

Cr²⁺:ZnSe laser as rare metal isotopes separation tool

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The base conception of using Cr²⁺:ZnSe laser as a tool for ¹⁷⁶Yb isotope optical enrichment together with experimental characteristic of laser source developed at the first stage of the project are presented. Output laser power of Cr²⁺:ZnSe laser with nonstationary active media and self-seeded unidirectional ring resonator run up to ~ 21.3 W with ~ 28.5 % slope and ~ 26.3 % optical efficiency.

Keywords: laser isotopes separation, Cr²⁺:ZnSe laser, ring resonator, nonstationary active media.

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Laser isotope separation is an effective (and, at times, the only available) method for enrichment of rare earth elements for medical applications [1,2]. Dye lasers, which are bulky and hard to operate but generate radiation within a wide range of the visible spectrum, are often used as radiation sources for these purposes. For example, the enrichment of ¹⁷⁶Yb, which is the raw material needed to produce the ¹⁷⁷Lu radionuclide used widely in nuclear medicine, involves three-stage ionization of Yb vapor by pulse-periodic radiation with wavelengths of 555.648, 581.067, and 582.782 nm [2]. Having examined the requirements for the laser radiation spectrum, one may conclude that dye lasers are a versatile and, at first glance, the only effective tool for optical separation of rare earth isotopes.

However, multiplying the given wavelengths by 4, we obtain the values of ~ 2222, ~ 2324, and ~ 2331 nm, respectively. It then becomes evident that the fourth harmonic of a Cr²⁺:ZnSe laser emitting within a wide (from 2 to 3 μm) region of the infrared spectrum may theoretically be used for isotope separation. To assess the possibility of application of a Cr²⁺:ZnSe laser as a tool for isotope separation, we list the other requirements for laser radiation: radiation power at the first, second, and third ionization stages: ~ 1 W, ~ 2 W, and ~ 8 W, respectively; pulse repetition rate: ~ 10 kHz; pulse duration: ~ 20 ns; radiation line width: no more than ~ 250 MHz with a positioning accuracy of ~ 50 MHz [1]. Since the efficiency of each stage of second-harmonic conversion is ~ 50 %, the requirements for radiation power must be raised fourfold.

Various approaches to the design of Cr²⁺:ZnSe lasers have been detailed in literature. Each of them allows one to fabricate a laser source that satisfies only a certain part of the above requirements. Specifically, high-power continuous-wave lasers based on a moving active medium with linear standing-wave resonators were demonstrated in [3,4]. The well-known effect of hole burning [5] makes them fundamentally incapable of providing the required width and accuracy of positioning of the lasing line.

Continuous-wave Cr²⁺:ZnSe lasers with unidirectional ring resonators generating radiation with a power of 160 and 754 mW with a spectral line width of ~ 100 and ~ 29 MHz were discussed in [6,7]. A pulse-periodic Cr²⁺:ZnSe laser pumped by a Ho:YAG laser and positioned in a linear resonator was demonstrated in [8]. Its radiation power and pulse repetition rate were 5.5 W and 2 kHz.

It is evident that a Cr²⁺:ZnSe laser needs to combine the main elements of all the above approaches for it to be potentially suitable for optical isotope separation. To satisfy the emission spectrum requirements, the laser must be positioned in a unidirectional traveling-wave ring resonator. Non-stationary active media need to be used in order to reach the required power level. Pulse-periodic operation may be established through the use of a Ho:YAG laser as a pump source.

The optical diagram of the prospective source is shown in Fig. 1. Pulse-periodic holmium laser 1 is a pump source for a Cr²⁺:ZnSe laser positioned in a unidirectional ring resonator. Pump radiation is focused by lens 2 into non-stationary active element 4 moving in the direction of vector **V**. The input and output flat faces of the active element have wedge angle γ . The resonator is formed by four flat mirrors: non-transmitting mirrors 7 and 8 with a high reflection coefficient in the lasing region and output 6 and dichroic 3 mirrors that have a high reflection coefficient for laser emission and are transparent for pump radiation. Lenses 5 and 9 are introduced into the resonator in order to align the dimensions of the pump beam and the resonator mode.

To ensure unidirectional circulation of radiation in the resonator and obtain the required spectral characteristics, seed radiation from continuous-wave single-frequency Cr²⁺:ZnSe laser 10a, which has one of the known designs [6,7], is introduced into the resonator through mirror 6. Acousto-optic filter 11, which serves to separate laser radiation from seed radiation, is mounted at the output of the laser source.

Let us assume that the active element moves with velocity V ; phase change $\Delta\varphi$ of radiation propagating round

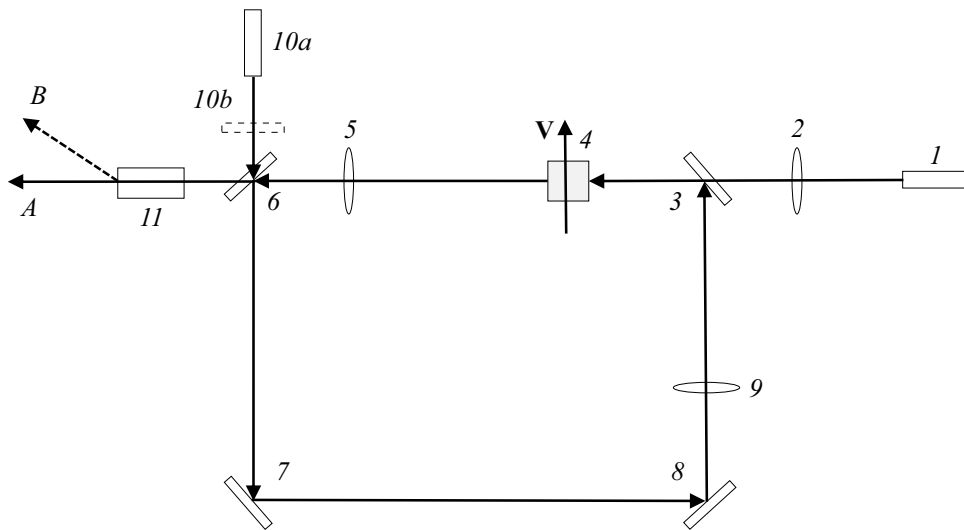


Figure 1. Optical diagram of a $\text{Cr}^{2+}:\text{ZnSe}$ laser.

the resonator will then depend on time t as

$$\Delta\varphi(t) = \frac{2\pi n}{\lambda} \text{tg}(\gamma) V t, \quad (1)$$

where $\lambda \sim 2.3 \mu\text{m}$ is the radiation wavelength and $n \sim 2.44$ is the refraction index of the active medium. With period ΔT written as

$$\Delta T = \frac{\lambda}{\text{tg}(\gamma) V n}, \quad (2)$$

radiation with wavelength λ will propagate round the resonator with a phase change being a multiple of 2π (i.e., will be the resonator mode). Period ΔT for the prototype with $V \sim 12.5 \text{ m/s}$ and $\gamma \sim 0.2^\circ$ discussed below is $\sim 21 \mu\text{s}$. Thus, at a pump pulse rate of $\sim 10 \text{ kHz}$, the interval between two lasing pulses will feature several time points where seed radiation corresponds to the longitudinal mode of the resonator. The power of seed radiation reflected from mirror 6 will decrease significantly at these points.

The time dependence of coefficient R_s of reflection of seed radiation may be determined using the following expression [9]:

$$R_s(t) = R + (1 - R) \frac{(1 - R)R_R + 2RR_R - 2\sqrt{RR_R} \cos(\Delta\varphi(t))}{1 + RR_R - 2\sqrt{RR_R} \cos(\Delta\varphi(t))}, \quad (3)$$

where R is the reflection coefficient of output mirror 6 and R_R is the reflection coefficient that incorporates losses in propagation round the resonator. Dependence $R_s(t)$ for parameters $R_6 \sim 50\%$ and $R_R \sim 70\%$ of the prototype discussed below is presented in Fig. 2.

It can be seen that the reflected power of seed radiation drops to a minimum at time $t = 0$ within several microseconds. Since the radiation beam of the $\text{Cr}^{2+}:\text{ZnSe}$ laser has a diameter of $\sim 300 \mu\text{m}$ and is located at a distance

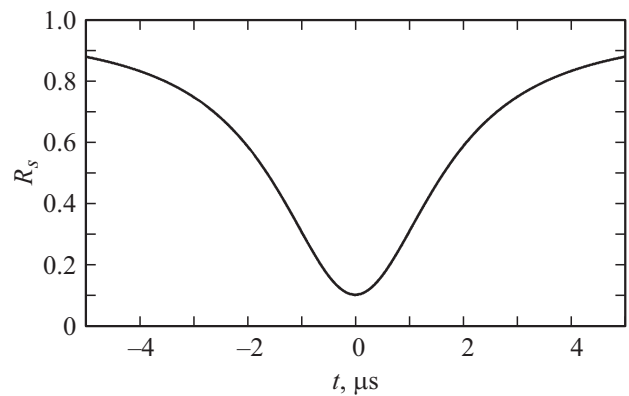


Figure 2. Calculated dependence of the reflection coefficient of the output mirror for seed radiation.

of $\sim 1 \text{ mm}$ from the piezoelectric element of acousto-optic filter 11, the filter switching time (with the speed of sound in paratellurite of $\sim 850 \text{ m/s}$ taken into account) is $\sim 1 \mu\text{s}$. Thus, controlling the power of seed radiation reflected from mirror 6 with a fast diode in position B, one may have just enough time to switch the filter, thereby directing generated radiation to position A, for synchronizing the pump pulse feed and establishing the required length of the resonator.

Thus, the optical diagram presented above allows one to reach the required level of radiation power by using a non-stationary active medium and to generate radiation with spectral characteristics equivalent to the seed signal by synchronizing the pump pulse feed with the establishment of the required length of the unidirectional ring resonator based on the level of seed radiation reflected from the output mirror.

At the first stage, a prototype $\text{Cr}^{2+}:\text{ZnSe}$ laser with an optical circuit largely corresponding to the one in Fig. 1 was constructed. A thulium fiber laser with an operating

wavelength of 1.908 μm was the pump source. The focal length of lens 2 was ~ 100 mm. The diameter of the pump radiation beam in the active element was ~ 0.3 mm. The active element was a Cr²⁺:ZnSe polycrystal with a thickness of ~ 7 mm fabricated at the Institute of Chemistry of High-Purity Substances of the Russian Academy of Sciences (Nizhny Novgorod) by the CVD method with post-growth doping in the process of diffusion under high-temperature isostatic pressing. Doping with Cr²⁺ ions was carried out from both sides of a disk; the doping depth of one layer was ~ 0.5 mm. The surfaces of the active element were polished and made anti-reflecting at pump and lasing wavelengths from 1.9 to 2.7 μm . According to experimental data, the active element absorbed $\sim 95\%$ of attenuated pump radiation. The motion velocity of the active medium was ~ 12.5 m/s.

Output mirror 6 had a reflection coefficient of $\sim 50\%$ in the lasing region. The focal length of lenses 5 and 9 was ~ 50 mm. The distances between the resonator elements were as follows: ~ 20 mm from 3 to 4, ~ 60 mm from 4 to 5, ~ 70 mm from 6 to 7, ~ 100 mm from 7 to 8, ~ 30 mm from 8 to 9, and ~ 40 mm from 9 to 3. The physical length of the resonator was ~ 340 mm. Calculations by the matrix method demonstrate that the diameter of the fundamental mode in the active element of the discussed resonator is ~ 250 μm . A thermal lens, which forms inevitably in the active element in the process of lasing, has little effect on the size of the fundamental mode.

A source of seed radiation was not used at this stage. To ensure unidirectional circulation of radiation in the resonator, flat mirror 10b was mounted at one of its outputs, sending radiation moving in the direction opposite to the working one back to the resonator. A similar method of self-seeding was used in [7].

Figure 3 shows the experimental and calculated dependences of output radiation power of the Cr²⁺:ZnSe laser on the pump radiation power with bidirectional (without mirror 10) and unidirectional propagation round the resonator. The calculated dependence of the lasing power of the Cr²⁺:ZnSe laser on the pump power was obtained with a total intracavity loss of 31.6%. This

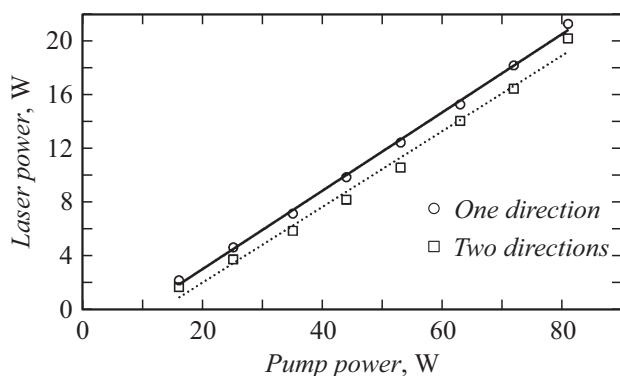


Figure 3. Calculated (line) and experimental (symbols) dependences of the lasing power of a Cr²⁺:ZnSe laser on the pump power with bidirectional and unidirectional propagation round the resonator.

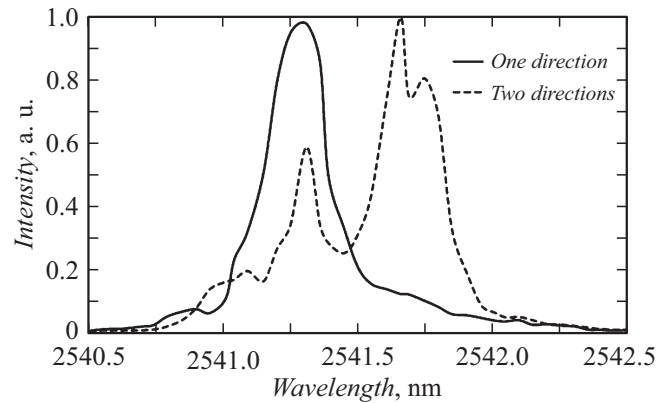


Figure 4. Lasing spectra with bidirectional and unidirectional propagation round the resonator.

high loss value may be attributed both to the quality of anti-reflection coatings on the active element and to non-resonant absorption.

The figure makes it evident that the maximum total lasing power with bidirectional propagation round the resonator was ~ 20.2 W at an optical efficiency of $\sim 24.9\%$ and a slope efficiency of $\sim 28.0\%$. The maximum lasing power with unidirectional propagation round the resonator was ~ 21.3 W at an optical efficiency of $\sim 26.3\%$ and a slope efficiency of $\sim 28.5\%$. It should be noted that the lasing power was divided approximately evenly between the two directions in the case of bidirectional propagation. The pattern was the same in calculations.

Figure 4 shows the lasing spectrum for unidirectional propagation and the total lasing spectrum for bidirectional propagation round the resonator. It is evident that the lasing spectrum with unidirectional propagation round the resonator is a single narrow peak ~ 0.3 nm in width. With bidirectional propagation, the lasing spectrum features several peaks with a total width of ~ 0.8 nm.

Thus, at the first stage of research, the feasibility of a fundamental element of the concept of application of a Cr²⁺:ZnSe laser for optical separation of rare earth metal isotopes was verified experimentally: the possibility of constructing a laser source based on a non-stationary Cr²⁺:ZnSe active medium in a ring resonator with unidirectional circulation of radiation in it established by seeding radiation through the output mirror of the resonator was proven.

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Conflict of interest

The authors declare that they have no conflict of interest.

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