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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Magnetic-field-induced enhancement 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=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 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 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 $(T_1T)^{-1}$ 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 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

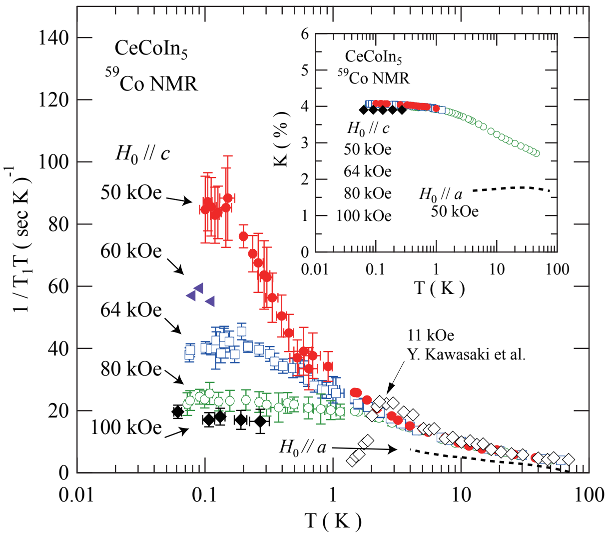

FIG. 1. $(T_1T)^{-1}$ for ^{59}Co NMR at several fields along the c-axis of CeCoIn$_5$. The broken lines are the data under $H_0 = 50$ kOe along the a axis. The results for 11 kOe along the c axis are taken from Ref. [17]. The inset shows the temperature dependence of Knight shifts (K) for ^{59}Co NMR at several fields along the c-axis of CeCoIn$_5$.

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 approaching field to $H_{c2}(0)$ 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 a single dynamical susceptibility is relevant near to the QCP. Then, $(T_1T)^{-1}$ is written as

$$(T_1T)^{-1} = \frac{k_\mathrm{B}}{(\gamma_\mathrm{e}\hbar)^2} \cdot 2(\gamma_\mathrm{n}A_\perp)^2 \sum_{\boldsymbol{q}} f_\perp^2(\boldsymbol{q}) \frac{\mathrm{Im}\chi_\perp(\boldsymbol{q},\omega_0)}{\omega_0}, \tag{1}$$

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 [18]. 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 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 $\chi_\mathrm{RPA}(\boldsymbol{q},\omega) = \chi_0(\boldsymbol{q},\omega)/\{1-\alpha_{\boldsymbol{q}}[\chi_0(\boldsymbol{q},\omega)/\chi_0(\boldsymbol{Q},\omega)]\}$, where $\chi_0(\boldsymbol{q},\omega)$ is the dynamical susceptibility of noninteracting quasi-particles and $\alpha_{\boldsymbol{q}}$ is an enhancement factor. $\chi_0(\boldsymbol{q},\omega)$ gives the well-known Korringa relation $T_1TK_\mathrm{s}^2 = (\hbar/4\pi k_\mathrm{B})(\gamma_\mathrm{e}/\gamma_\mathrm{n})^2 \equiv S$, with K_s being the spin part of K. Using $K_\mathrm{s} \propto (1-\alpha_{\boldsymbol{q}})^{-1}$, the modified Korringa relation for $\chi_\mathrm{RPA}(\boldsymbol{q},\omega)$ is obtained as $T_1TK_\mathrm{s}^2 = nSK(\alpha_{\boldsymbol{q}})^{-1}$, 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 [19]. To deduce the $4f$ electronic component, non-interactive electronic and lattice terms are subtracted by the value of $(T_1T)^{-1}$ for LaCoIn$_5$ [17]. 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 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 $K_{0,i}$ [18]. At 50 kOe, from this modified Korringa relation, the in-plane component of $K(\alpha_{\boldsymbol{q}})$ is found to increase rapidly as $T \to 0$, and 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 $(T_1T)_c^{-1}$ 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 $y_1 \equiv 2J_Q/(\pi^2T_A)$

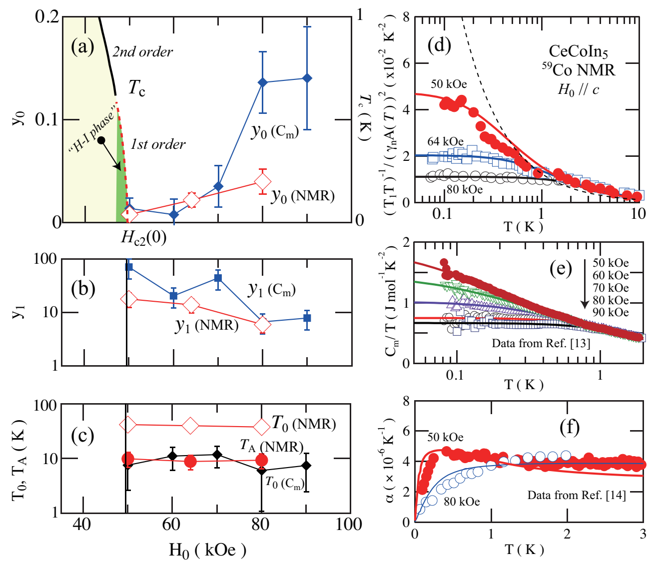

FIG. 2. Field dependence of (a) y_0, (b) y_1, (c) T_0 and T_A obtained from the SCR analysis of ^{59}Co NMR and the specific heat for CeCoIn$_5$ in the case of $H_0 \parallel c$. The schematic phase diagram for CeCoIn$_5$ is superimposed in the top-left panel, where a field-induced (H-I) phase is apparent just below $H_{c2}(0)$ [6]. To show the fitting results, the data and fitted curves for (d) the normalized nuclear relaxation rates $(T_1T)^{-1}$ by $(\gamma_n A)^2$ which are subtracted by the values for LaCoIn$_5$ [17], 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 kOe and 80 kOe.

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 $(2T_0)^{-1}\ln(1 + 1/y)(T \ll T_0)$, 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 three-dimensional (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 Fig. 2(d) and (e). The obtained SCR parameters are plotted against magnetic field in Fig. 2(a), (b), and (c). T_0 and T_A for CeCoIn$_5$ are about 40 K 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 DOS 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$_2$Si$_2$ and CeCu$_{5.9}$Au$_{0.1}$ [23]. Due to 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 approaches zero near $H_{\mathrm{c2}}(0)$.

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 kOe 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 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 also is excluded in recent measurement of volume thermal expansion [15]. Collectively, these results show that, as H_0 approaches $H_{\mathrm{c2}}(0)$ 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 $\geq$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 $T_{\mathrm{N}} = 3.8$ 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 $H_{\mathrm{c2}}(T \to 0)$. Our estimate is close to the zero-field value of ξ/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 $\xi_c/c \sim 0.87$ from INS, *i.e.*, $\xi_a/\xi_c = 2.4/(c/a)$ with the lattice anisotropy of $c/a \simeq 1.6$ in CeCoIn$_5$. Pa-

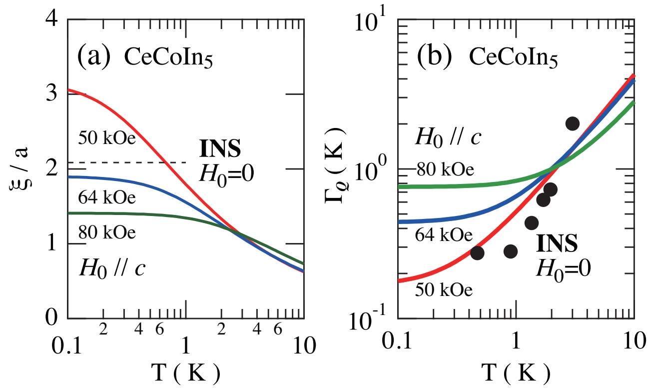

FIG. 3. Temperature dependence of (a) magnetic correlation length ξ/a and (b) spin fluctuation energy Γ_Q derived from an SCR analysis of $CeCoIn_5$ at 50 kOe, 64 kOe, and 80 kOe. The broken line in (a) indicates an estimate from inelastic neutron scattering (INS) at zero field. The closed circles in (b) are 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.

rameters 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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