Editors' Suggestion

Ultrahigh-resolution neutron spectroscopy of low-energy spin dynamics in UGe$_2$

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Studying the prototypical ferromagnetic superconductor UGe$_2$ we demonstrate the potential of the modulated intensity by zero effort (MIEZE) technique—a novel neutron spectroscopy method with ultrahigh energy resolution of at least 1 μeV—for the study of quantum matter. We reveal purely longitudinal spin fluctuations in UGe$_2$ with a dual nature arising from 5f electrons that are hybridized with the conduction electrons. Local spin fluctuations are perfectly described by the Ising universality class in three dimensions, whereas itinerant spin fluctuations occur over length scales comparable to the superconducting coherence length, showing that MIEZE is able to spectroscopically disentangle the complex low-energy behavior characteristic of quantum materials.

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I. INTRODUCTION

Ultraslow spin dynamics represent a key characteristic of quantum matter such as quantum spin liquids [1], electronic nematic phases [2], topological spin textures [3,4], non-Fermi liquid behavior [5–8], and unconventional superconductivity [9]. For the clarification of these phenomena spectroscopic methods with excellent momentum and energy resolution are required, as key characteristics emerge typically in the low milli-Kelvin range. In principle, neutron scattering is ideally suited for studies of the relevant spin excitations. However, the typical energy resolution of conventional neutron spectroscopy corresponds to Kelvin temperatures. Although techniques such as neutron spin-echo spectroscopy offer ultrahigh resolution of sub-μeV, they are incompatible with conditions that depolarize neutron beams such as ferromagnetism (FM), superconductivity, or large magnetic fields.

The discovery of superconductivity in the FM state of UGe$_2$ highlights the combination of scientific and experimental challenges that arise in the study of the complex low-energy behavior of quantum matter that characteristically emerges due to the competition of high-energy atomic energy scales [10]. Namely, actinide-based compounds such as UGe$_2$ are formidable model systems, where the hybridization of itinerant $d$ and localized $f$ electrons drives low-energy excitations that mediate a multitude of novel states [11–13]. Traditionally the concomitant subtle reconstruction of the electronic structure has been studied via the charge channel, which fails to provide the required high resolution [14,15]. Exploiting in contrast the spin channel recent advances in neutron spectroscopy provided new insights [16–18].

Using an implementation of neutron resonance spin echo (NRSE) spectroscopy that is insensitive to depolarizing conditions, namely the so-called modulated intensity by zero effort (MIEZE) [19], we report a study of UGe$_2$ in which we identify the enigmatic low-energy excitations as an unusual combination of fluctuations attributed normally either to itinerant or localized electrons in an energy and momentum range comparable to the superconducting coherence length and ordering temperature. As the superconductivity in UGe$_2$ represents a prototypical form of quantum matter, our study underscores also the great potential of the MIEZE technique in studies of quantum matter on a more general note.

At ambient pressure UGe$_2$ displays ferromagnetism with a large Curie temperature, $T_c = 53$ K, and a large ordered moment, $\mu_{FM1} = 1.2 \mu_B$ (FM1) [20,21]. Under increasing pressure FM order destabilizes, accompanied by the emergence of a second FM phase below $T_c < T_c$ where $\mu_{FM2} = 1.5 \mu_B$ (FM2). The FM2 and FM1 phases vanish discontinuously at $p_c \approx 12.2$ kbar and $p_c \approx 15.8$ kbar, respectively [21], while superconductivity emerges between $\approx 9$ kbar $< p_c$ and $p_c$. Evidence for a microscopic coexistence of FM order and superconductivity makes UGe$_2$ a candidate for p-wave pairing, where the Cooper pairs form spin triplets [20]. This p-wave superconductivity is believed to be mediated by an abundance of low-lying longitudinal spin fluctuations associated

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with a FM quantum phase transition (QPT), where transverse spin fluctuations are theoretically known to break spin-triplet pairing \[22\]. Neutron triple-axis spectroscopy (TAS) at ambient pressure indeed identified predominantly longitudinal spin fluctuations in UGe\(_2\) \[23\] but failed to provide insights into the character of the fluctuations in the momentum and energy range comparable to the superconducting coherence length and transition temperature, respectively.

Prior to our study the interplay of seemingly conflicting ingredients of the spectrum of spin fluctuations were unresolved. On one hand, the strong Ising anisotropy promotes longitudinal spin fluctuations as typically attributed to localized electrons in the presence of strong to spin-orbit coupling. This is contrasted, on the other hand, by the notion of Cooper pairs and a well developed, strongly exchange-split Fermi surface \[22,24\]. Consistent with this dichotomy characteristic of p-wave superconductivity, our ultrahigh resolution data reveals that the low-energy spin fluctuations of UGe\(_2\) reflect a subtle interplay of itinerant and local electronic degrees of freedom on scales comparable to the superconductivity.

II. EXPERIMENTAL METHODS

NRSE achieves extreme energy resolution by encoding the energy transfer \(\hbar \omega\) of the neutrons in their polarization as opposed to a change of wavelength. However, FM domains, Meissner flux expulsion, or applied magnetic fields typically depolarize the beam. We used therefore a novel NRSE technique, so-called MIEZE, implemented at the instrument RESEDA at the Heinz Maier-Leibnitz Zentrum (MLZ) \[25–27\]. Generating an intensity modulated beam by means of resonant spin flippers and a spin analyzer in front of the sample, amplitude of the intensity modulation assumes the role of the NRSE polarization. Because all spin manipulations are performed before the sample, beam depolarizing effects are no longer important. Using incident neutrons with a wavelength \(\lambda = 6\ \text{Å}\) and \(\Delta \lambda/\lambda \approx 10\%\) provided by a velocity selector, we achieved an energy resolution of \(\Delta E \approx 1\ \mu\text{eV}\). MIEZE in small angle neutron scattering (SANS) configuration also provides high momentum \(q\) resolution of approximately 0.015 Å\(^{-1}\). The MIEZE setup is described in the Supplemental Material \[28\].

A high-quality single crystal of UGe\(_2\) was grown by the Czochralski technique followed by an annealing similar to Ref. \[29\]. A cylindrical piece with nearly constant diameter of 7 mm and 16 mm length \((m = 6\ \text{g})\) with the crystallographic \(c\) axis approximately parallel to the cylinder axis was cut for the MIEZE experiments. The sample was oriented using neutron Laue diffraction so that \(c\) was perpendicular to the scattering plane. The Laue images also confirm a high-quality single-grain sample \[28\]. Neutron depolarization imaging measurements \[28\] of the same sample reveal that the magnetic properties of the crystal are completely homogeneous with a Curie temperature \(T_C = 52.68(3)\ \text{K}\) demonstrating that this sample is optimal for the investigation of critical spin fluctuations. Magnetic susceptibility measurements were performed on a small piece \((m = 36\ \text{mg})\) of the same sample in a Quantum Design magnetic property measurement system (MPMS).

FIG. 1. Magnetic intensity in UGe\(_2\) near the Curie temperature \(T_C = 52.7\ \text{K}\). (a) and (b) show two experimental configurations with the \(a\) axis parallel or perpendicular to the incident neutron beam \(n\), respectively, used to differentiate longitudinal from transverse spin fluctuations (see text). (c) and (d) show the observed energy-integrated intensities for \(n \parallel a\) and \(n \perp a\) as a function of temperature \(T\) and momentum transfer \(q\). The black dashed line marks \(T_C\).

III. RESULTS AND DISCUSSION

The magnetic cross section is related to the imaginary part of the dynamical magnetic susceptibility \(\chi''_{ij}(\mathbf{q}, \omega)\) via

\[
\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{k_f}{k_0} (\delta_{ij} - \hat{q}_i \hat{q}_j) F_{q} |n(\omega) + 1|^2 \chi''_{ij}(\mathbf{q}, \omega),
\]

where \(k_0\) and \(k_f\) are the wave vector of the incident and scattered neutrons, respectively. \(\hat{q}\) is a unit vector parallel to the scattering vector \(\mathbf{q}\) and \(n(\omega)\) is the Bose function. \(F_{\mathbf{q}}\) is the uranium magnetic form factor.

In Fig. 1 we show the temperature and \(q\) dependence of the energy-integrated intensity of the spin fluctuations in UGe\(_2\) that was obtained by switching the MIEZE setup off. Nonmagnetic background scattering obtained well above \(T_C\) was subtracted from all data sets shown. The temperature scan was carried out with the crystallographic \(a\) axis, which is the magnetic easy axis for UGe\(_2\) oriented parallel \((n \parallel a)\) and perpendicular \((n \perp a)\) to the incident neutron beam, respectively. Due to the term \(\delta_{ij} - \hat{q}_i \hat{q}_j\) in Eq. (1) neutron scattering is only sensitive to spin fluctuations that are perpendicular to \(\mathbf{q}\). Because in SANS configuration \(\mathbf{q}\) is approximately perpendicular to the incident neutron beam, this allows us to separate longitudinal \((\delta S_L)\) from transverse spin fluctuations \((\delta S_T)\) as illustrated in Figs. 1(a) and 1(b). For \(n \parallel a\) both \(\delta S_L\) and \(\delta S_T\) are perpendicular to \(\mathbf{q}\). As shown in Fig. 1(c) substantial magnetic intensity is observed for this configuration. In contrast, for \(n \perp a\) only \(\delta S_T\) is perpendicular to \(\mathbf{q}\) and the vanishingly small signal observed in this case [see Fig. 1(d)] can only come from transverse spin fluctuations. Because of the cylindrical shape of the sample differences in...
neutron transmission between the two orientations are negligible. As shown in the Supplemental Material [28], the small intensity observed for \( \mathbf{n} \perp \mathbf{a} \) arises from finite \( \mathbf{q} \) resolution, demonstrating that the critical fluctuations in UGe\(_2\) are solely longitudinal.

Inspecting the temperature dependence of the integrated intensity for \( \mathbf{n} \parallel \mathbf{a} \) [see Fig. 1(c)], a pronounced peak is centered at \( T_C = 52.7\) K due to the divergence of critical spin fluctuations. For low \( q \) and for \( T < T_C \) additional intensity is observed that increases like a magnetic order parameter. Figure 2 shows the \( q \) dependence of the intensity for a few temperatures below \( T_C \). Below \( q^* \approx 0.02\) Å\(^{-1}\) the intensity is well described by a \( q^{-4} \) dependence that is characteristic for scattering from FM domains that form below \( T_C \) [30,31]. To follow this so-called Porod scattering towards lower \( q \), we have performed a supporting SANS experiment on the instrument SANS-1 at MLZ (details are described in Ref. [28]) denoted with square symbols in Fig. 2. Observation of Porod scattering down to \( q_{\text{min}} = 0.004\) Å\(^{-1}\) implies the onset of long-range order over length scales \( > 2\pi/q_{\text{min}} \approx 1600\) Å. In Fig. 3, we show the temperature dependence of the intensity for selected \( q < q^* \). Near to \( T_C \) it evolves as \( M^2(T) \propto (1 - T/T_C)^{2\beta} \). We find that \( \beta = 0.32(1) \) describes our data perfectly in agreement with \( \beta^{\text{theo}} = 0.32 \) for a three-dimensional (3D) Ising system [32]. This is also in good agreement with \( \beta = 0.36(1) \) from neutron diffraction [33].

For \( q > q^* \) and for \( T \approx T_C \) the \( q \) dependence is described by a Lorentzian line shape characteristic of critical spin fluctuations with a correlation length \( \xi \). The corresponding dynamical magnetic susceptibility is

\[
\chi''(q, \omega) = \frac{\chi(q) \Gamma_q}{\Gamma_q^2 + \omega^2},
\]

\[
\chi(q) = \frac{\chi_0}{1 + (\xi q)^2},
\]

where \( \Gamma_q \) and \( \chi_0 \) are the momentum dependent relaxation frequency and the static magnetic susceptibility, respectively. Because of the longitudinal character of the spin fluctuations only \( \chi_{\text{uu}} \) is nonzero, and we have thus dropped the indices \( i, j \).

To investigate the critical scattering quantitatively, we subtract the Porod scattering (Fig. 2) from the observed intensities [Fig. 1(c)]. For our experimental conditions the quasistatic approximation [34,35] is valid and thus integrating Eq. (1) with respect to \( h\omega \), we obtain \( \frac{d\sigma}{d\omega} \propto T \chi(q) \) (see Supplemental Material [28]). We show \( \chi(q) \) obtained by dividing the observed intensity by \( T \) for various temperatures in Fig. 4(b). The solid lines are fits to Eq. (3) to extract the \( T \) dependence of \( \chi_0 \) and \( \xi \) shown in Figs. 5(a) and 5(b), respectively. For comparison we show the static magnetic susceptibility \( H/M \) determined by bulk magnetization measurements in Fig. 5(a) (blue squares) that scales perfectly with \( \chi_0 \).

We find that \( 1/\chi_0 \propto (1 - T/T_C)^\gamma \) with \( \gamma = 1.23(3) \) and \( 1/\xi = \kappa = \kappa_0(1 - T/T_C)^\nu \) with \( \kappa_0 = 0.31(2)\) Å\(^{-1}\) and \( \nu = 0.63(2) \) [solid lines Figs. 5(a) and 5(b)]. The critical exponents are in excellent agreement with a 3D Ising FM, for which \( \gamma^{\text{theo}} = 1.24 \) and \( \nu^{\text{theo}} = 0.63 \) [32]. Huxley et al. found 0.29 Å\(^{-1}\) in good agreement with our result. In contrast, they determined \( \nu = 1/2 \), consistent with a mean-field transition [23]. However, their study was limited to \( q > 0.03\) Å\(^{-1}\) and underestimates the divergence of the critical fluctuations.
FIG. 5. The inverse susceptibility $1/\chi_0$ and inverse correlation length $1/\xi$ as a function of temperature $T$ (near the Curie temperature $T_C = 52.7$ K), respectively, resulting from fits in Fig. 4. The blue squares in (a) denote the static easy-axis magnetic susceptibility $H/M$ determined with a magnetic field $H = 0.1$ T. The solid black and red lines are fits to determine the critical exponents for $\chi_0$ and $\xi$ (see text), and the shaded region denotes the uncertainty of the fit.

We now discuss the results of our MIEZE measurements. MIEZE measures the intermediate scattering function $S(q, \tau)$ that is the time Fourier transform of the scattering function $S(q, \omega) = 1/\pi [\Gamma_q(q, \omega) + 1] \chi''_{ij}(q, \omega)$ [cf. Eq. (1)]. In Fig. 6 we show $S(q, \tau)$ for various $q$ at $T_C$. $S(q, \tau)$ for all other measured temperatures are shown in Ref. [28]. Because the spin fluctuations have Lorentzian lineshape [see Eq. (2)] we fit $S(q, \tau)$ with an exponential decay [solid lines in Fig. 6]:

$$S(q, \tau) = \exp(-|\Gamma_q| \cdot \tau).$$

The resulting fluctuation frequency $\Gamma_q$ is shown in Fig. 7.

The momentum dependence of $\Gamma_q$ is described by the dynamical exponent $z$ via $\Gamma_q \propto q^z$. For $T \leq T_C$, we find that $\Gamma_q$ is fitted perfectly by $z = 2.0(1)$ [Fig. 7(a)]. This is in excellent agreement with predictions for a 3D Ising FM, for which $z_{\text{theo}} = 2$ [32]. For $T > T_C$, $\Gamma_q$ is also well described by $z = 2$, however, only above a crossover value of $q^0 = 0.04$ Å$^{-1}$. Below $q^0$, our data is best fit by $\Gamma_q = Aq^z$ with $z = 2.53(4)$ (see Fig. 7). This is consistent with $z = 5/2$ calculated for itinerant FMs within critical renormalization group theory [36] and confirmed for various $d$-electron FMs such as Fe [37], Ni [38], and Co [39]. Notably, typical values reported for $A$ are $3$–$350$ meV Å$^{-5/2}$ [37–40] consistent with $A = 200(2)$ meV Å$^{-5/2}$ that we find for UGe$_2$. As demonstrated in Fig. 7(a) for $T = 54$ K, the fit of $\Gamma_q$ with $z = 2.0(1)$ also describes the data of Huxley et al. [23] (black empty circles) perfectly. However, they conclude that $\Gamma_q$ remains finite for $q \to 0$ in contrast to our findings. This discrepancy is easily explained by considering that their experiment was
We show $\chi_q$ who carried out measurements for $T \ll \xi(z)\Gamma_q$ in agreement with the dynamical scaling prediction $[41]$. In the range over which $q^z$ with $z = 5/2$ is observed, this behavior is corroborated via three independent methods that are illustrated in Figs. 7–9.

IV. SUMMARY

Our results demonstrate that the spin fluctuations in UGe$_2$ exhibit a dual character associated with localized $5f$ electrons that are hybridized with itinerant $d$ electrons. Notably, as expected for a local moment FM with substantial uniaxial magnetic anisotropy all critical exponents determined from our results are in perfect agreement with the 3D Ising universality class $[32]$. Further, $\chi(q)\Gamma_q$ is approximately constant as a function of $q$ down to $q^0$ highlighting that the underlying spin fluctuations are localized in real space. In contrast, the dynamical exponent $z = 5/2$ and $\chi(q)\Gamma_q \to 0$ for $q \to 0$ observed below the crossover value $q^0$ are characteristic of itinerant spin fluctuations. Because the contribution of the conduction electrons to the total ordered moment is less than 3% $[33]$, below $T_C$ fluctuations of localized $f$ magnetic moments are dominant. Spin fluctuations with a dual character are consistent with the moderately enhanced Sommerfeld coefficient $\gamma = 34 \text{ mJ/K}^2\text{ mol of UGe}_2$ $[46,47]$ and a next-nearest-neighbor uranium distance $d_{U-U} = 3.85$ Å $[48]$ near to the Hill value of 3.5 Å $[49]$ that both suggest that the $5f$ electrons in UGe$_2$ are hybridized with the conduction electrons.

In conclusion, the dual nature of spin fluctuations revealed by our MIEZE measurements strongly supports the scenario of $p$-wave superconductivity in UGe$_2$. First, to promote strong longitudinal fluctuations requires strong Ising anisotropy that typically is a result of localized $f$ electrons with substantial spin-orbit coupling and is consistent with critical Ising exponents that we observe above $q^0$. Second, the theory for $p$-wave pairing assumes that it is the same itinerant electrons that are responsible for the coexisting FM and superconducting states $[22]$, highlighting that the low-energy itinerant spin fluctuations below $q^0$ discovered here are crucial to mediate $p$-wave superconductivity. The maximum superconducting...
critical temperature $T_c$ occurs at the QPT at $p_x$ [20,21]. Here a substantial increase of the Sommerfeld coefficient [50] and changes in the electronic structure observed near $p_x$ [51,52] suggest that the hybridization of $5f$ electrons and conduction electrons increases at $p_x$ and corroborates that spin fluctuations with a dual nature are relevant for p-wave superconductivity. This is supported by a theory based on competition of FM exchange and the Kondo interaction that results in a localized to itinerant transition at $p_x$ [53,54].

Further, we note that our findings of longitudinal critical fluctuations in UGe$_2$ are also consistent with the findings for UCoGe [55], which is another material that is a candidate for p-wave superconductivity. However, the results on UCoGe by Hattori et al. [55] were obtained by NMR measurements that are unable to probe spin fluctuations at finite $q$ and, in turn, are unable to observe an itinerant-to-localized crossover as we report it here. Similarly, TAS measurements of UCoGe by Stock et al. [56] lack the required momentum and energy transfer resolution.

Finally, we note that the crossover value $q^0$ corresponds to a length scale of approximately 160 Å. The superconducting coherence length of UGe$_2$ was estimated as $\xi^{SC} = 200$ Å [20], which shows that the spin fluctuations relevant to the p-wave pairing are present at $q < q^0$. This may explain why triple-axis measurements of the spin fluctuation near $p_x$ with limited resolution were inconclusive [57]. Although the pressure dependence of the crossover length scale $q^0$ remains to be determined to unambiguously associate it with the unconventional superconducting state in UGe$_2$, our results highlight that recent developments in ultrahigh resolution neutron spectroscopy are critical for the study of low-energy spin fluctuations that are believed to drive the emergence of quantum matter states. Here the fluctuations that appear at zero $q$ such as for ferromagnetic and electronic-nematic quantum states can immediately be investigated via the MIEZE SANS configuration used here. In addition, MIEZE can be extended in straightforward fashion to study quantum fluctuations arising at large $q$ [58], allowing for insights in antiferromagnetic QPTs and topological forms of order.

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[28] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.99.014429 for the characterization of the sample with neutron Laue diffraction and neutron depolarization analysis, as well as detailed information about the used MIEZE setup, and additional SANS measurements. Further, we describe approximations used for the analysis of energy-integrated magnetic critical scattering and resolution calculations. Finally, we explain how the MIEZE data was analyzed.

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