Evidence of frustrated magnetism in $CeAl_3$ from muon-spin-rotation spectroscopy

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Zero-, transverse-, and longitudinal-field muon-spin rotation (μ^+SR) spectroscopy was performed on polycrystalline $CeAl_3$. The onset of frustrated quasistatic magnetic correlations below about 2 K, becoming partly coherent below 0.7 K, was detected. The occurrence of different components in the μ^+SR signal with relative strengths showing a pronounced temperature dependence leads to the conclusion that these correlations develop in a spatially inhomogeneous, frustrated way. No evidence for a macroscopic phase transition was obtained by specific heat measurements and neutron-diffraction experiments on our sample. This indicates that the coherence length of the correlations must be very short even below 0.7 K. The effective magnetic moment was estimated to be about $0.5\mu_B$ per cerium atom. It is speculated that the observed gradual evolution of magnetism in $CeAl_3$ is driven by the temperature dependence of competing magnetic interactions rather than by a divergence in the single-ion susceptibility, similar to what was recently proposed for the heavy-electron compound U_2Zn_{17}.

I. INTRODUCTION

Since the discovery of a giant electronic specific heat coefficient $\gamma = 1.62$ J/mole K^2, an almost temperature-independent magnetic susceptibility below 1 K, and a T^2 variation of the electrical resistivity in the same temperature range,[1] $CeAl_3$ has been regarded as a prime example of substances in which itinerant f electrons form a Fermi liquid of quasiparticles with exceptionally large masses at low temperatures. Within the class of heavy-electron materials, most of which were discovered in the last five years, $CeAl_3$ [along with $CeCu_6$ (Ref. 2)] is exceptional in the sense that no sign indicating any cooperative phase transition could be found down to the lowest temperatures (10 mK).[3] This is surprising, because in analogy to isostructural RAl_3 compounds (R denotes a rare-earth element)[4] and in view of the high-temperature behavior of its magnetic susceptibility $\chi(T)$, a magnetic phase transition at low temperatures might be expected.

A rough analysis of $\chi(T)$ reveals the existence of three temperature regimes, namely a high-temperature Curie-Weiss regime with an effective moment of $2.55\mu_B$ per cerium atom and a paramagnetic Curie temperature of approximately $\Theta = -40$ K,[5] a transition regime between 1.5 and 30 K,[5] and a nearly temperature-independent Pauli-type regime at temperatures below 1.5 K.[1] At 0.7 K there is a very shallow maximum in the temperature dependence of the static magnetic susceptibility $\chi(T)$ which does, however, not seem to be connected with a macroscopic magnetic phase transition. Rauchschwalbe[6] recently reported low-field ac-susceptibility measurements which revealed a shoulder of $\chi_{ac}(T)$ at 1 K followed by a sharp increase towards lower temperatures. Since this upturn of $\chi_{ac}(T)$ can be suppressed already at $B = 10$ kG,[6] it was attributed to the presence of magnetic impurities in the sample.

In this paper we present a detailed μ^+SR study of the microscopic local magnetic susceptibility of polycrystalline $CeAl_3$ which unambiguously reveals the onset of a strange kind of magnetism below about 2 K. This unanticipated experimental result was detected for the very first time by zero-field μ^+SR measurements, and was already reported previously in the form of a Letter.[7]

There are numerous anomalous features in the thermodynamic and transport properties of $CeAl_3$ below 2 K that were found in earlier experiments. Since they might be related to our findings, we would like to quote some of them here. The ratio C_p/T, where C_p is the specific heat at constant pressure, shows a distinct maximum around 0.3 K,[8,9] which can be suppressed by the application of moderate external pressure.[10] The temperature dependence of C_p itself exhibits a shallow shoulder at 2 K.[11] In addition, there are broad minima in the elastic constants at about 0.6 K,[12] a change of sign in the thermal expansion coefficient at 0.7 K,[1] and maxima in the magnetoresistance at 20 kG below 0.7 K.[6,13,14] These and other[15] features which were exclusively investigated in polycrystalline samples were interpreted as resulting from the formation of a Fermi-liquid state of heavy quasiparticles, as we have already mentioned.

Recently it became possible to overcome the problems of the peritectic formation of $CeAl_3$ which, for a long time, made the production of $CeAl_3$ single crystals impossible.[16] Resistivity and magnetoresistance measurements[17] on single crystals which revealed pronounced anisotropies subsequently provided further evidence that some kind of magnetism starts to develop in $CeAl_3$ below about 1.6 K.

Several microscopic investigations have been performed on polycrystalline $CeAl_3$. Inelastic neutron-scattering experiments gave evidence for a crystal-field splitting of the $J=\frac{5}{2}$ Ce multiplet into three Kramers doublets.[18,19] At 5 K the two excited states $J_z=\pm\frac{5}{2}$ and $J_z=\pm\frac{1}{2}$ are separated from the $J_z=\pm\frac{3}{2}$ ground state[19] by 5.0 meV and 7.25 meV, respectively. Quasielastic neutron scattering was performed on $CeAl_3$ between 60 mK and 125 K.[20] The quasielastic spectral width was found to increase as $T^{1/2}$ above 1 K, but to remain almost constant below 1 K.

^{27}Al NMR measurements in an external field of 8 kG between 1.5 and 300 K (Ref. 21) have revealed a change of the linear scaling between the isotropic Knight shift and the bulk susceptibility at 10 K, and a broad maximum of the extracted 4f-spin-lattice relaxation time T_1^{4f} at about the same temperature. The Knight-shift behavior was attributed to a temperature-dependent transferred hyperfine field caused by the crystalline electric-field splitting, while the behavior of T_1^{4f} was interpreted as possible evidence for correlated spin fluctuations of the 4f electrons.[21] At temperatures below 10 K the anisotropic NMR linewidth was found to increase strongly with decreasing temperature. In order to avoid this inhomogeneous line broadening, which is due to the anisotropic Knight shift, a NMR study of $CeAl_3$ in the temperature range from 0.4 to 10 K at a low frequency of about 1 MHz was performed recently.[22] It revealed a rapid intrinsic line broadening of the spin echo spectra below about 1.2 K, which was interpreted as the onset of a complicated type of antiferromagnetism. In contrast, a previous nuclear orientation experiment at 10 mK (Ref. 23) gave no hint of the existence of static magnetic correlations.

Our paper is organized as follows. After a short description of the experiment in Sec. II, the experimental results which were obtained for different geometries of the external field are reported in Sec. III. In Sec. IV the data are discussed and possible explanations for the observed phenomena are given. In Sec. V we finally draw some conclusions and discuss the relevance of our data with respect to a general understanding of the heavy-electron ground state.

II. SAMPLE AND EXPERIMENT

We performed standard muon-spin-rotation (μ^+SR) spectroscopy experiments[24] in zero-, transverse- and longitudinal-field geometry. The experiments were carried out at the Paul-Scherrer-Institute in Villigen, Switzerland. Our sample consisted of two polycrystalline pieces of $CeAl_3$ with total dimensions of $20\times15\times3$ mm^3. The specimens were flattened on one side by electron spark erosion and then glued to the copper cold finger of a dilution refrigerator with top-loading access. In the dilution refrigerator, the temperature could be varied from 30 mK to 2 K. Additional measurements between 1.5 and 150 K were performed in a standard ^{4}He-bath cryostat. Thermometry relied on calibrated Allen-Bradley carbon resistors and germanium resistors.

Unfortunately, the single-crystal specimens which have been available up to now have linear dimensions of only about 1 mm.[16] This, in volume, is almost three orders of magnitude less than what is required for μ^+SR measurements.

A severe problem in studies of intrinsic magnetic properties of $CeAl_3$ may occur from the presence of foreign phases which order magnetically, namely $CeAl_2$ ($T_M=3.8$ K) (Ref. 25) and Ce_3Al_{11} ($T_M=3.2$ K and $T_M=6.2$ K).[26] Within the limits of x-ray analysis, our sample was free of these phases. This claim is particularly supported by specific heat data taken above 1 K which revealed a smooth $C_p(T)$ behavior. In addition, and most convincing, the μ^+SR spectra did not show any line broadening at the respective ordering temperatures.

III. RESULTS

A. Zero-field measurements

Three zero-field μ^+-polarization spectra taken in $CeAl_3$ at 0.05, 0.5, and 1 K are displayed in Fig. 1. Most interesting is the appearance of a spontaneous muon-spin-precession frequency below about 0.7 K indicating the presence of a quasistatic (fluctuation rate <1 MHz) internal magnetic field of nonzero average value at a frac-

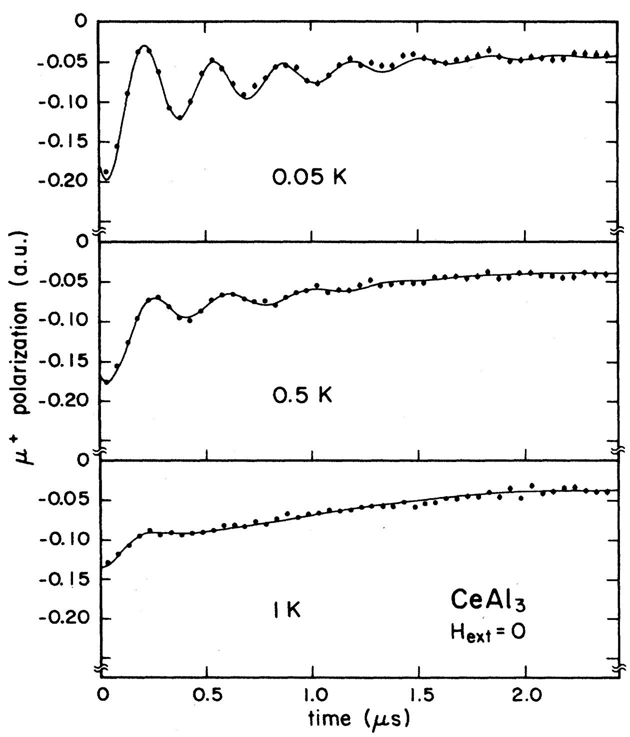

FIG. 1. μ^+ polarization as a function of time in $CeAl_3$ at 0.05, 0.5 and 1 K in zero external field. The solid lines represent three-component fits according to Eq. (1). Note that there is no oscillatory component, i.e., $N_1=0$, at 1 K.

tion of the muon sites.[7] A detailed analysis showed that the zero-field μ^+ signal, in fact, consists of three distinguishable time-dependent components below 0.7 K. Besides the oscillatory component which, according to chi-square analysis, exhibits a Lorentzian rather than a Gaussian relaxation, there are two nonoscillatory components of typical static Kubo-Toyabe form[27] which reflect muons stopped in two different environments, both with vanishing average magnetic field at the muon site. One of the latter must originate from the $CeAl_3$ target because of its large linewidth, which amounts to several inverse microseconds. The other component which has a temperature-independent linewidth of 0.6 μs^{-1} is of mixed origin, as indicated by the temperature dependence of its relative strength to be discussed below. It partly results from muons stopped in the copper cold finger of the dilution refrigerator and partly from muons stopped in $CeAl_3$. Since the muons stopped in copper and those representing the third $CeAl_3$ component appear to be subject to nearly the same magnetic field distribution, a separation of these components in zero field was not possible. This separation, however, could be achieved in transversely applied magnetic fields (see Sec. III B).

Before going into detail we would like to state that Fig. 1 already illustrates the main physical contents of this paper, namely the unexpected onset of quasistatic magnetic correlations below about 2 K. The presence of several components in the μ^+SR signal reveals that these correlations do not develop in a spatially homogeneous way over the sample. In order to prevent confusion, the reader is advised to interpret the different signal components as different magnetic surroundings of the muons rather than different crystallographic sites in the host lattice. The justification for this statement will be given later.

The zero-field μ^+ signal in $CeAl_3$ is described by the following equation:[24]

$$N(t)=N_0 e^{-t/\tau_\mu}[1+N_1 A e^{-\lambda t}\cos(2\pi\nu_\mu t+\Phi) + N_2 A G_z^{\mathrm{KT}}(\Delta_1,t) + N_3 A G_z^{\mathrm{KT}}(\Delta_2,t)]+B \ . \quad (1)$$

N_0 is a normalization constant, and $\tau_\mu = 2.2$ μs is the muon lifetime. B represents a time-independent background, A is the instrumental μ^+-decay asymmetry which was determined independently, and Φ is the phase angle. N_1, N_2, and N_3 are weight factors which measure the relative fractions of muons influenced by the three different distributions of local magnetic fields. The oscillatory component is characterized by its frequency ν_μ which is related to the average local magnetic field B_μ at the muon site by

$$\nu_\mu=(\gamma_\mu/2\pi)B_\mu \ , \quad (2)$$

with $\gamma_\mu/2\pi = 13.55$ MHz/kG, and by the parameter λ which is the Lorentzian linewidth. The general expression for the static Kubo-Toyabe (KT) term $G_z^{\mathrm{KT}}(\Delta,t)$ is given by[28]

$$G_z^{\mathrm{KT}}(\Delta,t)=\tfrac{1}{3}+\tfrac{2}{3}(1-\Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2) \ . \quad (3)$$

The linewidth Δ is related to the second moment of the isotropic local magnetic field distribution, which is assumed to be Gaussian, by

$$\Delta^2=\gamma_\mu^2\langle (B_{\mathrm{loc}}^i)^2\rangle , \quad i=x,y,z \ . \quad (4)$$

N_1 was found to be zero within the error bars above 0.7 K. Therefore, the data obtained between 0.7 and 2 K were successfully fitted using two Kubo-Toyabe functions instead of the three components of Eq. (1). The temperature dependence of the sum (N_1+N_2) is shown in Fig. 2. There is a continuous decrease from 65% at 30 mK to approximately zero at 2 K. N_3 increases by the same amount when the temperature is raised from 30 mK to 2 K. The sum $(N_1+N_2+N_3)$, which, according to Eq. (1), amounts to unity, was kept fixed in the analysis of the zero-field data.

It is important to note that the absolute values of N_1 and N_2 cannot be determined precisely by the fitting procedure of the zero-field data because of correlations among the various parameters of the model function represented by Eq. (1). The sum (N_1+N_2), however, can be obtained accurately. In order to get a qualitative picture of the respective temperature dependences of N_1 and N_2 below 0.7 K, a particular fitting procedure was applied which leads to the dashed-dotted and dashed lines in Fig. 2. In this procedure, N_1, ν_μ, λ, N_3, and Δ_2 were determined in a first step from a fit within the time interval 0.4–7 μs. In a second step N_2 and Δ_1 were determined from a fit within the total time interval 0.02–7 μs with all the other parameters obtained in the first step kept fixed. In this way the 50% fraction N_1 representing the oscillatory component turns out to be constant between 30 mK and 0.3 K. Above 0.3 K, it decreases, reaching a value of 20% at 0.7 K. The Kubo-Toyabe fraction N_2 amounts to 16% at 30 mK and increases to 20% at 0.7 K. At temperatures above 0.7 K, where N_1 is zero, N_2 continuously decreases to effectively zero at 2 K.

Figure 3 illustrates the weak temperature dependence

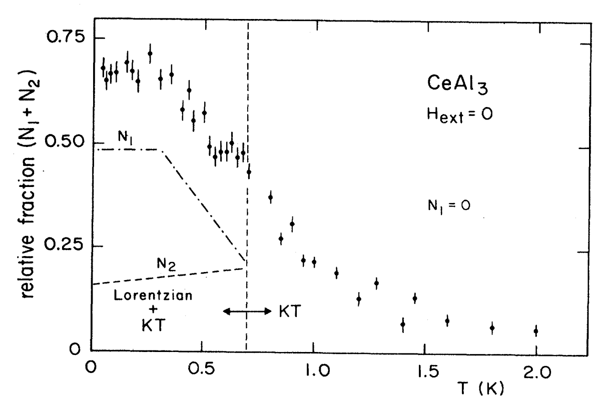

FIG. 2. Temperature dependence of the sum of the asymmetries (N_1+N_2) or the relative fractions of muons contributing to these components. Note that $N_1=0$ above 0.7 K. The dashed-dotted and the dashed lines below 0.7 K represent the values of N_1 and N_2, respectively. These values were obtained by our particular fitting procedure (see the text).

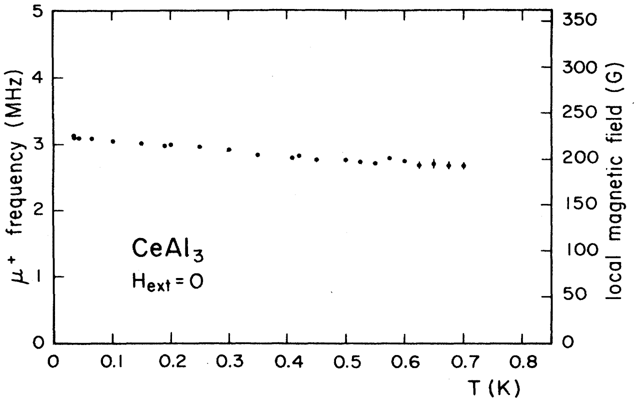

FIG. 3. Temperature dependence of the spontaneous μ^+ Larmor-precession frequency ν_μ or average local magnetic field B_μ at the muon site in zero external field.

of the muon Larmor-precession frequency ν_μ extracted from the oscillatory component below 0.7 K. A linear extrapolation of ν_μ to $T=0$ K gives a corresponding average local magnetic field of 220 G. When the temperature is raised to 0.7 K, this value decreases monotonously by about 20%.

In Fig. 4 the temperature dependence of the linewidth λ of the oscillatory component and the linewidths Δ_1 and Δ_2 of the Kubo-Toyabe components are shown. λ continuously increases from 1.8 μs^{-1} at 30 mK to 5 μs^{-1} at 0.7 K. Δ_1 decreases from 8.5 μs^{-1} at 30 mK to 6 μs^{-1} at 0.7 K, and keeps this value up to 2 K. The effective linewidth Δ_2 drawn as a dashed line is temperature independent, and amounts to about 0.6 μs^{-1}.

We would like to draw attention to the pronounced temperature dependences of the populations of the various μ^+SR-signal components. This effect turns out to be independent of the particular fitting procedure applied. The internal magnetic fields and their distributions, in contrast, are less temperature dependent. The justification of our particular fitting procedure in zero field, which may not be the only conceivable one, is finally obtained from the analysis of the transverse-field data to be discussed in the following subsection.

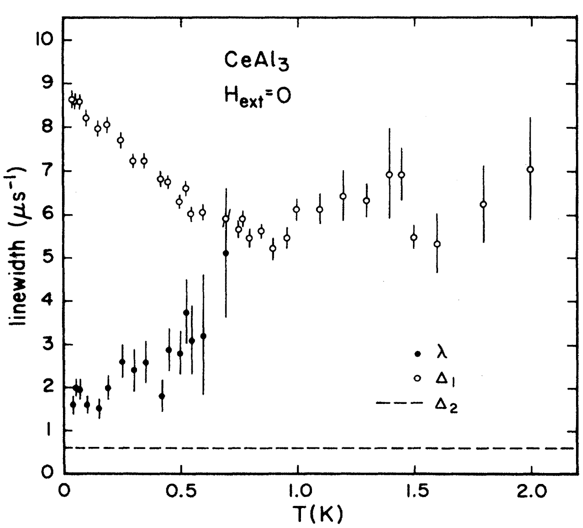

FIG. 4. Temperature dependence of the linewidths λ, Δ_1, and Δ_2 in $CeAl_3$. No external field was applied. Note that Δ_2 is an effective linewidth of two components which could not be separated in zero field (see the text).

B. Transverse-field measurements

Spectra in transverse external fields were taken between 100 mK and 2 K. In fields exceeding 1000 G, a three-component structure of the transverse-field μ^+ signal could be resolved. In contrast to the zero-field measurements, one can now clearly assign one of the three components to muons stopped in the copper cold finger of the dilution refrigerator because of its temperature-independent asymmetry A_{Cu}, frequency $\nu_{\mu,Cu}$, and its well-known Gaussian linewidth σ_{Cu}.[24] One immediately recognizes that according to the zero-field data, four μ^+-signal components in transversely applied fields below 0.7 K should be expected. All attempts, however, to fit the data taken below 0.7 K using a four-component model function failed, and also the Fourier-transformed spectra did not allow one to distinguish a fourth component. The reason for this will be discussed in Sec. IV.

The temperature dependences of the three asymmetries A_1, A_2, and A_{Cu} (normalized to the instrumental μ^+-decay asymmetry which was determined independently) or the relative fractions of muons contributing to the three components in an external field of 2000 G are displayed in Fig. 5. A_1 decreases from 60% at 0.1 K to 10% at 2 K, while A_2 increases by the same amount. The temperature-independent asymmetry A_{Cu} amounts to 25%. The corresponding μ^+ frequencies $\nu_{\mu,1}$, $\nu_{\mu,2}$, and $\nu_{\mu,Cu}$ are shown in Fig. 6. $\nu_{\mu,Cu}$ has a very small temperature-independent positive Knight shift $K_{\mu,Cu}$ of 60 ppm with respect to the externally applied field,[24] and therefore may be used for field calibration. Although $\nu_{\mu,1}$ seems to show a systematic temperature variation in Fig.

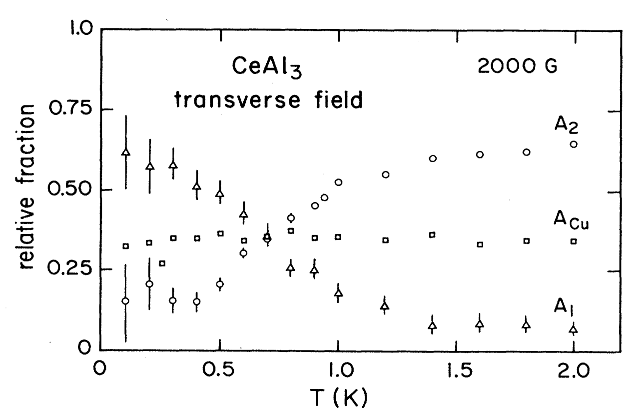

FIG. 5. Temperature dependence of the asymmetries A_1 and A_2 of the transverse-field μ^+ signal in $CeAl_3$. An external field of 2000 G was applied. A_{Cu} is the asymmetry of the background signal of the Cu target holder.

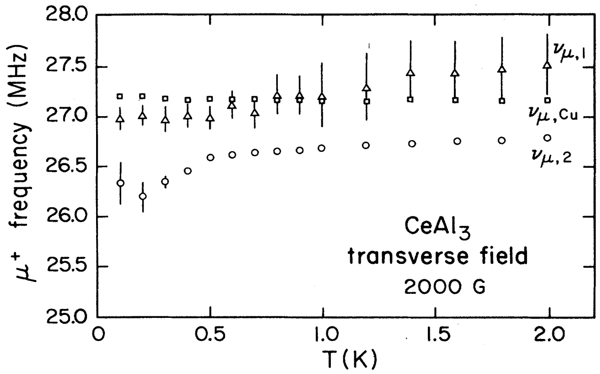

FIG. 6. Temperature dependence of the frequencies $\nu_{\mu,1}$ and $\nu_{\mu,2}$ of the transverse-field μ^+ signal in $CeAl_3$. An external field of 2000 G was applied. $\nu_{\mu,Cu}$ is the frequency of the background signal of the Cu target holder.

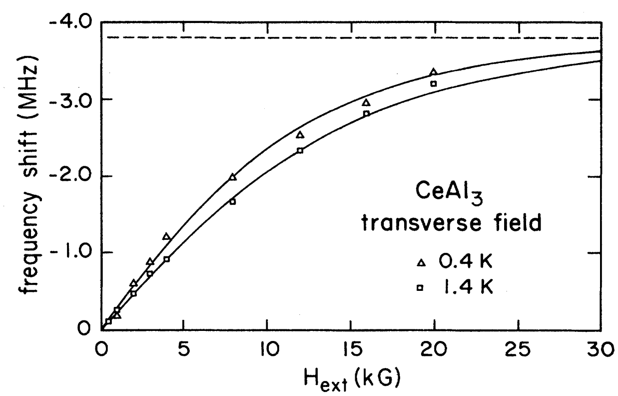

FIG. 8. Field dependence of the transverse-field frequency shift ($\nu_{\mu,2}-\nu_{\mu,Cu}$) in $CeAl_3$ at 0.4 and 1.4 K. The solid lines represent Brillouin functions B_J with $J=\frac{3}{2}$. The dashed line corresponds to the asymptotic value of 3.8($\pm$0.1) MHz.

6, it is believed that this is an artifact of the fitting program in the case that all fit parameters are left to vary freely. What can be stated with certainty is that $\nu_{\mu,1}$ is temperature independent between 0.1 and 2 K, and within its large error bars equals $\nu_{\mu,Cu}$. The frequency $\nu_{\mu,2}$ does not vary with temperature above 0.5 K, corresponding to a large negative Knight shift $K_{\mu,2}$ of about -2%. Below 0.5 K, $\nu_{\mu,2}$ clearly decreases, leading to a still larger Knight shift $K_{\mu,2}$ of about -3.2% as $T\rightarrow 0$ K. The quoted K_μ values are not corrected for demagnetization fields, which are estimated to be of the order of only some percent of the uncorrected values. Figure 7 illustrates the Gaussian linewidths σ_1, σ_2, and σ_{Cu} of the three components. The linewidth of the Cu component σ_{Cu} amounts to 0.40 μs^{-1} and is temperature independent, as expected. σ_1 is about 8 μs^{-1} at 100 mK and slightly decreases to 6 μs^{-1} at 2 K, whereas σ_2 decreases from 6 μs^{-1} at 100 mK to 0.45 μs^{-1} at 0.7 K without showing any further variation up to 2 K.

The magnetic field dependence of the frequency shift ($\nu_{\mu,2}-\nu_{\mu,Cu}$) at temperatures of 0.4 and 1.4 K is depicted in Fig. 8. It can be well approximated by a Brillouin function B_j (Ref. 29) with $J=\frac{3}{2}$ which is represented by the solid lines in Fig. 8. These data are compatible with an f moment between $0.4\mu_B$ and $0.5\mu_B$. The uncertainty of this result arises from the fact that for both temperatures the asymptotic value of 3.8($\pm$0.1) MHz (the dashed line in Fig. 8) could not be determined very accurately due to instrumental limitations of the external field strength.

The magnetic field dependences of the linewidths σ_1 at 0.4 K, σ_2 at 1.4 K, and σ_{Cu} (no difference between 0.4 and 1.4 K) are shown in Fig. 9. The special choice of the temperatures at which the field dependences of σ_1 and σ_2 are shown is implied by the fact that the relative fractions of these components dominate at the selected temperatures (see Fig. 5) and that σ_2 is still temperature independent at 1.4 K (see Fig. 7). σ_1 amounts to 6 μs^{-1} without showing any magnetic field dependence. In contrast, σ_2 linearly increases from 0.45 μs^{-1} at 1000 G to 4 μs^{-1} at 20 kG with increasing field. Also, σ_{Cu} exhibits a weak increase with increasing field from 0.40 μs^{-1} at 1000 G to 0.8 μs^{-1} at 20 kG. This latter field dependence is due to the inhomogeneity of the external field and should be sub-

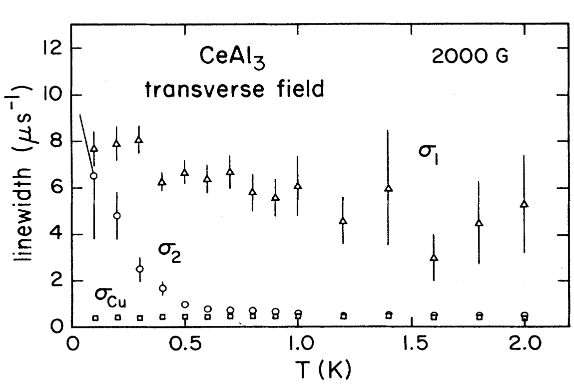

FIG. 7. Temperature dependence of the linewidths σ_1 and σ_2 of the transverse-field μ^+ signal in $CeAl_3$. An external field μ^+ signal of 2000 G was applied. σ_{Cu} is the linewidth of the background signal of the Cu target holder.

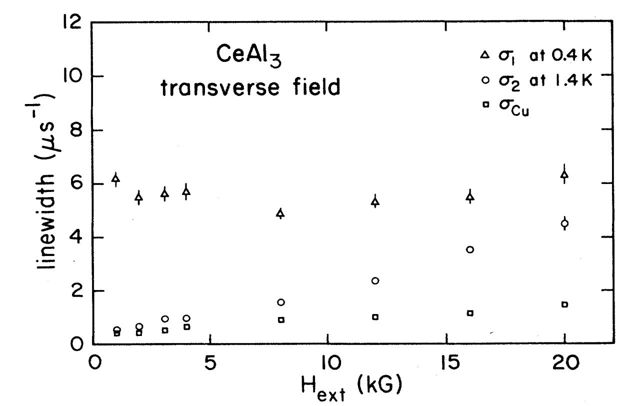

FIG. 9. Field dependence of the transverse-field linewidths σ_1 at 0.4 K and σ_2 at 1.4 K in $CeAl_3$. σ_{Cu} is the linewidth of the background signal of the Cu target holder which is temperature independent.

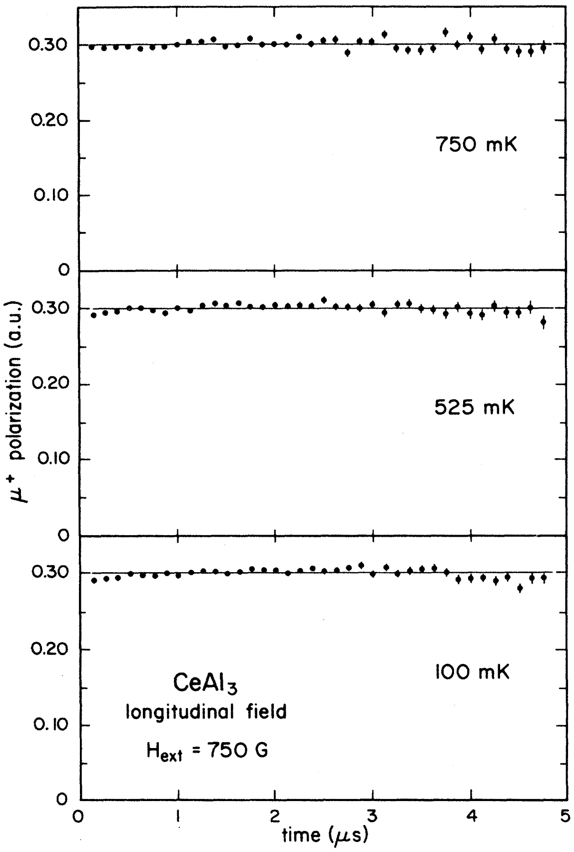

FIG. 10. μ^+ polarization as a function of time in $CeAl_3$ at 100, 525, and 750 mK. An external field of 750 G was applied longitudinally with respect to the initial μ^+ polarization.

tracted from the field dependence of σ_1 and σ_2. Since such corrections do not affect the field dependences of σ_1 or σ_2 in any substantial way, they were neglected here.

C. Longitudinal-field measurements

Measurements in longitudinal fields exceeding 750 G, which is three to four times the mean local field corresponding to the oscillatory component observed in zero external field, did not reveal a relaxing μ^+ polarization. This means that the line broadening of the different signal components in zero and transverse field must be exclusively of quasistatic origin (fluctuation rate <1 MHz) and that there is no spin-lattice relaxation by fluctuating magnetic fields on the muon time scale. For illustration three μ^+-polarization spectra taken at 100, 525, and 750 mK in a longitudinal field of 750 G are shown in Fig. 10.

IV. DISCUSSION

Since the central result of the presented μ^+SR study on $CeAl_3$, i.e., the observation of static magnetic correlations below about 2 K, has also been established in the meantime by two other experiments,[17,22] the most important question now is whether these correlations and the unconventional way they appear are driven by a truly intrinsic mechanism of this material or whether there are some extrinsic influences.

While it can be ruled out that the presence of foreign magnetic phases in our sample, i.e., $CeAl_2$ or Ce_3Al_{11}, is responsible for them, one might imagine that impurity rare-earth ions, e.g., Gd, induce local magnetic order in their vicinity. Noting, however, that at 30 mK about 65% of all muons in the sample are subject to these correlations, this trivial explanation can be safely discarded.

Furthermore, one has to consider to what extent static magnetic correlations may be influenced by the positive muon itself. Indeed, the muon provokes a lattice relaxation, thereby modifying the crystalline electric-field gradients acting on its nearest Ce neighbors and inducing quadrupolar effects. Consequently, one may expect a local variation of the electronic excitation spectrum of the Ce $4f$ electrons and possibly enhanced but certainly not static, magnetic correlations. Particularly, moment fluctuations of more distant Ce ions should cause a rapid relaxation of correlations among the muon's neighbor moments. Quadrupolar effects do not cause a splitting of the $J_z \doteq \pm m$ levels, and possible valence fluctuations would suppress coherence among ionic magnetic moments rather than stabilize it. The muon's own magnetic field at its nearest Ce neighbors amounts to less than 10 G, and is therefore too small to lead to any appreciable polarization.

These arguments make it very unlikely that the observed phenomena are influenced or even caused by extrinsic mechanisms, and strongly indicate an intrinsic phenomenon. Therefore, we interpret our μ^+SR measurements as follows. Well above 2 K, the muon spin is completely decoupled from the rapidly fluctuating Ce ionic moments and only subject to the weak static interaction with the ^{27}Al nuclear moments. This interaction causes a line broadening of $\sigma_2=0.45\ \mu s^{-1}$ in a transverse field of 2000 G and of roughly $\Delta_2=0.60\ \mu s^{-1}$ in zero external field. The observed ratio $\Delta_2/\sigma_2 \approx (1.5)^{1/2}$ (recall that Δ_2 is an effective linewidth) reveals that the zero-field second moment is influenced by the presence of a quadrupolar interaction between the ^{27}Al nuclei ($Q=0.149$ b) and the electric-field gradient produced by the positive muon,[24] and the transverse-field second moment is given only by the Zeeman interaction. In other words, the magnetic field range, where the quadrupolar interaction is negligible with respect to the Zeeman interaction, the so-called Van Vleck limit, is already reached here at a transverse field of 2000 G. At about 2 K quasistatic magnetic correlations start to develop among the f moments in a small volume of the sample. This gives rise to a broad magnetic field distribution with zero average at some muon sites, causing the strongly damped ($\Delta_1=6\ \mu s^{-1}$) Kubo-Toyabe component in zero field and the strongly damped ($\sigma_1=6\ \mu s^{-1}$) Gaussian component in a transverse field of 2000 G. According to Eq. (4), the static random local field components are of the order $\Delta/\gamma_\mu=440$ G. The experimental ratio $\Delta_1/\sigma_1=1$, which is constant up to fields of about 20 kG, can be easily understood by the following considerations. In the strong coupling limit, i.e., in the case that the exchange interaction between the f moments is much stronger than the Zeeman interaction, the equation

$$\langle (B^x_{\rm loc})^2 \rangle = \langle (B^y_{\rm loc})^2 \rangle = \langle (B^z_{\rm loc})^2 \rangle \quad (5)$$

holds for the second moments of the components of the random local magnetic fields in our polycrystalline target. Since the zero- and transverse-field relaxation functions can be written as

$$\begin{aligned} G_z^{\rm KT}(\Delta, t \to 0) &= \exp(-\Delta^2 t^2) \\ &= \exp\{-\tfrac{1}{2}\gamma_\mu^2 [\langle (B^x_{\rm loc})^2 \rangle + \langle (B^y_{\rm loc})^2 \rangle] t^2\} \end{aligned} \quad (6)$$

and

$$\begin{aligned} G_x(\sigma, t) &= \exp(-\tfrac{1}{2}\sigma^2 t^2) \\ &= \exp[-\tfrac{1}{2}\gamma_\mu^2 \langle (B^z_{\rm loc})^2 \rangle t^2] \ , \end{aligned} \quad (7)$$

one obtains with Eq. (5) the identity $\Delta = \sigma$.

With decreasing temperature, the total volume comprising these frustrated correlations increases. Below 0.7 K, the local order develops into an extended coherence, as indicated by the occurrence of a spontaneous Larmor precession of a part of the muons reflecting an average local field of about 220 G. The distribution of this average local field, which is characterized by the parameter λ, becomes narrower with decreasing temperature. In transverse fields above 1000 G, this change to extended magnetic coherence is not directly observable. A corresponding μ^+-signal component would have a linewidth of $\sigma = \gamma_\mu (2 \times 220 \text{ G}) \approx 6\ \mu s^{-1}$, and is therefore indistinguishable from the first component with $\sigma_1 = 6\ \mu s^{-1}$. Indeed, the relative fractions $(N_1 + N_2)$ in zero field and A_1 in a transverse field of 2000 G turn out to be identical within the experimental uncertainties below and above 0.7 K.

The fact that in magnetically correlated regions the transverse-field frequency $\nu_{\mu,1}$ roughly equals (note the remarks made above) the background frequency $\nu_{\mu,\rm Cu}$, whereas the frequency $\nu_{\mu,2}$, which is associated with muons stopped in a paramagnetic environment, exhibits a huge negative Knight shift $K_\mu \approx -2\%$ indicates that there are different hyperfine fields at the corresponding muon sites. In polycrystalline samples, only the isotropic part of K_μ can be measured. Assuming ideal polycrystallinity, the anisotropic parts are averaged out in the sense that they only lead to an asymmetric line broadening but not to a frequency shift.[30] Indeed, this inhomogeneous line broadening is observable in the linear field dependence of σ_2 shown in Fig. 9. *The reason for the reduced Knight shift in magnetically correlated regions is that the f moments cannot be aligned by the external magnetic field and therefore do not contribute to K_μ.*

A large negative μ^+ Knight shift in the paramagnetic regime of $CeAl_3$ which scales with the magnetic susceptibility between 6 and 130 K was already reported by Uemura *et al.*[31] In additional measurements we reproduced the Knight-shift data of these authors. In Fig. 11, our Knight-shift results and those of Uemura *et al.*[31] are

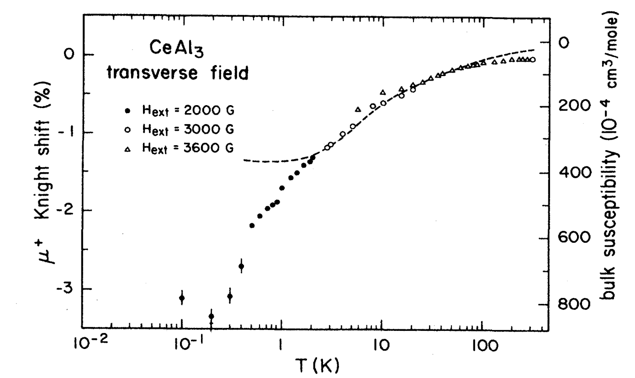

FIG. 11. Temperature dependence of the paramagnetic μ^+ Knight shift in $CeAl_3$. The data at $H_{\rm ext} = 3600$ G are taken from Uemura *et al* (Ref. 31). The dashed line represents the magnetic susceptibility of our sample.

shown together with the magnetic susceptibility of our sample (dashed line). Apparently, there is a scaling of the magnetic susceptibility of our sample and K_μ between 2 K and about 100 K. The deviation from this scaling above 100 K may perhaps be attributed to muon diffusion, while the deviation below 2 K indicates a change in the temperature dependence of the hyperfine coupling which occurs simultaneously with the development of the static magnetic correlations. The existence of large negative hyperfine fields can be explained in terms of a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between neighbor f moments mediated by a spatially oscillating polarization of itinerant electrons. The competition of the RKKY interaction with the Kondo interaction which is likely to occur in $CeAl_3$ cannot be monitored directly by our experiment. We note that the coupling constant of K_μ and χ, which roughly amounts to 2 kG/μ_B per cerium atom in $CeAl_3$, is comparable to values obtained in other $4f$ intermetallic compounds.[24] This strongly suggests localized f moments above 2 K, while below 2 K a change of the f-moment localization is indicated by the breakdown of the scaling between K_μ and χ with decreasing temperature shown in Fig. 11. Knight-shift measurements on single crystals are very desirable for a more conclusive interpretation of this interesting experimental result.

The increase of the linewidth σ_2 of the paramagnetic transverse-field component which occurs below 0.5 K is quite unexpected because there is no simultaneous change of the corresponding zero-field linewidth Δ_2 at this temperature. One should recall, however, that the paramagnetic volume fraction has already decreased to 20% at this temperature, and therefore this effect is probably not resolvable in the zero-field spectra. The most likely physical origin of this behavior is a change of the dynamics of the f-moment fluctuations in the still paramagnetic regions in the sample.

According to the longitudinal-field data there is no sign for dynamical line broadening of the other signal components. This is also supported by the temperature independence of σ_1 and Δ_1 near 2 and 0.7 K. Moment fluctuations are predicted for itinerant electrons by Fermi-liquid theory, and have been found in some other heavy-electron systems by neutron scattering.[3] If they really exist, they do not match the muon time window in the case of $CeAl_3$, and can be estimated to lie well above 10^{10} Hz.

As clearly outlined above, the simultaneous observation of different fractions of muons which are subject to different magnetic surroundings and the marked temperature dependence of the total magnetically correlated volume of the sample reflected by (N_1+N_2) or A_1 suggests that the coherence among the Ce moments develops in a spatially inhomogeneous way. This behavior can be compared to the spin freezing in a spin glass like *Cu*Mn, where similar μ^+SR results were obtained.[32] $CeAl_3$, of course, cannot be considered as a spin glass, which is characterized by randomly distributed magnetic moments whose spatially varying distances lead to frustrated magnetic interactions. This is particularly reflected by the Gaussian line shapes of the signal components observed in $CeAl_3$, because in a spin glass a Lorentzian distribution of the local field components acting on the muon is expected.[33] *Frustration effects, however, may arise from other causes and could in principle account for the observed magnetic phenomena, as we will show below.*

Unfortunately, the μ^+SR technique does not allow one to measure wave vectors. Therefore the magnitude of the correlated regions and the exact type of ordering cannot be determined here. In order to get at least an estimate of the magnitude of the involved effective static f moments and a qualitative understanding of the local magnetic structure, we performed dipolar-field calculations neglecting the presence of additional contact hyperfine fields. Figure 12 displays the hexagonal Ni_3Sn structure of $CeAl_3$. The room-temperature lattice constants are $a=6.545$ Å and $c=4.609$ Å.[34] The Ce atoms which are separated from one another by Al atoms within the planes have six nearest Ce neighbors outside the planes at a distance of about 4.426 Å and six next-nearest Ce neighbors within the planes at about 6.545 Å. The most probable interstitial sites for the positive muon in crystal coordinates are $\mu^+(1)$ at $(0,0,\frac{1}{2})$ and $\mu^+(2)$ at $(\frac{1}{3},\frac{2}{3},\frac{1}{2})$. They are marked in Fig. 12. $\mu^+(1)$ is of octahedral type and has six Al neighbors; $\mu^+(2)$ is of tetrahedral type with one Ce and three Al neighbors.

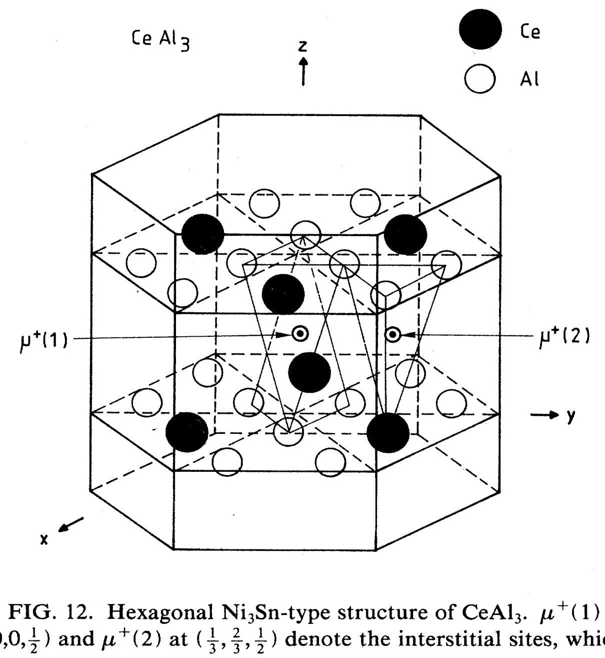

FIG. 12. Hexagonal Ni_3Sn-type structure of $CeAl_3$. $\mu^+(1)$ at $(0,0,\frac{1}{2})$ and $\mu^+(2)$ at $(\frac{1}{3},\frac{2}{3},\frac{1}{2})$ denote the interstitial sites, which are most probably occupied by the positive muon. The room-temperature lattice constants are $a=6.545$ Å and $c=4.609$ Å (Ref. 34).

Most significant information about the actual μ^+ site is obtained from calculations of the transverse-field linewidth produced by the quasistatic arrangement of nearest-neighbor ^{27}Al nuclear dipoles. The calculations were performed for the case of the Van Vleck limit, which applies here, as already pointed out. The calculated linewidths are 0.45 μs^{-1} at $\mu^+(1)$ and 0.23 μs^{-1} at $\mu^+(2)$. Because of the excellent agreement of σ_2 with the calculation for the site $\mu^+(1)$ above 0.7 K, this is considered to be the most probable muon site.

Secondly, we performed calculations of the zero-field linewidth Δ_1 under the assumption of a random magnetic field distribution caused by quasistatic nearest-neighbor Ce moments possessing an arbitrary magnetic moment of $1\mu_B$. Such an arrangement would occur in the case of frustrated magnetic interactions. The calculated values are 17 μs^{-1} at $\mu^+(1)$ and 297 μs^{-1} at $\mu^+(2)$. Also, these results imply an occupation of $\mu^+(1)$ rather than one of $\mu^+(2)$, because the normalization of the value 17 μs^{-1} calculated for $\mu^+(1)$ to the experimental value Δ_1 gives an effective f moment of $0.4\mu_B$, which is consistent with the moment deduced from the field dependence of $(\nu_{\mu,2}-\nu_{\mu,Cu})$ in transverse fields.

Furthermore, we performed dipolar-field calculations at $\mu^+(1)$ and $\mu^+(2)$ in order to find an explanation for the observation of the spontaneous Larmor-precession frequency in zero external field corresponding to a local field of 220 G. Thereby we considered both simple ferromagnetic alignments and antiferromagnetic stacking sequences of planes within which the moments are ferromagnetically coupled. The results of these calculations are listed in Table I together with the results of the linewidth calculations discussed above. It turns out that for a simple ferromagnetic alignment of nearest f neighbors having an arbitrary moment of $1\mu_B$, magnetic fields between 233 and 1407 (depending on the angle Φ between the moments and the a axis) can be obtained at $\mu^+(1)$. No net fields are found for antiferromagnetic stacking sequences of planes within which the moments are ferromagnetically coupled either parallel or perpendicular to the c axis at $\mu^+(1)$. It is obvious, however, that for more complicated antiferromagnetic arrangements there are net fields at $\mu^+(1)$ which are in magnitude comparable to those in the ferromagnetic case. Thus one can conclude that in the case of an occupation of $\mu^+(1)$, the effective f moment roughly lies between $0.1\mu_B$ and $1\mu_B$ under the assumption of ferromagnetic or complicated antiferromagnetic correlations. At $\mu^+(2)$, the fields calculated for the same arrangements lie between 5513 and 12 455 G. Note that at $\mu^+(2)$ the moments are asymmetrically distributed around the muon, and therefore a net field is obtained in any case. According to our calcula-

TABLE I. Dipolar field and linewidth calculations for an arbitrary moment of $1\mu_B$ per Ce atom in $CeAl_3$ at the most probable muon sites $\mu^+(1)$ and $\mu^+(2)$. The arrows denote the angle between the f moments and the hexagonal c axis. ϕ is the angle between the f moments and the hexagonal a axis and only given when required. For more details see the text.

	σ_{VV} (Al) (μs^{-1})	Δ (Ce) (μs^{-1})	B_{dip} (Ce) $\uparrow\uparrow$ (G)	B_{dip} (Ce) $\rightrightarrows$	B_{dip} (Ce) $\uparrow\downarrow$ (G)	B_{dip} (Ce) $\rightleftarrows$
$\mu^+(1)$ at $(0,0,\frac{1}{2})$	0.45	17	669	$\Phi=0°$: 1279 G $\Phi=30°$: 684 G $\Phi=60°$: 233 G $\Phi=90°$: 900 G $\Phi=120°$: 1407 G	0	0 G
$\mu^+(2)$ at $(\frac{1}{3},\frac{2}{3},\frac{1}{2})$	0.23	297	11 785	$\Phi=0°$: 5925 G $\Phi=30°$: 5783 G $\Phi=60°$: 6089 G $\Phi=90°$: 6510 G $\Phi=120°$: 6636 G	12 455	$\Phi=0°$: 6258 G $\Phi=30°$: 6343 G $\Phi=60°$: 6033 G $\Phi=90°$: 5610 G $\Phi=120°$: 5513 G

tions, an occupation of the site $\mu^+(2)$, which appears very unlikely as shown above, would yield an f moment which roughly lies between $0.01\mu_B$ and $0.05\mu_B$.

Since ferromagnetic correlations seem to be in contradiction to the behavior of the magnetic susceptibility at low temperatures, we suppose rather that a complicated antiferromagnetic arrangement of the magnetic moments occurs in $CeAl_3$ below 0.7 K. Our experimental results are consistent with an occupation of $\mu^+(1)$ and an effective f moment of $0.5(\pm 0.1)\mu_B$.

Frustration effects may principally occur in $CeAl_3$ as a consequence of antiferromagnetic intraplane interactions between Ce moments at a distance of 6.545 Å. This was recently pointed out by Coles *et al.*[35] in connection with an electron-spin resonance (ESR) study of the isostructural compound $GdAl_3$. In $GdAl_3$ a rapid increase of the ESR linewidth well above the antiferromagnetic ordering temperature was ascribed to such frustrated magnetic correlations. Analogously, we interpret our results in the sense that frustrated antiferromagnetic interactions are responsible for the broad field distribution with vanishing average which starts to develop below about 2 K, and to a certain fraction persists down to 30 mK.

We speculate that the driving mechanism of this gradual onset of magnetic order in $CeAl_3$ is the temperature dependence of the effective Ruderman-Kittel-Kasuya-Yosida interaction between the $4f$ moments, and not the single-ion susceptibility. Such a model was recently proposed for the interpretation of neutron-scattering data in U_2Zn_{17}.[36]

In a complementary study we were not able to find any hint for a macroscopic phase transition by searching for a distinct anomaly of the specific heat of our sample in the critical temperature range. Furthermore, no Bragg reflections could be found in a neutron-diffraction experiment.[37] The failure of these experiments is compatible with the model of magnetism proposed for $CeAl_3$ here, and emphasizes that the coherence length of the observed correlations must be very short, even below 0.7 K.

We would like to note here that the NMR results do not seem to contradict our model, although they were interpreted in a slightly different way.[22] Below about 1.2 K, the NMR spectra also consist of three resonance lines, namely a very narrow one and two differently broadened ones. In principle both methods, NMR and μ^+SR, give the same information concerning the T_2 line-broadening processes. However, to determine the relative weights of the different resonance lines using the Fourier analysis of NMR seems hopeless in the case of $CeAl_3$, but is of crucial importance here, as revealed by our experiments. Another problem occurring in the Fourier domain are baseline effects which could mask the gradual onset of magnetism between 1.2 and about 2 K in the NMR spectra. Furthermore, the possibility of performing μ^+SR in zero external field seems in our case to lead to new complementary physical information. Most significant support for the interpretation of our data is, of course, obtained from the magnetotransport measurements on single crystals which yielded a magnetic ordering temperature of 1.6 K.[17] This clearly reveals that there is not just a conventional antiferromagnetic phase transition at 1.2 K as might be concluded from the NMR data.

V. CONCLUSIONS

In conclusion, μ^+SR spectroscopy has provided unique information regarding the characterization of the electronic ground state of $CeAl_3$. The onset of frustrated quasistatic magnetic correlations below about 2 K which become partly coherent below about 0.7 K was monitored by zero- and transverse-field experiments. Longitudinal-field measurements confirmed the quasistatic nature of these correlations. From the simultaneous observation of different signal components whose relative fractions show a marked temperature dependence, it was concluded that these correlations develop in a spatially inhomogeneous frustrated way. Simple geometrical arguments can make the occurrence of frustration effects in this compound[35] plausible. The relevance of the data of $CeAl_3$ presented is emphasized by the fact that similar results were recently obtained by μ^+SR and neutron scattering in other heavy-electron compounds like UPt_3,[38,39] $CeCu_{2.1}Si_2$,[40] and $U_{0.967}Th_{0.033}Be_{13}$.[41] The lack of any obvious trivial reasons for the observed phenomena lets us conclude that the appearance of weak magnetism which may coexist with superconductivity[39–41] and involve the same kind of electrons is a general feature of heavy-electron systems. This feature has yet to be taken into account in any microscopic theory attempt to explain the ground state of these systems. Many of the low-temperature thermal and transport properties were so far interpreted as evidence for Fermi-liquid behavior. It must now be examined whether this weak magnetism can be incorporated in a Fermi-liquid-type theory of the heavy electrons. Microscopic evidence of magnetic correlations in the absence of distinct indications for a cooperative phase transition is a new, puzzling phenomenon in magnetism that appears to be of a general nature in heavy-electron physics.

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