Properties of excitonic states in MOCVD-grown Zn(Cd)Se/ZnMgSSe quantum wells with spreaded heterointerfaces

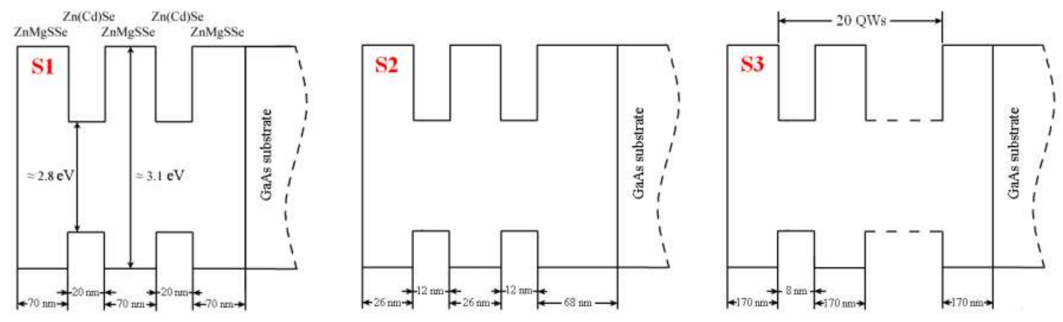
A.F. Adiyatullin[1,2], S.V. Shevtsov[1,2], A.N. Minnullin[2] and V.S. Krivobok[1,2*]

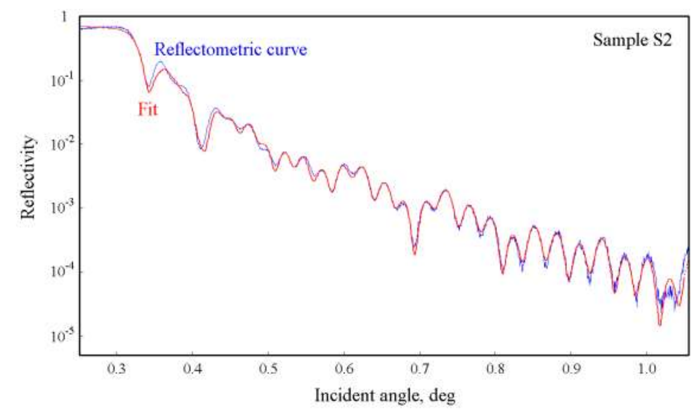
[1]P.N. Lebedev Physical Institute, Russian Academy of Sciences, Moscow, 119991 Russia; [2]Moscow Institute of Physics and Technology (State University), Dolgoprudny, 141700 Russia; *e-mail: krivobok@lebedev.ru

Motivation

- In contrast to the model QW heterointerface, real interface is often characterized by diffusion broadening and disorder, affecting the electronic and optical properties of QWs.
- Electronic properties of heterovalent ZnSe/GaAs interface depend strongly on the structure of Ga-Se and Zn-As bindings. They result in specific band bending promoting charge redistribution in the structure. QWs act as additional carrier reservoirs helping in understanding of ZnSe/GaAs interface features.
- Properties of real interfaces are of crucial interest for development of electronic devices. Being a wide-bandgap semiconductor, ZnSe is promising for several optoelectronic applications.

Samples





- MOVPE growth in a hydrogen atmosphere at 85 Torr and 450 °C.
- Samples with two QWs: the bottom one had been held at a high growth temperature longer than the top QW, which led to the different diffusion blurring of QWs.

S2: best fit of X-ray reflectometry curve reproduces interface blurring 1.5 nm for bottom QW and <0.5 nm for the top one.

Exciton emission spectra

Resonances in PL and photoreflectance spectra:

- Free excitons with heavy hole (***X***), light hole (***X_{lh}***), excited state for light-hole exciton (***X_{lh}^****) ***hh-lh*** splitting 25 meV, ***X_{lh}*** excitation energy 17 meV, corresponding to theoretical calculations
- Excitons bound on neutral donors (***D^0X***)
- Deep acceptor related transitions (***DA***)

Exciton-phonon interaction

- X line homogenous broadening

$$\Gamma_L(T) = \Gamma_0 + \Gamma_{ac}T + \frac{\Gamma_{LO}}{\exp(\hbar\omega_{LO}/kT)}$$

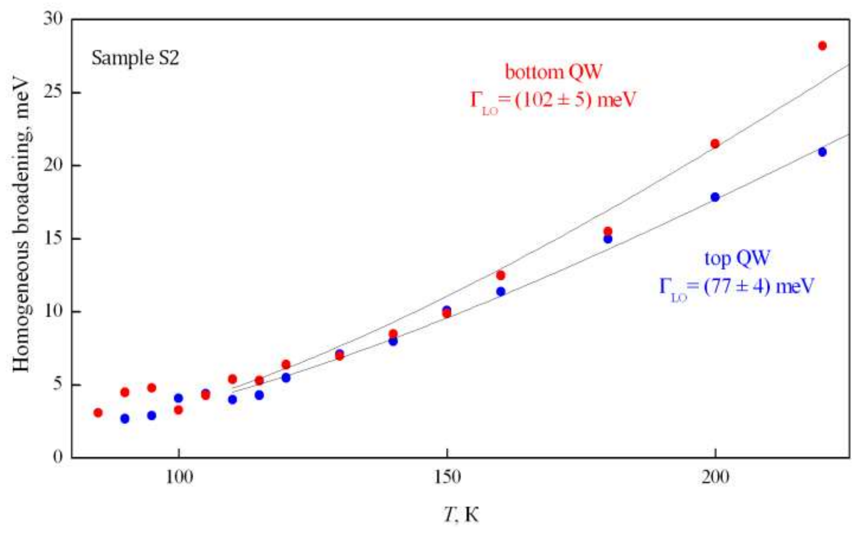
- Interface blurring results in enhancement of exciton-LO phonon interaction (Fröhlich mechanism)
- Different effective masses and barrier heights for e^- and h^+ in QW

⇩

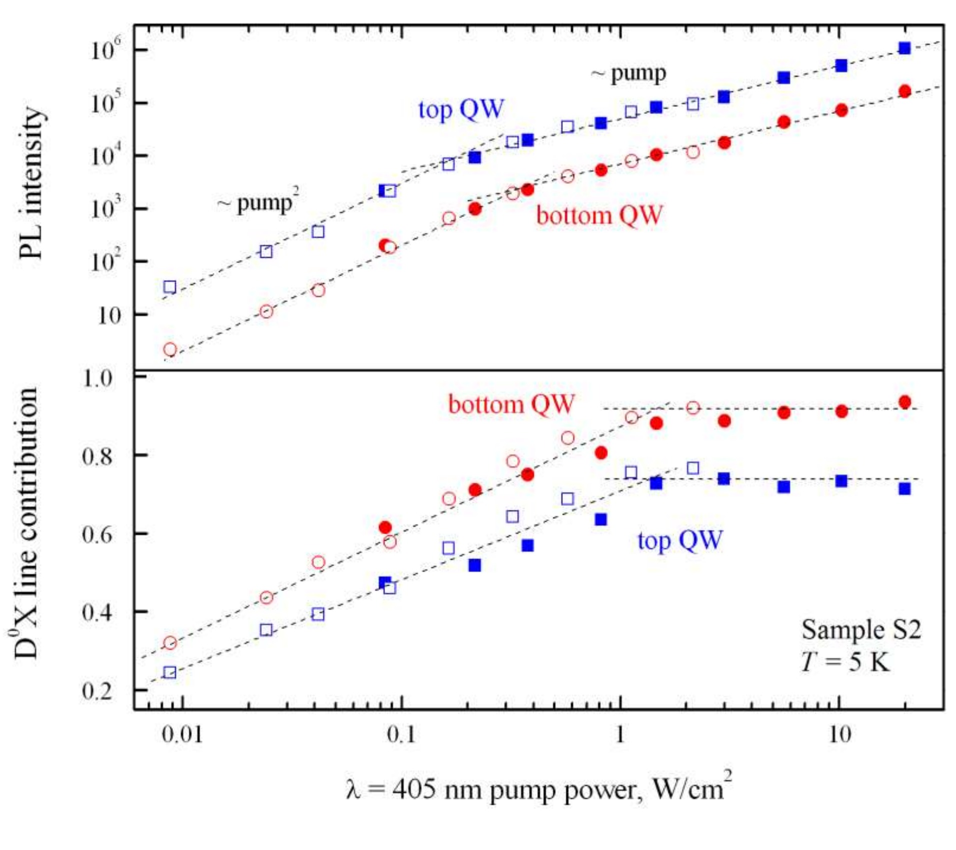
different degree of e^- and h^+ penetration in the barrier

⇩

local exciton polarization enhancing Fröhlich interaction



Nonradiative exciton decay channel



Total PL intensity on pumping:

- linear at high pump powers (constant quantum yield)
- superlinear at low ones (additional nonradiative decay channel)

D^0X line contribution to the PL signal:

- grows at low pump powers
- saturates at high ones

Nonradiative exciton decay channel is linked to bound exciton concentration.

Model of donors with neutral or charged states

- low pump powers: excitons are trapped by charged donors $D^+ + X \to D^0 + h^+$
- high pump powers: majority of charged donors are neutralized

Heterointerface influence on bound exciton binding energy

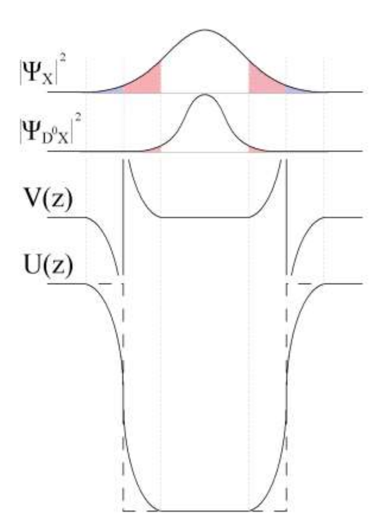
Heterointerface spreading is a small perturbation ***V*** of the ideal rectangular QW potential, leading to the shift of free and bound exciton energy levels:

$$\Delta E_X = \langle \Psi_X | V | \Psi_X \rangle, \Delta E_{D^0X} = \langle \Psi_{D^0X} | V | \Psi_{D^0X} \rangle$$

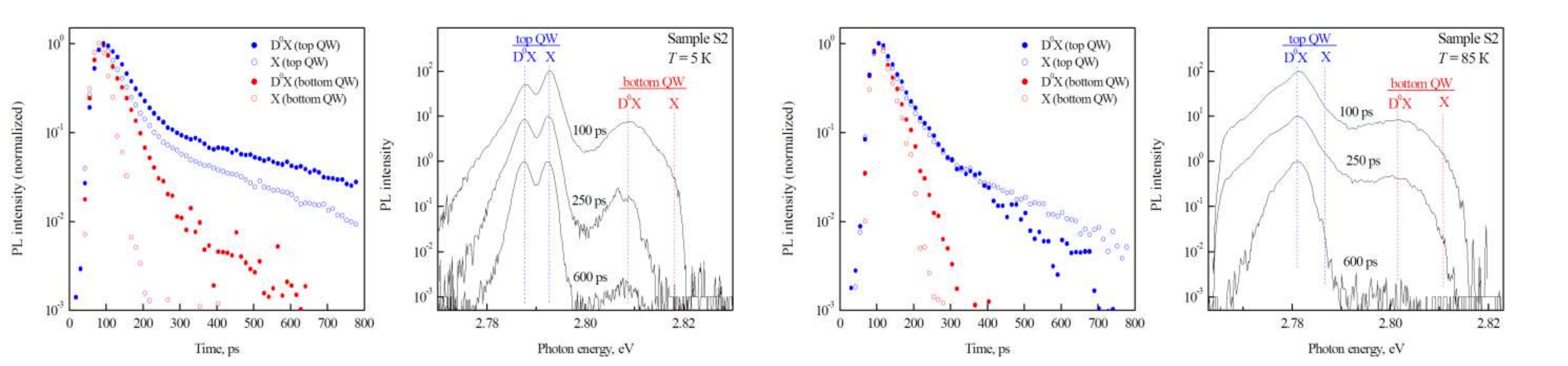
Ψ_{D^0X} is localized more than Ψ_X and is hardly affected by the perturbation. Total energy shift is determined by ΔE_X

$$\Delta E_X >> \Delta E_{D^0X} \Rightarrow \Delta E_{total} = \Delta E_X - \Delta E_{D^0X} \approx \Delta E_X$$

rising with heterointerfaces blurring.



PL dynamics



Bi-exponentional PL decay:

- ***t_{fast}*** ~ 30–50 ps – exciton trapping by ***D^+***
- ***t_{slow}*** ~ 200–400 ps – radiative lifetime, vanishes with temperature growth due to thermal escape of excitons

t_{slow} vanishes at lower temperature for the bottom QW

⇩

increase of defects and charged/neutral donors concentration with interface blurring

Broadening of exciton emission line

- Increase in pump power leads to the broadening of ***X*** line in PL spectra as well as corresponding resonances in reflectance spectra.
- According to Saha equation $n_X < 0.15\, n_e$

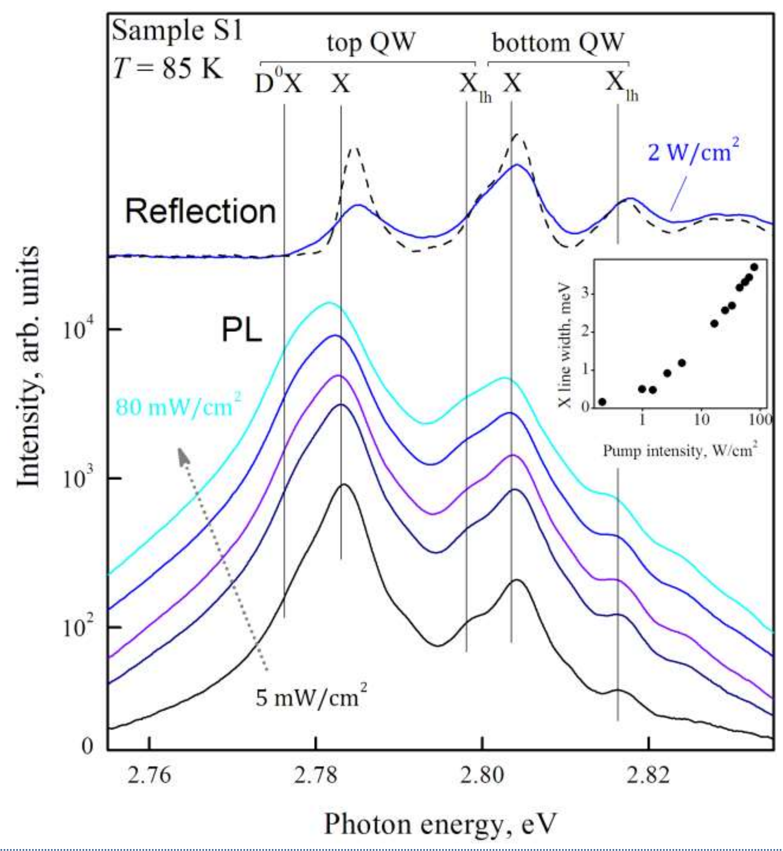
⇩

exciton-electron scattering plays a major role in ***X*** line broadening

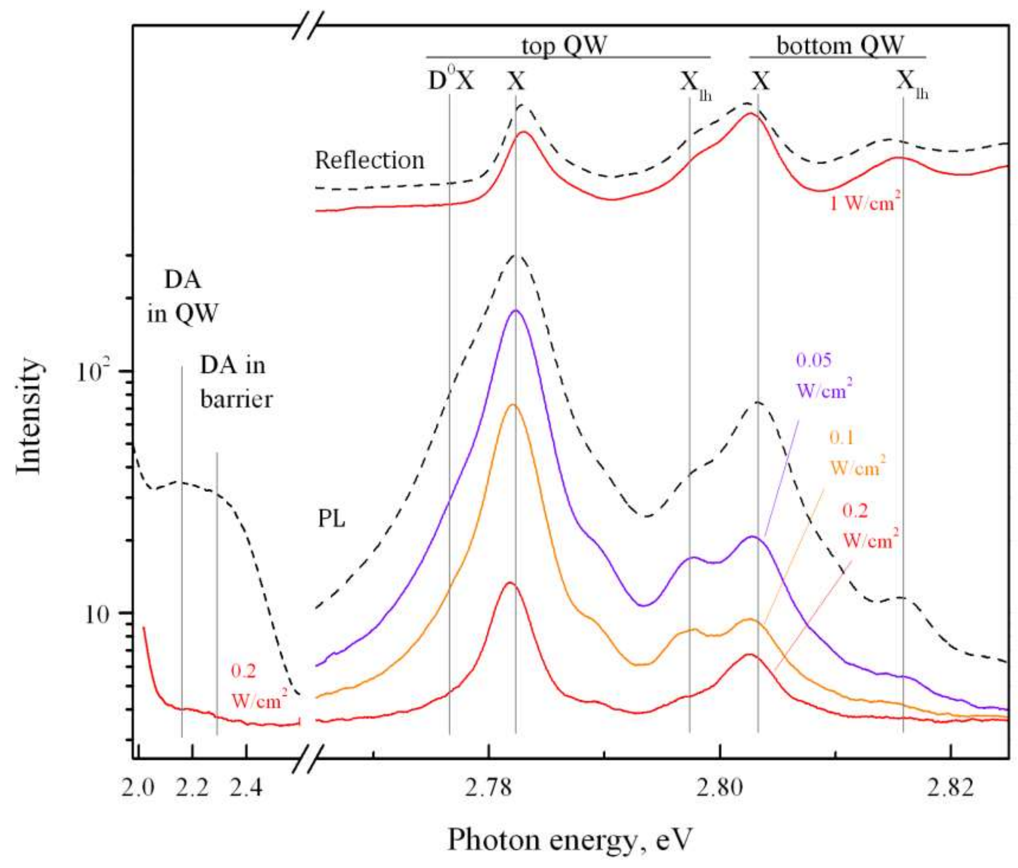
- From the X line broadening: $n_e \sim 10^{11}$ cm^{-2} Photoinduced carrier concentration: $n_e \sim 10^9$ cm^{-2}

⇩

increase in pump power leads to excess electron concentration in QW



Effect of below-barrier illumination on excitonic states



Effects at low pump powers and T ~ 100 K:

(1) drastic decrease of PL quantum yield;

(2) ***D^0X*** line disappearance;

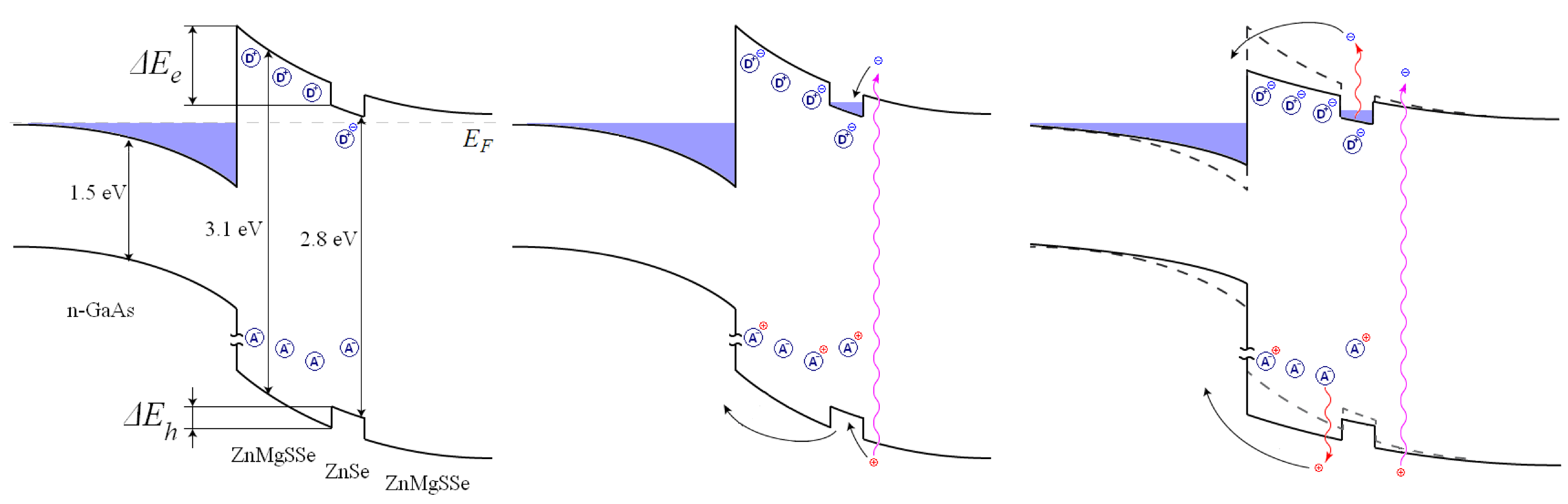
(3) ***X*** line narrowing.

Illumination leads to n_e decrease

⇩

- according to Saha equation for $D^+ + e^- \leftrightarrow D^0$ number of ***D^+*** (nonradiative decay centers) increases
- number of ***D^0*** decreases
- X line is narrowed due to electron-exciton scattering reduction

ZnMgSSe/GaAs heterointerface



- Interdiffusion of *Zn* and *Ga* atoms during the growth
- Band bending due to ionized *Ga* donors in ZnMgSSe layer
- Barrier in conductivity band preventing e^- flow to the substrate; no barrier for h^+
- Excess e^- concentration in QWs

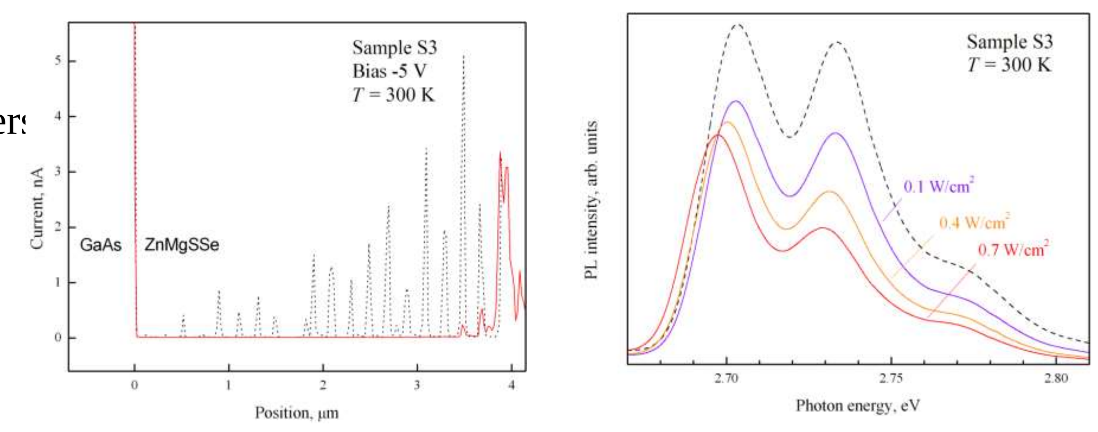
Possible effects of below-barrier illumination on excess n_e:

– decrease of the barrier height due to deep acceptor ionization

– free electron absorption

SSRM measurements with additional illumination

- In the dark: no current through ZnSe/ZnMgSSe layers
- Under the optical pumping: e^- current from the QWs, no current from barrier layers
- Additional below-barrier illumination: decrease in e^- electronic current from QWs as well as decrease in PL quantum yield



⇩

Optical pumping leads to the electron accumulation in QWs.

Additional below-barrier illumination causes decrease of excess electron concentration in QWs.

Conclusion

- ZnSe/ZnMgSSe QW heterointerfaces blurring leads to:
  1) enhancement of Fröhlich interaction due to the induced exciton polarization;
  2) increase in ***X***–***D^0X*** line distance due to different degree of localization of free and bound excitons on the heterointerface;
  3) increase in concentration of neutral and charged defects.
- The major exciton nonradiative decay channel is trapping on charged donors.
- Under excitation an excess electron concentration in QWs due to band bending on ZnMgSSe/GaAs interface is presented.
- Additional below-barrier illumination leads to the disappearance of excess electron concentration in QWs, resulting in
  1) dramatic decrease in PL quantum yield due to increase in number of ***D^+*** centers;
  2) disappearance of ***D^0X*** line caused by decrease in number of ***D^0*** centers;
  3) narrowing of ***X*** line due to decrease of exciton-electron scattering role.
- Charge redistribution processes are confirmed by the measurements of scanning spreading resistance microscopy with additional illumination.