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New potentials for the laser crystals with second and third optical nonlinearities

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Some new ideas for achieving stimulated-emission generation in laser crystals doped with Ln^{3+} ions are proposed. They are based on a pumping involving second harmonic generation and stimulated-Raman scattering phenomena. These ideas have been implemented experimentally.

The structural diversity of activated insulating crystals, their wide range of physical properties, and their essentially inexhaustible spectroscopic potential constitute a gold mine of opportunities for developing new operating laser schemes and principles for achieving stimulated emission (SE) in these crystals. Among the many known laser compounds [1], nonlinear (acentric) crystals are distinguished by a unique set of properties. In particular, they have already provided one of the most elegant advances in experimental physics and quantum electronics: crystalline lasers in which self-doubling of lasing frequency occurs (see, for example [2-5]). A distinctive feature of these lasers is that the SE is excited and its frequency is converted into the second harmonic in the same nonlinear laser element.

No less interesting is the "self-stimulated Raman scattering" which was recently observed: a frequency conversion of SE of the potassium-rare-earth tungstenates doped with Nd^{3+} ions [6,7]. Again, the effect occurs in a single laser crystal. In addition to the fundamental SE, it is accompanied by the generation of components at Stokes and anti-Stokes Raman-scattering frequencies.

These nonlinear-optics effects also underlie the new principles for achieving SE in activated crystals which we discuss below. Here, however, these effects are utilized in a completely different way. In the experiments which we just mentioned, the self-doubling (second harmonic generation) and stimulated-Raman conver-

sion resulted from intrinsic SE of laser crystals, and they led to an enrichment of the output spectra of these crystals [8]. In the operating schemes proposed below, in contrast, these effects are called on to play a different rôle: to convert the emission frequency of external pump lasers in order to match the active absorption bands of the lasing ions, i.e., to create the conditions required for achieving SE of these activator ions.

Our generation experiments in these new laser schemes used acentric LaBGeO_5 (space group C_3^2) and $\text{LiNbO}_3:\text{MgO}$ (C_{3v}^6) single crystals doped with Nd^{3+} ions and stimulated-Raman-active crystals of the $\text{KY}(\text{WO}_4)_2$ type (C_{2h}^6) doped with Er^{3+} ions, which are single-centered crystals with ordered structure. Fig. 1 explains the experimental conditions and the nonlinear-conversion processes involving absorption of external laser light in the matrix crystals and the laser channels for the excitation and generation of SE.

Let us look at some properties of these crystals. The $\text{LaBGeO}_5:\text{Nd}^{3+}$ crystal has an atomic neodymium concentration $C_{\text{Nd}}=1.4\%$. The angle between the geometric axis (F) and the crystallographic axis (c) is equal to the phase-matching angle, $Fc=\theta_m \approx 54^\circ$. The length is $l=12$ mm, and the diameter 4 mm. In the $\text{LiNbO}_3:\text{MgO}:\text{Nd}^{3+}$ crystal, the neodymium concentration is $C_{\text{Nd}} \approx 0.2\%$. In this case we have $\theta_m \approx 70^\circ$, $l=10$ mm, and a diameter of 5 mm. The $\text{KY}(\text{WO}_4)_2:\text{Er}^{3+}$ crystal has $C_{\text{Er}} \approx 30\%$, $F||b$, $l=40$ mm, and a diameter of 6 mm. The $\text{KGd}(\text{WO}_4)_2:\text{Er}^{3+}$ crystal has the same properties. The $\text{KEr}(\text{WO}_4)_2:\text{Er}^{3+}$ crystal has $C_{\text{Er}}=100\%$, $F||b$, $l=25$ mm, and a diameter of 6 mm. The $\text{KLu}(\text{WO}_4)_2:\text{Er}^{3+}$ crystal has $C_{\text{Er}}=25\%$, $F||b$, $l=30$ mm, and a diameter 6 mm. No anti-reflection coatings were applied to the plane-parallel ends of the laser elements in this stage of the research.

We have a few words regarding the "external" pump Q-switched lasers and hemispherical optical resonators used for the excitation of SE in our crystals containing Nd^{3+} (${}^4F_{3/2} \rightarrow {}^4I_{13/2}$ lasing channel) and Er^{3+} (${}^4I_{11/2} \rightarrow {}^4I_{13/2}$) in these new operating schemes. Two types of pulse pump lasers were used ($\tau_p=10-15$ ns, $E_p=10-15$ mJ): based on $\text{LaF}_3:\text{Nd}^{3+}$ ($\lambda=1.0406 \mu\text{m}$ (9609 cm^{-1})) and $\text{Al}_2\text{O}_3:\text{Ti}^{3+}$ ($\lambda=0.752 \mu\text{m}$

(13300 cm^{-1}) crystals. The polarized laser emission from these crystals was focused by a lens ($f=20\text{ cm}$) into the active elements through a plane mirror of the resonator (whose transmission at pumping wavelengths was about 80%). This mirror was pressed tightly against the active element. The spherical mirror differed in curvature. The radius of curvature was $r=5\text{ cm}$ for the experiments with the crystals containing Nd^{3+} ions, while it was $r=25\text{ mm}$ for the crystals with Er^{3+} activators. These mirrors had a high reflection coefficient (no less than 99%) both at the frequencies of the SE of the our activator ions and at the frequencies of the pump emission converted by the crystal-hosts. The spectra of the SE from these crystals was studied with the help of high-resolution grating monochromators and a GA-1M avalanche Ge-photodiode.

In these experiments we were most interested in the $\text{LaBGeO}_5:\text{Nd}^{3+}$ and $\text{KY}(\text{WO}_4)_2:\text{Er}^{3+}$ crystals. When the pump-power density exceeded a level of about 0.1 GW/cm^2 in the first of these crystals, the second harmonic ($\nu=19220\text{ cm}^{-1}$) excited SE of Nd^{3+} ions (by the absorption channels $^4\text{I}_{9/2} \rightarrow ^4\text{G}_{7/2,9/2}, ^2\text{K}_{13/2}$, followed by a nonradiative relaxation of the excitation to levels of metastable $^4\text{F}_{3/2}$ state, Fig. 1a). This crystal emitted π -polarized SE at the $1.3141\text{ }\mu\text{m}$ wavelength. The coefficient of the conversion of pump emission by the LaBGeO_5 crystal into second harmonic was about 5%. Despite the large quadratic nonlinear susceptibility $\chi^{(2)}$ of our second nonlinear crystal $\text{LiNbO}_3:\text{MgO}$, much higher pump densities were required to achieve SE of Nd^{3+} in that case.

In the monoclinic $\text{KY}(\text{WO}_4)_2:\text{Er}^{3+}$ crystal, SE was achieved on inter-Stark transitions ($\lambda_{\text{SE}}=2.6887$ and $2.8070\text{ }\mu\text{m}$) of the $3\text{-}\mu\text{m}$ self-saturating $^4\text{I}_{11/2} \rightarrow ^4\text{I}_{13/2}$ channel of Er^{3+} ions at a pump-power density of about 0.3 GW/cm^2 . As can be seen from Fig. 1b, Er^{3+} activators are excited in this case by emission at the first Stokes components, $\omega_s = \omega - \omega_{\text{RS}}$, of the pump emission at ω ($\omega_{\text{RS}} \approx 900\text{ cm}^{-1}$ is the frequency of the stimulated-Raman-active optical vibrational mode of the tetrahedral WO_4^{2-} complex of the crystal-host). In view of the high efficiency of stimulated-Raman conversion in crystals of this type, which ranges up to 60% [9], it can be assumed that their cubic nonlinear susceptibility $\chi^{(3)}$ is fairly high. Unfortunately, the efficiency of stimulated-Raman conversion in our experiments did not exceed 7-9%. The $3\text{-}\mu\text{m}$ SE of Er^{3+} ions in the three other monoclinic tungstenates was excited at approximately the same pump-power densities.

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FIG. 1. Simplified diagrams of the excitation of stimulated emission in spectrum-converting pumping processes in (a) nonlinear and (b) stimulated-Raman-active laser crystals doped with Ln^{3+} activator ions. The unconverted pump is represented by double arrows, and the converted pump by ordinary arrows. The heavy arrows show channels for stimulated emission of the Ln^{3+} ions, while the wavy arrows show radiationless processes.