Synchrotron X-ray study of charge density waves in $\alpha$-TaS$_3$

K. Inagaki, M. Tsubota, K. Ichimura, S. Tanda, K. Yamamoto\textsuperscript{1}, N. Hanasaki\textsuperscript{1}, Y. Nogami\textsuperscript{1}, N. Ikeda\textsuperscript{1}, T. Ito\textsuperscript{2}, and H. Toyokawa\textsuperscript{1}

Department of Applied Physics, Hokkaido University, Sapporo, Japan
\textsuperscript{1}Graduate School of Natural Science, Okayama University, Okayama, Japan
\textsuperscript{2}Japan Synchrotron Research Center, Hyogo, Japan

E-mail: kina@eng.hokudai.ac.jp

Abstract. We report a synchrotron X-ray study of charge density waves (CDW) in an $\alpha$-TaS$_3$ crystal. CDW of $\alpha$-TaS$_3$ has been known to undergo a commensurate-incommensurate transition at 100 K, below which the wavevector locks in with the pristine lattice. We exploited the beamline BL02B1 of SPring-8. Temperature dependence of the Bragg peak (002) and satellite peak (1-1 2)+\vec{q} was measured from 7 K to 180 K. We found that a new phase in a temperature range of 130-50 K, where two independent CDWs coexist. These waves are incommensurate and commensurate CDWs with longitudinal wave vectors $\vec{q}_c$=0.252$c^*$ and 0.250$c^*$, respectively. By lowering the temperature, intensity of the incommensurate CDW was decreased, while that of the commensurate CDW was increased. At 50 K, the incommensurate CDW was completely diminished. Based on the concept of discommensuration, we determined the dislocation configuration from the intensity of the two CDWs.

1. Introduction

Low-dimensional conductors undergo the Peierls transition by spontaneous modification of charge density with the wavenumber $2k_F$ [1]. This charge density wave (CDW) is generally independent from the pristine lattice constant, however, a remarkable change is made in CDW characteristics if the ratio of CDW wavenumber and the lattice constant is integer, namely commensurate [2]. Several systems are found to exhibit commensurate CDW, both in quasi-one dimensional materials (\textit{e.g.} $\alpha$-TaS$_3$ [3, 4]) and quasi-two dimensional materials (\textit{e.g.} 1T-TaS$_2$ [5]). It should be noted that the CDW wavenumber is not commensurate over the whole range of temperature. Commensurate CDW is only possible at low temperatures, while at higher temperatures the wavevector changes from the integer ratio to the lattice constant [4, 5]. Phase diagram of incommensurate-commensurate phase is found to have finer structure in a quasi-two dimensional material [5], however, details of the commensurate-incommensurate transition in quasi-one dimensional material have not been clarified mostly because of experimental resolution.

In this paper, we report our preliminary result on synchrotron X-ray study of CDW in $\alpha$-TaS$_3$. We measured temperature dependence of Bragg and satellite peak profiles down to 7.3 K. We found that at temperatures 50-130 K, the mixed state existed stably with coexistence of two kinds of CDWs, attributed to commensurate and incommensurate wavevectors. From the electric current dependence of the intensity of the two CDWs, we demonstrated the evolution of edge dislocations of CDW, i.e. discommensurations [6].
Figure 1. Bragg (top) and satellite (middle) profiles obtained at 180, 130, 80, 50, 30, and 7.3 K for an $\alpha$-TaS$_3$ crystal. Vertical axes in the intensity maps denote the $c^*$ direction. The cross-section of each satellite profile along the $c^*$ direction (white dotted line) are shown in the bottom panels. Here $2\theta_0$ is the center of the detector. The shape of the Bragg peak does not show any significant change, whereas that of the satellite peak evolves with decreasing temperature. The two peaks at 80 K are attributed to commensurate and incommensurate CDWs, denoted as c and ic, respectively.

2. Experimental

We investigated a quasi-one dimensional conductor $\alpha$-TaS$_3$ on its CDW properties. It undergoes a Peierls transition at $T_P \sim 218$ K to an incommensurate CDW. In contrast to similar materials, e.g. NbSe$_3$, the whole Fermi surface of $\alpha$-TaS$_3$ contribute to the transition. The single crystal of $\alpha$-TaS$_3$ used in this study was synthesized by the standard chemical vapor transport method. The sample dimensions were 5 mm $\times$ 12 $\mu$m $\times$ 27 $\mu$m. The X-ray study was performed at BL02B1 of SPring-8. The synchrotron beam of 0.5 mm in diameter was illuminated through a hole of the sample holder.

Diffraction was monitored with a two-dimensional detector, Pilatus 100K [7], located 1303.7 mm away from the rotation center of a four-circle diffractometer, providing an angular resolution of 0.007559 degrees, corresponding to one pixel ($172 \times 172 \mu$m$^2$) of the two-dimensional detector. The incident beam passed through a double-crystal monochromator. The wavelength was 0.61984(89) Å, which was calibrated with standard CeO$_2$ powder prior to the measurements. Before each measurement, several reciprocal points were scanned to assign the peaks in order to prevent from confusing with any spurious signals. The sample was cooled to 7 K by a refrigerator mounted on the diffractometer. Temperature stability was maintained within $\pm 0.01$ K throughout the temperature range of the measurement.

The scattered X-ray beam was detected with the two-dimensional detector. The sample was fixed during exposure, typically for a duration of 100 seconds. Figure 1 shows temperature evolution of Bragg (002) and satellite ($-1 \ 1 \ 2$) + $\vec{q}$ peaks obtained at 180, 130, 80, 50, 30 and 7.3 K. The position of the satellite peak was determined as $\vec{q} = (0.5 \ 0.125 \ 0.25)$, which agrees with the results of previous studies [3, 4] within experimental errors. We also confirmed that the intensity of the satellite peak rapidly decreases at temperatures higher than the Peierls
3. Results and discussion

The predominant feature in the satellite profiles is the change in the number of peaks (Fig. 1). When the system undergoes the Peierls transition at 218 K, a satellite peak begins to develop in intensity and correlation. The initial position of the peak lies around $q_c = (0.255 \pm 0.002)c^\ast$. When we reduced the temperature, a second peak emerged close to the first. This second peak appeared at 130 to 50 K, while at temperatures below 50 K the number of peaks went back to unity. The difference between the two $q$’s is only $1.3 \times 10^{-3}c^\ast$, or two pixels at the detector, however, this is still larger than the spatial resolution of the experiment (i.e. $\sim 1$ pixel). In addition, since no anomalies were found in the Bragg peaks throughout the whole temperature range, we can rule out the possibility of the presence of any artifacts.

The positions of the two $q$’s at 80 K were assigned as $q_c = (0.252 \pm 0.002)c^\ast$ and $q'_c = (0.250 \pm 0.002)c^\ast$, where $q$ and $q'$ denote the satellites for higher and lower temperatures, respectively. Hence, the CDWs at higher and lower temperatures are assigned as incommensurate and commensurate, respectively. In a previous X-ray study [4] it was shown that the wavevector at temperatures around $T_P$ is incommensurate, $q_c \sim (0.255 \pm 0.003)c^\ast$, and by reducing the temperature it becomes nearly commensurate below 100 K, where $q_c \sim (0.250 \pm 0.004)c^\ast$. The error bars were mainly the result of the resolution of the experimental setup used at that time. For example, the full width at half maximum of the diffraction peak was as large as $0.01c^\ast$ in the experiment described in Ref. [3], whereas our synchrotron study has an advantage as regards the sharpness of the beam, which leads to a resolution of better than $1 \times 10^{-3}c^\ast$. In this context, our data can be understood as a natural extension of the previous results.

Details of the incommensurate-commensurate transition was investigated through the intensity evolution of the two CDWs. Intensity of each peak was obtained by numerical integration from the two-dimensional profile. Figure 2.a shows the relative intensity of the commensurate CDW ($q'_c = 0.250c^\ast$) normalized by the total satellite intensity, $I_{ic}/(I_{ic} + I_c)$.  

![Figure 2](image_url)
The broken line in the figure is a guide for the eye. It is shown that at temperatures of 50-130 K, where two CDWs coexist, the intensity of the commensurate CDW increases at lower temperature. The main difference revealed in this study is that the incommensurate CDW is not locked to be commensurate, although its wavevector approaches that of the commensurate CDW. The commensurate CDW begins to appear separately at around 130 K and develops at lower temperatures.

Since the longitudinal wavevector $q_c$ is very close to being commensurate, it is important to discuss the possibility of discommensuration. If the wavenumber $2k_F$ is very close to a commensurate length $Ma$, where $M$ is an integer, and $a$ is the lattice constant, discommensuration will occur per length $l_s$ [6]. This length is related to the difference between the CDW wavevector and the commensurate length,

$$\delta q = 2\pi/Ml_s.$$  \hspace{1cm} (1)

In our case, $M = 4$ and $\delta q = q_c - q'_c = 0.002c^*$. This gives the mean separation between discommensurations $l_s = 418$ Å.

We illustrate the change in the transverse correlation length schematically in Fig. 2.b. As discussed above, there are discommensurations, or defects, in the incommensurate CDW. Since the two CDWs have different wavevectors, there must be a misalignment of wavefronts at the border between these CDWs. In our case, since $q$ in the incommensurate CDW is larger than in the commensurate CDW, it is possible for dislocations to be located between them. In Fig. 2.b, the solid lines represent the wavefront, and the hatched circles are edge dislocations in the phase. There are additional wavefronts between two discommensurations.

The observed change in the peak profile can be understood if the length of the discommensurations is increased, resulting in the expansion of the incommensurate CDW. At present, there is no microscopic mechanism that agrees quantitatively with the observed phenomena. However, it is worth noting that the edge dislocations in a CDW move easily in the transverse direction [8]. For a complete understanding we must determine the force exerted on the dislocations, for example, through experiments on sliding regime of the CDWs.

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