INVESTIGATION OF DIRAC AND 2D MATERIAL SURFACES WITH HELIUM ATOM SCATTERING



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Why use Helium Atoms?

- Helium atom scattering (HAS) is a powerful technique to analyse the surface properties \bullet
- Helium atoms are scattered by the electron density 2-3 Å above the first atomic layer due to the Pauli repulsion \rightarrow non-destructive, strictly surface sensitive technique
- Typical energy range of helium atoms: 5-200 meV
- Helium atoms are neutral and chemically inert therefore insensitive to surface charges \rightarrow investigation of insulting materials
- As a noble gas helium is chemically inert \rightarrow completely inert investigation

Charge Density Waves in TaS₂

Monitoring the phase transition in TaS₂

- TaS₂ exhibits multiple charge density wave (CDW) states [2]
- Transition from a nearly-commensurate (NC-CDW) phase to a commensurate (C-CDW) charge density wave phase at roughly T = 220 K [3]
- Debye-Waller measurement: elastic peak intensity plotted against the surface temperature

$$I(T_S) = I_0 \cdot e^{-2W(T_S)}$$

with $W(T_S)$ being the Debye-Waller (DW) exponent [3]

Experimental Setup

Helium atom scattering apparatus and scattering geometry

- Helium beam is generated in the source chamber by supersonic expansion through a nozzle
- Helium beam is scattered in the main chamber at UHV-conditions and detected in a quadrupole mass-spectrometer
- Chopper is added into the beam for energy dependent measurements (TOF)
- Fixed source-detector angle $\theta_{SD} = \theta_i + \theta_f = 91.5^\circ$



Interaction processes at the surface

- **Elastic diffraction:**
- no energy exchange between the helium atoms and surface
- Inelastic scattering:

- Debye-Waller exponent contains contributions of all phonon modes \rightarrow measure for the electron-phonon coupling constant [4]
- Increased sample temperature leads to more vibrational modes \bullet \rightarrow decreasing specular intensity [4]
- Very sharp, stepwise onset in the intensity at the transition temperature \bullet
 - \rightarrow discontinuous, first-order transition
 - \rightarrow rapid increase in electron-phonon coupling [3]





- incident helium atom looses or gains energy via creating or annihilating a phonon
- Selective adsorption resonance (SAR):

helium atom is temporarily trapped in a bound state of the interaction potential

Surface Diffusion in Reciprocal Space

Quasi-elastic helium atom scattering

- Only experimental method to continually monitor mobile species on both Å length and ps timescales [1]
- Helium atoms scattered from moving molecules on the surface can exchange energy with the surface atoms resulting in in a broadening around the elastically scattered peak (analogous to Doppler broadening) \rightarrow quasi-elastic broadening of elastic peak [1]
- Extent and rate of broadening depends on rate and mechanism of diffusion



Inciden

wave

Helium spin-echo (HeSE) spectroscopy

Using nuclear-spin polarisation of He³ to measure coherence [1]

Quasi-Freestanding Epitaxial Graphene w/ Gallium intercalation

- Using confinement heteroepitaxy (Chet) an atomicallythin (2D) metal can be intercalated [5] at an epitaxial graphene (EG)/ silicon carbide(SiC) interface [5]
- The process of intercalation modifies the physical properties of the metal (e.g. enhanced conductivity, unique Raman features) [5,6]
- Nice and clear diffraction pattern from graphene in helium scattering
- DW-measurement at specular peak:
- $\rightarrow \theta_D = 530 \text{ K}$

 \rightarrow no transition in e-phonon coupling observable

- Incident wave is split into two components which arrive with a time spread t_{SF} at the surface
- Coherence of two components after scattering is measured
- Scattered waves differ as a result of changes on the surface during t_{sF} \rightarrow loss of correlation [1]
- Final outcome: Intermediate scattering function (ISF) $I(\Delta K, t)$
- For simple models of diffusion, the ISF decays exponentially [1]

Surface temperature T_S (K)

40

Outlook

- TaS₂: theoretical calculations (CC-calculations) to obtain information about the \bullet surface corrugation and the surface interaction potential
- QFEG_{Ga}: measuring disorder-to order transition at 285 K (observed in Raman spectroscopy); measurements of other QFEG-materials
- surface diffusion measurements of various molecules on different surfaces (e.g. water on hexagonal boron nitride (h-BN))

References		Acknowledgement	
 [1] A.P. Jardine et al., Prog. Surf. Sci. 2009, 84, 323-379 [2] G. Brusdeylins, Surf. Sci. 1989, 211-212, 98-105. [3] G. Benedek, et al., Cond. Matter 2020, 5, 79 	[5] M.T. Wetherington et al., 2D materials 2021, 8, 041003 [6] N. Briggs et al.,Nature materials 2020, 19, 637-643	The research is supported by the Austrian Science Fund (FWF) within the project P34704.	FШF
[4] G. Benedek, et al., Phys. Chem. Lett. 2020, 11, 1927-1933			Der Wissenschaftsfonds.

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