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GROUND STATE OF $CeAl_3$ AT LOW TEMPERATURES[1]

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The results of our ^{27}Al–NMR studies of $CeAl_3$, at low frequencies and in the temperature range between 30 mK and 20 K, suggest that below 3 K gradually a new phase develops. This phase is characterised by short–range magnetic correlations among very small Ce moments, of the order of 0.05 μ_B, and coexists with the normal paramagnetic phase. Our results cast doubts on a simple Fermi–liquid description of the electronic ground state of $CeAl_3$.

1. Introduction

For a long time the low temperature electronic properties of $CeAl_3$ have been considered characteristic for a simple Fermi–liquid ground state with strongly renormalised parameters [1, 2]. No hints of a cooperative phase transition to a magnetically ordered state [2, 3] had been found down to 1 mK. Therefore, the results of subsequent μSR measurements revealing, below 2 K, the gradual development of magnetic correlations among very small Ce moments [4, 5] appeared as a surprise and triggered a series of investigations of the magnetic properties of this compound.

Results of different NMR studies have lead to different conclusions [6, 7]. While comparing data sets obtained on different materials it should be kept in mind, that the low temperature properties of $CeAl_3$ are extremely sensitive to sample preparation techniques and chemical composition. In view of this we have performed NMR measurements on a sample well–characterised by transport and thermal properties [8], and prepared from the same piece of material previously used for the μSR studies. A more detailed report of the results of our NMR studies may be found elsewhere [9].

2. Experimental Technique

We have measured ^{27}Al-NMR spectra and made spin–lattice relaxation rate T_1^{-1} experiments of randomly oriented and partially oriented $CeAl_3$ powder at several frequencies, between 1 and 10 MHz, in the temperature range between 35 mK and 20 K. The partial

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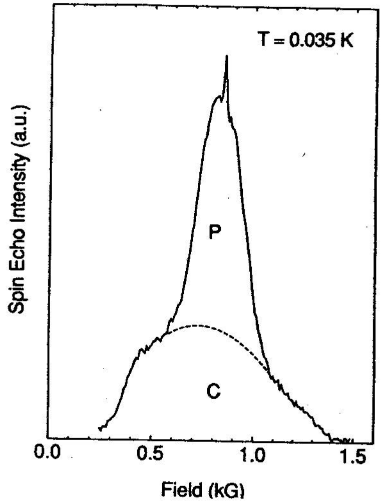

Fig.1: NMR spectrum of $CeAl_3$ at 0.035 K and a frequency of 0.968 MHz. The dashed line gives an estimate for the correlated phase (C). The normal paramagnetic contribution is denoted by (P).

orientation of our $CeAl_3$ powder was induced by the application of a large field below 1 K. The small crystallites were oriented along the c–axis of the hexagonal lattice, and the subsequent removal of the field and repeated thermal cycling did not significantly decrease the degree of orientation, which was approximately 60 % for all the measurements reported here. Standard spin–echo techniques were used in our NMR experiments, and we emphasise that the employed magnetic fields were kept at low values as not to spoil the delicately balanced ground state of $CeAl_3$. All our measurements were performed in a ^3He–^4He dilution refrigerator with the powdered sample in direct contact with the mixture either in the liquid (below 4.2 K) or the gaseous state (above 4.2 K). Considerable care was taken to minimize extraneous effects such as sample heating due to the application of the rf–pulses.

3. Results and Discussion

Above 3 K, the results of both the NMR spectra and $T_1^{-1}(T)$ indicate that our $CeAl_3$ sample contains a single phase of high–quality material, where the correlated metallic state invoking $4f$ electrons is gradually developing. This is reflected in the temperature dependence of the spin lattice relaxation rate, where the value of $(T_1T)^{-1}$ increases as the temperature decreases, scaling roughly with the C/T ratio of the specific heat C. In this temperature regime C/T also increases with decreasing$T$, reaching a maximum below 1 K. Above 3 K, our results for the NMR spectra and the relaxation rate [9, 10, 11, 12] agree very well with previously published results [6, 7, 13]. In particular, we confirm that above 10 K, Ce $4f$ electrons are in the $J_z = \pm 3/2$ crystal–field ground state with moments of the order 1 μ_B per Ce ion. The hyperfine field coupling at the Al sites is 2.5 kG per μ_B of Ce moment.

However, below 3 K both the spectra and the spin–lattice relaxation rate show important deviations from what may be expected for a simple Fermi liquid ground state. The low temperature NMR spectra (see Fig. 1) show clearly 2 different components: i) the spectrum associated with the normal paramagnetic phase (seen also above 3 K) and ii) a broad structure with a total width of the order of 1 kG. This total width is approximately field and temperature independent and cannot be explained in terms of a temperature dependent Knight shift or the onset of a quadrupolar interaction. The integrated intensity of the broad structure, expressed as a fraction $f(T)$ of the total area of the spectrum, increases with decreasing temperature at the expense of the paramagnetic contribution. This broad structure is associated with a magnetically novel phase of $CeAl_3$, which may be thought of as regions of correlated Ce moments. The static part of the correlated moments can be estimated from the total width of the spectrum of the correlated phase and the hyperfine field coupling. We obtain moments of the order 0.05 μ_B per Ce ion, too small to be seen in neutron scattering experiments. The development of the correlated phase follows approximately

$$f(T) = 0.6(1 - (T/3.2)^2). \qquad (1)$$

At the lowest temperature the correlated phase reaches 60 % of the total sample volume. We conjecture that this phase consists of very small regions, with linear dimensions of the order of 20 Å, which are embedded in the normal paramagnetic phase. As the temperature lowers, the number of these small regions increases, in a manner analogous to phase transitions occurring in some disordered magnetic materials. In these systems there is a temperature region of coexistence, of the paramagnetic and ordered phases, where small sections of the sample undergo individually a first order phase transition [14]. In other words, one may also speak of a progressive separation of two magnetically inequivalent phases. The results of the spin lattice relaxation rate measurements also indicate the presence of two different phases. The relaxation rate associated with the normal paramagnetic part follows, below 1 K, the expected Korringa law with $(T_1T)^{-1} \sim 70\,(\mathrm{K\,sec})^{-1}$. This value is roughly 100 times larger than the Korringa constant of Al metal. The relaxation associated with the correlated part is governed by a distribution of smaller relaxation rates. This distribution is characterized by a parameter τ, with $(\tau T)^{-1}$ decreasing with decreasing temperatures, even below 1 K. This result implies substantial differences between the dynamics of the $4f$ electrons in the two phases. In particular, it indicates an increasing degree of correlation among $4f$ moment fluctuations in the correlated phase. The relative portions of both phases, paramagnetic and correlated, obtained from the relaxation rate measurements agree well with the intensity ratio obtained from the spectra.

4. Conclusion

¿From the results of our NMR spectrum and spin-lattice relaxation rate measurements, we conclude that the ground state of $CeAl_3$, below 3 K, invokes two coexisting and very different magnetic phases, in agreement with conclusions drawn from previous μSR experiments.

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