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MODELLING THE CHARGE-DENSITY WAVE STATE OF $TaSe_2$ WITH A TIGHT-BINDING MODEL

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TaSe2 is a layered transition metal dichalcogenide (TMD) that exhibits charge density waves. In order to describe this electronic phase in the bulk crystal, we construct an effective two band model for the Fermi level states.

1. WHY TASE$_2$?

TMDs are a large group of materials that consist of strongly bonded layers of atoms, each of which interacts with the others via the weak van der Waals interaction. Each of these layers are made of planes of transition metal atoms between two planes of chalcogens. Here we focus on bulk TaSe2 in the so-called 2H hexagonal structure, corresponding to the stacking of transition metal/chalcogen “sandwiches” as seen below.

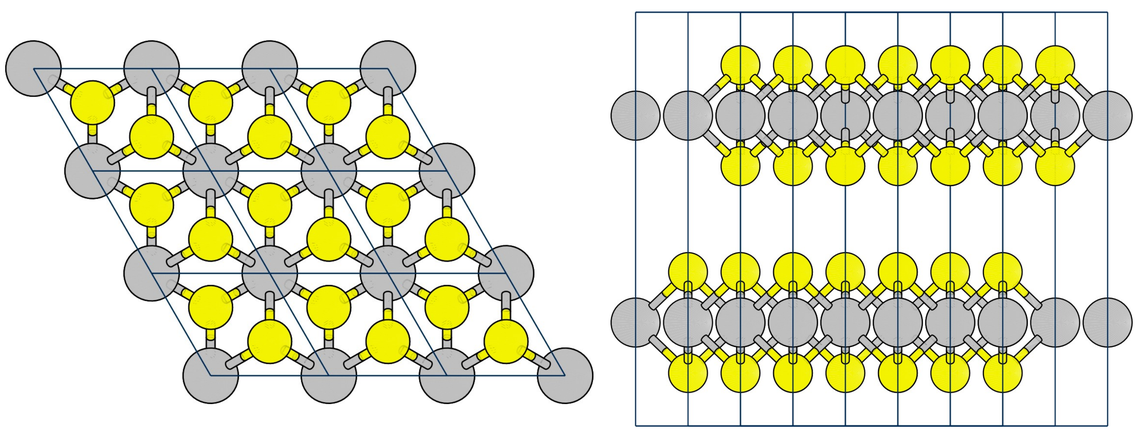
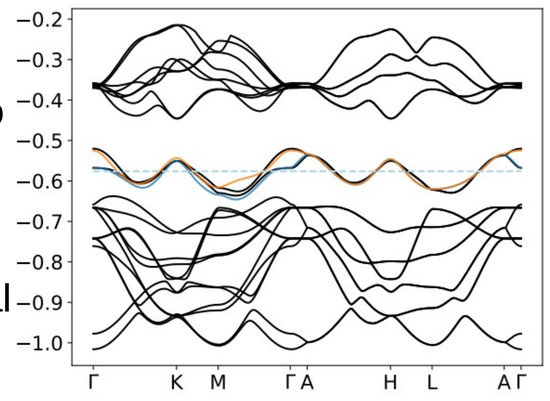


Figure 1: The crystal structure of Tase2, with Ta atoms shown in grey and Se atoms in gold. Note how the unit cell, shown as the blue lines contains two ”sandwiches” of three layers each.

TaSe2, like NbSe2, exhibits both commensurate and incommensurate charge density wave (CDW) states at around 100 K. Theoretical studies of NbSe2 by Flicker et al. revealed that the CDW transition is not driven by Fermi surface nesting, but instead by the momentum and orbital dependance of the electron-phonon coupling [1]. Their two-orbital model of NbSe2 describes the effective coupling between the d3z2−r2 orbitals of the two inequivalent niobium atoms and captures well the electronic structure in the vicinity of the Fermi level. Here, we follow their approach and aim to construct a two-orbital model for 2H-TaSe2.

2. AB INITIO CALCULATION

The tight-binding method used requires data to fit to in order to ascertain its free parameters (see Methods). For this we used density functional calculations, via QuantumEspresso [2].



The bandstructure produced is shown in black, *right.* We fit in the k_z=0 plane due to quasi-2d nature of $TaSe_2$. For fitting the two-orbital model we also use ARPES data below the Fermi energy to ensure an accurate model in a hybrid fit (see Methods) [3].

3. METHODS

The tight-binding model is constructed in an orbital basis using a Slater-Koster parametrisation. The Slater-Koster method has two sets of free parameters: the (two-centre) hopping amplitudes between atomic orbitals and the overlap integrals that correct for the non-orthogonality of orbitals on neighbouring atoms. Stated thusly,

$$H\,|n\rangle = E_n S\,|n\rangle$$

the problem consists of finding the matrices H, the Hamiltonian, and S, the overlap, and solving the eigenproblem. We consider each of the Ta atoms to contribute five d-orbitals, and each Se 3 p-orbitals. The Slater-Koster method, for this set of orbitals, yields 33 free parameters considering only on-site energies and first-nearest neighbours for each pair of non-equivalent atoms.

Using the orbital basis in the TB model we calculate the contribution of each orbital $|\psi_i\rangle$ to each eigenstate $|v_j\rangle$ as $|\langle\psi_i|v_j\rangle|^2$ for the Fermi bands. The contribution from the d_{z^2} orbitals dwarfs all others and justifies the use of an effective model in the basis of Bloch states constructed of these orbitals.

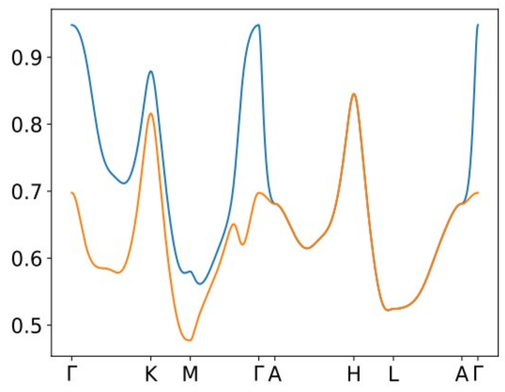


Figure 2: Fraction of total orbital contribution from the two isolated Fermi bands. These contributions dwarf that of any other orbital, justifying a two-orbital model.

We then construct a tight-binding model considering only these two orbitals on each Ta atom. To provide additional fitting parameters, we extend the model to include fifth-nearest-neighbour hoppings in-plane. Additionally, we consider the hopping integrals themselves the free parameters, rather than using the Slater-Koster formalism, as this minimises unknowns. We fit to both ARPES and DFT data to accurately model the material, shown in Figure 3.

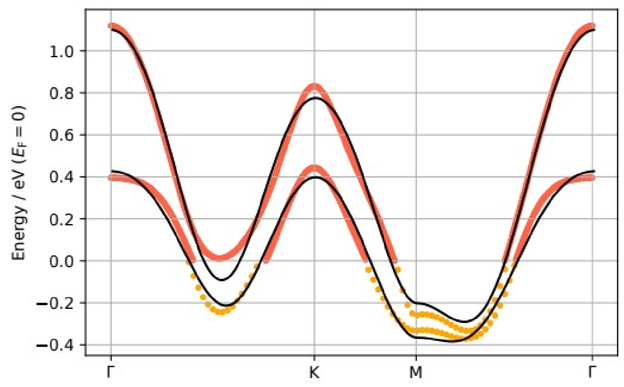


Figure 3: A fitted two-orbital model, shown in black, as compared to the ab initio calculations in red and ARPES measurements in yellow. This “hybrid” fitting approach yields accurate calculation of the CDW Q-vector.

4. CHARACTERISING THE CDW STATE

The CDW state in 2H-TaSe2 has a Q-vector = 2/3ΓM, i.e. one can expect to see folding in the bandstructure translated by this nesting vector when in the CDW state [3].

Once the two-orbital fit has been completed, it can be used to calculate the electronic susceptibility using the electron-phonon coupling expression of Varma et al. [4]. This susceptibility, projected along the direction of momentum transfer in the CDW state, is sharply peaked along the direction of the CDW wavevector Q = 2/3ΓM, as seen with peaks in Figure 4.

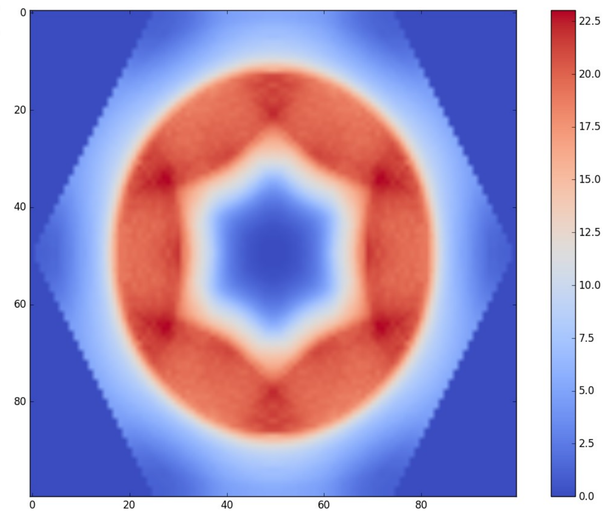


Figure 4: Calculated general electronic susceptibility in the CDW state. Note the peak in intensity at Q = 2/3ΓM, where a particular phonon will soften to zero frequency at the transition temperature, locking in the lattice distortion and transitioning to the CDW state.

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REFERENCES

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