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Study of the magnetic properties of $Ce_3Pd_{20}Si_6$ compound

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Abstract

We report μSR studies on a $Ce_3Pd_{20}Si_6$ polycrystalline sample. Zero-field measurements were undertaken to gain information on the magnetic ordering at low temperatures. Below 0.4 K the increase of the muon-spin depolarization rate reflects the development of a quasi-static ordering of magnetic moments of electronic origin probably randomly oriented. In transverse-field studies a clear frequency shift was observed. This fact may be ascribed to the increase of the total-magnetic moment of the superparamagnetic cube containing 8 Ce2 atoms and their ferromagnetic ordering with decreasing temperature.

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The recently synthesized [1] systems $Ce_3Pd_{20}X_6$ (X = Ge, Si) manifest unusual physical properties which categorize them as magnetic Kondo systems. Firstly, the electrical resistivity ρ in these compounds shows a Kondo-like behaviour, namely, below the temperature T_ρ, the temperature dependence $\rho(T)$ follows the relation [4] $\rho(T) = A - B\log(T)$, where A and B are numerical parameters; $T_\rho \simeq 10$ K for $Ce_3Pd_{20}Ge_6$ and $T_\rho \simeq 50$ K for $Ce_3Pd_{20}Si_6$. Secondly, the electronic specific heat coefficient is very large (about 8 J/Ce mol K^2) in $Ce_3Pd_{20}Si_6$ [2] and rather enhanced (about 0.7 J/Ce mol K^2) in $Ce_3Pd_{20}Ge_6$ [3]. Interestingly, $Ce_3Pd_{20}Si_6$ shows also magnetic anomalies both at rather high temperature $T_{magn} \simeq 50$ K [4,5] and at low temperature $\simeq 0.15$ K [2].

Nikiforov et al. [5] have developed a 'molecular magnetism' model to explain the coincidence of the characteristic temperatures T_ρ and T_{magn} in $Ce_3Pd_{20}Si_6$. According to this model, there are two relatively separated cerium subsystems in this compound. The $Ce_3Pd_{20}Si_6$ structure is presented in Fig. 1. Only the cerium positions are shown because the total number of atoms per unit cell is quite large (1 1 6). The sites of Ce1 form a face-centered 'large' cube with other atoms inside the cell. The atoms of Ce2 make up a 'small' cube with only Pd atoms inside.

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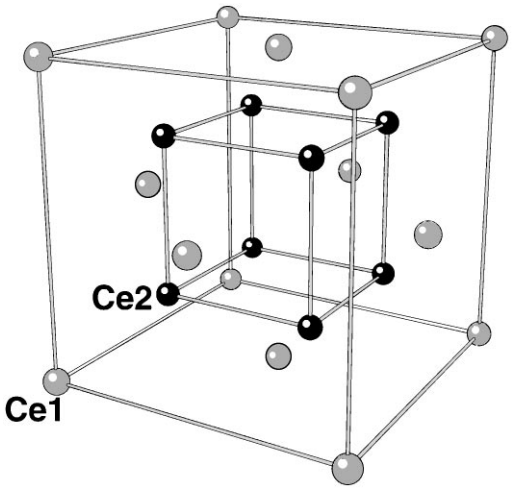

Fig. 1. The unit cell of $Ce_3Pd_{20}Si_6$. Only Ce positions are shown.

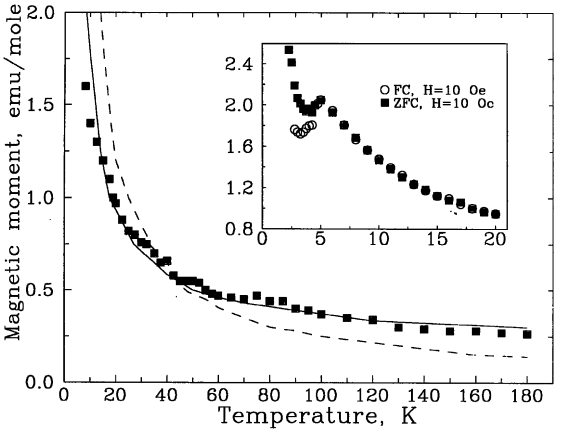

Fig. 2. Temperature dependence of the magnetization in $Ce_3Pd_{20}Si_6$.

One of the subsystems involves Ce^{3+} ions in Ce2 [1] positions which form 'small' cubes inside the unit cell. Within the framework of the 'molecular magnetism' model, Ce2 atoms should interact preferably inside the 'small' cubes. Ions of each cube make up magnetic 'molecules' with a magnetic moment that increases with decreasing temperature and undergo antiferromagnetic-like ordering at T_{magn}.

The second cerium subsystem consists of Ce1 ions. These ions are less magnetically active because they have Si as the nearest neighbors. In this model the Ce1 atoms may mostly play the role of Kondo scattering centers for the conduction electrons. Ordering in the Ce2 sublattice near T_{magn} removes the Ce1–Ce2 interactions which are competitive with the Kondo ones. This could explain the enhanced increase in electrical resistivity due to Kondo scattering of the conducting electrons in $Ce_3Pd_{20}Si_6$ after the magnetic transition.

The magnetic susceptibility measurements for $Ce_3Pd_{20}Si_6$ show a pronounced peak at 0.15 K which is ascribed to a magnetic phase transition [2]. Moreover, the existence of a remanent magnetic moment indicates that the ordering is of the spin-glass like type.

The polycrystalline samples of $Ce_3Pd_{20}Si_6$ were prepared by using a melting technique in an arc furnace in argon atmosphere as described in Ref. [1]. The samples were annealed under argon atmosphere (about 24 days). The crystal structure of the samples, determined by X-rays analysis, is the same as reported in Ref. [1].

The magnetic properties of $Ce_3Pd_{20}Si_6$ were studied by using a standard commercial DC SQUID magnetometer as a function of temperature (2–200 K) and of magnetic field (10 Oe to 30 kOe).

Fig. 2 shows the magnetic moment M versus the temperature T for the sample $Ce_3Pd_{20}Si_6$, which was cooled down to 5 K in zero-magnetic field (a magnetic field $H = 10$ Oe was then applied). The anomaly of $M(T)$ at $T_{m1} \simeq 50$ K is clearly present, though it appears more smooth in comparison with analogous measurements reported in Ref. [5]. It is important to note that $M(T)$ at high field (30 kOe) does not show any anomaly at T_{m1}. Another magnetic anomaly, namely the non-linear behaviour of the magnetization $M(H)$ at low fields (below 2 kOe), was also confirmed in the present magnetic measurements. The dashed line in Fig. 2 represents the contribution of $3N_A$ free cerium ions with a cerium effective magnetic moment $\mu_{eff} = 2.54\ \mu_B$. Remarkably, at high temperature (above 45 K) one observes $M(T) > M_{free}$, whereas at low temperature the opposite relationship $M(T) < M_{free}$ takes place. The calculations within the framework of 'molecular' magnetism model (solid line on Fig. 2) reasonably describe $M(T)$.

There is not a marked difference in $M(T)$ measured in zero- or field-cooling experiments for temperatures above 5 K. However, below 5 K distinct cooling conditions influence the behaviour of $M(T)$, as shown in the inset. The antiferromagnetic-like anomaly with a maximum of the magnetic moment at 5 K is more pronounced in the field-cooling experiment.

Zero-field μSR measurements were undertaken to gain information about the magnetic ordering at low temperatures. The experiments were performed on the surface-μ^+ beamline πM3 using the LTF-setup at the Paul Scherrer Institute (Villigen, Switzerland) [6].

The depolarization function was represented by a Gaussian function $G(t) = \exp(-\sigma^2 t^2)$. The temperature dependence of the depolarization parameter σ is shown in Fig. 3. We observe a partial recovery of the polarization in longitudinal-field measurements, with fields up to 25 kOe. This proves the dynamic nature of part of the muon-spin depolarization. Below 0.4 K an increase of the depolarization rate represents the development of quasi-static ordering of magnetic moments of electronic origin most probably randomly oriented.

Crystal-electric-field calculations show that the Ce moments may undergo frustrations, which could explain such a spin-glass behaviour. The depolarization caused by such a distribution should be described by a Kubo–Toyabe function with the typical $\frac{1}{3}$ recovery of polarization. The absence of full recovery is usually caused by the fluctuations of the magnetic moments.

We also performed transverse-field μSR measurements on the muon decay channel μE1 with the GPD-setup at the Paul Scherrer Institute (Villigen, Switzerland). The precession time spectra $A(t)$ are described by the sum of two signals $A(t) = A_s \exp(\lambda_s t)\cos(\omega_s t + \phi) + A_b \exp(\lambda_b t)\cos(\omega_b t + \phi)$, where the background signal $A_b(t)$ corresponds to the muon stopping in the sample holder and cryostat walls. The value of the external magnetic field during these measurements was controlled by the muon spin precession frequency of the background signal. The measurements were performed in external fields from 1 to 5 kOe. A clear frequency shift was seen at a temperature below 60 K. The temperature dependence of the magnetic field seen by the muons (at 3 kOe) is shown in Fig. 4. The dashed line on this corresponds to the external magnetic field $B = \omega_b/\gamma_\mu$. For other field values a similar behaviour was observed. These measurements were performed with two samples and similar results were obtained.

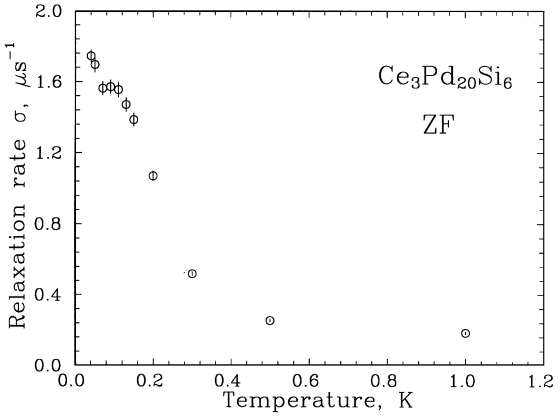

Fig. 3. Temperature dependence of the depolarization rate σ in zero field.

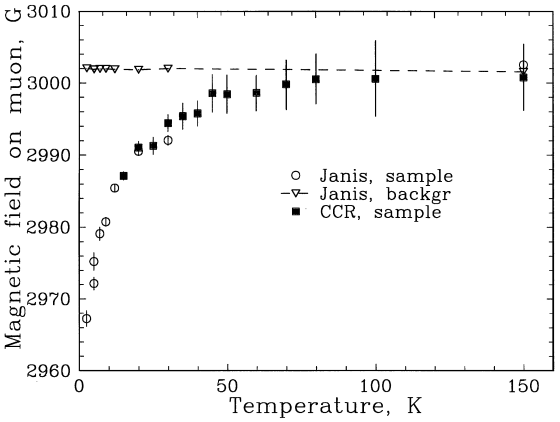

Fig. 4. Temperature dependence of the internal magnetic field acting on the muon in $Ce_3Pd_{20}Si_6$.

This fact may be ascribed to an increase of the total moments of the superparamagnetic cube (SPC) containing 8 Ce2 atoms and to their ferromagnetic ordering with decreasing temperature. The μ Knight-shift on Pd is too small to explain our results. Direct evidence of the superparamagnetic cube formation would be proved by measuring the

deviation of the frequency shift from the linear scaling with the external field due to the increase of the total moment of the cube. No such effect could be observed.

Test measurements with the nonmagnetic-related compound $La_3Pd_{20}Si_6$ were performed. The absence of muon-spin frequency shift in this sample stresses the Ce contribution in the origin of the frequency shift.

All our results agree with the model of 'molecular magnetism' based on the consideration of the 'magnetic cube' containing eight Ce2 ions coupled by ferromagnetic exchange interaction. The existence of ferromagnetic cubes explains why the experimental $M(T)$ exceeds the 'free ions' magnetic moment at high temperatures. However these cubes should interact with each other in an antiferromagnetic manner to provide the decrease of the total magnetic moment of Ce2 subsystem below approximately 50 K. We have obtained reproducible results with transition-like anomalies on two samples of $Ce_3Pd_{20}Si_6$. Further experiments at low magnetic fields are required for the direct evidence of the superparamagnetic cubes. It seems that all magnetic anomalies can be observed only at sufficiently low magnetic field.

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