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著者	KOSAKA Masashi
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Quadrupolar Interaction and Magnetic Properties of tetragonal Tm compounds

Masashi KOSAKA

Institute for Materials Research

Abstract

Chap. 1. Introduction

Many works on quadrupolar interaction and quadrupolar ordering in rare earth compounds with cubic structures have been performed extensively in the past decade, and quadrupolar orderings have been found out in paramagnetic states of TmCd, TmZn and so on. Only few studies, however, have been made so far on quadruploar interaction in tetragonal or hexagonal symmetry. The quadrupolar interaction naturally plays an important role in the compounds with these symmetries. For this point of view, our interest is focused on the quadrupolar interaction in the tetragonal rare earth compounds. The existence of quadrupolar ordering has been found out only in $TmAg_2$ which has a body centered tetragonal $MoSi_2$-type structure with a space group $I4/mmm(D_{4h}^{17})$[1]. In this work, we are concerned with $TmAu_2$ and $TmCu_2Si_2$ which are in the same space group of $TmAg_2$.

Chap. 2. Experimental

The sample compound was synthesized by a conventional argon arc technique, followed by annealing at 900 °C for a week. Single crystals were grown by the Czochralski method using a tri-arc furnace. The powder sample was used for inelastic neutron scattering and neutron diffraction. The single crystalline samples were used for magnetometric measurements, ultrasonic velocity measurements and specific heat measurements.

Chap. 3. Results and Discussions

Sec. 3-1. $TmCu_2Si_2$

The magnetic susceptibility as well as the neutron diffraction reveal that $TmCu_2Si_2$ undergoes a transition to an antiferromagnetic state at T_N=2.8K. The crystalline electric field (CEF) parameters are determined by inelastic neutron scattering and magnetic susceptibility measurements. It became clear that the Tm^{3+} CEF ground state of $TmCu_2Si_2$ is a non-magnetic Γ_3 singlet. Therefore, there are not quadrupolar ordering at very low temperature, because the quadrupolar interaction is weak. However, the result of temperature dependence of elastic constant with C_{66} and C_{44} mode are affected by an excited state

of a Γ_5 doublet which has a large quadrupole moment. The energy splitting between the ground state and Γ_5 is about 80K. Moreover, a first excited state of a non-magnetic Γ_4 singlet is close to ground state. The transition probability between the ground and the first excited state has a large value. Therefore, it was made clear that the mechanism of magnetic transition on $TmCu_2Si_2$ is a Van Vleck antiferromagnetic transition. The magnetic structure is a sinusoidally moment-modulated structure described by the propagation vector **Q**=(0.147, 0.147, 0).

Sec. 3-2. TmAu₂

The anomalies at 7.0K and 3.2K have been observed in temperature dependence of the specific heat. The occurrence of a nonmagnetic transition at T_Q=7.0K in $TmAu_2$ is interpreted as a ferroquadrupolar ordering within the orthorhombic γ-symmetry lowering mode, and an antiferromagnetic transition occur at T_N=3.2K. The CEF parameters are obtained by the inelastic neutron scattering and the magnetic susceptibility measurements. Both the magnetoelastic couplings and the quadrupolar interactions are then determined by the ultrasonic velocity experiment. The $(C_{11}-C_{12})/2$ mode exhibits a very pronounced softening which, at 7.0K, reaches ~50% of the high temperature value. Below T_Q, the ground state Γ_5 is splitted into two singlets accompanied with the quadrupolar ordering. Therefore, we think that the magnetic long range ordering in $TmAu_2$ is realized in the singlet ground state. The results indicate that the Van Vleck component which is a transition probability between the ground and the first excited state is very large at temperature region between T_Q and T_N. Therefore, we believe that the magnetic ordering in $TmAu_2$ is also the Van Vleck antiferromagnetic ordering, and the transition between the singlet ground state and the very low lying excited state plays a dominant role in the magnetic transition. Such a mechanism is the same to that of $TmCu_2Si_2$.

The magnetic structure in $TmAu_2$ which develops below T_N is a sinusoidally moment-modulated structure described by the propagation vector **Q**=(0.435, 0.347, 0) and the Tm moments aligned along the a-axis parallel to the quadrupole moment.

Chap. 4. Summary

The study reveals the existence of a quadrupolar ordering in the $TmAu_2$ intermetallics with the tetragonal symmetry. The results of magnetic and magnetoelastic measurement made it clear that the quadrupolar interaction plays an important role in magnetic properties of the tetragonal compound. This information will be applicable to understanding various anomalous behaviors appearing in tetragonal and hexagonal rare earth compound.

Reference

[1] P. Morin and J. Rouchy, Phys. Rev. B **48** (1993) 256.