Carrier Dynamics in Monolayer WS₂ 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 WS₂

• 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

TRTS Modes of Operation

• Pump scan
  • Yields time evolution of photo-carriers
  • ΔE/E₀ < 0 indicates a photo-induced increase in conductivity
  • Fast decay = 334 fs
  • Unbound electron/hole pairs rapidly forming excitons
  • Slow decay = 3.7 ps
  • Recombination and exciton-exciton annihilation

• Probe scan
  • Yields complex frequency-dependent conductivity
  • Full THz waveform recorded at various pump delays

Time Evolution of Photoconductivity

• Conductivity was probed at pump delays of tₚ = 1, 3, and 5 ps on resonance (584 nm) at 20 Kelvin with ~6 x 10¹⁴ photons/cm²

  - tₚ = 1 ps
  - tₚ = 3 ps
  - tₚ = 5 ps

- We model the THz photoconductivity as a sum of three oscillators

\[ \sigma(\omega) = \sum_{m=1}^{3} \frac{\omega m}{\omega_m + i \Gamma_m} \]

- There is no trion component for pump delays of tₚ = 1, 3, and 5 ps
- As tₚ increases:
  - The ratio C₁/C₂ increases
  - ω₁₋₂ shifts to lower frequencies

Conclusions

• The trend of C₁/C₂ suggests that the Drude response becomes more important relative to the plasma response at longer delay times
• We assign the source of the ω₁₋₂ resonance to a plasmonic response associated with particles with sizes similar to the THz wavelengths
• We see evidence of trion formation at early pump delays

References