IR luminescence of Fe²⁺: ZnSe single crystals excited by an electron beam

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Abstract. Spectral-kinetic characteristics of the cathodoluminescence of Fe^{2+} : ZnSe single crystals are studied at T = 300 and 80 K. These characteristics correspond to the IR luminescence spectrum and kinetics of Fe^{2+} ions in ZnSe crystals optically excited into the impurity absorption band. The obtained results open a real possibility of pumping Fe^{2+} : ZnSe lasers by hot electrons.

Keywords: Fe²⁺: ZnSe crystals, cathodoluminescence.

Iron-doped ZnSe crystals (Fe²⁺:ZnSe) are used as active media of tunable mid-IR lasers (4–5 μ m) [1–5]. The high gain coefficient [2] makes it possible to develop low-threshold [2] and high-power [5, 6] Fe²⁺:ZnSe lasers operating in both cw [7] and repetitively pulsed [5] regimes. A conventional optical pump source for Fe²⁺:ZnSe lasers is an Er:YAG laser operating at the wavelength $\lambda = 2.94 \mu$ m [8], which is widely used in scientific laboratories and medicine. High pulse energies of Fe²⁺:ZnSe lasers were achieved under pumping by HF lasers [3, 6].

Active ions in doped semiconductor matrices can be excited by hot electrons accelerated by an electric field (electron-impact excitation and ionisation [9]). These processes have been extensively studied in order to design electroluminescent light sources [10]. The creation of inverse population between the activator levels in a semiconductor by applying a voltage pulse to the active laser element is an important and challenging scientific and engineering problem, because this pumping method provides a high electricity-to-light conversion efficiency and makes it possible to achieve high gains [11].

As applied to the Fe^{2+} : ZnSe laser crystals, the first step on the way to realisation of impact excitation of dopant ions can be the observation and study of the IR (4–5 µm) luminescence of Fe^{2+} ions in ZnSe under action of a high-energy electron beam, which is the aim of the present work.

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Received 27 April 2016 *Kvantovaya Elektronika* **46** (6) 545–547 (2016) Translated by M.N. Basieva The Fe²⁺ impurity was introduced into the ZnSe crystal simultaneously from both sides by the diffusion method under the thermodynamic phase equilibrium conditions. Then, the crystal was polished so that the layers removed from both sides were identical. The thickness of the layer enriched with Fe²⁺ with the average concentration $C \approx 10^{19}$ cm⁻³ was 100 µm.

The cathodoluminescence of the crystals was excited by a pulsed electron gun with an accelerated electron energy of 36 keV and an average electron penetration depth of ~3 μ m. The pulsed electron beam current varied from 0.01 to 0.2 A, the beam diameter was from 1.5 to 10 mm, and the average electron-hole pair generation rate in the excited layer changed from 10²⁵ to 10²⁷ pairs per second per cm³. The luminescence was excited and observed at an angle of 45° to the sample surface. The cathodoluminescence radiation was coupled out of the chamber through a CaF₂ window and focused on the input slit of an MS2004 (SOL Instruments Ltd) monochromator. As a detector, we used a PD42NB (IoffeLED Ltd) diode with a spectral sensitivity within the range 3.15–4.75 μ m. A signal from the photodetector was sent to the input of a digital oscilloscope directly or after passing through a broadband USh-10 amplifier.

The luminescence spectrum of a Fe^{2+} : ZnSe single crystal excited by an electron beam at T = 80 K is shown in Fig. 1. It corresponds to the emission spectra of Fe^{2+} ions in ZnSe recorded previously by other authors upon optical excitation of the crystal into the impurity absorption band [12]; in par-



Figure 1. Normalised luminescence spectrum of Fe^{2+} : ZnSe crystal under electron-beam excitation at T = 80 K (points correspond to the experimental data).

ticular, the spectra in both cases have identical positions of luminescence maxima (near $3.9 \,\mu$ m), as well as the short- and long-wavelength edges.

The IR photoluminescence kinetics of Fe^{2+} : ZnSe crystals was studied by several scientific groups at different times [1, 12, 13]. The lifetime of the ${}^{5}T_{2} \rightarrow {}^{5}E$ laser transition is characterised by a strong temperature dependence and varies from 370 ns at room temperature to 100 µs at liquid nitrogen temperatures [1, 12, 13]. In addition, a pronounced dependence of the lifetime on the thickness of the active iron layer was observed in [12].

We measured the luminescence decay kinetics of the Fe²⁺: ZnSe crystal under electron-beam excitation. An electron-beam pulse with a duration $t_0 \approx 100$ ns had a rectangular shape at a current $I \approx 30$ mA and a voltage $U \approx 40$ kV. Figure 2a shows the time dependence of the luminescence signal of the Fe²⁺: ZnSe crystal under electron-beam excitation (T = 80 K). The luminescence kinetics is non-exponential, which is seen from the logarithmic time dependence of the signal intensity shown in Fig. 2b. At times exceeding 25 µs, the luminescence signal can be described by the function $\exp(-t/\tau)$ with the characteristic time $\tau = 35-40$ µs, which is shown by the straight line in Fig. 2b. This time agrees with the luminescence decay time observed previously upon optical excitation of iron ions in the ZnSe crystal at liquid nitrogen temperature $(40-100 \ \mu s \ [12, 13])$. In general, the decay curve is well described by the dependence $I_0(1 + t/\tau)^{-1}$ with $\tau =$ $2.5 \,\mu s$, which is typical for the case when the probability of emission of a luminescence photon is proportional to the concentration of excited centres.



Figure 2. (a) Time dependence of the Fe^{2+} : ZnSe luminescence intensity normalised to the maximum I/I_0 under electron-beam excitation at T = 80 K. (b) The same dependence on the logarithmic scale.

Figure 3 presents the time dependences of the luminescence signal intensity and its logarithm at room temperature. The luminescence kinetics is well described by the function $\exp(-t/\tau)$ with $\tau = 250$ ns (Fig. 3b). This time is close to the luminescence decay time of Fe²⁺: ZnSe at room temperature upon optical excitation of iron ions (355 ns [14]).



Figure 3. (a) Time dependence of the Fe^{2+} : ZnSe luminescence intensity normalised to the maximum I/I_0 at room temperature. (b) The same dependence on the logarithmic scale.

Thus, we observed the luminescence of Fe^{2+} : ZnSe crystals in the IR spectral region under electron-impact excitation. The luminescence maximum at liquid nitrogen temperature lies near 3.9 µm. The luminescence kinetics is recorded at liquid nitrogen and room temperatures. It is found that the luminescence kinetics at liquid nitrogen temperature has a nonexponential character, especially at the initial stages. The luminescence decay at room temperature is described by an exponential function with a time of 250 ns, which is close to the time reported in the literature for optical excitation of the luminescence. The obtained results allow us to conclude that the method of pumping Fe^{2+} : ZnSe IR lasers by electric current pulses is really promising.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant Nos 16-02-00741a, 15-59-31817 RT-omi, and 15-52-45024 IND-a), by the Programme for the Support of Leading Scientific Schools of the Russian Federation (Grant No. NSh-451.2014.2), and by the Programme of the Presidium of the Russian Academy of Sciences 'Fundamentals of Innovative Dual-Purpose Technologies for Homeland Defence' (Project 'Investigation of the possibility of creating high-efficiency IR (4–5 μ m) lasers based on iron-doped ZnSe crystals with hot-electron-impact excitation').

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