Middle infrared Fe$^{2+}$:ZnS, Fe$^{2+}$:ZnSe and Cr$^{2+}$:CdSe lasers: new results

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Abstract. The output energy of 10.6 and 3.25 J with a pulse duration of ~1 ms and a crystal temperature of 85 K was achieved in the Fe:ZnSe and Fe:ZnS lasers, respectively. At room temperature, the Fe:ZnSe laser energy was as high as 1.2 J with a pulse duration of 150 ns. A few optical schemas of the Cr:CdSe laser with laser diode pumping were realized.

1. Introduction
Lasers of middle infrared (mid IR) spectral range are perspective for various applications in laser location of objects in atmosphere, high resolution spectroscopy, metrology, monitoring of pollution of the environment, medicine. To date the works directed to increasing an average power up to 10 W and pulse energy up to 1 J or higher in the 4-5 $\mu$m range are of the greatest interest.

Among well-known lasers of this spectral range it is worth to mark out chemical DF lasers (toxic, bulky, ineffective at $\lambda > 4 \mu$m), quantum cascade lasers (high level technology, problem with obtaining high energy pulse) and optical parametric oscillators (unreliable as yet). In recent years, the lasers based on II-VI compound crystals doped Cr and Fe, which are free from disadvantages of mentioned lasers, are intensively developed.

To date the following results are achieved. At previous symposium, we reported on obtaining of 2.1 J in the pulsed Fe$^{2+}$:ZnSe laser at liquid nitrogen cooling of the active crystal pumped by an Er:YAG laser [1]. An average power of such a laser operating in a repetitively pulsed mode with cryogenic cooling of the crystal was increased up to 35 W [2]. The maximum energy of 253 mJ was obtained at room temperature of the crystal pumped by a HF laser [3].

In this talk, we report new results obtained by our team and other research groups using ZnSe:Fe crystals grown by our team in P.N. Lebedev Physical Institute. Also we present several optical schemas of Cr$^{2+}$:CdSe lasers pumped by laser diodes. We hope that the Cr:CdSe laser may be used as a pump for the Fe:ZnSe laser in future.

2. High energy pulsed Fe:ZnSe and Fe:ZnS lasers at pumping by Er:YAG laser radiation
Figure 1 presents the schema of the experimental setup for studying Fe:ZnS and Fe:ZnSe lasers upon pumping by a free-running flashlamp-pumped Er:YAG laser. This schema is described in detail in [1, 4].

![Figure 1. Schema of the experimental setup for studying Fe:ZnSe and Fe:ZnS lasers. PD is photodiode, BS is beam splitter, HR is high reflective mirror, OC is output coupler.](image)

The main results are presented in figure 2a. The most experiments were fulfilled at the pump pulse duration of 0.9 ms and the pump energy of 15 J [4]. The maximum output energy of the Fe:ZnSe laser was 4.9 J at the crystal temperature of 85 K and 1.3 J at $T = 245$ K. The last temperature may be controlled by a two-step thermoelectric cooler. The energy of 3.25 J was achieved with the ZnS:Fe crystal. Recently we were able to increase the pump energy up to 30 J at a little longer pump pulse duration of 1.3 ms. However we succeeded in increasing pulse energy of the Fe:ZnSe laser only by using a larger active crystal with smaller Fe concentration ($10^{15}$ cm$^{-3}$). At that case, a problem of inversion dumping by amplified spontaneous noise at large lateral size of pump beam spot (up to 16

![Figure 2. Dependences of the output energy of the pulsed Fe:ZnSe/ZnS laser on the absorbed pump energy at using of different active elements (a) and a ZnSe:Fe boule used in the last experiments with an active element length ($h$) of 17 mm (b). Crystal temperature and slope efficiency are indicated in figure 2a also.](image)
mm in diameter in our last experiments) was solved. The active element was fabricated from a boule presented in figure 2b. The maximum energy of the Fe:ZnSe laser was as high as 10.6 J at $T = 85$ K while the slope efficiency was 53% relative to absorbed input energy. In the near future, we hope to obtain a pulse energy of 5 J or higher with a thermoelectric cooler.

3. Fe:ZnSe laser with pumping by HF laser radiation
Pulse-periodic operation mode of the Fe:ZnSe laser was studied at pumping by an electric discharge HF laser in the work [5]. The main results of this study are presented in figures 3 and 4. The HF laser operated in the pulse periodic regime with a repetition rate of 100 and 200 Hz for a train with duration of 1 s. The crystal was at room temperature. Each pulse in the train had duration of about 100 ns at half maximum (see figure 3a). The pump energy was as high as 75 mJ. The laser pulse contained relaxation spikes and was delayed with respect to the pump pulse. This delay decreased with increasing pump energy (figure 3b).

![Figure 3](image1.png)  
Figure 3. Oscillograms of the pulses of the HF pump laser (1) and Fe:ZnSe laser (2) at pump pulse energies of 18.1, 23.8, and 58.7 mJ (a), as well as dependence of the laser pulse delay with respect to the pump pulse at a level of 0.1 of the intensity on the absorbed pump energy (b) (the points and the curve correspond to experiment and calculation, respectively).

![Figure 4](image2.png)  
Figure 4. Changes in the laser pulse energy (a, b) from pulse to pulse (each point is an individual pulse) in 1-s trains with different pulse repetition rates 100 (a) and 200 (b) Hz. Near- (insert a) and far-field (insert b) patterns of the Fe:ZnSe laser radiation.
The energy of an individual pulse was about 14 mJ at an absorbed energy of 58 mJ and weak
changed from pulse to pulse in the train at the repetition rate 100 Hz (figure 4a). At 200 Hz, a small
drop of the output energy of an individual pulse was observed because of crystal heating (figure 4b).
The average train power was 1.4 and 2.4 W at pulse repetition rates of 100 and 200 Hz, respectively.
The total train energies were, respectively, 1.4 and 2.4 J. Lateral distribution in the near-field pattern
was elliptic that was responsive to a form of pump spot with the average lateral size of 2.2 mm. The
total divergence angle at a level of 1/e of the maximum intensity was 3.8 mrad.

To achieve high pulse energy at room temperature of the crystal, the high energy pulsed HF laser
was used in work [6]. The maximum energy was obtained as high as 1.2 J at the input pulse energy of
4.8 J and the pulse duration of 150 ns (at half maximum).

4. Amplifier of nanosecond pulses based on ZnSe:Fe crystals
First experiments concerning amplification of nanosecond pulses in the ZnSe:Fe crystals were carried
out. In future we plan to develop this work to obtain high power femtosecond pulses [7]. A schema of
an experimental setup consisting of a master oscillator and an amplifier is presented in figure 5a.

![Figure 5](image)

**Figure 5.** Schema of a setup for studying amplification of nanosecond pulses in the ZnSe:Fe crystal
(a) and output – input characteristics of the amplifier for two different ZnSe:Fe crystals (b). AE is
active element, PM is power meter, PR is photoresistance, SM is spherical mirror.

The passively Q-switched Er:YAG flashlamp laser was used by us to pump the ZnSe:Fe crystals. It
generates laser pulses with duration of 40 ns and energy of 30 mJ. The incident pump beam was
divided by a beam splitter into two beams that were focused by a spherical mirror (radius of curvature
is 150 cm) in the ZnSe:Fe crystals which were placed in both the oscillator and the amplifier. The
pump energy directed to the oscillator and amplifier were 7 mJ and 23 mJ, respectively.

The master oscillator cavity was formed by a spherical mirror (radius of curvature is 50 cm) and a
well-polished end face of the ZnSe:Fe crystal (without any antireflective coating). The laser was
operated at room temperature with a central wavelength of 4.4 μm. The seed pulse from the oscillator
was focused into the ZnSe:Fe crystal using the spherical mirror with a curvature radius of 100 cm. The
FWHM pulse duration was 15 ns, and the energy was up to 360 μJ.

To obtain gain parameters we tested two ZnSe:Fe crystals with antireflection coating on a central
wavelength of 4.4 μm and the different doping of divalent iron ions in a ZnSe matrix. The samples
were cut into rectangular slabs of 5 × 5 mm² cross sections, the first one with a thickness of 4.5 mm
and $2.5 \times 10^{18}$ cm$^{-3}$ doping, and the second one with a thickness of 8 mm and $4.2 \times 10^{18}$ cm$^{-3}$ doping.
The diameters of the seed beam and pump beam were about 1.2 mm and 2 mm, respectively. The
remaining part of the pump was returned into the active medium.
Comparison of dependences of the output pulse energy on the seed pulse energy for two crystals was presented in figure 5b. The maximal 2.7 times amplified pulse was about 1 mJ for the crystal with Fe concentration of $4.3 \times 10^{18}$ cm$^{-3}$.

5. Cr: CdSe laser with pumping by laser diodes

High energy characteristics of the Fe:ZnSe laser described above was achieved using the bulky HF laser and flashlamp Er:YAG laser. For practical uses, a laser version with pumping by solid-state laser radiation, which in turn is pumped by laser diodes, is preferable one. It is desirable also that the all elements of a laser system operate at room temperature. At that case, the ZnSe:Fe crystal should be pumped by a short pulse train. There are some problems to create a repetitively pulsed Er-laser ($\lambda \approx 3$ μm) with nanosecond pulse duration and pumping by laser diodes. But several successful versions of a similar laser based on Tm ions were already developed. Unfortunately Tm-lasers emit at a wavelength close to 1.9 μm and cannot directly be used as a pump for the Fe:ZnSe laser. We offered to convert a radiation with wavelength of 1.9 μm to 2.9-3 μm using a laser based on a CdSe:Cr crystal [8]. A Tm-fibre laser was used for pumping of the Cr: CdSe laser. Recently we were able to start up the Cr:CdSe laser directly pumped by a fibre-coupled bar of laser diodes with radiation wavelength of 1.94 μm [9]. However both these schemas do not allow to create high power nanosecond pulses for pumping of the Fe:ZnSe laser at room temperature.

High enough power pulses may be obtained by a laser-diode pumped solid-state Tm-laser. One of possible schemas of the Cr:CdSe laser pumped by a Tm:YAP laser radiation ($\lambda = 1.99$ μm) realized in a work [10] with our crystals is presented in figure 6a. A Fabry-Perot etalon and an interference-

![Figure 6](image)

**Figure 6.** Schematic of the experimental setup (a), spectrum of laser radiation and transmission spectra of methane, ethane and water vapors (b) and dependences of the laser pulse energy on the pump energy at two values of laser wavelength: 2.8 and 3.3 μm (c).

polarization filter were placed inside a cavity to control spectrum near wavelength of 2.8 μm or 3.3
μm. An example of such a laser spectrum is presented in figure 6b. Here the spectra of absorption by methane, ethane and water vapours are shown also. It is seen that we can turn the laser spectrum such a manner that two of the most intense laser lines will be matched with the ethane and methane absorption lines, 3336.8 and 3334.3 nm, respectively. While the third intense line at 3331.8 nm not coinciding with any lines of ethane, methane or water may be taken as a reference line. Such a laser is of independent interest of application as a lidar.

Figure 6c presents dependences of the pulse energy of the Cr:CdSe laser on the pump energy at two values of the laser wavelength. At pulse duration of 100 ns and repetition rate of 2-5 kHz, the maximum energy was 0.65 and 0.4 mJ at wavelength of 2.8 and 3.3 μm, respectively.

For pumping of the Cr:CdSe laser was also used a repetitively pulsed Tm$^{3+}$:Lu$_2$O$_3$ ceramic laser with radiation at a wavelength of 2.066 μm [11]. At that case, lasing was realized at a wavelength of 2.92 μm with the parameters of output pulse energy, pulse duration and repetition rate being close to ones described above. We plan to continue this work to increase an energy in individual pulses of the Cr:CdSe laser and use a pulsed train for pumping the Fe:ZnSe laser.

Acknowledgements
This work was supported by Competitiveness Program of National Research Nuclear University MEPhI.

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