Effect of XeCl laser irradiation on the defect structure of Nd:YAG crystals

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\textbf{A B S T R A C T}

This paper presents the effect of XeCl laser irradiation on Nd:YAG single crystal samples with various number of pulses at different repetition rates and laser fluences. Effects of the irradiation on the optical and structural properties of the crystal are analyzed by UV–vis-NIR spectroscopy. Annihilation of some point defects of the crystal structure is observed following laser irradiation at a fluence of 100 mJ cm\(^{-2}\) with 100 and 500 pulses. Increasing the laser fluence and pulse numbers leads to saturation and new defects are found to be formed in the crystal. Additional absorption spectra of the irradiated samples show that oxygen vacancies in the Nd:YAG crystals are removed during the low-dose irradiation. The laser irradiation is compared to the thermal annealing process for Nd:YAG crystal modification. Additional absorption spectrum of an annealed sample reveals that induced negative absorption band at 236 nm is correlated with the annihilation of the oxygen vacancy center. Our results also demonstrate that XeCl laser treatment has several advantages upon annealing at high temperatures in the Nd:YAG crystal quality improvement. Thus, the present work can give a new approach to modify Nd:YAG crystals to be used in a wide variety of solid-state laser engineering.

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\textbf{1. Introduction}

Nd:YAG is one of the most important active materials in the solid-state lasers due to its peculiar optical and mechanical characteristics. Recently its potential for developing as a scintillation detector has been also demonstrated [1].

Point defects affect the optical and structural properties of the ionic crystals. These defects include vacancies, substitutionals (antisites, un-controlled and controlled impurities), interstitials, Frenkel and Schottky defects. Indeed, real crystals are not perfect and certain defect subsystems form in any crystal systems because of the growth method, laboratory conditions and applying different technological factors for the crystal growth. Properties of the defect subsystem depend on the purity of constituent materials, growth and annealing environment, dopants, etc.

The common feature of oxide crystals such as YAG is the oxygen vacancy formation due to the annealing in the vacuum, argon or nitrogen gas during the growth process. The oxygen vacancy formation in the argon or nitrogen gas is accompanied by changing the valence of transition metal impurity (for example Fe\textsuperscript{3+} \rightarrow Fe\textsuperscript{2+}). Also it is known that during the Nd:YAG crystal growth process, the transition metal impurities, in particular Fe and Cr, substitute Al position in YAG crystal structure [2].

Vacancies can act as electron traps and suppress the luminescence of the crystal. When an oxygen vacancy (V\textsubscript{O}\textsuperscript{2+}) captures one electron, it forms F\textsuperscript{+} center (V\textsubscript{O}F\textsuperscript{+}) provided that the F\textsuperscript{+} center captures another electron change to natural vacancy named F center (V\textsubscript{O}). These defects have several absorption bands in the spectral range of 200–800 nm [3].

Influence of ionizing radiations on the optical properties of laser crystals has been studied experimentally [4–10]. Exposure to high energy particles or photons (e.g. electron, proton, neutron beams and \(\gamma\)-ray) can result in active ion recharging, new defects formation, electron ionization and structural damage in the crystals. These radiations often worsen the optical and lasing properties of the crystals due to the color center absorption at laser wavelength and absorption of pump radiation [10]. However, the effective energy transport to the active ions by color centers and recharged impurity ions in some laser crystals has been reported in a few papers [4,7,9].

Except for the work of Bedilov et al. [8], only destructive effects of ionizing radiations on the performance of Nd:YAG crystals have been reported [11]. Ionizing radiations generate many free electrons and change the valence of active ions in the crystal, too. V\textsubscript{O}\textsuperscript{2+} captures free electrons and forms F\textsuperscript{−} and F centers; this process continues till system reaches a new equilibrium condition on
which the new defect subsystem reduces the quantum efficiency of the laser crystal.

In fact, for the lattice system of a specific crystal, there is a definite defect subsystem that is generated during crystal growth process. When the crystal is irradiated by ionizing radiation, by absorbing the high energy photons and particles many electrons of the crystal lattice can excite and ionize and then many free electrons are generated in the crystal structure. The valence of impurity ions can change in the presence of the free electrons. New defects such as vacancies, F type centers or color centers, antisite ions, interstitial defects can also be formed in the crystal structure. Then, the initial defect subsystem changes and after the irradiation, crystal system reaches the equilibrium. In the new equilibrium state, many new defects are added to the crystal system. These new defects, by absorbing the pump and laser irradiation, greatly affect the optical properties and the performance of the laser crystal during laser operation [9,12].

On the other hand, it is known that laser irradiation has a peculiar feature to modify the optical and structural properties of various insulator materials [13–15]. The band-gap energy of these materials is higher than ultraviolet (UV) photons, so multi-photon process is the main mechanism for pumping electrons from valence to conduction band in the laser interaction with ultra-pure insulators in wavelength range from infrared (IR) to UV. However, all real crystals have a subsystem of point defects which is that their energy states lie in the band gap. The crystal defects strongly enhance the coupling between the laser irradiation and insulator. Moreover, some defects may be generated during laser irradiation and increase the effective absorption of the crystals. Thus, laser interaction with defect structure can modify optical properties of the crystals by changing the defect subsystem.

In this paper, the effect of XeCl excimer laser irradiation on the optical properties of Nd:YAG laser crystals is studied. Laser induced absorption modifications as well as generation and annihilation of crystal defects are characterized by UV–vis-NIR spectroscopy. In contrast to ionizing radiations, potentials of XeCl excimer laser irradiation in Nd:YAG laser crystal quality improvement are demonstrated. Our results show that low-dose irradiation of nanosecond XeCl excimer laser modifies the optical and structural characteristics of the Nd:YAG crystal.

2. Experimental section

The samples are homemade Nd:YAG single crystal discs which were prepared as follows: Al₂O₃, Y₂O₃ and Nd₂O₃ powder raw materials of 99.99% purity were melted at 1970 °C in an iridium crucible and the bulk of Nd:YAG single crystal (1 at% Nd³⁺) with the dimension of 115 × 935 mm² was grown by the Czochralski method along the [111] crystallographic direction with 5 arcsec parallelism in a pure Ar atmosphere. The rotation rate of the seed was 24 rpm and the pulling rate was kept constant at 0.6 mm/h. A crystal rod of 100 × 12.5 mm² was obtained from the crystal bulk for the experiments. It was cut into thin discs with 12.5 mm in diameter and 0.7 mm in thickness (≤ 1 arcsec parallelism). The obtained thin disc samples were polished on both sides (≤ λ/4 at 632.8 nm flatness and 80–50 scratch & dig) and then cleaned with sulfachromic acid and deionized water in a ultrasonic chamber to prevent any influence of pollutions on experimental results.

Radiation of a pulsed excimer laser system (Lambda Physik X200, λ = 308 nm, τ = 20 ns) was directed normally to the surface.

![Fig. 1. Experimental setup of XeCl laser irradiation on Nd:YAG crystal samples. 300 mm focal length lens was used when more than 100 mJ/cm² laser fluence was required.](image)

![Fig. 2. (a)Transmission and (b) absorption spectra of un-irradiated sample.](image)
of Nd:YAG crystal samples under environmental conditions. Effects of laser fluence, pulse number and repetition rate are shown schematically in Fig. 1.

The single pulse damage threshold fluence of the Nd:YAG crystals at 308 nm was measured in our laboratory and obtained to be about 1500 mJ cm\(^{-2}\). Then, to study nondestructive effects of nanosecond excimer laser irradiation, the samples were irradiated at the laser fluences of 100 and 500 mJ cm\(^{-2}\) with 100, 500 and 1000 pulses at 1, 10 and 25 Hz repetition rates.

The laser fluence was calculated by measuring the energy and laser spot size on the sample surface. In the case of 100 mJ cm\(^{-2}\) laser fluence, the laser output energy and spot size were 300 mJ and 30 \(\times\) 10 mm\(^2\) rectangle, respectively. For obtaining a laser fluence of 500 mJ cm\(^{-2}\), a plano-concave lens of 300 mm focal length was used. The sample was placed at a distance from the lens so that laser spot size on the sample surface becomes 60 mm\(^2\).

One sample was annealed for 3 h at 400 °C for thermal relaxation and then 3 h at 1100 °C for changing the defect structure in the air. The optical absorption of samples was recorded by a (Cary 500) UV–vis-NIR spectrophotometer (200–800 nm) before and after the irradiation. Defects in oxide crystals can be studied by the optical absorption spectrum. Change of the defect energy state leads to new optical absorption [16].

To determine the type and amount of changes in defect structure, the value of laser induced additional absorption (AA) is calculated by [4]

$$\Delta k = \ln(T_1/T_2)/d$$  \hspace{1cm} (1)

where \(k\) is the absorption coefficient, \(d\) is the sample thickness and \(T_1\) and \(T_2\) are the crystal transmissions before and after irradiation, respectively.

### Table 1

<table>
<thead>
<tr>
<th>The samples</th>
<th>Pulse number (mJ/cm(^2))</th>
<th>Laser fluence (mJ/cm(^2))</th>
<th>Repetition rate (Hz)</th>
<th>Absorption relative to un-irradiated sample</th>
</tr>
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<tbody>
<tr>
<td>1</td>
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<td>1</td>
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<td>2</td>
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<tr>
<td>6</td>
<td>100</td>
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3. Result and discussion

Fig. 2(a) and (b) shows the transmission and absorption spectra, respectively of an un-irradiated sample. XeCl laser irradiation parameters are summarized in Table 1. To study the effect of the laser pulse numbers, the sample nos. 1, 2 and 3 were irradiated at the fluence of 100 mJ cm\(^{-2}\) and 1 Hz repetition rate with 100, 500 and 1000 pulses, respectively.

Fig. 3(a) shows the comparison of the UV–vis optical transmission spectra of the sample nos. 1, 2 and un-irradiated sample. Also the comparison between sample no. 3 and un-irradiated sample was shown in Fig. 3(b). Optical transmissions of sample nos. 1 and 2 increased while it decreased for the sample no. 3 in comparison to the un-irradiated sample.

It has been shown that increase and decrease of optical absorption in the entire spectral range are the characteristic features of generation and annihilation of point defects in the materials, respectively [12]. Reduced absorption of sample nos. 1 and 2 is shown in Fig. 3(a) from which it can be seen that the XeCl laser at low-dose irradiation has a curing effect on the Nd:YAG crystal structure by removing some of its intrinsic point defects.

We observed that optical transmission and so the curing effect increases initially, reaches a maximum at a certain number of pulses (\(\sim 550\) pulses at the fluence of 100 mJ cm\(^{-2}\) and 1 Hz repetition rate) and then gradually decreases with increasing the number of pulses. After surface irradiation with a certain number of pulses (\(\sim 700\) pulses at the fluence of 100 mJ cm\(^{-2}\) and 1 Hz repetition rate) the curing effect would vanish. It can be proposed that new defects are found to be formed in the crystal by receiving more laser pulses (Fig. 3(b)).

In addition, the decrease of the optical absorption of the irradiated Nd:YAG in the UV region (Fig. 3(a)) causes to reduce the heat generation in the crystal during the flash lamp pumping process. Thus, application of the low-dose XeCl laser irradiated samples as an active medium in the flash lamp pumped solid state laser has an important advantage due to reduced thermal effects.

In flash lamp–pumped laser systems, the broad spectral distribution of the pump source causes a certain amount of background absorption by the laser host material, particularly in the UV regions of the lamp spectrum. Absorption of lamp radiation by impurity atoms and defects can further increase heating. Thus, annihilation of defects and reduced absorption in UV spectral range decrease the heat generation during the pumping process.

![Fig. 3](image-url)
In the following, Nd:YAG crystal sample nos. 4 and 5 were irradiated at a fluence of 100 mJ cm$^{-2}$ with 100 pulses at 10 and 25 Hz repetition rates. Fig. 4 shows the comparison among three transmission spectra of sample nos. 1, 4 and 5. As shown in Fig. 4, the effect of crystal structure curing by excimer laser irradiation is more pronounced at higher repetition rates; it may be due to the temperature increase of the sample during the laser treatment and incubation effect.

In the following step, the effect of XeCl laser fluence on the optical absorption of Nd:YAG crystal was examined. To achieve this purpose, sample no. 6 was irradiated at a fluence of 500 mJ cm$^{-2}$ and 1 Hz repetition rate with 100 pulses. A comparison of transmission spectra of sample nos. 1 and 6 with un-irradiated sample is shown in Fig. 5. This profile shows that optical transmission of the sample no.6 has reduced relative to un-irradiated sample due to the generation of new defects in the XeCl laser irradiated crystal at this regime of laser fluence.

To compare the effect of XeCl laser irradiation with annealing at high temperature, one sample was annealed for 3 h at 400 °C and then for 3 h at 1100 °C. As mentioned above, annealing removes some oxygen vacancies in the Nd:YAG crystals and annihilation of the point defects creates negative bands in additional absorption spectrum. Then, there should be a negative band related to the annihilation of the oxygen vacancies in the additional absorption spectrum of the annealed sample, but it is observed no negative band in the additional absorption spectrum except the 236 nm negative absorption band. Thus, we can conclude that the 236 nm negative band is just related to the annihilation of the oxygen vacancy center.

Fig. 6 shows additional absorption spectra of the sample nos. 1, 2, 4 and 5 which become more transparent after XeCl laser irradiation. A broad negative AA band at 236 nm is also observed for all these irradiated samples. As demonstrated in this work, the 236 nm absorption band in additional absorption spectrum is related to the annihilation of the oxygen vacancy centers in Nd:YAG crystals.

The physical mechanism of the relation between 236 nm absorption band and annihilation of oxygen vacancies is based on the created energy levels in the band gap of the crystal related to the vacancy defects. The existence of a vacancy induces distortion of the local geometry around it. An oxygen vacancy ($V_{\text{O}}^{\text{2+}}$) behaves like a double positively charged vacancy. So, in comparison with the original interaction between the oxygen and surrounding ions, there are only weak electrostatic attractions or repulsions between a vacancy and its surrounding ions, which explains the distortion of the surrounding ions. The nearest two Y and two Al ions surrounding the vacancy are displaced away from

Removing some oxygen vacancy centers [17]. To determine the type and value of annealing and XeCl laser induced annihilated and generated defects, additional absorption (AA) spectra of the irradiated and annealed samples were obtained.

Fig. 6 shows the AA absorption spectrum of the annealed Nd:YAG crystal with the broad absorption band centered at 256 nm, which has been observed in air annealed Nd:YAG and YAG crystals [16]. It has been attributed to charge transfer from oxygen ion to Fe$^{3+}$ ion. Generally, some uncontrolled Fe$^{3+}$ and Fe$^{2+}$ ions often exist as impurities in Nd:YAG crystals. XRF analysis also detects about 400 ppm of iron ions in the Nd:YAG crystal samples which were used in the experiments. Some oxygen vacancies can be filled by oxidizing Fe$^{2+}$ impurity during annealing in air and then the number of Fe$^{3+}$ ions localized near the oxygen ion will increase.

In addition, Fig. 6 shows a sharp negative absorption band at 236 nm that has not been reported earlier to the best of our knowledge. As mentioned above, annealing removes some oxygen vacancies in the Nd:YAG crystals and annihilation of the point defects creates negative bands in additional absorption spectrum. Then, there should be a negative band related to the annihilation of the oxygen vacancies in the additional absorption spectrum of the annealed sample, but it is observed no negative band in the additional absorption spectrum except the 236 nm negative absorption band. Thus, we can conclude that the 236 nm negative band is just related to the annihilation of the oxygen vacancy center.
the vacancy site. Therefore, the new defect energy levels are induced around the VB or CB edges. This charge localization is due to the Al 3s, 3p orbitals, the Y-5p, 4d orbitals, and the partial O 2p orbital extending to the area of the vacancy. Although the Al 3s, 3p and the Y 4d, 5p orbitals constitute the CB in perfect YAG, such localization of the Al 3s, 3p and the Y 4d, 5p orbitals will shift the energy level downward from the CB edge, resulting in the formation of a defect level in the band gap. The optical absorption spectrum is important for studying defects in YAG. The above mentioned defect states will lead to new optical absorption.

In Ref. [16] results of the plane wave density functional theory (DFT) calculation of various point vacancies in crystal YAG have been presented. First-principle method that is a powerful theoretical tool for acquiring accurate defect levels and optical properties of charged vacancies in metal oxides has been used to theoretically characterize the point vacancy in YAG and to explain the experimental optical absorption spectrum. The results of Ref. [16] about oxygen vacancy are as follows: “A charged vacancy introduced new defect states in the band gap. For the oxygen vacancy, \( V_{O}^{2+} \) induced a shallow level near the bottom of the CB”. These theoretical results are in good agreement with our experimental results.

Additional absorption spectrum of sample no. 6 is shown in Fig. 8. The sharp additional absorption peak at 236 nm is related to the generation of a large number of oxygen vacancies in the laser treated Nd:YAG crystal. It is known that oxygen vacancy can be formed more easily than the other defects in oxide crystals [12]; hence after vanishing the curing effect, increased energy absorption of the laser irradiation by Nd:YAG crystal leads to the oxygen vacancy generation.

Fig. 8 also shows AA band at \(~212\) nm accompanied by the negative AA band at \(~220\) nm. These two peaks which were reported previously (in Ref. [3]) are correlated with F and \( F^{+} \) center in YAlO₃ (YAP) crystals, respectively [3]. Then, it also can be attributed to F and \( F^{+} \) centers in Nd:YAG due to YAG and YAP crystal constituent elements similarity. The responsible process can be expressed as: \( F^{+} + e^{-} \rightarrow F \).

High-dose laser irradiation creates a large number of electrons and holes in the optical crystals. As grown \( F^{+} \) centers can capture one of the excited electrons and change to the F centers. Then, decreasing the value of \( F^{+} \) center concentrations leads to the negative AA value at \(~220\) nm while the increasing of F center concentrations causes the positive band at \(~212\) nm.

The AA spectrum of sample no. 3 is presented in Fig. 9. The characteristic behavior of defects in this sample is somewhat different from that of the previous sample. In this case, there is a continuum absorption band in the region of 234–348 nm.

At higher dose irradiation, in addition to oxygen vacancies and F centers, other defects such as \( O^{-} \) hole centers, active impurities in different valences, antisites, aggregated and compound defects may be formed. Thus, the continuum absorption band in Fig. 9 belongs to the complicated defect subsystem. However, the degree of laser inducing crystal defect after irradiation strongly depends on its initial quality, the type of active impurities and concentration of dopants.

As mentioned above, Nd:YAG crystal annealing at high temperatures in air is a method to improve its quality for using as an active medium. A broad absorption band at 256 nm correlated with \( \text{Fe}^{3+} \) was observed after annealing. But this peak was not seen in the case of the laser irradiated crystals. \( \text{Fe}^{2+} \) ions oxidation helps to the oxygen vacancy annihilation during annealing process as follows.

It has been shown that a divalent impurity in the Nd:YAG crystal tends to be placed near an oxygen vacancy to form neutral aggregate in the garnet crystal lattice [18]. On the other hand, at

![Fig. 7. Additional absorption spectra of samples no. 1 (100 mJ cm⁻², 100 pulses, 1 Hz), no. 2 (100 mJ cm⁻², 500 pulses, 1 Hz), no. 4 (100 mJ cm⁻², 100 pulses, 10 Hz) and no. 5 (100 mJ cm⁻², 100 pulses, 25 Hz).](image)

![Fig. 8. Additional absorption spectrum of the sample no. 6 (500 mJ cm⁻², 100 pulses, 1 Hz).](image)

![Fig. 9. Additional absorption spectrum of sample no. 3 (100 mJ cm⁻², 1000 pulses, 1 Hz).](image)
high temperatures Fe$^{2+}$ ion oxidizes and fills the adjacent oxygen vacancy. An oxygen ion of the adjacent oxygen vacancy can fill the oxygen vacancy and oxidize Fe ion. Thus, the oxygen vacancy moves away from Fe ion. Then, due to the local charge imbalance, this state is unstable and oxygen vacancy gets away and migrates in the crystal until it reaches the surface of the Nd:YAG crystal and would be oxidized. Indeed, Fe ion is a primary stimulant in this process.

We believe that the mechanism of oxygen vacancy annihilation by laser irradiation is different from annealing. The energy required to migrate an oxygen vacancy in the Nd:YAG crystal, regarding its position relative to the other ions is in the range of 1.5–3 eV [19]. When an oxygen vacancy receives a photon of $\hbar\nu \sim 4.02$ eV, it can move inside the crystal, reach the surface and finally be oxidized. Each laser pulse includes too many photons and can remove many oxygen vacancies. In addition, the captured electrons at some oxygen vacancies (F and F$^-$ centers) receive the energy of photons and recombine with a hole such as O$^-$ hole center and then the remaining oxygen vacancy gets removed as well. While in annealing, each oxygen vacancy with increasing temperature up to the melting point of Nd:YAG crystal receives the energy of $\sim 0.1$ eV and it cannot migrate in the crystal alone in the absence of active ions (here Fe$^{2+}$ ions).

Also, we believe that the sharpness of the 236 nm band in the case of annealed sample is related to the annihilation of the oxygen vacancies located adjacent to Fe$^{2+}$ ion while in the case of laser treatment the oxygen vacancies in all energy states (regarding different locations relative to other ions such as Al, Y,$\ldots$) can annihilate and for this reason the 236 nm band becomes broad in the additional absorption spectrum of laser irradiated samples.

Annihilation of oxygen vacancies by laser irradiation continues till this process is saturated and further absorbed energies generate new defects in the crystal. Multi-photon absorption can ionize or excite electron to the conduction band; then many O$^-$ hole centers produce and trapping centers capture free electrons and holes, and active impurities change their valence. When an oxygen vacancy receives a photon of $\hbar\nu > 4.02$ eV, it can be removed from the crystal surface and an oxygen vacancy remains there.

Consequently, the defect structure in the crystal gradually changes and new defects appear with the new optical absorption bands. In this condition, the effective energy absorption of the crystal and also the coupling between the laser irradiation and the crystal increases till the breakdown happens in the crystal.

A comparison of additional absorption quantity correlated with annihilation of oxygen vacancies in the case of annealing (Fig. 6 $\Delta k \sim -0.5$) and laser treatment (Fig. 7 $\Delta k \sim -10$) reveals that the oxygen vacancy centers are removed partially by annealing while all the oxygen vacancy centers, F$^+$ and F centers centers can be possibly annihilated in the case of laser irradiation by controlling the irradiation parameters. In addition, annealing is a time consuming process and requires certain temperature program for increasing temperature and slow cooling processes, whereas using laser beam makes the experiment much faster and cheaper.

On the other hand, the curing effect of the as grown defects of Nd:YAG crystal that take place at low-dose irradiation is due to low photon energy of XeCl excimer laser ($\sim 4.2$ eV) in comparison to ionizing radiation (typically $\sim 1$ MeV). No curing effect can occur in crystal interaction with high energy photons and particles due to ionization effect of crystal ions or atoms even at low-dose irradiation.

In fact, the required energy for electron excitation from valence to conduction band in the YAG crystal is $\sim 7$ eV (band gap of the crystal) and the energy of one particle or photon of ionizing radiation (typically $\sim 1$ MeV) produces many free electrons in the crystal structure. The free electrons may be captured by any trapping center in the crystal or excite another electron or ion. Therefore, by irradiating the crystal with ionizing radiation, many new defects are formed in the crystal while in the case of XeCl laser irradiation, each photon has the energy of 4.2 eV that is insufficient to ionize electron or excite it to the conduction band. As discussed above, this energy can help movement and annihilation of the oxygen vacancies.

4. Conclusion

Summarizing, we demonstrated a potential of XeCl excimer laser irradiation to improve performance of the Nd:YAG crystal in the laser and the scintillation detector applications.

The effect of the pulse numbers, repetition rates and laser fluences on the crystal modification was examined. It was observed that higher repetition rates have a more pronounced curing effect. It was also shown that at low-dose irradiation, by increasing the pulse numbers and laser fluences, the curing effect increases. On further increasing the dose of the laser irradiation new defects generate in the crystal.

XeCl laser induced the absorption coefficient reduction, especially in the UV region, leading to reduced thermal effects due to reduced pump absorption of point defects. In addition, the curing effect due to the point defect annihilation improves optical and structural features of the Nd:YAG crystals.

The additional absorption spectrum of the annealed sample can be demonstrated as the negative absorption band at the 236 nm is related to the partial removal of some oxygen vacancy centers in the Nd:YAG crystal. The comparison of the additional absorption spectra of the laser irradiated samples with the annealed sample reveals that the number of annihilated oxygen vacancies by the laser irradiation is remarkably more than by the thermal treatment.

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