}, after a temperature-independent contribution $M_0$ was subtracted from the measured $M(T)$ ($\Delta M = M - M_0$). $\Delta M/H$ sharply increases as $T$ is lowered through $T_C$ for $x = 0.32 – 0.36$ where $T_C$ is determined both through a linear fit, as shown in Fig. 1(a), and the Arrott-Noakes analysis as discussed below. For $x_{\text{crit}} = 0.375$ (where $T_C \to 0$), $\Delta M/H$ shows only a small increase down to the lowest measured temperature of 2 K, consistent with the complete suppression of FM order. Isothermal magnetization measurements at $T = 2$ K confirm that Ni\textsubscript{1−x}Rh\textsubscript{x} is a soft ferromagnet without a measurable hysteresis (Fig 1(b)). We cannot rule out a very small antiferromagnetic component or canting close to $x_{\text{crit}}$, although magnetization suggests that FM correlations dominate, as evidenced by an abrupt increase of $M(H)$ at the lowest field (Fig. 1(b)) and adherence to Arrott-Noakes scaling all the way up to $x_{\text{crit}}$. Future neutron scattering and nuclear magnetic resonance measurements will shed light on this issue. For the $x = 0.32$ sample, which orders near 100 K, the inverse magnetic susceptibility $H/\Delta M$ exhibits Curie-Weiss-like behavior between 150 and 300 K, from which we derive a paramagnetic (PM) effective moment $\mu_{\text{PM}} = 1.97\mu_B$/f.u. (see SM). For the same sample, $\Delta M$ is small at 7 T ($\sim 0.22\mu_B$/f.u.), and the Rhodes-Wohlfarth ratio, $\mu_{\text{PM}}/\mu_{\text{sat}} = 9$, much larger than unity, is indicative of itinerant moment behavior in Ni\textsubscript{1−x}Rh\textsubscript{x} [35]. An earlier study indicated spin glass behavior in Ni\textsubscript{1−x}Rh\textsubscript{x} [36]. However, our AC magnetic susceptibility measurements, presented in the SM, show no evidence for spin glass behavior near $T_C$. Such a discrepancy may be due to different purity of starting materials or sample homogeneity.

For ferromagnets, the equation of state at $T_C$ is given by $\Delta M \sim H^{1/\delta}$ [31]. From linear fits of $\log(\Delta M)$ vs. $\log(\mu_0 H)$, as shown by the dashed line in Fig. 1(c), we determine that $T_C = 96$ K and $\delta \sim 3.5$ for the $x = 0.32$ sample. We applied the same analysis for all samples with $x = 0.30 – 0.37$. The critical exponents $\beta$ and $\gamma$ were determined by applying Arrott-Noakes scaling to the isotherms measured in the vicinity of $T_C$ (see SM for details) [31]. The composition dependence of all three exponents, $\delta$, $\beta$, and $\gamma$, is summarized in Fig. 1(d). The Widom relation $\gamma/\beta = \delta - 1$ is obeyed over the entire range of Rh concentrations investigated here, a self-consistent check of the scaling analysis. At $x = 0.30$, which is well below $x_{\text{crit}}$, the exponents $\beta = 0.5$, $\gamma = 1.3$, and $\delta = 3.1$ are close to the expected mean-field values. With increasing $x$, the exponents deviate from the mean-field values and approach $\beta = 0.6$, $\gamma = 0.7$, and $\delta = 2.3$ at $x = 0.37$, just below $x_{\text{crit}}$. A similar evolution of the critical exponents with chemical substitution was observed in Sr\textsubscript{1−x}Ca\textsubscript{x}RuO\textsubscript{3}, where it was proposed that disorder resulted in enhanced quantum fluctuations near $x_{\text{crit}}$ [37].

Zero field $\mu$SR measurements were performed on six samples of Ni\textsubscript{1−x}Rh\textsubscript{x} with $x = 0.30 – 0.39$, in order to determine whether the magnetic order takes place via a first- or second-order process. Hallmarks of a first-order transition are phase separation or an abrupt change of ground state [22, 38]. Conversely, in the case of a second-order transition, the size of the ordered moment is expected to continuously decrease without phase separation. $\mu$SR allows an independent measure of both the
local order parameter and the magnetic volume fraction, \( f_{\text{mag}} \), and can thus unambiguously distinguish between these scenarios. Representative muon decay asymmetry spectra, \( P(t) \), are plotted in Fig. 2(a) for \( x = 0.32 \) at various temperatures below and above \( T_C \). Above \( T_C \), \( P(t) \) is essentially non-relaxing, as expected in a PM state. The onset of magnetic order is signaled by a fraction of the asymmetry undergoing rapid relaxation at early times. The compositional dependence of \( P(t) \) at \( T = 2 \) K is presented in Fig. 2(b). This comparison reveals that the samples with the highest Rh concentrations, \( x = 0.375 \) and 0.39 (\( \geq x_{\text{crit}} \), blue and purple symbols), exhibit only weak relaxation down to the lowest measured temperatures, thus confirming the absence of magnetic order for these compositions. The samples with \( x < x_{\text{crit}} \) exhibit sharp relaxation associated with magnetic order. The \( P(t) \) data for all compositions and temperatures is well-described by the dynamic Kubo-Toyabe function [7]:

\[
P(t) = (1 - f_{\text{mag}}) \cdot e^{-\lambda t} + f_{\text{mag}} \cdot G_{\text{DKT}}(t, \sigma, \nu)
\]

where \( \lambda \) and \( \sigma \) are the relaxation rates for the non-magnetic and magnetic fractions of the sample, respectively, and \( \nu \) is the hopping rate. The temperature dependence of \( f_{\text{mag}} \) is presented in Fig. 2(c), revealing no evidence for phase separation: \( f_{\text{mag}} \) remains 100% up to Rh concentrations of \( x = 0.36 \) and drops to 0% at \( x_{\text{crit}} = 0.375 \). With increasing Rh concentration, the Kubo-Toyabe minimum moves to increasing times as can be seen in Fig. 2(b), consistent with a decreasing ordered moment. This suggests that the suppression of magnetic order in \( \text{Ni}_{1-x}\text{Rh}_x \) occurs via a continuous second-order process.

Next we show evidence for divergent thermodynamic parameters in \( \text{Ni}_{1-x}\text{Rh}_x \). Figure 3(a) shows the electronic specific heat \( C_{el}/T \) around \( x_{\text{crit}} = 0.375 \), where the phonon contribution has been subtracted from the measured specific heat. For concentrations that are both far above and far below \( x_{\text{crit}} \) (\( x \leq 0.15 \) and \( x \geq 0.6 \), \( C_{el}/T \) is nearly temperature-independent at low temperatures, as expected for a Fermi liquid (FL) [27]. Close to \( x_{\text{crit}} \), \( C_{el}/T \) diverges logarithmically on cooling. The fastest divergence occurs at \( x_{\text{crit}} = 0.375 \), where \( C_{el}/T = a_0 \log(T_0/T) \) between 0.1 and 3 K (solid line in Fig. 3(a)), such that \( a_0 \) is maximum at the QCP (red diamonds in Fig. 4). This logarithmic divergence was previously reported in \( \text{Ni}_{0.62}\text{Rh}_{0.38} \) [39] and has also been observed in other QCP systems [9–11, 40]. For \( x > x_{\text{crit}} \), \( C_{el}/T \) levels off at the lowest temperatures, consistent with non-Fermi-liquid (NFL) to FL crossover. This is similar to other FM and antiferromagnetic quantum critical systems [1, 3–5].

QCPs are characterized by an accumulation of magnetic entropy \( S_{mag} \) as a function of the control parameter at low, but finite temperatures. In \( \text{Ni}_{1-x}\text{Rh}_x \), this is underscored by the dependence of the specific heat pa-
rameter $a_0$ on $x$ (red diamonds in Fig. 4), given that $S_{mag}$ is commensurate to $a_0$, which, in turn, is maximum at the QCP. At the same time, $S_{mag}$ is related to the volume thermal expansion $\alpha_V$ through the Maxwell relation $\alpha_V = -V^{-1} \partial S_{mag}/\partial p$ (where $p$ is pressure), and the divergence of $\alpha_V/T$ has been taken as proof of the QCP in heavy fermion systems, such as CeCu$_{6-x}$Au$_x$ [41], CeNi$_2$Ge$_2$, and YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$ [42]. Our data shows that at $x = 0.39$ (just above the QCP), zero-field $\alpha_V/T$ diverges logarithmically between 10 and 0.1 K (diamonds in Fig. 3(b)). This is indicative of NFL behavior in proximity to the QCP [43]. The data show no hysteresis between heating (open) and cooling (full) measurements, ruling out any history-dependent spin glass effects. The length measurements on Ni$_{1-x}$Rh$_x$ with $x = 0.39$ reached the resolution limit of the dilatometer of $\Delta L \geq 10^{-3}$ Å at the lowest measured temperatures, resulting in an enhanced scattering below $\sim 0.2$ K. The application of a magnetic field of 4 T reduces $\alpha_V/T$ to a nearly constant value below 4 K, indicating a recovery of the FL behavior (squares in Fig. 3(b)). This recovery of FL behavior is consistent with what has been observed in field-dependent specific heat measurements for Ni$_{0.62}$Rh$_{0.38}$ [39].

An additional probe for a QCP is the Grüneisen ratio $\Gamma = \alpha_V/C_{el} \sim 1/E^* \cdot \partial E^*/\partial p$. $\Gamma$ reveals the hydrostatic pressure dependence of the dominating, characteristic energy scale $E^*$ (e.g., the energy related to the conduction band splitting at the Fermi energy, which is proportional to the spontaneous magnetization [44]). At a QCP, $E^*$ vanishes, and $\Gamma$ is expected to diverge with decreasing $T$ [43]. In the low temperature range for the $\alpha_V$ measurements, the phonon contribution is negligible. The calculated $\Gamma$ is depicted in the inset of Fig. 3(b), showing logarithmic divergence over two decades in temperature from $T = 10$ K to 0.1 K. The fact that $\Gamma \sim -\log T$ suggests either that the quantum critical behavior in Ni$_{1-x}$Rh$_x$ extends to a finite pressure interval (rather than a point) [43], or that the system lies within a disordered quantum Griffiths phase [45].

We summarize the $T_C - x$ phase diagram of Ni$_{1-x}$Rh$_x$ in Fig. 4. Magnetization $M(T, H)$ and $\mu$SR measurements reveal the suppression of $T_C$ with increasing Rh concentration up to $x_{crit} = 0.375$ (black symbols). The magnetically-ordered volume fraction remains 100% up to $x_{crit}$, while the magnitude of the ordered moment per formula unit continuously decreases, as expected for a second order transition [19]. In addition, the FM QCP is also revealed by the divergence of $C_{el}/T$, $\alpha_V/T$, and $\Gamma$ in the low temperature limit, associated with NFL behavior that extends up to $\sim 10$ K.

Finally, we compare our results with other Ni$_{1-y}M_y$ ($M = Al, Si, V, Cr, Mn, Cu, Zn, Pd, and Sb$) alloys. Non-magnetic $M$ metals dilute the Ni magnetic moment and therefore suppress the FM order. Magnetic susceptibility measurements on these alloys are sensitive to sample preparation [36, 50]. In the absence of a spin glass state or short range order, the enhancement of $C_{el}/T$ has been observed for all $M$ where $T_C \rightarrow 0$ [50–52]. This commonality can be understood in terms of enhanced spin fluctuations and does not necessarily indicate quantum critical fluctuations. A noteworthy member of this family is Ni$_{1-y}V_y$ where V substitution results in quantum Griffiths effect that competes with critical behavior without reaching a QCP [53, 54]. By contrast, Ni$_{1-x}$Rh$_x$ is the first member of the Ni$_{1-y}M_y$ family where divergent $\alpha_V/T$ and $C_{el}/T$ result in divergent $\Gamma$ [43], demonstrating the presence of a FM QCP. In fact, for most ferromagnets, when a dilution occurs in the magnetic sublattice, short-range order or spin glass behavior is observed [5]. The only exception is the $5f$-electron system Th$_{1-x}$U$_x$Cu$_2$Si$_2$ that the FM transition remains continuous at the critical concentration, where NFL behavior is observed [55].

One plausible scenario to account for the FM QCP in Ni$_{1-x}$Rh$_x$ is the aforementioned BKV theory [6, 16, 17]. The current study utilized polycrystalline samples and the residual resistivity ratio (not shown), which is often taken as a gauge of the amount of disorder, is small and comparable among the whole series of Ni$_{1-x}$Rh$_x$. To test if the FM quantum criticality in Ni$_{1-x}$Rh$_x$ fulfills the universality class in the strong disorder regime of the BKV theory, the growth of single crystals is imperative and is the subject of an ongoing study. Ni$_{1-x}$Rh$_x$ shows the first occurrence of a FM QCP with dilution of the $d$-electron magnetic sublattice. This is in contrast with chemical substitution on the non-magnetic sublattice in other FM QCP systems [9, 10, 18–21]. In particular, due
to its chemical simplicity, Ni$_{1-x}$Rh$_x$ is an ideal platform for future studies and our work establishes a new approach to explore FM quantum criticality.

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