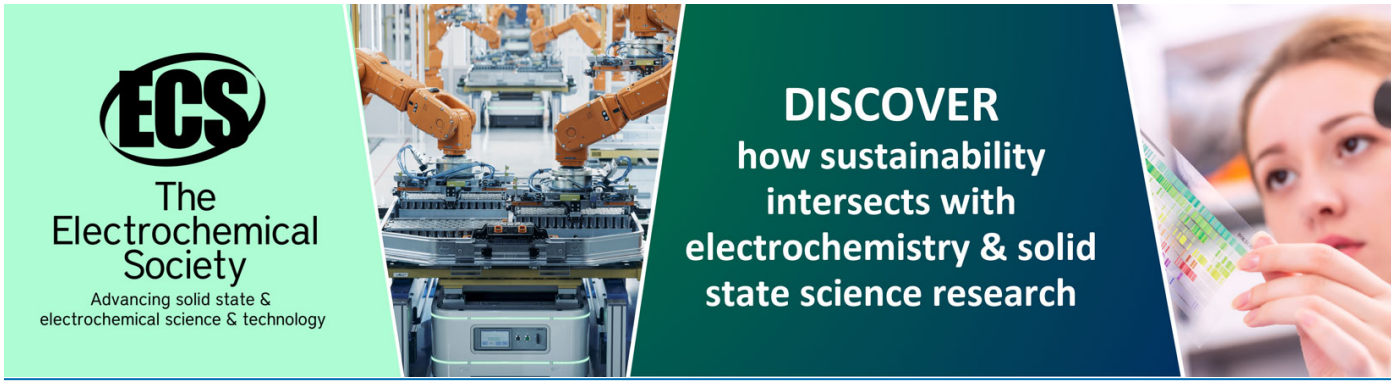
Spontaneous strain in ferroquadrupolar phase of $TmAu_2$



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Spontaneous strain in ferroquadrupolar phase of $TmAu_2$

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Abstract. We present the results of a study of the spontaneous strain accompanied by a ferroquadrupolar ordering in $TmAu_2$. The strain has been investigated by means of thermal expansion, magnetostriction, and neutron diffraction measurements on a single crystalline $TmAu_2$. In the ferroquadrupolar ordered phase, the strain shows a strong enhancement by applying magnetic field. This implies that structural orthorhombic domains caused by the ferroquadrupolar ordering are transformed into a single-domain structure by a magnetic field.

1. Introduction

It is widely recognized that quadrupolar interactions play an important role in the f-electron system. When the system has a degeneracy in the crystalline electric field (CEF) ground state associated with the quadrupole moment, and the quadrupolar interaction is stronger than magnetic interactions, a quadrupolar ordering can occur in a paramagnetic phase. The type of the quadrupolar ordering is divided into two categories of a ferroquadrupolar and an antiferroquadrupolar ordering, according to the sign of its quadrupolar interaction. In the case of the ferroquadrupolar ordering, a structural phase transition occurs simultaneously, which causes a spontaneous strain in ferroquadrupolar ordered phase.

The binary thulium compound $TmAu_2$ crystallizes in the tetragonal $MoSi_2$-type structure, which shows the ferroquadrupolar and an antiferromagnetic ordering at $T_Q = 7.0$ K and $T_N = 3.2$ K, respectively [1]. At T_Q, the structural phase transition from a tetragonal to an orthorhombic phase occurs. The strain caused by the ferroquadrupolar ordering is an orthorhombic γ-symmetry-lowering mode, and then the ordering parameter has been determined to be $\langle O_2^2 \rangle$.

In this paper, we present the results of thermal expansion, magnetostriction, and neutron diffraction measurements using a single crystalline $TmAu_2$.

2. Experimental

A single crystal was prepared using the tetra-arc Czochralski method. The detail of the sample preparation was reported in our previous work [1]. Used as sample is $TmAu_2$ in the form of rectangular parallelepiped with the size of $3.20 \times 3.25 \times 3.65$ mm^3. Thermal expansion measurements were performed using the sensitive three-terminal capacitance method. The capacitive dilatometer for these measurements has been constructed with reference to the design

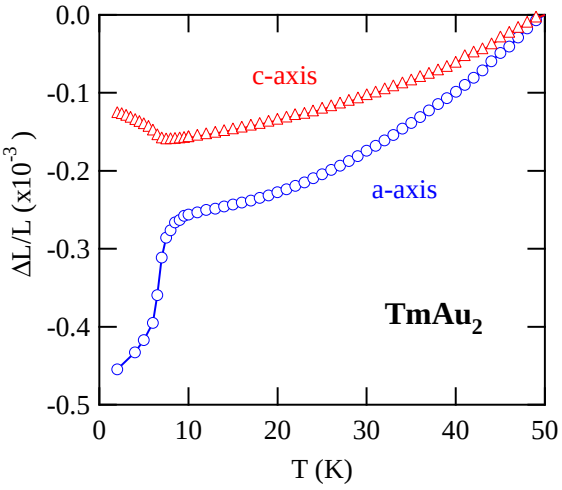

Figure 1. Temperature dependence of the thermal expansion of $TmAu_2$ along the a and c axes under zero magnetic field.

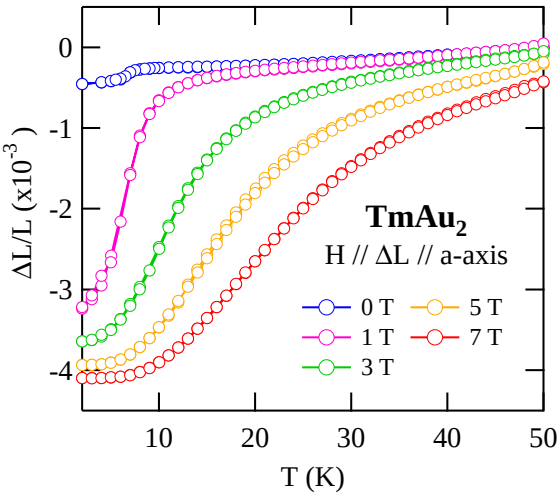

Figure 2. Temperature dependence of the thermal expansion of $TmAu_2$ along the a axis at various magnetic fields.

of the apparatus proposed by Schmiedeshoff *et al.* [2]. We have installed the dilatometer in a commercial Quantum Design (QD) physical property measurement system (PPMS). The neutron diffraction experiments were carried out using the IMR single-crystal diffractometer installed at the thermal neutron guide of the JRR-3M reactor at the Japan Atomic Energy Agency (JAEA). The diffraction patterns were recorded using neutrons of wave length 1.527 Å.

3. Results and Discussion

All thermal expansion data in this paper are normalized at 50 K under zero magnetic field. Thermal expansion measurements were performed in the process of decreasing and increasing temperature as well as magnetic field. A distinguishable hysteresis has not been observed in our measurements. Figure 1 shows the temperature dependence of the thermal expansion $\Delta L/L$ along the a and c axes at zero field. The both thermal expansion curves decrease monotonically with decreasing temperature, and show anomalies around $T_Q = 7.0$ K. The $\Delta L/L$ along the c axis turns to increasing below T_Q, whereas the $\Delta L/L$ along the a axis displays a large drop. It is well known that the crystal structure of $TmAu_2$ transforms from the paramagnetic tetragonal phase to the ferroquadrupolar ordered orthorhombic phase. The observed large change in $\Delta L/L$ along the a is considered to be in qualitative agreement with this fact. The value of the change in thermal expansion along the a axis around T_Q is estimated to be about 0.1×10^{-3}.

Figure 2 shows the temperature dependence of the $\Delta L/L$ along the a axis under several magnetic fields. Notice that, the anomaly corresponding to the ferroquadrupolar ordering develops extremely when a 1 T magnetic field is applied, the change in $\Delta L/L$ is estimated to be 2.65×10^{-3}. This behavior is considered to be associated with a formation of the orthorhombic domain structure. In this case, many orthorhombic domains are expected to occur at the structural phase transition. We previously reported that the orthorhombic distortion $2(a-b)/(a+b)$ increased gradually with lowering temperature, reaching 2.7×10^{-3} at 4.2 K [3]. This value was estimated from the temperature dependence of the lattice parameter a, which was determined by neutron powder diffraction. The magnitude of the change in $\Delta L/L$ at 1 T is almost the same as this distortion ratio. Neutron powder diffraction measurements, which are microscopic experiments, are not affected by the existence of structural domains. Therefore, the

spontaneous strain at zero magnetic field must be averaged over these domains. These domains are considered to be modified into a single-domain via the coupling between the quadrupole moment and the magnetic moment induced by a magnetic field. In addition, the anomaly demonstrates a tendency to shift toward higher temperatures and to broaden with increasing magnetic field. This behavior is similar to that of the temperature dependence of the specific heat. By applying magnetic fields, the broadening of the specific heat anomaly was observed around T_Q and the beginning of the ferroquadrupolar ordering appeared to move toward higher temperatures [1].

Shown in Fig. 3 is the isothermal magnetostriction along the a and c axes at several temperatures. The initial slope of the magnetostriction along the a axis indicated with circles is drastically altered between 10 K and 5 K. On the other hand, the data along the c axis are almost constant even at 2 K. These results also indicate that the realignment of the orthorhombic multi-domain structure by applying magnetic fields.

In ordered to characterize the multi-domain structure in the ferroquadrupolar ordered phase, we have measured the splits of nuclear reflections by neutron diffraction on a single crystalline $TmAu_2$. Figure 4 and 5 show the contour map of (200) and (110) nuclear reflections at 5.2 K on the reciprocal $a^* - a^*$ plane, respectively. The indexes of h and k are determined based on these nuclear reflections in the paramagnetic phase. Below T_Q, the (200) reflection splits into four peaks, while the (110) reflection is divided into three peaks. To distribute internal stress caused by the orthorhombic distortion, the high temperature tetragonal phase appear to be converted into an assemblage of orthorhombic domains with domain walls oriented in the (110) or $(1\bar{1}0)$ planes. This plays a role in lowering the elastic energy at the structural phase transition. Switching the a and the b axes in every small domain is advantageous for relieving stress. Such situations are schematically illustrated in Fig. 6, in which the broken lines indicate the tetragonal lattice before the structural phase transition occurs. The inset shows the real lattice on the $a-a$ plane, the solid lines express the switching behavior of the orthorhombic axes with the boundary of the (110) plane. Assuming the situation described above, there should be four kinds of orthorhombic domains. The crystallographic axes of domain individuals are slightly

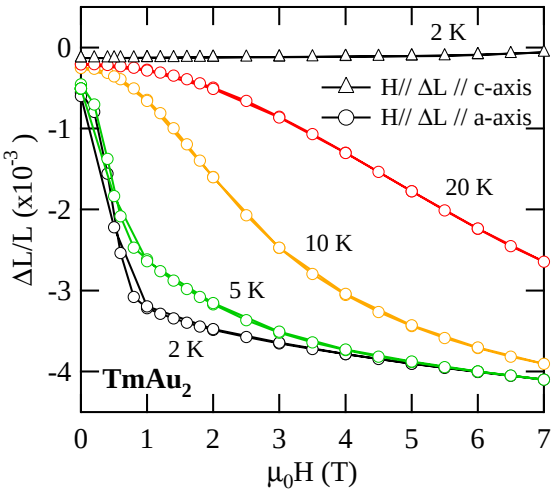

Figure 3. Magnetostriction of $TmAu_2$ along the a and c axes at various temperatures in magnetic fields applied along the same direction of $\Delta L/L$ measurements.

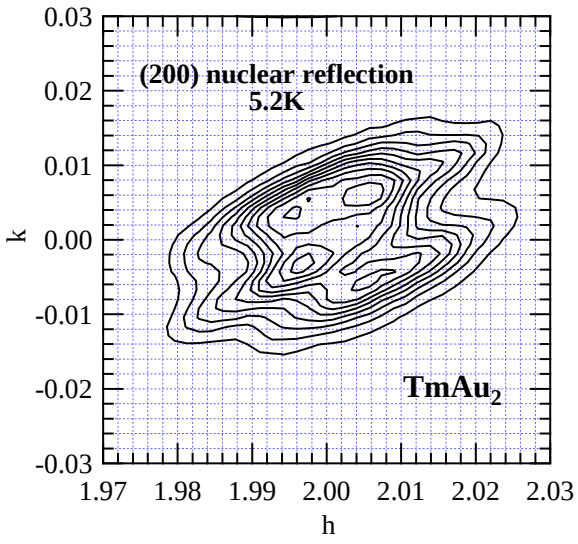

Figure 4. The (200) nuclear reflection of $TmAu_2$ at 5.2 K.

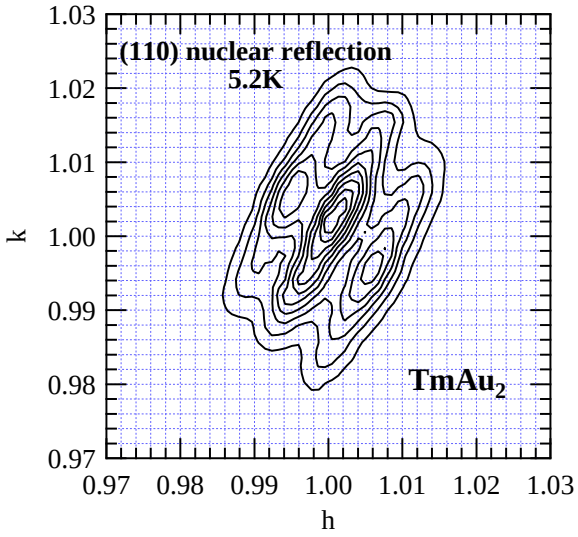

Figure 5. The (110) nuclear reflection of $TmAu_2$ at 5.2 K.

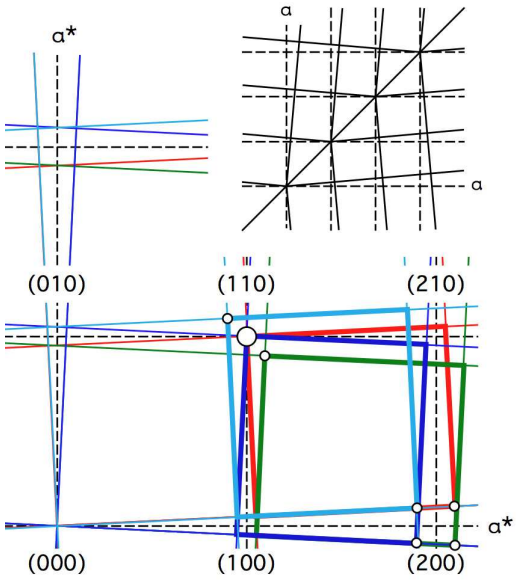

Figure 6. Schematic geometry of four orthorhombic domains in the ferroquadrupolar ordered phase on the reciprocal $a^* - a^*$ plane.

inclined from 90° each other due to the small difference between the length of a and b axes. The reciprocal lattice in Fig. 6 represents a schematic geometry of four individual orthorhombic domains. An instructive example was observed in high-T_C cuprate reported by Sueno *et al.* [4] and Saito *et al.* [5]. The origin of the similar nuclear reflection splittings was discussed in Ref. 4. They found that the twinning, which means the orthorhombic domain, occurs at a structural transition from a tetragonal phase in cooling process.

4. Conclusion

We have found that orthorhombic domains can be controlled by magnetic fields via the coupling between the quadrupole moment and the magnetic moment induced by a magnetic field. This technique makes it possible to compare the magnitude of the change in lattice constant and thermal expansion accompanied by the ferroquadrupolar ordering. The single-crystal neutron diffraction measurement reveals the existence of the multi-domain structure and its strain.

Acknowledgments

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