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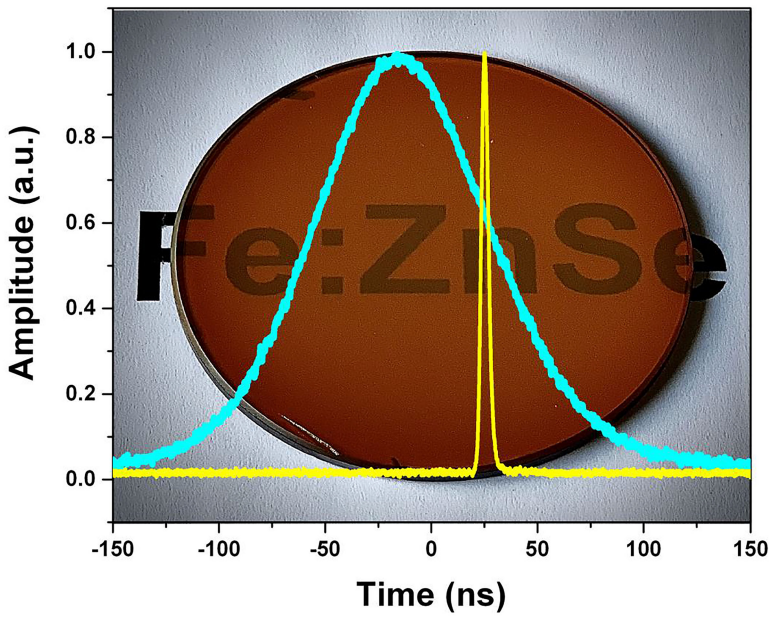

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Abstract: We investigated room-temperature pulsed lasing in heavily doped Fe:ZnSe single crystals. The active elements were pumped by a Q-switched Cr^{3+}:Yb^{3+}:Ho^{3+}:YSGG laser operating at 2.87 μm. Our results show that the generation of short laser pulses has a deep high-frequency modulation associated with relaxation dynamics in Fe:ZnSe. The lasing regime obtained in this study provides a straightforward way to generate mid-IR single nanosecond pulses at moderate pump energies. Moreover, we found a relation between the lasing pulse duration and the concentration of Fe^{2+} doping ions, and we experimentally demonstrated pulse shortening in heavily doped active crystals. Single-pulse lasing with an FWHM pulse duration of $\sim$2.8 ns was achieved in ZnSe crystals doped with $2.3 \cdot 10^{19}$ cm^{-3} Fe^{2+} ions. The demonstrated single-pulse lasing regime is applicable for seeding high-power mid-IR laser systems.

Index Terms: Mid-infrared radiation, Fe:ZnSe laser, nanosecond pulse.

1. Introduction

Lasers based on optically pumped Fe:ZnSe crystals have been studied for many years [1]–[26]. These solid-state sources allow for the generation of powerful coherent radiation in the spectral range of 4–5 μm, which is important for several practical applications (molecular spectroscopy, physical experiments at high radiation flux densities, environmental monitoring, medical applications, etc.). Fe:ZnSe and other active media based on chalcogenides doped with transition metal ions are characterized by a broad gain bandwidth [7], [8]. This property allows the generation of

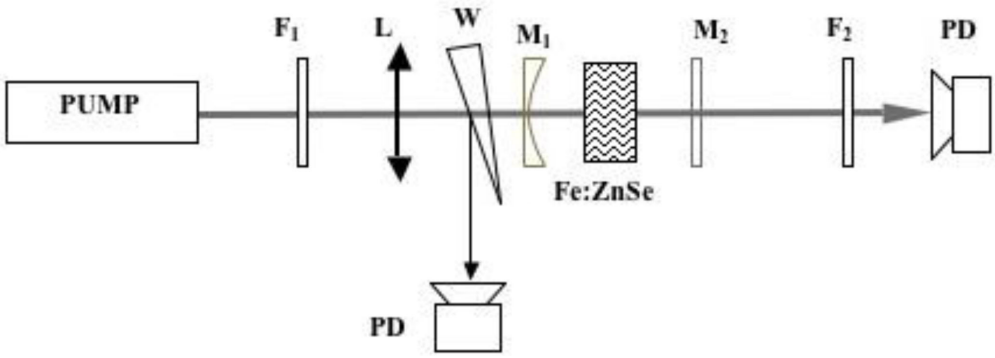

Fig. 1. The scheme of the experimental setup: M_1, M_2—cavity mirrors of the Fe:ZnSe laser; F_1, F_2—optical filters; L—a spherical lens; W—an optical wedge; PD—photodetectors; Fe:ZnSe—the active element; PUMP—a Cr^3:Yb^{3+}:Ho^{3+}: YSGG Q-switched laser.

femtosecond pulses in the mid-IR range. In the spectral range of 2–3 μm, this problem has been solved using Cr:ZnSe, Cr:ZnS, and Cr:CdSe crystals as active elements [7], [8]. Recently, possible methods of generating and amplifying femtosecond pulses in the spectral region of 4–5 μm have also been actively studied in Fe:ZnSe crystals [15], [16]. As noted by the authors of [8], powerful nanosecond pulses centered at 4–5 μm are also of interest for various practical applications. In [8], 5-ns pulses were generated in a Fe:ZnSe laser at room temperature upon excitation of the active element with a Cr:ZnSe short-pulse laser.

Here, we report the room-temperature generation of single few-nanosecond-long pulses in heavily doped Fe:ZnSe [17] pumped with a Q-switched solid-state laser. The demonstrated lasing regime opens up possibilities for developing novel master oscillators for high-power mid-IR laser systems.

2. Experimental Setup

The study was carried out on a single Fe:ZnSe crystal with a concentration of dopant Fe^{2+} ions of $n_t = 2.3 \cdot 10^{19}$ cm^{-3}. The thickness of the active element (active medium length) was 2.5 mm. For comparison, we also studied the lasing properties of a Fe:ZnSe active element with a thickness of 5 mm and a lower dopant concentration ($n_t = 6.4 \cdot 10^{18}$ cm^{-3}). Both crystals were grown from the melt by the vertical Bridgman method and AR-coated at wavelengths of 2.7–4.8 μm.

Figure 1 provides an optical scheme of the experimental setup. The laser cavity was 10.5 cm long and formed by a concave dichroic mirror, M_1 (the reflection coefficient was $\rho = 98\%$ in the range $\lambda = 4$–5 μm, and the transmission coefficient was T = 90% at $\lambda_p = 2.87$ μm), with a curvature radius of 50 cm and a flat output mirror, M_2 ($\rho = 60\%$ at $\lambda = 4$–5 μm). An active element was installed perpendicular to the optical axis at a distance of 5 cm from the output mirror. The Cr^{3+}:Yb^{3+}:Ho^{3+}:YSGG acousto-optically Q-switched laser was used as a pump source providing 110 ns (FWHM) pulses with a maximum energy of 6.5 mJ with a repetition rate of 2 Hz at 2.87 μm. The Fe:ZnSe crystal was pumped at an angle of $\sim 3°$ to monitor the pump power transmitted through the crystal. The pump beam diameter on the crystal surface was 650 μm (corresponding to the $1/e^2$ level). The pump radiation was focused by a spherical lens L (f = 210 mm). A calibrated filter F_1 was used to attenuate the pump radiation. F_2 is an interference filter that cuts off the pump radiation (the transmittance at 2.87 μm $\sim$ 5%).

The temporal profiles of the pump and the Fe:ZnSe laser power were obtained using a fast photodetector (response time 1.5 ns, Vigo System Ltd.) connected to a 300-MHz digital storage oscilloscope. The output pulse energy was measured by Molectron calorimeters. The pulse-to-pulse energy fluctuation was less than 15% for both the pump and Fe:ZnSe lasers. We used the

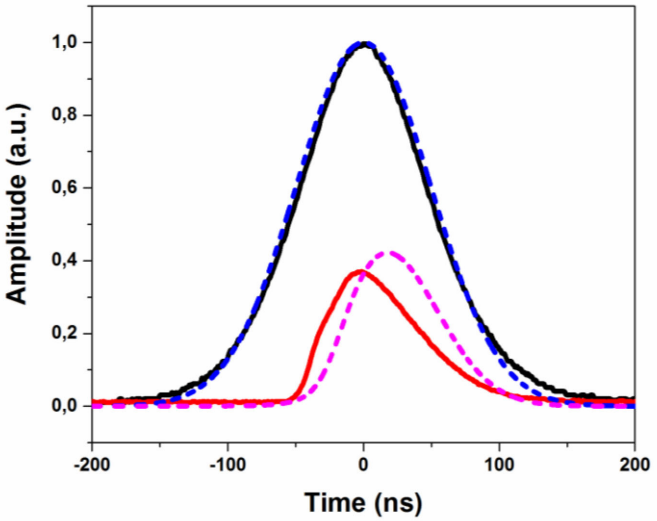

Fig. 2. The waveforms of the incident laser pulses (black and blue) and the pulses transmitted through the Fe:ZnSe crystal pump (red and magenta; the Fe^{2+} ion concentration is $n_t = 2.3 \cdot 10^{19}$ cm^{-3}). Solid lines—experiment, dashed lines—simulation.

calorimeters for the pulse energy measurement, which were installed in place of photodetectors. Each measurement was an average of 20 pulses. All the experiments were carried out with the active elements at room temperature.

3. Results and Discussion

The heavily doped single crystal used in this work is not transparent (the absorption coefficient is 22.3 cm^{-1}) for the pump wavelength (2.87 μm) at low radiation intensities. A simple estimate from the absorption cross section in Fe:ZnSe [7] shows that the low-intensity radiation's penetration depth into the crystal does not exceed 450 μm. The solid lines in Figure 2 show the waveforms of the incident ($W_{inc} = 2.7$mJ) and transmitted pump power through the crystal. The waveforms were obtained at the direct incidence of the Cr^{3+}:Yb^{3+}:Ho^{3+}:YSGG laser radiation on the crystal surface; the cavity was absent. As shown in Figure 2, the transmitted pump pulse was delayed by 60 ns with respect to the input pulse. The delay decreases with increasing pump energy, while the transmission of the crystal increases. Let us consider pump-induced photobleaching in the Fe:ZnSe crystal in detail.

To simulate the photobleaching of the active element numerically, we employ the approach described in [19] and [27]. We assume a pump pulse with Gaussian temporal and spatial profiles. Namely, the intensity of the pump pulse had the form of

$$I_{in}(r,t) \sim exp[-4(ln(2))\,t^2/\tau_p{}^2]\,exp[-2r^2/r_0{}^2], \quad (1)$$

where $\tau_p = 110$ ns is the FWHM pump pulse duration and $r_0 = 325$ μm is the $1/e^2$ beam radius. The crystal transmission over time was determined by numerically solving the equation

$$d/dt\,(ln(T)) = \sigma_{abs}\,(W_0/h\nu_p)\,/(\pi^{3/2}\cdot r_0{}^2\cdot\tau_p)\,exp[-(t/\tau_p)^2]\,exp[-(r^2/r_0{}^2)]\,(1-T) - 1/\tau\,ln(T/T_0) \quad (2)$$

where $T = T(r, t)$ is the transmittance, $T_0 = exp[-\sigma_{abs}\cdot n_t\cdot l]$ is the initial transmittance, $\sigma_{abs} = 0.97\cdot10^{-18}$ cm^2 is the absorption cross section at room temperature at a wavelength of $\lambda_p = 2.87$ μm, n_t is the Fe^{2+} ion concentration, $\tau = 265$ ns is the upper-state lifetime (measured similarly to [17]), $l = 2.5$ mm is the crystal thickness, h is Planck's constant, ν_p is the pump radiation frequency, and W_0 is the pump radiation energy. The output power over time was determined by

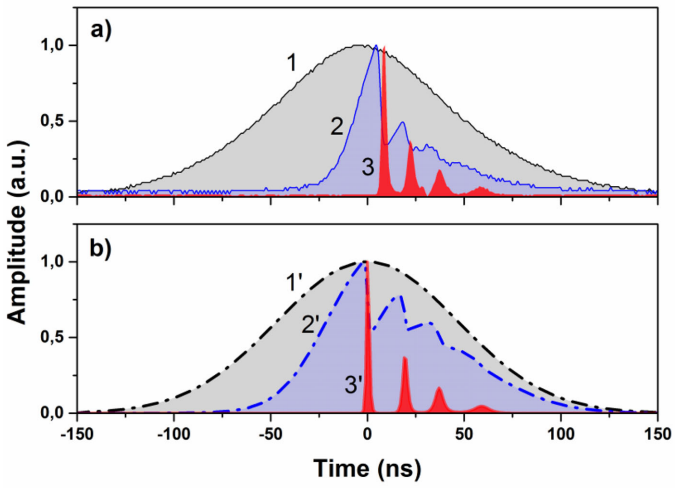

Fig. 3. (a) experimental waveforms of the incident pump pulses (1), transmitted pump pulses (2), and Fe:ZnSe lasing pulses (3); (b) The simulated waveforms of the incident pump pulses (1'), transmitted pump pulses (2'), and Fe:ZnSe lasing pulses (3').

integrating the transmitted pump pulse intensity $I_{out}(r,t) = T(r,t)\, I_{in}(r,t)$ over the beam cross section:

$$P_{out}(t) = \int_S [I_{out}(r,t)\, dS] \tag{3}$$

Our experiments on the transmission of pump radiation and the numerical simulation of this process demonstrate the possibility of obtaining lasing in heavily doped Fe:ZnSe single crystals at pump intensities sufficient for photobleaching.

Experimentally, we achieved lasing in the studied Fe:ZnSe crystal at a pump energy of 2.5 mJ. Figure 3(a) shows the experimental waveforms of the pump pulse (1), the pump pulse transmitted through the crystal (2), and the pulses generated in the Fe:ZnSe laser (3). The lasing began after the moment of Fe:ZnSe photobleaching, producing a series of short spikes. There is a clear temporal coincidence between the dips and peaks in the intensity of the profiles of the transmitted pump and lasing pulses, correspondingly. Note that for larger pump spot diameters (6–8 mm), we did not observe such high-frequency and deep laser pulse modulation for heavily doped Fe:ZnSe single crystals [17].

We simulated the lasing process by solving a system of rate equations for the population inversion $n(t) = n_2(t) - n_1(t)$ and the number of photons in the cavity $\phi(t)$ [28]:

$$n'(t) = (\sigma_{abs} \cdot I_a(t)/h\nu_p) \cdot (n_t - n(t)/2) - (2\sigma_{em} \cdot c/V) \cdot \phi(t)\, n(t) - (n_t + n(t))/\tau, \tag{4}$$

$$\phi'(t) = (V_a \cdot \sigma_{em} \cdot c/V) n(t)\, \phi(t) - (\phi(t) - \phi_0)/\tau_c, \tag{5}$$

$$n(0) = -n_t, \phi(0) = 0 \tag{6}$$

where $\sigma_{em} = 1.1 \cdot 10^{-18}$ cm^2 is the room-temperature emission cross section at a wavelength of $\lambda = 4.5\ \mu$m, h is Planck's constant, ν_p is the pump radiation frequency, $I_a(t) = I_p(t)\,[1 - \exp[-(n_t - n(t))/2)\, \sigma_{abs} \cdot l)]$ is the absorbed pump intensity, $I_p(t)$ is the pump intensity, $\tau_c = -2L / (c \cdot \ln[R_1 R_2 (1 - t_i)^2])$ is the photon lifetime in the laser cavity (L is the cavity length, R_1 and R_2 are the reflectivity of the mirrors, and $t_i \approx 0.95$ is cavity loss), c is the speed of light in vacuum, V_a is the volume filled by the mode inside the active medium, V is the volume filled by the mode inside the cavity, l is the Fe:ZnSe crystal thickness, and ϕ_0 are extra photons that are initially present in the cavity to allow laser action to start [28]. (6) describes the initial conditions of this system.

Figure 3(b) shows the waveforms of the incident pump pulses (1') and the pump pulses transmitted through the Fe:ZnSe crystal (2'); the Fe:ZnSe lasing pulse (3') was obtained by calculation.

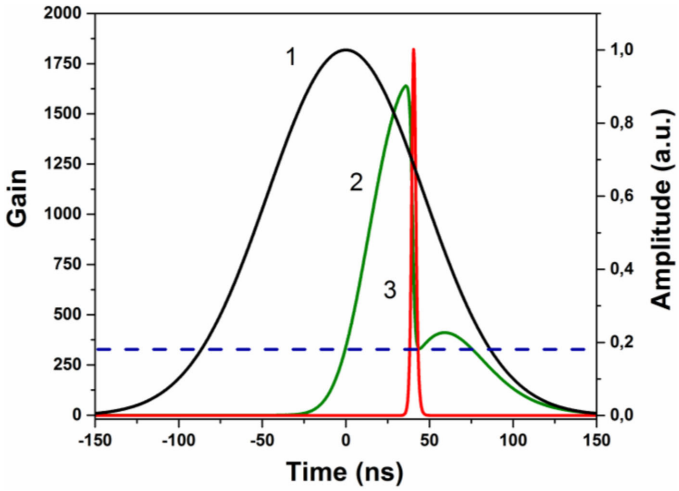

Fig. 4. The simulated waveforms of the incident pump pulse (1), the gain coefficient (2), and the Fe:ZnSe lasing pulse (3). The dashed line marks the level of losses.

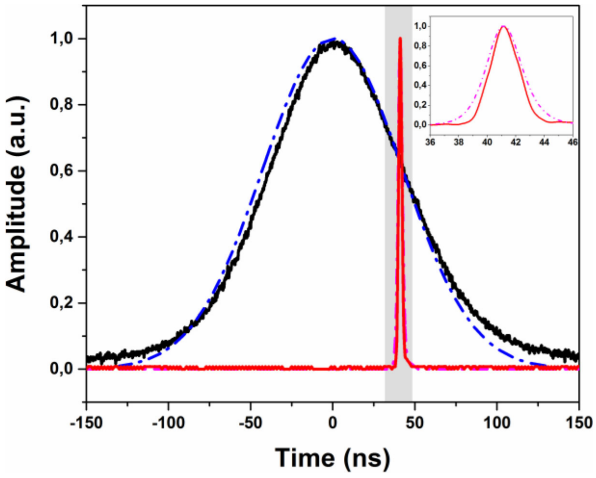

Fig. 5. The waveforms of the pump laser pulse (black and blue) and the ns pulse generated in the Fe:ZnSe laser (red and magenta; the Fe^{2+} ion concentration is $n_t = 2.3 \cdot 10^{19}$ cm^{-3}). The inset shows the zoomed-in view of a single-pulse lasing obtained in Fe:ZnSe. Solid lines—experiment, dash-dotted lines—simulation.

Figure 3 shows substantial agreement between the experimental and simulation data. The observed lasing suggested that it might be possible to achieve single-pulse lasing in heavily doped Fe:ZnSe by reducing the pump energy.

Figure 4 shows the simulated waveforms of the incident pump pulse (1), the gain coefficient (2), and the Fe:ZnSe lasing pulse (3). The pump energy was reduced from 2.5 to 1.5 mJ, which led to the generation of a single pulse. As can be seen from the presented dependences, the generation begins at the moment when the gains exceed the losses. In this case, the population inversion increases continuously due to the pump (consequently, the gain increases as well). As a result, when a high lasing power is reached, the gain significantly exceeds the losses, and the power grows at a rate of $\sim 1.6 \cdot 10^3$ times per round trip of the cavity (0.7 ns). As the lasing power continues to increase, upper laser level depletion due to the stimulated emission is the main contributor to the inversion change. This leads to a sharp drop in the lasing power, producing a single short pulse.

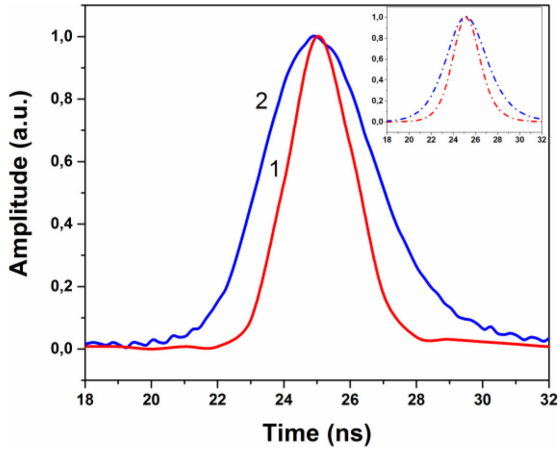

Fig. 6. The waveforms of a single-pulse lasing obtained in Fe:ZnSe with different concentrations of Fe^{2+} doping: $n_t = 2.3 \cdot 10^{19}$ cm^{-3} (1) and $n_t = 6.4 \cdot 10^{18}$ cm^{-3} (2). The inset shows the simulated waveforms of a single-pulse lasing obtained in Fe:ZnSe with different concentrations of Fe^{2+}. Solid lines - experiment, dash-dotted lines – simulation.

Figure 5 compares the pump and laser pulse waveforms obtained from the simulation and experiment.

As shown in Figure 5, at a lower pump power, the generation appears in the form of a single pulse with an FWHM duration of ∼2.8 ns and ∼20 μJ energy. Such a single pulse can be further amplified in Fe:ZnSe, excited (e.g., by a pulsed HF laser [4]) to the energies required for light–matter interaction experiments.

The obtained model also shows that the duration of a single nanosecond pulse depends on the doping concentration. For example, Figure 6 shows the waveforms of a single-pulse lasing obtained with a less-doped Fe:ZnSe crystal ($n_t = 6.4 \cdot 10^{18}$ cm^{-3}) installed in the same laser cavity. As shown, the doping decrease leads to an increase in the pulse duration up to ∼5 ns.

4. Conclusion

We performed a simulation and an experimental study of lasing in Fe:ZnSe single crystals with a high concentration of iron ions $n_t = 2.3 \cdot 10^{19}$ cm^{-3}. We propose a simple method for few-nanosecond pulsed lasing with a Fe:ZnSe laser. The FWHM duration of the obtained pulses was ∼2.8 ns. The demonstrated lasing regime can be used for seeding high-power mid-IR laser systems.

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