

^{29}Si nuclear spin-lattice relaxation in $\text{CePtSi}_{1-x}\text{Ge}_x$ near a magnetic instabilityBen-Li Young,^{1,2,*} D. E. MacLaughlin,¹ M. S. Rose,¹ K. Ishida,^{1,3,†} O. O. Bernal,⁴ H. G. Lukefahr,⁵ K. Heuser,⁶ and G. R. Stewart⁷¹*Department of Physics, University of California, Riverside, California 92521-0413, USA*²*Condensed Matter and Thermal Physics, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*³*Department of Physical Science, Graduate School of Engineering Science, Osaka University, Osaka 560-8531, Japan*⁴*Department of Physics and Astronomy, California State University, Los Angeles, California 90032-8206, USA*⁵*Whittier College, Whittier, California 90608, USA*⁶*Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany*⁷*Department of Physics, University of Florida, Gainesville, Florida 32611, USA*

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^{29}Si nuclear spin-lattice relaxation measurements have been performed in the heavy-fermion alloys $\text{CePtSi}_{1-x}\text{Ge}_x$, $x=0$ and 0.1 , in order to study spin dynamics near a magnetic instability. The spin-relaxation curves for both $x=0$ and $x=0.1$ are found to fit to a stretched exponential slightly better than a single exponential function, which suggests that the relaxation rates are inhomogeneous to some extent, due to structural disorder. Within experimental resolution, the relaxation curve is in agreement with the theoretical curve calculated from the Kondo-disorder model. The temperature-dependent relaxation rate $1/T_1$ follows the Korringa relation in CePtSi below 4 K, but not in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. This means that Fermi-liquid and non-Fermi-liquid excitations appear in $x=0$ and $x=0.1$ samples, respectively, which agrees with the results from specific heat experiments. The effective moments of Ce^{3+} ions in $\text{CePtSi}_{1-x}\text{Ge}_x$ for directions parallel and perpendicular to the c axis, calculated from the crystalline electric field (CEF) levels, explain the anisotropy in the observed susceptibilities, hyperfine coupling constants, and spin-lattice relaxation rates. The magnitude of the CEF-corrected Korringa products for the two samples shows the same order as the expected value for a Fermi gas, which indicates that no obvious spin-correlated fluctuations or magnetic order are present. This seems to disagree with the Griffiths phase model, in which magnetic clusters are spatially-extended objects containing many spins.

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I. INTRODUCTION

CePtSi is generally considered to be a nonmagnetic heavy-fermion system,^{1,2} although sample-dependent properties attributed to impurities have been observed.^{3,4} The substitution of Si by Ge alters the hybridization between $4f$ and conduction electrons, and tunes the system toward an anti-ferromagnetic quantum critical point (QCP).^{5,6} Specific heat experiments at low temperatures⁵ have revealed typical Fermi-liquid (FL) behavior (constant specific heat coefficient $\gamma_0 = C/T \sim 800$ mJ/mol K²) in CePtSi , but non-Fermi-liquid (NFL) behavior $C/T \propto -\ln T$ in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. Like many other NFL systems in the neighborhood of a QCP,⁷ $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ exhibits unusual electronic ground-state properties. Quantum criticality has been intensively studied in the similar NFL compound $\text{CeCu}_{6-x}\text{Au}_x$,⁸ where a NFL state also appears at a critical concentration $x=0.1$. It is believed that unusual spin fluctuations at a $T=0$ quantum phase transition (QPT) give rise to NFL behavior in this system.⁹

Nuclear magnetic resonance (NMR) is a local probe of spin, and therefore is very helpful to understand spin dynamics in a NFL state. $\text{CePtSi}_{1-x}\text{Ge}_x$ has a very similar phase diagram¹⁰ to that of $\text{CeCu}_{6-x}\text{Au}_x$,⁸ but Cu (nuclear spin $I=3/2$) NMR in $\text{CeCu}_{6-x}\text{Au}_x$ is complicated by quadrupolar splitting, two isotopes (^{63}Cu and ^{65}Cu), and signals from inequivalent Cu sites in the unit cell. ^{29}Si is a spin $I=1/2$ nucleus (no quadrupole moment), and all ^{29}Si sites in the

tetragonal unit cell of $\text{CePtSi}_{1-x}\text{Ge}_x$ are equivalent,¹ so that ^{29}Si NMR spin relaxation data from this system can be analyzed easily and accurately. Our ^{29}Si NMR linewidth analysis in CePtSi and $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ (Ref. 11) revealed that a spatially inhomogeneous distribution of the f local-moment susceptibility develops at low temperatures in both materials. Disorder-driven NFL models^{12,13} were able to account for the increased low-temperature ^{29}Si linewidth.

^{29}Si NMR spin-lattice relaxation experiments have been carried out in CePtSi and $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. In order to investigate further the effect of disorder on the spin fluctuations, the possibility of the inhomogeneous relaxation in the two samples is considered in this article. Several important features were observed, which may be summarized as follows.

First, the relaxation rates are spatially inhomogeneous in both samples, because the relaxation functions are found to exhibit upward curvature on a semi-logarithmic plot.¹⁴ A theoretical relaxation function based on the Kondo-disorder model¹² fits the experimental results within the experimental resolution.

Second, the spin-lattice relaxation rate $1/T_1$ at low temperatures exhibits Korringa behavior ($T_1 T = \text{const}$) in CePtSi , but not in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. This result is consistent with the Fermi-liquid and non-Fermi-liquid excitations observed in specific heat experiments⁵ for $x=0$ and $x=0.1$, respectively. The behavior of $1/T_1$ in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ is not in agreement with the predicted power laws ($1/T_1 \sim T^{1/3}$ and $1/T_1 \sim T^{1/4}$)

or $1/T_1 = \text{const}$ in QCP-based theories.¹⁵⁻¹⁷ We obtain instead $1/T_1 \sim T^{0.7-0.73}$ at low temperatures.

Third, anisotropy in $1/T_1$ and the transferred hyperfine coupling constant A is observed in both CePtSi and CePtSi_{0.9}Ge_{0.1}. The strong Ce³⁺ crystalline electric field (CEF) level splitting in Ce³⁺ ions is the probable origin of this anisotropy.

Fourth, anisotropy in the Korringa products K^2T_1T , where K is the ²⁹Si Knight shift, and deviations from the free-electron Korringa product could also be explained by taking into account the effective moment μ_{eff} calculated for the Ce³⁺ CEF ground states.¹⁸ The Korringa products after CEF μ_{eff} correction are the same order of magnitude as the value expected for a Fermi gas, which suggests that neither the magnetic order nor spin correlation have a strong effect on relaxation behavior in the two materials.

Last, an anomaly in the magnetic field dependence of $1/T_1$ is observed in CePtSi_{0.9}Ge_{0.1} around 2.7 Tesla, but not in CePtSi. However, the mechanism for this field dependence is not clear at present.

The article is organized as follows. Experimental techniques and procedures are briefly described in Sec. II. Various aspects of the experimental results are discussed in Sec. III. Anisotropy and a comparison of $1/T_1$ with QCP-based theories are discussed in Sec. IV. Finally, in Sec. V we summarize our conclusions.

II. EXPERIMENTAL DESCRIPTION

²⁹Si spin-lattice relaxation experiments were performed in field-aligned powder samples^{11,19} of CePtSi and CePtSi_{0.9}Ge_{0.1} at a NMR frequency of 25.5 MHz (~ 3 Tesla) over the temperature range from 1.4 to 20 K. The samples are the same ones used previously for the measurements of susceptibility and NMR spectrum experiments as described in Ref. 11. Relaxation functions were measured for applied fields \mathbf{H}_0 perpendicular and parallel to the tetragonal c axis. The field dependence of the spin-lattice relaxation was also measured in both samples at 2.1 K for fields $\mathbf{H}_0 \parallel c$ between 1.8 and 4.8 Tesla. A standard NMR nuclear spin saturation-recovery technique, using either a single rf pulse or a short comb of rf pulses, was used for the spin-lattice relaxation measurements.²⁰ The number of saturating rf pulses and the pulse width were found not to modify the nuclear magnetization recovery curve significantly. In order to ensure irradiating the NMR lines uniformly, but to avoid sample heating, a moderate number (3) of saturation pulses was used. Alternating 180° phase-flipped $\pi/2$ - and π -pulse spin echo sequences²⁰ were used to cancel any spurious signals from the rf pulses.

III. EXPERIMENTAL RESULTS

A. Spin-lattice relaxation functions

In a spin-lattice relaxation process the NMR probe nucleus is relaxed by the fluctuation of local magnetic fields, which in paramagnetic solids commonly arise from Fermi contact, dipolar, orbital, and transferred hyperfine interactions between nuclear and electron spins.²¹ In heavy-fermion

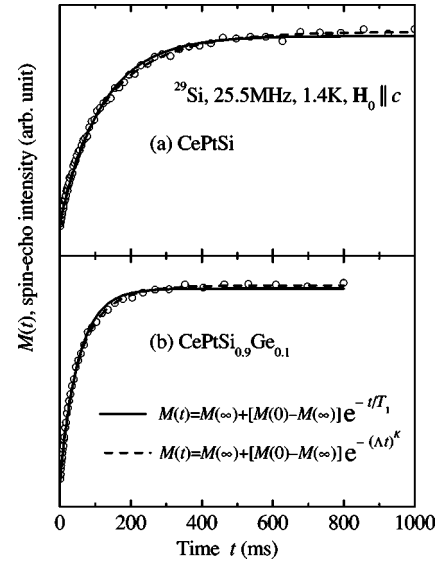


FIG. 1. ²⁹Si nuclear magnetization recovery curves $M(t)$ (circles) for (a) CePtSi and (b) CePtSi_{0.9}Ge_{0.1} measured at 1.4 K with the NMR field $\mathbf{H}_0 \parallel c$ axis at a frequency of 25.5 MHz. Solid curves: exponential fits. Dashed curves: stretched-exponential fits.

systems, fluctuations of the $4f$ or $5f$ electrons usually dominate the relaxation. In the motional narrowing limit, nuclear spins are expected to relax exponentially in a uniform system, even though several relaxation mechanisms are often involved.²²

In a system with magnetic disorder (e.g., a spin glass), spin fluctuation rates are expected to vary randomly over the material, so that the nuclear spin-lattice relaxation rate is distributed. The nuclear magnetization recovery function $M(t)$ then is the spatial average result of the local relaxation functions, i.e., $M(t)$ recovers after saturation according to²³

$$1 - \frac{M(t)}{M(\infty)} = \int_0^\infty P(1/T_1) e^{-t/T_1} d(1/T_1), \quad (1)$$

where $P(1/T_1)$ is the distribution function for the relaxation rate $1/T_1$. Instead of a single exponential $\exp(-t/T_1)$ for a uniform system, the sample-averaged relaxation function, therefore, tends to have a sub-exponential form (an upward curvature in a semi-log plot). Often this behavior can be parametrized by a stretched exponential form $\exp[-(\Lambda t)^K]$, where $K < 1$ characterizes the spread in relaxation rates and Λ is a characteristic relaxation rate.¹⁴ ($1/\Lambda$ is the time for the nuclear magnetization to relax to $1/e$ of its initial value.) For example, spin-lattice relaxation via dilute paramagnetic impurities with no spin diffusion in the nuclear spin system exhibits $\exp[-(\Lambda t)^{1/2}]$ behavior.²⁴

Figure 1 shows typical ²⁹Si relaxation curves (circles) in CePtSi and CePtSi_{0.9}Ge_{0.1} at 1.4 K for $\mathbf{H}_0 \parallel c$ at a frequency of 25.5 MHz. The curves are fit to single exponential [$\propto \exp(-t/T_1)$, solid curve] and stretched-exponential [$\propto \exp[-(\Lambda t)^K]$, dashed curve] functions, respectively. The stretched-exponential function is found to fit the data slightly better. At this temperature, the exponent $K \approx 0.85$ is obtained for both samples, which indicates a resolved but relatively

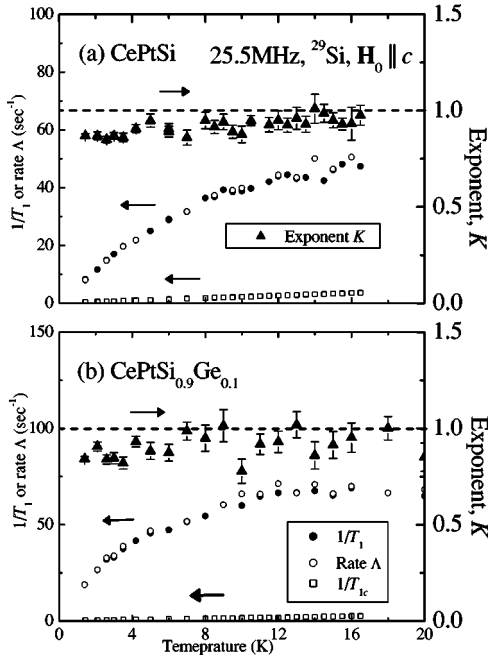


FIG. 2. Temperature dependence of the exponent $K(T)$ (triangles), relaxation rates Λ (open circles), and $1/T_1$ (filled circles) in (a) CePtSi and (b) CePtSi_{0.9}Ge_{0.1} for the $\mathbf{H}_0 \parallel \mathbf{c}$ axis at 25.5 MHz. Rectangles: estimated $1/T_{1c}$ (see the text).

narrow distribution of the relaxation rates. Since the bandwidth of the NMR rf pulses used to saturate the nuclear spins covers most of the spectrum linewidth for $\mathbf{H}_0 \parallel \mathbf{c}$, the effects of cross relaxation due to incomplete saturation, also known to relax nuclear spins nonexponentially,²⁰ are minimized.

The temperature dependencies of $K(T)$, $\Lambda(T)$, and $1/T_1$ are given in Fig. 2. The sub-exponential rate $\Lambda(T)$ (open circles) is found to be nearly the same as the exponential rate $1/T_1$ (filled circles). The exponent $K(T)$ (triangles) is close to unity at high temperatures, and decreases slightly at low temperatures. This suggests that inhomogeneous relaxation develops gradually at low temperatures in both materials, but does not dominate the form of the relaxation function.

In our previous NMR linewidth study,¹¹ strongly enhanced line broadening at low temperatures gave evidence for a magnetic disorder in the static magnetic susceptibility in both samples. In order to investigate the effects of inhomogeneity on the relaxation rate, we have compared the relaxation curves with the observed NMR linewidth as described below.

Since the NMR Larmor frequency ω (\sim MHz) is much smaller than the f -electron spin fluctuation rate Γ (\sim THz), the nuclear spin-lattice relaxation rate due to a single f ion has the form²⁵

$$1/T_1 \propto TA^2\chi/\Gamma, \quad (2)$$

where A is the hyperfine coupling constant and χ is the static susceptibility. This relation suggests that any magnetic disorder in the local susceptibility can result in an inhomogeneous relaxation rate. If the distribution function $P(1/T_1)$ of the relaxation rate is known, the ensemble average of the relax-

ation curve then can be calculated via Eq. (1).

The nuclear spin, in general, is coupled to several f spins, so that, if there is no correlation between the fluctuations of these spins, the relaxation rate $W \equiv 1/T_1$ is a sum over the individual contributions w_i , i.e., $W = \sum_i w_i$. The derivation of the distribution function $P(W)$ for the relaxation rate is similar to the solution of the random walk problem, and gives the expression^{11,26}

$$P(W) = \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} P_n(w_1, w_2, \dots, w_n) \times \delta\left(W - \sum_i w_i\right) dw_1 dw_2 \dots dw_n = \int_{-\infty}^{\infty} d\tau e^{2\pi i W \tau} \left(\int_{-\infty}^{\infty} P(w) e^{-2\pi i w \tau} dw \right)^n, \quad (3)$$

where $P(w)$ describes the distribution of the individual contributions and n is the number of near-neighbor f ions. Again no correlation is assumed, so that the distribution function $P_n(w_1, w_2, \dots, w_n)$ becomes the product of the $P(w_i)$: $P_n(w_1, w_2, \dots, w_n) = P(w_1)P(w_2) \cdots P(w_n) = [P(w)]^n$.

In single-ion Kondo physics, the local nuclear spin-lattice relaxation rate w has the form²⁷

$$w \equiv T_1^{-1}(T; T_K) = \begin{cases} C_1 T/T_K^2, & T < T_K, \\ C_2 \sqrt{T_K/T}, & T > T_K, \end{cases} \quad (4)$$

where C_1 and C_2 are temperature-independent constants. The above relation can be easily obtained from Eq. (2), if the approximations $\Gamma \propto 1/\chi \sim T_K$ for $T < T_K$, and $\Gamma(T) \propto \sqrt{T/T_K}$ (Ref. 28) and $\chi \sim 1/T$ for $T > T_K$ are made. We set $C_2/C_1 = 1/T_K$ to ensure the continuity of T_1^{-1} at $T = T_K$.

In the Kondo-disorder model, T_K is distributed, so that w is distributed. Therefore, the distribution function $P(W)$ in Eq. (3) can be calculated in a similar fashion as the Knight shift distribution described in Ref. 11, where the distributions of the Kondo temperatures $P(T_K)$ in CePtSi and CePtSi_{0.9}Ge_{0.1} have been estimated from the susceptibility data.

Figure 3(a) shows an example of the predicted $P(W)$ from the Kondo-disorder model for ²⁹Si NMR in CePtSi_{0.9}Ge_{0.1} at 3.5 K and 25.5 MHz with the NMR field $\mathbf{H}_0 \parallel \mathbf{c}$. The calculated spin-lattice relaxation curves from Eq. (1) for the Kondo-disorder model is given in Fig. 3(b). Often it is difficult to resolve the NMR long-time relaxation tails after the nuclear magnetization has relaxed to $\sim 10^{-1} - 10^{-2}$ of its initial value. Within experimental resolution, however, we find that the experimental data (circles) agree with the model curve (solid curve).

In the calculation of theoretical relaxation curves, we made the important assumption that the spin fluctuations are not spatially correlated. Indeed, from our $1/T_1$ analysis, we conclude that spatial correlation is not significant in CePtSi and CePtSi_{0.9}Ge_{0.1} (see Sec. III D).

In an inhomogeneous spin environment, spin diffusion between the slowly- and rapidly-relaxed nuclear spins may take place in addition to the spin-lattice relaxation process, and modify the relaxation curve.²⁹ This spin diffusion effect is

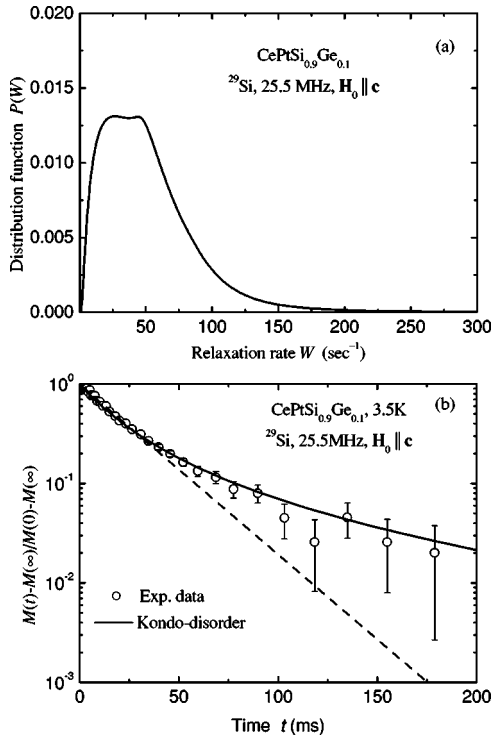


FIG. 3. (a) The distribution function $P(W)$ of ^{29}Si nuclear spin-lattice relaxation rates W for $\mathbf{H}_0 \parallel \mathbf{c}$ at 3.5 K and 25.5 MHz predicted by the Kondo-disorder model. (b) A comparison of the relaxation curves from the experiment (circles) and Kondo-disorder model calculation (solid line). Dashed line denotes the single exponential function.

usually not easy to characterize experimentally, so that it is often difficult to separate the f -spin relaxation mechanism from diffusion effects. This problem seems not to be important in our samples, however, because the ratio of the diffusion length L to the lattice spacing d , i.e., $L/d \sim \sqrt{T_1/50T_2}$, where T_2 is the transverse relaxation time,³⁰ is found to be smaller than unity. Thus spin diffusion does not change the inhomogeneous nuclear magnetization appreciably during a time T_1 . The low natural abundance (5%) of the ^{29}Si nuclei is also unfavorable to spin diffusion. We conclude that relaxation behavior in CePtSi and CePtSi_{0.9}Ge_{0.1} is consistent with the disorder models.

Muon spin relaxation experiments for CePtSi and CePtSi_{0.9}Ge_{0.1} have been reported in Ref. 31, in which stretched exponential behavior for muon decay was observed in both samples. This gives us an independent evidence of the existence of magnetic disorder.

B. Relaxation rate $1/T_1$

In Sec. III A we have seen that a stretched-exponential function describes the low-temperature relaxation curves better in both CePtSi and CePtSi_{0.9}Ge_{0.1}, and a distribution of the spin relaxation rates is inferred. Therefore, the use of an average relaxation rate rather than a single rate is more appropriate for the discussion. In general, the initial slope of a relaxation curve defines the average relaxation rate, which can be understood by taking the time derivative in Eq. (1).

Since the exponent K obtained from the stretched-exponential fit is not too far from unity in the two samples, the difference between the initial slopes from the experimental relaxation curve (or stretched exponential) and single exponential function are minor. Therefore, the exponential relaxation rate $1/T_1$ shown in Fig. 2 will be taken as the average value in the following discussion.

As mentioned in the previous section, several mechanisms are usually involved in a nuclear spin-lattice relaxation process. If there is no correlation in these relaxation mechanisms, the spin-relaxation rate is expected to be the sum of the individual rates due to each mechanism. In a heavy-fermion system, $1/T_1$, in general, can be written as $1/T_1 = 1/T_{1c} + 1/T_{1f}$, where $1/T_{1c}$ is the relaxation rate due to the fluctuations of conduction electrons (via the Fermi contact interaction), and $1/T_{1f}$ is the contribution from f -electron local moments (via the transferred hyperfine interaction)³² (except for systems with light nuclei such as ^9Be in CeBe₁₃ and UBe₁₃, where the dipolar interaction is also important²¹). In concentrated f -electron systems, conduction electrons often relax nuclear spins slowly at low temperatures, so that the spin-lattice relaxation is dominated by the f spins.³³

In metals, the conduction-electron relaxation rate $1/T_{1c}$ obeys the relation³⁴

$$1/T_{1c} \propto N(E_F)^2 T, \quad (5)$$

where $N(E_F)$ is the conduction-electron density of states at Fermi energy. The product $(T_{1c}T)^{-1}$ is proportional to $N(E_F)^2$, and is independent of temperature. In a free-electron metal, $1/T_{1c}$ is related to the Knight shift K_0 due to the conduction electrons by the Korringa relation,

$$K_0^2 T_{1c} T = (\hbar/4\pi k_B)(\gamma_e/\gamma_n)^2 \equiv S_0, \quad (6)$$

where γ_e and γ_n are the gyromagnetic ratios of the electron and the NMR nucleus, respectively. However, in systems with strong electron-electron interactions, spatial spin correlations, magnetic order, or other effects (e.g., spin-orbital interactions and CEF effects), deviations from the free-electron Korringa product S_0 are often seen, because such effects may affect the Knight shift and $1/T_1$ differently.^{35,36}

In strongly-correlated f -electron systems at low temperatures, if the f moments are completely quenched by the Kondo effect and the system enters a Fermi-liquid state, the nuclear spin then relaxes to the quasiparticles near the Fermi surface. The Korringa relation is also expected, except that the Korringa product may be enhanced.¹⁸ Kuramoto and Müller-Hartmann³⁷ have given a theoretical proof of the Korringa relation in heavy-fermion systems at low temperatures as long as interactions between f moments are negligible.

An estimate of $1/T_{1c}$ can be obtained from the Korringa product S_0 if K_0 is known. The intercept in a plot of Knight shift K versus susceptibility χ (with temperature an implicit parameter) can give a good estimate of K_0 , because K_0 is independent of temperature. The estimated $1/T_{1c}$ in CePtSi and CePtSi_{0.9}Ge_{0.1} is shown in Fig. 2 (rectangles). As expected, $1/T_{1c}$ is small enough compared to $1/T_1$ so that it can be ignored in this temperature range, and the observed relaxation rate $1/T_1$ is a good measure of $1/T_{1f}$.

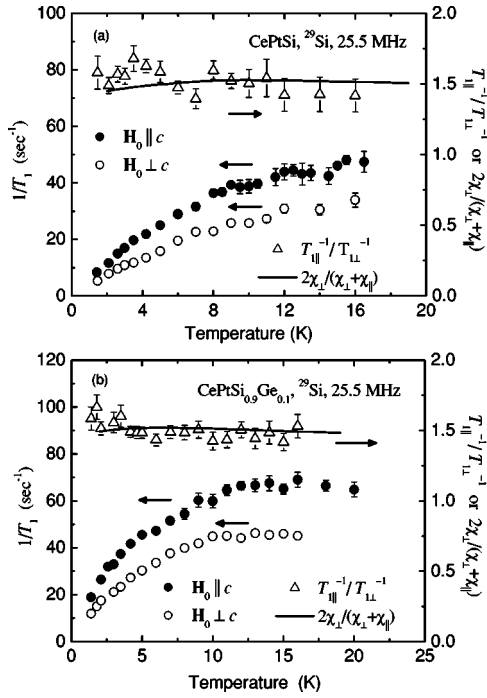


FIG. 4. Spin-lattice relaxation rates ($1/T_1$) measured in (a) CePtSi and (b) CePtSi_{0.9}Ge_{0.1}, for the NMR fields $\mathbf{H}_0 \parallel c$ (filled circles) and $\mathbf{H}_0 \perp c$ (open circles). Triangles: ratio of $T_{1\parallel}^{-1}$ to $T_{1\perp}^{-1}$. Curve: ratio of $2\chi_{\perp}$ to $(\chi_{\perp} + \chi_{\parallel})$, where χ_{\perp} and χ_{\parallel} are the ab -plane and c -axis susceptibilities, respectively.

The temperature dependence of $1/T_1$ for the NMR fields $\mathbf{H}_0 \parallel c$ (filled circles) and $\mathbf{H}_0 \perp c$ (open circles) is shown in Figs. 4(a) and 4(b) for CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively. The relaxation rate $1/T_{1\parallel}$ for $\mathbf{H}_0 \parallel c$ is found to be greater than the relaxation rate $1/T_{1\perp}$ for $\mathbf{H}_0 \perp c$. Nevertheless, $1/T_{1\parallel}$ and $1/T_{1\perp}$ have a similar temperature dependence in the two samples. They both decrease with T at low temperatures, and extrapolate to zero at $T=0$. The saturation of $1/T_1$ around 15 K is an indication of the breakup of the Kondo resonance; at higher temperatures the f spins are weakly interacting with the conduction electrons. These features have been seen in many other heavy-fermion compounds.³⁸

Figures 4(a) and 4(b) also reveal that CePtSi_{0.9}Ge_{0.1} exhibits a faster ²⁹Si relaxation rate than CePtSi. $1/T_1$ of ⁶³Cu in CeCu_{6-x}Au_x also shows a greater value for a larger doping x .³⁹ This may be understandable from the disorder enhanced $1/T_1$ as proposed by Shastry and Abrahams.⁴⁰

In the plot of the temperature dependence of $(T_1 T)^{-1}$ given in Fig. 5, we find that CePtSi and CePtSi_{0.9}Ge_{0.1} exhibit different behavior at low temperatures. For CePtSi, $(T_1 T)^{-1}$ reaches a constant below 3 K for both directions of \mathbf{H}_0 ; this is Korringa relaxation and indicates FL behavior. As for CePtSi_{0.9}Ge_{0.1}, a continuing increase of $(T_1 T)^{-1}$ with decreasing temperature is seen at the lowest temperature for both field directions, which obviously does not follow the Korringa relation and is not expected for a FL state. Although this may simply reflect the reduction of T_K as the QCP is approached (which would also explain the faster rate noted above), it might be due to at least in part to the devel-

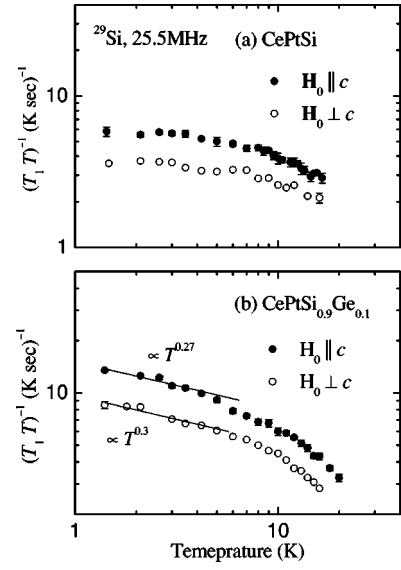


FIG. 5. Plots of $1/(T_1 T)$ versus temperature for (a) CePtSi and (b) CePtSi_{0.9}Ge_{0.1}. Filled circles: $\mathbf{H}_0 \parallel c$. Open circles: $\mathbf{H}_0 \perp c$. Solid lines are fits to the power law at low temperatures.

opment of NFL fluctuations. We delay a further discussion to Sec. IV. The FL and NFL behavior observed in these spin relaxation data for CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively, are consistent with the results from the specific heat experiments.^{5,6}

C. Anisotropic spin-lattice relaxation

From the discussion of Sec. III B, we have seen that $1/T_1$ is anisotropic in CePtSi and CePtSi_{0.9}Ge_{0.1}. Surprisingly, the ratio $T_{1\parallel}^{-1}/T_{1\perp}^{-1}$ (triangles) is found to be nearly temperature independent, and approximately coincides with the ratio $2\chi_{\perp}/(\chi_{\perp} + \chi_{\parallel}) \approx 1.5$ (curves) as shown in Fig. 4 for the two materials, where χ_{\perp} and χ_{\parallel} are the ab -plane and c -axis susceptibilities, respectively. The reason for comparing $T_{1\parallel}^{-1}/T_{1\perp}^{-1}$ with $2\chi_{\perp}/(\chi_{\perp} + \chi_{\parallel})$ is that the nuclear relaxation rate is dominated by spin fluctuations perpendicular to the NMR field, which in turn are related to the susceptibility in that direction.⁴¹

In order to investigate this anisotropy further, frequency shift measurements have been carried out in CePtSi and CePtSi_{0.9}Ge_{0.1} for $\mathbf{H}_0 \parallel$ and $\perp c$. The frequency shifts determined from the center peak of ²⁹Si NMR spectra are plotted against the magnetic susceptibility (with temperature an implicit parameter) in Fig. 6.

We find that the frequency shift curves are linear in the susceptibility at high temperatures (small susceptibilities), but become curved at low temperatures (high susceptibilities). Similar frequency shift anomalies have been observed in other Ce compounds.⁴²⁻⁴⁴ The deviation from the linear relation $K=A\chi$ suggests that the hyperfine coupling is temperature dependent (via CEF effects,⁴⁴ valence fluctuations,⁴⁵ or the onset of Kondo screening⁴⁶).

The slopes of the linear parts of the K - χ plot show strong anisotropy in the hyperfine coupling constants for the two samples, and yield hyperfine coupling constants as follows:

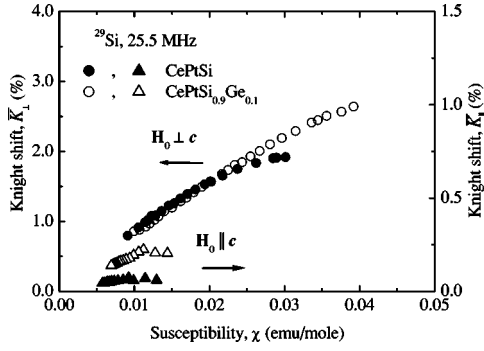


FIG. 6. Plots of ^{29}Si NMR Knight shifts as a function of magnetic susceptibility in CePtSi (filled symbols) and CePtSi $_{0.9}\text{Ge}_{0.1}$ (open symbols). Circles: $\mathbf{H}_0 \perp c$. Triangles: $\mathbf{H}_0 \parallel c$.

$A_{\parallel} \approx 0.42 \pm 0.03 \text{ kOe}/\mu_B$ and $A_{\perp} \approx 3.78 \pm 0.08 \text{ kOe}/\mu_B$ for CePtSi, and $A_{\parallel} \approx 0.98 \pm 0.04 \text{ kOe}/\mu_B$ and $A_{\perp} \approx 3.99 \pm 0.04 \text{ kOe}/\mu_B$ for CePtSi $_{0.9}\text{Ge}_{0.1}$. In heavy-fermion compounds, the frequency shift is dominated by the transferred hyperfine interaction between f electrons and non- f -ion nuclei, which comes from the s - f hybridization and is usually taken to be isotropic. Since the dipolar interaction can be anisotropic, the magnetic dipolar fields between Si and its surrounding Ce $^{3+}$ ions have been calculated in order to estimate their contribution to the anisotropic hyperfine coupling. We find that the coupling constants due to the dipolar interactions are $A_{\parallel, \text{dip}} \approx -0.2 \text{ kOe}/\mu_B$ and $A_{\perp, \text{dip}} \approx 0.36 \text{ kOe}/\mu_B$. Clearly, the dipolar interactions alone are too small (and in one case of the wrong sign) to account for the anisotropy.

Although the spin polarization due to the s - f exchange and RKKY interactions is isotropic according to the models, in practice anisotropy in the transferred hyperfine coupling has been observed in several f -electron compounds, e.g., CeCu $_2\text{Si}_2$ (Ref. 43) and UPt $_3$ (Ref. 47). Since the $4f$ orbital of Ce $^{3+}$ lies inside the $5s$, $5p$, $5d$, and $6s$ orbitals, the transferred hyperfine coupling directly from the s - f hybridization may be less important than from the spin polarization of these outer shells. This non- s contribution is therefore considered to be responsible for the anisotropy.⁴³ On the other hand, Ohama *et al.*⁴³ proposed that the s - f hybridization itself is possibly anisotropic, if the CEF splittings of the conduction electrons overlapping with the f orbital are taken into account. The crystalline electric field is known to be strong in CePtSi and CePtSi $_{0.9}\text{Ge}_{0.1}$, and therefore could be responsible for the hyperfine coupling anisotropy to some extent. In addition, we find that the ratios of $A_{\perp}/A_{\parallel} \sim 9$ for CePtSi and ~ 4 for CePtSi $_{0.9}\text{Ge}_{0.1}$ are close to the ratios of $(\mu_{\text{eff}})_{\perp}/(\mu_{\text{eff}})_{\parallel}$ in the two samples, where $(\mu_{\text{eff}})_{\perp}$ and $(\mu_{\text{eff}})_{\parallel}$ are the effective moments calculated from the CEF ground states for the ab plane and along the c axis, respectively. As discussed in Sec. III D, this behavior is expected in, and seems to support, the CEF scenario.

According to NMR spin-lattice relaxation theory, the relaxation rates $(1/T_1)_{\parallel}$ and $(1/T_1)_{\perp}$ for NMR fields parallel and perpendicular to the crystal c axis, respectively, are given by⁴¹

$$T_{1\parallel}^{-1} = 2\hbar \gamma_N^2 k_B T \sum_i [A_{\perp, i}^2 \chi''_{\perp, i}(\omega)/\omega] \quad (7)$$

and

$$T_{1\perp}^{-1} = \hbar \gamma_N^2 k_B T \sum_i [A_{\perp, i}^2 \chi''_{\perp, i}(\omega)/\omega + A_{\parallel, i}^2 \chi''_{\parallel, i}(\omega)/\omega], \quad (8)$$

where $\chi''(\omega)$ is the imaginary susceptibility. The sums include the contributions from the surrounding f ions near the NMR nucleus under the condition of uncorrelated f -spin fluctuations. Equations (7) and (8) reflect the fact that only the fluctuation fields perpendicular to \mathbf{H}_0 contribute to relaxation. If spin correlation is important, the q dependence of the hyperfine coupling constant $A(q)$ and dynamic susceptibility $\chi''(q, \omega)$ must be considered, and the sums in Eqs. (7) and (8) are often expressed in q space.³⁶

The imaginary part of the dynamic susceptibility is often taken to have the Lorentzian form⁴⁸ $\chi''(\omega) = \chi\omega\Gamma/(\omega^2 + \Gamma^2)$, where Γ is the f -spin fluctuation rate and χ is the static susceptibility. Since the NMR Larmor frequency ω is usually much smaller than the spin fluctuation rate Γ , the dynamic susceptibility can be approximated by $\chi''(\omega) \approx \chi\omega/\Gamma$. Suppose that there are no spatial correlations between f -ion spin fluctuations, the relaxation rates $T_{1\parallel}^{-1}$ and $T_{1\perp}^{-1}$ for $\mathbf{H}_0 \parallel$ and $\perp c$ directions, respectively, are given by⁴¹

$$T_{1\parallel}^{-1} = 2\hbar \gamma_n^2 k_B T \sum_i [A_{\perp, i}^2 \chi_{\perp, i}/\Gamma_{\perp, i}] \quad (9)$$

and

$$T_{1\perp}^{-1} = \hbar \gamma_n^2 k_B T \sum_i [A_{\perp, i}^2 \chi_{\perp, i}/\Gamma_{\perp, i} + A_{\parallel, i}^2 \chi_{\parallel, i}/\Gamma_{\parallel, i}], \quad (10)$$

where χ_i and Γ_i are the static susceptibility and fluctuation rate, respectively, for the i th near-neighbor f spin. In order to simplify the problem, we introduce the effective near-neighbor numbers $(n_{\text{eff}, 1/T_1})_{\alpha}$, $\alpha = \parallel$ and \perp , such that $\sum_i A_{\alpha, i}^2 \chi_{\alpha, i}(T)/\Gamma_{\alpha, i} = (n_{\text{eff}, 1/T_1})_{\alpha} A_{0, \alpha}^2 \chi_{\alpha}(T)/\Gamma_{0, \alpha}$, where $A_{0, \alpha}$ is the average hyperfine coupling constant due to a single f ion, and χ_{α} and $\Gamma_{0, \alpha}$ represent the average susceptibility and spin fluctuation rate, respectively.

If there are no spin correlations and no disorder in χ_i and Γ_i , the effective number $n_{\text{eff}, 1/T_1}$ is expected to be the number n_0 of nearest-neighbor f ions to the relaxing nucleus because the transferred hyperfine coupling is short-ranged. However, $n_{\text{eff}, 1/T_1} \neq n_0$ suggests the presence of spatial correlation or, alternatively, disorder in the f moments. In the case of ferromagnetic fluctuations, $n_{\text{eff}, 1/T_1}$ is expected to be close to n_0^2 , because the spins fluctuate coherently so that the near-neighbor f spins can be thought of as a single giant spin with n_0 times larger value of the coupling constant $A_{\alpha, i}$.

According to Eqs. (9) and (10), the ratio of the anisotropic relaxation rates can be written as

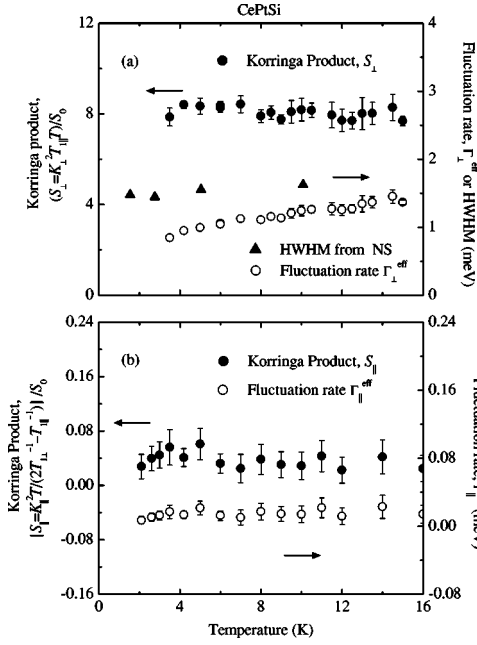


FIG. 7. Temperature dependence of the Korringa products (filled circles) and the fluctuation rates (open circles) for CePtSi in directions of the (a) \perp and (b) \parallel c axis. Triangles: HWHM from quasi-elastic neutron scattering spectra in CePtSi.

$$\frac{T_{1\parallel}^{-1}}{T_{1\perp}^{-1}} = \frac{2(n_{\text{eff},1/T_1})_{\perp} A_{0,\perp}^2 \chi_{\perp} / \Gamma_{0,\perp}}{(n_{\text{eff},1/T_1})_{\perp} A_{0,\perp}^2 \chi_{\perp} / \Gamma_{0,\perp} + (n_{\text{eff},1/T_1})_{\parallel} A_{0,\parallel}^2 \chi_{\parallel} / \Gamma_{0,\parallel}}. \quad (11)$$

Comparing this to our previous result $T_{1\parallel}^{-1}/T_{1\perp}^{-1} \approx 2\chi_{\perp}/(\chi_{\perp} + \chi_{\parallel})$, this suggests that

$$\Gamma_{0,\perp}/\Gamma_{0,\parallel} \approx (n_{\text{eff},1/T_1})_{\perp} A_{0,\perp}^2 / [(n_{\text{eff},1/T_1})_{\parallel} A_{0,\parallel}^2], \quad (12)$$

for CePtSi and CePtSi_{0.9}Ge_{0.1}. The hyperfine couplings are known to be strongly anisotropic in the two samples, i.e., $A_{\perp} > A_{\parallel}$, as mentioned previously. Therefore the spin fluctuations are also anisotropic, and $\Gamma_{0,\perp} > \Gamma_{0,\parallel}$ according to Eq. (12), if no correlation appears in the spin fluctuations so that $(n_{\text{eff},1/T_1})_{\perp} = (n_{\text{eff},1/T_1})_{\parallel}$.

D. The Korringa products

So far we have considered the shape of the relaxation curves, the temperature dependence of $1/T_1$, and the anisotropic relaxation. In order to further investigate the electron interactions or possible spin correlations in CePtSi and CePtSi_{0.9}Ge_{0.1}, the Korringa product $K^2 T_1 T$ is also considered. Figures 7 and 8 show the Korringa products (filled circles) for $\mathbf{H}_0 \parallel \mathbf{c}$ and $\perp \mathbf{c}$ in CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively. These products are expressed in units of the free-electron Korringa product $S_0 \equiv (\hbar/4\pi k_B)(\gamma_e/\gamma_n)^2$ [Eq. (6)]. According to Ref. 49, the definitions of the Korringa products, $S_{\perp} \equiv K_{\perp}^2 T_{1\parallel} T$ and $S_{\parallel} \equiv TK_{\parallel}^2 / (2T_{1\perp}^{-1} - T_{1\parallel}^{-1})$, are appropriate for a discussion of the anisotropy. The denominator $(2T_{1\perp}^{-1} - T_{1\parallel}^{-1})$ in S_{\parallel} can be thought as an effective $T_{1\perp}^{-1}$, which is defined in such a form in order to have the similar definition as $T_{1\parallel}^{-1}$ in Eq. (9).

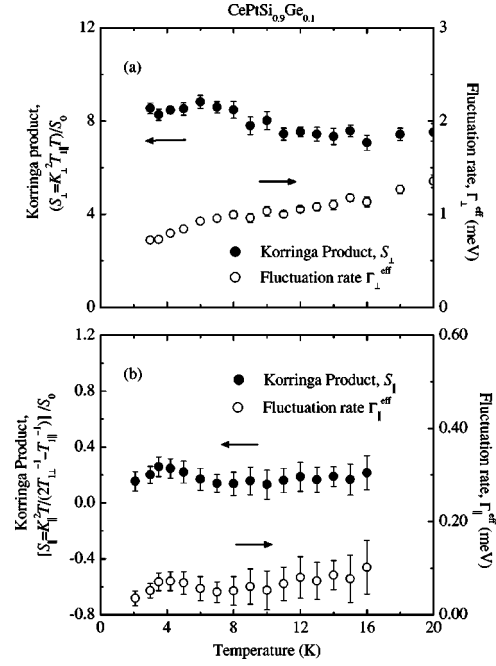


FIG. 8. Temperature dependence of the Korringa products (filled circles) and the fluctuation rates (open circles) for CePtSi_{0.9}Ge_{0.1} in directions of the (a) \perp and (b) \parallel c axis.

We find that CePtSi has a nearly constant value of $S_{\perp} \approx 8S_0$ for $T < 16$ K. CePtSi_{0.9}Ge_{0.1} has a similar S_{\perp} , but it shows a slight increase below 8 K. However, small values of $S_{\parallel} \approx 0.04S_0$ and $S_{\parallel} \approx 0.2S_0$ are obtained for CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively. There are obviously quantitative differences between the observed values and the free-electron Korringa product. Anisotropy again is seen in the Korringa products for the two samples. However, we will see that the effective moment calculated from the CEF ground states explains the magnitude of and anisotropy in the Korringa products.

Kuramoto and Müller-Hartmann³⁷ have proved that the Korringa-Shiba relation⁵⁰ holds in heavy-fermion systems if no interactions exist between the f moments. In cases with orbital degeneracy or crystal-field energy splitting, the Korringa-Shiba relation is generalized to²¹

$$\lim_{\omega \rightarrow 0} \chi_{\alpha}''(\omega)/\omega = 3\pi\chi_{\alpha}^2(0)/[N(\mu_{\text{eff}})_{\alpha}^2] \quad (T \rightarrow 0), \quad (13)$$

where

$$(\mu_{\text{eff}})_{\alpha}^2/\mu_B^2 = 3 \sum_{i,j} |\langle i | g_j J_{\alpha} | j \rangle|^2 / N, \quad \alpha = \parallel \text{ or } \perp. \quad (14)$$

Here μ_{eff} and μ_B represent the effective moment and Bohr magneton, respectively, g_j is the f -ion g factor, and N is the degeneracy of the ground state. The states $|i\rangle$ and $|j\rangle$ are the eigenstates in the ground-state multiplet.

The Knight shift K_{α} is given by $K_{\alpha} = \sum_i A_{\alpha,i} \chi_{\alpha,i}$ ($\alpha = \parallel$ or \perp), summed over individual contributions from near-neighbor f moments. As discussed in Sec. III C, we also introduce an effective number $n_{\text{eff},K}$ such that $K_{\alpha} = (n_{\text{eff},K})_{\alpha} A_{0,\alpha} \chi_{\alpha}$. $n_{\text{eff},K}$ is expected to be close to the number

of near-neighbor f ions but unlike $n_{\text{eff},1/T_1}$, $n_{\text{eff},K}$ is not expected to be affected by the presence of a spin correlation (except in a magnetically ordered phase).

By applying the Korringa-Shiba relation [Eq. (13)] and the Knight shift, it is not difficult to show that Eqs. (7) and (8) give the Korringa products

$$S_{\perp} \equiv K_{\perp}^2 T_{\parallel} T = (n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\perp} \eta_{\perp} S_0, \quad (15)$$

and

$$S_{\parallel} \equiv TK_{\parallel}^2/(2T_{\perp}^{-1} - T_{\parallel}^{-1}) = (n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\parallel} \eta_{\parallel} S_0, \quad (16)$$

where $\eta_{\alpha} \equiv N(\mu_{\text{eff}}^2/\alpha\mu_B^2 = \sum_{i,j} |\langle i|g_J J_{\alpha}|j\rangle|^2/2)$, $\alpha = \perp$ or \parallel . Surprisingly, the effective moment μ_{eff} is involved in the Korringa product. Since there is a tetragonal CEF effect on Ce^{3+} ions in CePtSi and CePtSi_{0.9}Ge_{0.1}, anisotropy is expected for μ_{eff} , which then may be related to the observed anisotropic Korringa products.

The ground states in CePtSi and CePtSi_{0.9}Ge_{0.1} are the Γ_7 doublets¹¹ $|\Gamma_7^{(1)}(\pm)\rangle = a|\pm 5/2\rangle + \sqrt{1-a^2}|\mp 3/2\rangle$, where the constant a is 0.64 for CePtSi and 0.77 ± 0.04 for CePtSi_{0.9}Ge_{0.1}. It should be noted that the uncertainty of a in CePtSi_{0.9}Ge_{0.1} comes from the susceptibility fits (see Ref. 11), whereas a for CePtSi is obtained more accurately from neutron scattering (NS) experiments.²⁸ As $T \rightarrow 0$, the effective moments are found to have the forms

$$(\mu_{\text{eff}})_{\parallel}^2 = 3(8a^2 - 3)^2 g_J^2 \mu_B^2/4, \quad (17)$$

and

$$(\mu_{\text{eff}})_{\perp}^2 = 15a^2(1 - a^2) g_J^2 \mu_B^2. \quad (18)$$

We obtain $(\mu_{\text{eff}})_{\parallel} = 0.2\mu_B$ and $(\mu_{\text{eff}})_{\perp} = 1.63\mu_B$ for CePtSi, and $(\mu_{\text{eff}})_{\parallel} = (1.29 \pm 0.37)\mu_B$ and $(\mu_{\text{eff}})_{\perp} = (1.63 \pm 0.04)\mu_B$ for CePtSi_{0.9}Ge_{0.1}.

After taking μ_{eff} into account and comparing the experimental Korringa products with Eqs. (15) and (16), we find

$$\text{CePtSi: } (n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\parallel} \approx 2.9,$$

$$(n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\perp} \approx 9,$$

and

$$\text{CePtSi}_{0.9}\text{Ge}_{0.1}: (n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\parallel} \approx 0.4 \pm 0.2,$$

$$(n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\perp} \approx 9 \pm 0.01.$$

We assume that $(n_{\text{eff},K})_{\perp} = (n_{\text{eff},K})_{\parallel}$, and these effective numbers are expected to be between 4 and 6, which are the numbers of near-neighbor Ce^{3+} ions within the first and second shells, respectively, around the ²⁹Si site (Ref. 11). However, we recall that this assumption may not be appropriate for $n_{\text{eff},1/T_1}$ if correlation appears in the spin fluctuations. As a consequence, we obtain

$$\text{CePtSi: } (n_{\text{eff},1/T_1})_{\parallel} \approx 5.5 - 12,$$

$$(n_{\text{eff},1/T_1})_{\perp} \approx 1.8 - 4,$$

and

$$\text{CePtSi}_{0.9}\text{Ge}_{0.1}: (n_{\text{eff},1/T_1})_{\parallel} \approx 40 - 90,$$

$$(n_{\text{eff},1/T_1})_{\perp} \approx 1.8 - 4.$$

Within the range of the uncertainties, $(n_{\text{eff},1/T_1})_{\perp}$ is found to be slightly smaller than or equal to the expected near-neighbor number for the two samples. This suggests that no significant spatial correlation exists in the ab -plane spin fluctuations. The values of $(n_{\text{eff},1/T_1})_{\parallel}$ have large uncertainties. In CePtSi, $(n_{\text{eff},1/T_1})_{\parallel}$ varies from close to the expected value to twice as large as the expected value. CePtSi_{0.9}Ge_{0.1}, however, shows an unrealistically large value of $(n_{\text{eff},1/T_1})_{\parallel}$. This makes us suspect that the CEF parameter $a=0.77$ in CePtSi_{0.9}Ge_{0.1}, obtained from fits to susceptibility data, might not be accurate. In order to get a value of $(n_{\text{eff},1/T_1})_{\parallel} \approx 4$, the effective moment $(\mu_{\text{eff}})_{\parallel}$ would have to be $0.39\mu_B$ for CePtSi_{0.9}Ge_{0.1}. In this case, $a=0.663$ from Eq. (17). However, we find that $(\mu_{\text{eff}})_{\perp}$ is still close to $1.63\mu_B$ from Eq. (18). From the derivatives,

$$d(\mu_{\text{eff}})_{\parallel}/da = 8\sqrt{3}g_J\mu_B a, \quad (19)$$

$$d(\mu_{\text{eff}})_{\perp}/da = \sqrt{15}g_J\mu_B(1 - 2a^2)/\sqrt{1 - a^2}, \quad (20)$$

we note that $(\mu_{\text{eff}})_{\parallel}$ is much more sensitive to the CEF parameter a than $(\mu_{\text{eff}})_{\perp}$. Since a was obtained from fits to the ab -plane susceptibility,¹¹ the value might not be determined accurately due to the insensitivity of $(\mu_{\text{eff}})_{\perp}$ to a . Therefore, a value of a for CePtSi_{0.9}Ge_{0.1} closer to 0.663 than 0.77 gives a reasonable explanation of the spin-relaxation results.

Hashi *et al.* have examined the Korringa products in several heavy-fermion compounds, and concluded that the Korringa product in a heavy-fermion system with a nonmagnetic ground state has the same order of magnitude as the Korringa-Shiba value. More than an order of magnitude deviation from the expected value has been attributed to the development of magnetic correlation between the localized moments.¹⁸ On the other hand, Moriya has pointed out that the electron-electron interaction alone is also able to produce an enhanced Korringa product.³⁶

If there is no correlation in a uniform conduction-electron sea between spins, the effective numbers of near neighbors are expected to be $n_{\text{eff},1/T_1} = n_{\text{eff},K} = 4 - 6$ in CePtSi and CePtSi_{0.9}Ge_{0.1}. By taking the prefactor $(n_{\text{eff},K}^2/n_{\text{eff},1/T_1})_{\alpha} \eta_{\alpha}$ in Eqs. (15) and (16) into account, the theoretical Korringa products are expected to be $S_{\parallel}/S_0 \approx 0.06 - 0.08$ and $S_{\perp}/S_0 \approx 3.6 - 5.3$ for CePtSi, and $S_{\parallel}/S_0 \approx 0.2 - 0.3$ and $S_{\perp}/S_0 \approx 3.6 - 5.3$ for CePtSi_{0.9}Ge_{0.1}. These values are compared with our experimental results in Table I. We find that S_{\perp} and S_{\parallel} are the same order of magnitude as the theoretical values. Small deviations from the theoretical Korringa product are usually seen experimentally,³⁴ due to the electron-electron exchange interaction,³⁶ weak correlations between local moments, or disorder effects that may enhance the relaxation rate.⁴⁰ However, an order of magnitude difference in the Korringa product suggests a strong magnetic correlation. Therefore our results confirm that no significant magnetic correlation or ordering is present in CePtSi and CePtSi_{0.9}Ge_{0.1}.

TABLE I. A comparison of Korringa products from experiments and theory for CePtSi and CePtSi_{0.9}Ge_{0.1}.

Korringa products	CePtSi	CePtSi _{0.9} Ge _{0.1}
(A) $S_{\parallel}(\text{Exp.}) = TK_{\parallel}^2 / (2T_{1\perp}^{-1} - T_{1\parallel}^{-1})$	$\sim 0.04S_0$	$\sim 0.2S_0$
(B) $S_{\parallel}(\text{Theo.}) = (n_{\text{eff},K}^2 / n_{\text{eff},1/T_1})_{\parallel} \eta_{\parallel} S_0$	$(0.06 - 0.08)S_0$	$(0.2 - 0.3)S_0$
(A)/(B)	0.5 - 0.7	0.7 - 1
(C) $S_{\perp}(\text{Exp.}) = K_{\perp}^2 T_{1\parallel} T$	$\sim 8S_0$	$\sim 8S_0$
(D) $S_{\perp}(\text{Theo.}) = (n_{\text{eff},K}^2 / n_{\text{eff},1/T_1})_{\perp} \eta_{\perp} S_0$	$(3.6 - 5.3)S_0$	$(3.6 - 5.3)S_0$
(C)/(D)	1.5 - 2.2	1.5 - 2.2

E. Spin fluctuation rate

The nuclear spin-lattice relaxation rate is determined by the electronic spin fluctuation rate [cf. Eqs. (9) and (10)]. It is straightforward to show that the average fluctuation rates $\Gamma_{0,\alpha}$ can be expressed in terms of the Korringa product by

$$\hbar\Gamma_{0,\alpha} = (n_{\text{eff},1/T_1} / n_{\text{eff},K}^2)_{\alpha} \frac{2k_B \gamma_n^2 S_{\alpha}}{\chi_{\alpha}}, \quad (21)$$

where $\alpha = \parallel$ or \perp . If the effective fluctuation rate $\Gamma_{\alpha}^{\text{eff}}$ is defined by $\hbar\Gamma_{\alpha}^{\text{eff}} \equiv 2k_B \gamma_n^2 S_{\alpha} / \chi_{\alpha}$, we obtain the relation

$$\Gamma_{0,\alpha} = (n_{\text{eff},1/T_1} / n_{\text{eff},K}^2)_{\alpha} \Gamma_{\alpha}^{\text{eff}}. \quad (22)$$

The effective f -moment fluctuation rates Γ^{eff} calculated from the Korringa products are given in Figs. 7 and 8 (open circles). CePtSi and CePtSi_{0.9}Ge_{0.1} show much faster fluctuations in the ab plane than along the c axis. The fluctuation rate obtained from the half width at half maximum (HWHM) in the quasielastic NS spectra for CePtSi [triangles in Fig. 7(a)] has been compared with $\Gamma_{\perp}^{\text{eff}}$ from the NMR experiments. The NS rates are higher, and the discrepancy between the NS and NMR data increases at lower temperatures. If the fluctuation rate from neutron scattering is $\Gamma_{0,\perp}$, according to Eq. (22) we find that $(n_{\text{eff},1/T_1} / n_{\text{eff},K}^2)_{\perp}$ has to be greater than unity, which, however, does not seem to happen. We notice that the forms of the samples used in NS and NMR are different. A random powder of CePtSi was used in the NS experiments, whereas a field-aligned powder sample was measured in the NMR experiments. NS gives the average fluctuation rate in all directions, but the NMR data gives the fluctuation rate only in the ab plane. In addition, neutron scattering and NMR probe different domains of the fluctuation spectrum (\geq GHz for NS and \sim MHz for NMR). This intrinsic difference between the two techniques may also need to be taken into account. Cox²¹ has pointed out that $1/T_1$ in NMR is a sum over all wave vectors of the dynamic susceptibility, and is expected to be less sensitive than NS to the coherent effects in the long-wavelength limit.

The anisotropy of the fluctuation rate is shown in Fig. 9. Fluctuations in the ab plane are faster than fluctuations along the c axis. The ratios $\Gamma_{\parallel}^{\text{eff}} / \Gamma_{\perp}^{\text{eff}}$ are found to be roughly equal to the ratios $A_{\parallel}^2 / A_{\perp}^2 = 0.012$ and 0.06 for CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively, where A_{α} , $\alpha = \parallel$ or \perp , is obtained from the slope of the K - χ plot, and is equivalent to $n_{\text{eff},K} A_0$. From Eqs. (12) and (22), it is not difficult to see that $\Gamma_{\parallel}^{\text{eff}} / \Gamma_{\perp}^{\text{eff}} \approx A_{\parallel}^2 / A_{\perp}^2$ is expected for the two samples. From Eq.

(22), we obtain $\Gamma_{\parallel}^0 / \Gamma_{\perp}^0 \approx 0.037$ for CePtSi and $\Gamma_{\parallel}^0 / \Gamma_{\perp}^0 \approx 0.14$ for CePtSi_{0.9}Ge_{0.1}.

F. Field dependence of $1/T_1$

The field dependence of the spin-lattice relaxation has been measured in CePtSi and CePtSi_{0.9}Ge_{0.1} at 2.1 K over the frequency range 16–45 MHz (or, equivalently, the field range 19–53 kOe). The results are shown in Fig. 10. CePtSi_{0.9}Ge_{0.1} exhibits an anomaly in the field dependence around 27 kOe, whereas CePtSi has no measurable field dependence. In some heavy-fermion systems, e.g., CeRu₂Si₂,⁵¹ metamagnetic transitions are seen in applied fields. However, a metamagnetic transition is excluded here, because CePtSi_{0.9}Ge_{0.1} does not show any nonlinear jump in the magnetization versus the field plot at 2 K. Since CePtSi_{0.9}Ge_{0.1} is close to a QCP, the applied field necessary for NMR may modify the spin dynamics if there is a critical magnetic field in this range. The effects of field induced or suppressed QCP, for instance, have been studied in CeCu_{5.9}Au_{0.1}, where 3 Tesla is enough to suppress the NFL behavior at the QCP.⁸ Dynamic susceptibility experiments at fields in the neighborhood of 27 kOe would be helpful to understand the possible field effect in CePtSi_{0.9}Ge_{0.1}.

IV. DISCUSSION

A. Anisotropy

In Sec. III C, we discussed the observed anisotropy in the ²⁹Si nuclear spin-lattice relaxation and f -spin fluctuation

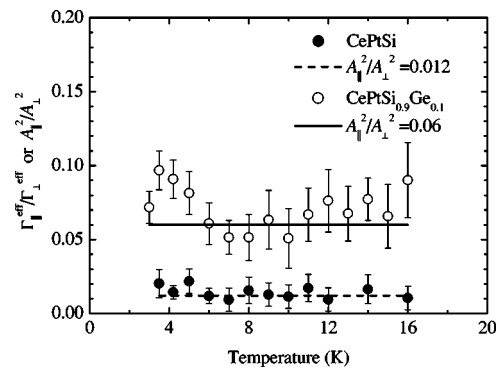


FIG. 9. Ratios of the fluctuation rates $\Gamma_{\parallel}^{\text{eff}} / \Gamma_{\perp}^{\text{eff}}$ for CePtSi (filled circles) and CePtSi_{0.9}Ge_{0.1} (open circles). Dashed line: $A_{\parallel}^2 / A_{\perp}^2 = 0.012$. Solid line: $A_{\parallel}^2 / A_{\perp}^2 = 0.06$.

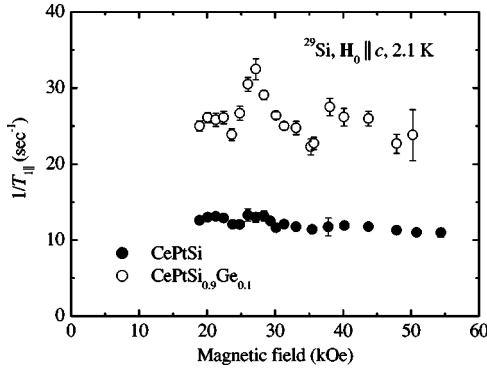


FIG. 10. Field dependence of the spin-lattice relaxation measured in CePtSi (filled circles) and CePtSi_{0.9}Ge_{0.1} (open circles) at 2.1 K.

rates in CePtSi and CePtSi_{0.9}Ge_{0.1}. The relaxation rates for fields along the *c* axis are faster than for fields in the *ab* plane. However, the fluctuation rates are found to be much greater for fluctuations in the *ab* plane than for fluctuations along the *c* axis. This seems to contradict our understanding of the relaxation, because the faster spin fluctuations in the *ab* plane are expected to relax nuclear spin more slowly for fields along the *c* axis. But from Eqs. (9) and (10), we see that anisotropy of the static susceptibility and hyperfine coupling constant must also be considered in order to explain the anisotropy of the relaxation and fluctuation rates. Similar anisotropic properties have been observed in the heavy-fermion compound CeRu₂Si₂.⁴¹

As mentioned in Sec. III D, the dynamic susceptibility can be approximately expressed as $\chi'' \approx \chi\omega/\Gamma_0$. Combining this with the Korringa-Shiba relation [Eq. (13)], the average fluctuation rate is given by the relation

$$\Gamma_{0,\alpha} \approx N(\mu_{\text{eff}})_{\alpha}^2/3\pi\chi_{\alpha}, \quad \alpha = \parallel \text{ or } \perp. \quad (23)$$

This expression gives $\Gamma_{0,\parallel}/\Gamma_{0,\perp} \approx 0.045$ and 0.17 for CePtSi and CePtSi_{0.9}Ge_{0.1}, respectively, which are close to the results (0.037 for CePtSi and 0.14 for CePtSi_{0.9}Ge_{0.1}) reported in Sec. III E. Since μ_{eff} and χ are dominated by the CEF splitting, we infer from Eq. (23) that the anisotropy of the spin fluctuation is due to crystal field effects. In addition, the previous result $\Gamma_{\parallel}^{\text{eff}}/\Gamma_{\perp}^{\text{eff}} \approx A_{\parallel}^2/A_{\perp}^2$ also suggests that the hyperfine coupling constant is also strongly affected by the CEF effect.

B. Disorder-driven relaxation

In Sec. III B, the absence of a Korringa relation at low temperatures in CePtSi_{0.9}Ge_{0.1} has been considered as evidence for non-Fermi-liquid behavior in this material down to ~ 1.5 K. From our NMR linewidth study,¹¹ disorder-driven NFL models were found to be able to explain the NFL behavior in CePtSi_{0.9}Ge_{0.1}. In our study of NMR spin-lattice relaxation, inhomogeneous relaxation rates also suggest the presence of magnetic disorder in the material. In order to understand disorder effects on the NFL-like relaxation in CePtSi_{0.9}Ge_{0.1}, we consider the effect of a distribution of Kondo temperatures on the Korringa relaxation in the single-ion Kondo-disorder model.

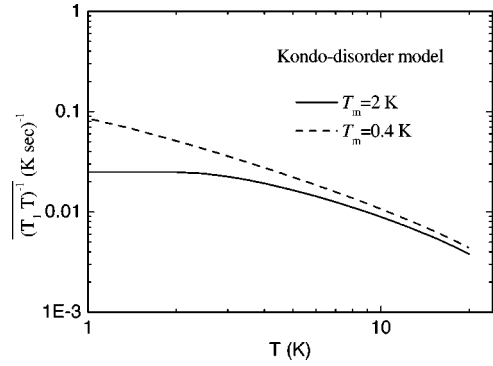


FIG. 11. Temperature dependence of $(T_1 T)^{-1}$ calculated by Kondo-disorder model for cases of $T_m=0.4$ (dashed curve) and 2 (solid curve) (see the text).

We assume a simplified rectangular distribution function $P(T_K)$ of the Kondo temperature, i.e.,⁵²

$$P(T_K) = \begin{cases} 1/(T_M - T_m), & T_m < T_K < T_M, \\ 0, & \text{otherwise,} \end{cases} \quad (24)$$

where T_m and T_M are minimum and maximum values of T_K , respectively. From Eqs. (4) and (24), the calculation of the average relaxation rate $\overline{T_1^{-1}}$ is straightforward. Two different results at low temperatures are obtained: for $T_m > T \rightarrow 0$, $\overline{T_1^{-1}} \propto T/(T_M T_m)$ (the Korringa relation), and for $T_M > T > T_m \sim 0$, $\overline{T_1^{-1}} \propto (3 - 2\sqrt{T_m/T} - T/T_M)/(T_M - T_m)$, i.e., $\overline{T_1^{-1}}$ exhibits non-Korringa-like behavior. The minimum distributed Kondo temperature T_m determines the relaxation behavior at low temperatures. This result clearly demonstrates the essential concept of the Kondo-disorder model that a finite probability of finding the unquenched local moments having T_K lower than the physical temperature, prevents the system from entering to the FL state. Figure 11 illustrates the two situations for $T_m=0.4$ K (dashed curve) and 2 K (solid curve). For both cases $T_M=20$ K. These values of T_m are roughly estimated from the predicated Kondo temperature distribution given in Ref. 11 for CePtSi_{0.9}Ge_{0.1} and CePtSi, respectively. Comparing Fig. 5 with Fig. 11, we find that the Kondo-disorder model is able to account qualitatively for the temperature dependence of $\overline{T_1^{-1}}$.

The Griffiths phase model¹³ also provides a form for the temperature dependence of the average relaxation rate, i.e.,

$$\overline{T_1^{-1}} \propto \omega^{-2+\lambda} T \tanh(\omega/T), \quad (25)$$

where $\lambda \lesssim 1$ is a characteristic exponent for the Griffiths phase. A frequency-dependent (or field-dependent) $\overline{T_1^{-1}}$ is predicted by this model. In the temperature and frequency ranges of our NMR experiments, $\omega/T \ll 1$. Thus $\tanh(\omega/T) \sim \omega/T$, and the above equation can be approximated to $\overline{T_1^{-1}} \propto \omega^{-1+\lambda}$, which is temperature-independent. Our experimental results (Fig. 4) do not agree with this relation. In addition, the field dependence of $\overline{T_1^{-1}}$ in CePtSi_{0.9}Ge_{0.1} (Fig. 10) does not obey Eq. (25). Thus the Griffiths phase model seems to disagree with our relaxation data.

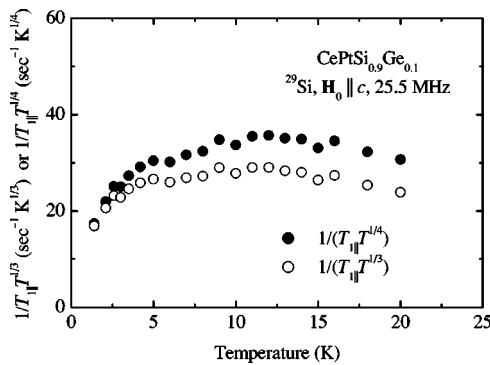


FIG. 12. Temperature dependence of $1/T_1 T^{1/4}$ (filled circles) and $1/T_1 T^{1/3}$ (open circles) in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$.

C. Other mechanisms

Other than disorder-driven mechanisms, several theoretical models for the spin dynamics near a QCP have been studied. Since $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ is close to a quantum phase transition, and comparing Figs. 5(a) and 5(b), the reduced Kondo energy scale due to this QPT may simply explain the suppressed Korringa behavior in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ at low temperatures.

From scaling analyses of the NFL alloys $\text{UCu}_{3.5}\text{Pd}_{1.5}$ and $\text{U}_{0.2}\text{Y}_{0.8}\text{Pd}_3$, Tselvik and Reizer¹⁵ proposed that collective bosonic modes of the fluctuations near a QCP dominate the low-temperature properties. The predicted fluctuation spectrum $\omega \propto q^3$ gives a power law for the NMR relaxation rate $1/T_1 \propto T^\alpha$, $\alpha=1/3$. Sengupta and Georges¹⁶ considered the mean-field behavior in the QCP regime, and the associated NFL behavior with a $T=0$ spin-glass transition. They also found that the NMR relaxation rate could have a NFL temperature dependence with $\alpha=1/4$. Figure 12 shows the temperature dependence of $1/T_{\parallel} T^{1/3}$ and $1/T_{\parallel} T^{1/4}$ in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. It is clear that neither of the above power laws for $1/T_1$ is satisfied at low temperatures. The fits to the low-temperature power law shown in Fig. 5(b) give $\alpha=0.7-0.73$ instead. This suggests a different relaxation mechanism in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$.

Recently, Si *et al.*¹⁷ have proposed a new type of QPT, in which the fluctuations of the order parameters at the critical point are localized in space, instead of spatially extended as expected in a conventional QPT. This so-called local criticality has successfully explained properties of the NFL heavy-fermion alloy $\text{CeCu}_{6-x}\text{Au}_x$, particularly neutron scattering data.⁵³ According to their theory, both the Kondo and RKKY energy scales vanish at the critical point. In addition, the local QPT facilitates two-dimensional magnetic fluctuations.

As mentioned in the Introduction, $\text{CePtSi}_{1-x}\text{Ge}_x$ has a similar phase diagram to that of $\text{CeCu}_{6-x}\text{Au}_x$. There is also a possibility for $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ to be considered quasi-two-dimensional in the spin and/or spatial domains, because spins in the *ab* plane have much greater fluctuation rates as discussed in Sec. III E, and ²⁹Si has four nearest-neighbor Ce^{3+} located in the *ab* plane of the tetragonal crystal structure. The theory of Si *et al.*¹⁷ is designed to accommodate these circumstances. However, this theory predicts a temperature-independent $1/T_1$ in the relevant temperature range, in disagreement with our data [Fig. 4(b)].

Recently, Walstedt *et al.* reported a power law for ⁶³Cu $1/T_1$ in $\text{CeCu}_{5.9}\text{Au}_{0.1}$ with $\alpha=0.75$,⁵⁴ which is close to our result in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. Their argument for the disagreement with Si's theory is that the nuclear spin-lattice relaxation process may be dominated by the magnetic fluctuations in the region away from the neutron scattering Bragg peaks in the Brillouin zone.⁵⁴ It is worth noting that various power laws for $1/T_1$ have been found experimentally in these NFL materials: $\text{Sc}_{0.7}\text{U}_{0.3}\text{Pd}_3$ ($\alpha=0$),⁵⁵ YbRh_2Si_2 ($\alpha=0.5$),⁵⁶ and $\text{CeCu}_{5.9}\text{Au}_{0.1}$ ($\alpha=0.5$ and 0.75).^{39,54} In $\text{U}_{0.2}\text{Y}_{0.8}\text{Pd}_3$, $1/T_1 \propto T \ln T$.⁵⁷

YbRh_2Si_2 is a heavy-fermion compound close to a QCP but without chemical doping.^{58,59} Ishida *et al.*⁶⁰ have reported a field dependence of T_1 in this material, and the temperature-independent T_1 as suggested by Si's local criticality tends to occur only in the extremely low-field muon spin relaxation (μSR) experiments. In addition, no magnetic disorder is observed in their NMR and μSR experiments.^{56,60} These results suggest that the static magnetic field (~ 3 Tesla) in our NMR experiments might possibly suppress the quantum fluctuations in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. On the other hand, many of the QCP theories do not take magnetic disorder into account. Magnetic inhomogeneity is inferred in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ from our NMR experiments, and this disorder could also affect the correlation of order parameters at a quantum critical phase transition, so that the predicted power laws or temperature-independent T_1 are suppressed in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$.

V. CONCLUSIONS

²⁹Si NMR spin-lattice relaxation measurements have been performed in CePtSi and $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ for applied fields in the *ab* plane and along the *c* axis. Slightly sub-exponential relaxation behavior is observed at low temperatures in the two samples, suggesting an inhomogeneous distribution of relaxation rates due to magnetic disorder. A comparison of the experimental relaxation curves with the theoretical curves based on the Kondo-disorder model shows qualitative agreement.

The Korringa relation is observed in CePtSi but not in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$, which means that the former has FL-like low-energy excitations and the latter has NFL excitations at least down to ~ 1.4 K. This result is in agreement with that from the specific heat experiments. The non-Korringa temperature dependence of $1/T_1$ in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ has been compared with the weak power laws or temperature-independent $1/T_1$ predicted from the QCP-based theories, but none of them is satisfied. This is evidence against a relaxation mechanism involving quantum critical fluctuations in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. The static magnetic field required for our NMR experiments might suppress the quantum fluctuations as in YbRh_2Si_2 .^{56,60} On the other hand, the Kondo-disorder model is able to reproduce the non-Korringa behavior and to give qualitative agreement with the $1/T_1$ data in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. This suggests that the relaxation in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$ is dominated by disorder, which may also suppress quantum critical fluctuations.⁶¹

Anisotropy in the relaxation rates and the hyperfine coupling constants is seen in CePtSi and $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. The *ab*

plane has a greater Ce^{3+} fluctuation rate and hyperfine coupling constant than the c axis. A strong CEF effect on Ce^{3+} ions in the two samples is believed to be responsible for much of this anisotropy. The Korringa products in the two samples also show anisotropy. The effective moments μ_{eff} calculated from the lowest CEF-split levels are found to be able to explain this anisotropy. The corrected Korringa products are of the same order as that for a free electron gas, which suggests that spin-spin correlations and magnetic order are not dominant in CePtSi and $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$. This appears to be evidence against the Griffiths phase model in these materials, because the expected spin correlations in magnetic clusters suggested in this model are not seen. An anomaly in the field dependence of the relaxation rate is

observed in $\text{CePtSi}_{0.9}\text{Ge}_{0.1}$, but not in CePtSi . The cause of this anomaly is not clear at this moment. Other experimental techniques such as the dynamic susceptibility measurements at different fields may be helpful.

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