Observation of charge density wave melt down in time resolved ARPES of single layer VSe_2

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Introduction

Though the charge density wave (CDW) systems are one of the most studied system in condensed matters physics, the detail mechanism of CDW still remains unclear. Experiments have shown that the electron-phonon coupling or other factors may play important role in CDW in higher dimensional systems than the Fermi surface (FS) nesting [1]. Group V transition metal dichalcogenide (TMD) VSe_2 shows charge density wave (CDW) transition at 110 K with the CDW ordering ($4 \times 4 \times 3$) in its bulk form. Recent experiments have shown that the charge ordering gets enhance in the monolayer (ML) VSe_2 with an increased transition temperature ~ 140 K with very different charge order structure. Moreover, in CDW phase almost the whole Fermi surface vanishes forming CDW gap [2, 3]. It is very clear that the nature and the mechanism of CDW in bulk and ML VSe_2 are very different. Here we use time and angle resolved photoelectron spectroscopy (trARPES) to understand the electron dynamics in both CDW (88 K) and normal phase (200 K) of ML VSe_2. We have also modelled the ARPES intensity to extract different many-body and transient parameters from our data.

Experimental detail

ML VSe_2 samples were prepared using molecular beam epitaxy (MBE) on bilayer graphene on SiC substrate. The samples were Se capped before transporting in air. The samples were de-capped in ultra high vacuum (UHV) prior to experiment. For the trARPES measurements, we used a 29.6 eV probe pulse with a time-resolution of 40 fs for the main photoemission and a 1.55 eV pump pulse. The schematic of our sample and the trARPES measurements are shown in figure 1(a). We have measured the electronic structure along $\bar{\Gamma}$ - $\bar{M}$ direction as shown in the inset. Figure 1(b) shows the ARPES spectra from the sample at 200 K before the arrival of pump pulse, i.e. delay (t) < 0 and the lines are the calculated density functional theory (DFT) bands. The intensity evolution in the boxes 'a' and 'b' are shown in figure 2 (f) and (g).

Simulation of ARPES intensity

The ARPES intensity (I) can be expressed as the product of the matrix element (M), the quasi particle spectral function (A) and the Fermi Dirac distribution function (f) as shown in eq. (1). For interacting systems, the spectral function can be written as eq. (2), where ϵ is the non-interacting band structure and Σ_1 and Σ_2 are the real and imaginary parts of the self-energy respectively.

$$I(k,\omega) \propto |M^k_{f,i}|^2 . A(k,\omega) . f(\omega) \quad \ldots..(1)$$

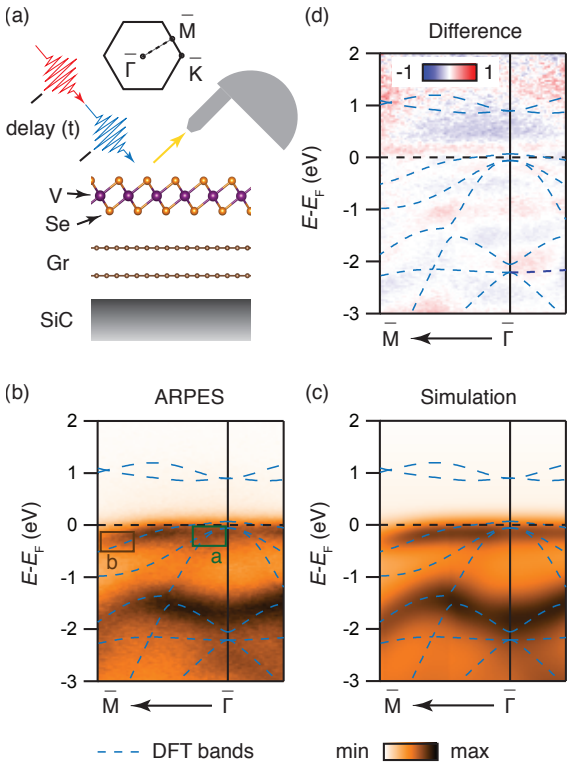

Figure 1 (a) Schematic of monolayer VSe_2 sample on bi-layer graphene and time resolved ARPES. Inset shows the monolayer Brillouin zone and the measurement direction, $\bar{\Gamma}$ - $\bar{M}$. (b) ARPES spectra collected at 200 K sample temperature before arrival of pump pulse. (c) Simulated spectra. (d) Plot of (data – simulation) / data.

$$A(k,\omega) = \frac{\pi^{-1} \times |\Sigma_2(k,\omega)|}{[\omega - \epsilon^b_k - \Sigma_1(k,\omega)]^2 + [\Sigma_2(k,\omega)]^2} \quad \ldots..(2)$$

Here we have used DFT calculated bands as the bare bands and a modified BCS type self-energy [4] (eq. (3)) is used to include the many body effects and the phase transition. All the parameters are extracted by fitting the simulated spectra to the ARPES data.

$$\Sigma(k,\omega) = \Sigma_0 - \frac{1-z}{z}\omega - i\Gamma_1 + \frac{\Delta^2}{\omega + \epsilon_k + \Sigma_0 + \frac{1-z}{z}\omega + i\Gamma_0} \quad \ldots.(3)$$

Figure 1(c) shows the simulated spectra and the corresponding relative error is shown in figure 1(d). Our simulated spectrum shows excellent match with the ARPES data. We find that our sample is fairly correlated with a band renormalization factor 0.52 for the band close to the Fermi level. Rest of the bands are reproduced well without including any self-energy correction.

Electron dynamics

The evolution of the electronic structure at different time delay is shown in figure 2. The electronic temperature (T_e) is extracted from the simulation as the temperature in the Fermi Dirac distribution function which is different from the sample temperature due to the pumping. The change in electronic temperature is plotted for both the sample temperatures in figure 2(a). In both cases it shows a rapid increase in T_e after the arrival of the pump pulse followed by a relatively slower decay. The decay constants for the CDW and the normal phases are found to be 207 fs and 293 fs respectively. Also, the maximum change in T_e for the normal phase is 1660 K whereas, for the CDW phase its only 960 K. We will be focusing our discussion for two regions – I: t ~ 40 fs, at the peak of the excitation and II: t ~ 3000 fs.

The ARPES spectrum for t < 0 fs (onset) is subtracted from the spectrum at t ~ 40 fs and the difference spectrum for the both the phases are shown in figure 2(b) and (c). The blue and red colors show the excess holes and electrons compare to the onset data respectively. At peak excitation both the data looks similar – excited electrons just above E_F without any sharp features and holes just below E_F. Such scenario can be explained considering the rapid decay of the hot electrons and holes due to scattering and Auger processes. But, a very different scenarios can be observed in region II. The difference spectra at t ~ 3 ps is plotted in figure 2(d) and (e). For the normal phase the excess holes are concentrated away from $\bar{\Gamma}$ point and the electrons around the $\bar{\Gamma}$. This can be explained by considering a lower electronic temperature in our simulation. But completely opposite behavior is observed for the CDW phase and we needed to consider a transition from the gapped phase to non-gapped phase due to pumping for proper simulation.

The melting of the CDW picture can also be verified by comparing the evolution of the intensity of the top filled band with pump-probe delay at different momentum. This band has dominant V *d* character and forms a hole pocket at $\bar{\Gamma}$. It crosses the E_F at about 0.2 $Å^{-1}$ in the normal phase. So below the transition temperature it is expected to get back folded intensity close to $\bar{\Gamma}$. The change in integrated intensity close to $\bar{\Gamma}$ (box 'a' in figure 1(b)) and away from $\bar{\Gamma}$ (box 'b' in figure 1(b)) are plotted in figure 2(f) and (g) respectively. We can see the evolution of intensity is same at both the sample temperatures in box 'b' (fig. 2(g)) but, for box 'a', at 88 K sample temperature, the intensity change is higher compare to the sample at 200 K. This extra intensity can be attributed to the back-folded spectral weight, which appears close to $\bar{\Gamma}$ and vanishes due to the melting of CDW after the arrival of the pump pulse.

We can also see even after 3 ps the charge order is not recovered and the system is still excited - the electronic temperature remains stable at a value higher than the sample temperature in both phases. This points toward some bottleneck in the decay mechanism of the hot carriers. One may need to consider the substrate induced effects to explain this long timescale behavior. This is especially crucial as earlier studies has shown the substrate may affect the charge ordering in ML VSe_2 [3].

Conclusions

We are able to simulate the ARPES intensity very accurately and extract different parameters related to the many-body effects and electron dynamics using this simulation. The hot electron dynamics in ML VSe_2 is very different in CDW and normal phase. We were able to melt the charge ordering using pumping. The observed long timescale behavior may be due to the substrate and needs further study.

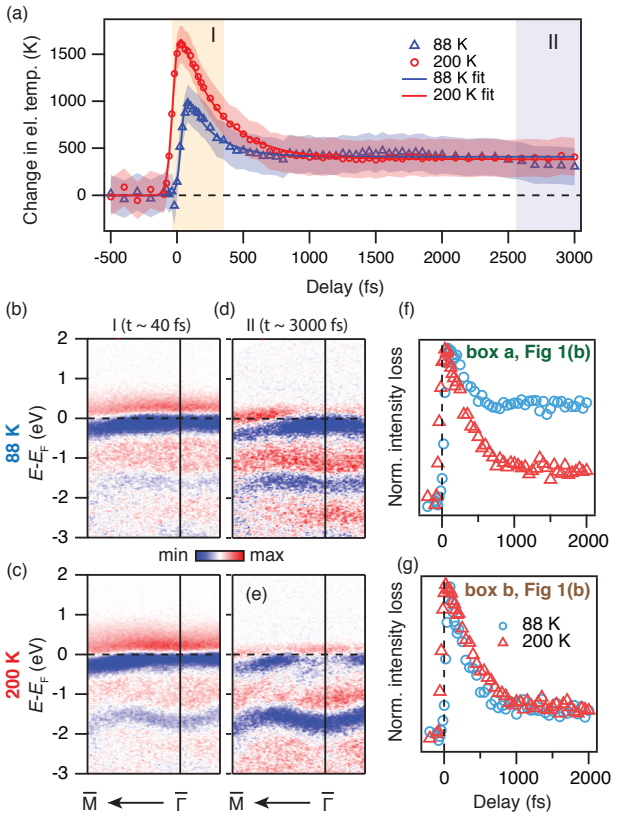

Figure 2(a) Change in electronic temperature for the sample at 88 K and 200 K with different time delay between pump and probe pulses. The shaded region shows the uncertainty in extracted temperatures. (b), (c) Difference spectra between 40 fs and onset at 88 K and 200 K sample temperature respectively. (d), (e) Corresponding difference spectra at 3 ps time delay. (f), (g) integrated intensity loss (normalized) in box 'a' and 'b' respectively (see figure 1(b)).

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References

1. Zhu, X. *et. al*. PNAS 112, 2367–2371 (2015).
2. Feng, J. *et al.* Nano Lett 18, 31 (2018).
3. G. Duvjir *et. al.* Nano Lett 18, 5432 (2018).
4. M. R. Norman *et al.*, Phys. Rev. B 57, R11093 (1998).