Luminescence of excitons
&
Highly excited semiconductors

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Chapter 7 & Chapter 8

Classification of luminescence by intensity

Low-fluence (Weak) excitation (0.01- 10 W/cm^2): gas-discharge lamp, incandescent lamp, continuous-wave gas laser

→ - Recombination of **free excitons** (FE or X; FE-LO phonon)
→ - Recombination of a **bound exciton** (BE): (D^0-X; A^0-X; D^+-X; A^-X, isoelectronic impurity)
✓ - Recombination of **donor-acceptor pairs** (D^0-A^0)
✓ - Recombination of **free hole/electron** with **neutral donor/acceptor** (h-D^0; e-A^0; e-h)

1 sun = 0.1 mW/cm^2 = 0.1 W/cm^2 = 1 kW/m^2

High-fluence (Strong) excitation (1 kW-10 MW/cm^2):

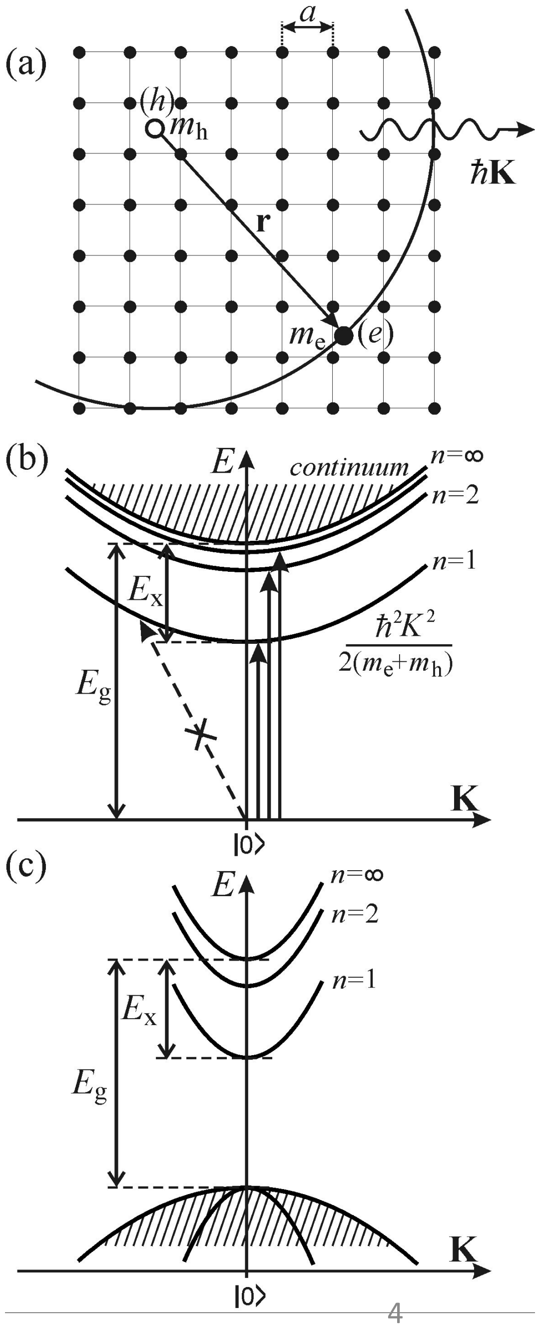
→ - Radiative decay of **excitonic molecule** (EM or XX)
✗ - Luminescence from inelastic collisions of excitons (X-X collisions)
✗ - Luminescence of electron-hole liquid (EHL) or electron-hole plasma (EHP)
✗ - Bose-Einstein condensation of excitons or excitonic molecules

These excitations can occur simultaneously, in these two chapters we focus on low-fluence excitation and neglected luminescence from excitons

Excitons

- Electron and hole not independent, treated as one quasi particle
  - Energy < E_g because of Coulomb interaction
- Three types
  1. Frenkel exciton
     - Distance about one unit cell
     - Molecular crystals
  2. Charge transfer exciton
     - A bit larger radius
     - Ionic crystals
  3. **Wannier exciton**
     - Separation over many lattice constants
     - Moves freely, delocalized wavefunction (if not bound, we will discuss later)
     - Free excitons
- **Bound excitons**

Wannier exciton



- Radius estimeted to be a_x =100*a_b = 5nm
- Binding energy estimated to be 13meV
  - Low T needed
- Proper treatment if wavefunction → radius and binding energy
- Exitons with kinetic energy
  - Bound exitons have less kinetic energy, localized
- Dispersion relation E(**k**)
  - Here we do not have a free electron approximation

$$E_{(n)}(\mathbf{K}) = E_g - E_{X(n)} + E_{\text{kin}} = E_g - \frac{(m_r/m_0)}{\varepsilon^2}\frac{Ry(\text{H})}{n^2} + \frac{\hbar^2K^2}{2(m_e+m_h)}. \tag{7.5}$$

Wannier exciton – Absorption spectrum

Direct bandgap

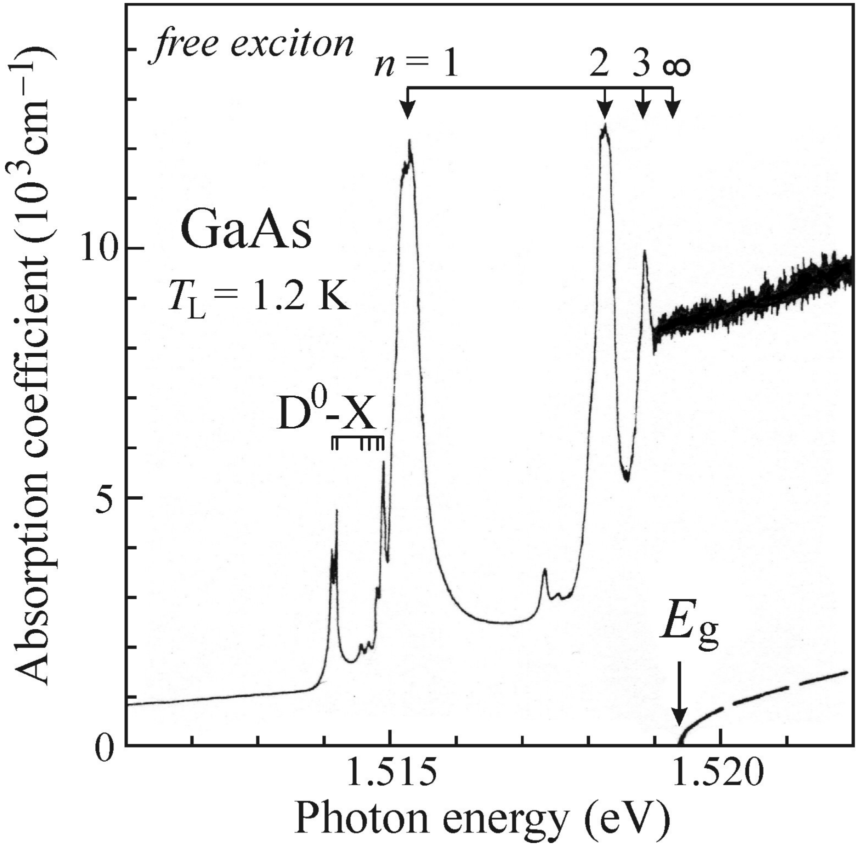
- Dipole approximation

$$\alpha(h\nu) \approx (h\nu - E_g)^{1/2}, \quad h\nu \geq E_g$$

- T = 0K, no kinteic energy → only vertical transitions (**k**)
- As in hydrogen atom, absorption line series

$$h\nu = E_g - \frac{E_X}{n^2}, \quad n = 1, 2, 3, \ldots \infty;$$

- Intensity is proportional to $1/n^3$
- Also, enhanced absorption above Eg



Wannier exciton – Absorption Spectrum

Indirect bandgap

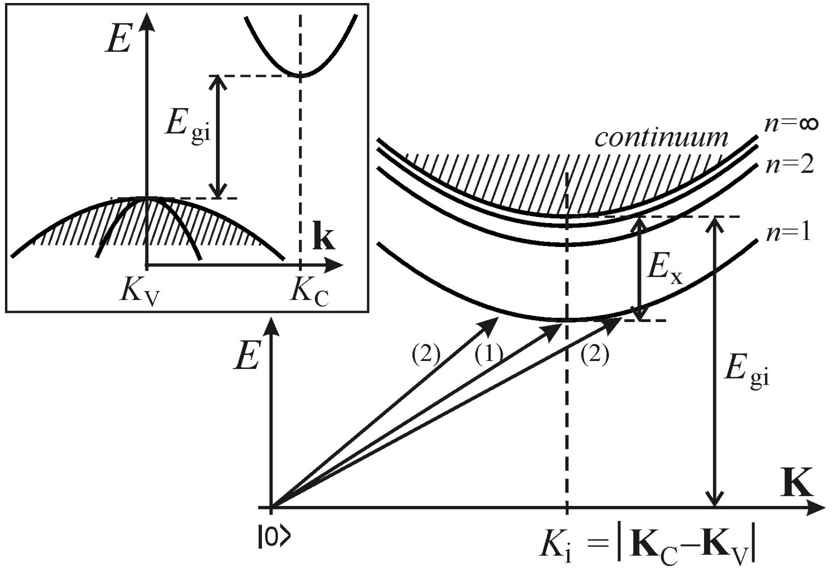
- Group veolcity of e and h equal

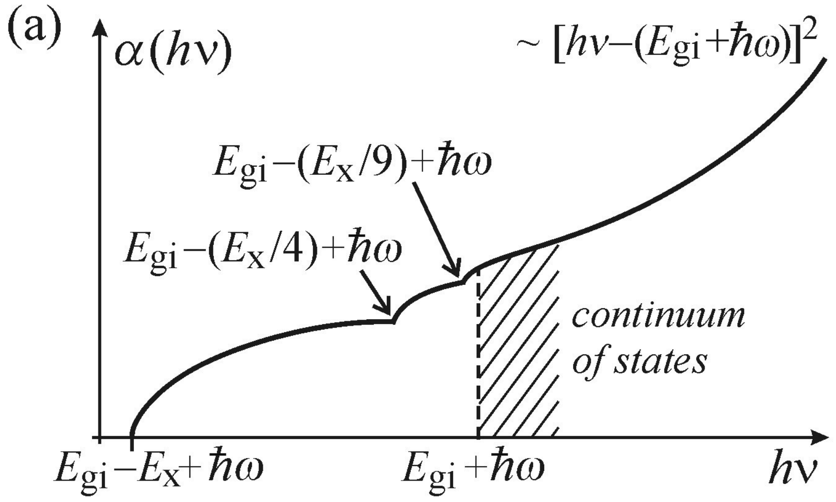
$$\mathbf{V}_\mathrm{g} = \frac{1}{\hbar}\frac{\partial E}{\partial \mathbf{k}}$$

- Minimum energy if $K_\mathrm{i} = |\mathbf{K}_\mathrm{c} - \mathbf{K}_\mathrm{v}|.^2$
- Onset of absorption $h\nu = E_\mathrm{gi} - E_\mathrm{X} + \hbar\omega$
- Transition (1) and (2) equally probable → density of states important

$$[E - (E_\mathrm{gi} - E_{\mathrm{X}(n)})]^{1/2}$$

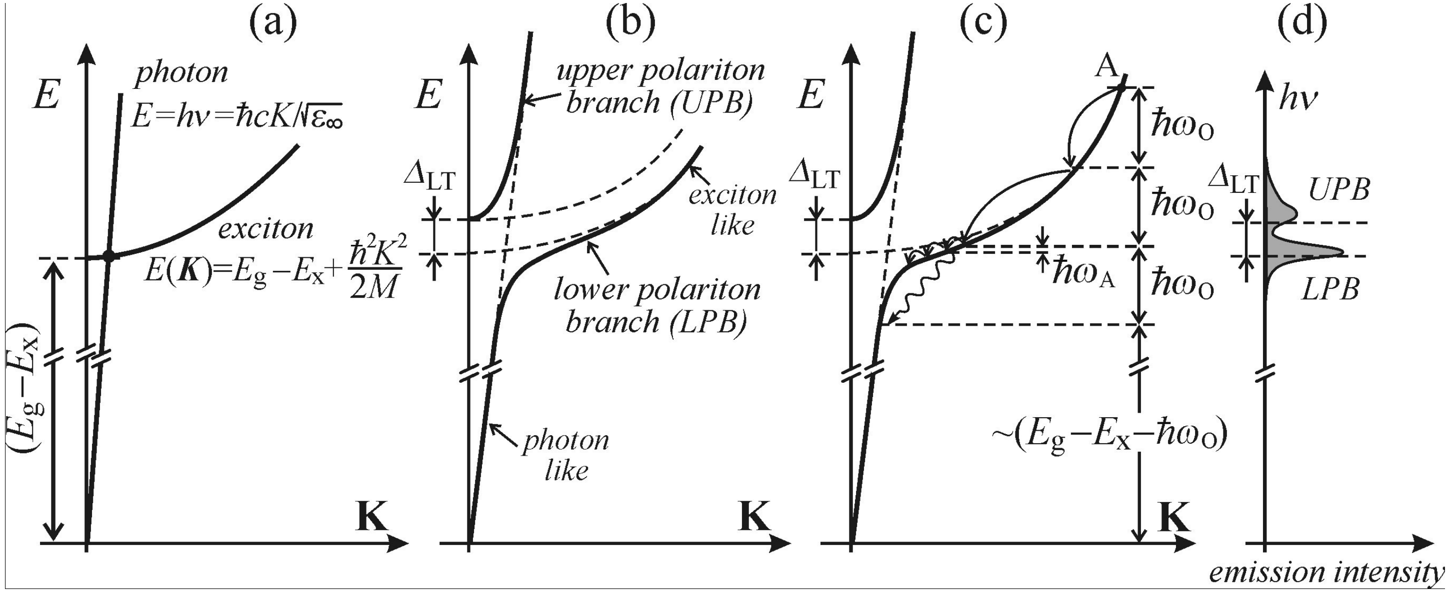
- Note: excitons with non-zero kinetic energy possible!

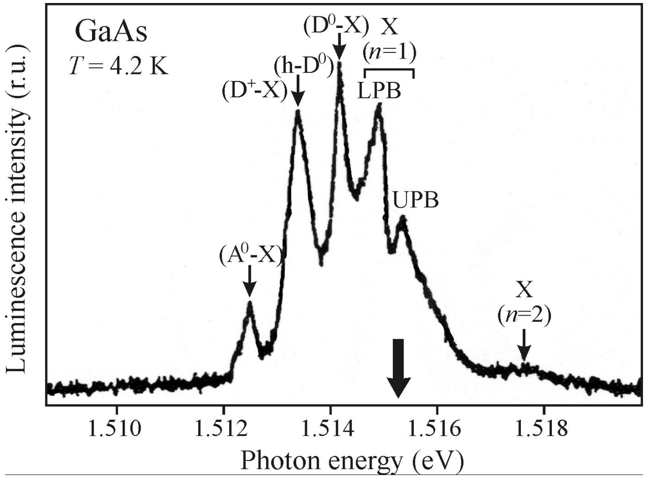




Resonant luminesence : direct E_g : free exciton-polaritons

- Absorption != emission
- Strong reabsorption
- Photon-Exciton dispersion curves combined
- Degeneracy lifted : band splitting
- No simple equation





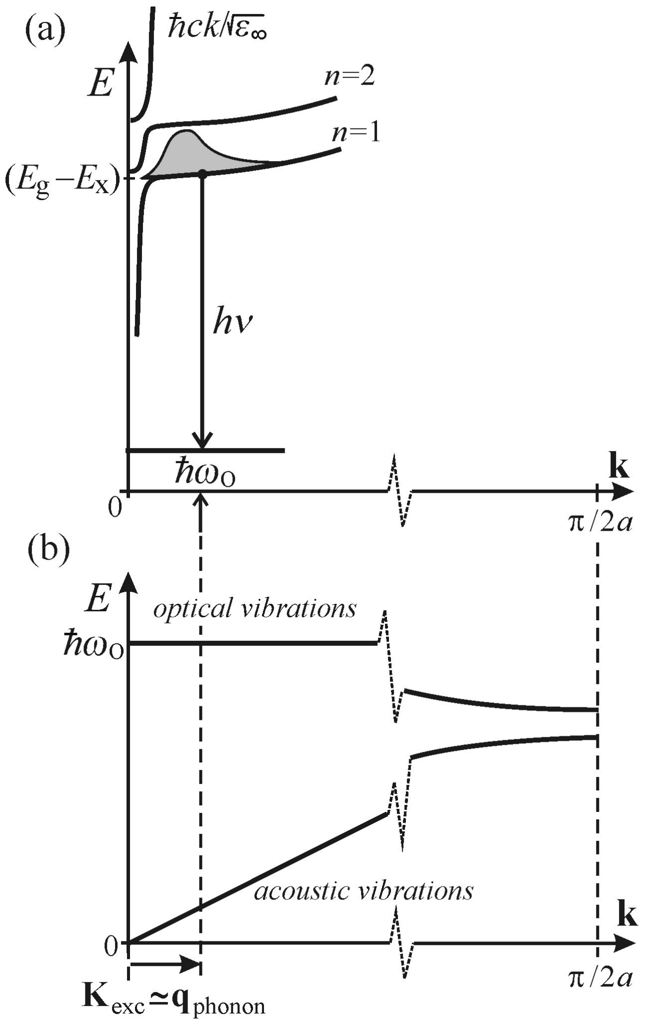
Luminesence : direct E_g : free excitons and OP

$$h\nu_{\mathrm{X}-m\,\mathrm{LO}} \approx (E_{\mathrm{g}} - E_{\mathrm{X}}) - m\,\hbar\omega_0, \quad m = 1, 2, 3, \ldots$$

- Very strong lines, why?
  - Energy of photon < Eg
  - All excitons regardless of **k** can participate
- Lineshape Maxwell-Boltzmann + varriaiton
  - Probability of phonon creation

$$I_{\mathrm{sp}}^{(m)}(h\nu) \approx (h\nu - [(E_{\mathrm{g}} - E_{\mathrm{X}}) - m\hbar\omega_0])^{1/2} \times \exp\left[-\frac{h\nu - [(E_{\mathrm{g}} - E_{\mathrm{X}}) - m\hbar\omega_0]}{k_{\mathrm{B}}T}\right]$$

$$W^{(m)}(m\mathbf{q}_{\mathrm{phonon}} \approx K_{\mathrm{exc}}),$$



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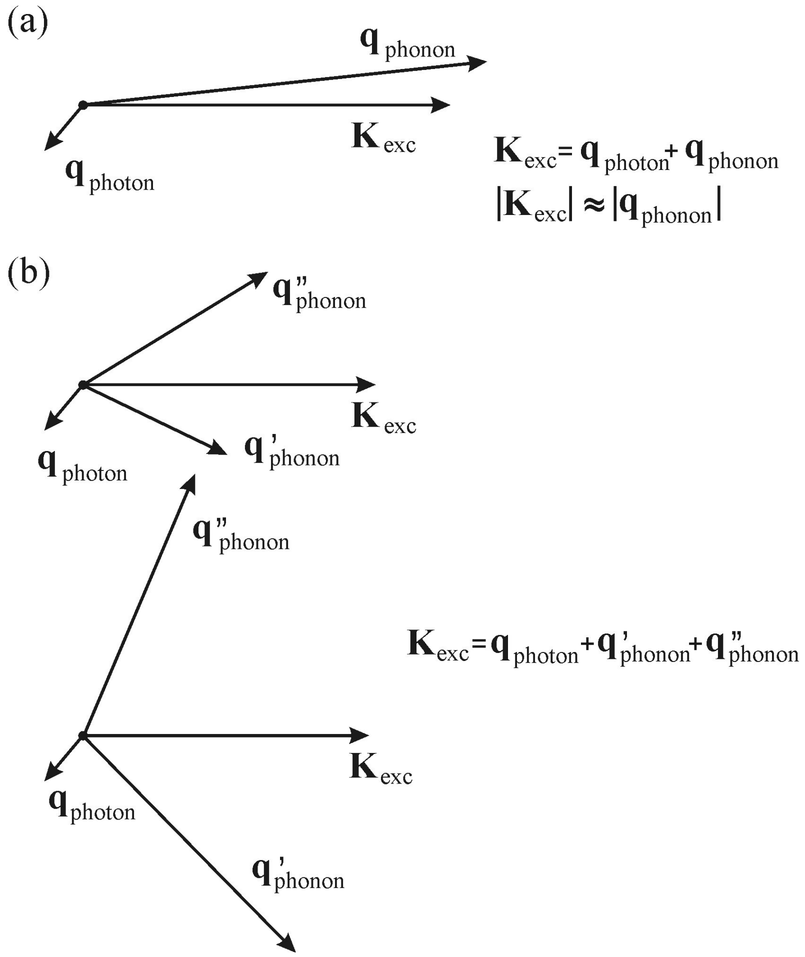
$$\times\exp\left[-\frac{h\nu - [(E_g - E_X) - m\hbar\omega_0]}{k_B T}\right]$$

$$W^{(m)}(m\mathbf{q}_{phonon} \approx K_{exc}),$$

- **m = 1** → phonons with ƛ similar to linear size of exciton are more probable $W^{(1)}(\mathbf{q}_{phonon} \approx \mathbf{K}_{exc}) \sim K_{exc}^2$

$$I_{sp}^{(1)}(h\nu) \approx (h\nu - [(E_g - E_X) - \hbar\omega_0])^{3/2}$$

$$\exp\left[-\frac{h\nu - [(E_g - E_X) - \hbar\omega_0]}{k_B T}\right]$$



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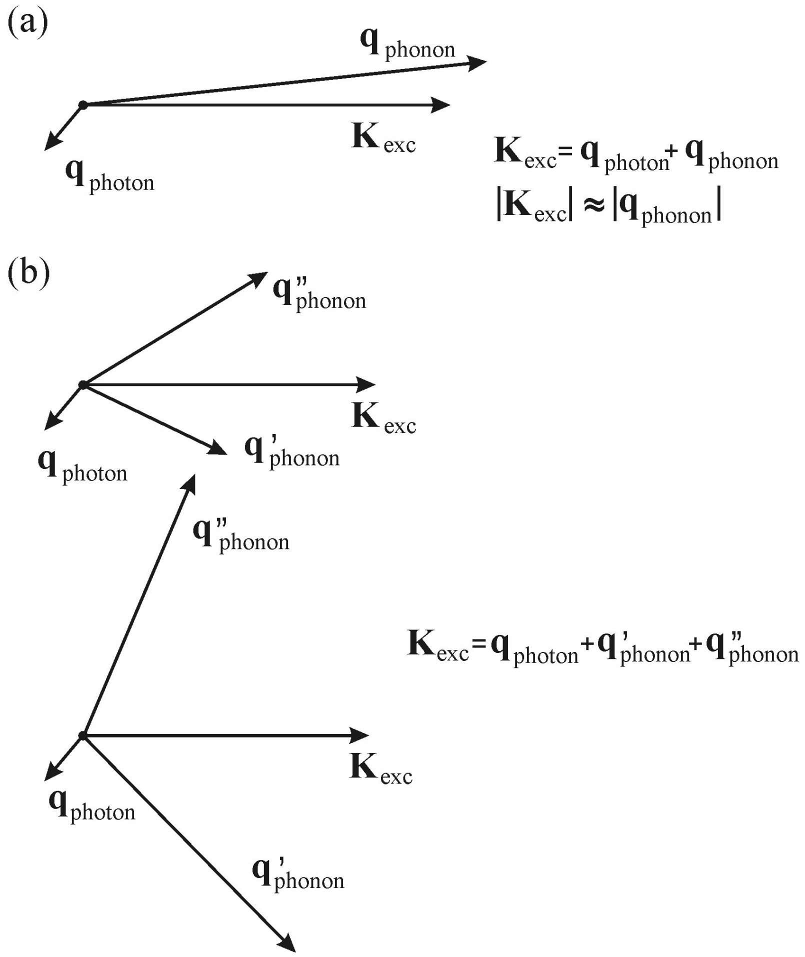
$$W^{(m)}(m\mathbf{q}_{\mathrm{phonon}} \approx K_{\mathrm{exc}}),$$

- **m = 2 →** many possibilities, Maxwell like

$$I_{\mathrm{sp}}^{(2)}(h\nu) \approx (h\nu - [(E_{\mathrm{g}} - E_{\mathrm{X}}) - 2\hbar\omega_0])^{1/2} \exp\left[-\frac{h\nu - [(E_{\mathrm{g}} - E_{\mathrm{X}}) - 2\hbar\omega_0]}{k_{\mathrm{B}}T}\right]$$



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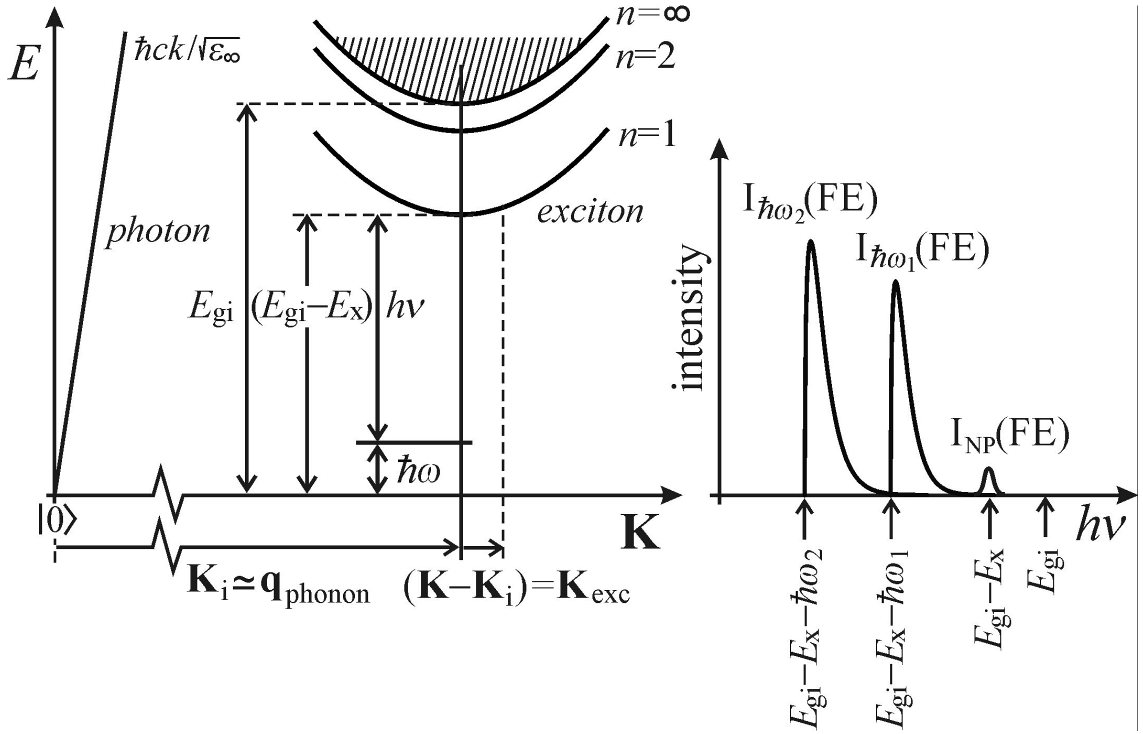
$$h\nu_{\mathrm{X}-m\,\mathrm{LO}} \approx (E_{\mathrm{g}} - E_{\mathrm{X}}) - m\,\hbar\omega_0, \quad m = 1, 2, 3, \ldots$$

- Free exciton recombination is a **linear** function of excitation intensity
- Aucustic phonons have too small momentum
- Has to be optical ones – are they allowed?
  - Symmetry argument : YES

Luminesence : indirect E_g : free excitons

- Exciton and phonon dispersion curves don’t cross – no polaritons
- Phonons assist: whole populaiton can recombine
- Probability doesn’t depend on phonon momnetum → Maxwell-Boltzmann



$$I^{(1)}_{\mathrm{sp(i)}}(h\nu) \cong (h\nu - [(E_{\mathrm{gi}} - E_{\mathrm{X}}) - \hbar\omega])^{1/2}\exp - \frac{h\nu - [(E_{\mathrm{gi}} - E_{\mathrm{X}}) - \hbar\omega]}{k_{\mathrm{B}}T}$$

Luminesence : indirect E_g : free excitons

- Which phonon will participate?
  - Selection rules based on symmetry
  - Si: LO, TO, LA, TA but LA and LO degenrate at X
- Impurities in crystals make transitions without phonon possible
- Linear depencence on pump intensity

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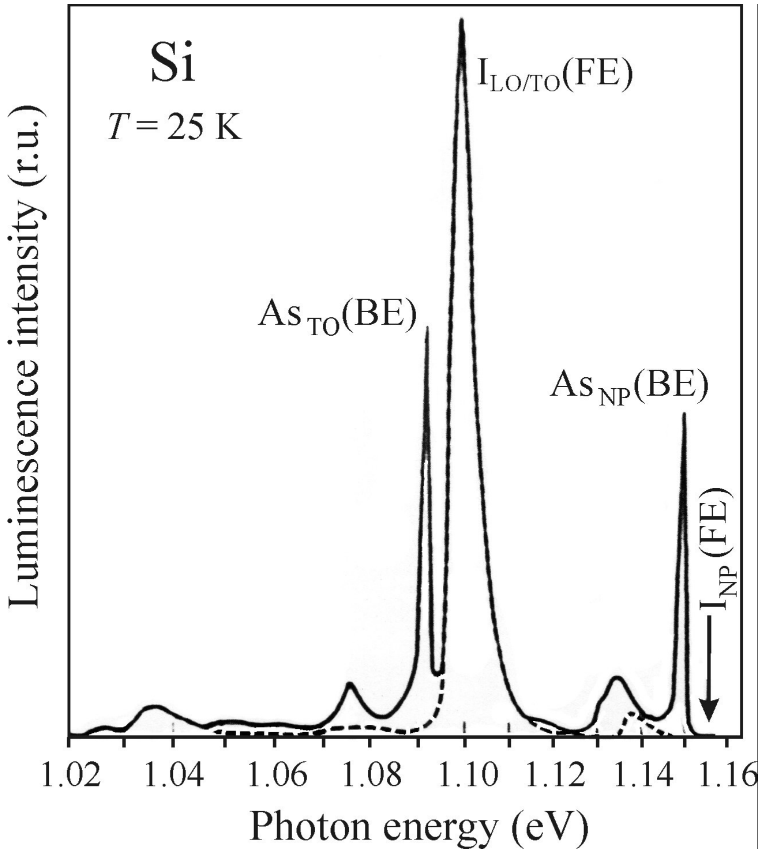
Bound Excitons

- Localized, looses kinetic energy
- Extrinsic
- Dominates free excition
  - Large radius: likely to encounter a trap
  - Giant strength from all unit cells within the volume
- PL can give us defects!
- No kinetic energy --> no broadeing with T
- Linear dependency on intensity until saturation
- Lower hv compared to free excition, no kinetic energy



Bound Excitions : shallow impurities

e	electron,
h	hole,
$\oplus e$	neutral donor; also D^0,
$\ominus h$	neutral acceptor; also A^0,
FE	free exciton,
$\oplus$	ionized donor; also D^+,
$\ominus$	ionized acceptor; also A^-,
$\oplus\, eh$	exciton bound to ionized donor; also $(D^+–X)$ or (D^+, X),
$\ominus\, eh$	exciton bound to ionized acceptor; also $(A^-–X)$ or (A^-, X),
$\oplus\, eeh$	exciton bound to neutral donor; also $(D^0–X)$ or (D^0, X),
$\oplus\, heh$	exciton bound to neutral acceptor; also $(A^0–X)$ or (A^0, X).

$$\left.\begin{array}{l} \oplus + \mathrm{FE} \rightarrow \oplus\, eh + D_1 \\ \oplus\, eh \rightarrow \oplus + h\nu_{\mathrm{BE}}. \end{array}\right\} \qquad h\nu_{\mathrm{BE}} = \mathrm{FE} - D_1 \cong (E_g - E_X) - D_1$$

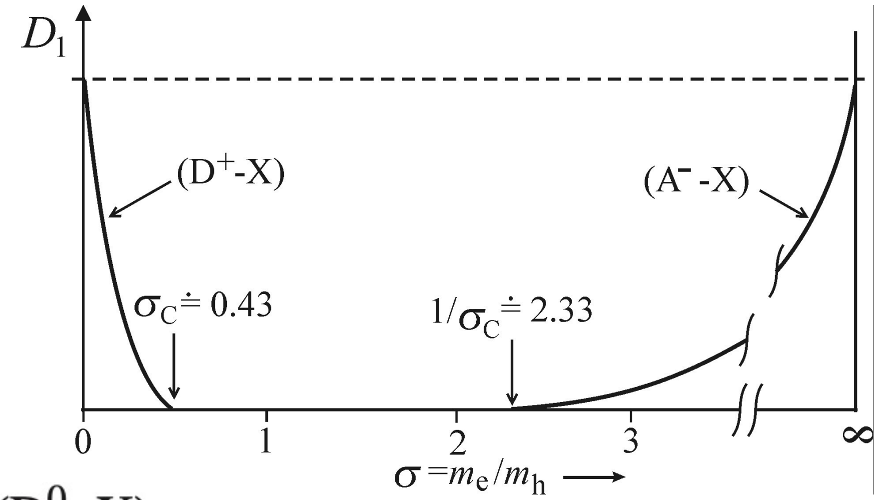
Bound Excitions : shallow impurities

D1

- Ionized

$\oplus\, eh \ldots\ldots\ldots\ldots\ldots$ exciton bound to ionized donor; also $(\mathrm{D}^+\text{–}\mathrm{X})$ or $(\mathrm{D}^+, \mathrm{X})$,

$\ominus\, eh \ldots\ldots\ldots\ldots\ldots$ exciton bound to ionized acceptor; also $(\mathrm{A}^-\text{–}\mathrm{X})$ or $(\mathrm{A}^-, \mathrm{X})$,



  - D1 hard to compute theoretically
    - Effective mass
      - Light hole in +eh will break away from the neutral +e
      - Heavy = small kinetic energy
    - A—X unlikely holes are usually heavier

E_{BX}

- Neutral : stable for any effective mass ratio

$\oplus\, eeh \ldots\ldots\ldots\ldots\ldots$ exciton bound to neutral donor; also $(\mathrm{D}^0\text{–}\mathrm{X})$ or $(\mathrm{D}^0, \mathrm{X})$,

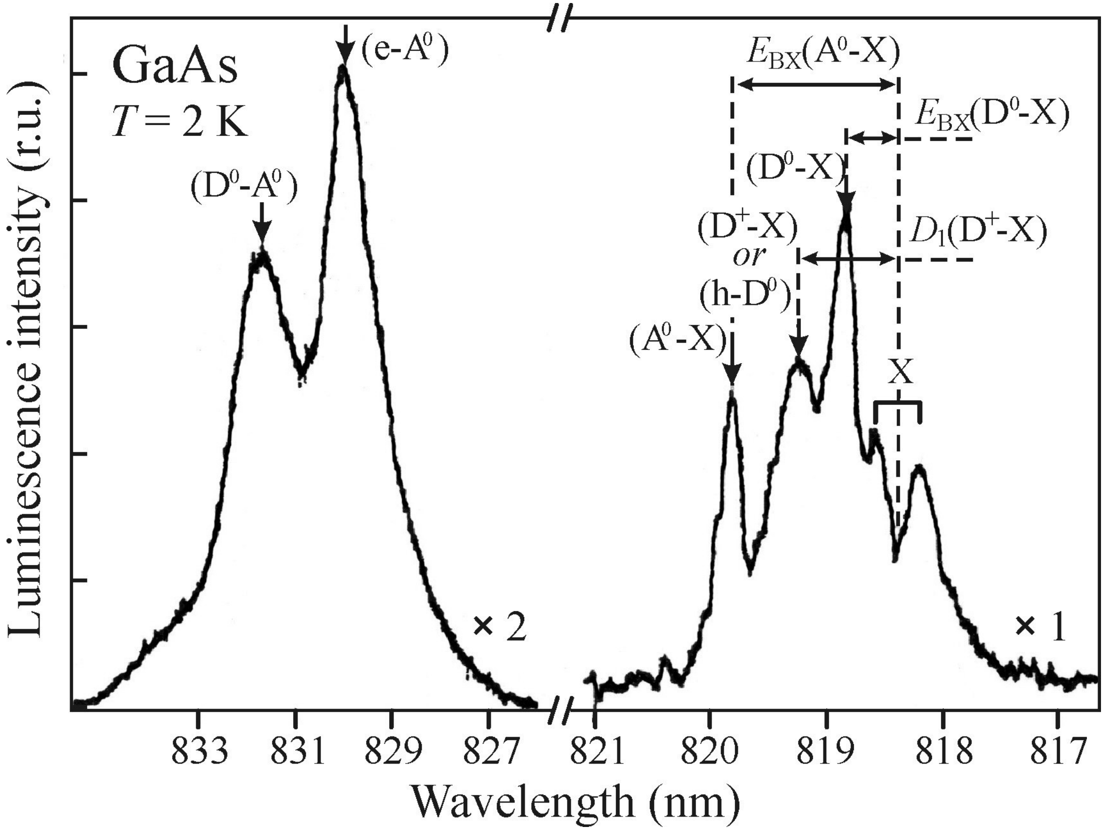
$\oplus\, heh \ldots\ldots\ldots\ldots\ldots$ exciton bound to neutral acceptor; also $(\mathrm{A}^0\text{–}\mathrm{X})$ or $(\mathrm{A}^0, \mathrm{X})$.

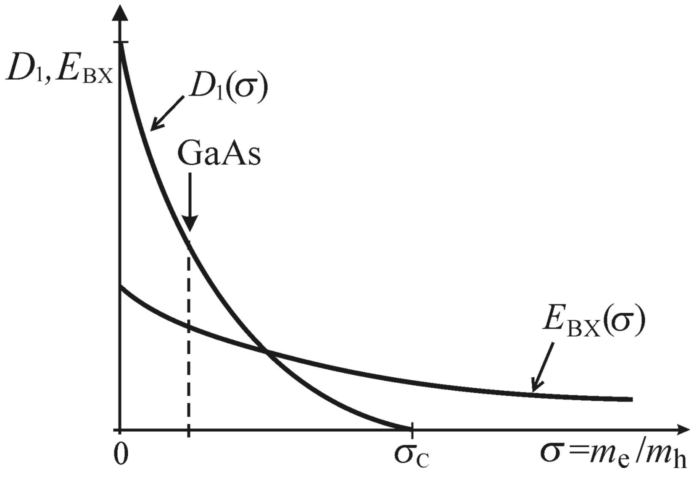
$$h\nu_{\mathrm{BE}} = \mathrm{FE} - E_{\mathrm{BX}} = (E_{\mathrm{g}} - E_{\mathrm{X}}) - E_{\mathrm{BX}}$$ Direct

$$h\nu_{\mathrm{BE}} = \mathrm{FE} - E_{\mathrm{BX}} = (E_{\mathrm{gi}} - E_{\mathrm{X}} - \hbar\omega) - E_{\mathrm{BX}}$$ Indirect

Bound Excitions : shallow impurities

- Here: D1 > E_{BX}
  - D1(D^+-X) emission more redshifted than E_{BX}(D^0-X)
- This is material dependent

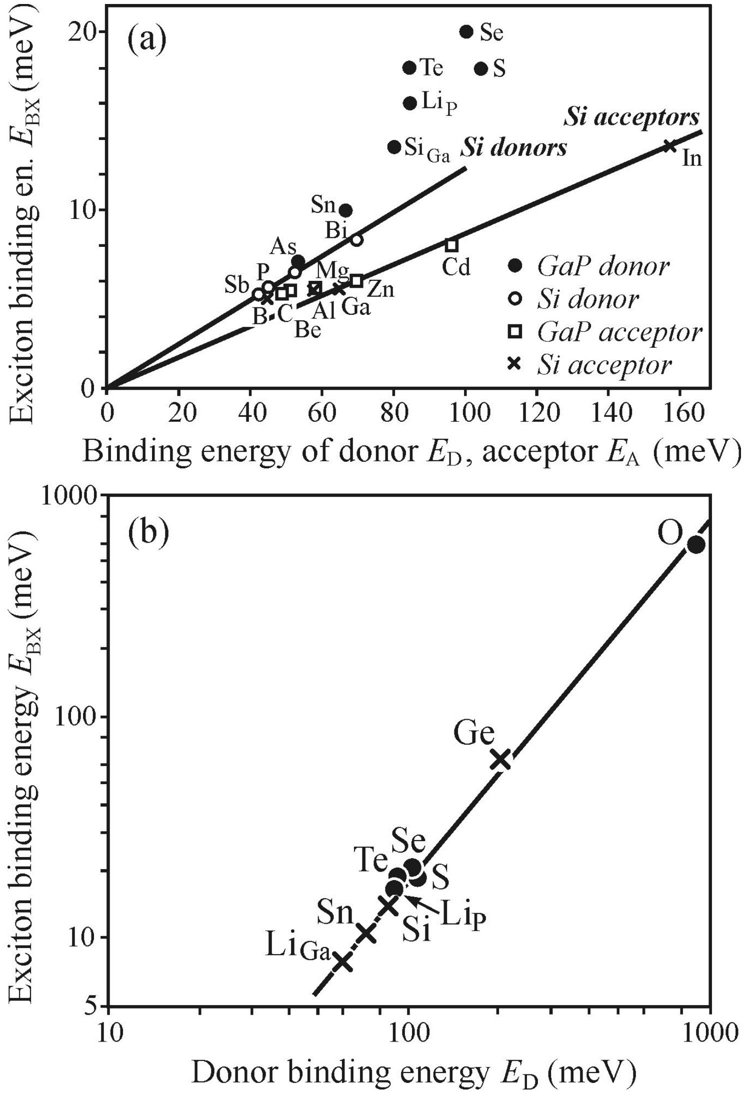




Bound Excitons : shallow impurities

- Hayne’s rule
  - Not only effective masses, also atom itself!
  - Si: a = 0, b = 0.1

$$E_{\mathrm{BX}} = a + b\, E_{\mathrm{D}}$$



Bound multiexciton complexes

- Shift of emission line by sum of all binding energies

$$\oplus e + \mathrm{FE} \rightarrow \oplus e(eh) + E_{\mathrm{BX}}^{(1)}$$

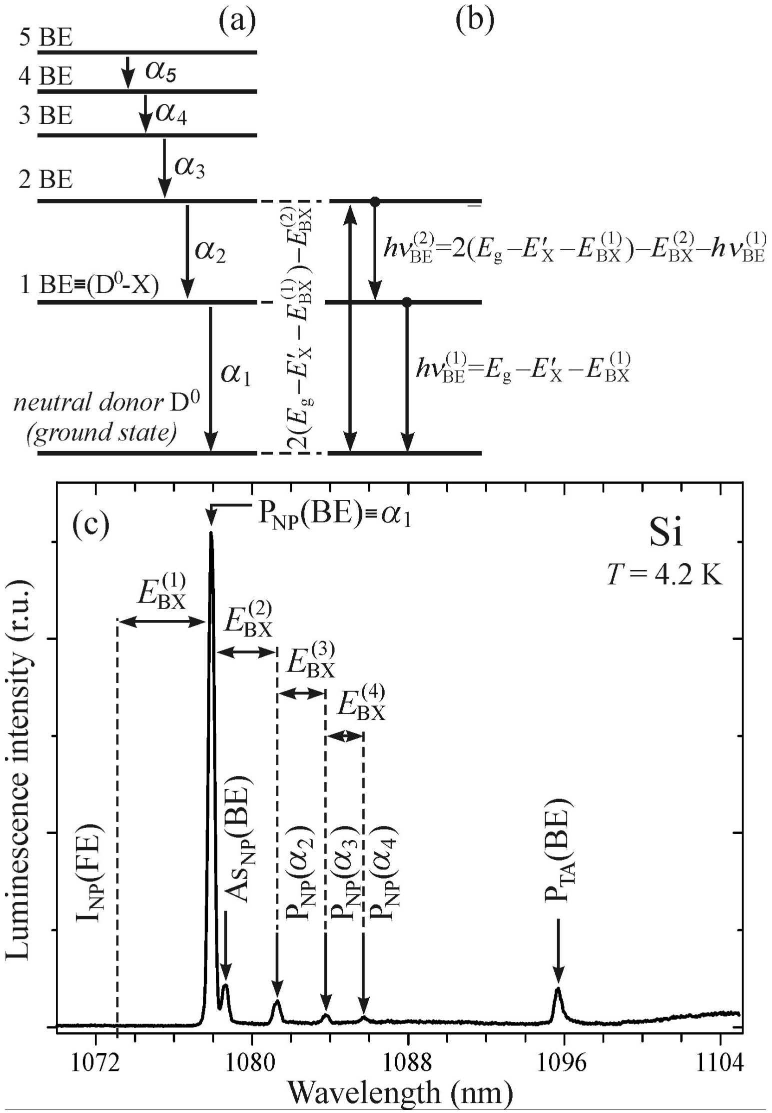
$$\oplus e(eh) - E_{\mathrm{BX}}^{(1)} + \mathrm{FE} \rightarrow \oplus e(2eh) + E_{\mathrm{BX}}^{(2)}$$

$$\oplus e(2eh) - \left(E_{\mathrm{BX}}^{(1)} + E_{\mathrm{BX}}^{(2)}\right) + \mathrm{FE} \rightarrow \oplus e(3eh) + E_{\mathrm{BX}}^{(3)}$$

$$\vdots$$

$$\oplus e\left((m-1)\ eh\right) - \sum_{j=1}^{(m-1)} E_{\mathrm{BX}}^{(j)} + \mathrm{FE} \rightarrow \oplus e(m\,eh) + E_{\mathrm{BX}}^{(m)}. \quad (7.24\mathrm{a})$$

- Intensity decreases for higher m
- More probable in indirect because longer lifetime



Quantitative analysis of shallow imputities in Si

- BE/FE
- Challenges:
  - Detector range
  - High S/N
  - High resolution
  - Need low excitation

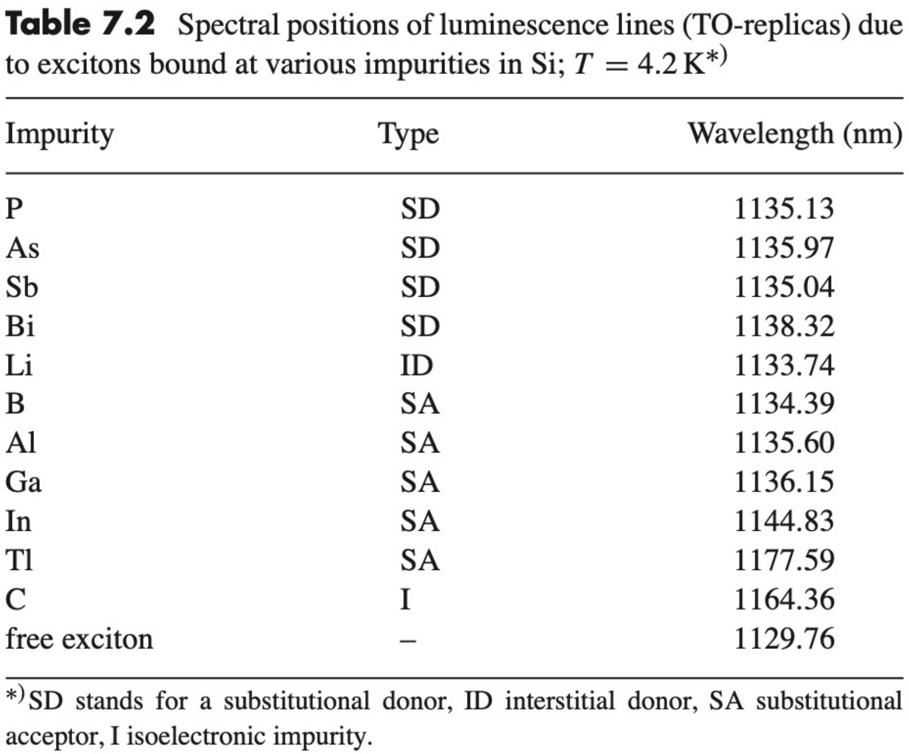
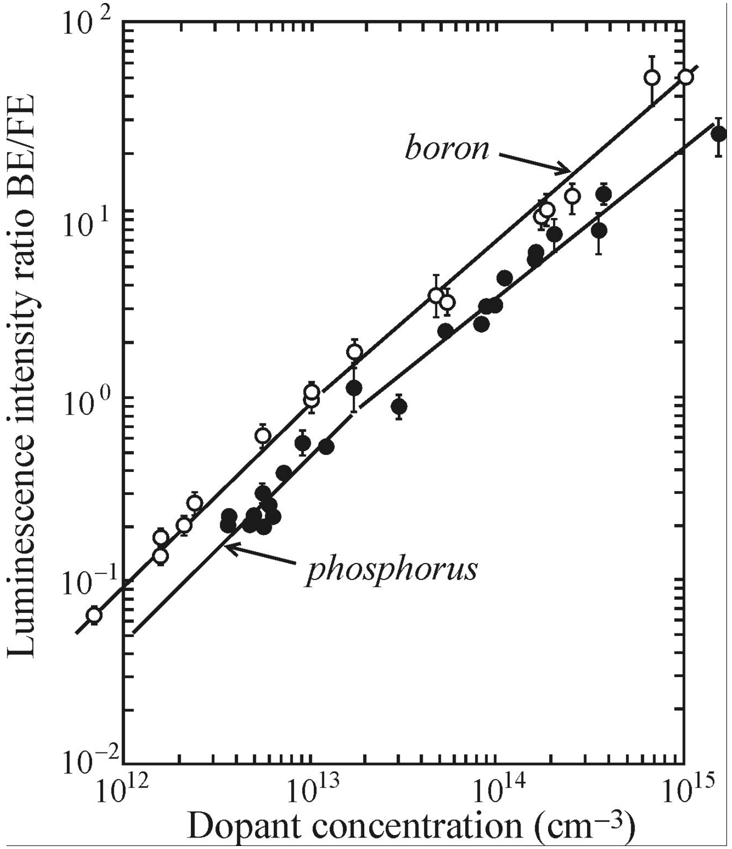


Table 7.2 Spectral positions of luminescence lines (TO-replicas) due to excitons bound at various impurities in Si; $T = 4.2\,\mathrm{K}^{*)}$

Impurity	Type	Wavelength (nm)
P	SD	1135.13
As	SD	1135.97
Sb	SD	1135.04
Bi	SD	1138.32
Li	ID	1133.74
B	SA	1134.39
Al	SA	1135.60
Ga	SA	1136.15
In	SA	1144.83
Tl	SA	1177.59
C	I	1164.36
free exciton	–	1129.76

*) SD stands for a substitutional donor, ID interstitial donor, SA substitutional acceptor, I isoelectronic impurity.



Bound Excitons : isoelectric impurities

- Impurities from the same group
  - Same number of valence electrons
  - N for P in GaP & I for Br in AgBr
- Electronegativity
- Lineshape
  - Short range forces : no broadening
  - Exciton-phonon interaction
    - Weak --> narrow line (small reaction from surrounding lattice)
    - Strong --> broader line (larger ineraction with surroinding lattice)
- Effective enhancement!

Iodide impurity emission

$$z_{\mathrm{ef}} = \frac{\mathrm{Iod}_{\mathrm{LO}}\,(\mathrm{BE})}{\mathrm{I}_{\mathrm{TO}}\,(\mathrm{FE})}\;\frac{\tau_{\mathrm{Iod}}}{\tau_{\mathrm{FE}}} \approx 10^4 \times \left(10^3/3\right) \cong \boxed{3 \times 10^6 (!)}$$

Free exciton emission

Bound Excitons : isoelectric impurities

- Why such effective enhancement?
  - Trapping
  - No Auger recombination
  - Strong localization in x -> strong delocalization in k
    - Δx Δk ≤ 1/2
    - Can stretch all the way to the direct Eg!

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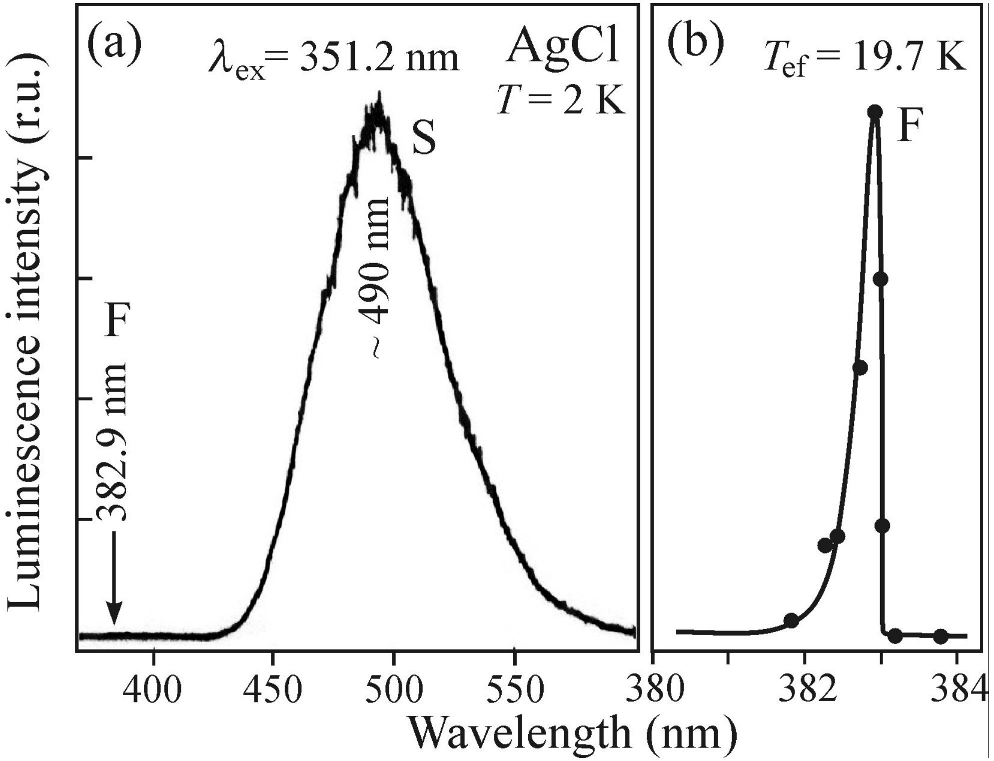
Self trapped excitons

- Strong exciton phonon interaction
- A moving exciton polarize its surroundings (hole is the heavier, drives)
  - → moves slower → more polarization → slows down even more → more polarization → stands still = localized
  - Local energy minima



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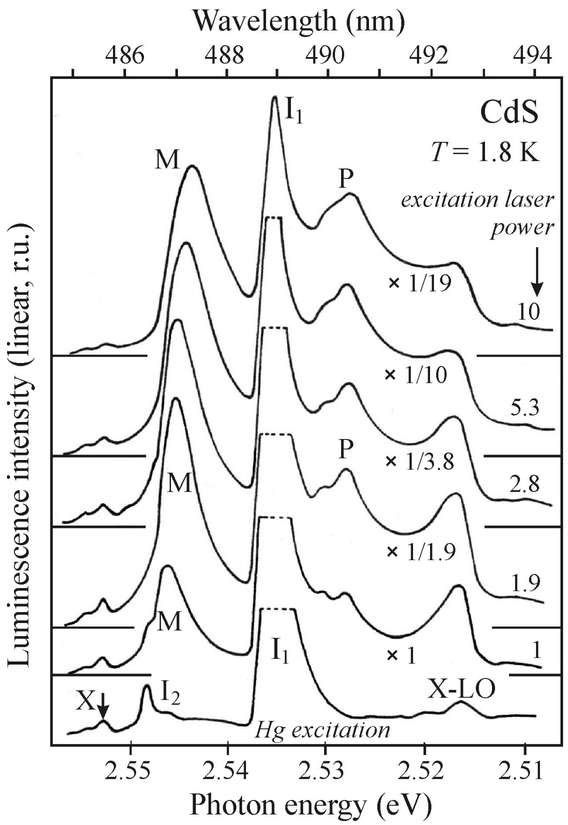
Highly Excited Semiconductors



- Until now only 0.01 – 10W/cm^2
- Now: 1k-1MW/cm^2
  - Pulsed laser
- New emission lines:
  - Radiative decay of excitonic molecules (=biexcitons) **M**
  - *Collissions between excitons* ***P***
  - *Luminescence of electron hole liquid or electron hole plasma*
  - *Bose Enistein condensation of excitons or biexcitons*

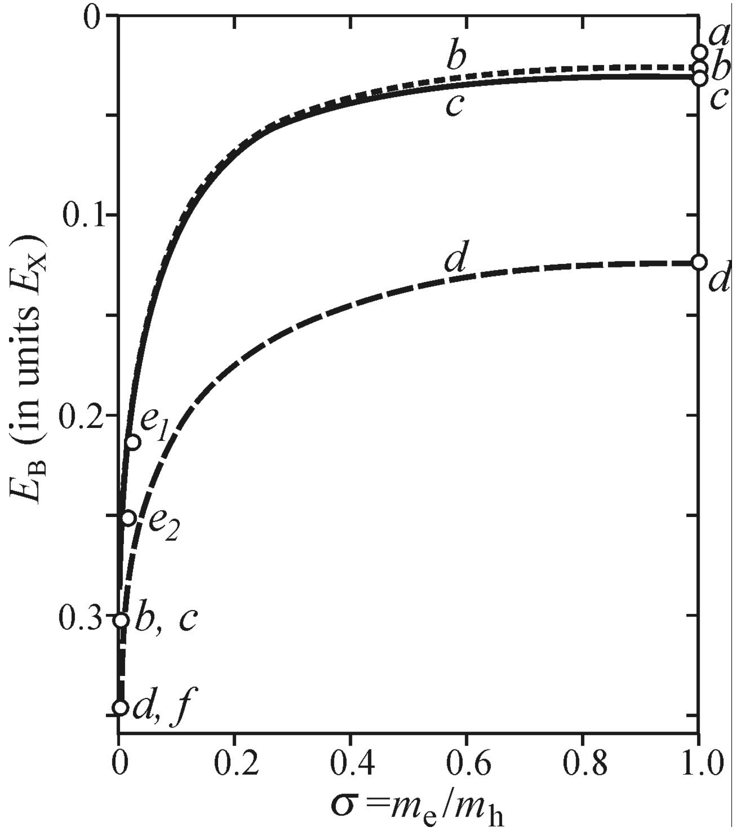
Not today ☹

Highly Excited Semiconductors

- When power is increased the concentration of free excitons increase
- When the mean separation is about two times the separation between electron and hole → interaction
- Reality: density not homogenous but rapid decrease from surface
    - Two photon excitation
- Indirect E_g → lower threshold
- Mechanical threshold!

Excitonic molecules / Biexcitons

- Fusion of two excitons into one quasi-particle
- Stability depends on ratio between effective masses
- Typical values 0.5-0.1 → $E_b = 0.1E_x$
  - Less resistant to thermal dissociation
  - 1.3-50K needed
- Radiative recombination:
  - Biexcition → free exciton (n=1 state) and one photon
  - M line



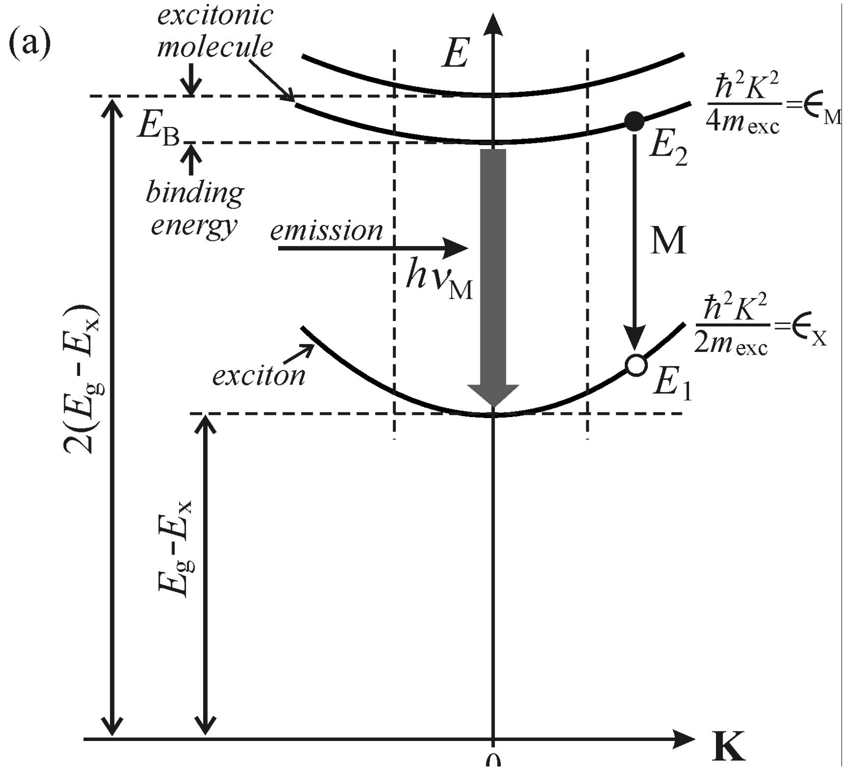
Biexitons EM

Direct semiconductor

- Fermi's golden rule

$$I_{\mathrm{sp}}^{\mathrm{M}}(h\nu) \sim \rho_{\mathrm{M}}(h\nu)\, f_{\mathrm{M}}(h\nu) |M_{\mathrm{M}}|^2.$$

- M_M independent of hv
- Effective temperature T_M
- Population factor f_M : Even n umber of fermions : total spin integer : bosons : don't have to take population factor into account
- Inverse Maxwell-Boltzmann distribution
- Recombining exciton takes a recoil **k**



$$\left\{\begin{array}{l} 2\left(E_{\mathrm{g}} - E_{\mathrm{X}}\right) - E_{\mathrm{B}} = h\nu_{\mathrm{M}} + \left(E_{\mathrm{g}} - E_{\mathrm{X}}\right) \\ h\nu_{\mathrm{M}} = \left(E_{\mathrm{g}} - E_{\mathrm{X}}\right) - E_{\mathrm{B}} \end{array}\right.$$

$$\boxed{\begin{aligned} I_{\mathrm{sp}}^{\mathrm{M}}(h\nu) \cong {} & \left[\left(E_{\mathrm{g}} - E_{\mathrm{X}} - E_{\mathrm{B}}\right) - h\nu\right]^{1/2} \\ & \exp\left\{-\left[\left(E_{\mathrm{g}} - E_{\mathrm{X}} - E_{\mathrm{B}}\right) - h\nu\right] / k_{\mathrm{B}} T_{\mathrm{M}}\right\} \end{aligned}}$$

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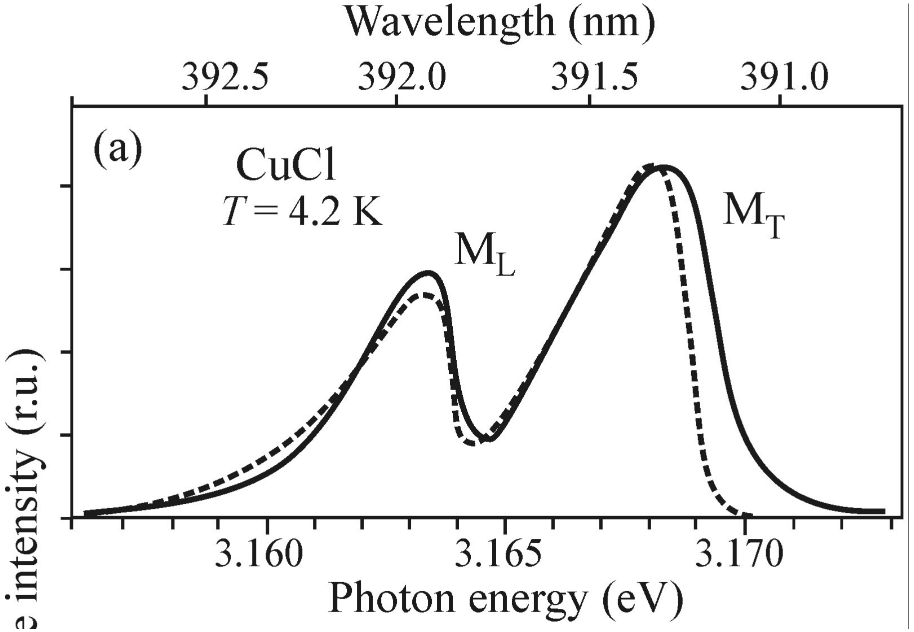

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- Effective temperature T_M
- Population factor f_M : Even n umber of fermions : total spin integer : bosons : don't have to take population factor into account
- Inverse Maxwell-Boltzmann distribution
- Recombining exciton takes a recoil **k**

$$\left\{\begin{aligned} &2\,(E_{\mathrm{g}} - E_{\mathrm{X}}) - E_{\mathrm{B}} = h\nu_{\mathrm{M}} + (E_{\mathrm{g}} - E_{\mathrm{X}}) \\ &h\nu_{\mathrm{M}} = (E_{\mathrm{g}} - E_{\mathrm{X}}) - E_{\mathrm{B}} \end{aligned}\right.$$

$$\boxed{\begin{aligned} I_{\mathrm{sp}}^{\mathrm{M}}(h\nu) \cong {} & [(E_{\mathrm{g}} - E_{\mathrm{X}} - E_{\mathrm{B}}) - h\nu]^{1/2} \\ & \exp\{-[(E_{\mathrm{g}} - E_{\mathrm{X}} - E_{\mathrm{B}}) - h\nu]\,/k_{\mathrm{B}}T_{\mathrm{M}}\} \end{aligned}}$$

Biexitons EM

Indirect semiconductor

- Phonon assistance
- Can't use joint density of states

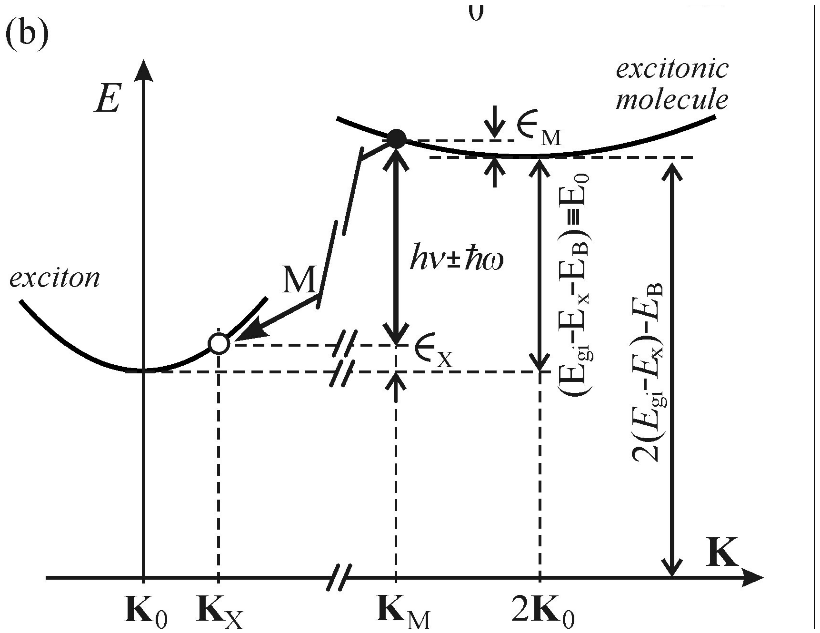
$$I_{\rm in}^{\rm M}(h\nu) \cong \int \rho_{\rm M}(\epsilon_{\rm M}) f_{\rm M}(\epsilon_{\rm M}) \rho_{\rm X}(\epsilon_{\rm X}) f_{\rm X}(\epsilon_{\rm X}) \left| M_{\rm in}^{\rm M} \right|^2 d\epsilon_{\rm M}$$

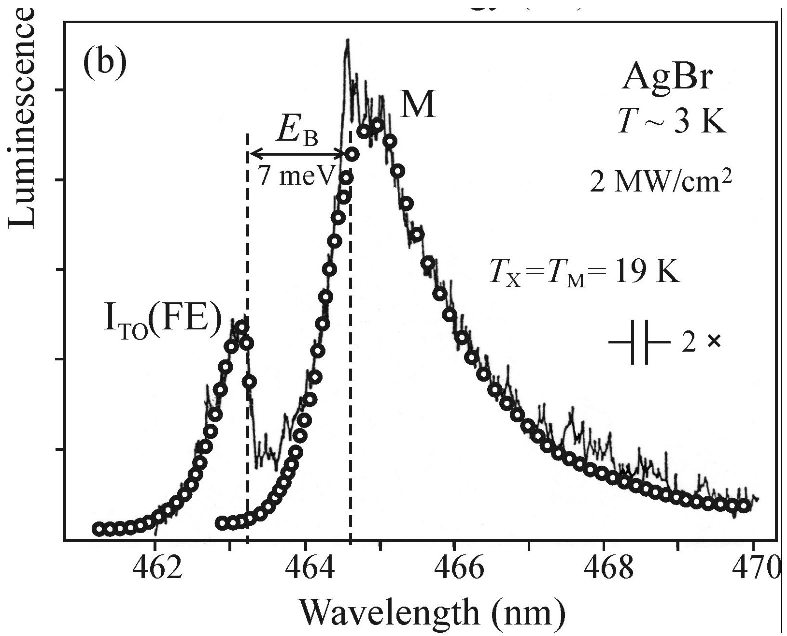
- Parabolic approximation of densities of states
- Lower integtion limit $\overline{h\nu} = h\nu + \hbar\omega - E_0.$
- Bosons in groundstate : ignore f_x

$$f_{\rm M}(\epsilon_{\rm M}) \approx \exp(-\epsilon_{\rm M}/k_{\rm B}T_{\rm M})$$

- Matrix element not constant: changes with k

$$\boxed{I_{\rm in}^{\rm M}(h\nu) \approx \int_{\overline{h\nu}}^{\infty} \sqrt{\epsilon_{\rm M}}\sqrt{\epsilon_{\rm M} - \overline{h\nu}}\, f_{\rm M}(\epsilon_{\rm M}) f_{\rm X}(\epsilon_{\rm X}) \left| M_{\rm in}^{\rm M} \right|^2 d\epsilon_{\rm M}.}$$





Biexitons EM

Intensity dependence

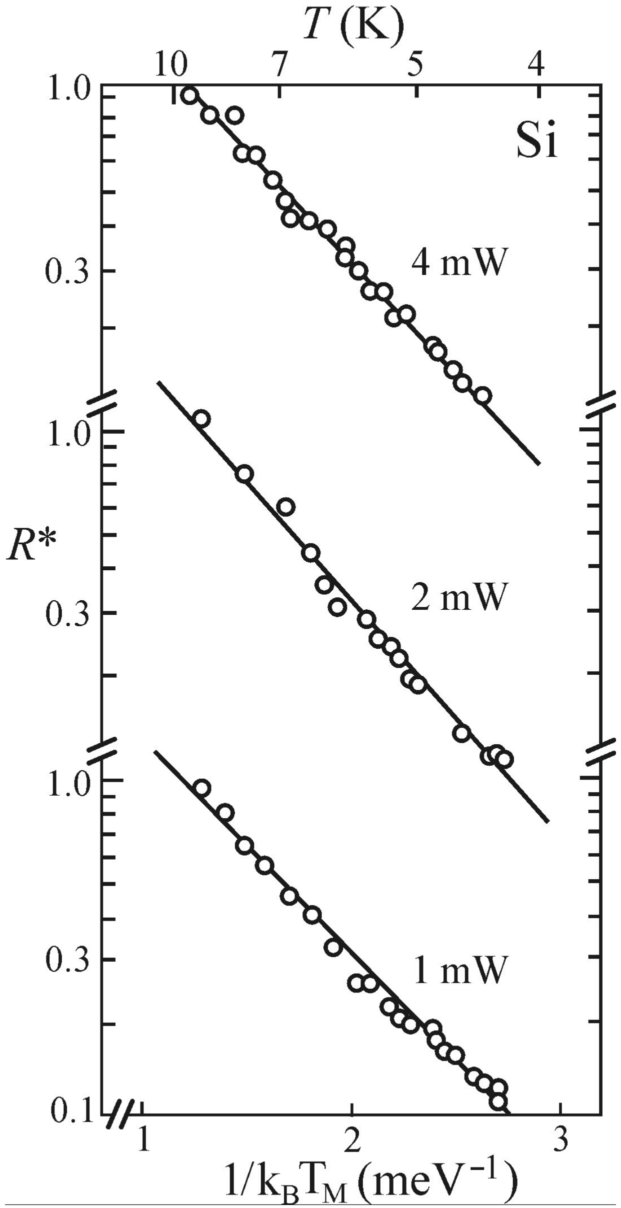
- Expect quadratic depence: two excitons have to be created
- n = 1.4 – 1.6 in experiments $I^{\mathrm{M}} \sim I_{\mathrm{ex}}^{n}$,
- Model with characteristic material dependent excitation intensity

$$I^{\mathrm{M}} \approx \mathrm{const}(\sqrt{1 + I_{\mathrm{ex}}/I_0} - 1)^2$$

Biexitons EM

Binding energy

- Direct: difference between FE and BE
- Indirect: Same but use fit of EM to get energy position at dispersion curve minimum
- Also: themordynamic by FE and M position as function of temperature
  - M line drops quicker than FE line when T increases, lower binding energy



Summary

- Free excitons
  - Direct bandgap: FE or X or FE-LO phonon
    - FE polariton or (optical) phonon assisted
  - Indirect bandgap:
    - Phonon assisted
  - Linear function of excitation intensity
- Bound excitons
  - Shallow impurities (donors/acceptors)
    - Narrow line
  - Isoelectric impurities
    - Narrow line if low electron-phonon coupling
  - No temperature broadening
  - Linear function of excitation intensity until saturation
  - Lower hv but stronger intensity
- Biexcitons (Excitonic molecules)
  - Superlinear function of intensity
  - Sort of inverse Maxwell-Boltzmann distribution

Classification of luminescence by intensity

Low-fluence (Weak) excitation (0.01- 10 W/cm^2): gas-discharge lamp, incandescent lamp, continuous-wave gas laser

➡ - Recombination of **free excitons** (FE or X; FE-LO phonon)

➡ - Recombination of a **bound exciton** (BE): (D^0-X; A^0-X; D^+-X; A^-X, isoelectronic impurity)

✓ - Recombination of **donor-acceptor pairs** (D^0-A^0)

✓ - Recombination of **free hole/electron** with **neutral donor/acceptor** (h-D^0; e-A^0; e-h)

1 sun = 0.1 mW/cm^2 = 0.1 W/cm^2 = 1 kW/m^2

High-fluence (Strong) excitation (1 kW-10 MW/cm^2):

➡ - Radiative decay of **excitonic molecule** (EM or XX)

✗ - Luminescence from inelastic collisions of excitons (X-X collisions)

✗ - Luminescence of electron-hole liquid (EHL) or electron-hole plasma (EHP)

✗ - Bose-Einstein condensation of excitons or excitonic molecules

These excitations can occur simultaneously, in these two chapters we focus on low-fluence excitation and neglected luminescence from excitons