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Abstract. For the analysis of ZnO luminescence, a set of rate equations (SRE) is proposed. It contains a set of parameters that characterize processes participating in luminescence: zone–zone excitation, excitons formation and recombination, formation and disappearance of photons, surface plasmons (SP), and phonons. It is shown that experimental ZnO microstructure radiation intensity dependence on photoexcitation levels can be approximated by using SRE. This approach was applied for the analysis of ZnO microfilm radiation with different thicknesses of Ag island film covering. It was revealed that the increase of cover thickness leads to an increase of losses and a decrease of the probability of photon-to-SP conversion. In order to take into account visible emission, rate equations for level populations in the bandgap and for corresponding photons and SPs were added to the SRE. By using such an SRE, it is demonstrated that the form of visible luminescence intensity dependence on excitation level (P) like $P^{1/3}$, as obtained elsewhere, is possible only if donor–acceptor pairs exist. The proposed approach was also applied for consideration of experimental results obtained in several papers taking into account the interpretation of these results based on assumptions about the transfer of electrons from the defect level in the ZnO bandgap to metal and then to the conduction band. [DOI: 10.1117/1.JNP.10.016001]

Keywords: zinc oxide; luminescence enhancement; surface plasmon resonance.

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1 Introduction

ZnO is a promising material for various optoelectronic applications. It is a direct-gap semiconductor whose bandgap width equals ~3.3 eV at room temperature (RT) and whose exciton binding energy equals 59 meV, which provides ultraviolet (UV) recombination luminescence existence at RT and higher. Defects and impurities in ZnO create energy levels in the bandgap, which provides visible radiation. In addition, it is possible to grow various morphologies of ZnO microstructures. There are numerous applications of ZnO in different areas, particularly in optics—as a material for UV photon detectors, light-emitting diodes, and lasers.

The analysis of regularities of UV and visible ZnO radiation and determination of radiation optimization methods, in particular due to surface plasmon resonance (SPR), is of great interest. There is a vast amount of papers devoted to experimental investigations of zinc oxide radiation. However, these papers do not include detailed interpretation of the results obtained.

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To the best of our knowledge, dependence of photoluminescence enhancement under the influence of SPR on pumping level was observed only in Ref. 1, but in this paper no interpretation of this dependence was given. In Ref. 2, it was shown that the dependence of visible radiation intensity on pumping level (P) appears as $P^{1/3}$, but it was not determined in what case that is correct. In Refs. 3–6, it was discovered that when UV and visible radiation present simultaneously in the ZnO film, the intensity of the visible part is significantly higher than that of the UV part. However, a metal covering (Ag or Au) of the film changes the form of the spectra: UV radiation intensity becomes significantly higher than the visible radiation intensity. The authors suggested an interpretation of this effect but did not analyze its dependence on pumping level.

In the course of ZnO luminescence investigation, a detailed study of the SPR influence on the exciton recombination luminescence intensity of ZnO films covered by Ag island films was carried out. Different excitation sources were used. In case of a He–Cd laser, luminescence enhancement compared to the same ZnO sample without an Ag cover was observed. Nevertheless, while using the Nd:YAG laser for the excitation of the same samples, luminescence quenching was observed, and enhancement could be achieved only with a significant decrease of the excitation level. It was shown that the dependence of exciton luminescence intensity on pumping level in the presence of an Ag cover is significantly nonlinear, whereas the dependence without the Ag cover is, as a rule, linear (see e.g., Ref. 2). This nonlinearity leads to the following effect: while the excitation increases, the luminescence enhancement decreases and finally turns into quenching.

A simplified approach is proposed for the description of ZnO luminescence and the SPR influence on it. This approach is based on the set of rate equations (SRE), which describes processes taking part in the luminescence.

Application of this SRE for the analysis of ZnO microfilms luminescence allows us to:

1. Obtain calculated dependences of UV luminescence intensity on pumping level close to experimental ones.
2. Estimate the influence of Ag film thickness on the luminescence enhancement due to SPR.
3. Check the hypothesis from Refs. 3–6 of the existence of electron transfer from the level in the ZnO bandgap to the ZnO conduction band through metal nanoparticles, which leads to a sharp enhancement of UV luminescence intensity with the decrease of the visible one. Using SRE, it was discovered that this effect could exist only under low pumping levels.
4. Demonstrate that the dependence of visible luminescence intensity on pumping level (P) following a sublinear power law with the exponent $1/3$ $(P^{1/3})^2$ will exist only in case of two levels in the ZnO bandgap, that is, in the case of donor–acceptor pairs (DAP) existence.

2 Ultraviolet ZnO Radiation

2.1 *Description of the Set of Rate Equations*

The analysis of the UV luminescence of various ZnO films with Ag cover using / allows us to estimate the variation of different characteristics, such as losses, closeness of luminescence maximum wavelength to SPR, and others. In order to use the proposed SRE, it is necessary to consider that all process participants have a corpuscular nature. Using SRE, we describe the time dependence of concentrations for electrons in the conduction band (n_1), holes in the valence band (n_3), excitons (n_4), photons caused by exciton recombination and forming plasmons (n_2), photons which simplistically describe the field produced by plasmons in the nanoscale region between the Ag cover and ZnO (n_5), and phonons caused by heating (n_6).

It should be noted that a part of this investigation has already been reported by Ryzhkov et al.[7]. But in that article, phonons caused by heating of the ZnO/Ag system due to pumping were not taken into account. In the course of further investigation, it was discovered that the maximum of the ZnO UV luminescence spectra shifts to the long wavelength region with the increase of pumping energy. Most likely, that shift is caused by the bandgap narrowing

with temperature growth. The value of the shift equals approximately 4 to 5 nm (~40 meV). Taking into account that the bandgap decreases by ~0.35 meV with 1 K heating[8] (in the case of $T > 200$ K), it is possible to estimate that the ZnO/Ag system temperature rises up to ~380 K. This results in the necessity of addition of the equation for phonons in the SRE. The SRE for exciton recombination (UV) radiation is as follows:

$$\begin{cases} \frac{dn_1}{dt} = Pe^{\frac{-(t-10)^2}{26.4}} - (M+B)n_1n_3 \\ \frac{dn_2}{dt} = Hn_4 + A(1-L_An_6)n_5n_4 - Cn_2 - Sn_2 \\ \frac{dn_3}{dt} = Pe^{\frac{-(t-10)^2}{26.4}} - (M+B)n_1n_3 \\ \frac{dn_4}{dt} = Mn_1n_3 - Rn_4 - A(1-L_An_6)n_5n_4 \\ \frac{dn_5}{dt} = N_0(1-L_Nn_6)Sn_2 - JN_0(1-L_Nn_6)Sn_2(1+L_Wn_6)n_2 - Tn_5 \\ \frac{dn_6}{dt} = \alpha Pe^{\frac{-(t-10)^2}{26.4}} + Vn_2 + JSn_2(1+L_Wn_6)n_2 - En_6 \end{cases},$$

where t refers to time in nanoseconds, n_i refers to concentrations in 10^{18} cm^{-3}, P refers to the excitation level, and the multiplier attached to P is the time dependence of the excitation pulse that corresponds to the experiment. Rough estimation shows that an energy density of the excitation pulse of 1 mJ/cm^2 corresponds to $P = 3.3$.

Let us fix upon the system parameters in more detail. In the equations for n_1 and n_3, parameter B refers to the interband recombination probability, and parameter M refers to the exciton formation probability. These two equations coincide, and a difference of results can be caused only by different initial conditions. In the equation for excitons (n_4), parameter R is the probability of exciton spontaneous recombination, parameter A refers to the probability of exciton recombination under the influence of the resonance field formed by plasmons, and parameter L_A refers to the influence of system heating on parameter A. In the equation for photons produced by exciton recombination (n_2), parameter H is the radiation exciton recombination probability ($H < R$) and parameters C and S are the probabilities of photon escape from the system and interaction of photons with the Ag covering with plasmon formation, respectively.

In the equation for photons which simplistically describes field produced by plasmons (n_5), parameter N_0 is a coefficient that refers to the plasmon field increasing against the field of photons formed by exciton recombination that form plasmons, parameter L_N refers to the influence of system heating on parameter N_0, parameter T is the characteristic of plasmon radiation decay, parameter J is the characteristic of Joule's losses in the Ag cover, which are determined by Ag electrical resistivity, and parameter L_W refers to the temperature dependence of Ag electrical resistivity. In the equation for phonons (n_6), parameter α refers to the relation of the part of photoexcitation converted into heat to the part of photoexitation that was used in ZnO excitation, parameter E is the probabilty of phonon attenuation, and parameter V is the probability of direct conversion of a photon into a phonon.

Let us consider the terms of the equation that determines Joule's losses in more detail. Photons caused by exciton recombination get inside the Ag cover. The rate of their input equals $\gamma = Sn_2$ (per unit time). They produce electron vibrations (plasmons). Those vibrations actually appear to be an electrical current. The second power of the electrical current is determined by the concentration of input photons: $I^2 \sim n_2$. Losses (with heating power of $I^2R \sim n_2$) arise in the Ag cover. Dividing this value by the energy of the photon ($h\nu$), it is possible to estimate a photon's lifetime before its conversion into heat: $1/\tau = I^2R/h\nu \sim n_2$. So, the rate of photon input in Ag and the rate of conversion into heat are proportional to n_2, and, consequently, Joule's losses equal JSn_2^2 and significant nonlinearity appears in the equation. Hence, the field in the narrow region at the interface between ZnO and Ag, simplistically described by photons n_5, will be formed by the remaining plasmons. The rate of input of such plasmons is proportional to $Sn_2(1 - Jn_2)$. As is known, electric field strength in the interface is much higher than field strength inside the metal.[9] So, in the equation the n_5 term which describes the plasmon input rate should be multiplied by $N_0 > 1$. As expression JSn_2^2 describes heat losses, it appears in the equation for phonons (n_6) with a positive sign.

2.2 *Application of Set of Rate Equations for the Analysis of ZnO Ultraviolet Luminescence*

The SRE was solved using MathCAD by the Runge–Kutta numerical method. The result of the calculation is the sum of photon concentrations $I_2 = \Sigma_t n_2^t$ at all (50,000) points of time in which the calculation was performed. This value is proportional to the integrated luminescence intensity. The calculation time was defined by the duration of the photoexcitation laser pulse. For the uncovered part of the film, parameters which correspond to the exciton luminescence lifetime of ~100 ps and efficiency of ~5% were taken. Those values are rather close to the experimental ones. The values equal $H = 0.5$, $R = C = 10$. We also set B and M to 20.

Let us consider estimations of the parameters that take into account the influence of heating (phonons) on the process in consideration. As was mentioned in the introduction, the heating of the ZnO/Ag system in the experiment was ~100 deg. By virtue of the fact that the temperature dependence of the resistance is $R = R_0[1 + \beta(T - T_0)]$, where for Ag $\beta \approx 0.004$, the maximum change of resistance must be around 40%. That is why the value of L_W equals 0.3. Parameters L_A and L_N should take into account the widening of the Ag extinction spectrum and, as a result, resonance degradation. As the width of this spectrum is big even at low temperatures, heating does not play significant role in this process. Solutions of the SRE using values of L_A and L_N equaling 0.3 and 1 correspondingly gave the same result for I_2. So for those parameters the value 0.3 was taken. For parameters α, E, and V, more or less arbitrary values were taken ($\alpha = 10$, $E = 1000$, and $V = 1$). Preliminary calculations have shown that the variation of those parameters' values (staying in a constant order of magnitude) does not significantly affect the result.

For the comparison of experimental dependencies of luminescence intensity on excitation level $I(P)$ with the calculated ones, those dependences should be plotted in the same scales. To get the same scale on the X axis, we should take into account that, as was mentioned above, an energy density of the excitation pulse of 1 mJ/cm^2 corresponds to $P = 3.3$. To make the same scale along the Y axis we should add one more simplification: we should assume that the calculated linear dependence $I(P)$ obtained using parameters for the uncovered part of the film coincides with the experimental dependence.

2.3 *Comparison with the Experiment*

The proposed approach was applied to two types of ZnO films with different manufacturing methods. The first film was prepared by thermal evaporation; the second film was prepared by magnetron sputtering. On the first film, there were three regions: uncovered ZnO and ZnO covered with 10- and 20-nm Ag film (effective thickness was determined using the suggestion of uniform Ag distribution on a semisphere in the course of thermal deposition). On the second film, there were two regions: uncovered ZnO and ZnO covered with ~10-nm Ag film. The comparison between the experimental and calculated dependences $I(P)$ for the first ZnO film is shown in Fig. 1. Calculated curves for 10- and 20-nm Ag approximate the experimental data rather well. Those calculated curves differ from each other only by the value of parameters J

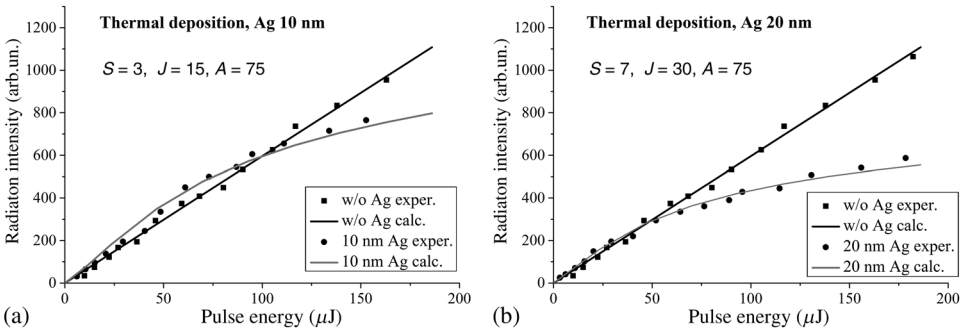

Fig. 1 Comparison of experimental and calculated dependences of luminescence intensity on excitation level for the first ZnO/Ag sample (thermal deposition): (a) Ag thickness is 10 nm and (b) Ag thickness is 20 nm.

(which determines losses in Ag cover) and S (the probability of interaction of photons with Ag cover with plasmon formation). All other parameters are equal for both curves. It turns out that with the increase of Ag film thickness, losses and the probability of interaction of photons with the Ag covering with plasmon formation become higher.

In Fig. 2, the dependence $I(P)$ for the second film is shown. It can be seen that in this case significantly greater enhancement of luminescence intensity is possible. In comparison with the first film, here anotably greater value of parameter A, which characterizes the probability of exciton recombination under the influence of the resonance field of plasmons, and lower value of losses (J) was obtained.

From our point of view, such an approach to the analysis of the dependence of luminescence intensity on excitation level, giving the opportunity to estimate characteristics of samples, gives possibilities for optimization of sample structure for the highest possible luminescence intensity enhancement due to SPR.

3 Visible ZnO Radiation

The proposed approach can be used for zinc oxide visible radiation consideration along with UV radiation. For this purpose the SRE is supplemented with the rate equations for the population of energy levels in the bandgap, caused by defects, for photons of visible radiation caused by transitions between the levels mentioned above and the valence band, and (in the presence of SPR) for photons of visible radiation which we use for simplified description of the field produced by plasmons in the nanoscale region between the Ag cover and ZnO. As a result, in the case of a single defect level in the ZnO bandgap, SRE consists of nine equations. In the case of existence of DAP in ZnO, in other words in the case of two energy levels in the ZnO bandgap, the SRE consists of 10 equations.

3.1 *Dependence of Visible Radiation on Excitation Level*

In Ref. 2, it was experimentally demonstrated that for visible radiation of ZnO the dependence of radiation intensity on excitation level could be approximated with high accuracy by the following relation: $I(P) \sim P^{1/3}$. Mathematical modeling of the dependence of visible radiation intensity on excitation level using the suggested SRE revealed that the dependence like $I(P) \sim P^{1/3}$ can be obtained only for the case of DAP existence, that is, two energy levels existing in the bandgap [Fig. 3(b)]. If only one defect level exists in the bandgap, the dependence $I(P)$ would look like $I(P) \sim P^k$, where $1/3 < k < 1$ [Fig. 3(a)].

Thus, utilization of the proposed simplified approach to mathematical modeling in combination with experimental dependences $I(P)$ can give additional information about the origin of ZnO visible radiation.

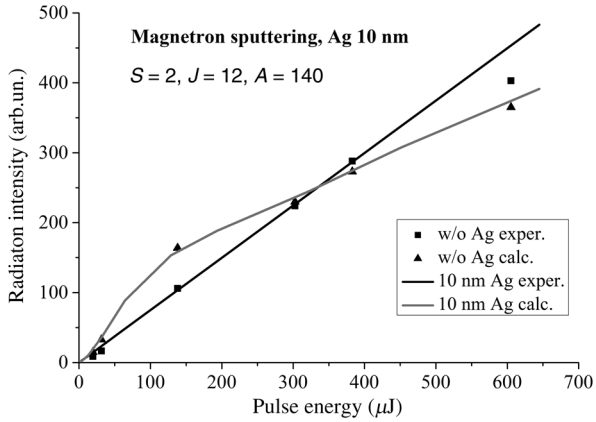

Fig. 2 Comparison of experimental and calculated dependences of luminescence intensity on excitation level for the second ZnO/Ag sample (magnetron sputtering).

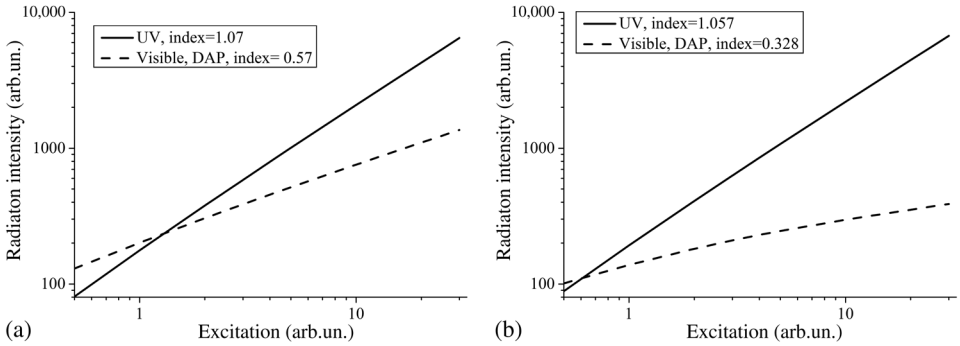

Fig. 3 Calculated dependences of luminescence intensity on excitation level for (a) one donor energy level in bandgap and (b) two energy levels in bandgap (DAP).

3.2 *Analysis of Surface Plasmon Resonance Influence on Ultraviolet and Visible Luminescence*

This approach can also be used for analysis of regularities in the influence of SPR on UV and visible radiation existing simultaneously. In Refs. 3–6, it was discovered that when UV and visible radiation are present simultaneously in the ZnO film the intensity of the visible part is significantly higher than that of the UV part. However, a metal covering (Ag or Au) of the film changes the form of its spectra: UV radiation intensity become significantly higher than visible radiation intensity. The authors suggest the following interpretation of this effect: (1) electron transfer from energy levels in the ZnO bandgap to metal particles, (2) electron excitation inside metal particles by visible radiation (with plasmon formation), and (3) further transfer of excited electrons to the ZnO conduction band.

It seems that it is possible to make the assumption that the metal covering increases the intensity of UV radiation due to SPR and decreases visible emission intensity due to effective annealing of the ZnO surface and consequently decreases the number of defects. This assumption will be verified as follows.

In our opinion, it is interesting to perform mathematical modeling of UV and visible ZnO emission together, taking into account such processes and discovering conditions that should cause such a luminescence spectrum transformation. As measurements in Refs. 3–6 have been carried out under continuous pumping, we should use a stationary variant of SRE (all derivatives equal zero and pumping level P is constant):

$$\begin{cases} P-(B+M)n_1n_3-B_2n_1(n_0-n_6)+Wn_6=0 \\ Hn_4+An_4n_5-Cn_2-Sn_2=0 \\ P-(B+M)n_1n_3-B_1n_3n_6-A_1n_3n_6n_8=0 \\ Mn_1n_3-Rn_4-An_4n_5=0 \\ N_0Sn_2(1-Jn_2)-Tn_5=0 \\ B_2n_1(n_0-n_6)-B_1n_3n_6-A_1n_3n_6n_8-Wn_6=0 \\ \alpha B_1n_3n_6+A_1n_3n_6n_8-C_1n_7-S_1n_7=0 \\ N_{01}S_1n_7(1-J_1n_7)-T_1n_8=0 \end{cases}.$$

Here, we have added equations for electron concentration on energy level in the bandgap (n_6), for visible photons concentration (n_7), and for concentration of photons, which simplistically describe the field produced by plasmons on visible emission frequencies in the nanoscale region between the metal cover and ZnO (n_8). We should mention that in this variant of the SRE we have excluded the rate equation for phonons due to negligible heating on the system under low power He–Cd continuous pumping.

Parameters A, C, S, J, N_0, and T with index "1" have the same meaning for visible emission as the corresponding parameters for UV emission. Additional parameters are as follows: B_1 refers to the probability of electron transition to the valence band from energy levels in the bandgap, B_2 refers to the probability of electron transition from the conduction band to energy levels in the bandgap, n_0 refers to the total number of levels in the bandgap (defect

concentration), α refers to the degree of radiating transitions of electrons from energy levels in the bandgap to the valence band, and W refers to the probability of electron transfer from energy levels in the bandgap to metal and the probability of transfer of excited electrons from metal to the ZnO conduction band. The SRE was solved using Newton's method, and dependences of UV radiation intensity (n_2) and visible radiation intensity (n_7) on pumping level P were analyzed.

The following situations have been considered:

1. Absence of SPR ($S = S1 = 0$).
2. Presence of SPR.

   2a. Electron transfer is "turned on" ($W = 30$) without taking into account potential growth of SPR parameters ($A = A1 = 50$) and decreasing number of defects ($n_0 = 0.25$).

   2b. Electron transfer is excluded ($W = 0$). However, due account is taken of the eventual increase of SPR parameters ($A = A1 = 200$) as well as a decreasing number of defects ($n_0 = 0.15$).

   2c. Both processes are taken into account.

Photoexcitation in experiments[3–5] was performed by a continuous He–Cd laser at 325-nm wavelength. Rough estimation shows that in this case a pumping level of 100 W/cm^2 (100 mW, pumping spot diameter equals 0.3 mm) corresponds to $P = 0.02$. That is why calculation were performed for P starting from 0.01.

In Fig. 4, calculated dependences of the relation between visible and UV radiation (n_7/n_2) on P for all four cases are shown.

Here, we can clearly see that the sharp decrease observed in experiments of the relation between intensities of visible and UV radiation in the case of a ZnO metal cover is relevant in the model only when there is an electron transfer from the level in the ZnO bandgap through the metal into the conduction band ($W \neq 0$). So, SPR and the decrease of visible emission intensity due to effective annealing of the ZnO surface and consequently a decrease of defect number without taking into account electron transfer cannot explain the experiment.

Figure 5 demonstrates the dependences of the enhancement of UV and visible radiation on the excitation level in the case of a ZnO metal cover. Three cases are examined:

1. Taking into account electron transfer from the level in the bandgap through the metal into the ZnO conduction band with the simultaneous growth of SPR parameters and the decrease of defect concentration;
2. Without taking it into account; and
3. Without electron transfer ($W = 0$), however, with the increase of SPR parameters and decrease of defect concentration.

Figure 5 clearly demonstrates the role of parameter W. Under excitation level $P \sim 1$, $W \neq 0$ leads to a small enhancement of UV luminescence. Under low excitation levels, $W \neq 0$ leads to

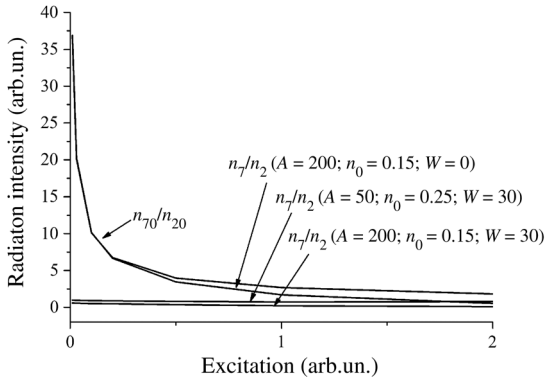

Fig. 4 Calculated dependences of relation between intensities of visible and UV radiation on excitation level.

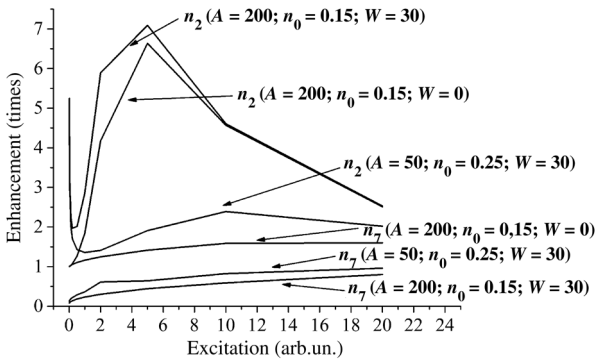

Fig. 5 Calculated dependences of UV and visible luminescence enhancement on excitation level.

sharp growth of UV luminescence enhancement. In addition, enhancement is high in the case of higher SPR parameters and smaller defect concentration but under high excitation level.

Consequently, results of the calculation qualitatively match experimental results.[3–6] But we have obtained additional information: sharp growth of the UV luminescence enhancement with simultaneous quenching of visible luminescence due to ZnO metal covering is possible only under low excitation levels.

4 Conclusion

For the analysis of the influence of SPR on the UV and visible luminescence intensity of ZnO films and the origin of visible luminescence of such films, a simplified approach was proposed. This approach is based on the SRE which allows us to calculate the dependence of radiation intensity on excitation level. The SRE includes the set of parameters that describe processes determining luminescence.

Comparison of experimental and calculated dependences of UV radiation intensity on excitation level allows estimating parameters that characterize ZnO films with or without an Ag cover. It was shown that when Ag cover thicknesses increases (from 10 to 20 nm), losses also increase.

It was found out that it is possible to make a conclusion about the visible radiation origin based on the analysis of the dependence of radiation intensity on excitation level. Namely, if visible radiation is caused by the existence of DAP in ZnO, the dependence will look like $I(P) \sim P^{1/3}$; in other cases the index of power will be higher than $1/3$ (but <1).

The proposed approach was applied for consideration of experimental results obtained in Refs. 3–6, taking into consideration their interpretation of these results based on assumptions about the transfer of electrons from the defect level in the ZnO bandgap to metal and then to the conduction band in ZnO. The results of the performed calculations using modified SRE revealed that the effects observed in these papers can exist under only low pumping levels.

In conclusion, it is worth noting that the proposed approach can be applied not only to ZnO but also to other semiconductor materials as well.

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