Research Activities

– Off-line Experiments –

Tracking charge-density wave formation and detangling dichroic signatures in VSe_2

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Keywords: Transition-metal dichalcogenides (TMD), Charge Density Wave (CDW), Laser-ARPES

Group V transition-metal dichalcogenides (TMDs) have the chemical formula MX_2, where M=V, Nb, Ta and X = S, Se, Te. The metallic variants are well-known systems for supporting charge density wave (CDW) formation [1].

The effects of the CDW on the electronic structure of 2H-$NbSe_2$ and 2H-$TaSe_2$ are well understood [2,3], with clear signatures in spectroscopic techniques. VSe_2 is not as well studied. Its CDW is known largely from transport measurements, with only weak potential signatures visible in ARPES thus far. Its thermodynamic preference for the 1T-structure as opposed to the 2H-structure, as well as its complex three-dimensional Fermi surface means that the resulting CDW phase could be rather different from that of its 2H-structured sister compounds.

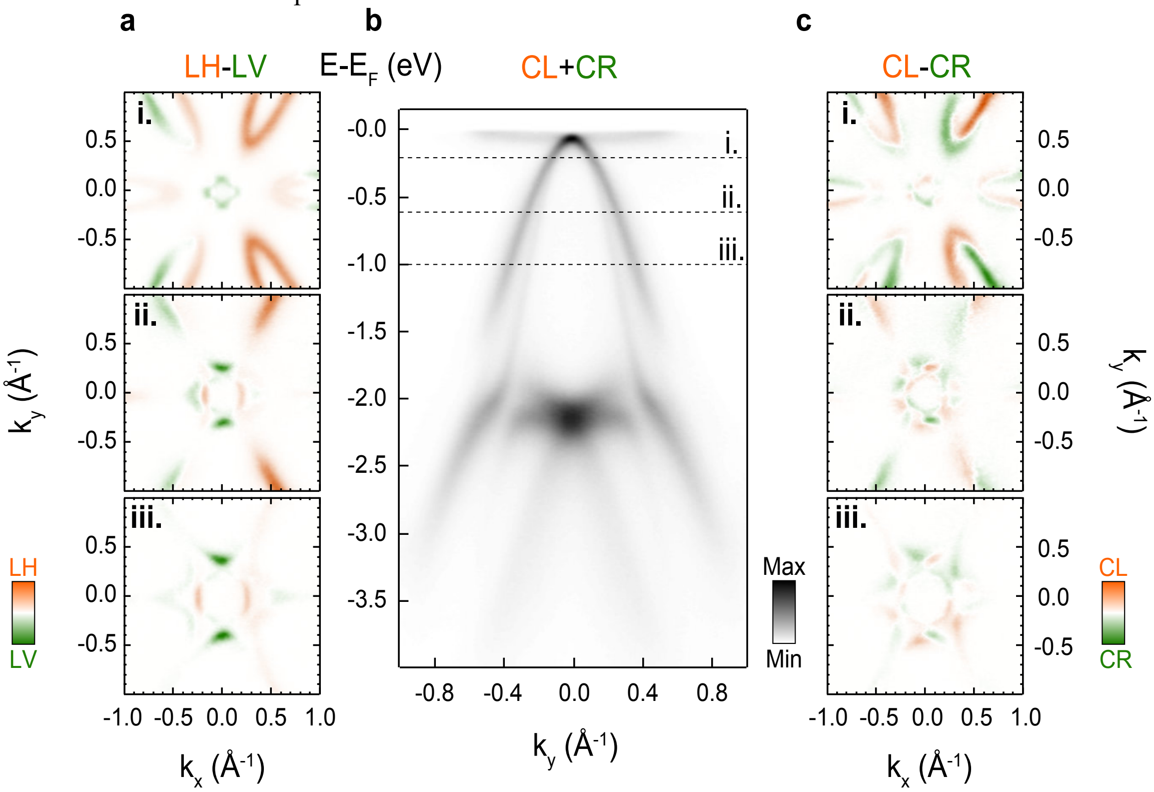

FIGURE 1. Previous APRES data taken in the CDW phase of 1T-VSe_2. The data was collected at the CASIOPEE beamline of SOLEIL, Paris, with 110eV light in linear horizontal (LH), linear vertical (LV), circular left (CL) and circular right (CR) polarized light.

Our recent ARPES measurements (Figure 1) additionally show extremely complex dichroic signatures with both linearly and circularly polarized light of both the flat transition metal derived conduction bands and the dispersive chalcogen derived valence bands, further motivating the need for a higher resolution spectroscopic study.

A dataset obtained with the 6eV laser source at HiSOR is shown in Fig. 2, probing the chalcogen p-valence states only. This is qualitatively similar to the p-bands measured in synchrotron ARPES (Fig. 1). However, the measured linewidth of the bands shown here is on the order of 100meV, much larger than the intrinsic instrumental resolution [4]. We attribute this to a non-uniform cleaved surface, with a potentially curved surface or spatially-dependent local workfunction variations leading to substantial extrinsic band broadening within our probing light spot area.

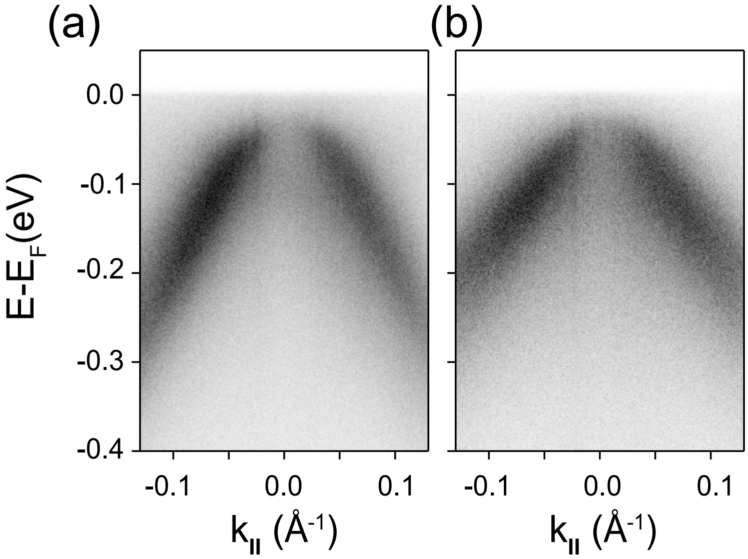

FIGURE 2. Low temperature phase of VSe_2 probed with 6eV light in (a) horizontal and (b) linear polarization.

Moreover, matrix elements lead to a consistent suppression of the spectral weight at the Γ point, unlike at the higher photon energies shown in Fig. 1 (but similar to features observed in some other transition-metal dichalcogenides [5]). This is true irrespective of the probing light polarization (Fig. 2). Ultimately, together, these two effects hindered our efforts to track any changes in the band structure through the charge density wave transition (T_C = 110K).

In conclusion, a change of band dispersion was observed between linearly vertical and linearly horizontal polarized light, but poor linewidths intrinsic to our samples coupled with a matrix element effect suppressing weight at the VBM meant that neither the subtleties of the mechanism behind complex dichroic signatures, nor that of the charge density wave formation could be addressed.

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Observation of *d* electron quantum-well states in Pd(100) ultrathin films

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Keywords: Pd, ultrathin film, quantum well states.

When the film thickness of the metal corresponds to the Fermi wave number, the quantum well (QW) states are formed in the thin film[1,2]. It is reported that the QW states formed in metal thin films bring peculiar physical properties periodically as a function of the film thickness [3]. It was also confirmed that in the Pd (100) ultrathin films, ferromagnetism appears periodically depending on the film thickness[4,5]. In terms of the Stoner model, which is the simplest model of the ferromagnetic expression in metals, it is believed that density of states around ε_F in the Pd(100) thin film increase by the quantum confinement of the *d* electron[6], and the thickness dependent ferromagnetism appears. Due to the possibility of developing new magnetic nanoscale materials whose magnetism can be controlled, this topic is of great technical concern.

ARPES is an ideal tool for the study of QW by which it may be able to directly observe subbands caused by QW states. In particular, it is expected that high resolution measurement can be performed in a Laser ARPES apparatus using a laser with high coherency as a light source. In this research, we aim to directly discuss the relation between magnetism and QW state by observing the QW state of *d* electron formed in Pd (100) ultrathin film by ARPES.

All experiments were performed at the HiSOR Laser ARPES apparatus. We used Cu(100) single crystal as a substrate of Pd ultrathin films to avoid charging. To get a clean and flat surface, we repeated cycle of Ar^+ sputtering subsequent annealing at 900 K in the ultra-high vacuum chamber until clean Auger electron spectroscopy (AES) and sharp low energy electron diffraction (LEED) spots are obtained. Pd was deposited on the Cu(100) substrate at the rate of 0.5 monolayers(ML) per minutes at room temperature. We estimated thickness of Pd ultrathin films by means of a reflected high energy electron diffraction (RHEED) oscillation technique [Fig. 1(a)].

Figure 1(b) shows LEED image of Pd film on Cu(100) substrate. Four-fold sharp spots were observed, indicating the epitaxial grew of Pd on Cu(100).

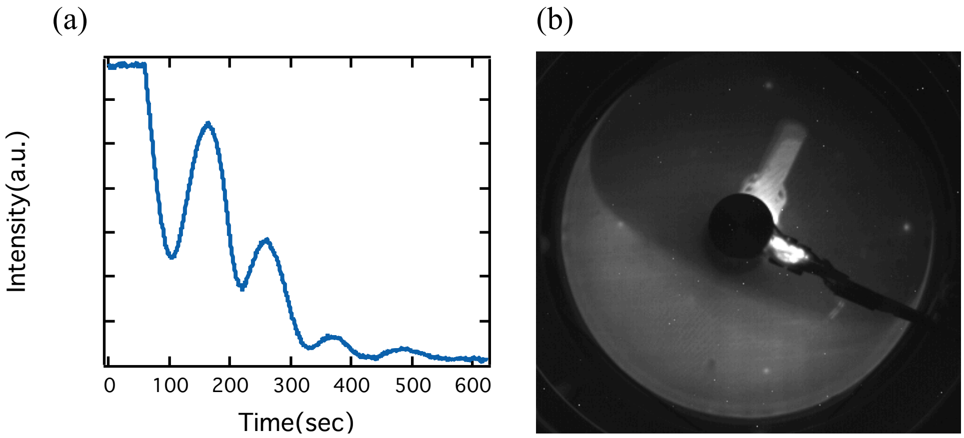

FIGURE 1. (a) RHHED oscillation during the deposition of the Pd on Cu(100) substrate. The oscillatory change in the RHHED intensity was observed, and the film thickness was estimated from the period of oscillation. (b) LEED image of the Pd film of Cu (100) substrate. Sharp spots were observed.

Figure 2 shows image plots of the obesrved band disperisons along the ΓM line of the 5.2 ML Pd film taken with $h\nu$ = 1.4 eV at 30 K. Clear band dispersion was not observed. It may be due to that the photoionization cross section became small by using the low photon energy light hν = 1.4 eV. Therefore, It is expected that clear band dispersion can be observed using higher photon energy light.

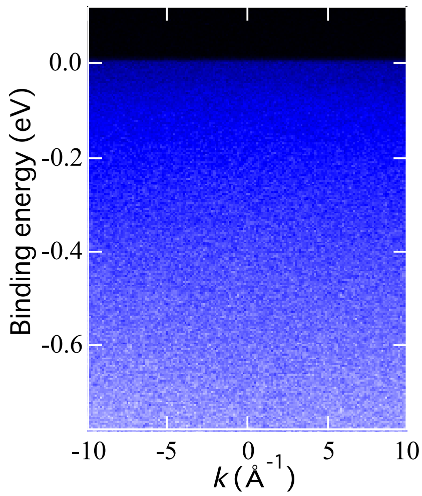

FIGURE 2. Band dispersion of the 5.2ML Pd film taken with $h\nu$ = 1.4 eV at 30 K. Clear band dispersion was not observed. This nay be due to that the photoionization cross section became small by using the low photon energy.

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Micro-ARPES study of a Weyl semimetal candidate $MoTe_2$

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Keywords: Weyl semimetal, Fermi arc, Topological surface states, $MoTe_2$, Micro-ARPES.

Three-dimensional (3D) topological Weyl semimetals are a new class of quantum materials that show peculiar electronic structures, unique quantum phenomena such as a chiral anomaly, anomalous Hall effect and negative magnetoresistance, and have a broad potential for device applications [1-7]. The electronic states of Weyl semimetals are characterized by the topology of the bulk bund structure, which leads to the presence of bands with liner dispersion, like graphene, around the nodes called Weyl points. Together with this graphene like property, Weyl semimetals show unique spin texture originating from the Weyl points that act as monopoles in the momentum space. These monopoles-like Weyl points always show up in pair, and the plane between a pair of Weyl points has Chern number C=1. Owing to this unique bulk electronic structure, the surface states of Weyl semimetals, which are topologically protected, show unclosed Fermi surface called Fermi arc at the Fermi level (E_F). This Fermi arc is a hallmark of 3D Weyl semimetals. The Weyl semimetals can be further classified into two types, the type-I in which the Lorentz symmetry is approximately preserved and the type-II in which this symmetry is broken and have tilted Weyl cones with both electron and hole pockets at the E_F. Due to the difference in these two types, the Weyl Fermions of type-II leads to further intriguing physical properties [8].

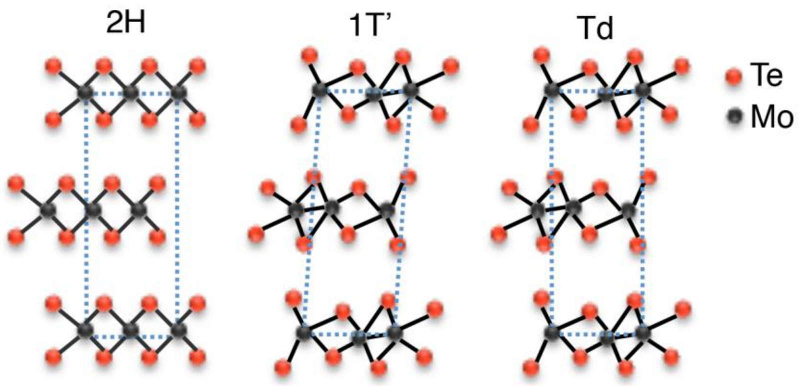

FIGURE 1. Crystal structures of $MoTe_2$ crystals. The most stable 2H has a semiconducting electronic states and the 1T' has a semimetal band structure. Td appears by cooling the 1T' phase.

Recently, type-II Weyl semimetals has been proposed to exist in layered transition metal dichalcogenides, such as $MoTe_2$ [8-12]. The most stable crystal structure of $MoTe_2$ is the hexagonal 2H phase that shows a semiconducting electronic property, and the next most stable structure is the monoclinic 1T' phase that shows a semimetal electronic states. The orthorhombic noncentrosymmetric Td phase, which appears by cooling the centrosymmetric 1T' down below 250 K, is proposed to be a Weyl semimetal. The Td-$MoTe_2$ would have 8 Weyl points that are located at binding energies of 6 meV and 50 meV above E_F, and the Fermi arc is predicted to be looked between the hole and electron pockets of the Fermi surface.

However, also several angle-resolved photoelectron spectroscopy (ARPES) measurements have been performed on the Td-$MoTe_2$, no strong evidence on the Fermi arc has been reported so far. The main reason for the lack of observation would be come from the quality of the sample. That is, it is difficult to grow 1T'- $MoTe_2$ that is large enough for normal ARPES measurements (ARPES using light whose spot size is in the order of 100 μm).

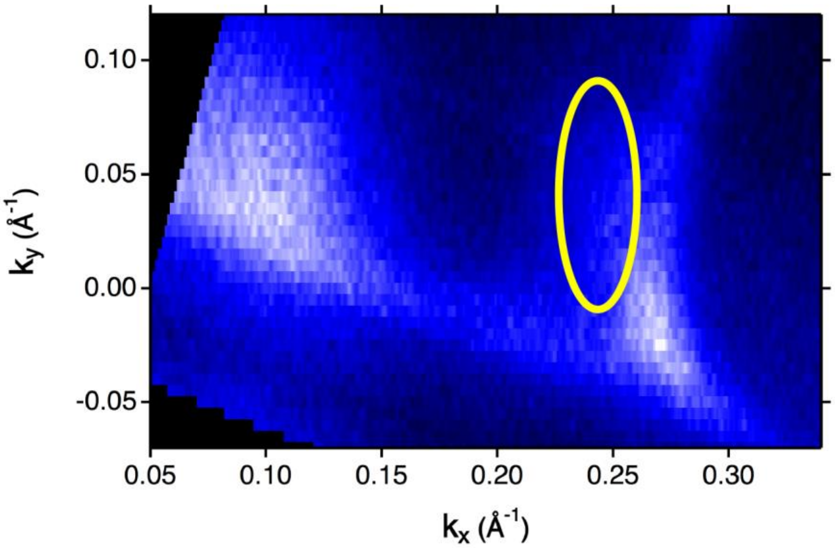

FIGURE 2. The Fermi surface of the Td-$MoTe_2$ single crystal obtained using the laser-based micro-ARPES apparatus at HiSOR.

In order to solve this problem and to observe the Fermi arc and/or the topological surface states, we have performed laser-based micro-ARPES measurements at HiSOR. The light spot of this apparatus is approximately 5 μm. By checking the ARPES spectra at different sample positions, we notice that our Td-$MoTe_2$ sample contains a lot of domains, and that the size of one single crystal domain is approximately 30x30 μm^2. Figure 2 shows the Fermi surface of the Td-$MoTe_2$ sample, measured after cleaving in ultra high vacuum. We clearly observed the electron and hole pockets, and trace of the Fermi arc (indicated by the yellow circle in Fig. 2). Although the Fermi arc is not clear (its intensity comparable to those reported in the literature), we conclude that $MoTe_2$ would be a type-II Weyl semimetal based on the band dispersion of this "Fermi arc". In this paper, we will first compare the present results and those reported previously, and will discuss this "Fermi arc" and the topological surface state in more details.

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