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 terms of the quasi-critical features which emerge from a very small difference between the ground-state energies of the spin-liquid and the magnetic phases. © *1995 American Institute of Physics.*

1. In the field of strongly correlated electron systems the class of compounds called "Kondo insulators" or "Kondo semiconductors" attract special interest. CeNiSn is considered the most prominent and instructive material among them.[1] The classifying names were coined because of the observation of a sharp increase in the 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 to be inadequate in view of the recent measurements down to less than 0.1 K, using highest-purity, single-crystal samples available today.[2] The increase in the 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] The compound CeNiSn possesses a carrier density which is reasonable for a metallic compound. Furthermore, its density of states (DOS) is characteristic of 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 the specific heat (see, for example, Ref. 1), remain unaffected.

A recent paper[6] proposes the rather general idea that the most characteristic feature of the systems like CeNiSn is the presence of a low-lying crystalline electric field (CEF) which is 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 dependences, but also the restoration of the normal HF behavior in the presence of a strong magnetic field. Simple arguments led to the conclusion that the hybridization produces only a partially open gap in momentum space (a pseudogap). Under these conditions the coexistence of normal HF behavior at very low temperatures and the pseudogap properties at intermediate temperatures is a natural consequence. Study of the behavior of CeNiSn at very low temperatures is therefore of special significance in order to obtain information on the HF state itself, since the influence of the pseudogap on the HF properties is weak, except for the simple effect of a partial decrease of the DOS. We will show that the μSR experiments, carried out down to 0.01 K, provide 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 will concentrate on the results of the measurements of a single crystal whose a axis is parallel to the muon beam. The eternal field is always applied along the beam axis, i.e., we deal with the condition $B\|a$ 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 the a axis.[9,10] The compound CeNiSn crystallizes in an orthorhombic lattice[1] (space group $Pn2_1a$). 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 of the relaxation behavior in a paramagnet near the magnetic transition temperature (see, for example, Ref. 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 a lowering of 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 belief that we have observed the manifestation of a very small difference in the ground-state energy of the spin-liquid phase and a magnetically ordered phase that accounts for a quasi-critical behavior. This should be equivalent to the appearance of an effective Curie temperature, which is 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, nonuniform demagnetizing fields and in the absence of any dipole interaction with the surrounding nuclei—as is the case in CeNiSn). A localized muon senses local spin fluctuations. This means that τ_S can be determined from the decay (in time) of the spin correlation function $\langle S^i(R=0,t)\cdot S^i(R=0,0)\rangle$. Within the MFA the result can be found directly via the (k,ω) Fourier transform of the general magnetic susceptibility:[12]

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

The details of a more general analysis of the critical situation are given, for example, in Ref. 13.

Figure 1 shows a fit of Eqs. (1) and (2) to the temperature dependence of λ. The data are well reproduced and the fit is

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

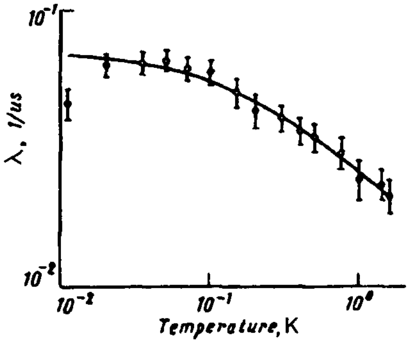

FIG. 1. Fit (as explained in the text) to the temperature dependence of the muon spin relaxation rate λ, measured in a 1-kG transverse field. The data are taken from Ref. 7.

The small value 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 follows:

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

where B is the applied field. Expression (4), together with Eq. (1) and the value of T_* given in (3), explains the experimental data for small magnetic fields quite well. Curve *1* in Fig. 2 shows the temperature dependence of the shift measured in $B_L=200$ 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 an approximate way:

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

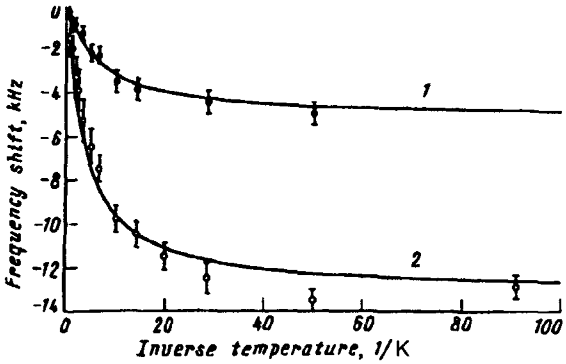

FIG. 2. Fits (as explained in the text) to the temperature dependence of the muon Knight shift in 200 G (curve *1*) and 1000 G (curve *2*). Note that an inverse temperature scale is used. The data are taken from Ref. 8.

TABLE I. 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	-	-

where C is a constant. This expression gives the correct result in the two limiting cases; specifically, when $B \to 0$ and when the second term in the denominator dominates over the first term (see Ref. 14). Curve *2* in Fig. 2 shows a fit of the temperature dependence of the frequency shift at $B_L = 1$ 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.

Equations (5) and (2) predict a decrease of the muon spin relaxatioin rate with increasing field. This trend has indeed been seen in measurements of λ in longitudinal fields, as Table I demonstrates. We immediately see that the scale of the characteristic magnetic field is $B \approx 1$ kG. The data from Table I also show very clearly the decrease of the 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.

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

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

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

The predicted sharp reduction in the field dependence of K_μ at $B \gtrsim 1$ kG and $T < 0.1$ K is shown in Fig. 3. The line represents the behavior predicted by Eqs. (4) and (5), with T_* taken from Eq. (3). At high temperatures (≈ 1 K) the expected linear dependence of K_μ is still not recovered (see Ref. 8). This behavior cannot be explained in the same fashion as above, because the model is not applicable in this temperature region. The characteristic values of T and B allow us to conclude that the effective moment of the Ce ions is not small.

We would also like to point out that the interpretation of the dependence of λ on B_L given above assumes that the fast relaxation limit $\omega_\mu \tau_S \ll 1$ (ω_μ is the muon Larmor frequency) is always valid and that the field changes only τ_S. Usually, one discusses the dependence of λ on B_L in the opposite case; specifically, with increasing field, one approaches the limit $\omega_\mu \tau_S = 1$ and thus observes a reduction in the depolarization rate. This approach, which was used in Ref. 7, led to very slow rates and thus to smaller

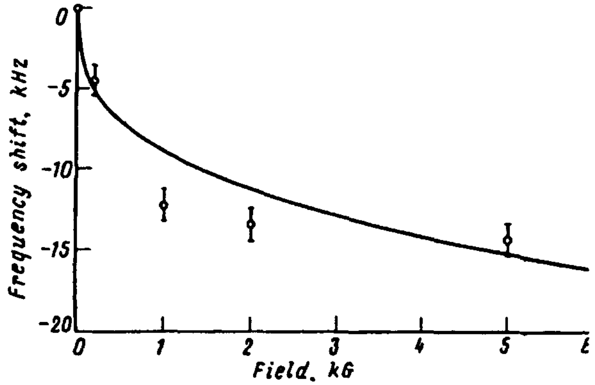

FIG. 3. Fit (as explained in the text) to the field dependence of the muon Knight shift, measured at 33 mK. The data are taken from Refs. 7 and 8.

magnetic moments of 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 are within the fast limit, $\omega_\mu \tau_S \ll 1$, allows us to ignore the dispersion of the relaxation time. This might not be the case, however, for the intrinsic relaxation of the electron spins. This presumably is the explanation of the strong dependence of λ on the applied field in the transverse field configuration, as presented in Ref. 8. In this case the relaxation rate λ is connected with the fluctuations of the magnetic field acting on the muon in the direction of the applied field, which is parallel to the *a* axis. We have already mentioned that the spin configuration has a quasi-Ising charcter, with the predominant spin orientation along the *a* axis. For the decrease of the instantaneous nonuniform distribution of the local magnetic field at the muon we need a reorientation of the electron spins in an applied magnetic field. In the framework of real processes and if $\omega_e \tau_S > 1$, the relaxation rate will then behave as follows:

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

Although this relation can qualitatively explain the strong increase of the transverse relaxation rate with the 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 cannot absolutely exclude the scenario in which the spin impurities play a role. For example, in such a case we could explain alternatively the behavior of the frequency shift as a function of the magnetic field. 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 Ref. 7 that impurities cannot be the primary cause of the observed μSR signal and, in particular, if they are present in small clusters, as described in Ref. 2. The μSR measurements of the quasi-ternary materials[7] $Ce_{1-x}La_x$ NiSn and $CeNi_{1-x}Pt_xSn$ [unpublished] (where x is in the range where the pseudogap is suppressed) gave essentially similar results as those for undoped CeNiSn. Hence, we are reasonably confident that the intrinsic low-temperature behavior of CeNiSn has been observed.

4. Clearly, the analysis given above has a qualitative character. Nevertheless, the reasonable correlation between the theoretical estimates and the unusual experimental results seems to support the initial idea about the very small difference between the ground-state energies of the spin-liquid and the magnetically ordered phases in CeNiSn. As a result, a picture of the 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 favor of a small energy difference between such a phase and a magnetically ordered one (for the Kondo lattice case—see, for example, Ref. 15). No experimental evidence has so far been obtained. In this respect, 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 here are outside the fluctuation region. On the other hand, many studies of ordinary magnetic substances demonstrate that mean field results for the critical region give correct descriptions in a wide temperature interval above T_C (see, for example, Ref. 11). For the system under consideration there is an additional favorable factor: The value of T_* is small compared to the effective interaction in the spin subsystem. The latter is on the order of the Kondo temperature (T_K) or is at least in the range between T_K and T_*.

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