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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^{59}Co nuclear spin-lattice relaxation has been measured for the heavy-fermion superconductor $CeCoIn_5$ in a range of applied fields directed parallel to the c axis. An enhanced normal-state relaxation rate, observed at low temperatures and fields just above $H_{c2}(0)$, is taken as a direct measure of the dynamical susceptibility and provides microscopic evidence for an antiferromagnetic instability. The results are well described using the self-consistent renormalized theory for two-dimensional antiferromagnetic spin fluctuations, and parameters obtained in the analysis are applied to previously reported specific heat and thermal expansion data with good agreement.

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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 $CeTIn_5$ ($T = \mathrm{Co, Rh, Ir}$) 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 $CeCoIn_5$, induce 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 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 the 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 relaxation 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 platelike 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 $(T_1T)^{-1}$ and K for ^{59}Co NMR are shown in Fig. 1. There is a very prominent low-temperature enhancement of $(T_1T)^{-1}$ 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 $(T_1T)^{-1}$ suggests strong AFM SF, of which quantitative analyses provide an insight into the criticality near $H_{c2}(0)$, as presented later. At all fields, $(T_1T)^{-1}$ monotonically increases on cooling over the temperature range $T < 100$ K. At the lowest temperatures, $(T_1T)^{-1}$ crosses over to a saturation

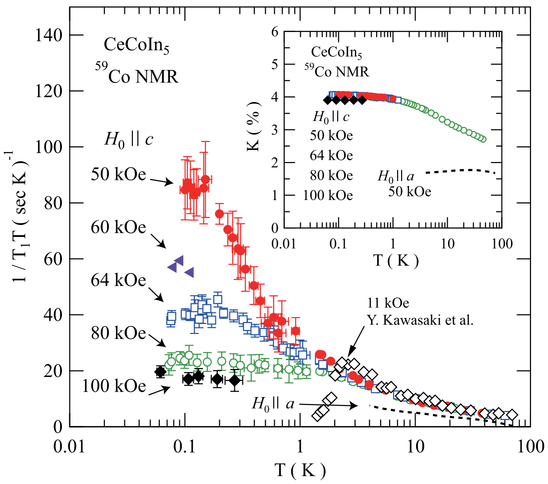

FIG. 1 (color online). $(T_1T)^{-1}$ for ^{59}Co NMR at several fields along the c axis of $CeCoIn_5$. The broken lines are the data under $H_0 = 50$ kOe along the a axis. The results for 11 kOe along the c axis are taken from Ref. [19]. The inset shows the temperature dependence of Knight shifts (K) for ^{59}Co NMR at several fields along the c axis of $CeCoIn_5$.

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 ~150 mK. The saturated behavior in K and $(T_1T)^{-1}$ is consistent with the FL behavior as observed in the macroscopic physical quantities. A tiny increase of K_c with field approaching $H_{c2}(0)$ is seen below ~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 that a single dynamical susceptibility is relevant near the QCP. Then, $(T_1T)^{-1}$ is written as

$$(T_1T)^{-1} = \frac{k_B}{(\gamma_e\hbar)^2} 2(\gamma_n A_\perp)^2 \sum_{\boldsymbol{q}} f_\perp^2(\boldsymbol{q}) \frac{\mathrm{Im}\chi_\perp(\boldsymbol{q}, \omega_0)}{\omega_0}, \quad (1)$$

where γ_n and γ_e are the nuclear and electronic gyromagnetic ratios, A_i is the transferred hyperfine coupling constant, $f_i(\boldsymbol{q})$ is the hyperfine form factor, $\mathrm{Im}\chi_i(\boldsymbol{q}, \omega_0)$ is the imaginary part of the dynamical susceptibility, ω_0 is the nuclear Larmor frequency, and the suffix $\perp$ refers to the component perpendicular to the quantization axis. The hyperfine coupling constants are obtained from the relationship $K_i = A_i\chi_{a,c} + K_{0,i}$, with $K_{0,i}$ independent of temperature. Values of A_i and $K_{0,i}$ were reported previously [17]. The form factor $f^2(\boldsymbol{q}) = 4\cos^2(q_zc/2)$ for the Co site and has no anisotropy with respect to the a and c axes. Therefore, $f^2(\boldsymbol{q})$ does not affect the sensitivity to strictly 2D-AFM SF.

First, let us consider a mean-field approximation. Within a random-phase approximation (RPA), the dynamical susceptibility for weakly correlated quasiparticles can be simplified as $\chi_{\mathrm{RPA}}(\boldsymbol{q}, \omega) = \chi_0(\boldsymbol{q}, \omega)/\{1 - \alpha_{\boldsymbol{q}}[\chi_0(\boldsymbol{q}, \omega)/\chi_0(\boldsymbol{Q}, \omega)]\}$, where $\chi_0(\boldsymbol{q}, \omega)$ is the dynamical susceptibility of noninteracting quasiparticles and $\alpha_{\boldsymbol{q}}$ is an enhancement factor. $\chi_0(\boldsymbol{q}, \omega)$ gives the well-known Korringa relation $T_1TK_s^2 = (\hbar/4\pi k_B)(\gamma_e/\gamma_n)^2 \equiv S$, with K_s being the spin part of K. Using $K_s \propto (1 - \alpha_{\boldsymbol{q}})^{-1}$, the modified Korringa relation for $\chi_{\mathrm{RPA}}(\boldsymbol{q}, \omega)$ is obtained as $T_1TK_s^2 = nSK(\alpha_{\boldsymbol{q}})^{-1}$, with $K(\alpha_{\boldsymbol{q}}) \equiv (1 - \alpha_{\boldsymbol{q}})^2\langle 1 - \alpha_{\boldsymbol{q}}\{\chi_0(\boldsymbol{q})/\chi_0(0)\}\rangle^{-2}$, where $n = 2$ is the number of nearest magnetic atoms and $\langle \cdots \rangle$ means an average over the Fermi surface [18]. To deduce the $4f$ electronic component, noninteractive electronic and lattice terms are subtracted by the value of $(T_1T)^{-1}$ for $LaCoIn_5$ [19]. Since $(T_1T)^{-1}$ 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 $(T_1T)^{-1}$ along the a and c axes. Namely, the in-plane and out-of-plane components of $(T_1T)^{-1}$ are obtained from $(T_1T)_c^{-1}/2$ and $(T_1T)_a^{-1} - (T_1T)_c^{-1}/2$, respectively. K_s is estimated by subtracting $K_{0,i}$ [17]. At 50 kOe, from this modified Korringa relation, the in-plane component of $K(\alpha_{\boldsymbol{q}})$ is found to increase rapidly as $T \rightarrow 0$, and is much larger than 1 at the lowest temperature. The out-of-plane component of $K(\alpha_{\boldsymbol{q}})$ is found to be nearly T independent and close to 1. $K(\alpha_{\boldsymbol{q}}) \gg 1$ 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 $(T_1T)_c^{-1}$, as shown in Fig. 1.

In order to treat $\chi(Q)$ at finite temperature, couplings among the $\boldsymbol{q}$ modes of SF should be considered in a self-consistent fashion, beyond RPA, considering a specific $\boldsymbol{q}$ 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 ω 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 $\boldsymbol{Q}$, where $\boldsymbol{q}_B$ 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 $y_0 \equiv [2T_A\chi(Q,0)]^{-1}$ and $y_1 \equiv 2J_Q/(\pi^2 T_A)$ by

$$y = y_0 + y_1 \int_0^{x_c} x\left[\ln u - \frac{1}{2u} - \psi(u)\right]dx, \qquad (2)$$

with $u = (y + x^2)/t$ and $t = T/T_0$, where $\psi(u)$ is the digamma function and x_c is the reduced cutoff wave vector of order unity. Here, y_0 is a measure of proximity to the QCP, $y_0 = 0$ defining the QCP, and y_1 reflects the strength of dispersion of the effective RKKY exchange interaction J_Q. To deduce the four parameters T_0, T_A, y_0, and y_1, $1/T_1$ and the reported specific heat have been fitted to simulations based on y calculated self-consistently from Eq. (2). $(T_1T)_c^{-1}$ normalized by $(\gamma_n A_a)^2$ and magnetic specific heat C_m/T can be calculated from $(2\pi T_A T_0 y)^{-1}$ and $(2T_0)^{-1}\ln(1 + 1/y) \times (T \ll T_0)$, respectively. Thus, $1/T_1$ directly measures the temperature dependence of $\chi(Q) = [2T_A y(T)]^{-1}$. Note that the observed logarithmic T behavior of C_m/T in the low temperature cannot be explained by a 3D AFM SCR scheme, in which it is proportional to $(a - b\sqrt{T})$ with constants a and b.

The magnetic specific heat C_m/T has been analyzed previously using a similar 2D SCR model [13], but parameters from that analysis cannot reproduce the NMR $1/T_1$, as shown by the broken curve using the reported parameters for 50 kOe in Fig. 2(d). It is noted that the previous value of $T_0 = 0.4$ K is beyond the applicable T range, which should be $T \ll T_0$. Therefore, we have fit those data again to the SCR model using an order of magnitude larger $T_0 \approx 10$ K. The fitting results are shown in Figs. 2(d) and 2(e). The obtained SCR parameters are plotted against the magnetic field in Figs. 2(a)–2(c). T_0 and T_A for $CeCoIn_5$ are about 40 and 10 K, respectively, and show no clear field dependence. y_1 is 6 for 100 kOe, 14 for 64 kOe, and 18 for 50 kOe, a field dependence that may be related to a slight increase of density of states reflected in K. These values of T_0, T_A, and y_1 are similar to those of other Ce-based heavy-fermion materials $CeRu_2Si_2$ and $CeCu_{5.9}Au_{0.1}$ [23]. Because of the uncertainty introduced by a large nuclear Schottky contribution at high fields, the values of these parameters deduced from C_m/T differ slightly from NMR values; nevertheless, y_0 values obtained from the 2D-AFM SCR model fit to both NMR $1/T_1$ and C_m/T approach zero near $H_{c2}(0)$.

In order to confirm the validity of these SCR parameters, the T dependence of the linear thermal expansion coefficient $\alpha = L^{-1}dL/dT$ has been calculated using the same parameters obtained from fits to NMR data. The thermal expansion coefficient is proportional to $T_0(dy/dT)/(y_1T_A)$ [24]. The simulated curves for 50 and 80 kOe are shown in Fig. 2(f). Again, these are not fits but are calculations scaled to the experimental data [14]. This good reproduction of the experimental data attests to the applicability of

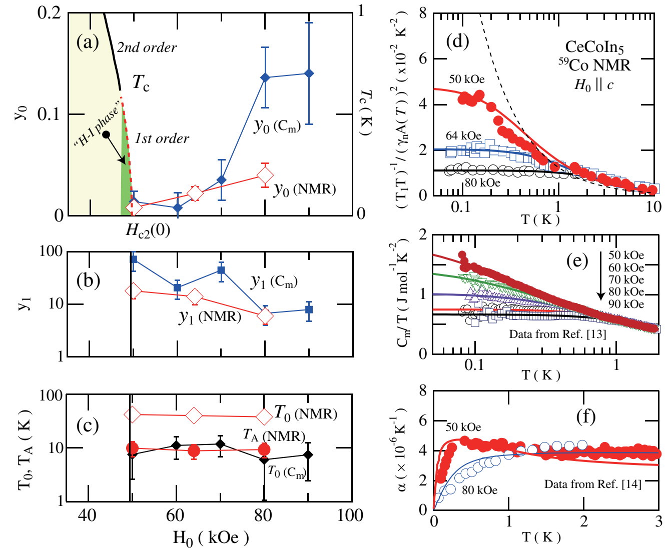

FIG. 2 (color online). Field dependence of (a) y_0, (b) y_1, (c) T_0 and T_A obtained from the SCR analysis of ^{59}Co NMR and the specific heat for $CeCoIn_5$ in the case of $H_0 \parallel c$. The schematic phase diagram for $CeCoIn_5$ is superimposed in the top-left panel, where a field-induced (H-I) phase is apparent just below $H_{c2}(0)$ [6]. To show the fitting results, the data and fitted curves are plotted for (d) the normalized nuclear relaxation rates $(T_1T)^{-1}$ by $(\gamma_n A)^2$ which are subtracted by the values for $LaCoIn_5$ [19] and (e) the magnetic specific heat C_m/T under several fields along c axis. The data for specific heat are taken from Ref. [13]. (f) The linear thermal expansion coefficient α, from Ref. [14], and SCR curves drawn by using the same parameters obtained from NMR data at 50 and 80 kOe.

the 2D-AFM SCR model in $CeCoIn_5$. Moreover, because the sharp decrease of α 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 is also excluded in recent measurements 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 ξ/a, which can be calculated in units of the in-plane lattice parameter a from $(\sqrt{4\pi y})^{-1}$. As shown in Fig. 3(a), ξ/a at 50 kOe is ≥ 3 at the lowest T, while it is only $\sim$1.4 at 80 kOe. In $CeRhIn_5$, ξ/a is estimated to be $\sim$5 just above the Néel temperature $T_N = 3.8$ K [25]. Similarly, Cd-doped $CeCoIn_5$ induces long-range AFM order where $\xi/a \sim 4$ [26]. Therefore, ξ/a at 50 kOe, Fig. 3(a), indicates that $CeCoIn_5$ 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

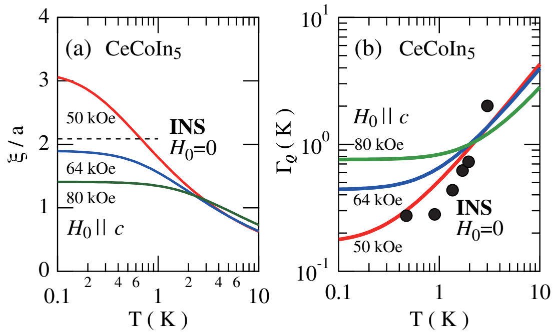

FIG. 3 (color online). Temperature dependence of (a) magnetic correlation length ξ/a and (b) spin fluctuation energy Γ_Q derived from a SCR analysis of $CeCoIn_5$ at 50, 64, and 80 kOe. The broken line in (a) indicates an estimate from inelastic neutron scattering (INS) at zero field. The closed circles in (b) are Γ at $\boldsymbol{Q} = (1/2, 1/2, 1/2)$ obtained by INS at zero field. These data are divided by $\sqrt{3}$ to compare the in-plane component.

confirmed by the out-of-plane component of $\xi_c/c \sim 0.87$ from INS, i.e., $\xi_a/\xi_c = 2.4/(c/a)$ with the lattice anisotropy of $c/a \simeq 1.6$ in $CeCoIn_5$. Parameters derived from fits to the SCR model also give the characteristic spin fluctuation energy Γ_Q, computed from $2\pi T_0 y$. As seen in Fig. 3(b), this Γ_Q agrees well with that obtained from INS [27]. Though Γ_Q shows no apparent field dependence above ~2 K, the energy scale of magnetic excitations decreases below ~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 ~1 K in $CeCoIn_5$ and that a magnetic field finely tunes this scale to order ~0.1 K.

In conclusion, we have demonstrated from microscopic measurements that the field-induced QCP in $CeCoIn_5$, for $H_0 \parallel c$, 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 $H_0 \parallel a$ 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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