NEUTRON SCATTERING STUDY ON MAGNETIC ORDER AND MAGNETIC EXCITATIONS OF A LOCALIZED URANIUM COMPOUND $U_3Pd_{20}Si_6$*

Y. Koike†, N. Metoki‡, Y. Haga

Advanced Science Research Center, JAERI, Tokai, Ibaraki, 319-1195, Japan

K.A. McEwen

Department of Physics and Astronomy, UCL
Gower Street, London WC1E 6BT, UK

M. Kohgi, R. Yamamoto

Department of Physics, Tokyo Metropolitan University
Hachioji, Tokyo 192-0397, Japan

N. Aso

Neutron Scattering Laboratory, ISSP, University of Tokyo
Ibaraki 319-1106, Japan

T. Komatsubara, N. Kimura and H. Aoki

Center for Very Low Temperature Science, Tohoku University
Sendai 980-8545, Japan

(Received July 10, 2002)

The magnetic structure in a localized $5f$ system $U_3Pd_{20}Si_6$ was studied by means of neutron diffraction. We revealed a remarkable collinear structure and found a new type of spin–flop transition due to the collinear coupling. Spin wave excitation was observed in a whole Brillouin zone. We concluded that the low energy quasi-elastic response is the excitation of the quasi-particles due to hybridization between $5f$ and conduction electrons.

PACS numbers: 61.12.–q, 75.30.–m

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

† JST Corporation, Kawaguchi, Saitama 332-0012, Japan.

‡ Department of Physics, Tohoku University, Aoba, Sendai 980-8578, Japan.

1. Introduction

Actinide based intermetallic compound is one of the most attractive systems in the field of strongly correlated electron systems [1]. Although most uranium compounds exhibit itinerant character of $5f$ electrons, an increasing number of uranium compounds which display localized moments are discovered. We found a new localized $5f$ intermetallic compound $U_3Pd_{20}Si_6$ very recently [2]. A crystalline electric field (CEF) excitation [3,4] provides direct evidence for the localized nature with the $5f^2(U^{4+})$–Γ_5 triplet ground state [2,4]. Our previous study [3] reported a successive magnetic ordering; uranium spins on the simple cubic $8c$ lattice order antiferromagnetically below $T_N = 19\,K$, while the fcc–$4a$ sites show ferromagnetic ordering below $T_C = 2\,K$. The purpose of this study is to reveal the magnetic structures and excitations in this localized $5f$ system.

2. Experimantal

Neutron scattering experiments were carried out on triple-axis spectrometers, TAS-2 ($E_i = 13.7\,meV$) and LTAS ($E_f = 3.5\,meV$) at the research reactor JRR-3 in Japan Atomic Energy Research Institute, JAERI.

3. Result and discussion

Fig. 1 shows the H–T phase diagram [5] determined in our neutron diffraction study [6]. In "AF"-phase only uranium spins on $8c$ sites order antiferromagnetically. The uranium spin on $4a$ site has eight nearest neighbors on $8c$ site with the same number of up and down spins, hence the exchange field is canceled out. It is the reason why the $4a$ sublattice remains paramagnetic.

The $4a$ site shows ferromagnetic ordering below T_C, coexisting with the $8c$ antiferromagnetic ordering in the "AF+FM"-phase. Our neutron scattering study revealed a collinear structure with the moment direction parallel to $\langle 100 \rangle$. We found that the antiferromagnetic moment follows the direction of the inter-penetrating ferromagnetic moment, when a weak external field is applied. This is direct evidence for the collinear interaction between the $4a$ and $8c$ sites. Higher order exchange and/or quadrupole interactions are considered as the candidate for this collinear coupling.

The $8c$ spins exhibit a new type of spin–flop transition for $H \sim 5\,T$, due to the uniaxial anisotropy induced by the collinear interaction.

As summarized in the table of Fig. 1, the moment size on $8c$ site $1.8\mu_B/U$ is consistent with Γ_5 ground state ($2\mu_B/U$), while $4a$ ferromagnetic moment shows a strong suppression of about $1\mu_B/U$ for $H = 0$. The $4a$ moment recovers to $1.64\mu_B/U$ for $H = 10\,T$. In "AF+FM"-phase a molecular field is

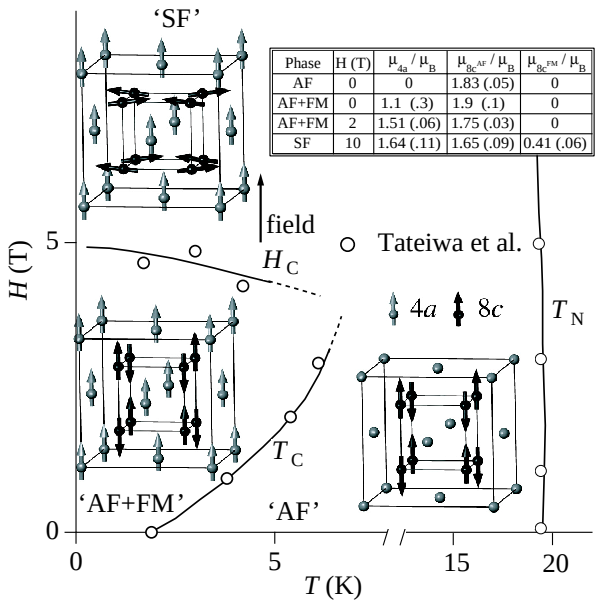

Fig. 1. H–T phase diagram for the magnetic structures of $U_3Pd_{20}Si_6$. Inset shows the magnetic moment in each phase.

induced on 8c site from the ferromagnetically ordered 4a site. This molecular field might perturb the 8c antiferromagnetic order. Therefore, the 4a ferromagnetic ordering could be suppressed so as not to disturb 8c antiferromagnetic ordering and the 4a moment would be recovered to the saturation moment under external field. The magnetic moment consistent to the ground state indicate that this compound is a prototype of the localized $5f$ magnet with $5f^2$ configuration. The image plots of the magnetic excitations around the zone center $Q = (111)$ are shown in Fig. 2. We observed clear antiferromagnetic and ferromagnetic spin wave excitations, indicating the strong localized character of $5f$ electrons. The existence of a clear spin wave in the whole Brillouin zone is quite unusual in uranium intermetallic compounds. The molecular field from 4a site splits antiferromagnetic spin wave on 8c site into two modes in "AF+FM"-phase. A dispersive ferromagnetic spin wave above T_C would be the result of the suppression of the 4a ferromagnetic ordering. This strong ferromagnetic fluctuation is consistent with a broad specific heat anomaly around T_C [2].

We found a low energy quasi-elastic component observed around the antiferromagnetic zone center. A similar low energy component has been observed in a heavy fermion superconductor UPd_2Al_3 [7,8] and an itinerant

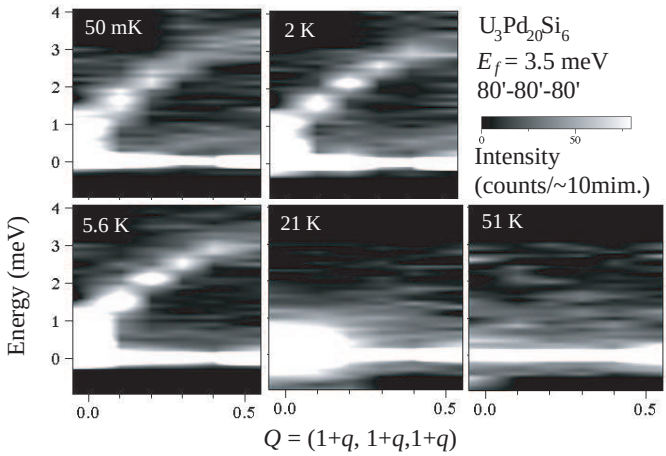

Fig. 2. Image plot of the magnetic excitations.

$5f$ antiferromagnet UGa_3 [9]. This component is considered as the excitation of the quasi particles due to the hybridization between $5f$ and conduction electrons. Although a phenomenological mean field coupling model is applicable for the magnetic excitations, the microscopic origin and mechanism for the remarkable two component magnetic excitation remain open question. Further experimental and theoretical studies are necessary in order to understand the nature of the quasi particles in $5f$ intermetallic compounds.

REFERENCES

[1] See, *e.g.*, V. Sechovsky and L. Havela, *Handbook of Magnetic Materials*, Ed. K.H.J. Buschow, Elsevier Science, Amsterdam 1998, vol. 11, p. 1.

[2] N. Tateiwa *et al., J. Phys. Soc. Jpn.* **69**, 1517 (2000).

[3] N. Tateiwa *et al., J. Phys. Soc. Jpn.* **70**, 2425 (2001).

[4] K. Kuwahara *et al., Physica B* **312**, 899 (2002).

[5] N. Tateiwa *et al., J. Phys. Soc. Jpn.* **70**, 1853 (2001).

[6] Y. Koike *et al.*, accepted to *Phys. Rev. Lett.*

[7] N. Metoki *et al., Phys. Rev. Lett.* **80**, 5417 (1998).

[8] N. Bernhoeft *et al., Phys. Rev. Lett.* **81**, 4244 (1998).

[9] S. Coad *et al., Physica B* **281**, 200 (2000).