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LOW TEMPERATURE MAGNETIC PROPERTIES OF CeNiSn

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It is shown that the results of recent μSR experiments on CeNiSn down to 11 mK can be understood in a picture of quasi-critical features emerging from a very small difference between ground state energies of the spin-liquid and the magnetic phases.

1. In the field of strongly correlated electron systems the class of compounds named "Kondo insulators" or "Kondo semiconductors" attract special interest. CeNiSn is considered the most prominent and instructive material amongst them [1]. The classifying names were coined on account of the observation of a sharp upturn in electrical resistivity at low temperatures which was explained by the presence of a narrow charge gap of the order of 5 K. However, these classifying terms appear not to be adequate in view of recent measurements down to less than 0.1 K using the highest purity single crystalline samples available today [2]. The upturn in resistivity is now absent and below 1 K a T^2 dependence is observed [3]. Other recent findings include a linear temperature dependence of the specific heat [4] and a Korringa law for the ^{119}Sn nuclear spin relaxation rate below 1 K as seen by NMR [5]. CeNiSn possesses a carrier density which is quite reasonable for a metallic compound and furthermore the density of states (DOS) is characteristic for a moderately heavy heavy fermion (HF) system. The established behavior at higher temperatures, especially a T^3 dependence of the NMR relaxation rate and a T^2 dependence of specific heat (see, for example [1]) remain unaffected.

A recent paper [6] proposes the rather general idea that the most characteristic feature of systems like CeNiSn is the presence of a low-lying crystalline electric field (CEF) level embedded inside the HF band. The hybridization of the CEF state with the continuous spectrum of HF excitations predetermines the appearance of spectral properties which allows one to understand not only the various anomalous temperature dependencies, but also the restoration of normal HF behavior in the presence of a strong magnetic field. Simple arguments led to the conclusion that the hybridization produces an only partially open gap in momentum space (pseudogap). Under these aspects the coexistence of normal HF behavior at very low temperatures and pseudogap properties at intermediate temperatures are a natural outcome. In consequence, studies of the behavior of CeNiSn at very low temperature are of special significance to obtain information on the HF state in itself, since then the influence of the pseudogap on the HF properties is weak, except for the simple effect of a partial decrease of the DOS. As will be shown, the μSR experiments carried out down to 0.01 K provide just such information.

2. A presentation of the μSR measurements under discussion as well as experimental details can be found in two separate publications [7, 8]. We concentrate

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on the results from measurements on a single crystal with the a-axis parallel to the muon beam. The external fields was always applied along the beam axis, that is, we deal with the condition $B \parallel a$ throughout. Consequently, switching from longitudinal to transverse μSR geometry required the rotation of the muon spin. This geometrical feature is relevant, because the a-axis stands out magnetically. For example, neutron studies have shown that the spin configuration is quasi one-dimensional with predominant spin orientation along a [9, 10]. CeNiSn crystallizes in an orthorhombic lattice (space group $Pn2_1a$) [1]. It has been shown that we deal with a muon which rests on its stopping site throughout its lifetime. The stopping site of the muon is not exactly known but an interstitial hole at the center of the pentagon of ions in the $z = 1/4$ ionic plane is the most likely place. However, the knowledge of the exact stopping place is of little relevance for the following interpretation of the μSR data. The most important features revealed in the μSR experiments are the following: (a) Static magnetic order is absent. (b) The transverse muon spin relaxation rate λ rises with decreasing temperature in a fashion typical for the relaxation behavior in a paramagnet near the magnetic transition temperature (see, for example [11]). (c) The electronic spin correlations responsible for the muon depolarization are of dynamic nature even at lowest temperatures. (d) The muon Knight shift K_μ exhibits first an increase in magnitude with lowering the temperature and then saturation at very low temperatures. In addition, the dependence on applied field is not linear, as should be the case for a free paramagnet.

3. The μSR results just summarized, together with the unusual sensitivity of all results to temperature changes at very low T as well as to relatively small magnetic fields led us to the conjecture that one has observed the manifestation of a very small difference in the ground state energy of the spin-liquid phase and a magnetically ordered phase that causes a quasi-critical character of behavior. This should be equivalent to the appearance of an effective Curie temperature, located formally in the negative temperature region ($-T_*$). Consequently, in the mean field approximation (MFA) the static magnetic susceptibility acquires the form

$$\chi(T) = \frac{A}{T + T_*}. \tag{1}$$

The muon spin depolarization rate λ in the motionally narrowed regime is proportional to the relaxation time τ_S of the fluctuating electron spin system (for weak non-uniform demagnetizing fields and in the absence of any dipole interaction with surrounding nuclei – as is the case in CeNiSn). A localized muon senses local spin fluctuations. This means that τ_S can be obtained from the decay in time of the spin correlation function $< S^i(R = 0, t) \cdot S^i(R = 0, 0) >$. Within the MFA the result can be found directly via the (k, ω)–Fourier transform of the general magnetic susceptibility [12]:

$$\lambda \propto \tau_S \propto \sqrt{\chi}(T). \tag{2}$$

Details of a more general analysis of the critical situation are given, for example, in [13].

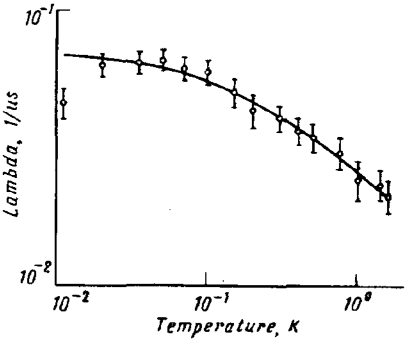

Fig.1

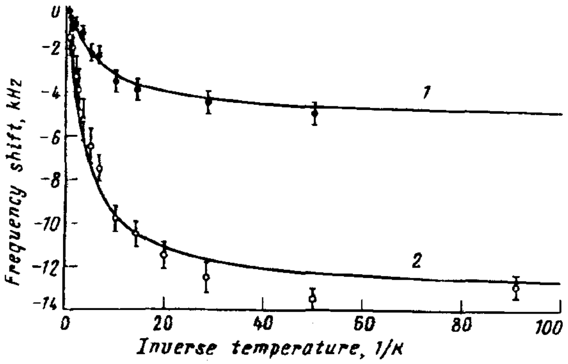

Fig.2

Fig.1. Fit (as explained in text) to the temperature dependence of the muon spin relaxation rate λ measured in 1 kG transverse field. Data are from [7]

Fig.2. Fits (as explained in text) to the temperature dependence of the muon Knight shift in 200 G (curve *1*) and 1000 G (curve *2*). Note that an inverse temperature scale is used. Data are from [8]

In Fig. 1 a fit of eqs. (2) and (1) to the temperature dependence of λ is shown. The data are well reproduced and the fit returns:

$$T_* = 0.15(2)\,\mathrm{K}. \tag{3}$$

The smallness of T_* must be emphasized. This value is just a reflection of the small energy difference between the spin-liquid and the magnetic states. It is intimately connected to the quasi-critical picture used.

The muon Knight shift is defined as

$$K_\mu \propto B\chi(T) \tag{4}$$

with B being the applied field. Expression (4) together with eq. (1) and using the value for T_* given in (3), explains the experimental data for small magnetic fields quite well as curve 1 in Fig. 2 demonstrates for the temperature dependence of the shift measured in $B_L = 200\,\mathrm{G}$.

The small value of T_* makes the results sensitive to fairly moderate applied magnetic fields B. The static magnetic susceptibility at finite B and T in MFA can be expressed in the following approximate form:

$$\chi(T,B) \approx \frac{A}{(T+T_*) + \mathrm{C}\cdot T_*^{1/3}(\mu_B B)^{2/3}} \tag{5}$$

where C is some constant. This expression gives the correct result in the two limiting situations, namely when $B \to 0$ and when the second term in the denominator dominates over the first one (see [14]). Curve *2* in Fig. 2 shows a fit to the temperature dependence of the frequency shift at $B_L = 1\,\mathrm{kG}$ on the basis of eqs. (4) and (5). For the constant we find the estimate C=1.33 when $\mu_B B$ is expressed in temperature units (0.067 K/kG).

The combination of eqs. (5) and (2) predicts a decrease of muon spin relaxation rate with increasing field. This trend had indeed been seen in measurements of

λ in longitudinal fields as Table demonstrates. One immediately notices the scale of characteristic magnetic field to be $B \approx 1$ kG. The data from Table also expose very clearly the drop of relaxation rate between T=0.04 K and T=0.25 K (in correlation with the value of T_* as given in (3)) or when B approaches ~ 1 kG.

Longitudinal field dependence of λ in (μs^{-1}) at very low temperatures

B_L (G)	11 mK	40 mK	250 mK
10	–	0.020	–
20	–	0.020	0.005
100	0.025	0.023	0.003
500	0.014	–	–
1 000	0.002	0.001	–
10 000	0.000	–	–
50 000	0.000	–	–

It should be mentioned at this point that magnitudes of $\lambda \sim 0.005\ \mu s^{-1}$represent about the lower limit of what can be measured reliably in a standard μSR experiment. Hence measurements of details of the variation of λ with B_L are beyond experimental feasibility.

From eq. (5) we further conclude that for $T < T_*$ and already for rather small magnetic fields, the applied field will strongly influence K_μ. Instead of the usual linear field dependence we now have:

$$K_\mu \propto B^{1/3}. \tag{6}$$

The thus predicted sharp reduction in the field dependence of K_μ at $B \gtrsim 1$ kG and $T < 0.1$ K is displayed in Fig. 3. The line represents the behavior predicted by eqs. (4) and (5) with T_* taken from eq. (3). At high temperatures ($\approx$ 1 K) the expected linear dependence of K_μ is still not recovered (see [8]). This can not be explained in the same fashion as above because the model is not applicable in this temperature region. The found characteristic values of T and B allow to estimate that the effective moment on the Ce ions is not small.

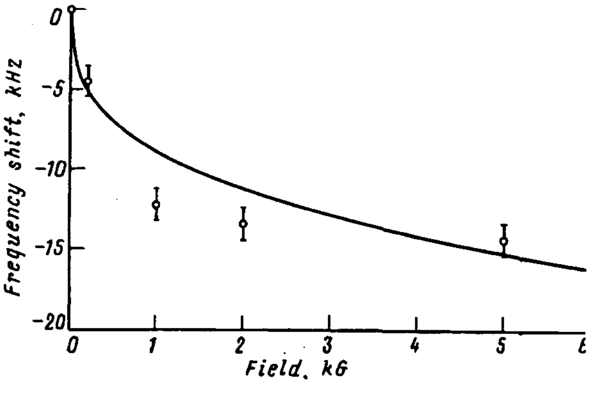

Fig.3. Fit (as explained in text) to the field dependence of the muon Knight shift measured at 33 mK. Data are from [7, 8]

We would also like to point out that the above interpretation of the dependence of λ on B_L assumes that the fast relaxation limit $\omega_\mu \tau_S \ll 1$ (ω_μ is muon Larmor frequency) is always valid and that the field alters only τ_S. Usually one discusses a dependence of λ on B_L in the opposite situation, namely that with increasing field one approaches the limit $\omega_\mu \tau_S = 1$ and thus observes a reduction in depolarization rate. This approach has been used in [7] and led to very slow rates and in consequence to smaller magnetic moments on Ce. In view of the consistent picture of the temperature dependence of λ and K_μ with quasi-critical behavior, the application of eq. (5) seems justified here.

The fact that we move within the fast limit $\omega_\mu \tau_S \ll 1$ allows one to ignore the dispersion of relaxation time. It might not be the case, however, for the intrinsic relaxation of electronic spins. This presumably is the explanation of the strong dependence of λ on applied field in the transverse field configuration as presented in [8]. In this case the relaxation rate λ is connected with fluctuations of the magnetic field acting on the muon in the direction of the applied field which was parallel to the a-axis. We had already mentioned that the spin configuration has quasi-Ising character with the predominant spin orientation along the a-axis. For the decrease of the instantaneous non-uniform distribution of the local magnetic field at the muon, one needs a reorientation of electronic spins in the applied magnetic field. Then, in the framework of real processes and if $\omega_e \tau_S > 1$, the relaxation rate starts to behave as

$$\lambda \propto \omega_e^2 \tau_S.$$

Although this could qualitatively explain the strong increase of the transverse relaxation rate with magnetic field, it is clear that the real picture is more complicated and requires more elaborate considerations.

Finally, we briefly address the question of impurities. In principle we can not absolutely exclude the scenario where spin impurities play a role. For example, in such a case we could explain the behavior of the frequency shift as function of magnetic field alternatively. However, we were not able to find a consistent picture which allows the explanation of all experimental results as a whole. Moreover, it was argued in [7] that impurities can not be the primary cause of the observed μSR signal and, in particular, not if they are present in small clusters as described in [2]. μSR measurements on the quasi-ternary materials $Ce_{1-x}La_xNiSn$ [7] and $CeNi_{1-x}Pt_xSn$ [unpublished] (with x being in a range where the pseudogap is suppressed) gave essentially similar results as undoped CeNiSn. Hence we are reasonably confident that the intrinsic low temperature behavior of CeNiSn has been observed.

4. Undoubtedly, the above analysis has qualitative character. Nevertheless, the reasonable correlation between theoretical estimates and the unusual experimental results, seems to support the initial idea about the very small difference between ground state energies of spin-liquid and magnetically ordered phases in CeNiSn. In consequence the picture of quasi-critical features at very low temperature emerges. The origin of the spin-liquid state at $T = 0$ is one of the most subtle problems in the physics of HF systems. In the past, there have been some arguments in a favour of a small energy difference between such a phase and a magnetically ordered one (for the Kondo lattice case – see, for example [15]). There was until now no experimental evidence for that. Apparently, in this aspect CeNiSn is a very attractive object.

It is worthwhile to make a few additional remarks. We have purposely used above the mean field results because all events considered are outside the fluctuation region. On the other hand, numerous studies of traditional magnetic substances demonstrate that mean field results for the critical region prove to give correct descriptions in a wide temperature interval above T_C (see, for example, [11]). For the system under consideration there is an additional favorable factor for that, namely the small value of T_* compared to an effective interaction in the spin subsystem. The latter is of the order of the Kondo temperature (T_K) or at least intermediate between T_K and T_*.

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