Experimental report

19/09/2016

Proposal:	**5-32-829**		**Council:**	4/2016
Title:	The study of the low-temperature CDW Mott state of TaS2			
Research area:	Physics			

This proposal is a new proposal

Main proposer:	**Je-Geun PARK**
Experimental team:	marie KRATOCHVILOVA Lucile MANGIN THRO
Local contacts:	Andrew WILDES

Samples: 1T-TaS2

Instrument	Requested days	Allocated days	From	To
D7	14	5	15/06/2016	20/06/2016

Abstract:

1T-TaS2 is a prototypical quasi-2D metallic compound revealing strong electron-phonon (e-p) coupling responsible for charge-density-wave (CDW) instabilities. The compound undergoes three successive CDW transitions; first, to an incommensurate CDW phase below 540 K and then to a nearly commensurate CDW phase at 350 K and finally to a commensurate one below ~160 K. The nature of this commensurate CDW phase is under intensive investigation. In particular, it is very interesting that 12 out of 13 Ta4+ d-electrons form some kind of molecular orbitals in David star patterns, freeing one 5d-electron. The behavior of this orphan spin at low temperatures receives currently attention of the scientific community. For example, our susceptibility data measured on a high-quality powder TaS2 sample exhibit a strong Curie-Weiss tail, suggesting that some kind of short-range correlations might as well exist at low temperatures. Thus we propose here to experimentally test this scenario by using polarized neutron option of D7 and specifically we want to measure its short range correlations.

Experimental report – proposal No. 75339: The study of the low-temperature CDW Mott state of TaS_2.

Low-dimensional electron systems realized in layered materials often form so-called charge density waves (CDW); a phenomena which have attracted enormous attention recently. The 1T-type tantalum disulfide is a well-known quasi-2D metal which undergoes a series of first-order phase transitions to CDW, Mott and superconducting phases (see e.g. [1] and references therein).

The compound has a lamellar structure formed by sheets of edge-linked TaS_6 octahedra with the weak Van der Waals bonding within the sheets. The Ta-atoms form a hexagonal lattice which exhibits a series of structural modulations as temperature decreases [2]. The high-temperature unmodulated structure is trigonal with *P-3m1* symmetry (a = 3.36 Å, c = 5.90 Å). Below ~540 K, an incommensurate CDW phase is established. Upon further cooling, the structure changes to a nearly commensurate CDW at ~350 K. Finally, below ~160 K we observe the commensurate CDW phase forming a hexagonal pattern of David-star clusters of 13 Ta-atoms. The formation of the commensurate CDW phase involves Mott-Hubbard physics leading to intricate electron–phonon and electron-electron interactions [3]. As each Ta^{4+} ion provides one 5*d*-electron, there are 13 5*d*-electrons per each David star. Out these 13 electrons 12 electrons form 6 covalent bonds leaving one 5*d*-electron free, i.e. S = 1/2 per each David star. What happens to this orphan spins with a large spin-orbit interaction is currently an interesting question. There is a suggestion that it may form a highly correlated phase such as a magnetic short-range ordering. The main aim of this proposal was to directly measure the expected short-range correlations in this interesting CDW Mott phase.

We have measured approx. 4.2 g of 1T-TaS_2 powder in a double-wall Al cylinder can using λ = 4.8 Å at 1.5K only during the whole measurement time of 5 days due to a very weak signal. The data were corrected to the background using cadmium, for finite polarization using quartz and normalized to the absolute values using vanadium. The nuclear, magnetic and nuclear spin-incoherent scattering cross sections were separated as a function of both momentum and energy transfer using the 6-point 'xyz'-polarization analysis [4]. The measurement was performed in the 1:8 flipping ratio.

The diffraction revealed several small Bragg peaks shown in Fig. 1 which do not belong to the major 1T-TaS_2 phase; however, the spurious phase was not identified.

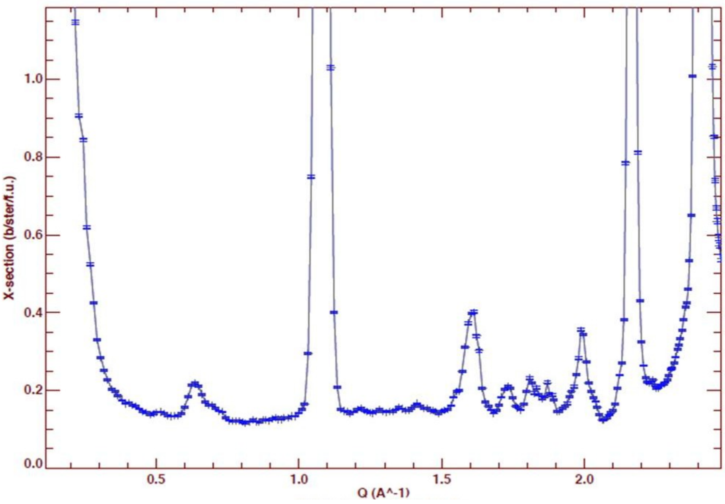

Fig. 1: The total scattering from 1T-TaS_2 at 1.5K. The small peaks at 0.6, 1.6 and 2 $Å^{-1}$ do not belong to the major TaS_2 phase.

We have observed a possible broad bump from Q = 0.7 to 1.2$Å^{-1}$ (assuming the background level is shifted slightly below zero value), however as the estimated magnetic signal coming from the expected orphan spin short-range order was close to the limits of the D7 instrument, the results of this experiment are inconclusive and need to be verified by another type of experiment, such as μSR (see Fig. 2).

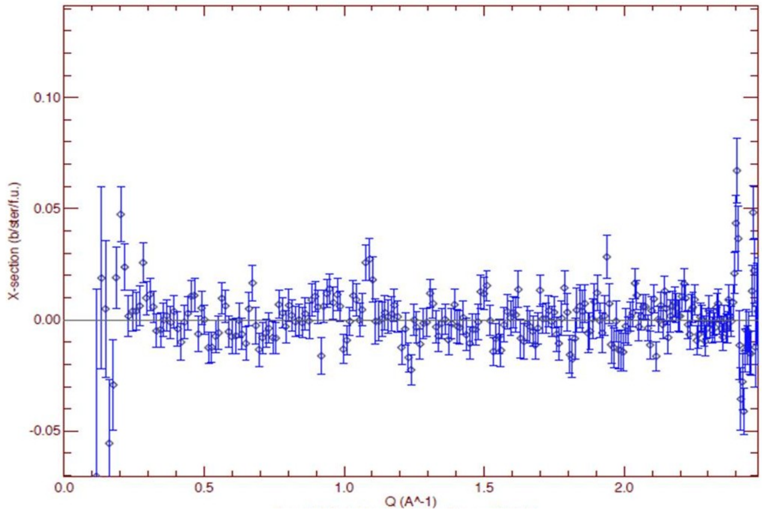

Fig. 2: The magnetic scattering from 1T-TaS_2 separated using the 6p method at 1.5K.

References:

[1] Sipos, B., Kusmartseva, A. F., Akrap, A. Berger, H., Forro, L., Tutis, E., Nat. Mat. 7, 960–965 (2008).

[2] Spijkerman, A., de Boer, J. L., Meestsma, A., Wiegers, G. A. &Smaalen, S., Phys. Rev. B 56,13757–13767 (1997).

[3] Zhu, X., Cao, Y., Zhang, J., Plummer, E. W., Guo, J., PNAS 112, 8, 2367–2371 (2015).

[4] Stewart, J. R., Deen, P. P., Andersen, K. H., Schober, H., Barthélémy, J.-F., Hillier, J. M., Murani, A. P., Hayes T. and Lindenau B., J. Appl. Cryst. 42, 69-84 (2009).