Onset of Magnetic Correlations in $CeAl_3$ below 2 K

S. Barth and H. R. Ott
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule–Hönggerberg, CH-8093 Zürich, Switzerland

F. N. Gygax, B. Hitti, E. Lippelt, and A. Schenck
Institut für Mittelenergiephysik, Eidgenössische Technische Hochschule–Zürich, CH-5234 Villigen, Switzerland

and

C. Baines, B. van den Brandt, T. Konter, and S. Mango
Schweizerisches Institut für Nuklearforschung, CH-5234 Villigen, Switzerland
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The results of zero-, transverse-, and longitudinal-field muon-spin rotation measurements in polycrystalline $CeAl_3$ are reported. They provide evidence for static magnetic correlations in $CeAl_3$ below 2 K. From the magnitude of the local magnetic field at the muon site it was estimated that the effective Ce^{3+} moments may be of the order of $0.05\mu_B$. The magnetic correlations appear as a spatially inhomogeneous phenomenon, and are partly coherent below 0.7 K. No evidence for a cooperative phase transition is found.

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Usually, intermetallic compounds whose low-temperature thermal and transport properties are dominated by heavy-mass f electrons are divided into three classes according to the type of their electronic ground state, i.e., normal conducting and paramagnetic, magnetically ordered, or superconducting.[1] Recently it was discovered that static magnetic order involving exceptionally small moments may develop in heavy-electron systems at low temperatures.[2] In UPt_3, however, antiferromagnetic correlations appear without a discernible magnetic phase transition.[3] Apparently there is a delicate balance among the various ground states, and the above classification scheme probably needs some adjustment. In this Letter we present experimental data which give direct evidence for the occurrence of static magnetic correlations in the prototype heavy-electron system $CeAl_3$.

In $CeAl_3$ so far no sign of a phase transition has been observed down to the lowest temperatures it has been investigated, i.e., 10 mK.[4] Around 1.5 K its magnetic susceptibility changes from the Curie-Weiss behavior with an effective moment of $2.55\mu_B$/Ce that is observed at elevated temperatures[5] to an almost temperature-independent Pauli-type susceptibility below 0.5 K.[6] In between, a very shallow maximum in $\chi(T)$ at 0.65 K was observed.[6] The ratio c_p/T, where c_p is the specific heat at constant pressure, shows a distinct maximum between 0.3 and 0.5 K,[7,8] which can be suppressed, however, by application of moderate external pressure.[9] These and various other anomalies[6,10,11] in thermal and transport properties below 1.5 K were interpreted as being due to the formation of a Fermi-liquid state of quasiparticles with large effective masses.[12]

So far only few data are available from investigations of $CeAl_3$ by microscopic methods. NMR measurements[13] performed in $CeAl_3$ between 1.5 and 300 K show a change in slope in the relation between the ^{27}Al isotropic Knight shift and the bulk susceptibility below 10 K. Also, at around 10 K a broad maximum of the extracted $4f$-spin-lattice relaxation time (T_1^{4f}) was observed. The Knight-shift behavior was attributed to a temperature-dependent transferred hyperfine field caused by crystalline electric-field splitting of the $^2F_{5/2}$ cerium $4f$-electron multiplet while the behavior of T_1^{4f} was interpreted as possible evidence for correlated fluctuations of the $4f$ electrons.[13] Neutron-diffraction experiments[14] performed between 60 mk and 125 K revealed that the quasielastic spectral width increases as $T^{1/2}$ above 1 K, but is effectively constant below 1 K. Neither in these studies nor in a nuclear orientation experiment[15] was any tendency towards static magnetic correlations found.

In order to obtain further microscopic information on magnetic properties of $CeAl_3$ at very low temperatures, we have investigated this substance by muon-spin rotation spectroscopy.[16] The experiment was carried out at the Swiss Institute for Nuclear Research in Villigen, Switzerland, using the new low-temperature facility. The centerpiece of this muon-spin–rotation spectrometer is a ^3He-^4He dilution refrigerator with top-loading capability which allows us to attain temperatures down to about 5 mK. Our sample consisted of two polycrystalline pieces of $CeAl_3$ attached to the copper cold finger of the cryostat. For good thermal contact the specimens were flattened on one side by electron spark erosion and then glued to the sample holder. Sample temperatures were varied from 35 mK to 2 K. Since $CeAl_3$ forms incongruently and single-phase material is only obtained by long-time annealing, one often has to deal with the presence of two foreign phases which order magnetically, namely $CeAl_2$ ($T_M = 3.8$ K)[17] and Ce_3Al_{11} ($T_M = 3.2$ K and $T_M = 6.2$ K).[18] According to x-ray analysis, our sample was free of these phases and the muon-spin–rotation spectra did not suffer from any line-broadening effects at the above-mentioned ordering temperatures.

Our measurements showed several very surprising and unanticipated results. The most striking result was the detection of a spontaneous muon spin-precession frequency in zero external field below 0.7 K, indicating the presence of a quasistatic (spin fluctuation rate ≤ 1 MHz) internal magnetic field of nonzero average value. Typical μ^+-asymmetry spectra taken at 50 mK, 0.5 K, and 1 K are displayed in Fig. 1. Clearly visible is the oscillatory component at 50 mK and 0.5 K, while at 1 K only a nonoscillatory decay of the initial spin polarization was found. A detailed analysis showed that the μ^+ signal, in fact, consists of three time-dependent components. Besides the oscillatory component there are two nonoscillatory components, one of which originates from the $CeAl_3$ target while the second results from muons stopped in the copper sample holder and will not be considered further in the following discussion.

The signal from muons stopped in the $CeAl_3$ sample is well reproduced below 0.7 K by the following fit function:

$$N(t) = N_0 e^{-t/\tau_\mu}\{N_1[1 + A e^{-\lambda t}\cos(\omega_\mu t + \phi)] + N_2[\tfrac{1}{3} + \tfrac{2}{3}(1 - \Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2)]\} + B \,. \quad (1)$$

The first term in the curly braces describes coherently precessing muons, indicating a field distribution with nonzero average, and the second term is of typical static Kubo-Toyabe form,[19] reflecting a wide spread of local magnetic fields with zero average. N_1 and N_2 are weight factors which are measures of the relative fractions of muons being subject to the two different field distributions.

The fraction contributing to the oscillatory component turned out to be constant and about 50% between 35 mK and 0.3 K. Above 0.3 K it starts to decrease, reaching a value of 20% at about 0.7 K. N_2 is nearly temperature independent and about 20% below 0.7 K. The temperature dependence of the sum of the corresponding asymmetries is shown in Fig. 2. Above 0.7 K the two components in Eq. (1) can no longer be distinguished in the fitting procedure. Therefore the data were fitted with good success by a single Kubo-Toyabe relaxation function (see the bottom of Fig. 1). The asymmetry of this single-component μ^+ signal decreases continuously to effectively zero when the temperature is raised to 2 K (see Fig. 2). This behavior indicated that there is another temperature-dependent fraction of muons that are only depolarized by their weak static interaction with the ^{27}Al nuclei, giving rise to another Kubo-Toyabe signal. This, however, could not be extracted from the data because of the copper background signal.

The linewidth λ of the precession signal is nearly constant and about 1.5 μs^{-1} below 0.3 K. Above 0.3 K it starts to increase reaching a value of 6 μs^{-1} at 0.7 K. Δ,

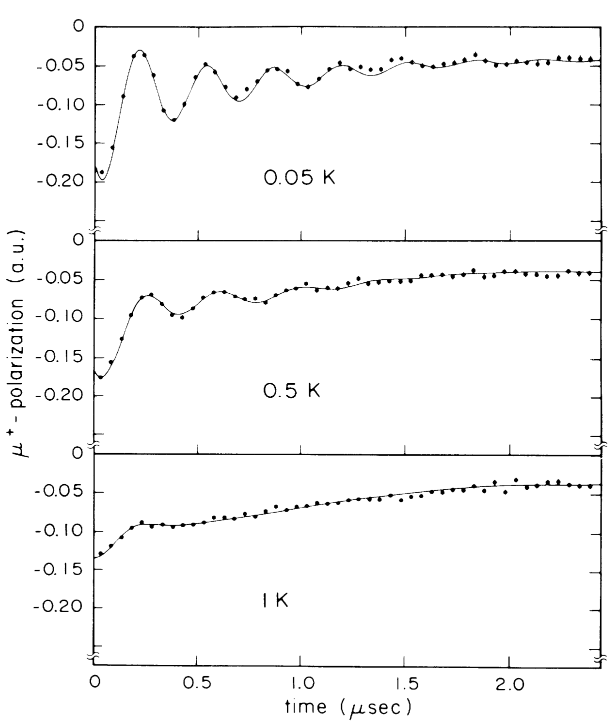

FIG. 1. μ^+ polarization as a function of time in $CeAl_3$ at 0.05, 0.5, and 1 K. No external magnetic field was applied. The solid line is a three-component fit for 0.05 and 0.5 K and a two-component fit for 1 K.

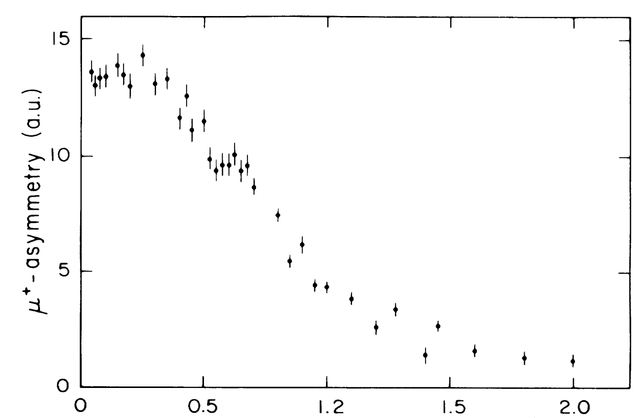

FIG. 2. Temperature dependence of the total μ^+ asymmetry in zero external field extrapolated to $t = 0$.

however, is nearly constant and about 6 μs^{-1} between 35 mK and 2 K.

Figure 3 illustrates the temperature dependence of the muon's Larmor precession frequency ω_μ extracted from the oscillatory component. At 35 mK, the lowest temperature of our measurements, the frequency reflects an average local magnetic field of 220 G at the muon site. When the temperature is raised to 0.7 K this value decreases by about 20%.

Additional measurements in longitudinally applied large fields did not reveal a relaxing μ^+ polarization. This excludes spin-lattice relaxation by fluctuating magnetic fields and underlines that the zero-field line broadening must exclusively be of quasistatic origin, i.e., correlations must survive longer in time than 1 μs. The absence of dynamically induced relaxation is also indicated by the constant linewidth of the Kubo-Toyabe component. We emphasize that, independent of the model of interpretation, only the population of distinctly evolving and relaxing polarization components varies considerably with temperature. Relaxation rates and the precession frequency are much less dependent on temperature.

While it can be ruled out that the presence of foreign magnetic phases is responsible for the observed phenomena, one question that arises immediately is whether they are an intrinsic property of $CeAl_3$ or whether they are only induced by the presence of the positive muon. Indeed, the muon will provoke a lattice relaxation, modify crystalline electric fields acting on its nearest Ce^{3+} neighbors, or induce quadrupolar effects. This may result in a local variation of the electronic energy excitation spectrum of Ce^{3+} and possibly enhanced but nonstatic magnetic correlations. A single muon-induced splitting of the lowest-lying Kramers doublet is unlikely a possible source for static magnetic correlations among a large number of Ce^{3+} moments. In addition, the particular temperature dependence in the present data, which may be correlated with the shallow maximum of the magnetic susceptibility mentioned above, points strongly to an intrinsic property.

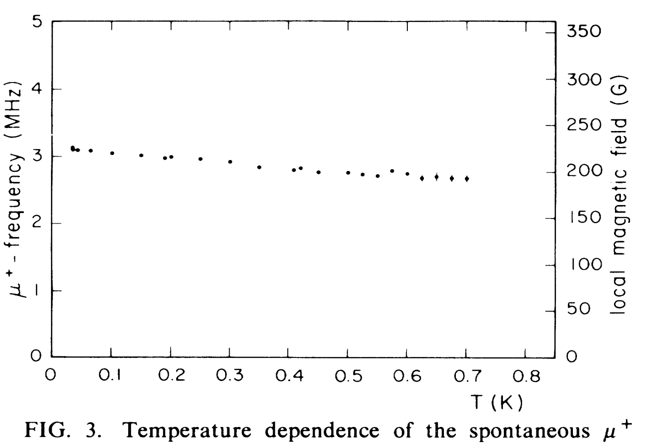

FIG. 3. Temperature dependence of the spontaneous μ^+ Larmor precession frequency or local magnetic field at the muon site in zero external field.

We interpret the results of our measurements as follows. Well above 2 K, the muon is totally decoupled from the cerium electronic moments. Around 2 K, static magnetic correlations of very short range start to develop among the $4f$ moments in a small volume of the sample. This gives rise to a broad magnetic field distribution with zero average at some of the muon sites causing the Kubo-Toyabe relaxation of about 6 μs^{-1}. With further decreasing temperature, the moment-correlated volume increases and a growing part of it even develops some coherence giving rise to a net magnetic field of 220 G at a part of the muon sites. The range of this coherence cannot be determined in this experiment. From the simultaneous observation of different fractions of muons which are obviously subject to different magnetic surroundings and the marked temperature dependence of the total μ^+ asymmetry which is a measure of the total moment-correlated volume of the sample, we conclude that the coherence among the Ce moments develops in a spatially inhomogeneous way.

In order to estimate the order of magnitude of the involved $4f$-electron moments we made dipolar-field calculations, neglecting the possible presence of presumably small contact hyperfine fields. Both ferromagnetic and antiferromagnetic alignments of neighboring Ce moments in the basal planes were considered. As possible muon sites in the hexagonal $CeAl_3$ lattice of the Ni_3Sn type,[20] we considered octahedral interstitial sites formed by six Al atoms, tetrahedral sites formed by three Al atoms and one Ce atom, and octahedral sites formed by four Al atoms and two Ce atoms. In our calculations, taking into account all possible sites, we added the contributions of ten shells of nearest neighbors. The results imply an order of magnitude of about $0.05\mu_B$/Ce which is, of course, a much smaller value than might be expected considering any possible doublet ground state of the crystal-field split $J=\frac{5}{2}$ multiplet. In addition, second-moment calculations show that $0.05\mu_B$/Ce could be made responsible for the linewidth of 6 μs^{-1} of the Kubo-Toyabe component. Such a severely reduced moment would make it difficult to detect these correlations by another method.

We were not able to find any hint of a phase transition by searching for a distinct anomaly in the specific heat of this sample in the critical temperature range, indicating that these correlations appear without a cooperative phase transition and as a spatially inhomogeneous, perhaps percolative, phenomenon. One may speculate that the gradual onset of magnetic order is driven by the temperature dependence of the effective Ruderman-Kittel-Kasuya-Yosida coupling between the $4f$ moments as was recently proposed for U_2Zn_{17}.[21] We do not have any explanation for the reduced small effective moments but would like to point out that similar small moments were recently indicated by muon-spin rotation measurements in UPt_3.[22] From this it seems that the appearance of small effective moments which exhibit static correlations is a rather general feature of heavy-electron systems and has to be taken into account as a characteristic property in the attempts to explain the ground state of these systems.

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