Extreme quantum behavior of positive muons in CeAl₂ below 1 K

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(Received 26 June 2001; published 19 December 2001)

A self-consistent analysis of zero-field μ SR data in CeAl₂ in the magnetically ordered state below $T_N \approx 3.8$ K leads to the following conclusions: (i) it is possible to confirm the nonchiral spiral (double- $\mathbf{q}$) magnetic structure found in neutron scattering [E. M. Forgan, B. D. Rainford, S. L. Lee, J. S. Abell, and Y. Bi, *J. Phys.: Condens. Matter* **2**, 10 211 (1990)], (ii) below 300 mK, each μ^+ enters into a quasilocalized state, tunneling between six g sites (Wyckoff notation), forming a ring-type arrangement, (iii) above 40 mK slow transitions (8–10 MHz) between different ring states imply that the quasilocalized states move across the crystal lattice in a quantum-mechanically driven manner, and (iv) above 300 mK about 50% of the implanted μ^+ enter into a different itinerant state, characterized by a much faster propagation rate ($\sim 10^8$ s⁻¹ at 0.9 K), again based on a quantum-mechanical process.

DOI: 10.1103/PhysRevB.65.024444

PACS number(s): 75.25.+z, 66.35.+a, 76.75.+i, 66.30.Jt

I. INTRODUCTION

In this work we demonstrate that two quite different aspects combine to produce, at first sight, a very puzzling and unusual μ SR signal in the magnetically ordered state of the cubic Laves phase ($C15$) compound CeAl₂ (space group $Fd\bar{3}m$). The first aspect concerns the complex antiferromagnetic structure of CeAl₂ below $T_N \approx 3.8$ K, which, on the basis of neutron-diffraction experiments,^{1–6} was pictured very differently as time progressed. Initially thought to be an incommensurately modulated single- $\mathbf{q}$ structure with $\mathbf{q} = (\frac{1}{2} + \delta, \frac{1}{2} - \delta, \frac{1}{2})$, $\delta = 0.11$ (Refs. 1 and 2) (which brought up the problem of understanding the origin of a modulated moment value in a system with a well-defined Kramers doublet as the lowest state of the crystal electric-field (CEF)-split $^2F_{5/2}$ multiplet of Ce³⁺), it was followed by the suggestion that the structure is in fact a triple- $\mathbf{q}$ structure³ (which was refuted immediately in Ref. 4), then followed about 10 years later by the completely different view that the structure is a nonchiral spiral (double- $\mathbf{q}$) structure with the moments on the two different Ce sublattices rotating in opposite senses and the axes of the spirals oriented along the $[00\bar{1}]$ -type axis.⁵ The spirals or helices are generated by two basis vectors $\mathbf{m}_{q1}$ (parallel to $[\frac{1}{2}\frac{1}{2}\frac{1}{2}]$) and $\mathbf{m}_{q2}$ (parallel to $[-\frac{1}{2}\frac{1}{2}\frac{1}{2}]$) (the angle between them is 70.5°) and are therefore elliptical in shape (ratio of the principal axes = $\sqrt{2}$). The advantage of this structure is that in contrast to the single- $\mathbf{q}$ structure the values of the moments vary only moderately. The ordered moment at position $\mathbf{R}_i$ is then given by⁵

$$\mathbf{m}_i(\mathbf{R}_i) = \mathbf{m}_{q1} \cos(2\pi \mathbf{q}_1 \cdot \mathbf{R}_i + \varphi_1) + \mathbf{m}_{q2} \cos(2\pi \mathbf{q}_2 \cdot \mathbf{R}_i + \varphi_2), \quad (1)$$

with $\mathbf{q}_1 = (\frac{1}{2}, \frac{1}{2} - \delta, \frac{1}{2} + \delta)$, $\mathbf{q}_2 = (-\frac{1}{2}, \frac{1}{2} - \delta, \frac{1}{2} + \delta)$, $|\mathbf{m}_{q1}| = |\mathbf{m}_{q2}| = m_0 = 0.65\mu_B$, and $\delta = 0.11$ as before.^{1,2,4} This structure leads to six inequivalent domains. In a subsequent neutron study⁶ the helical structure was further refined and it

was found that the angle between $\mathbf{m}_{q1}$ and $\mathbf{m}_{q2}$ changed from 70.5° just below T_N to 81.5° at $T = 0$ K. Given all the evidence it seems that the double- $\mathbf{q}$ helical structure describes correctly the situation of the magnetic order in CeAl₂.

Nevertheless it was deemed important to check on the magnetic structure with a local probe technique like μ SR. And here we encounter the second aspect. With the help of the μ^+ , the internal field distribution in the ordered state can be measured and compared with the expected distribution, assuming the suggested magnetic structure (see, e.g., Ref. 7). This procedure usually requires the knowledge of the μ^+ site or sites, and works only if the implanted muons are immobile, i.e., occupy a single fixed lattice site. The latter condition is usually fulfilled if the ordered state develops at a low temperature. At low temperatures (i.e., below 10 K) muon diffusion in intermetallics is almost always absent. Examples of persisting μ^+ diffusion at low temperatures, which, if present, are governed by quantum effects,⁸ are rare and are restricted to high-purity metals such as Cu, Al,⁸ and Sc.⁹ To our knowledge the only intermetallic compound in which a quantum behavior of the μ^+ was observed is UNi₂Al₃.¹⁰ In this hexagonal compound the μ^+ was found to be in a tunneling state connecting the six m or k sites forming a hexagon around the high-symmetry b site. A similar tunneling state may have been seen in isostructural GdNi₅ below 80 K.¹¹ As we will see below, such a tunneling state over an array of six sites, forming a hexagon, seems also to be present in CeAl₂ and, moreover, appears to migrate through the lattice even much below 1 K. Hence the low-temperature zero-field (ZF)- μ SR signal is the result of a spatially and temporally averaged internal field distribution. In an earlier transverse field study above T_N it was concluded that the μ^+ most likely occupy the (2-2) or g sites.¹² This was confirmed by ZF measurements on the present crystal (to be published elsewhere). The network of g sites (there are 96 in the unit cell) is displayed in Fig. 1.

FIG. 1. Crystal structure of CeAl₂ showing the network of the 96 g sites, which can be viewed as consisting of 16 interconnected hexagon configurations.

II. MEASUREMENTS

Our zero-field [and one longitudinal field (LF)] measurements were performed on the low-temperature μ SR spectrometer on the π M3 beam line of the Paul Scherrer Institute (PSI) down to 50 mK. A few measurements were also carried out on the general purpose spectrometer (GPS) on the same beam line down to 1.85 K. The sample was a single crystal exhibiting a sharp onset of the antiferromagnetic (AF) state at 3.8 K, as evidenced by the results of measurements of the specific heat $C(T)$.¹³ In the GPS the sample could be rotated around a [100] axis perpendicular to the initial polarization $\mathbf{P}_\mu(0)$, which was oriented parallel to the beam momentum. Preliminary results were reported in Ref. 14. More detailed considerations and additional measurements provoked major revisions of the interpretation to be presented in this report. In Fig. 2 we show two typical μ^+ SR signals, observed in the forward direction with respect to the beam momentum, displaying the time evolution $P(t)$ of the spin polarization of implanted μ^+ along that direction at 50 mK and 1 K in zero applied field. The spectrum of the Fourier transform of the 50-mK signal is presented below in Figs. 6–8. The appearance of an oscillating component demonstrates directly the presence of a more or less static field at the μ^+ , reflecting a magnetically ordered state. In the range 0.05–0.3 K the signal is well described by the expression

$$P(t) = A_0 [A_1 \exp(-\frac{1}{2}\sigma^2 t^2) J_0(\omega t) + A_2 \exp(-\lambda_2 t)], \quad (2)$$

with $A_1 + A_2 = 1$ and A_0 the effective decay asymmetry of the μ^+ . $J_0(\omega t)$ is the zeroth-order Bessel function, which is usually encountered if the internal field distribution, sensed by the μ^+ , is sinusoidally modulated.⁷ Although $J_0(\omega t)$ is intrinsically damped it has to be multiplied by a Gaussian damping factor in order to fit the data. We find $\sigma \approx 18 \mu\text{s}^{-1}$

FIG. 2. The time evolution of the μ^+ polarization $P(t)$ observed in ZF in the forward direction with respect to the μ^+ -beam momentum (a) at 50 mK and (b) at 1 K. The solid lines represent fits of Eq. (2) to the data. The dashed lines represent the individual components.

below 0.4 K. This fit function is to be viewed as a purely empirical function without any direct physical meaning. In particular, the ratio $A_2/A_1 \approx 0.22$ is definitely smaller than the value of 0.5 expected if the μ^+ were exposed to a distribution of randomly oriented internal fields. In this case an average of $\frac{1}{3}$ of the implanted μ^+ will sense a field oriented parallel to the muon spins. Hence they will not precess but may relax in the presence of fluctuating fields. Since, as mentioned, CeAl₂ adopts a multidomain magnetic structure with essentially randomly oriented internal fields at the μ^+ positions, we indeed expect a value of $A_2/A_1 = \frac{1}{3}/\frac{2}{3} = \frac{1}{2}$.^{7,15}

The absence of a time-independent “ $\frac{1}{3}$ ” term can be understood if some slow dynamics is involved, which exposes the μ^+ to fields of varying directions and values. This may arise from some μ^+ motion, slow enough as not to suppress the oscillating feature of the first term and to allow for a decoupling of the μ^+ spin from the internal fields in the presence of a strong external field $H_{\text{ext}} > B_i$ (see below).

Above 0.3 K $P(t)$ adopts a different behavior rather abruptly [see the 1-K signal in Fig. 2(b)]. In order to fit the data properly, a third term $A_3 \exp(-\lambda_3 t)$ has to be added to Eq. (2) and we find $A_3 \approx A_1 + A_2$ up to 3.6 K, i.e., about 50% of the implanted μ^+ are exposed to a different situation, while the other 50% continue to show the behavior seen for $T \lesssim 0.3$ K. The overall temperature dependence of the frequency $\omega/2\pi$ is displayed in Fig. 3 together with the square root of the intensity $I[(111)^+]$ of the neutron-diffraction $(111)^+$ satellite.² Both should reflect the temperature dependence of the value of the ordered moment, but obviously the frequency $\omega/2\pi$ decreases significantly faster than $\sqrt{I[(111)^+]}$. Very intriguing is the temperature dependence

FIG. 3. Temperature dependence of the average precession frequency $\omega/2\pi$. The dashed line reflects the temperature dependence of the ordered moment according to neutron data (Ref. 2).

of λ_3 , as shown in Fig. 4. It exhibits a drastic increase on approaching 0.4 K from above and is almost temperature independent above 1.5 K.

In order to test whether the third component is of a static or dynamic origin a LF, i.e., a field parallel to $\mathbf{P}_\mu(0)$, of 0.2 T, was applied at 0.9 K. The observed signal is displayed in Fig. 5. It is perfectly fitted by the expression

$$P(t) = A_0[(A_1 + A_2)\exp(-\sqrt{\lambda'}t) + A_3\exp(-\lambda_3 t)], \quad (3)$$

where $A_1 + A_2$ and A_3 are practically unchanged from their zero-field values, but components 1 and 2 are now merged into a single one with $\lambda' = 0.023(4) \mu\text{s}^{-1}$, and λ_3 has dropped from ~ 32 in ZF to $\sim 8.4 \mu\text{s}^{-1}$ in 0.2 T. The now only weakly time-dependent first term in Eq. (3) proves essentially the quasistatic nature of the internal fields, responsible for components 1 and 2 in zero field (decoupling of the μ^+ spin from the internal fields). Its “empirical” stretched exponential shape reflects probably the fact that the applied field of 0.2 T is not very much larger than the internal field of ~ 0.11 T and hence the decoupling is not complete. Note that a stretched exponential function may mimic a two-component situation. The second term in Eq. (3) corresponds obviously to the third component in zero field and proves their dynamic origin. The reduced value of λ_3 is discussed in Sec. III.

FIG. 4. Temperature dependence of the relaxation rate λ_3 . The inset displays the temperature dependence of the hopping rate $1/\tau_c$.

FIG. 5. Time evolution of the μ^+ polarization in a longitudinal field of 0.2 T at 0.9 K, observed in the forward direction. Note the difference with respect to Fig. 2(b). The solid line represents a fit to Eq. (3) to the data, with dashed lines representing the individual components.

III. DISCUSSION

In the following we focus attention on the μSR spectra below 0.3 K. In a first step we calculate the field distribution for either the single- or the double- $\mathbf{q}$ structure at the different possible μ^+ sites. It turns out that only the g -site assumption together with the double- $\mathbf{q}$ structure allows to reproduce the experimental data sufficiently well. In a second step the possibility of μ^+ motion, applying a strong-collision (Markovian) model,¹⁶ will be taken into account.

For the calculation of the field distribution over the 96 g sites we consider only the dipolar fields generated by the ordered Ce moments and we neglect a possible contact hyperfine field (this will mainly be justified by the outcome of the analysis). The dipolar field $\mathbf{B}(\mathbf{r}_\mu)$ at a given μ^+ position $\mathbf{r}_\mu$, arising from the ordered moments $\mathbf{m}_i(\mathbf{R}_i)$ at position $\mathbf{R}_i$, is, as usual, given by

$$\mathbf{B}(\mathbf{r}_\mu) = \sum_i \left[\frac{3(\mathbf{m}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} - \frac{\mathbf{m}_i}{r_i^3} \right], \quad (4)$$

where $\mathbf{r}_i = \mathbf{R}_i - \mathbf{r}_\mu$. In the case of a single- $\mathbf{q}$ structure,

$$\mathbf{m}_i(\mathbf{R}_i) = \mathbf{m}_0 \cos(2\pi\mathbf{q} \cdot \mathbf{R}_i + \varphi), \quad (5)$$

and Eq. (4) can be rewritten as⁷

$$\mathbf{B}(\mathbf{r}_\mu) = \mathbf{C} \cdot \cos(2\pi\mathbf{q} \cdot \mathbf{r}_\mu + \varphi) + \mathbf{S} \cdot \sin(2\pi\mathbf{q} \cdot \mathbf{r}_\mu + \varphi) \quad (6)$$

with

$$\mathbf{C} = \sum_i \left(\frac{3(\mathbf{m}_0 \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} - \frac{\mathbf{m}_0}{r_i^3} \right) \cos(2\pi\mathbf{q} \cdot \mathbf{r}_i),$$

and

$$\mathbf{S} = \sum_i \left(\frac{3(\mathbf{m}_0 \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} - \frac{\mathbf{m}_0}{r_i^3} \right) \sin(2\pi\mathbf{q} \cdot \mathbf{r}_i).$$

$\mathbf{C}$ and $\mathbf{S}$ are lattice sums, which are the same for all equivalent μ^+ positions $\mathbf{r}_\mu$ and need to be calculated only once. Equation (6) describes an ellipse in the $(\mathbf{C}, \mathbf{S})$ plane. For an incommensurate $\mathbf{q}$ structure the argument $2\pi\mathbf{q} \cdot \mathbf{r}_\mu$ will as-

sume all possible values between 0 and 2π and in Eq. (6) we may replace it by a continuous variable α . All possible $\mathbf{B}$ at equivalent μ^+ sites are thus determined by all points on the curve described by Eq. (6). In this case the spectral distribution of B is given by

$$S(B) = \frac{2}{\pi} \frac{B}{(B^2 - B_{\min}^2)^{1/2} (B_{\max}^2 - B^2)^{1/2}}. \quad (7)$$

In the case of the double- $\mathbf{q}$ structure, where $m(R_i)$ is given by Eq. (1), we cannot define a single variable α unless the difference between the two $\mathbf{q}$ vectors is a reciprocal-lattice vector. However, in our case this is indeed realized because $\Delta\mathbf{q} = \mathbf{q}_1 - \mathbf{q}_2 = (1,0,0)$, and we arrive again at an equation of the form of Eq. (6). One can now generate a μ SR signal in a forward/backward direction with respect to $\mathbf{P}_\mu(0)$ by calculating

$$P_0(t) = A_0 \frac{1}{2\pi N} \sum_{n=1}^N \int_0^{2\pi} \frac{1}{B_n^2(\alpha)} \{ [\mathbf{P}_\mu(0) \cdot \mathbf{B}_n(\alpha)]^2 + [\mathbf{P}_\mu(0) \times \mathbf{B}_n(\alpha)]^2 \cos[\gamma_\mu B_n(\alpha)t] \} d\alpha, \quad (8)$$

where the sum runs over all possible N inequivalent μ^+ positions. In order to take into account the multidomain structure the powder average $\langle P_0(t) \rangle$ of $P_0(t)$ is also calculated. For comparison with the data we generate with Eq. (8) a realistic “noisy” μ SR signal, taking into account the μ^+ decay and imposing statistical fluctuations. Finally we compare the Fourier transforms of generated and experimental spectra.

We first consider the double- $\mathbf{q}$ structure. Figure 6(a) displays $S(\omega/2\pi)$, calculated from the generalized Eq. (7), the inset shows the calculated $\langle P_0(t) \rangle$, and Fig. 6(b) shows the power of the Fourier transform $P_{\text{cal}}(\omega/2\pi)$ of the “noisy” $\langle P_0(t) \rangle$, superimposed on the Fourier transform (power) $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$ of the 50-mK signal. Despite the fact that the generated $\langle P_0(t) \rangle$ [see inset in Fig. 6(a)] contains, as expected, a $\frac{1}{3}$ term (which is not seen in the data and will not contribute to the simulated Fourier transform) it is evident that the Fourier transform $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$ is distinctly broader and shifted to lower frequencies, see Fig. 6(b). Note the increased weight on the low-frequency side as compared to $P_{\text{cal}}(\omega/2\pi)$, which arises from the time-dependent second term in Eq. (2). In order to get rid of the time-independent $\frac{1}{3}$ term in the simulated signal we apply the aforementioned strong-collision model by solving numerically the integral equation¹⁶

$$P(t) = P_0(t)e^{-\nu t} + \nu \int_0^t P_0(t-t')P(t')e^{-\nu t'} dt', \quad (9)$$

where ν is the fluctuation (collision) rate and $P_0(t)$ is given by Eq. (8). In the strong-collision model one assumes that the correlation between the local fields at the μ^+ before and after the fluctuation/collision event is completely lost. With $\nu \approx 8$ –10 MHz, the “ $\frac{1}{3}$ term” relaxes significantly and is roughly accounted for by the second term in Eq. (2). The Fourier transform of the “noisy” $\langle P(t) \rangle$ now reveals the same width as the experimental $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$, but still

FIG. 6. (a) Spectral distribution of the precession frequencies (or internal fields), $S(\omega/2\pi)$, across the g sites calculated for the double- $\mathbf{q}$ structure of Ref. 5. The inset shows the corresponding calculated $\langle P_0(t) \rangle$, and (b) describes Fourier power of the calculated “noisy” $\langle P_0(t) \rangle$, $P_{\text{cal}}(\omega/2\pi)$, and of the 50-mK signal $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$ (shaded).

peaks at the same, too high frequency ($\sim 22 \text{ MHz}$) as before (not shown). In order to bring the measured and the generated spectra and, correspondingly, their Fourier transforms into agreement, a reduction of the ordered moment of about 40% has to be assumed. Such a reduction is in strong conflict with the neutron results and we have to search for another explanation.

Among the various considered possibilities, e.g., the inclusion of contact hyperfine fields in the calculations or other μ^+ sites, only the conjecture that the μ^+ wave function is spread out over six g sites forming a hexagon around the crystallographic c site [generic position $(\frac{1}{8}\frac{1}{8}\frac{1}{8})$] gives a satisfactory explanation. There are 16 such rings in the unit cell. Each ring is associated with an effective field, which is assumed to be given by the average over the fields at the six ring sites. The further calculations for this configuration follow Eqs. (4)–(9), properly adapted. The results are displayed in Fig. 7. Figure 7(a) shows $S(\omega/2\pi)$ and, in the inset, $\langle P(t) \rangle$, for various fluctuation rates ν ranging from 0 to 12 MHz. In Fig. 7(b) we show the Fourier transforms of $\langle P(t) \rangle$ with $\nu = 0$ and 10 MHz together with the Fourier transform of the 50-mK data. We note a nearly perfect agreement between $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$ and the calculated $P_{\text{cal}}(\omega/2\pi)$ for $\nu = 10 \text{ MHz}$ without having to rescale the ordered moment taken from Ref. 5. The inset in Fig. 7(b) shows the experimental $P(t)$ at 50 mK together with the calculated $\langle P(t) \rangle$ for $\nu = 8$ and 10 MHz. We have achieved a similarly good agreement at other temperatures by taking into account the tem-

FIG. 7. (a) Spectral distribution of the precession frequencies if averages over the hexagon configurations are taken. The calculations are again based on the double- $\mathbf{q}$ helical structure. The inset shows the corresponding calculated $\langle P(t) \rangle$ for various fluctuation rates ranging from $\nu=0$ to 12 MHz. (b) Fourier power of the calculated “noisy” $\langle P_0(t) \rangle$ for $\nu=0$ (short dashed line) and 10 MHz (solid line) and, again the 50-mK signal (shaded). The inset compares the experimental $P(t)$ at 50 mK with calculated $\langle P(t) \rangle$ for 8 and 10 MHz.

perature dependence of the ordered moment, i.e., of $\sqrt{I[(111)^+]}$ (see Fig. 3). At least up to 2.5 K the fluctuation rate ν stays close to 10 MHz. Above that temperature ν may increase somewhat, but due to the reduced average precession frequency it is impossible to extract quantitative estimates. Note that in the limit of very fast diffusion across all g sites and as a result of the overall cubic symmetry of CeAl_2 the dipole fields at the μ^+ would average to zero. The faster decay of $\omega/2\pi$, as compared to $\sqrt{I[(111)^+]}$, with rising temperature is probably related to the ring-state motion, which also leads to a reduced average field, but is much too slow to lead to a zero-average field during the μ^+ lifetime.

The same calculations have been made on the basis of the single- $\mathbf{q}$ structure of Barbara *et al.*^{1,2,4} The results are displayed in Fig. 8. Figure 8(a) shows the calculated $S(\omega/2\pi)$ and, in the inset, $\langle P_0(t) \rangle$, for the case where the μ^+ rest at single g sites. Assuming the extended state over the hexagon of six g sites and $\nu=0$, the calculations result in $S(\omega/2\pi)$ and $P_{\text{cal}}(\omega/2\pi)$ as shown in Fig. 8(b). Simply changing the fluctuation rate to $\nu=10$ MHz results in the diagrams of Figs. 8(b), showing $P_{\text{cal}}(\omega/2\pi)$ in comparison with the measured $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$ and, in the inset $\langle P_0(t) \rangle$. Inde-

FIG. 8. Compilation of predictions assuming the single- $\mathbf{q}$ structure of Refs. 1, 2 and 4: (a) Spectral distribution of the precession frequencies across the g sites. The inset shows the corresponding $\langle P_0(t) \rangle$. (b) Spectral distribution if the extended hexagon state is assumed. The inset shows the Fourier power of the generated “noisy” $\langle P(t) \rangle$. (c) Fourier power of the “noisy” $\langle P(t) \rangle$ if the ring state and a fluctuation rate of $\nu=10$ MHz are assumed. The inset shows the corresponding $\langle P(t) \rangle$ without noise.

pendent of the actually chosen ν , the peak of $P_{\text{exp}}(\omega/2\pi)$ is always above the peak of the experimental $P_{\text{exp}}(\omega/2\pi, 50 \text{ mK})$.

Summarizing the present analysis we arrive at the conclusion that for $T \leq 0.3 \text{ K}$ all μ^+ and for $T > 0.3 \text{ K}$ about 50% of the μ^+ are in an tunneling state extended over six g sites, which form a hexagonlike ring and that this configuration migrates through the lattice with an essentially temperature-independent rate of 8–10 MHz. Both phenomena are, no doubt, quantum effects.

Finally, we briefly consider the additional relaxation component that appears above 0.3 K. The field dependence of λ_3 , measured at 0.9 K, may be explained via the well-known expression for the spin-lattice relaxation rate¹⁷

$$\lambda = \gamma_{\mu}^2 \Delta B^2 \tau_c / (1 + \omega^2 \tau_c^2). \quad (10)$$

At 0.9 K, $\lambda_3(\text{ZF})/\lambda_3(\text{LF}) \approx 32/8.4 \approx 3.8$ and we estimate a correlation time of $\tau_c \approx 10^{-8}$ s and $\Delta B \approx 0.67$ kG. The latter value is of the same order of magnitude as the width $\Delta B = 0.45$ kG of the calculated $S(\omega/2\pi)$ over the g sites. We interpret $1/\tau_c \approx 10^8 \text{ s}^{-1}$ as a μ^+ hopping rate. The estimated value of ΔB indicates that it is not the extended (tunneling) state that diffuses but that the μ^+ are localized momentarily at individual g sites. For extracting the temperature dependence of the hopping rate from Eq. (10) we have to take into account the temperature dependence of ΔB^2 , which we take to follow that of $I[(111)^+]$. The inset in Fig. 4 shows the resulting $\tau_c(T)$. The curve is similar to $\tau_c(T)$ previously reported for μ^+ in Cu at low T .⁸ First, when lowering the temperature down from T_N , τ_c decreases or, respectively, the hopping rate τ_c^{-1} increases. This behavior is predicted by the quantum theory of small-polaron propagation by incoherent tunneling at low temperatures.⁸ However, in the present case the covered temperature range is too narrow to allow for the determination of the precise form of the temperature dependence. The decrease of τ_c^{-1} below 2 K, like in Cu below ~ 0.2 K, indicates a suppression of incoherent tunneling and, unlike in the case of Cu, is followed by a transition into the extended state below 0.4 K, thereby joining the other 50% of implanted μ^+ , which are generally in this state up to at least T_N . The appearance of two essentially equal fractions of μ^+ in rather different motional states above 0.3 K is surprising and so far we cannot offer any explanation for it. It should be mentioned that preliminary results from ZF and transverse field measurements above T_N also signal the presence of two μ^+ fractions with different behavior. True long-range diffusion is observed to set in only above 80 K, as was also seen in the earlier work.¹²

In any case our observations indicate that extreme quantum behavior is involved. This is conceivable because of the small distances between two nearest-neighbor g sites in the hexagon configuration with $d_{\text{intra}} = 1.23$ Å, and between two different rings with $d_{\text{inter}} = 1.42$ Å. Since $d_{\text{inter}} > d_{\text{intra}}$ it may be understood why a tunneling state within a single ring is formed at all. Nevertheless, d_{inter} is short enough to allow for a certain transfer probability of the tunneling states between neighboring rings.

It is now instructive to compare the present results with what is known about the motional behavior of protons (hydrogen) in $C15$ Laves phase compounds, such as in TaV_2H_x and ZrCr_2H_x , which has been studied by various methods.¹⁸ The diffusive motion of H can be described in terms of at least two jump processes with different frequency scales. The faster process implies H motion within the g -site hexagons, whereas the slower process corresponds to jumps from one hexagon to another. For $\text{TaV}_2\text{D}_{1.65}$ the distance between the nearest-neighbor g sites within the hexagonal ring is 1.00 Å, whereas the minimum distance between g sites on different rings is 1.50 Å. For ZrCr_2H_x the corresponding distances are 1.13 and 1.21 Å, respectively. The slow diffusion process is due to thermal activation and is observed only at temperatures above 200 K. The fast local hopping, however, exhibits a non-Arrhenius behavior, which, on the NMR frequency scale, survives down to 10 K. The observed strong isotope effects (D vs H) at low T suggest a quantum origin of the intraring motion. This picture of proton motion in $C15$ Laves phase compounds is quite similar to our conclusions concerning the μ^+ behavior in CeAl_2 , except that the much lighter mass of the μ^+ generally favors a quantum behavior as indicated by the nonthermally activated interhexagon motion at low temperatures.

In summary we conclude that the μ^+ implanted into the intermetallic compound CeAl_2 exhibit an extreme quantum-type behavior at low temperatures. The μ^+ wave function is spread out over six g sites forming a ringlike state and, in addition, this extended tunneling state migrates to neighboring-ring configurations with a temperature-independent rate up to at least 3 K. Above 0.3 K, about 50% of the implanted μ^+ enter into a different, faster motional state across the g sites without forming the ringlike tunneling state. But also this motion is not thermally activated. The separation into two differently behaving μ^+ fractions above 0.3 K is not understood. As a by-product we confirm the double- $\mathbf{q}$ helical AF structure of the Ce moments, previously demonstrated using neutron-scattering data.⁵

ACKNOWLEDGMENTS

We thank the PSI μSR -facility group, in particular Chris Baines, and the accelerator crew for providing excellent measuring conditions.

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