# Exotic non-magnetic order accompanied by antiferromagnetic short range order in $URu_2Si_2$

Yoshihito Miyako,[*1] Hiroshi Amitsuka,[*2] Naoto Metoki,[*3] Masugu Sato,[*3] and Kazuhiro Marumoto[*4]
[*1] *Graduate School of Science, Osaka University*
[*2] *Graduate School of Science, Hokkaido University*
[*3] *Advanced Science Research Center, Japan Atomic Energy Research Institute*
[*4] *Department of Applied Physics, School of Engineering, Nagoya University*

Some feature of $URu_2Si_2$ shows a phase transition at 17.5 K similar to spin density wave and superconductivity coexists below 1.5 K. The phase transition at 17.5 K is reviewed from the view point of quadrupolar ordering.

$URu_2Si_2$ is one of most interesting heavy fermion compounds which have been studied extensively.[1)] At early stage of research, neutron scattering and muon spin relaxation($\mu$SR) experiments have demonstrated the existence of weak magnetic ordering in heavy fermion compound $URu_2Si_2$ ($T_o$ = 17.5 K) which exhibits a superconducting phase transition at lower temperatures. Kohori *et al.* performed NMR experiments[2)] on $^{29}$Si in $U(Ru_{1-x}Rh_x)_2Si_2$, but they could not find any evidence to support the existence of static magnetic order in $URu_2Si_2$.

Related to this phase transition, we have suggested quadrupolar ordering from the measurement of nonlinear susceptibility[3)] instead of static antiferromagnetic ordering. Since our study of the nonlinear susceptibility, the phase transition at $T_o$ has been a controversial issue for the last 10 years. Many explanations have been proposed for the phase transition. One of them is the type-I antiferromagnetic order, which is claimed to be static for high quality single crystal of $URu_2Si_2$. However, the proposal of quadrupolar ordering was supported by experiments on the magnetic field dependence of $T_o$ and the staggered ordered magnetic moment[4)] and on the point contact spectroscopy[5)] and by the recent neutron scattering experiment under pressure.[1)]

We found a first order antiferromagnetic phase transition at 1.5 GPa ($P_c$) in a single crystal of $URu_2Si_2$ by the neutron scattering experiment under hydrostatic pressure $P$. The staggered magnetic moment increased with pressure and the first order phase transition occurs at $P_c$ with a jump of the staggered moment into 0.4 $\mu_B$ from 0.25 $\mu_B$ as shown in Fig. 1, without any change of the antiferromagnetic structure above and below $P_c$.

The antiferromagnetic state above $P_c$ is an Ising type as described in a previous paper.[1)] The ordered moment aligns along the $c$-axis. These results prove that the antiferromagnetic order with the wave vector $q(001)$ becomes static by pressure and below the $P_c$ the antiferromagnetic order is parasitic. The correlation length also increases with pressure from 180 A at 2 K and at ambient pressure to more than $10^3$ A at $P > 0.6$ GPa. A heavy fermion state evolves below the peak temperature, $T_{\max}(\chi)$, of the susceptibility which is around 50 K. In thermodynamic measurements of

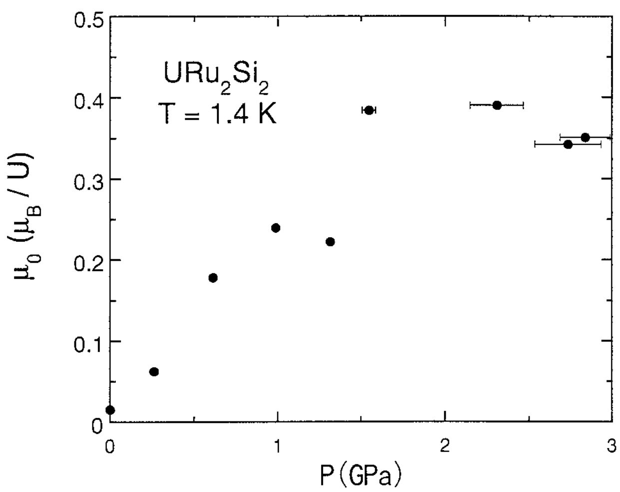


Fig. 1. A staggered magnetic moment measured at 1.4 K by a neutron scattering experiment for a single crystal of $URu_2Si_2$ as a function of the hydrostatic pressure.

$URu_2Si_2$, there is a clear anomaly in the specific heat at $T_o$ and the phase transition at $T_o$ is accompanied by lattice instability. The entropy change due to the phase transition is 0.2 Rln2/mol. K. The anomaly at $T_o$ in the resistivity is quite similar to that of a SDW transition in rare earth metals and this suggests a gap opening in the Fermi surface in the $a^*$- and $c^*$-axis of the reciplocal lattice space.

An antiferro-quadrupolar order in Fig. 2 is compatible with an anomalous increase of the resistivity as well as the specific heat anomaly at $T_o$.

Thus, the pressure induced phase transition is a possible phase transition from an exotic phase, like quadrupolar order, accompanied by a parasitic antiferromagnetic short range order into a static type-I antiferromagnetic order.

The non-linear susceptibility of $URu_2Si_2$ is derived from the magnetic field dependence of the magnetization using the for-

* H. Amitsuka and T. Sakakibara also proposed the $\Gamma_{5t}^{(2)}$ (or $\Gamma_{5t}^{(1)}$) doublet ground state before the Ref. 9 for $U_xTh_{1-x}Ru_2Si_2$ dilute alloy system in Ref. 10.

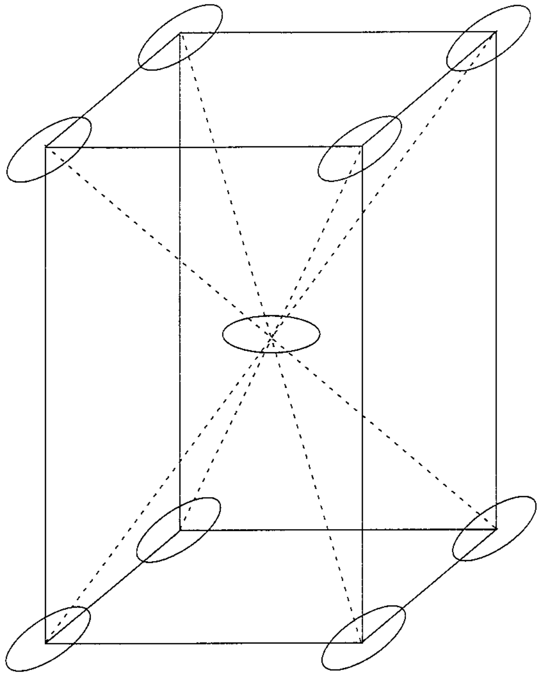

Fig. 2. Schematical figure of the antiferro-quadrupolar order.

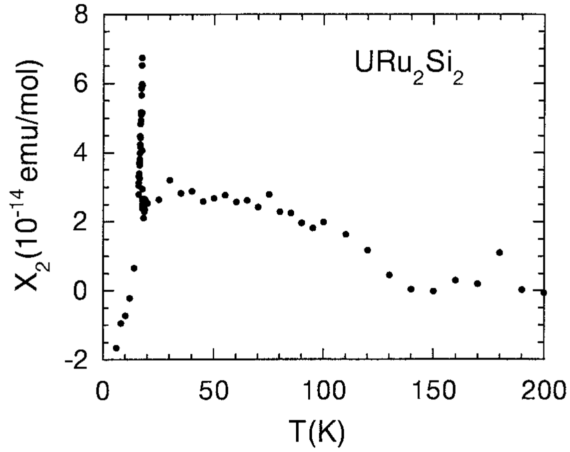


Fig. 3. Temperature dependence of the non-linear susceptibility, $\chi_2$.

mula of $M = \chi_0 H + \chi_2 H^3$, where $\chi_0$ and $\chi_2$ are the linear and non-linear susceptibility and H is an applied magnetic field. $\chi_0$ has a kink at $T_o$, but $\chi_2$ seems to show a critical behavior, as shown in Fig. 3. For magnetic phase transitions, which are ferromagnetic, antiferromagnetic and spin glass phase transitions, $\chi_2$ has a negative sign in the paramagnetic phase. Therefore, the result in Fig. 3 predicts a possible non-magnetic phase transition. The temperature dependence of $\chi_2$ closely follows that of the lattice thermal expansion coefficient, $\alpha_c - \alpha_a$ of $c/a$, where $a$ and $c$ are the lattice constant of $URu_2Si_2$.[3)] Thus, the critical behavior of $\chi_2$ seems to be dominated by a phase transition associated with lattice instability. The short range order seems to develop from around 50 K which is much higher than $T_o$, as can be seen in Fig. 3. Neutron scattering experiments[6)] have also predicted the existence of a large intensity of the inelastic magnetic excitation which has a large peak at $T_o$ corresponding to $2.5\,\mu_B$ and the inelastic excitation starts at 50 K or higher temperatures. Holland-Moritz *et al.*[6)] suspected non-magnetic quadrupole interactions or a charge-density wave as a possible origin for the observed spatial correlation and the triggering mechanism for the magnetic order.

Due to the large intensity of the inelastic neutron scattering, singlet — singlet crystalline field splitting model was proposed for the behaviors mentioned above.

Niuwenhuys[7)] analyzed the magnetic susceptibility and magnetization of $URu_2Si_2$ and $UPt_2Si_2$ by crystalline field theory assuming that U is in the 4+ state having $J = 4$, the $^3H_4$ spin-orbit ground state. A singlet-singlet-doublet-singlet crystal field level scheme was used for both compounds. This model explains well the susceptibility and the magnetization of $UPt_2Si_2$, but not for $URu_2Si_2$. $UPt_2Si_2$ is $CaBe_2Ge_2$ type structure and the crystalline field level splittings were actually deduced to be singlet-50 K-singlet-12 K-doublet-14 K-singlet from the inelastic neutron scattering experiment.[8)] The inelastic neutron scattering peaks were observed because of the absence of a heavy fermion character ($\gamma = 37\,\mathrm{mJ/mol.K^2}$) in $UPt_2Si_2$. The antiferromagnetic ordered moment is $1.9\,\mu_B$ and the magnetic structure can be described as a type-I antiferromagnet. On the contrary, the susceptibility and the magnetization of $URu_2Si_2$ were not well fitted by Niuwenhuys's crystalline field model. It is most reasonable to assume a non-Kramers doublet ground state to explain the strong anisotropy of the susceptibility. This model can explain the large inelastic neutron scattering above $T_o$ as the development of quardrupolar short range ordering from around 50 K.

We assumed before[9)] the doublet, $\Gamma_{t5}^{(2)}$ (or $\Gamma_{t5}^{(1)}$), — singlet, $\Gamma_{t1}^{(1)}$, splitting of $J$ multiplet $^3H_4$ for diluted uranium system $U_xLa_{1-x}Ru_2Si_2$ to explain the experiments of the susceptibility, magnetization and specific heat*. The $\Gamma_{t5}^{(2)}$ state has a $J_z$ component and no $J_{x,y}$ components, and is expressed as follows:

$$\begin{aligned}\Gamma_{t5}^{(2)} &= \phi(+) + i\phi(-)/2^{1/2} \\ &= \phi(+Q) \text{ and } \phi(+) - i\phi(-)/2^{1/2} \\ &= \phi(-Q),\end{aligned}$$

where

$$\langle\phi(+)\ J_z\ \phi(+)\rangle = +m, \quad \langle\phi(-)\ J_z\ \phi(-)\rangle = -m,$$
$$\langle\phi(+)\ J_z\ \phi(-)\rangle = \langle\phi(-)\ J_z\ \phi(+)\rangle = 0$$

and

$$\langle\phi(+Q)\ J_x^2 - J_y^2\ \phi(+Q)\rangle = +Q/2,$$
$$\langle\phi(-Q)\ J_x^2 - J_y^2\ \phi(-Q)\rangle = -Q/2.$$

In the ordered state of the quadrupole moment, the ground-state wave function changes from $\phi(+Q)$ to $\phi(-Q)$ with respect to the U site at the body center and the corner of the body centered tetragonal crystal structure. The energy splitting between the $\phi(+Q)$ and $\phi(-Q)$ states is in proportion to $Q$ and the inelastic neutron scattering intensity between the two states is in proportion to $m$. The magnetic moment, $2\,\mu_B\,m$, is estimated to be $1.8\,\mu_B$ from a high field magnetization measurement of U impurity in $LaRu_2Si_2$. The inelastic neutron scattering intensity becomes smaller above $T_o$ because the lifetime of the splitted energy states, $\phi(+Q)$ and

$\phi(-Q)$ becomes shorter.

We can understand qualitatively the quadrupolar ordering and the temperature dependence of the inelastic neutron scattering intensity, and an Ising-type antiferromagnetic state above $P_c$ in terms of the $\Gamma_{t5}^{(2)}$ ground-state model. However, this model does not explain the rather small ordered staggered moment ($0.4\,\mu_{\rm B}$) above $P_c$, although there could be a change in the Kondo temperature $T_K$. Furthermore, although $U_xLa_{1-x}Ru_2Si_2$ diluted alloy shows a Fermi liquid behavior at low temperatures, $U_xTh_{1-x}Ru_2Si_2$ exhibits a non-Fermi liquid behavior in the susceptibility and specific heat.[10] The origin for these different behaviors is not well understood although the importance of the contribution of the crystalline field excited level was predicted.[11]

This paper is dedicated to the memory of the retirement of Professor A. Ito from Ochanomizu University.

## References

1) H. Amitsuka, M. Sato, N. Metoki, M. Yokoyama, K. Kuwahara, T. Sakakibara, H. Morimoto, S. kawarazaki, Y. Miyako, and J. A. Mydosh: Phys. Rev. Lett. **83**, 5114 (1999).
2) Y. Kohori, Y. Noguchi, T. Kohara, K. Asayama, H. Amitsuka, and Y. Miyako: Solid State Commun. **82**, 479 (1992); T. Kohori: private communication.
3) Y. Miyako, S. Kawarazaki, H. Amitsuka, C. C. Paulsen, and K. Hasselbach: J. Appl. Phys. **70**, 2680 (1991); Y. Miyako, H. Amitsuka, S. Kunii, and T. Kasuya: Physica B **186/188**, 236 (1993); Y. Miyako, H. Amitsuka, S. Kawarazaki, T. Taniguchi, and T. Shikama: Jpn. J. Appl. Phys., Ser. 8, 230 (1993).
4) S. A. M. Mentink, T. E. Mason, S. Sullow, G. J. Niuwenhuys, A. A. Menovsky, and J. A. Mydosh: Phys. Rev. B **53**, R6014 (1996).
5) J. G. Rodrigo, F. Guinea, S. Vieira, and F. G. Aliev: Phys. Rev. B **55**, 14318 (1997).
6) E. Holland-Moritz, W. Schlabitz, M. Loewenhaupt, and U. Walter: Phys. Rev. B **39**, 551 (1989).
7) G. J. Niuwenhuys: Phys. Rev. B **35**, 5260 (1987).
8) M. B. Walker, Z. Tun, W. J. L. Buyers, A. A. Menovsky, and W. Que: Physica B **199/200**, 165 (1994).
9) K. Marumoto, T. Takeuchi, and Y. Miyako: Phys. Rev. B **54**, 12194 (1996).
10) H. Amitsuka and T. Sakakibara: J. Phys. Soc. Jpn. **63**, 736 (1994).
11) M. Koga and H. Shiba: J. Phys. Soc. Jpn. **64**, 4345 (1995).