Observation of Trions in Monolayer WS_2 via Time-Resolved Terahertz Spectroscopy

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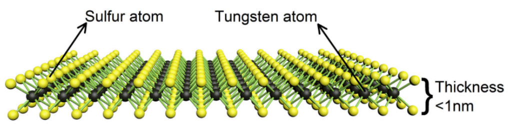
Introduction

Carrier Dynamics: How charges behave in a material under the influence of an electric field

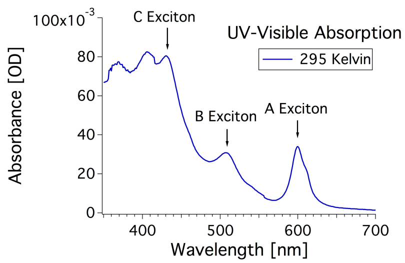
Monolayer: A single layer of molecules

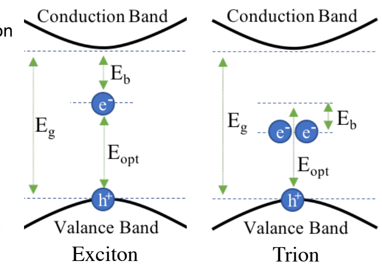


Device Applications: High-speed optoelectronics
Field-effect transistors
Photovoltaics

Properties of WS_2

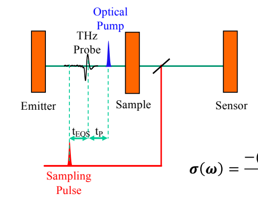
- Reduced dielectric screening results in the existence of tightly bound excitons at room temperature
- Strong Coulombic interactions support charged excitons (trions)



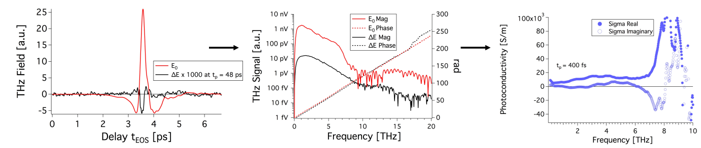


Time-Resolved Terahertz Spectroscopy (TRTS)

- TRTS probes the photo-generated excited state of a material with sub-picosecond resolution
- Yields complex frequency-dependent conductivity

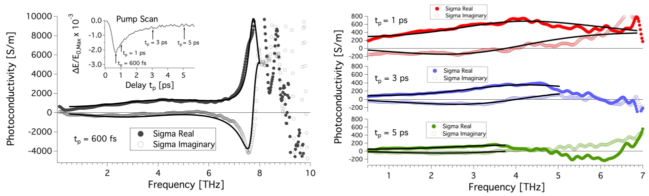


$$\sigma(\omega) = \frac{-(n+1)}{Zd}\frac{\Delta E(\omega)}{E_0(\omega)}$$



Time Evolution of Photoconductivity

- Conductivity was probed at pump delays t_p = 0.6, 1, 3, and 5 ps at 20 Kelvin with ~ 6 x 10^{14} photons/cm^2 of 584 nm



- Trions have been predicted[2,3] and observed[1,4,5,6,7] to have binding energies of about 25-40 meV (~ 6.0-9.7 THz) in WS_2
- The resonant feature in the conductivity at 7.75 THz (32meV) indicates the formation of trions in our sample
- We model the THz photoconductivity as a sum of three oscillators

$$\sigma(\omega) = \sum_{m=1}^{3} \frac{i\, C_m\, \omega}{\omega^2 - \omega_{0m}^2 + i\,\omega\,\gamma_m}$$

$C_m \equiv$ Spectral weight

$\gamma_m \equiv$ Linewidth (THz)

$\omega_{0m} \equiv$ Resonant frequecncy (THz)

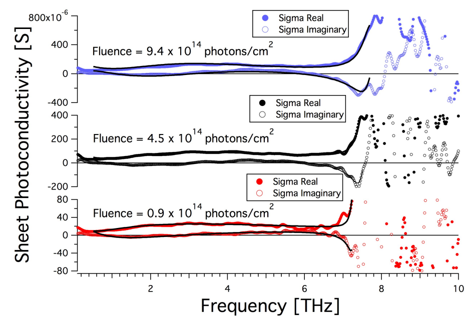
m = 1 → Drude Response m = 2 → Plasma Response m = 3 → Trion Response

Pump Delay	C_1	γ_1	ω_{01}	C_2	γ_2	ω_{02}	C_3	γ_3	ω_{03}
t_p = 600 fs	1.5e16	25.0	0	4.4e16	41	4.7	2.6e16	2.93	7.75
t_p = 1 ps	7.8e15	27.0	0	1.9e16	35	4.5	0	0	0
t_p = 3 ps	3.9e15	44.9	0	7.5e15	26	4.3	0	0	0
t_p = 5 ps	2.1e15	41.1	0	3.3e15	27	4.1	0	0	0

- ➢ There is no trion component for pump delays of t_p = 1, 3, and 5 ps
- ➢ As t_p increases, ω_{02} shifts to lower frequencies

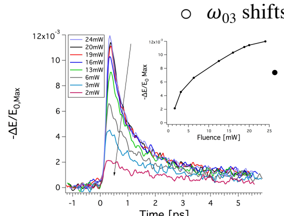
Fluence-Dependent Photoconductivity

- Conductivity was probed on resonance (A exciton) at 20 Kelvin (557 nm pump) at various fluences 400 fs after excitation



Fluence	C_1	γ_1	ω_{01}	C_2	γ_2	ω_{02}	C_3	γ_3	ω_{03}
9.4e14	6.0e8	3.8	0	4.3e9	32	3.2	2.3e9	3.55	7.75
4.5e14	2.6e8	2.6	0	3.7e9	42	3.3	1.0e9	2.31	7.56
0.9e14	6.5e7	1.0	0	1.3e9	53	3.2	9.9e7	1.02	7.29

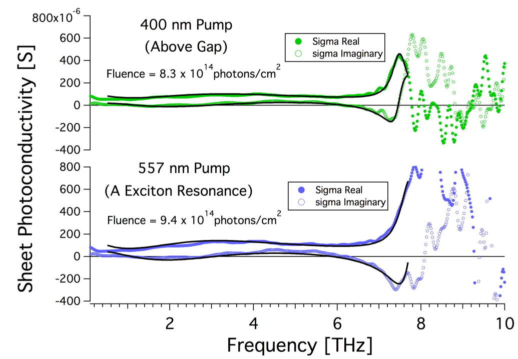
- ➢ Trion component observed at all fluences
- ➢ As t_p increases:
  - ○ No trend in ω_{02}
  - ○ ω_{03} shifts to lower frequencies



- As fluence increases, the change in the THz field saturates
- ➢ Free charge carriers are no longer being generated

Above and Below Resonant Excitation

- Conductivity was probed above the bandgap (400 nm pump) and on resonance (557 nm) at 20 Kelvin



Conclusions

- We attribute the Drude response to the promotion of trapped defect electrons to the conduction band
- We assign the source of the ω_{02} resonance to a plasmonic[8] response associated with particles with sizes similar to the THz wavelengths
- We assign the source of the ω_{03} resonance to the dissociation of trions into free electrons and excitons

References

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