Introduction

Single layer (SL) tantalum disulfide (TaS_2) is a genuine two-dimensional (2D) metal, which belongs to a class of transition metal dichalcogenides (TMDCs) with strongly correlated electrons and therefore complex many-body interactions. It is hitherto unknown how the transition from the bulk to the SL limit affects these interactions, and ultimately the electron dynamics. Here, we provide the first direct femtosecond study of this 2D metal in a van der Waals heterostructure with graphene using time- and angle-resolved photoemission spectroscopy (TR-ARPES) at the Artemis facility. We simulate the measured spectral function in order to extract the electronic dispersion and temperature. We find that 40 fs after optical excitation of the SL-TaS_2, the electrons reach an exceptionally high temperature of 3080 K. The elevated electronic temperature is accompanied by a surprising renormalization of the electronic structure bandwidth, driven by interactions of the hot electron gas. Upon exploring the excitation and its temporal evolution for different fluences and sample temperatures, we find an ultrafast single-exponential decay of hot electrons, regardless of the different parameters employed.

A two-dimensional metal

For this experiment, SL-TaS_2 was grown on bilayer graphene on a doped silicon carbide substrate. The growth process yielded the 1H polymorph. The electronic structure of 1H-TaS_2 is characterized by a partially occupied band around the Fermi level in contrast to the semiconducting SL-TMDCs, such as MoS_2 and WS_2. Similar, however, to SL-MoS_2 on graphene, the SL-TaS_2 on graphene adopts multiple domain orientations due a relatively weak van der Waals interaction between the SL-TMDC and the underlying graphene layers [1].

Bulk TaS_2 hosts a plethora of intriguing properties including, charge density waves (CDWs), superconductivity (SC), and Mott states [2, 3]. At the SL limit, TaS_2 can adopt two different polymorphs: 1T and 1H. Using a physical vapour deposition approach, we find the SL-TaS_2 assumes a 1H structure when grown on bilayer graphene. Using TR-ARPES to access the time domain, we explore the electronic temperature of distinct states of the SL-TaS_2 and determine the lifetime of these excited carriers. In order to analyze the data, we simulate the (E,k,t)-dependent photoemission intensity obtained in the experiment, directly allowing us to extract the Fermi-Dirac function and electronic dispersion including many-body effects.

TR-ARPES measurements were achieved using optical pulses generated by a 1 kHz Ti:sapphire amplifier laser system with a fundamental wavelength of 785 nm. By seeding an optical parametric amplifier (He-Topas), we obtained *s*-polarized pulses with a photon energy of 2.05 eV for optical excitation of the SL-TaS_2. Photoemission of electrons along the high symmetry Γ-K direction in the Brillouin zone (BZ) was achieved with a *p*-polarized high harmonic probe pulse with an energy of 25 eV.

A snapshot of excited carriers

The first objective of the experiment was to extract the lifetime and investigate the thermalization of the excited electrons. In Fig. 1 we show the pump-probe TR-ARPES experiment of SL-TaS_2 on bilayer graphene on silicon carbide. Fig. 1a reveals a snapshot of the SL-TaS_2 band structure in the Γ-K direction of its BZ, see Fig. 1b, before the arrival of the pump pulse, i.e. t<0. The same bands are shown in Fig. 1c, 40 fs after the arrival of the pump pulse. The difference spectrum is plotted in Fig. 1d. The difference plot shows the intensity difference between the spectrum obtained before optical excitation and the spectrum acquired after optical excitation — in this case, 40 fs after the arrival of the pump pulse. To first order approximation, the red and blue false colour scale can be interpreted as the electrons and holes we induce with the pump pulse. The experiment was carried out for different pump fluences in order to tune the distribution of hot carriers and thereby the electronic temperature (see Fig. 1e). Note that the excited distributions can always be described by a Fermi-Dirac function (straight lines in panel (e)), demonstrating that carriers thermalize within our time-resolution of 40 fs.

In Fig. 1f, the time dependence of the observed difference signals reveals an ultrafast decay of 170 fs for the transient signal, which is expected for a metallic state where the phase space is large for both electron-phonon and electron-hole damping processes.

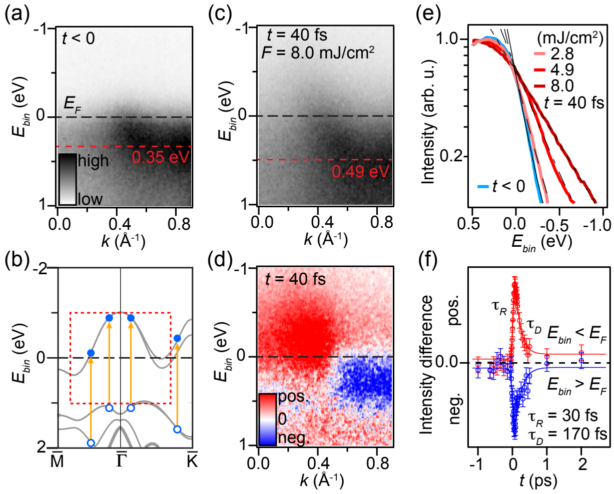

Fig. 1. TR-ARPES measurement of the 1H-TaS_2 dispersion around the Fermi level. (a) Measured spectrum before optical excitation, i.e. t<0. (b) Calculated dispersion from Ref. [4] with possible excitation processes. (c) TR-ARPES data acquired 40 fs after arrival of the pump pulse. (d) Difference spectrum obtained by subtracting panel (a) from (b). Red (blue) corresponds to excited electrons (holes). (e) Plot of the momentum-integrated intensity over the measured region in panel (a) and (c) for the stated time delays and pump fluence. Time-dependent intensity difference (markers) summed above and below the Fermi level.

Spectral function simulations

In order to simultaneously extract the electronic temperature and band dispersion from the TR-ARPES data for the SL TaS_2, a photoemission intensity simulation procedure was needed. Most importantly, the photoemission intensity is proportional to the spectral function and the Fermi Dirac distribution. The spectral function describes the bare bandstructure and the electronic self-energy which accounts for dispersion renormalization and spectral linewidths. The Fermi-Dirac distribution directly measures the electronic temperature T_e. From Fig. 1, it can be seen that the SL-TaS_2 state around the Fermi level is parabolical and thus we assume the dispersion can be modeled as a simple parabolic band as for a free 2D electron gas. This model allows us to re-write the photoemission intensity in terms of a set of fitting parameters such that we can simulate the measured spectra and fit these parameters at each fluence and time-delay. This fitting procedure (lower panels of Fig. 2) reproduces the experimental data (upper panels of Fig. 2) exceptionally well.

Hot electrons

By following the response in the spectral function with femtosecond time resolution, we have access to the statistical distribution of hot electrons. Our quantitative analysis method permits us to conclude that the thermalization to a hot Fermi-Dirac distribution having a temperature higher than 3000 K takes place on an ultrafast time scale of 40 fs; see Fig. 2(d) and

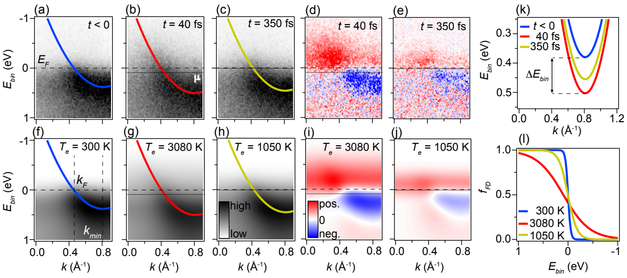

Fig. 2. Snapshot of the excited carriers in SL-TaS2 on graphene on silicon carbide and spectral function simulations generated from the 2D fitting procedure. (a)-(c) TR-ARPES spectra acquired at the stated time delays for pump energy of 2.05 eV and fluence of 7.8 mJ/cm2, and a sample temperature of 300 K. (d)-(e) Difference spectra obtained by subtracting panel (a) from (b)-(c). (f)-(j) Simulated spectra corresponding to the experimental data in panels (a)-(e). The parabolic dispersions extracted from the simulations are shown in each case in (a)-(c) and (f)-(h) and have been plotted together in (k) with a t-dependent shift marked as ΔE_{bin}*. (l) FD functions obtained from the simulation with the given electronic temperatures* T_e*, which have also been provided in each case in (f)-(j). Note that an error bar of ±200 K accompanies* T_e*.*

(i). As a result, the temperature of the hot electrons can be determined immediately upon excitation and its temporal evolution followed for different fluences and sample temperatures. We find that an ultrafast single-exponential decay of the hot electron temperature is found regardless of these different conditions (see Fig. 3(a)). The decay times seem to depend on the initial density of excited carriers – a higher density leads to a faster decay. Intriguingly, we also observe a strong renormalization of the electronic bandwidth on the order of 100 meV. As seen in Fig. 3(b), the renormalized bandwidth is correlated with the electronic temperature. We believe that our measurement provides access to the temperature dependence of the electronic self-energy. Comparison with a bare interacting 2D electron gas reveals that this temperature dependence can be quite strong [5] – a property that is carried over to our more complex 2D TaS_2.

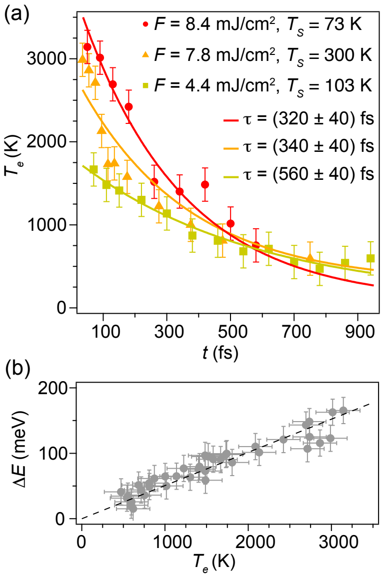

Fig. 3. (a) Time dependent dynamics of the fitted electronic temperature at the given pump laser fluence and sample temperature T_S*. (b) Electronic temperature dependence of* ΔE_{bin} *corrected by the change in chemical potential combining all data sets obtained for a wide range of time delays, fluences and sample temperatures. The dashed line is a guide to the eye tracking the extracted* ΔE_{bin} *values before taking into account the chemical potential.*

Conclusions

Using the tunable pump pulses and high harmonic UV pulses available for TR-ARPES experiments at the Artemis facility we have been able to determine a decay time of 170 fs for the excited electrons. 40 fs after optical excitation of the SL-TaS_2, the electrons reach an exceptionally hot temperature of 3080 K. Upon exploring the excitation and its temporal evolution for different fluences and sample temperatures, we find that bandwidth of the electronic dispersion renormalizes due to the elevated electronic temperatures. This renormalization is a fingerprint of the electronic self-energy, which is modified due to the temperature-dependence of electron-electron interactions in the 2D metal.

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