Successive phase transitions in a Kagomé-like heavy-fermion compound, CePdAl

2003 J. Phys.: Condens. Matter 15 S2267

(http://iopscience.iop.org/0953-8984/15/28/366)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) S2267–S2270 PII: S0953-8984(03)62617-2

Successive phase transitions in a Kagomé-like heavy-fermion compound, CePdAl

M Nishiyama[1], A Oyamada[1], S Maegawa[1], T Goto[1] and H Kitazawa[2]

[1] Graduate School of Human and Environment Studies, Kyoto University, Yoshida-Nihonmatsu-cho, Kyoto 606-8501, Japan
[2] National Institute for Materials Science (NIMS), 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

Received 12 November 2002
Published 4 July 2003
Online at stacks.iop.org/JPhysCM/15/S2267

Abstract

The heavy-fermion compound CePdAl exhibits an antiferromagnetic phase transition at 2.7 K (T_N). Ce atoms in CePdAl form a distorted Kagomé lattice in the c-planes. The ordered spin structure in the Kagomé plane is of a partially disordered type, which has a third paramagnetic spins. The $\boldsymbol{k}$-vector is $(1/2, 0, \tau \approx 0.35)$, which is incommensurate along the c-axis. Frustration effects in the Kagomé plane are responsible for this partially disordered structure, although the origin of the incommensurability is not clear at present. Whether or not a magnetic phase transition occurs at lower temperatures is a question of considerable interest. ^{27}Al nuclear magnetic resonance measurements have been performed at low temperatures to investigate the transition below T_N. There is no remarkable change in the spin-echo spectra below T_N down to 0.5 K. On the other hand, the spin–lattice relaxation rate shows a small anomaly around 1 K. This strongly suggests that there is another phase transition below T_N. Some possible origins of successive transitions are discussed.

1. Introduction

Spin fluctuations have recently attracted attention as the basis for quantum critical behaviour as well as the origin of the cooper pair attraction in high-T_c superconductors. Magnetic systems which have geometrically competitive interactions exhibit large spin fluctuations. Such magnetic systems are called geometrically frustrated magnets. Kagomé lattice antiferromagnets have been well known as two-dimensional geometrically frustrated systems.

CePdAl shows heavy-fermion behaviour with a geometrically frustrated Kagomé-like lattice [1]. The crystal structure of CePdAl is hexagonal ZnNiAl type, with space group $P\bar{6}2m$. The Ce ions form a Kagomé-like lattice in the c-plane, in which elemental triangles of the Kagomé lattice rotate on a C_3 axis of symmetry, and the hexagons of the Kagomé

0953-8984/03/282267+04$30.00

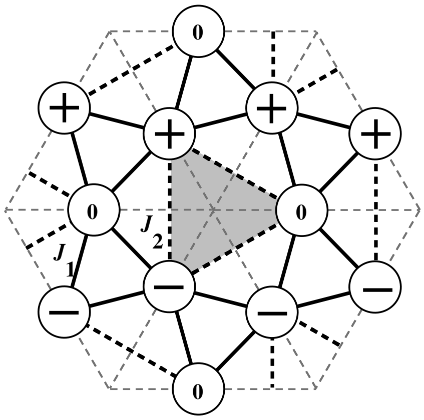

Figure 1. The magnetic structure of CePdAl. Ce atoms form the Kagomé-like lattice. J_1 and J_2 indicate the nearest-neighbour and next-nearest-neighbour interactions, respectively. The shaded area indicates the frustrated triangle [2, 3].

lattice are distorted. The lattice parameters are $a = 7.2$ Å and $c = 4.2$ Å [2]. The nearest-neighbour distance between Ce atoms is 3.7 Å in the c-plane and 4.2 Å along the c-axis. The ion–ion distance in the c-plane is smaller than that along the c-axis. CePdAl shows antiferromagnetic ordering at $T_N = 2.7$ K. The ordered spin structure observed in neutron diffraction measurements [3] is shown in figure 1.

One third of the spins remains paramagnetic in the ordered spin structure, and the magnetic propagation vector $\boldsymbol{k}$ is $(1/2, 0, \tau \approx 0.35)$, which is incommensurate along the c-axis.

The ordered spin structure in the Kagomé plane is of a partially disordered type as mentioned above. This is quite similar to the spin structure in Ising-type triangular lattice antiferromagnets, although the phase transition occurs at lower temperature and the spins are in a ferrimagnetic state. Thus, partially disordered spins reflect the presence of geometrical frustration. This peculiar spin structure has been explained by means of a model Hamiltonian which includes ferromagnetic nearest-neighbour interactions, antiferromagnetic next-nearest-neighbour interactions in the lattice plane, and Kondo screening [4]. In this model, the nearest-neighbour interactions are ferromagnetic and do not compete with other nearest-neighbour interactions. The next-nearest-neighbour interactions are antiferromagnetic. They form triangle networks and compete with each other (see figure 1). The frustration is mainly caused by antiferromagnetic next-nearest-neighbour intra-plane interactions in CePdAl. Therefore, it is supposed that spins remain partially disordered at low temperatures owing to both the frustration effect and the Kondo screening.

On the other hand, the incommensurate spin structure is not explained by the above-mentioned model. Whether or not a magnetic phase transition occurs at lower temperatures is a question of considerable interest. In this paper, we report NMR measurements performed down to 500 mK to investigate the existence of an extra phase transition below T_N.

2. Experimental results and discussion

The spin-echo spectra and the spin–lattice relaxation rate (T_1^{-1}) of ^{27}Al ($I = 5/2$) were obtained using a phase coherent-type spectrometer. The magnetic field was applied along the c-axis of a free oriented powder sample. The operating frequency was around 14 MHz.

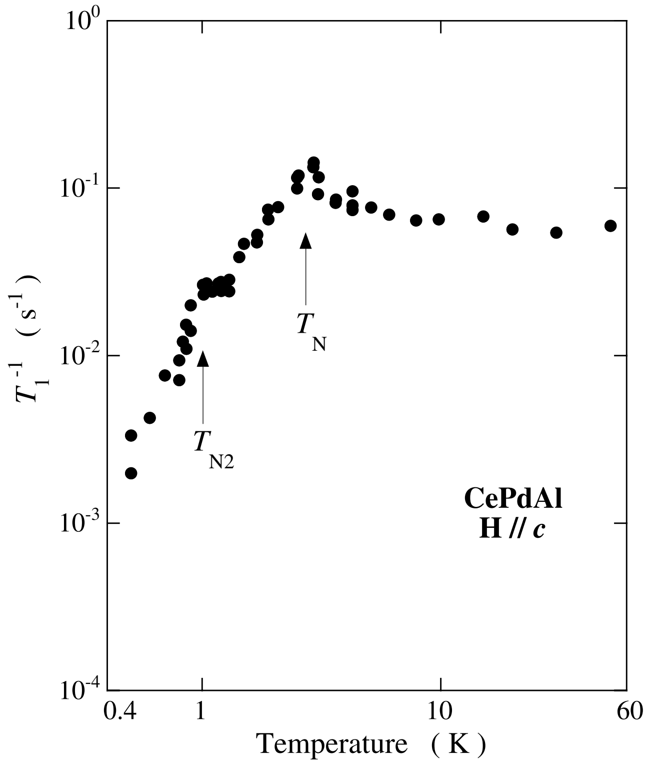

Figure 2. The spin–lattice relaxation rate of ^{27}Al around 14 MHz.

The temperature dependence of T_1^{-1} is shown in figure 2. T_1^{-1} in the high-temperature region increases slightly as the temperature approaches T_N, and shows a maximum at T_N. The rate above T_N exhibits a similar temperature dependence to Heisenberg Kagomé lattice antiferromagnet, jarosite-type compounds [5]. Jarosites show long-range magnetic ordering at $T_N \sim 60$ K, and the magnetic structure is a 120° spin configuration because of geometrical frustration. At low temperatures, T_1^{-1} decreases sharply and is explained by a two-magnon process. Above T_N, the temperature dependence of T_1^{-1} is approximately proportional to $\exp(-\Delta/T)$, which is a weak dependence compared with common critical behaviour. This suggests that short-range spin correlations may develop as the temperature approaches T_N, and the spins are strongly fluctuating. CePdAl and jarosites are similar in terms of the two-dimensional geometrically frustrated system. Short-range spin correlation may develop as the temperature approaches T_N. This confirms that two-dimensionality and geometrical frustration play important roles in the spin fluctuations of CePdAl. Below T_N, T_1^{-1} decreases sharply as the temperature decreases and shows a small anomaly around $T_{N2} \sim 1$ K. This strongly suggests that there exists another phase transition below T_N.

The spin-echo spectra above T_N separate into five uniformly spaced peaks through the electric quadrupole effect at the ^{27}Al site. Below T_N, the spin-echo spectra split into seven peaks, and each peak becomes broad as shown in figure 3. These results indicate that the magnetic ordering is not so simple. The spin-echo spectra are quite similar above and below T_{N2}. This behaviour suggests that the spin structure is not changed much at T_{N2}. Thus, the 4f moment at a third of the Ce atoms remains paramagnetic at the lowest temperatures. The phase

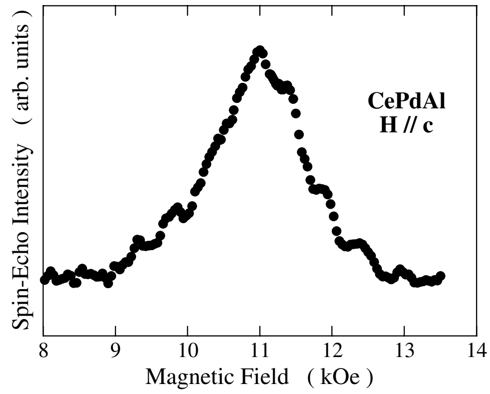

Figure 3. The ^{27}Al spin-echo spectrum of CePdAl at 1.43 K obtained using an NMR frequency of 12.36 MHz.

transition around T_{N2} is suggested to be a lock-in transition, in which the magnetic propagation vector $\boldsymbol{k}$ along the c-axis is locked into a certain value.

In conclusion, CePdAl and the jarosites show similar dynamical properties above T_N. Below T_N, jarosites show long-range order, whereas the 4f moments at a third of the Ce atoms in CePdAl remain paramagnetic. We find a second phase transition around 1 K, where the incommensurate $\boldsymbol{k}$-vector along the c-axis may undergo a slight change.

References

[1] Kitazawa H, Matsushita A, Matsumoto T and Suzuki T 1994 *Physica* B **199/200** 28

[2] Hulliger F 1993 *J. Alloys Compounds* **196** 225

[3] Dönni A, Ehlers G, Maletta H, Fischer P, Kitazawa H and Zolliker M 1996 *J. Phys.: Condens. Matter* **8** 11213

[4] Núñez-Reguerio M D, Lacroix C and Canals B 1997 *Physica* C **282–287** 1885

[5] Nishiyama M, Morimoto T, Maegawa S, Inami T and Oka Y 2001 *Can. J. Phys.* **79** 1511