INDICATION FOR A DELOCALIZED PRECURSOR μ^+ STATE IN CeAl_3

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The low temperature ($\lesssim 0.6$ K) ZF- μ SR signal in the heavy electron compound CeAl_3 displays a three component structure with one component revealing the precession of μ^+ in a static internal field of 220 G. Surprisingly the phase of this signal is shifted by the order of 90° which indicates the presence of a precursor state. The precursor state manifests itself by a quickly decaying nonoscillating component. A simple two state model with zero average field in the precursor state and 220 G in the final state can describe the results except for the magnitude of the phase shift.

1. Introduction

The possibility that implanted μ^+ in the course of thermalization first populate a metastable non polaronic and highly excited state before decaying into the self trapped small polaron-like state has been considered by Emin [1] and Browne and Stoneham [2] more than a decade ago. However, no experimental evidence for this possibility has ever been found as far as we know. In this contribution we will present results obtained in the heavy electron compound CeAl_3 which, for the first time, seem to reveal the presence of a possibly delocalized precursor state before the μ^+ settles down into a particular interstitial site.

A few years ago ZF- μ SR-measurements on CeAl_3 revealed unexpectedly in part of the sample volume a magnetically ordered state below 1 K which is associated with an average field of ~ 220 G at the μ^+ -site as deduced from the observed precession signal [3,4]. The ordered domains seem to grow from zero volume above 1 K to about 80% of the volume for $T \rightarrow 0$ K. No cooperative phase transition is seen and in fact the internal field at the

Fig. 1. ZF- μ SR signals in CeAl_3 at (a) 0.01 K, (b) at 0.2 K. Also separately shown is the oscillating component following from the fit. Note the pronounced phase shifts.

μ^+ stays nearly independent of temperature as long as ordered domains remain visible. A subsequent NMR study on CeAl_3 by Nakamura et al. [5] revealed a sharp phase transition at $\sim 1\text{K}$ which result could not be confirmed by a later NMR investigation by Wong and Clark [6]. Meanwhile at Geneva one has successfully grown monocrystalline CeAl_3 which samples all showed clear cut magnetic phase transitions as evidenced e.g. from electronic specific heat measurements [7]. Transition temperatures, however, can vary widely from sample to sample. These developments induced us to apply ZF- μ SR once more to a new polycrystalline CeAl_3 -sample which was prepared at the University of Geneva .

Fig. 2. Temperature dependence of the fitted amplitudes A_{slow} , A_{fast} and A_ω . Also shown is $A_\omega \cdot \cos(180 - \varphi)$, $A_{max} - A_{1/3}$ and $A = A_\omega \cos(180 - \varphi) + A_{slow} + A_{fast}$, where, as expected, $A \simeq A_{max} - A_{1/3}$.

Fig. 3. Temperature dependence of the fitted relaxation rates λ_{slow} , λ_{fast} and λ_ω .

2. Results

Fig. 1a shows a typical ZF- μ SR-spectrum from CeAl_3 at 0.01 K obtained by using the Low Temperature μ SR-Facility (LTF) at PSI. As found previously [3,4] the signal consists of three components: two non oscillating but differently relaxing components and one oscillating component, reflecting again the presence of an internal field of ~ 220 G at the μ^+ position which is nearly independent of temperature up to 0.5 K. Fig. 2 and 3 display the temperature dependencies of the respective amplitudes (A_{slow} , A_{fast} , A_{ω}) and relaxation rates (λ_{fast} , λ_{slow} , λ_{ω}). It can be seen that the amplitude A_{slow} of the slowly decaying non oscillating signal rises steeply with temperature and accounts essentially for all implanted μ^+ for $T \geq 0.6$ K. As previously we identify this component with paramagnetic domains in the sample. The amplitude A_{fast} of the rapidly decaying and non oscillating component decreases almost by the same proportion as A_{slow} increases with rising temperature. A new features, also present but overlooked in the older data [3,4], is noticed when analysing the phase ϕ of the oscillating signal. As Fig. 5 shows the phase is temperature dependent and is shifted by 75° to $\sim 120^\circ$ (see Fig. 1b) from the value (180°) expected for the forward telescope. The corresponding shift is also seen in the backward telescope. The shift is independent of whether a gaussian or exponential decay function was used for the various components. It was not possible to describe the fast component in terms of a precession signal. Also the precessing component could not be splitted into more than one frequency. The phase shift implies that the contribution of the oscillating component to the zero time total asymmetry is only $A_{\omega} \cdot \cos(180 - \phi)$ and quite small. Nevertheless the sizable A_{ω} (see Fig. 2) indicates that quite some polarization must be transfered in time from the "fast" component with amplitude A_{fast} to the oscillating signal. In the previous work the fast component had been assigned to a spin glass like phase. The new analysis implies a quite different picture.

3. Two state model

Obviously the oscillating component evolves somehow from the rapidly decaying but non oscillating component. This induced us to consider the following model: Initially, after near thermalization, the μ^+ is found in a state with zero average field out of which it decays into a state with an average field of ~ 220 G. Assuming for simplicity that the first state

decays exponentially into the second state with rate $1/\tau_0$ and considering the fact that our sample is polycrystalline we write for the time dependent polarization of the μ^+ :

$$\begin{aligned}
 P(t) &= A_0 \{ \exp(-\lambda_{fast} t) \\
 &+ \frac{1}{4\pi} \frac{1}{\tau_0} \int_0^t \int_0^\Omega [\cos^2 \theta - \sin^2 \theta \exp(-\lambda_\omega(t-t')) \cos \omega(t-t')] \\
 &\exp(-(\frac{1}{\tau_0} + \frac{1}{T_2})t') dt' \sin \vartheta d\vartheta d\varphi \} \\
 &= A_0 \left\{ \left(1 - \frac{1}{3\tau_0 \lambda_{fast}} - \frac{2}{3} \frac{1}{\tau_0} \frac{\cos \varphi}{\sqrt{(-\lambda_{fast} + \lambda_\omega)^2 + \omega^2}} \right) \exp(-\lambda_{fast} t) \right. \\
 &\left. + \frac{2}{3} \frac{1}{\tau_0} \frac{\exp(-\lambda_2 t)}{\sqrt{(-\lambda_{fast} + \lambda_\omega)^2 + \omega^2}} \cos(\omega t - \varphi) + \frac{1}{3} \frac{1}{\tau_0 \lambda_{fast}} \right\} \quad (1)
 \end{aligned}$$

with

$$\tan \varphi = \frac{\omega}{\lambda_{fast} - \lambda_\omega} \quad , \quad (2)$$

Fig. 4. Temperature dependence of the fitted phase of the oscillating signal. Also shown is the phase that is calculated from Eq. 2 by using the values for λ_{fast} and λ_ω .

$$\lambda_{fast} = \frac{1}{\tau_0} + \frac{1}{T_2} \quad (3)$$

Here we have assumed that the first state may not only decay into the second state but may also show spin relaxation with a rate $\frac{1}{T_2}$. In addition we have assumed that the second oscillating state will relax with a rate λ_ω . The amplitude A_0 is given by $A_{max} - A_{slow}$ ($A_{max} \cong 25\%$). Hence

$$A_{fast} = \left(1 - \frac{1}{3\tau_0\lambda_{fast}} - \frac{2}{3} \frac{1}{\tau_0} \frac{\cos\varphi}{\sqrt{(-\lambda_{fast} + \lambda_\omega)^2 + \omega^2}} \right) A_0 \quad (4)$$

$$A_\omega = \frac{2}{3} \frac{1}{\tau_0} \frac{1}{\sqrt{(-\lambda_{fast} + \lambda_\omega)^2 + \omega^2}} A_0 \quad (5)$$

$$A_{1/3} = \frac{1}{3} \frac{1}{\tau_0\lambda_{fast}} A_0 \quad (6)$$

The last amplitude reflects those μ^+ whose spin happens to be parallel to the internal field. The phase φ can be calculated from λ_{fast} and λ_ω according to Eq. 2. The calculated values are shown also in Fig. 4. As can be seen measured and calculated φ show a very similar trend with temperature but are different by $\sim 50^\circ$. We do not have a good explanation for this discrepancy except to suggest that the decay function of the first state may not be simply exponential. Other decay functions may fit the first component equally well. In principle phase shifts of around 90° are not impossible if $\lambda_{fast} \simeq \lambda_\omega$.

In order to extract $1/\tau_0$ we make only use of λ_{fast} , λ_ω and the ratio A_ω/A_{fast} applying Eqs. 4,5 and 2. The result is wheresshown in Fig. 5. The emerging temperature dependence of τ_0 looks certainly strange, in particular its tendency to first decrease with increasing temperature. A more reasonable behaviour is followed by $1/T_2(T)$ (see Fig. 5). Next we calculate $A_{1/3}$ from Eq. 6. It is plotted in Fig. 2 as the difference $A_{max} - A_{1/3}$. It roughly accounts for the missing asymmetry when comparing A_{max} and $A_{fast} + A_{slow} + A_\omega \cos\varphi$.

The slow "paramagnetic" component, of course, should also emerge from the same precursor state. Applying the same two state model one can show that the precursor state has no effect on the signal in the final state as long as $\frac{1}{\tau_0} \gg \frac{1}{T_2}$.

Fig. 5. Temperature dependence of τ_0 and T_2^{-1} , extracted from λ_{fast} , λ_w and A_w/A_{fast} by use of Eqs. 2,4,5.

4. Summary

Zero field data on CeAl_3 below 0.6 K reveal clearly that the oscillating signal in the magnetically ordered state emerges from a short lived precursor state. We tried to explain the data in terms of a two state model in which an initially formed state with zero average field decays exponentially into the final state associated with an average field of 220 G at the μ^+ . Within this model we find a severe inconsistency between the observed phase shift of the oscillating signal (around 90°) and the amplitudes and relaxation rates of both the precursor and the final state. This issue remains unsolved. The transition rates between the two states are of the order of 10^7 s^{-1} , displaying a weak but puzzling temperature dependence. We suggest that the initial state is associated with rapidly moving μ^+ , which in this way “feel” essentially a zero average field. This state could be a highly excited metastable state which possibility had been postulated already more than a decade ago by Emin [1] and Browne and Stoneham [2].

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