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High-power diode-pumped alkali lasers*

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1. Introduction

In recent years, diode-pumped alkali vapor lasers have been attracting great interest from both the scientific and the military establishments, because there is reason to hope that such lasers will generate in the near future very high-power cw radiation (100 kW and higher) with an efficiency of no less than 50%, while being compact enough and simple in design and operation.

The physical principle of the operation of alkali vapor lasers is very simple (Fig. 1). Pump radiation is resonantly absorbed at the transition from the $ns_{1/2}$ ground state of an alkali metal atom to the $np_{3/2}$ state (the D₂ line) (n = 2, 3, 4, 5, 6 for lithium, sodium, potassium, rubidium, and cesium, respectively). One of the necessary conditions for the laser's operation is a sufficiently high buffer gas pressure. Collisions with buffer gas particles cause transitions between the finestructure components $np_{3/2}$ and $np_{1/2}$ of the excited energy level. The distance between these components is not very large (less than or equal to the thermal energy $k_{\rm B}T$), and therefore the probability of these transitions is quite high. At high buffer gas pressures (on the order of a few hundred mmHg), the equilibrium Boltzmann distribution is established between the excited $np_{3/2}$ and $np_{1/2}$ states for a time shorter than the lifetime of the state. According to this distribution, the population of the $np_{1/2}$ state is higher than that of the $np_{3/2}$ state by the Boltzmann factor exp ($\Delta E/k_{\rm B}T$), where ΔE is the energy separation between the $np_{3/2}$ and $np_{1/2}$ levels. Notice that collision transitions from excited levels to the ground state are negligibly weak, even at high pressures (at

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Figure 1. Diagram of operating energy levels and optical transitions in alkali metal atoms. The straight and wavy arrows denote pump-induced and laser transitions, respectively. The curved arrows indicate collision-induced transitions. The vertical columns show the relative level populations.

least for a number of buffer gases, in particular, inert gases), so that here we are dealing with the possibility of the establishment of the Boltzmann equilibrium for a separate group of adjacent energy levels.

If we now provide the rise in the pump radiation intensity to an extent that it equalizes the populations of the ground state and the $np_{3/2}$ level, the population of the $np_{1/2}$ level will become higher than that of the ground state by the same Boltzmann factor. Thus, we obtain population inversion at the $np_{1/2}-ns_{1/2}$ transition, i.e., the condition for lasing in this transition.

The present paper deals with the history of this problem and its development to the current state of the art; then, the prospects of and options for generating high-power radiation in different pumping schemes are estimated, and relations are obtained for physical parameters determining the lasing process.

2. Historical excursion and the state of the art

The idea of producing population inversion by the method described in the Introduction was first proposed by B A Glushko at the Institute of Physical Research, Academy

of Sciences of Armenian Republic in 1981 [1]. In this paper, the first experiments were also reported on lasing at the D_1 line in potassium vapor pumped by a laser radiation to the D_2 line. A short time later, paper [2] was published, in which lasing was achieved by the same method in the superluminescence regime in sodium vapor by using helium as the buffer gas. In addition, in Ref. [2] the lasing mechanism was described theoretically.

The further development of investigations in this area is reflected in papers [3–6]. In Ref. [3], more detailed experiments on lasing in potassium vapor were performed. In Ref. [4], experiment [2] was repeated, and then the same authors showed that lasing in sodium vapor was considerably enhanced by using molecular gas (ethane) as the buffer gas [5]. Ethane was also utilized as the buffer gas in experiments with rubidium vapor [6].

All the experiments considered above were performed using pumping by coherent laser radiation. In this connection, although this effect was interesting from the physical point of view, no prospects for its practical applications were expected because laser radiation was converted to almost the same radiation with the spectrum only slightly shifted to the red.

The situation drastically changed when Krupke [7] proposed a fruitful idea based on the great progress in the technology of manufacturing laser diodes. The idea was to use laser diodes for pumping to D₂ transitions of alkali metal atoms. Modern laser diodes have a number of unique parameters, first and foremost high efficiency. The radiation power of a single diode, despite its small size (the emitting surface area is a few fractions of a centimeter squared) reaches a few hundred watts. The emission spectrum of a laser diode is relatively narrow (several reciprocal centimeters). It is possible to produce a high power density (more than 1 kW cm⁻²) sufficient to equalize the level populations of alkali metal atoms under operating conditions. Finally, laser diodes emitting in the near-IR spectral region, where the resonance absorption lines of alkali K (0.77 µm), Rb (0.79 μ m), and Cs (0.85 μ m) atoms are located, have the best parameters.

Laser diodes are not themselves sources of truly coherent radiation, but they serve as excellent light sources for pumping lasers of different types. Thus, by using laser diodes for pumping alkali vapors to the D_2 transition, we obtain lasing at the closely spaced D_1 line, i.e., in fact the incoherent radiation of laser diodes will be converted into coherent radiation.

Because diode-pumped alkali vapor lasers can be scaled, they can be expected to become sources of high-power coherent radiation (more than 100 kW) for wide industrial and military applications. One of the reasons for optimism in this respect is that active gas media offer known advantages. The active gas medium is optically and thermally stable (no damage or chemical degradation appears, as in solids and liquids, respectively). The parameters of a gas medium (concentrations of the active and buffer components, temperature, volume, and geometry) can be readily controlled. A gas medium offers more possibilities for cooling, in particular, by using gas circulation. The relative quantum defect of energy in the conversion of a pump photon to a laser photon in the lasing mechanism discussed here is very small (5% in cesium, 2% in rubidium, and 0.5% in potassium), which provides the high lasing efficiency. This circumstance is especially important for minimizing heat generation.



Figure 2. Principal scheme of a transversely pumped laser (a) and a twostage system (master oscillator and amplifier) (b).

We will show below that alkali vapor lasers can emit record high powers per unit volume of the active medium due to a small quantum defect of energy and the high quantum yield (the ratio of the number of laser photons to the number of pump photons) under optimal conditions and at high enough concentrations of active atoms.

After issuing Krupke's proposal, many papers devoted to investigations of alkali vapor lasers were published (see Refs [8–29] as key publications). The first studies were carried out using laser pumping. The dependence of the lasing efficiency in the D₁ transitions under various conditions was investigated in detail and theoretical models were simultaneously developed. Then, researchers began to implement direct diode pumping in experiments, and soon diodepumped lasers began to demonstrate promising results. A high enough lasing efficiency (68%) was achieved in cesium vapor for practically significant output power of 10 W [17]. The maximum output power (145 W) for diode-pumped alkali vapor lasers was achieved for the lasing efficiency of 28% in Ref. [26].

The output powers presented above are, of course, much lower than optimistic predictions (a few hundred kW) for lasers of this type. This is mainly explained by the fact that scientific studies are performed with one module oriented to using one laser diode (it is not improbable, however, that not all the information is published in journals because this field of research attracts the attention of the military).

To obtain considerably higher output powers, it is necessary to scale the process. The lasers under study are quite promising for scaling. In our opinion, the transverse pumping scheme is optimal for scaling (Fig. 2a). The output power in this geometry increases proportionally to the laser length. In practice, it is appropriate to utilize a separate master oscillator unit generating radiation with the required properties (coherent and monochromatic with the specified frequency and geometrical parameters of the beam). Then, this radiation picks up power in an amplifier by preserving the specified properties (Fig. 2b).

3. Basic relations

The change in the N_1 , N_2 , and N_3 populations of levels 1, 2, and 3 (see Fig. 1) caused by pump radiation, atomic collisions, and generated radiation is described by the equations

$$\frac{\mathrm{d}N_3}{\mathrm{d}t} = -(A_{31} + v_{32})N_3 + v_{23}N_2 + w_\mathrm{p}(N_1 - \alpha N_3),$$

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} = -(A_{21} + v_{23})N_2 + v_{32}N_3 + w_\mathrm{L}(N_1 - N_2),$$

$$N_1 + N_2 + N_3 = N. \tag{1}$$

Here, A_{31} and A_{21} are the spontaneous emission rates (the first Einstein coefficients) for the 3–1 and 2–1 transitions; the collision frequencies v_{32} and v_{23} describe the collision mixing of levels 3 and 2; w_p and w_L are the probabilities of stimulated transitions caused by pump radiation and laser radiation, respectively; $\alpha = g_1/g_3$ is the ratio of the statistical weights of levels 1 and 3 ($\alpha = 1/2$ for alkali atoms), and N is the total concentration of active atoms.

We assume that the spectrum of pump radiation has an arbitrary width, while laser radiation is monochromatic. Then, the transition probabilities are as follows:

$$w_{\rm p} = |G_{\rm p}|^2 \int \frac{g(\omega) \Gamma_{\rm p}}{\Gamma_{\rm p}^2 + (\omega - \omega_{31})^2} \, \mathrm{d}\omega \,, \quad \int g(\omega) \, \mathrm{d}\omega = 1 \,,$$
$$w_{\rm L} = |G_{\rm L}|^2 \frac{\Gamma_{\rm L}}{\Gamma_{\rm L}^2 + \Omega_{\rm L}^2} \,, \quad G_{\rm p} = \frac{E_{\rm p} d_{31}}{2\hbar} \,, \quad G_{\rm L} = \frac{E_{\rm L} d_{21}}{2\hbar} \,. \tag{2}$$

Here, d_{31} , d_{21} , and ω_{31} , ω_{21} are the matrix elements of the dipole moment and frequencies of the 3–1 and 2–1 transitions, respectively; G_p and G_L are the Rabi frequencies for pump and laser radiations, respectively; E_p and E_L are the corresponding electric-field amplitudes; Γ_p and Γ_L are the homogeneous half-widths of the pump and laser radiation lines (we assume here that the collision broadening considerably exceeds the Doppler broadening), and $g(\omega)$ is the spectral density of pump radiation normalized to unity.

According to the principle of detailed balance, the collision frequencies v_{32} and v_{23} are related by the expression

$$v_{23} = \frac{g_3}{g_2} v_{32} \exp\left(-\frac{\Delta E}{k_{\rm B}T}\right), \quad \Delta E = E_3 - E_2 = \hbar\omega_{32}.$$
 (3)

This is the basic relation determining the emergence of lasing at the 2-1 transition.

By solving equations (1) under stationary conditions, we can easily obtain the population differences characterizing lasing and absorption of pump radiation (the natural generalization of results [2] taking into account the statistical weights of appropriate energy levels, nonmonochromaticity of pumping, and the influence of laser radiation on the internal states of atoms):

$$N_{2} - N_{1} = \frac{N}{1 + \kappa_{p} + \kappa_{L} + b\kappa_{p}\kappa_{L}} \left(\kappa_{p} \frac{v_{32} - \alpha\Gamma_{2}}{v_{32} + (1 + \alpha)\Gamma_{2}} - 1\right),$$

$$N_{1} - \alpha N_{3} = \frac{N}{1 + \kappa_{p} + \kappa_{L} + b\kappa_{p}\kappa_{L}} \left(\kappa_{L} \frac{\Gamma_{3} - \alpha v_{23}}{v_{23} + 2\Gamma_{3}} + 1\right),$$

$$b = \frac{(1 + 2\alpha)(A_{21}v_{32} + A_{31}\Gamma_{2})}{(v_{32} + (1 + \alpha)\Gamma_{2})(v_{23} + 2\Gamma_{3})},$$
(4)

were $\Gamma_3 = A_{31} + v_{32}$ and $\Gamma_2 = A_{21} + v_{23}$ are the total spontaneous emission and collision decay frequencies of levels 3 and 2; κ_p and κ_L are the so-called saturation parameters (here, each of them characterizes the degree of equalization of populations at the 3–1 or 2–1 transition in the absence of the second field):

$$\kappa_{\rm p} = w_{\rm p} \, \frac{v_{32} + (1+\alpha)\,\Gamma_2}{A_{21}v_{32} + A_{31}\Gamma_2} \,, \quad \kappa_{\rm L} = w_{\rm L} \, \frac{v_{23} + 2\Gamma_3}{A_{31}v_{23} + A_{21}\Gamma_3} \,. \tag{5}$$

As follows from the first relation in Eqn (4), to achieve efficient lasing, it is essential to strive for satisfying the conditions

$$\nu_{23}, \nu_{32} \ge A_{31}, A_{21}, \kappa_{p} \ge \frac{\nu_{32} + (1+\alpha)\nu_{23}}{\nu_{32} - \alpha\nu_{23}} = \frac{1+3\exp\left[-\Delta E/(k_{B}T)\right]}{1-\exp\left[-\Delta E/(k_{B}T)\right]}$$
(6)

[here, we used relation (3) with $g_3 = 4$ and $g_2 = 2$). The first condition in Eqn (6) is provided with a large margin in the experiments performed (buffer gas pressure of about 1 atm and higher). The second condition at the power density of ~ 1 kW cm⁻² provided by diode pumping is also virtually fulfilled. Under conditions (6), pumping produces ultimately high population inversion in the laser transition. When lasing is still absent ($\kappa_L = 0$), this population inversion equals

$$N_2 - N_1 = N \frac{1 - \exp\left[-\Delta E/(k_B T)\right]}{1 + 3 \exp\left[-\Delta E/(k_B T)\right]}.$$
(7)

For rubidium and cesium, $\Delta E = 237.5 \text{ cm}^{-1}$ and $\Delta E = 554.1 \text{ cm}^{-1}$, respectively, and therefore at operating temperatures of 400–500 K, the inversion produced is very large, providing the rapid rate of lasing development.

The broader the pump radiation spectrum, the more difficult it is to fulfill the second condition in Eqn (6). The emission linewidth of laser diodes is several cm^{-1} . The optimum will be achieved by decreasing the laser spectral width by approximately an order of magnitude.

If lasing has developed to the level at which

$$\kappa_{\rm L} \gg \frac{2\nu_{32} + \nu_{23}}{\nu_{32} - \alpha\nu_{23}} = 2 \frac{1 + \exp\left[-E/(k_{\rm B}T)\right]}{1 - \exp\left[-\Delta E/(k_{\rm B}T)\right]}, \qquad (8)$$

the quantum yield (the ratio of the numbers of generated and absorbed photons) becomes the maximum possible, i.e., equal to unity. This can be easily verified by using relations (4) under conditions (6) and (8). The number of emitted laser photons per unit time and unit volume is $w_L(N_2 - N_1)$. The ratio of this number to the number $w_p(N_1 - \alpha N_3)$ of absorbed pump photons gives just the quantum yield. Under stipulated conditions, we have

$$\frac{w_{\rm L}(N_2 - N_1)}{w_{\rm p}(N_1 - \alpha N_3)} = 1.$$
(9)

The achievement of the quantum yield close to unity is another advantage of the type of lasers discussed here.

It is obvious that under conditions (6) and (8) the fraction of spontaneously emitted photons is small compared to the fraction of generated photons. Let us assume that the former comprises 10%. Now we can estimate the important quantity like output laser energy per unit volume. From the expression for $w_L(N_2-N_1)$ at $A_{31} \approx A_{21} \approx 3 \times 10^7 \text{ s}^{-1}$, we find that one atom can emit more than 10⁸ laser photons per second. To provide 1 kW of radiation power (10²² photons per second), 10^{14} atoms are required. For a working concentration of active atoms equal to 10^{14} cm^{-3} (this value can be even higher in the conditions of transverse pumping), this power can be provided by a lasing volume of 1 cm³. Based on these estimates, one may conclude that alkali vapor lasers might be quite compact.

4. Other lasing schemes

We considered the most efficient modern lasers operating due to the collision production of population inversion in the transition to the ground state. These lasers operate according the three-level scheme. Moreover, variants are possible in



Figure 3. Four-level scheme of an alkali vapor laser. The straight and wavy arrows denote pump-induced and laser transitions, respectively. The curved arrow indicates collision-induced transitions.

which the physical mechanism is the same, but the number of operating levels is different.

In patent [30] (see also paper [31]), the four-level scheme was proposed for alkali metal atoms (Fig. 3), in which a higher lying state (for example, the (n + 2) s state) is excited in two pumping stages. Inelastic collisions of active atoms with a buffer gas transfer the population of the (n + 2) s level to the (n + 1) p level with the opposite parity, so that the (n + 1) p -ns transition appears as allowed.

At high enough pressures, the (n + 1) p level can be populated more strongly than the ground *n*s level, resulting in the appearance of amplification and subsequent lasing in the (n + 1) p – *n*s transition. This scheme is of interest because lasing occurs in the visible spectral region at the frequency almost equal to the sum of pump photon frequencies. However, there is also a disadvantage, because the energy gap between the (n + 2) s and (n + 1) p levels is rather large (considerably larger than the mean thermal energy of particles), and therefore the population transfer between these levels due to collisions is hindered.

We suggest an idea that the buffer gas of molecules with the vibrational quantum energy close to the energy of the desired transition be used. Then, resonance energy transfer will occur in atom-molecule collisions and the efficient collision population of the (n + 1) p level can be expected.

Laser diodes available today can reliably provide the equalization of populations of the *n*s and (n + 2)s levels, even in the two-stage process, so that efficient lasing in the visible region can occur in alkali metal vapors.

It was shown that the number of levels can be reduced to two, and nevertheless lasing can be achieved under the action of collisions and incoherent pumping (Fig. 4) [32–36]. A key factor in this situation is the inequality between the absorption and stimulated emission probabilities (second Einstein coefficients), which is realized during frequent collisions and when radiation interacting with the two-level system is considerably detuned from the resonance. In the case of blue detuning from the resonance and a high enough pump intensity, the population inversion is produced in the 2–1 transition, resulting in the appearance of amplification at the resonance frequency and subsequent lasing. The process is



Figure 4. Two-level laser scheme. The straight and wavy arrows denote pump-induced and laser transitions, respectively. The vertical columns show the relative level populations.

described by the equations

1.1.1

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} = -A_{21}N_2 + w_{\mathrm{L}}(N_1 - N_2) + w_{\mathrm{p}}(N_1 - \xi N_2),$$

$$N_1 + N_2 = N. \tag{10}$$

Here, the notions were introduced:

$$\begin{split} w_{\rm p} &= |G_{\rm p}|^2 \, \frac{\Gamma_{\rm oc}}{\Omega_{\rm p}^2} \,, \quad w_{\rm L} = |G_{\rm L}|^2 \, \frac{\Gamma}{\Gamma^2 + \Omega_{\rm L}^2} \,, \\ \xi &= \exp\left(-\frac{\hbar |\Omega_{\rm p}|}{k_{\rm B}T}\right) , \quad \Omega_{\rm p} = \omega_{\rm p} - \omega_{\rm 21} \,, \quad \Omega_{\rm L} = \omega_{\rm L} - \omega_{\rm 21} \,. \end{split}$$

The frequency detuning Ω_p of pump radiation is assumed positive and large ($\Omega_p \gg \Gamma$); therefore, pump radiation can have a broad spectrum and may thus be treated as monochromatic. The frequency detuning Ω_L of laser radiation is close to zero, and Γ_{oc} is the optical collision constant [35–37] (the analog of the absorption line half-width Γ). The rest of the notation is identical to that introduced above.

We find from equations (10) the level population differences determining the amplification of generated radiation and the absorption of pump radiation (stationary conditions):

$$N_{2} - N_{1} = \frac{N}{1 + \kappa_{p} + \kappa_{L}} \left(\kappa_{p} \frac{1 - \xi}{1 + \xi} - 1 \right),$$

$$N_{1} - \xi N_{2} = \frac{N}{1 + \kappa_{p} + \kappa_{L}} \left(\kappa_{L} \frac{1 - \xi}{2} + 1 \right).$$
 (11)

As above, κ_p and κ_L are the saturation parameters for pump and laser radiations, respectively:

$$\kappa_{\rm p} = w_{\rm p} \, \frac{1+\xi}{A_{21}} \,, \quad \kappa_{\rm L} = w_{\rm L} \, \frac{2}{A_{21}} \,.$$
(12)

One can see from the first relation in Eqn (11) that, if the pump intensity in high enough $[\kappa_p > (1 + \xi)/(1 - \xi)]$, the

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population inversion will appear in the 2–1 transition. For a stronger condition, viz.

$$\kappa_{\rm p} \gg \frac{1+\xi}{1-\xi} \,, \tag{13}$$

the level population inversion reaches the maximum value (lasing has not appeared yet) equal to

$$N_2 - N_1 = N \frac{1 - \xi}{1 + \xi} , \qquad (14)$$

which is determined only by the pump frequency detuning from the resonance. Theoretically, the inversion can be complete (for $\xi \rightarrow 0$); however, one should bear in mind that the fulfillment of condition (13) becomes more complicated with increasing detuning. Note also that this condition can be more easily satisfied with increasing buffer gas pressure.

The quantum yield in the lasing scheme under consideration is determined by the relation

$$\frac{w_{\rm L}(N_2 - N_1)}{w_{\rm p}(N_1 - \xi N_2)} = \frac{1 - \xi - (1 + \xi)/\kappa_{\rm p}}{1 - \xi + 2/\kappa_{\rm L}} \,. \tag{15}$$

If generated radiation is amplified so that, along with condition (13) for pump radiation, the condition $\kappa_L \ge 2/(1-\xi)$ is fulfilled for generated radiation, one can easily see that the quantum yield becomes unity.

Lasing in the two-level scheme was examined experimentally in sodium vapor pumped by pulsed laser radiation [32, 34, 35]. The most efficient lasing was achieved at the buffer gas pressure of 5 atm. Lasing is beginning to manifest itself at the pump power density of about 1 MW cm⁻². This value remains to be reached with incoherent pumping at present. However, we can hope that such pulsed radiation sources will appear in the near future. An advantage of this lasing method is that there are no constrains on the pump-spectrum width.

Another option for obtaining lasing in a two-level system is based on the equalization of populations of levels 1 and 2 by high-power pump radiation tuned to the resonance with the 1-2 atomic transition. In the case of frequent atomic collisions, amplification conditions are realized in the red wing of the line due to the same reason of inequality between the absorption and stimulated emission probabilities. Equation (10) under these conditions is transformed into [36]

$$\frac{dN_2}{dt} = -A_{21}N_2 + w_p(N_1 - N_2) + w_L(\xi N_1 - N_2),$$

$$N_1 + N_2 = N,$$

$$w_p = |G_p|^2 \int \frac{g(\omega)\Gamma}{\Gamma^2 + (\omega - \omega_{21})^2} d\omega,$$

$$\int g(\omega) d\omega = 1, \quad w_L = |G_L|^2 \frac{\Gamma_{oc}}{\Gamma^2 + \Omega_L^2}.$$
 (16)

The level population differences characterizing the amplification of generated radiation and the absorption of pump radiation are described by the expressions

$$N_{2} - \xi N_{1} = \frac{N}{1 + \kappa_{p} + \kappa_{L}} \left(\kappa_{p} \, \frac{1 - \xi}{2} - \xi \right),$$
$$N_{1} - N_{2} = \frac{N}{1 + \kappa_{p} + \kappa_{L}} \left(\kappa_{L} \, \frac{1 - \xi}{1 + \xi} + 1 \right).$$
(17)

The signal amplification in the case of the red detuning occurs if $\kappa_p > 2\xi/(1-\xi)$, which can be quite easily realized in practice if ξ noticeably differs from unity. When the stronger inequality $\kappa_p \ge 2\xi/(1-\xi)$ and the similar condition $\kappa_L \ge (1+\xi)/(1-\xi)$ for generated radiation are simultaneously fulfilled, the quantum yield of lasing determined by the ratio $w_L(N_2 - \xi N_1)/[w_p(N_1 - N_2)]$ tends, as can be readily convinced, to unity.

The specific feature of the above-considered two-level scheme of lasing is as follows. To achieve the equalization of the level populations during pumping is no more difficult than in the three-level scheme, but the gain proves to be rather small and the amplifying medium should be long for attaining the radiation intensity at which the quantum yield is close to unity.

5. Conclusions

The development of alkali vapor lasers undoubtedly shows considerable promise. The results already obtained in this area are quite impressive, although not all the advantages of this lasing method have been used so far. In particular, the theoretically possible ultimately high quantum yield has not been reached to date. The lasing efficiency will increase as the output power of laser diodes increases and (or) the width of their emission spectrum decreases. The employment of transverse pumping will easily provide scaling, thereby increasing the entire output power.

The four-level lasing scheme with two-stage diode pumping gives promise that high-power alkali vapor lasers emitting in the visible spectral range will be created.

In the case of the two-level system, the possibility of achieving highly efficient lasing is related to the development of sufficiently high-power pulsed incoherent radiation sources in the near future. The spectrum of this pumping radiation can be rather broad (up to a few hundred cm^{-1}).

Unfortunately, as far as we know, experimental developments in alkali vapor lasers are not at all being pursued at present at academic institutions in Russia. It is advantageous to eliminate this shortcoming, because these lasers can find important industrial and special applications.

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