Multicomponent fluctuation spectrum at the quantum critical point in CeCu$_6$–$_x$Ag$_x$

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Quantum critical points (QCPs) are widely accepted as a source of a diverse set of collective quantum phases of matter. The basic nature of a QCP is manifested in the critical fluctuation spectrum which in turn is determined by the adjacent phases and associated order parameters. Here we show that the critical fluctuation spectrum of CeCu$_{5.8}$Ag$_{0.2}$ cannot be explained by fluctuations associated with a single wave vector. Interestingly, when the critical fluctuations at wave vectors corresponding to the incommensurate antiferromagnetic order adjacent to the QCP are separated they are found to be three dimensional and to obey the scaling behavior expected for long wavelength fluctuations near an itinerant antiferromagnetic QCP. Without this separation, $E/T$ scaling with a fractional exponent is observed. Together these results demonstrate that a multicomponent fluctuation spectrum is a previously unexplored route to obtaining $E/T$ scaling at a QCP.

INTRODUCTION

The QCPs found in heavy fermion materials serve as prototypes of quantum criticality.$^{1–3}$ However, several fundamental characteristics remain unexplained preventing a general understanding of quantum critical phenomena. For example, a growing number of heavy fermion materials exhibit logarithmically diverging masses of electronic quasiparticles.$^{1,4–8}$ In the critical concentrations of UC$_{u_x}$Pd$_x$ and CeCu$_{5.8}$Au$_{x}$ energy over temperature ($E/T$) scaling of the critical dynamics is observed with a fractional exponent.$^{9,10}$ In CeCu$_{6}$Au$_{x}$, the fractional exponent also explains the temperature dependence of magnetic susceptibility.$^{10,11}$ indicating that the QCP is distinct from the conventional framework proposed by Hertz, Millis, and Moriya (HMM).$^{12–14}$ Recently, the debate as to the understanding of these phenomena has intensified.$^{15–22}$

This renewed debate is particularly significant since the hallmark of the unconventional quantum criticality, the unusual $E/T$ scaling, is explained by disparate mechanisms such as the breakdown of a local energy scale,$^{15–17}$ coupling between quasiparticles and order parameter fluctuations,$^{18,19}$ or by topological excitations.$^{20,21}$ Beyond these cases, $E/T$-scaling provides a stringent test of theories across a spectrum of quantum materials including highly frustrated magnets, spin liquids, high $T_c$ superconductors, non-Fermi liquid behavior, and quantum phase transitions.$^{22–26}$

As $E/T$ scaling of the dynamic susceptibility is almost universally used to validate theories of quantum criticality, additional more stringent experimental tests are essential. Here, we approach the aforementioned questions by studying the spin dynamics of the critical composition of CeCu$_{5.8}$Ag$_{0.2}$, a material closely related to the well known quantum critical system CeCu$_{6}$Au$_{x}$. Similar to CeCu$_{5.8}$Au$_{0.2}$, CeCu$_{6}$Ag$_{x}$ is a member of the CeCu$_{5.8}$Ag$_{0.2}$ family derived from the heavy fermion compound CeCu$_6$.$^{27,28}$

The basic properties of CeCu$_{5.8}$Au$_{0.2}$ and CeCu$_{6}$Ag$_{x}$ are nearly identical. Both systems display long range antiferromagnetic order above the critical composition, which is characterized by amplitude modulated Ce-moments with an incommensurate wavevector.$^{27,28}$ At the QCP, CeCu$_{5.8}$Ag$_{0.2}$ materials display a logarithmic divergence of the heat capacity over temperature ($C/T$) suggesting physics beyond the HMM model.$^{30}$ In CeCu$_{5.8}$Ag$_{0.2}$, the divergence of the Gruneisen ratio is much weaker than the predictions of HMM model.$^{30}$ Hence, CeCu$_{6}$–Ag$_{x}$ is an ideal model system to investigate the microscopic origins of the quantum critical behavior.

In this paper, we study the quantum critical behavior of CeCu$_{5.8}$Ag$_{0.2}$ through inelastic neutron scattering (INS) measurements. These measurements reveal a complex spin fluctuation spectrum which can be parameterized as consisting of fluctuations occurring at multiple reciprocal space positions. When separated from the other components of the spectrum, fluctuations corresponding to the ordering wave vector in magnetically ordered compositions are found to be three dimensional and scalable according to the predictions of the HMM model. Furthermore, in order to contextualize our new results with the previous work on CeCu$_{5.8}$Au$_{0.2},$$^{10,11}$ the imaginary part of the dynamic susceptibility, $\chi$, as a function of temperature, $T$, energy transfer, $E$, and momentum transfers, $Q$, is scaled with the phenomenology previously described in refs $^{10,11}$ This analysis yields a fractional exponent $\alpha = 0.73(1)$, which is very close to the value of 0.75(5) found for CeCu$_{5.8}$Au$_{0.2}.$$^{10}$ Together, these observations demonstrate that a multicomponent fluctuation spectrum is a mechanism for generating $E/T$ scaling with a fractional exponent. Given the similarity between the physical properties of CeCu$_{5.8}$Au$_{0.2}$ and CeCu$_{6}$–Ag$_{x}$, the results presented here likely have broader implications.

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RESULTS

Sample preparation and the inelastic neutron scattering (INS) measurements are described in the Methods section and additional details are given in the supplementary information (SI). The quality of the sample was checked by energy dispersive x-ray spectroscopy (EDS) and neutron diffraction, which confirm that the sample is homogeneous. In addition, thermodynamic measurements show that heat capacity over temperature \( C/T \) logarithmically diverges with temperature, indicating that the sample is close to the QCP (Fig. S2). Several methods were used to extract the magnetic signal from the measured spectra, the details of which are described in the Supplementary information.

Figure 1 shows the evolution of the magnetic scattering in the \((H\ 0\ L)\) scattering plane with \( E \) and \( T \). Magnetic scattering is observed in the form of a diffuse pattern centered at \( Q_x = (1 0 0) \) instead of the incommensurate position \( Q_{1} = (0.65, 0, 0.3) \) where the magnetic Bragg peak is observed in the magnetically ordered side of the QCP. The intensity of the scattering is maximum at \( (100) \) but does not strongly vary in the nearby region of reciprocal space. With increasing \( E \), the intensity of the scattering decreases and becomes more diffuse as shown in Fig. 1a–d. A qualitatively similar situation arises with increasing \( T \). As shown in Fig. 1a, e–h, the pattern of scattering remains essentially unchanged until 1 K but becomes more diffuse at higher temperatures. A more detailed analysis is presented in the following sections.

Figure 2 shows the scattering in the \((H\ K\ 0)\) scattering plane. In addition to the magnetic scattering described above, scattering centered at \( Q_x = (0 1 0) \) (Fig. 2a) is observed. The correlation length along the \( b \)-axis was determined with a fit of a Lorentzian function to a cut along \((1\ K\ 0)\), which is shown in Fig. 2b. The fit yields \( \xi_b = 28(1) \) Å which, as discussed in more detail below, is comparable to the real space correlation lengths in the \( ac \)-plane.

A central question concerning the nature of the quantum critical behavior in CeCu\(_{6}\)Ag\(_{x}\) is: What type of spin-spin correlations in real space give rise to a butterfly shaped pattern and the observed \( E/T \) scaling? The magnetic order in CeCu\(_{6}\)Ag\(_{x}\) is incommensurate with a wave-vector \( Q_x = (0.65, 0, 0.3) \) and is nearly independent of Ag composition.\(^{27}\) Hence, the critical scattering is expected at the wave-vector \((0.65, 0, 0.3)\) and equivalent positions in reciprocal space, e.g., \((1.35, 0, 0.3)\). However, fluctuations at only these spots are inconsistent with the maximum intensity occurring at \( Q_x = (1 0 0) \). A previous proposal to explain the spin excitation spectrum of CeCu\(_{6}\)Au\(_{x}\), was that the real space correlations are two dimensional\(^{31}\) resulting in rod shaped structures in reciprocal space. While the overlap of rods is not apparent from Fig. 1, we have explored this possibility with and without including considerations of finite correlation lengths (see Supplementary materials). As described in SI and Fig. 3, models having fluctuations only at a single wave-vector do not reproduce the data presented here (see Supplementary materials). Instead this analysis implies that there are additional fluctuations centered at the wave-vector \( Q_x = (1 0 0) \). The overlap of these fluctuations renders the observed pattern of diffuse magnetic scattering.

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**Fig. 1** Constant energy slices of the INS data from CeCu\(_{6}\)Ag\(_{0.2}\) collected with CNCS. a–d The \( E \) dependence of the scattering in \((H\ 0\ L)\) scattering plane at 0.25 K. e–h The \( T \) dependence of the magnetic scattering in the interval \( E = (0.1, 0.3) \) meV. In all panels, data collected at 50 K is used as the background and is subtracted from the data. Each pixel in the plot represents the integrated intensity in the area of dimension \(0.05^2\) r.l.u\(^2\). r.l.u denotes reduced lattice units. Note: The black circles represent regions where the magnetic Bragg reflections occur in the antiferromagnetically ordered compositions of CeCu\(_{6}\)Ag\(_{x}\).

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**Fig. 2** Inelastic neutron scattering data collected with MACS at 0.3 K with 50 K data subtracted to remove background. a Constant \( E \) slice at 0.25 meV showing the magnetic scattering in the \((H\ K\ 0)\) scattering plane. Each pixel in the plot represents the integrated intensity in the area of dimension \(0.05^2\) r.l.u\(^2\). b Cut along \((1\ K\ 0)\). The green and blue dotted lines represent fits of the Lorentzian function to the fluctuations centered at \((0 0 0)\) and \((1 \pm 1 0)\), respectively. The red line is the sum of Lorentzian components.
Lorentzian functions were used, one corresponding to \( Q_1 \) and symmetry equivalent wave-vectors and the other to \( Q_2 \). This parameterization provides a good description of the data for all temperatures and energy transfers. We note that the model deviates from the experimental data near \( Q = (1, 0, \pm 0.5) \) (Fig. 3f), indicating that the spectrum may be more complex than our current parameterization. However, the key point is that this minimal parameterization allows the fluctuations at \( Q_1 \) to be separated and analyzed independently of the remainder of the spectrum. An example of the parameterization at 0.25 K is shown in Fig. 3b, which consists of the 2D-Lorentzian components corresponding to \( Q_1 \) and \( Q_2 \). The cuts are also shown in Fig. 3e, f. This parameterization also allows estimation of correlation lengths and energy dependence of scattering at \( Q_1 \) and \( Q_2 \). As shown in Fig. 4a, the spectral weight of \( Q_1 \) is quasielastic below 0.5 meV. This is in accord with the observed incommensurate antiferromagnetic ground state associated with \( Q_1 \) as opposed to a commensurate antiferromagnetic ground state associated with \( Q_2 \). The correlation lengths along parallel and perpendicular axes are of the same order over the entire spectrum (see Supplementary materials). At 0.25 K and at 0.2 meV, parallel, \( \xi_p \), and perpendicular, \( \xi_\perp \), components of the correlation lengths of the fluctuation at \( Q_1 \) are 47.2 Å and 69.3 Å, respectively. The correlation lengths being of the same order of magnitude indicates that the fluctuation at \( Q_1 \) is clearly three dimensional. On the other hand, the fluctuation at \( Q_2 \) appears to be inelastic with a small gap (Fig. 4b). Similarly, at 0.25 K and at 0.2 meV, the components of correlation lengths for the fluctuation at \( Q_2 \) along the \( a \) and \( c \)-axis are \( \xi_a = 35(3) \) Å and \( \xi_c = 9.3(5) \) Å.

Taken together, the data and analysis presented here indicate the presence of at least three distinct spin fluctuations in CeCu5.8Ag0.2: \( Q_1 = (0.65, 0.3, 0) \), \( Q_2 = (1, 0, 0) \), and \( Q_0 = (0, 1, 0) \). The incommensurate fluctuations \( \{Q\} \) are characteristic of the magnetically ordered side of CeCu5.8Ag0.2. As noted in earlier reports, the commensurate fluctuations \( Q_2 \) also appear in the parent compound CeCu6, which is the paramagnetic end member of the series. Interestingly, the \( Q_2 \)-fluctuations are gapped with small energy transfer, similar to that reported in CeCu6. The intensities and the energy scale of these fluctuations are very similar, indicating that all likely play a significant role in the thermodynamic properties. The spectral weights of \( Q_1 \) and \( Q_2 \) fluctuations are different but of the same order of magnitude. For instance, at the base temperature of 0.25 K and \( e = 0.25 \) meV, the weight of \( Q_1 \)-fluctuations averaged over the Brillouin zone is 75% of the \( Q_2 \)-fluctuations. Thus it is natural to expect their presence influences the critical properties of the QCP. Understanding the critical behavior of this rich fluctuation spectrum is thus important. For the remainder of the paper, we focus on the critical behavior of the fluctuations \( Q_1 \) and \( Q_2 \), since these are most closely related to each other and to the magnetically ordered region of the phase diagram. There is a natural relationship between these two types of fluctuations: both fluctuations represent the tendency to form antiferromagnetic order with \( Q_1 \) and \( Q_2 \) reflecting the tendency for incommensurate (commensurate) order.

For a 3D QCP, the HMM model predicts that \( \chi^c \) scales as a function of \( E_{\text{T}1/2}^{3/2} \). After the separation of the fluctuations, when only the \( Q_1 \) contribution to \( \chi^c \) is scaled as a function of energy and temperature, \( E_{\text{T}1/2}^{3/2} \)-scaling is observed in CeCu5.8Ag0.2. As shown in Fig. 4c, the quantity \( \chi^c T^{3/2} \) at different temperatures collapses onto a single curve which can be fit with the equation \( \chi^c = T^{-3/2} (\eta/T)^{3/2} \). This shows that the incommensurate component of the fluctuation spectrum of CeCu5.8Ag0.2 is consistent with the HMM model.

For a quantitative comparison of our measurements with previous studies in the related compound CeCu6.5Au0.5, we analyzed our measurements with the phenomenology used previously to examine the critical dynamics of CeCu5.8Ag0.2.10,11 This phenomenology relates the magnetic susceptibility with \( E, T, \ldots \).
Fig. 4 Spectral weights of (a) $Q_1 = (0.65 \pm 0.3)$ (b) $Q_2 = (1 0 0)$ extracted from the fit of 2D-Lorentzian function. c Scaling analysis of the component of $\chi''$ near $Q_1$ using the HMM approach. This component of $\chi''$ is extracted from the fit of a sum of overlapping Lorentzians as shown in Fig. 3.

Fig. 5 Scaling analysis of CeCu$_{5.8}$Ag$_{0.2}$. a–c $\chi''/T^\alpha$ scaled as a function of $E/T$ in different regions of reciprocal space. The solid lines are a fit of Eq. (2) to the data. A global fit of Eq. (2) yields the scaling exponent $\alpha = 0.73(1)$. d $q$-dependence of $\theta$ determined from the fit shown in panels a–c. Near $\mathbf{q} = \mathbf{Q} - \mathbf{Q}_0 = 0$, $\theta(q) \approx q^2$ (red line). e $\chi''$ at 0.25 K scaled as a function of $E/\theta(q)$. $\chi''$ at different $Q$s collapses onto a single curve showing $E/\theta(q)$-scaling. Each color represents a point in the (H 0 L) scattering plane. The solid line is a fit of Eq. (2) at zero $T$ to the data. f The variation in the goodness of fit parameter $\chi^2$ with $\alpha$. 
and $Q$ and is rooted in the Curie-Weiss law.\textsuperscript{10} Within this picture, the dynamic susceptibility, $\chi(Q, E, T)$ can be written as,

$$\chi(Q, E, T) = \frac{C}{q^\alpha + (T - E)^\beta}$$

where $\delta(Q - Q_0)$ captures the wave-vector dependence of the magnetic fluctuations similar to the Curie–Weiss temperature. $\alpha$ is a scaling exponent independent of $Q$, $E$, and $T$. The mean field limit is given by $\alpha = 1$. This equation yields $E/T$ scaling at the critical wave-vector and $E/(\theta Q)$-scaling at zero temperature.\textsuperscript{19}

To make contact with the above picture, we performed a global fit of the imaginary part of Eq. (2) to $\chi''$ at several $Q$, $E$ and $T$ without the separation of the fluctuations. The quantity $\delta(Q - Q_0)$ was included as a fitting parameter without assumption of a particular functional form. The fit yields $\alpha = 0.73(1)$ (see Fig. 5f for $\alpha$ as a function of the goodness of fit). The measurement and fit for several regions of the $(H 0 L)$ scattering plane are presented in Fig. 5a–c. For ease of comparison among different regions of the $Q$-space, $\chi''$ versus the dimensionless ratio $E/T$ is plotted. Consistent with Eq. (2), $\chi''$ at several temperatures collapses onto a single curve displaying $E/T$ scaling at $Q_0 = (1 0 0)$, which is the center of the diffuse structure (Fig. 5a). The overlap becomes less pronounced as the magnitude of $q = (Q - Q_0)$ increases. The deviation from $E/T$-scaling at higher $q$ is captured by the $\delta(q)$ term of Eq. (2). The values of $\delta(q)$ as a function of $q = (Q - Q_0)$ are presented in Fig. 5d, which shows that $\delta(q)$ varies quadratically near the center of the pattern ($q = 0$). As an alternative test, we performed $E/(\theta Q)$ scaling at 0.25 K. As shown in Fig. 5e, $\chi''$ at 0.25 K is well fit by Eq. (2) and the quantity $\chi''(\theta Q)^\alpha$ at different $q$ collapses onto a single curve.

The observed $E/T$ scaling with the exponent 0.73(1) implies that the QCP is unconventional. This observation is very similar to the related system CeCu$_5.8$Ag$_{0.2}$,\textsuperscript{10,11} The similarity of the $E/T$ scaling shown here, thermodynamic properties,\textsuperscript{8,36,37} and microscopic details of the magnetic order\textsuperscript{25,38} demonstrate that the QCP in CeCu$_5.8$Ag$_{0.2}$ is virtually identical to the QCP in CeCu$_5.8$Au$_{0.2}$.

**DISCUSSION**

There are important implications of the above analysis. Firstly, while the $E/T$ scaling, as well as the anomalous exponent of 0.73 is tantalizingly close to the expectations of several theoretical models,\textsuperscript{15,18–21} the critical magnetic fluctuations being 3D and suggests that these models, which as currently formulated to rely on 2D correlations, are inapplicable to the QCP studied here. Second, the $E/T$ scaling of the critical fluctuations indicates that long wavelength fluctuations of the order parameter play an important role. In addition, the $E/(\theta Q)$ scaling and the associated implication that fluctuations at $Q_1$ conform to the HMM model suggests that the separation is fundamentally correct. Thus the analysis presented here demonstrates that in addition to the theoretical predictions described above, overlap of multiple fluctuations provides an alternative explanation to the observed $E/T$-scaling. Although, the existence of multiple magnetic fluctuations is indeed a common feature of Ce-based QCPs,\textsuperscript{39,40} CeCu$_5.8$Ag$_{0.2}$ differs from other Ce-based QCPs as the fluctuations are not well-separated in reciprocal space. In this scenario, an important follow up question is whether the coupling between different fluctuations gives rise to a fractional exponent as described in the literature.\textsuperscript{41} Additional measurements as a function of composition and field are required to answer this question.

The results presented here provide explanation to some of the longstanding questions in the family of CeCu$_5.8$T$_x$ ($T =$ Ag, Au) system. Earlier studies show that some members of these family possibly host a structural QCP.\textsuperscript{27,42–44} In CeCu$_5.8$Au$_{0.2}$ the structural QCP nearly coincides with the magnetic QCP raising a possibility that the underlying multiritical point\textsuperscript{45} gives rise to an anomalous $E/T$-scaling. However, our work demonstrates that the $E/T$-scaling also occurs in CeCu$_6.4$Ag$_{0.6}$ where the structural and magnetic QCP are well-separated. This verifies that the magnetic QCP in CeCu$_6.4$T$_x$ is not influenced by the structural QCP. Another interesting question in CeCu$_5.8$T$_x$ family is why the field tuned QCPs are characteristically different from the composition tuned QCPs.\textsuperscript{34–36} The compositionally tuned QCPs in CeCu$_5.8$T$_x$ are unconventional,\textsuperscript{10,11,36} while the field tuned QCPs in both systems appear to adhere to the framework of three dimensional HMM model.\textsuperscript{14,37} In the case of the field tuned QCP achieved by suppressing an antiferromagnetically ordered phase,\textsuperscript{14,37} the HMM scaling was found at the position of a magnetic ordering wave vector—which is an equivalent wave vector to where we find HMM scaling in CeCu$_6.4$Ag$_{0.6}$. Thus the results here show that the nature of the QCPs are likely to be the same, and independent of whether the tuning mechanism is field or composition.

The correlation lengths remain finite at the QCP despite the critical part of the fluctuation spectrum exhibiting HMM scaling. This is unexpected especially considering that the heat capacity measurements show a logarithmic divergence of heat capacity over temperature indicating a strong divergence in the thermodynamic average of these fluctuations (see Supplementary materials), and the spectrum fits with the phenomenological scaling which implies that both the correlation length and time diverge at the QCP. However, an important point here is that there are more than one type of low energy fluctuation contributing to the heat capacity and other bulk thermodynamic measurements. Thus the true behavior of the QCP may be masked in measurements which measure all thermally accessible fluctuations. In addition, it has been established from earlier studies on other QCP systems that the correlation lengths remain finite while $C/T$ shows a strong divergence.\textsuperscript{45–47} Such non diverging correlation lengths has been attributed to the presence of intrinsic disorder or the phase transition becoming weakly first order\textsuperscript{45–47} and similar explanations may be relevant for CeCu$_5.8$Ag$_{0.2}$.

In summary, we have studied the critical behavior of CeCu$_5.8$Ag$_{0.2}$ with extensive INS measurements of the fluctuation spectrum as a function of $T$, $E$, and $Q$. Our analysis over a large region of reciprocal space shows that there are at least three magnetic fluctuations with similar spectral weights. The portion of fluctuation spectrum corresponding to the magnetically ordered region of the phase diagram scales as $E/T$\textsuperscript{1/2} demonstrating that the critical behavior in CeCu$_5.8$Ag$_{0.2}$ is consistent with the conventional 3D antiferromagnetic QCP. Further, when analyzed as a whole, $\chi''$ displays a phenomenological scaling with an anomalous fractional exponent of 0.73(1). The results here then indicate that an additional means of generating $E/T$ at a QCP is complex fluctuation spectrum.

**METHODS**

**Material growth**

A single crystal of CeCu$_5.8$Ag$_{0.2}$ was grown using the Czochralski technique. Starting elements Ce (Ames Laboratory, purity = 99.998%), Cu (Alpha Aesar, purity = 99.999%), Ag (Alpha Aesar, purity = 99.9999%) were melted in a stoichiometric proportion. The Czochralski process was performed in a tri-arc furnace with a graphite hearth. The furnace was continuously purged with ultra high purity argon during the growth. The hearth was rotated with a constant speed of 100 revs/min. A seed rod was pulled with a constant vertical speed of 20 mm/h.

**Inelastic neutron scattering measurements**

Inelastic neutron scattering measurements in the (H 0 L) scattering plane of CeCu$_5.8$Ag$_{0.2}$ were carried out using the cold neutron chopper spectrometer (CNCs) of the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL). The measurement was performed with an incident energy ($E$) of 2.5 meV, which results in an elastic energy resolution of 0.07 meV. An additional measurement at 50 mK was collected in a...
DATA AVAILABILITY

Data presented in this study are available from authors upon reasonable request.

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AUTHOR CONTRIBUTIONS

L.P. and A.D.C. conceived the research project. L.P. and D.M. synthesized the single crystal for the measurement. A.F.M. and F.R. performed preliminary characterization of the sample. L.P., L.W., G.E., Y.Q. and A.D.C. performed the INS measurements. L.P. analyzed the data with help of J.M.L. and A.D.C. L.P., J.M.L. and A.D.C. prepared the paper with input from all authors.
COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
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