Optical Properties of Composite Structure Based on ZnO Microneedles and Alq3 Thin Film

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Research Article

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Abstract

The composite material based on ZnO microneedles and Alq_3 thin film has been obtained. The photoluminescence study shows a tenfold enhancement in the band-edge UV emission (390 nm) of ZnO microneedles and 2x enhancement in visible emission of the hybrid composite, when excited by 266 nm laser beam. This enhancement can be explained by the interaction between ZnO and Alq3 molecules and the energy transfer from ZnO to Alq_3 molecule.

1 Introduction

Organic light-emitting diode (OLED) technologies are at the center of attention of scientists working in the field of optoelectronics, due to their wide practical use. The serious problems of OLED based devices such as degradation of properties due to the influence of oxygen and water molecules, relatively low glass transition temperatures, low mobility of charge carriers due to the amorphous nature of solids made from organic molecules remain unsolved [1]. Besides, the problem to obtain white light is simply still actual [2]. One of the promising ways to address these problems is the use of nanocomposites to create hybrid organic-inorganic LEDs. To optimize such hybrid systems with controlled optoelectronic properties, it is necessary and important to have a deep understanding of the processes of electronic energy transfer between organic and inorganic subsystems [3].

Tris(8-hydroxyquinoline)aluminium (Alq_3) is a very stable and widely used electron-transport and light emitting material in organic optoelectronic devices [4]. This material is thermally stable, has a high glass-transition temperature of 172°C, and can easily be thermally deposited to form pin hole–free amorphous thin films due to its intrinsic polymorphic phase behavior [5]. Semiconductor materials based on ZnO are now considered as the best alternative to indium tin oxide, since they are much cheaper and non-toxic [6]. This material is also light emitting [7]. There are many companies in the world involved into production of transparent and electrically conductive ZnO-based oxides for the needs of electronics [8]. The most challenging problem of the electrooptic devices based on ZnO homojunction is the lack of stable and reliable *p*-type doping [7]. In this situation, ZnO-based LEDs usually are fabricated by combining of *n*-type ZnO with a *p*-type semiconductor other than ZnO, for example, Cu_2O, ZnTe, $SrCu_2O_2$, AlGaN, GaN or *p*-type conduction polymers [7, 9]. To our knowledge, there are quite a number of publications devoted separately to ZnO and Alq_3, while composite materials based on them have been studied by only a few research teams from India [10–15], China [16, 17], Swiss [18], Republic of Korea [19] and Japan [20]. They consider different combinations of this composite such as amorphous or polycrystalline AlQ_3 or ZnO doped with ZnO [10–12] or AlQ_3 [13, 14], composite nanowires [19], heterostructures [16, 17, 20], or AlQ_3 imbedded into porous ZnO [15, 18], and enhancement of photoluminescence intensity of AlQ_3 and ZnO has been observed.

In this paper, we report the data concerning fabrication and luminescent properties of the composite structure based on ZnO microneedles and Alq_3 thin film.

2 Experimental

The ZnO microneedles were obtained on Si (100) substrates by a solid-vapor-phase process in a horizontal tube furnace at a heating temperature of 500°C in air atmosphere using pure metallic Zn powder (> 98 % purity) [21, 22].

Alq_3 organic layers with the thickness less than 50 nm were thermally deposited in 10^{-4} Pa vacuum on Si substrates without, and, for composite structure fabrication, covered by ZnO microneedles. Alq_3 powder (99.995 % purity) was purchased from Sigma-Aldrich Corporation and purified by recrystallization. Thickness control during the process was provided by quartz crystal deposition rate controller.

The X-ray diffraction (XRD) measurements were carried out using STOE STADI P diffractometer with linear position sensitive detector in transmission Bragg–Brentano geometry (Cu $K_{\alpha 1}$ radiation at λ = 0.15406 nm, Ge (111) monochromator, detector scanning step: 0.480° 2θ, accumulation time: 320 s, 2θ angle resolution: 0.015◦, 2θ range: 10–65 deg).

The surface morphology and the local chemical composition of the samples were analyzed using a PEMMA-102-02 (SELMI, Ukraine) scanning electron microscope.

The photoluminescence (PL) spectra were measured using a portable fiber optic spectrometer AvaSpec-ULS2048L-USB2-UA-RS (Avantes BV, (Apeldoorn, Netherlands) with an input slit of 25 μm, a diffraction grating of 300 lines/mm and a resolution of 1.2 nm. The accumulation time was 1000 msec. The samples were excited by Nd:YAG laser (266 nm, max output power – 1 μJ, light spot diameter ~ 1 mm, pulse duration < 1 ns, max repetition rate – 10 kHz).

3 Results And Discussion

Up to now, various growth approaches have been used for the synthesis of Alq_3 nano- or microstructures [23–31]. Partially, different variants of vacuum evaporation of Alq_3 have resulted in the creation of fine nanowires and microcrystals with a clear hexagonal morphology [23, 26, 29]. The problem is that Alq_3 molecule has two different geometric isomers: meridional (*mer*-) and facial (*fac*-) [23]. Five crystalline phases of Alq_3, α-, β-, γ-, δ- and ε-, have been observed [23]. α- and β- phases have two *mer*-Alq_3 molecules in a unit cell, while δ-phase has four *fac*-Alq_3 per unit cell [23]. Also, the morphology of vacuum deposited organic films depends strongly on the substrate [32]. Zinc oxide nanostructure was employed as catalyst in organic syntheses and transformations [33]. ZnO microneedles can be expected to act as catalysts and nucleation centres for the growth of specific Alq_3 structures.

X-ray diffraction pattern of the initial Alq_3 powder is shown in Fig. 1. The X-ray diffraction analysis of OLEDs materials, including Alq_3, is often rather difficult, because the crystals of OLEDs materials are sometimes disordered and contaminated by other polymorphs. We compared this pattern with the XRD patterns of α-, β-, γ-, δ- and ε-Alq_3 [27, 28, 34–37]. The powder exhibits quite similar XRD pattern to that of

α-phase Alq_3. This is important to know since photoluminescence properties for different phases of Alq_3 may vary.

The morphology of the investigated ZnO microneedles and ZnO-AlQ_3 composite is presented in Fig. 2. ZnO microneedles with an average diameter of about 0.5–6 μm and height of ~ 4 μm were grown on the (100) silicon substrate. Figure 2, a confirm the hexagonal nature of grown ZnO structures. The morphology of Alq_3 film deposited on ZnO microneedles is presented in Fig. 2, b. These structures with the thickness of 50 nm seem to cover outside surfaces of ZnO microneedles.

The room-temperature PL spectrum of the ZnO microneedles (Fig. 3) consists of the two weak bands in the ultraviolet (UV) and visible regions. The UV band at 390 nm is typical for ZnO and arises due to recombination of the free excitons, bound excitons and transitions in the donor-acceptor pairs [38]. The wide green band in the range from approximately 450 nm to 650 nm is caused by defects, first of all, by uncontrolled impurities and stoichiometry defects [38].

The room-temperature PL emission spectrum of pure Alq_3 film with the same thickness as in composite film (they were obtained at the same deposition process) exhibits a characteristic green emission at around 525 nm [28, 39] when excited at 266 nm is shown in Fig. 3. Alq_3 is characterized by crystallization in polymorphism both under vapor deposition and solvent evaporation [31].

As you can see in Fig. 3, the intensity of the UV PL band of the composite is more than 10 times higher than that of ZnO microneedles, and the intensity of the band with a maximum at 525 nm is approximately twice as high than that of this band in the PL spectrum of the Alq_3 thin film. It can be seen that both bands are not relatively narrow and shoulders of both bands do not increase the intensity of each other. Our results correlate satisfactorily with the data reported in Refs. [10–12, 14]. According to that data, compared with PL of pure Alq_3 and ZnO, PL of the composite sample based on ZnO and Alq_3 has higher intensities due to the processes of energy transfer between inorganic and organic materials. According to [14], when composite material based on ZnO and Alq_3 is excited with 266 nm wavelength, both ZnO and Alq_3 molecules are excited simultaneously. The excited state energy of Alq_3 molecule can be absorbed by the luminescent quencher and then the absorbed energy may eventually be non-radiatively transferred to ZnO, giving rise to an increase in UV emission (band edge emission) of ZnO in the composite.

However, in the case of the energy transfer the luminescence intensity of component from which energy transfer occurs, has to be decreased. But, in our case, the luminescence enhancement of ZnO and Alq_3 is observed in both bands. To address the question about energy transfer between inorganic and organic counterparts in our system, excitation functions of Alq3 and the composite have been measured (see Fig. 4). They characterize the efficiency of energy transformation in the considered system, taking into account the efficiency of absorption, luminescence quantum yield, and luminescence spectrum shift. In the case of the absence of changes in quantum yield and spectral shifts, excitation functions correspond to the absorption spectra. The shape of the obtained function for pure Alq3 is similar to the reported

literature [40]. In the excitation function of the composite both components can be excited simultaneously (Fig. 4). Probability of excitations is changed over the spectrum, as a result the ratio of the enhanced emission will depend on the excitation wavelength. Since no emission of Alq3 in the region of 390 nm is observed, curve 3 in Fig. 4 belongs only to radiation of ZnO microneedles. The energy in the region of 390 nm ZnO band can be transferred to Alq3. However, comparing energy diagrams of ZnO (4.2 and 7.6 eV [41]) and Alq3 (3.2 and 5.7 eV [42]), it can be seen that conditions for energy transfer from Alq3 to ZnO UV band and even from this ZnO band to Alq3 are not appropriate. Nevertheless, the radiative energy transfer can be observed, when Alq3 absorbs UV luminescence of ZnO. But, luminescence lifetime of ZnO UV band (1.56 ns) [43] is an order of magnitude less than that of Alq3 (12 ns) [44]. As a result, this process can't be efficient. The shape of excitation function registered at 520 nm is slightly differed for composite as compared to pure Alq3 film. It means that some energy transfer from ZnO green band to Alq3 occurs. It can be noted that some difference in PL intensity of Alq3 band may also be due to the difference in Alq3 morphology (pure Alq3 film is amorphous and Alq3 in composite may form specific structures with higher quantum yield). No evidence of energy transfer from AlQ3 to ZnO is observed.

On the other side, the observed enhancement of luminescence intensity of both components under Alq_3 deposition can be caused by mutual influence of each other. It has been found that passivation of ZnO films by coatings with metal oxides [45] and doping with hydrogen [46] result in the strong UV ZnO luminescence enhancement. Alq_3 contains oxygen atoms and a lot of hydrogen atoms to promote this process. Besides, interaction of Zn ions with Alq_3 was found to enhance luminescence of Alq_3 [29]. The luminescence intensity of green band is also enhanced that can be noticed from the shape changes of Alq_3 band.

4 Conclusion

The composite material based on ZnO microneedles and Alq_3 thin film has been obtained. The photoluminescence study shows a tenfold enhancement in the band-edge UV emission (390 nm) of ZnO and doubled intensity of the visible emission in the hybrid composite, when excited by 266 nm wavelength. Probability of excitations is changed over spectrum, as a result the ratio of the enhanced emission depends strongly on the excitation wavelength. This enhancement can be explained by the interaction between ZnO and Alq3 molecules, and the energy transfer from ZnO to Alq_3 molecule. This composite is a possible candidate for the emitting layer of an OLED device.

Declarations

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Figures

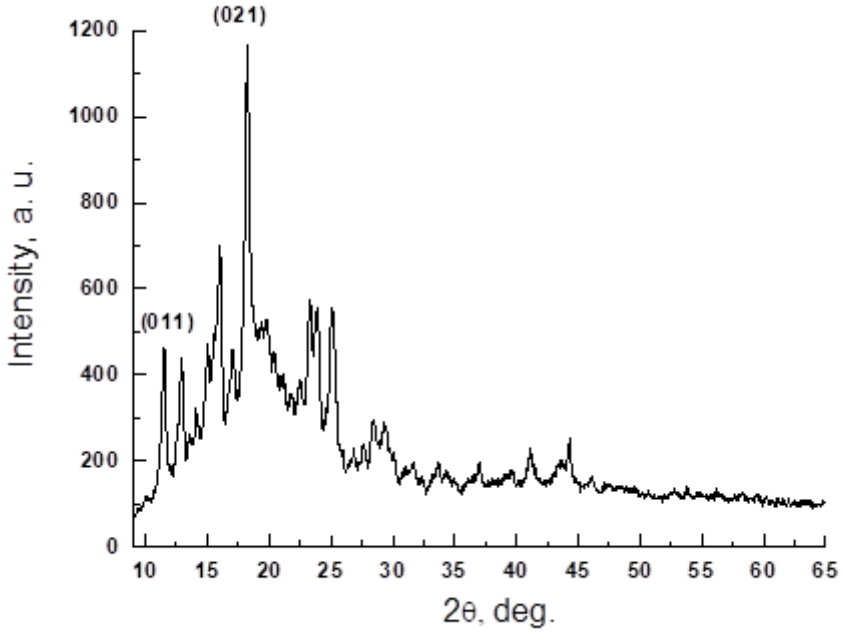

Figure 1

X-ray diffraction profile of the original Alq3 powder.

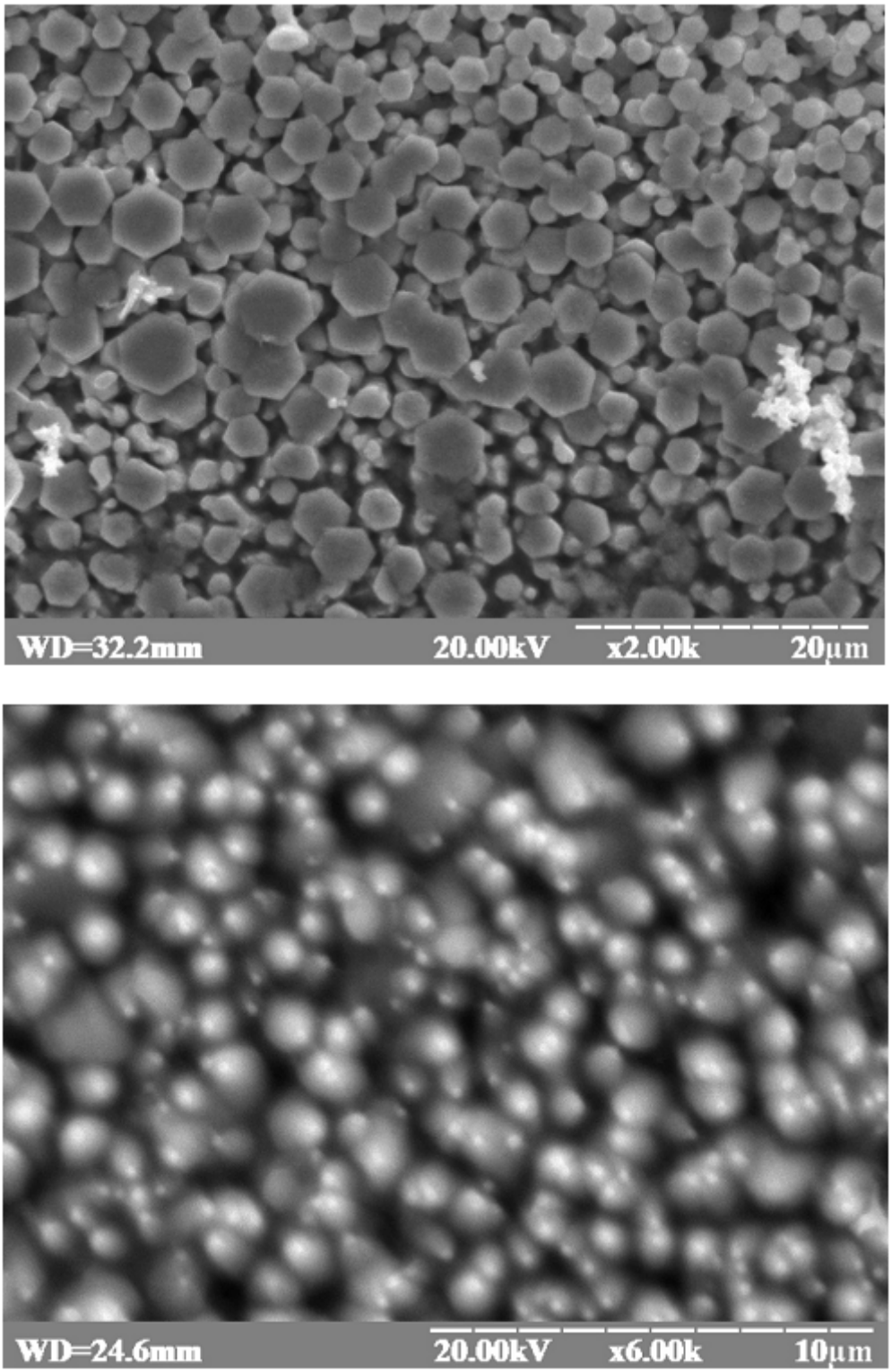

Figure 2

Microphotograph of composite structure based on ZnO microneedles (a) and Alq3 thin film (b).

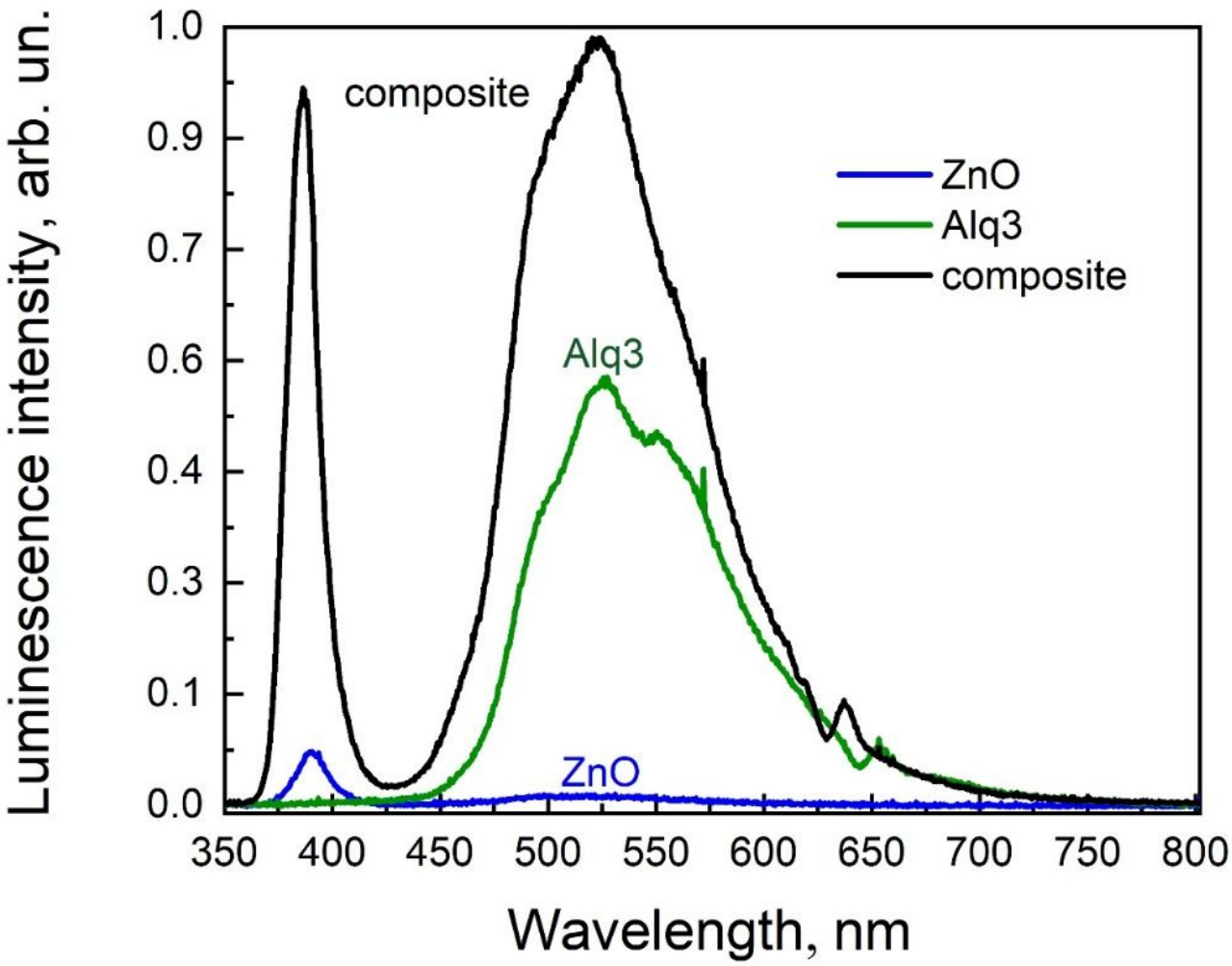

Figure 3

Room-temperature PL emission spectra of Alq3 thin film, ZnO microneedles and composite structure based on ZnO microneedles and Alq3 thin film.

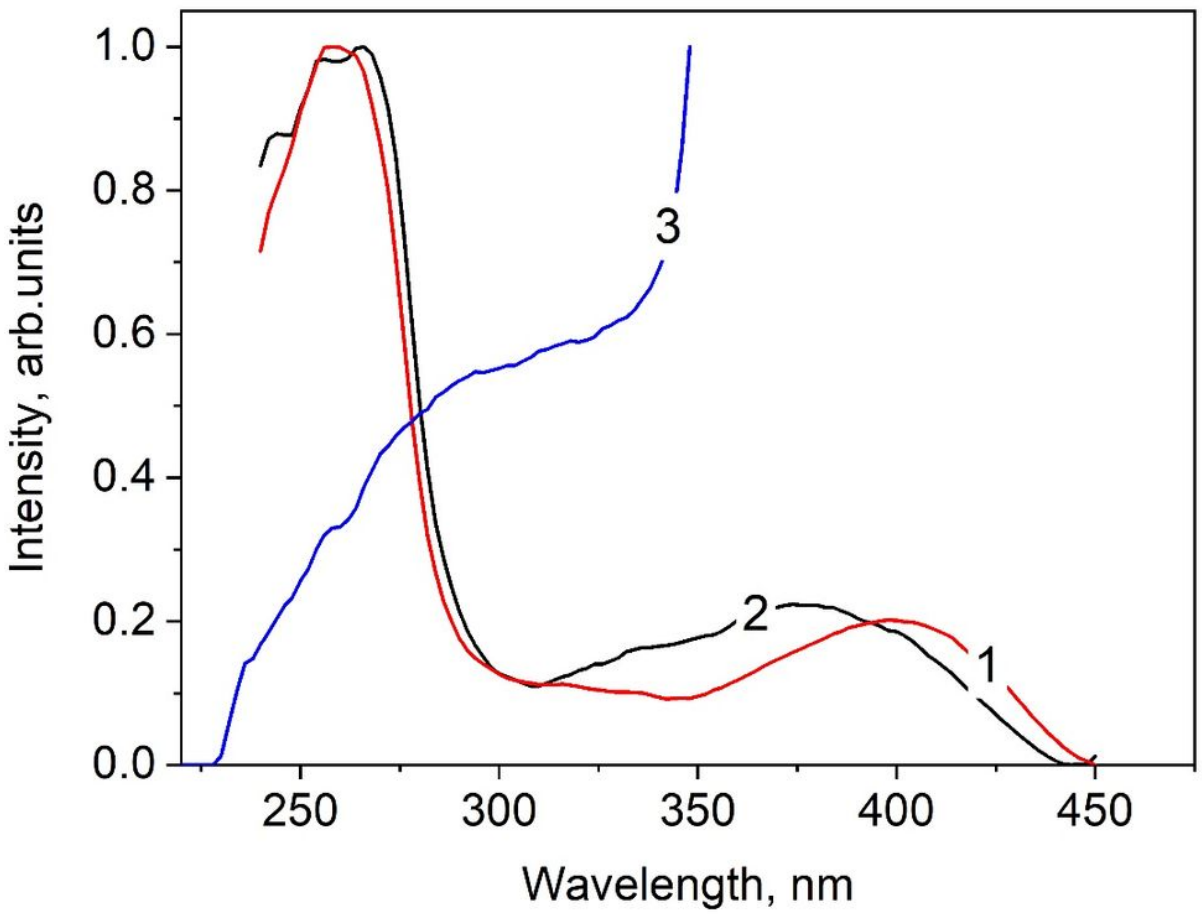

Figure 4

Normalized excitation functions of Alq3 (1) and Alq3+ZnO (2, 3) film (registration at 520 (1, 2) and 390 (3) nm).