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Luminescence of Cr-doped β-Ga_2O_3 thin films

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The results of investigation of optical excitation, photoluminescence (PL) and cathodoluminescence (CL) spectra in Cr-doped β-Ga_2O_3 thin films are presented. The broad bands due to the generation of electron-hole pairs and transitions from the $^4A_2$ ground level to the $^4T_1$ and $^4T_2$ excited states in Cr^{3+} ions are observed in the photoexcitation spectra. The R-lines and phonon repetitions of R-lines are observed on the luminescence spectra against the background of a broad structureless band with a maximum around 700 nm, caused by the $^4T_2$ – $^4A_2$ transitions in Cr^{3+} ions. The crystal field force Dq was determined and the Stokes and anti-Stokes repetitions of R-lines were interpreted.

Key words: gallium oxide, activator, thin films, photoluminescence, cathodoluminescence.

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Introduction

The study of the optical properties of the luminescence centers created by Cr^{3+} ions at the photo- and cathode excitation is of unconditional interest to researchers, as a whole number of crystal phosphors with this activator is used in practice. Among them, β-Ga_2O_3:Cr^{3+} occupies an important place as in single-crystal [1–3] so and thin-film or nanocrystalline [4–6] states. The interest in these compounds does not decrease due to the expansion of the areas of their practical application, where new requirements are put forward for the properties of phosphors. Such an area, for example, is the study of PDP (plasma, display, panel) flat screens, which are actively developing [7].

In addition, β-Ga_2O_3 is an almost ideal matrix for the introduction of Cr^{3+} ions, since it has an ionic radius of 0.62 Å, which is almost identical to the radius of the Ga^{3+} ion in octahedral coordination [8]. However, despite the large number of works, the question of the nature of such luminescence centers and the effect on them of the real structural parameters of the β-Ga_2O_3 lattice remains incompletely studied. The properties of real crystal lattices are determined both by the method and the conditions of obtaining the investigated samples. In this regard, in this work, the excitation and luminescence spectra of β-Ga_2O_3:Cr^{3+} thin films obtained by radio-frequency (HF) ion-plasma sputtering were investigation. This method is considered optimal for obtaining multicomponent dielectric and semiconductor thin films [9].

I. Experimental technique

Thin films of β-Ga_2O_3:Cr^{3+} with a thickness 0.3–0.8 μm were obtained by RF ion-plasma sputtering on substrates of υ-SiO_2 fused quartz. RF sputtering was carried out in an atmosphere of argon in the system using the magnetic field of external solenoids for compression and additional ionization of the plasma column. The raw material for the manufacture of the target was β-Ga_2O_3 with grade «ОСЧ» (the purity 99.99%). The doping impurity in form of Cr_2O_3 oxide with a concentration of 0.05 wt.% was added to the target material. After deposition of the thin films, the thermal treatment in air atmosphere at 1000-1100 °C was held. X-ray diffraction studies of the obtained thin films showed , as for undoped β-Ga_2O_3 films, the presence of a polycrystalline structure with a predominant orientation in the (400), (002), (111) and (512) planes. In more detail, the diffraction patterns for β-Ga_2O_3 thin films were described by us earlier in [10].

The investigation of excitation spectra of luminescence and photoluminescence (PL) was carried

out on a CM-2203 spectrofluorimeter with a Hamamatsu R928 recording head in the spectral range from 220 to 800 nm.

The cathodoluminescent (CL) properties were studied in the mode of pulsed electronic excitation (τ=3 μs, f = 20 Hz). The measurement of the luminescence spectra was carried out on the installation mounted on the basis of the СФ-4А spectrophotometer. The spectra were measured using a ФЭУ-79 photomultiplier, the signal from which was fed through a resonant amplifier and recorded by milliammeter, and was also transmitted through an analog-to-digital interface converter to an IBM/PC computer for recording the spectrum of luminescence. The wavelength scanning by monochromator was carried out using a stepper motor, which was controlled by a computer through a control unit. The investigation of luminescence spectra was carried out in the wavelength range from 300 to 800 nm at a temperature of 295 K.

II. Results and discussion

Our studies show that unannealed β-Ga_2O_3:Cr^{3+} thin films are characterized by weak luminescence and photoexcitation of both intrinsic and impurity luminescence. Therefore, the research was carried out on β-Ga_2O_3:Cr^{3+} thin films annealed in air atmosphere.

The revealed effect of the presence and composition of the thermal treatment atmosphere on the output of intrinsic and impurity luminescence shows the important role of crystal lattice defects in the processes of capture and recombination of charge carriers in β-Ga_2O_3:Cr^{3+} thin films. In particular, the investigation of the electrical conductivity of β-Ga_2O_3:Cr^{3+} thin films [11] shows the influence of the obtaining conditions on the concentration of shallow donor levels caused by interstitial gallium atoms and deep donor levels caused by oxygen vacancies. Based on this, Cr3+-centers with the closest environment are likely to be excited, mainly due to the migration mechanism from point defects, which perform the role of sensitization centers. Such luminescence centers can be located in places of accumulation of point defects, for example, near dislocations, grain boundaries or the surface.

Typical PL and CL spectra of β-Ga_2O_3:Cr^{3+} thin films are shown in Fig. 1. As can be seen from the obtained results, the PL and CL spectra of the obtained thin films, annealed in air atmosphere, consist of the intrinsic luminescence of the β-Ga_2O_3 matrix, which is manifested in a broad band with a maximum in the region of 400 nm (3.10 eV) and the activator luminescence of Cr^{3+}, which is manifested in the luminescence band in the red region of the spectrum with a maximum near 700 nm (1.77 eV). Characteristically, the PL spectra are dominated by intrinsic luminescence, and the CL spectra are dominated by impurity Cr^{3+} luminescence in the "red" region of the spectrum (Fig. 1). Earlier in [12–14], it was shown that the intrinsic luminescence band of β-Ga2O3 thin films is complex and contains two luminescence bands with maximum in the region of 395 and 415 nm (3.15 and 3.00 eV).

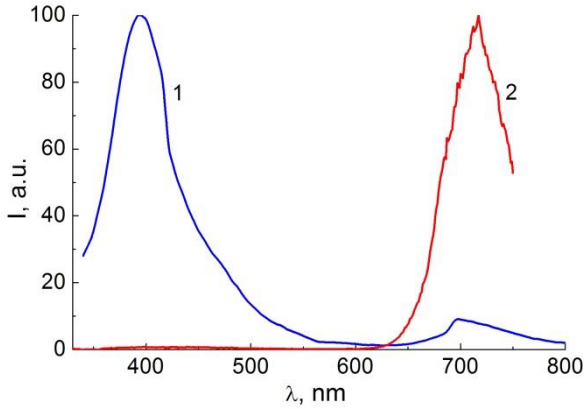

Fig. 1. The (1) PL (λ_{ex} = 240 nm) and (2) CL spectrums of β-Ga_2O_3:Cr^{3+} thin films after thermal treatment in air atmosphere, T = 295 K.

The typical excitation spectra of intrinsic and impurity luminescence of β-Ga_2O_3:Cr^{3+} thin films are shown in Fig. 2. Note that for both types of luminescence, the excitation band with a maximum in the region of 240 nm (5.15 eV) dominates, and an even more intense excitation band is observed in the short-wavelength region. In addition, two weaker excitation bands with maximums in the region of 440 nm (2.82 eV) and 600 nm (2.07 eV) appear in the photoexcitation spectra of impurity luminescence of Cr^{3+} ions. The most intense luminescence excitation band of β-Ga_2O_3:Cr^{3+} thin films with a maximum in the region of 240 nm, is due to absorption by the base lattice with the generation of electron-hole pairs [14–16]. The weakly of luminescence excitation bands are not observed at excitation of pure β-Ga_2O_3 thin films [14–16] and are associated with activator luminescence. The main term of the Cr^{3+} ion is the orbital singlet $^4A_2$ (t_2^3). The transitions $^4A_2 \rightarrow {}^4T_2$ (t_2^2 $^3T_1$,*e*) (U), $^4A_2 \rightarrow {}^4T_1$ (t_2^2 $^3T_1$,*e*) (Y) and $^4A_2 \rightarrow {}^4T_1$ (t_2,e^2 $^3A_2$) (V) in the Cr^{3+} ion give three broad bands in the absorption spectra: respectively U-, Y- and V-bands. In many crystals, including those with an octahedral environment of Cr^{3+} ions, which typical of β-Ga_2O_3:Cr^{3+} thin films [17], the third band V falls into the region of its own absorption and is not observed [18, 19]. Based on this, the two photoexcitation bands we discovered in the spectrums of β-Ga_2O_3:Cr^{3+} thin films can be associated with the U-band (with a maximum in the region of 600 nm) and the Y-band (with a maximum in the region of 440 nm).

The energy difference between the $^4T_2$ (t_2^2 *e*) and $^4A_2$ (t_2^3) levels is equal to 10 D_q [20]. In this regard, the crystal field strength Dq was determined on the basis of the first broad luminescence excitation band $^4A_2$ (t_2^3) $\rightarrow$ $^4T_2$ (t_2^2 *e*) (U-band). Calculations for β-Ga_2O_3:Cr^{3+} thin films annealed in air atmosphere give value of D_q =1667 cm^{-1}. It should be noted that the calculations of the value of D_q for single-crystal samples of β-Ga_2O_3:Cr^{3+} [21] give the value of D_q = 1680.7 cm^{-1}. As we can see, the obtained results are quite close to each other, and the slight weakening of the crystal field in β-Ga_2O_3:Cr^{3+} thin films relative to single crystal samples

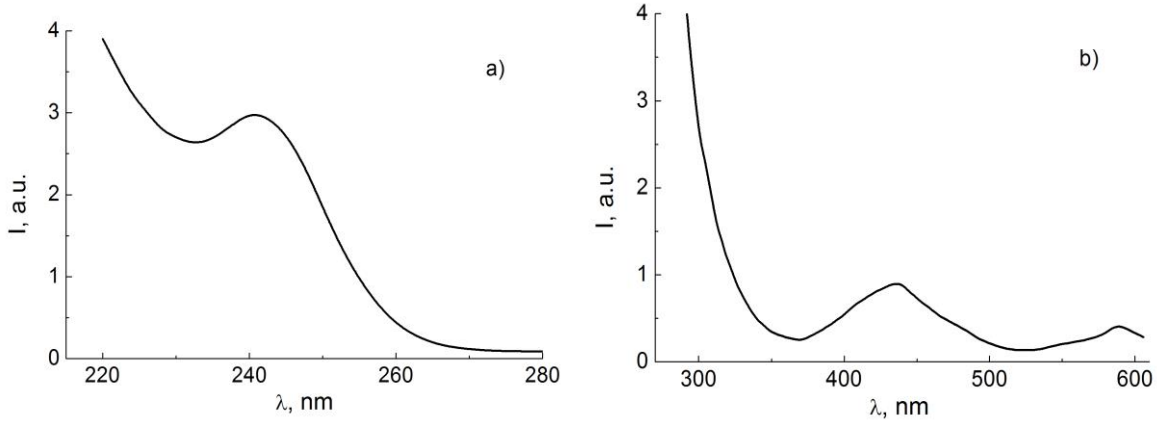

Fig. 2. The luminescence excitation spectrum of β-Ga_2O_3:Cr^{3+} thin films for light emission (a) of 400 nm and (b) 700 nm, T = 295 K.

can be explained by the presence of a larger number of oxygen vacancies in the films and the lower packing density of the films compared to single crystal samples. Such a situation is typical for films obtained under low pressure conditions, in particular, by RF sputtering [9].

Gallium oxide has a monoclinic structure and Cr^{3+} ions in the β-Ga_2O_3 structure are in an octahedral oxygen environment [17]. In such an environment with an inversion center, electric dipole transitions are forbidden by parity. Therefore, the luminescence spectrum can be caused either by the displacement of chromium ions from the inversion center, or by interaction with the vibrations of the crystal lattice. As can be seen from Fig. 3, the luminescence spectrum of β-Ga_2O_3 thin films consists of a number of lines and bands of different intensities, which are superimposed on a wide structureless band resulting from the radiative transition $^4T_2 \rightarrow {}^4A_2$ [18]. These lines and bands are caused by different electronic transitions in Cr^{3+} ions. According to [22, 23], in the spectrums of β-Ga_2O_3:Cr^{3+} at 689.7 nm and at 696.6 nm, two R-lines of different intensities are observed, caused by electronic transitions to the $^4A_2$ ground state from two sublevels of the ^{2}E-state which split in the crystal field ($\bar{E}$ i $2\bar{A}$) (so-called phononless or zero-phonon transitions). On the obtained CL spectrums of β-Ga_2O_3:Cr^{3+} thin films (Fig. 3), these bands appear as local maximums against the background of a structureless broad band caused by the $^4T_2 \rightarrow {}^4A_2$ transition. In the longer-wavelength region, from the R-lines on the background of this band, several weak lines due to phonon repetitions of the same non-phonon transitions (vibrational or vibronic bands) appear, which are shifted relative to the R-lines in the long-wavelength region of the spectrum (Stokes bands). In β-Ga_2O_3:Cr^{3+} thin films, such side phonon bands appear at 703, 708, 716, 720, 734, 745, and 756 nm.

If we take into account that the Stokes bands arise due to the exchange of part of the energy of the $^2E \rightarrow {}^4A_2$ transitions into thermal vibrations of the crystal lattice, so due to the superposition of the energy of the $^2E \rightarrow {}^4A_2$ electronic transitions with the energy of atomic vibrations, anti-Stokes phonon lines which accompanying of R-lines arise, which in β-Ga_2O_3:Cr^{3+} thin films appear at 683 nm. The typical emission bands in the luminescence spectrums of β-Ga_2O_3:Cr^{3+} thin films and their interpretation are given in table. 1. Note that the obtained results are in good agreement with the investigation of vibrational spectrums of single crystals and β-Ga_2O_3:Cr^{3+} thin films made in [24 – 27].

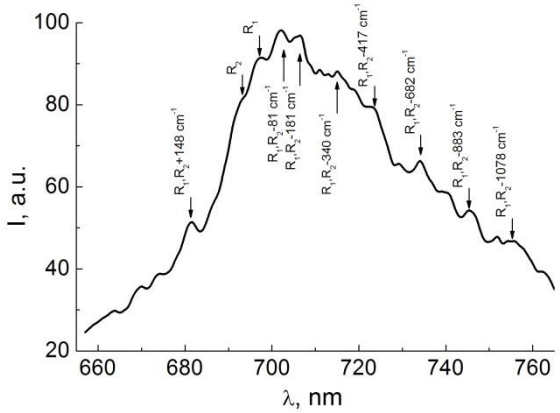

Fig. 3. The CL spectrum of activator luminescence of β-Ga_2O_3:Cr^{3+} thin films and typical emission bands.

Table 1.
The bands in the CL spectrum of β-Ga_2O_3:Cr^{3+} thin films (T = 295 K)

The maximum of emission bands, nm	Interpretation
683	R_1,R_2 + 148 cm^{-1}
690	R_2
699	R_1
703	R_1,R_2 – 81 cm^{-1}
708	R_1,R_2 – 181 cm^{-1}
716	R_1,R_2 – 340 cm^{-1}
720	R_1,R_2 – 417 cm^{-1}
734	R_1,R_2 – 682 cm^{-1}
745	R_1,R_2 – 883 cm^{-1}
756	R_1,R_2 – 1078 cm^{-1}

We also note that for the R-lines in β-Ga_2O_3:Cr^{3+} thin films, the maximums of which are located at 690 and 699 nm, there is a slight long-wave shift relative to the maximums at 689.7 and 696.6 nm, typical of single-crystal samples. According to the Tanabe-Sugano diagram [18], this situation indicates a weakening of the force of the local crystal field and is in good agreement with the force of the crystal field Dq determined by us above. At the

same time, the greater expansion of these lines in β-Ga_2O_3:Cr^{3+} thin films relative to single-crystal samples indicates an increase in the range of changes in this force. It is logical to expect a decrease in the force of the local crystal field with an increase in the number of vacancies of ligand atoms around chromium atoms and a decrease in the packing density of thin films relative to single crystal samples. In addition, as a result of the uneven distribution of such vacancies throughout the volume of the sample, an increase in the range of changes in the force of the local crystal field is observed in the thin films.

Conclusions

The made investigation show that of β-Ga_2O_3:Cr^{3+} thin films can be used as phosphors with a red emission at electronic excitation. On the basis of the photoexcitation spectrums, the force of the crystal field D_q was determined and it was shown that it is somewhat smaller than in β-Ga_2O_3:Cr^{3+} single-crystal samples. This situation is associated with a greater number of oxygen vacancies in the thin films relative to single-crystal samples. In the CL spectrums of β-Ga_2O_3:Cr^{3+} thin films, we observe a broad structureless luminescence band with a maximum near of 700 nm, which corresponds to electron-vibrational transitions of $^4T2 \rightarrow {}^4A_2$. Against the background of this band, both R-lines (E $\rightarrow$ $^4A_2$ transition) and phonon repetitions of R-lines (Stokes and anti-Stokes) were analyzed. The obtained spectrums were analyzed in connection with the force of the crystal field.

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Люмінесценція тонких плівок β-Ga_2O_3 легованих хромом

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Наведено результати досліджень спектрів оптичного збудження та свічення фотолюмінесценції (ФЛ) і катодолюмінесценції (КЛ) в тонких плівках β-Ga_2O_3, легованих хромом. У спектрах фотозбудження спостерігаються широкі смуги, зумовлені генерацією електронно-діркових пар та переходами з основного рівня $^4A_2$ на збуджені стани $^4T_1$ та $^4T_2$ в іонах Cr^{3+}. На спектрах люмінесценції на фоні широкої безструктурної смуги з максимумом в околі 700 нм, зумовленої переходами $^4T_2$ – $^4A_2$ в іонах Cr^{3+}, спостерігаються R-лінії та фононні повторення R-ліній. Проведено визначення сили кристалічного поля Dq та інтерпретацію стоксових та антистоксових повторень R-ліній.

Ключові слова: оксид галію, активатор, тонка плівка, фотолюмінесценція, катодолюмінесценція.