Emission at the excitonic resonances in GaAs quantum wells: The Excitons *vs* plasma problem revisited

Richarj Mondal, Bipul Pal and Bhavtosh Bansal*

Indian Institute of Science Education and Research Kolkata, Mohanpur, Nadia 741252, WB, India

Summary

- **Study of density-dependent ionization-equilibrium using photoluminescence spectroscopy**
- PL intensity ratio between exciton and band-edge peak measures exciton ionization equilibrium density (following Kira and Koch)
- Coulomb-correlated-plasma contribution is not important at 4K but dominates PL at exciton energy after ~100K.
- Approach to Mott transition directly observed through excitation-correlation PL dynamics
- Exciton formation times compete with radiative lifetimes at power

Kira-Koch Luminescence Formula[1]

Free carrier contribution

Exciton contribution

$$I_{PL}(\omega) \propto Im\left[\sum_{\lambda} \frac{|\phi_\lambda(r=0)|^2\left(N_\lambda^{carrier} + N_\lambda^{ex}\right)}{E_\lambda - \hbar\omega - i\delta_\lambda}\right]$$

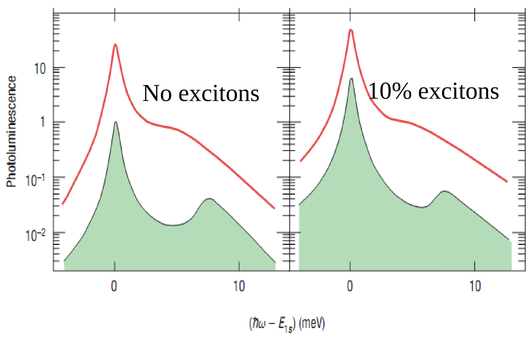
Elliott-like formula for luminescence[1]

- **Plasma can contribute to emission at exciton energy**

Q. How significant is this contribution?

Q. How do we estimate the true exciton population?

- Intensity ratio between the 1s exciton and band-edge emission can give a clue?
- Study of the Mott transition in this light

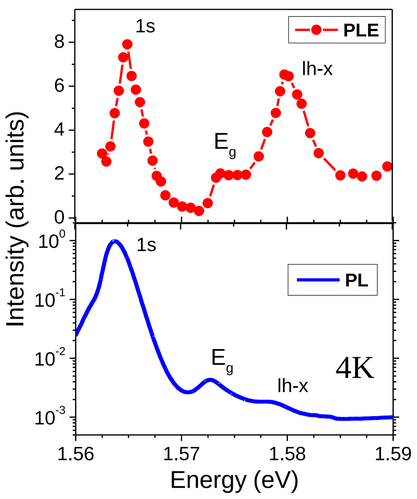


Calculations by Kira, Koch and Coworkers

Relevant references:

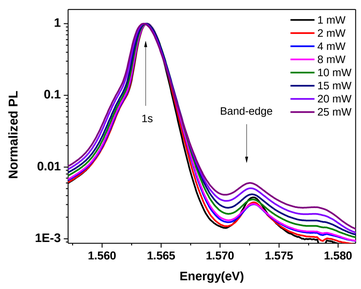
1. M. Kira and S. W. Koch Semiconductor Quantum Optics (Cambridge 2012)
2. S. Chatterjee, et al. Phys Rev Lett 92, 067402 (2004).
3. J. Szczytko, et al. Phys. Rev B 71, 195313 (2005).
4. Literature on excitonic Mott transition

Experimental

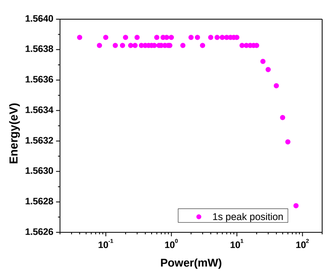


- **Special Sample:** Both 1s exciton and band-edge emission seen in PL => Can estimate relative populations from the PL spectra
- 8nm, multi-quantum well GaAs sample
- Experiments: PL, PLE, pump-probe reflectivity and dephasing dynamics
- Here we will only study the ratios of 1s to band-edge emission intensities. At low temperature, ratio >100

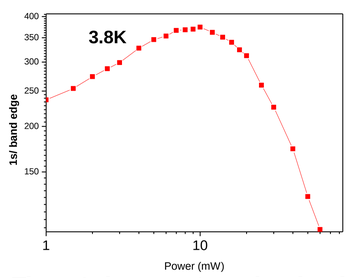
Excitation power-dependent PL spectra



Normalized PL spectra for different power



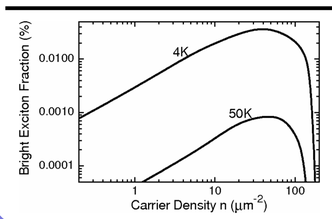
Red shift with power, heating?



The ratio between 1s peak to band-edge peak intensity as function of power.

Observations:

- Qualitatively PL spectra look similar as excitation power is increased but are quantitatively different.
- As the excitation power increased that ratio first increase up to a certain power. Why? Then it falls sharply. Why?
- Small red-shift with excitation power (Bandgap renormalization or heating?)



Calculated bright exciton fraction in Chatterjee, et al. PRL 92, 067402 (2004). Our *experimental* trend is similar. But we claim that the bright exciton fraction is orders of magnitude larger. Their analysis is flawed due to the use of KMS relation. Discussion on why **KMS relation should not be used to convert PL into absorption**: R. Bhattacharya, et al. Appl Phys Lett 100, 222103 (2012).

Simple analysis seems to work

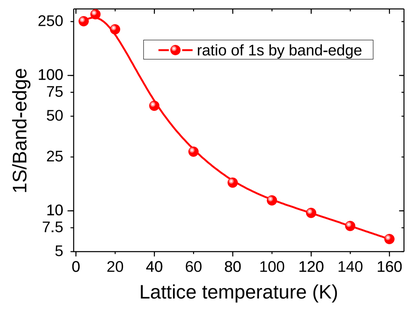
If 1s to band-edge peak PL emission intensity ratio is due to Coulomb-correlated plasma only

$$R = \frac{|\phi^{1s}(r=o)|^2 \int |\phi^{1s}(k)|^2 f_e(k) f_h(k) dk}{|\phi^{free}(r=o)|^2 f_e(k=0) f_h(k=0)} \leq 16$$

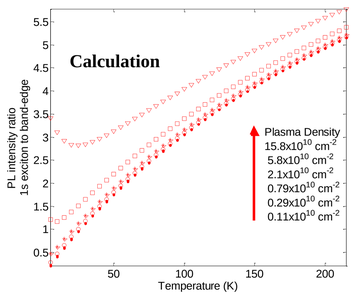
; $|\phi^{free}(r=o)|^2$: Sommerfeld factor

Qualitatively

- Kira-Koch analysis suggests that we definitely have real excitons if R > 16
- Experimentally $R > 100$ ⇒ emission at low temperature is mostly from excitons



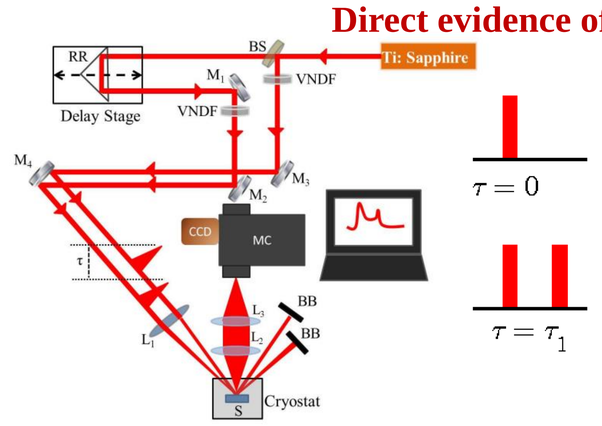
The ratio between the 1s to band-edge PL intensity as a function of temperature



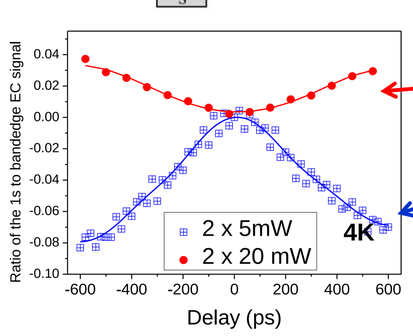
Theoretical plot of the ratio (1s/band-edge) as a function of temperature for different plasma densities

- Binding energy of 1s excition is about 9 meV ≈104 K => most excitions are ionized at 160 K
- At higher temperature, essentially only e-h plasma exists with no bound states
- Kira-Koch argument seems relevant at high temperature (why we see exciton peak at high temperature)
- Our calculated value of the ratio approaches ~ 5 same as experimental value at higher temperature

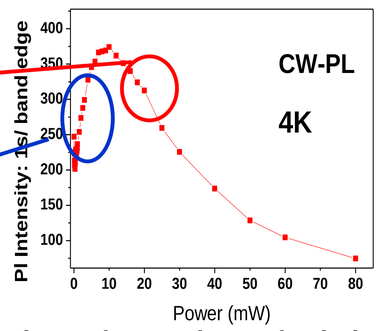
Picosecond excitation-correlation study of ionization ratio Direct evidence of Mott transition



- Picosecond excitation correlation PL spectroscopy is a two-beam time-resolved correlation technique Suitable for study of nonlinearity
- The number of photon incident on the sample is constant whatever the delay between the two pulses
- Spectrally resolved and time–integrated PL spectra is measured at given delay using CCD



This is the time evolution of the ratio between 1s to band-edge.



The ratio between the 1s to band-edge as a function of power

Results

- EC signal ratio at low power increases as the delay approaches zero => exciton population increases with carrier density. **Reverse of Mott transition:** Competition between exciton formation and recombination times
- EC signal ratio reverses at higher power=> **Mott transition**
- Power-dependent heating is not important for this measurement
- Exciton formation times are within about an order of magnitude of recombination times (detailed analysis in progress)

Laboratory at IISER Kolkata, India