Magnetic-Field-Induced Enhancements of Nuclear Spin-Lattice Relaxation Rates in the Heavy-Fermion Superconductor CeCoIn$_5$ Using $^{59}$Co Nuclear Magnetic Resonance

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Among heavy-fermion systems, there is a growing number of examples of the emergence of unconventional superconductivity near a magnetic-nonmagnetic boundary tuned toward a zero temperature (quantum) critical point (QCP), raising the possibility of a connection between these phenomena [1]. In particular, the CeTh$_5$ (T=Co, Rh, Ir) materials, the so-called Ce115 family, have served as instructive examples by motivating the need to understand phenomena around an antiferromagnetic (AFM) QCP, including the observation of non-Fermi liquid (NFL) behavior, and their relationship to superconductivity [2]. CeCoIn$_5$ is a d-wave heavy-fermion superconductor with $T_c = 2.3$ K [3], and is thought to be located at the slightly positive pressure side of an AFM QCP at zero magnetic field [4]. Indeed, slight Cd-substitutions for In, which act as a negative chemical pressure in CeCoIn$_5$, induces long-range AFM order [5]. One of the several intriguing properties of CeCoIn$_5$ is the discovery of the “Q phase” at low temperatures just below the first-order upper critical field $H_{c2}$ boundary in the a-b plane [6, 7]. Though possibly reflecting the emergence of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [6], the “Q phase” supports incommensurate spin density wave order that coexists spatially with superconductivity [8].

Another important finding is a possible QCP induced by a magnetic field applied along tetragonal c-axis. Although several phase diagrams have been proposed on the basis of resistivity [9–12], specific heat [13], linear thermal expansion [14], and volume thermal expansion [15] measurements, a common feature of these proposals is that an extrapolation of the normal-state boundary between Fermi-liquid (FL) and NFL behaviors to $T \to 0$ intersects the field axis near $H_{c2}(T \to 0) = 49.5$ kOe. The cyclotron mass, determined by de Haas-van Alphen experiments, also is enhanced at $H_{c2}(0)$ [16]. Because these macroscopic physical properties do not probe spin-dynamics directly, the relationship of the field-induced critical behavior to magnetic fluctuations has not been established. In the case that there is an association with spin dynamics, nuclear magnetic resonance (NMR) relaxation provides a direct probe of their role as a consequence of the hyperfine coupling. In this Letter, we report on the $(H_0, T)$-dependences of the nuclear relax-ation rate $(1/T_1)$ and Knight shift $(K)$ for $^{59}$Co in the normal state of CeCoIn$_5$. A critical increase of $1/T_1$ for $^{59}$Co NMR is observed at fields $H_0 \sim H_{c2}(0)$, for $H_0 \parallel c$. As will be shown, $1/T_1(H_0, T)$ can be understood consistently as arising from 2D-AFM spin-fluctuations (SF), and provide microscopic evidence for a 2D-AFM instability near $H_{c2}(0)$.

A plate-like single crystal ($\sim 2 \times 1 \times 0.3$ mm$^3$) of CeCoIn$_5$ was used for $^{59}$Co NMR measurements. Alignment of the crystal relative to the applied field $H_0$ was checked by nuclear quadrupole splittings of the $^{59}$Co (nuclear spin 7/2) NMR spectrum. Measurements of $K$ and $1/T_1$ were performed by scanning temperature, using the central transition $(1/2 \leftrightarrow -1/2)$ under several applied fields above $H_{c2}(0)$. CeCoIn$_5$ has a tetragonal (HoCoGa$_5$-type) layered structure, which can be thought of as layers of CeIn$_3$ separated by layers of CoIn$_2$ along the c axis. Crystallographically, the Co sites are unique in this structure.

The temperature dependences of $(T_1 T)^{-1}$ and $K$ for $^{59}$Co NMR are shown in Fig. 1. There is a very prominent low-temperature enhancement of $(T_1 T)^{-1}$ along the c axis near $H_{c2}(0)$, although the corresponding increase of $K$ along c is not observed near $H_{c2}(0)$. This enhancement of $(T_1 T)^{-1}$ suggests strong AFM SF, of which quantitative analyses provide an insight into the criticality near $H_{c2}(0)$, as presented later. At all fields, $(T_1 T)^{-1}$ monotonically increases on cooling over the temperature range, $T < 100$ K. At the lowest temperatures, $(T_1 T)^{-1}$ crosses over to a saturation regime, with the crossover increasing in temperature at higher fields. In the case of $H_0 = 50$ kOe, which is nearest to $H_{c2}(0)$, the saturation is only...
seen below $\sim 150$ mK. The saturated behavior in $K$ and $(T_1 T)^{-1}$ is consistent with the FL behavior as observed in the macroscopic physical quantities. A tiny increase of $K_s$ with approaching field to $H_0(0)$ is seen below $\sim 1$ K, which comes from a small increase of spin polarization given by the magnetization along the $c$ axis [7].

In order to extract quantitative information, as well as a context for comparing to previously reported measurements, we analyze the data within the framework of the spin fluctuation theory. For that purpose, we assume a single dynamical susceptibility is relevant near to the QCP. Then, $(T_1 T)^{-1}$ is written as

$$(T_1 T)^{-1} = \frac{k_B}{(\gamma_n h)^2} \cdot 2(\gamma_n A_{1\perp})^2 \sum_q f_{1\perp}(q) \frac{\text{Im} \chi_{1\perp}(q, \omega_0)}{\omega_0},$$

where $\gamma_n$ and $\gamma_e$ are the nuclear and electronic gyromagnetic ratios, $A_{1\perp}$ is the transferred hyperfine coupling constant, $f_1(q)$ is the hyperfine form factor, $\text{Im} \chi_{1\perp}(q, \omega_0)$ is the imaginary part of the dynamical susceptibility, $\omega_0$ is the nuclear Larmor frequency and the suffix $\perp$ the nuclear $Larmor$ frequency and the suffix $A$ the $Co$ site and has no anisotropy with respect to the $a$ and $c$ axes. Therefore, $f^2(q)$ does not affect the sensitivity to strictly two dimensional (2D) AFM SF.

First, let us consider a mean-field approximation. Within a random-phase approximation (RPA), the dynamical susceptibility for weakly correlated quasi-particles can be simplified as $\chi_{RPA}(q, \omega) = \chi_0(q, \omega)/(1 - \alpha_q [\chi_0(q, \omega)/\chi_0(Q, \omega)])$, where $\chi_0(q, \omega)$ is the dynamical susceptibility of noninteracting quasi-particles and $\alpha_q$ is an enhancement factor. $\chi_0(q, \omega)$ gives the well-known Korringa relation $T_1 TK_{q}^{2} = (\hbar / 4\pi K_B)(\gamma_n / \gamma_0)^2 \equiv S$, with $K_s$ being the spin part of $K$. Using $K_s \propto (1 - \alpha_q)^{-1}$, the modified Korringa relation for $\chi_{RPA}(q, \omega)$ is obtained as $T_1TK_{q}^{2} = nSK(\alpha_q)^{-1}$, with $K(\alpha_q) \equiv (1 - \alpha_q)^2(1 - \alpha_q(\chi_0(q)/\chi_0(0)))^{-2}$ where $n = 2$ is the number of nearest magnetic atoms, and $(\cdot \cdot \cdot)$ means an average over the Fermi surface [19]. To deduce the $4f$ electronic component, non-interactive electronic and lattice terms are subtracted by the value of $(T_1 T)^{-1}$ for LaCoIn$_5$ [17]. Since $(T_1 T)^{-1}$ responds to the perpendicular component of SF from Eq. (1), the respective dynamical susceptibility of in-plane and out-of-plane can be obtained by a geometrical decomposition of $(T_1 T)^{-1}$ along $a$ and $c$ axes. Namely, the in-plane and out-of-plane components of $(T_1 T)^{-1}$ are obtained from $(T_1 T)^{-1}/2$ and $(T_1 T)^{-1} - (T_1 T)^{-1}/2$, respectively. $K_s$ is estimated by subtracting $K_{0, i}$ [18]. At 50 kOe, from this modified Korringa relation, the in-plane component of $K(\alpha_q)$ is found to increase rapidly as $T \rightarrow 0$, and much larger than 1 at the lowest temperature. The out-of-plane component of $K(\alpha_q)$ is found to be nearly T-independent and close to 1. $K(\alpha_q) \gg 1$ for the in-plane component indicates AFM correlations at the lowest temperatures. The observations are consistent with easy-plane AFM SF in the low temperatures. Here, the important finding from RPA is a remarkable $T$-dependence of in-plane $\chi(Q)$. In addition, an unusual $H_0$-dependence of in-plane $\chi(Q)$ is also indicated as well by $(T_1 T)^{-1}$ shown in Fig. 1.

In order to treat $\chi(Q)$ at finite temperature, couplings among the $q$-modes of SF should be considered in a self-consistent fashion, beyond RPA, considering a specific $q$-mode only. In such a framework, the dynamical susceptibility can be treated quantitatively by the self-consistent renormalization (SCR) theory [20–22], which has been applied successfully to characterize the nature of SF in many heavy-fermion materials [23, 24]. In the SCR model, the dynamical susceptibility is characterized by two energy scales, $T_0$ and $T_A$, which correspond to the magnetic fluctuation energy in $\omega$- and $q$-spaces, respectively. The $q$ dependence of the effective RKKY interaction $J_Q$ is expressed as $J_Q = J_{Q+q} = 2T_A(|q|/|q_B|)^2$ around the AFM wave vector $Q$, where $q_B$ is the zone-boundary vector. We consider the in-plane SF only in the SCR scheme, using the dimensionless inverse static susceptibility $y = (2T_A \chi(Q))^{-1}$. Here, the out-of-plane component is assumed to be negligibly small due to a weak correlation between planes. The dynamical susceptibility in the 2D-AFM case can be written as $(2T_A \chi(Q + q, \omega))^{-1} = y + (q/q_B)^2 - i\omega/(2\pi T_0)$. Then, the self-consistent equation for $y$ is given using two more parameters $y_0 = (2T_A \chi(Q, 0))^{-1}$ and $y_1 = 2J_Q/(\pi^2 T_A)$.
(3D) AFM SCR scheme, in which it is proportional to temperature cannot be explained by a three-dimensional heat have been fitted to simulations based on terms and exchange interaction previously using a similar 2D SCR model [13], but parameters are not fits but are calculations scaled to the experimental data [14]. This good reproduction of the experimental data attests to the applicability of the 2D-AFM SCR model in CeCoIn5. Moreover, because the sharp decrease of \( \alpha \) below \( \sim 0.3 \) K for 80 kOe can be explained within the 2D-AFM scheme, there is no need to postulate a dimensional crossover from 3D to 2D [14]. A possible dimensional crossover also is excluded in recent measurement of volume thermal expansion [15]. Collectively, these results show that, as \( H_0 \) approaches \( H_{c2}(0) \) from above, the distance from the QCP \( (y_0) \) becomes increasingly small \( (y_0 = 0.04 \) for 80 kOe, 0.022 for 64 kOe) and is nearly zero \( y_0 = 0.008 \) (but still finite) at 50 kOe.

The SCR model also provides an estimate of the in-plane spin correlation length \( \xi/a \), which can be calculated in units of the in-plane lattice parameter \( a \) from \( (1/\sqrt{\pi y})^{-1} \). As shown in Fig. 3(a), \( \xi/a \) at 50 kOe is \( \geq 3 \) at the lowest \( T \), while it is only \( \sim 1.4 \) at 80 kOe. In CeRhIn5, \( \xi/a \) is estimated to be \( \sim 5 \) just above the Néel temperature \( T_N \) = 3.8 K [25]. Similarly, Cd-doped CeCoIn5 induces long-range AFM order where \( \xi/a \sim 4 \) [26]. Therefore, \( \xi/a \) at 50 kOe, Fig. 3(a), indicates that CeCoIn5 is on the threshold of a long-range AFM ordering just near \( H_{c2}(T \to 0) \). Our estimate is close to the zero-field value of \( \xi/a \sim 2.1 \) extracted from inelastic neutron scattering (INS) experiments [27]. We note that a quasi-2D nature of SF is confirmed by the out-of-plane component of \( \zeta/a \sim 0.87 \) from INS, i.e., \( \zeta/a/\xi_a = 2.4/(c/a) \) with the lattice anisotropy of \( c/a \approx 1.6 \) in CeCoIn5. Pa-

\[ y = y_0 + y_1 \int_0^x \left[ \ln u - \frac{1}{2u} - \psi(u) \right] dx, \]
rameters derived from fits to the SCR model also give the characteristic spin fluctuation energy $\Gamma_Q$, computed from $2\pi T_0q_y$. As seen in Fig. 3(b), this $\Gamma_Q$ agrees well with that obtained from INS [27]. Though $\Gamma_Q$ shows no apparent field dependence above $\sim 2$ K, the energy scale of magnetic excitations decreases below $\sim 2$ K as $H_0$ approaches $H_{c2}(0)$. Therefore, our results provide evidence for an energy scale of low-lying magnetic excitations that is $\sim 1$ K in CeCoIn$_5$ and that a magnetic field finely tunes this scale to order $\sim 0.1$ K.

In conclusion, we have demonstrated from microscopic measurements that the field-induced QCP in CeCoIn$_5$, for $H_0 || c$, exists and that the driving force for this QCP is quasi-2D-AFM SF. Although these experiments are unable to determine if the QCP is located exactly at $H_{c2}(0)$, they are consistent with resistivity [12] and volume thermal expansion experiments [15] that locate the QCP just below $H_{c2}(0)$. The relationship of this QCP to the field-induced “H-I phase” (“Q phase”) for $H_0 || a$ remains an open question. At a minimum, a microscopic understanding of the “H-I phase” will need the presupposition of the existence of quasi-2D-AFM QCP, as considered in some theoretical models [28].

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FIG. 3. Temperature dependence of (a) magnetic correlation length $\xi/a$ and (b) spin fluctuation energy $\Gamma_Q$ derived from an SCR analysis of CeCoIn$_5$ at 50 kOe, 64 kOe, and 80 kOe. The broken line in (a) indicates an estimate from inelastic neutron scattering (INS) at zero field. The closed circles in (b) are $T(0)$. Therefore, our results provide evidence for an energy scale of low-lying magnetic excitations that is $\sim 1$ K in CeCoIn$_5$ and that a magnetic field finely tunes this scale to order $\sim 0.1$ K.


