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Higher-order collinear interaction and magnetic excitation in the 5f localized system $U_3Pd_{20}Si_6$

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Abstract
The magnetic structure and excitation in a localized 5f system $U_3Pd_{20}Si_6$ were studied by means of neutron scattering. Our neutron diffraction experiments on a single-domain sample uniquely determined a remarkable collinear structure due to higher-order exchange interaction. We discovered a new type of spin-flop transition with collinear coupling. The localized nature of uranium 5f electrons with a $5f^2$ Γ_5 ground state was clearly demonstrated by the observed crystalline electric field excitation. We observed beautiful ferromagnetic and antiferromagnetic spin-wave excitation over a whole Brillouin zone. Surprisingly, however, a clear low-energy quasi-elastic component was also observed around the antiferromagnetic zone centre. This low-energy quasi-elastic response is concluded to be the excitation of the quasi-particles due to hybridization between 5f and conduction electrons. The observation of this localized and itinerant dual nature in magnetic excitation indicates the possibility of partial localization based on an itinerant and two localized 5f electrons proposed theoretically.

1. Introduction

Actinide-based intermetallic compounds attract strong interest because of the variety of their magnetic and electronic properties. In particular, the recent great discoveries of

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ferromagnetic superconductivity in UGe_2 [1] and URhGe [2], and high-temperature heavy-fermion superconductivity in $PuCoGa_5$ [3] and its derivatives [4] have attracted much attention in the field of the strongly correlated electron systems. The 5f electrons in most uranium-based intermetallic compounds are believed to show itinerant character due to the hybridization with conduction electrons. Very recently a microscopic description of heavy quasi-particles in U-based compounds has been proposed. It is based on the assumption that some of the 5f states may be itinerant while the others remain localized. Within this scenario a recent band calculation for UPt_3 and UPd_2Al_3 reproduces the dHvA frequencies and effective mass very well [5]. However, the existence of localized 5f electrons in UPt_3, and even in UPd_2Al_3, remains controversial, because of the absence of the crystalline electric field (CEF) and J-multiplet excitations which are direct evidence for the localized character of 5f electrons.

Apart from in UPt_3, partial localization of the 5f state is in a general sense a possible and highly interesting scheme. In order to reveal the dual nature of U-based 5f-electron systems, we studied the magnetic structure and excitations in a localized 5f intermetallic compound $U_3Pd_{20}Si_6$. Up to now only a few uranium compounds have been reported to be localized 5f systems. Recently we found a new uranium intermetallic compound $U_3Pd_{20}Si_6$ [6]. The localized character was suggested by the Curie–Weiss behaviour in the magnetic susceptibility, as well as the CEF excitation [7, 8]. $U_3Pd_{20}Si_6$ is the first example of a localized 5f system with a magnetic ground state, a Γ_5 and $5f^2$ configuration, while the antiferro-type quadrupolar ordering in UPd_3 has been intensively studied by McEwen and co-workers [9]. It should be noted that these prototype compounds have the same $5f^2$ configuration, with one 5f electron remaining somewhere. It is also noteworthy that, so far, no U-based localized system with a $5f^3$ configuration has been reported.

In this paper we review the remarkable collinear structure and a new type of spin-flop transition due to the higher-order collinear interaction [10]. We observed beautiful ferromagnetic and antiferromagnetic spin-wave excitation over a whole Brillouin zone [11]. The quasi-elastic response is concluded to be the excitation of the quasi-particles, namely the itinerant part, due to hybridization between 5f and conduction electrons. Our observation indicates a clear dual nature of magnetic excitation in the localized 5f system $U_3Pd_{20}Si_6$.

2. Experimental details

Neutron scattering experiments were carried out at the research reactor JRR-3 in Japan Atomic Energy Research Institute (JAERI). Elastic scattering was measured on the triple-axis spectrometer TAS-2 ($E_i = 13.7$ meV). A tiny single crystal (0.5 mm thick and 5 mm in diameter) was mounted with the [1, 0, 0] and [0, 1, 1] axes in the horizontal scattering plane. A magnetic field was applied either along the vertical, $\boldsymbol{H}_\perp \parallel [0, \bar{1}, 1]$, or horizontal, $\boldsymbol{H}_\parallel \parallel \boldsymbol{Q}$, direction. The sample was cooled down to 0.2 K with liquid-He cryogen-free dilution refrigerator [12]. The vertical field was applied up to 10 T using a liquid-He cryogen-free superconducting magnet [13]. These liquid-He cryogen-free systems were developed by JAERI.

Neutron inelastic scattering spectra were measured with the use of TAS-1 at the final energy $E_f = 14.7$ meV and LTAS with $E_f = 3.5$ meV. A large single crystal with total mass 10 g was cut into two pieces and mounted with the [1, 0, 0] and [0, 1, 1] axes in the horizontal scattering plane within the accuracy of 0.1°. The samples were cooled down to 100 mK with a Oxford Kelvinox dilution refrigerator.

Single-crystalline $U_3Pd_{20}Si_6$ was grown by the Czochralski pulling method under an Ar gas atmosphere using a tri-arc furnace. The detail of the sample preparation has been published elsewhere [7].

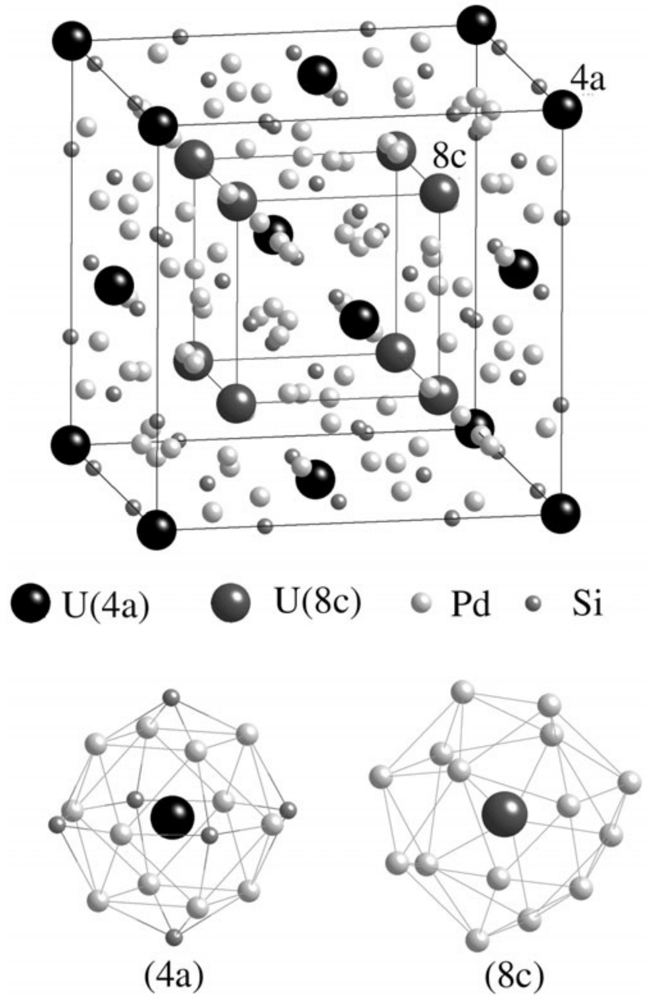

Figure 1. The crystal structure of $U_3Pd_{20}Si_6$.

3. Experimental results

The crystal structure of $U_3Pd_{20}Si_6$ is shown schematically in figure 1. $U_3Pd_{20}Si_6$ crystallizes in an ordered derivative of the $Cr_{23}C_6$-type cubic structure with space group $Fm\bar{3}m$. The crystal structure is very complicated, containing 116 atoms in a large unit cell with the lattice constant 12.175 Å. However, if we pay attention only to uranium atoms and ignore the other elements, the uranium lattice is rather simple. There are two different crystallographic sites for uranium atoms, 4a and 8c sites. Uranium atoms on 4a sites have the face-centred-cubic (fcc) structure, while the 8c sites form a simple cubic lattice with a half-unit of the crystallographic unit cell. The nearest-neighbour U–U distance is 5.27 Å. This value is the interatomic distance between the 4a and 8c site. The nearest-neighbour distances within the 8c and 4a sublattices are 6.09 and 8.61 Å, respectively. Each uranium atom is surrounded by ligand Pd and Si atoms. This cage-like structure and the large uranium interatomic distances would be important for the localized character of this compound.

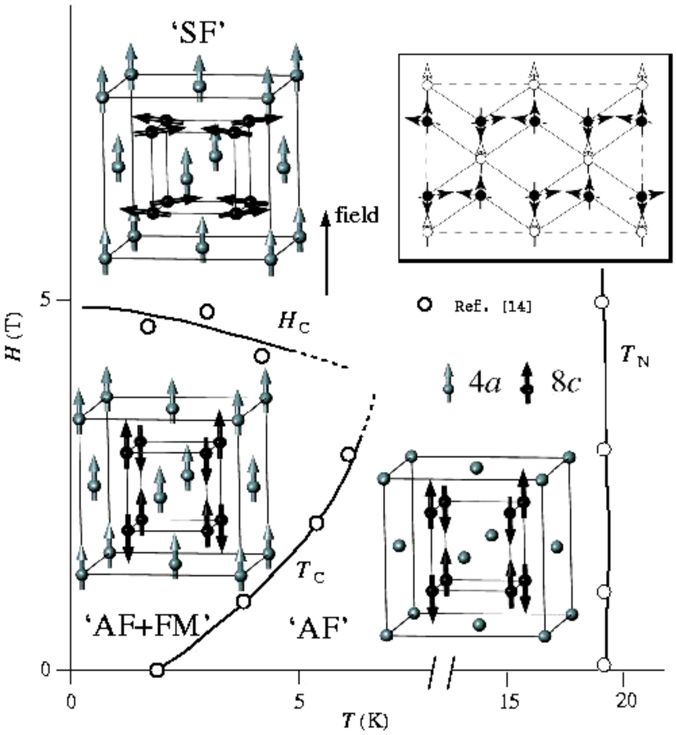

Figure 2. The H–T magnetic phase diagram of $U_3Pd_{20}Si_6$.
(This figure is in colour only in the electronic version)

Figure 2 summarizes the H–T phase diagram for the magnetic structures of $U_3Pd_{20}Si_6$ [10]. These magnetic structures described here were uniquely determined by means of neutron diffraction for a single-domain sample under a magnetic field. There are three ordered phases. In the 'AF' phase, uranium spins on the 8c sites order in the G-type antiferromagnetic structure. Note that the uranium moments on 4a sites remain paramagnetic. The magnetic moment is parallel to $\langle 100 \rangle$. In the 'AF + FM' phase, the ferromagnetic ordering on the 4a sites shows a collinear coupling with 8c antiferromagnetic ordering, which enables us to get a single-domain sample under a weak external field. The 8c spins exhibit a spin-flop transition for $H \sim 5$ T, and the 'SF' phase becomes stable for higher magnetic fields.

Heisenberg exchange interaction between the nearest-neighbour 4a and 8c sublattices is cancelled out, which is why the 4a and 8c spins order at different temperatures. The transition temperatures, $T_C = 2$ and $T_N = 19$ K, indicate the strength of the intra-site interactions. In the 'AF + FM' phase, we found that the antiferromagnetic moment follows the direction of the interpenetrating ferromagnetic moment, when the 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 candidates for providing this collinear coupling.

The 8c spins exhibit a spin flop for $H \sim 5$ T. This spin-flop transition is unusual, because (i) it is not expected with cubic anisotropy and (ii) a spin flop of the 8c sites is observed when both 4a and 8c spins are ordered. We concluded that it is a new type of spin-flop transition with collinear interaction which induces uniaxial anisotropy on the 8c sites.

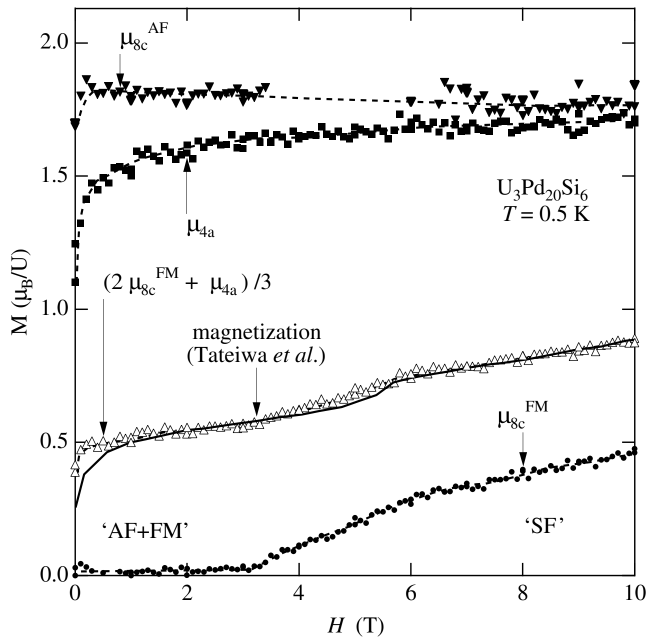

Figure 3. The field dependence of the ferromagnetic and antiferromagnetic components on 8c sites, μ_{8c}^{FM} and μ_{8c}^{AFM}, respectively, and the ferromagnetic moment on 4a sites, μ_{4a}, for $H \parallel [1, 1, 0]$.

The field ($H \parallel [1, 1, 0]$) dependence of the magnetic moment obtained by our neutron diffraction experiments is shown in figure 3. The magnetic moment at 8c sites, $([\mu_{8c}^{FM}]^2 + [\mu_{8c}^{AFM}]^2)^{1/2} \simeq 1.8\ \mu_B/U$ is in good agreement with the $5f^2$ Γ_5 ground state (2 μ_B). The ferromagnetic component of the 8c sites exhibits a steep increase in the vicinity of the spin-flop field. μ_{4a} is strongly suppressed for small external field but recovers in value in the Γ_5 ground state. The total moment per uranium atom $(\mu_{4a} + 2\mu_{8c}^{FM})/3$ obtained by the neutron diffraction study is in perfect agreement with the result of a recent magnetization measurement [14].

The strong suppression of the 4a ferromagnetic moment could be explained by the inter-site coupling. Below T_C, a constant molecular field on the 8c sites is expected from the 4a sites. This molecular field might perturb the antiferromagnetic structure of 8c spins. Therefore, the 4a ferromagnetic ordering could be suppressed and so not disturb the 8c antiferromagnetic ordering. Note that the intra-site interaction in 8c sites is much stronger than that in 4a sites due to the large difference in U–U distance. In this situation, the magnetic moment would recover to the saturation value with field application.

The CEF excitation in $U_3Pd_{20}Si_6$ was observed by Tateiwa *et al* [7] for the first time. Kuwahara *et al* [8] observed the CEF excitation with the HET spectrometer at ISIS. They revealed that the observed CEF excitation spectra can be explained in terms of the $5f^2$ configuration with a Γ_5 ground state both for 4a and 8c sites. The result for the CEF excitation is consistent with the specific heat and the softening of the elastic constant c_{44} around the magnetic ordering temperature.

The spin-wave excitation of $U_3Pd_{20}Si_6$ was reported for the first time by Aso *et al* [11]. $U_3Pd_{20}Si_6$ exhibits very beautiful antiferromagnetic and ferromagnetic spin-wave excitations due to 8c and 4a sites, respectively, over a whole Brillouin zone. The existence of the clear spin-wave excitation in $U_3Pd_{20}Si_6$ is rather 'unusual' for uranium intermetallic compounds. In most U-based intermetallic compounds magnetic excitations are often very weak, broad and

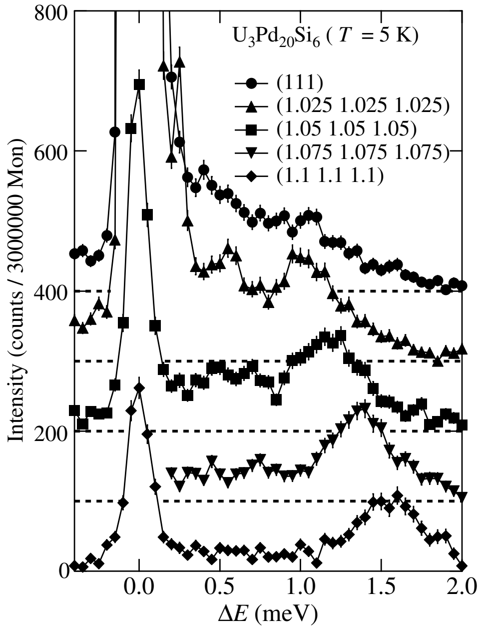

Figure 4. The high-resolution neutron inelastic scattering spectra of $U_3Pd_{20}Si_6$.

spread out in Q–ω space. Therefore the very sharp spin-wave excitation is also attributed to the localized character of 5f electrons in $U_3Pd_{20}Si_6$.

Figure 4 shows high-resolution neutron inelastic scattering profiles of $U_3Pd_{20}Si_6$ around the zone centre (1, 1, 1) measured at 5 K. The inelastic peak at $\Delta E = 1.5$ meV for $Q = (1.1, 1.1, 1.1)$ is the antiferromagnetic spin-wave excitation. The spin-wave excitation energy decreases with Q approaching the (1, 1, 1) zone centre. We expect the gap about 1 meV from extrapolation of the excitation energy. Surprisingly, however, it is clearly seen that a weak quasi-elastic response is observed in a very narrow Q-region around the zone centre. The two-component scattering profile around the antiferromagnetic zone centre is very similar to the low-energy magnetic response in the heavy-fermion superconductor UPd_2Al_3. In UPd_2Al_3, a very broad spin-wave excitation and a quasi-elastic component were observed around the $(0, 0, 1/2)$ magnetic zone centre [15]. The quasi-elastic component showed a superconducting gap in the superconducting state [16]; hence this low-energy response was concluded to be the magnetic excitation due to the heavy quasi-particles in UPd_2Al_3 [17]. A similar low-energy response has been observed in UPt_3 [18] and UGa_3 [19]. Therefore the existence of the quasi-elastic component in $U_3Pd_{20}Si_6$ involves there being quasi-particles even in this localized system.

At present we have no idea how to explain the existence of the dual natures of the magnetic excitation—namely: the clear localized nature of the CEF and spin-wave excitation; and the itinerant nature of the quasi-elastic low-energy response due to quasi-particles. It is significant that the localized character with the $5f^2$ Γ_5 ground state in $U_3Pd_{20}Si_6$ is well established experimentally; it coexists with the quasi-elastic component. A possible explanation might be a partial localization based on the coexistence of the localized and itinerant parts of the 5f electrons. Further theoretical and experimental studies are necessary in order to clarify the behaviour of the 5f electrons in uranium-based intermetallic compounds.

In conclusion, we have discovered remarkable collinear magnetic ordering and a new type of spin-flop transition due to the higher-order collinear interaction. We observed a clear localized and itinerant dual nature of the magnetic excitation, indicative of partial localization based on an itinerant and two localized 5f (Γ_5) electrons.

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