PHYSICAL REVIEW B **81**, 094434 (2010)

Spin dynamics in CeB_6 studied by muon spin relaxation

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(Received 14 December 2009; revised manuscript received 2 March 2010; published 29 March 2010)

The dynamics of magnetic fluctuations in the paramagnetic and antiferroquadrupolarly (AFQ) ordered phases of single crystal CeB_6 was investigated by means of muon spin relaxation experiments. Applied magnetic fields up to 2.5 T were used. Since in the AFQ phase the contact hyperfine coupling that contributes to the fluctuating field amplitudes at the three *d*-type interstitial muon sites strongly depends on the temperature and the applied field, as shown by the corresponding Knight-shift results, this feature was taken into account in determining the magnetic fluctuation rates. The extracted correlation time τ_c of the magnetic fluctuations depends only weakly on the applied field, its temperature dependence demonstrates an accelerated slowing down of the spin dynamics in parallel with the development of the AFQ order.

DOI: 10.1103/PhysRevB.81.094434 PACS number(s): 75.25.Dk, 75.20.Hr, 76.30.Kg, 76.75.+i

I. INTRODUCTION

Multipolar interactions between *d*- and *f*-shell ions, in competition with dipolar forces, have since long been known to be the origin of phase transitions in the solid state associated with ordering of the ionic quadrupole moments, known also as orbital ordering.[1] The revival of interest in this field is due to recent discoveries of a number of ordered phases in rare earth and actinide compounds with intriguing properties and, in particular, with "hidden" order parameters related to the orientational arrangement of aspherical ionic charge distributions. The possibility for a quadrupolar (hexadecapolar or other even rank multipolar) ordering is *a priori* given if the ground-state sublevel, resulting from the free ion *J*-multiplet under the action of the crystal field, still has orbital degrees of freedom. By removal of the orbital degeneracy different aspherical ionic charge distributions can arise, and the multipolar interactions between these, when not preempted by a magnetic ordering, bring about a structure with oriented multipoles. If not coupled to a lattice distortion, this order remains hidden as it does not appear in neutron diffraction, and although the observation of some multipolar structures has recently become possible by x-ray scattering, the unambiguous assessing of the oriented multipolar structures remains in most cases difficult.

In cubic crystal fields a ground state with *both* orbital and spin degrees of freedom is possible for *f*-shell ions with half-integer $J \geq 5/2, L > 0$ multiplets. In this cases (e.g., $J=5/2$ for Ce^{3+}, 9/2 for Nd^{3+}, Np^{4+}) the orbital and spin degeneracies left by the cubic crystal field at the lowest energy level allow a fascinatingly rich variety of possible order parameters and their combinations.[2] A prominent example of coexistence and competition of various multipolar interactions is provided by the phase transitions in CeB_6, initiating a large body of recent theoretical and experimental work. The CeB_6 crystal has the CsCl structure with Ce cubes containing uniformly oriented boron octahedrons in their centers (space group $Pm\bar{3}m$, no. 221). The cubic crystal field splits the $4f^1$ $^2F_{5/2}$ ground sextet of the free Ce^{3+} ions into a quartet Γ_8 that becomes the ground state and an excited Γ_7 doublet at a distance[3] of $\approx$46 meV (530 K). For low-temperature phenomena the doublet can be left out of consideration, but the four states of the quartet Γ_8 alone are sufficient for the formation of magnetic (dipolar, octupolar) as well as time-even (quadrupolar, hexadecapolar) moments, with as many as 15 possible order parameters.[2]

At temperatures $T > T_Q$ CeB_6 is a paramagnet (PM), where $T_Q = 3.2$ K in the absence of a magnetic field. The PM state, phase I, has been thought to be of typical Kondo type, yet recent data[4,5] contradict this interpretation. On lowering T, at T_Q a transition to an ordered phase II occurs which, in the absence of magnetic field, is nonmagnetic and no lattice distortion has been observed.[6,7] (The hypothesis of an as yet unobserved small change in the boron octahedra has however also been evoked.[8]) At the still lower temperature T_N a second transition takes place to a complex antiferromagnetic (AFM) structure,[7,9] phase III, which under an external magnetic field $\vec{B}$ is further divided into two or more phase regions depending on the orientation of the field. Application of a magnetic field in phase II leads to the appearance of both ferromagnetic and ordered AFM moments coexisting with a hidden order of the same type as that present at $B=0$ but oriented according to the field direction.

Theoretical analysis[10–12] led to the conclusion that phase II has an antiferroquadrupolar (AFQ) order, with atomic positions as in phase I but with the Ce lattice divided into two sublattices differing in the orientations of the Ce^{3+} quadrupole moments of type (O_{xy}, O_{yz}, O_{zx}). This order appears when, due to the quadrupolar forces, each ionic Γ_8 quartet splits into two Kramers doublets with two different values for quadrupolar orientation, and in the ground state of the system, at complete order, all ions of a given sublattice are characterized by the one or the other Kramers state. For $B > 0$ the splitting of the doublets gives rise to an additional AFM structure with four sublattices, the structure of the AFQ and AFM orders depend on the orientation of $\vec{B}$. The unusual shape of the boundary between the AFQ and PM phases in the *T*-*B* plane, showing an increase in T_Q with increasing B [up to $B \approx 35$ T (Ref. 13)], has been explained[14,15] by the interaction of the quadrupoles with field-induced octupole moments of the same (Γ_5) rotational symmetry, stabilizing the AFQ phase. This theory explains also the variation in T_Q with applied pressure.[16]

The AFQ order appears also in $Ce_xLa_{1-x}B_6$ compounds, but only in sufficiently high, composition dependent magnetic fields, and neutron-diffraction results[17] indicate a similar situation in $Ce_{0.7}Pr_{0.3}B_6$. Recent data led to the conclusion[18–20] that phase IV of $Ce_xLa_{1-x}B_6$, replacing the AFQ phase at zero and low fields, is due to a spontaneous ordering of Ce octupoles, accompanied by some sort of random short-range magnetic order.[21]

Besides the indirect support for the predicted AFQ order given by macroscopic data, neutron-scattering,[9,22] resonance methods [NMR,[23] EPR (Refs. 24 and 25], and muon spin rotation,[26,27] the AFQ sublattices were also directly observed by x-ray scattering at synchrotron beamlines.[28–34] On lowering the temperature, at T_Q the intensity of resonant x-ray scattering starts to increase in the $[\frac{1}{2}\frac{1}{2}\frac{1}{2}]$ direction[28] consistent with the predicted AFQ order, and the temperature dependence of the AFQ order parameter could be described as

$$[(T_Q - T)/T_Q]^{2\beta}, \tag{1}$$

with $\beta \approx 0.33-0.37$. This value of the critical exponent was confirmed also by EPR data.[25] The absolute intensities from the AFQ superlattices were measured by nonresonant x-ray scattering,[31,33,35] and comparison with the calculated charge structure factors[8] showed that the ions in the AFQ phase carry not only quadrupolar but also an equally important amount of hexadecapole moments, besides the field-induced octupole moments for $B>0$.

While muon spin relaxation (μSR) is not a tool for measuring the electric fields of ionic multipoles, it provides information on the AFQ phase by probing its magnetic response, in particular, by measuring the Knight shifts and the fluctuation rate of the local magnetic field at the μ^+ site.[36] The μSR Knight shift data showed that the coupling tensor for the nondipolar (contact) part of the hyperfine interaction is anisotropic in the AFQ and also in the PM phase, and its temperature dependence for fields $0.1<B<2.5$ T could be described[26] by $\{1-(T/T_Q)^{\gamma}\}^{\beta}$ with $\beta \approx 0.5$ and $\gamma=1$ or 2 depending on the orientation of the field. This clearly shows that the contact interaction, due to the *anisotropic* Ruderman-Kittel-Kasuya-Yoshida (RKKY) mechanism, reflects the development of the AFQ order parameter. The onset of the T dependence in the μSR data well above T_Q, up to ≈ 10 K, confirmed the presence of some precursory effect in the PM phase, as indicated also in previous work.[37]

Previous μSR data also indicated[27] that at the weak $B=0.01$ T field the fluctuation rate τ_c^{-1} of the local magnetic fields in the PM phase correlates well with an assumed Kondo-type resistivity $\rho(T)$. Recent measurements[4] show, however, a temperature dependence $\rho \propto T^{-0.4}$ in this phase. To explain the new data for resistivity, magnetoresistance, and other transport properties in the PM and also in the AFQ phases the hypothesis of formation of a spin-density wave (SDW) near and below T_Q was put forward.[4,5]

As to the dynamics of the magnetic moments near and below T_Q, no singular behavior of the μSR relaxation rate λ was found at the transition. Previous work[27] seemed to indicate that τ_c *decreases* as the temperature goes below T_Q but this behavior would be difficult to understand. The reason for this paradox, as conjectured in Ref. 27, lies in the fact that this result was obtained without accounting for the variation of the strength of the hyperfine coupling between the μ^+ and the ion-electron system for $T<T_Q$.

In the present work the spin dynamics in the AFQ phase is reinvestigated by duly taking into account the field and temperature dependence of the hyperfine coupling parameters determined from previous Knight-shift data[26] on the same single crystals. The results give information on the temperature and field dependence of the correlation time τ_c of the magnetic fluctuations over the entire range $T_N(B)<T<300$ K and for $0.08<B<2.5$ T, indicating, in particular, a slowing down of spin dynamics in the AFQ phase.

II. EXPERIMENTAL SETUP, SAMPLES

The μSR experiments were performed at the Swiss Muon Source (SμS) of the Paul Scherrer Institute. In the temperature range of 100 mK–8 K the Low Temperature Facility (LTF) was used that allows to apply magnetic fields up to 3 T. The field strengths in these experiments were $B=0.08$, 0.2, 0.4, 0.6, 1.5, 2.0, and 2.5 T. Results for higher temperatures (up to 200 K) were obtained by use of the General Purpose Spectrometer (GPS) at an applied field of 0.6 T. Originally the experimental setup was aimed at measuring the μ^+ Knight shift across the phase diagram of CeB_6. To this end, the polarization of the incoming muons was, by means of a spin rotator, turned from the direction parallel to the beam axis toward a perpendicular orientation, so that the precession of the μ^+ spins around the applied field $\vec{B}$ could be observed (the field was directed along the beam axis). Since the spin rotator produced a rotation of only about 50°, a sizable longitudinal component of the polarization along $\vec{B}$ was preserved. The time dependence of this component was monitored simultaneously with the precession signal in detectors placed forward and backward with respect to the sample position along the beam axis.

In the LTF we used three pieces of flat (0.15-mm-thick) rectangular single crystals of CeB_6 covering an area of 17.5×8.6 mm^2 that were glued to the silver sample holder on the cold finger. The normal of the plate coincided with the [001] crystal axis. In the GPS instrument a cylindrical sample was used with its axis parallel to the [110] crystal axis and mounted so that the cylinder axis was perpendicular to both $\vec{B}$ and the beam direction. On rotating the cylinder, both the [001] or $[1\bar{1}0]$ crystal axes could be brought parallel to $\vec{B}$. Both samples were high-quality CeB_6 crystals grown at the Department of Physics, Tohoku University, by Prof. S. Kunii.

As usual in a μSR experiment, the evolution of the μ^+ polarization $P_\mu(t)$ was monitored via the time-dependent decay asymmetry of the nearly 100% spin-polarized implanted μ^+ by observing the positrons form the muon decay as a function of the elapsed μ^+ lifetime. The positron rate in the forward and backward detectors can be written[36] as

$$\frac{dN_{e^+}(t)}{dt} = \frac{1}{4\pi\tau_\mu} N_0 \exp(-t/\tau_\mu)[1 \pm aP_\mu(t)]\Omega. \tag{2}$$

Here a is the effective decay asymmetry ($a=\alpha P_0 \cos 50°$, where P_0 is the beam polarization, $\alpha \approx 1/3$), $\tau_\mu=2.197$ μs is

the average μ^+ lifetime, Ω is the solid angle covered by the detector, and $P_\mu(t)$ describes the evolution of the polarization of the implanted μ^+ $[P_\mu(0)=1]$.

Spin-lattice relaxation of the muon spin leads to an exponential decay,

$$P_\mu(t) = \exp(-\lambda t), \tag{3}$$

where λ is the relaxation rate, or $T_1=\lambda^{-1}$ is the spin-lattice relaxation time. Since only μ^+ implanted in the sample are subject to the spin-lattice relaxation mechanism and develop a time-dependent polarization, the muons stopped outside the sample (in the silver sample holder or cryostat walls) can be ignored (in any case, the portion of these muons in the GPS instrument is less than 1%, and in the LTF also small).

It will be seen that our measured data for the Knight shift, reported earlier,[26] represent an important *input* in the present investigation of *spin dynamics*. Note that the present data on μ^+ spin relaxation were obtained simultaneously with the Knight-shift data, hence under identical conditions.

For evaluation of our data the knowledge of the differential magnetic susceptibility $\bar{\chi}_B(T)=dM/dB$ of the crystal was also needed. Therefore, we have measured $M(B,T)$ between 1.7 and 10 K at all applied fields on a small piece of CeB_6 cut from one of the flat crystals used in the LTF facility. These measurements were made by a Quantum Design Physical Properties (QDPP) instrument.

III. THEORETICAL CONSIDERATIONS FOR DATA ANALYSIS

Our aim is to learn about the dynamical behavior of the magnetism in the AFQ phase as a function of temperature at different applied magnetic fields by measuring the spin-lattice relaxation rate λ of the test particles μ^+ which reside at given interstitial sites. The relaxation of the μ^+ polarization is caused by the fluctuating part $\vec{B}_1(t)$ of the internal field $\vec{B}_i=\vec{B}_{i,0}+\vec{B}_1(t)$ at the μ^+ site, $\langle\vec{B}_1(t)\rangle=0$. The source of the field fluctuations are the fluctuating $4f$ moments of the Ce^{3+} ions and the associated fluctuating spin polarization of the conduction electrons. That the time dependence of $\vec{B}_i$ seen by the μ^+ arises *solely* from these fluctuations is shown by the observed splitting[26] of the transverse-field (TF) μSR signal, proving that μ^+ diffusion as the origin of time dependence of $\vec{B}_i$ can be ruled out. Under the usual assumption that the correlation function of the fluctuating local field has an exponential decay characterized by a correlation time τ_c,

$$\langle\vec{B}_1(t)\vec{B}_1(0)\rangle = \langle B_1^2\rangle\exp(-t/\tau_c), \tag{4}$$

the relaxation rate $\lambda=1/T_1$ of the μ^+ polarization is given by[38]

$$\lambda = \gamma_\mu^2 \sum_{neighbor\ k} \langle(\vec{B}_{1\perp,k})^2\rangle \frac{\tau_c}{1+\omega^2\tau_c^2}. \tag{5}$$

Here $\vec{B}_{1\perp,k}$ is the contribution from atomic site k to the component $\vec{B}_{1\perp}$ of the fluctuating field $\vec{B}_1$ *perpendicular* to the external field $\vec{B}$ at the μ^+ site, $\langle\ \rangle$ denotes averaging, and $\omega=\gamma_\mu B$ is the Larmor frequency ($\gamma_\mu=2\pi\times135.54$ MHz/T). It has been assumed that the fields originating from different atomic sites fluctuate independently.

The field $\vec{B}_{1,k}$ is induced by fluctuating moments $\widetilde{\delta m}_k=\widetilde{m}_k-\langle\widetilde{m}_k\rangle$ at the position $\vec{r}_k$ of ion k, and is a sum of two terms,[39]

$$\vec{B}_{1,k} = \hat{A}_{dip,k}\widetilde{\delta m}_k + (A_{con}^{\perp} + A'_{con}\cos^2\theta_k)\widetilde{\delta m}_k, \tag{6}$$

where $\hat{A}_{dip,k}$ is the dipolar coupling tensor giving the hyperfine field at the μ^+ site $\vec{r}_\mu$ induced by a magnetic dipole $\widetilde{\delta m}_k$ and the second term stands for the contact hyperfine interaction. Here θ_k is the angle between the vector $\vec{r}_{\mu,k}=\vec{r}_k-\vec{r}_\mu$ and the vector $\widetilde{\delta m}_k$, the quantities $A_{con}^{\perp}$ and $A'_{con}=A_{con}^{\parallel}-A_{con}^{\perp}$ are scalars with $A_{con}^{\perp,\parallel}$ giving the contact coefficient for $\theta=90°$ and $\theta=0°$. While in normal metals the contact coupling parameter is isotropic and independent of temperature, experiments on f-electron metals showed[39] both the anisotropy (expressed by the $\cos^2\theta$ term) and a temperature variation in $A_{con}^{\perp,\parallel}$ in these systems. In view of this, instead of specifying the contact term as originating from a usual RKKY mechanism as in simple metallic systems, the notation "*con*" refers generally to all hyperfine contributions not accounted for by the direct dipolar interaction. In particular, the partly delocalized $4f$ electrons will also contribute to this term. In order to find the correlation time τ_c as a function of temperature T and applied field $\vec{B}$ in both the PM and the AFQ phase $T<T_Q$ we need, according to Eq. (5), the mean values $\langle\vec{B}_{1\perp,k}^2\rangle$ depending on the dipolar and contact hyperfine coupling parameters.

The tensors $\hat{A}_{dip,k}=r_{\mu,k}^{-3}(-\hat{I}+3\hat{r}_{\mu,k}\otimes\hat{r}_{\mu,k})$ are uniquely determined by the vector $\vec{r}_{\mu,k}$ (here $\hat{I}$ is the unit tensor and $\hat{r}_{\mu,k}=\vec{r}_{\mu,k}/r_{\mu,k}$), and the information on the contact hyperfine parameters $A_{con}^{\perp,\parallel}$ and their temperature dependences is available from the observed μSR Knight-shift $K(\hat{b})$ obtained on the same samples. By definition, and using Eq. (6), one has

$$K(\vec{b}) = \frac{\vec{b}\vec{B}_{i,0}}{B} = \frac{\sum_k \{\vec{b}\hat{A}_{dip,k}\hat{\chi}\vec{B} + (A_{con}^{\perp} + A'_{con}\cos^2\theta_k)]\vec{b}\hat{\chi}\vec{B}\}}{B}, \tag{7}$$

where $\vec{b}=\vec{B}/B$, $\hat{\chi}$ is the local susceptibility tensor, here actually a scalar χ due to the cubic symmetry at the ionic sites, $\vec{B}_{i,0}$ is the static field produced by the moments $\langle\widetilde{m}_k\rangle=\hat{\chi}\vec{B}$ and $\cos\theta_k=(\hat{r}_{\mu,k}\vec{b})$. Specifically, if $\vec{B}$ is oriented along a cubic unit vector $\vec{e}_i$, Eq. (7) gives

$$K(\vec{e}_i) = \sum_{neighbor\ k} \hat{A}_{dip,k}^{ii} + [A_{con}^{\perp} + A'_{con}(\hat{r}_{\mu,k}\vec{e}_i)^2]\chi, \quad i=1,2,3 \tag{8}$$

for each μ^+ site $\vec{r}_\mu$. We show below that the Knight shifts measured along a cubic axis for two magnetically different μ^+ sites give all the quantities needed for determining $\langle\vec{B}_{1\perp,k}^2\rangle$.

As to $\hat{A}_{dip,k}$, it is known[42] that in CeB_6 the μ^+ reside at the interstitial sites d, halfway between two nearest neighbor Ce ions. In the presence of the field $\vec{B}$ there are, however, three

magnetically different d sites: type d_3 at the position $(00\frac{1}{2})$, d_2 at $(0\frac{1}{2}0)$, and d_1 at $(\frac{1}{2}00)$. In what follows only the contributions from the two nearest-neighbor ions will be taken into account, the contribution from the more distant neighbors proved to be negligibly small. Thus, the sums over k reduce to two terms for each μ^+ site. For site d_3, for example, one has for each of the two neighbors

$$\hat{A}_{dip}(d_3) = \begin{pmatrix} A_{dip}^{\perp} & 0 & 0 \\ 0 & A_{dip}^{\perp} & 0 \\ 0 & 0 & A_{dip}^{\parallel} \end{pmatrix} = a_{dip} \cdot \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{pmatrix}, \tag{9}$$

where $a_{dip}=\xi^{-3}=0.1045$ T μ_B^{-1}, ($\xi=2.07$ Å is the μ^+-Ce^{3+} distance), and $\hat{A}_{dip}(d_i)$ for sites d_1 and d_2 are obtained by cyclic permutations of the diagonal elements.

Turning now to the contact hyperfine parameters, in the special case of $\vec{B}$ oriented along a cubic unit vector $\vec{e}_i$ we have, in view of the collinear Ce-μ^+-Ce configurations, $(\hat{r}_{\mu,k}\vec{b})=1$ for the d_i site with $\hat{r}_{\mu,k}\|\vec{b}$, and 0 for the two other sites. Since Knight-shift measurements provided the parameters for the two kinds of d sites separately,[26] both $A_{con}^{\perp}(T)$ and $A_{con}^{\parallel}(T)$ could be determined.

With Eq. (6), the mean square of the field component $\vec{B}_{1\perp,k}$ perpendicular to the direction $\vec{b}$ of the applied field, arising from the fluctuating moment $\overrightarrow{\delta m}_k$ at atom k is

$$\langle \vec{B}_{1\perp,k}^2 \rangle = \langle \{ \hat{A}_{dip} \overrightarrow{\delta m}_k - \hat{b}(\hat{b}\hat{A}_{dip}\overrightarrow{\delta m}_k) + [A_{con}^{\perp} + A'_{con}(\hat{r}_{\mu,k}\widehat{\delta m}_k)^2] \times [\overrightarrow{\delta m}_k - \hat{b}(\hat{b}\,\overrightarrow{\delta m}_k)] \}^2 \rangle, \tag{10}$$

where $\widehat{\delta m}_k$ is the unit vector parallel to $\overrightarrow{\delta m}_k$. One sees that the quantity to be averaged is quadratic in the components of $\overrightarrow{\delta m}$, thus proportional to $(\delta m)^2$, and contains second, fourth, and sixth powers of the direction cosines of $\overrightarrow{\delta m}$. Assuming isotropy of the moment fluctuations, the averaging is straightforward. Taking into account the (equal) contributions from the two nearest neighbors of each μ^+ sites and performing the average over the three sites d_i one obtains

$$\begin{aligned} \sum_k \langle \vec{B}_{1\perp,k}^2 \rangle &= \frac{4}{3}\langle (\delta m)^2 \rangle \left\{ \frac{1}{3}[2(A_{dip}^{\perp})^2 + (A_{dip}^{\parallel})^2] + (A_{con}^{\perp})^2 \right. \\ &\quad \left. + \frac{2}{3}A_{con}^{\perp}A'_{con} + \frac{1}{5}A'^2_{con} + \frac{2}{5}A'_{con}\left(A_{dip}^{\parallel} + \frac{2}{3}A_{dip}^{\perp} \right) \right\} \\ &= \langle A^2 \rangle \langle (\delta m)^2 \rangle, \end{aligned} \tag{11}$$

where the "effective coupling parameter" $\langle A^2 \rangle$ defined by the second equation relates the mean square fluctuations of the hyperfine field at the μ^+ site to the momentum fluctuations in the ion-electron system. One sees that $\langle A^2 \rangle$ is independent of the orientation of the applied field $\vec{B}$ but depends, for $T<T_Q$, on the temperature via the observed temperature dependences[26] of $A_{con}^{\perp,\parallel}$,

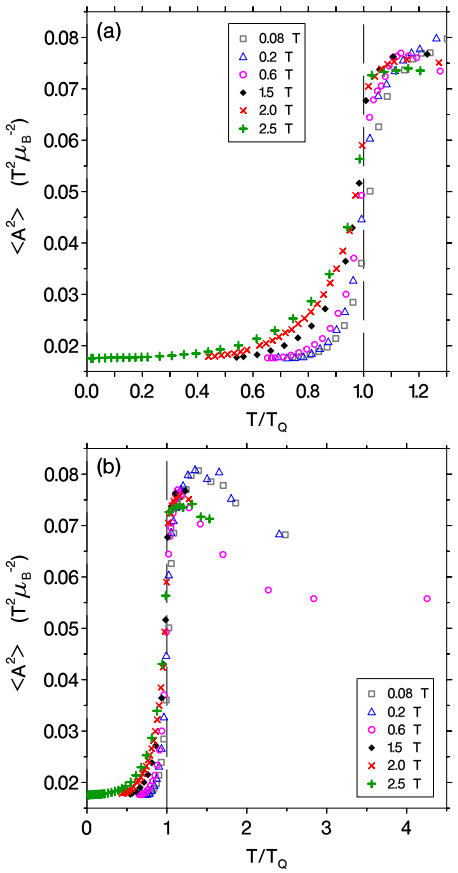

FIG. 1. (Color online) Effective coupling parameter $\langle A^2 \rangle$, Eq. (11), as a function of temperature T/T_Q for different field strengths B.

$$A_{con}^{\perp,\parallel}(T) = A_{con}^{\perp,\parallel}(T_Q) + A_{con}^{\perp,\parallel}(0)[1 + (T/T_Q)^{\gamma_{\perp,\parallel}}]^{0.5}, \quad T < T_Q, \tag{12}$$

where for $B>0.1$ T one has $\gamma_{\parallel}=2$, $\gamma_{\perp}=1$. The temperatures $T_Q(\vec{B})$ and the values for $A_{con}^{\perp,\parallel}(0)$ are listed in Ref. 26.

Using our Knight-shift data[26] the effective coupling parameter $\langle A^2 \rangle$ is plotted in Fig. 1 for different applied fields $\vec{B}\|[001]$. One sees that $\langle A^2 \rangle$ depends only weakly on temperature for $T>T_Q$ and becomes a constant for $T \gtrsim 3T_Q$, but on lowering T below T_Q it drops rapidly to one fourth of its value at T_Q, as a result of the interplay of the constant dipolar and varying contact part, Eq. (12), of the coupling.

Equation (5) with use of Eq. (11) takes the form

$$\lambda = \gamma_\mu^2 \langle A^2 \rangle \langle (\overrightarrow{\delta m})^2 \rangle \frac{\tau_c}{1+\omega^2\tau_c^2}, \tag{13}$$

so that for determining $\tau_c(T,B)$ we have to calculate $\langle (\delta m)^2 \rangle$ as a function of T and B. It is well known that this quantity is related to the linear magnetic response of the system by the fluctuation-dissipation theorem[40] as

$$\langle (\delta m)^2 \rangle = \frac{2k_BT}{\pi} \int_0^\infty \frac{\chi''(\omega)}{\omega} \left(\frac{\hbar\omega}{2k_BT} \coth \frac{\hbar\omega}{2k_BT} \right) d\omega, \tag{14}$$

where $\chi''(\omega)$ is the imaginary part of the magnetic susceptibility as a function of the frequency ω of the perturbing field,

and the mean value on the left-hand side refers to the *unperturbed* state. [For that one has $\langle\vec{m}\rangle=0$ and $\langle(\delta m)^2\rangle=\langle m^2\rangle$, which gives the usual form of Eq. (14).] For CeB_6 indeed $\langle\vec{m}\rangle=0$ for $B=0$ in both the PM and AFQ states, but for strong fields, where the perturbation (with respect to the $\langle\vec{m}\rangle=0$ state) goes well beyond the linear-response region, this fact has to be taken into account.

Consider first weak applied fields, where the shift of T_Q with B is negligible. For any real system χ'' becomes negligibly small[40] above some maximum frequency ω_m. If at a given T the factor $f(\omega/T)=\frac{\hbar\omega}{2k_BT}\coth\frac{\hbar\omega}{2k_BT}$ in the integral remains near to 1 (which is the case for $\frac{\hbar\omega_m}{2k_BT}\ll 1$), this factor can be ignored and the relation connecting the real and imaginary parts χ' and χ'' of χ leads to the classical limit of Eq. (14),

$$\langle(\delta m)^2\rangle \approx \chi(0)k_BT, \quad [\hbar\omega_m/(2k_BT)\ll 1], \tag{15}$$

with the static susceptibility $\chi(0)$. The validity of Eq. (15) at a given temperature T depends on the value of ω_m, *a priori* unknown in this case. In lack of measured data a reasonable estimate of ω_m can be given by the (at the outset unknown) τ_c as $\omega_m\approx 2\pi\tau_c^{-1}$, and the condition $\hbar\omega_m/(2k_BT)\approx hk_B^{-1}(\tau_cT)^{-1}<1$ takes the form

$$\tau_cT > 4.80\times 10^{-11}\ \text{K s}. \tag{16}$$

Some estimates for τ_cT in the case of CeB_6 can be given by either the linewidth found in quasielastic neutron scattering[22] or by the data of earlier μSR work.[27] These show that a minimum value $\tau_cT\gtrapprox 2-3\times 10^{-11}$ K s may be reached at T_Q but the inequality Eq. (16) is certainly true as T increases. Thus for $T>T_Q$, apart from the immediate neighborhood of T_Q, the use of Eq. (15) seems *a priori* reasonable. We use therefore Eq. (15), calculate τ_c and check *a posteriori* the consistency of the procedure by calculating the function $f(\omega_m/T)$ with $\omega_m=2\pi\tau_c^{-1}$. The result plotted in Fig. 2 shows that $1<f(\omega_m/T)<1.2$ everywhere except at $T=T_Q$, where a sharp maximum appears within a narrow temperature interval of $\Delta T\approx 1$ K. Thus, except for this interval ΔT, the calculation is consistent for all fields and temperatures. Since $f(x)=x\coth x\approx 1+x^2/3$ for $x\ll 1$, neglecting this factor within ΔT will lead to an underestimate of $\langle(\delta\vec{m})^2\rangle$ and, by Eq. (13), an overestimate of τ_c.

For higher applied fields other complicating circumstances appear. Not only the shift of T_Q with B becomes significant but χ characterizing the state $\langle\vec{m}\rangle=0$ is no longer relevant for determining the momentum fluctuations. To calculate $\langle(\delta m)^2\rangle=\langle(m-\langle m\rangle)^2\rangle$ in this case one has to start from the equilibrium state *under the effect of* $\vec{B}$, where $\langle m\rangle\neq 0$, and use in Eq. (14) the differential susceptibility $\bar{\chi}_B=(dM/dB)$, which is now the linear-response function of this state for small variations in B. (Also, for higher fields the isotropy of the fluctuations can no longer be perfect since by $\langle\vec{m}\rangle\neq 0$ the cubic symmetry of the equilibrium state is broken. We assume that for the moderately high fields $B\leq 2.5$ T used here the difference in the mean values $\langle(\delta m_\alpha)^2\rangle$ remains nevertheless sufficiently small.)

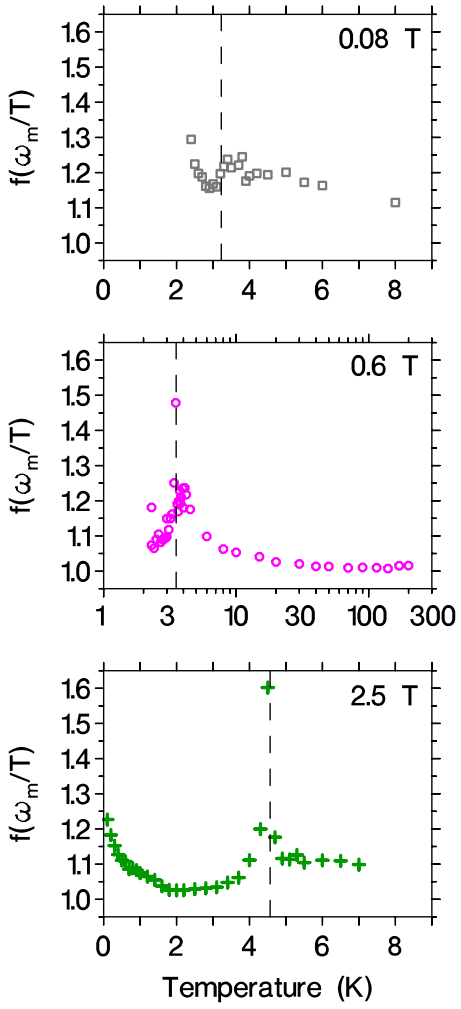

FIG. 2. (Color online) Variation in the function $f(\omega_m/T)$ (see text) for different applied fields. The deviation from unity is appreciable at the immediate neighborhood of T_Q (vertical lines).

IV. RESULTS AND ANALYSIS

The results for the longitudinal muon spin relaxation rate λ at several applied field values are displayed as a function of temperature in Figs. 3–5. The data at 0.08, 0.2, 0.6, and 1.5 T clearly reflect the transition at T_N into the AF phase III by an abrupt drop of λ, as is generally seen in AF compounds[41] and indicates the freezing of spin fluctuations in the ordered state. In contrast, λ at $B=2.0$ T shows a smooth decline through $T_N\approx 1.75$ K (except that the data point just above T_N may reflect the closeness to the phase III—phase II boundary), while the drop seen at 2.6 K is at first sight puzzling but, as shown below, of no significance. The transition into the AF phase is also reflected in the TF μSR signal. Above T_N this signal consists[42] of *two* well split precession components arising from the different Knight shifts associated with the magnetically nonequivalent interstitial d sites occupied by the μ^+. Below T_N a multifrequency signal appears,[43] generally very fast damped. This allows one to obtain an independent determination of T_N, which agrees well with the one marking the drop of λ. For the high field $B=2.5$ T no phase III exists, and the transverse-field signal

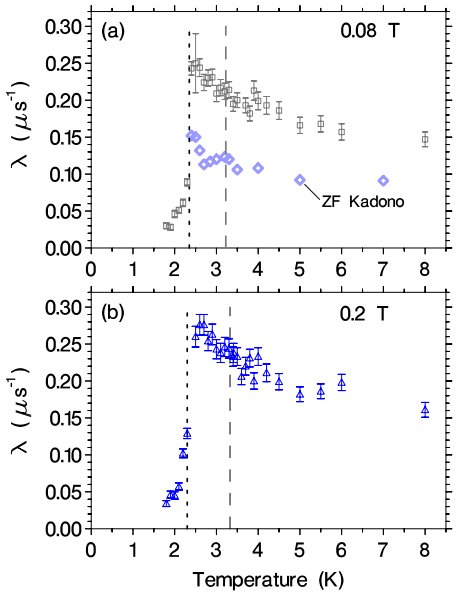

FIG. 3. (Color online) Exponential relaxation rate λ at low applied fields B vs temperature T. The transition temperatures T_Q and T_N are indicated by dashed and dotted lines, respectively. The near-zero-field (ZF) results of Ref. 27 are also shown for comparison.

shows the splitting into two components down to the lowest temperatures.[26] The transition between phase I and II does not seem to be reflected in λ to any significant degree, certainly no anomalies or discontinuity can be seen at T_Q.

In Fig. 5 the temperature dependence of λ is plotted for $B=0.6$ T in the whole measured temperature range. The figure also shows that λ does not depend on the orientation of the field $\vec{B}$ with respect to the crystal axes over the entire temperature range. We also mention that the TF-μSR signal displayed a damping that was for $T \geq T_Q$ best described by the product $\exp(-\frac{1}{2}\sigma^2)\exp(-t/T_2)$, indicating static (inhomogeneous) line broadening as well as dynamically induced (homogeneous) broadening. We found that $\lambda \equiv T_1^{-1} \approx T_2^{-1}$ implying very rapidly fluctuating internal fields. This is consistent with the further observation that above 2.5 K λ is essentially field independent up to field strengths of 2.5 T (see also below).

Also shown are in Fig. 5 the data[27] taken for the low longitudinal field (0.01 T). The present λ's are overall about a factor 2 larger. This difference must be due to the different samples: the data of Ref. 27 were taken on a polycrystalline sample, whereas in the present measurements single crystals of CeB_6 were used.

Since the static susceptibility is an important input for evaluating our data, it seemed appropriate to measure this quantity for different fields and temperatures also on a sample used in these μSR experiments. The measured $M(T)=\chi B$ values are shown in Fig. 6, they agree well with the data available in the literature[23–25,44] [note the nonlinear field dependence of $M(B)$ below T_Q]. From these data the differential susceptibility $\bar{\chi}_B(T)$ was determined by numerical derivation, results for different fields are displayed in Fig. 7 and are consistent with the data of Ref. 5. [For $B=2.5$ T below 1.7 K an extrapolation for $\bar{\chi}_B(T)$ was used, in order to analyze also the low temperature data for λ which, at that field strength, refer everywhere to phase II.] However, these calculations carry some numerical uncertainties, in particular, close to T_Q.

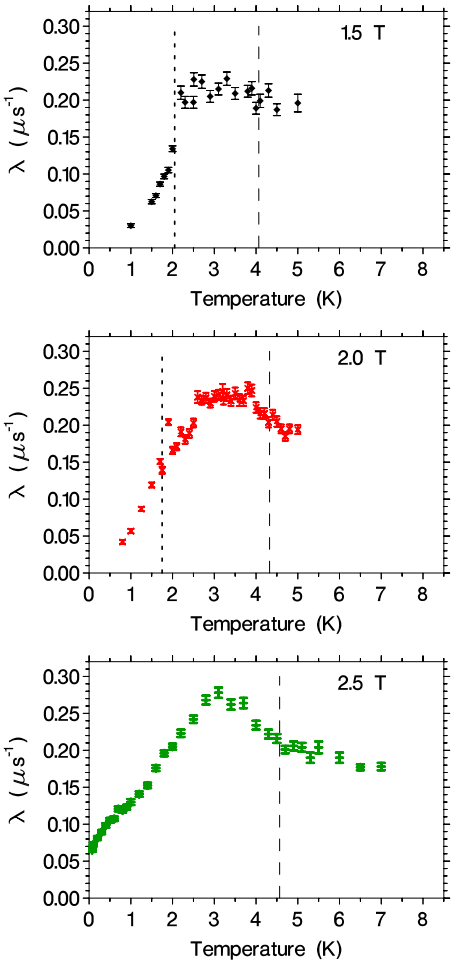

FIG. 4. (Color online) Relaxation rate λ as in Fig. 3 but for the higher fields 1.5, 2.0, and 2.5 T.

The observed field independence of λ above T_Q implies, by Eq. (5), that $(\omega\tau_c)^2 \ll 1$. In fact, for the highest field of 2.5 T one has $\omega = 2.13 \times 10^9$ s^{-1} and, as seen below, $\tau_c < 0.3 \times 10^{-10}$ s within the measured temperature range, therefore this condition is fulfilled both above and below T_Q.

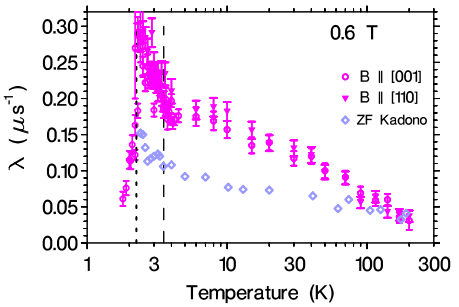

FIG. 5. (Color online) Relaxation rate λ at a field strength $B=0.6$ T for the broad temperature range $2 < T < 300$ K at different field orientations. The $B=0.01$ T (ZF) data of Ref. 27 measured at a polycrystalline sample are also shown.

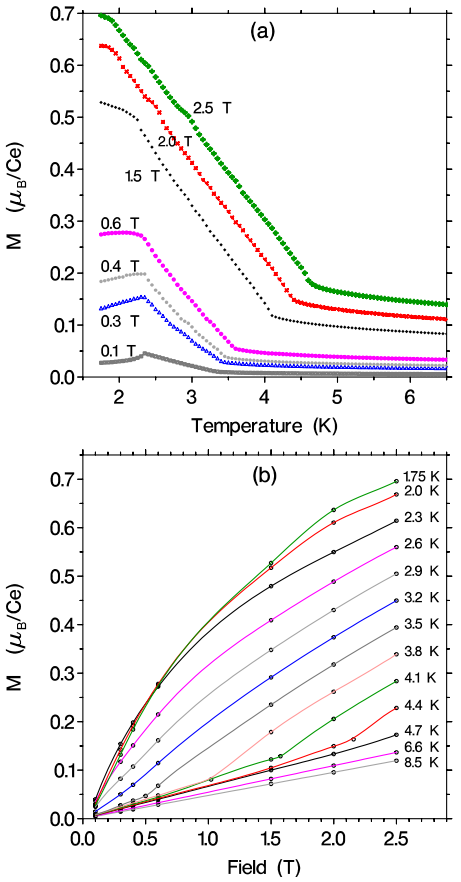

FIG. 6. (Color online) Magnetic moment $M(B)$ per Ce atom (a) vs temperature and (b) vs applied field B.

Therefore, according to Eqs. (13) and (15) of the previous section the μ^+ spin relaxation rate λ is given by the expression

$$\lambda(B,T) = \gamma_\mu^2 \langle A^2 \rangle \bar{\chi}_B k_B T \tau_c. \tag{17}$$

The effective coupling parameter $\langle A^2 \rangle$ was evaluated in Sec. IV and displayed in Fig. 1. With the data for $\lambda(B,T)$ and

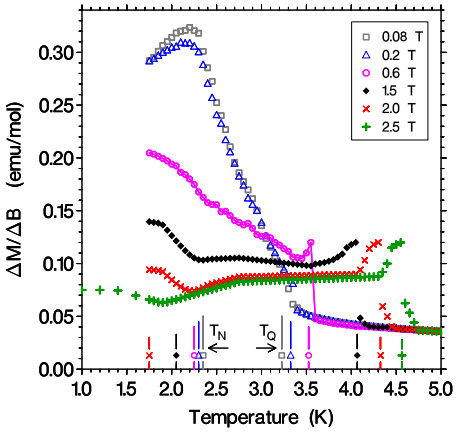

FIG. 7. (Color online) Differential susceptibility $\bar{\chi}_B$ vs temperature for different applied fields. The transition temperatures T_Q and T_N are also indicated.

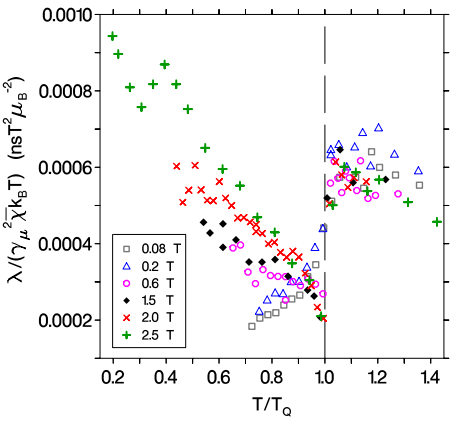

FIG. 8. (Color online) The quantity $\lambda(B,T)(\gamma_\mu^2 \bar{\chi}_B k_B T)^{-1} \propto \langle A^2 \rangle \tau_c$ as a function of temperature near T_Q.

$\bar{\chi}_B(T)$ and in the knowledge of $\langle A^2 \rangle$ as a function of B and T the correlation time was deduced by the above equation. As a first step, Fig. 8 shows $\lambda(B,T)(\bar{\chi}_B k_B T)^{-1} \propto \langle A^2 \rangle \tau_c$. One sees that all anomalies in Figs. 3 and 4 have disappeared, in particular, the maximum of λ at 3 K ($T/T_Q \approx 0.66$) for $B=2.5$ T. This can be taken as an evidence that indeed $\lambda \propto \bar{\chi}_B \cdot T$. On the other hand the local maximum at $T/T_Q = 0.4$ for $B=2.5$ T reflects the behavior of $\bar{\chi}_B(T)$ and can be an artifact of the possible systematic uncertainties in calculating this quantity. These uncertainties may also be responsible for the drop of $\lambda(B,T)(\bar{\chi}_B k_B T)^{-1}$ at T_Q. It is interesting to note that below T_Q the slope in Fig. 8 changes from positive for low B to negative for high B. We will see that this distinction has largely disappeared in the behavior of τ_c.

Finally, the results for $\tau_c(B,T)^{-1}$ are displayed in Fig. 9(a) in a log-log plot for the full temperature range. This global view gives the impression that all data follow, independently of the applied field, more or less the same temperature dependence. This is not strictly the case, as seen in Figs. 9(b) and 9(c) showing the range $T/T_Q < 2.5$. Below T_Q the low field (0.08 and 0.2 T) data show a tendency to level off as T_N is approached, in contrast to the data for higher B. The differences may in part be traced back to the behavior of $\bar{\chi}_B(T)$ but also to the fact that close to T_Q the function $f(\omega/T)$ deviates significantly from unity (Fig. 2) so that Eq. (15) is not strictly valid. Figure 9 also shows that even for the highest field $B=2.5$ T one has, except for the point at 0.02 K, overall $\tau_c < 1.7 \times 10^{-10}$, i.e., $\omega^2 \tau_c^2 < 0.13$ and much less for weaker fields, hence the term $\omega^2 \tau_c^2$ in the denominator of Eq. (17) can indeed be neglected.

V. DISCUSSION

Equation (17) implies that in determining $\tau_c(T,B)$ the variation in the coupling parameter $\langle A^2 \rangle$ is decisive. Comparing Figs. 1 and 9 it is conspicuous that below T_Q both $\langle A^2 \rangle$ and τ_c vary strongly with the temperature, but their opposite variations result in a relatively weak temperature dependence of λ (Figs. 3 and 4), with no clear anomaly at T_Q but simulating the peaks at 3 K at the field of 2.5 T and the drop below 2.6 K at 2 T. In other words, the increase in τ_c with the

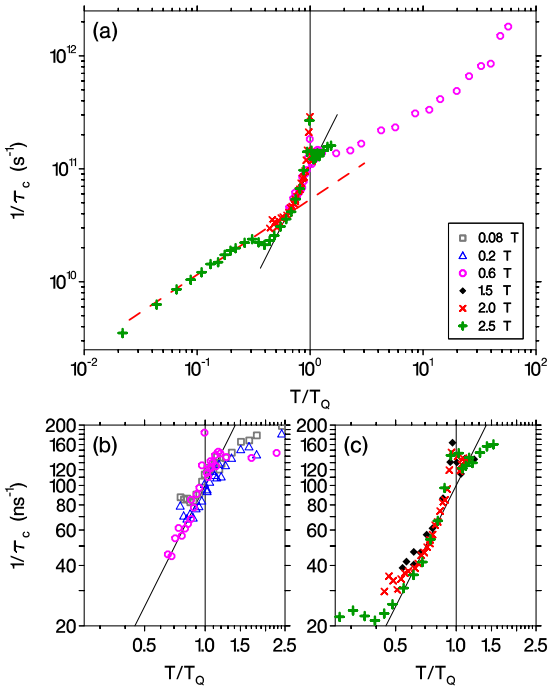

FIG. 9. (Color online) Reciprocal correlation time τ_c^{-1} of the moment fluctuations vs the temperature T/T_Q for different applied field strengths B. The solid and dashed lines correspond to $\tau_c^{-1} \propto T^2$ and $\tau_c^{-1} \propto T^{2/3}$, respectively.

development of the AFQ order has little influence on the μ^+ relaxation time $\propto\lambda^{-1}$ because of the simultaneous weakening of the effective hyperfine coupling $\langle A^2\rangle$. (A remark that the variation in $\langle A^2\rangle$ in the AFQ phase may be an important factor in determining τ_c was made in Ref. 27 since keeping $\langle A^2\rangle$ constant lead to a *decrease* in τ_c with decreasing T, a result difficult to understand.)

A point to be considered is the possibility that what is observed is partly a muon-induced effect. It is indeed known that in some intermetallic rare earth and actinide compounds [e.g., in $PrNi_5$ (Ref. 45) and UNi_2Al_3 (Ref. 46), but not in general], the presence of the muon may induce significant changes in the local atomic susceptibility by modifying the crystalline electric field splitting or the exchange interaction of the neighbor pairs straddling the μ^+. The latter can be seen, for example, by a change in the Curie temperature (see, e.g., Ref. 47). However, as discussed in Ref. 23, if there is a muon-induced change in the local susceptibility in CeB_6 at low temperatures, it is of relatively minor importance compared to the dramatic temperature dependence of A_c in phase II. For CeB_6, the perfect scaling of the muon Knight shift with the bulk susceptibility above 10 K also implies that the Curie temperature and therefore the exchange coupling between the Ce ions in the vicinity of the μ^+ is not significantly affected by the presence of the muon. This is further confirmed by the precise equality of the field dependent T_Q values, Ref. 26, obtained via the muon Knight shift and the data given in the literature.

In using $\langle A^2\rangle$ calculated on the basis of Knight-shift data we have assumed that the hyperfine coupling is the *same* for both the static moments $\vec{m_k}$ induced by a *static* field, and for

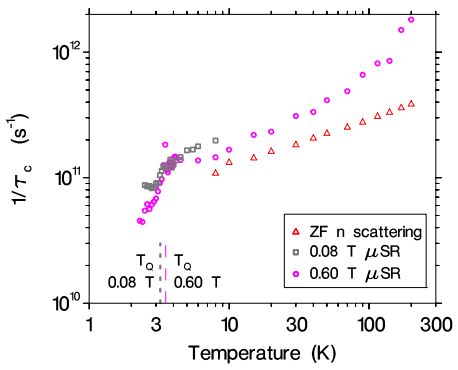

FIG. 10. (Color online) Comparison of the present values of τ_c^{-1} for $B=0.08$ T and 0.6 T with the values deduced from quasielastic neutron scattering (Ref. 22).

the rapidly fluctuating $\vec{\delta m_k}$. This implies merely the absence of a mechanism delaying the propagation of the field fluctuation, a most plausible assumption. The dipolar coupling $\hat{A}_{dip}$ being constant, the decrease in $\langle A^2\rangle$ with decreasing T is entirely due to the variation in $A_{con}^{\perp,\parallel}$. This variation for $T<T_Q$ was seen[26] to weaken and even overbalance the coupling $\hat{A}_{dip}$, resulting in the fact that both Knight shifts K_1 and K_2 (for the two magnetically inequivalent μ^+ sites) *go through zero* (or, for some B values, approach closely zero) as T decreases.

As to the physical mechanism behind the variation in the hyperfine coupling $A_{con}(T)$, an explanation was given in Ref. 39. It was shown that, due to the *anisotropy* of the RKKY interaction in the *s-f* electron system, ordering of the ionic electric multipoles must necessarily result in a temperature dependent and anisotropic contact hyperfine coupling. In the context of the relationship between the anisotropic RKKY interaction and quadrupolar ordering it was also shown[48] that, for ions such as Ce^{3+} with crystal field Γ_8 ground states, the anisotropy of the RKKY coupling leads to a specific quadrupolar interaction between the ions with preferred orientations for the quadrupole moments with respect to the crystal axes. An exact *quantitative* treatment of the effect of AFQ ordering on the anisotropy of the RKKY interaction and thereby on the contact hyperfine coupling in CeB_6 is still difficult, first because of the inherent complexity of the correlated *s-f* electron system and second, since higher-order multipole moments besides the quadrupolar ones seem also to be involved in the AFQ order.[8,14,15] While anisotropic and temperature dependent contact terms have been observed[43] in several heavy-fermion systems, the peculiar feature for CeB_6 is that this temperature dependence follows the evolution of the AFQ order parameter. Recent indications on the formation of some specific magnetic structures, polarons or a SDW in the AFQ phase,[4,5] if confirmed by future *microscopic* studies, may modify the picture based primarily on the ordering of electric multipoles, however these proposed magnetic structures have not been seen in magnetic neutron-diffraction patterns.[9]

In Fig. 10 the present results for τ_c^{-1} are compared to values extracted from the broadening of the quasielastic peak in neutron scattering[22] data available for $8<T<200$ K. One

sees that the values derived from the neutron data are somewhat smaller and show a weaker temperature dependence. The reason for this numerical disagreement between the muon and neutron data may in part be due to the difficulty to deconvolute the shape of the quasielastic line at the given experimental resolution. The value $\tau_c^{-1}=3.3\times10^{10}$ s^{-1} obtained at $T=4$ K by EPR (Ref. 24) is also reasonably near to our data.

Clearly, as Fig. 9(a) demonstrates, the spin dynamics shows a marked change at T_Q. The $B=2.5$ T data, which extend to much lower temperatures since phase III is absent, show a further change in temperature dependence below $T/T_Q\cong0.3$. In this range one has approximately $\tau_c^{-1}\propto T^{2/3}$, whereas above this temperature, and above T_N for all other applied fields, one has roughly $\tau_c^{-1}\propto T^2$. Above T_Q, in the PM phase, the monotonic increase in τ_c^{-1} with T was attributed to a Kondo-type behavior.[27]

Assuming that the relaxation processes arising from the interaction of the Ce^{3+} moments with the conduction electrons has no singular behavior at T_Q, the abrupt variation in the slope of τ_c^{-1} at T_Q can be qualitatively explained by the change in the rate of the *phonon processes*, associated by the particular form of the ionic wave functions required by the AFQ order.

Intuitively it is clear that freezing of the quadrupolar degrees of freedom by the removal of orbital degeneracy of the ionic ground state can reduce the motional freedom of the dipolar moments as well, slowing down thereby the dipolar relaxation. This is indeed the case for CeB_6 below T_Q. To see this one has to remind that, *above* T_Q, the orbital degeneracy of the Γ_8 levels allows direct one and two-phonon processes for spin relaxation via transitions *within* the quartet. However, the formation of the two AFQ sublattices at T_Q changes this situation, since the lowest-lying ionic states at sublattice A (or B) are now Kramers doublet K_A (K_B), split out of the Γ_8 space by the quadrupolar ordering. The rate of direct phonon processes is now abruptly reduced[49] and Orbach processes become important, requiring participation of an orbitally different, intermediate excited state. For an ion in sublattice A with quadrupolar orientation determined by doublet K_A the components of K_B are just such excited states, with a rotated, "out-of-order" quadrupole orientation, since by the AFQ order this orientation corresponds to the ionic ground state in sublattice B. The excitation energy Δ for these intermediate states is of the order of T_Q. Strictly speaking, this situation is fully realized only at complete AFQ order, but the transition to the Orbach regime does begin at T_Q and is completed gradually with the increase in the AFQ order parameter as the temperature decreases. This gradual removal of the direct processes and transition to the Orbach regime seems to be behind the accelerated decrease in τ_c^{-1} as T is lowered down to T_N. For the highest field $B=2.5$ T the phase transition at T_N is absent, at $T\approx0.3T_Q$ the transition to the Orbach regime seems to be completed and this continues to govern spin relaxation down to the lowest measured temperatures.

The above localized exciton-like picture of a single Ce^{3+} ion being excited to the level of the out-of-order Kramers doublet, i.e., the formation of individual quadrupolar excitations, is somewhat oversimplified. The elementary excitations in a complete AFQ order are expected to be collective "quadrupolar" modes, similar to spin waves or "librational" phonons in molecular crystals related to the rotational degrees of freedom. Indeed, the model calculations[50] for CeB_6 have indicated that the wave functions of the excitations are superposed from individual excited ionic states of mixed, multipolar character, where besides the quadrupolar degrees of freedom the dipolar and octupolar ones are also involved. The excitations form several "acoustical" (Goldstone-type) and optical branches, and the spin-relaxation processes should imply creation and annihilation of these collective multipolar modes. Below T_Q these processes gradually replace the direct phonon processes that become strictly forbidden for $B=0$ and even for $B\neq0$ are reduced by a factor[49] of the order of $(B\mu_B/\Delta)^2$. The above argument is qualitative, but for an accurate description of the functional form of the temperature variation in τ_c^{-1} more information based on as yet not available, experimental investigations of the multipolar excitations are needed. Also, the role of the predicted[51] short-range ferromagnetic correlations in the relaxation processes have to be clarified.

A further complication may be introduced by the well known fact that in phase II the external field induces a simple AFM order. (This AFM order is not seen in our μSR measurements, due to the high symmetry of the μ^+ site, at which the static internal fields arising from the AFM moments cancel.) The AFM order will certainly lead to a partial suppression of magnetic fluctuations. Along with the AFM order some of the fluctuations of the $4f$ moments may be correlated and would cancel at the μ^+ sites, having no effect on the μ^+ spin relaxation.

Further, one has to remind that the ionic Γ_8 quartets can carry also octupole moments and, in fact, field-induced octupole moments are important in stabilizing the AFQ phase (Sec. I). Yet, in the analysis of the local field *fluctuations* we considered only magnetic fields due to the randomly fluctuating *dipolar* moments in the CeB_6 system [see Eqs. (5) and (6)]. One has to ask, therefore, how important is the contribution from the *fluctuating octupole* moments to the average field fluctuation $\Sigma\langle(\vec{B}_{1\perp,k})^2\rangle$ at the μ^+ site. The answer is given by the fact that, according to a reasonable estimate,[52] the field amplitude $B_1^{(oct)}$ in CeB_6 at the μ^+, due to the neighboring Ce^{3+} octupoles, is about an order of magnitude smaller than the amplitude of the dipolar field of these ions. Therefore, the part of octupolar origin in the average of the *squared* field at the μ^+ is indeed negligibly small and can be left out of consideration.

Finally, no critical or significant change in the spin dynamics is evident on approaching from above the magnetically ordered phase III. The mentioned leveling off in τ_c^{-1} at $0.8T_Q$, if real and not an artifact of our analysis, would rather point to a slight acceleration on approaching T_N.

VI. CONCLUSION

Muon spin relaxation experiments were used to study the dynamics of magnetic fluctuations in the paramagnetic and antiferroquadrupolar ordered phases of single crystal CeB_6. A particular feature in this case is that the contact hyperfine

coupling that determines, besides the dipolar coupling, the fluctuating field amplitudes at the μ^+ site, is anisotropic and depends strongly both on the applied field and the temperature in the AFQ phase, as shown by the corresponding Knight-shift results. This variation in the hyperfine coupling was taken into account in determining the spin fluctuation rate. The extracted correlation time τ_c of the fluctuations was found to be essentially field independent for fields $B < 2.5$ T used in the experiments. On lowering the temperature below T_Q, the steeper variation in τ_c^{-1} demonstrates an accelerated slowing down of the spin dynamics. This behavior is explained by the increasingly dominant Orbach processes involving multipolar collective excitations, in parallel with the development of the AFQ order.

ACKNOWLEDGMENTS

The authors are very much indebted to S. Kunii for providing the high-quality single crystal, and to Yu. M. Kagan for a valuable discussion. We thank PSI and the Laboratory for Muon Spin Spectroscopy for providing beam time and excellent measuring conditions.

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