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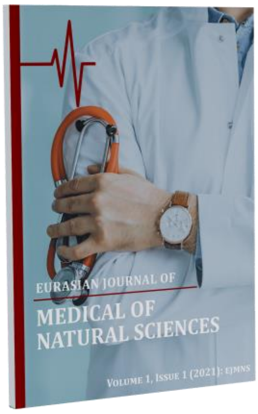

MATERIALS FOR ACTIVE MEDIA OF MINIATURE SOLID-STATE LASERS

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ABSTRACT

The article studies a new class of laser materials - neodymium scandoborates, which can be used to create miniature lasers for communication and information processing. Such a crystal can be used as a highly efficient highly concentrated medium for miniature medium power lasers. Crystals of this group have a high non-linear dielectric susceptibility; therefore, with a certain orientation of the crystal relative to the propagation of laser radiation, infrared radiation of a non-dim laser can be converted into visible radiation.

It is known that miniature, but rather powerful pulsed or continuous lasers are considered the most suitable for communication and information processing. For this, solid-state lasers with high efficiency are used.

The requirement for a high efficiency is imposed on lasers of all types, but in the case of solid-state lasers with optical pumping, it is especially important, since under lamp pumping, due to poor matching between the emission spectra of the lamp and the absorption of the active element, the efficiency can be as low as 0, 01% and the maximum value is 12%. True, the efficiency of semiconductor lasers reaches approximately 80%, but due to the large divergence of the laser beam, their use in some areas is very limited. Gas lasers are too large and cannot be used as miniature ones either. Therefore, where miniature lasers with sufficiently high powers (0.1-10 Wt) and low laser beam divergence are needed, solid-state lasers should be used.

The most common classical laser emitting in the near infrared region of the spectrum (1.06 μm) is a neodymium yttrium-aluminum garnet laser. The working particles in it are Nd^{3+} neodymium ions, and the laser operates according to the so-called four-level scheme.

Yttrium-aluminum garnet $Y_3Al_5O_{12}:Nd^{3+}$ crystals have an exceptional set of properties that make them a very suitable material for solid-state lasers. They are transparent in a very wide spectral region (0.2-5 μm), mechanically strong, have high radiation stability, and in terms of thermal conductivity they are only slightly inferior to Al_2O_3 corundum, whose thermal conductivity is approximately the same as that of copper. The crystal structure of yttrium aluminum garnet (YAG) allows the introduction of significant concentrations of Nd^{3+} ions. At present, the technology of growing YAG single crystals is well developed. The YAG laser has a low generation threshold. Thus, it seemed that this material is ideal for creating high-performance lasers. However, it turned out that because of the so-called concentration quenching of luminescence, it cannot be used for miniature high-efficiency lasers. To understand what is the matter, one will have to consider those processes and phenomena that occur in crystals containing impurities of rare-earth ions, to which YAG belongs: Nd^{3+}.

In order to miniaturize the active element, it is necessary to have crystals with active particle concentrations of -10^{21}-10^{22} cm^{-3}. In the case of YAG:Nd, an increase in the neodymium concentration above 10^{19} cm^{-3} leads to a sharp drop in the probabilities of radiative transitions $^4F_{3/2}$ - Al_j, which means that the generation threshold increases sharply and, most importantly, the efficiency drops sharply. At first glance, this phenomenon, called the concentration extinguishing of luminescence (CEL), is incomprehensible, since the electronic states at such concentrations of neodymium are practically independent of its concentration in the crystal. Studies have shown that the cause of CEL was the interaction of Nd^{3+} ions with each other.

As already mentioned, an excited ion can be considered as a system of oscillators with frequencies $v_{mn} = (E_m\text{-}E_n)/h$. In the case of Nd^{3+} ions in YAG, the crystal field splits the AIj multiplets so strongly that the transitions $^4F_{3/2}$-$^4I_{15/2}$ and $^4I_{9/2}$-$^4I_{15/2}$, as well as correspond to the same frequencies, that is, such oscillators are in resonance. This means that if unexcited Nd^{3+} is located near the excited Nd^{3+} ion, then due to the electromagnetic interaction, part of the excitation energy will be transferred to the neighbor, so that both ions will be excited to the levels of ^4Ij multiplets, from which the excitation energy is resonantly transferred to lattice vibrations, and, thus, the excitation dies for emission.

The probability of the implementation of the described process strongly depends on the distance between the excited ion (donor) and the unexcited ion (acceptor), it is proportional to $1/r^6$ (r is the distance between the interacting ions). Therefore, at low concentrations of impurity ions, such a process is unlikely; it turns out to be essential for YAG and other crystals with a strong crystalline field, when the concentrations of active ions reach certain critical values. For YAG : Nd^{3+}, this concentration is approximately $5 \cdot 10^{19}$ cm^{-3}. The described process is called cross-relaxation.

In addition to this process, there is another, which also eventually leads to the loss of excitation - the so-called migration of excitation. Excitation from one ion is transferred to another, nearby, due to resonant interaction. For Nd^{3+} ions, this occurs at the frequencies of the $^4F_{3/2}$ - $^4I_{9/2}$ and $^4I_{9/2}$ - $^4F_{3/2}$ transitions. The excitation, as it were, jumps from one ion to another, migrating through the crystal until either the above process occurs, or the excitation is transferred to another inactive impurity and, thus, will be lost for radiation.

In high-purity crystals, the main cause of CEL is cross-relaxation, and it is precisely this that sets the limit for increasing the concentration of the active impurity in YAG crystals and others with a strong crystalline field. Careful consideration of the Stark splittings of the ^4I_j multiplets showed that a decrease in these splittings can bring the $^4F_{3/2}$-$^4I_{15/2}$ and $^4I_{9/2}$-$^4I_{15/2}$ transitions out of resonance. Hence it followed that it was necessary to find crystals in which the strength of the crystalline electric field at those lattice sites where neodymium ions are located would be small and, moreover, the maximum lattice vibration frequency would also be such that the phonon energies with this frequency would not be sufficient to introduce purely electronic transitions into resonance.

Note that in free ions (atoms) optical transitions, that is, transitions with absorption or emission of a photon, between levels of the same configuration, in this case $4f^3$ do not occur (they are forbidden by parity): in the absorption and emission spectra of frequencies, corresponding to these transitions, no. If the ion (atom) is in a crystal, then the crystal field, in particular, its dynamic low-symmetry component, changes the states of the ion in such a way that these transitions turn out to be very probable.

It follows from these data that in crystals (YAG, $YA1O_3$), in which neodymium ions are directly surrounded by O^{2-} oxygen ions, the Stark splitting of multiplets 4Ij; there are significantly more splittings in those crystals in which the nearest environment of neodymium ions is formed by oxygen ions that are part of phosphate, borate, tungstate and other groups, the bond in which is covalent in nature. For many of these crystals, the lattice sites where the active neodymium ion can be located are low-symmetric, and the maximum background energy is low. It is these crystals that are the most suitable laser active media with a high concentration of Nd^{3+} ions. Such crystals include complex phosphates and borates. It turned out to be possible to obtain crystals of these compounds only from a solution in a melt. The crystal growth rate in this technology does not exceed 1–2 mm per day, and the optical quality of the crystals is low. Another such crystal is yttrium aluminum borate $YAl_3(BO_3)_4 : Nd^{3+}$. Its spectral-luminescent properties are unique. Lasers built on these crystals and crystals with complete replacement of yttrium ions by neodymium ions, that is, on $NdAl_3(BO_3)_4$ crystals, have low lasing thresholds and high efficiency. However, due to the very complex technology for obtaining these crystals, lasers with them have not been widely used.

NEODIUM SCANDOBORATES - A NEW CLASS OF LASER MATERIALS

After analyzing the physicochemical properties of complex borates, researchers [1] found that the $YSc_3(BO_3)_4$ compound is suitable for obtaining its single crystals directly from a melt in air using the Czochralski method with growth rates of 3–4 mm per hour. Indeed, the prediction was justified: $Y_{1-y}Nd_xSc_3(BO_3)_4$ crystals were obtained [1, 2] and studied in detail [2, 3]. Studies have shown that the spectral-luminescent and generation properties of crystals of this family are not inferior to either aluminoborates or phosphates. Having discovered this, a group of scientists studied the entire class of rare earth scandoborates, that is, stable compounds of the type

$R_{j1-x}R''Scl_{1-y}Cr_y(BO_3)_4$;

here R_j= Y, La, Ce, Nd; R"= Gd, Eu, Lu. It turned out that these compounds form crystals of two different types of crystal structure, and the type of structure, and, consequently, the properties depend on the size of the rare-earth ions included in this crystal. If this size less than a certain critical value, then the compound crystallizes in the so-called trigonal syngony.In this case, the Rj ion is surrounded by six oxygen ions belonging to the BO_3 group with a covalent bond, so that the crystalline electric field has a low symmetry and is relatively weak. These circumstances determine the high efficiencies of laser transitions, on the one hand, and the detuning of almost all cross-relaxation transitions, on the other. This means that such a crystal can be used as a highly efficient highly concentrated medium for miniature lasers of medium power. In addition, it turned out that crystals of this group have a high non-linear dielectric susceptibility, so that for a certain orientation of the crystal relative to the propagation of laser radiation, infrared radiation of a non-dim laser can be converted into visible radiation. That is, a miniature laser is created on such crystals, emitting in the green region of the spectrum. Experiments have confirmed this.

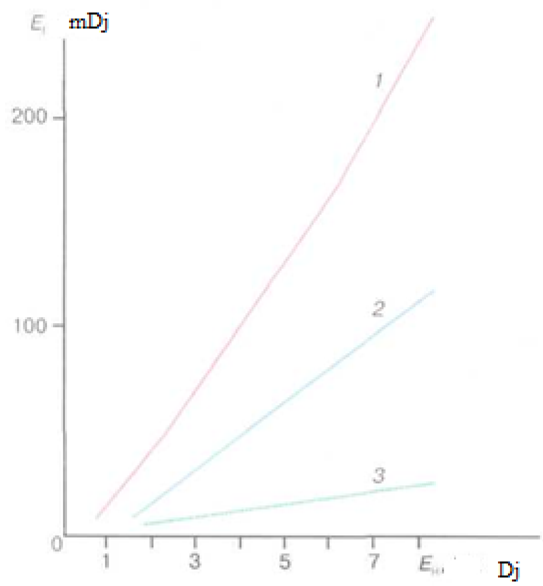

Fig. 1. Dependence of the output energy Er on the pump energy En in the free-running mode of lanthanum-scandium borate with chromium and neodymium (1),ISAG:Cr,Nd (2), and YAG:Nd(3).

One of the important drawbacks of all neodymium laser materials is the low average absorption coefficient of light in that part of the visible region of the spectrum where conventional pump lamps emit. This disadvantage is compensated by the introduction of additional impurities into the crystal, which strongly absorb light in this region of the spectrum and transfer energy to working ions, that is, neodymium ions. Scandoborates of rare earths allow the replacement of scandium ions by chromium Cr^{3+} ions, which significantly increases the efficiency of the laser as a whole. On fig. Figure 1 shows the dependence of the radiation energy of a neodymium scandoborate laser sensitized with chromium on the lamp pump energy in comparison with the same parameters of the most efficient garnet lasers. Recently, information has appeared in the literature that a total efficiency of 67% has been obtained with a scan-doborate neodymium laser pumped by a semiconductor LED. Thus, at present, the best material for medium-power solid-state miniature lasers seems to be neodymium scandoborate sensitized with chromium ions for lamp pumping or without a sensitizer for diode pumping.

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